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Spatial distribution and source identification of traditional and emerging persistent toxic substances in the offshore sediment of South Korea

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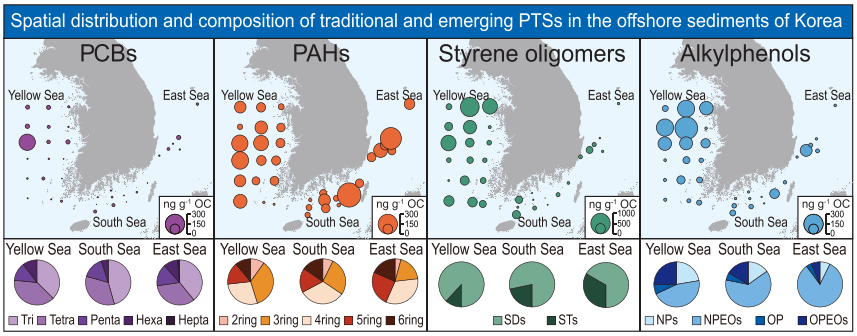
HIGHLIGHTS

- First investigated the extensive PTSs pollution in offshore environments of South Korea
- Distributions and sources of PTSs in three seas generally reflected oceanographic settings.
- Sedimentary PCBs and PAHs showed no significant difference in the three sea areas.
- SOs and APs concentrations were significantly greater in the Yellow Sea than other two seas.
- Major source of PAHs in the Yellow Sea was found to be coke production (77%).

GRAPHICAL ABSTRACT



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ABSTRACT

While the coastal pollution of persistent toxic substances (PTSs) has been widely documented, information on offshore environments remains limited. Here, we investigated the spatial distribution and sources of PTSs in the offshore sediments (n = 34) of South Korea. Sediment samples collected from the Yellow Sea (n = 18), the South Sea (n = 10), and the East Sea (n = 6), in 2017–18 were analyzed for a total of 71 PTSs. Target compounds include 31 PCBs, 15 PAHs, 9 emerging PAHs (e-PAHs), 10 styrene oligomers (SOs), and 6 alkylphenols (APs). Sedimentary PCBs showed relatively low concentrations with no significant difference across the three seas (0.16–6.9 ng g^{-1} normalized organic carbon, OC). Low-chlorinated PCBs (tri- and tetra Cl-CBs) were predominant (mean: 77%), primarily indicating atmospheric inputs. PAHs widely accumulated in the three seas with low to moderate level (22–250 ng g^{-1} OC), and dominated by high molecular weight PAHs (4–6 rings). PMF analysis revealed coast-specific PAHs sources; i.e., originated from mainly coke production (77%) in the Yellow Sea, vehicle emissions (68%) in the South Sea, and fossil fuel combustion (49%) in the East Sea. SOs showed significant contamination than other PTSs, with elevated concentrations in the Yellow Sea (mean: 350 ng g^{-1} OC). APs showed a similar regional distribution to SOs, but concentrations were much lower (mean: 17 ng g^{-1} OC). SOs and APs seemed to be introduced from rivers and estuaries on the west coast of Korea, where industrial and municipal activities are concentrated, then might be transported to offshore through tide or currents. Overall, the novel data presented for various PTSs in offshore Korean sediments warrant the necessity of a long-term monitoring effort and urgent management practice to protect marine ecosystem.

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1. Introduction

Persistent toxic substances (PTSs) originate from various anthropogenic activities, and are transported to the marine environment through wastewater, rivers, and the atmosphere (Yoon et al., 2020). These PTSs pose potential hazards to marine ecosystems (Wolska et al., 2012). PTSs mainly accumulate in coastal sediments, because of their hydrophobicity and particle adsorption properties. However, less hydrophobic PTSs are transported farther to the offshore regions through the atmosphere and via ocean currents. For example, high concentrations of PTSs, such as polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs), have been detected in the remote sites of the Arctic (Dachs et al., 2002; Galbán-Malagón et al., 2012). In addition, PTSs are highly persistent and accumulate both in coastal sediments and deep-sea bottoms (Montuori et al., 2016; Sobek and Gustafsson, 2014; Webster et al., 2011). To date, studies on the distribution and sources of PTSs have primarily focused on coastal sites, and offshore environment has received little attention for the sediment PTSs study until lately. Thus, it is necessary to monitor a wide area extending from the coastal areas to offshore sites in order to accurately understand the behavior and fate of PTSs in marine environment.

Recent concerns of PTSs identified on the coasts of South Korea encompass PCBs, PAHs, styrene oligomers (SOs), and alkylphenols (APs) (Hong et al., 2016a; Lee et al., 2017; Yoon et al., 2017, 2019). Although PCBs designated as persistent organic pollutants (POPs) in 2001 by the Stockholm Convention were banned in the early 1980s globally, they are still detected in marine environments, including from Korean seas (Hong et al., 2016b). PAHs have negative effects on aquatic organisms, including genotoxicity and developmental toxicity. Consequently, the United States Environmental Protection Agency (US EPA) prioritizes 16 PAHs (traditional PAHs). Although PAHs are managed as controlled substances, they continue to be present at great concentrations in the marine environment, due to their multiple sources (Gong et al., 2018; Kannan et al., 2005; Neff, 2002). Since 2007, APs have been completely banned from domestic usage in South Korea, but they were frequently reported to occur in river and coastal sediment, thus remains under significant concern (Hong et al., 2016b; MOE, 2014; Lee et al., 2017; Soares et al., 2008). SOs have been recently reported in coastal sediments (Hong et al., 2016a). SOs negatively impact marine organisms, such as genotoxicity and reproductive toxicity; yet, monitoring data remain much limited, impeding effective management (Kwon et al., 2014; Ohyama et al., 2001; Tatarazako et al., 2002).

As more than 40% of the population in South Korea is concentrated along the coastline, human impacts on the coastal environment would be inevitable (MOE, 2008). Since the 1960s, coastal pollution and environmental deterioration caused by traditional PTSs have long been recognized due to the rapid industrialization and increased population density (Ahn et al., 1995; Choi et al., 2009; Hong et al., 2006; Koh et al., 2005; Moon et al., 2008; Yim et al., 2007). Consequently, interest and research on PTSs in the coastal areas of South Korea have sharply grown since the 1990s (Khim and Hong, 2014). Yet, few studies have focused on offshore pollution, despite the persistence and long-range transport potential of PTSs. In fact, the extent of PTS contamination in the coastal environment of Korea would vary primarily depending on oceanographic settings and the size and type of surrounding human activities. In particular, Lake Sihwa in the Yellow Sea, Masan Bay in the South Sea, and Ulsan Bay and Yeongil Bay in the East Sea are seriously contaminated by PTSs (Khim and Hong, 2014).

More recently, numerous emerging toxic substances have been identified and introduced in Korean coastal waters (Cha et al., 2019; J. Kim et al., 2019). Examples include 11H-benzo[*b*]fluorene (11BF), benzo[*b*]naphtho[2,3-d]furan (BBNF), benzo[*b*]naphtho[2,1-d]thiophene (BBNT), 5-methylbenzo[*a*]anthracene (5MBA), and benzo[*j*]fluoranthene (BjF). These substances can cause cancer, genotoxicity, and developmental toxicity (Cha et al., 2019; J. Kim et al., 2019; Mimura and Fujii-Kuriyama, 2003). In addition, benzo[*e*]pyrene (BEP) and 1,12-dimethylbenzo[*c*]phenanthrene (BCP) are potential toxic substances, while 2-methylphenanthrene (2MA) and 9-ethylphenanthrene (9EP) are mutagenic substances (Gibson et al., 1978; LaVoie et al., 1983; Onodera et al., 1994). Thus, to understand the potential toxicity and fate of these emerging-PAHs (e-PAHs) in marine environments, monitoring studies are indeed necessary and timely significant (Amamiya et al., 2019; Kwon et al., 2015).

Here, we investigated the distribution characteristics and potential sources of traditional (PCBs, PAHs, and APs) and emerging (e-PAHs and SOs) PTSs in the offshore surface sediment of South Korea. The objectives were to i) determine the distribution of PTSs in the sediment of the three Korean seas, ii) identify potential sources of PTSs, and iii) compare the degree of offshore sedimentary contamination of target PTSs with those reported in coastal regions of Korea and elsewhere. Our results are expected to provide baseline data and information on PTSs in offshore sediment that will be of use to future research and management.

2. Materials and methods

2.1. Study area, sampling, and sample preparation

The present study focused on the three seas around the Korean Peninsula, namely the Yellow Sea, the South Sea, and the East Sea. These three seas represent somehow typical and independent characteristics in oceanographic settings and geomorphological features (Khim et al., 2021). First, the Yellow Sea, westward from the mainland, is a shallow water system (mean: 51 m), characterized by macrotidal environment (4–10 m) with extensively developed coastal wetlands due to larger river inputs of sediments and organic matters. The South Sea, southward from the mainland, shows a mesotidal environment (1.3–4.3 m), having many semi-closed bays and islands, but slightly deeper (mean: 71 m) than the Yellow Sea. Finally, the East Sea, eastward from the mainland, is a small marginal sea with relatively deep waters (mean: 1497 m; max: 2985 m) compared to the other two seas, and featured by the typical rocky shore and beaches along the coast.

A total of thirty-four sediment samples were collected in the Yellow Sea (n = 18), the South Sea (n = 10), and the East Sea (n = 6) during August 2017, August 2018, and May 2017, respectively (Fig. 1). In the East Sea, due to the deep water depth, wide-area sampling was unavailable, and sediment samples were collected from a total of six sites, from shallow to deep water depth. Each station of the Yellow Sea was classified into three groups (A: inner; B: middle; and C: outer) depending on the distance from the land (Fig. 1). Surface sediments (0–2 cm) were collected using a box corer on board. Sediment samples were homogenized and stored in pre-cleaned 125 mL glass jars, and were immediately frozen at −20 °C until laboratory analysis. Mud contents in sediments were measured from partial analysis by wet-sieving (Buchanan, 1984). All samples were freeze-dried and sieved (2-mm mesh) in the laboratory before analyses for organic matters and target PTSs.

2.2. Analysis of organic carbon and nitrogen and their stable isotopes

Sedimentary organic carbon (OC) and total nitrogen (TN) were analyzed using the previously reported methods (Kim et al., 2018). To measure OC and carbon stable isotopes in sediment samples, inorganic carbon was removed by adding 1 M hydrochloric acid. TN and nitrogen stable isotopes were analyzed using freeze-dried sediment without acid treatment. OC, TN, and their stable isotopes were analyzed by use of an elemental analyzer coupled with an isotope ratio mass spectrometer (EA-IRMS). Isotopic compositions were expressed as δ notation, based on the following Eq. (1):

$$\delta^{13}\mathrm{C} \text{ or } \delta^{15}\mathrm{N}\ (\text{‰}) = \left[\left(\mathrm{R}_{\mathrm{sample}}/\mathrm{R}_{\mathrm{reference}}\right) - 1\right] \times 1000 \quad (1)$$

where R is the ratio of $^{13}C/^{12}C$ or $^{15}N/^{14}N$ of a given sample and reference material. Vienna Peedee Belemnite and atmospheric N_2 were used

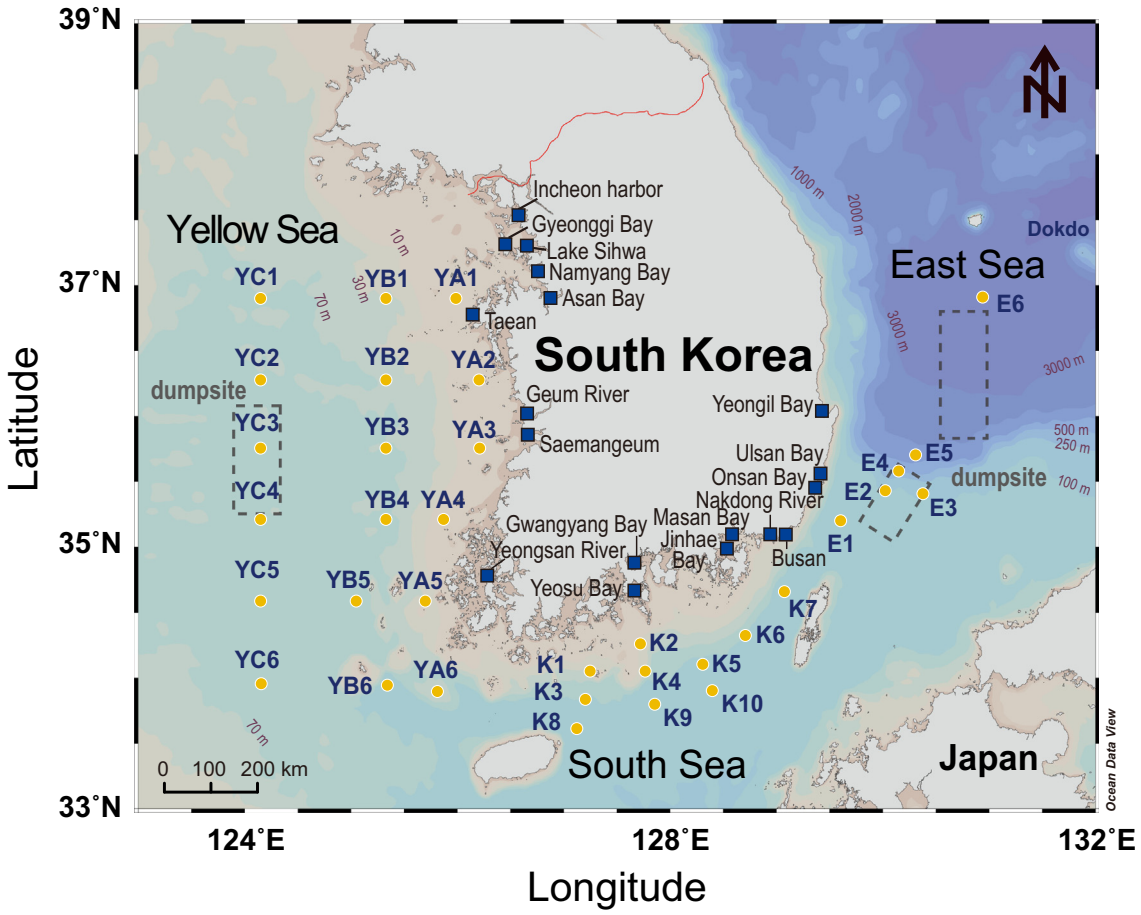


Fig. 1. Map showing the study area and sampling locations around South Korea in the Yellow Sea (n = 18), the South Sea (n = 10), and the East Sea (n = 6). Coastal location (blue square) refers to the data available for a regional comparison presented in Fig. 5. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

as reference materials for carbon and nitrogen, respectively. Analytical errors were 0.05‰ for $\delta^{13}C$ and 0.1‰ for $\delta^{15}N$, and were estimated from working standards (CH6 for $\delta^{13}C$ and N1 for $\delta^{15}N$, International Atomic Energy Agency, Vienna, Austria).

2.3. Analyses of PTSs

Standard materials of 31 PCB congeners were purchased from Sigma-Aldrich (Saint Louis, MO). Fifteen traditional PAHs were obtained from ChemService (West Chester, PA), nine e-PAHs from Sigma-Aldrich, 10 SOs from Wako Pure Chemical Ind. (Osaka, Japan) and Hayashi Pure Chemical Ind. (Osaka, Japan), and six APs from Sigma-Aldrich.

Analyses of PTSs in sediments were conducted following the methods described elsewhere, with minor modifications (Hong et al., 2016a; Lee et al., 2017; Cha et al., 2019; J. Kim et al., 2019; An et al., 2020). Ten grams of freeze-dried sediment samples were extracted, and placed for 16 h on a Soxhlet extractor with dichloromethane (DCM, Burdick & Jackson, Muskegon, MI). For quality control, 12 surrogate standards were added before extraction. These included ^{13}C-labeled CB-28, -52, -101, -153, -138, -180, and -209 for PCBs; acenaphthene-d10 (Ace-d10), phenanthrene-d10 (Phe-d10), chrysene-d12 (Chr-d12), and perylene-d12 (Pery-d12) for PAHs and SOs; and bisphenol A-d16 for APs. Organic extracts were concentrated with rotary evaporator and were exchanged to hexane (Burdick & Jackson). Activated copper powder (Merck, Darmstadt, Germany) was added to remove elemental sulfur for 1 h. The extracts were subsequently cleaned and fractionated through an open column chromatography with activated silica gel (70–230 mesh, Sigma-Aldrich). The first fraction (F1) was collected to obtain PCBs by the elution with 30 mL hexane. The second fraction (F2) was collected to obtain PAHs and SOs by the elution of 60 mL hexane:DCM (8:2, v/v). The third fraction (F3) was collected to obtain APs by the elution of 50 mL 60% DCM in acetone (J.T. Baker, Center Valley, PA). Each fraction was concentrated to 1 mL under N_2 gas flow, and 100 ng internal standard (2-fluorobiphenyl, ChemService) was added before the instrumental analyses.

Target PTSs were quantified using an Agilent 7890B GC coupled with a 5977B mass spectrometer detector (MSD, Agilent Technologies, Santa Clara, CA). Instrumental conditions for analyzing PTSs are presented in Tables S1 and S2 of the Supplementary Materials. Method detection limits (MDLs) were calculated as 3.707 × standard deviations of standard materials quantified seven times. MDLs were 0.01–0.03 ng g^{-1} dry weight (dw) for PCBs, 0.07–0.89 ng g^{-1} dw for PAHs, 0.06–0.52 ng g^{-1} dw for e-PAHs, 0.11–0.89 ng g^{-1} dw for SOs, and 0.08–3.7 ng g^{-1} dw for APs. The average recovery of surrogate standards for PCBs was 106%, 99%, 102%, 109%, 110%, 114%, and 115% for ^{13}C-labeled CB-28, -52, -101, -153, -138, -180, and -209, respectively. Surrogate recoveries for Ace-d10, Phe-d10, Chr-d12, and Pery-d12 were 72%, 76%, 88%, and 90%, respectively. Average recovery of bisphenol A-d16 was 75% (see Table S1 for details). Concentrations of PCBs in the offshore sediments of South Korea were partially reported in a previous study (Y.N. Kim et al., 2019).

2.4. Positive matrix factorization receptor model

The EPA positive matrix factorization receptor (PMF) model (Ver. 5.0) was utilized to estimate the sources of PAHs. This model is a type of factor analysis used to apportion the sources of PAHs (Paatero and

Tapper, 1993). PMF uses a generic factorization method to quantify the contribution of source compositions (Larsen and Baker, 2003; Norris et al., 2014). Each contribution and composition of factors are derived by minimizing the objective function Q. Q is based on Eq. (2).

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{X_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}}\right]^2 \quad (2)$$

where x_{ij} is the concentration of species j in sample i, u_{ij} is the uncertainty of x_{ij}, and p is the number of factors. The relative contribution of factor k is g_{ik}, and f_{kj} is the profile of each source. Based on PMF user guidelines, uncertainties (*Unc*) were calculated using Eqs. (3) and (4).

$$Unc = 5/6 \times MDL \quad (3)$$

$$Unc = \sqrt{(ErrorFraction \times concentration)^2 + (0.5 \times MDL)^2} \quad (4)$$

When PTS concentrations were below the MDL, *Unc* was calculated using Eq. (3), and when PTS concentrations exceeded the MDL, *Unc* was calculated using Eq. (4). The *Error Fraction* was calculated as the standard deviation of compound *j*. In this study, datasets with 15 PAHs × 34 sites were used as input data for the PMF model. In the model analysis, the category of phenanthrene (Phe) was considered weak, because of its low correlation with the predicted value ($r^2 = 0.21$). All other PAHs had strong correlations ($r^2 > 0.90$). After the base model was run 100 times, the Q values with 2–5 factors were calculated, and Q_{True}/Q_{Exp} values were 2.4, 1.2, 1.3, and 1.5, respectively. In this study, the 3-factor solution was selected that exhibited the lowest Q (robust) value (1.2). To evaluate the accuracy, the modeled PAH concentration was compared to the measured concentration. The coefficient of determination formula was 0.99 (R^2) in the linear regression, indicating that the PMF model result is satisfactory.

2.5. Statistical analysis

SPSS 24.0 (SPSS Inc., Chicago, IL) was used to confirm the relationship between physicochemical properties and PTS concentrations. Principal component analysis (PCA) was used with varimax rotation. We also performed Bartlett's and Kaiser-Meyer-Olkin (KMO) tests. When the index exceeded 0.6, the correlation matrix was considered suitable for PCA analysis.

3. Results and discussion

3.1. Distribution and sources of sedimentary organic matter

OC content in the sediment from the offshore regions of the three seas (Yellow Sea, South Sea, and East Sea) ranged from 0.12 to 0.82% (mean: 0.37%), 0.22 to 1.5% (mean: 0.87%), and 0.24 to 3.2% (mean: 2.3%), respectively (Fig. 2a). Relatively greater OC content was detected in the East Sea sites, due to the smaller grain size and higher mud content (MC) (Table S3 and Fig. S1a), supporting the previous studies (Lee et al., 2008; Song et al., 2014). Significantly greater OC content in the East Sea than the Yellow Sea and the South Sea ($p < 0.05$) was attributed to higher export production and lower inputs of terrestrial minerals (i.e., substances were less diluted in the East Sea) (Lee et al., 2008) (Fig. 2a). OC contents in sediments were also greater in the outer sites (YC group) of the Yellow Sea (Fig. 2a). The central part of the Yellow Sea contains areas of mud deposits, due to its hydrodynamic properties and oceanographic settings. These phenomena led to relatively high OC content in this region (Qiao et al., 2017). TN content of the Yellow Sea, the South Sea, and the East Sea were 0.02–0.14% (mean: 0.06%), 0.04–0.14% (mean: 0.09%), and 0.07–0.35% (mean: 0.27%), respectively (Table S3). The C/N ratio (ratio of OC and TN; ranging from 6 to 20) was used to determine the origin of organic matter (Fig. S2a). It is indicated that sedimentary organic matter in the offshore areas of South Korea mainly originated from marine phyto- and zooplankton (Nakai et al., 1982).

The $\delta^{13}C$ values in the sediments of the Yellow Sea, the South Sea, and the East Sea ranged from −22.3 to −20.5‰ (mean: −21.6‰), from −21.6 to −21.1‰ (mean: −21.3‰), and from −21.6 to −20.9‰ (mean: −21.2‰), respectively. $\delta^{15}N$ values in sediments ranged from 4.4 to 7.0‰ (mean: 5.9‰), 6.3 to 8.1‰ (mean: 7.1‰), and 6.2 to 7.7‰ (mean: 6.9‰) in the Yellow Sea, the South Sea, and the East Sea, respectively (Table S3). $\delta^{13}C$ values originating from marine organic matter were previously reported to range from −22 to −20‰, while those originating from terrestrial organic matter ranged from −27 to −25‰ (Peters et al., 1978). Thus, sedimentary organic matter tends to originate from marine sources (Fig. S2b). In comparison, $\delta^{15}N$ values previously ranged from −0.9 to 4.4‰ for anthropogenic organic matter and 2.9 to 9.8‰ for organic matter of marine origin (Liénart et al., 2017). Thus, $\delta^{15}N$ in the offshore sediments of South Korea obtained from the present study (4.4–8.1‰) were considered to be of marine origin, in general. However, relatively lower $\delta^{15}N$ in the sediments of the Yellow Sea seemed to be more affected by the anthropogenic organic matter input, due to the large influx of freshwater through estuaries, compared to the other seas (Wang et al., 2003).

3.2. Distribution and composition of PTSs

PTS concentrations in the offshore sediments were significantly correlated with OC content ($p < 0.05$) (Fig. S3). Comparing PTS contamination in the offshore sediments between the three seas based on dry weight might not be appropriate due to the regional heterogeneity of OC in particles of different size classifications (Huang et al., 2011). As shown in Fig. 2a, OC content significantly differed in the sediments of the three sea areas; thus, OC-normalized concentrations of PTSs were used to assess and compare the extent of contamination in the present study.

3.2.1. PCBs

PCB concentrations in the sediments of the offshore areas in the Yellow Sea, the South Sea, and the East Sea were 0.37–6.9 ng g^{-1} OC (mean: 1.4 ng g^{-1} OC), 0.16–1.6 ng g^{-1} OC (mean: 0.62 ng g^{-1} OC), and 0.59–1.7 ng g^{-1} OC (mean: 1.0 ng g^{-1} OC), respectively (Fig. 2b and Table S4). PCB concentrations in the offshore sediments of South Korea might not impact benthic organisms; they did not exceed sediment guidelines of the Canadian Council of Ministers of the Environment (CCME, interim marine sediment quality guidelines: 21.5 μg kg^{-1} dw; probable effect levels: 189 μg kg^{-1} dw) (CCME, 2002). Low-chlorinated PCBs, such as tri- and tetra-CBs frequently dominated the offshore sediments around Korea, with detection rates of 53% and 30%, respectively, whereas the detection rates of high-chlorinated PCBs, such as penta- to hepta-CBs, were low. Low-chlorinated PCBs have relatively high volatility compared to high-chlorinated PCBs, and can travel through the atmosphere to remote areas (Wania and Mackay, 1996). Thus, the sedimentary PCBs in the offshore areas of South Korea were likely introduced from neighboring countries through atmospheric deposition (Y.N. Kim et al., 2019).

3.2.2. Traditional PAHs

PAH concentrations were 22–180 ng g^{-1} OC (mean: 102 ng g^{-1} OC) in the Yellow Sea, 35–250 ng g^{-1} OC (mean: 88 ng g^{-1} OC) in the South Sea, and 83–220 ng g^{-1} OC (mean: 130 ng g^{-1} OC) in the East Sea (Fig. 2c and Table S5). In the Yellow Sea, PAHs were more concentrated in sediments of outer areas (YC group) compared to coastal areas (YA group). The distribution of PAHs found in the Yellow Sea showed a similar pattern reported in the previous studies (J. Li et al., 2015; Lin et al., 2011, 2017; Wang et al., 2020; Xu et al., 2019). The central parts of the Yellow Sea contain mud deposits where hydrodynamic effects cause introduced organic toxic substances to accumulate (J. Li et al.,

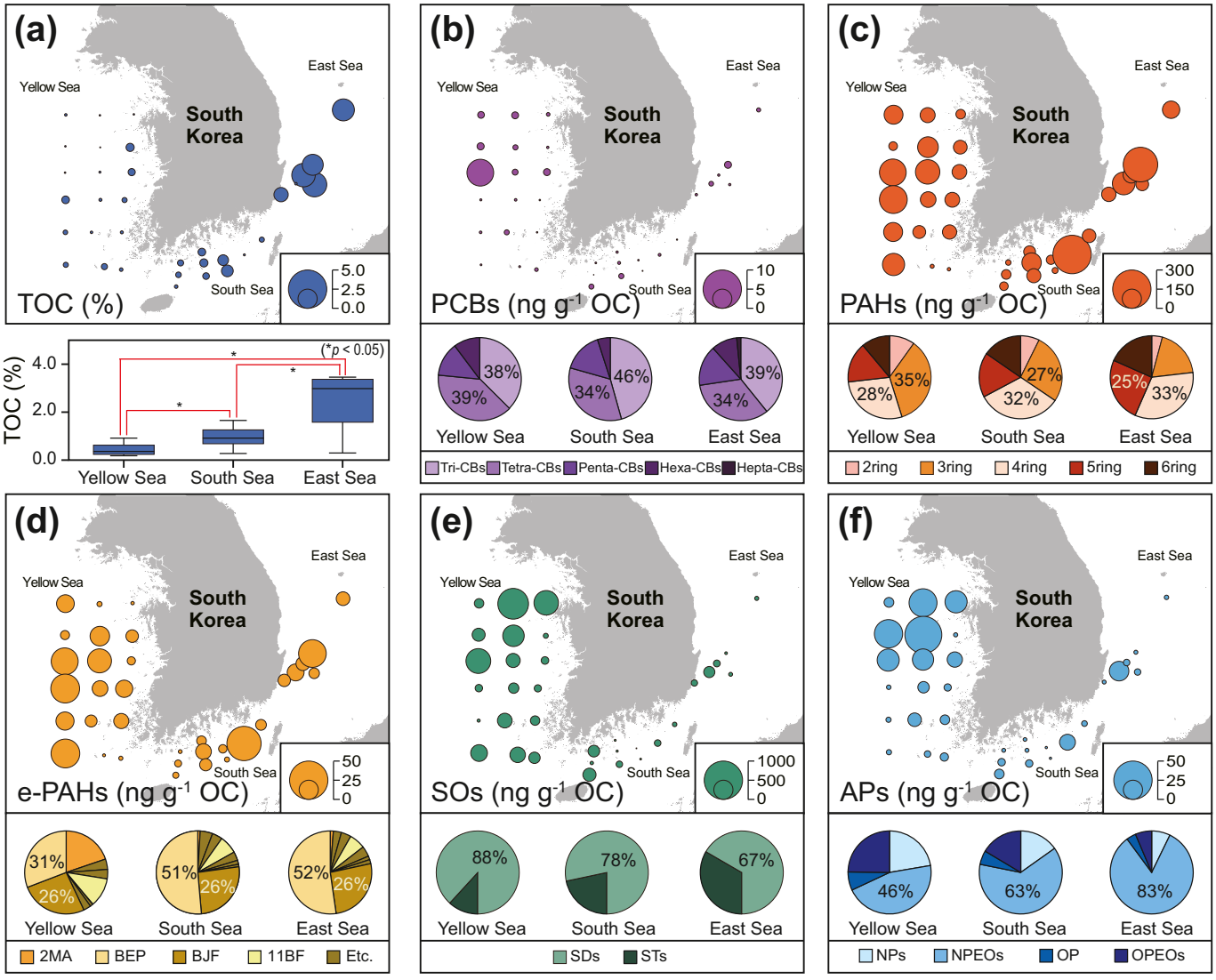


Fig. 2. Distributions and composition of PTSs in the sediments of the Yellow Sea, the South Sea, and the East Sea. (a) Organic carbons, (b) PCBs, (c) PAHs, (d) e-PAHs, (e) SOs, and (f) APs. On the map, organic carbon is expressed as a percentage, while PTSs are expressed as normalized organic carbon concentration. Compositions of each PTS are given as percentages. Kruskal-Wallis test was used to examine differences and the asterisk (*) represents significant differences ($p < 0.05$).

2015; Lin et al., 2011, 2017; Qiao et al., 2017). The relatively high concentration of PAHs in the sediment at K7 might be influenced by the surrounding industrial complexes and cities (Han et al., 2016). PAH concentrations in the East Sea were highest at site E5 (220 ng g^{-1} OC). However, it should be noted that the concentrations of sedimentary PAHs across the three seas were not statistically different (Fig. S1). Overall, PAHs were widely distributed in the sediments of offshore areas around South Korea.

Meantime, the composition of PAHs differed among the three seas. For instance, Phe (mean: 19 ng g^{-1} OC) was the greatest PAH found in the sediments of the Yellow Sea, whereas indeno[1,2,3-*cd*]pyrene (IcdP) dominated in the sediments of the South Sea (mean: 13 ng g^{-1} OC) and the East Sea (mean: 25 ng g^{-1} OC) (Table S5). In all three sea areas, high molecular weight (HMW)-PAHs (4–6 rings) accounted for a large proportion of total PAHs (Yellow Sea: 55%; South Sea: 66%; East Sea: 80%) (Table S5). This result indicated that combustion sources were dominant, including fossil fuel combustion and vehicle emissions (Gong et al., 2018; Kannan et al., 2005; Neff, 2002). In comparison, 3 ring-PAHs were prevalent in the sediments of the Yellow Sea (contributing 35%), while 4 ring-PAHs (32%) were prevalent in the South Sea. Similarly, 4 ring PAHs (33%) 5 ring-PAHs (25%) were prevalent in the East Sea (Fig. 2c). Overall, in the three seas, PAHs in sediments seemed to mainly originate from combustion sources; however, the specific sources in each sea differed slightly.

3.2.3. Emerging PAHs

e-PAH concentrations in sediments ranged from 3.4 to 38 ng g^{-1} OC (mean: 19 ng g^{-1} OC) in the Yellow Sea, 4.5 to 44 ng g^{-1} OC (mean: 14 ng g^{-1} OC) in the South Sea, and 14 to 35 ng g^{-1} OC in the East Sea (mean: 21 ng g^{-1} OC) (Fig. 2d and Table S6). e-PAHs in offshore sediments showed a similar distribution pattern to that of traditional PAHs ($p < 0.05$). BEP and BjF accounted for the highest proportion of PAHs, on average (Fig. 2d). In the Yellow Sea region, 3 ring e-PAHs were the most prevalent (39%), with 2MA occurring in the greatest contribution. In the South Sea and the East Sea, 5 ring e-PAHs (such as BEP) had the highest percentages (51% and 52%, respectively). Previously, 2MA was shown to originate from diesel and wood combustion (Fine et al., 2001, 2002; Westerholm et al., 2001), while BEP originated from vehicle emissions (Khalili et al., 1995), and BjF from fossil fuel combustion (Ledesma et al., 2000). Thus, the major sources of e-PAHs in the offshore sediments of the South Sea and the East Sea were likely vehicle emissions and fossil fuel combustion, while diesel and wood combustion potentially represented the main sources in the Yellow Sea.

3.2.4. Styrene oligomers

SO concentrations in offshore sediments ranged from 80 to 790 ng g^{-1} OC (mean: 350 ng g^{-1} OC) in the Yellow Sea, with the greatest concentration occurring at site YB1 (Fig. 2e and Table S7). In the South Sea, SO concentrations ranged from 50 to 340 ng g^{-1} OC (mean: 140 ng g^{-1} OC), with the greatest concentration occurring at site KC1. In the East Sea, SO concentrations ranged from 69 to 290 ng g^{-1} OC (mean: 150 ng g^{-1} OC). SO concentrations in the sediments of the Yellow Sea were significantly greater than those found in the South Sea and the East Sea (Fig. S1). SOs are mainly derived from polystyrene plastics, and are introduced by runoff from rivers, rather than atmospheric deposition (Kwon et al., 2015). Of note, there are many large rivers flowing into the Yellow Sea. Consequently, relatively greater concentrations of SOs were detected in the offshore sediments of the Yellow Sea (H.G. Choi et al., 2011) compared to other seas. Meantime, styrene dimers (SDs) were the dominant group of SOs in all three seas (Yellow Sea: 88%; South Sea: 78%; East Sea: 67%). SDs are the products of styrene trimers (STs) decomposition (Tian et al., 2020). Because all sites evaluated were located in regions that were 6.4–190 km offshore (average 80 km) from mainland, STs introduced to the sea were assumed to decompose during transportation to form SDs.

3.2.5. Alkylphenols

AP concentrations in offshore sediments ranged from 4.0 to 150 ng g^{-1} OC (mean: 24 ng g^{-1} OC) in the Yellow Sea, 1.6–35 ng g^{-1} OC (mean: 8.0 ng g^{-1} OC) in the South Sea, and 2.9–23 ng g^{-1} OC (mean: 8.1 ng g^{-1} OC) in the East Sea (Fig. 2f and Table S8). Sedimentary AP concentrations did not exceed the sediment guidelines of the CCME (1000 μg kg^{-1} dw) (CCME, 1999), and were not at a level that could present an ecological risk. The distribution patterns of APs in offshore sediments were similar to those of SOs ($r = 0.75$, $p < 0.05$), suggesting the common sources and transport pathways for these PTSs. APs are considered to be indicators of sewage-derived organic matter, and are introduced to the marine environment through rivers (B. Li et al., 2015; Loos et al., 2007). The relatively higher concentrations of APs in the sediments of the Yellow Sea were attributed to the high influx of freshwater from rivers, similar to the case for SOs. The composition of APs in sediments indicated a high contribution of nonylphenol ethoxylates (NPEOs) (Yellow Sea: 46%; South Sea: 63%; East Sea: 83%). Nonylphenols are the degradation products of NPEOs; thus, the high proportion of NPEOs detected in this study likely enhanced nonylphenol concentrations and associated potential risks (Isobe et al., 2001).

3.3. Source identification of PTSs

Potential sources of sedimentary PCBs in South Korea have been described previously (Y.N. Kim et al., 2019). Major sources of PCBs in the offshore sediment of South Korea included a mixture of Aroclors 1016, Aroclors 1042, and Kanechlor 300, as well as combustion sources (Ikonomou et al., 2002; Y.N. Kim et al., 2019; Pedersen et al., 2015). However, it should be noted that the sources of PCBs found in offshore sediments in the present study differed slightly among the three seas (Fig. S4).

Further, a PMF model was used to identify the sources of PAHs in the offshore sediment of South Korea (Fig. 3a). Fluorene (Flu), fluoranthene (Fl), pyrene (Py), and benzo[*g,h,i*]perylene (BghiP) were the main components of Factor 1, and likely originated from vehicle emissions (Khalili et al., 1995; Nielsen, 1996). Anthracene (Ant) and Fl were identified as the main components of Factor 2, and likely originated from coke production (Harrison et al., 1996; Kong et al., 2012). The main components Factor 3 included benzo[*b*]fluoranthene (BbF) and BghiP, and likely originated from fossil fuel combustion (D. Yang et al., 2013). Using the three factors, we elucidated the sources for each site across the three seas (Fig. 3b). Sedimentary PAHs in the Yellow Sea were associated with coke production (77%), followed by fossil fuel combustion (14%) and vehicle emissions (9%). They likely originated from industrial complexes and coal-fired power plants located on the west coast of South Korea (H.G. Choi et al., 2011). The major sources of PAHs in the South Sea were likely vehicle emissions (68%), followed by fossil fuels (21%) and coke production (11%). Finally, sedimentary PAHs in the East Sea, likely originated from fossil fuels (49%) and vehicle emissions (48%).

Due to the high urbanization and industrialization of the southeastern areas of South Korea (including Busan, Ulsan, and Pohang), various anthropogenic pollutants could be introduced to the marine environment (H.G. Choi et al., 2011). Most fossil fuels and vehicle emissions of sedimentary PAHs in the East Sea could be explained by nearby sources. In the previous studies, biomass combustion sources did not contain or accounted for a very small proportion of high molecular weight PAHs, including BbF, benzo[*k*]fluoranthene, benzo[*a*]pyrene, dibenz[*a,h*]anthracene, BghiP, and IcdP (McGrath et al., 2001; Ravindra et al., 2008; Singh et al., 2013; B. Yang et al., 2013). Thus, the contribution of biomass combustion sources to PAHs in offshore sediments of South Korea is suggested to be small.

To identify the sources of SOs, the ratio of 2,4,6-triphenyl-1-hexene (ST1) and *cis*-1,2-diphenylcyclobutane (SD2) was used (Tian et al.,

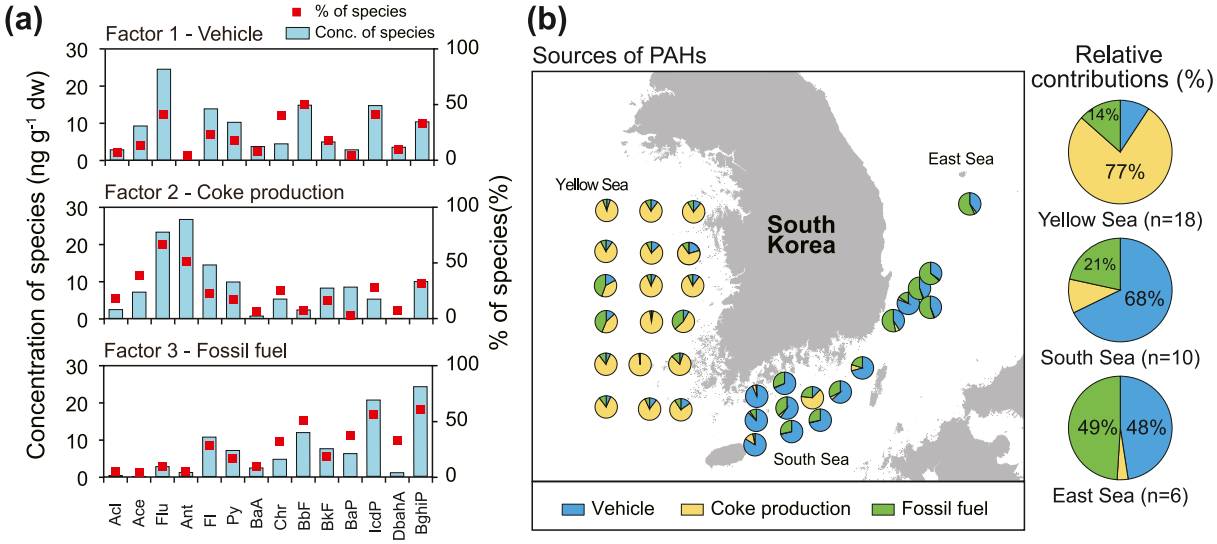


Fig. 3. Source identification and source contribution of PAHs in the sediments analyzed in the present study. (a) Factor profiles and (b) factor scores resulting from the PMF model used for identification of the potential sources of PAHs in the offshore sediments of South Korea.

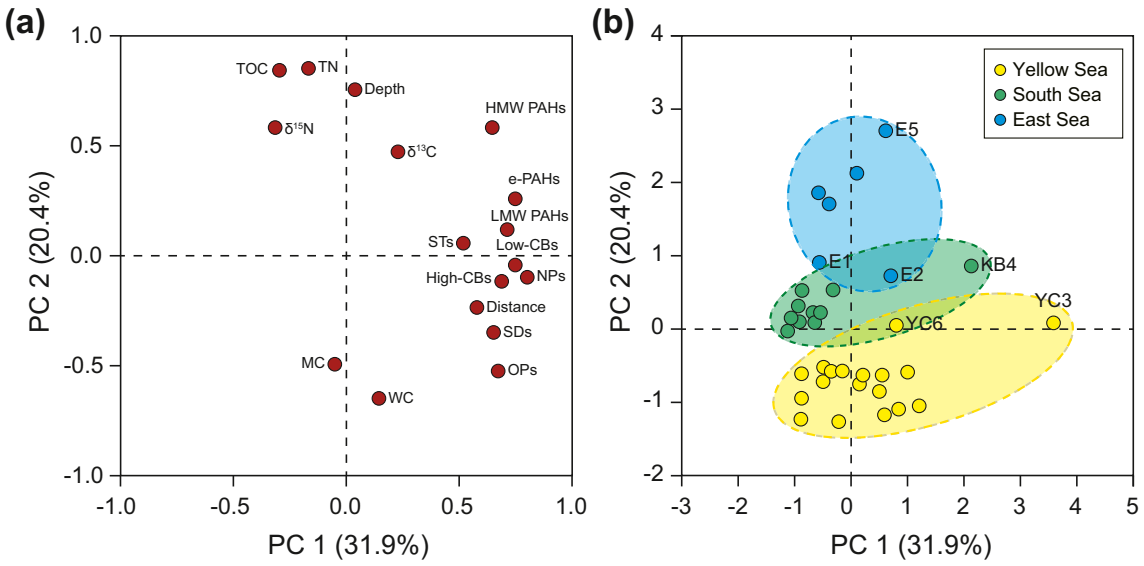


Fig. 4. PCA analysis based on physicochemical properties and PTS concentrations in the study area.

2020). If the ratio exceeds 2, they are considered to be aged expanded polystyrene (EPS); otherwise, they are considered to be fresh EPS. Most sites (94%) produced ST1:SD2 ratios exceeding 2 (Fig. S5); thus, aged EPS was the major source of SOs. AP sources were identified using the ratio of NPs and NPEOs (Isobe et al., 2001). If the ratio is less than 1, it means a fresh input of APs. When we calculated the NPs/(NP1EOs + NP2EOs) ratio, it was below 1 at all sites, indicating fresh inputs of APEOs. However, it should also be noteworthy that this exceptional result was attributed to the very low concentrations of APs with varied compositions.

Last, PCA was used with the physicochemical properties and individual PTS compounds to elucidate regional characteristics. PC1 represented 31.9% of the total variance, while PC2 represented 20.4%. Together, these components explained 52.3% of the total variance (Fig. 4). PC1 had a high positive correlation with PTSs and a weak negative correlation to OC, TN, depth, and $\delta^{13}C$ (Fig. 4a). Meantime, PC2 had a high positive correlation with HMW-PAHs and some other parameters (depth, $\delta^{13}C$, $\delta^{15}N$, OC, and TN), but a weak positive correlation with e-PAHs, low molecular weight PAHs (LMW-PAHs), and STs. There was a strong negative correlation with octylphenols (OPs) and MC, water content (WC), and SDs, while NPs and PCBs had weak negative correlations. In general, PCA groups across the environmental parameters and PTSs detected in offshore sediments of South Korea reflected oceanographic settings of the three seas (Fig. 4b). For example, the Yellow Sea group was located in a negative direction at PC2, and was more affected by PCBs, STs, and APs compared to the other seas. Station YC3 in the Yellow Sea group had a higher correlation with PC1 compared to the other stations, indicating that it was significantly affected by PTSs. Station YC3, which was significantly affected by PTSs, was located within the waste dumpsite, and showed higher degree of pollution than the other sites in the Yellow Sea (Kim et al., 2009). Of note, the South Sea group was located between the Yellow Sea and the East Sea groups. In the South Sea group, KB4 was distinguished from other stations. This station had a strong positive correlation with PC1, which was attributed to it being located closer to industrial cities compared to the other South Sea stations (Han et al., 2016). The East Sea group was located in the positive direction of PC2 compared to the other two groups. The East Sea had high OC and TN content, with PAHs and STs having potentially strong effects.

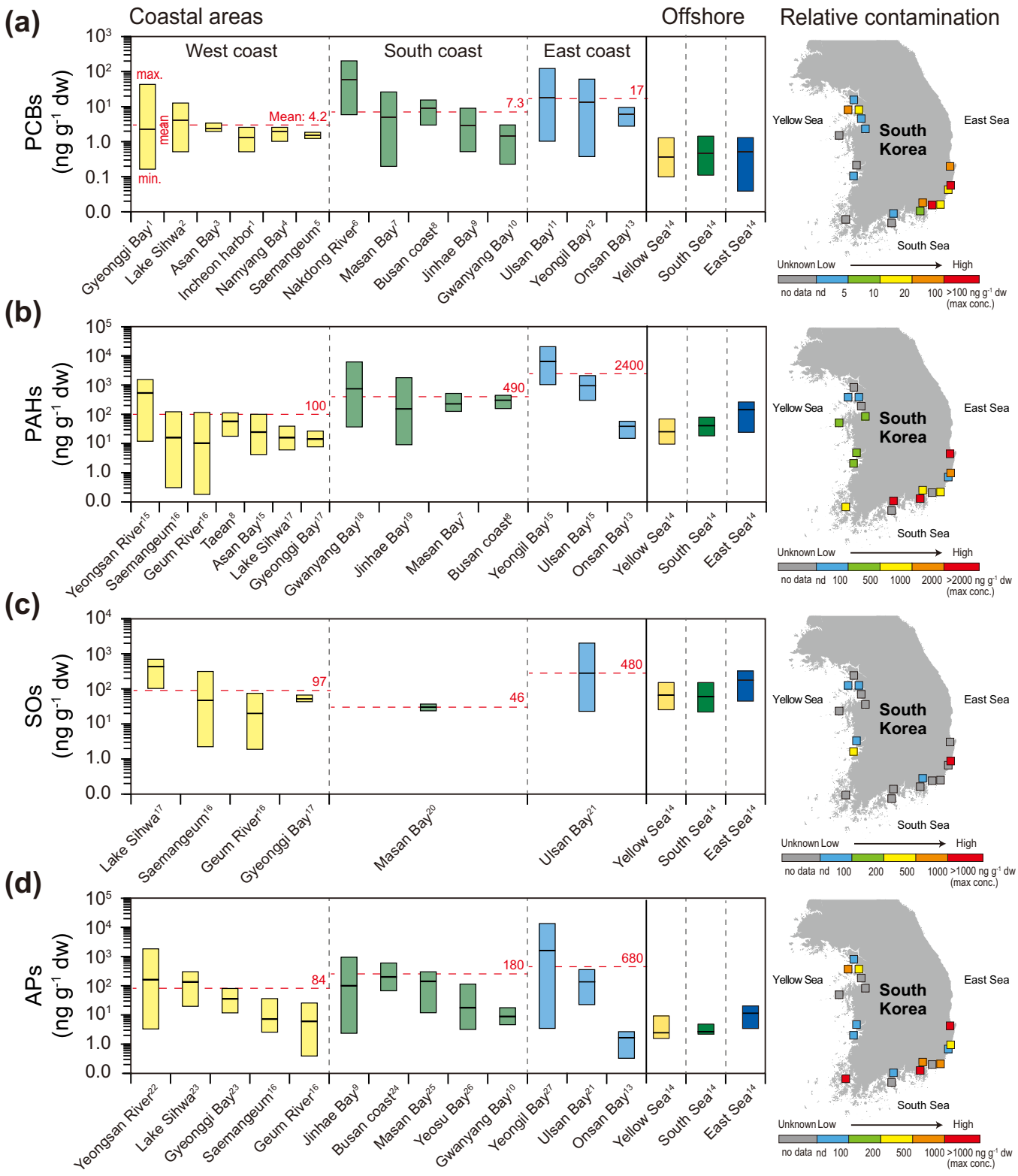
3.4. Comparison with previous studies

The concentrations of PTSs in offshore sediments obtained from the present study were compared to previously reported data in the coastal areas of South Korea (An et al., 2020; Cho et al., 2004; Choi et al., 2001, 2007, 2009, 2011a, 2011b, 2014; Hong et al., 2005, 2009, 2010, 2014, 2016a; Jeong et al., 2001; Jung et al., 2012; Khim et al., 1999; Kim et al., 2014; Koh et al., 2002, 2005; Lee et al., 2001, 2020; Woo et al., 2006; Yim et al., 2007, 2014; Yoon et al., 2017) (Fig. 5) and other countries (Table 1). To facilitate comparison with previous studies, the PTS concentrations in this study were expressed as dry weight basis. PCB concentrations in the Yellow Sea and the South Sea in the present study were relatively lower or similar to those previously reported in the Yellow Sea (Duan et al., 2013) and the Korea Strait (Guerra et al., 2019). PCB concentrations in our study were similar or lower compared to those documented in the Adriatic Sea, the Barents Sea, the Bohai Sea, the Mediterranean Sea, and the Indian Sea (Cheng et al., 2015; Combi et al., 2016; Mandalakis et al., 2014; Wang et al., 2015; Zaborska et al., 2011). However, PCB concentrations were higher in the Baltic Sea (Sobek et al., 2015) compared to the Yellow Sea and the South Sea, and were similar to those in the East Sea.

PAHs concentration in the Yellow Sea was previously reported as 90 ng g^{-1} dw (Liu et al., 2012), on average, but was comparatively lower in the present study (44 ng g^{-1} dw). However, PAH concentrations in the Yellow Sea and South Sea were generally higher than those of the Mediterranean Sea (Mandalakis et al., 2014). PAHs in the Bohai Sea, the White Sea, and the Barents Sea were similar or lower than the Yellow Sea and the South Sea (Hu et al., 2011; Savinov et al., 2000; Zaborska et al., 2011). PAH concentrations in the Adriatic Sea, the Kara Sea, and the South China Sea were 2–3 times higher compared to those in the Yellow Sea and South Sea, but were lower compared to the East Sea (Table 1).

SO concentrations were compared with previous studies that conducted at relatively far distances from mainland, because few studies have been conducted on offshore area. All three sea areas in the present study had higher SO concentrations compared to those reported in the previous studies. In particular, the East Sea had SO concentrations that were more than 100 times higher than those reported in the previous studies (Hong et al., 2016a; Tian et al., 2020; Yoon et al., 2017, 2019). Despite the large number of target compounds, AP concentrations were lower in the Yellow Sea and the South Sea in this study compared to previous studies. AP concentrations were higher in the East Sea compared to the Adriatic and Baltic seas, but were lower compared to previous studies conducted in the Pearl River Estuary (China) and the South China Sea.

Relatively higher PTS pollution levels in the East Sea were attributed to a high pollution state in the corresponding coastal region. This region



References

[1] Lee et al. (2001); [2] Hong et al. (2009); [3] Khim et al. (1999); [4] Choi et al. (2011b); [5] Choi et al. (2014); [6] Jeong et al. (2001); [7] Jung et al. (2012); [8] Choi et al. (2011a); [9] Hong et al. (2014); [10] Koh et al. (2005); [11] Choi et al. (2001); [12] Hong et al. (2005); [13] Koh et al. (2002); [14] This study; [15] Yim et al. (2007); [16] Yoon et al. (2017); [17] Hong et al. (2016a); [18] Woo et al. (2006); [19] Yim et al. (2014); [20] Lee et al. (2020); [21] An et al. (2020); [22] Choi et al. (2007); [23] Hong et al. (2010); [24] Choi et al. (2009); [25] Hong et al. (2009); [26] Cho et al. (2004); [27] Kim et al. (2014).

Fig. 5. Comparison between concentrations of (a) PCBs, (b) PAHs, (c) SOs, and (d) APs in sediments of coastal areas and three seas of South Korea.

Table 1
PTS concentrations in the offshore sediments of South Korea in comparison to previously reported data.

PTSs	Sampling regions	Sampling year	#of site	#of target chemicals	Concentration (ng g^{-1} dw)			References
					Min	Max	Mean	
PCBs	Adriatic Sea	2014	48	5	<LOD	4.3	1.0	Combi et al. (2016)
	Baltic Sea	2010	11	155	0.7	28	2.1	Sobek et al. (2015)
	Barents Sea	2000	92	7	<LOD	3.5	0.93	Zaborska et al. (2011)
	Bohai Sea	2012–2013	34	37	0.16	1.7	0.53	Wang et al. (2015)
	Mediterranean Sea	2006–2011	52	33	0.04	1.2	0.26	Mandalakis et al. (2014)
	Indian Ocean	2011	10	32	0.14	0.51	0.23	Cheng et al. (2015)
	Korea Strait	2010	1	209	–	–	0.65	Guerra et al. (2019)
	Yellow Sea	2011	42	24	0.01	3.1	0.72	Duan et al. (2013)
	Yellow Sea	2017	18	31	0.1	1.3	0.37	This study
	South Sea	2018	10	31	0.10	1.1	0.49	This study
	East Sea	2017	6	31	0.29	4.9	2.3	This study
PAHs	Adriatic Sea	2014	48	16	39	570	170	Combi et al. (2016)
	Barents Sea	2000	4	12	35	130	83	Zaborska et al. (2011)
	Bohai Sea	2006	2	16	32	200	92	Hu et al. (2011)
	Mediterranean Sea	2006–2011	52	16	6	41	26	Mandalakis et al. (2014)
	Kara Sea	1993–1994	10	30	70	260	170	Dahle et al. (2003)
	South China Sea	1993	11	10	25	280	150	Yang (2000)
	White Sea	1994	11	27	16	230	96	Savinov et al. (2000)
	Yellow Sea	2007	8	18	27	220	90	Liu et al. (2012)
	Yellow Sea	2017	18	24	14	150	44	This study
	South Sea	2018	10	24	31	150	75	This study
	East Sea	2017	6	24	40	700	340	This study
SOs	Arabian Gulf	2017	3	10	0.9	6.0	2.8	Yoon et al. (2019)
	Gyeonggi Bay	2015	4	10	20	35	25	Hong et al. (2016a)
	Saemangeum Coast	2014	28	10	0.4	260	22	Yoon et al. (2017)
	Lake Sihwa	2015–2016	9	10	<LOD	150	57	Tian et al. (2020)
	Yellow Sea	2017	18	10	38	250	100	This study
	South Sea	2018	10	10	38	240	94	This study
	East Sea	2017	6	10	69	550	290	This study
APs	Adriatic Sea	2014	48	1	<LOD	40	10	Combi et al. (2016)
	Baltic Sea	2011–2012	5	2	<LOD	270	8.9	Koniecko et al. (2014)
	South China Sea	2002	24	2	61	580	270	Chen et al. (2005)
	Yellow Sea	2017	18	6	3.1	20	5.7	This study
	South Sea	2018	10	6	4.7	8.4	5.9	This study
	East Sea	2017	6	6	6.0	27	17	This study

<LOD: limit of detection.

is considered to be the main source of PTS pollutions, as the industrial and municipal activities are highly concentrated in the adjacent inland cities and bays (Hong et al., 2014; Khim et al., 2001; Koh et al., 2002; Moon et al., 2001; Yim et al., 2007). In particular, the Ulsan Bay and Yeongil Bay showed the highest levels of PTS contamination in sediments of South Korea (Fig. 5). Overall, contamination of PTSs in offshore sediments reported in the present study was lower than those of coastal areas in South Korea, but PTSs tended to accumulate in mud dominant areas near hotspots.

The compositions of PCBs and PAHs in offshore sediments of South Korea were compared with other regions using PCA (Fig. S6). The compositions of sedimentary PCBs in South Korea were similar to those of the Adriatic Sea (Combi et al., 2016), Bohai Sea (Wang et al., 2015), Indian Ocean (Cheng et al., 2015), and Korea Strait (Guerra et al., 2019), in general. The compositional patterns of PCBs in some stations, such as YB6 and YC6, were found to be similar to that of a previous study conducted in the Yellow Sea (Duan et al., 2013). However, the compositions of sedimentary PCBs in this study were somewhat different from the Mediterranean Sea (Mandalakis et al., 2014). The compositions of PAHs in sediments showed a region-specific pattern. East Sea was located in the positive direction of PC1 compared to the Yellow Sea. Yellow Sea and South Sea were located in the positive direction of PC2, while the East Sea was located in the middle. These results indicated that the East Sea was more affected by HMW-PAHs compared to the other two seas. The compositions of sedimentary PAHs in the Yellow Sea were similar to those of the Bohai Sea (Hu et al., 2011), South China Sea (Yang, 2000), and Kara Sea (Dahle et al., 2003). PAHs compositions of the East Sea were similar to the Mediterranean Sea (Mandalakis et al., 2014) and the White Sea (Savinov et al., 2000). This difference is thought to be due to the region-specific sources of PAHs in each sea area.

4. Conclusions

The present study is novel in that the distribution characteristics and sources of traditional PTSs (PCBs, PAHs, and APs) and emerging PTSs (SOs and e-PAHs) in offshore sediments were extensively investigated in South Korea. The distribution of PTSs fairly well reflected the oceanographic settings and physical environments of each sea and proximate sources in a site-specific manner. Relatively high concentrations of PTSs were observed in the mud deposit area of the Yellow Sea, which was attributed to their hydrodynamic characteristics. In comparison, the South Sea and the East Sea were seemingly influenced by nearby coastal and/or inland sources. PCBs originated from a mixture of industrial products and combustion. PAH sources differed with sea area. The dominant source of PAHs was coke production in the Yellow Sea, vehicle emissions in the South Sea, and fossil fuel combustion in the East Sea. When compared with previous studies conducted on offshore sediments in other countries, the Yellow Sea and the South Sea had relatively low traditional PTSs levels. In contrast, PTS levels were comparatively higher in the East Sea compared to other seas. Due to insufficient monitoring studies in offshore sediments globally, comparisons of e-PTSs were difficult to obtain. Therefore, more monitoring studies are required to elucidate the distributions and fate of various PTSs and their impacts on the marine ecosystem in the future. The results of this study are expected to provide baseline information for PTSs pollution in offshore marine environments in South Korea and elsewhere in the world.

CRediT authorship contribution statement

Youngnam Kim: Conceptualization, Investigation, Formal analysis, Statistical analyses, Visualization, Writing – original draft. **Seongjin**

Hong: Conceptualization, Writing – original draft, Writing – review & editing, Visualization, Project administration, Funding acquisition, Supervision. **Junghyun Lee:** Conceptualization, Writing – review & editing, Visualization. **Seo Joon Yoon:** Investigation, Formal analysis. **Yoonyoung An:** Investigation, Formal analysis. **Min-Seob Kim:** Investigation, Formal analysis. **Hee-Dong Jeong:** Investigation, Formal analysis. **Jong Seong Khim:** Conceptualization, Writing – review & editing, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.147996.

References

Ahn, I.-Y., Kang, Y.-C., Choi, J.-W., 1995. The influence of industrial effluents on intertidal benthic communities in Panweol, Kyeonggi Bay (Yellow Sea) on the west coast of Korea. Mar. Pollut. Bull. 30, 200–206.

Amamiya, K., Saido, K., Chung, S.-Y., Hiaki, T., Lee, D.S., Kwon, B.G., 2019. Evidence of transport of styrene oligomers originated from polystyrene plastic to oceans by runoff. Sci. Total Environ. 667, 57–63.

An, Y., Hong, S., Yoon, S.J., Cha, J., Shin, K.-H., Khim, J.S., 2020. Current contamination status of traditional and emerging persistent toxic substances in the sediments of Ulsan Bay, South Korea. Mar. Pollut. Bull. 160, 111560.

Buchanan, J.B., 1984. Sediment analysis. In: Holme, N.A., McIntyre, A.D. (Eds.), Methods for the Study of Marine Benthos. Blackwell, Oxford, pp. 41–65.

Canadian Council of Ministers of the Environment (CCME), 1999. Canadian sediment quality guidelines for the protection of aquatic life: Polycyclic aromatic hydrocarbons (PAHs). Canadian Environmental Quality Guidelines. Canadian Council of Ministers of the Environment, Winnipeg.

Canadian Council of Ministers of the Environment (CCME), 2002. Canadian Sediment Quality Guidelines for the Protection of Aquatic Life Summary Tables. CCME, Winnipeg, MB.

Cha, J., Hong, S., Kim, J., Lee, J., Yoon, S.J., Lee, S., Moon, H.-B., Shin, K.-H., Hur, J., Giesy, J.P., 2019. Major AhR-active chemicals in sediments of Lake Sihwa, South Korea: application of effect-directed analysis combined with full-scan screening analysis. Environ. Int. 133, 105199.

Chen, B., Mai, B.-X., Duan, J.-C., Luo, X.-J., Yang, Q.-S., Sheng, G.-Y., Fu, J.-M., 2005. Concentrations of alkylphenols in sediments from the Pearl River estuary and South China Sea, South China. Mar. Pollut. Bull. 50, 993–997.

Cheng, Z., Lin, T., Xu, W., Xu, Y., Li, J., Luo, C., Zhang, G., 2015. A preliminary assessment of polychlorinated biphenyls and polybrominated diphenyl ethers in deep-sea sediments from the Indian Ocean. Mar. Pollut. Bull. 94, 323–328.

Cho, H.S., Kim, Y.O., Seol, S.U., 2004. A study on the pollution of nonylphenol in surface sediment in Gwangyang bay and Yeosu sound. Environ. Int. 13, 561–570.

Choi, J., Matsuda, M., Kawano, M., Min, B., Wakimoto, T., 2001. Accumulation profiles of persistent organochlorines in waterbirds from an estuary in Korea. Arch. Environ. Contam. Toxicol. 41, 353–363.

Choi, M.-K., Choi, H.-G., Moon, H.-B., Yu, J., Kang, S.-K., Choi, S.-K., 2007. Sources and distributions of organic wastewater compounds on the Mokpo Coast of Korea. Fish. Aquat. Sci. 10, 205–214.

Choi, M.-K., Moon, H.-B., Yu, J., Eom, J.-Y., Choi, H.-G., 2009. Butyltin contamination in industrialized bays associated with intensive marine activities in Korea. Arch. Environ. Contam. Toxicol. 57, 77–85.

Choi, H.G., Moon, H.-B., Choi, M., Yu, J., 2011a. Monitoring of organic contaminants in sediments from the Korean coast: spatial distribution and temporal trends (2001–2007). Mar. Pollut. Bull. 62, 1352–1361.

Choi, J.-Y., Lee, S.-G., Bang, J.-H., Yang, D.-B., Hong, G.-H., Shin, K.-H., 2011b. On the distribution of PCBs and organochlorine pesticides in fish and sediment of the Asan Bay. Ocean Polar Res. 33, 45–53.

Choi, J.-Y., Yang, D.-B., Hong, G.H., Shin, K.H., 2014. Distribution and bioaccumulation of polychlorinated biphenyls and organochlorine pesticides residues in sediments and Manila clams (Ruditapes philippinarum) from along the Mid-Western coast of Korea. Mar. Pollut. Bull. 85, 672–678.

Combi, T., Pintado-Herrera, M.G., Lara-Martin, P.A., Miserocchi, S., Langone, L., Guerra, R., 2016. Distribution and fate of legacy and emerging contaminants along the Adriatic Sea: a comparative study. Environ. Pollut. 218, 1055–1064.

Dachs, J., Lohmann, R., Ockenden, W.A., Méjanelle, L., Eisenreich, S.J., Jones, K.C., 2002. Oceanic biogeochemical controls on global dynamics of persistent organic pollutants. Environ. Sci. Technol. 36, 4229–4237.

Dahle, S., Savinov, V.M., Matishov, G.G., Evenset, A., Næs, K., 2003. Polycyclic aromatic hydrocarbons (PAHs) in bottom sediments of the Kara Sea shelf, Gulf of Ob and Yenisei Bay. Sci. Total Environ. 306, 57–71.

Duan, X., Li, Y., Li, X., Zhang, D., Li, M., 2013. Polychlorinated biphenyls in sediments of the Yellow Sea: distribution, source identification and flux estimation. Mar. Pollut. Bull. 76, 283–290.

Fine, P.M., Cass, G.R., Simoneit, B.R., 2001. Chemical characterization of fine particle emissions from fireplace combustion of woods grown in the northeastern United States. Environ. Sci. Technol. 35, 2665–2675.

Fine, P.M., Cass, G.R., Simoneit, B.R., 2002. Chemical characterization of fine particle emissions from the fireplace combustion of woods grown in the southern United States. Environ. Sci. Technol. 36, 1442–1451.

Galbán-Malagón, C., Berrojalbiz, N., Ojeda, M.-J., Dachs, J., 2012. The oceanic biological pump modulates the atmospheric transport of persistent organic pollutants to the Arctic. Nat. Commun. 3, 1–9.

Gibson, T.L., Smart, V.B., Smith, L.L., 1978. Non-enzymic activation of polycyclic aromatic hydrocarbons as mutagens. Mutat. Res. Mol. Mut. 49, 153–161.

Gong, X., Xiao, L., Zhao, Z., Li, Q., Feng, F., Zhang, L., Deng, Z., 2018. Spatial variation of polycyclic aromatic hydrocarbons (PAHs) in surface sediments from rivers in hilly regions of Southern China in the wet and dry seasons. Ecotoxicol. Environ. Saf. 156, 322–329.

Guerra, R., Pasteris, A., Righi, S., Ok, G., 2019. Historical record of polychlorinated biphenyls (PCBs) in the continental shelf of the Korea Strait. Chemosphere 237, 124438.

Han, G.M., Hong, S.H., Shim, W.J., Ra, K.T., Kim, K.T., Ha, S.Y., Jang, M., Kim, G.B., 2016. Assessment of persistent organic and heavy metal contamination in Busan coast: application of sediment quality index. Ocean Polar Res. 38, 171–184.

Harrison, R.M., Smith, D., Luhana, L., 1996. Source apportionment of atmospheric polycyclic aromatic hydrocarbons collected from an urban location in Birmingham, UK. Environ. Sci. Technol. 30, 825–832.

Hong, S.H., Yim, U.H., Shim, W.J., Oh, J.R., 2005. Congener-specific survey for polychlorinated biphenlys in sediments of industrialized bays in Korea: regional characteristics and pollution sources. Environ. Sci. Technol. 39, 7380–7388.

Hong, S., Yim, U., Shim, W., Li, D., Oh, J., 2006. Nationwide monitoring of polychlorinated biphenyls and organochlorine pesticides in sediments from coastal environment of Korea. Chemosphere 64, 1479–1488.

Hong, S.H., Munschy, C., Kannan, N., Tixier, C., Tronczynski, J., Héas-Moisan, K., Shim, W.J., 2009. PCDD/F, PBDE, and nonylphenol contamination in a semi-enclosed bay (Masan Bay, South Korea) and a Mediterranean lagoon (Thau, France). Chemosphere 77, 854–862.

Hong, S., Won, E.-J., Ju, H.-J., Kim, M.-S., Shin, K.-H., 2010. Current nonylphenol pollution and the past 30 years record in an artificial Lake Shihwa, Korea. Mar. Pollut. Bull. 60, 308–313.

Hong, S.H., Han, G.M., Yim, U.H., Lim, D.-i, Ha, S.Y., Kim, N.S., Shim, W.J., 2014. Integrative assessment of sediment quality in terms of chemical contamination in Jinhae Bay, South Korea. Ocean Sci. J. 49, 265–278.

Hong, S., Lee, J., Lee, C., Yoon, S.J., Jeon, S., Kwon, B.-O., Lee, J.-H., Giesy, J.P., Khim, J.S., 2016a. Are styrene oligomers in coastal sediments of an industrial area aryl hydrocarbon-receptor agonists? Environ. Pollut. 213, 913–921.

Hong, S., Yoon, S.J., Lee, Y., Khim, J.S., 2016b. Chaper 8. Persistent toxic substances in sediments of Korean coastal waters: a review. In: Loganathan, et al. (Eds.), Persistent Organic Chemicals in the Environment: Status and Trends in the Pacific Basin Countries I ACS Symposium Series. American Chemical Society, Washington, DC 2016.

Hu, L., Guo, Z., Shi, X., Qin, Y., Lei, K., Zhang, G., 2011. Temporal trends of aliphatic and polyaromatic hydrocarbons in the Bohai Sea, China: evidence from the sedimentary record. Org. Geochem. 42, 1181–1193.

Huang, Y.-J., Lee, C.-L., Fang, M.-D., 2011. Distribution and source differentiation of PAHs and PCBs among size and density fractions in contaminated harbor sediment particles and their implications in toxicological assessment. Mar. Pollut. Bull. 62, 432–439.

Ikonomou, M.G., Sather, P., Oh, J.-E., Choi, W.-Y., Chang, Y.-S., 2002. PCB levels and congener patterns from Korean municipal waste incinerator stack emissions. Chemosphere 49, 205–216.

Isobe, T., Nishiyama, H., Nakashima, A., Takada, H., 2001. Distribution and ehavior of nonylphenol, octylphenol, and nonylphenol monoethoxylate in Tokyo metropolitan area: their association with aquatic particles and sedimentary distributions. Environ. Sci. Technol. 35, 1041–1049.

Jeong, G.H., Kim, H.J., Joo, Y.J., Kim, Y.B., So, H.Y., 2001. Distribution characteristics of PCBs in the sediments of the lower Nakdong River, Korea. Chemosphere 44, 1403–1411.

Jung, J.-H., Hong, S.H., Yim, U.H., Ha, S.Y., Shim, W.J., Kannan, N., 2012. Multiple in vitro bioassay approach in sediment toxicity evaluation: Masan Bay, Korea. Bull. Environ. Contam. Toxicol. 89, 32–37.

Kannan, K., Johnson-Restrepo, B., Yohn, S.S., Giesy, J.P., Long, D.T., 2005. Spatial and temporal distribution of polycyclic aromatic hydrocarbons in sediments from Michigan inland lakes. Environ. Sci. Technol. 39, 4700–4706.

Khalili, N.R., Scheff, P.A., Holsen, T.M., 1995. PAH source fingerprints for coke ovens, diesel and, gasoline engines, highway tunnels, and wood combustion emissions. Atmos. Environ. 29, 533–542.

Khim, J.S., Hong, S., 2014. Assessment of trace pollutants in Korean coastal sediments using the triad approach: a review. Sci. Total Environ. 470–471, 1450–1462.

Khim, J.S., Kannan, K., Villeneuve, D.L., Koh, C.H., Giesy, J.P., 1999. Characterization and distribution of trace organic contaminants in sediment from Masan Bay, Korea. 1. Instrumental analysis. Environ. Sci. Technol. 33, 4199–4205.

Khim, J.S., Lee, K., Kannan, K., Villeneuve, D., Giesy, J., Koh, C., 2001. Trace organic contaminants in sediment and water from Ulsan Bay and its vicinity, Korea. Arch. Environ. Contam. Toxicol. 40, 141–150.

Khim, J.S., Lee, C., Song, S.J., Bae, H., Noh, J., Lee, J., Kim, H.G., Choi, J.W., 2021. Marine biodiversity in Korea: a review on macrozoobenthic assemblages, distributions, and human impacts on long-term community changes. Oceanogr. Mar. Biol. Annu. Rev. (In press).

Kim, P.-G., Park, M.-E., Sung, K.-Y., 2009. Distribution of heavy metals in marine sediments at the ocean waste disposal site in the Yellow Sea, South Korea. Geosci. J. 13, 15–24.

Kim, S., Lee, S., Kim, C., Liu, X., Seo, J., Jung, H., Ji, K., Hong, S., Park, J., Khim, J.S., 2014. In vitro and in vivo toxicities of sediment and surface water in an area near a major steel industry of Korea: endocrine disruption, reproduction, or survival effects combined with instrumental analysis. Sci. Total Environ. 470, 1509–1516.

Kim, D., Kim, J.-H., Kim, M.-S., Ra, K., Shin, K.-H., 2018. Assessing environmental changes in Lake Shihwa, South Korea, based on distributions and stable carbon isotopic compositions of n-alkanes. Environ. Pollut. 240, 105–115.

Kim, J., Hong, S., Cha, J., Lee, J., Kim, T., Lee, S., Moon, H.-B., Shin, K.-H., Hur, J., Lee, J.-S., Giesy, J.P., Khim, J.S., 2019a. Newly identified AHR-active compounds in the sediments of an industrial area using effect-directed analysis. Environ. Sci. Technol. 53, 10043–10052.

Kim, Y.N., An, Y., Yoon, S.J., Jeong, H.D., Khim, J.S., Hong, S., 2019b. Spatial distributions of polychlorinated biphenyls (PCBs) in surface sediments of coastal and offshore areas of South Korea. J. Hydrol. Soc. Kor. 8, 19–27.

Koh, C.-H., Khim, J.S., Villeneuve, D.L., Kannan, K., Giesy, J.P., 2002. Analysis of trace organic contaminants in sediment, pore water, and water samples from Onsan Bay, Korea: instrumental analysis and in vitro gene expression assay. Environ. Toxicol. Chem. 21, 1796–1803.

Koh, C.-H., Khim, J.S., Villeneuve, D.L., Kannan, K., Johnson, B.G., Giesy, J.P., 2005. Instrumental and bioanalytical measures of dioxin-like and estrogenic compounds and activities associated with sediment from the Korean coast. Ecotoxicol. Environ. Saf. 61, 366–379.

Kong, S., Lu, B., Ji, Y., Bai, Z., Xu, Y., Liu, Y., Jiang, H., 2012. Distribution and sources of polycyclic aromatic hydrocarbons in size-differentiated re-suspended dust on building surfaces in an oilfield city, China. Atmos. Environ. 55, 7–16.

Koniecko, I., Staniszewska, M., Falkowska, L., Burska, D., Kielczewska, J., Jasinska, A., 2014. Alkylphenols in surface sediments of the Gulf of Gdansk (Baltic Sea). Water Air Soil Pollut. 225, 2040.

Kwon, B.G., Saido, K., Koizumi, K., Sato, H., Ogawa, N., Chung, S.-Y., Kusui, T., Kodera, Y., Kogure, K., 2014. Regional distribution of styrene analogues generated from polystyrene degradation along the coastlines of the North-East Pacific Ocean and Hawaii. Environ. Pollut. 188, 45–49.

Kwon, B.G., Koizumi, K., Chung, S.-Y., Kodera, Y., Kim, J.-O., Saido, K., 2015. Global styrene oligomers monitoring as new chemical contamination from polystyrene plastic marine pollution. J. Hazard. Mater. 300, 359–367.

Larsen, R.K., Baker, J.E., 2003. Source apportionment of polycyclic aromatic hydrocarbons in the urban atmosphere: a comparison of three methods. Environ. Sci. Technol. 37 (9), 1873–1881.

LaVoie, E.J., Tulley-Freiler, L., Bedenko, V., Hoffmann, D., 1983. Mutagenicity of substituted phenanthrenes in Salmonella typhimurium. Mutat. Res. Genet. Toxicol. 116, 91–102.

Ledesma, E., Kalish, M., Nelson, P., Wornat, M., Mackie, J., 2000. Formation and fate of PAH during the pyrolysis and fuel-rich combustion of coal primary tar. Fuel 79, 1801–1814.

Lee, K.-T., Tanabe, S., Koh, C.-H., 2001. Contamination of polychlorinated biphenyls (PCBs) in sediments from Kyeonggi Bay and nearby areas, Korea. Mar. Pollut. Bull. 42, 273–279.

Lee, T., Hyun, J.-H., Mok, J.S., Kim, D., 2008. Organic carbon accumulation and sulfate reduction rates in slope and basin sediments of the Ulleung Basin, East/Japan Sea. Geo-Mar. Lett. 28, 153–159.

Lee, J., Hong, S., Yoon, S.J., Kwon, B.-O., Ryu, J., Giesy, J.P., Allam, A.A., Al-Khedhairy, A.A., Khim, J.S., 2017. Long-term changes in distributions of dioxin-like and estrogenic compounds in sediments of Lake Sihwa, Korea: revisited mass balance. Chemosphere 181, 767–777.

Lee, J., Hong, S., Kim, T., Lee, C., An, S.-A., Kwon, B.-O., Lee, S., Moon, H.-B., Giesy, J.P., Khim, J.S., 2020. Multiple bioassays and targeted and nontargeted analyses to characterize potential toxicological effects associated with sediments of Masan Bay: focusing on AhR-mediated potency. Environ. Sci. Technol. 54, 4443–4454.

Li, J., Dong, H., Zhang, D., Han, B., Zhu, C., Liu, S., Liu, X., Ma, Q., Li, X., 2015a. Sources and ecological risk assessment of PAHs in surface sediments from Bohai Sea and northern part of the Yellow Sea, China. Mar. Pollut. Bull. 96, 485–490.

Li, B., Cao, J., Xing, C., Wang, Z., Cui, L., 2015b. Assessing estrogenic activity and reproductive toxicity of organic extracts in WWTP effluents. Environ. Toxicol. Pharmacol. 39, 942–952.

Liénart, C., Savoye, N., Bozec, Y., Breton, E., Conan, P., David, V., Feunteun, E., Grangeré, K., Kerhervé, P., Lebreton, B., Lefebvre, S., L'Helguen, S., Mousseau, L., Raimbault, P., Richard, P., Riera, P., Sauriau, P.-G., Schaal, G., Aubert, F., Aubin, S., Bichon, S., Boinet, C., Bourasseau, L., Bréret, M., Caparros, J., Cariou, T., Charlier, K., Claquin, P., Cornille, V., Corre, A.-M., Costes, L., Crispi, O., Crouvoisier, M., Czamanski, M., Del Amo, Y., Derriennic, H., Dindinaud, F., Durozier, M., Hanquiez, V., Nowaczyk, A., Devesa, J., Ferreira, S., Fornier, M., Garcia, F., Garcia, N., Geslin, S., Grossteffan, E., Gueux, A., Guillaudeau, J., Guillou, G., Joly, O., Lachaussée, N., Lafont, M., Lamoureux, J., Lecuyer, E., Lehodey, J.-P., Lemeille, D., Leroux, C., Macé, E., Maria, E., Pineau, P., Petit, F., Pujo-Pay, M., Rimelin-Maury, P., Sultan, E., 2017. Dynamics of particulate organic matter composition in coastal systems: a spatio-temporal study at multi-systems scale. Prog. Oceanogr 156, 221–239.

Lin, T., Hu, L., Guo, Z., Qin, Y., Yang, Z., Zhang, G., Zheng, M., 2011. Sources of polycyclic aromatic hydrocarbons to sediments of the Bohai and Yellow Seas in East Asia. J. Geophys. Res.-Atmos. 116, D23305.

Lin, Y., Deng, W., Li, S., Li, J., Wang, G., Zhang, D., Li, X., 2017. Congener profiles, distribution, sources and ecological risk of parent and alkyl-PAHs in surface sediments of Southern Yellow Sea, China. Sci. Total Environ. 580, 1309–1317.

Liu, L.-Y., Wang, J.-Z., Wei, G.-L., Guan, Y.-F., Zeng, E.Y., 2012. Polycyclic aromatic hydrocarbons (PAHs) in continental shelf sediment of China: implications for anthropogenic influences on coastal marine environment. Environ. Pollut. 167, 155–162.

Loos, R., Hanke, G., Umlauf, G., Eisenreich, S.J., 2007. LC–MS–MS analysis and occurrence of octyl-and nonylphenol, their ethoxylates and their carboxylates in Belgian and Italian textile industry, waste water treatment plant effluents and surface waters. Chemosphere 66, 690–699.

Mandalakis, M., Polymenakou, P.N., Tselepides, A., Lampadariou, N., 2014. Distribution of aliphatic hydrocarbons, polycyclic aromatic hydrocarbons and organochlorinated pollutants in deep-sea sediments of the southern Cretan margin, eastern Mediterranean Sea: a baseline assessment. Chemosphere 106, 28–35.

McGrath, T., Sharma, R., Hajaligol, M., 2001. An experimental investigation into the formation of polycyclic-aromatic hydrocarbons (PAH) from pyrolysis of biomass materials. Fuel 80, 1787–1797.

Mimura, J., Fujii-Kuriyama, Y., 2003. Functional role of AhR in the expression of toxic effects by TCDD. Biochim. Biophys. Acta Gen. Subj. 1619, 263–268.

Ministry of Environment (MOE), 2008. Enforcement Rules of the Environmental Impact Assessment Act. Ministry of Environment, Republic of Korea Available at. http://www.me.go.kr/ (last updated in 2020).

Ministry of Environment (MOE), 2014. Designation of Restricted Substances and Prohibited Substances. Ministry of Environment, Republic of Korea Available at. http://www.me.go.kr/ (last updated in 2018).

Montuori, P., Aurino, S., Garzonio, F., Sarnacchiaro, P., Nardone, A., Triassi, M., 2016. Distribution, sources and ecological risk assessment of polycyclic aromatic hydrocarbons in water and sediments from Tiber River and estuary, Italy. Sci. Total Environ. 566, 1254–1267.

Moon, H.-B., Park, H.-G., Kim, S.-S., Lee, P.-Y., 2001. Level and origin of polycyclic aromatic hydrocarbons (PAHs) in sediments from Ulsan Bay, Korea. Envrion. Sci. Bull. Kor. Environ. Sci. 10, 113–119.

Moon, H.-B., Choi, H.G., Lee, P.Y., Ok, G., 2008. Congener-specific characterization and sources of polychlorinated dibenzo-p-dioxins, dibenzofurans and dioxin-like polychlorinated biphenyls in marine sediments from industrialized bays of Korea. Environ. Toxicol. Chem. 27, 323–333.

Nakai, N., Ohta, T., Fujisawa, H., Yoshida, M., 1982. Paleoclimatic and sea-level changes deduced from organic carbon isotope ratios, C/N ratios and pyrite contents of cored sediments from Nagoya Harbor, Japan. Quat. Res. 21, 169–177.

Neff, J.M., 2002. Bioaccumulation in Marine Organisms: Effect of Contaminants From Oil Well Produced Water. Elsevier Ltd, p. 450.

Nielsen, T., 1996. Traffic contribution of polycyclic aromatic hydrocarbons in the center of a large city. Atmos. Environ. 30, 3481–3490.

Norris, G., Duvall, R., Brown, S., Bai, S., 2014. EPA Positive Matrix Factorization (PMF) 5.0 Fundamentals and User Guide, Prepared for the US Environmental Protection Agency, Washington, DC, by the National Exposure Research Laboratory. Research Triangle Park (EPA/600/R-14/108; STI-910511-5594-UG, April).

Ohyama, K., Nagai, F., Tsuchiya, Y., 2001. Certain styrene oligomers have proliferative activity on MCF-7 human breast tumor cells and binding affinity for human estrogen receptor. Environ. Health Perspect. 109, 699–703.

Onodera, S., Igarashi, K., Fukuda, A., Ouchi, J., Suzuki, S., 1994. Mutagenic potentials of anthracene and phenanthrene compounds during water disinfection with chlorine. Eisei kagaku 40, 233–243.

Paatero, P., Tapper, U., 1993. Analysis of different modes of factor analysis as least squares fit problems. Chemom. Intell. Lab. Syst. 18, 183–194.

Pedersen, K.B., Lejon, T., Jensen, P.E., Ottosen, L.M., 2015. Chemometric analysis for pollution source assessment of harbour sediments in Arctic locations. Water Air Soil Pollut. 226, 150.

Peters, K.E., Sweeney, R.E., Kaplan, I.R., 1978. Correlation of carbon and nitrogen stable isotope ratios in sedimentary organic matter 1. Limnol. Oceanogr. 23, 598–604.

Qiao, S., Shi, X., Wang, G., Zhou, L., Hu, B., Hu, L., Yang, G., Liu, Y., Yao, Z., Liu, S., 2017. Sediment accumulation and budget in the Bohai Sea, Yellow Sea and East China Sea. Mar. Geol. 390, 270–281.

Ravindra, K., Sokhi, R., Van Grieken, R., 2008. Atmospheric polycyclic aromatic hydrocarbons: source attribution, emission factors and regulation. Atmos. Environ. 42, 2895–2921.

Savinov, V.M., Savinova, T.N., Carroll, J., Matishov, G.G., Dahle, S., Næs, K., 2000. Polycyclic aromatic hydrocarbons (PAHs) in sediments of the White Sea, Russia. Mar. Pollut. Bull. 40, 807–818.

Singh, D.P., Gadi, R., Mandal, T.K., Saud, T., Saxena, M., Sharma, S.K., 2013. Emissions estimates of PAH from biomass fuels used in rural sector of Indo-Gangetic Plains of India. Atmos. Environ. 68, 120–126.

Soares, A., Guieysse, B., Jefferson, B., Cartmell, E., Lester, J., 2008. Nonylphenol in the environment: a critical review on occurrence, fate, toxicity and treatment in wastewaters. Environ. Int. 34, 1033–1049.

Sobek, A., Gustafsson, Ö., 2014. Deep water masses and sediments are main compartments for polychlorinated biphenyls in the arctic ocean. Environ. Sci. Technol. 48, 6719–6725.

Sobek, A., Sundqvist, K.L., Assefa, A.T., Wiberg, K., 2015. Baltic Sea sediment records: unlikely near-future declines in PCBs and HCB. Sci. Total Environ. 518–519, 8–15.

Song, Y., Choi, M.S., Lee, J.Y., Jang, D.J., 2014. Regional background concentrations of heavy metals (Cr, Co, Ni, Cu, Zn, Pb) in coastal sediments of the South Sea of Korea. Sci. Total Environ. 482, 80–91.

Tatarazako, N., Takao, Y., Kishi, K., Onikura, N., Arizono, K., Iguchi, T., 2002. Styrene dimers and trimers affect reproduction of daphnid (*Ceriodaphnia dubia*). Chemosphere 48, 597–601.

Tian, Z., Kim, S.-K., Hyun, J.-H., 2020. Environmental distribution of styrene oligomers (SOs) coupled with their source characteristics: tracing the origin of SOs in the Environment. J. Hazard. Mater. 398, 122968.

Wang, B.-d, Wang, X.-l, Zhan, R., 2003. Nutrient conditions in the Yellow Sea and the East China Sea. Estuar. Coast. Shelf Sci. 58, 127–136.

Wang, G., Peng, J., Yang, D., Zhang, D., Li, X., 2015. Current levels, composition profiles, source identification and potentially ecological risks of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in the surface sediments from Bohai Sea. Mar. Pollut. Bull. 101, 834–844.

Wang, P., Mi, W., Xie, Z., Tang, J., Apel, C., Joerss, H., Ebinghaus, R., Zhang, Q., 2020. Overall comparison and source identification of PAHs in the sediments of European Baltic and North Seas, Chinese Bohai and Yellow Seas. Sci. Total Environ. 737, 139535.

Wania, F., Mackay, D., 1996. Peer reviewed: tracking the distribution of persistent organic pollutants. Environ. Sci. Technol. 30, 390A–396A.

Webster, L., Russell, M., Walsham, P., Phillips, L.A., Hussy, I., Packer, G., Dalgarno, E.J., Moffat, C.F., 2011. An assessment of persistent organic pollutants in Scottish coastal and offshore marine environments. J. Environ. Monit. 13, 1288–1307.

Westerholm, R., Christensen, A., Törnqvist, M., Ehrenberg, L., Rannug, U., Sjögren, M., Rafter, J., Soontjens, C., Almén, J., Grägg, K., 2001. Comparison of exhaust emissions from Swedish environmental classified diesel fuel (MK1) and European Program on Emissions, Fuels and Engine Technologies (EPEFE) reference fuel: a chemical and biological characterization, with viewpoints on cancer risk. Environ. Sci. Technol. 35, 1748–1754.

Wolska, L., Mechlińska, A., Rogowska, J., Namieśnik, J., 2012. Sources and fate of PAHs and PCBs in the marine environment. Environ. Sci. Technol. 42, 1172–1189.

Woo, S., Kim, S., Yum, S., Yim, U.H., Lee, T.K., 2006. Comet assay for the detection of genotoxicity in blood cells of flounder (Paralichthys olivaceus) exposed to sediments and polycyclic aromatic hydrocarbons. Mar. Pollut. Bull. 52, 1768–1775.

Xu, Y., Liu, T., Zhu, X., Ji, G., 2019. Quantitative analysis of genetic associations in the biodegradative pathway of PAHs in wetland sediments of the Bohai coast region. Chemosphere 218, 282–291.

Yang, G.-P., 2000. Polycyclic aromatic hydrocarbons in the sediments of the South China Sea. Environ. Pollut. 108, 163–171.

Yang, D., Qi, S., Zhang, Y., Xing, X., Liu, H., Qu, C., Liu, J., Li, F., 2013a. Levels, sources and potential risks of polycyclic aromatic hydrocarbons (PAHs) in multimedia environment along the Jinjiang River mainstream to Quanzhou Bay, China. Mar. Pollut. Bull. 76, 298–306.

Yang, B., Zhou, L., Xue, N., Li, F., Li, Y., Vogt, R.D., Cong, X., Yan, Y., Liu, B., 2013b. Source apportionment of polycyclic aromatic hydrocarbons in soils of Huanghuai Plain, China: comparison of three receptor models. Sci. Total Environ. 443, 31–39.

Yim, U.H., Hong, S.-H., Shim, W.-J., 2007. Distribution and characteristics of PAHs in sediments from the marine environment of Korea. Chemosphere 68, 85–92.

Yim, U.H., Hong, S.H., Ha, S.Y., Han, G.M., An, J.G., Kim, N.S., Lim, D.-i, Choi, H.-W., Shim, W.J., 2014. Source-and region-specific distribution of polycyclic aromatic hydrocarbons in sediments from Jinhae Bay, Korea. Sci. Total Environ. 470, 1485–1493.

Yoon, S.J., Hong, S., Kwon, B.-O., Ryu, J., Lee, C.-H., Nam, J., Khim, J.S., 2017. Distributions of persistent organic contaminants in sediments and their potential impact on macrobenthic faunal community of the Geum River Estuary and Saemangeum Coast, Korea. Chemosphere 173, 216–226.

Yoon, S.J., Hong, S., Kim, T., Lee, J., Kwon, B.-O., Allam, A.A., Al-khedhairy, A.A., Khim, J.S., 2019. Occurrence and bioaccumulation of persistent toxic substances in sediments and biota from intertidal zone of Abu Ali Island, Arabian Gulf. Mar. Pollut. Bull. 144, 243–252.

Yoon, S.J., Hong, S., Kim, S., Lee, J., Kim, T., Kim, B., Kwon, B.-O., Zhou, Y., Shi, B., Liu, P., Hu, W., Huang, B., Wang, T., Khim, J.S., 2020. Large-scale monitoring and ecological risk assessment of persistent toxic substances in riverine, estuarine, and coastal sediments of the Yellow and Bohai seas. Environ. Int. 137, 105517.

Zaborska, A., Carroll, J., Pazdro, K., Pempkowiak, J., 2011. Spatio-temporal patterns of PAHs, PCBs and HCB in sediments of the western Barents Sea. Oceanologia 53, 1005–1026.

<Science of the Total Environment>

Supplementary materials for

Spatial distribution and source identification of traditional and emerging persistent toxic substances in the offshore sediment of South Korea

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References

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Supplementary Tables

Table S1. Target compounds, abbreviations, and monitoring ions in the instrumental analysis, and method detection limits and recoveries of surrogate standards.

Target compounds	Abbreviation	Monitoring ions		Method detection limit (ng g^{-1} dw)
		Quantification ion	Confirmation ion	
Polychlorinated biphenyl (PCBs)				
2,4,4'-Trichlorobiphenyl	CB28	256	258	0.03
2,2',5,5'-Tetrachlorobiphenyl	CB52	292	290	0.02
2,2',4,5'-Tetrachlorobiphenyl	CB49	292	290	0.02
2,2',3,5'-Tetrachlorobiphenyl	CB44	292	290	0.03
3,4,4'-Trichlorobiphenyl	CB37	256	258	0.02
2,4,4',5-Tetrachlorobiphenyl	CB74	292	290	0.02
2,3',4',5-Tetrachlorobiphenyl	CB70	292	290	0.01
2,3',4,4'-Tetrachlorobiphenyl	CB66	292	290	0.03
2,3,4,4'-Tetrachlorobiphenyl	CB60	292	290	0.03
2,2',4,5,5'-Pentachlorobiphenyl	CB101	326	324	0.02
2,2',4,4',5-Pentachlorobiphenyl	CB99	326	328	0.02
2,2',3,4,5'-Pentachlorobiphenyl	CB87	292	290	0.02
3,3',4,4'-Tetrachlorobiphenyl	CB77	326	256	0.01
2,2',3,3',4-Pentachlorobiphenyl	CB82	338	340	0.03
2,3',4,4',5-Pentachlorobiphenyl	CB118	326	328	0.03
2,3,4,4',5-Pentachlorobiphenyl	CB114	326	328	0.02
2,2',4,4',5,5'-Hexachlorobiphenyl	CB153	360	362	0.02
2,3,3',4,4'-Pentachlorobiphenyl	CB105	326	324	0.02
2,2',3,3',5,6,6'-Heptachlorobiphenyl	CB179	396	398	0.03
2,2',3,4,4',5'-Hexachlorobiphenyl	CB138	360	362	0.02
2,3,3',4,4',6-Hexachlorobiphenyl	CB158	326	328	0.03
3,3',4,4',5-Pentachlorobiphenyl	CB126	360	362	0.02
2,3,4,4',5,6-Hexachlorobiphenyl	CB166	394	396	0.02
2,2',3,4',5,5',6-Heptachlorobiphenyl	CB187	394	396	0.02
2,2',3,4,4',5',6-Heptachlorobiphenyl	CB183	360	362	0.02
2,2',3,3',4,4'-Hexachlorobiphenyl	CB128	440	442	0.02
2,3,3',4,4',5-Hexachlorobiphenyl	CB156	360	362	0.02
2,2',3,4,4',5,5'-Heptachlorobiphenyl	CB180	394	396	0.03
3,3',4,4',5,5'-Hexachlorobiphenyl	CB169	360	362	0.03
2,2',3,3',4,4',5-Heptachlorobiphenyl	CB170	394	396	0.03
2,3,3',4,4',5,5'-Heptachlorobiphenyl	CB189	476	478	0.03
Polycyclic aromatic hydrocarbons (PAHs)				
Acenaphthylene	Acl	152	151	0.15
Acenaphthene	Ace	153	154	0.11
Fluorene	Flu	166	165	0.19
Phenanthrene	Phe	178	176	0.21
Anthracene	Ant	178	176	0.13
Fluoranthene	Fl	202	200	0.07
Pyrene	Py	202	200	0.89
Benzo[*a*]anthracene	BaA	228	226	0.05
Chrysene	Chr	228	226	0.11
Benzo[*b*]fluoranthene	BbF	252	253	0.09
Benzo[*k*]fluoranthene	BkF	252	253	0.09
Benzo[*a*]pyrene	BaP	252	253	0.14
Indeno[*1,2,3-cd*]pyrene	IcdP	276	138	0.13
Dibenz[*a,h*]anthracene	DbahA	278	276	0.12
Benzo[*g,h,i*]perylene	BghiP	276	138	0.14

Table S1. Continued.

Target compounds	Abbreviation	Monitoring ions		Method detection limit ($ng\ g^{-1}$ dw)
		Quantification ion	Confirmation ion	
Emerging PAHs (e-PAHs)				
2-Methylphenanthrene	2MP	192	191	0.17
9-Ethylphenanthrene	9EP	191	206	0.06
Benzo[*b*]naphtho[*2,3-d*]furan	BBNF	218	189	0.32
11H-Benzo[*b*]fluorene	11BF	216	215	0.23
Benzo[*b*]naphtho[*2,1-d*]thiophene	BBNT	234	235	0.12
5-Methylbenzo[*a*]anthracene	5MBA	256	241	0.20
1,12-Dimethylbenzo[*c*]phenanthrene	BCP	242	241	0.34
Benzo[*j*]fluoranthene	BJF	252	253	0.52
Benzo[*e*]pyrene	BEP	252	250	0.28
Styrene oligomers (SOs)				
1,3-Diphenylpropane	SD1	105	196	0.19
cis-1,2-Diphenylcyclobutane	SD2	78	208	0.19
2,4-Diphenyl-1-butene	SD3	104	208	0.89
trans-1,2-Diphenylcyclobutane	SD4	78	208	0.11
2,4,6-Triphenyl-1-hexene	ST1	117	194	0.63
1e-Phenyl-4e-(1-phenylethyl)-tetralin	ST2	129	207	0.66
1a-Phenyl-4e-(1-phenylethyl)-tetralin	ST3	129	207	0.31
1a-Phenyl-4a-(1-phenylethyl)-tetralin	ST4	129	207	0.70
1e-Phenyl-4a-(1-phenylethyl)-tetralin	ST5	207	105	0.41
1,3,5-Triphenylcyclohexane	ST6	117	104	0.88
Alkylphenols (APs)				
4-*tert*-Octylphenol	t-OP	207	221	0.12
4-*tert*-Octylphenol monoethoxylate	t-OP1EO	251	265	0.61
4-*tert*-Octylphenol diethoxylate	t-OP2EO	295	309	0.08
Nonylphenols	NPs	207	221	3.7
Nonylphenol monoethoxylates	NP1EOs	251	265	0.45
Nonylphenol diethoxylates	NP2EOs	295	309	1.5
Internal standard				
2-Fluorobiphenyl	IS	172	171	

Surrogate standards	Abbreviation	Quantification ion	Confirmation ion	Surrogate recovery (%, mean ± SD)
Polychlorinated biphenyl (PCBs)				
^{13}C-labeled CB 28		268	270	106 ± 7
^{13}C-labeled CB 52		304	302	99 ± 6
^{13}C-labeled CB 101		326	328	102 ± 6
^{13}C-labeled CB 153		372	374	109 ± 7
^{13}C-labeled CB 138		360	362	110 ± 6
^{13}C-labeled CB 180		406	408	114 ± 7
^{13}C-labeled CB 209		510	512	115 ± 8
Polycyclic aromatic hydrocarbons(PAHs)				
Acenaphthene-d10	Ace-d10	164	162	72 ± 12
Phenanthrene-d10	Phe-d10	188	189	76 ± 9
Chrysene-d12	Chr-d12	240	236	88 ± 11
Perylene-d12	Pery-d12	264	270	90 ± 11
Alkylphenols (APs)				
Bisphenol A-d16	BPA-d16	368	386	75 ± 17

Table S2. GC/MSD conditions for analyzing polychlorinated biphenyls, polycyclic aromatic hydrocarbons, styrene oligomers, and alkylphenols in this study.

Instrument	**Agilent 7890B GC / 5977B MSD**	
Column	DB-5ms (30 m × 250 μm × 0.25 μm)	
Gas flow	1 $mLmin^{-1}$ He	
Injection mode	Splitless	
Injection volume	1 μL	
Oven temperature program	PCBs	60 °C (hold 1 min) → 5 °C min^{-1} to 140 °C (hold 1 min) → 30 °C min^{-1} to 200 °C (hold 1 min) → 4 °C min^{-1} to 250 °C (hold 5 min) → 10 °C min^{-1} to 300 °C (hold 1 min)
	PAHs	60 °C (hold 2 min) → 6 °C min^{-1} to 300 °C (hold 13 min)
	SOs	60 °C (hold 2 min) → 6 °C min^{-1} to 300 °C (hold 3 min)
	APs	60 °C (hold 5 min) → 10 °C min^{-1} to 100 °C → 20 °C min^{-1} to 300 °C (hold 6 min)

Table S3. Overview of sampling locations and properties of sediment samples collected from the offshore of South Korea.

Sea	Sites	Latitude (°N)	Longitude (°E)	Sampling depth (m)	Water Contents (%)	Mud Contents (%)	OC (%)	TN (%)	$\delta^{13}C$ (‰)	$\delta^{15}N$ (‰)
Yellow Sea	YA 1	36°56.34'	125°58.94'	38	25	46	0.24	0.04	-22.0	4.40
	YA 2	36°33.00'	126°24.00'	16	23	11	0.17	0.02	-21.3	5.54
	YA 3	35°51.18'	126°14.42'	28	27	6.8	0.13	0.03	-20.5	5.68
	YA 4	35°19.61'	125°46.97'	31	56	98	0.75	0.12	-21.3	6.97
	YA 5	34'43.28'	125°43.13'	33	49	88	0.47	0.07	-21.4	6.07
	YA 6	34°01.28'	125°42.23'	69	41	96	0.52	0.07	-22.0	6.71
	YB 1	36°55.29'	125°12.38'	52	28	5.9	0.13	0.02	-21.6	4.36
	YB 2	36°19.09'	125°12.68'	62	26	6.9	0.12	0.02	-22.0	5.55
	YB 3	35°51.16'	125°12.23'	73	27	16	0.21	0.04	-22.0	5.62
	YB 4	35°20.05'	125°12.84'	74	25	18	0.30	0.03	-21.9	6.35
	YB 5	34°43.25'	125°00.27'	93	33	41	0.23	0.06	-21.4	5.90
	YB 6	34°04.55'	125°18.31'	110	34	97	0.62	0.08	-22.3	6.84
	YC 1	36°55.73'	124°24.09'	76	25	14	0.15	0.02	-21.9	4.47
	YC 2	36°20.28'	124°23.44'	76	53	85	0.82	0.12	-21.8	6.69
	YC 3	35°50.64'	124°23.00'	83	54	77	0.70	0.14	-21.9	6.38
	YC 4	35°19.93'	124°23.87'	85	50	69	0.50	0.11	-21.7	6.07
	YC 5	34°43.27'	124°23.48'	77	39	46	0.28	0.07	-21.3	5.55
	YC 6	34°06.67'	124°24.30'	80	43	61	0.34	0.07	-21.4	6.25
South Sea	K 1	34°09.00'	127°06.00'	32	28	11	0.80	0.04	-21.3	7.47
	K 2	34°21.00'	127°36.00'	18	55	86	1.0	0.13	-21.2	7.46
	K 3	33°55.30'	127°03.00'	66	33	22	0.73	0.07	-21.3	7.16
	K 4	34°09.00'	127°39.00'	38	56	88	1.1	0.12	-21.5	6.97
	K 5	34°12.00'	128°06.00'	73	51	71	1.4	0.14	-21.6	6.83
	K 6	34°18.00'	128°36.00'	94	42	35	0.66	0.10	-21.1	6.95
	K 7	34°39.00'	129°00.00'	75	-	-	0.22	0.05	-21.1	6.90
	K 8	33°42.00'	127°00.00'	110	28	20	0.49	0.05	-	8.10
	K 9	33°54.00'	127°42.00'	83	42	69	0.84	0.11	-21.5	6.30
	K 10	34°00.00'	128°12.00'	92	40	31	1.5	0.09	-21.5	6.94
East Sea	E 1	35°26.26'	129°28.78'	70	-	-	1.9	0.19	-21.0	6.20
	E 2	35°38.01'	129°49.27'	140	-	-	0.24	0.07	-20.9	7.72
	E 3	35°43.16'	129°54.08'	610	72	93	3.1	0.34	-21.3	6.52
	E 4	35°47.76'	129°59.71'	1100	-	-	3.2	0.34	-21.3	6.80
	E 5	35°59.29'	130°12.51'	1400	-	-	2.7	0.35	-21.6	7.73
	E 6	37°01.36'	130°55.55'	2000	73	90	2.9	0.31	-21.3	6.68

-: Not analyzed.

Table S4. Concentrations of polychlorinated biphenyls in offshore sediments of South Korea.

Sea	Sites	Concentration of 31 PCBs (ng g^{-1} OC)															
		28	52	49	44	37	74	70	66	60	101	99	87	77	82	118	114
Yellow Sea	YA 1	0.32	< LOD	0.30	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YA 2	0.40	< LOD	0.06	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YA 3	0.46	0.39	0.20	0.07	< LOD	0.08	< LOD	< LOD	0.05	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YA 4	< LOD	0.13	0.14	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YA 5	0.16	< LOD	0.21	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YA 6	0.55	0.19	0.20	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 1	0.63	< LOD	0.77	< LOD	0.38	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 2	0.40	< LOD	0.52	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 3	1.24	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 4	0.11	< LOD	0.26	< LOD	< LOD	< LOD	0.12	0.16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 5	0.44	0.35	0.34	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YB 6	< LOD	0.08	< LOD	< LOD	0.06	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 1	0.64	< LOD	0.43	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 2	1.06	< LOD	0.51	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 3	5.32	0.38	0.56	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 4	< LOD	0.10	0.10	< LOD	< LOD	< LOD	< LOD	0.26	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 5	0.31	0.13	< LOD	< LOD	< LOD	< LOD	< LOD	0.08	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	YC 6	< LOD	< LOD	0.15	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
South Sea	K 1	< LOD	< LOD	< LOD	< LOD	0.30	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 2	0.11	0.05	< LOD	< LOD	0.26	< LOD	0.05	0.12	0.19	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 3	0.10	0.05	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 4	0.42	0.06	0.04	< LOD	0.30	< LOD	0.04	0.09	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 5	0.10	0.04	< LOD	< LOD	0.20	< LOD	0.02	0.05	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 6	0.47	0.24	< LOD	< LOD	< LOD	< LOD	< LOD	0.21	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 7	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.07	0.09	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 8	0.08	< LOD	0.17	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 9	0.35	0.04	0.12	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	K 10	0.10	0.02	0.06	< LOD	0.14	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
East Sea	E 1	0.15	0.07	0.08	0.03	< LOD	< LOD	0.03	0.05	0.08	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	E 2	0.57	0.16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	E 3	0.13	0.07	0.03	0.03	< LOD	0.02	< LOD	0.03	0.02	< LOD	0.02	< LOD	< LOD	< LOD	< LOD	0.03
	E 4	0.22	0.03	0.03	< LOD	0.09	< LOD	< LOD	0.03	0.28	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
	E 5	1.30	0.02	0.06	< LOD	< LOD	0.02	< LOD	0.02	< LOD	0.02	< LOD	< LOD	< LOD	< LOD	0.03	< LOD
	E 6	0.37	0.07	0.09	0.02	< LOD	0.02	< LOD	0.02	0.26	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD

< LOD: Limit of detection.

Table S4. Continued.

Sea	Sites	Concentration of 31 PCBs (ng g^{-1} OC)															
		153	105	179	138	158	126	166	187	183	128	156	180	169	170	189	ΣPCBs
Yellow Sea	YA 1	< LOD	0.33	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.95
	YA 2	0.06	0.10	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.62
	YA 3	0.06	0.28	< LOD	0.05	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.15	< LOD	< LOD	1.8
	YA 4	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.10	< LOD	< LOD	0.37
	YA 5	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.37
	YA 6	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.94
	YB 1	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.8
	YB 2	0.39	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.3
	YB 3	< LOD	0.27	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.5
	YB 4	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.65
	YB 5	< LOD	0.18	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.3
	YB 6	< LOD	0.35	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.49
	YC 1	0.26	0.41	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.7
	YC 2	0.28	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.9
	YC 3	0.65	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	6.9
	YC 4	0.09	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.55
	YC 5	0.09	0.08	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.69
	YC 6	0.16	0.16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.47
South Sea	K 1	< LOD	0.16	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.46
	K 2	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	< LOD	< LOD	0.82
	K 3	< LOD	0.07	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.22
	K 4	< LOD	0.08	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	< LOD	< LOD	1.1
	K 5	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.03	< LOD	< LOD	0.44
	K 6	< LOD	0.34	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.36	< LOD	< LOD	1.6
	K 7	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.16
	K 8	< LOD	0.18	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.43
	K 9	< LOD	0.07	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	< LOD	< LOD	0.62
	K 10	< LOD	0.03	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.03	< LOD	< LOD	0.38
East Sea	E 1	0.02	0.06	< LOD	< LOD	< LOD	0.03	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.04	< LOD	< LOD	0.64
	E 2	< LOD	0.49	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	1.2
	E 3	0.03	0.04	< LOD	0.03	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.05	< LOD	< LOD	0.53
	E 4	0.02	0.04	< LOD	0.02	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	0.03	< LOD	< LOD	0.79
	E 5	0.05	0.07	< LOD	0.06	< LOD	< LOD	< LOD	0.02	< LOD	0.02	< LOD	< LOD	0.04	< LOD	< LOD	1.7
	E 6	0.02	0.04	< LOD	0.03	< LOD	< LOD	< LOD	< LOD	< LOD	0.02	< LOD	< LOD	0.06	< LOD	< LOD	1.0

< LOD: Limit of detection.

Table S5. Concentrations of traditional polycyclic aromatic hydrocarbons in offshore sediments of South Korea.

Sites	Concentration of 15 PAHs (ng g^{-1} OC)															
	Acl	**Ace**	**Flu**	**Phe**	**Ant**	**Fl**	**Py**	**BaA**	**Chr**	**BbF**	**BkF**	**BaP**	**IcdP**	**DbahA**	**BghiP**	**Σ15PAHs**
YA 1	1.3	1.6	5.5	19	1.2	11	7.6	< LOD	1.9	< LOD	7.4	6.2	< LOD	< LOD	< LOD	63
YA 2	1.5	2.5	6.9	25	1.1	11	8.4	11	5.0	2.5	4.9	8.5	0.4	< LOD	< LOD	89
YA 3	2.3	4.3	11	29	1.8	12	11	< LOD	4.0	1.7	5.0	7.0	0.2	< LOD	5.1	94
YA 4	0.3	0.7	2.3	7.6	0.2	13	7.0	1.2	4.6	6.4	8.6	5.3	20	1.3	17	96
YA 5	0.8	1.4	4.4	13	0.2	12	7.3	1.2	4.6	3.7	8.5	3.9	15	< LOD	15	91
YA 6	< LOD	0.9	2.8	7.2	0.3	3.1	2.1	0.1	1.2	0.4	1.9	2.0	< LOD	< LOD	< LOD	22
YB 1	1.2	3.0	7.8	23	21	13	7.8	< LOD	3.9	< LOD	8.5	7.8	< LOD	< LOD	< LOD	97
YB 2	2.5	3.8	12	36	1.7	17	13	16	7.7	2.7	11	5.0	1.0	< LOD	4.2	130
YB 3	1.5	2.2	6.6	26	1.1	20	13	1.1	8.0	6.2	16	20	9.0	< LOD	27	160
YB 4	1.4	2.3	5.2	19	0.6	14	9.1	0.7	5.4	2.5	10	11	4.9	< LOD	14	100
YB 5	0.6	2.3	6.7	16	1.0	9.0	6.4	0.4	3.9	2.5	8.9	9.2	4.7	< LOD	12	84
YB 6	0.4	0.7	2.5	8.8	0.2	3.2	2.7	< LOD	1.2	0.7	1.3	2.1	< LOD	< LOD	2.1	26
YC 1	2.2	3.9	9.7	31	1.5	18	13	5.8	5.7	< LOD	5.4	17	5.5	< LOD	4.0	120
YC 2	0.5	0.9	2.3	9.5	0.3	7.0	4.6	0.5	2.9	1.8	6.0	6.8	4.1	< LOD	8.8	56
YC 3	0.8	1.3	3.7	12	0.6	17	10	2.6	7.2	14	14	10	39	7.4	33	170
YC 4	0.9	1.4	3.7	15	0.7	18	12	2.6	7.6	15	16	9.2	41	0.2	36	180
YC 5	1.1	1.7	4.5	17	0.9	14	10	1.1	5.9	4.7	13	14	10	1.4	23	120
YC 6	1.3	1.9	6.0	20	0.8	17	11	1.6	8.9	11	15	9.1	11	0.2	28	140
K 1	0.4	0.7	2.8	7.1	0.2	4.8	3.9	1.0	1.4	3.1	1.6	1.0	3.0	0.7	3.2	35
K 2	0.4	0.8	3.2	9.0	1.0	10	5.8	2.4	2.8	11	3.7	3.2	12	2.0	10	77
K 3	0.5	1.2	3.7	9.1	0.5	6.3	4.5	1.3	2.2	6.3	9.6	1.4	7.3	1.3	6.8	62
K 4	0.6	1.4	3.9	11	12	14	8.6	3.4	4.7	17	7.0	4.7	17	3.2	16	120
K 5	0.3	0.7	2.0	6.6	0.5	6.8	4.8	1.7	2.4	8.7	3.6	2.3	10	0.7	8.5	60
K 6	2.2	3.3	8.1	32	1.8	30	19	7.0	10	31	15	9.0	39	6.5	35	250
K 7	0.6	1.5	4.1	12	0.5	8.8	6.5	2.6	3.1	11	4.8	3.6	14	2.6	12	88
K 8	0.7	1.5	5.1	14	0.4	5.7	4.5	1.1	1.9	4.9	2.4	1.1	5.2	1.1	5.6	55
K 9	0.6	1.4	3.9	11	0.6	9.6	6.1	2.6	3.8	13	4.1	3.5	15	2.6	13	91
K 10	0.3	0.4	1.6	4.8	0.3	3.6	2.5	1.1	1.5	5.0	2.1	1.5	6.1	2.6	5.4	39
E 1	0.3	0.8	1.9	6.1	1.0	10	7.6	4.1	3.3	12	5.8	6.2	17	2.8	13	92
E 2	0.7	2.5	9.6	22	1.0	16	10	4.4	5.6	17	6.4	6.4	23	3.6	19	150
E 3	0.5	1.2	2.0	7.4	0.8	9.7	7.6	3.7	4.0	14	5.6	6.8	21	3.5	17	100
E 4	0.4	0.8	1.9	6.5	0.9	7.7	5.6	2.9	3.1	11	4.1	5.3	16	2.8	13	83
E 5	1.3	1.3	3.1	17	1.4	19	14	7.3	8.3	34	12	9.9	52	8.7	35	220
E 6	0.4	1.3	2.0	7.1	1.1	11	11	3.9	4.4	16	5.6	7.2	21	4.1	17	110

< LOD: Limit of detection.

Table S6. Concentrations of emerging polycyclic aromatic hydrocarbons in offshore sediments of South Korea.

Sites	Concentration of e-PAHs (ng g^{-1} OC)									
	2MA	**9EP**	**BBNF**	**11BF**	**BBNT**	**5MBA**	**BCP**	**BJF**	**BEP**	**Σe-PAHs**
YA 1	2.2	0.1	0.6	1.2	< LOD	< LOD	< LOD	< LOD	< LOD	4.1
YA 2	3.3	0.6	0.6	2.0	< LOD	< LOD	0.4	4.5	4.8	16
YA 3	4.0	0.7	0.6	1.4	< LOD	< LOD	0.4	3.3	< LOD	10
YA 4	1.7	0.6	< LOD	1.5	< LOD	0.5	< LOD	9.0	8.7	22
YA 5	2.3	0.7	< LOD	2.3	< LOD	< LOD	< LOD	6.6	6.9	19
YA 6	1.0	0.3	< LOD	0.8	< LOD	< LOD	< LOD	1.5	1.4	5.0
YB 1	3.1	0.7	0.6	1.0	< LOD	< LOD	0.7	< LOD	0.3	6.4
YB 2	5.2	0.7	0.7	2.0	< LOD	< LOD	2.8	7.7	6.0	25
YB 3	3.9	0.7	0.5	2.3	< LOD	0.3	1.4	9.1	13	31
YB 4	2.7	0.8	0.5	2.6	< LOD	0.4	0.4	5.0	7.9	20
YB 5	2.5	0.4	0.5	1.2	< LOD	0.3	< LOD	5.0	5.4	15
YB 6	0.9	0.2	< LOD	< LOD	< LOD	< LOD	< LOD	1.0	1.3	3.4
YC 1	4.6	0.8	0.6	2.3	< LOD	< LOD	1.1	2.7	11	23
YC 2	1.8	0.4	< LOD	1.3	< LOD	< LOD	< LOD	3.7	4.2	11
YC 3	3.0	0.9	< LOD	2.8	< LOD	0.3	0.9	12	14	34
YC 4	3.7	1.1	0.5	3.8	< LOD	1.0	0.5	11	16	38
YC 5	3.1	1.0	0.6	2.5	< LOD	< LOD	< LOD	5.9	9.8	23
YC 6	4.2	1.0	0.7	4.0	< LOD	0.7	0.4	12	14	37
K 1	< LOD	0.3	< LOD	0.4	< LOD	< LOD	< LOD	1.3	2.5	4.5
K 2	< LOD	0.6	0.6	1.0	0.3	0.2	< LOD	3.1	6.6	12
K 3	< LOD	0.5	< LOD	0.5	0.4	< LOD	< LOD	2.4	4.7	8.5
K 4	0.2	0.9	1.0	1.4	0.8	0.3	< LOD	5.4	11	21
K 5	< LOD	0.4	0.4	0.6	0.4	0.2	< LOD	3.1	5.6	11
K 6	0.2	2.2	1.7	2.9	1.4	0.7	< LOD	12	23	44
K 7	< LOD	0.7	0.6	0.9	0.4	0.2	0.7	3.2	7.1	14
K 8	< LOD	0.5	< LOD	0.9	0.3	< LOD	< LOD	1.9	3.7	7.3
K 9	0.2	0.7	0.6	0.9	0.7	0.2	< LOD	4.0	8.2	16
K 10	< LOD	0.3	< LOD	0.3	0.2	< LOD	< LOD	1.7	3.5	6.0
E 1	0.2	0.5	0.7	1.2	0.5	0.2	1.1	4.1	7.4	16
E 2	< LOD	1.0	0.8	1.4	0.8	0.4	< LOD	5.5	12	22
E 3	0.3	0.5	0.7	1.1	0.8	0.2	< LOD	4.6	9.5	18
E 4	0.2	0.4	0.5	0.9	0.6	0.2	< LOD	3.4	7.3	14
E 5	0.4	0.8	1.4	1.8	1.6	0.4	< LOD	9.7	19	35
E 6	0.2	0.5	0.6	1.1	0.7	0.2	< LOD	4.9	9.3	18

< LOD: Limit of detection.

Table S7. Concentrations of styrene oligomers in offshore sediments of South Korea.

Sites	Concentration of SOs (ng g^{-1} OC)										
	SD1	**SD2**	**SD3**	**SD4**	**ST1**	**ST2**	**ST3**	**ST4**	**ST5**	**ST6**	**ΣSOs**
YA 1	4.5	3.6	550	7.8	13	3.1	3.8	3.6	3.2	36	630
YA 2	1.0	0.6	110	1.7	3.2	1.4	1.1	1.6	0.8	4.9	130
YA 3	1.1	1.4	180	1.9	3.5	0.6	0.8	1.1	1.8	2.7	200
YA 4	1.3	1.0	160	2.5	4.3	1.6	2.0	2.8	0.8	11	190
YA 5	1.7	1.8	210	4.1	6.8	2.2	1.6	1.9	1.3	15	240
YA 6	2.1	1.5	260	3.7	6.2	2.1	1.4	1.3	0.8	17	290
YB 1	5.5	10	670	10	13	4.7	2.0	2.5	1.9	76	790
YB 2	5.2	3.8	440	10	16	4.3	3.9	4.2	2.1	60	550
YB 3	3.0	1.9	300	5.7	7.9	2.9	3.8	4.2	1.8	31	360
YB 4	2.7	1.7	240	4.2	8.1	2.1	2.9	3.5	0.9	7.4	270
YB 5	2.8	5.1	340	5.7	9.9	2.9	2.1	2.4	4.2	7.0	380
YB 6	1.1	1.1	390	2.9	3.1	0.7	0.7	0.9	0.9	2.3	400
YC 1	2.8	4.3	210	5.2	8.8	2.8	1.6	1.8	1.8	8.0	240
YC 2	4.4	2.6	280	8.1	13	1.0	2.7	3.6	2.0	19	340
YC 3	5.7	4.0	580	9.7	16	1.1	3.5	3.8	1.2	11	640
YC 4	1.3	2.3	120	2.8	11	3.4	7.4	11	2.4	28	190
YC 5	1.6	0.8	54	2.5	4.8	1.6	1.5	0.3	0.4	13	80
YC 6	1.1	2.5	410	3.7	4.1	1.0	1.0	1.2	2.0	4.9	430
K 1	1.3	1.2	70	1.5	3.7	0.2	0.5	0.5	0.4	8.5	88
K 2	1.4	0.5	41	1.0	3.7	0.3	1.8	2.4	0.5	6.0	59
K 3	1.6	1.0	32	1.3	4.2	0.3	0.7	0.8	0.3	8.5	51
K 4	1.4	1.0	110	1.6	3.8	0.5	1.4	1.9	0.5	9.9	130
K 5	1.2	0.4	100	1.2	3.1	0.2	0.9	1.2	0.4	8.6	120
K 6	4.3	3.2	1000	5.8	13	2.1	6.4	9.7	5.6	60	1100
K 7	1.1	0.6	69	1.8	4.1	1.0	1.9	2.5	0.6	16	99
K 8	2.2	1.1	130	2.3	6.3	0.4	0.9	0.9	0.5	10	160
K 9	1.5	0.6	18	1.4	4.1	0.3	1.3	1.8	0.3	9.8	39
K 10	0.8	0.2	22	0.6	2.5	0.2	0.7	0.9	0.2	4.6	32
E 1	1.0	1.0	110	1.2	2.6	0.5	10	2.1	1.6	34	160
E 2	4.3	1.7	140	4.5	13	0.7	18	1.0	2.4	99	290
E 3	0.5	0.4	130	0.7	1.9	0.3	7.6	11	1.8	24	180
E 4	0.4	0.5	63	0.6	1.1	0.2	5.9	1.2	0.9	29	100
E 5	0.6	0.9	46	3.1	3.6	1.3	4.4	6.1	1.7	1.8	69
E 6	0.6	0.6	71	0.8	1.7	0.3	6.7	1.4	1.0	22	110

Table S8. Concentrations of alkylphenols in offshore sediments of South Korea.

Sites	Concentration of APs (ng g^{-1} OC)						
	NPs	**NP1EOs**	**NP2EOs**	**t-OP**	**t-OP1EO**	**t-OP2EO**	**ΣAPs**
YA 1	5.3	4.0	5.7	1.6	4.4	2.2	23
YA 2	< LOD	2.4	2.4	0.6	1.2	0.4	7.0
YA 3	< LOD	1.9	< LOD	0.3	1.7	0.1	4.0
YA 4	< LOD	2.8	2.9	0.5	1.4	0.5	8.1
YA 5	< LOD	2.7	2.3	0.9	3.7	0.1	9.7
YA 6	< LOD	1.7	3.3	1.2	2.2	0.8	9.2
YB 1	< LOD	15	8.4	2.0	5.2	2.8	33
YB 2	8.6	16	9.8	5.7	9.4	3.0	53
YB 3	6.0	6.3	6.5	3.2	7.1	1.6	31
YB 4	3.9	3.1	5.1	1.1	3.7	0.1	17
YB 5	3.9	3.7	5.2	1.3	4.2	1.5	20
YB 6	< LOD	1.8	2.1	1.1	2.1	0.5	7.6
YC 1	< LOD	2.5	3.5	1.0	2.7	1.4	11
YC 2	11.3	7.3	5.6	1.0	6.3	1.1	33
YC 3	80	39	17	5.1	6.0	2.4	150
YC 4	< LOD	1.4	1.7	0.5	1.5	0.5	5.6
YC 5	< LOD	1.7	1.9	0.6	1.8	0.5	6.5
YC 6	< LOD	1.6	1.5	0.6	2.3	0.1	6.1
K 1	< LOD	2.0	1.6	0.4	1.1	0.3	5.4
K 2	< LOD	2.7	1.5	0.2	0.7	0.1	5.2
K 3	< LOD	2.0	2.3	0.4	0.9	0.1	5.7
K 4	< LOD	3.6	2.5	0.2	0.8	0.2	7.3
K 5	< LOD	1.9	< LOD	0.3	< LOD	< LOD	2.2
K 6	4.6	13	12	1.7	3.1	0.6	35
K 7	< LOD	1.7	< LOD	0.5	1.4	0.5	4.1
K 8	< LOD	3.0	2.7	0.8	2.5	0.1	9.1
K 9	< LOD	1.8	1.5	0.2	1.1	0.1	4.7
K 10	< LOD	1.3	< LOD	0.2	< LOD	0.1	1.6
E 1	< LOD	2.4	< LOD	0.4	< LOD	0.1	2.9
E 2	< LOD	11	7.3	2.2	2.3	0.5	23
E 3	< LOD	5.6	1.8	0.2	< LOD	0.1	7.7
E 4	< LOD	5.9	< LOD	< LOD	< LOD	0.1	6.0
E 5	< LOD	2.8	1.7	< LOD	< LOD	0.1	4.6
E 6	< LOD	4.2	< LOD	< LOD	< LOD	0.1	4.3

< LOD: Limit of detection

Supplementary Figures

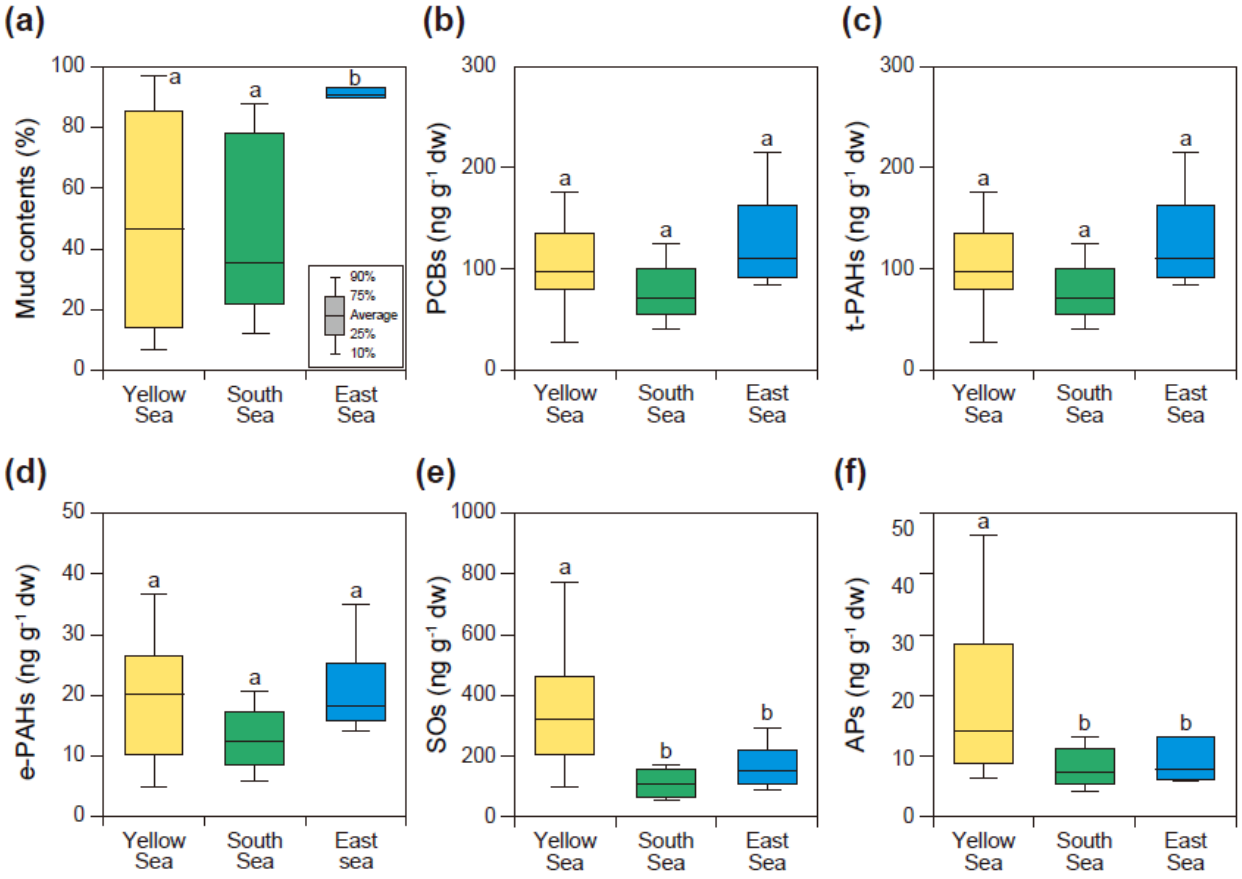

Fig. S1. Comparisons of (a) mud contents, concentrations of (b) polychlorinated biphenyls (PCBs), (c) traditional polycyclic aromatic hydrocarbons (t-PAHs), (d) emerging polycyclic aromatic hydrocarbons (e-PAHs), (e) styrene oligomers (SOs), and (f) alkylphenols (APs) in sediments from three sea areas of South Korea. The differences in mud contents and concentrations of PTSs in sediments between the three sea areas were examined for significance ($p < 0.05$) using the Kruskal-Wallis test. Different letters show statistical significance.

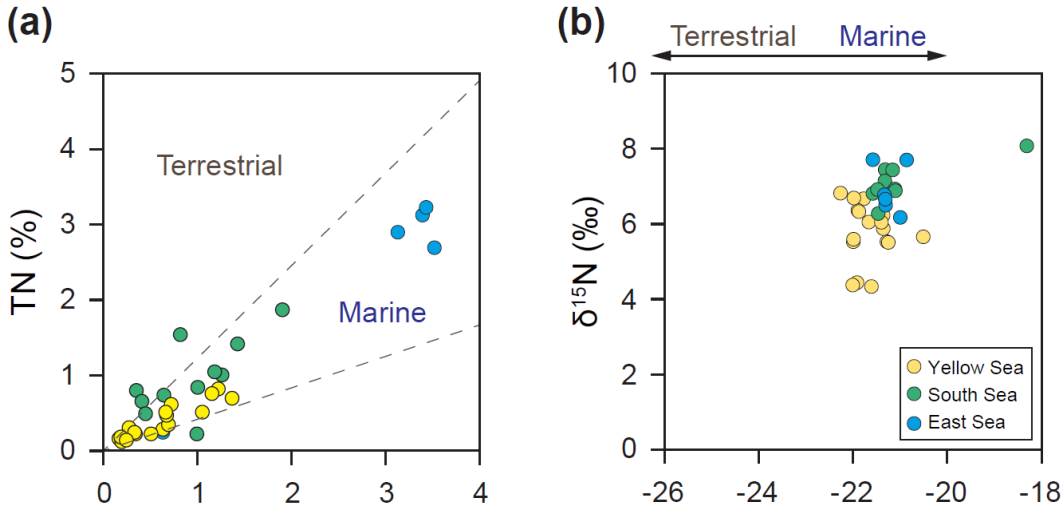

Fig. S2. Source identification of organic matter in offshore sediments of South Korea using (a) C/N ratio and (b) carbon and nitrogen stable isotope ratios.

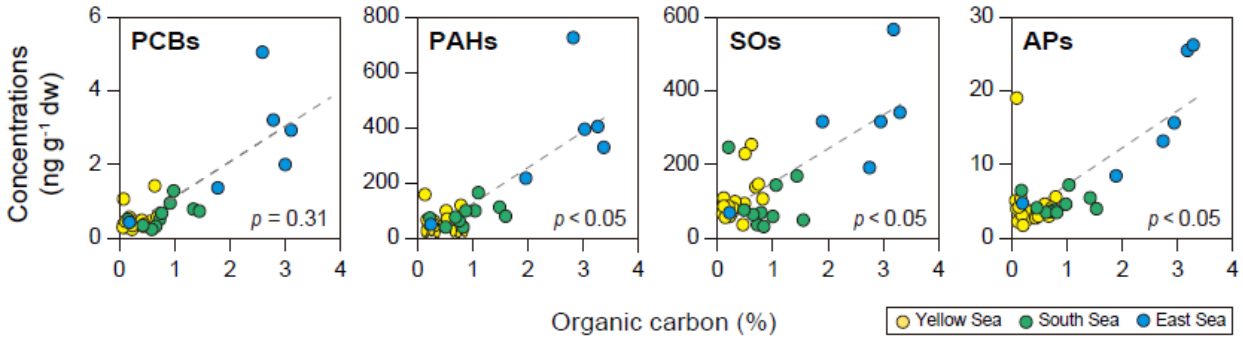

Fig. S3. Correlations between organic carbon contents (%) and polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), styrene oligomers (SOs), and alkylphenols (APs) in offshore sediments of South Korea.

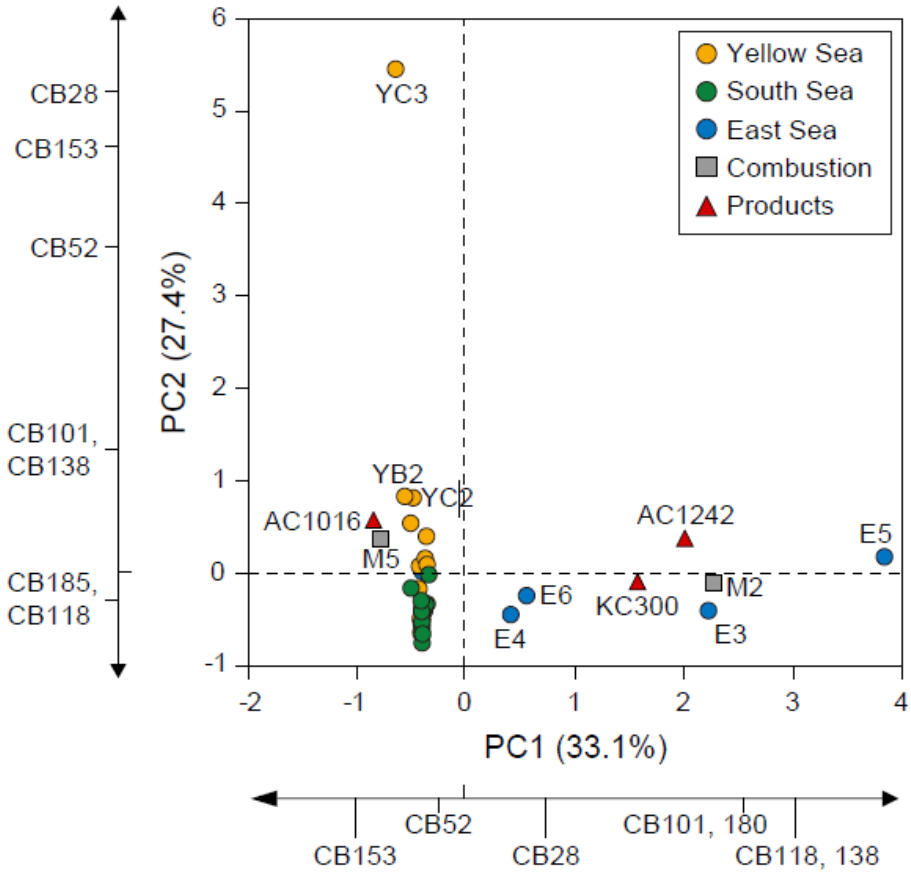

Fig. S4. Result of principal component analysis (PCA) of individual PCBs in sediments collected from the offshore areas of South Korea and in representative sources (Data from Kim et al., 1999).

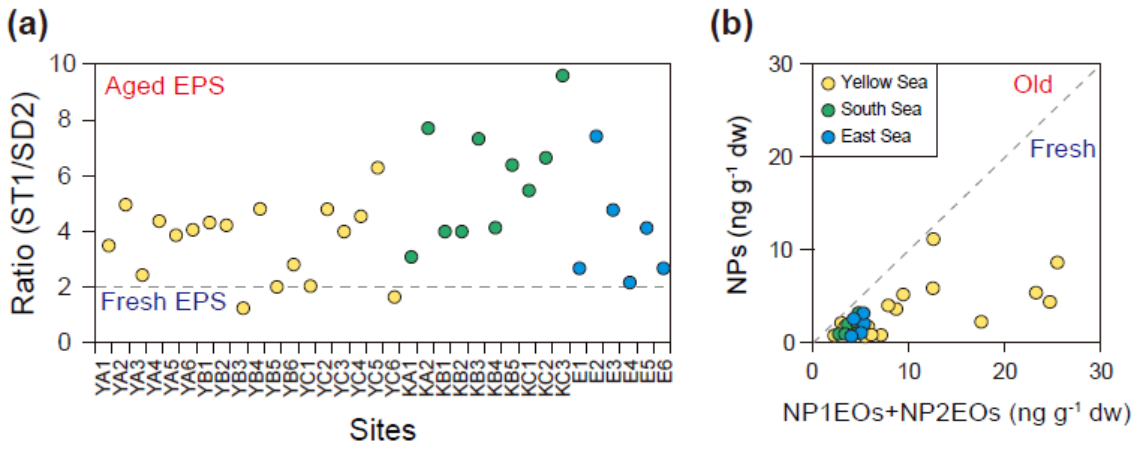

Fig. S5. Source identification of (a) styrene oligomers and (b) alkylphenols in offshore sediments of South Korea using diagnostic ratios.

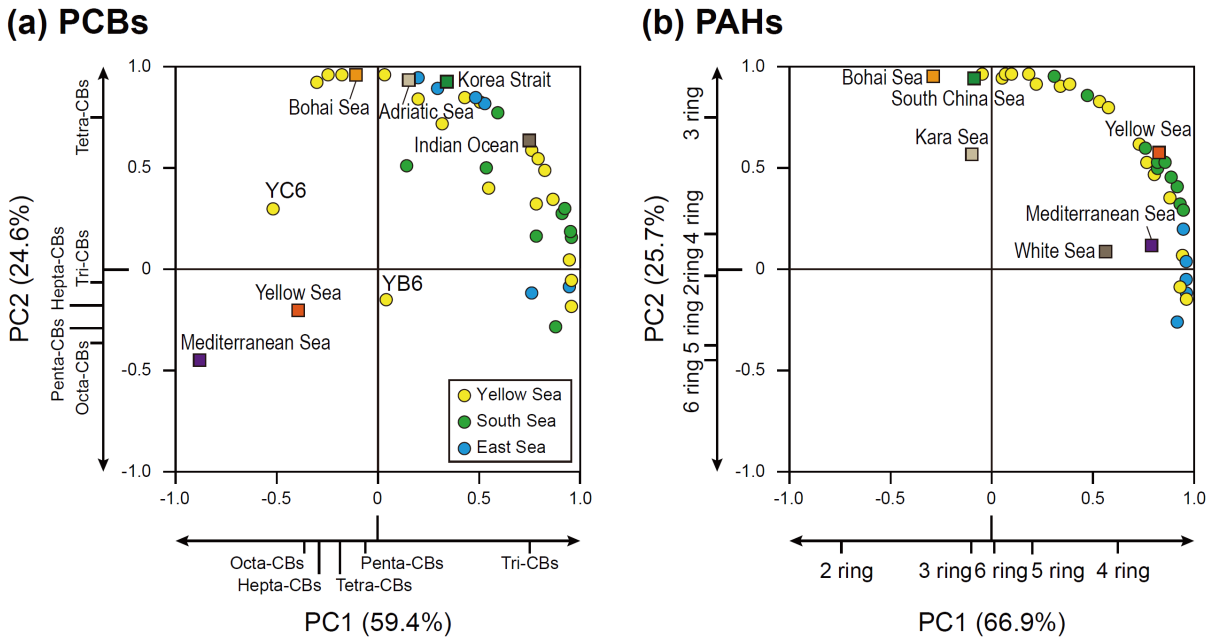

Fig. S6. Principal component analysis using compositions of PCBs and PAHs in sediments obtained from the present study and previous studies (Cheng et al., 2015; Combi et al., 2016; Dahle et al., 2003; Duan et al., 2013; Guerra et al., 2019; Hu et al., 2011; Liu et al., 2012; Mandalakis et al., 2014; Savinov et al., 2000; Wang et al., 2015; Yang, 2000).

References

Cheng, Z., Lin, T., Xu, W., Xu, Y., Li, J., Luo, C., Zhang, G., 2015. A preliminary assessment of polychlorinated biphenyls and polybrominated diphenyl ethers in deep-sea sediments from the Indian Ocean. Mar. Pollut. Bull. 94, 323–328.

Combi, T., Pintado-Herrera, M.G., Lara-Martin, P.A., Miserocchi, S., Langone, L., Guerra, R., 2016. Distribution and fate of legacy and emerging contaminants along the Adriatic Sea: A comparative study. Environ. Pollut. 218, 1055–1064.

Dahle, S., Savinov, V.M., Matishov, G.G., Evenset, A., Næs, K., 2003. Polycyclic aromatic hydrocarbons (PAHs) in bottom sediments of the Kara Sea shelf, Gulf of Ob and Yenisei Bay. Sci. Total Environ. 306, 57–71.

Duan, X., Li, Y., Li, X., Zhang, D., Li, M., 2013. Polychlorinated biphenyls in sediments of the Yellow Sea: Distribution, source identification and flux estimation. Mar. Pollut. Bull. 76, 283–290.

Guerra, R., Pasteris, A., Righi, S., Ok, G., 2019. Historical record of polychlorinated biphenyls (PCBs) in the continental shelf of the Korea Strait. Chemosphere 237, 124438.

Hu, L., Guo, Z., Shi, X., Qin, Y., Lei, K., Zhang, G., 2011. Temporal trends of aliphatic and polyaromatic hydrocarbons in the Bohai Sea, China: evidence from the sedimentary record. Org. Geochem. 42, 1181–1193.

Kim, Y., An, Y., Yoon, S.J., Jeong, H.D., Khim, J.S., Hong, S., 2019. Spatial Distributions of Polychlorinated Biphenyls (PCBs) in Surface Sediments of Coastal and Offshore Areas of South Korea. J. Hyrdo. Soc. Kor. 8, 19–27. (in Korean)

Liu, L.-Y., Wang, J.-Z., Wei, G.-L., Guan, Y.-F., Zeng, E.Y., 2012. Polycyclic aromatic hydrocarbons (PAHs) in continental shelf sediment of China: implications for anthropogenic influences on coastal marine environment. Environ. Pollut. 167, 155-162.

Mandalakis, M., Polymenakou, P.N., Tselepides, A., Lampadariou, N., 2014. Distribution of aliphatic hydrocarbons, polycyclic aromatic hydrocarbons and organochlorinated pollutants in deep-sea sediments of the southern Cretan margin, eastern Mediterranean Sea: a baseline assessment. Chemosphere 106, 28–35.

Savinov, V.M., Savinova, T.N., Carroll, J., Matishov, G.G., Dahle, S., Næs, K., 2000. Polycyclic aromatic hydrocarbons (PAHs) in sediments of the White Sea, Russia. Mar. Pollut. Bull. 40, 807–818.

Wang, G., Peng, J., Yang, D., Zhang, D., Li, X., 2015. Current levels, composition profiles, source identification and potentially ecological risks of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in the surface sediments from Bohai Sea. Mar. Pollut. Bull. 101, 834–844.

Yang, G.-P., 2000. Polycyclic aromatic hydrocarbons in the sediments of the South China Sea. Environ. Pollut. 108, 163–171.