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Integrated assessment of persistent toxic substances in sediments from Masan Bay, South Korea: Comparison between 1998 and 2014

Junghyun Lee [a, 1], Seongjin Hong [b, 1], Bong-Oh Kwon [a], Seung Ah Cha [c], Hee-Dong Jeong [d], Won Keun Chang [e], Jongseong Ryu [c], John P. Giesy [f], Jong Seong Khim [a, *]

[a] School of Earth and Environmental Sciences & Research Institute of Oceanography, Seoul National University, Seoul, Republic of Korea
[b] Department of Ocean Environmental Sciences, Chungnam National University, Daejeon, Republic of Korea
[c] Department of Marine Biotechnology, Anyang University, Ganghwagun, Incheon, Republic of Korea
[d] East Sea Fisheries Research Institute, National Institute of Fisheries Science, Gangneung, Republic of Korea
[e] Korea Maritime Institute, Busan, Republic of Korea
[f] Department of Veterinary Biomedical Sciences & Toxicology Centre, University of Saskatchewan, Saskatoon, SK, Canada

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ABSTRACT

Complexity of anthropogenic influences on coastal ecosystems necessitates use of an integrated assessment strategy for effective interpretation and subsequent management. In this study a multiple lines of evidence (LOE) approach for sediment assessment, that combined use of chemistry, toxicity, and benthic community structure in the sediment quality triad was used to assess spatiotemporal changes and potential risks of persistent toxic substances (PTSs) in sediments of Masan Bay highlighting "long-term changes" between 1998 and 2014. Specific target objectives encompassed sedimentary PTSs (PAHs, alkylphenols (APs), and styrene oligomers), potential aryl hydrocarbon receptor (AhR; H4IIE-*luc* assay)- and estrogen receptor (ER; MVLN assay)-mediated activities, and finally several ecological quality (EcoQ) indices of benthic community structure. Concentrations of target PTSs in Masan Bay sediments were generally less by half in 2014 compared to those measured in 1998. Second, AhR-mediated potencies in sediments also decreased during this time interval, whereas ER-mediated potencies increased (+3790%), indicating that there has been substantial ongoing, input of ER agonists over the past 16 years. Potency balance analysis revealed that only 3% and 22% of the AhR- and ER-mediated potencies could be explained by identified known chemicals, such as PAHs and APs, respectively. This result indicated that non-targeted AhR and ER agonists had a considerable presence in the sediments over time. Third, EcoQ indices tended to reflect PTSs contamination in the region. Finally, ratio-to-mean values obtained from the aforementioned three LOEs indicated that quality of sediments from the outer region of the bay had recovery more during the period of 16-years than did the inner region. Overall, the results showed that even with the progress supported by recent efforts from the Korean governmental pollution control, PTSs remain a threat to local ecosystem, especially in the inner region of Masan Bay.

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1. Introduction

To understand risks that persistent toxic substances (PTSs) pose for the coastal environment, it is important to examine the occurrence, fate, and distribution of PTSs in sediments (Khim and Hong, 2014). Sediments require special attention because they can accumulate greater concentrations of PTSs than the water column. Sediments act not only as sinks, but also as secondary sources of contaminants for biota, a situation that can disrupt benthic community ecology (Hollert et al., 2002). They also serve as a source of PTS to higher trophic levels by serving as the first step and some time the largest in trophic biomagnification.

Chemical analyses can be combined with toxicological effects to monitor status of contaminated sediments, particularly in areas known to be affected by priority toxicants. However, *in vitro* cell bioassays can explain only a small portion of overall toxic potencies

* Corresponding author. School of Earth and Environmental Sciences & Research Institute of Oceanography, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea.
E-mail address: jskocean@snu.ac.kr (J.S. Khim).
[1] These authors contributed equally to this work.

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in extracted samples (Hong et al., 2015). *In vitro* assessment of cytotoxicity is limited with respect to predicting *in vivo* toxicity, which requires assessment of tissue-specific toxicity, adaptive response and metabolic conversion to predict effects on fitness of individuals (Fent, 2001). Since toxicity is a function of both exposure and response and rates of damage relative to rates of repair, *in vitro* bioassays cannot capture much of the important properties of PTS. In particular, *in vitro* studies conducted in the laboratory cannot elucidate the exposure side of the hazard relationship. Furthermore, chemical and toxicological analyses cannot capture community-level effects observed in natural systems (de Castro-Catala et al., 2016). The utility of such results is enhanced when it can be integrated more closely with ecological data. Hence, to obtain a fuller understanding, it is preferable to bring together different lines of evidence (LOE), such as chemistry, toxicity, and benthic community structure.

To predict ecological effects, studies have tended to focus on structures of communities of benthic organisms (Wernersson et al., 2015). Benthic organisms represent ecological receptors ideal for assessment of sediment quality because they are exposed both directly and indirectly by contaminated sediments (Chapman et al., 2013). Generally, responses of benthic organisms to environmental factors are assessed by use of biotic indices of numbers of taxa and individuals, diversity and tolerance, which provide an integrated evaluation of alterations caused by exposure to multiple stressors. However, it is disadvantageous that values of biotic indices are not always sensitive to effects of some stressors and there are concerns regarding subjectivity of interpretations of indices (Lim et al., 2007).

Integrated evaluations of benthic community structure together with chemistry and tests of toxic potencies of sediments can provide a more comprehensive assessment of PTS-induced contamination than the integration of only chemistry and toxicity data (McPherson et al., 2008). An adaptation of the ratio-to-reference (Long and Chapman, 1985) and ratio-to-maximum values (DelValls and Chapman, 1998) methods known as the ratio-to-mean values (RTMV) method can be used to integrate the chemical, toxicological, and ecological data (Chapman, 1990). The RTMV method is appreciated for its simplicity and visual presentation of data, despite simplification and loss of detail during reduction of data into a single index. The RTMV method is also useful for time-series monitoring, particularly in terms of enabling changes to be summarized by time and location (Cesar et al., 2009).

Masan Bay, located on the southern coast of Korea, is a semi-enclosed bay with restricted water exchange (Fig. 1). In the 1970s, large amounts of contaminants from nearby industrial complexes were discharged without appropriate treatment into Masan Bay. Thus, Masan Bay has been identified as a hot spot for coastal pollution from significant loading of land-driven pollutants. A doubling of the human population in the region from the 1990s to 2014 has further contributed to the pollution of Masan Bay (Fig. 1). With the goal of improving local water quality, in 1982, the South Korean government designated Masan Bay as a special management area, dredged out contaminated sediments and implemented the Total Pollution Load Management System (TPLMS) in 2007 (Fig. 1) (Lee et al., 2016). As a result, water quality of Masan Bay has improved, but studies continue to report severe contamination with metals and PTSs, including polycyclic aromatic hydrocarbons (PAHs), alkylphenols (APs), and polychlorinated biphenyls (Hong et al., 2009; Lee et al., 2016; Yim et al., 2014). In particular, PAHs and APs have long been identified as prominent contaminants in Masan Bay and considered as priority PTSs in coastal sediments to cause adverse effects on marine benthic organisms (Khim and Hong, 2014; Lee et al., 2016; Neff, 2002).

Prior studies of contamination of sediments by PTSs in Masan Bay have been focused on diagnosis of causes of toxicity and monitoring of status and trends (see Supplemental Materials, see Tables S1, S2, and S3). Although there is some evidence of significant reductions in concentrations of PTS in sediments from Masan Bay (Jin et al., 2016), there has yet to be a comprehensive report on emerging PTSs and their potential adverse effects on organisms. For example, styrene oligomers (SOs) are emerging pollutants resulting from plastic degradation in marine environments and the usage of plastics has been increasing, bringing attention to SO pollutants (GBI, 2012). SOs have been reported to cause adverse effects on aquatic organisms (Tatarazako et al., 2002), while their occurrence and distribution in coastal sediments are seldom documented. Our recent study demonstrated that SOs were widely distributed in sediments of coastal environments, with relatively great concentrations that comparable to PAHs (Hong et al., 2016a). Accordingly, the historical occurrence and distributions of these chemicals remain question and would be of significant concern.

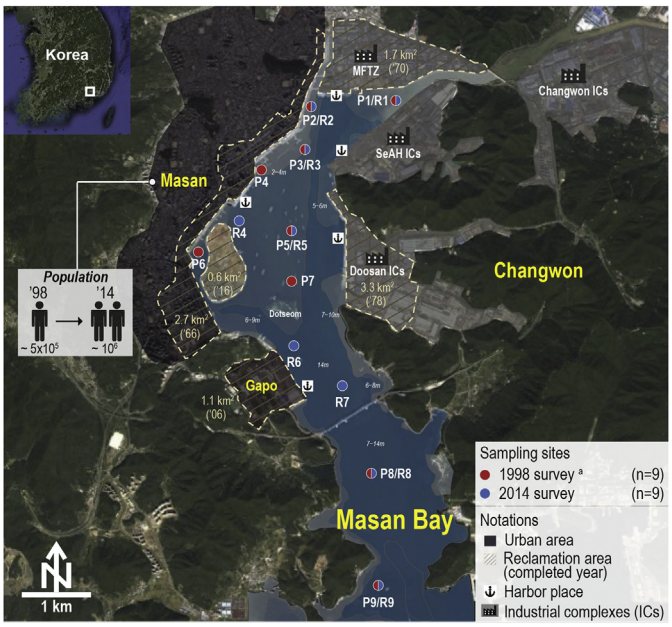

Fig. 1. Map showing locations from which sediments were collected in the inner- (P1-P4) and outer- (P5-P9) region sediment sampling sites in 1998 and the inner- (R1-R3) and outer- (R4-R9) region sediment sampling sites in 2014 ([a] Data from Khim et al., 1999a for chemical contamination, Khim et al., 1999b for toxic effects, and Ryu et al., 2016 for benthic community quality).

The purpose of the present study was to assess ecotoxicological effects of PTSs in sediments from Masan Bay by use of an integrated approach, combining chemical analyses, *in vitro* bioassays, and *in situ* investigations of benthic communities. Specific aims were to: (1) investigate spatio-temporal changes in classic PTSs, namely PAHs and APs, and emerging chemicals, namely SOs, in the sediments from Masan Bay; (2) evaluate the aryl hydrocarbon receptor (AhR)- and estrogen receptor (ER)-mediated potencies associated with sediment extracts; (3) assess the risk for benthic community being associated with residual contamination in the Masan Bay; and (4) seek common trends in integrated sediment assessment data over a recent 16-year period.

2. Materials and methods

2.1. Sampling and sample preparations

Two surveys of quality of sediments in Masan Bay were

conducted 16 years apart; sediment samples were collected in May 1998 and in May 2014 at almost the same locations (Fig. 1). At each time point, samples were collected from nine, including seven benthic community sites, termed P1-P7 and R1-R7, respectively. These sites represent both inner (P1-P4 and R1-R3; average water depth, $\leq$ 5 m) and outer (P5-P9 and R4-R9; average water depth, $\geq$ 5 m) regions of the bay.

Results of analysis to identify and quantify chemicals (except for SOs), *in vitro* bioassay data, and benthic community data from 1998 were obtained from previous studies (Khim et al. 1999a and 1999b; Ryu et al. 2016). Although sampling sites were not exactly the same and bay dynamics can shift sediment and associated contaminants over time, it is useful to evaluate these data with respect to changes in chemical concentrations over time. To address historical occurrences of SOs as well as long-term changes in concentrations of SOs in this study, archived samples from 1998 were re-analyzed for SOs levels together with samples collected in 2014.

All samples were transferred immediately to the laboratory, freeze-dried, and stored at −20 °C until analyzed. Freeze-dried samples of 1988 were kept at −20 °C in a freezer in our laboratory for 16 years. To avoid technical and/or methodological errors in chemical analyses for the use of archived samples that collected in 1998, the 1998 samples were newly extracted and analyzed together with the 2014 samples. Detailed descriptions of sample preparation for chemical and bioassay analyses have been published previously (Hong et al., 2012 and 2015). In brief, 10 g of freeze-dried sediment samples were extracted by dissolving them in 350 mL of dichloromethane (Burdick & Jackson, Muskegon, MI) in a Soxhlet extractor for 16 h. To remove elemental sulfur, the extracts were treated with activated copper powder (Sigma Aldrich, Saint Louis, MO) and concentrated into 1 mL. For the *in vitro* assays, the aliquot of the extract was exchanged in dimethyl sulfoxide (DMSO, Sigma-Aldrich) using differential volatilization.

2.2. Chemical analysis

Concentrations of PAHs, APs, and SOs in extracts of sediments were quantified by use of an Agilent 7890 gas chromatograph equipped with a 5975C mass-selective detector (MSD, Agilent Technologies, Santa Clara, CA); instrument settings used for the detection of PAHs, APs, and SOs are provided in detail in Table S4. A total of 16 PAHs, 6 APs, 4 styrene dimers (SDs), and 6 styrene trimers (STs) (full chemical names and abbreviations are provided in Table S5) were analyzed according to previously reported methods (Hong et al., 2016a). Limits of detection for PAHs, APs, and SOs in sediments, calculated as 3.707 × standard deviations of the standard samples, were 0.2–1.3 ng g^{-1} dry mass (dm), 0.1–0.9 ng g^{-1} dm, and from 0.2 to 0.9 ng g^{-1} dm, respectively (Table S5). Four surrogate standards (SS) were used to assess PAHs, APs, and SOs recovery rates. Mean recovery rates of five SS were generally within acceptable ranges (83–102% for PAHs and SOs; 77% for APs; see Table S5).

For source appointment of sedimentary PAHs and SOs, principal component analysis (PCA) was performed based on the concentrations of 16 PAHs and 10 SOs, respectively. PCA was performed with normalized chemical concentrations of individual chemicals. The statistical analyses were conducted in SPSS 23.0 software (SPSS Inc., Chicago, IL).

2.3. In vitro bioassays

H4IIE-*luc* bioassays were performed to detect AhR-mediated activities according to previously reported methods (Hong et al., 2012). H4IIE-*luc* bioassay results (expressed as mean relative luminescence units) were converted to percentages of the maximum 2,3,7,8-tetrachlorodibenzo-*p*-dioxin response (% $TCDD_{max}$), where 300 pM TCDD was considered 100% $TCDD_{max}$. AhR-mediated potencies were expressed as a TCDD equivalent concentration (pg TCDD-EQ g^{-1} dm) for direct comparison to instrumentally-derived TCDD equivalent concentrations (TEQs). All samples were assayed in triplicate.

An MVLN bioassay was used to evaluate ER-mediated potencies in organic extracts of the sediments (Khim et al., 1999b). Luciferase activity was determined after 72 h of exposure as described previously (Villeneuve et al., 2002). MVLN bioassay responses were converted to relative response units expressed as the percentage of the maximum response (%$E2_{max}$) observed for 1235 nM 17β-estradiol (E2). Significant responses were defined as those that were at least three times the standard deviation of the mean of the solvent controls. The E2 standard equivalent concentration (pg E2-EQ g^{-1} dm) was also calculated using the same method. All samples were assayed in triplicate.

2.4. Potency balance analysis

A potency balance analysis between bioassay-derived concentrations (TCDD-EQs and E2-EQs) and instrument-derived concentrations (TEQs and EEQs) in the sediments were conducted to determine the contribution of each known chemical to total induced dioxin-like and estrogenic activities. TEQs were calculated as a TEQ sum by multiplying the concentration of individual PAHs with previously established relative potency values (RePs) (Villeneuve et al., 2002; Table S6). EEQ values for APs were summed from the chemical concentrations of 4-*tert*-octylphenol (4-*t*-OP) and nonylphenols (NPs) multiplied by their previously established RePs (Villeneuve et al., 1998).

2.5. Benthic community analysis

Duplicate sediment samples were collected with a van Veen grab (surface area, 0.1 m^2). Of note, pooled samples were used for species identification and individual organism counting to minimize site-specific variation or possible technical errors in grab sampling. For ecological quality (EcoQ) assessment, we employed one simple index, the Shannon-Wiener diversity index (H′), and four multivariate indices, namely the Ecological Quality Ratio (EQR), Benthic Quality Index (BQI), Azti Marine Biotic Index (AMBI), and Multivariate-AMBI (M-AMBI). More details about these multivariate indices are provided in Table S7 and in previous publications (Blanchet et al., 2008; Ryu et al., 2016). Pearson's correlation coefficients were determined to assess potential correlations between PTS contamination and toxic effects on the benthic community. Statistical analyses were performed in SPSS 23.0 (SPSS Inc., Chicago, IL).

2.6. Integrated approach: RTMV method

The RTMV method is an integrative approach wherein a data matrix is used to convert values for each variable of interest in each LOE to non-dimensional values by dividing the value obtained by the arithmetic mean obtained for all stations (Cesar et al., 2009). RTMVs were combined through the calculation of a mean, thus producing a single new value for each LOE. These single new values were plotted in three-axis graphs to producing a triangular pyramid reflecting each class at each survey time point, providing a visual representation of sediment quality.

3. Results and discussion

3.1. Distributions of PTSs in sediments

Mean concentrations of PAHs were 4.6×10^2 ng g^{-1} dm (range, 90–1.1×10^3 ng g^{-1} dm) in 1998 and 91 ng g^{-1} dm (range, 58–1.9×10^2 ng g^{-1} dm) in 2014 (Fig. 2a), demonstrating an 80.1% reduction and a statistically significant decline ($p < 0.05$). Reductions in concentrations of PAHs in sediments from outer regions (P5-P9 and R4-R9) were significantly more pronounced than those of the inner regions (P1-P4 and R1-R3) ($p < 0.05$, Fig. 2a). These decreasing trends seemed to be associated with South Korea's implementation of pollution control measures and management of toxic substances. For example, new environmental quality and dioxin emission standards were established in 1998, waste incinerator flue gas has been regulated since 1999, and TPLMS has been implemented since 2007 (Chang et al., 2012; Lee et al., 2016). Although these environmental regulations and chemical controls do not manage PAHs levels directly, they control land-derived pollutants, which are major causes of coastal and marine pollution. The environmental regulations and pollution controls were apparently more effective for reducing contamination of outer regions, relative to inner regions of the bay. Greater concentration of PAHs was observed at both sampling time points, presumably due to the geographical characteristics (i.e., a long, narrow inlet) limiting flushing, thereby resulting in localized PAHs sedimentation in the inner regions (Li et al., 2008).

Based on loadings of the 16 PAHs analyzed, the total variances of principal components (PCs) 1 and 2 were 67% and 16%, respectively (Fig. 2d). PC 1 had a high positive loading for 4–6 ring PAHs [benzo[*a*]anthracene (BaA), chrysene (Chr), benzo[*b*]fluoranthene (BbF), benzo[*k*]fluoranthene (BkF), BaP, indeno[*1,2,3-c,d*]pyrene (InP), and benzo[*g,h,i*]perylene)] and was thus selected to represent vehicular emission sources (Li et al., 2015). PC 2 was heavily loaded with compounds related to coal combustion, including phenanthrene, fluoranthene, and pyrene (Li et al., 2015; Fig. S1). The compositional profiles of PAHs were in principle similar for both sampling years but showed greater inner region variance in 1998. The predominant PAHs found in inner region sediments in 1998 were consistent with coal combustion derivatives. Overall, industrial waste (combustion of diesel and gasoline engines) can be considered the major origin of PAHs in sediments from Masan Bay (Rogge et al., 1993).

Patterns of distribution reductions in concentrations of APs in sediments of Masan Bay were similar to those of PAHs; and mean concentrations of APs in sediment likewise decreased from 1998 (5.8×10^2 ng g^{-1} dm; range, 1.8×10^2–1.1×10^3 ng g^{-1} dm) to 2014 (39 ng g^{-1} dm; range, 18–72 ng g^{-1} dm) (Fig. 2b). NPs were used as detergents, wetting agents, dispersing agents, and emulsifiers in various industrial, domestic, and household applications throughout the latter half of the twentieth century (Hong and Shin, 2011). The South Korean government named NPs as priority chemicals in 2001, banning their use in kitchen cleaners in 2002, and then designating them as restricted chemicals and prohibiting their use for all domestic applications in 2007. The use of NPs in paints and ink binders was banned in 2010 (MOE, 2007). Our observation that means concentrations of APs declined by 93% between 1998 and 2014 (Fig. 2b, $p < 0.01$) indicates that these regulations appear to have been quite effective. The mean concentrations of APs declined significantly between 1998 and 2014 in both inner and outer bay regions. Direct comparison of APs data with past levels was difficult because historical alkylphenol ethoxylate data are lacking. Nonetheless, the presence of NP precursors in sediments collected from inner regions in 2014 suggests that fresh APs input to the bay is still occurring.

A dramatic reduction (97%) in mean concentrations of SOs in sediments was observed between 1998 (4.9×10^3 ng g^{-1} dm; range, 3.0×10^3–1.0×10^4 ng g^{-1} dm) and 2014 (1.3×10^2 ng g^{-1} dm; range, 73–3.5×10^2 ng g^{-1} dm) (Fig. 2c). Concentrations of SOs

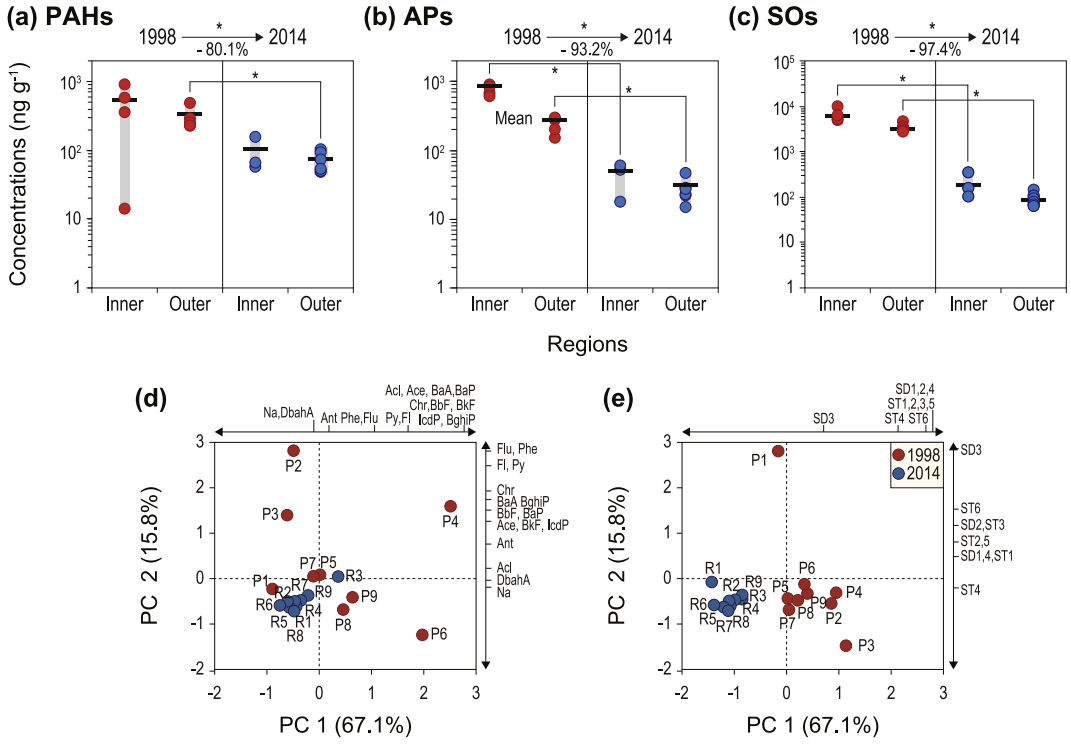

Fig. 2. Concentrations of (a) PAHs, (b) APs, and (c) SOs in sediment from Masan Bay in 1998 and 2014. Cluster results from PCA of (d) PAH and (e) SO concentrations in sediments collected from 18 locations in Masan Bay. Percentages of variability in each data accounted for by PC 1 and PC 2 are shown in the graph ($^*p < 0.05$).

in sediments from Masan Bay in the presently reported 1998 and 2014 are greater than respective mean values from similar time points (1998 mean, 2.2×10^2 ng g^{-1} dm; 2015 mean, 34 ng g^{-1} dm) for sediments from Lake Sihwa in South Korea (Hong et al, 2016a; Lee et al., 2017). Results of PCA indicated that environmental management practices, such as TPLMS, have been successful in reducing land-based pollution loads. PC 1 and PC 2 accounted for 67.1% and 15.8% of the dataset variability, respectively (Fig. 2e), and when sites were ordinated based on PCA scores, all samples except P1 were divided into sampling years 1998 and 2014. SD3 was the predominant (70%) chemical pollutant at P1, whereas STs were the predominant pollutants at other sites (Fig. S2). Relatively large contributions of STs might indicate recent inputs of fresh materials because ST1 emerges early in polystyrene decomposition after mechanical breakdown (Saido et al., 2014). However, our understanding of SO compound is limited because few studies have been conducted on the distributions and relative compositions of SOs in coastal marine sediments (Kwon et al., 2014). Additional complementary studies are needed to identify the origins and fates of these compounds. Furthermore, continuous pollutant monitoring is needed and regulation of new materials should be considered.

3.2. *In vitro* potencies in sediments

Mean concentrations of TCDD-EQ in sediments of the Masan Bay decreased from 5.9×10^2 pg g^{-1} dm (range, 39–2.1×10^3 pg g^{-1} dm) in 1998 to 75 pg g^{-1} dm (range, 50–1.6×10^2 pg g^{-1} dm) in 2014 (Fig. 3a); mean concentrations of AhR-mediated potencies in sediments also decreased significantly between 1998 and 2014 ($p < 0.05$). These TCDD-EQs exceeded both US (<25 pg g^{-1} dm, possible-effect level; US EPA, 1993) and Canadian (<0.85 pg g^{-1} dm; CCME, 2002) sediment quality guidelines for dioxin-like compounds at both time points. In contrast to concentration distribution patterns of PAHs, AhR-mediated potencies in 1998 were greater in sediments from outer regions than in sediments from inner regions. With the exception of outer region samples in 1998, AhR-mediated potencies were generally well correlated with PAHs concentrations in sediments (Fig. 3b). These results suggested that the aforementioned actions implemented to reduce the release of chemicals (including AhR agonists) into the bay have been effective. Indeed, dioxin emissions decreased by 88% from 2001 to 2011 (MOE, 2012). The results of a direct comparison between bioassay-derived TCDD-EQ and instrument-derived TEQ potency balance analyses conducted to identify chemical-specific contributions to total induced AhR-mediated activities in sediments are presented in Table 1. Known AhR agonists such as PAHs and SOs explained only a small portion of TCDD-EQs, ~2.7% in 1998 and ~2.2% in 2014 (Table 1), revealing the apparent presence of possible unknown AhR agonists in sediments. For instance, several untargeted AhR active agonists such as dioxins and furans, some co-planar PCBs and PCNs, four- to five-ring PAHs and/or their derivatives (e.g., oxy-, nitro-, sulfur-, alkyl-, cyano-, amino-, or methylated PAHs) in sediments of Masan Bay might explain the unidentified proportion of dioxin-like activities (Barron et al., 2004; Horii et al., 2009; Kannan

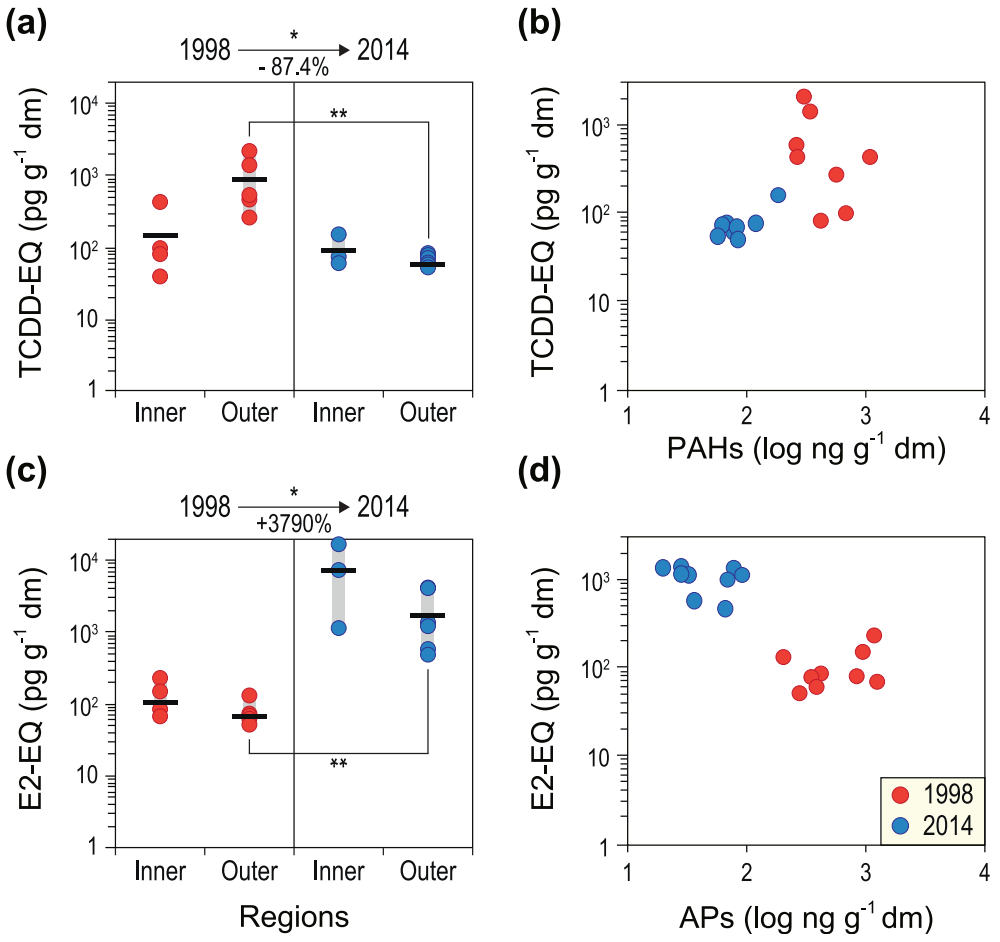

Fig. 3. Spatiotemporal distributions of (a) TCDD-EQ and (c) E2-EQ biological responses in inner- and outer-region Masan Bay sediment samples for 1998 and 2014. (b and d) Scatter plots showing dose-response relationships between chemical contaminant levels and biological responses in sediment samples collected from Masan Bay ($^*p < 0.05$).

Table 1
Comparison of instrument-derived equivalents and bioassay-derived equivalents in 1998 and 2014 sediment samples from Masan Bay, South Korea.

Sampling year	Region	No. sites	Instrument-derived equivalents		Bioassay-derived equivalents [a]		Potency balance analysis [b]	
			TEQ [c]	EEQ [d]	TCDD-EQ	E2-EQ	TEQ/ TCDD-EQ	EEQ/ E2-EQ
			Min.-Max. (Mean)	Min.-Max. (Mean)	Mean	Mean	Min.-Max. (Mean)	
			(pg g^{-1} dm)		(pg g^{-1} dm)		(%)	
1998 [e]	Inner	7	0.3–5.7 (2.6)	9.8–14.8 (12.3)	159	127	0.9–2.7 (1.7)	6.1–21.9 (12.1)
	Outer	7	1.2–4.3 (2.1)	2.5–4.8 (4.0)	942	78.3	0.1–0.9 (0.4)	1.9–7.0 (5.7)
2014	Inner	7	0.6–2.9 (1.5)	0.2–0.8 (0.4)	100	8580	0.8–1.8 (1.4)	n.d. [f]-0.1(0.04)
	Outer	7	0.6–1.4 (0.8)	0.2–0.7 (0.4)	62.6	1530	0.8–2.2 (1.4)	n.d.-0.1 (0.02)

[a] Bioassay-derived values obtained from sample dose-response relationships generated by testing samples at multiple dilution levels.
[b] Values are the percentages of instrument-derived values relative to bioassay-derived values.
[c] TEQ values of PAHs were summed from concentrations of BaA, Chr, BbF, BkF, BaP, IcdP, and DBahA multiplied by the ReP values reported in Villeneuve et al., 2002.
[d] EEQ values of APs were summed from concentrations of NPs and 4-*t*-OP multiplied by the ReP values reported in Villeneuve et al., 1998.
[e] Data from Khim et al. 1999a and 1999b.
[f] n.d.: Below detection limits.

et al., 2000; Khim et al., 1999a; Trilecova et al., 2011).

Mean concentrations of E2-EQs in sediments were 1.0×10^2 pg g^{-1} dm (range, 51–2.2×10^2 pg g^{-1} dm) in 1998 and 3.9×10^3 pg g^{-1} dm (range, 4.9×10^2–1.7×10^4 pg g^{-1} dm) in 2014 (Fig. 3c), evidencing an approximately 40-fold increase over about 16 years. Although concentrations of APs known to be ER agonists decreased, overall ER-mediated potencies increased between 1998 and 2014. Thus, the correlation between ER-mediated potencies and concentrations of APs showed an opposite tendency relative to AhR activity (Fig. 3d). ER-mediated potencies expressed as instrument-derived EEQs were at 22% in 1998, but only 0.1% in 2014 (Table 1). These results indicated that other untargeted ER agonists might exist in sediments of Masan Bay. For example, several unmeasured chemicals such as pesticides (DDT, *o*,*p*′-DDD, and *o*,*p*′-DDE), kepone, and parabens have been reported to show ER binding affinity, which might be present in sediments of Masan Bay (Gadio et al., 1997; Legler et al., 1999; Routledge et al., 1998). In other words, APs were not a major ER agonist in Masan Bay sediments. Overall, the portion of unknown AhR agonists and ER agonists increased over the sampling interval. Unknown toxic chemicals may contribute to the total induced toxicity in coastal sediments. Thus, complementary research, such as effect-directed analysis, may lead to a better understanding of new substances and identification of appropriate countermeasures (Hong et al., 2016b).

3.3. Macrobenthic community

The number of species (taxa) and species density differed between the two sampling years ($p < 0.05$, Fig. 4a and b). In 1998, a total of 14 taxa were found with a mean density of 241 ind. m^{-2}, whereas in 2014, 45 taxa were found with a mean density of 1800 ind. m^{-2}. Few samples were collected in some sites and the range of mean densities in the inner region varied broadly in 1998 (Table S8). It seemed that contamination of PTSs was serious in sediments from 1998, potentially leading to deterioration of some inner-region sites. Outer regions presented greater diversity than inner regions, indicating that benthic biodiversity reflected the geographical and contamination gradients of the semi-enclosed Masan Bay system (Khim et al., 1999a; Khim and Hong, 2014). The aforementioned numbers of species observed in Masan Bay sediments were lower than values previously recorded for the southeastern coastal area (means in 1998 and 2014 were 43 and 123, respectively; MOF, 2014; Ryu et al., 2016), suggesting that the benthic community in Masan Bay had not yet fully recovered by 2014. Indeed, some polychaete species such as *Capitella capitata* and *Lumbrineris longifolia*, that are well-known opportunistic species and organic pollution indicators were dominant across our sampling sites (Tables S3 and S8; Bae et al., 2017).

The five EcoQ indices calculated based on our benthic community data showed some improvement in ecological quality over the study time interval. The H′ index results for all sites were moderate or bad in 1998, but good or poor in 2014 (Fig. 4c). Likewise, EQRs for the sites shifted from bad or poor in 1998 to poor or moderate in 2014 (Fig. 4d). Among the five indices, these two indices best-reflected contamination of PTSs in sediment (Fig. 4 and Fig. S3). The AMBI, by comparison, seemed to be less sensitive than the other multivariate indicators to sedimentary pollution, yielding bad to good values in 1998, where other indicators indicated poor or bad outputs (Fig. S3). Averaging of index grading indicated that the ecological qualities of the inner and outer regions in 1998 were 4.45 (poor) and 3.87 (moderate), respectively, improving to 3.2 (moderate) and 2.54 (good), respectively, by 2014. These EcoQ assessment-based results indicated that Masan Bay sediment quality improved over time, albeit under the constraints of the geographical setting and PTSs contamination of the bay. This conclusion is further reinforced by the increased biodiversity observed over the study time interval.

Pearson correlation analysis demonstrated a multitude of significant correlations among the presently developed chemical analysis, toxic effect, and benthic community quality variables (see Table S9 for coefficient values and significance levels). Notably, all of the chemical concentration values correlated significantly with EQR and BQI ($p < 0.05$, Table S9), suggesting that these indices of benthic community composition are responsive to chemical concentration changes. However, results of neither of the toxicity bioassays were correlated with any of the EcoQ indices. This pattern of greater sensitivity to sedimentary toxicants in resident benthos than *in vitro* toxicity bioassays is consistent with similar analyses of US estuaries (Hyland et al., 1999). Overall, the EcoQ results were useful for demonstrating general pollution of sediment.

3.4. Integrated approach: RTMV

The data obtained from chemical contamination, toxicity effect, and benthic community (EQR and BQI) data were integrated by use of the RTMV method to identify how conditions within the sampled sites changed over the study time interval. Comparing RTMVs calculated for samples with respect to time (1998 vs. 2014) and region (inner vs. outer) revealed an overall decrease of RTMV from

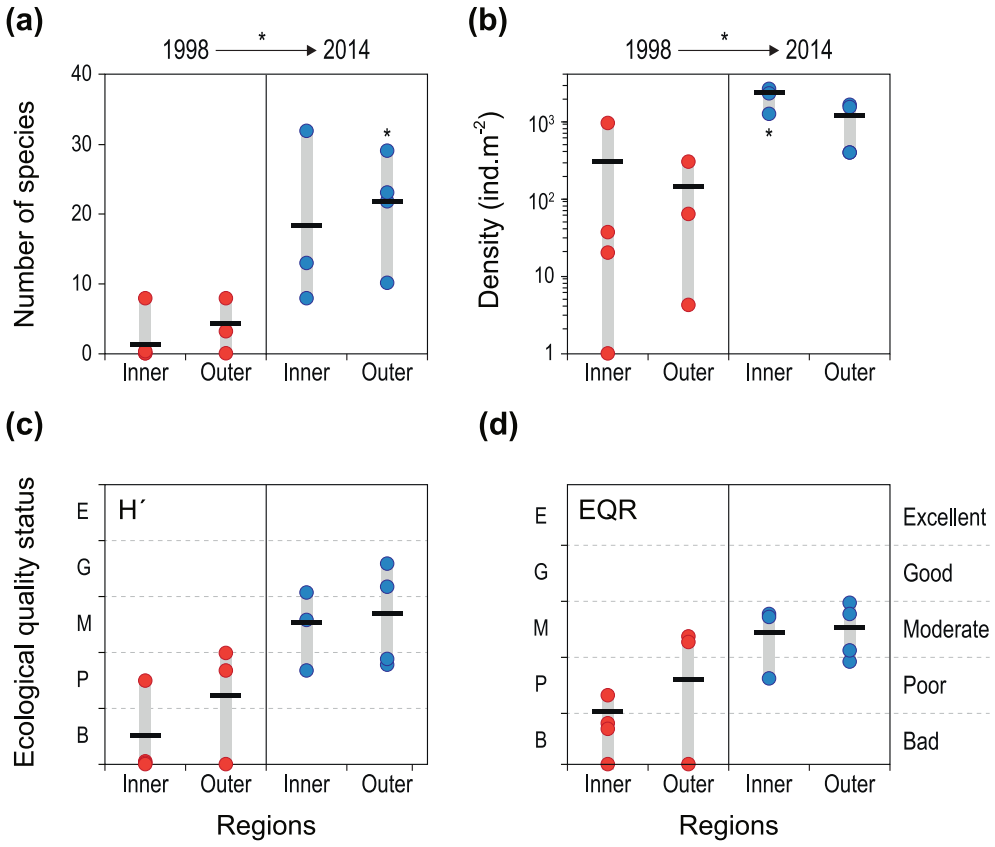

Fig. 4. Recovery of benthic community health in Masan Bay. Comparisons of (a) numbers of taxa (species) and (b) distributions (density, ind. m^{-2}) of benthic communities in Masan Bay between 1998 and 2014. Comparisons of EcoQ status as represented by (c) the Shannon-Wiener (H′) index and (d) the EQR in Masan Bay between 1998 and 2014 ($^{*}p < 0.05$).

1.34 in 1998 to 0.78 in 2014, a pronounced outer-region RTMV decrease from 1.28 in 1998 to 0.44 in 2014 ($p < 0.01$), and a no significant inner-region decrease from 1.39 in 1998 to 1.12 in 2014. It was demonstrating better and more improved conditions in the outer regions than in the inner regions. Chemical contamination, toxic potency, and benthic community index RTMVs each decreased in the outer regions from 1998 to 2014; meanwhile, during the same time interval, only the chemical contamination and benthic community index RTMVs decreased in the inner regions, while the inner-region toxicity RTMV rose sharply (Fig. 5). Presumably, because of the geographical features of the semi-closed bay, it is taking more time for sediment quality to recover in the inner regions than in the outer regions. In the inner regions, chemical contamination was the most pressing aspect of bay pollution in 1998, whereas toxic effect had emerged as the more important concern in 2014. Meanwhile, in the outer regions, the values of three factor were of similar magnitude in 1998, whereas benthic community quality and toxic effect RTMVs were of notably greater magnitude than the chemical contamination RTMV in 2014.

The inner-region and outer-region chemical contamination RTMVs fell dramatically from 1998 (2.59 and 1.23, respectively) to 2014 (0.22 and 0.14, respectively). The APs and SOs components of the chemical contamination RTMVs showed more pronounced reductions than did the PAHs component in both the inner and outer

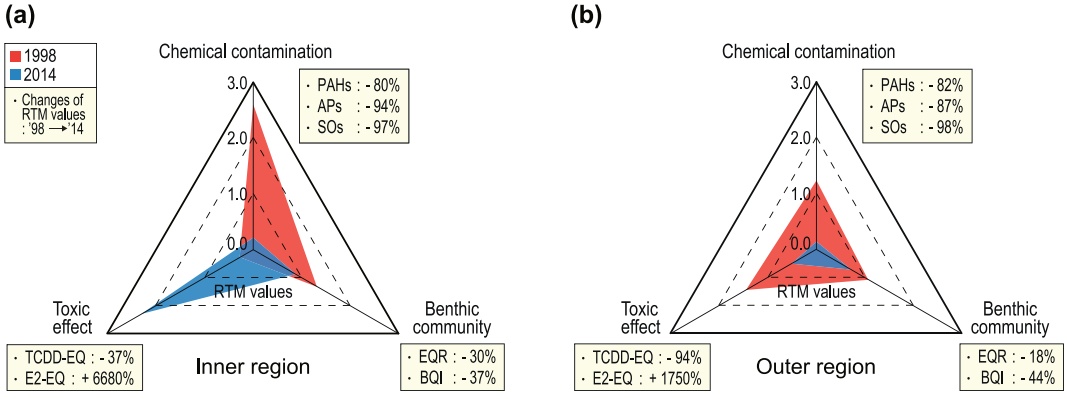

Fig. 5. Sediment quality triads of normalized to RTMVs obtained for chemical contamination, toxic effects, and benthic community quality of sediment samples from (a) inner and (b) outer regions in 1998 and 2014.

regions (Fig. 5), perhaps because PAHs continue to be generated and transported unceasingly through the atmosphere, whereas APs are now strictly controlled. Previous studies on PAHs and APs in sediments of Masan Bay also revealed that the RTMVs exhibited a generally decreasing trend both inner and outer regions during the last decade (Fig. S4). Overall, these reduced RTMVs for chemical contamination reflect effective legislative actions.

RTMVs of toxic effect increased from 0.86 in 1998 to 1.40 in 2014, but opposite tendencies were observed for the inner versus outer regions. The RTMVs obtained for sediments from inner regions increased about eight-fold over the ~16-year period, while those from outer regions decreased by almost two thirds. RTMVs for E2-EQs were increased in both inner and outer regions. However, these increasing RTMVs were offset by decreasing RTMVs for TCDD-EQs in outer regions. These results indicate that unknown ER agonists have been, and likely are still, accumulating more in inner regions than in outer regions and/or that there are ER agonist sources near inner-region sites.

The RTMV pattern of the benthic indices was similar to that observed for our chemical analysis, although less dramatic changes of the study time interval. These results show that while chemical concentrations can be reduced rapidly, it takes more time for benthic communities to recover. Other conditions such as metal contaminations and/or hypoxia in bottom water were considered as anthropogenic pressures on the benthic community. Results of previous study conducted in Masan Bay indicated that benthic ecological quality generally reflected the pollution gradient of metals (Ryu et al., 2016). Similar to the temporal decrease of target organic PTSs, some metal concentrations, such as Cu, Zn, and Pb, reported in sediments of Masan Bay showed a decreasing trend during the last decade (Table S10). Thus, in addition to the important work of regulating the use and emission of chemicals, ongoing assessments of ecological impact are also needed. Overall, RTMVs of chemical contamination and benthic community decreased while the toxicity RTMV increased, indicating that there are unmeasured chemicals or conditions with the potential to cause degradation of inner-region in Masan Bay ecosystem.

4. Conclusions

This study is the first to provide an integrated assessment of relatively long-term changes in the sediment quality of Masan Bay. The following findings regarding the spatio-temporal distributions of PTSs, AhR- and ER-mediated potencies, and risks to the benthic community in Masan Bay were obtained:

- Concentrations of target PTSs (PAHs, APs, and SOs) decreased significantly from 1998 to 2014.
- Greater concentrations of PTSs were found at inner, compared to outer, region sites presumably due to the geographical and oceanographic settings of the bay.
- AhR-mediated potency decreased over time, while ER-mediated potency increased, indicating continuous inputs of unknown ER agonists.
- Increased species number and density findings in 2014 compared with 1998 provide evidence of benthic community recovery; among the five indices, EQR and BQI tended to best reflect PTSs contamination.
- An integrated analysis of three LOEs (chemistry, toxicology, and benthic community) revealed a gradient of contamination, with higher levels occurring towards the inner region and sediment quality showing more recovery during the study time interval in the outer region than in the inner region.

Results of this study document relatively long-term changes of an important class of environmental pollutants and associated biological responses in severely contaminated coastal sediments and provides a baseline from which evaluate future actions intended to improve sediment quality in the Masan Bay.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2018.02.064

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<Supplementary Materials>

Integrated assessment of persistent toxic substances in sediments from Masan Bay, South Korea: Comparison between 1998 and 2014

Junghyun Lee [1], Seongjin Hong [1], Bong-Oh Kwon, Seung Ah Cha, Hee-Dong Jeong, Won Keun Chang, Jongseong Ryu, John P. Giesy, Jong Seong Khim [*]

Supplementary Tables

Supplementary Figures

[1] These authors contributed equally to this work.

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

Supplementary Tables

Table S1. Summary of reported PAH, AP, and SO concentrations in Masan Bay sediments.

Regions	Sampling Year	Site Number (n)	Target chemicals	Concentrations (ng g^{-1} dw)				References
				Min.	Max.	Mean	Median	
In land	2000	8	16 PAHs	33.4	481	216	157	Koh et al., 2005
	2000	8	NP	84.7	1070	395	315	Koh et al., 2005
Bay	1997	20	16 PAHs	207	2670	680	-[a]	Yim et al., 2005
	1998	28	16 PAHs	41.5	1100	354	312	Khim et al., 1999a
	1998	28	NP+OP	122	4070	527	346	Khim et al., 1999a
	1998	7	10 SOs	3110	10200	4940	3810	***This study***
	2000	1	16 PAHs	-	-	55.4	-	Moon et al., 2001
	2004	18	NP+ NP1EO+ NP2EO	131	2810	581	421	Li et al., 2008
	2005	20	16 PAHs	123	1670	928	-	Moon et al., 2008
	2005	20	NP	39.7	1208	411	-	Moon et al., 2008
	2006	5	NP	24	504	248	206	Hong et al., 2009
	2006-2007	5	NP+ NP1EO +NP2EO	49.4	124	71.1	63.0	Choi et al., 2009
	2010	9	NP	28.4	589	245	157	Al-Odaini et al., 2015
	2012	21	16 PAHs	171	707	309	292	Jung et al., 2012
	2012	21	NP+ NP1EO +NP2EO	142	2190	574	483	Jung et al., 2012
	2013	20	16 PAHs	47.9	151	83.7	-	Jin et al., 2016
	2014	29	16 PAHs	-	-	175	-	Yim et al., 2014
	2014	7	16 PAHs	58.1	191	90.7	80.2	***This study***
	2014	7	APs	17.8	71.7	39.3	32.9	***This study***
	2014	7	10 SOs	73.1	353	128	100	***This study***

[a] -: No data.

Table S2. Summary of reported AhR- and ER-mediated potencies in Masan Bay sediments.

Regions	Sampling Year	Site Number (n)	Endpoint	AhR- or ER- activity			References
				Min.	Max.	Mean	
In land	2000	8	TCDD $_{max}$	37.8	93.5	64.6	Koh et al., 2005
		8	E2 $_{max}$	9.3	113	44.9	Koh et al., 2005
	2003	15	TCDD $_{max}$	0.1	93	35	Yoo et al., 2006
Bay	1998	9	TCDD $_{max}$	39.4	2080	594	Khim et al., 1999b
		9	E2 $_{max}$	51.4	221	99.7	Khim et al., 1999b
	2014	7	TCDD-EQ	49.8	161	75.1	***This study***
		7	E2-EQ	486	17000	3880	***This study***

Table S3. Summary of reported temporal occurrence of dominant macrozoobenthos species found in Masan Bay.

Phylum	Species	Year					
		1980-1981	1987-1990	1998	2004	2010-2012	2014
Annelida	*Paraprionospio patiens*	✓	✓	✓	✓	✓	✓
	Nectoneanthes latipoda	✓					
	Chone sp.	✓		✓			
	Nephtys sp.	✓					
	Capitella capitata		✓		✓	✓	✓
	Chaetozone setosa		✓	✓	✓		
	Lumbrineris longifolia		✓	✓	✓	✓	✓
	Cirratulus cirratus			✓			
	Glycinde sp.			✓			
	Sigambra tentaculata			✓			
	Tharyx sp.			✓	✓		✓
	Euchone analis				✓		
	Glycera chirori				✓		
	Heteromastus filiformis				✓	✓	✓
	Prionospio cirrifera				✓		
	Magelona japonica						✓
	Polydora ligni						✓
	Spiochaetopterus koreana						✓
	Sternaspis scutata						✓
Mollusca	*Theora lata*						✓
	Raetellops pulchella						
	Macoma tokyoensis						
Amphipoda	*Corophium sp.*						
Urochordata	*Ciona intestinalis*						
Reference		a	b	c	d	e	f

a. Hong and Lee., 1983, b. Lim and Hong., 1997, c. Paik and Yun., 2000; Lim et al.,2007; Ryu et al., 2016, d. Choi et al., 2005, e. KORDI.,2010; Seo et al., 2015, f. ***This study***.

Table S4. Gas chromatograph-mass-selective detector instrument conditions for determination of PAHs, APs, and SOs.

GC/MSD system	Agilent 7890A GC and 5975C MSD	
Column	DB-5MS (30 m long × 0.25 mm i.d × 0.25 μm film thickness)	
Gas flow	1.0 mL/min (He)	
Injection mode	Splitless	
MS temperature	180 °C	
Detector temperature	230 °C	
Injection volume	2 μL	1 μL
Oven temperature	1. 60 °C hold 2 min 2. Increase 6 °C/min to 300 °C 3. 300 °C hold 13 min	1. 60 °C hold 5 min 2. Increase 10 °C/min to 100 °C 3. Increase 20 °C/min to 300 ° C 4. 300 °C hold 6 min
Target Compounds	PAHs, SOs	APs

Table S5. Abbreviations of chemicals and quality assurance/quality control data for sedimentary PAHs, APs, and SOs measured in the present study.

Organic Chemicals		**Method detection limit**	**Surrogate recovery**
Full name	**Abbreviation**	**(ng g dw^{-1})**	**(%, n=7)**
PAHs			
Naphthalene	Na	0.68	
Acenaphthylene	Acl	0.78	
Acenaphthene	Ace	0.89	
Fluorene	Flu	1.32	
Phenanthrene	Phe	0.95	
Anthracene	Ant	0.40	
Fluoranthene	Fl	0.95	
Pyrene	Py	0.90	
Benzo[*a*]anthracene	BaA	0.62	
Chrysene	Chr	0.95	
Benzo[*b*]fluoranthene	BbF	0.66	
Benzo[*k*]fluoranthene	BkF	0.86	
Benzo[*a*]pyrene	BaP	0.69	
Indeno[*1,2,3-cd*]pyrene	IcdP	0.72	
Dibenz[*a,h*]anthracene	DbahA	0.17	
Benzo[*g,h,i*]perylene	BghiP	0.61	
APs			
4-*tert*-octylphenol	4-*t*-OP	0.07	
4-*tert*-octylphenol monoethoxylate	4-*t*-OP1EO	0.09	
4-*tert*-octylphenol diethoxylate	4-*t*-OP2EO	0.09	
Nonylphenol	NPs	0.93	
Nonylphenol monoethoxylate	NP1EOs	0.45	
Nonylphenol diethoxylate	NP2EOs	0.76	
Bisphenol A-d16	BPA-d16		76.8 ± 18.7 [a]
SOs			
1,3-Diphenylpropane	SD1	0.34	
cis-1,2Diphenylcyclobutane	SD2	0.65	
2,4-Diphenyl-1-butene	SD3	0.94	
trans-1,2-Diphenylcyclobutane	SD4	0.23	
2,4,6-Triphenyl-1-hexene	ST1	0.57	
1e-Phenyl-4e-(1-phenylethyl)-tetralin	ST2	0.53	
1a-Phenyl-4e-(1-phenylethyl)-tetralin	ST3	0.30	
1a-Phenyl-4a-(1-phenylethyl)-tetralin	ST4	0.49	
1e-Phenyl-4a-(1-phenylethyl)-tetralin	ST5	0.32	
1,3,5-Triphenylcyclohexane	ST6	0.34	
Acenaphthene-d10	Ace-d10		82.9 ± 10.1
Phenanthrene-d10	Phe-d10		102 ± 22.2
Chrysene-d12	Chr-d12		97.6 ± 14.6
Perylene-d12	Pery-d12		92.9 ± 20.4

[a] Mean ± SD.

Table S6. Relative potency values of measured PAHs and alkylphenols for the AhR-mediated (H4IIE-*luc* cell) and RE-mediated (MVLN cell) activities, respectively.

Target compound	**H4IIE-*luc* cell** [a]	**MVLN cell** [b]
Benzo[*a*]anthracene	1.9×10^{-6}	
Chrysene	2.3×10^{-6}	
Benzo[*b*]fluoranthene	5.1×10^{-6}	
Benzo[*k*]flouranthene	1.4×10^{-4}	
Benzo[*a*]pyrene	1.6×10^{-6}	
Indeno[*1,2,3-c,d*]pyrene	1.5×10^{-5}	
Dibenz[*a,h*]anthracene	4.6×10^{-6}	
4-*tert*-octylphenol		1.9×10^{-5}
Nonylphenol		1.3×10^{-5}

[a] Villeneuve et al., 2002.

[b] Villeneuve et al., 1998.

Table S7. Definition of benthic community quality index levels.

Biotic indices	Abbreviation	Ecological status				
		Bad	Poor	Moderate	Good	Excellent
Ecological index						
Shannon-Wiener diversity index	H´[a]	< 1	1-2	2-3	3-4	> 4
Multivariate index						
Ecological Quality Ratio	EQR	< 0.2	0.2-0.43	0.43-0.65	0.65-0.80	> 0.80
AZTI Marine Biotic Index	AMBI	> 5.5	4.3-5.5	3.3-4.3	1.2-3.3	0-1.2
Multivariate-AMBI	M-AMBI[b]	< 0.2	0.20-0.41	0.41-0.62	0.62-0.83	> 0.83
Benthic Quality Index	BQI[c]	< 3.6	3.6-7.2	7.2-10.8	10.8-14.4	> 14.4

[a] Blanchet et al., 2008.
[b] Muxika et al., 2007.
[c] Rosenberg et al., 2004.

Table S8. Faunal list of macrobenthos species with abundance found in Masan Bay.

Pylum	Name	1998							2014						
		P1	P2	P3	P4	P5	P6	P7	R1	R2	R3	R4	R5	R6	R7
Annelida	*Amage* sp.										12			1	9
	Aricidea simplex										3		2		
	Capitella capitata	159				5		2		190	11	15			
	Chone teres													1	
	Cirratulus cirratus										1			1	1
	Dorvillea sp.									4	15	1		4	1
	Eteone sp.									1	6	1	1		1
	*Euchon*e sp.														5
	Glycera chirori										1				1
	Glycera convoluta														3
	Glycera onomichensis										1				
	Glycinde sp.														
	Goniada sp.										3		1	1	6
	Heteromastus filiformis														1
	Lumbrineris heteropoda												1		
	Lumbrineris longifolia					1				1	152		16	10	31
	Magelona japonica														
	Neanthes succinea					1				2	1				
	Nectoneanthes multignatha									5	5	1	6	2	9
	Nectoneanthes oxypoda	12		9	5	10		8							
	Nephtys polybranchia					1					1				
	Nereis longior									1	4		2	1	6
	Ophiodromus sp.									1	10	1	15	7	8
	Paraprionospio pinnata	1				15		3					5	11	117
	Paraprionospio sp.	7				40	1	3			2		1	1	4
	Parougia caeca	30													
	Polydora sp.													1	
	Polynoidae indet.													1	
	Prionospio elegantula									65	15	2	1	1	1
	Prionospio membranacea										1			1	29
	Scolelepis sp.										1				1
	Sigambra tentaculata									1	5		31	14	5
	Spiochaetopterus koreana									12	60	7	13		3
	Thelepus sp.												1	2	1
	indet.								2		3	1			
Sipunculida	indet.								6		4	2		2	3
Nemertina	*Lineus* sp. 1					1					3	2		2	2
Cnidaria	*Anthopleura kurogane*	14													
Mollusca	*Arcidae* indet.										1				1
	Hastula sp.								4						
	Hydatina albocincta										1	1			
	Macoma sp.								39	2	10	4	1		3
	Musculus senhausia								18		13	9		1	
	Philine orientalis	2													
	Raetella pulchella														1
	Ruditapes philippinarum								13						
	Scapharca broughtonii												1		
	Theora lata					1			20	12	184	276	61	18	100
Arthropoda	*Chionidae* indet.										2				
	Corophium sp.								457	7	123		3		26
	Gaprella sp.								1						
	Grandidierella sp.										13		5	12	9
	indet.												1		
Arthropoda	*Nebalia bipes*	5									4	5		1	1
	Oratosquilla oratoria	1													
	Number of species	9	0	1	1	9	1	4	9	14	33	15	20	23	30
	Total species	231	0	9	5	75	1	16	560	304	671	328	168	96	389
	Mean density (indiv./m^2)	1155	0	45	25	375	5	80	2800	1520	3355	1640	840	480	1945

Table S9. Pearson correlation analysis results for associations among chemical contamination levels, toxicity bioassay outcomes, and environmental quality parameters for Masan Bay benthic macrobenthic community.

		Chemical			**Bioassay**		**Macrobenthic Community**						
		PAH	APs	Sos	TCDD-EQ	E2-EQ	# of species	Density	H'	BQI	AMBI	M-AMBI	EQR
Chemical	PAH		0.54	0.48	0.17	-0.26	-0.63	-0.56	0.72	0.74	-0.05	0.74	0.77
	APs	+		0.95	-0.08	-0.38	-0.64	-0.50	0.74	0.74	0.21	0.55	0.74
	SOs		++		0.09	-0.42	-0.64	-0.50	0.70	0.75	0.32	0.39	0.65
Bioassay	TCDD-EQ					-0.16	-0.19	-0.27	0.00	0.26	-0.15	-0.36	-0.16
	E2-EQ						0.63	0.64	-0.41	-0.47	-0.07	-0.17	-0.27
Macrobenthic Community	# of species	+	+	+		+		0.82	-0.83	-0.89	-0.19	-0.58	-0.69
	Density	+				+	++		-0.58	-0.69	-0.09	-0.46	-0.55
	H'	++	++	++			++	+		0.91	0.28	0.80	0.89
	BQI	++	++	++			++	++	++		0.20	0.62	0.75
	AMBI											0.16	0.29
	M-AMBI	++	+				+		++	+			0.94
	EQR	++	++	+			++	+	++	++		++	

+ $p < 0.05$, ++ $p < 0.01$, two-tailed correlation.
Acronyms for corresponding all parameters and criteria are as in Table S7.

Table S10. Summary of concentrations of metals in sediments of Masan Bay reported previously.

Sampling year	Cr	Co	Ni	Cu	Zn	Pb	Hg	References
	(mg kg $^{-1}$)						(ug kg^{-1})	
1998	68.0	14.0	32.0	97.0	360.0	67.0		Ryu et al., 2016
2002	73.3			60.3	263.7	51.3		Woo et al., 2007
2005	67.1	11.5	28.8	43.4	206.3	44.0		Hyun et al., 2007
2006	79.8		29.6	75.1	314.5	66.5		Cho et al., 2015
2010		14.8	32.4	25.3	218.3	48.4	109.1	Lim et al., 2013
2010	71.4	13.4	32.5	39.4	232.0	55.4		Ra et al., 2013

Supplementary Figures

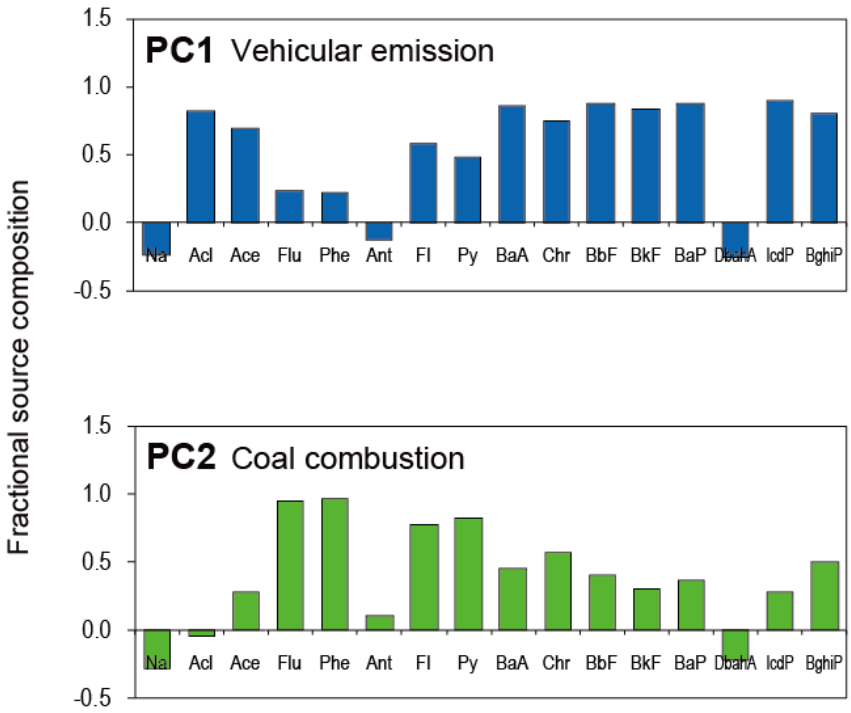

Fig. S1. PCA after Varimax rotation for selected PAHs in Masan Bay sediments.

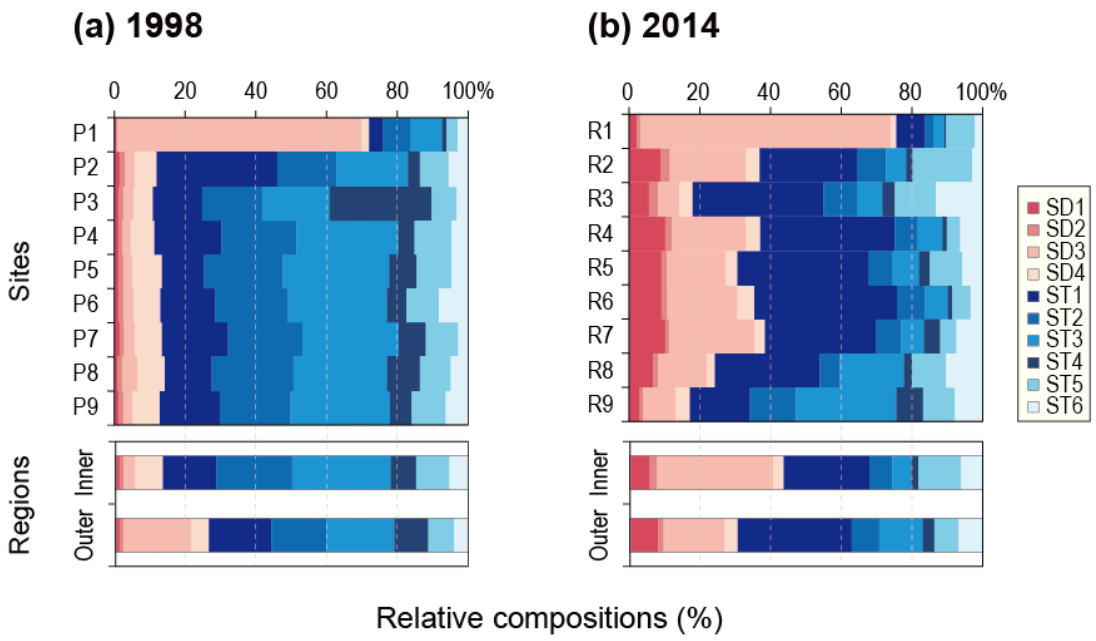

Fig. S2. Comparison of the relative SOs compositions (10 component SOs) at each sampling site and at inner-region versus outer-region sites within Masan Bay.

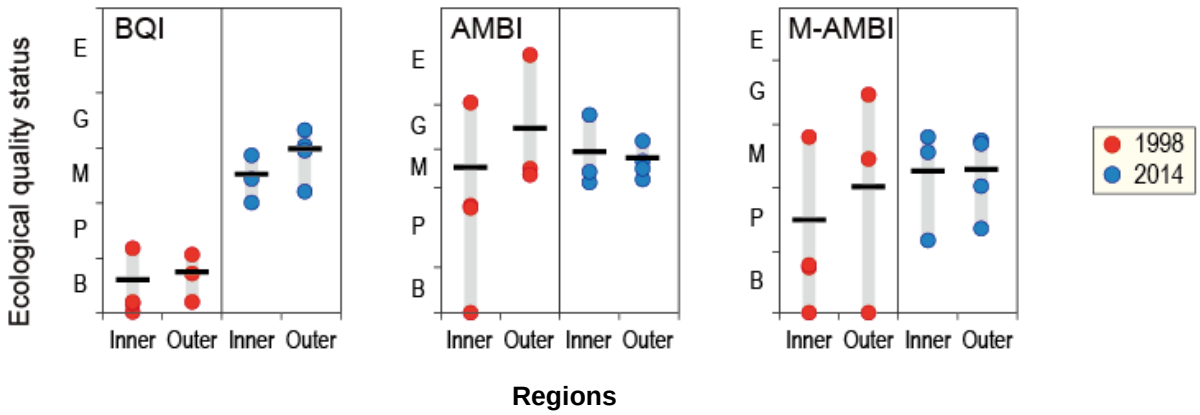

Fig. S3. Ecological quality (EcoQ) status as indicated by Benthic Quality Index (BQI), Azti Marine Biotic Index (AMBI), and Multivariate-AMBI (M-AMBI) indices.

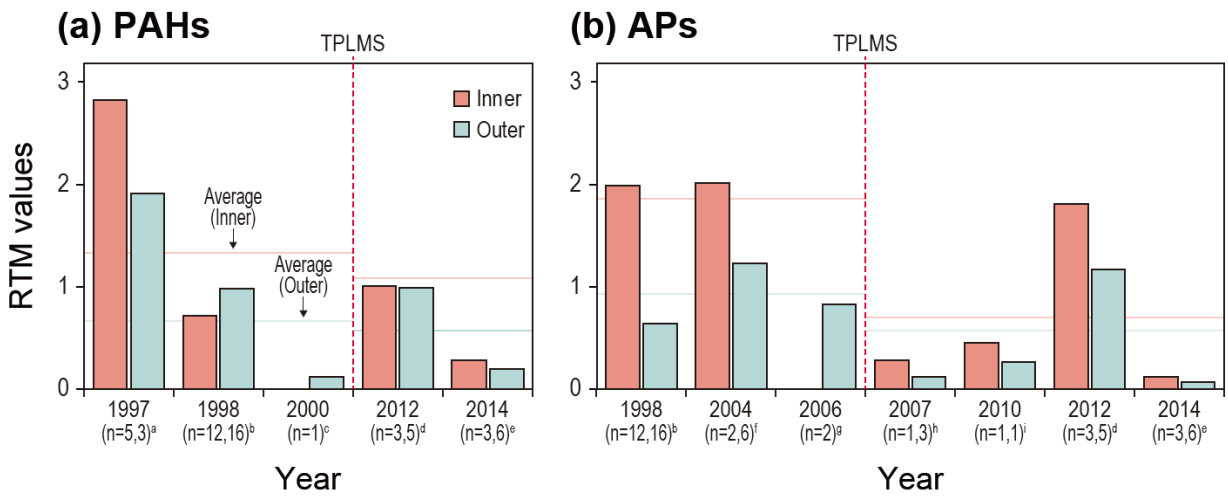

Fig. S4. Comparisons of normalized RTMVs for (a) PAHs and (b) APs in sediments of inner and outer regions of Masan Bay obtained from this study and previous studies (Data from [a] Yim et al., 2005; [b] Khim et al., 1999a; [c] Moon et al., 2001; [d] Jung et al., 2012; [e] This study; [f] Li et al., 2008; [g] Hong et al., 2009; [h] Choi et al., 2009; and [i] Al-Odaini et al., 2005).

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