Science of the Total Environment 792 (2021) 148371

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Large-scale sediment toxicity assessment over the 15,000 km of coastline in the Yellow and Bohai seas, East Asia

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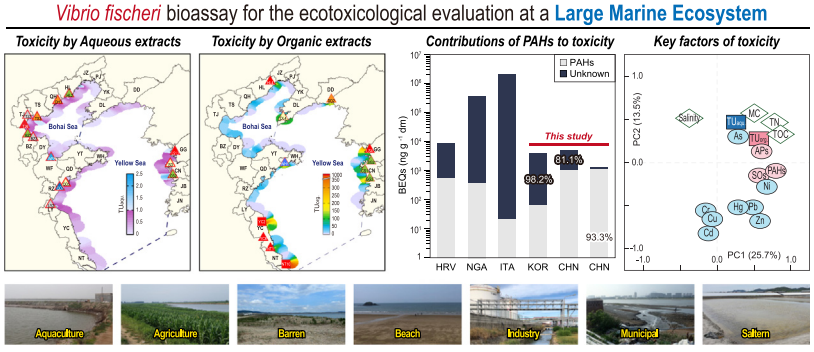
HIGHLIGHTS

- A large scale assessment successfully mapped overall sediment toxicity in Yellow and Bohai seas.
- *Vibrio fischeri* bioassay profiled varied sediment toxicity between aqueous and organic forms.
- Organic toxicity correlated to measured PTSs and reached 1600 TU g^{-1} in Nantong, China.
- Benzo[*a*]anthracene and phenanthrene exhibited a significant contribution to *V. fisheri* toxicities.
- Compared to other countries, *V. fischeri* toxicities of Yellow and Bohai seas were relatively low.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 19 November 2020
Received in revised form 7 June 2021
Accepted 7 June 2021
Available online 10 June 2021

Guest Editor: Wenhua Liu

Keywords:
Marine environment
In vitro bioassay
Bioluminescent bacteria

ABSTRACT

The Yellow and Bohai seas have long been contaminated by persistent toxic substances (PTSs) from numerous (un)known anthropogenic sources. In this study, we used *Vibrio fischeri* bioassay to evaluate ecotoxicological profiles associated with sedimentary PTSs contamination at a large marine ecosystem (LME) scale. A total of 125 surface sediments collected from the coastal areas of the Yellow and Bohai seas were analyzed both for aqueous and organic extracts. Not surprisingly, the results indicated site-dependent toxicities, but most sites were identified as non-toxic to *V. fischeri*. For aqueous extracts and organic extracts, 13% and 8% of samples, respectively exhibited marginal toxicity, while 0% and 2% of samples exhibited moderate toxicity. However, it should be noted that organic extracts (mean TU = 56) induced stronger toxicities than aqueous samples (mean TU = 0.4). This result generally back-supported the high toxicity potentials associated with sedimentary sink of organic pollutants. Several PTSs measured in the samples indicated a significant contribution to the observed *V. fischeri* toxicities. Of note, polycyclic aromatic hydrocarbons (PAHs; $r = 0.28$, $p < 0.05$), styrene oligomers ($r = 0.41$, $p < 0.01$), and alkylphenols ($r = 0.38$, $p < 0.05$) showed significant associations to the observed bacterial inhibition.

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https://doi.org/10.1016/j.scitotenv.2021.148371

Persistent toxic substances
Heavy metals

Among PAHs, benzo[*a*]anthracene and phenanthrene exhibited a significant contribution to the observed *V. fischeri* toxicities. Meantime, salinity which reflects the distance from the point sources of land-driven pollutants along the rivers and estuaries in the Yellow and Bohai seas was a key environmental variable representing the sample toxicities. Overall, the present study provides baseline information for evaluating the potential sediment toxicity to implement responsible coastal management at an LME scale, and elsewhere.

1. Introduction

The Yellow Sea (YS) and Bohai Sea (BS) are the semi-enclosed sea of the Korean peninsula and northeast China, encompassing the most rapidly developing industries in East Asia (Khim et al., 2018a, 2018b; Kim et al., 2020; Meng et al., 2017). More than 20 large cities are existing in this region, which supported almost 25% of the Korean and Chinese population, with increasing coastal activities (Lin et al., 2011; MOF, 2015). Anthropogenic toxic substances are introduced into the YS and BS through more than hundreds of small and large rivers and multiple sources and pathways, which have significantly deteriorated the coastal ecosystems. The very region has long been recognized as global hotspots for heavy metals (HMs) (Tian et al., 2020), organic matter (Zhao et al., 2017), and persistent toxic substances (PTSs) (Khim and Hong, 2014; Khim et al., 2018a; Meng et al., 2017; Qin et al., 2011; Tian et al., 2020; Wang et al., 2015; Yang et al., 2020) which pose risks to numerous marine organisms inhabiting the Yellow Sea coastal environments. Consequently, the YS and BS are experiencing severe ecological disturbances, which are becoming more and more prominent (Li et al., 2020).

A series of our coastal pollution studies in the YS and BS since the late 2000s broadly documented the pollution status of sediment, soil, and water. Most recently, we reported the contamination status of some classic environmental pollutants such as HMs (Tian et al., 2020) and typical and/or emerging PTSs, including polycyclic aromatic hydrocarbons (PAHs), styrene oligomers (SOs), and alkylphenols (APs) (Shi et al., 2021; Yoon et al., 2020). These studies successfully demonstrated the sources and site-specific distributions of target pollutants, with respect to land-use type and salinity, in the given region at a LME scale. Meantime, these studies further questioned the ecotoxicological impacts associated with contaminated sediments as the majority of target contaminants in several hotspots far exceeded the reported sediment quality guidelines (Tian et al., 2020; Yoon et al., 2020).

It is generally recommended that sediment toxicity is estimated by both chemical analysis and bioassay, as toxicity has to be defined as a biological response to a particular test exposure (Brack et al., 1999; Chapman et al., 2002; Khim et al., 1999). When the concentrations of the chemicals are below sediment quality guidelines, the harmfulness of individual chemicals would be low. However, contaminants exist as complexes in sediment, reacting individually, or with agents that negatively affect organisms (Khim et al., 1999; Koh et al., 2002). Thus, biological assessment would be necessary to address integrated sample toxicities, particularly for contaminated sediment mixtures. One of the earliest sediment toxicity studies in the Yellow Sea utilized the H4IIE-*luc* bioassay at a regional scale (Hong et al., 2012). We further reported the long-term trends of potential toxicities in sediments along the west coast of Korea, said the eastern part of the Yellow Sea, based on several bioassays (Kim et al., 2020). This study employed three in vitro bioassays, including H4IIE-*luc*, MVLN, and *Vibrio fischeri* bioassay, where an assessment of the long-term associations between chemicals and biological effects was highlighted. H4IIE-*luc* and MVLN bioassays were performed to measure "mechanism-based toxicity (i.e., receptor binding affinity)" of aryl hydrocarbon receptor (AhR) and estrogen receptor (ER)- active chemicals, respectively (Lee et al., 2020). The *V. fischeri* assay, which is related to the energy metabolism of a bacterium, was used for "baseline toxicity" measurement (Jacobs et al., 1993). In agreement, a series of our pollution and ecosystem health studies in the Yellow Sea repeatedly acknowledged the need for multiple assessments of diverse biological effects for contaminated sediments (Lee et al., 2020).

The *V. fischeri* bioassay is a useful tool when evaluating multiple samples, because of its efficiency in terms of speed, economy, and precision (Beg et al., 2001; Long et al., 2002). In addition, it would be more suitable for assessing the potential risk of a wide spectrum of chemicals in environmental samples compared to other in vitro bioassays (Lee et al., 2020; Lee et al., 2017). In particular, *V. fischeri* bioassay would be a benefit to test both aqueous (Demuth et al., 1993) and organic extracts (Yang et al., 2016) simultaneously, when testing the mixture toxicities of sediments. Altogether, the *V. fischeri* bioassay would be appropriate for assessing potential sediment toxicities as screening of the large samples targetting a broad spectrum of environmental chemicals.

In the present study, we first report the potential ecotoxicological effects associated with contaminated coastal sediments in the YS and BS at a LME scale. We specifically aimed to (1) screen and map the potential toxicities of aqueous and organic extracts of coastal sediments, (2) determine the relationships between toxicological and chemical data, (3) calculate the contributions of individual PAHs to total inhibition bioluminescence, and (4) compare our results with those of previous studies to clarify the current status of PTSs contamination in YS and BS. The present work would support and aid our limited understanding and assessment of ecotoxicological effects of chemical pollutions in the YS and BS. Further, it would provide a better management solution towards ecosystem health monitoring in the given LME and elsewhere.

2. Materials and methods

2.1. Study area and sampling

The study area broadly encompasses three major regions in an oceanographic setting, say Yellow Sea of Korea (YSK), Yellow Sea of China (YSC), and BS. YSK encompasses four provinces located along the west coast of Korea; Gyeonggi, Chungnam, Jeonbuk, and Jeonnam, in the southern direction. YSC includes three provinces in China; Liaoning from the northern coast of the Yellow Sea, Shandong, and Jiangsu from the western part of the Yellow Sea. BS encompasses three provinces and one city; western Liaoning, Hebei, Tianjin, and northern Shandong, counterclockwise (Fig. 1). Details on sampling area and locations are provided in Table S1 of the Supplementary Materials, including geographic location, land-use type, salinity, mud contents (MC), total nitrogen (TN), and total organic carbon (TOC). Land-use type was delineated as the main use type of adjacent land at the time of the investigation and was supplemented by our recent publication (Yoon et al., 2020). Of note, the land-use type of the stations and nearby regions includes industrial, municipal, agricultural, beaches, aquaculture, salt, and barren land.

A total of 125 surface sediments were collected using stainless steel devices from freshwater ($n = 31$), brackish ($n = 63$), and seawater ($n = 31$) locations along the coasts of the YS and BS in June–July 2018 (Fig. 1). At each site, the top 2 cm of sediments, which could reflect current contamination status (Qiao et al., 2017), were placed in precleaned glass bottles and transported immediately to the laboratory and stored at −20 °C until further analysis (Yoon et al., 2020).

2.2. Sample preparation for two extracts

Sediments were extracted by two methods; one for aqueous extraction and the other for organic extraction. Aqueous extraction was carried out in accordance with the standard method of the Ministry of

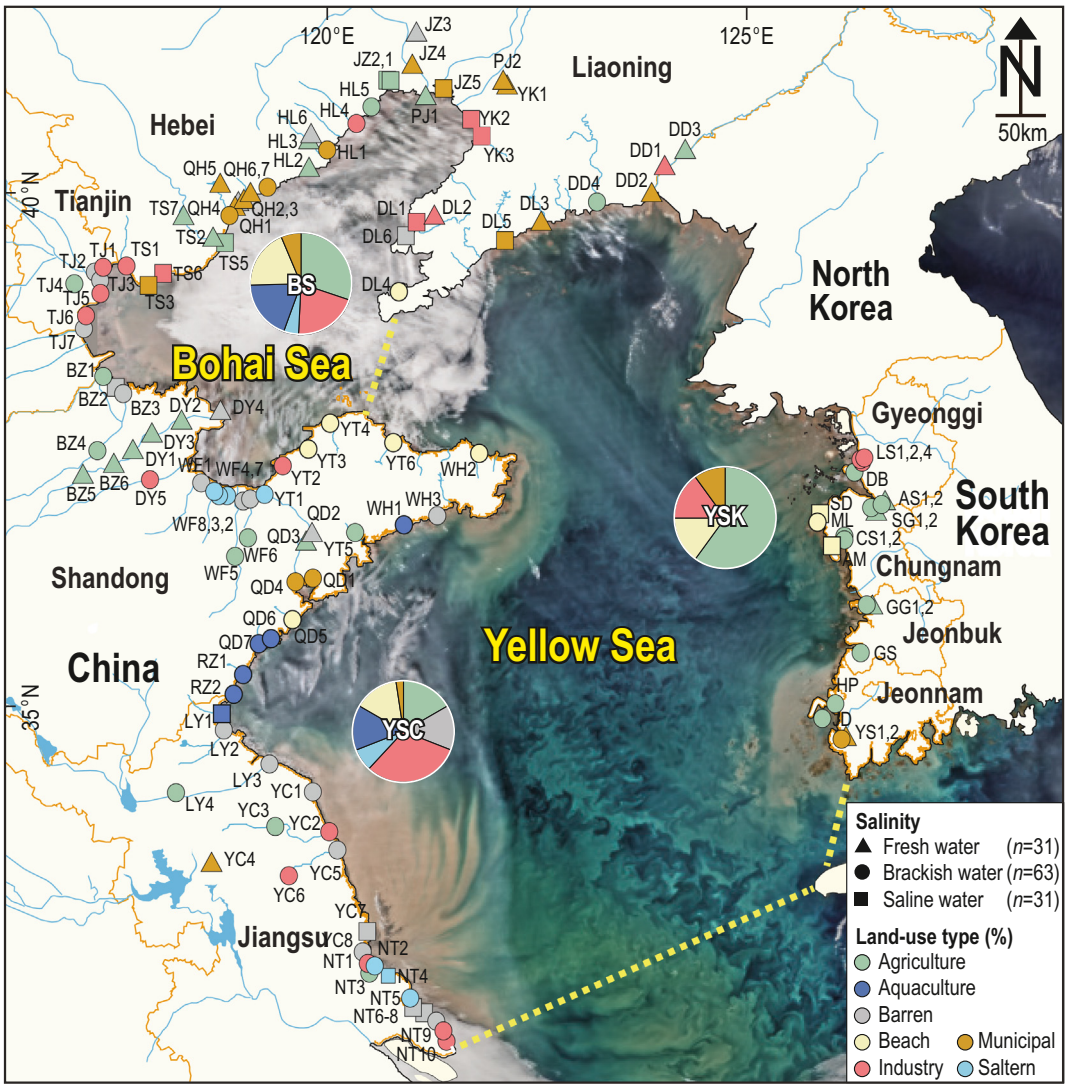

Fig. 1. Map showing the sampling sites in the Yellow and Bohai seas (Total = 125 sites). Land use types were classified as "Agriculture", "Aquaculture", "Barren", "Beach", "Industry", "Municipal", and "Saltern" based on dominant surrounding activity. Sites in the Yellow Sea of Korea (YSK) included Lake Sihwa (LS1, 2, 4), Daebu (DB), Asan (AS1, 2), Sapgyo (SG1, 2), Sinduri (SD), Manlipo (ML), Anmyeon (AM), Cheonsu (CS1, 2), Geumgang (GG1, 2), Gomso (GS), Hampyeong (HP), Jeungdo (JD), and Yeongsan (YS1, 2) (n = 20). Sites in the Yellow Sea of China (YSC) included Dandong (DD1–4), Dalian (DL3, 5), Lianyungang (LY1–4), Nantong (NT1–10), Qingdao (QD1–7), Rizhao (RZ1, 2), Weihai (WH1–3), Yangcheng (YC1–8), and Yantai (YT5, 6) (n = 42). Sites in the Bohai Sea (BS) included Binzhou (BZ1–6), Dalian (DL1, 2, 4, 6), Dongying (DY1–5), Huludao (HL1–6), Jinzhou (JZ1–5), Panjin (PJ1, 2), Qinhuangdao (QH1–7), Tianjin (TJ1–7), Tangshan (TS1–7), Weifang (WF1–8), Yingkou (YK1–3), and Yantai (YT1–4) (n = 63). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Maritime Affairs and Fisheries of South Korea (MOF, 2018). Due to differences in the sampling amount, 119 samples were performed by aqueous extraction. In brief, aqueous extracts were prepared by mixing 3.0 g of wet sediment with 30 mL of a proper diluent solution (buffered K_2HPO_4–KH_2PO_4 0.1 M in 2% NaCl) with magnetic stirring at 200 rpm for 6 h. The supernatant was separated by centrifugation (2000 rpm for 3 min), followed by filtration through 1.0 μm filters. The set of aqueous extracts were used to determine the concentrations of HMs in sediments along with bacterial toxicity testing.

Detailed procedures for sample preparation of organic extracts (OEs) have been reported previously (Hong et al., 2016; Khim et al., 1999; Lee et al., 2019a). Approximately 30 g of freeze-dried sediments were extracted for 16 h on a Soxhlet extractor with 300 mL dichloromethane (DCM) (Burdick & Jackson, Muskegon, MI). Elemental sulfur in OEs was removed by adding activated copper (Sigma Aldrich, Saint Louis, MO) for 1 h. The extracts were concentrated to 4 mL with a rotary evaporator, followed by a gentle stream of nitrogen gas (~10 g sediment equivalent (SEq) mL^{-1}). OEs were divided into two aliquots (3 mL and 1 mL) for bioassay and chemical analyses, respectively. For the bioluminescence inhibition bioassay, the solvents of OEs were subsequently exchanged to dimethyl sulfoxide (DMSO, Sigma-Aldrich).

2.3. Bioluminescence inhibition bioassay

The toxicity of aqueous and organic extracts of sediments was measured based on the decrease of bioluminescence of the marine bacteria *V. fischeri* (NRRL-B-11177), which reflects inhibition of cellular activity. The *V. fischeri* bioassay was conducted following the standard method by the Ministry of Oceans and Fisheries (MOF) of South Korea (MOF, 2018) and Lee et al. (2019b). Briefly, the freeze-dried luminescent bacteria were supplied and cultivated with reactivation solution (1 mL) from NeoEnBiz Inc. (Bucheon, Korea). N-Tox model 200 (NeoEnBiz Inc.) with the thermostat unit was used for measuring bioluminescence. For OEs, the DMSO concentration was set to 1% for all dilution series, at which level the background DMSO toxicity was negligible. The diluent solution by the same salinity (20 psu) was used in this study. Each experiment consisted of four controls and four replicates. Reference toxicant ($ZnSO_4 \cdot 7H_2O$) solution was used to ensure validity across all

tests (ISO, 1998). The inactivation ratio was determined by the inhibition of bioluminescence after 30 min exposure to the sample solution, and the EC50 values with 95% confidence limits were calculated from the results by the dilution series of exposures. The details on bioassay procedures are presented in Table S2.

2.4. Chemical analyses

Our previous studies reported the concentrations of HMs and PTSs (PAHs, SOs, and APs) in the same sediments which *V. fischeri* bioassay was performed (Tian et al., 2020; Yoon et al., 2020). Details of procedure (including QA/QC) and instrumental conditions for PTSs analyses were provided in Table S3. The concentrations of chemicals reported in the previous studies are summarized in Table S4, and compared with the observed toxicities from the present study. Meantime, we selected 50 of the total 125 stations based on their regional composition and concentrations of HMs in the sediment to analyze soluble HM concentrations in aqueous extracts, which have not been measured previously (Table S5). To prevent the concentrations of HMs in the aqueous extracts from changing, the aqueous extracts for HMs analysis were treated with nitric acid (HNO_3), stored at a pH below 2, and analyzed within two weeks using Inductively Coupled Plasma Mass Spectrometry (ICP/MS) equipment following the method described elsewhere (Kim et al., 2020).

2.5. Data analyses

Bioluminescence inhibition of *V. fischeri* was calculated as the median effect concentration (EC50) with 95% confidence limits using the log-linear model. EC50 is the concentration of sediments or sediment solvents representing 50% bioluminescence inhibition compared to the control. EC50 was expressed as mg wet sediment per mL aqueous extracts for aqueous extraction, and μl DMSO per mL solvent for organic extraction. The toxicity criteria of the EC50 for aqueous extracts and organic extracts were separated into four levels based on Bombardier and Bombardier and Bermingham (1999). EC50 was replaced by a toxic unit (TU), which was calculated based on the EC50, to obtain an intuitive understanding of how bioluminescence inhibition increases with an increasing level of risk. The TU was used for the data analysis (Eq. (1)) (Rosado et al., 2016). If EC50 could not be measured, due to low bioluminescence inhibition caused by sediment extracts, TU was set as zero. The toxicity criteria range of TU for aqueous extracts and organic extracts are presented in Table S6.

$$\text{Toxic unit (TU)} = 1/\text{EC50} \times 100 \quad (1)$$

Significant differences in toxicity with respect to region, salinity, and land-use type were identified by the Kruskal-Wallis test and Mann-Whitney test using SPSS 24.0 (SPSS Inc., Chicago, IL). The relationships between toxicity and concentrations of HMs, PTSs, and environmental variables were determined by performing the Spearman correlation analysis using SPSS 24.0. To interpret the correlation analysis results comprehensively, principal components analysis (PCA) was performed with PRIMER 6. PCA is an analytical method that uses correlations between variables to present information of multiple variables that are correlated with each other as a comprehensive characteristic of each variable without significant loss.

2.6. Potency balance analysis

To determine the contribution of detected PAHs to observed *V. fischeri* toxicity, potency balance analysis was performed between bioassay-derived equivalent concentrations (BEQs) and instrument-derived EQs (CEQs) (Hu et al., 2015; Neale et al., 2020). EC50 of the maximum level achieved from previous studies were used for 10 PAHs [naphthalene (Na), acenaphthylene (Acl), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Fl), benzo[*a*]anthracene (BaA), chrysene (Chr), and benzo[*a*]pyrene (BaP)] (Jacobs et al., 1993; Lee et al., 2013). A geometric mean of the reported EC50s for the *V. fischeri* toxicity of individual PAHs was calculated (Table 1). BaA showed the lowest EC50 indicating that the BaA would likely be a major toxicant; BaA was chosen as the standard compound to quantify toxic potency of the samples in the present study. BEQs of two extracted samples were calculated relative to BaA (BEQ) by dividing the concentration of BaA that caused 50% inhibition of bioluminescence of *V. fischeri* by the volume of extracts of samples that produced equivalent inhibition bioluminescence (50%) of *V. fischeri*. The relative potency values (RePs) of each chemical were calculated by dividing the concentration of BaA, which produced 50% inhibition of bioluminescence of *V. fischeri* by concentrations of individual compounds that caused 50% inhibition of bioluminescence (Hu et al., 2015). CEQs were calculated as the sum of measured concentrations for individual compounds in sediment multiplied by their RePs. The RePs of 10 PAHs were listed in Table 1.

3. Results and discussion

3.1. Toxicity of aqueous extracts of sediments

Aqueous extracts of sediments generally showed low bacterial toxicity across sampling stations with 0–2.5 TU (mean = 0.4 TU) (Fig. 2 and Table S7). Among 119 samples tested, 87% were "non-toxic" ($n = 104$), 13% were "marginally toxic" ($n = 15$), and no samples were "moderately toxic" or "highly toxic" (Bombardier and Bermingham, 1999). Significant toxicity was primarily detected in the inner part of the BS, parts of the YSK (Gyeonggi and South Chungcheong provinces), and parts of the YSC (Qingdao, QD; Lianyungang, LY) (Fig. 2a). In general, the *V. fischeri* toxicity data of aqueous extracts did not differ statistically across the region, salinity, and land-use type even though BS contained stations with higher toxicity (Fig. 2a, b). Meantime, high variation between sampling sites was observed (Fig. 2). The high site-specific variations in bacterial toxicities could be simply explained by several hotspot stations. For example, the maximum toxicity of 2.5 TU was measured in QH3 (Qinhuangdao), which is the freshwater and municipal site located in BS. This was followed by CS2 (Cheonsu), which is brackish water and agricultural site, located in the YSK. At present, large spatial variations in sedimentary aqueous toxicity found in the Yellow and Bohai seas could be combined effects of multiple environmental conditions. In turn, the results might indicate the aqueous fraction of sediments could be potentially toxic regardless of certain specific drivers in the lotic system (van Beelen, 2003).

In general, the *V. fischeri* bioassay has been known to have great sensitivity in detecting the toxic levels of metal present in elutriates, although the results are contrary (Codina et al., 2000; Dutka and Kwan, 1981). One possible explanation for the lacking inhibition in *V. fischeri* bioluminescence is that the sediments from the Yellow and Bohai seas do not reach metal contaminants to such a degree that the aqueous extracts become toxic enough (Hagner et al., 2018). It should be noted that some stations showed relatively low toxicity, even though HMs concentrations in sediments exceeded the corresponding Threshold Effect Levels (TELs) (Fig. S1 and Table S4) (Tian et al., 2020). In favor of this hypothesis are the results which showed a low extractability when highly polluted the sediment. This is because only a small part of the total metals was detected in aqueous extracts (Table S5). Bioavailable metal concentrations in aqueous extracts are significantly lower than those of sediment ones. Of note, the concentrations of six HMs in aqueous extracts, except for Cu and Ni, were below the criterion continuous concentration (CCC) (Fig. S2) (US EPA, 2009). Thus, *V. fischeri* toxicity above marginal levels in this study appears to be due to the influence of other water-soluble substances or environmental variables, not due to HMs. Another possible explanation is that it is called a hormetic response, a phenomenon known to cause a stimulatory

Table 1
Concentrations and relative potency values for bioluminescence inhibition of PAHs and results for potency balance analysis between instrument-derived CEQs and bioassay-derived BEQs in the sediments of Yellow and Bohai seas.

Target compounds	Abbreviations	EC 50[a] (mg L^{-1})	EC 50[b] (mg L^{-1})	Mean values	ReP values
Naphthalene	Na	0.68	0.53	0.61	0.43
Acenaphthene	Ace	0.74	0.44	0.59	0.44
Acenaphthylene	Acl	0.31	–[c]	0.31	0.84
Fluorene	Flu	3.23	0.73	1.98	0.13
Phenanthrene	Phe	0.53	0.20	0.36	0.72
Anthracene	Ant	33.40	n.c.[d]	33.40	0.01
Fluoranthene	Fl	2.02	0.37	1.20	0.22
Pyrene	Py	–	n.c.	–	n.c.
Benzo[*a*]anthracene	BaA	0.26	n.c.	0.26	1.0
Chrysene	Chr	1.49	–	1.49	0.17
Benzo[*a*]pyrene	BaP	8.11	n.c.	8.11	0.03

Potency balance analysis		Yellow Sea (Korea)		Yellow Sea (China)		Bohai Sea (China)	
CEQ concentrations (Instrument-derived equivalents, ng CEQ g^{-1} dm)		2.74–13.51 (7.0)[e]		0.9–10,707 (343)		2.1–1439 (91)	
BEQ concentrations (Bioassay-derived equivalents, ng BEQ g^{-1} dm)		44.7–21,849 (3580)		46.6–41,270 (4735)		48.1–4800 (1148)	
Contributions (CEQ/BEQ × 100 (%))		0.02–9.4 (1.8)		0.09–246 (18.9)		0.30–1991 (93.3)	
Top 3 chemicals with great contribution							
	1st	Na	1.3%	BaA	7.1%	BaA	47.7%
	2nd	Phe	0.3%	Phe	6.4%	Phe	15.6%
	3rd	Fl	0.1%	Fl	1.9%	Chr	14.5%

[a] Data from Jacobs et al. (1993).
[b] Data from Lee et al. (2013).
[c] No data.
[d] n.c.: Not calculated.
[e] Min–Max (Mean).

response compared to the control in the presence of sub-inhibitory levels of toxicants (Christofi et al., 2002; Lee et al., 2020).

Many HMs in the sediment exists as insoluble fractions and/or remain strongly adsorbed on sediments. Consequently, HMs would have low bioavailability and minimally impact organisms (Ocampo-Duque et al., 2008). Altogether, the toxicity result of aqueous extracts produced in batch might not adequately reflect the sediment composition and/or concentration of the site under investigation. For this

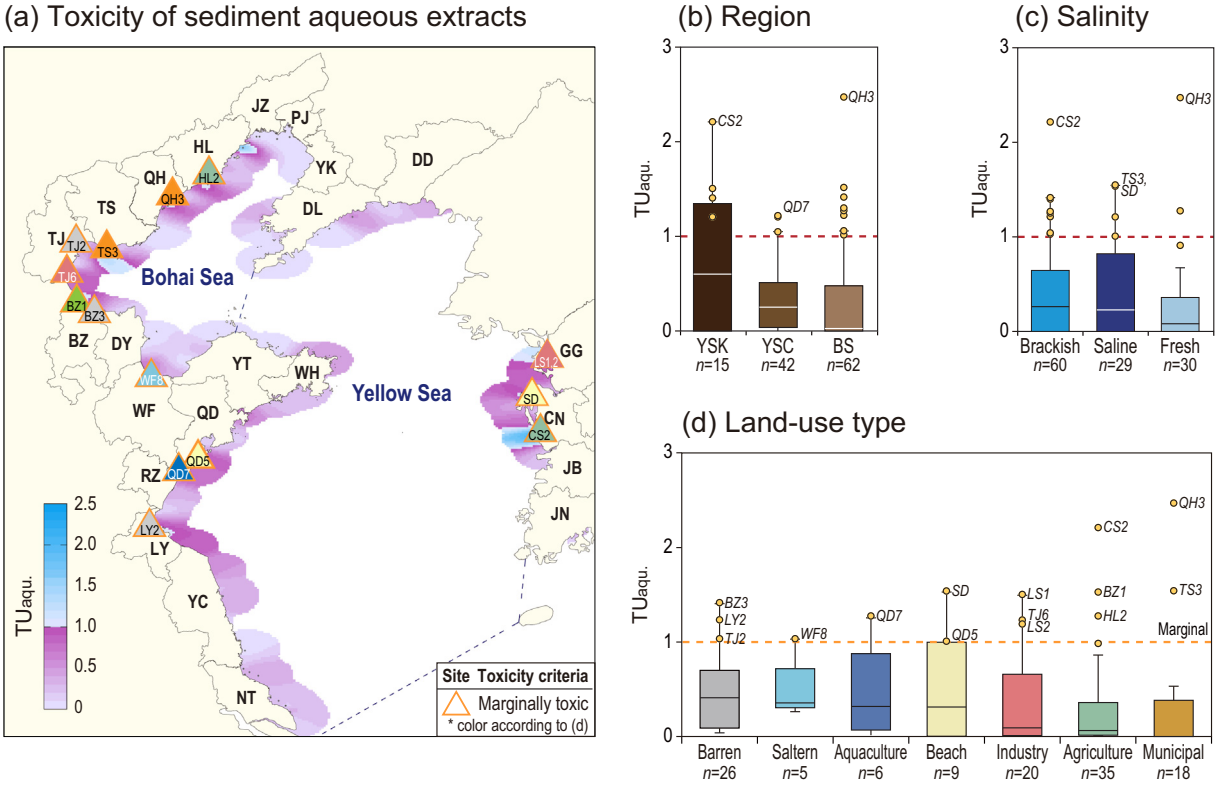

Fig. 2. Bioluminescence inhibition by *Vibrio fischeri* for the aqueous extracts of sediments (a) in the Yellow and Bohai seas with respect to (b) region, (c) salinity, and (d) land-use type. Panels: (b) Yellow Sea of Korea (YSK), Yellow Sea of China (YSC), and Bohai Sea (BS). Lower than 1 toxic unit (TU) is "non-toxic", greater than 1 TU is "marginally toxic", greater than 10 TU is "moderately toxic", and greater than 100 TU is "highly toxic"; however, no moderately toxic or highly toxic samples were detected for aqueous extracts in this study. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

reason, when applied spearman correlation analysis, no statistically significant relationship between soluble metals and TU was shown ($p > 0.05$) (Fig. S2). However, when correlation analysis was performed with the reviewed results in the same study area without converting to TU, the luminescence inhibition showed a significant correlation with concentrations of Cd, As, and Hg (Fig. S3). It was confirmed that the toxicity values as the detection of stimulatory responses at lower doses result in an atypical concentration-response curve, and EC50 value did not show the accurate estimation of toxicity (Davoren et al., 2005). In other words, in cases with relatively low toxic effects, this implies that inhibition rates are reasonable compared to TU.

3.2. Toxicity of organic extracts of sediments

The result of toxicity testing for the organic extracts of sediments, compared to aqueous data, revealed distinct spatial distributions and patterns across regions, salinity, and land-use type (Fig. 3). First, the toxicity generally increased with a maximum of 1600 TU (mean = 56 TU) (Fig. 3 and Table S7) that surpasses the toxicity criteria of Environment Canada (2002), viz., TU > 100. However, the proportion of "non-toxic" samples (90%; $n = 113$) for organic extracts was similar to that for aqueous ones. Next, 7.5% were "marginally toxic" ($n = 10$), 1.7% were "moderately toxic" ($n = 2$), and no samples were classified as "highly toxic". By region, great toxicity was detected at Gyeonggi and South Chungcheong provinces in YSK, Dandong (DD), Weihai (WH), Yangcheng (YC), and Nantong (NT) in YSC, and Huludao (HL) and Dalian (DL) in BS (Fig. 3a). Although there was no statistically significant difference in toxicity with respect to the region (Fig. 3b), toxicity criteria of organic extracts in BS are far reduced compared to those of aqueous ones (Fig. 2). In addition, the toxicity of sediment OEs from brackish water stations was significantly different compared to those from seawater stations. Interestingly, most of the *V. fischeri* toxicity in OEs of sediments was found in the stations of brackish water, with the exception of DD2. These results seemed to reflect direct influence from the land-derived pollutants (Abdo Alkhadher et al., 2016). Physical variables, such as water flow, salinity, organic carbon contents, and grain sizes of sediments might also affect the introduction and accumulations of PTSs in coastal sediments. In particular, salinity is a major factor controlling distribution coefficients for organic and inorganic pollutants in the estuarine environments (Chapman and Wang, 2001). Thus, it is not surprising that the toxicity results are strongly associated with the variation of salinity in the study areas. In the present study, the toxicity difference due to the inflow and accumulation of PTSs associated with region-specific physical factors could not be fully considered. Further study is needed to determine physical factors controlling behavior and fate of PTSs in sediments and their potential adverse effects on organisms (Jarque et al., 2016; Romeo et al., 2015).

The examination of bacterial toxicity data for sediment organic extracts by land-use type also clearly demonstrated their close association (Fig. 3d). First of all, industrial and agricultural areas exhibited relatively greater toxicity, followed by municipal and agricultural areas. While, saltern and barren lands showed no significant toxicities, of which aqueous toxicities were the greatest (Fig. 2d and Fig. 3d). The toxicity of industrial areas showed a statistically significant difference from those of beach ($p < 0.01$) and barren ($p < 0.05$) sites. Among the industrial stations, NT10 (Nantong) and YC2 (Yancheng) showed the most toxic responses over 1000 TU (moderately toxic) in this study. Those stations were located in the inland creeks where point sources from industry, agriculture, and domestic wastewaters prevailed. Of note, Yancheng Municipality has strongly promoted the local economy, implementing large aquaculture complexes for decades. Sedimentary pollution with antibiotics in the water column of the very area has also been recently documented (Xie et al., 2020). As for Nantong, large accumulations of PAHs have been documented in recent years of which sources were identified from the large influx of organic materials generated by industrial activities (Yoon et al., 2020).

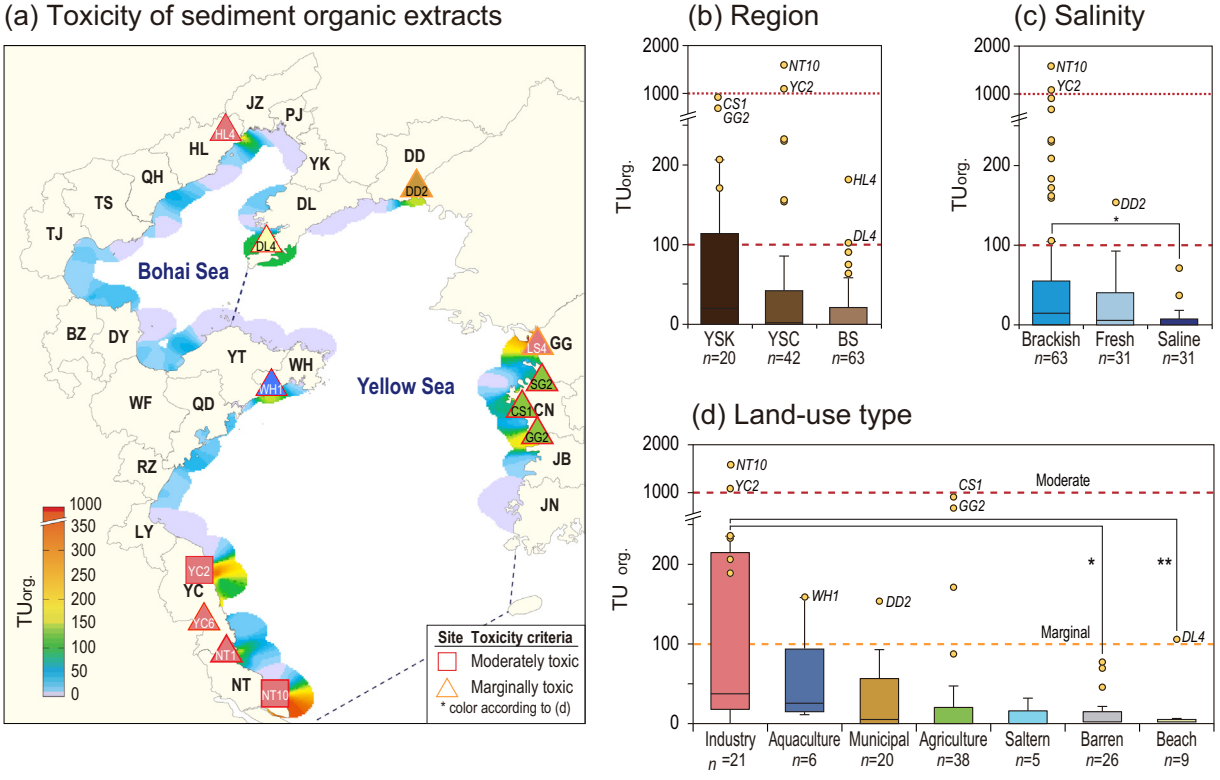

Fig. 3. Bioluminescence inhibition by *Vibrio fischeri* for organic extracts of sediments (a) in the Yellow and Bohai seas with respect to: (b) region, (c) salinity, and (d) land-use type. Lower than 100 toxic units (TU) is "non-toxic", greater than 100 TU is "marginally toxic". (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

It is not surprising that soluble hazardous substances in sediments would lead to lower concentrations remaining in the sediment (Ocampo-Duque et al., 2008). Accordingly, the *V. fischeri* bioassay exhibited greater sensitivity to organic extracts compared to aqueous extracts (Demuth et al., 1993; Grant and Briggs, 2002; Ocampo-Duque et al., 2008). The toxicity difference between aqueous and organic extracts originally, at least partly, comes from the extractible compounds in samples; thus, selection of extraction methods is critical (Vaal et al., 2000; Lee et al., 2020). Besides, such variability could also vary depending on the test organisms; thus, it is recommended to use toxicity test batteries with diverse species at different trophic levels. Finally, multiple endpoints for sediment toxicity testing would be a benefit to consider different exposure routes and species-dependent sensitivities to target toxicants (Chapman et al., 2002; Giesy and Hoke, 1989; Lee et al., 2020).

3.3. Relationship between bacterial toxicity and chemicals

Aqueous samples exhibited very low bacterial toxicities; thus, cause-effect assessment of water-soluble contaminants measured in this study could not be anticipated. While, several classes of PTSs which occurred in sediment organic fractions could be examined for the casual association to the observed toxicities of sediment organic extracts. Significant Spearman's rank correlation analysis was applied to address this specific question targeting PAHs, SOs, and APs. Not surprisingly, there were statistically significant relationships between bacterial inhibition and the measured PTSs in the sediments (PAHs, $r = 0.28$, $p < 0.05$; SOs, $r = 0.41$, $p < 0.01$; APs, $r = 0.38$, $p < 0.05$) (Fig. 4). Among the PTSs examined, *V. fischeri* toxicities were more significantly correlated to concentrations of fluoranthene (Fl), pyrene (Py), 2,4-diphenyl-1-butene (SD3), 4-*tert*-octylphenol (*t*-OP), 4-*tert*-octylphenol monoethoxylate (*t*-OP1EO), and nonylphenol ethoxylates ($p < 0.05$) (Fig. 4 and Table S8).

Potency balance between BEQs and CEQs was assessed to determine chemical-specific contribution to total induced bacterial toxicity targeting the organic extracts of sediments (Table 1). Total CEQs ranged from 0.9–10,707 ng g^{-1} in samples with the greatest mean CEQ of 343 ng g^{-1} in the YSC. Ten PAHs explained maximum 93% of the BEQs in the BS. Thus, the *V. fischeri* toxicity detected in the sediments from BS seemed to be due to traditional PAHs rather than emerging toxic chemicals. The major contributor of *V. fischeri* toxicity in BS was BaA (47.7%), followed by Phe (15.6%) and Chr (14.5%) (Table 1). In YSC, BaA and Phe showed great contributions with 7.1% and 6.4%, respectively. In the case of YSK, the overall toxicity contribution of PAHs was low, and the contribution of Na (1.3%) and Phe (0.3%) was relatively high. Overall, BaA and Phe contributed the most to total BEQ concentrations in the study areas, due to their great RePs and high concentrations in sediments. BaA is known to primarily originate from coal combustion (Kong et al., 2012); thus, this result suggests that the use of coal in China would be a major source of PAHs in coastal environments. Phe primarily originates from vehicle emissions (Ravindra et al., 2008), suggesting that vehicle emission might be a major source of PAHs pollution in both countries.

The results of the potency balance analysis for bacteria toxicity in sediments and sludge from various countries were compared (Fig. S4). Although Phe and BaA generally contributed a large proportion, its degree varied greatly among regions. Of note, toxicity varied depending on the sample matrices and types of the surrounding environment. Overall, the 10 PAHs had relatively minor contributions to the organic extracts of sediments in Nigeria, Italy, and YSK. Thus, it is needed to identify the causative chemicals in sediments whether PAHs lined toxicity is associated with site-specificity and/or matrix-dependency in the future.

We further tried to identify the key environmental factors that influence the sediment toxicities on *V. fischeri*. PCA was performed using all the data available from the present and previous works (Yoon et al., 2020). TU for aqueous and organic extracts, concentrations of soluble metals and PTSs, and environmental variables (salinity, MC, TN, and TOC) were used to determine the principal components or variables (Fig. 5a). The two principal components of PC1 and PC2 explained 25.7% and 13.5% of the total variances, respectively. TU for organic extracts was located in the same quadrant as concentrations of PAHs, SOs, and APs, MC, TOC, and TN, reflecting their associations.

The PCA encompassing both aqueous and organic extracts data seemed to mask their independent associations; thus, separate analyses were further made (Fig. 5b and c). As expected, the toxicity of aqueous extracts is associated with the soluble HMs in sediment (Fig. 5b), and the toxicity of organic extracts is directly linked to the PTSs (Fig. 5c). One particular feature of the PCA between aqueous and organic toxicities would have prevailed salinity impacts that evidenced by a strong negative correlation between salinity and TU_{org}. Also, the impact of land-use types explained the toxicities of sedimentary organic matter and the degree of PTSs contamination with increased power of explanation in PC1 (43.1%) (Fig. 5c). For example, the near-shore/near-source locations YC6, NT10, and HL4 were closely grouped in the score plot (high on PC1, low on PC2). Overall, the PCA results were consistent with the generalized idea of a strong association between the majority of bacterial toxicity and detected concentrations of PTSs in the Yellow and Bohai seas (Yoon et al., 2020). However, the potential biological

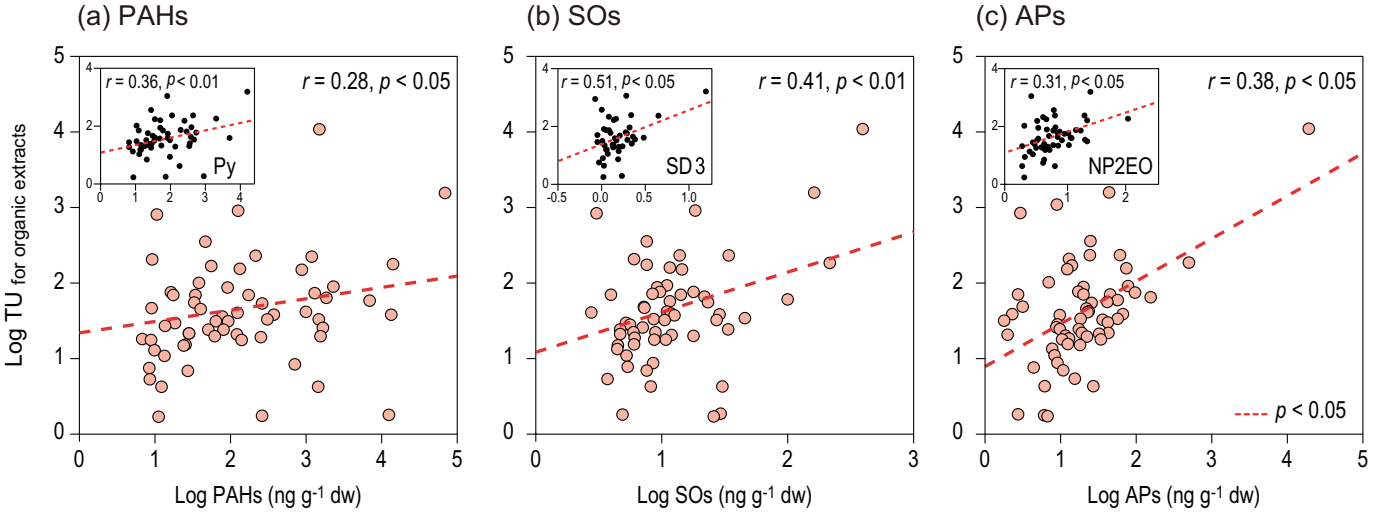

Fig. 4. Spearman rank correlation between the concentration of (a) polycyclic aromatic hydrocarbons (PAHs), (b) styrene oligomers (SOs), (c) alkylphenols (APs) in sediments, and toxic unit (TU) for organic extracts. A concentration below the limit of detection was replaced with a value of half of the limit of detection before statistical analyses were performed.

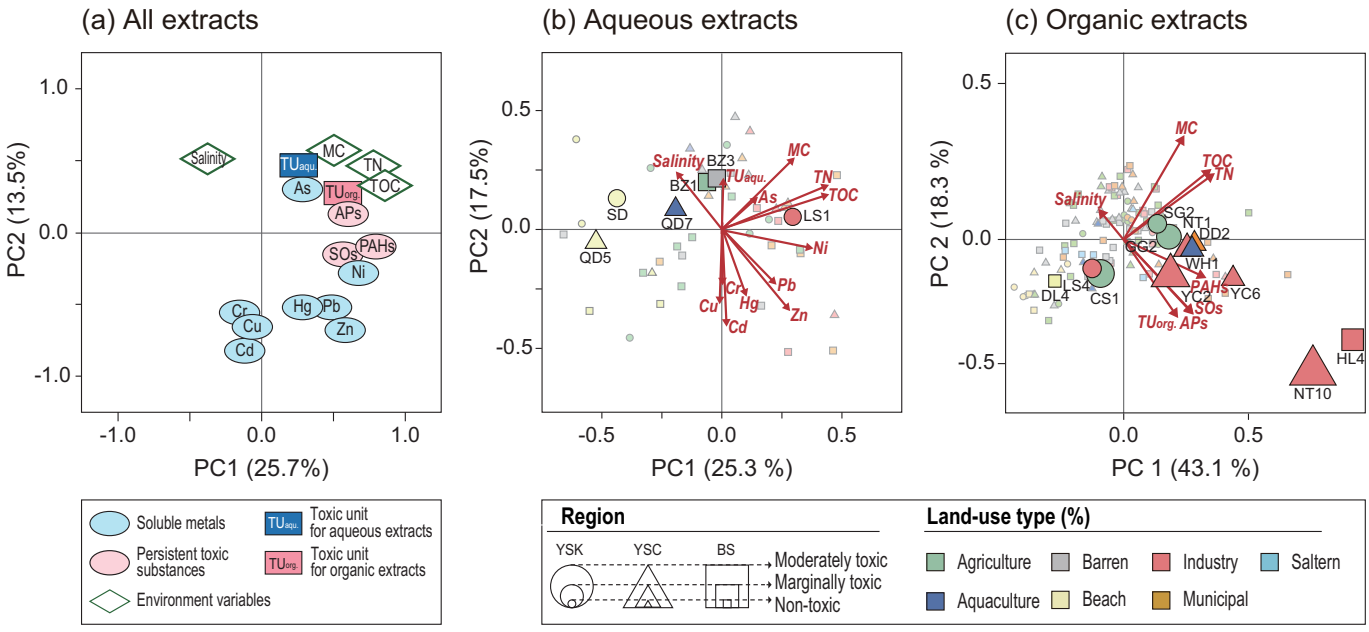

Fig. 5. Principal component analysis (PCA) ordination of (a) concentrations of soluble heavy metals, polycyclic aromatic hydrocarbons (PAHs), styrene oligomers (SOs), and alkylphenols (APs), bioluminescence inhibition for two extracts of sediments ($TU_{aqueous\ extracts}$ and $TU_{organic\ extracts}$), and four environmental variables (salinity; total organic carbon, TOC; total nitrogen, TN; mud contents, MC). (b) PCA for soluble heavy metals, $TU_{aqu.}$ and four environmental variables. (c) PCA for PAHs, SOs, APs, $TU_{org.}$ and four environmental variables.

effects of organic extraction on inorganic pollutants, such as hydrogen sulfide and ammonia in the sediments, were not considered.

3.4. Comparison with previous studies

Several previous studies have successfully utilized the *V. fischeri* bioassay for the assessment of potential toxicity associated with contaminated sediments. As for a solid comparison between our results and the previous works, we analyzed the reported *V. fischeri* toxicity data of which studies assessed either aqueous and/or organic extracts of sediments (Table S9). While, it should be noted that the previous studies examined limited localities focusing on specific sites. Also, each study focused on either aqueous or organic extracts of sediments; thus, potential toxicity cross two phases or possible combined effects were not well addressed (van Beelen, 2003).

For aqueous extracts, Rosado et al. (2016) conducted a study in two estuaries in Spain, Odiel in the west and Tinto in the east, finding "highly toxic" to *V. fischeri* with a maximum of 8700 TU, although littoral samples showed relative low toxicity compared to those in estuarine samples (Table S9). In contrast, potential toxicities of sediments collected from the coastal areas of the YS and BS were noticeably low, by two to five orders of magnitude, compared to those reported in Spain, except Wang et al. (2009). The study found stronger correlations between bacterial toxicity and bioavailable metals, especially pyritic metals such as Zn, Cu, Cd, Pb, and Fe, which were originated from inland mining activities. Meanwhile, the TU values found in the Yellow and Bohai seas (mean = 68 TU), on average, were greater than those of the Atlantic coast of Spain (mean < 0.01 TU) (Table S9).

For organic extracts, bioluminescence inhibition of industrialized coastal sediments from Ulsan Bay, Masan Bay, and Siheung in Korea has been documented (Table S9). These coastal areas have long been recognized as coastal pollution hotspots in Korea contaminated by numerous environmental contaminants, including HMs and PTSs (Khim and Hong, 2014). The highest average toxicity was found in sediments from Masan Bay, followed by the Siheung and Ulsan (Choi et al., 2010). The adverse effects on bioluminescent bacteria from Masan were lower in 2016 compared to 2006. Samples collected in 2006 from Siheung showed toxicity levels comparable to those of polluted sites in Masan and exceeded toxicity thresholds suggested by Environment Canada (2002). For example, bioluminescence inhibition at Nantong (NT1) and Yancheng (YC6) of the present study (235 TU and 232 TU, respectively) was within a similar range compared to those of Yulin, China (min. 230 TU), being marginally toxic. Meantime, bioluminescence inhibition at Nantong (NT10) and Yancheng (YC2) of this study (1600 TU and 1100 TU, respectively) was similar to the maximum bioluminescence inhibition reported in Masan (909 TU). The minimum bioluminescence inhibition in Ulsan Bay (18 TU) was similar to those of Qinhuangdao (QH7) and Weifang (WF5) in the BS. Overall, toxicity values for organic extracts found in the present study were similar to or generally lower than those in other polluted sediments around the world. Dramatic reduction in concentrations of PTSs in sediments was associated with a decrease in toxicity of extracts for *V. fischeri*, although unexplained portions remained. Therefore, continuous monitoring of environmental contaminants, especially for various emerging PTSs, would be needed for better and appropriate management and environmental regulation in the future.

4. Conclusions

V. fischeri bioassay has been successfully utilized on screening and toxicity mapping at a LME scale in the Yellow and Bohai seas. The bioluminescence inhibition of *V. fischeri* fairly well reflected the type and degree of sedimentary PTSs contaminations. Relatively high sediment toxicities were observed in the stations of brackish water, which might attribute to land-driven pollutants. When compared with previous studies in other countries, the Yellow and Bohai seas showed relatively low potential toxicity. The results of potency balance analysis revealed that the toxicity contribution of individual PAHs was different in the YSK, YSC, and the BS. Major toxic PAHs with great contributions were found to be Na in YSK, BaA and Phe in YSC, and BaA in BS. Overall, results of the present study could be used as useful baseline data for prioritizing effect-based chemicals of concern in the coastal sediments of the Yellow and Bohai seas. Further tests such as toxicity identification evaluations would be beneficial to advance compound-specific management in the watershed and estuarine environments of the Yellow and Bohai seas.

CRediT authorship contribution statement

Kyuwon Hwang: Conceptualization, Formal analysis, Statistical analyses, Visualization, Writing – original draft. **Junghyun Lee**: Conceptualization, Formal analysis, Visualization, Writing – original draft. **Inha Kwon**: Investigation, Formal analysis. **Shin Yeong Park**: Data review, formal analysis. **Seo Joon Yoon**: Investigation. **Jongmin Lee**: Investigation. **Beomgi Kim**: Investigation. **Taewoo Kim**: Investigation. **Bong-Oh Kwon**: Writing – review & editing, Project administration. **Seongjin Hong**: Conceptualization, Writing – review & editing. **Moo Joon Lee**: Investigation, Formal analysis. **Wenyou Hu**: Investigation, Project administration, Funding acquisition. **Tieyu Wang**: Investigation, Project administration, Funding acquisition. **Kyungsik Choi**: Investigation, Project administration. **Jongseong Ryu**: Investigation, Project administration. **Jong Seong Khim**: Writing – review & editing, Project administration, Funding acquisition, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

This work was supported by the projects entitled “Development of Advanced Science and Technology for Marine Environmental Impact Assessment (grant number 20210427)” and “Ecosystem-Based Analysis and Decision-Making Support System Development for Marine Spatial Planning (grant number 20170325)” funded by the Ministry of Oceans and Fisheries of Korea (MOF), South Korea granted to Jong Seong Khim. This study was also supported by the Scientific Program for International Cooperation of Guangdong Province between China and Korea (grant number 2021A0505030071).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.148371.

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Supplementary materials for

Large-scale sediment toxicity assessment over the 15,000 km of coastline in the Yellow and Bohai seas, East Asia

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This PDF file includes:

Number of pages: 21

Number of Supplementary Tables: 9, Tables S1 to S9

Number of Supplementary Figures: 4, Figs. S1 to S4

References

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Supplementary Tables

Table S1. Description of the sample sites and parameters of sediments in the Yellow and Bohai seas.

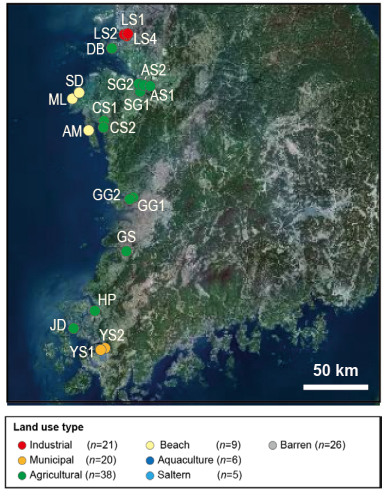
Sampling map	Region	Site	Latitude (°N)	Longitude (°E)	Land-use type	Salinity (‰)	Mud contents (%)	TN (%)	TOC (%)
	***Yellow Sea (Korea)***								
	Anmyeon	AM	36.5401	126.3265	Beach	30.5	0.00	0.005	0.03
	Asan	AS1	36.8933	126.9123	Agricultural	0.2	0.00	0.031	0.02
		AS2	36.9154	126.9052	Agricultural	24.9	75.65	0.106	0.66
	Chenosu	CS1	36.5981	126.4632	Agricultural	1.5	2.63	0.041	0.43
		CS2	36.2142	126.5355	Agricultural	29.6	63.14	0.073	1.04
	Daebu	DB	37.2142	126.5855	Agricultural	22.5	61.31	0.107	1.06
	Geumgang	GG1	36.0225	126.7422	Agricultural	0.1	92.87	0.083	1.31
		GG2	36.0085	126.7353	Agricultural	12.7	89.44	0.104	1.60
	Gomso	GS	35.5728	126.6636	Agricultural	24.9	84.97	0.063	0.71
	Hampyeong	HP	35.0890	126.3538	Agricultural	31.3	1.13	0.043	0.82
	Jeungdo	JD	34.9690	126.1662	Agricultural	32.3	98.72	0.043	1.04
	Lake Sihwa	LS1	37.3348	126.6895	Industrial	22.2	83.39	0.107	0.80
		LS2	37.3257	126.6571	Industrial	28.6	78.64	0.128	1.38
		LS4	37.3249	126.6556	Industrial	28.8	30.84	0.018	0.13
	Manlipo	ML	36.7838	126.1364	Beach	31.2	0.00	0.003	0.02
	Sinduri	SD	36.8385	126.1834	Beach	31.3	1.80	0.028	0.10
	Sapgyo	SG1	36.8788	126.8272	Agricultural	0.2	0.00	0.002	0.05
		SG2	36.8951	126.8191	Agricultural	27.2	86.09	0.145	1.00
	Yeongsan	YS1	34.7821	126.4441	Municipal	26.7	95.72	0.050	0.98
		YS2	34.7866	126.4627	Municipal	0.2	89.80	0.050	1.14

Table S1. Continued.

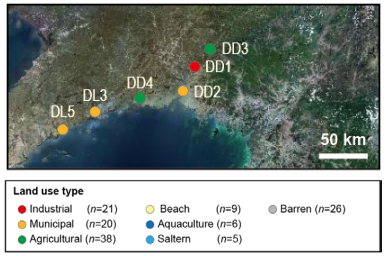
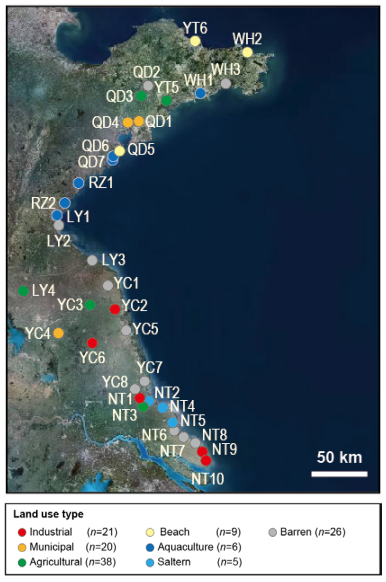
Sampling map	Region	Site	Latitude (°N)	Longitude (°E)	Land-use type	Salinity (‰)	Mud contents (%)	TN (%)	TOC (%)
	***Yellow Sea (China)***								
	Dandong	DD1	40.1775	124.4469	Industrial	0.1	38.76	0.148	1.76
		DD2	39.9436	124.2828	Municipal	0.4	89.70	0.137	1.80
		DD3	40.3344	124.7136	Agricultural	0.1	16.79	0.082	0.57
		DD4	39.8383	123.6528	Agricultural	9.9	65.26	0.092	0.96
	Dalian	DL3	39.6633	122.9939	Municipal	31.8	91.79	0.144	1.53
		DL5	39.4817	122.5592	Municipal	34.6	86.29	0.102	0.97
	Lianyungang	LY1	34.9023	119.1961	Aquaculture	46.6	60.09	0.039	0.39
		LY2	34.7963	119.2244	Barren	30.7	97.58	0.126	1.57
		LY3	34.5026	119.7720	Barren	33.7	98.42	0.116	0.97
		LY4	34.1537	118.8366	Agricultural	0.5	90.35	0.076	0.75
	Nantong	NT1	32.6031	120.9437	Industrial	2.3	98.60	0.118	1.60
		NT2	32.5577	121.0457	Barren	17.2	85.37	0.070	1.19
		NT3	32.5140	120.9660	Agricultural	1.4	61.58	0.042	0.22
		NT4	32.4919	121.2226	Barren	44.6	0.72	0.012	0.08
		NT5	32.2016	121.3851	Saltern	28.1	86.79	0.074	1.00
		NT6	32.1535	121.4562	Barren	43.1	93.29	0.077	1.12
		NT7	32.1014	121.6039	Barren	44.6	75.57	0.038	0.21
		NT8	32.0292	121.7411	Barren	30.8	95.55	0.075	0.95
		NT9	31.9337	121.8257	Industrial	33.3	92.41	0.066	1.53
		NT10	31.8490	121.8521	Industrial	2.0	77.17	0.071	1.31
	Qingdao	QD1	36.2609	120.3259	Municipal	18.5	95.61	0.124	1.83
		QD2	36.7802	120.4099	Barren	0.5	68.23	0.049	1.65
		QD3	36.2609	120.3259	Agricultural	0.3	1.74	0.136	7.00
		QD4	36.2353	120.1206	Municipal	18.6	84.20	0.054	2.01
		QD5	35.8568	120.0477	Beach	21.2	0.52	0.002	0.02
		QD6	35.7684	119.9262	Aquaculture	4.4	84.42	0.101	1.34
		QD7	35.7405	119.9111	Aquaculture	14.2	1.54	0.014	0.32

Table S1. Continued.

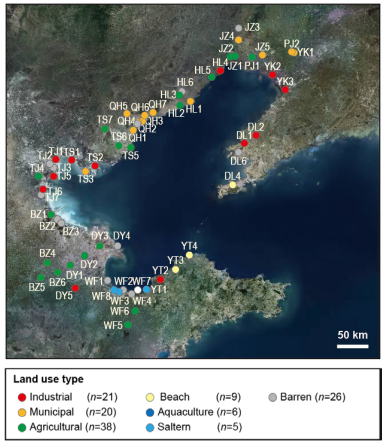
Sampling map	Region	Site	Latitude (°N)	Longitude (°E)	Land-use type	Salinity (‰)	Mud contents (%)	TN (%)	TOC (%)
	Rizhao	RZ1	35.2980	119.4482	Aquaculture	13.8	24.67	0.016	0.11
		RZ2	35.0782	119.3033	Aquaculture	24.3	17.49	0.029	0.15
	Weihai	WH1	36.8266	121.4636	Aquaculture	19.9	86.30	0.161	1.40
		WH2	37.4296	122.2754	Beach	26.9	1.26	0.005	0.03
		WH3	36.9321	121.8657	Barren	20.9	0.00	0.009	0.06
	Yancheng	YC1	34.1128	120.3239	Barren	15.0	78.49	0.055	0.35
		YC2	33.8160	120.4768	Industrial	0.8	90.71	0.075	0.63
		YC3	33.8934	120.0150	Agricultural	0.6	70.16	0.117	0.77
		YC4	33.4793	119.1460	Municipal	0.4	80.65	0.067	0.27
		YC5	33.7400	120.5499	Barren	20.2	65.14	0.023	0.17
		YC6	33.3674	120.0770	Industrial	0.6	69.77	0.198	1.51
		YC7	32.8821	120.9646	Barren	41.7	73.65	0.063	0.78
		YC8	32.6933	120.8959	Barren	4.0	95.41	0.067	0.35
	Yantai	YT5	36.6543	120.7688	Agricultural	1.8	0.00	0.007	0.04
		YT6	37.5753	121.2966	Beach	28.0	8.09	0.007	0.06
	***Bohai Sea (China)***								
	Binzhou	BZ1	38.2637	117.8511	Agricultural	31.6	74.66	0.041	0.49
		BZ2	38.2006	118.0047	Barren	33.3	91.58	0.079	0.66
		BZ3	38.1460	118.0528	Barren	15.7	86.90	0.045	0.28
		BZ4	37.5010	117.8540	Agricultural	0.8	77.83	0.112	2.58
		BZ5	37.2497	117.7231	Agricultural	0.4	90.04	0.030	0.84
		BZ6	37.3350	118.0576	Agricultural	0.3	65.33	0.015	0.06
	Dalian	DL1	39.6208	121.5214	Industrial	34.9	84.78	0.066	1.60
		DL2	39.6947	121.7400	Industrial	0.4	68.33	0.070	1.55
		DL4	38.9844	121.5103	Beach	0.8	2.14	0.008	0.06
		DL6	39.5058	121.4033	Barren	36.0	46.58	0.015	0.16
	Dongying	DY1	37.4851	118.2691	Agricultural	0.5	84.73	0.034	0.51
		DY2	37.6046	118.5384	Agricultural	0.3	25.64	0.007	0.02
		DY3	37.7481	118.8214	Agricultural	0.3	56.38	0.011	0.05

Table S1. Continued.

Sampling map	Region	Site	Latitude (°N)	Longitude (°E)	Land-use type	Salinity (‰)	Mud contents (%)	TN (%)	TOC (%)
		DY4	37.7615	119.1706	Barren	0.5	94.67	0.027	0.19
		DY5	38.1363	118.4322	Industrial	1.6	74.16	0.106	1.56
	Huludao	HL1	40.2697	120.4622	Municipal	33.7	76.60	0.134	2.40
		HL2	40.1747	120.2614	Agricultural	0.2	87.98	0.184	1.73
		HL3	40.3703	120.2583	Agricultural	0.0	89.84	0.115	1.36
		HL4	40.7469	120.9347	Industrial	9.1	90.16	0.341	3.12
		HL5	40.5919	120.7694	Agricultural	7.5	61.06	0.070	1.05
		HL6	40.4192	120.2956	Barren	0.3	0.00	0.062	0.45
		HL6	40.4192	120.2956	Barren	0.3	0.00	0.062	0.45
	Jinzhou	JZ1	40.9242	121.1867	Agricultural	37.5	39.16	0.021	0.48
		JZ2	40.9181	121.2436	Agricultural	39.2	83.24	0.037	0.27
		JZ3	41.4531	121.4594	Barren	0.3	8.79	0.011	0.02
		JZ4	41.1753	121.3764	Municipal	0.4	20.73	0.029	0.50
		JZ5	40.9092	121.8192	Municipal	36.5	90.46	0.086	0.87
	Panjin	PJ1	40.8822	121.5714	Agricultural	0.5	86.62	0.088	0.89
		PJ2	41.0239	122.4339	Municipal	0.4	45.82	0.095	0.97
	Qinhuangdao	QH1	39.6789	119.2911	Municipal	24.2	6.48	0.019	0.05
		QH2	39.7814	119.4136	Barren	8.1	0.00	0.002	0.04
		QH3	39.8394	119.5133	Municipal	0.5	23.84	0.119	2.02
		QH4	39.8017	119.4419	Municipal	0.3	0.00	0.277	2.48
		QH5	39.9800	119.2108	Municipal	0.2	40.35	0.146	1.99
		QH6	39.9203	119.5667	Municipal	0.4	78.20	0.274	4.67
		QH7	39.9653	119.7694	Municipal	22.5	30.71	0.117	3.39
	Tianjin	TJ1	39.2000	117.7641	Industrial	2.2	79.64	0.099	1.50
		TJ2	39.1640	117.6623	Barren	0.7	83.81	0.116	1.22
		TJ3	39.0938	117.7298	Barren	22.0	59.98	0.065	0.59
	Tangshan	TS1	39.0203	117.4578	Industrial	3.1	99.61	0.082	2.38
		TS2	39.1522	118.5342	Industrial	37.3	79.10	0.025	1.01
		TS3	39.0436	118.3642	Municipal	38.0	49.12	0.058	0.93
		TS5	39.4308	119.2800	Agricultural	35.3	32.52	0.066	0.43
		TS6	39.4606	119.1347	Agricultural	0.3	6.99	0.117	0.26
		TS7	39.6619	118.7869	Agricultural	0.3	81.13	0.056	6.74

Table S1. Continued.

Sampling map	Region	Site	Latitude (°N)	Longitude (°E)	Land-use type	Salinity (‰)	Mud contents (%)	TN (%)	TOC (%)
	Weifang	WF1	37.2751	118.9848	Barren	5.2	32.65	0.050	0.36
		WF2	37.1354	119.2870	Saltern	23.5	19.32	0.032	0.13
		WF3	37.1401	119.1434	Saltern	29.7	14.81	0.037	0.70
		WF4	37.0765	119.4793	Barren	25.6	71.59	0.386	0.35
		WF5	36.5802	119.3846	Agricultural	0.8	0.00	0.022	0.03
		WF6	36.7421	119.5374	Agricultural	1.7	34.83	0.034	0.61
		WF7	37.0921	119.5599	Barren	33.7	26.60	0.042	0.20
		WF8	37.1330	119.1860	Saltern	37.0	21.93	0.019	0.11
	Yingkou	YK1	40.9964	122.4639	Municipal	0.3	72.78	0.067	1.36
		YK2	40.6900	122.1292	Industrial	35.3	76.98	0.071	0.86
		YK3	40.4250	122.2844	Industrial	38.5	98.56	0.134	1.42
	Yantai	YT1	37.1286	119.7277	Saltern	28.2	18.81	0.013	0.09
		YT2	37.4017	119.9493	Industrial	27.4	11.32	0.026	1.74
		YT3	37.5518	120.2482	Beach	17.4	0.00	0.002	0.09
		YT4	37.7493	120.5242	Beach	27.8	0.00	0.004	0.02

Table S2. Description of test conditions and test acceptability criteria for luminescent bacteria test.

Parameters	Test conditions
Test type	Luminescent bacteria test for liquid phase sample
Sample type	Aqueous extracts and organic extracts of sediment samples
Test species	Freeze-dried *Vibrio fischeri*; NRRL B-11177 (N-Tox VF100®)
Diluent	20‰ NaCl
Test chamber	96–well plates
Test volume	100 µl (max)
Temperature	15 ℃
Test duration	30 min.
Instrument	N-Tox Model 200
Endpoint	Bioluminescence inhibition rates after 30 min. exposure
Test acceptability criteria	1) Variation of natural light emission rate between 0 and 30 min. must be 0.6–1.8 2) The following reference toxicants cause 20–80% inhibition by 30 min.

Table S3. GC-MSD instrumental conditions and QA/QC data for PAHs, SOs, and APs analyses.

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 mL $min.^{-1}$ (He)					
Injection mode	Splitless					
MS temperature	180 °C					
Detector temperature	230 °C					
Oven temperature	60 °C hold 2 min. Increase 6 °C $min.^{-1}$ to 300 °C 300 °C hold 13 min.				60 °C hold 5 min. Increase 10 °C $min.^{-1}$ to 100 °C Increase 20 °C $min.^{-1}$ to 300 °C 300 °C hold 6 min.	
Elution solvents	50 mL of 20% DCM in hexane				50 mL of 60% DCM in acetone	
Injection volume	2 μL				1 μL	
Target compounds	***Polycyclic aromatic hydrocarbons (PAHs)***		***Styrene oligomers (SOs)***		***Alkylphenols (APs)***	
	Naphthalene	Na	1,3-Diphenylpropane	SD1	4-*tert*-Octylphenol	*t*-OP
	Acenaphthylene	Acl	*cis*-1,2-Diphenylcyclobutane	SD2	4-*tert*-Octylphenol-monoethoxylate	*t*-OP1EO
	Acenaphthene	Ace	2,4-Diphenyl-1-butene	SD3		
	Fluorene	Flu	*trans*-1,2-Diphenylcyclobutane	SD4	4-*tert*-Octylphenol diethoxylate	*t*-OP2EO
	Phenanthrene	Phe	2,4,6-Triphenyl-1-hexene	ST1	Nonylphenol	NP
	Anthracene	Ant	1e-Phenyl-4e-(1-phenylethyl)-tetralin	ST2	Nonylphenol monoethoxylate	NP1EO
	Fluoranthene	Fl	1a-Phenyl-4e-(1-phenylethyl)-tetralin	ST3	Nonylphenol diethoxylate	NP2EO
	Pyrene	Py	1a-Phenyl-4a-(1-phenylethyl)-tetralin	ST4		
	Benzo[*a*]anthracene	BaA	1e-Phenyl-4a-(1-phenylethyl)-tetralin	ST5		
	Chrysene	Chr	1,3,5-Triphenylcyclohexane	ST6		
	Benzo[*b*]fluoranthene	BbF				
	Benzo[*k*]fluoranthene	BkF				
	Benzo[*a*]pyrene	BaP				
	Indeno[1,2,3-*cd*]pyrene	IcdP				
	Dibenz[*a,h*]anthracene	DbahA				
	Benzo[*g,h,i*]perylene	BghiP				
Method detection limits	0.27–0.90 ng g^{-1} dry weight (dw)		0.24–0.91 ng g^{-1} dw		0.10–0.91 ng g^{-1} dw	
Chemicals for surrogate and range of recovery (mean)	Acenaphthene-d_{10}	Ace-d10	68%–96% (80%)		Bisphenol A-d_{16}	BPA-d16 62%–90% (78%)
	Phenanthrene-d_{10}	Phe-d10	75%–105% (93%)			
	Chrysene-d_{12}	Chr-d12	90%–121% (110%)			
	Perylene-d_{12}	Pery-d12	69%–98% (88%)			

Table S4. Concentrations of persistent toxic substances (organic chemicals and heavy metals) in sediments of the Yellow and Bohai seas.

Sea		Organic chemicals (ng g^{-1} dw) [a]			Heavy metals (mg kg^{-1} dw) [b]							
Country		**PAHs**	**SOs**	**APs**	**Cd**	**Pb**	**Cu**	**Zn**	**Cr**	**Ni**	**As**	**Hg**
Yellow Sea	Korea	6.36–57 (22.27)	3.03–34 (10.28)	2.98–92 (18)	0.01–0.20 (0.09)	15.40–51.20 (30.96)	4.80–33.20 (16.85)	15–124 (71.79)	9.20–83.60 (49.41)	3.74–42 (21.79)	3.30–12.10 (6.52)	0–0.08 (0.02)
	China	2.06–60720 (1894)	0.87–164 (13.74)	0.53–98 (21.69)	0.06–0.92 (0.16)	13.70–60.50 (26.69)	4.83–83 (23.24)	30.30–380 (82.58)	27–93 (62.07)	9.67–41.70 (25.75)	2.82–65.90 (11.39)	0–0.43 (0.05)
Bohai Sea	China	6.23–18100 (730)	0.69–218 (16.13)	0.59–504 (23.73)	0.06–1.79 (0.18)	15.20–96.70 (25.85)	7.46–70.10 (22.54)	26.30–383 (76.37)	21.60–146 (55.02)	6.06–45.20 (22.77)	2.25–18.70 (7.19)	0.01–0.75 (0.07)

[a] Data from Yoon et al. (2020).

[b] Data from Tian et al. (2020).

Table S5. Concentrations of heavy metals in aqueous extracts of sediments collected from freshwater (F; *n*=18), brackish water (B; *n*=22), and seawater (S; *n*=10) areas in the Yellow and Bohai seas.

Sea (Country)		Sal.*	Concentrations							
Sites			Cd (μg L^{-1})	Pb (μg L^{-1})	Cu (μg L^{-1})	Zn (μg L^{-1})	Cr (μg L^{-1})	Ni (μg L^{-1})	As (μg L^{-1})	Hg (ng L^{-1})
Yellow Sea (Korea)										
Lake Sihwa	LS1	B	0.09	0.04	32	6.2	0.44	19	6.7	1.3
Sapgyo	SG1	F	0.75	0.09	6.3	8.6	0.56	1.8	1.8	1.3
	SG2	B	0.03	0.06	1.4	2.0	0.30	2.4	4.5	1.1
Sinduri	SD	S	0.10	0.04	7.2	1.3	0.19	0.94	2.6	2.3
Anmyeon	AM	S	0.06	0.05	6.7	1.5	0.15	1.4	2.5	2.2
Cheonsu	CS1	B	0.01	0.05	0.24	0.84	0.10	1.5	2.0	0.4
Geumgang	GG1	F	0.91	0.19	0.96	2.0	0.19	4.9	2.6	0.7
	GG2	B	0.65	0.08	5.6	5.0	0.21	2.5	3.4	1.8
Yellow Sea (China)										
Dandong	DD1	F	0.84	0.46	15	15	0.75	5.2	7.5	4.7
	DD2	F	0.10	0.08	5.2	8.7	0.21	3.1	4.4	1.1
Dalian	DL3	S	0.05	0.22	3.9	2.0	0.26	2.2	3.6	0.77
	DL5	S	0.08	0.21	6.3	1.8	0.30	2.3	4.4	0.44
Yantai	YT6	B	0.27	0.16	32	2.8	0.23	3.1	5.8	0.77
Qingdao	QD1	B	0.02	0.15	18	3.9	1.06	9.9	5.2	0.25
	QD2	F	0.03	0.10	0.17	2.2	0.51	5.8	2.3	0.60
	QD4	B	0.11	0.09	4.9	1.3	1.2	2.0	2.0	0.67
	QD5	B	0.12	0.05	6.4	2.1	0.37	2.4	1.7	1.4
	QD7	B	0.06	0.13	9.7	1.7	0.24	5.7	5.2	0.30
Rizhao	RZ2	B	0.05	0.32	6.2	1.5	0.20	2.6	4.0	0.19
Lianyungang	LY1	S	0.03	0.15	3.1	1.5	0.12	3.1	4.1	0.43
	LY3	S	0.01	0.10	0.30	1.8	0.18	3.9	4.1	1.0
Yancheng	YC8	B	0.06	0.05	5.0	1.7	0.14	2.1	5.0	0.76
Nantong	NT1	B	0.04	0.07	2.2	1.9	0.01	3.3	4.8	0.31
	NT10	B	0.03	0.13	4.0	12	0.08	5.3	3.0	0.62

*Sal.: Salinity.

Table S5. Continued.

Sea (Country) Sites		Sal.*	Concentrations							
			Cd (μg L^{-1})	Pb (μg L^{-1})	Cu (μg L^{-1})	Zn (μg L^{-1})	Cr (μg L^{-1})	Ni (μg L^{-1})	As (μg L^{-1})	Hg (ng L^{-1})
Bohai Sea (China)										
Dalian	DL4	B	0.25	0.09	25	3.3	0.41	4.8	2.5	0.53
	DL6	S	0.15	0.49	6.5	2.4	0.80	4.1	1.4	0.28
Yingkou	YK3	S	0.24	0.09	7.4	5.9	0.20	3.0	4.2	2.7
Panjin	PJ1	F	0.18	0.16	1.1	3.2	0.53	7.0	18	3.1
	PJ2	F	0.03	0.55	1.9	6.9	0.92	7.2	5.7	1.1
Huludao	HL3	F	0.43	0.06	6.3	5.4	0.15	8.1	6.1	2.8
Qinhuangdao	QH1	B	0.51	0.10	3.8	6.2	0.27	2.9	2.6	0.57
	QH2	B	0.09	0.04	4.0	1.7	0.20	0.5	1.6	1.3
	QH4	F	0.05	0.25	1.8	20	0.06	9.6	0.42	210
	QH5	F	0.30	0.18	8.6	8.7	0.09	2.4	1.1	0.83
	QH6	F	0.17	0.12	1.7	2.9	0.17	7.4	9.1	1.8
Tangshan	TS6	F	11	0.16	29	5.2	0.50	4.9	4.1	2.3
	TS7	F	0.14	0.67	4.2	32	0.24	11	0.75	0.83
Tianjin	TJ1	B	0.07	0.15	2.0	4.4	0.24	7.5	2.1	1.9
	TJ4	B	0.05	0.31	2.7	2.3	0.55	2.2	2.9	0.06
	TJ7	B	0.07	0.22	2.1	1.9	0.48	3.0	3.6	0.69
Binzhou	BZ1	S	0.10	0.09	4.3	4.0	0.39	3.8	2.91	0.22
	BZ2	S	0.06	0.13	0.60	2.7	0.09	5.8	2.03	0.80
	BZ3	B	0.03	0.07	15	1.5	0.31	6.3	5.1	0.96
	BZ5	F	0.43	0.11	5.8	1.2	0.21	1.7	0.92	0.67
	BZ6	F	0.53	0.28	2.6	3.8	0.57	1.8	1.6	1.1
Dongying	DY2	F	0.11	0.05	1.5	7.2	0.12	0.80	1.20	8.6
	DY3	F	0.19	0.27	3.6	2.1	0.29	2.7	2.6	0.91
	DY4	F	0.24	0.24	4.8	5.8	6.5	11	4.0	1.5
Yantai	YT2	B	0.29	2.9	91	4.5	0.8	7.3	6.5	2.5
	YT4	B	0.11	0.05	7.2	4.9	1.8	1.3	1.7	3.9

*Sal.: Salinity.

Table S6. Toxicity criteria for aqueous extracts and organic extracts of sediments based on the Bombardier and Bermingham (1999).

Criteria	Toxic unit (TU)	
	Aqueous extracts	**Organic extracts**
Non-toxic	< 1	< 100
Marginally toxic	1–10	100–1000
Moderately toxic	10–100	1000–10000
Highly toxic	> 100	> 10000

Table S7. Summary of toxic unit for aqueous and organic extracts of sediments in the Yellow Sea.

Site information	Aqueous extracts (toxic unit)				Organic extracts (toxic unit)			
	Min.–Max.	**Median**	**Mean**	***n***	**Min.–Max.**	**Median**	**Mean**	***n***
Regions								
Yellow Sea of Korea (YSK)	0–2.2	0.35	0.65	15	0–840	12	90	20
Yellow Sea of China (YSC)	0–1.2	0.26	0.34	42	0–1600	3.6	95	42
Bohai Sea of China (BS)	0–2.5	0.04	0.34	62	0–180	0	18	63
Salinity range								
Freshwater (<5 psu)	0–2.5	0.07	0.29	30	0–150	0.07	23	31
Brackish water (5–30 psu)	0–2.2	0.26	0.39	60	0–1600	15	95	64
Seawater (>30 psu)	0–1.5	0.23	0.44	29	0–77	0.23	9.0	31
Land-use type								
Agriculture	0–2.2	0.06	0.33	35	0–840	0	44	38
Aquaculture	0–1.2	0.31	0.45	6	11–160	26	51	6
Barren	0–1.4	0.41	0.45	26	0–77	0	13	26
Beach	0–1.5	0.31	0.47	9	0–100	0	14	9
Industry	0–1.4	0.06	0.32	20	0–1600	37	190	22
Municipal	0–2.5	0.04	0.33	18	0–150	5.3	24	20
Saltern	0–1.0	0.34	0.47	5	0–32	0	6.5	5
All sites	0–2.5	0.18	0.38	119	0–1600	0.18	56	130

Table S8. Spearman rank correlation between concentration of PAHs, SOs, and APs in sediments and toxic unit for organic extracts ($TU_{organic\ extracts}$).

Target compounds	**$TU_{organic\ extracts}$**	
	r	***P*-value**
16 PAHs	***0.28***	***< 0.05***
Naphthalene	0.21	0.12
Acenaphthylene	0.54	0.22
Acenaphthene	0.10	0.75
Fluorene	0.16	0.42
Phenanthrene	0.23	0.09
Anthracene	0.26	0.1
Fluoranthene	***0.29***	***< 0.05***
Pyrene	***0.36***	***< 0.01***
Benz[*a*]anthracene	0.18	0.22
Chrysene	0.12	0.43
Benzo[*b*]fluoranthene	0.07	0.65
Benzo[*k*]fluoranthene	-0.05	0.78
Benzo[*a*]pyrene	-0.02	0.88
Indeno[1,2,3-*cd*]pyrene	-0.02	0.93
Dibenzo[*a*,*h*]anthracene	0.08	0.69
Benzo[*g*,*h*,*i*]perylene	0.05	0.78
10 SOs	***0.41***	***< 0.01***
1,3-Diphenylpropane	0.24	0.26
cis-1,2-Diphenylcyclobutane	-0.08	0.81
2,4-Diphenyl-1-butene	***0.51***	***< 0.05***
trans-1,2-Diphenylcyclobutane	0.15	0.59
2,4,6-Triphenyl-1-hexene	0.29	0.14
1e-Phenyl-4e-(1-phenylethyl)-tetralin	0.22	0.48
1a-Phenyl-4e-(1-phenylethyl)-tetralin	0.30	0.15
1a-Phenyl-4a-(1-phenylethyl)-tetralin	-0.11	0.69
1e-Phenyl-4a-(1-phenylethyl)-tetralin	0.07	0.83
1,3,5-Triphenylcyclohexane	0.31	0.54
6 APs	***0.38***	***< 0.05***
4-*tert*-Octylphenol	***0.30***	***< 0.05***
4-*tert*-Octylphenol monoethoxylate	***0.28***	***< 0.05***
4-*tert*-Octylphenol diethoxylate	0.20	0.12
Nonylphenol	0.26	0.13
Nonylphenol monoethoxylate	***0.30***	***< 0.05***
Nonylphenol diethoxylate	***0.31***	***< 0.05***

Table S9. Toxic unit values from bioluminescence inhibition tests for aqueous and organic extracts of sediments obtained from the present study and previous studies.

Country	Region	Sampling year	Sample matrix	# of samples	EC50			Toxic unit (TU)			References
					v/v (%)	mg sediment ml^{-1} extract	µl DMSO ml^{-1} solvent	Min	Max	Mean	
Aqueous extracts											
Czech Republic	Brno	2007	Sediment	3	√			2.1	31	12	Beklova et al. (2010)
Poland	-	-	Biochar	6	√			0.9	7.1	3.8	Gondek et al. (2017)
Portugal	-	-	Soil	2	√			1.3	3.3	2.3	Bastos et al. (2014)
Spain	Atlantic coast	-	Sediment	11		√		< 0.01	< 0.01	< 0.01	Riba et al. (2004)
	Iberian Peninsula	-	Sediment	4	√			18	1470	567	Garcia-Ordiales et al. (2019)
	Huelva estuary	2013	Sediment	16		√		54	8700	3531	Rosado et al. (2016)
Finland	Petäjävesi	-	Sediment	3	√			3.4	8.5	6.0	Hyötyläinen and Oikari (1999)
Korea	-	-	Sludge	11	√			n.c.	4.8	2.2	Park et al. (2005)
	-	-	Sludge	6		√		0.4	1.4	0.7	Park et al. (2006)
	-	-	Soil	1	√			-	-	8.3	Eom et al. (2007)
	Yellow Sea	2018	Sediment	15		√		n.c.	2.2	0.7	***This study***
China	Yellow Sea	2018	Sediment	42		√		n.c.	1.2	0.3	***This study***
	Bohai Sea	2018	Sediment	62		√		n.c.	2.5	0.3	***This study***
	Guiyu	2006	Sediment	28	√			n.c.	909	89	Wang et al. (2009)
Organic extracts											
Croatia	Rovinj	-	Sediment	8	√			0.2	7.2	3.1	Bihari et al. (2006)
	Gulf of Rijeka	-	Seawater	6		√		2.0	8.3	5.8	Bihari et al. (2007)
Nigeria	Niger Delta	-	Sediment	9		√		8	222	37	Olajire et al. (2005)
Korea	Siheung	2006	Sediment	5		√		1639	5556	3995	Choi et al. (2010)
		2006	Sediment	5			√	333	1111	798	Choi et al. (2010)
	Ulsan	2006	Sediment	5		√		89	1515	506	Choi et al. (2010)
		2006	Sediment	5			√	18	303	101	Choi et al. (2010)
	Masan	2006	Sediment	5		√		1124	7143	4507	Choi et al. (2010)
		2006	Sediment	5			√	227	1429	909	Choi et al. (2010)
		2016	Sediment	2		√		66.7	111	89	Lee et al. (2020)
	-	-	Sludge	6		√		0.4	1.4	0.7	Park et al. (2006)
	Yellow Sea		Sediment	20			√	n.c.	840	90	***This study***
China	Yellow Sea		Sediment	42			√	n.c.	1587	95	***This study***
	Bohai Sea		Sediment	63			√	n.c.	185	18	***This study***
	Yulin	-	Wastewater	5	√			1.2	315	205	Ma et al. (2017)

[a] n.c.: Not calculated.

Supplementary Figures

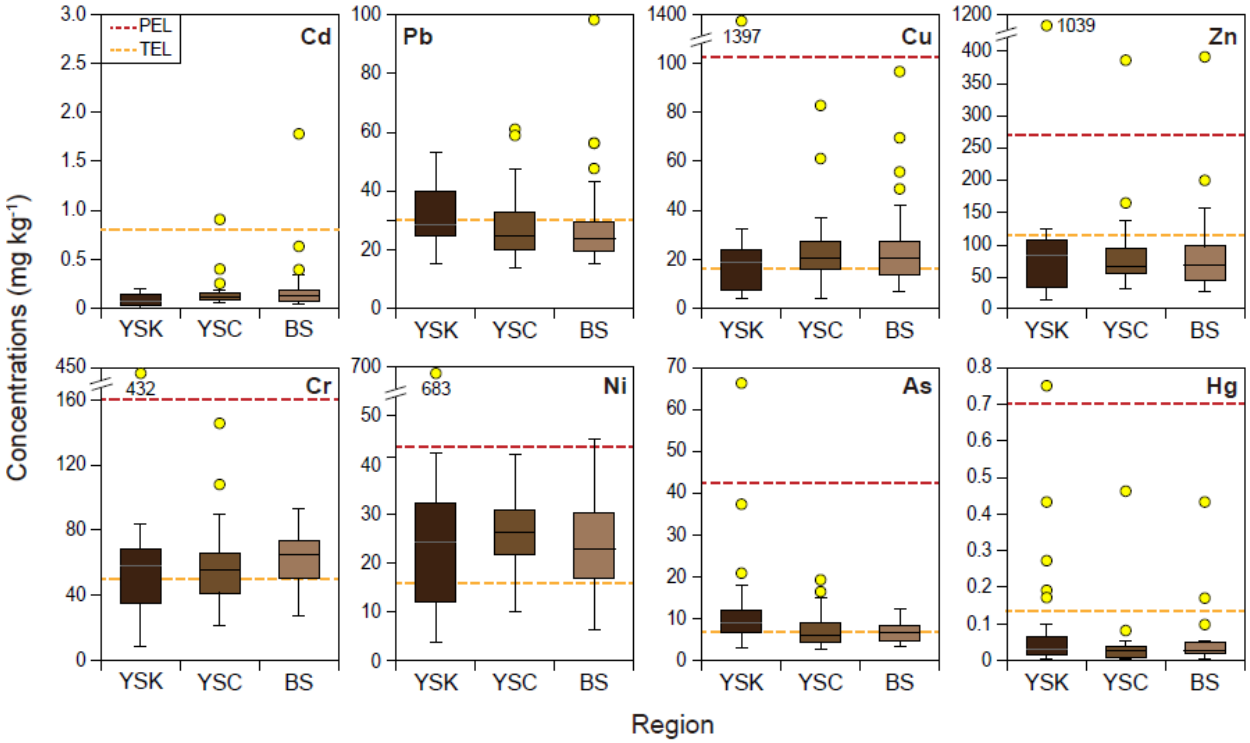


Fig. S1. Box plots for the concentrations of eight heavy metals (Cd, Pb, Cu, Zn, Cr, Ni, As, and Hg) in sediments relative to regions by the Yellow Sea of Korea (YSK), Yellow Sea of China (YSC), and Bohai Sea (BS). Each dot represents raw data of measured heavy metals. Yellow and red dotted lines indicated existing sediment quality guidelines (TEL: threshold effect level, PEL: probable effect level, guideline from Long et al. (1995)).

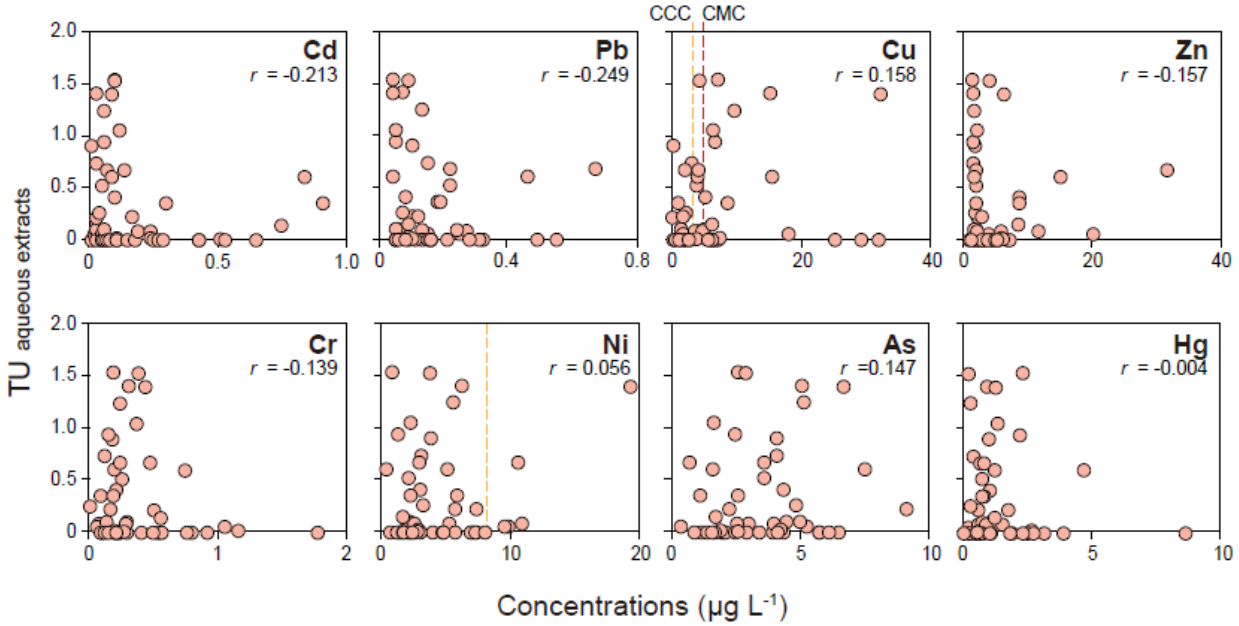


Fig. S2. Spearman correlation analysis between concentrations of eight soluble heavy metals (Cd, Pb, Cu, Zn, Cr, Ni, As, and Hg) and toxic unit (TU) for aqueous extracts of sediments. Red and orange dotted lines represent criterion maximum concentration (CMC, acute) and criterion continuous concentration (CCC, chronic) of National Recommended Aquatic Life Criteria of US EPA (2009).

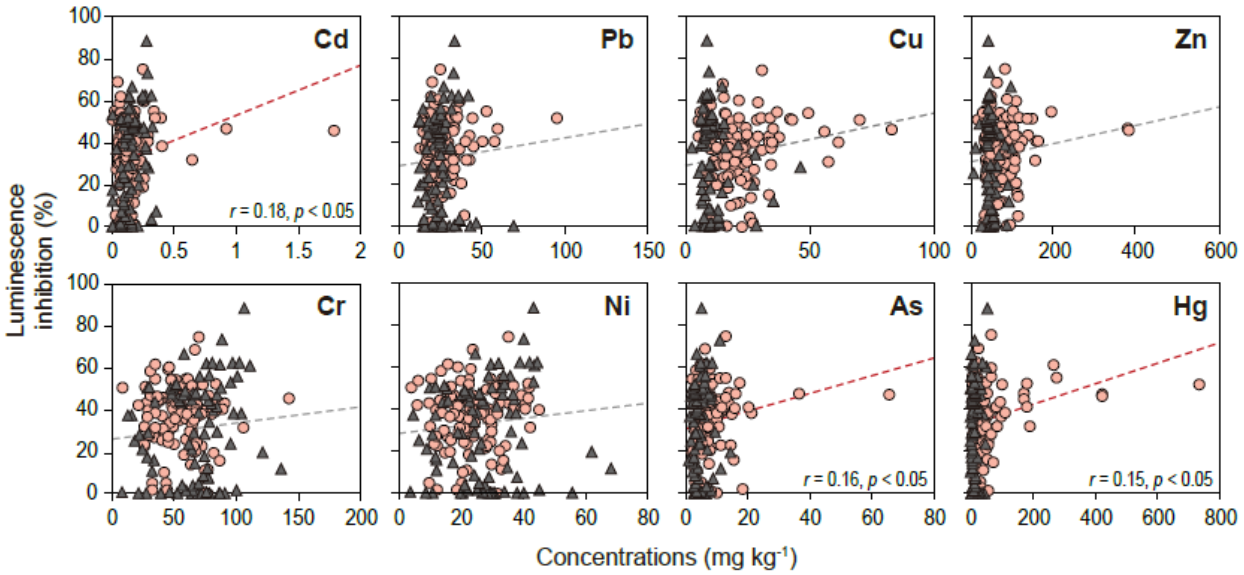


Fig. S3. Spearman correlation analysis between concentrations of eight heavy metals (Cd, Pb, Cu, Zn, Cr, Ni, As, and Hg) and luminescence inhibition of aqueous extracts from sediments. Reference data obtained from Kim et al. (2020).

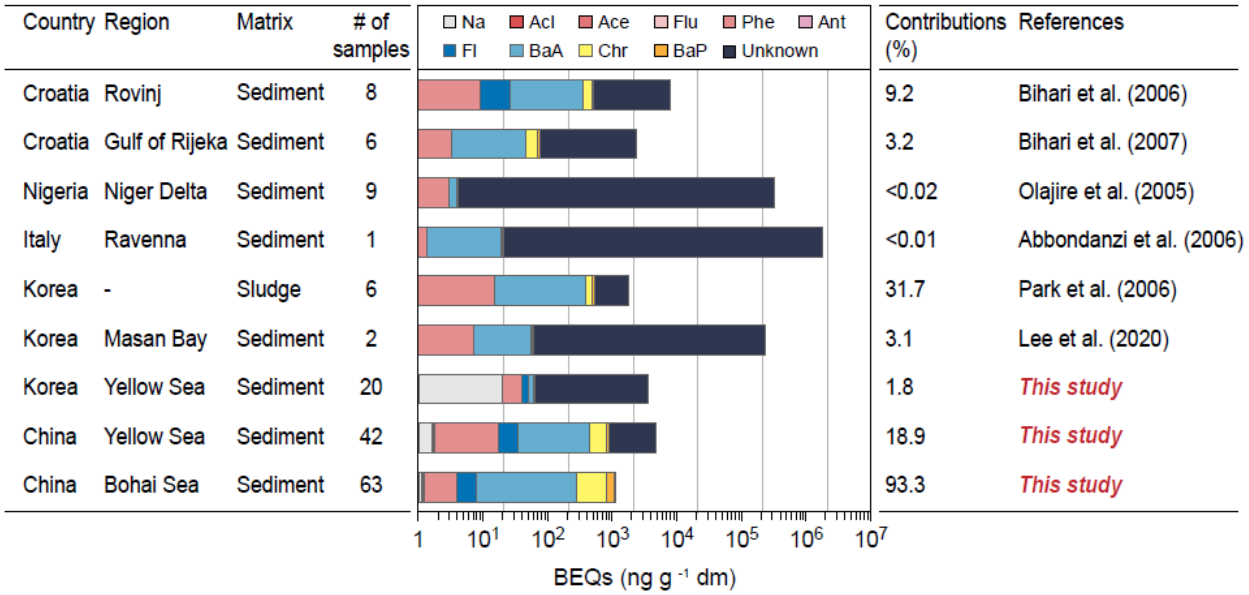


Fig. S4. Toxicity contribution of 10 PAHs to bioassay-derived BEQs in sediments obtained from the present study and previous studies.

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