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Assessment of trace pollutants in Korean coastal sediments using the triad approach: A review

Jong Seong Khim *, Seongjin Hong

School of Earth and Environmental Sciences & Research, Institute of Oceanography, Seoul National University, Seoul 151-742, Republic of Korea

HIGHLIGHTS

- Chemistry, toxicology, and ecology studies for sediment assessment are reviewed.
- Hot-spots of sediment contaminations closely associated with surrounding activities.
- Potential biological effects associated with sediments well reflected chemistry data.
- Benthic community responses tell the type and degree of sediment contamination.
- Sediment triad approach towards integrated assessment was well exampled in Korea.

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ABSTRACT

Here we summarize and review the previous efforts on sediment assessment together with major scientific findings that were conducted in the Korean coastal waters since late 1990s. Towards integrated triad analysis, sediment data (>1700 samples) reported from the Korean coasts were collected and reviewed of which data collectively includes three components of chemical, toxicological, and ecological measures. First, the chemistry data suggested widespread and historic distribution of sedimentary pollutants along the Korean coasts. Spatial distributions suggested that their sources were independent of each other, while some localized areas (highlighted for Lake Shihwa, Masan Bay, and Ulsan Bay) and zones with extremely high pressures of certain pollutants were also identified. The mass balance analyses and/or direct correlations linking triad components reflected a general agreement between endpoints. The benthic community responses given by species occurrence and diversity also reflected the type and degree of sediment contamination, however, could not be fully explained by the known target chemicals. Overall, the triad assessment of trace pollutants in Korean coastal sediments seemed to be useful and much powerful when all the components are fully addressed.

Contents

* Corresponding author at: School of Earth and Environmental Sciences & Research, Institute of Oceanography, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 151-742, Republic of Korea. Tel.: +82 2 880 6750; fax: +82 2 872 0311.
E-mail address: jskocean@snu.ac.kr (J.S. Khim).

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1. Introduction

Halogenated aromatic hydrocarbons (HAHs) such as polychlorinated dibenzo-*p*-dioxins and -dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), and polycyclic aromatic hydrocarbons (PAHs) are classes of compounds to act through an aryl hydrocarbon receptor (AhR) mediated mechanism and called dioxin-like compounds (Safe, 1990; Van den Berg et al., 2006; Zacharewski, 1997). Synthetic HAHs have been released to and thus widely distributed in the environment and finally sink in sediment in aquatic ecosystem, thus the sediment assessment is of continuing concern. Similarly, alkylphenols (APs) such as nonylphenols (NPs) and octylphenols (OPs) are degradation products of alkylphenol ethoxylates (APEOs) which are widely used in surfactants since the 1940s (White et al., 1994). In addition, butyltin compounds (BTs) such as tributyltin (TBT) have been widely used as biocidal additives in paint ships since the 1970s (Huggett et al., 1992). These chemicals tend to occur in sediment or suspended particles and can bioaccumulate and biomagnify in the food chain due to their highly lipophilic nature and resistance to biotransformation (Loonen et al., 1996; Schecter et al., 2005). Many of them have been reported to elicit estrogenic responses in aquatic organisms (Nimrod and Benson, 1996), thus certain chemicals such as NPs and BTs are of particular concern for human and/or wildlife.

These compounds elicit a variety of toxic responses in aquatic wildlife and laboratory animals which include mortality, weight loss, thymic atrophy, impairment of immune system, hepatotoxicity, reproductive alterations, birth defects, behavioral changes, and carcinogenicity (Neff, 1979; Nimrod and Benson, 1996; Safe, 1990; Sanderson et al., 1997). Some of these effects are thought to act through an AhR and/or estrogen receptor (ER) mediated mechanism (Safe, 1990; Van den Berg et al., 2006; Zacharewski, 1997). The increase of concern for wildlife is in a large part because adverse effects are often observed in wildlife before they are noticed in the human population. As a result increased research in wildlife toxicology is intended to create a better understanding of the responses of wildlife species and laboratory animals to environmental contamination. Today, environmental contamination with HAHs and endocrine disrupting chemicals (EDCs) and its threat to wildlife and human being has become a global issue, including South Korea.

1.1. Triad approach

To assess the environmental hazards of persistent organic pollutants (POPs) and EDCs to aquatic wildlife, it is important to examine the occurrence, fate, and distribution of contaminants in both sediment and water column. However, the procedures using instrumental analysis are usually time consuming and expensive due to the exhaustive clean-up required to remove all the interferences and the use of sophisticated and expensive instruments like high resolution gas chromatograph (HRGC). In addition, due to the complex nature of environmental mixtures, it is difficult to understand and/or predict potential effects of environmental contaminants based on instrumental analysis alone (Sanderson and Giesy, 1998). These limitations have bought the development of new and alternative methods which apply biological techniques to determine these compounds (Diaz-Ferrero et al., 1997; Sanderson and Giesy, 1998).

In vitro bioassays provide useful information which can complement instrumental analysis to provide a more complete characterization of environmental mixture samples (Giesy et al., 2002). Because they measure the ability of a sample to alter a biochemical or physiological process, in vitro bioassays can provide an indication of the biological relevance of a sample, for example, both additive and non-additive interactions between compounds can be accounted for. Using successive iterations of chemical fractionation and in vitro bioassay, followed by instrumental analysis, specific compounds, or classes of compounds, eliciting a particular biological response can be identified from complex mixtures containing multiple active and inactive compounds. Additionally potential non-additive interactions can be detected and defined. Finally, when applied along with experimentally determined toxic equivalency factors (TEFs) or relative potencies (REPs), bioassay-derived potency can be compared to theoretical potency (viz. instrumental derived potency) in a mass balance analysis (Sanderson et al., 1996).

The sediment quality triad approach is an effect-based approach that incorporates measures of various chemicals, potential toxicities, and benthic community structures (Fig. 1) (Chapman, 1986, 1989, 1990).

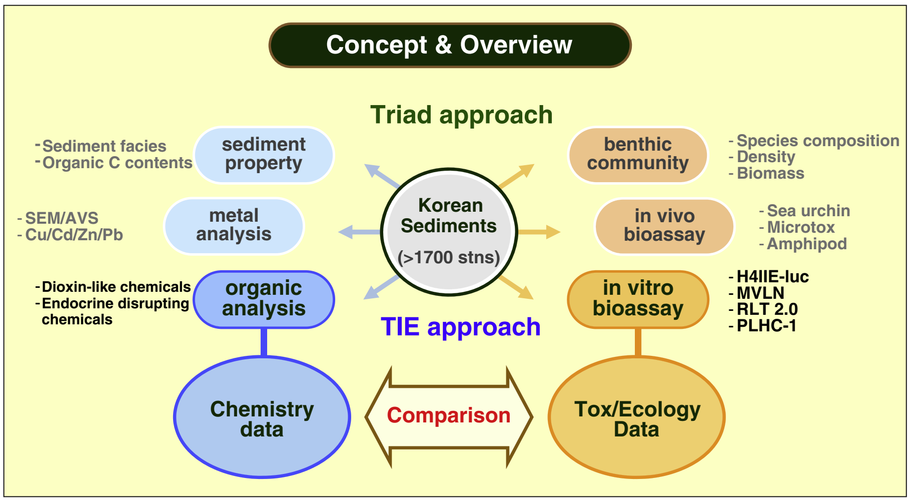

Fig. 1. The concept and overview of the sediment assessment study (triad approach).

The method provides a direct assessment of sediment quality and can be applied to all chemicals of concern, although it does not prove a cause-and-effect relationship between the concentrations of individual chemicals and adverse biological effects. Further this method could be supported by the lines of evidence approach which further considers the broad spectrum of physiological components to resident aquatic communities and their interactions (Chapman and Hollert, 2006). For example, the characterization of biomagnification looking for the chemical allocation through the food chain in aquatic environment or determining causation by sediment toxicity identification and evaluation (TIE), which has been also collectively applied on the sediment assessment of Korea (Fig. 1), would be worthy of addition.

1.2. Review framework

The coastal sediment contamination by POPs and EDCs in Korea has been extensively monitored only after the mid-1990, although this issue has long been of significant concern world-wide during the past several decades. However, a rapid growth in research activities has been shown during the past 15 years reporting sedimentary POPs study in Korea, which covers >66 studies with survey at a total of >1700 locations for hundreds of environmental contaminants (Table 1 and Fig. 2) (Cho et al., 2004; Choi et al., 1997, 2001a, 2002, 2003, 2007, 2009a,b,c, 2010, 2011a,b; Hong et al., 2001, 2003, 2004, 2005, 2006a,b, 2009a,b, 2010, 2012a,b; Im et al., 2002; Jeong et al., 2001, 2012; Jung et al., 2007; Kannan et al., 2007; Khim et al., 1999a,b,c, 2001a,b; Kim et al., 1997, 1998, 2008, 2011a,b; Kim and Park, 2001; Koh et al., 2001, 2002, 2004, 2005, 2006a,b; Lee et al., 2001a,b, 2012; Li et al., 2004a,b,c, 2008; Moon et al., 2001, 2008a,b, 2012a; Naile et al., 2011; Oh et al., 2003; Shim et al., 1999, 2004, 2005, 2010; Terauchi et al., 2009; Woo et al., 2006; Yim et al., 2005, 2007). The Korean government has also forced to investigate the environmental occurrence of many classes (>90 compounds) of POPs and EDCs in various media such as water, soil, air, and sediment during the periods of 1999–2003. Now, the issue of the environmental pollution by POPs and EDCs in Korea becomes one of the hot issues to the public and governmental people as well as the scientists.

Considering the strong dependence of coastal communities on marine resources (e.g. fishery products) in Korean peninsula, the management of coastal environment from POPs contamination would be of great need and concern nationwide. Although the production and/or use of several POPs, such as PCBs and organochlorine pesticides (OCPs) have been banned in Korea since the 1970s, these compounds continue to persist in the coastal environment of Korea (Hong et al., 2006b; Kim et al., 2007) and further enter into the food chain (Choi et al., 2001b; Khim et al., 2000; Kim et al., 2002; Moon et al., 2010). At the beginning stage of the POPs study in Korea, most studies were limited to report the environmental levels of classical POPs such as PCBs (Lee et al., 2001a; Oh et al., 2003) and OCPs (Lee et al., 2001b). Later, several studies have identified the PAHs as prominent contaminants in sediments from Korean coastal areas (Kim et al., 1997; Koh et al., 2002). Meantime, the elevated concentration of APs such as NPs and OPs and BTs such as TBTs has been frequently reported in lake (Hong et al., 2010; Koh et al., 2006a; Li et al., 2004a,b, 2008), bays (Khim et al., 1999a; Kim et al., 2011a; Li et al., 2008; Shim et al., 2004), and coastal sediments (Hong et al., 2010; Koh et al., 2006a; Li et al., 2004a,c, 2008; Shim et al., 2005).

Despite the potential for direct and accidental releases of these pollutants into Korean coastal areas, still little was known for the potential biological effects over the broad range of compounds that were introduced in Korea. During the course of our study, H4IIE-luc (Sanderson et al., 1996) and MVLN (Pons et al., 1990) bioassays have been employed to aid the understanding of the potential biological effects associated with chemicals in sediment extracts (Table 1). They have been effectively used as a screen for detecting dioxin-like and estrogenic contaminants in extracts of sediment, surface waters, and animal tissues (Giesy et al., 2002). These bioassays could help in characterizing the total load and potency of dioxin-like and estrogenic chemicals associated with sediment samples from Korean coastal waters.

In the present study, we collected and reviewed major findings that determined the contamination of POPs and EDCs in Korean coastal sediments since the late 1990s up to date. Both instrumental and bioanalytical data obtained from >120 sites along the west, south, and east coasts of Korea have been scrutinized to overview POPs contamination and their potential biological effects. The present article was aimed to specifically document the 1) concentration and distribution of major groups of POPs and EDCs (PCDD/Fs, PCBs, OCPs, PAHs, NPs, and BTs), 2) potential biological effects mainly AhR and ER activities associated with sediments, 3) comparison of instrumental analyses and bioassays, and 4) benthic infaunal communities in Korean coastal sediments, especially targeting the hotspot areas including Lake Shihwa, Masan Bay, and Ulsan Bay for future monitoring and risk assessment use. Overall, the present study would provide the most up-to-date information and understanding on the POPs contamination in Korean sediments in terms of triad approach, which guides future direction and research priority needs into the routine POPs monitoring, assessments, and management practices.

2. Site descriptions: hot-spot areas

The Lake Shihwa, Masan Bay, and Ulsan Bay were the most frequently studied (over half among the total studies) in the Korean coasts as hot-spot areas of sediment contaminations and biological effects (Table 1 and Fig. 2), thus selected as model sites for the descriptive review. The Lake Shihwa was an artificial lake which had been isolated after construction of sea dike (12.7 km) in 1994 for freshwater supply to industrial and agricultural regions (Table 2 and Fig. 2). Due to the insufficient wastewater treatment facilities and increasing pollutant loads from the surrounding industrial complexes and cities, Lake Shihwa environments had been rapidly deteriorated (Khim et al., 1999c; Li et al., 2004c). Consequently, the Korean government abandoned its original plan to change Lake Shihwa into a freshwater lake and constructed a water gate in 1999 for the purpose of seawater circulations (Li et al., 2004a). Although water quality has improved in the vicinity of the gate, various organic pollutants such as PCDD/Fs (Moon et al., 2012a), polybrominated diphenyl ethers (PBDEs) (Moon et al., 2012b), PCBs (Khim et al., 1999c; Lee et al., 2001a), OCPs (Lee et al., 2001b), PAHs (Khim et al., 1999c; Koh et al., 2005), perfluorinated alkyl compounds (PFAs) (Rostkowski et al., 2006), and NPs (Hong et al., 2010; Li et al., 2004a,c) have been found in sediments from the inner region of Lake Shihwa and surrounding inland creeks in recent years.

Next, the Masan Bay is a long and narrow inlet of semi-closed bay, located on the south coast of Korea (Table 2 and Fig. 2). Because of geographical characters, Masan Bay has shown a slow rate of water exchange and a trapping effect on contaminants discharged from surrounding industrial complexes and cities. Masan and Changwon industrial complexes, including petrochemical, heavy metal, electrical, and plastic industries, as well as heavily populated cities, are located adjacent to the bay (Hong et al., 2009b; Khim et al., 1999a; Moon et al., 2008b). Masan Bay has been listed as a Special Management Coastal Zone (SMCZ) by the Korean government since 2000, in association with increased contaminant levels (Choi et al., 2009a,b). The POPs and EDCs in Masan Bay environments have been frequently reported with relatively great concentrations compared to other regions (Hong et al., 2003; Khim et al., 1999a; Kim et al., 2011a; Li et al., 2008; Moon et al., 2008b; Yim et al., 2005). These studies showed that Masan Bay was heavily polluted by the trace organic chemicals originating from surrounding industrial complexes and municipal cities.

Finally, the Ulsan Bay that is the highly industrialized region located at the east coast of Korea is considered to be one of the most contaminated areas in Korea. The largest petrochemical industrial complexes had been developed in Ulsan since the middle of the 1970s. The potential toxic effects of hazardous chemicals emitted from petrochemical

Table 1
List of sediment assessment studies in the Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Chemical analysis						Bioassay		References
				PCDD/Fs	PCBs	OCPs	PAHs	NPs	BTs	AhR	ER	
West Sea												
A. Lake Shihwa	Inland	2000	8		V	V	V	V		V	V	Koh et al. (2005)
		2000	18					V				Li et al. (2004a)
		2008	12	V								Moon et al. (2012a)
	Lake	1996	4		V	V						Lee et al. (2001a, 2001b)
		1998	11		V	V	V	V		V	V	Khim et al. (1999c)
		2000	12					V				Li et al. (2004a)
		2002	10					V				Li et al. (2004c)
		2005	1	V								Terauchi et al. (2009)
		2006–2007	5					V				Choi et al. (2009c)
		2008	7					V				Hong et al. (2010)
		2008	16	V								Moon et al. (2012a)
1. Gyeonggi Bay	Bay	1995	67				V					Kim et al. (1997)
		1995	40						V			Kim et al. (1998)
		1996	47		V	V						Lee et al. (2001a, 2001b)
		2000	13		V							Hong et al. (2005)
		2003	33		V	V	V	V	V			Hong et al. (2009a)
		2008	6	V								Moon et al. (2012a)
		2008	7					V				Hong et al. (2010)
2. Incheon Harbor	Bay	1996	7		V	V						Lee et al. (2001a, 2001b)
3. Namyang Bay	Bay	1996	5		V	V						Lee et al. (2001a, 2001b)
4. Asan Bay	Bay	2005	1	V								Terauchi et al. (2009)
		2005	6		V	V						Choi et al. (2011b)
5. Taean oil spill area	Coast	2009	50				V			V		Hong et al. (2012b)
6. Gunsan	Coast	2008	10				V	V	V			Lee et al. (2012)
7. Saemangeum	Coast	2002	13			V						Hong et al., 2006a
8. Mokpo	Coast	2006	29					V				Choi et al. (2007)
South Sea												
B. Masan Bay	Inland	2000	8		V	V	V	V		V	V	Koh et al. (2005)
	Bay	1992	11	V								Im et al. (2002)
		1996	10						V			Choi et al. (1997)
		1997	20		V	V						Hong et al. (2003)
		1997	20				V					Yim et al. (2005)
		1998	28		V	V	V	V		V	V	Khim et al. (1999a, 1999b)
		1998	1	V								Oh et al. (2003)
		2004	21	V								Kannan et al. (2007)
		2004	18					V				Li et al. (2008)
		2004	21						V			Kim et al. (2011a)
		2005	20	V			V	V				Moon et al. (2008b)
		2005	1	V								Terauchi et al. (2009)
		2006	5	V				V				Hong et al. (2009b)
		2006–2007	5					V				Choi et al. (2009c)
9. Gwangyang Bay	Bay	1996	12						V			Kim and Park (2001)
		1999–2000	6					V				Cho et al. (2004)
		2000	6		V							Hong et al. (2005)
		2000–2002	35	V								Moon et al. (2008a)
		2001	11		V	V	V	V		V	V	Koh et al. (2005)
		2001	15					V				Li et al. (2004b)
		2001	23		V	V						Hong et al. (2004)
		2001	35						V			Choi et al. (2002)
		2001	17						V			Shim et al. (2004)
		2003	13				V					Woo et al. (2006)
		2006	3	V								Kim et al. (2008)
		2006–2010	8	V								Jeong et al. (2012)
10. Busan Bay	Bay	2000	10		V							Hong et al. (2005)
		2000	5		V				V			Choi et al. (2001a, 2003)
		2000–2002	21	V								Moon et al. (2008a)
		2002–2003	3	V								Jung et al. (2007)
		2002–2004	40						V			Choi et al. (2009b)
		2005	1	V								Terauchi et al. (2009)
		2006–2007	5					V				Choi et al. (2009c)
		2007	27						V			Choi et al. (2010)
11. Jinhae Bay	Bay	1995	55						V			Shim et al. (1999)
		2000	5		V				V			Choi et al. (2001a, 2003)
		2000–2002	26	V								Moon et al. (2008a)
		2002–2003	15	V								Jung et al. (2007)
		2002–2004	35						V			Choi et al. (2009b)
		2007	16		V	V	V	V	V			Shim et al. (2010)
12. Yeosu Bay	Bay	1999–2000	9					V				Cho et al. (2004)
13. Gohyeon Bay	Bay	2003	18						V			Kim et al. (2011b)
14. Okpo Bay	Bay	1995	20						V			Hong et al. (2001)
15. Haengam Bay	Bay	2005	1	V								Terauchi et al. (2009)

(continued on next page)

Table 1 (*continued*)

Regions	Type	Sampling year (y)	Site number (n)	Chemical analysis						Bioassay		References
				PCDD/Fs	PCBs	OCPs	PAHs	NPs	BTs	AhR	ER	
16. Nakdong River	River	1999	18		V							Jeong et al. (2001)
	Estuary	2002–2003	11	V								Jung et al. (2007)
East Sea												
C. Ulsan Bay	Inland	1999	6		V	V	V	V		V	V	Khim et al. (2001a, 2001b)
	Inland	1999	5		V	V	V	V		V	V	Khim et al. (2001a, 2001b)
	River	1999	4		V	V	V	V		V	V	Khim et al. (2001a, 2001b)
	Bay	1996	3	V								Oh et al. (2003)
		1997	30				V					Koh et al. (2001)
		1999	16		V	V	V	V		V	V	Khim et al. (2001a, 2001b)
		2000	11		V							Hong et al. (2005)
		2000	6		V				V			Choi et al. (2001a, 2003)
		2000	22				V					Moon et al. (2001)
		2000–2002	20	V								Moon et al. (2008a)
		2002–2003	9	V								Jung et al. (2007)
		2002–2004	39						V			Choi et al. (2009b)
		2005	1	V								Terauchi et al. (2009)
		2006–2007	5					V				Choi et al. (2009c)
17. Yeongil Bay	Inland	2001	6	V	V		V			V	V	Koh et al. (2004)
	Bay	1996	1	V								Oh et al. (2003)
		2000	9		V							Hong et al. (2005)
		2000	26		V	V	V	V		V	V	Koh et al. (2006a, 2006b)
		2000	5		V				V			Choi et al. (2001a, 2003)
		2000–2002	20	V								Moon et al. (2008a)
		2005	1	V								Terauchi et al. (2009)
18. Onsan Bay	Inland	1999	6		V	V	V	V		V	V	Koh et al. (2002)
	River	1999	3		V	V	V	V		V	V	Koh et al. (2002)
	Bay	1999	13		V	V	V	V		V	V	Koh et al. (2002)
		2005	1	V								Terauchi et al. (2009)
19. Uljin	Coast	2005	1	V								Terauchi et al. (2009)
Nationwide												
20. Nationwide	Bay & coast	1997	20						V			Shim et al. (2005)
		1997–2002	138		V	V						Hong et al. (2006b)
		2000–2001	117				V					Yim et al. (2007)
		2001–2005	20–25						V			Choi et al. (2009a)
		2001–2007	25	V	V		V					Choi et al. (2011a)
		2006–2007	15					V				Choi et al. (2009c)
		2008	8	V						V		Hong et al. (2012a) and Naile et al. (2011)

industries to aquatic organisms and human health are of serious concern (Khim et al., 2001a). Various organic chemicals in sediments have been detected widely in Ulsan Bay and its vicinity (Choi et al., 2003; Hong et al., 2005; Khim et al., 2001a,b; Koh et al., 2001; Moon et al., 2008a; Oh et al., 2003). Additional descriptions on the backgrounds, social and legal histories, major environmental concerns, and study efforts in the Lake Shihwa, Masan Bay, and Ulsan Bay are presented in Table 2.

3. Distributions and sources of organic contaminants

Data reported for the sedimentary POPs and EDCs concentrations were collected for >1700 samples from 22 coastal regions of Korea from the published international and domestic journal articles (total of 66 papers) for review (see Table 1, Figs. 2 and 3; summarized in Table S1–S6 of the Supplemental materials). Spatio-temporal distributions and sources of PCDD/Fs, PCBs, OCPs, PAHs, NPs, and BTs in Korean coastal sediments are discussed in this section. Based on the number of studies, efforts on the POPs and EDCs in Korean sediments are frequently given in the regions of Lake Shihwa, Masan Bay, and Ulsan Bay, where relatively great concentrations were found compared to other regions across the coasts.

3.1. Lake Shihwa

All of the target organic contaminants have been detected in Lake Shihwa sediments (Fig. 3), where relatively greater concentrations were found in the inland creeks than in the lake. Previous studies repeatedly found that high degree of contamination in the creeks was mainly affected by the heavy industrial and municipal activities (Koh et al., 2005; Li et al., 2004a; Moon et al., 2012a,b). Among the organic contaminants, PCDD/Fs and NPs were found to be uppermost of which concentrations exceeded the sediment quality guidelines (probable effect concentrations). Especially, concentrations of NPs were remarkably great in inland creek sediments, showing the greatest levels nationwide (Choi et al., 2009c; Hong et al., 2010; Li et al., 2004c).

However, it should be noted that the temporal trends of NPs in the lake have shown a decreasing trend particularly after 2002. This trend seemed to be associated with the effort of restriction of NPs, banning on household uses of NPEOs since 2002, or general water quality improvement by exchanging water gate since 1999 (Hong and Shin, 2011). Meanwhile, sedimentary PCB and OCP concentrations in some locations also exceeded the lower guidelines (viz., threshold effect concentrations), but such temporal trends could not be clearly found due to limited data points.

Overall, studies on POPs and EDCs in sediments of Lake Shihwa suggested that sediment qualities would have been improved by the pollution control and/or restriction of chemicals in recent years. However, emerging contaminants such as PBDEs and PFCs in inland sediments of Lake Shihwa were recently reported to be serious (Moon et al., 2012a,b; Rostkowski et al., 2006), not parallel reviewed in the present study. Thus continuing monitoring efforts and environmental management planning in this particular concern of area, Lake Shihwa, would be highly recommended.

3.2. Masan Bay

In Masan Bay, the concentrations of sedimentary POPs and EDCs were relatively great, similar to Lake Shihwa (Fig. 3). In particular,

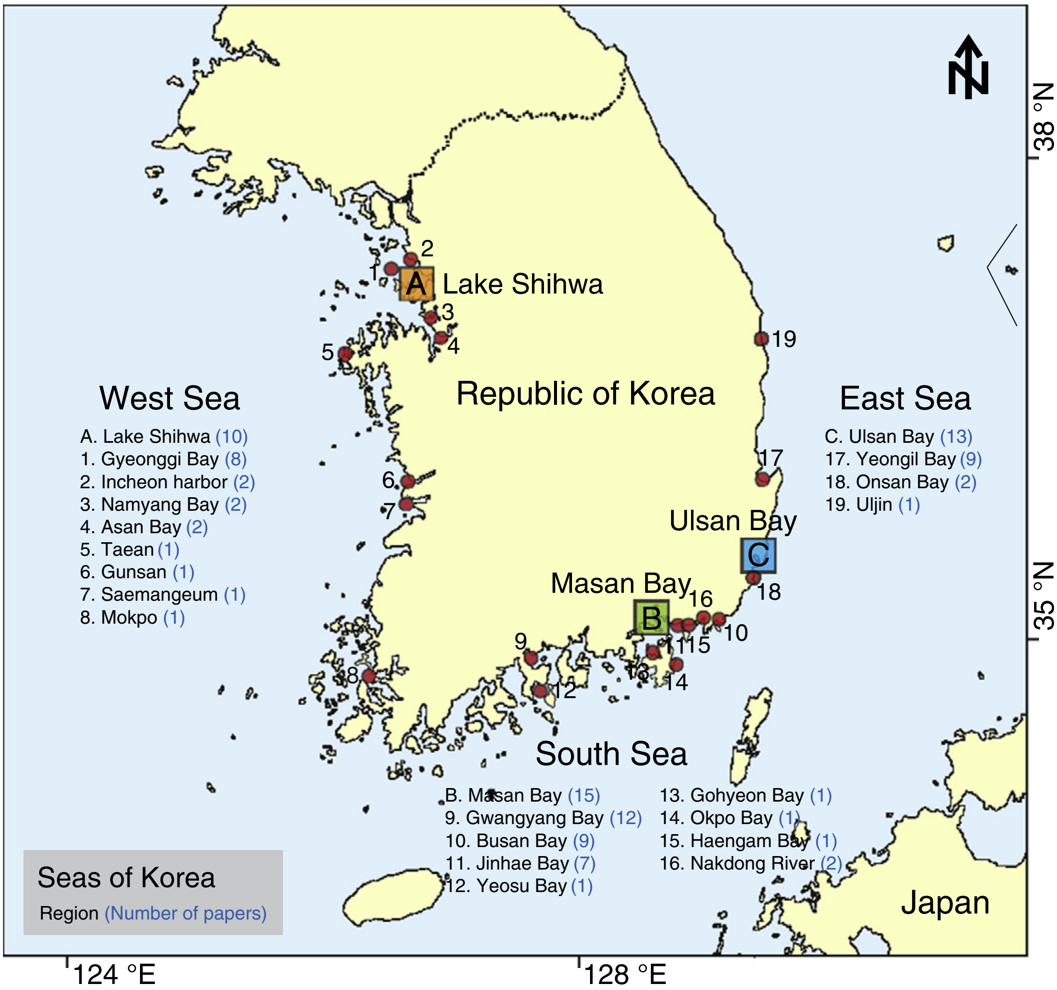

Fig. 2. Map showing the locations of study area for the sediment assessment in the Korean coastal waters reviewed in the present study; number of studies given in parenthesis. Three regions of Lake Shihwa (A), Masan Bay (B), and Ulsan Bay (C) are highlighted for the descriptive review.

PCDD/Fs in Masan Bay sediments have long been detected at levels above the upper sediment quality guidelines (probable effect levels) since 1992. Such chemicals seem to be present with long residence time in the Masan Bay environment due to a slow rate of water exchange according to the geographical feature of semi-enclosed system. Previous studies indicated that the major pollution pressures in Masan area would be industrial wastewater treatment plants (WWTPs) discharge and diffuse sources such as surface runoff (Hong et al., 2009b; Khim et al., 1999a; Li et al., 2008; Moon et al., 2008b).

Temporal trends of NPs have shown to decrease in Masan Bay sediments over 15 years, while PCDD/Fs and PAHs were found to be with the similar range during the corresponding period. Recently, a total pollutant load management system (TPLMS), as part of the implementation of water quality improvement, has been launched in Masan Bay since 2007. TPLMS project comprises a total of 800 km of combined sewer networks that convey sewage to two central WWTPs that have been replaced by an advanced biological treatment facility with an extended capacity of 500,000 ton d^{-1} (82% of total freshwater input) (Chang et al., 2012). TPLMS project has been successfully conducted to improve the general water quality, targeting COD and SS reductions, in Masan Bay.

3.3. Ulsan Bay

Concentrations of target organic contaminants in sediments of Ulsan Bay were found to be generally similar, except for OCPs and NPs, compared to those of Lake Shihwa and Masan Bay (Fig. 3). Among the given chemicals, PCBs, PAHs, and BTs were found to be relatively great in sediments of Ulsan Bay, which are mainly originating from surrounding industrial areas such as the petrochemical plant, a shipyard, and a mechanical complex (Choi et al., 2003; Moon et al., 2008a). The NP concentrations in Ulsan Bay sediments were generally low compared to those in other industrialized bays in Korea (Choi et al., 2009c), however. Due to the limited information, temporal trends of POPs and EDCs in sediments in Ulsan could not be fully addressed this time.

3.4. Other bay and coastal areas

The great concentrations of POPs and EDCs in sediments were not limited to the hot-spot areas of Lake Shihwa, Masan Bay and Ulsan Bay, but they were also found in other highly industrialized regions (Fig. 3). According to the previous reports, the notable priority chemicals in other regions were found to be: i) PCBs and OCPs in Gyeonggi Bay (Lee et al., 2001a,b), ii) PAHs in Gwangyang Bay (Woo et al., 2006), iii) PCBs in Busan Bay (Hong et al., 2005), iv) PCDD/Fs in Pohang area (Koh et al., 2004), and v) BTs in Busan Bay (Choi et al., 2003, 2009b, 2010), Jinhae Bay (Choi et al., 2003, 2009b; Shim et al., 1999, 2010), Gohyeon Bay (Kim et al., 2011b), and Okpo Bay (Hong et al., 2001). Overall, the distributions and sources of POPs and EDCs in Korean coastal sediments are shown to be in site-specific and source-dependent manners. The PCDD/Fs, PCBs, and PAHs were generally found to be uppermost in sediments nearby industrial complexes,

Table 2
Site descriptions on the hot-spot areas in the Korean coastal waters (for descriptive review).

Regions	Lake Shihwa	Masan Bay	Ulsan Bay
Location	West coast	South coast	East coast
Geography	Artificial lake	Semi-closed bay	Bay
Surrounding activities	Cities (population) –Ansan: 714,956 –Shiheung: 441,147 –Hwasung: 539,522 Industrial complex (chemical, wood, textile, metal, foodstuff, papers, and printing)	City –Changwon: 1,107,955 Industrial complex (petrochemical, heavy metal, electrical, and plastics)	City –Ulsan: 1,145,096 Industrial complex (petrochemical and chemicals)
Causes of pollution	Developed industrial complex (1986) Construction of sea dike (isolation,1994) Wastewater loads	Developed industrial complex (1970) Poor water circulation Wastewater loads	Developed industrial complex (1975) Wastewater loads
Environmental management	Established SWQ management (1996) Renunciation of irrigation water (1998) Designation of SMCZ (2000) Implementation of SCR EMMP (2007) Implementation of TPLMS (2011)	Designation of red tide management (1982) Sediment dredging (1994) Designation of SMCZ (2000) Upgrade of WWTP (2003) Implementation of TPLMS (2007)	Designation of SMCZ (2000) Plan for coastal management (2008)
Major environmental concerns	Anoxic condition in inner parts Heavy metals and organic pollutions Surrounding inland creeks	Red tide outbreak Nitrogen and phosphorous Wastewater treatment outfall	Dioxin-like activity Industrial area Sediment pollution
Chemicals of concerns[a]	PCDD/Fs and PBDEs	DDTs, PCBs, and *p,p′*-DDD	DDTs, *p,p′*-DDD, and PAHs
Number of papers[b]	31 (1997 to present)	62 (1988 to present)	16 (1985 to present)
Water	6	12	2
Sediment	15	22	9
Biota	9	20	3
Etc.	1	8	2

SWQ: special water quality.
SMCZ: Special Management Coastal Zone.
SCR EMMP: Shihwa coastal reservoir environmental management master plan.
TPLMS: total pollutant load management system.
WWTP: wastewater treatment plant.
[a] Referred from Khim and Koh (2011) and Moon et al. (2012a,b).
[b] Referred from SCOPUS (www.scopus.com).

elevated concentrations of BTs were found in the large scale shipyard, and NPs were mainly detected in inland creeks surrounding industrial complexes and municipal cities. The concentrations of some trace organic chemicals in Korean coastal sediments did exceed the existing sediment quality guidelines, thus could have potential adverse effects on benthic organisms.

4. Potential biological effects: AhR and ER activities

Several studies reported the potential biological activities associated with AhR and ER active agonists in Korean coastal sediments adopting in vitro H4IIE-luc and MVLN bioassays, respectively. All available data of in vitro bioassays were collected for >200 sediments from 7 coastal regions of Korea in the published literature (total of 12 papers) (see Table 1 and Fig. 2). Dioxin-like and estrogenic activities in sediments were quantitatively expressed as TCDD-EQ (2,3,7,8-TCDD) and E2-EQ (17-β-estradiol) concentrations (Khim et al., 2001b).

The potential AhR and ER activities in inland areas were comparable and/or slightly greater than those in lake and/or bays, in geographical manner (Fig. 4). Concentrations of TCDD-EQs and E2-EQs in Korean coastal sediments showed that their distributions were in chemical- and source-dependent manners. For example, great concentrations of E2-EQs were found in the Lake Shihwa indicating that elevated concentrations of EDCs were present in corresponding sediments (Khim et al., 1999c; Koh et al., 2005). The MVLN bioassay result well reflected the high degree of NP contaminations, particularly in inland creeks of Lake Shihwa (Choi et al., 2011c; Hong et al., 2010; Li et al., 2004a,c). In Masan Bay, however, bay sediments showed greater biological responses compared to inland sediments (Khim et al., 1999a,b; Koh et al., 2005), which was not the case for Lake Shihwa. It was indicated that the elevated TCDD-EQs and E2-EQs in Masan Bay sediments seemed to be affected by point sources in outer locations and/or possible long-term accumulation in sediments due to trapping effect in semi-closed bay system (Hong et al., 2003, 2009b; Li et al., 2008; Moon et al., 2008b).

Similar to Lake Shihwa, the greatest TCDD-EQs were found in the inland sediments of the Ulsan Bay (Khim et al., 2001a,b). It could be explained by great concentrations of known dioxin-like chemicals, such as PCDD/Fs, PCBs, and PAHs, in the inland creek sediments in the vicinities of industrial complexes. However, the ER-binding activities in Ulsan Bay sediments were relatively low compared to those in other regions that seemed to reflect the low to moderate levels of NP contaminations (Choi et al., 2009c). Similarly, great TCDD-EQ and low E2-EQ concentrations were found in Yeongil Bay. Previous studies conducted in Yeongil Bay collectively suggested that the majority of dioxin-like activities was explained by the PCDD/Fs, dioxin-like PAHs, and coplanar PCBs in the order (Koh et al., 2004, 2006a,b). And those chemicals were mainly originated from the surrounding iron and steel manufacturing industries in this region (Choi et al., 2008; Fang et al., 2011).

Other in vitro and/or in vivo bioassays for potential toxicity associated with Korean coastal sediments together with chemical analyses of POPs and EDCs have been reported occasionally (Ji et al., 2011; Jung et al., 2012; Lee et al., 2011; Seo et al., 2008; Won et al., 2012; Woo et al., 2006). The results of these potential toxicity tests indicated that biological responses such as genotoxic effect, endocrine disruption, and enzyme activities were significantly induced in highly polluted sediments affected by industrial and municipal activities and/or oil spill. Overall, the potential toxic effects associated with the Korean coastal sediments generally well reflected the chemical concentrations and compositions of which sources are closely related with the surrounding anthropogenic activities.

5. Comparisons of chemical analyses and in vitro bioassays

The results of chemical analyses and in vitro bioassays in Korean coastal sediments were compared with all of the reviewed data

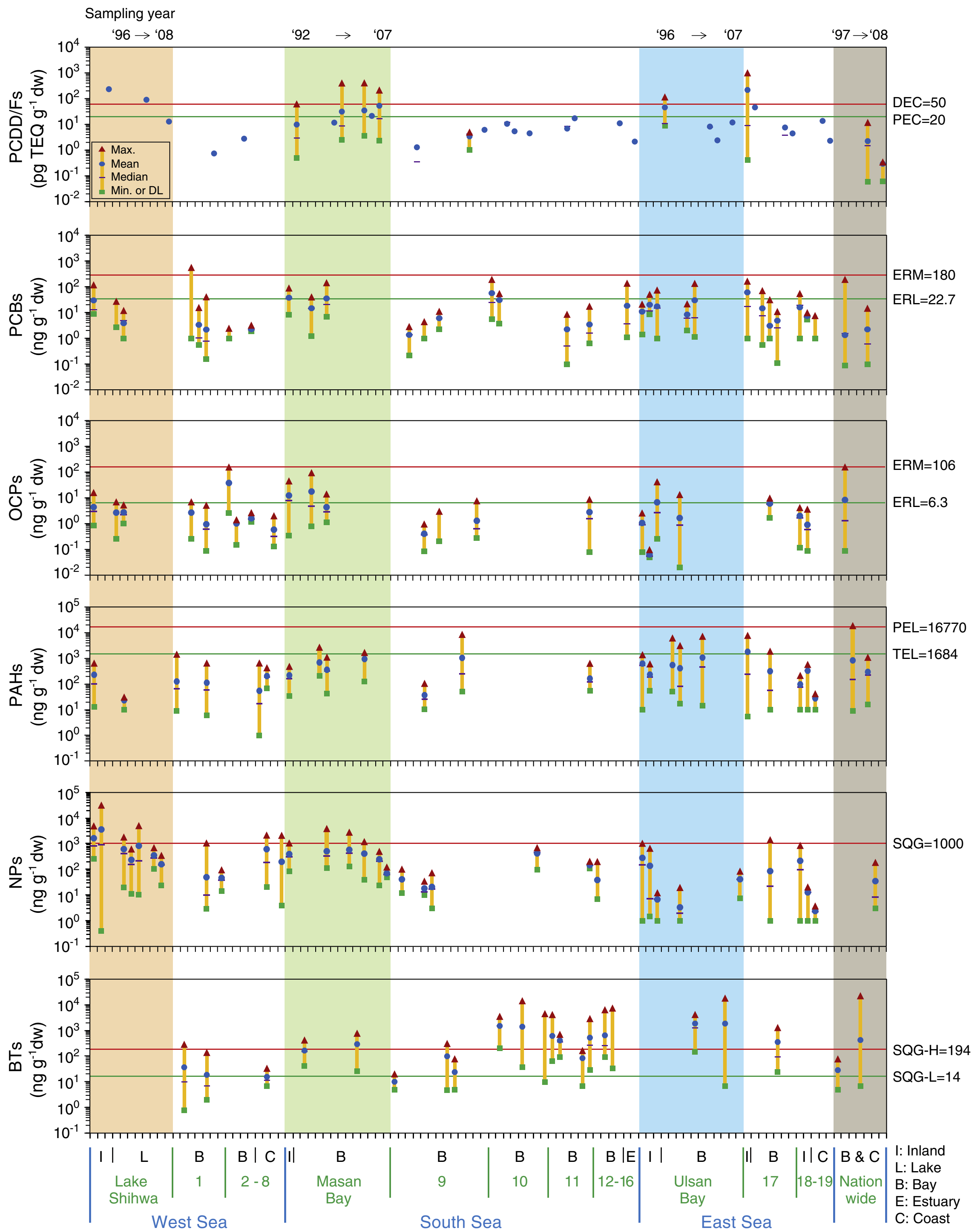

Fig. 3. Concentrations and distributions of target compounds of PCDD/Fs, PCBs, OCPs, PAHs, NPs, and BTs in sediments from the Korean coastal waters. Studies of Lake Shihwa (red), Masan Bay (green), Ulsan Bay (blue), and nationwide monitoring (gray) are highlighted. Location ID given refers to Table 1 and Fig. 2. Horizontal solid lines indicate the existing sediment quality guidelines for each chemical groups (red: probable effect concentrations (DEC, ERM, PEL, and SQGs-H); green: threshold effect concentrations (PEC, ERL, TEL, and SQG-L)).

(Table 1 and Fig. 5). The H4IIE-luc responses (%$TCDD_{max}$) were significantly correlated with the concentrations of known AhR agonists such as PCBs and PAHs in sediments (Fig. 5a). This result revealed that total induced dioxin-like activities in sediments were partly explained by the concentrations of dioxin-like PCBs and PAHs in the Korean coastal sediments. The correlation of MVLN responses (%$E2_{max}$) and NP concentrations was shown to be weak for the same set of sediment samples (Fig. 5b). It is indicated that endocrine disruption potentials

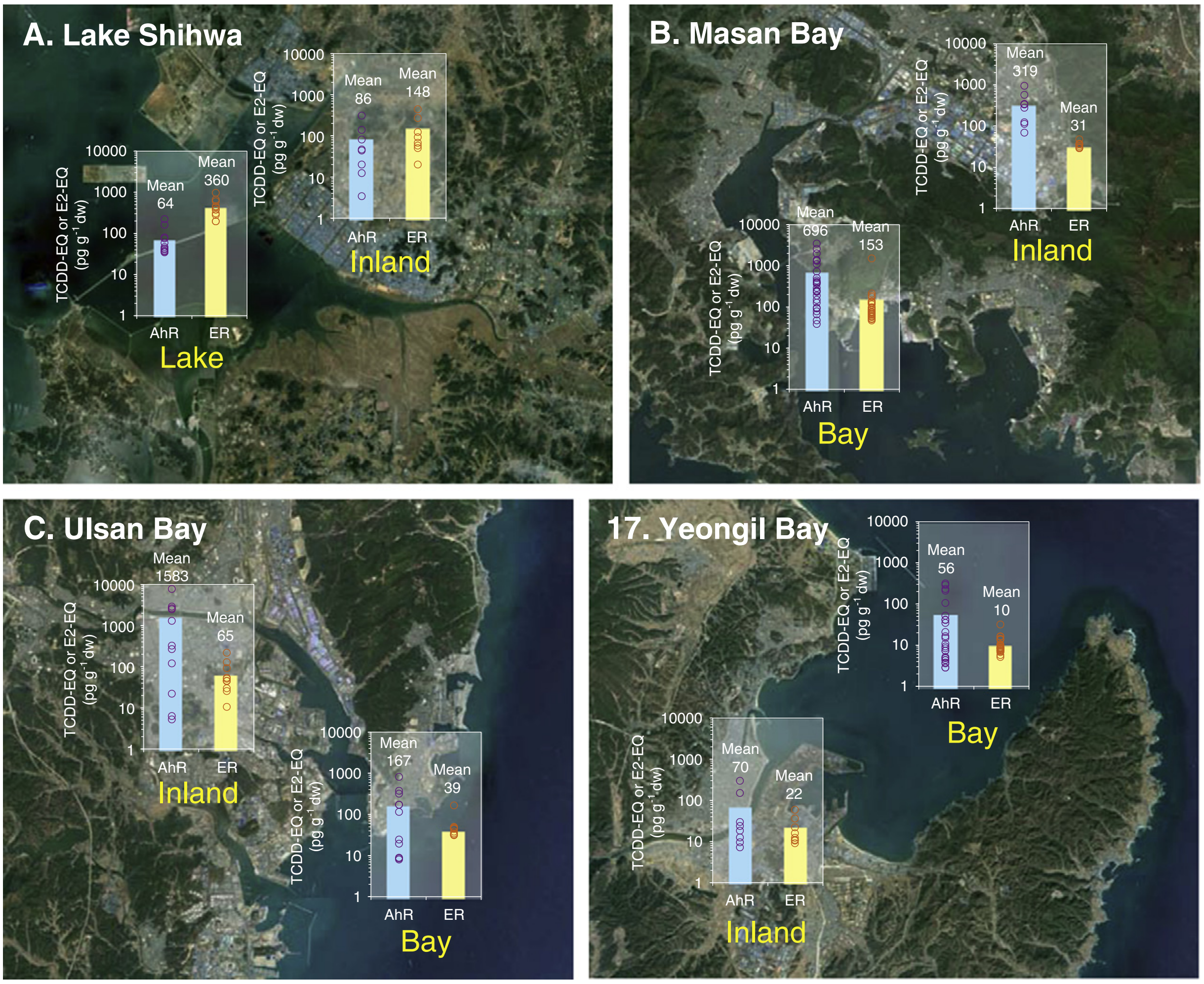

Fig. 4. Concentrations of TCDD-EQs and E2-EQs in sediment samples from the inland, lake, and bay of the Lake Shihwa (A), Masan Bay (B), Ulsan Bay (C), and Yeongil Bay (17).

in sediments could not be fully explained solely by the NP concentrations, suggesting that other unknown ER active compounds are present.

The comparison between bioassays and chemical analyses can be used to identify the contribution of each chemical in sediments to the total toxic potentials by a potency based mass balance analysis (Giesy et al., 2002; Hong et al., 2012a; Khim et al., 2001b; Koh et al., 2004). To do this, assay-specific relative potencies are multiplied by the concentrations of individual known chemicals and reported as 2,3,7,8-TCDD equivalents (TEQs) and 17-β-estradiol equivalents (EEQs) (Giesy et al., 2002). Earlier, the mass balance analysis was successfully applied to evaluate the dioxin-like activities in Hyeongsan River (inland river of Yeongil Bay) sediments (Koh et al., 2004). The sum of the TEQs of PCDD/Fs, co-planar PCBs, and dioxin-like PAHs accounted for majority of the bioassay TCDD-EQs in sediments. The 2,3,4,7,8-PeCDF was the most contributing congener (~60%) in the Hyeongsan River sediment, followed by 1,2,3,7,8-PeCDD (~12%) and 2,3,7,8-TCDF (~7.4%). However,

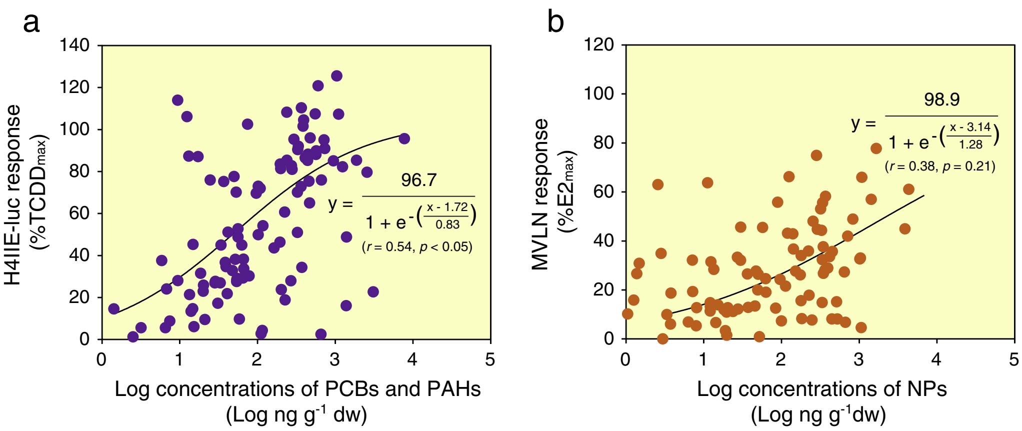

Fig. 5. Comparison between chemical analysis and biological response in sediments from the Korean coastal waters. (a) Relationship between PCB and PAH concentrations vs. H4IIE-luc responses and (b) relationship between NP concentrations vs. MVLN responses.

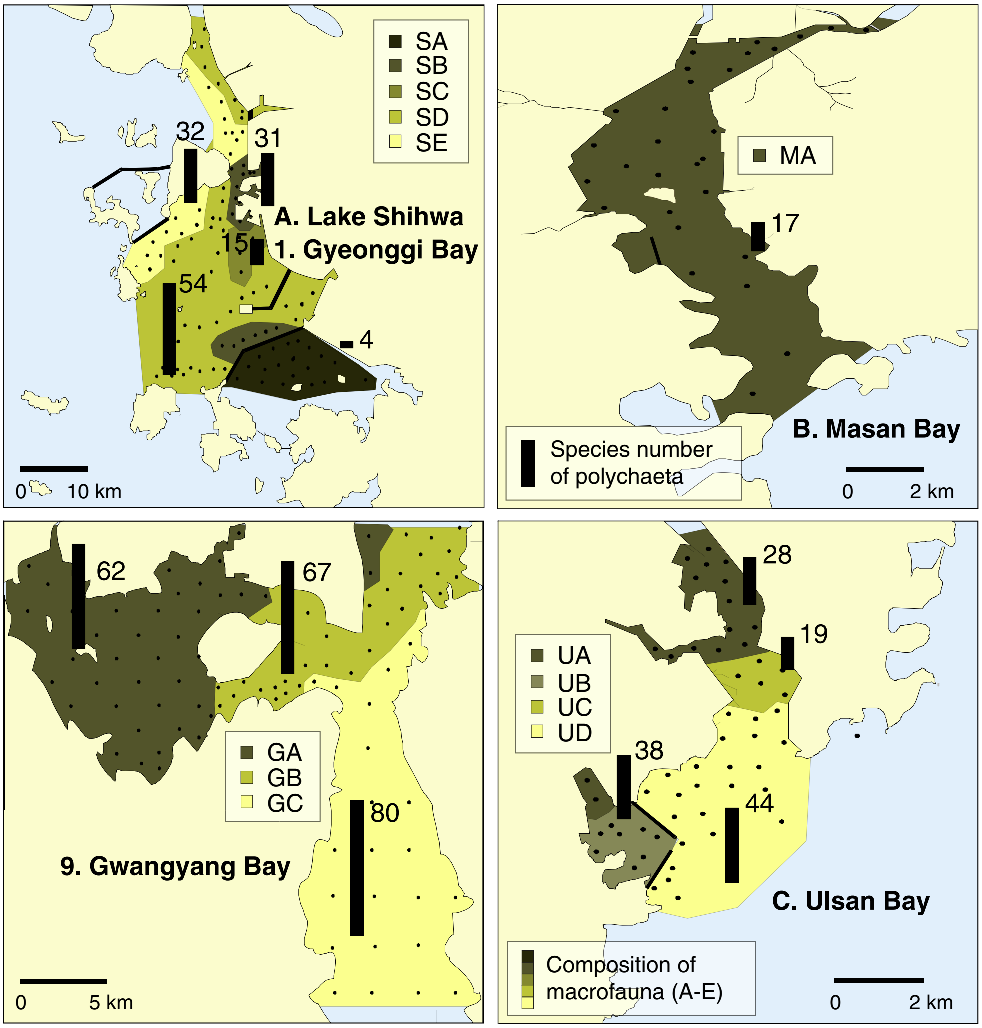

Fig. 6. Benthic community mapping (1–5 groups) by cluster analysis based on composition of macrofauna in the Lake Shihwa (A), Gyeonggi Bay (1), Masan Bay (B), Gwangyang Bay (9), and Ulsan Bay (C) (black bars indicate the total number of polychaete species in given group).

sometimes due to the complex nature of environmental samples together with limited instrumental support, causative agents of observed bioassay responses were not fully completed in sediments of west coasts (Hong et al., 2012a) and Taean oil spill areas (Hong et al., 2012b). Thus, more complementary studies (e.g., iterative bioassay derived fractionations) are needed to be narrowed down to characterize the causative agents and possible interactions between them.

6. Macrobenthic communities

One of the key components for sediment triad assessment would be benthic community responses as targeting infaunal biological responses in the bottom sediment (Chapman and Hollert, 2006). During the series of our studies, the benthic community structure including species composition, biomass and density of macrobenthic organisms has been investigated (Figs. 1 and 6). Here, we present some of these results from the selected regions of Lake Shihwa, Gyeonggi Bay, Masan Bay, Gwangyang Bay, and Ulsan Bay that were surveyed in the late 1990s.

The benthic communities have been identified by group(s) (A to E depending on the regions) based on the cluster analysis with macrofaunal compositions (Fig. 6). Spatial gradient in biological diversity has demonstrated the distinct patterns of benthic communities, coinciding with geographical locations from inner to outer bay, of which gradient was apparently related with the degree of water circulation. However, it should be noted that the varying community similarities found in the different regions also reflected the general spatial distributions of organic contaminants in sediments from Lake Shihwa and Gyeonggi Bay (Kim et al., 1997; Lee et al., 2001a,b), Masan Bay (Hong et al., 2009b; Khim et al., 1999a; Li et al., 2008; Yim et al., 2005), Gwangyang Bay (Koh et al., 2005; Woo et al., 2006), and Ulsan Bay (Khim et al., 2001a; Koh et al., 2001, 2002). These results indicated that benthic community structures have been affected by the amount as well as residence time of organic contaminants, in general.

Further the polychaete populations in the given group(s) also reflected the degree of contamination as the number of species generally decreased from more polluted (viz., inner locations) to lesser polluted ones (say outer locations). Similarly, one study also showed that deterioration of the macrobenthic communities was closely associated with the WWTP discharge, identified as major pollution source in the Masan Bay (Moon et al., 2008b). The PCDD/Fs was found to be the major contaminants influencing the structure of the macrobenthic community in this case.

However, macrobenthic community structures sometimes did not clearly explain concentrations and/or activities of known target chemicals. For example, in one recent study that examined the effect of oil spill in Taean area, it shows that the elevated crude-derived hydrocarbons could successfully explain the great AhR-mediated activities in sediments, but the study lacks evidence towards the recovery of macrobenthic communities two years after the Hebei Spirit oil spill (Hong et al., 2012b).

The difficulties in matching the sediment chemistry and benthic community structure, among the triad components, could be explained by various reasons such as i) insufficient dissolved oxygen in water column (Seitz et al., 2009), ii) high organic carbon contents (Hyland et al., 2005), iii) heavy metal pollutions (Ryu et al., 2011), iv) bioavailability of organic contaminants (Janssen et al., 2011), and v) tolerance and threshold of organic contaminants to organisms (Critto et al., 2005; Wildhaber and Schmitt, 1998). Recently, Chapman and Hollert (2006) suggested additional lines of evidence approach such as biomagnification, benthos colonization, and habitat morphology not limited to three components of sediment chemistry, sediment bioassays, and benthic community analysis, thus further expansion of effect assessment would be of advanced benefit.

7. Conclusions: Current understandings and limitations

Many studies on chemical, toxicological, and ecological measures for sediment assessment have been conducted in the Korean coastal waters since the late 1990s, but not much integrated and/or balanced to date. At the beginning, many studies, mostly chemistry work, focused on coastal to offshore marine sediments, considering a final sink of target trace pollutants, then highlighted the identification and characterization of sources by broadening study gardens to inlands and upstream regions. More recently, studies have been extended to the bigger ecosystem of the Yellow Sea, focusing on multimedia fate and transport between Korea and China. Studies on POPs and EDCs indicated that sediment contaminations were relatively low to moderate but their occurrence and concentrations tell widespread and persistent distribution across the Korean coasts, while some localized zones with extremely high pressures of certain pollutants were also identified and temporal concentrations have shown slight decreases during the decades. Later, toxicological studies targeting exposure assessment of potential dioxin-like and estrogenic chemicals associated with sediments reflected a general good agreement between biological responses and chemical concentrations, in composition and potency wise. Although the benthic ecology data is still limited until now, some available data on macrofaunal community responses given by species occurrence and diversity could reflect the type and degree of sediment contaminations, in general. However, concentrations and activities of known target chemicals could not fully account for the biological and ecological responses at all times. Overall, we found that the triad approach with multiple lines of evidence towards integrated assessment of trace pollutants in coastal environment of Korea would be of increasing demands and concerns to fully address individual components and their association.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2013.07.052.

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< Supplemental Materials>

Assessment of trace pollutants in Korean coastal sediments using the triad approach: A review

Jong Seong Khim*, Seongjin Hong

Supplemental Materials: Tables

*Corresponding Author. *E-mail address:* jskocean@snu.ac.kr (J.S. Khim).

Supplemental Materials: Tables

Table S1. Concentrations of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Target chemicals	Total PCDD/Fs (pg TEQ g^{-1} dw)				References
					Min.	Max.	Mean	Median	
A. Lake Shihwa	In land	2008	12	PCDD/Fs+CoPCBs[a]			240		Moon et al., 2012a
	Lake	2005	1	PCDD/Fs+CoPCBs			93		Terauchi et al., 2009
		2008	16	PCDD/Fs+CoPCBs			13		Moon et al., 2012a
1. Gyeonggi Bay	Bay	2008	6	PCDD/Fs+CoPCBs			0.76		Moon et al., 2012a
4. Asan Bay	Bay	2005	1	PCDD/Fs+CoPCBs			2.9		Terauchi et al., 2009
B. Masan Bay	Bay	1992	11	PCDD/Fs	0.50	64	10	3.0	Im et al., 2002
		1998	1	PCDD/Fs+CoPCBs			12		Oh et al., 2003
		2004	21	PCDD/Fs	2.6	420	32	9.0	Kannan et al., 2007
		2005	20	PCDD/Fs+CoPCBs	3.6	420	36		Moon et al., 2008a
		2005	1	PCDD/Fs+CoPCBs			22		Terauchi et al., 2009
		2006	5	PCDD/Fs	2.4	220	55	17	Hong et al., 2009b
9. Gwangyang Bay	Bay	2000-2002	35	PCDD/Fs+CoPCBs			1.3	0.40	Moon et al., 2008b
		2006	3	PCDD/Fs+CoPCBs	1.0	5.2	3.5	4.3	Kim et al., 2008
		2006	8	PCDD/Fs			5.1		Jeong et al., 2012
		2007	8	PCDD/Fs			6.7		Jeong et al., 2012
		2008	8	PCDD/Fs			5.6		Jeong et al., 2012
		2009	8	PCDD/Fs			5.8		Jeong et al., 2012
		2010	8	PCDD/Fs			8.5		Jeong et al., 2012
10. Busan Bay	Bay	2000-2002	21	PCDD/Fs+CoPCBs			11	12	Moon et al., 2008a
		2002-2003	3	PCDD/Fs+CoPCBs			5.5		Jung et al., 2007
		2005	1	PCDD/Fs+CoPCBs			4.6		Terauchi et al., 2009
11. Jinhae Bay	Bay	2000-2002	26	PCDD/Fs+CoPCBs			7.0	8.6	Moon et al., 2008a
		2002-2003	15	PCDD/Fs+CoPCBs			18		Jung et al., 2007
15. Haengam Bay	Bay	2005	1	PCDD/Fs+CoPCBs			11		Terauchi et al., 2009
16. Nakdong River	Estuary	2002-2003	11	PCDD/Fs+CoPCBs			2.2		Jung et al., 2007
C. Ulsan Bay	Bay	1996	3	PCDD/Fs+CoPCBs	9.0	120	47	11	Oh et al., 2003
		2000-2002	20	PCDD/Fs+CoPCBs			8.4	8.0	Moon et al., 2008a
		2002-2003	9	PCDD/Fs+CoPCBs			2.5		Jung et al., 2007
		2005	1	PCDD/Fs+CoPCBs			12		Terauchi et al., 2009
17. Youngil Bay	In land	2001	6	PCDD/Fs+CoPCBs	0.43	1000	220	9.4	Koh et al., 2004
	Bay	1996	1	PCDD/Fs+CoPCBs			47		Oh et al., 2003
		2000-2002	20	PCDD/Fs+CoPCBs			7.8	3.8	Moon et al., 2008a
		2005	1	PCDD/Fs+CoPCBs			4.6		Terauchi et al., 2009
18. Onsan Bay	Bay	2005	1	PCDD/Fs+CoPCBs			14		Terauchi et al., 2009
19. Uljin	Coast	2005	1	PCDD/Fs+CoPCBs			2.3		Terauchi et al., 2009
20. Korean coasts	Coast	2001-2007	25	PCDD/Fs	0.06	12	2.3	1.5	Choi et al., 2011a
		2008	8	PCDD/Fs	0.10	0.40	0.30	0.30	Naile et al., 2011

[a] Co-PCBs: coplanar polychlorinated biphenyls.

Table S2. Concentrations of polychlorinated biphenyls (PCBs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Congener number (n)	Total PCBs (ng g^{-1} dw)				References
					Min.	Max.	Mean	Median	
A. Lake Shihwa	Inland	2000	8	98	9.0	120	30	13	Koh et al., 2005
	Lake/River	1996	4	101	2.7	28			Lee et al., 2001a
		1998	11	98	< 0.99	12	3.9	4.9	Khim et al., 1999c
1. Gyeonggi Bay	Bay	1995	54	101	< 0.99	580			Lee et al., 2001a
		2000	13	22	0.60	16	3.4	1.1	Hong et al., 2005
		2003	33	116	0.20	42	2.2	0.80	Hong et al., 2009a
3. Namyang Bay	Bay	1996	5	101	< 0.99	2.5			Lee et al., 2001a
4. Asan Bay	Bay	2005	6	22	1.9	3.3	2.3	2.1	Choi et al., 2011b
B. Masan Bay	Inland	2000	8	98	8.5	92	39	35	Koh et al., 2005
	Bay	1997	20	22	1.2	41	15	14	Hong et al., 2003
		1998	28	98	6.9	150	36	21	Khim et al., 1999a
9. Gwangyang Bay	Bay	2000	6	22	0.20	2.9	1.4	1.4	Hong et al., 2005
		2001	11	98	< 0.99	4.5	0.41		Koh et al., 2005
		2001	23	89	2.3	11	6.2		Hong et al., 2004
10. Busan Bay	Bay	2000	10	22	5.7	200	58	25	Hong et al., 2005
		2000	5	20	3.9	56	31	31	Choi et al., 2001a
11. Jinhae Bay	Bay	2000	5	20	< 0.10	8.8	2.3	0.52	Choi et al., 2001a
		2007	16	22	0.64	18	3.5	1.6	Shim et al., 2010
16. Nakdong River	Lake/River	1999	18	43	1.1	140	19	3.8	Jeong et al., 2001
C. Ulsan Bay	Inland	1999	6	98	1.4	22	11	12	Khim et al., 2001a
	Inland	1999	5	98	8.7	52	20	12	Khim et al., 2001a
	Bay	1999	16	98	< 1	77	18	15	Khim et al., 2001a
		2000	11	22	2.1	22	8.5	6.2	Hong et al., 2005
		2000	6	20	1.2	140	31	6.4	Choi et al., 2001a
17. Yeongil Bay	In land	2001	6	98	< 1	170	62	17	Koh et al., 2004
	Bay	2000	9	22	0.60	73	15	7.8	Hong et al., 2005
		2000	26	98	< 0.99	32	3.1		Koh et al., 2006a
		2000	5	20	0.11	11	4.9	2.6	Choi et al., 2001a
18. Onsan Bay	Inland	1999	6	98	< 1.0	56	16	20	Koh et al., 2002
	Lake/River	1999	3	98	5.5	10	7.5	6.9	Koh et al., 2002
	Bay	1999	13	98	< 1.0	7.8			Koh et al., 2002
20. Korean coasts	Bay & Coast	1997-2002	138	18	0.10	200	1.4	1.6	Hong et al., 2006b
		2001-2007	25	23	< 0.10	15	2.3	0.61	Choi et al., 2011a

Table S3. Concentrations of organochlorine pesticides (OCPs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Target chemicals	Total OCPs (ng g^{-1} dw)				References
					Min.	Max.	Mean	Median	
A. Lake Shihwa	Inland	2000	8	CHLs[a]+DDTs[b]+HCHs[c]+HCB[d]	0.90	17	4.5	3.0	Koh et al., 2005
	Lake/River	1996	3	CHLs+DDTs+HCHs	0.26	7.2	2.7		Lee et al., 2001b
		1998	11	CHLs+DDTs+HCHs+HCB	1.0	5.5	2.8	2.3	Khim et al., 1999c
1. Gyeonggi Bay	Bay	1996	47	CHLs+DDTs+HCHs	0.26	7.2	2.7		Lee et al., 2001b
		2003	33	DDTs+HCHs	0.10	5.3	1.0	0.6	Hong et al., 2009a
2. Incheon harbor	Bay	1996	7	CHLs+DDTs+HCHs	2.6	160	39		Lee et al., 2001b
3. Namyang Bay	Bay	1996	5	CHLs+DDTs+HCHs	0.15	1.5	1.0		Lee et al., 2001b
4. Asan Bay	Bay	2005	6	DDTs+HCHs	1.2	2.7	1.7	1.5	Choi et al., 2011b
7. Saemanguem	Coast	2002	13	CHLs+DDTs+HCHs+HCB	0.13	2.1	0.60	0.3	Hong et al., 2006a
B. Masan Bay	Inland	2000	8	CHLs+DDTs+HCHs+HCB	0.40	47	13	8.1	Koh et al., 2005
	Bay	1997	20	CHLs+DDTs+HCHs+HCB+etc.[e]	0.81	98	18	4.8	Hong et al., 2003
		1998	28	CHLs+DDTs+HCHs+HCB	1.1	15	4.5	2.9	Khim et al., 1999a
9. Gwangyang Bay	Bay	2001	11	CHLs+DDTs+HCHs+HCB	0.10	1.0	0.40	0.4	Koh et al., 2005
		2001	23	CHLs+DDTs+HCHs+HCB+etc.	0.20	3.1			Hong et al., 2004
11. Jinhae Bay	Bay	2007	16	CHLs+DDTs+HCHs	0.28	7.9	1.3	0.65	Shim et al., 2010
C. Ulsan Bay	Inland	1999	6	CHLs+DDTs+HCHs+HCB	0.10	9.0	2.9	1.6	Khim et al., 2001a
		1999	5	CHLs+DDTs+HCHs+HCB	0.10	2.7	1.1	0.90	Khim et al., 2001a
	Lake/River	1999	4	CHLs+DDTs+HCHs+HCB	0.10	0.10	0.10	0.10	Khim et al., 2001a
	Bay	1999	16	CHLs+DDTs+HCHs+HCB	0.30	43	6.9	2.7	Khim et al., 2001a
17. Yeongil Bay	Bay	2000	26	CHLs+DDTs+HCHs+HCB	0.10	14	1.7	0.90	Koh et al., 2006a
18. Onsan Bay	Inland	1999	6	CHLs+DDTs+HCHs+HCB	1.7	10	6.2	5.9	Koh et al., 2002
	Lake/River	1999	3	CHLs+DDTs+HCHs+HCB	0.12	4.3	2.1	1.7	Koh et al., 2002
	Bay	1999	13	CHLs+DDTs+HCHs+HCB	0.10	3.7	0.92	0.61	Koh et al., 2002
20. Korean coasts	Bay & Coast	1997-2002	138	CHLs+DDTs+HCHs+HCB+etc.	0.10	160	8.5	1.3	Hong et al., 2006b

[a] CHLs: chlordanes.
[b] DDTs: bis(*p*-chlorophenyl)ethane.
[c] HCHs: hexachlorocyclohexanes.
[d] HCB: hexachlorobenzene.
[a] etc.: Aldrine, dieldrine, endrine, endosulfan II, and mirex.

Table S4. Concentrations of polycyclic aromatic hydrocarbons (PAHs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Number of chemicals (n)	Total PAHs (ng g^{-1} dw)				References
					Min.	Max.	Mean	Median	
A. Lake Shihwa	In land	2001	8	16	13	640	230	100	Koh et al., 2005
	Lake/River	1998	11	16	< 10	31	23	25	Khim et al., 1999c
1. Gyeonggi Bay	Bay	1995	67	23	9.0	1400	120	65	Kim et al., 1997
		2003	33	24	6.0	650	110	58	Hong et al., 2009a
5. Taean oil spill area	Coastal	2009	50	16	< 1	650	54	17	Hong et al., 2012b
6. Gunsan coast	Coastal	2008	10	16	68	430	200	190	Lee et al., 2012
B. Masan Bay	In land	2000	8	16	33	480	220	160	Koh et al., 2005
	Bay	1997	20	16	210	2700	680		Yim et al., 2005
		1998	28	16	42	1100	350	310	Khim et al., 1999a
		2005	20	16	120	1700	930		Moon et al., 2008b
9. Gwangyang Bay	Bay	2001	11	16	10	110	36	25	Koh et al., 2005
		2003	13	16	51	8400	1000	250	Woo et al., 2006
11. Jinhae Bay	Bay	2007	16	24	54	630	160	120	Shim et al., 2010
C. Ulsan Bay	Inland	1999	6	16	< 10	1400	620	520	Khim et al., 2001a
		1999	5	16	54	600	230	180	Khim et al., 2001a
	Bay	1997	30	23	50	6100	540		Koh et al., 2001
		1999	16	16	17	3100	410	82	Khim et al., 2001a
		2000	22	16	14	7100	1100	460	Moon et al., 2001
17. Yeongil Bay	In land	2001	6	16	5.3	7700	1800	240	Koh et al., 2004
	Bay	2000	26	16	< 10	1900	310	56	Koh et al., 2006a
18. Onsan Bay	Inland	1999	6	16	< 10	210	96	74	Koh et al., 2002
	Lake/River	1999	3	16	< 10	570	330	330	Koh et al., 2002
	Bay	1999	13	16	< 10	42	27	27	Koh et al., 2002
20. Korean Coasts	Bay & Coast	2000-2001	117	16	8.8	18000	820	150	Yim et al., 2007
		2001-2007	25	16	16	1100	290	220	Choi et al., 2011a

Table S5. Concentrations of nonylphenols (NPs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Target chemicals	Total NPs (ng g^{-1} dw)				References
					Min.	Max.	Mean	Median	
A. Lake Shihwa	In land	2000	8	NP[a]	250	4900	1600	810	Koh et al., 2005
		2000	18	NP	< 0.4	32000	3600	900	Li et al., 2004a
	Lake	1998	11	NP	20	1800	620	410	Khim et al., 1999c
		2000	12	NP	11	620	240	160	Li et al., 2004a
		2002	10	NP	10	5100	830	210	Li et al., 2004c
		2002	10	NP	16	2500	770	210	Li et al., 2004c
		2006-2007	5	NP+NP1EO[b]+NP2EO[c]	100	700	350	280	Choi et al., 2009c
		2008	7	NP+NP1EO+NP2EO	24	350	160	150	Hong et al., 2010
		2008	7	NP+NP1EO+NP2EO	< 2	1000	210	76	Hong et al., 2010
1. Gyeonggi Bay	Bay	2003	33	NP	3.0	1100	50	10	Hong et al., 2009a
		2008	7	NP+NP1EO+NP2EO	14	97	47	38	Hong et al., 2010
		2008	7	NP+NP1EO+NP2EO	6.5	37	16	16	Hong et al., 2010
6. Gunsan coast	Coast	2008	10	NP	21	2200	610	190	Lee et al., 2012
8. Mokpo coast	Coast	2006	29	NP+NP1EO+NP2EO	4	2100	200		Choi et al., 2007
B. Masan Bay	In land	2000	8	NP	85	1100	390	320	Koh et al., 2005
	Bay	1998	28	NP	110	3900	510	330	Khim et al., 1999a
		2004	18	NP+NP1EO+NP2EO	130	2800	580	420	Li et al., 2008
		2005	20	NP	40	1200	410		Moon et al., 2008b
		2006	5	NP	24	500	250	210	Hong et al., 2009b
		2006-2007	5	NP+NP1EO+NP2EO	49	120	71	63	Choi et al., 2009c
9. Gwangyang Bay	Bay	1999-2000	6	NP	12	100	41		Cho et al., 2004
		2001	11	NP	9.9	35	18	13	Koh et al., 2005
		2001	15	NP	3.1	74	21	18	Li et al., 2004b
10. Busan Bay	Bay	2006-2007	5	NP+NP1EO+NP2EO	98	700	420	460	Choi et al., 2009c
11. Jinhae Bay	Bay	2007	16	NP	110	210	150	150	Shim et al., 2010
12. Yeosu Bay	Bay	1999-2000	9	NP	7.0	200	39		Cho et al., 2004
C. Ulsan Bay	Inland	1999	6	NP	< 1	1000	280	150	Khim et al., 2001a
		1999	5	NP	1.5	670	140	7.3	Khim et al., 2001a
	River	1999	4	NP	< 1	12	6.8	6.9	Khim et al., 2001a
	Bay	1999	16	NP	< 1	20	3.4	2.0	Khim et al., 2001a
		2006-2007	5	NP+NP1EO+NP2EO	7.5	86	42	41	Choi et al., 2009c
17. Yeongil Bay	Bay	2000	26	NP	< 1	1400	86	22	Koh et al., 2006a
18. Onsan Bay	Inland	1999	6	NP	< 1	860	220	96	Koh et al., 2002
	Lake/River	1999	3	NP	< 1	21	13	12	Koh et al., 2002
	Bay	1999	13	NP	< 1	3.8	2.4	2.3	Koh et al., 2002
20. Korean Coasts	West coast	2006-2007	6	NP+NP1EO+NP2EO	3.0	63	24	9.6	Choi et al., 2009c
	South coast	2006-2007	5	NP+NP1EO+NP2EO	7.9	190	44	8.5	Choi et al., 2009c
	East coast	2006-2007	4	NP+NP1EO+NP2EO	2.3	140	37	3.9	Choi et al., 2009c

[a] NP: nonylphenols.
[b] NP1EO: nonylphenol monoethoxylate.
[c] NP2EO: nonylphenol diethoxylate.

Table S6. Concentrations of butyltins (BTs) in Korean coastal waters (for review).

Regions	Type	Sampling year (y)	Site number (n)	Target chemicals	Total BTs (ng g^{-1} dw)[d]				References
					Min.	Max.	Mean	Median	
1. Gyeonggi Bay	Bay	1995	40	MBT[a]+DBT[b]+TBT[c]	0.80	300	38	10	Kim et al., 1998
		2003	33	MBT+DBT+TBT	2.0	140	19	7.0	Hong et al., 2009a
6. Gunsan coast	Coast	2008	10	MBT+DBT+TBT	6.8	34	16	12	Lee et al., 2012
B. Masan Bay	Bay	1996	10	MBT+DBT+TBT	43	440	170	150	Choi et al., 1997
		2004	21	MBT+DBT+TBT	27	800	300		Kim et al., 2011a
9. Gwangyang Bay	Bay	1996	12	TBT	< 5.0	21	10		Kim and Park, 2001
		2001	35	MBT+DBT+TBT	< 4.9	320	100		Choi et al., 2002
		2001	17	TBT	< 5.0	81	24		Shim et al., 2004
10. Busan Bay	Bay	2000	5	MBT+DBT+TBT	210	3700	1500	1600	Choi et al., 2003
		2002-2004	40	DBT+TBT	38	15000	1400		Choi et al., 2009b
		2007	27	TBT	9.8	4700			Choi et al., 2010
11. Jinhae Bay	Bay	1995	55	MBT+DBT+TBT	66	4300	630		Shim et al., 1999
		2000	5	MBT+DBT+TBT	93	730	420	370	Choi et al., 2003
		2002-2004	35	DBT+TBT	< 7.0	170	220		Choi et al., 2009b
		2007	16	MBT+DBT+TBT	29	3000	540	280	Shim et al., 2010
13. Gohyeon Bay	Bay	2003	18	MBT+DBT+TBT	95	6600	660	260	Kim et al., 2011b
14. Okpo Bay	Bay	1995	20	MBT+DBT+TBT	34	7700			Hong et al., 2001
C. Ulsan Bay	Bay	2000	5	MBT+DBT+TBT	150	4300	1900	1300	Choi et al., 2003
		2002-2004	39	DBT+TBT	< 7.0	19000	1900		Choi et al., 2009b
17. Yeongil Bay	Bay	2000	5	MBT+DBT+TBT	24	1300	360	95	Choi et al., 2003
20. Korean Coasts	Bay & Coast	1997	20	MBT+DBT+TBT	< 5.0	80	29		Shim et al., 2005
		2001	20	TBT	< 7.0	1700	240		Choi et al., 2009a
		2002	20	TBT	< 7.0	23000	1300		Choi et al., 2009a
		2003	25	TBT	< 7.0	850	170		Choi et al., 2009a
		2004	25	TBT	< 7.0	1500	250		Choi et al., 2009a
		2005	25	TBT	< 7.0	1900	170		Choi et al., 2009a

[a] MBT: mono-*n*-butyltin.
[b] DBT: di-*n*-butyltin.
[c] TBT: tri-*n*- butyltin.
[d] Note: To convert from ng Sn g^{-1} to ng g^{-1}, multiply by 2.44.