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Traditional and new POPs in environments along the Bohai and Yellow Seas: An overview of China and South Korea

Jing Meng [a, b, 1], Seongjin Hong [c, 1], Tieyu Wang [a, b, *], Qifeng Li [a, b], Seo Joon Yoon [d], Yonglong Lu [a, b], John P. Giesy [e, f], Jong Seong Khim [d, **]

[a] State Key Lab of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
[b] University of Chinese Academy of Sciences, Beijing 100049, China
[c] Department of Ocean Environmental Sciences, Chungnam National University, Daejeon 34134, Republic of Korea
[d] School of Earth and Environmental Sciences & Research Institute of Oceanography, Seoul National University, Seoul 08826, Republic of Korea
[e] Department of Veterinary Biomedical Sciences & Toxicology Centre, University of Saskatchewan, Saskatoon, SK, Canada
[f] Department of Zoology & Center for Integrative Toxicology, Michigan State University, East Lansing, MI, USA

HIGHLIGHTS

- Spatio-temporal distribution of classic and emerging POPs along Yellow Sea reviewed.
- Hot spots for POPs pollution and associated sources or local activities characterized.
- Sources of target POPs were independent each other both in local and national scale.
- OCPs, PCBs and PFASs were widely contaminated groups in China and South Korea.

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ABSTRACT

Rapid economic growth during the past two decades in the region surrounding the Bohai and Yellow Seas has resulted in severe pollution. Large amounts of monitoring data on persistent organic pollutants (POPs) in various environmental media have been accumulated, which allows us to conduct a fairly comprehensive assessment of the region around the Bohai and Yellow Seas to elucidate spatial patterns of pollution on a regional scale. This review summarized distributions of traditional and new POPs, including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), hexabromocyclododecanes (HBCDs), and perfluoroalkyl substances (PFASs), in various environmental media. In general, due to their physico-chemical properties (poor solubility in water), OCPs and PCBs were mainly detected in sediments, PBDEs and HBCDs were mainly detected in sediments and soils. PFASs, which have greater solubility, were mainly detected in the hydrosphere. For conventional POPs, such as OCPs and PCBs, Bohai Bay and Haihe River in China, Gyeonggi Bay and Lake Sihwa in South Korea were found to be most polluted areas. While for new POPs, such as PBDEs, HBCDs and PFASs, some areas were heavily polluted due to local production and applications. Estuarine and coastal areas of the Bohai Sea were more severely contaminated by POPs than coastal regions of the Yellow Sea. Overall, the present review will guide identification of key areas for strengthening risk assessment of POPs and management practices.

1. Introduction

Persistent organic pollutants (POPs) have been of great concern during the last few decades due to: a) persistence, b) long-range transport, c) potential to be bioaccumulated by organisms, and d) potential toxic effects on humans and wildlife (Nadal et al., 2015). The Stockholm Convention on POPs aims to protect humans and

* Corresponding author. State Key Lab of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China.
** Corresponding author.
E-mail addresses: wangty@rcees.ac.cn (T. Wang), jskocean@snu.ac.kr (J.S. Khim).
[1] These authors contributed equally to this work.

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the environment from hazardous and persistent chemicals by reducing or eliminating their production and emission. The convention, which was adopted in 2001, came into force in 2004. The initial list included twelve chemicals called the “dirty dozen”, which included organic chlorinated pesticides (OCPs) (including dichlorodiphenyltrichloroethanes, DDTs) and polychlorinated biphenyls (PCBs) (UNEP, 2001). In 2009, nine new chemicals were added and came into force one year later. The added chemicals included hexachlorocyclohexanes (HCHs), polybrominated diphenyl ethers (PBDEs) and perfluoroalkyl substances (PFASs) (e.g., perfluorooctane sulfonic acid (PFOS) and its salts, perfluorooctane sulfonyl fluoride) (UNEP, 2009). Hexabromocyclododecanes (HBCDs) were also listed in Annex A of the Stockholm Convention on POPs in 2013 (UNEP, 2013).

HCHs and DDTs are two main components of OCPs that historically have been used extensively in agriculture and sanitation (Zhang et al., 2013a). PCBs are widely used in transformers, capacitors, and printing ink, among which trichlorobiphenyl with pentachlorobiphenyl is the most prevalent congener (Erickson and Kaley, 2011). PBDEs, often produced as penta-, octa-, and deca-BDE commercial mixtures, are major constituents of brominated flame retardants (BFRs) broadly used in electronics and household products (La Guardia et al., 2006). Penta-BDE and octa-BDE have been banned due to the adverse effects on biota and their increasing concentrations in the environment. However, deca-BDE is still being produced and used (Zhu et al., 2009). HBCDs have become an important alternative BFR after production and use of PBDEs were restricted (Xia et al., 2011). They are primarily added into expanded and extruded polystyrene which are used in building insulation, and are also employed in electrical equipment, great impact polystyrene and textile coatings (POPRC, 2011). PFASs have been widely used in the past six decades as additives in surface coatings of carpets, leather, papers, and textiles, as well as in firefighting foams, pesticides, paints, etc. PFOS and perfluorooctanoic acid (PFOA) are the most frequently detected PFASs in the environments (Kannan, 2011).

Distributions of POPs in environmental media are strongly dependent on their physico-chemical properties (Wania and Mackay, 1996). POPs have generally hydrophobic nature with log K_{ow} values ranging from 3 to 7 (Jones and De Voogt, 1999; Kelly et al., 2007). Thus, POPs have low water solubility, great bioaccumulation potential, low rates of degradation, and high adsorption onto sediments. When POPs are transported from river to estuary, due to the great ionic strength, solubility of POPs decreases in seawater compared to that in freshwater. Consequently, adsorption of POPs onto suspended particles and sediments increases in estuarine areas (Hong et al., 2013; Turner and Millward, 2002; Turner and Rawling, 2001). Thus, POPs are largely scavenged by adsorption onto suspended particles, less delivered to remote regions by oceanic current, and accumulated in sediments in estuarine and coastal areas. Finally, POPs can persist in coastal sediments for long periods of time and can cause adverse effects on benthic organisms. However, water soluble POPs, such as PFASs occurred in water column and can be delivered to remote regions by oceanic currents (Hong et al., 2013, 2015; Naile et al., 2010).

The Yellow and Bohai Seas, together with surrounding coastal areas, are major commercial zones and have undergone massive urbanization and industrialization. As one of the most developed regions, they are surrounded by several metroplexes, such as the Chinese cities of Beijing, Tianjin, Dalian and Qingdao, and Korean cities including Seoul, Incheon, Asan, Gunsan, and Mokpo. Intensive anthropogenic activities in these regions have severely deteriorated environmental quality, especially along the coast (Hong et al., 2012, 2016; Naile et al., 2010, 2011, 2013). Several rivers, including the Liaohe, Haihe and Yellow from China, and the Han, Geum and Youngsan from South Korea, empty into the Bohai and Yellow Seas, respectively. Industrial and municipal wastewater from surrounding cities is also discharged. In addition, the semi-enclosed terrain makes exchange of water between the Bohai and Yellow Seas and the open ocean relatively slow. Thus, pollutants tend to be contained in these rather enclosed areas. The Bohai and Yellow Seas together, have become one of the most heavily polluted sea areas and thus act as a sink and possible long-term source of pollutants including POPs. In addition, the Bohai and Yellow Seas are also one of the most important fishery stocks, and they provide amounts of seafood to peripheral cities. Therefore, pollution as a potential source of exposure of humans, has caused concerns in the past decades. This review will support basic information to master overall pollution status and assess ecological risks, while help scientists identify future research direction.

2. Review methodology

Contamination of environmental media by POPs along the Bohai and Yellow Seas in China and South Korea, respectively, have been extensively studied during the last 20 years. Large amounts of monitoring data on POPs have been accumulated which allow us to conduct a fairly comprehensive assessment of coastal areas of China and South Korea and elucidate spatial patterns of pollution on a regional scale.

In the present study, we collected and reviewed results of previous studies on POPs in environmental media, such as sediments, soils, and water from the Bohai and Yellow Seas, specifically including Bohai Sea and north Yellow Sea from Dalian Bay to Haizhou Bay in China, and West Sea of South Korea from north Incheon Harbor to south Youngsan River. POPs focused on: a) chlorinated POPs (OCPs and PCBs), b) brominated POPs (PBDEs and HBCDs), and c) fluorinated POPs (PFASs) (Table 1). A total of 101 previous studies on distributions of POPs in environmental media were reviewed (details in Tables S1 and S2 in Supplementary Materials).

Study efforts on each chemical group were generally comparable except for new POPs such as PBDEs and HBCDs. Among the environmental media, POPs in sediments have been extensively studied, followed by soils and water. In particular, because of their low solubility in water and hydrophobic characteristics, studies of POPs except for PFASs in water samples were scarce. Thus, in the present study, distributions of POPs focused on OCPs, PCBs, PBDEs and HBCDs in sediments and PFASs in water collected from the area in and surrounding the Bohai and Yellow Seas. At the beginning of studies of POPs, most studies were limited to reporting concentrations of traditional POPs such as OCPs and PCBs. Later, several studies have reported information on new POPs, such as PBDEs, HBCDs and PFASs.

In both China and South Korea, studies of POPs in sediments have shown a preponderance of certain areas, such as greatly polluted regions (Table 1 and Fig. 1). Fifteen regions in areas surrounding the Bohai Sea and north Yellow Sea in China, including: Dalian Bay, Liaohe River, Daliao River, Shuangtaizi River, Daling River, Liaodong Bay, Haihe River, Tianjin, Bohai Bay, Yellow River, Xiaoqing River, Laizhou Bay, Qingdao, Jiaozhou Bay, and Haizhou Bay and nine regions of the West Sea of South Korea, including: Incheon Harbor, Gyeonggi Bay, Lake Sihwa, Namyang Bay, Asan Bay, Taean Coast, Geum River, Saemangeum Coast, and Youngsan River have been studied.

In the present study, we collected and reviewed major findings that determined pollution of POPs in environmental media (sediments, soils and water) of the Bohai and Yellow Seas during the past two decades. The present article is aimed to specifically document spatial distribution, sources and potential risks of five groups of

Table 1
Summary of studies of chlorinated and brominated POPs in sediments and fluorinated POPs in water of regions along the Bohai and Yellow Seas in China and South Korea (locations are presented in Fig. 1).

Countries and regions	Types	Survey year	Chlorinated POPs		Brominated POPs		Fluorinated POPs
			OCPs	PCBs	PBDEs	HBCDs	PFASs
Chinese Coasts							
C1. Dalian Bay	B	1996	V				
C2. Liaohe River	E, R	2004–2005, 2009–2010, 2012, 2015	V	V	V		V
C3. Daliao River	B, E	1996, 2001–2007	V	V	V		
C4. Shuangtaizi River	E	2013	V	V			
C5. Daling River	R	2009, 2011					V
C6. Liaodong Bay	E	2008, 2013					V
C7. Haihe River	E, R	2004, 2006, 2007–2010	V	V	V		V
C8. Tianjin	E	2009, 2010			V	V	
C9. Bohai Bay	B, C, E	2004, 2006–2013	V	V	V		V
C10. Yellow River	E	2012	V				
C11. Xiaoqing River	R	2014					V
C12. Laizhou Bay	B, E, R	2007, 2009–2010, 2014			V	V	
C13. Qingdao	C	2006, 2007		V			
C14. Jiaozhou Bay	B, C	1996, 2006–2007, 2009, 2014	V	V		V	
C15. Haizhou Bay	C	2001		V			
Korean Coasts							
K1. Incheon Harbor	B	1996, 2001–2003, 2005–2007	V	V	V	V	
K2. Gyeonggi Bay	B, R	1996, 2000–2005, 2008–2009, 2012	V	V	V	V	V
K3. Lake Sihwa	I, L	1996, 1998, 2000, 2005, 2008–2012	V	V	V	V	V
K4. Namyang Bay	B	1996	V				
K5. Asan Bay	B	2001–2003, 2005, 2008, 2010–2012	V	V			V
K6. Taean Coast	C	2001–2003, 2008, 2010–2012	V	V			V
K7. Geum River	E, R	2001–2004, 2008–2012	V	V			V
K8. Saemangeum Coast	C, R	2001–2003, 2006–2007, 2009	V	V			V
K9. Youngsan River	B, C, E, I, R	2001–2003, 2006–2012	V	V			V
Data present in			Fig. 2	Fig. 3	Fig. 4a	Fig. 4b	Fig. 5

B: bay; C: coast; E: estuary; I: inland; L: lake; R: river.

POPs in the environment for future monitoring and risk assessment use. Overall, the present review would provide the most up-to-date information, list of studies on POPs in the Bohai Sea and Yellow Sea regions, and better understanding on pollution of POPs. In addition, this study will guide identification of hotspot areas and prioritization of research needed concerning monitoring, assessments, and management practices of POPs.

3. Chlorinated POPs in the environment

3.1. OCPs

Technical HCHs and DDTs were widely used in China from the 1950s until their production was officially banned in 1983 (Gong et al., 2007). Studies on OCPs in the Bohai and Yellow Seas started in the 1990s (Lu et al., 2007b). Occurrences of OCPs in sediments from rivers, estuaries, intertidal zones, bays, and seas have been widely investigated, especially in adjacent rivers and estuaries (Fig. 2). Overall, concentrations in sediments from rivers were higher than those from the open ocean. OCPs from rivers flowing into Bohai Bay were relatively higher (Li et al., 2013), followed by those from Jiaozhou Bay (Wang et al., 2007). In the northern Bohai Sea and its coastal area, concentrations of HCHs and DDTs were generally less than 100 ng g^{-1} dw (Gao et al., 2015; Li et al., 1998; Wang et al., 2007; Wang, 2013; Yuan et al., 2015). In coastal area of Bohai Bay, concentrations of HCHs and DDTs were generally higher and mainly exceeded 100 ng g^{-1} dw (Gan, 2010; Li et al., 2013; Qin et al., 2010; Wang, 2010; Zhao et al., 2010). Increasing trend of HCHs and decreasing trend of DDTs were observed from 2004 to 2010. While in Bohai Bay, concentrations of HCHs and DDTs decreased and all those were less than 50 ng g^{-1} dw (Gan, 2010; Hu et al., 2014; Liu et al., 2012; Wang, 2010; Zhang, 2009a). HCHs and DDTs generally both showed decreased trend from 2004 to 2012. In Jiaozhou Bay and its coastal area, while concentrations of HCHs and DDTs were less than 100 ng g^{-1} dw (Cao, 2008; Zhang, 2009b; Zhou et al., 2012). Concentrations of HCHs and DDTs varied greatly among areas. Relatively great concentrations were found in sediments from Haihe River (Zhao et al., 2010). The highest concentration of HCHs was detected in lower reaches of the Haihe River, with a value of 11,806 ng g^{-1} dw (Li et al., 2013). DDTs in sediments were generally lower than that of HCHs, with the highest concentration of 1417 ng g^{-1} dw in coastal area of Bohai Bay (Ma et al., 2001). When comparing concentrations of residues in sediments with quality standards, HCHs from the most study areas were higher than probable effect level (PEL, 0.99 ng g^{-1} dw) (Feng et al., 2011), maybe posing certain harm to environment and wildlife. Most DDTs were lower than effect range median (ERM, 46.1 ng g^{-1} dw) (Feng et al., 2011), but these sites with higher than ERM would be subjected to frequently adverse biological effects, such as the Haihe River and Bohai Bay.

In South Korea, HCHs and DDTs were banned for agricultural use in 1969 under the Agro Chemicals Control Act (ACCA) (Kim and Yoon, 2014). Along the west coast of South Korea, HCHs and DDTs were widely distributed in sediments (Fig. 2) (Choi et al., 2010, 2011a, 2011b, 2014; Hong et al., 2006a, 2009; Khim et al., 1999; Koh et al., 2005; Lee et al., 2001b). Greater concentrations of HCHs were found in inland sediments of Lake Sihwa (0.55–10.7 ng g^{-1} dw) (Koh et al., 2005). Concentrations of HCHs were relatively great in sediments of industrialized and urbanized regions, with some concentrations of HCHs exceeded the PEL. Although their production and use had mostly been banned since the late 1960s, DDTs were still found in various marine samples and were widely distributed along the coasts of South Korea. In South Korea, approximately 758 tons and 1320 tons of DDT were produced and imported from 1949 to 1971. This study found that coastal sediments of the West Sea were widely contaminated by DDTs (Fig. 2), which was the major organochlorine pesticide. Incheon Harbor was identified as an area of concern because of

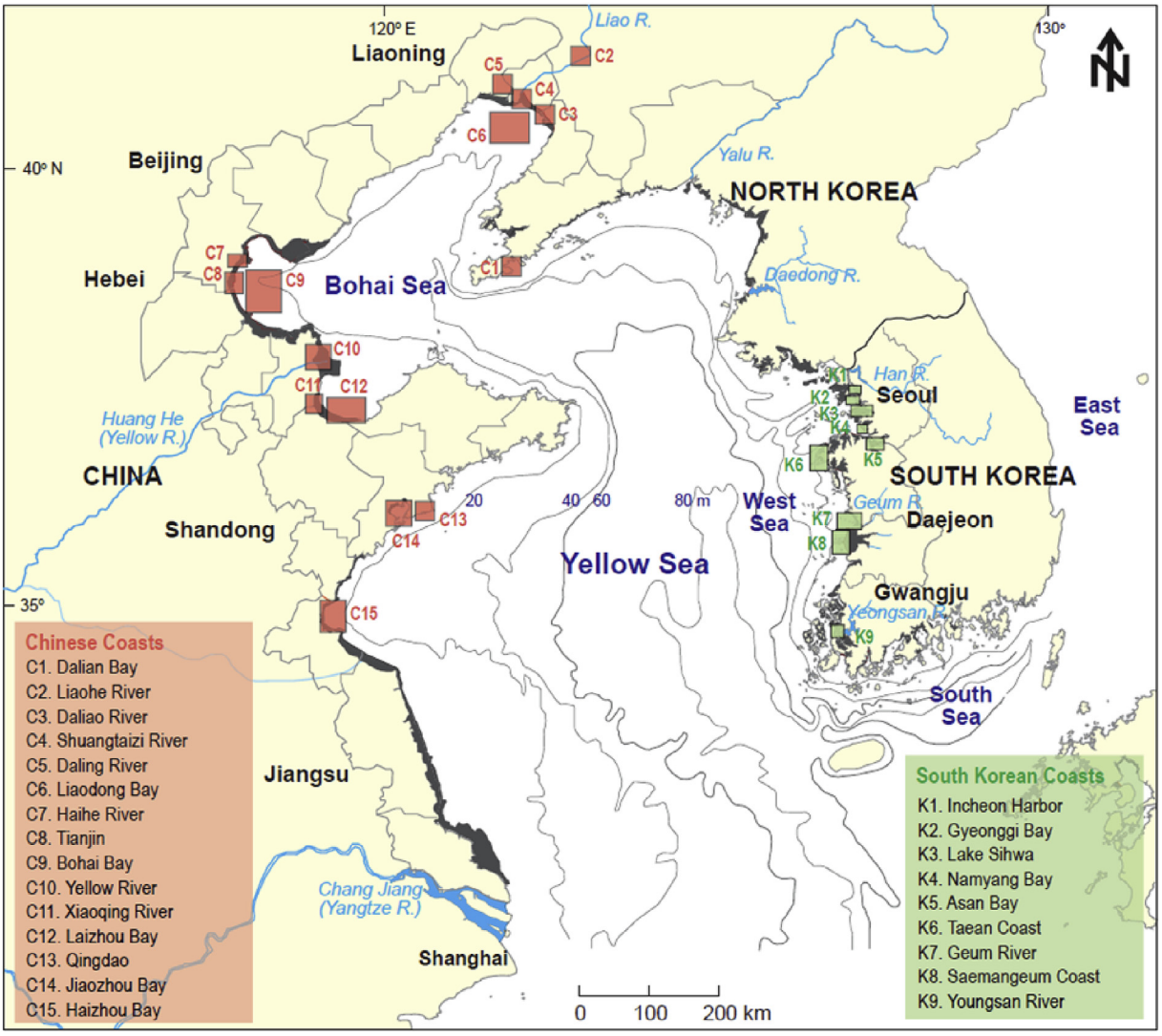

Fig. 1. Map showing the locations of the study areas along the Bohai Sea and Yellow Sea in China and South Korea.

contamination with DDTs (Lee et al., 2001b), but concentrations did not exceeded the ERM. Temporal trends of OCPs contaminations in sediments could not be evaluated due to the limited monitoring data. Overall, concentrations of OCPs in sediments of the West Sea of South Korea were generally less than those in portions of the Bohai and Yellow Seas in China.

In soils, OCPs were only investigated in Haihe River Basin, Daling River Basin, Yellow River Delta and Qingdao (Table S3) (Da, 2014; Geng, 2006; Hou et al., 2013; Li, 2010; Tao et al., 2008; Wang, 2013; Zhang, 2011). The highest concentration was measured in the Haihe River Basin, with a value of 12,549 ng g^{-1} dw for HCHs and 2033 ng g^{-1} dw for DDTs, which was mainly due to samples from the surrounding chemical industrial zone (Hou et al., 2013). Another study also reported higher OCPs in Haihe River Basin and showed that current residues of OCPs were 10-fold less than those from the 1980s (Tao et al., 2008). Based on National Environmental Quality Standards for Soils of China, soil are classified as three levels: first level (residual concentration $\leq$50 ng g^{-1} dw), second level (residual concentration from 50 to 500 ng g^{-1} dw), and third level (residual concentration from 500 to 1000 ng g^{-1} dw) for HCHs and DDTs (MEP China, 1995). Concentrations of HCHs and DDTs in some soils from Haihe River Basin exceeded the second level, meaning that posed risks on plants and environment in farmland, orchards and other agricultural land. Concentrations in some soils even exceeded the third level, which could not be used as land for agriculture and forestry production. HCHs and DDTs in soils from the Daling and Yellow River Basins, costal area of Bohai Sea and Qingdao were all less than 100 ng g^{-1} dw (Da, 2014; Geng, 2006; Li, 2010; Wang, 2013; Zhang, 2011). Meanwhile, although studies on OCPs in soil samples along the west coast of South Korea were scarce (Kim et al., 2014), concentrations of OCPs were generally less than those in regions around the Bohai Sea (Table S3).

Studies of OCPs in riverine or marine waters were scarce and mainly focused on estuaries and bays (Table S4). Generally, concentrations of OCPs in estuaries were higher than those from bays. In the Daling, Yellow and Haihe River Estuaries, concentrations of OCPs were greatest in the Haihe River Estuary, for HCHs (1290–5900 ng L^{-1}) or DDTs (520–4840 ng L^{-1}), followed by those from the Daling River Estuary (Da, 2014; Wang, 2010, 2013). In Jinzhou, Bohai, Liaozhou and Jiaozhou Bays, concentrations of OCPs in marine water from Bohai Bay was the most severe (Wang, 2010; Wang et al., 2013b; Xu et al., 2007; Yao et al., 2013). Only one study reported contamination of water by OCPs in Korea, focusing on the Saemangeum Coast (Hong et al., 2006a). The results showed that contamination there was less than that in China, but similar with that in Jiaozhou Bay, which is at the same latitude.

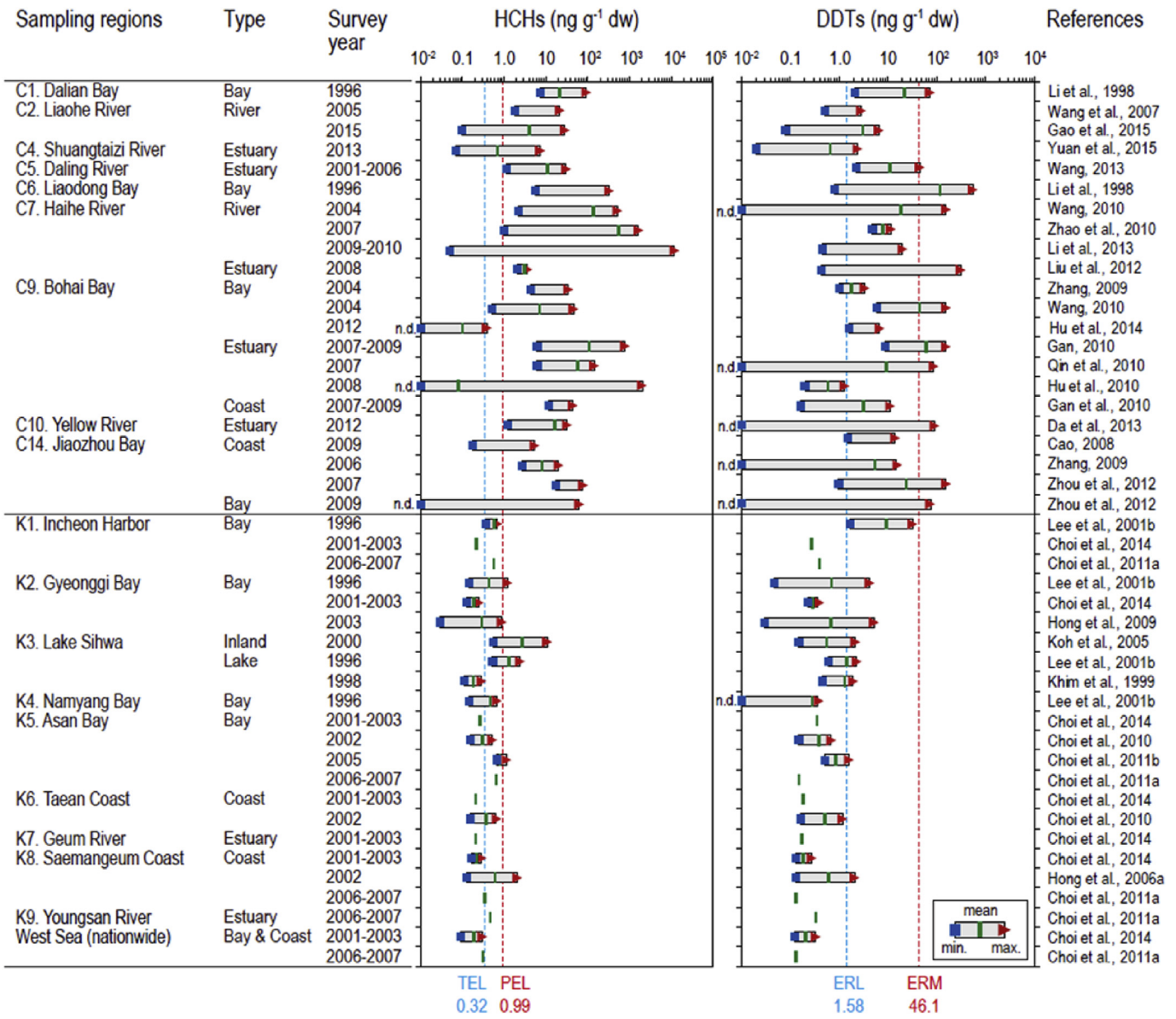

Fig. 2. Distributions of organochlorine pesticides (HCHs and DDTs) in sediments along the Bohai Sea and Yellow Sea in China and South Korea (threshold effect level (TEL) and probable effect level (PEL) for HCHs from Feng et al. (2011); effect range low (ERL) and effect range median (ERM) for DDTs from Feng et al. (2011)).

3.2. PCBs

Over the last 30 years, PCBs have been detected worldwide in various environmental media. In China, approximately 10,000 tons of PCBs were produced from 1965 to 1974, after which production of PCBs was banned. A total of 9000 tons as trichlorobiphenyl and 1000 tons as pentachlorobiphenyl were produced (Xing et al., 2005). A series of studies were conducted across whole area covered in this review. In general, concentrations of PCBs in sediments were 10-fold less than those of OCPs (Fig. 3). Concentrations of PCBs in coastal areas of Liaodong Bay varied greatly. The highest concentration was detected in the Liaohe River (1075.61 ng g^{-1} dw), flowing into Liaodong Bay (Lu, 2015; Zhang et al., 2010). Other studies reported concentrations of PCBs less than 50 ng g^{-1} dw in coastal areas of Liaodong Bay (Men et al., 2014; Wang et al., 2007; Yuan et al., 2015; Zhao et al., 2011). A decreasing trend of PCBs in this area could be found from 2004 to 2012. In Bohai Bay and its coastal areas, higher concentrations of PCBs were detected in the Haihe River, which were as great as 253 ng g^{-1} dw (Gan, 2010; Hu et al., 2014; Li et al., 2013; Liu, 2010; Wang, 2010; Zhang, 2009a; Zhao et al., 2010, 2012). Concentrations of PCBs in Jiaozhou Bay and its coastal areas were similar with those from Bohai Bay and its coastal areas (Cao, 2008; Guo et al., 2011; Wang et al., 2010). In Haizhou Bay, the least concentrations of PCBs were comparable with the least concentrations from three other bays (Zhang et al., 2014). In the broader areas of the sea, sediments were less contaminated by PCBs (Duan et al., 2013; Liu et al., 2008; Ma et al., 2001; Pan et al., 2010; Wang et al., 2015; Zhang et al., 2007). PCBs in coastal areas of the Bohai and Yellow Seas were reported at an early stage of industrial development in the region. Concentrations of PCBs in Dalian, Tianjin, Qingdao, and Lianyungang, which were also the more developed cities in studied area (Ma et al., 2001), were generally less than the ERL (22.7 ng g^{-1} dw), adverse biological effects of which would be *di minimis* (Zhao et al., 2010). Concentrations of PCBs from some sites in the Liaohe and Haihe Rives and Bohai, Qingdao, and Jiaozhou Bays were detected with higher

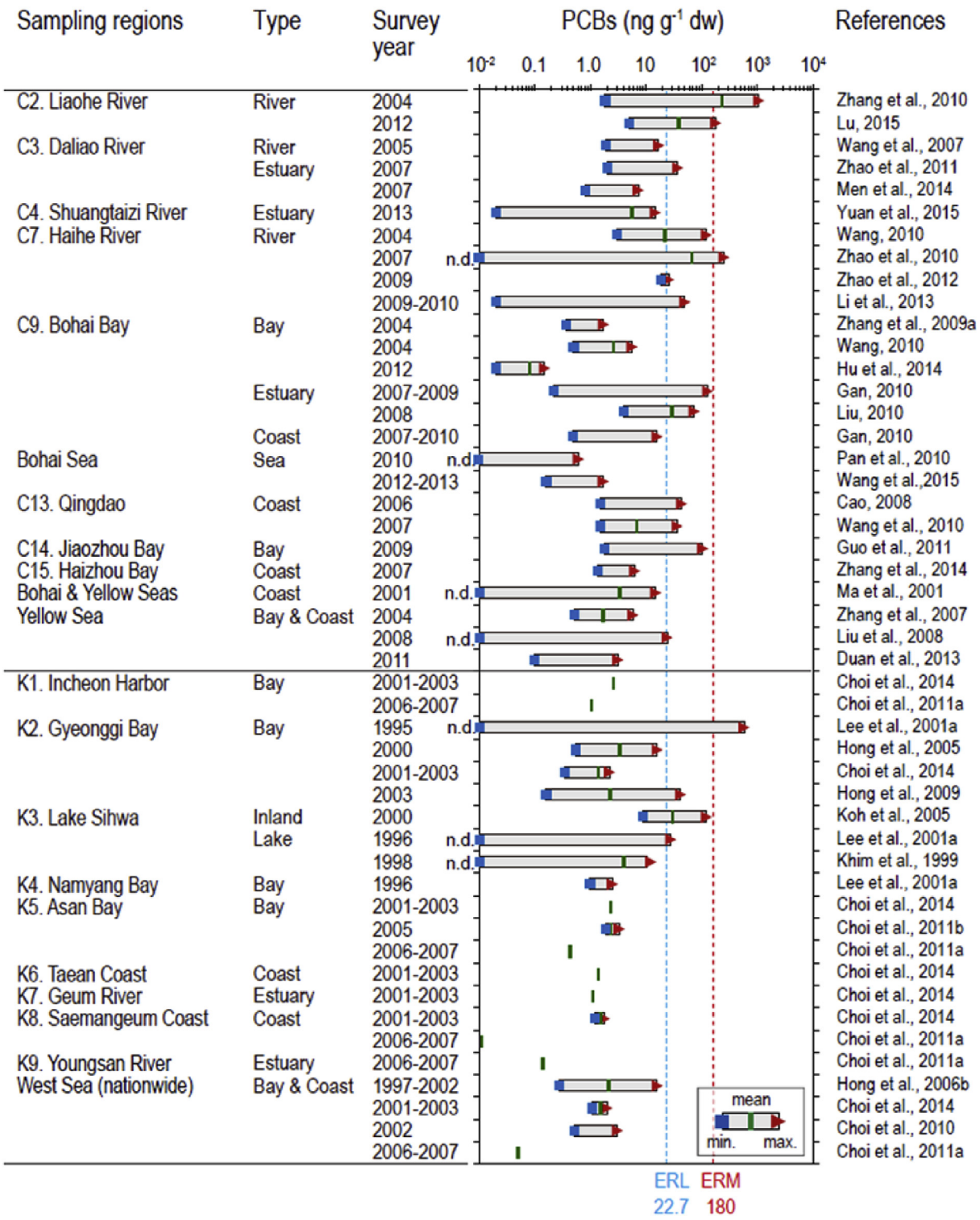

Fig. 3. Distributions of PCBs in sediments along the Bohai Sea and Yellow Sea in China and South Korea (effects range low (ERL) and effects range median (ERM) for PCBs from Zhao et al. (2010)).

concentrations than the ERL, and some from Liaohe River even significantly higher than the ERM (180 ng g^{-1} dw), which would be predicted to cause adverse biological effects.

In South Korea, despite the ban on use of PCBs as dielectric fluid in capacitors and transformers since the 1979 under the Electric Utility Act (EUA) (Kim and Yoon, 2014), PCBs were frequently detected in sediments (Fig. 3) (Choi et al., 2010, 2011a, 2011b, 2014; Hong et al., 2005, 2006b, 2009; Koh et al., 2005; Lee et al., 2001a). Greatest concentrations of PCBs were frequently found in the greatly industrialized regions, such as Incheon Harbor (1.0–580 ng g^{-1} dw) on the west coast of South Korea, which was similar to the highest concentration detected in China which was associated with adverse effects (Lee et al., 2001a). Concentrations of PCBs in sediments from some sites exceeded the ERM (Zhao et al.,

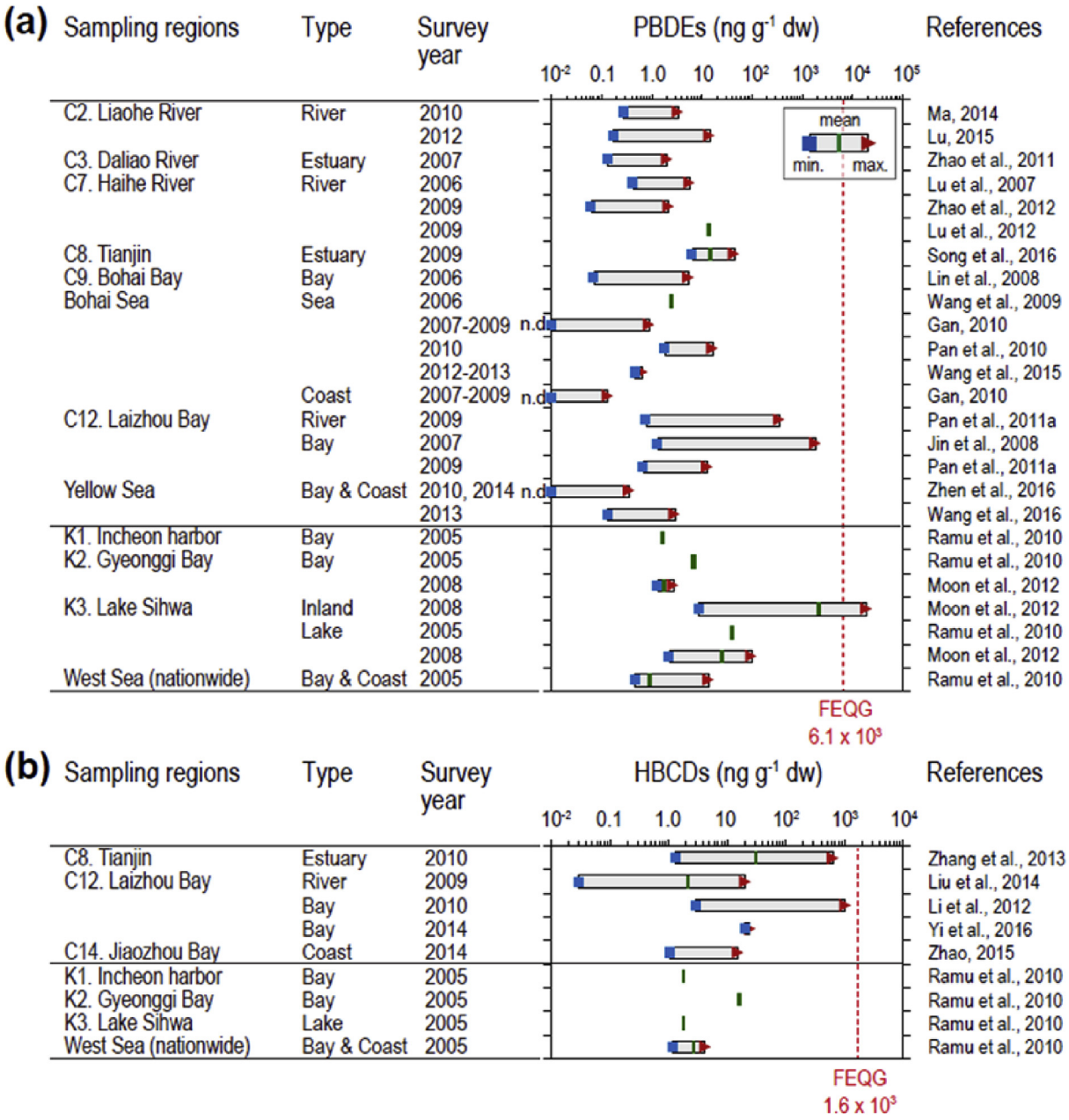

Fig. 4. Distributions of (a) PBDEs and (b) HBCDs in sediments along the Bohai Sea and Yellow Sea in China and South Korea (federal environmental quality guidelines (FEQG) for PBDEs and HBCDs from Environment Canada (2013, 2016)).

2010). Temporal trends of sedimentary PCBs in Gyeonggi Bay showed slightly decreasing trend from 1995 to 2003 (Choi et al., 2014; Hong et al., 2005, 2009; Lee et al., 2001a). Overall, concentrations of PCBs in sediments were generally comparable between the west coast of South Korea and Bohai Sea regions of China.

PCBs in soils were investigated in relatively developed areas of China, such as Tianjin, Qingdao, Dalian and the Yellow River Delta (Table S5). In these four areas, the highest concentrations of PCBs were detected in Tianjin, where concentrations were as great as 373 ng g^{-1} dw (Hou et al., 2013), followed by those in Yellow River Delta (Zhang, 2011). In Tianjin, Dagu Chemical Co., Ltd was recognized as the main source. In Qingdao and Dalian, concentrations of PCBs were all less than 15 ng g^{-1} dw, but no obvious primary source was identified (Geng, 2006; Wang et al., 2008). A study of PCBs in soils from across China was conducted in 2005, concentrations ranged from 0.14 to 1.84 ng g^{-1} dw (Ren et al., 2007). Concentrations of PCBs in soils from coastal areas of the Bohai and Yellow Seas were moderate and the tri-PCB homologue was dominant, followed by di-PCB homologue. According to Environmental Quality Standards for Soils (revised draft), the second level criteria for agricultural and industrial lands are 100 and 1500 ng g^{-1} dw, respectively, which means that it is possible for adverse effects to occur at concentrations exceeding that concentration (MEP China, 2008). Based on this guideline, most sites could be used as agricultural land, while some could be used as other land use purposes and did not need remediation. One study reported concentrations of PCBs in soils from South Korea, which showed lesser contamination than that observed in China (Kim et al., 2016).

PCBs in river or marine waters are only investigated in the northern Bohai Sea, including the Daliao and Haihe Rivers and

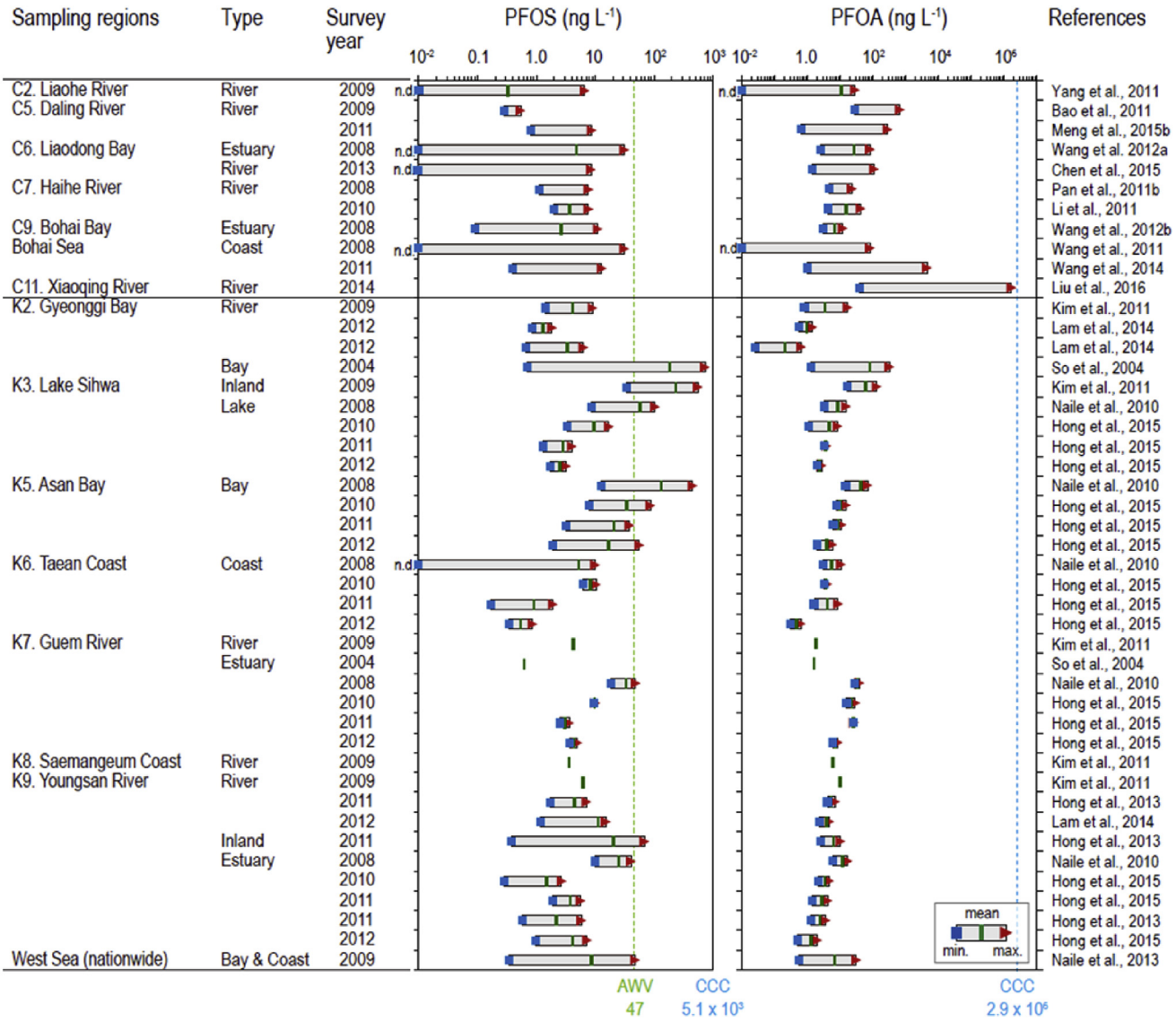

Fig. 5. Distributions of PFOS and PFOA in surface water samples along the Bohai Sea and Yellow Sea in China and South Korea (avian wildlife values (AWV) and criteria continuous concentration (CCC) for PFOS and PFOA from Giesy et al. (2010)).

Jinzhou and Bohai Bays (Table S5). Generally, concentrations of total PCBs in water from Jinzhou Bay and the Haihe River Estuary were greater, with concentrations of 215.4–3161 ng L^{-1} and 310–3110 ng L^{-1}, respectively (Wang, 2010; Yao et al., 2013). Concentrations of PCBs in water from Bohai Bay were 10-fold less than those from Jinzhou Bay and the Haihe River Estuary (Wang et al., 2013b; Wang, 2010). Concentrations of PCBs from lower reaches of the Haihe River to Bohai Bay were analyzed (Wang, 2010). Concentrations of PCBs in water from the Haihe River were significantly greater than those from Bohai Bay. The highest concentrations of PCBs were detected in the estuary. This study through continues monitoring showed that an increasing trend of detected PCBs was observed from 2004 to 2006. This result indicated a new source. Long-term air transportation and unintentional production were major sources. Concentrations of PCBs in water from the Daliao River Estuary were least (Men et al., 2014). In these areas, concentrations of PCBs in water were generally greater than were concentrations of OCPs (mainly HCHs and DDTs), which was likely due to the phase-out of HCHs and DDTs.

4. Brominated POPs in environment

4.1. PBDEs

Several studies of PBDEs in sediments from areas surrounding the Bohai Sea, from the Liaohe River Basin in the north to Laizhou Bay in the south have been conducted (Fig. 4a). The most severely polluted area was Laizhou Bay due to manufacture of PBDEs and Bohai Bay due to use and disposal of electronic products. The greatest concentration of PBDEs was detected in the Bailong River, Laizhou Bay, with concentrations as great as 1800 ng g^{-1} dw (Jin et al., 2008; Pan et al., 2011a). In areas adjacent to Bohai Bay, rivers flowing into the Bohai Sea were most heavily contaminated by PBDEs, with the highest concentration of 42.79 ng g^{-1} dw (Gan, 2010; Lu et al., 2007a, 2012; Pan et al., 2010; Song et al., 2016; Zhao et al., 2012). Three studies were conducted to investigate PBDEs in sediment in the Liaohe River Basin. Similar concentrations were detected among the studies with concentrations less than 15 ng g^{-1} dw (Lu, 2015; Ma, 2014; Zhao et al., 2011). In addition, less

concentrations of PBDEs were detected in marine sediments from coastal areas of the Bohai Sea (Lin et al., 2008; Wang et al., 2009). Bohai and Laizhou Bays were more polluted than other areas of the Bohai Sea. Only one study was carried out to investigate concentrations of PBDEs in marine sediments from the Yellow Sea and similar concentrations were detected with those from Bohai Sea (Wang et al., 2016). Principal component analysis (PCA) reveled that PBDEs in the Yellow Sea were mainly from the continental runoff (69%) and atmospheric deposition (31%). No concentrations of PBDEs exceeded the sediment quality guideline (6143 ng g^{-1} dw), meaning no potential risks (Environment Canada, 2013).

Contamination of sediments by PBDEs was widespread along the Korean coast (Moon et al., 2012; Ramu et al., 2010). Results of previous studies revealed that greatest concentrations of PBDEs in sediments were observed in creeks flowing into Lake Sihwa (8.5–18,700 ng g^{-1} dw) (Moon et al., 2012) (Fig. 4a). In particular, concentrations of PBDEs detected in the Lake Sihwa near industrial complexes exceeded the federal environmental quality guidelines (FEQG) (Environment Canada, 2013) and were greatest compared with worldwide values. Although PBDEs in Korean sediments were less studied and thus temporal distribution trends could not be evaluated, significant reductions in concentrations of PBDEs in sediments were observed during recent years. This reduction seems to be the result of a recent ban on the use and production of commercial PBDEs in Korea. More monitoring of PBDEs and novel brominated flame retardants or other non-brominated replacements in coastal environments is needed.

There were large differences in concentrations of PBDEs in soils from coastal areas of the Bohai Sea (Table S6). The most seriously contaminated areas were in the vicinity of sites where PBDEs were manufactured or used for making electronics or where electronic devices were dismantled. Several studies that have investigated PBDEs in soils around manufacturing sites of Laizhou Bay revealed that BDE-209 was the predominant congener (Deng et al., 2016; Jin et al., 2011; Liu et al., 2014a; Li et al., 2015). In these studies, the greatest concentration of PBDEs was 226,906 ng g^{-1} dw. At sites where electronics were dismantled, such as Tianjin, concentrations of PBDEs ranged from 1.34 to 343.88 ng g^{-1} dw (Liu, 2013). A series of studies investigated PBDEs in soils from various provinces on the Bohai Rim (Liu, 2013). Soils from north China were collected from Beijing, Tianjin, Shandong, Hebei and Shanxi. Concentrations of PBDEs were between 0.01 and 948.84 ng g^{-1} dw. There was an increasing trend from inland to coast and the most severe polluted areas were distributed in Shandong, especially Laizhou Bay (Chen et al., 2012; Yuan, 2013). Concentrations of PBDEs in soils from the west coast of South Korea were less than those from near the Bohai and Yellow Seas of China (Kim et al., 2014).

4.2. HBCDs

Based on regulatory restrictions on uses of PBDE formulations, usage of HBCD has increased during the past decade with a concomitant increase in concentrations of HBCDs in environmental media. However, information on concentrations of HBCDs in environment in Bohai and Yellow Sea regions in both China and South Korea was scarce. Some studies investigated concentrations of HBCDs in sediment and soils (Fig. 4b and Table S7). Generally, concentrations of HBCDs were not high, except those in samples from manufacturing sites near Laizhou Bay. Laizhou Bay area is the center of the brominated chemical industry in China, with many BFR manufacturing facilities located in the industrial park of Weifang city near Laizhou Bay (Li et al., 2012). HBCDs in soils and sediments from on-site and off-site at HBCDs production and processing plants were investigated (Yi et al., 2016). Concentrations of HBCDs in soils collected on-site and off-site were 96.1–560.4 ng g^{-1} dw and 0.11–31.1 ng g^{-1} dw, respectively. Two samples of sediments were also collected around plants, and concentrations were 20.4 ng g^{-1} dw and 24.2 ng g^{-1} dw, respectively. Another study reported relatively higher concentrations of HBCDs in environment near manufacturing facilities in Laizhou Bay area (Liu et al., 2012). Concentrations in soils and sediment were 0.88–6901 ng g^{-1} dw and 2.93–1029 ng g^{-1} dw, respectively. Relatively greater concentrations of HBCDs were detected in sediments from rivers and harbor of Tianjin, with values of 1.35–634 ng g^{-1} dw (Zhang et al., 2013b). In these studies areas, γ-HBCD was predominant, which was consistent with the pattern of distributions of diastereomer in industrial products.

HBCDs in soils from coastal cities along the Bohai and Yellow Seas were reported (Zhang et al., 2016). Concentrations of HBCDs ranged from 0.12 ng g^{-1} dw to 363 ng g^{-1} dw. Generally, soils from coastal cities along Bohai Sea were more seriously polluted than those along Yellow Sea. Among all 21 cities, the highest mean concentrations of HBCDs were 34.6 ng g^{-1} dw in Weifang, 12.3 ng g^{-1} dw in Cangzhou and 11.1 ng g^{-1} dw in Tianjin. Mean concentrations from the other 18 cities were all less than 10 ng/g dw from other 18 cities. In addition, some studies were carried out in the Yellow River Delta, rivers flowing into Laihou Bay and Jiaozhou Bay area, and less HBCDs were detected (Liu et al., 2014b; Yuan, 2013; Zhao, 2015). The highest concentration of HBCDs did not exceed the FEQG for sediment (1600 ng g^{-1} dw) but was close (Environment Canada, 2016). Therefore, with continuous production and accumulation of HBCDs in sediment, it will likely cause harm near these sites of production. Worldwide, including in the South Korea, few studies of occurrences of HBCDs have been reported (Fig. 4b). Results indicated that relatively great concentrations of HBCDs were found in Gyeonggi Bay near industrial complexes and harbors (Ramu et al., 2010). However, detected HBCDs concentrations in sediments did not exceeded the FEQG (Environment Canada, 2016). More complementary studies on sources, distributions, and potential toxic effects on marine organisms of emerging contaminants HBCDs are needed for assessment and management.

5. Fluorinated POPs in environment

PFASs were widely distributed in water of rivers and marine environments of the Bohai and Yellow Seas due to their solubility in water and negligible vapor pressures, when dissolved in water (Fig. 5, Tables S8 and S9). There were abundant studies on PFASs in water and sediment of rivers of the Bohai Rim. Concentrations of PFOS and PFOA in water from the northern Bohai coastal areas ranged from n.d. to 30.9 and n.d.–81.7 ng L^{-1}, respectively (Wang et al., 2011), while those in south Bohai costal area were 0.40–12.78 and 0.96–4534 ng L^{-1}, respectively (Wang et al., 2014). Corresponding concentrations of PFASs in sediments were also reported. Concentrations of PFOS and PFOA in sediment from the northern Bohai coastal areas were n.d.–1.97 and n.d.–0.54 ng g^{-1} dw, respectively (Wang et al., 2011), while those in southern Bohai coastal areas were 0.03–0.44 and 0.005–29.02 ng g^{-1} dw, respectively (Zhu et al., 2014). The greatest concentration was observed in the Xiaoqing River, which was likely due to the presence of local fluorine chemical industries. Pollution of PFASs in water and sediment of Liaodong Bay basin was investigated, and results showed that PFASs in the Daling River were relatively great (Chen et al., 2015).

Occurrences and fates of PFASs in marine sediments from the Bohai and Yellow Seas were reported (Gao et al., 2014). In the Bohai Sea, concentrations of PFOS and PFOA were n.d.–0.15 and 0.06–2.70 ng g^{-1} dw, among which relatively high concentrations were found near Liaodong Bay. In the Yellow Sea, concentrations of

PFOS and PFOA were n.d.–0.40 and nd–1.52 ng g^{-1} dw, which was due to fluorine chemical plants in the Daling and Xiaoqing River basins. Concentrations of PFASs were investigated in the surroundings of fluorine chemical plants in the Daling River basin (Bao et al., 2011). Concentrations of PFOS and PFOA in water from streams in the Daling River basin were 0.28–0.54 and 27.2–668 ng L^{-1}, respectively, while those in sediment were n.d. and 0.18–18 ng g^{-1} dw, respectively. Sediment cores and overlying water were also collected to investigate pollution history in Daling River basin (Meng et al., 2015b). Concentrations of PFOS and PFOA in surface water were 0.80–8.7 and 0.63–284 ng L^{-1}, respectively. Concentrations of PFOA in water from the Xiaoqing River basin ranged from 38.6 to 1,707,290 ng L^{-1} (Liu et al., 2016). The greatest concentrations of PFASs in sediment were detected in the Xiaoqing River, of which PFOA was dominant and its concentrations were between 3.86 and 456.20 ng g^{-1} dw (Sun et al., 2016). In addition, studies in the Liaohe, Haihe and Yellow Rivers were also carried out (Li et al., 2011; Pan et al., 2011b; Wang et al., 2012a, 2012b; Yang et al., 2011; Zhao et al., 2013). Since there was no obvious production of PFASs and related products, PFOS and PFOA in water were less than 100 ng L^{-1} and those in sediment were less than 10 ng g^{-1} dw in these rivers. It was noted that concentrations of PFOA in China were generally higher than that of PFOS. Concentrations of both PFOS and PFOA in water did not exceed water quality guidelines, avian wildlife values (AWV) or criteria continuous concentration (CCC), which indicates that there would be no harm to wildlife or aquatic organisms expected (Giesy et al., 2010).

In recent years, on the west coast of South Korea, concentrations of waterborne PFASs have generally decreased (Hong et al., 2015; Naile et al., 2010, 2013). Distributions of concentrations of PFASs indicated that they were greater in freshwater than they were in seawater, which suggested that PFASs originated from point sources of surrounding inland areas rather than from non-point sources (Naile et al., 2013). Greatest concentrations of PFOS and PFOA in water were found in Gyeonggi Bay, followed by Lake Sihwa, and Lake Asan (Fig. 5) (Hong et al., 2013, 2015; Kim et al., 2011; Lam et al., 2014; Naile et al., 2010; So et al., 2004). Concentrations of PFOA in some water samples from these sites exceeded the AWV and would have been expected to cause some adverse effects. Temporal trends in concentrations of PFASs in water samples indicated that concentrations have decreased since 2008 (Hong et al., 2013; Naile et al., 2010, 2013). Relative contributions of PFOS decreased, while those of shorter-chain PFASs such as PFBA have increased in recent years, which indicated that recent global restrictions on uses of some PFASs have resulted in less production and subsequent releases to the environment. In 2010, the government of South Korea designated PFASs (PFOS and its salts and PFOSF) as "restricted chemicals" in the "Persistent Organic Pollutants Control Act (PCA)". The PCA covers the regulations of commercial use, waste management, and emission control, etc. (Kim and Yoon, 2014). Overall, concentrations of PFOS in water were generally greater in the Bohai and Yellow Sea regions of China than those from the west coast of South Korea. Concentrations of PFOA in Chinese coasts were greater than those of South Korea. Some sediment samples were collected from coastal areas of the West Sea, Han River and Yeongsan River (Lam et al., 2014; Naile et al., 2013). In general, concentrations of PFOS and PFOA from these rivers were small. Concentrations of PFOA and PFOS from coastal areas of the West Sea were similar to those in China, except PFOA, concentrations of which were greater at some manufacturing sites, such as watershed of the Xiaoqing River in China.

Studies on PFASs in soils were scarce, but almost cover whole coastal areas of Bohai Sea and West Sea of South Korea (Table S9). Generally, concentrations of PFOS and PFOA in soils were small and most were less than 10 ng g^{-1} dw (Meng et al., 2015a; Wang et al., 2011, 2012b). Relatively more studies have been conducted in the area surrounding Liaodong, Bohai, and Laizhou Bays. Relatively greater concentrations were detected in these three areas, which was likely due to production of PFASs (Liaodong Bay and Laizhou Bay) and numerous industrial applications (Bohai Bay) (Pan et al., 2011b; Wang et al., 2012b, 2013a). In South Korea, concentrations of PFOA and PFOS in soils from coastal areas of the West Sea were greater than those from inland regions (Lam et al., 2014; Naile et al., 2013).

6. Future research direction

In the present review, spatial distributions and potential risks of several POPs in environmental media along the Bohai and Yellow Seas were summarized. Results of data review suggested that sources of different POPs were independent and some hotspot areas with greater production of certain chemicals were identified (Table 2). Bohai Bay and Haihe River in China and Gyeonggi Bay and Lake Sihwa in South Korea were found to be the hot spot areas for traditional POPs pollution along the Bohai Sea and Yellow Sea regions. Meanwhile, new POPs such as PBDEs, HBCDs, and PFASs were greatly polluted in some regions, for example, Laizhou Bay, Tianjin, Xiaoqing River, and Asan Bay. Patterns of POPs pollution (e.g., chemicals of concerns) were distinguished between China and South Korea. Temporal trends of sedimentary POPs were found to decrease in recent years that seemed to be associated with chemical controls and regulations, mainly for banned OCPs and PCBs, and PFASs in South Korea. Studies on new POPs such as PBDEs and HBCDs in multimedia samples were still limited. In addition, environmental quality guidelines of POPs for protection of the coastal ecosystem have not been established in both China and South Korea. We suggest that the future research directions of POPs based on the current understandings and limitations, include: 1)

Table 2
Summary of results of the review, including key sources of POPs and hotspot areas along the Bohai and Yellow Seas in China and South Korea.

POPs	General distribution	Areas of concern in Chinese Coasts			Areas of concern in Korean Coasts		
		1st	2nd	3rd	1st	2nd	3rd
Chlorinated POPs							
OCPs	China ≫ Korea	C7. Haihe River	C9. Bohai Bay	C14. Jiaozhou Bay	K1. Incheon Harbor	K2. Gyeonggi Bay	K3. Lake Sihwa
PCBs	China~Korea	C2. Liaohe River	C7. Haihe River	C9. Bohai Bay	K2. Gyeonggi Bay	K3. Lake Sihwa	
Brominated POPs							
PBDEs	China < Korea	C12. Laizhou Bay	C8. Tianjin	C9. Bohai Bay	K3. Lake Sihwa	K2. Gyeonggi Bay	
HBCDs	China > Korea	C12. Laizhou Bay	C8. Tianjin				
Fluorinated POPs							
PFOS	China < Korea	C9. Bohai Bay	C6. Liaodong Bay		K2. Gyeonggi Bay	K3. Lake Sihwa	K5. Asan Bay
PFOA	China > Korea	C11. Xiaoqing River	C9. Bohai Bay		K2. Gyeonggi Bay	K3. Lake Sihwa	K5. Asan Bay

systematic monitoring of new POPs for baseline data; 2) establishment of sediment quality guidelines; 3) monitoring of alternatives for POPs; 4) identification of unknown toxic chemicals (non-targeted screening); 4) determination of ecotoxicological effects; 6) determination of mixture effects of POPs and other pollutants; and 7) approach of multiple lines of evidence for accurate risk assessment. Overall, the present review will guide identification for areas of concerns and prioritization of research on POPs pollution assessment and management practices in both China and South Korea.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.11.108.

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<Supplementary Materials>

Traditional and new POPs in environments along the Bohai and Yellow Seas: An overview of China and South Korea

Jing Meng [a,b,1], Seongjin Hong [c,1], Tieyu Wang [a,b,*], Qifeng Li [a,b], Seo Joon Yoon [d], Yonglong Lu [a,b], John P. Giesy [e,f], Jong Seong Khim [d,**]

[a] *State Key Lab of Urban and Regional Ecology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China*

[b] *University of Chinese Academy of Sciences, Beijing 100049, China*

[c] *Department of Ocean Environmental Sciences, Chungnam National University, Daejeon 34134, Republic of Korea*

[d] *School of Earth and Environmental Sciences & Research Institute of Oceanography, Seoul National University, Seoul 08826, Republic of Korea*

[e] *Department of Veterinary Biomedical Sciences & Toxicology Centre, University of Saskatchewan, Saskatoon, SK, Canada*

[f] *Department of Zoology & Center for Integrative Toxicology, Michigan State University, East Lansing, MI, USA*

*Corresponding authors.

E-mail address: wangty@rcees.ac.cn (T. Wang) & jskocean@snu.ac.kr (J.S. Khim).

Supplementary Tables

Supplementary Tables

Table S1. Summary of studies of POPs in environmental media along the Bohai and Yellow Seas of China.

Sampling regions	Type	Sampling year	Sample number	Target POPs						References*
		(y)	(n)	DDTs	HCHs	PCBs	PBDEs	HBCDs	PFASs	
Sediment										
1. Dalian Bay	Bay	1996	30	V	V					Li et al., 1998
2. Liaohe River	River	2004	14			V				Zhang et al., 2010
		2009	20						V	Yang et al., 2011
		2010	19				V			Ma, 2014
		2012	15			V	V			Lu, 2015
		2015	26	V	V					Gao et al., 2015
3. Daliao River	River	2005	12	V	V	V				Wang et al., 2007
	Estuary	2007	14				V			Zhao et al., 2011
		2007	35			V				Men et al., 2014
4. Shuangtaizi River	Estuary	2013	60	V	V	V				Yuan et al., 2015
5. Daling River	Estuary	2001-2006	30	V	V					Wang, 2013
	River	2009	6						V	Bao et al., 2011
		2011	7						V	Meng et al., 2015b
6. Liaodong Bay	Bay	1996	26	V	V					Li et al., 1998
	River	2013	34						V	Chen et al., 2015
7. Haihe River	River	2004	10	V	V	V				Wang, 2010
		2006	20				V			Lu et al., 2007
		2007	25	V	V	V				Zhao et al., 2010
		2008	23						V	Pan et al., 2011b
		2009	66			V	V			Zhao et al., 2012
		2009	10	V	V	V	V			Lu et al., 2012
		2009-2010	17	V	V	V				Li et al., 2013
		2010	24						V	Li et al., 2011
	Estuary	2008	19	V	V					Liu et al., 2012
8. Tianjin	Estuary	2009	11				V			Song et al., 2016
		2010	51					V		Zhang et al., 2013
9. Bohai Bay	Bay	2004	27	V	V	V				Zhang, 2009a
		2004	10	V	V					Wang, 2010
		2006	16				V			Lin et al., 2008
		2007-2009	22	V	V	V	V			Gan, 2010

		2012	20	V	V	V				Hu et al., 2014
	Estuary	2007-2009	9	V	V	V	V			Gan, 2010
		2007	6	V	V					Qin et al., 2010
		2008	19	V	V	V				Liu, 2010
		2008	8						V	Wang et al., 2012b
	Coast	2007-2009	4	V	V	V	V			Gan, 2010
Bohai Sea	Sea	2006	16				V			Wang et al., 2009
		2010	52			V	V			Pan et al., 2010
		2011-2012	29						V	Gao et al., 2014
		2012-2013	34			V	V			Wang et al., 2015
	Coast	2008	42						V	Wang et al., 2011
		2011	36						V	Zhu et al., 2014
10. Yellow River	Estuary	2012	34	V	V					Da, 2014
11. Xiaoqing River	River	2014	10						V	Sun et al., 2016
12. Laizhou Bay	River	2009	36				V			Pan et al., 2011a
		2009	36					V		Liu et al., 2014
	Estuary	2009	26						V	Zhao et al., 2013
	Bay	2007	1				V			Jin et al., 2008
		2009	26				V			Pan et al., 2011a
		2009	24						V	Zhao et al., 2013
		2010	6					V		Li et al., 2012
		2014	2					V		Yi et al., 2016
13. Qingdao	Coast	2006	17			V				Cao, 2008
		2007	29	V	V	V				Wang et al., 2010
14. Jiaozhou Bay	Coast	2006	17	V	V					Cao, 2008
		2007	30	V	V	V				Zhang, 2009
		2007	21	V	V					Zhou et al., 2012
		2014	25				V	V		Zhao, 2015
	Bay	2009	21	V	V					Zhou et al., 2012
		2009	12			V				Guo et al., 2011
15. Haizhou Bay	Coast	2007	20			V				Zhang et al., 2014
Bohai & Yellow Seas	Coast	2001	20	V		V				Ma et al., 2001
Yellow Sea	Bay & Coast	2004	33			V				Zhang et al., 2007
		2008	82	V		V				Liu et al., 2008
		2011	42			V				Duan et al., 2013
		2013	24				V			Wang et al., 2016
	Sea	2011-2012	61						V	Gao et al., 2014
Soil										

Dalian	Urban	2007	14			V					Wang et al., 2008
5. Daling River	Estuary	2001-2002	29	V	V						Wang, 2013
6. Liaodong Bay	Coast	2008	14							V	Wang et al., 2013a
7. Haihe River	River	2004	302	V	V						Tao et al., 2008
8. Tianjin	Industrial	2008	70	V	V	V					Hou et al., 2013
	Suburban	2008	86							V	Pan et al., 2011b
	Electronic dismantle	2012	12				V				Liu, 2013
9. Bohai Bay	Estuary	2008	8							V	Wang et al., 2012b
10. Yellow River	River	2008	22	V	V	V					Zhang, 2011
	Estuary	2008	20				V				Chen et al., 2012
		2012	26	V	V						Da, 2014
		2012	23				V	V			Yuan, 2013
12. Laizhou Bay	Coast	2007	5				V				Jin et al., 2011
	Manufacture	2010	15					V			Li et al., 2012
Bohai Sea	Coast	2005-2008	264	V	V						Li, 2010
		2008	31							V	Wang et al., 2011
		--	79							V	Meng et al., 2015a
13. Qingdao	Urban	2005	60	V	V	V					Geng, 2006
Shandong province	Manufacture	2011	23				V				Li et al., 2015
		2011	12								Liu, 2013
		2013	25					V			Yi et al., 2016
		2016	9				V				Deng et al., 2016
	Urban	2011	27				V				Liu, 2013
Hebei province	Urban	2011	28				V				Liu, 2013
Tianjin & Beijing	Urban	2011	7				V				Liu, 2013
Shanxi province	Urban	2011	25				V				Liu, 2013
Bohai & Yellow Seas	Coast	2013	188					V			Zhang et al., 2016
North China		2011	87				V				Liu et al., 2014
Water											
2. Liaohe River	River	2009	20							V	Yang et al., 2011
3. Daliao River	Estuary	2007	12			V					Men et al., 2014
5. Daling River	Estuary	2001-2006	30	V	V						Wang, 2013
		2009	6							V	Bao et al., 2011
		2011	7							V	Meng et al., 2015b
6. Liaodong Bay	Estuary	2008	10							V	Wang et al., 2012a
	Bay	2012	15	V	V	V					Yao et al., 2013
	River	2013	35							V	Chen et al., 2015
7. Haihe River	Estuary	2004	10			V					Wang, 2010

		2006	10	V	V			Wang, 2010
	River	2008	23				V	Pan et al., 2011b
		2010	24				V	Li et al., 2011
9. Bohai Bay	Bay	2004	10			V		Wang, 2010
		2006	10	V	V			Wang, 2010
		2011	9	V	V	V		Wang et al., 2013b
	Coast	2008	36				V	Wang et al., 2011
		2011	35				V	Wang et al., 2014
10. Yellow River	Estuary	2012	34	V	V			Da, 2014
11. Xiaoqing River	River	2014	10				V	Liu et al., 2016
12. Laizhou Bay	Bay	2005	44	V	V			Xu et al., 2007
14. Jiaozhou Bay	Bay	2005	44	V	V			Xu et al., 2007

*Details of references are in the main text.

Table S2. Summary of studies of POPs in environmental media along the west coast of South Korea.

Sampling regions	Type	Sampling year	Sample number	Target POPs						References*
		(y)	(n)	DDTs	HCHs	PCBs	PBDEs	HBCDs	PFASs	
Sediment										
1. Incheon harbor	Bay	1996	7	V	V					Lee et al., 2001b
		2001-2003	1	V	V	V				Choi et al., 2014
		2005	1				V	V		Ramu et al., 2010
		2006-2007	1	V	V	V				Choi et al., 2011a
2. Gyeonggi Bay	Bay	1996	54			V				Lee et al., 2001a
		1996	47	V	V					Lee et al., 2001b
		2000	13			V				Hong et al., 2005
		2001-2003	7	V	V	V				Choi et al., 2014
		2003	33	V	V	V				Hong et al., 2009
		2005	1				V	V		Ramu et al., 2010
		2008	6				V			Moon et al., 2012
3. Lake Shihwa	Inland	2000	8	V	V	V				Koh et al., 2005
		2008	12				V			Moon et al., 2012
	Lake	1996	4			V				Lee et al., 2001a
		1996	3	V	V					Lee et al., 2001b
		1998	11	V	V	V				Khim et al., 1999
		2005	1				V	V		Ramu et al., 2010
		2008	16				V			Moon et al., 2012
4. Namyang Bay	Bay	1996	5			V				Lee et al., 2001a
		1996	5	V	V					Lee et al., 2001b
5. Asan Bay	Bay	2001-2003	1	V	V	V				Choi et al., 2014
		2002	3	V	V					Choi et al., 2010
		2005	6	V	V	V				Choi et al., 2011b
		2006-2007	1	V	V	V				Choi et al., 2011a
6. Taean Coast	Coast	2001-2003	1	V	V	V				Choi et al., 2014
		2002	5	V	V					Choi et al., 2010
7. Guem River	Estuary	2001-2003	1	V	V	V				Choi et al., 2014
8. Saemangeum Coast	Coast	2001-2003	3	V	V	V				Choi et al., 2014
		2002	13	V	V					Hong et al., 2006a
		2006-2007	1	V	V	V				Choi et al., 2011a
9. Youngsan River	Estuary	2006-2007	1	V	V	V				Choi et al., 2011a
West Sea (nation wide)	Bay & Coast	1997-2002	35			V				Hong et al., 2006b

		2001-2003	6	V	V	V				Choi et al., 2014
		2002	8			V				Choi et al., 2010
		2005	2				V	V		Ramu et al., 2010
		2006-2007	1	V	V	V				Choi et al., 2011a
		2009	12						V	Naile et al., 2013
	River (Inland)	2012	10						V	Lam et al., 2014
West Sea	Bay & Coast	2009	13						V	Naile et al., 2013
Soil										
Gyeonggi province	Inland	nd	6			V				Kim et al., 2016
Gyeonggi province	Industrial & urban	2012	6		V		V		V	Kim et al., 2014
Chungnam province	Urban & agricultural	2012	5		V		V		V	Kim et al., 2014
Jeonbuk province	Urban & agricultural	2012	3		V		V		V	Kim et al., 2014
Jeonnam province	Urban & agricultural	2012	3		V		V		V	Kim et al., 2014
Water										
2. Gyeonggi Bay	River	2009	5						V	Kim et al., 2011
		2010	5						V	Kim et al., 2014
		2012	3						V	Lam et al., 2014
		2012	4						V	Lam et al., 2014
	Bay	2004	4						V	So et al., 2004
3. Lake Shihwa	Inland creek	2009	4						V	Kim et al., 2011
	Lake	2008	4						V	Naile et al., 2010
		2010	4						V	Hong et al., 2015
		2011	3						V	Hong et al., 2015
		2012	3						V	Hong et al., 2015
5. Asan Bay	Osan Stream	2010	4						V	Kim et al., 2014
	Bay	2008	4						V	Naile et al., 2010
		2010	4						V	Hong et al., 2015
		2011	4						V	Hong et al., 2015
		2012	4						V	Hong et al., 2015
6. Taean Coast	Coast	2008	3						V	Naile et al., 2010
		2010	3						V	Hong et al., 2015
		2011	3						V	Hong et al., 2015
		2012	3						V	Hong et al., 2015
7. Guem River	River	2009	1						V	Kim et al., 2011
		2010	11						V	Kim et al., 2014
	Estuary	2004	1						V	So et al., 2004
		2008	2						V	Naile et al., 2010
		2010	2						V	Hong et al., 2015

		2011	2				V	Hong et al., 2015
		2012	2				V	Hong et al., 2015
8. Saemangeum Coast	River	2009	1				V	Kim et al., 2011
	River & Coast	2002	18	V	V			Hong et al., 2006
9. Youngsan River	River	2009	1				V	Kim et al., 2011
		2010	5				V	Kim et al., 2014
		2012	3				V	Lam et al., 2014
	Estuary	2008	2				V	Naile et al., 2010
		2010	2				V	Hong et al., 2015
		2011	2				V	Hong et al., 2015
	Estuary area	2011	8				V	Hong et al., 2013
	Artificial Lake	2011	3				V	Hong et al., 2013
	Inland creek	2011	7				V	Hong et al., 2013
	Estuary	2012	2				V	Hong et al., 2015
West Sea (nation wide)	Bay & Coast	2009	15				V	Naile et al., 2013

*Details of references are in the main text.

Table S3. Hexachlorocyclohexanes (HCHs) and dichlorodiphenyltrichloroethanes (DDTs) (ng g^{-1} dw) in soils from coastal areas of Bohai and Yellow Seas in China and South Korea.

Location	Sampling time	HCHs min-max (mean)	DDTs min-max (mean)	Reference
China				
Tianjin	2008	2.1-12,549 (945)	n.d.-2,033 (88.4)	(Hou et al., 2013)
Haihe River Basin	2004	0.02-349 (--)	0.40-2,350 (--)	(Tao et al., 2008)
Yellow River Delta	2008	0.01-18.52 (3.34)	0.10-9.80 (1.41)	(Zhang, 2011)
Yellow River Estuary	2012	0.28-1.32 (0.35)	0.17-10.46 (0.63)	(Da, 2014)
Daling River Estuary	2001-2002	0.89-43.47 (5.18)	0.85-127.27 (30.24)	(Wang, 2013)
Coastal area of Bohai Sea	2005-2008	0.98-21.09 (4.45)	2.97-97.59 (24.35)	(Li, 2010)
Qingdao	2005	0.41-9.67 (4.01)	3.88-79.55 (26.51)	(Geng, 2006)
Korea				
Gyeonggi Province	2012	n.d.-0.12 (0.07)		(Kim et al., 2014)
Chungnam Province	2012	n.d.-0.05 (0.04)		(Kim et al., 2014)
Jeonbuk Province	2012	n.d.-0.26 (0.16)		(Kim et al., 2014)

n.d.: not detected; --: not available.

Table S4. HCHs and DDTs (ng L^{-1}) in water from rivers and marine areas of Bohai and Yellow Seas in China and South Korea.

Location	Sampling time	HCHs min-max (mean)	DDTs min-max (mean)	Reference
China				
Daling River Estuary	2001-2006	0.33-507.41 (52.72)	1.40-69.23 (14.13)	(Wang, 2013)
Yellow River Estuary	2012	-- (13.71)	-- (0.08)	(Da, 2014)
Haihe River Estuary	2006	1,290-5,900 (2,180)	520-4,840 (2,410)	(Wang, 2010)
Bohai Bay	2006	220-1,240 (590)	110-910 (430)	(Wang, 2010)
Bohai Bay	2011	50-130 (--)	70-210 (--)	(Wang et al., 2013b)
Liaodong Bay	2012	4.17-136.8 (--)*		(Yao et al., 2013)
Laizhou Bay	2005	n.d.-3.8 (--)*		(Xu et al., 2007)
Jiaozhou Bay	2005	0.1-3.9 (--)*		(Xu et al., 2007)
Korea				
Saemangeum Coast	2002	0.02-1.43 (0.36)	n.d.-8.19 (2.64)	(Hong et al., 2006)

*: Concentrations of OCPs; n.d.: not detected; --: not available.

Table S5. Polychlorinated biphenyls (PCBs) in soils (ng g^{-1} dw) and water (ng L^{-1}) from rivers and marine areas of Bohai and Yellow Seas in China and South Korea.

Location	Medium	Sampling time	PCBs min-max (mean)	Reference
China				
Tianjin	Soil	2008	n.d.-373 (46.2)	(Hou et al., 2013)
Qingdao	Soil	2005	3.06-14.88 (8.04)	(Geng, 2006)
Dalian	Soil	2007	1.3-4.8 (2.8)	(Wang et al., 2008)
Yellow River Delta	Soil	2008	9.3-124.2 (36.7)	(Zhang, 2011)
Daliao River Estuary	Water	2007	5.51-40.28 (--)	(Men et al., 2014)
Liaodong Bay	Water	2012	215.4-3,161 (--)	(Yao et al., 2013)
Haihe River	Water	2004	310-3,110 (760)	(Wang, 2010)
Bohai Bay	Water	2004	60-710 (210)	(Wang, 2010)
Bohai Bay	Water	2011	90-220 (--)	(Wang et al., 2013b)
Korea				
Gyeonggi Province	Soil	2016	0.11-0.22 (0.15)	(Kim et al., 2016)

n.d.: not detected; --: not available.

Table S6. Polybrominated diphenyl ethers (PBDEs) in soils (ng g^{-1} dw) from rivers and marine areas of Bohai and Yellow Seas in China and South Korea.

Location	Sampling time	PBDEs min-max (mean)	Reference
China			
Manufacture, Shandong	2016	73,216-226,906 (--)	(Deng et al., 2016)
North China	2011	0.08-8,264.87 (202.27)	(Liu et al., 2014)
Manufacture, Shandong	2011	17,000-146,000 (58,700)	(Li et al., 2015)
Coastal area, Laizhou Bay	2007	73-2,629 (--)	(Jin et al., 2011)
Yellow River Delta	2008	n.d.-18.26 (0.84)	(Chen et al., 2012)
Yellow River Delta	2012	0.26-3.72 (0.94)	(Yuan, 2013)
Manufacture, Shandong	2012	2.93-8,345.54 (879.25)	(Liu, 2013)
Electronic dismantling site, Tianjin	2012	1.34-343.88 (40.37)	(Liu, 2013)
Shandong	2011	0.011-948.84 (41.57)	(Liu, 2013)
Hebei	2011	0.019-35.09 (1.65)	(Liu, 2013)
Tianjin & Beijing	2011	0.012-3.32 (0.71)	(Liu, 2013)
Shanxi	2011	0.003-15.07 (0.78)	(Liu, 2013)
Korea			
Gyeonggi Province	2012	0.25-2.94 (1.13)	(Kim et al., 2014)
Chungnam Province	2012	0.01-0.26 (0.12)	(Kim et al., 2014)
Jeonbuk Province	2012	0.16-0.30 (0.21)	(Kim et al., 2014)
Jeonnam Province	2012	0.04-0.36 (0.25)	(Kim et al., 2014)

n.d.: not detected; --: not available.

Table S7. Hexabromocyclododecanes (HBCDs) in soils (ng g^{-1} dw) from rivers and marine areas of Bohai and Yellow Seas in China.

Location	Sampling time	HBCDs min-max (mean)	Reference
Manufacture, Shandong	2013-2014	0.11-560 (--)	(Yi et al., 2016)
Manufacture, Laizhou Bay	2010	0.88-6901 (--)	(Li et al., 2012)
Yellow River Delta	2012	0.82-12.14 (3.73)	(Yuan, 2013)
Coastal area, Bohai & Yellow Seas	2013	0.12-363 (--)	(Zhang et al., 2016)

--: not available.

Table S8. Perfluorooctane sulfonic acid (PFOS) and perfluorooctanoic acid (PFOA) in sediment (ng g^{-1} dw) from rivers and marine areas of Bohai and Yellow Seas in China and South Korea.

Location	Sampling time	PFOS min-max (mean)	PFOA min-max (mean)	Reference
China				
Coastal area, north Bohai Sea	2008	n.d.-1.97 (--)	n.d.-0.54 (--)	(Wang et al., 2011)
Coastal area, south Bohai Sea	2011	0.03-0.44 (--)	0.005-29.02 (--)	(Zhu et al., 2014)
Liaodong Bay Basin	2013	0.02-1.12 (--)	0.44-20.8 (--)	(Chen et al., 2015)
Bohai Sea	2011-2012	n.d.-0.15 (--)	0.06-2.70 (--)	(Gao et al., 2014)
Yellow Sea	2011-2012	n.d.-0.40 (--)	n.d.-1.52 (--)	(Gao et al., 2014)
Daling River	2009	n.d.	0.18-18 (--)	(Bao et al., 2011)
Daling River	2011	n.d.-0.47 (--)	0.02-0.55 (--)	(Meng et al., 2015b)
Xiaoqing River	2014	--	3.86-456.20 (--)	(Sun et al., 2016)
Haihe River	2010	1.76-7.32 (5.2)	0.92-3.69 (1.8)	(Li et al., 2011)
Haihe River	2008	0.15-0.79 (--)	n.d.-0.25 (--)	(Pan et al., 2011b)
Estuaries, Bohai Bay	2008	n.d.-4.3 (--)	n.d.-1.5 (--)	(Wang et al., 2012b)
Liaohe River	2009	0.04-0.48 (0.15)	0.02-0.18 (0.08)	(Yang et al., 2011)
Estuaries, Laizhou Bay	2009	n.d.-1.6 (--)	n.d.-76.9 (--)	(Zhao et al., 2013)
Laizhou Bay	2009	n.d.-0.06 (--)	0.07-1.8 (--)	(Zhao et al., 2013)
Korea				
Coastal area, West Sea	2009	0.10-5.80 (1.50)	0.10-2.40 (1.00)	(Naile et al., 2013)
North Han River	2012	0.01-0.07 (0.04)	n.d.-0.09 (0.04)	(Lam et al., 2014)
South Han River	2012	0.02-0.05 (0.18)	0.03-0.28 (0.07)	(Lam et al., 2014)
Yeongsan River	2012	0.05-0.11 (0.07)	n.d.-0.05 (0.02)	(Lam et al., 2014)

n.d.: not detected; --: not available.

Table S9. PFOS and PFOA in soils (ng g^{-1} dw) from coastal areas of Bohai and Yellow Seas in China and South Korea.

Location	Sampling time	PFOS min-max (mean)	PFOA min-max (mean)	Reference
China				
Suburban districts, Tianjin	2008	0.02-2.36 (0.19)	n.d.-0.51 (0.19)	(Pan et al., 2011b)
Estuaries, Bohai Bay	2008	n.d.-9.4 (1.76)	n.d.-0.93 (0.20)	(Wang et al., 2012b)
Coastal area, north Bohai Sea	2008	n.d.-0.7 (0.58)	n.d.-0.47 (0.21)	(Wang et al., 2011)
Coastal area, Liaodong Bay	2008	n.d.-0.42 (--)	n.d.-0.32 (--)	(Wang et al., 2013a)
Coastal area, Bohai Sea	--	n.d.-9.37 (--)	n.d.-13.30 (--)	(Meng et al., 2015a)
Korea				
Coastal area, West Sea	2009	0.10-1.70 (0.82)	0.10-3.40 (2.20)	(Naile et al., 2013)
Gyeonggi Province	2012	0.05-0.69 (0.42)	0.08-0.64 (0.34)	(Kim et al., 2014)
Chungnam Province	2012	--	-- (0.21)	(Kim et al., 2014)
Jeonbuk Province	2012	-- (0.13)	-- (0.06)	(Kim et al., 2014)
Jeonnam Province	2012	0.09-0.91 (0.50)	--	(Kim et al., 2014)

n.d.: not detected; --: not available.

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