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Distributions of persistent organic contaminants in sediments and their potential impact on macrobenthic faunal community of the Geum River Estuary and Saemangeum Coast, Korea

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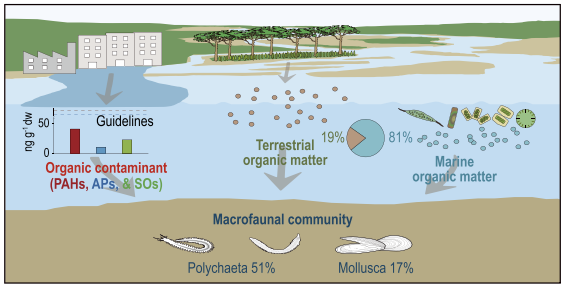
HIGHLIGHTS

- Persistent organic contaminants in estuarine and coastal sediments were surveyed.
- Distribution of organic contaminants were affected by surrounding activities.
- Organic contaminants mainly originated from near industrial and municipal areas.
- Terrestrial organic matter found the inner estuary and near the watergate regions.
- Origin of sedimentary organic matter can be controlled macrofaunal community.

GRAPHICAL ABSTRACT



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ABSTRACT

Over the last 30 years, the Geum River Estuary and Saemangeum Coast have been subject to major environmental changes, including dike construction, reclamation, and development of industrial complexes. This study aimed to: 1) investigate the occurrence of polycyclic aromatic hydrocarbons (PAHs), alkylphenols (APs), and styrene oligomers (SOs), 2) identify the sources of sedimentary organic matter, and 3) determine key environmental factors controlling the macrozoobenthos community structure. A total of 58 surface sediments were collected from the estuary and coastal area in 2014. Specific persistent organic contaminants (POCs), including 24 PAHs, 6 APs, and 10 SOs were measured. PAHs, APs, and SOs were detected in the sediments at all sites, with concentrations varying among sites. Although POCs concentrations were generally below the Canadian sediment quality guidelines, relatively greater concentrations of POCs were found at some sites adjacent to industrial complexes and the estuarine area. Sediment organic carbon, total nitrogen, and the stable carbon isotope ratio ($\delta^{13}C$) were determined. Some sites near watergate had about 2–3‰ lighter $\delta^{13}C$ values compared to other areas, indicating that these sites are affected by terrestrial organic matter. The number of species in the macrofaunal

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Organic matter
Macrofaunal community

community was significantly correlated with $\delta^{13}C$ values ($p < 0.001$), positively, suggesting that the origin of sedimentary organic matter is important for controlling the macrozoobenthos distribution. Overall, this research provides information about the level and sources of sediment pollution, the origins of organic matter, and the relationships with the macrofaunal community.

1. Introduction

The Geum River Estuary has been subject to major changes since the construction of a sea dike in 1990 and the establishment of the Gunsan-industrial complex in 1992 (Kim et al., 2006). Likewise, the Saemangeum Coast located to the south of the Geum River Estuary undergone major changes following the construction of the Saemangeum sea dike between 1991 and 2010 (Lee and Ryu, 2008; Lie et al., 2008). Gunsan is a major city in southwestern Korea that is located near the Geum River Estuary and Saemangeum Coast. Over the years, this city has expanded to support a manufacturing industry and an international trade port (Yi and Ryu, 2015), which might cause local contamination with persistent organic contaminants (POCs) (Hong et al., 2012). Moreover, dike construction and reclamation induced erosion and change to in surface sediments (Lie et al., 2008). These activities might alter the biogeochemical conditions of particulate organic matter in coastal areas and benthic environments (Lee et al., 2012).

Toxic chemicals, such as polycyclic aromatic hydrocarbons (PAHs), alkylphenols (APs), and styrene oligomers (SOs) have been found near industrial complexes and in waste from cities and large harbors in Korea (Khim et al., 1999; Koh et al., 2006; Hong et al., 2016). PAHs are ubiquitous POCs in various environments, particularly intensively used areas, such as dockyards, harbors, estuaries, and shallow coastal zones exposed to anthropogenic effects (Lipiatou et al., 1997; Rogers, 2002). PAHs are generally accumulated with relatively great concentrations in sediments due to their hydrophobic nature. PAHs can have also high degree of biota-sediment accumulation factors by being accumulated in coastal benthic organisms (Gewurtz et al., 2000). PAHs have long been considered as one group of major toxic contaminants found in coastal sediments to cause adverse effects on aquatic wildlife (Neff, 1979, 2002). Alkylphenol ethoxylates (APEOs), which include nonylphenol polyeothoxylates (NPEOs) and octylphenol polyeothoxylates (OPEOs), are extensively used as nonionic surfactants (White et al., 1994). APEOs can be degraded into products such as nonylphenol and octylphenol through biological and photochemical degradation (Li et al., 2013). However, the degradation products, endocrine disruptors, have harmful effects, including population decrease and the feminization of several aquatic species (Giesy and Snyder, 1998; Chen and Yen, 2013).

SOs have been reported as new pollutants in highly developed coastal areas (Hong et al., 2016). SOs are known decomposition chemicals that originate from polystyrene plastic materials, and have been listed as new contaminants of increasing concern; yet, few studies have reported the distribution of SOs (Saido et al., 2014; Kwon et al., 2015; Hong et al., 2016). SO analogues originate from the thermal decomposition of polystyrene at temperatures of 240–300 °C (Kwon et al., 2014). SOs have been reported to cause estrogenic effect *in vitro* and reproduction toxicity on daphnids (Ohyama et al., 2001; Tatarazako et al., 2002). Thus, sedimentary SOs may cause potential adverse effects on ecosystem, but effects on benthic community are not well known. Altogether, study on the occurrences and distributions of PAHs, APs, and SOs in sediments and benthic community responses would remain in question.

In general, sediment organic carbon (SOC) is positively correlated with organic contaminants (Warren et al., 2003). The SOC content and origin of organic matter is affected by inputs of freshwater. The origin of organic matter is generally determined by using the SOC to sediment nitrogen (SN) ratio and stable carbon isotopes (Sampei and Matsumoto, 2001; Meksumpun and Meksumpun, 2002). It is important to determine the origin because terrestrial organic matter is connected with an increase in macrofaunal community biomass and density (Hermand et al., 2008).

However, there have been few studies on organic contaminants, the sources of organic matter, and factors affecting the benthic macrofaunal community inhabiting the sediment of the Geum River Estuary and Saemangeum Coast. In the present study, we aimed to 1) investigate distribution of PAHs, APs, and SOs, 2) identify sources by analysis of chemical compositions, 3) investigate sources of sedimentary organic matter by use of carbon stable isotope ratio, and finally 4) determine macrobenthic community responses against sedimentary contamination.

2. Material and methods

2.1. Sampling areas and strategy

The Geum River Estuary and Saemangeum Coast are located on southwestern part of Korea. These areas are affected by three watergates. The inner part of the Saemangeum sea dike is influenced by freshwater originating from the Mangyeong and Dongjin Rivers. The Geum River Estuary and the Saemangeum Coast surround the industrial and domestic areas of the cities of Gunsan and Seocheon. Fifty-eight sediment samples were collected from the Geum River Estuary and Saemangeum Coast (Fig. 1). Thirty sediment samples were collected from the Saemangeum Coast in September 2014. The other samples were collected from the Geum River Estuary in December 2014. Surface sediments were collected for chemical analysis by use of Van Veen grab sampler. Upper 2 cm of the sediments were collected. All sediment samples for chemical analyses were transported with dry-ice and stored at −20 °C until analysis. For macrofaunal community, duplicate sediment samples were collected at each site. Macrofauna were separated by using a 1 mm mesh sieve on site, and were fixed with 5% buffered formalin.

2.2. Sample preparation

To analyze the organic chemicals, the samples were freeze-dried and homogenized and were extracted with 350 mL dichloromethane (DCM, Burdick & Jackson, Muskegon, MI) using Soxhlet extractor for 16 h. Five surrogate standards (SS, acenaphthene-d10, phenanthrene-d10, chrysene-d12, perylene-d12, and bisphenol A-d16) were added before extraction. The extracts were concentrated and replaced with hexane (Burdick & Jackson) using a rotary evaporator and activated copper (Sigma Aldrich, Saint Louis, MO) was added to remove elemental sulfur. The extracts were purified and fractionated by passing it through 8 g silica gel (70–230 mesh, Sigma Aldrich). The first fraction (F1) contained PAHs and SOs, and

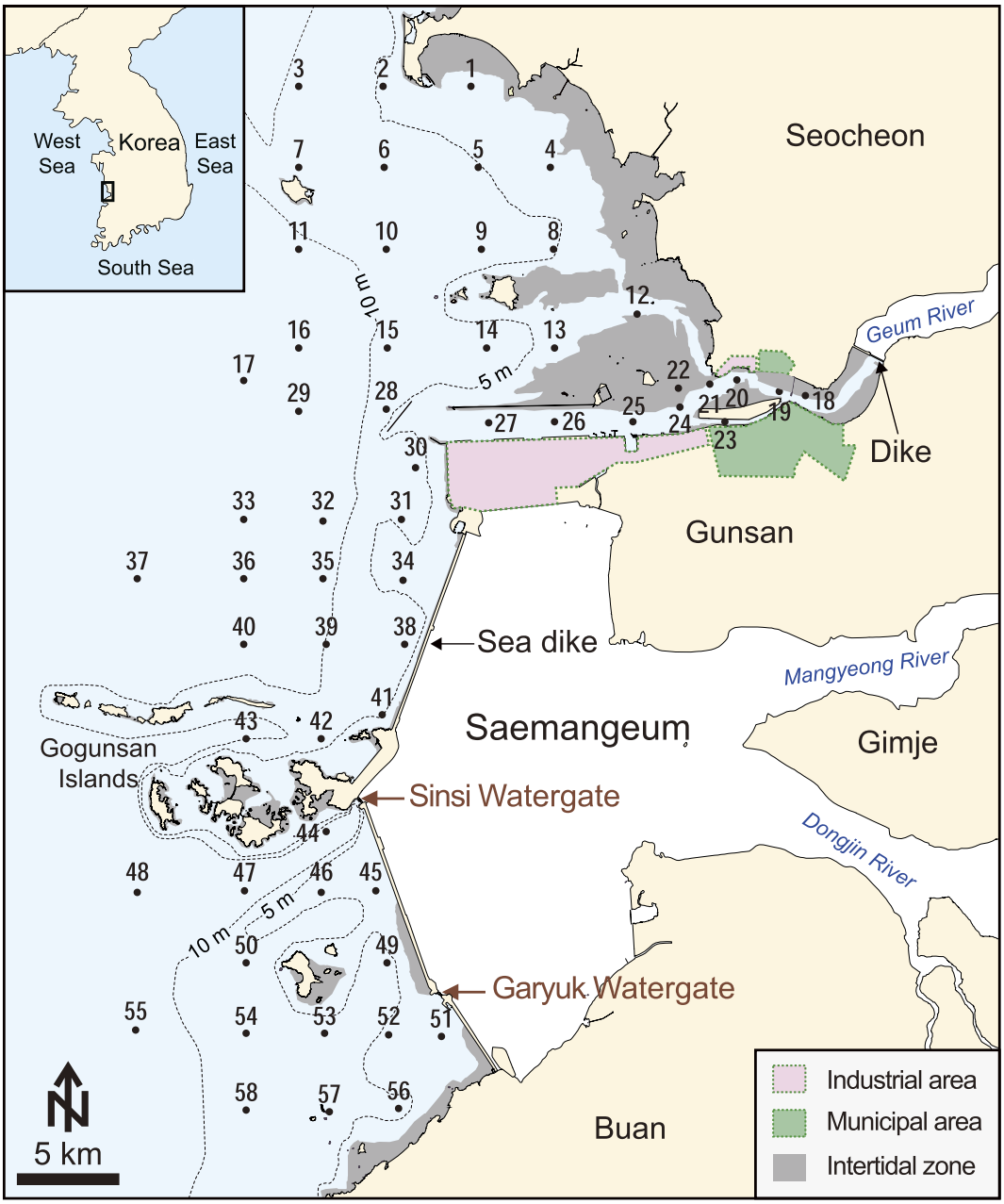

Fig. 1. Map showing the sampling sites of the surface sediment at the Geum River Estuary and the Saemangeum Coast.

was eluted with 50 mL hexane:DCM (80:20, v/v). The remaining alkylphenols were eluted in the second fraction (F2) with 50 mL DCM:acetone (J.T. Baker, Center valley, PA) (60:40, v/v). F1 and F2 were concentrated to 1 mL under nitrogen flow, and internal standard (2-fluorobiphenyl) was added. The mixture was transported in a vial for instrumental analysis.

To analyze SOC, SN, and stable carbon isotopes, the samples were freeze-dried and homogenized. Approximately 50 mg of sediment was weighed using a microbalance (Mettler Toledo, Columbus, OH) and packed in a tin capsule for SN analysis. A portion of the sample was acidified with 1 M hydrochloric acid (HCl, Sigma Aldrich) to eliminate inorganic carbon and rinsed with distilled water for SOC and stable carbon isotope analyses. The acidified samples were repeatedly freeze-dried, weighed, and packed.

2.3. Measurement of PAHs, APs, and SOs

The concentration of PAHs, APs, and SOs was determined using an Agilent 7890A gas chromatograph equipped with a mass-selective detector (MSD, Agilent Technologies, Santa Clara, CA). Details on the instrument condition for PAHs, SOs, and APs are provided in Tables S1 and S2 of the Supplementary Materials (S).

2.4. Measurement of the carbon stable isotope ratio

The elemental content (SOC and SN) and stable carbon isotope ratio ($\delta^{13}C$) were measured using an Elemental Analyzer (EA)-Isotope Ratio Mass Spectrometer (IRMS, Elementar, Hanau, Hesse). High purity carbon dioxide was used as reference gas, while helium and oxygen was used as the carrier and combustion gas,

Table 1
Overview of the results for chemical and ecological analyses in the sediments of the Geum River Estuary and the Saemangeum Coast, Korea.

Analysis		Unit	Range	Sediment (n = 58)
Sediment property	Mud content	%	Min.– Max.	2.4–99
			Mean	29
Persistent Organic Chemicals	PAHs[a]	ng g^{-1} dw	Min.– Max.	2.9–158
			Mean	39
	APs[b]	ng g^{-1} dw	Min.– Max.	0.6–46.0
			Mean	9.5
	SOs[c]	ng g^{-1} dw	Min.– Max.	0.3–261
			Mean	14
Organic matter	Sediment organic carbon	%	Min.– Max.	0.1–1.3
			Mean	0.3
	Sediment nitrogen	%	Min.– Max.	0.01–0.15
			Mean	0.03
	Carbon stable isotope ratio ($^{13}C/^{12}C$)	‰	Min.– Max.	−19.9–−23.9
			Mean	−21.6
Macrofunal community	Total species number	number	Min.– Max.	3–51
			Mean	11
	Total density	indiv/m^2	Min.– Max.	15–2350
			Mean	569
	Total biomass	g WW/m^2	Min.– Max.	0.3–1266
			Mean	101

[a] Concentration of PAHs was used the sum of 16 parent PAHs, 2-Na, 1-Na, 1,3-Na, 1-Flu, 3-Phe, 2-Phe, 3-Chr, and perylene.
[b] Concentration of APs was used the sum of OP, OP1EO, OP2EO, NP, NP1EO, and NP2EO.
[c] Concentration of SOs was used the sum of SD1, SD2, SD3, SD4, ST1, ST2, ST3, ST4, ST5, and ST6.

respectively. To quantify SOC and SN, sulfanilamide was used as the external standard. Stable carbon isotope ratios were expressed in ‰ delta notation as (Eq. (1)):

$$\delta^{13}C\ (‰) = \left[\frac{R_{sample}}{R_{reference}} - 1\right] \times 1000 \quad (1)$$

where $R_{sample}/R_{reference}$ are the ratios ($^{13}C/^{12}C$) of the sample and reference, respectively. The isotope value was reported in relation to the Vienna Pee Dee Belemnite. To calibrate $\delta^{13}C$, the international isotope standards, IAEA-CH-3 and IAEA-CH-6, were used.

2.5. Macrofauna analysis

After transferring the macrofauna to the laboratory, the samples were re-sieved using a 1 mm screen to rinse away formalin the before analysis. The macrofaunal samples were sorted, identified to the species level, and counted using a dissection microscope. The grain size of the sediments were analyzed using the dry sieve and pipette method.

2.6. Quality assurance and quality control

Method detection limit (MDL) was calculated as 3.707 × standard deviation of standard. The MDLs of PAHs, SOs, and APs ranged from 0.23 to 1.4 ng g^{-1}, from 0.28 to 0.94 ng g^{-1}, and from 0.09 to 0.97 ng g^{-1}, respectively. The mean recoveries of five SS were generally within the acceptable range (78–111% for PAHs and SOs; 75% for APs; detailed in Table S3). In the carbon and nitrogen stable isotope analysis, the analytical precision of the IAEA-CH-3 and IAEA-CH-6 were 0.2‰ and 0.1‰, respectively.

2.7. Data analysis

In this study, 24 PAHs, 6 APs, 10 SOs, organic carbon, nitrogen, and stable carbon isotope were investigated in the surface sediments (full names of the chemicals and abbreviations are shown in Tables S1 and S2). Principal component analysis (PCA) was performed using the normalized values of the environmental parameters and macrofaunal community data. SPSS 23.0 (SPSS INC., Chicago, IL) and SigmaPlot 13.0 software were used for the statistical analyses.

3. Results and discussion

3.1. Spatial distribution of persistent organic contaminants

The concentration of organic contaminants in the sediments is shown in Table 1 and Fig. 2. The mean concentration of PAHs, APs, and SOs ranged from 39.6, 9.46, and 14.3 ng g^{-1} dry weight (dw) respectively (details in Tables S4, S5, and S6). Looking for the distributions of POCs, most of target compounds were concentrated (top 20%) in the mouth of the Geum River Estuary to the Gogunsan Islands along the Saemangeum sea dike, indicating hot spots along this line probably due to the industrial and municipal activities from Gunsan area (Fig. 2). The mean PAHs concentrations of top 10%, top 10–20%, and < top 20% were 129 ng g^{-1} dw, 80 ng g^{-1} dw, and 23 ng g^{-1} dw, respectively. The concentrations of detectable PAHs varied among the sites and regions, while APs showed narrow groupings in terms of range of concentrations (top 10%, top 10–20%, and < top 20%; 25 ng g^{-1} dw, 14 ng g^{-1} dw, and 6.9 ng g^{-1} dw, respectively). The range of SOs concentrations was also relatively small (top 10%, top 10–20%, and < top 20%; 87 ng g^{-1} dw, 19 ng g^{-1} dw, and 4.2 ng g^{-1} dw), in general. Relatively smaller concentrations of POCs were detected at several long distanced sites from the coastline. These results indicated that the industrial and municipal activities were the main sources of POCs, and seemed to cause relatively great accumulation in nearby bottom sediments (Ashley and Baker, 1999). The greater POCs concentrations in some remote sites may be affected by geological hydrodynamic conditions and resuspension by dredge activity for port (Chen et al., 2006). Overall, spatial distributions of POCs indicated that POCs were mainly accumulated in sediments of adjacent sources due to point-sources and could be transported to limited area.

The distribution pattern of POCs was similar to those of SOC and mud content. The mud content was positively correlated with POCs ($r = 0.27–0.67$, $p < 0.05$). The SOC was significantly correlated with PAHs and APs ($r = 0.31–0.70$, $p < 0.01$). Unlikely PAHs and APs, no significant correlation was observed between SOs concentration and SOC ($r = 0.14$, $p = 0.74$). Correlation between $\delta^{13}C$ and POCs was

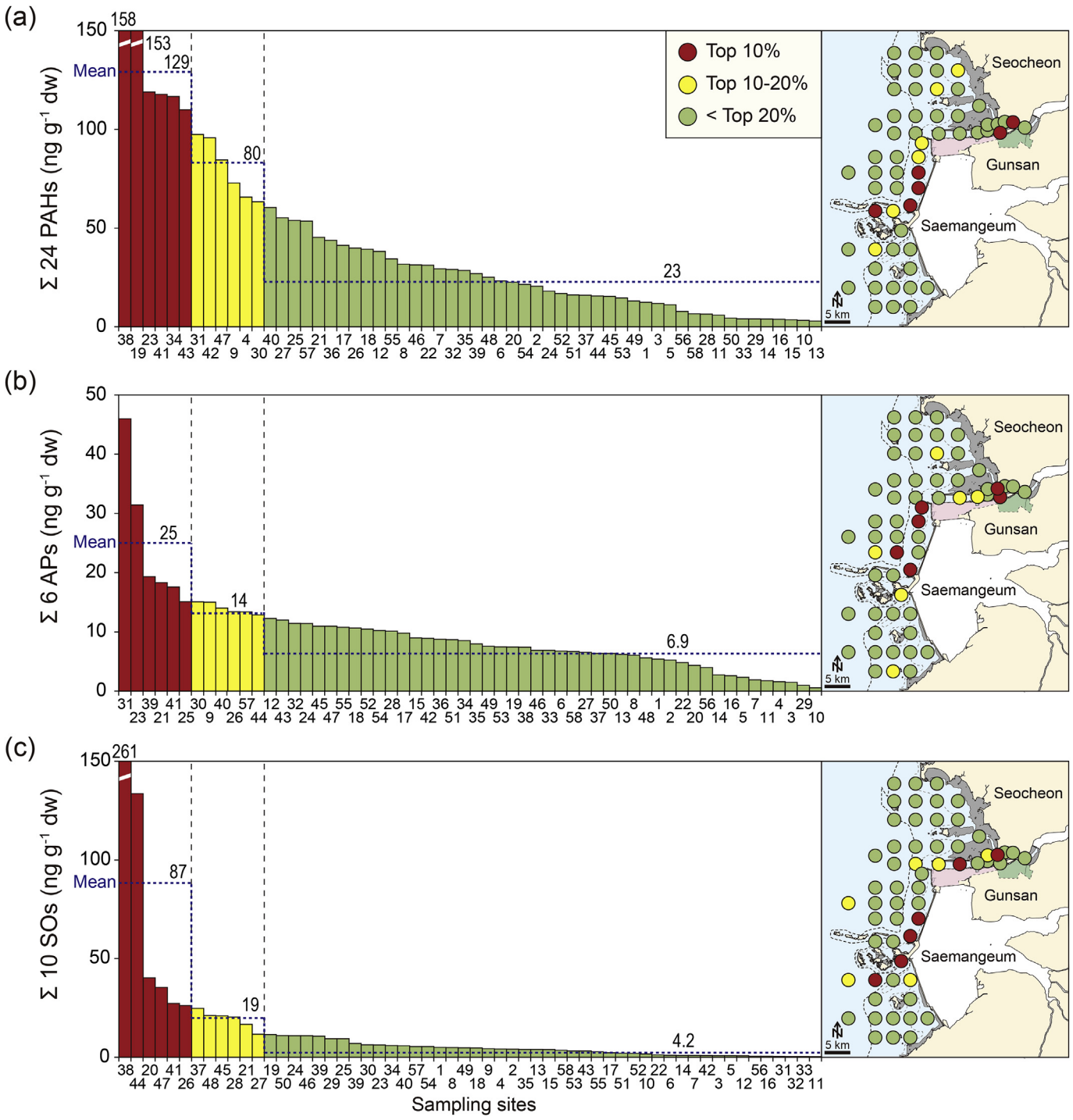

Fig. 2. Spatial distributions of (a) 24 PAHs, (b) 6 APs, and (c) 10 SOs in sediments of the Geum River Estuary and Saemangeum Coast. Red, yellow, and green areas mean the top 10%, between the top 10 and 20%, and below the top 20%, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

not significant ($r = 0.04–0.22$, $p > 0.05$). PAHs and APs were highly correlated with SOC (Xu et al., 2006; Liu et al., 2013), but correlation between SOs and SOC was not found probably due to lack of data on measured SOs (10 chemicals) at this time. This result indicated that concentrations of POCs were not apparently affected by the origin of organic matter. Of note, the concentration of SOs was correlated with sediment grain size rather than SOC. The major reason for lack of correlation between POC and $\delta^{13}C$ might be attributed to POC sources being located in the outer region of estuary. Due to limited information of SOs in sediment, further characterization of SOs accumulation would remain in question. In anyhow, the overall distributions of POCs in the Geum River Estuary and Saemangeum Coast were collectively affected by general sediment characteristics, particularly the organic matter.

The concentrations of PAHs, APs, and SOs in the sediments of the present study area were compared with the results of previous

studies in the coastal areas affected by industrial complexes and domestic areas of Korea. Overall, similar or less concentrations of PAHs and APs were detected in the sediments of the Geum River Estuary and the Saemangeum Coast compared to reports for sediments in other coastal areas, such as Gyeonggi Bay, Kwangyang Bay, Masan Bay, and Yeongil Bay (Koh et al., 2005, 2006; Moon et al., 2008; Hong et al., 2009). Moreover, the greatest concentrations of PAHs and APs in the sediments of the Geum River Estuary and Saemangeum coasts were below the Interim marine sediment quality guidelines (CCME, 2001). Thus, the contamination level of PAHs and APs in the sediments at present study area was much lower compared to other coastal sediments in Korea. In SOs (10 chemicals), only one study has been previously conducted in Korea in Gyeonggi Bay (Hong et al., 2016). The concentrations of SOs obtained in present study were similar to those obtained in the previous study. Thus, the contamination level of PAHs, APs, and SOs in the Geum River Estuary and Saemangeum Coast was moderate to low compared to other areas in Korea. This results indicated that the sediment conditions of the areas assessed by the present study were better compared to other coastal areas in Korea.

3.2. Composition and sources of persistent organic contaminants

The composition of PAHs in the sediments of the Geum River Estuary and Saemangeum Coast differed among samples (Fig. 3a). Out of all sites, the top 10% and top 10–20% contained PAHs of high molecular weight (HMW), representing 75% of total PAHs in both groups. However, HMW accounted for about 59% of sites below the top 20%. HMW PAHs (4–6 rings) appeared to be affected by pyrogenic sources (Gschwend and Hites, 1981; Budzinski et al., 1997). Low molecular weight PAHs (2–3 rings) might be derived from air-water exchange and atmospheric deposition, due to their relatively high volatility (Tobiszewski and Namieśnik, 2012). Thus, this composition indicated that industrial complex affected as main

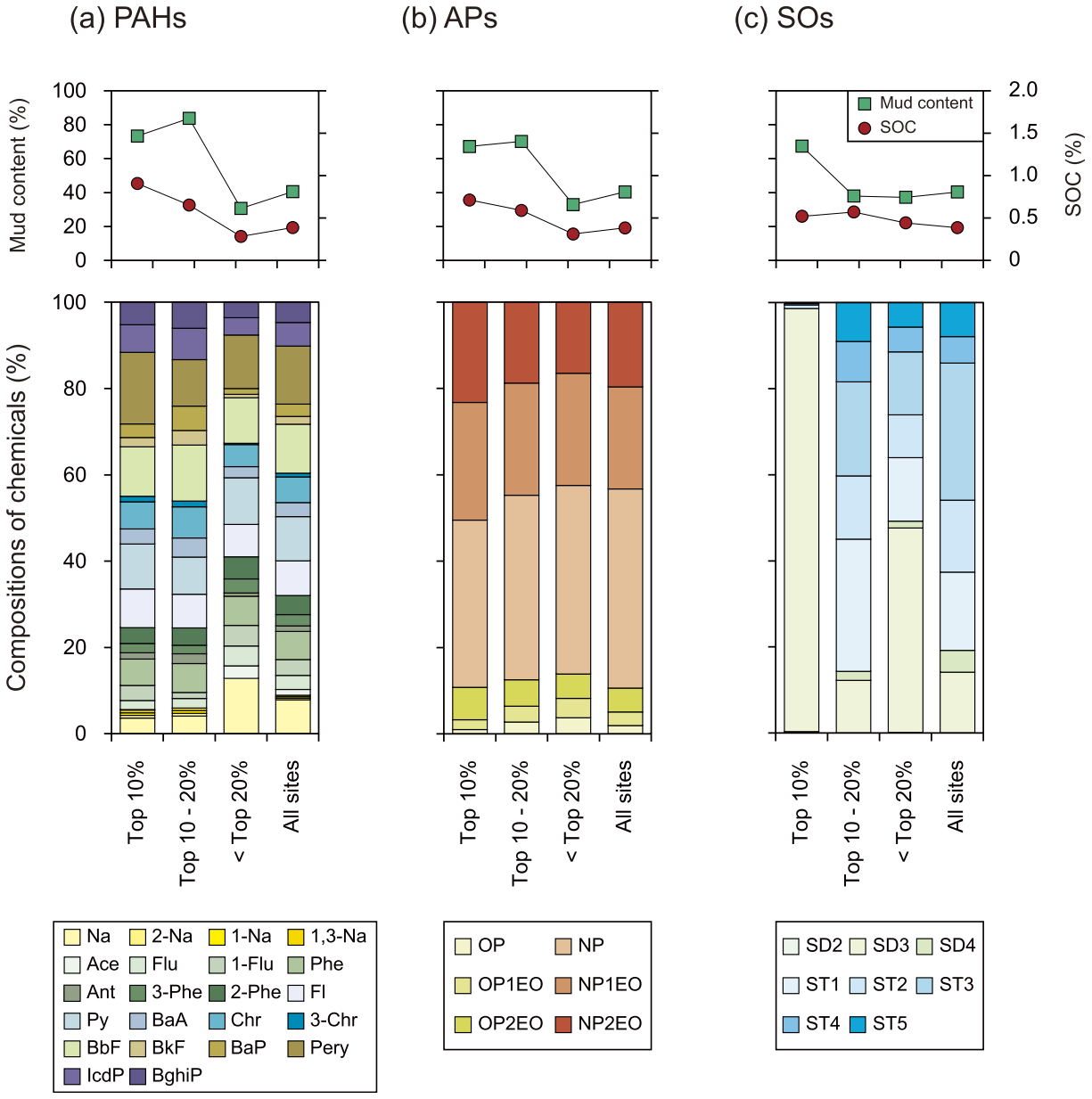

Fig. 3. Relative compositions of (a) PAHs, (b) APs, and (c) SOs in sediments of the Geum River Estuary and the Saemangeum Coast. The mud contents and SOC contents were given.

source. To analyze the sources of PAHs, diagnostic ratios were used as the principal method (Fig. S1). The results suggested that the sources of PAHs in the study area primarily originated from pyrogenic sources. These results correspond with the dominance of HMW PAHs, which are affected by pyrogenic sources. Pyrogenic sources might originate from the surrounding activities. Consequently, the PAHs in the sediment layer of the Geum River Estuary and Saemangeum Coast were affected by pyrogenic sources originating from sources in the surrounding area, such as the industrial complex.

Unlike PAHs, the composition of APs was similar across sites of great and less concentrations (Fig. 3b). Overall, NPs and nonylphenol monoethoxylate (NP1EO) were the dominant compounds accounting for 43% and 26% of APs in all sites, respectively. This result supports previous studies (Duan et al., 2014; Dong et al., 2015). NP and OP were the degradation chemicals of NP and OP polyethoxylates. In this study, more NP was detected compared to NP1EO + nonylphenol diethoxylate (NP2EO); however, lower OP was calculated compared to octylphenol monoethoxylate + octylphenol diethoxylate (Fig. S1). Thus, fresh input consists of OP derivatives; but, concentrations were negligible. Lesser NP1EO and NP2EO concentrations might have been influenced by their limited uses in household products being banned since 2002 in Korea. Overall, NP derivatives were dominant, with fresh inputs of OP derivatives being detected. This result indicated that although a small quantity of fresh input was detected, regulation of their uses seemed to be effective in more recent years.

The compositions of SOs in the sediments of the Geum River Estuary and Saemangeum Coast varied among sites, mostly because of site 38 (top 1) (Fig. 3c). At site 38, SD3 (98%) was the dominant chemical (Table S6). However, for other sites in the top 10%, ST1

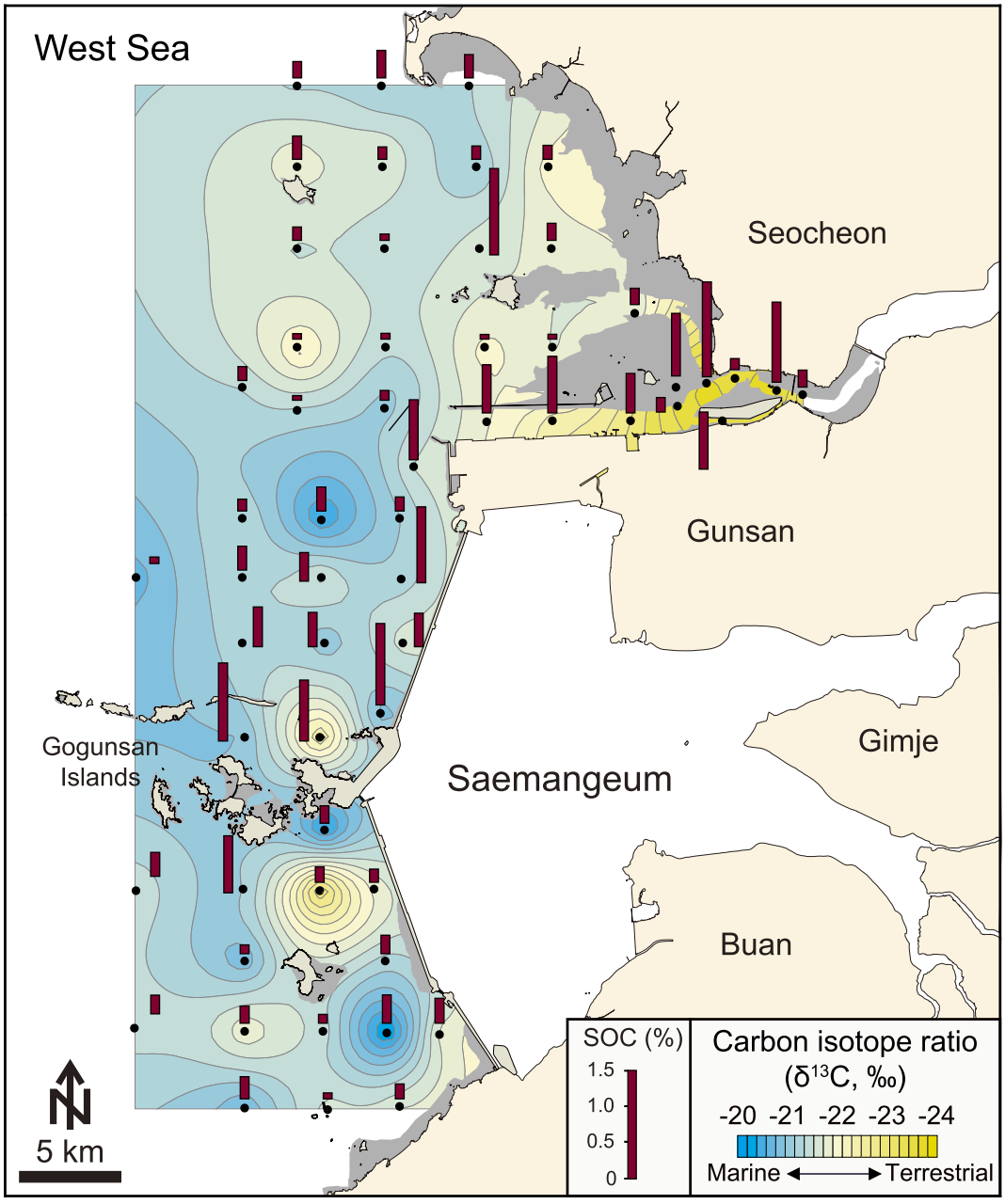

Fig. 4. Spatial distributions of $\delta^{13}C$ values and total organic carbons in sediments of the Geum River Estuary and the Saemangeum Coast.

(26%) was the dominant chemical, followed by ST3 (25%), ST2 (16%), and SD3 (11%). Likewise, in sites top 10–20%, ST3 was the dominant chemical with 26%, followed by ST1 (22%), ST2 (18%), and SD3 (11%). In sites below the top 20%, a similar pattern was found. The great concentration of SD3 found at site 38 might be affected by adjacent industrial sources, because the greatest concentration of PAHs was also found at site 38. SD1 and ST6 were not detected in all sites. Relatively great composition of STs may indicate a fresh input of SOs because styrene monomer would be detected after decomposition of polystyrene (Kwon et al., 2014). Thus, this result might indicate that fresh input of SOs would exist in the study area. A recent study performed by Hong et al. (2016) first investigated 10 SOs in lake and coastal sediments (same as present study) with relatively great contributions of SDs. However, the current study found that STs were the predominant chemical group among the SOs. This difference of SOs composition might be explained by the sources, i.e., by direct inputs (Hong et al., 2016) and/or specific decomposition mechanism of polystyrene at nearby sources. Overall, the lack of reports on the 10 SOs in the sediment means that more studies are required on their composition and distribution in coastal sediments.

3.3. Sources and distribution of organic matter

The $\delta^{13}C$ values in the sediments of the Geum River Estuary and Saemangeum Coast ranged between −23.9‰ and −19.9‰, with a mean value of −21.6‰ (Table 1 and Fig. 4). The $\delta^{13}C$ values of marine organic matter and terrestrial organic matter clearly differ. Previous studies reported lower $\delta^{13}C$ values for organic matter of terrestrial origin compared to the $\delta^{13}C$ values for marine origin. Terrestrial organic matter had a $\delta^{13}C$ value ranging between −25‰ and −27‰ (Schubert and Calvert, 2001; Lehmann et al., 2002). In comparison, the value for marine organic matter ranges between −22‰ and −20‰ (Peters et al., 1978; Meyers, 1994). The mouth of the Geum River, Site 42, and 46 (located near Shinsi-watergate) had relatively lighter $\delta^{13}C$ values (<−22.0‰). Relatively heavier $\delta^{13}C$ values were detected at other sites. In the West Sea, Korea, benthic particulate organic matter and suspended particulate organic matter were found to range between −13‰ and −22‰ (Suh and Shin, 2013). This results indicate that the organic matter of the sediments in sites near to freshwater inputs is of terrestrial origin (DeLaune, 1986; Chmura et al., 1987). In Saemangeum Coast, the marine origin of organic matter prevailed which could be explained by the weak freshwater input as well as seawater circulation at Sinsi- and Garyuk Watergates. The difference in hydrodynamics on water flow followed by the spatial variations of organic matter between Geum River Estuary and Saemangeum Coast seemed to cause dissimilar species composition of macrozoobenthos between two regions, in general (Hermand et al., 2008). Overall, the organic matter in the coastal area of the Geum River Estuary and Saemangeum Coast was mostly of marine origin; however, the organic matter of some sites located near the watergate was of terrestrial origin.

The SOC and SN contents ranged between 0.06% and 1.27% (mean: 0.38%) and 0.01%–0.15% (mean: 0.04%), respectively (Table 1, Fig. 4, and Fig. S2). High SOC contents were detected at sites located along the domestic and industrial area, from within the Geum River Estuary to the Gogunsan Islands. Lower SOC contents were detected at all other sites, except site 9 located near an island. The distribution of SN was similar pattern as SOC and organic chemical concentrations across sites. Previous studies reported that organic carbons tend to contain smaller particles (Guo et al., 2009), with similar results being obtained for sites with a high composition of mud content in our study (Fig. 3). Sedimentation

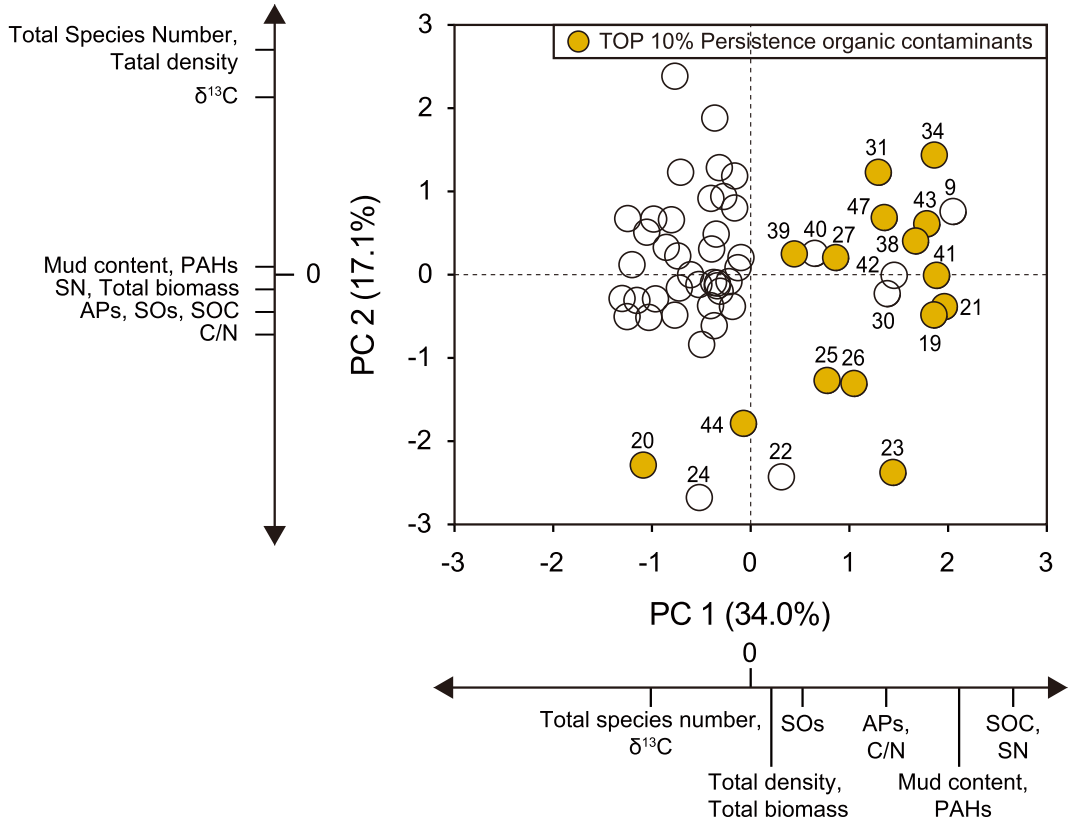

Fig. 5. Principal component analysis (PCA) for factors controlling the macrofaunal community.

conditions might be affected by the complex hydrodynamic and water current conditions of the study area. Overall, the SOC and SN contents in the study area appeared to be correlated with sediment grain size.

The SOC/SN ratios ranged from 4.15 to 17.93 (mean: 9.53; Fig. S3). The SOC/SN ratios indicated that most organic matter (81%) was of marine origin (SOC/SN between 4 and 12), while only a small percentage (19%) was of terrestrial origin (SOC/SN > 12) (Holligan et al., 1984; Stein, 1991). The highest SOC/SN values (>12) were detected in mouth of the Geum River Estuary and in the vicinity of the watergate of the Saemangeum sea dike. This result was consistent with the $\delta^{13}C$ value, indicating that the sedimentary organic matter in the Geum River Estuary and Saemangeum Coast is partially affected by terrestrial inputs.

3.4. Association of POCs contamination to the macrofaunal community

The present study detected 10,878 individuals from 186 macrofaunal species belonging to four major taxonomic groups. In all sites, Polychaeta was main group (51%), followed by Arthropoda (26%), Mollusca (17%), and Echinodermata (3%). Polychaeta was well known as an opportunistic species and indicator of organic enrichment (Martínez-Lladó et al., 2007; Seo et al., 2014). *Heteromastus filiformis* was the single most abundant species of all counted individuals (18.9%). The next dominant species was *Sinocorophium sinensis* (4.7%), followed by *Chaetopterus* sp. (4.0%), *Spiochaetopterus costarum* (3.7%), and *Photis brevipes* (3.7%). *Heteromastus filiformis, Sinocorophium sinensis,* and *Spiochaetopterus costarum* are opportunistic species that are used as indicators of organic pollution (Pearson and Rosenberg, 1978; Hong et al., 1997). The presence of these species indicates that the sediments of the Geum River Estuary and Saemangeum Coast have been subject to organic enrichment. Thus, present study focused on identifying the factors that control the macrofaunal community in the Geum River Estuary and Saemangeum Coast.

Principal component analysis (PCA) was performed to identify the factors controlling the macrofaunal community, by using all available parameters (macrofaunal community, POCs, organic matter, and mud content) (Table 1 and Fig. 5). The PCA divided the sites into two groups. The first group included the industrial and domestic area, the area near the sea dike, and the Gogunsan Islands. These sites had greater concentrations (top 10%) of POCs, larger species ranges, and a greater density of macrofauna. The second group was characterized by sites that had relatively high species numbers and density, as well as high $\delta^{13}C$ values. The results indicated that the parameters of the macrofaunal community were more strongly affected by $\delta^{13}C$ compared to POCs, mud content, SOC, or SN. However, PC1 and PC2 only explained 51% of variance; thus, the high correlation factors with the macrofaunal community should be treated with caution.

The correlation of the macrofaunal community with environmental parameters was used to determine the coefficient of determination and *p*-value (Table S7). The total number of species was significantly correlated with $\delta^{13}C$ ($r^2 = 0.52$, $p < 0.01$); however, there was only a low correlation with the other parameters. The total density and biomass were poorly correlated with all parameters, except for total biomass and $\delta^{13}C$ ($p < 0.05$). In the macrofaunal community, the number of Polychaeta species was highly correlated with $\delta^{13}C$ ($r^2 = 0.45$, $p < 0.01$), followed by Arthropoda and Mollusca (Fig. S4). Overall, the macrofaunal community (particularly Polychaeta) was more highly correlated with $\delta^{13}C$ compared to all other parameters.

Table 2
Characteristics of sites 17 to 30 classified based on their environmental parameters and macrofaunal community.

Sites	17	29	28	30	27	26	25	22	24	21	23	20	19	18
Environmental parameters														
$\delta^{13}C$ (‰)	−21.2	−21.2	−20.9	−21.4	−21.8	−22.3	−22.8	−23.5	−23.5	−23.7	−23.6	−23.4	−23.9	−23.0
Total organic carbon	0.2	0.1	0.1	0.8	0.6	0.8	0.5	0.8	0.2	1.3	0.8	0.1	1.1	0.3
Polycyclic aromatic hydrocarbons	41.3	4.0	6.4	63.4	55.2	39.9	53.9	31.2	18.1	45.3	119.0	22.6	153.1	39.3
Alkylphenols	9.8	1.0	10.1	15.1	6.6	13.4	15.1	4.8	11.4	18.3	31.4	4.4	7.5	10.7
Styrene oligomers	2.3	9.5	20.4	6.3	11.7	26.2	9.5	1.2	11.0	16.7	6.3	40.3	11.5	4.7
Macrofaunal community														
Total species number	44	36	39	22	30	14	16	6	5	12	3	3	12	11
Total individuals number	463	243	349	144	181	107	77	18	17	413	3	23	284	212
Total biomass (g wet weight)	29.2	1.7	4.7	2.3	3.0	3.7	1.3	0.4	4.2	1.8	0.1	52.9	253.2	117.5
Dominant species[a] (%)														
(P) *Chaetopterus* sp.	**34.6**	-	–	–	-	–	–	–	-	–	–	–	–	–
(A) *Gammaropsis japonicus*	9.9	-	–	–	-	–	–	–	-	–	–	–	–	–
(A) *Photis brevipes*	**20.7**	2.5	2.3	–	**29.8**	-	–	-	–	-	-	-	-	-
(P) *Spiochaetopterus costarum*[b,c]	–	**36.6**	**47.3**	–	–	–	–	–	–	–	–	–	–	–
(A) *Urothoe brevicornis*	1.5	**23.5**	4.0	–	–	–	–	–	–	–	–	–	–	–
(P) *Sigambra tentaculata*[b]	–	0.8	0.3	7.6	–	3.7	–	–	–	0.20	–	–	–	–
(M) *Hydatina albocincta*	0.2	2.1	2.9	3.5	0.6	1.9	–	-	-	–	–	–	–	–
(P) *Heteromastus filiformis*[b,c]	1.1	3.7	5.4	**53.5**	4.4	**52.3**	18.2	**44.4**	**47.1**	8.5	–	–	3.5	1.4
(P) *Nectoneanthes multignatha*	0.2	0.4	0.9	0.7	19.3	–	11.7	–	–	–	–	–	0.4	–
(P) *Sternaspis scutata*	–	–	0.3	0.7	–	9.3	11.7	–	–	0.2	–	–	–	–
(M) *Theora fragilis*	–	–	–	–	–	13.1	–	–	–	–	–	–	–	–
(A) *Sinocorophium sinensis*[c]	–	–	–	–	–	–	**20.8**	–	–	**86.4**	–	13.0	15.1	**43.9**
(P) *Glycinde* sp.	–	–	–	0.7	–	4.7	14.3	–	–	1.0	–	–	1.1	–
(P) *Glycera chirori*	0.4	1.2	2.3	–	4.4	–	5.2	11.1	**35.3**	0.5	–	–	0.4	0.5
(P) *Neanthes japonica*[d]	–	–	–	–	–	–	–	–	–	0.5	-	–	1.4	0.5
(P) *Prionospio membranacea*[c]	–	–	–	–	–	–	–	–	–	0.2	**33.3**	13.0	-	**22.6**
(M) *Potamocorbula amurensis*	–	–	–	–	–	–	–	–	–	–	–	**73.9**	**27.5**	**22.2**
(A) *Cirripedia* sp.	–	–	–	–	–	–	–	–	–	–	–	–	**47.9**	4.7

[a] Species in bold indicate >20% individuals in all species and acronyms: (P) Polychaeta, (A) Arthropoda, and (M) Mollusca.
[b] Opportunistic species.
[c] Organic polluted or enriched indicators.
[d] Brackish water species.

The distribution of the macrofaunal community was affected by $\delta^{13}C$ along the Geum River Estuary (between sites 17 and 30), and differed with increasing distance from the mouth of the estuary (Table 2). High numbers of species were found at sites that contained heavier $\delta^{13}C$ values and that were located far away from mouth of Geum River Estuary (Site 17, 29, 28, 30, and 27). But, the numbers of individuals and biomass were similar, or lower, further away from the mouth when compared to closer to the mouth. A low number of species was detected at sites, which were supplied with terrestrial organic matter ($\delta^{13}C < -23$). But, high biomass was detected near dike due to *Potamocorbula amurensis*, which is an invasive species (Lowe et al., 2000). This high biomass dominated because the estuarine area was highly affected by the river input of terrestrial phytodetritus (Hermand et al., 2008). Many Polychaeta species were dominated in sites affected by terrestrial organic matter because they exploit terrestrial organic matter (Darnaude et al., 2004). Furthermore, opportunistic species and organic pollution indicator species (such as *Spiochaetopterus costarum*, *Sigambra tentaculata*, *Sinocorophium sinensis*, and *Prionospio membranacea*) dominated at the sites affected by terrestrial organic matter. Thus, input of terrestrial organic matter caused enriched environment and influenced the macrobenthic faunal assemblages in the given area. Therefore, in general, origin of sedimentary organic matter was an important factor controlling the macrofaunal community at this time.

4. Conclusions

The present study was performed to investigate occurrence, distribution, and sources of POCs, origin of organic matters, and key environmental factors controlling the macrozoobenthos community structure. Overall, concentrations of POCs in the Geum River Estuary and Saemangeum Coast were generally less than suggested sediment quality guidelines and other highly industrialized areas in Korea. However, several hotspot sites showed relatively great concentrations of POCs, indicating localized point sources associated with land-use and inland activities. It was evidenced that terrestrial organic matter was mainly found in the inner estuary and several sites adjacent to the watergate. The macrofaunal community structure was primarily influenced by the terrestrial organic matter in the sediments when comparing a number of biological parameters including the number of species and species compositions. Although the present study suggests that the terrestrial organic matter is a key factor that controls the macrofaunal community, further study would be acknowledged at a larger and/or long-term scale to confirm key or limiting factors controlling the macrofaunal community change against the environmental settings.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2017.01.031.

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<Supplementary Materials>

Distributions of persistent organic contaminants in sediments and their potential impact on macrobenthic faunal community of the Geum River Estuary and Saemangeum Coast, Korea

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Supplemental Tables

Supplemental Figures

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Table S1. GC/MSD instrumental conditions for PAHs and SOs analyses.

GC/MSD system	Agilent 7890A GC and 5975C MSD
Column	DB-5MS (30 m long, 0.25 mm i.d., 0.25 μm film thickness)
Gas flow	1 mL/min He
Injection mode	Splitless
Injection volume	2 μL
MS temperature	180 °C
Detector temperature	230 °C
Oven temperature	60 °C hold 2 min Increase 6 °C/min to 300 °C 300 °C hold 13 min
Target PAHs, alkyl-PAHs and SOs	Naphthalene (Na), 2-Methylnaphthalene (2-Na), 1-Methylnaphthalene (1-Na), 1,3-Dimethylnaphthalene (1,3-Na), Acenaphthylene (Acl), Acenaphthene (Ace), Fluorene (Flu), 1-Methylfluorene (1-Flu), Phenanthrene (Phe), 3-Methylphenanthrene (3-Phe), 2-Methylphenanthrene (2-Phe), Anthracene (Ant), Fluoranthene (Fl), Pyrene (Py), Benzo[*a*]anthracene (BaA), Chrysene (Chr), 3-Methylcrysene (3-Chr), Benzo[*b*]fluoranthene (BbF), Benzo[*k*]fluoranthene (BkF), Benzo[*a*]pyrene (BaP), Perylene (Pery), Indeno[*1,2,3-cd*]pyrene (IcdP), Dibenz[*a,h*]anthracene (DbahA), Benzo[*g,h,i*]perylene (BghiP), 1,3-Diphenylproane (SD1), *cis*-1,2Diphenylcyclobutane (SD2), 2,4-Diphenyl-1-butene (SD3), 2,4,6-Triphenyl-1-hexene (SD4), 2,4,6-Triphenyl-1-hexene (ST1), 1e-Phenyl-4e-(1-phenylethyl)-tetralin (ST2),1a-Phenyl-4e-(1-phenylethyl)-tetralin (ST3), 1a-Phenyl-4a-(1-phenylethyl)-tetralin (ST4), 1e-Phenyl-4a-(1-phenylethyl)-tetralin (ST5), and 1,3,5-Triphenylcyclohexane (isomer mix) (ST6)

Table S2. GC/MSD instrumental conditions for APs analysis.

GC/MSD system	Agilent 7890A GC and 5975C MSD
Column	DB-5MS (30 m long, 0.25 mm i.d., 0.25 μm film thickness)
Gas flow	1 mL/min He
Injection mode	Splitless
Injection volume	1 μL
MS temperature	180 °C
Detector temperature	230 °C
Oven temperature	60 °C hold 5 min Increase 10 °C/min to 100 °C Increase 20 °C/min to 300 ° C
Target Alkyl phenols	300 °C hold 6 min 4-tert-Octylphenol (OP), 4-tert-Octylphenol monoethoxylate (OP1EO), 4-tert-Octylphenol diethoxylate (OP2EO), Nonylphenol (NP), Nonylphenol-monoethoxylate (NP1EO), and Nonylphenol diethoxylate (NP2EO)

Table S3. QA/QC data for sedimentary PAHs, SOs, and APs measured in the present study.

Compounds	Abbreviations	Method detection limit (ng g dw^{-1}, n = 7)	Surrogate recovery (%, n = 58)
PAHs and SOs			
Naphtalene	Na	0.43	
2-Methylnaphthalene	2-Na	0.80	
1-Methylnaphthalene	1-Na	0.78	
1,3-Dimethylnaphthalene	1,3-Na	0.65	
Acenaphthylene	Acl	0.85	
Acenaphthene	Ace	0.84	
Fluorene	Flu	0.80	
1-Methylfluorene	1-Flu	1.40	
Phenenthrene	Phe	0.79	
Anthracene	Ant	0.41	
3-Methylphenanthrene	2-Phe	0.28	
2-Methylphenanthrene	3-Phe	0.73	
Fluoranthene	Fl	0.78	
Pyrene	Py	0.90	
Benzo[a]anthracene	BaA	0.60	
Crysene	Chr	0.72	
3-Methylchrysene	3-Chr	0.24	
Benzo[b]fluoranthene	BbF	0.69	
Benzo[k]fluoranthene	BkF	0.71	
Benzo[a]pyrene	BaP	0.59	
Perylene	Pery	0.66	
Indeno[1,2,3-cd]pyrene	IcdP	0.43	
Dibenz[a,h]anthracene	DbahA	0.27	
Benzo[g,h,i]perylene	BghiP	0.33	
1,3-Diphenylproane	SD1	0.34	
cis-1,2-Diphenylcyclobutane	SD2	0.65	
2,4-Diphenyl-1-butene	SD3	0.94	
trans-1,2-Diphenylcyclobutane	SD4	0.28	
2,4,6-Triphenyl-1-hexene	ST1	0.57	
1e-Phenyl-4e-(1-phenylethyl)-tetralin	ST2	0.53	
1a-Phenyl-4e-(1-phenylethyl)-tetralin	ST3	0.30	
1a-Phenyl-4a-(1-phenylethyl)-tetralin	ST4	0.49	
1e-Phenyl-4a-(1-phenylethyl)-tetralin	ST5	0.32	
1,3,5-Triphenylcyclohexane (isomer mix)	ST6	0.34	
Acenaphthene-d10	Ace-d12		77.9 ± 19.1[a]
Phenanthrene-d10	Phe-d10		111.3 ± 28.2
Crysene-d12	Chr-d12		93.6 ± 20.6
Perylene-d12	Pery-d12		85.9 ± 20.4
APs			
4-tert-Octylphenol	t-OP	0.09	
Iso-Nonylphenol	NPs	0.97	
4-tert-Octylphenol-mono-ethoxylate	t-OP1EO	0.10	
Iso-Nonylphenol-mono-ethoxylate	NP1EOs	0.49	
4-tert-Octylphenol-di-ethoxylate	t-OP2EO	0.10	
Iso-Nonylphenol-di-ethoxylate	NP2EOs	0.88	
Bisphenol A-d16	BPA-d16		74.8 ± 18.7

[a] Mean ± SD

Table S4. Concentrations of PAHs and alkyl-PAHs in sediments of the Geum River Estuary and Saemangeum Coast.

Sites	**Polycyclic aromatic hydrocarbons (ng g^{-1} dry mass)**																								**ΣPAHs**
	Na	2-Na	1-Na	1,3-Na	Acl	Ace	Flu	1-Flu	Phe	3-Phe	2-Phe	Ant	Fl	Py	BaA	Chr	3-Chr	BbF	BkF	BaP	Pery	IcdP	DbahA	BghiP	
1	1.0	-	-	-	-	-	2.0	0.9	1.2	-	0.4	-	1.6	1.4	0.6	-	-	1.9	-	-	1.4	-	-	-	12
2	1.3	-	-	-	-	-	2.2	1.2	2.5	-	0.8	-	2.6	2.7	0.9	1.2	-	2.9	-	-	2.2	-	-	-	21
3	1.2	-	-	-	-	-	2.2	0.8	-	-	0.4	-	1.4	1.8	0.6	-	-	1.9	-	-	1.4	-	-	-	12
4	2.2	-	-	-	-	-	1.9	0.7	3.0	1.6	0.9	-	8.5	9.1	4.2	5.1	0.5	7.5	2.8	4.6	4.8	4.5	-	3.9	66
5	1.6	-	-	-	-	-	1.9	-	1.5	-	0.7	-	1.0	1.0	-	-	-	2.1	-	-	1.4	-	-	-	11
6	1.1	-	-	-	-	-	1.8	0.8	1.8	-	1.6	-	1.4	7.2	0.9	1.7	-	2.7	-	-	2.2	-	-	-	23
7	1.4	-	-	-	-	-	2.0	0.8	2.0	-	0.7	-	3.4	2.6	1.3	1.8	-	4.3	-	-	4.0	2.6	-	2.4	29
8	1.9	-	-	-	-	-	2.0	0.8	2.5	-	0.9	-	4.1	2.3	1.3	2.0	-	4.5	-	-	4.4	2.6	-	2.3	32
9	3.1	-	1.2	-	-	-	2.6	1.4	5.3	2.7	1.7	0.8	2.0	5.2	3.7	4.9	0.5	10.6	3.1	3.3	8.4	6.7	-	5.6	73
10	1.6	-	-	-	-	-	1.7	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.3
11	1.5	-	-	-	-	-	1.7	-	-	-	0.3	-	1.3	-	-	-	-	-	-	-	1.1	-	-	-	5.9
12	2.0	-	-	-	-	-	2.3	0.7	2.8	-	0.9	-	5.4	3.8	1.7	2.1	-	4.6	1.4	1.7	4.2	2.6	-	2.0	38
13	1.1	-	-	-	-	-	1.8	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	2.9
14	1.2	-	-	-	-	-	1.9	0.7	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.9
15	1.2	-	-	-	-	-	1.7	0.7	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.6
16	1.3	-	-	-	-	-	1.6	0.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	3.8
17	5.6	-	-	-	-	-		5.9	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	41
18	1.3	-	-	-	-	-	2.6	0.9	2.4	1.1	0.9		5.9	3.6	1.5	2.1	-	4.2	-	-	8.1	2.5	-	2.2	39
19	5.5	-	-	-	-	-	4.0	1.7	7.6	2.6	2.1	0.8	17.8	18.1	7.2	8.1	0.7	15.0	5.0	6.1	34.3	9.0	-	7.4	153
20	5.5	-	-	-	-	-	2.2	0.7	1.2	0.5	0.4	-	2.1	2.3	0.8	1.1	-	2.1	-	-	3.7	-	-	-	23
21	3.1	-	-	-	-	-	-	0.6	2.0	0.8	0.8	-	4.1	4.9	1.8	2.3	-	4.9	1.5	1.5	11.7	2.7	-	2.7	45
22	2.4	-	-	-	-	21.8	-	0.7	-	-	0.4	0.9	-	1.5	-	-	-	-	-	-	3.5	-	-	-	31
23	7.0	-	-	-	-	-	3.7	1.7	3.7	1.7	1.6	-	8.6	8.4	3.4	3.9	0.5	9.9	2.6	3.0	49.9	5.2	-	4.4	119
24	5.0	-	-	-	-	-	2.0	-	-	0.4	0.3	-	1.9	2.3	1.0	1.3	-	2.6	-	-	1.1	-	-	-	18
25	3.7	-	-	-	-	-	2.8	1.2	2.3	1.2	1.0	-	4.5	5.0	2.1	2.7	-	5.7	1.6	1.9	12.2	3.2	-	2.9	54
26	4.2	-	-	-	-	-	2.6	0.8	1.9	0.9	0.7	-	3.0	3.6	1.5	1.9	-	4.0	-	-	9.7	2.6	-	2.3	40
27	5.0	-	-	-	-	-	2.7	0.8	2.8	0.9	0.9	-	5.4	5.3	2.4	3.6	-	7.5	2.2	2.0	5.1	4.3	-	4.3	55
28	1.5	-	-	-	-	-	2.0	0.9	-	-	-	-	1.0	-	-	-	-	-	-	-	1.0	-	-	-	6.4
29	1.3	-	-	-	-	-	1.9	0.7	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	4
30	2.4	-	-	-	-	-	2.9	1.3	3.4	2.2	1.1	-	6.3	6.0	2.7	3.7	-	8.4	2.5	2.5	8.8	5.0	-	4.1	63

Abbreviations: Na: Naphthalene; 2-Na: 2-Methylnaphthalene; 1-Na: 1-Methylnaphthalene; 1,3-Na: 1,3-Dimethylnaphthalene; Acl: acenaphthylene; Ace: acenaphthene; Flu: fluorene; 1-Flu: 1-Methylfluorene; Phe: phenanthrene; 3-Phe: 3-Methylphenanthrene; 2-Phe: 2-Methylphenanthrene; Ant: anthracene; Fl: fluoranthene; Py: pyrene; BaA: benzo[a]anthracene; Chr: chrysene; 3-Chr: 3-Methylchrysene; BbF: benzo[b]fluoranthene; BkF: benzo[k]fluoranthene; BaP: benzo[a]pyrene; Pery: Perylene; IcdP: indeno[1,2,3-cd]pyrene; DbahA: dibenz[a,h]anthracene; BghiP: benzo[g,h,i]perylene.
-: below detection limits.

Table S4. (Continued).

Sites	Polycyclic aromatic hydrocarbons (ng g^{-1} dry mass)																								ΣPAHs
	Na	2-Na	1-Na	1,3-Na	Acl	Ace	Flu	1-Flu	Phe	3-Phe	2-Phe	Ant	Fl	Py	BaA	Chr	3-Chr	BbF	BkF	BaP	Pery	IcdP	DbahA	BghiP	
31	5.7	1.0	-	-	-	-	-	1.0	6.1	1.3	2.1	5.5	6.1	5.9	3.5	7.9	1.8	13.6	2.2	3.3	17.5	7.4	-	5.4	98
32	4.2	-	-	-	-	-	-	-	2.1	-	0.9	2.8	1.8	1.6	1.0	2.2	0.5	3.9	-	-	3.6	2.4	-	2.1	29
33	2.9	-	-	-	-	-	-	-	-	-	0.3	0.8	-	-	-	-	-	-	-	-	-	-	-	-	4
34	2.9	1.0	-	-	-	-	1.7	1.8	7.1	1.5	3.4	7.2	9.7	16.1	4.2	9.4	2.2	16.5	2.6	3.9	11.4	7.8	-	6.3	117
35	2.7	-	-	-	-	-	-	1.0	2.2	0.5	0.8	2.1	1.6	2.7	0.9	2.2	0.5	4.1	-	-	2.7	2.4	-	2.1	29
36	2.0	-	-	-	-	8.7	-	0.8	2.1	0.6	0.5	1.4	2.2	2.1	1.3	2.5	0.8	5.9	-	1.4	5.1	3.4	-	3.0	44
37	2.9	-	-	-	-	-	-	0.7	1.6	-	0.6	1.5	1.2	2.0	-	1.4	-	2.4	-	-	1.7	-	-	-	16
38	4.0	1.5	1.5	2.0	-	-	2.8	5.7	10.5	2.5	3.2	6.9	10.6	11.7	5.6	13.1	3.0	24.2	3.3	5.1	15.3	14.6	-	11.1	158
39	3.6	-	-	-	-	-	-	1.1	2.2	-	1.4	3.4	1.0	5.1	-	-	-	1.8	-	-	7.5	-	-	-	27
40	2.4	-	-	-	-	-	-	1.1	4.1	1.2	1.0	2.2	4.5	4.0	2.5	5.6	1.3	9.8	-	2.4	7.9	5.5	-	5.1	61
41	5.1	1.3	1.5	1.6	-	1.0	2.0	14.9	10.4	1.1	6.3	13.6	5.3	17.2	2.5	5.2	1.4	8.3	1.1	2.6	7.1	4.4	-	3.9	118
42	3.0	0.8	0.8	1.0	-	-	1.5	1.0	6.7	1.4	1.6	3.2	7.6	8.2	4.4	7.6	2.0	14.1	1.8	4.6	10.9	7.4	-	6.2	96
43	3.7	1.3	1.3	1.6	-	-	1.8	1.3	8.2	1.9	-	-	17.4	9.2	4.0	9.3	2.0	15.0	2.1	3.6	10.5	8.5	-	7.4	110
44	2.8	-	-	-	-	-	-	-	1.6	-	-	1.0	1.3	1.3	-	1.6	-	2.7	-	0.8	2.4	-	-	-	16
45	4.2	-	-	-	-	-	-	1.9	1.8	-	-	-	1.4	4.5	-	0.7	-	0.9	-	-	-	-	-	-	15
46	2.5	-	-	-	-	-	-	-	2.5	-	-	1.5	2.5	2.6	1.3	2.8	-	5.0	0.8	1.3	3.8	2.7	-	2.1	31
47	3.2	1.2	1.2	1.8	-	-	1.6	1.2	7.7	1.5	2.2	9.6	7.0	7.2	2.7	5.6	1.5	8.1	3.6	9.0	1.2	3.7	-	3.8	85
48	2.6	-	-	-	-	-	-	5.7	2.6	-	1.8	4.4	1.2	5.0	-	0.7	-	1.1	-	-	-	-	-	-	25
49	2.7	-	-	-	-	-	-	0.9	1.2	-	-	1.0	0.9	1.1	-	1.0	-	1.6	-	-	2.6	-	-	-	13
50	3.6	-	-	-	-	-	-	-	0.8	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	4.4
51	4.5	-	-	-	-	-	-	-	1.3	-	0.9	2.4	0.9	1.1	-	1.1	-	1.7	-	-	2.3	-	-	-	16
52	4.1	-	-	-	-	-	-	1.3	1.5	-	1.5	3.3	0.8	4.4	-	-	-	-	-	-	-	-	-	-	17
53	4.9	-	-	-	-	-	-	0.8	1.1	-	1.2	2.2		1.6	-	0.9	-	0.9	-	-	0.9	-	-	-	15
54	5.2	-	-	-	-	-	-	1.0	1.9	-	1.2	3.0	1.3	2.0	-	1.5	-	2.4	-	-	2.0	-	-	-	22
55	4.9	-	-	-	-	-	0.8	0.8	2.3	-	1.2	2.7	1.9	2.0	1.0	2.2	-	4.2	0.8	1.2	4.0	2.4	-	2.0	34
56	3.5	-	-	-	-	-	-	-	1.0	-	-	-	-	-	-	0.8	-	1.3	-	-	1.2	-	-	-	7.8
57	5.7	-	-	-	-	-	1.0	12.1	4.4	-	5.7	11.8	1.9	10.3	-	-	-	0.7	-	-	-	-	-	-	54
58	4.1	-	-	-	-	-	-	-	0.9	-	-	-	-	-	-	-	-	0.8	-	-	0.8	-	-	-	6.6

Table S5. Concentrations of APs in sediments of the Geum River Estuary and Saemangeum Coast.

Sites	Alkylphenols (ng g^{-1} dry mass)						
	4-tert-octylphenol	4-tert-octylphenol monoethoxylate	4-tert-octylphenol diethoxylate	Nonylphenol	Nonylphenol monoethoxylate	Nonylphenol diethoxylate	**ΣAPEOs**
1	0.12	0.19	0.14	3.91	1.07	-	5.4
2	0.13	0.17	0.14	3.75	1.08	-	5.3
3	0.15	0.19	0.16	-	1.00	-	1.5
4	0.09	0.22	0.15	-	1.15	-	1.6
5	0.11	0.25	0.16	-	1.84	-	2.4
6	0.10	0.21	0.14	5.29	1.05	-	6.8
7	0.16	0.23	0.20	-	1.36	-	1.9
8	0.11	0.24	0.15	3.89	1.67	-	6.1
9	0.17	0.35	0.47	6.25	3.72	4.08	15
10	-[a]	0.10	-	-	0.48	-	0.6
11	0.12	0.24	0.13	-	1.29	-	1.8
12	0.17	0.51	0.32	6.30	3.01	1.99	12
13	0.11	0.37	0.21	3.91	1.64	-	6.2
14	0.16	0.46	0.23	-	1.91	-	2.8
15	0.14	0.48	0.30	6.02	2.07	-	9.0
16	0.14	0.43	0.29	-	1.77	-	2.6
17	0.89	0.18	3.27	2.77	1.86	0.87	9.8
18	-	0.30	0.41	5.96	2.14	1.86	11
19	-	0.25	-	-	2.79	4.42	7.5
20	-	0.18	0.13	-	1.68	2.37	4.4
21	-	0.42	0.65	4.84	4.76	7.65	18
22	0.53	0.39	0.66	1.71	0.59	0.96	4.8
23	0.15	0.34	0.45	19.76	4.48	6.26	31
24	-	0.18	0.12	7.72	1.55	1.86	11
25	-	0.34	0.22	6.17	3.41	4.99	15
26	0.08	0.39	0.32	4.96	4.26	3.43	13
27	-	0.24	0.36	-	3.05	2.92	6.6
28	0.15	0.64	0.32	4.09	3.04	1.90	10
29	-	-	-	1.1	-	-	1.1
30	0.22	0.55	0.44	6.53	4.60	2.77	15

-: below detection limits.

Table S5. (Continued).

Sites	Alkylphenols (ng g^{-1} dry mass)						
	4-tert-octylphenol	4-tert-octylphenol monoethoxylate	4-tert-octylphenol diethoxylate	Nonylphenol	Nonylphenol monoethoxylate	Nonylphenol diethoxylate	**ΣAPEOs**
31	0.24	0.93	1.78	15.38	19.72	7.94	46
32	0.63	0.62	3.13	3.95	1.62	1.49	11
33	0.21	0.57	0.35	3.67	1.19	0.92	6.9
34	0.15	0.89	0.41	2.67	1.89	2.56	8.6
35	0.39	0.18	0.24	1.86	2.55	2.78	8.0
36	0.33	0.59	0.27	4.55	2.18	0.86	8.8
37	0.48	0.98	0.12	2.17	1.18	1.43	6.4
38	0.12	0.35	0.20	3.60	0.94	2.23	7.4
39	0.27	1.00	7.32	3.88	1.45	5.40	19
40	0.30	0.99	0.65	5.96	2.77	3.37	14
41	0.82	0.36	0.67	7.13	6.45	2.16	18
42	0.73	0.25	0.23	4.36	1.66	1.69	8.9
43	0.19	0.26	0.56	6.28	2.48	2.25	12
44	0.39	0.22	0.26	8.65	1.54	1.83	13
45	0.13	0.19	0.23	5.12	4.38	0.95	11
46	0.29	0.15	0.33	3.17	1.72	1.25	6.9
47	0.93	0.26	-	2.87	3.96	2.95	11
48	0.32	0.29	1.28	1.99	1.14	0.62	5.6
49	0.34	0.22	0.17	3.32	2.16	1.37	7.6
50	0.33	0.18	0.31	3.17	1.51	0.85	6.4
51	0.12	0.13	0.25	4.89	1.98	1.35	8.7
52	0.92	0.17	0.19	4.85	1.63	2.74	11
53	0.11	0.15	0.16	4.82	1.37	0.88	7.5
54	0.34	0.12	0.23	4.83	3.28	1.45	10
56	0.45	0.12	0.55	5.17	2.78	1.73	11
57	0.11	0.31	0.45	2.85	0.12	0.15	4.0
58	0.48	0.13	2.50	6.35	2.96	1.00	13

Table S6. Concentrations of SOs in sediments of the Geum River Estuary and Saemangeum Coast.

Sites	**Styrene oligomers (ng g^{-1} dry mass)**										**ΣSOs**
	SD1	SD2	SD3	SD4	ST1	ST2	ST3	ST4	ST5	ST6	
1	-	-	-	0.45	0.95	1.15	1.72	-	0.72	-	4.99
2	-	-	-	0.45	0.71	0.94	1.41	-	0.58	-	4.10
3	-	-	-	0.40	-	-	0.43	-	-	-	0.84
4	-	-	-	0.49	0.80	1.02	1.33	-	0.53	-	4.18
5	-	-	-	0.29	-	-	0.52	-	-	-	0.80
6	-	-	-	0.37	-	-	0.67	-	-	-	1.04
7	-	-	-	0.40	-	-	0.55	-	-	-	0.94
8	-	-	-	0.39	0.83	1.19	1.88	-	0.67	-	4.95
9	-	-	-	0.46	0.91	0.79	1.54	-	0.63	-	4.32
10	-	-	-	0.29	-	0.61	0.78	-	-	-	1.68
11	-	-	-	-	-	-	0.33	-	-	-	0.33
12	-	-	-	-	-	-	0.78	-	-	-	0.78
13	-	-	-	0.36	0.75	0.88	1.43	-	0.54	-	3.95
14	-	-	-	0.29	-	-	0.68	-	-	-	0.97
15	-	-	-	0.34	0.68	0.88	1.54	-	0.45	-	3.90
16	-	-	-	-	-	-	0.52	-	-	-	0.52
17	-	-	1.78	-	-	-	0.50	-	-	-	2.28
18	-	-	1.51	0.76	0.58	0.77	1.12	-	-	-	4.74
19	-	-	-	0.54	2.05	2.50	3.94	1.27	1.17	-	11.47
20	-	-	1.25	1.27	6.40	8.41	13.11	4.73	5.09	-	40.27
21	-	-	1.14	0.80	2.97	3.31	5.13	1.52	1.82	-	16.68
22	-	-	-	-	-	0.71	0.50	-	-	-	1.21
23	-	-	1.18	0.54	1.36	1.28	1.35	-	0.57	-	6.28
24	-	-	-	0.58	1.73	2.34	3.71	1.33	1.28	-	10.98
25	-	-	0.98	0.54	1.89	1.81	2.53	0.84	0.89	-	9.46
26	-	-	1.07	0.88	4.36	5.40	8.45	2.92	3.15	-	26.24
27	-	-	-	0.56	2.02	2.52	3.96	1.31	1.36	-	11.73
28	-	-	-	0.71	3.32	4.36	7.05	2.40	2.57	-	20.41
29	-	-	-	0.48	1.50	2.11	3.36	1.03	0.98	-	9.46
30	-	-	-	0.77	1.14	1.45	2.17	-	0.79	-	6.32

-: below detection limits.

Table S6. (Continued).

Sites	Styrene oligomers (ng g^{-1} dry mass)										ΣSOs
	SD1	SD2	SD3	SD4	ST1	ST2	ST3	ST4	ST5	ST6	
31	-	-	-	-	-	-	0.45	-	-	-	0.45
32	-	-	-	-	-	-	0.43	-	-	-	0.43
33	-	-	-	-	-	-	0.41	-	-	-	0.41
34	-	-	1.28	-	1.32	0.86	1.23	0.54	0.68	-	5.91
35	-	-	-	-	1.11	0.80	1.10	0.94	-	-	3.95
36	-	-	1.95	-	3.14	1.81	2.39	0.67	0.70	-	10.65
37	-	-	2.23	1.05	6.62	4.68	5.68	2.32	2.23	-	24.81
38	-	0.94	256.81	-	-	2.14	0.78	-	0.62	-	261.30
39	-	-	1.20	-	1.89	1.23	1.46	0.64	0.59	-	7.00
40	-	-	1.24	-	1.46	1.52	1.25	-	0.40	-	5.86
41	-	-	5.66	0.76	6.66	3.21	6.25	2.74	1.98	-	27.27
42	-	-	-	-	-	-	0.87	-	-	-	0.87
43	-	-	1.31	-	1.11	-	0.83	-	-	-	3.25
44	-	-	16.35	2.82	41.04	19.61	29.19	12.61	12.05	-	133.67
45	-	-	6.51	0.35	4.87	2.52	3.78	1.62	1.36	-	21.02
46	-	-	1.18	0.28	2.23	1.75	3.25	1.31	0.96	-	10.96
47	-	-	3.89	0.69	10.78	5.51	7.95	3.62	2.97	-	35.41
48	-	-	3.45	0.50	5.96	3.15	4.66	1.73	1.74	-	21.18
49	-	-	1.09	-	1.25	0.73	1.36	-	0.40	-	4.83
50	-	-	1.71	0.29	2.58	1.70	2.67	1.17	0.90	-	11.02
51	-	-	1.37	-	-	-	0.69	-	-	-	2.06
52	-	-	1.41	-	-	-	0.61	-	-	-	2.02
53	-	-	1.62	-	0.66	-	0.97	-	-	-	3.25
54	-	-	1.87	-	0.90	-	1.86	0.80	-	-	5.43
56	-	-	1.15	-	-	-	1.12	0.53	-	-	2.80
57	-	-	-	-	-	-	0.55	-	-	-	0.55
58	-	-	3.43	-	0.76	-	0.93	-	0.33	-	5.46

Table S7. Correlation between environmental parameters and the macrofaunal community.

Environmental parameters	Species number (number)		Density (individual/m^2)		Biomass (g wet weight/m^2)	
	r^2	*p*-Value	r^2	*p*-Value	r^2	*p*-Value
PAHs (ng g^{-1})	0.01	0.41	0.03	0.24	0.05	0.09
APs (ng g^{-1})	0.01	0.49	0.01	0.69	0.01	0.41
SOs (ng g^{-1})	0.01	0.64	0.01	0.40	0.01	0.90
TOC (%)	0.06	0.07	0.01	0.57	0.01	0.42
TN (%)	0.03	0.21	0.03	0.20	0.03	0.23
C/N	0.06	0.08	0.05	0.09	0.06	0.07
$\delta^{13}C$ (‰)	0.52	<0.01**	0.05	0.11	0.10	0.01*
Mud content (%)	0.01	0.73	0.06	0.07	0.01	0.46

* $p < 0.05$, ** $p < 0.01$

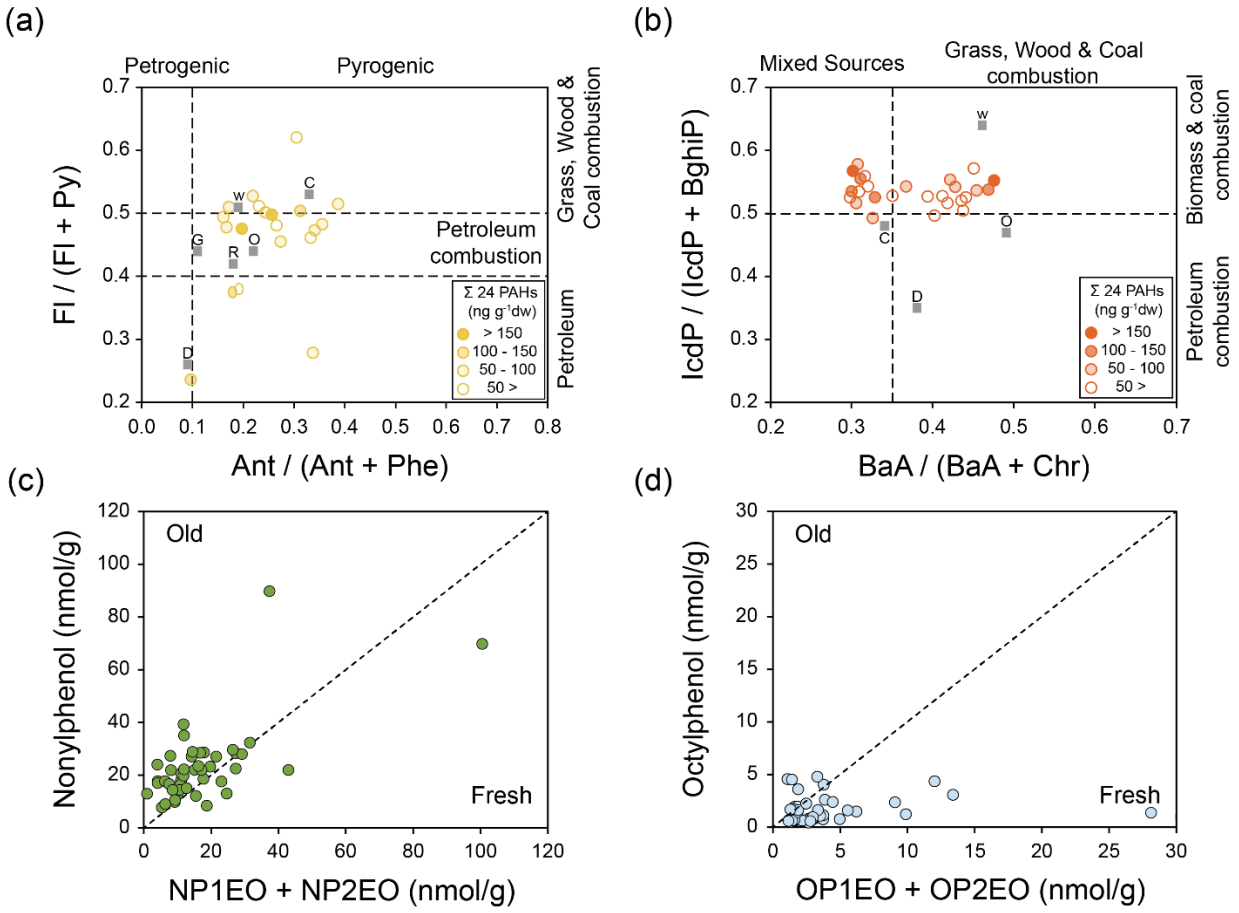

Fig. S1. Diagnostic ratios for prediction of sources of chemicals between (a) Ant/(Ant+Phe) and Fl/(Fl+Py), (b) BaA/(BaA+Chr) and IcdP/(IcdP+BghiP), (c) NP1EO+NP2EO and NP, and (d) OP1EO+OP2EO and OP in sediments of the Geum River Estuary and Saemangeum Coast.

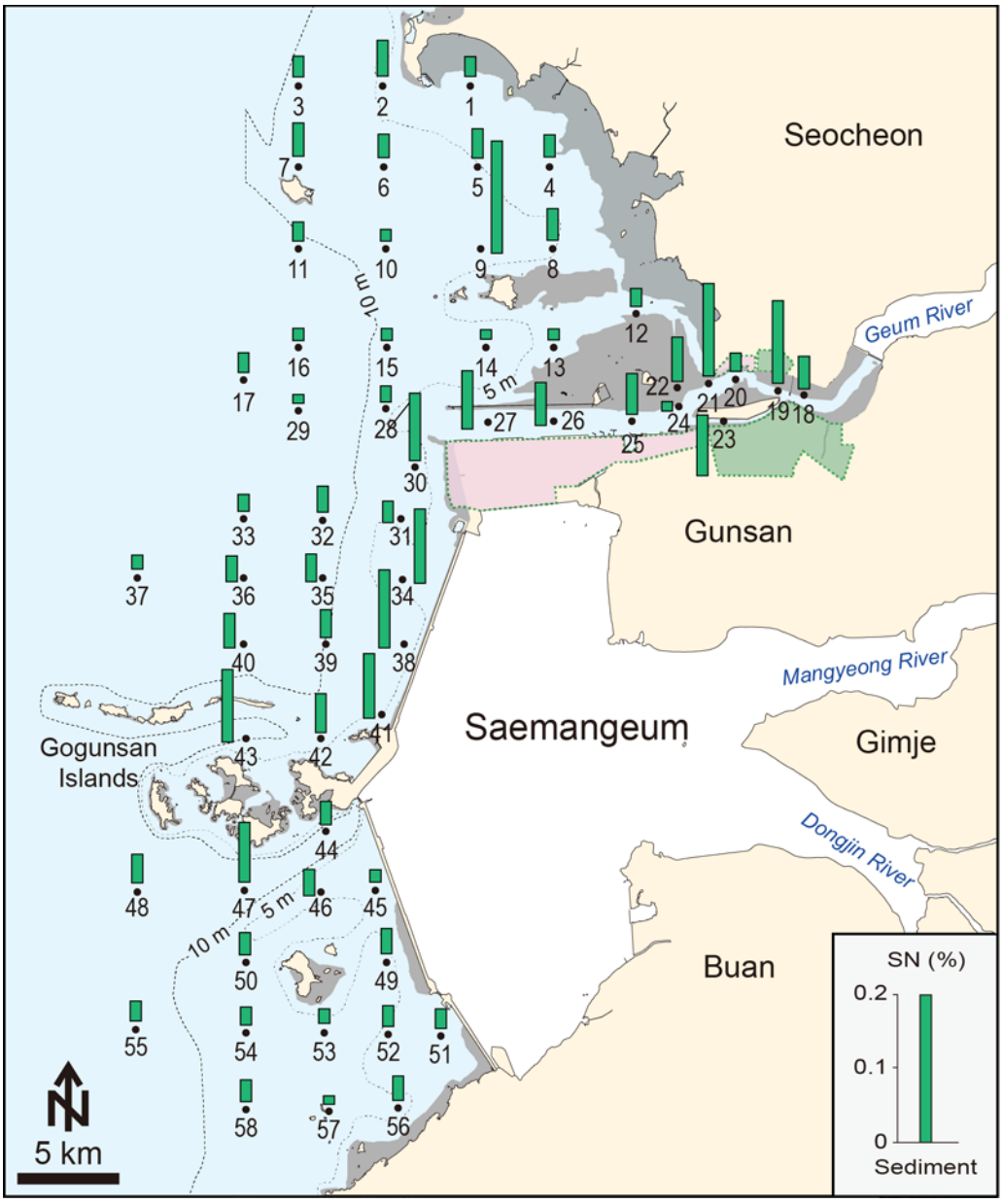

Fig. S2. Spatial distributions of total nitrogen contents in sediments of the Geum River Estuary and Saemangeum Coast.

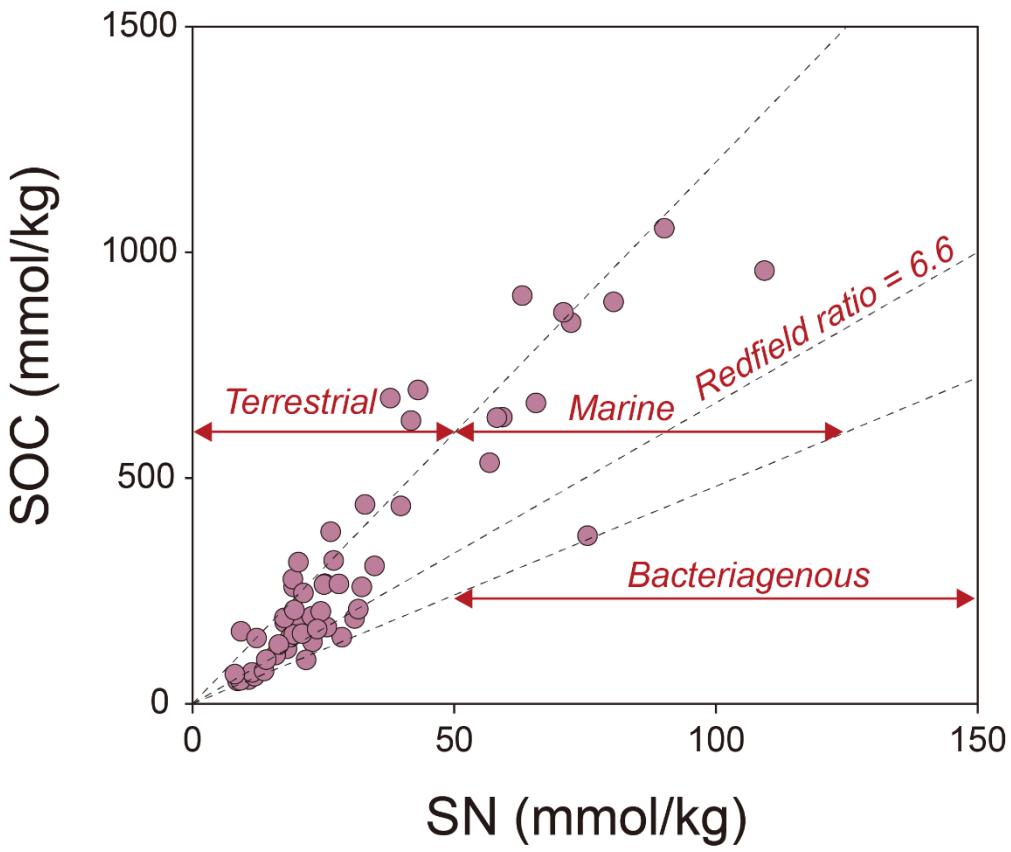

Fig. S3. Scatter plots of sediment organic carbon and nitrogen contents for prediction of sources of organic matter in sediments of the Geum River Estuary and the Saemangeum Coast.

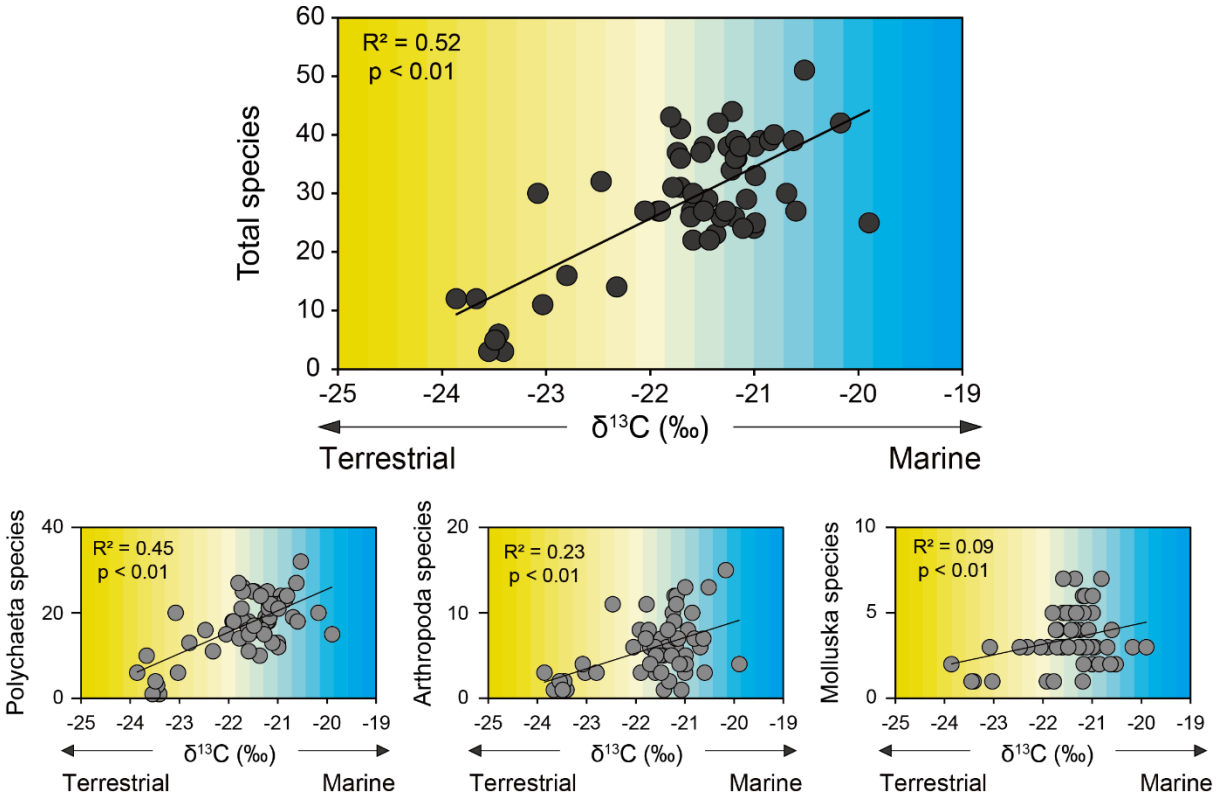

Fig. S4. Correlations between macrofaunal community and $\delta^{13}C$ values in sediments of the Geum River Estuary and Saemangeum Coast.