

# SPATIAL AND TEMPORAL TRENDS OF PERSISTENT ORGANIC POLLUTANTS (POPs) AND RELATED COMPOUNDS IN SEDIMENT AND BIVALVES FROM KOREAN COASTAL WATERS DURING 2001-2012

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## Introduction

Persistent organic pollutants (POPs) such as polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), dichlorodiphenyltrichloroethane (DDTs) and related compounds such as polycyclic aromatic hydrocarbons (PAHs) and tributyltin (TBT) are ubiquitous toxic contaminants in the environment. These compounds have received considerable attention due to their characteristics such as persistence, toxicity and bioaccumulation potentials. POPs are released from various sources such as incinerators, chemical industries, municipal wastewater effluents and agricultural activities<sup>1</sup>. PAHs are generated by combustion of fossil fuels including automobile exhaust, residential heating and oil production. TBT has been extensively used for industrial and agricultural purposes such as antifouling paint on ships, harbor structures, fungicides and insecticides<sup>2</sup>. Because of adverse health effects of these contaminants, it is necessary to monitor toxic organic pollutants including POPs in marine environment.

Korean peninsula is surrounded by the sea on the western, southern and eastern sides. Urbanization and industrialization have developed along the coastline since the 1960s. In particular, there are large industrial complexes and harbors along the coastline in the South Sea and East Sea. Thus, large amounts of various anthropogenic pollutants are released into marine environment of Korea. Korean government has designated the coastal areas close to the industrial complexes as the 'Specialized Management Coastal Zone'<sup>3,4</sup>. Monitoring of these contaminants in sediment and bivalve are useful to identify the contamination sources and to establish strategies for managing these contaminants. However, limited information is available on the contamination status of POPs and related compounds in Korean coastal waters. The objectives of this study were to investigate the contamination status of POPs and related compounds and to evaluate spatial and temporal trends of contaminants during 2001-2012 in Korean coastal waters.

## Materials and methods

Surface sediments and bivalves were collected at 25 locations along the coastlines during 2001–2012 (5 locations for East Sea, 12 locations for South Sea, and 8 locations for West Sea). The sediment and bivalve samples collected were transported to the laboratory and stored in a freezer at –20°C until extraction. The shells of bivalves were removed and freeze-dried for the next steps. Experimental procedures and instrumental conditions of POPs and related compounds in the samples were described elsewhere<sup>5-8</sup>.

## Results and discussion

### *Contamination status of POPs and related compounds*

The concentrations of PCDD/Fs, PCBs, DDTs, PAHs and TBT in sediments and bivalves from Korean coastal regions in 2012 are summarized in Table 1. POPs and related compounds were detected in most of the sediments and bivalves samples, suggesting widespread contamination of these contaminants in Korean coastal water. The total concentrations of PCDD/Fs, PCBs, DDTs, PAHs and TBT in sediments ranged from nd (not detected) to 0.89 (mean: 0.24) pgTEQ/g dry weight (dw), from 0.13 to 23.0 (1.86) ng/g dw, from nd to 13.8 (1.47) ng/g dw, from 11.0 to 1413 (204) ng/g dw and from 0.32 to 109 (16.6) ngSn/g dw, respectively. The total concentrations of PCDD/Fs, PCBs, DDTs, PAHs and TBT in bivalves ranged from nd to 0.26 (mean: 0.03) pgTEQ/g wet weight (ww), from nd to 71.5 (11.5) ng/g ww, from nd to 52.6 (8.43) ng/g ww, from 43.2 to 301 (108) ng/g dw and from 3.66 to 611 (90.1) ngSn/g dw, respectively. Among target contaminants measured in our study, PAHs showed the higher levels in sediments and bivalves. The total concentrations of PCBs, DDTs and TBT in bivalves were 2–6 times higher than those measured in sediments, implying bioaccumulation potentials for these contaminants in

coastal waters of Korea. However, the total concentrations of PAHs in bivalves were lower than those measured in sediments, because lower bioaccumulation potentials in the marine food web.

Table 1. Concentrations of PCDD/Fs, PCBs, DDTs, PAHs and TBT in sediment and bivalves from Korean coastal regions measured in 2012

	Sediment			Bivalve		
	mean $\pm$ SD	min	max	mean $\pm$ SD	min	max
PCDD/Fs(pgTEQ/g dw)	0.24 $\pm$ 0.25	nd <sup>a</sup>	0.89	0.03 $\pm$ 0.06 <sup>b</sup>	nd	0.26 <sup>b</sup>
PCBs(ng/g dw)	1.86 $\pm$ 4.53	0.13	23.0	11.5 $\pm$ 14.8	nd	71.5
DDTs(ng/g dw)	1.47 $\pm$ 2.94	nd	13.8	8.43 $\pm$ 13.5	nd	52.6
PAHs(ng/g dw)	204 $\pm$ 292	11.0	1413	108 $\pm$ 69.0	43.2	301
TBT(ngSn/g dw)	16.6 $\pm$ 26.6	0.32	109	90.1 $\pm$ 145	3.66	611

<sup>a</sup>not detected; <sup>b</sup>wet weight basis.

#### *Spatial distribution of POPs and related compounds measured in 2012*

The spatial distribution of POPs and related compounds in sediment and bivalves from Korean coastal waters is presented in Figure 1. The overall spatial distribution of POPs and related compounds was similar between sediment and bivalve samples. The highest concentrations of PCDD/Fs, PCBs, DDTs, PAHs and TBT were found in sediment and bivalves from southeastern coastal regions of Korea, which are located close to industrial complexes and larger harbor such as Ulsan, Busan, Jinhae and Masan Bays. Our results suggest industrial complexes and shipping activities are major contamination sources of POPs, PAHs and TBT in Korean coastal waters.

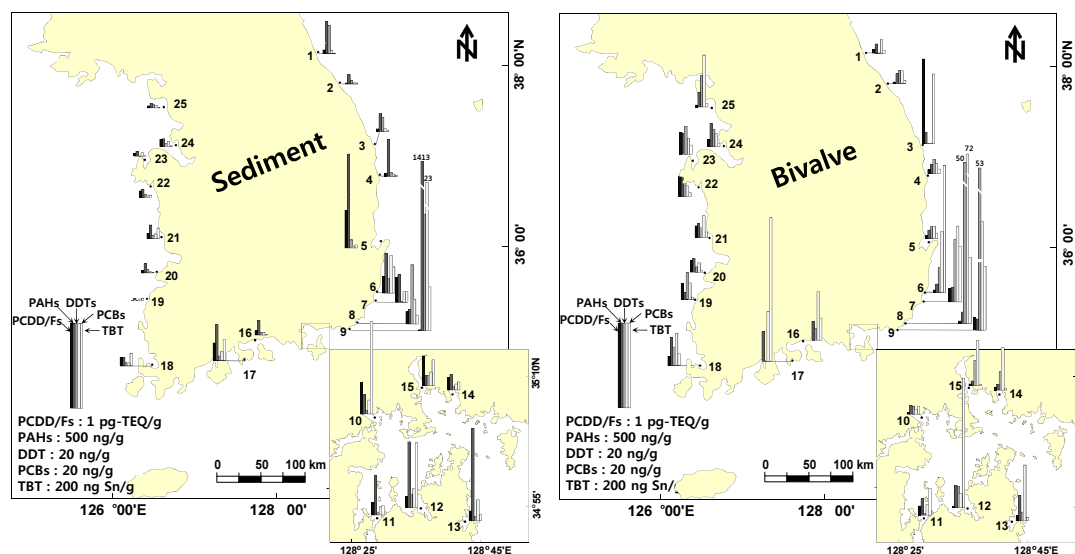


Figure 1. Spatial distribution of POPs and related compounds in sediment and bivalves collected from Korean coastal regions in 2012.

#### *Temporal trends of POPs and related compounds during 2001–2012*

The temporal trends of POPs and related compounds in sediment and bivalves from Korean coastal region are shown in Figure 2. Significant decreasing trend in the concentrations of PCDD/Fs in sediment and TBT in bivalves, due to strong regulation on contamination sources of these contaminants. In 1999, the emission of flue gas from municipal solid waste incinerators (MSWIs), regarded as a major source of PCDD/Fs, has been regulated by the Ministry of Environment. In 2003, the use of TBT-based antifouling paints was banned in Korea. Our results indicate that the strong regulation on chemicals is effective to reduce contamination by toxic

chemicals in marine environment. The concentrations of PCBs, DDTs and PAHs did not show any temporal trends in Korean coastal waters. Although PCBs and DDTs were banned since the 1980s, chemicals were detected in coastal waters due to longer half-lives in the environment. The major sources of PAHs in marine environment are combustion processes and oil spillage, implying on-going source of PAHs in Korean coastal waters.

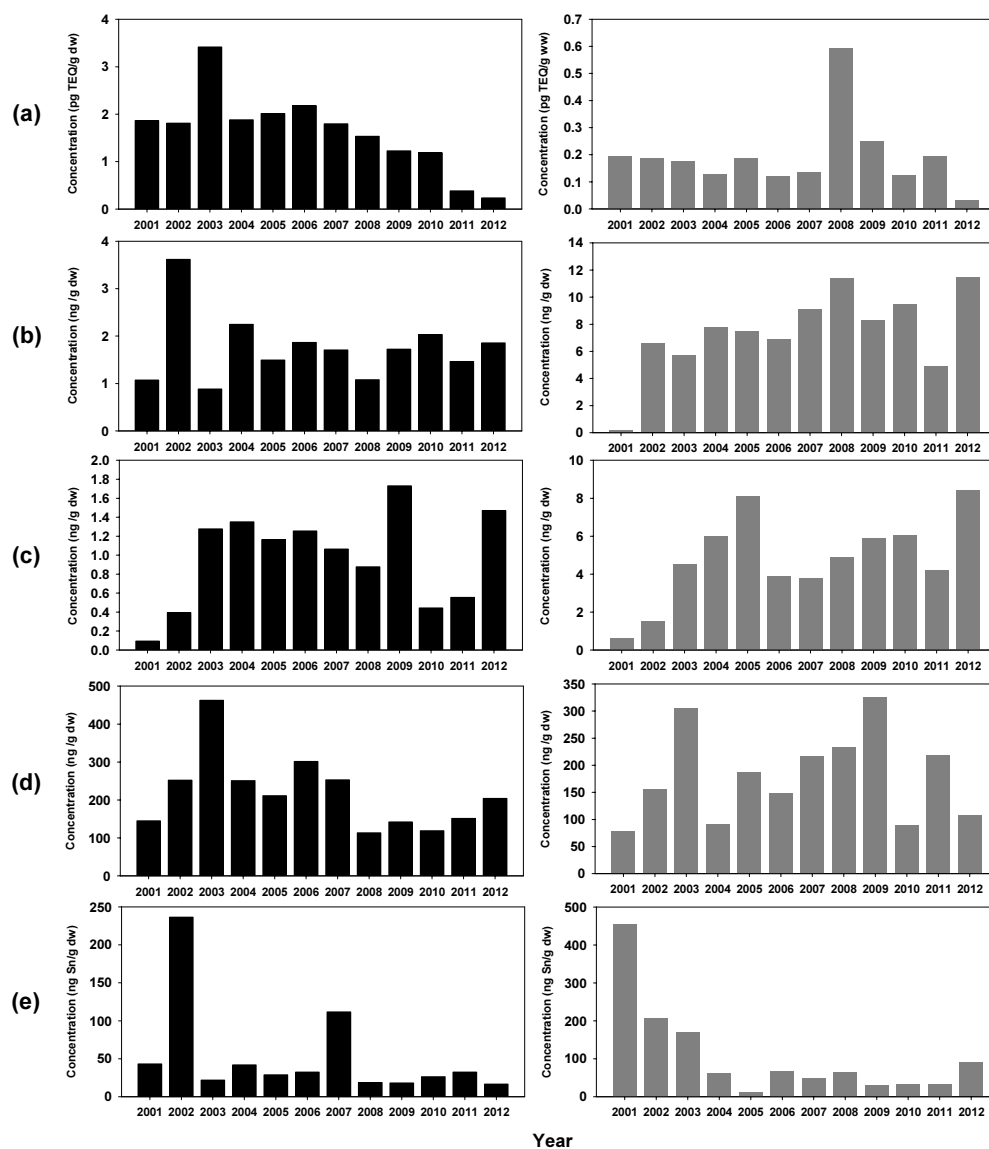


Figure 2. Temporal trends of average concentrations for (a) PCDD/Fs, (b) PCBs, (c) DDTs, (d) PAHs, and (e) TBT in sediment and bivalves collected from Korean coastal waters during 2001–2012. Black bars (left side) represent sediment levels and gray bars (right side) represent bivalve levels.

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