# MONITORING OF ORGANIC CONTAMINANTS IN VARIOUS CULTURING GROUNDS FROM THE KOREAN COAST: SPATIAL DISTRIBUTION AND TEMPORAL TRENDS (2010-2012)

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## Introduction

Persistent organic pollutants (POPs) including polychlorinated dibenzo-*p*-dioxins and furans (PCDD/Fs), dioxin-like polychlorinated biphenyls (DLPCBs), and polybrominateddiphenyl ethers (PBDEs) have been regulated by the Stockholm Convention to protect human health and the environment[1,2]. In Korea, the routine monitoring network of POPs in marine environments have been conducted, which investigated the POPs levels in sediments and organisms from locations near urban area, big harbor, and industrial complex[3,4]. However, POPs in sediment and culturing organisms in various culturing grounds along the Korean coasts have not been points of interests even though highly dense aquaculture activities have been conducted from inshore areas. Therefore, the objective of this study was to investigate the concentrations and distribution patterns of PCDD/Fs, DLPCBs and PBDEs in sediment and organisms from various culturing grounds in the Korean coasts as well as temporal trends of POPs from 2010 to 2012.

## Materials and methods

The sediment and organism samples from culturing grounds were collected at 13 locations in 2010 and 2011, 15 locations in 2012 throughout the coastal areas of Korea. The samples were stored in a cooler box with ice, and immediately transported to the laboratory. Samples were kept in a freezer at -20℃ until extraction. The analytical procedure for PCDD/Fs, DLPCBs is described elsewhere (Moon et al., 2008)[5], and PBDEs were analyzed following the method described by Moon et al. (2007)[6].

## Results and discussion

*Concentration of PCDD/Fs, DLPCBs and PBDEs*

Concentrations of PCDD/Fs, DLPCBs and PBDEs in sediment and organisms from culturing grounds along the Korean coasts are shown in Fig. 1. The concentrations of organic contaminants in sediments ranged from 0.17 to 1.98 pg TEQ/g dw for PCDD/Fs, from 0.01 to 0.71 pg TEQ/g dw for DLPCBs, and from 0.28 to 10.9 ng/g dw for PBDEs, respectively. The concentrations of PCDD/Fs were below the probable effect level (21.5 pg $WHO_{1998}$-TEQ/g dw) of the Canadian Sediment Quality Guidelines[7] but higher than the threshold effect level (0.85 pg $WHO_{1998}$-TEQ/g dw) at 5 - 7 locations (38 - 54%) from 2010 to 2012. S8 and S9 showed relatively higher concentrations of PCDD/Fs, DLPCBs and PBDEs than other locations. These locations are large commercial harbor and located near big cities and effluent discharging point of wastewater treatment plant. Therefore, the main sources of POPs in sediments and culturing organisms might be industrial activities and intensive shipping traffic, which could also raise the amounts of suspended particulate matters containing POPs in coastal waters. The concentrations of organic contaminants in culturing organisms ranged from 0.00 to 0.15 pg TEQ/g ww for PCDD/Fs, from 0.00 to 0.13 pg TEQ/g ww for DLPCBs, and from 0.03 to 0.35 ng/g ww for PBDEs. The concentrations of dioxins (PCDD/Fs+DLPCBs) in culturing organisms were below the restricted level (6.5 pg $WHO_{2005}$-TEQ/g ww) of the European Union[8]. Overall concentration ranges in sediments and culturing organisms, especially bivalves of this study were lower than those of other countries[9-13].

*Distribution patterns of PCDD/Fs, DLPCBs and PBDEs*

The distribution patterns of PCDD/Fs and DLPCBs in coastal sediments and culturing organisms are shown in Fig. 2. O8CDD was a predominant congener in sediment (77 ± 7 %) and culturing organisms (59 ± 15 %), indicating the effect of chemical pentachlorophenol (79 ± 16 %)[14]. The distribution patterns were dominated by PCB118 (sediment, 47 ± 13 %; culturing organisms, 54 ± 7 %) and -105 (sediment, 17 ± 5 %; culturing organisms, 19 ± 4 %) and similar with commercial PCBs products such as aroclor, kanechlor, clophen and

sovol[15]. These results were in accordance with Yao et al. (2008)[16] and Okumura et al. (2004)[17]. BDE 209 was the predominant congener in both sediment and culturing organism samples. In sediments, BDE 209 accounted for over 70 % of the total PBDE concentrations, suggesting high use amount of commercial deca-BDE product in surrounding areas. BDE 209 accounted for over 27 % of the total PBDE concentration in bivalves. In comparison to sediments, the less highly brominated congeners of PBDE such as BDEs 47 (13 ± 10 %), 99 (5 ± 5 %), and 100 (3 ± 3 %) showed greater contribution to total PBDE concentrations in bivalves. However, BDE 209 also had high contributions despite of non-bioavailable characteristic to aquatic organisms, due to their large molecular size[18]. Moon et al. (2007)[6] suggested the effect of filter-feeding activity taking PBDEs absorbed on small particles of culturing organisms and the re-consideration on bioavailability of BDE 209.

*Temporal trends of PCDD/Fs, DLPCBs and PBDEs in sediments and culturing organisms from 2010 to 2012*
Temporal trends of PCDD/Fs, DLPCBs and PBDEs in sediment and culturing organisms are shown in Fig. 3. In sediment samples, concentration variation of DLPCBs showed slightly increasing trend between 2010 and 2012 ($p< 0.05$), but PCDD/Fs, PBDEs did not show any significant trends. In culturing organism samples, concentration variation of DLPCBs did not show any significant trend but PCDD/Fs, PBDEs showed decreasing trends between 2010 and 2012 ($p< 0.05$). Munschy et al. (2008)[1] reported that the concentration decrease of banned or restricted persistent organohalogen contaminants in environmental media has been observed on a global scale, and decreasing trend is related to the severe restriction or phasing out of these compounds and more efficient control of emissions.

**Acknowledgements**
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Table 1.Characteristics of the collected sediment and culturing organisms in this study.

| **Sediment** | | | | **Culturing Organisms** | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Sampling location | | Sample name | Moisture (%) | Species | Length (cm) | Height (cm) | weight (g) | Moisture (%) | Lipid (g/100g) |
| West Sea | Garorim Bay | W1 | 28±6.6 | a | 3.6±0.2[a] | 2.5±0.2[a] | 9.4±1.7[a] | 84±1.3[a] | 1.2±0.2[a] |
| | Chunsu Bay | W2 | 38 ± 13 | a | 3.9±0.4[a] | 2.3±0.5[a] | 10±3.4[a] | 81±0.9[a] | 1.3±0.1[a] |
| | Gomso Bay | W3 | 30±7.2 | a | 3.8±0.2[a] | 2.6±0.2[a] | 11±2.6[a] | 80±3.3[a] | 1.4±0.3[a] |
| South Sea | Wando | S1 | 41±5.4 | h | 214[h] | - | - | 85[h] | 0.4[h] |
| | Deukryang Bay | S2 | 47±6.0 | b | 3.0[b] | 2.3[b] | 7.3[b] | 79[b] | 2.7[b] |
| | Yeoja Bay | S3 | 50±4.5 | b | 3.3[b] | 2.5[b] | 9.2[b] | 79[b] | 1.7[b] |
| | Gamak Bay | S4 | 53±8.6 | c, d | 5.7±1.8[c] | 6.3±2.2[c] | 37±5.4[c] | 75±2.5[c] | 1.5±0.5[c] |
| | | | | | 6.0[d] | 3.0[d] | 9.2[d] | 80[d] | 2.5[d] |
| | Jinju Bay | S5 | 52±6.4 | | 4.6±0.4[c] | 7.7±1.9[c] | 35±13[c] | 75±3.8[c] | 2.1±2.7[c] |
| | Jaran Bay | S6 | 62±6.1 | c, f | 4.±0.4[c] | 8.8±0.9[c] | 46±8.0[c] | 84±2.1[c] | 2.0±0.6[c] |
| | | | | | 6.4±1.3[f] | 4.1±1.0[f] | 41±22[f] | 87±2.2[f] | 1.3±0.6[f] |
| | Hansan Bay | S7 | 54±13.0 | c, f | 4.8±0.2[c] | 9.2±1.1[c] | 54±9.7[c] | 84±2.1[c] | 1.7±0.4[c] |
| | | | | | 7.4±0.1[f] | 4.6±0.2[f] | 48±19.5[f] | 83±8.4[f] | 0.7±0.2[f] |
| | Jinhae Bay | S8 | 60±5.8 | c, d, g | 5.0±0.7[c] | 9.7±1.9[c] | 57±33[c] | 82±3.5[c] | 1.7±0.6[c] |
| | | | | | 5.8±1.0[d] | 3.0±0.5[d] | 15±6.6[d] | 83±2.2[d] | 1.7±0.3[d] |
| | | | | | 8.2[g] | - | 11[g] | 85[g] | 1.4[g] |
| | Gijang coast | S9 | 53±1.3 | h, i | 82±33[h] | - | - | 87±4.7[h] | 0.1±0.1[h] |
| | | | | | 175±6.8[i] | - | - | 85±9.4[i] | 0.2±0.1[i] |
| East Sea | Guryongpo coast | E1 | 59±1.1 | f | 5.6±1.4[f] | 3.2±0.7[f] | 19±9.1[f] | 80±1.7[f] | 2.2±0.6[f] |
| | Yeongdeok coast | E2 | 52±2.3 | f | 6.4±0.3[f] | 3.9±0.3[f] | 33±9.9[f] | 85±3.0[f] | 1.9±0.3[f] |
| | Goseung | E3 | 49.0 | e | 11[e] | 11[e] | 118[e] | 80[e] | 1.1[e] |

Bivalve, [a]*Ruditapesphilippinarum*; [b]*Scapharcasubcrenata*; [c]*Crassostreagigas*; [d]*Mytilusedulis*; [e]*Patinopectenyessoensis*; Cephalopod, [f]*Halocynthiaroretzi*; [g]*Styelaclava*; Seaweed,[h]*Undariapinnatifida*; [i]*Laminaria japonica*

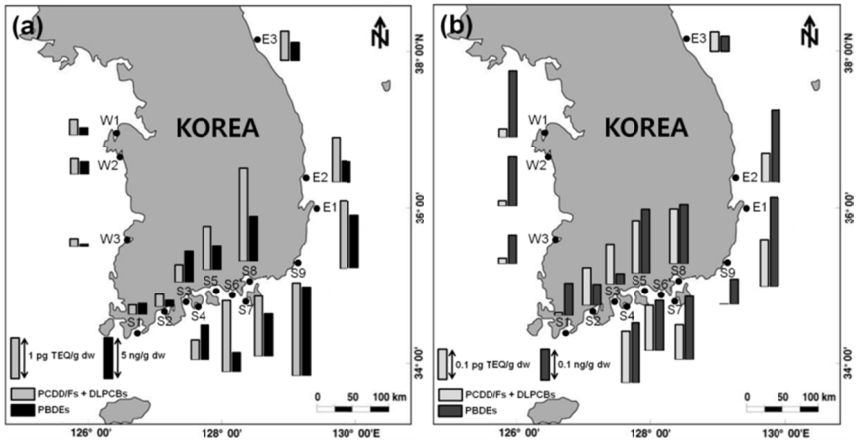


Fig. 1.Concentrations of PCDD/Fs, DLPCBs, and PBDEs in (a) sediment and (b) culturing organisms in this study.

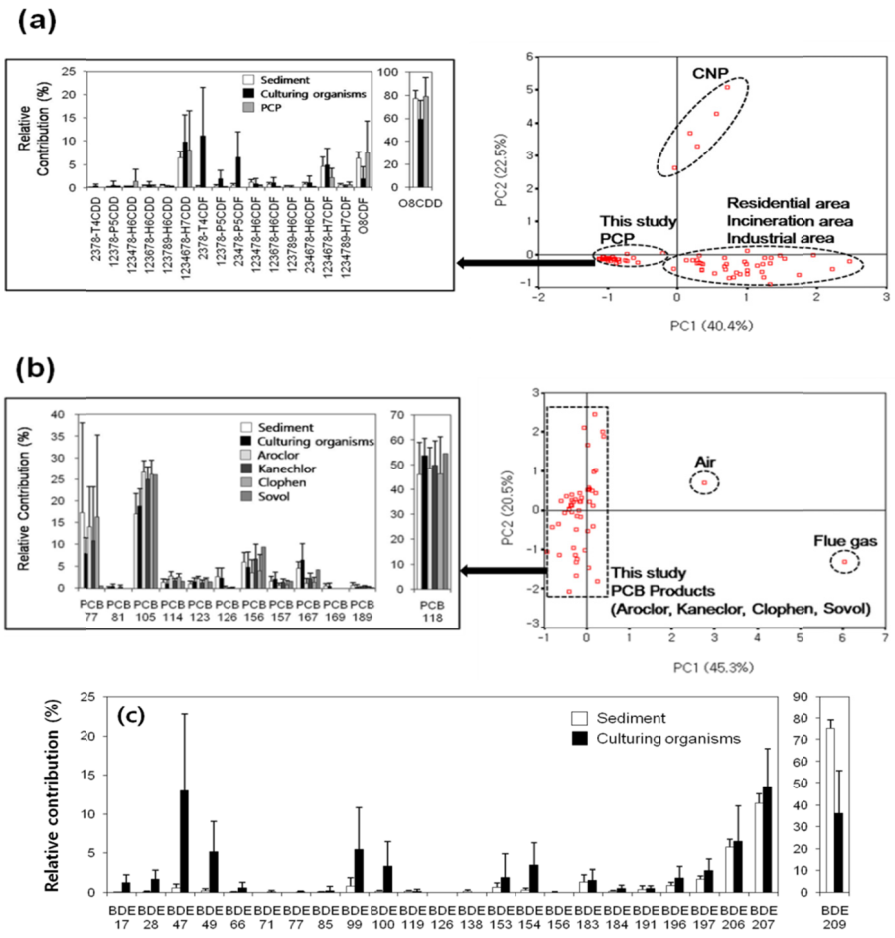


Fig. 2.Principal component analysis of (a) PCDD/Fs, (b) DLPCBs, and (c) PBDEs congener pattern in sediment and culturing organisms from culturing grounds, PCDD/Fs and DLPCBs sources.

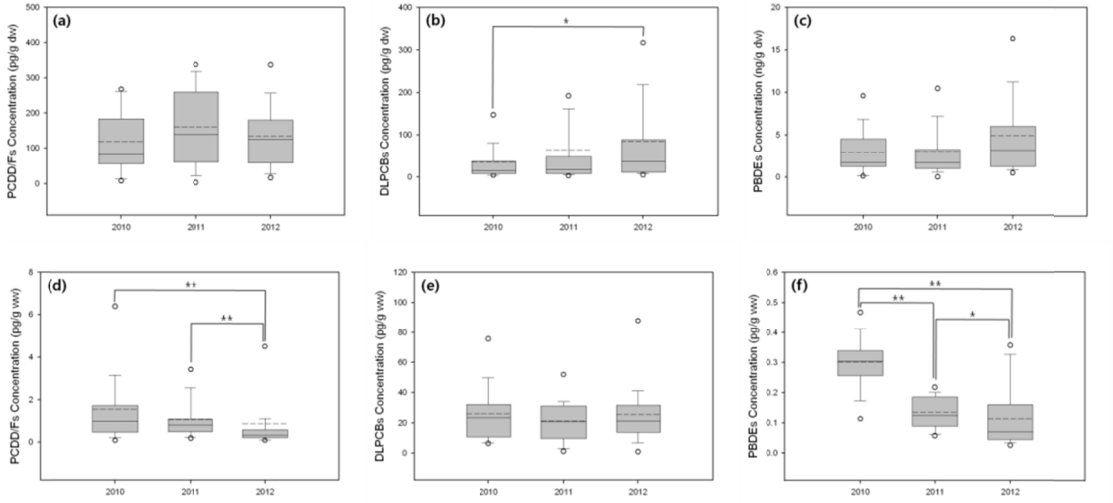


Fig. 3.Concentrations of PCDD/Fs, DLPCBs, and PBDEs in sediment and culturing organisms from 2010 to 2012.*$p$< 0.05. **$p$< 0.005