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Assessment of persistent toxic substances in sediments of Gyeonggi Bay, Korea: Distributions, sources, and potential ecological risks☆

Hyunseo Song [a,1], Taewoo Kim [a,1], Junghyun Lee [b], Seo Joon Yoon [a], Beomgi Kim [a], Youngnam Kim [c], Seongjin Hong [c], Jong Seong Khim [a,d,e,*]

[a] School of Earth and Environmental Sciences, Seoul National University, Seoul 08826, Republic of Korea
[b] Department of Environmental Education, Kongju National University, Gongju 32588, Republic of Korea
[c] Department of Marine Environmental Sciences, Chungnam National University, Daejeon 34134, Republic of Korea
[d] Research Institute of Oceanography, Seoul National University, Seoul 08826, Republic of Korea
[e] Center for Convergence Coastal Research, Seoul National University, Siheung-si, Gyeonggi-do 15011, Republic of Korea

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ABSTRACT

Persistent toxic substances (PTSs) from anthropogenic activities are a growing concern for marine ecosystems. In addition, the specific sources and ecological consequences of PTSs, particularly in coastal regions influenced by industrial and urban developments, remain insufficiently understood. This study evaluated the distribution, sources, and risks of 54 PTSs in Gyeonggi Bay. Polycyclic aromatic hydrocarbons (PAHs) ranged from 22.0 ng g^{-1} dw to 2710 ng g^{-1} dw, and alkylphenols (APs) peaked at 21,500 ng g^{-1} dw in source-dominated areas. Elevated levels were observed in Incheon Port and Lake Sihwa, from industrial and urban wastewater discharges. PMF modeling identified fossil fuel combustion as the main source of PAHs and natural and agriculture for metal(loid)s. Ecological risk assessments revealed significant contributions of metal(loid)s (49.1 %) and APs (39.3 %), with nonylphenols and arsenic posing the highest risks. These findings highlight the need for continuous monitoring and stricter regulations to mitigate the impacts of PTSs in marine ecosystems.

1. Introduction

Gyeonggi Bay, a representative semicircular bay with large and small islands on the west coast of Korea, is an area with low water level and large tidal range, and the tidal flats are well developed. With its high biological productivity, Gyeonggi Bay serves as an essential spawning and nursery ground for marine organisms, contributing to the sustainability of marine populations in the region. Incheon, one of Korea's major port cities bordering the bay, has experienced rapid industrialization and urbanization, exacerbating pollution levels. Similarly, Lake Sihwa, originally constructed to supply freshwater for agricultural and industrial purposes, has suffered from deteriorated water quality due to inadequate wastewater treatment and pollutant discharges from nearby industrial and urban areas (Lee et al., 2017). To address these issues, the Korean government designated both Incheon and Lake Sihwa as specially managed sea areas in 2000, followed by the construction of a tidal power plant and water gates to improve seawater circulation. Despite these efforts, high concentrations of persistent toxic substances (PTSs) remain prevalent in the region (Khim and Hong, 2014; Hong et al., 2016; Hong et al., 2019). Most studies have predominantly focused on key areas such as Lake Sihwa and have often centered on general contamination patterns or individual pollutants. While Gyeonggi Bay holds significant ecological importance, comprehensive studies that examine multiple pollutant types, their sources, and ecological risks across a broader spatial scale remain lacking.

PTSs, which mainly come from anthropogenic activities such as industrial discharges, urban wastewater, and agriculture, accumulate in marine sediments through riverine and wastewater inputs (Kim et al., 2021; Yoon et al., 2022). Polycyclic aromatic hydrocarbons (PAHs), alkylated PAHs (A-PAHs), styrene oligomers (SOs), alkylphenols (APs), and metal(loid)s exhibit hydrophobicity, high affinity for sediment particles, and long-distance transport and thus persist in marine

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* Corresponding author at: School of Earth and Environmental Sciences & Research Institute of Oceanography, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 151-742, Republic of Korea.

E-mail address: jskocean@snu.ac.kr (J.S. Khim).

[1] These authors contributed equally to this work.

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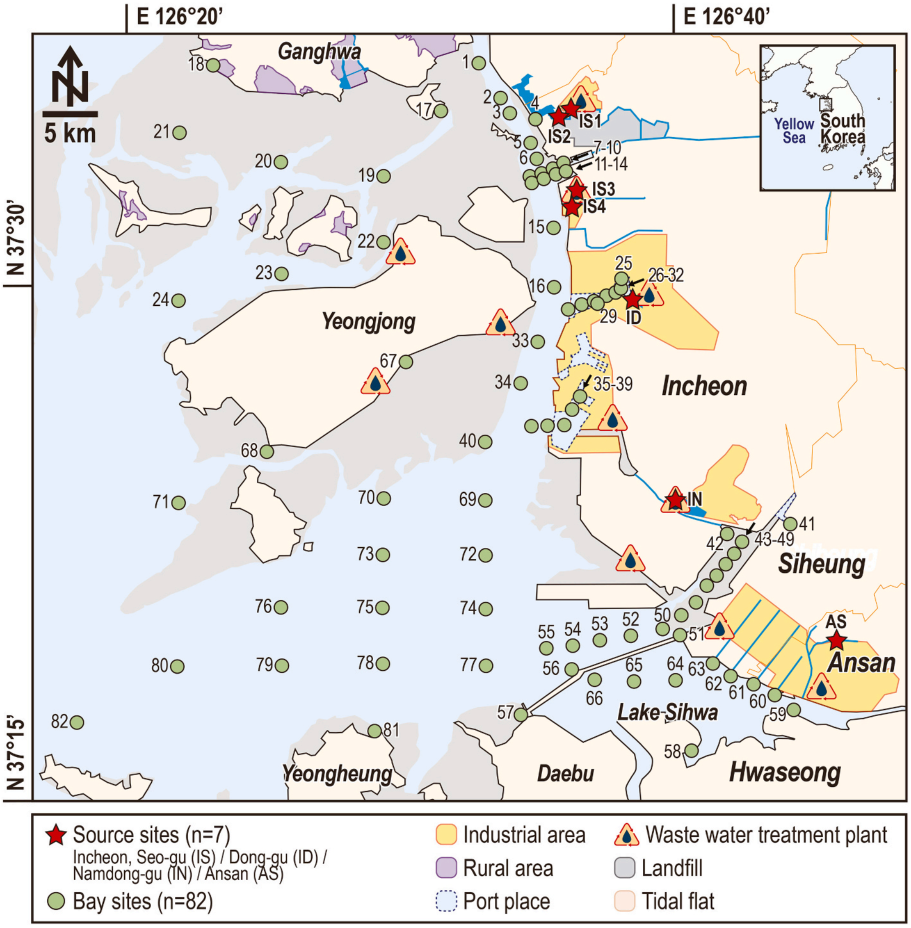


Fig. 1. Map showing the sampling sites of surface sediments in Gyeonggi Bay, Korea.

sediments (Dinç et al., 2021; Kwon and Moon, 2019; Montuori et al., 2016). These PTSs cause bioaccumulation and biomagnification, which in turn lead to reproductive, endocrine, and habitat disruptions among marine organisms (Christophoridis et al., 2009; Çelebi et al., 2024; Güzel and Canlı, 2023; Hong et al., 2012; Lee et al., 2018).

Among these PTSs, PAHs are of particular concern due to their carcinogenic and immune-disruptive properties. These pollutants arise from both natural sources, like wildfires, and anthropogenic activities, including fossil fuel combustion and vehicle emissions (Srogi, 2007; Ravindra et al., 2008; Mojiri et al., 2019; Patel et al., 2020). A-PAHs, typically found in oil-contaminated regions, serve as indicators of hydrocarbon pollution and exhibit significant toxicity (Mu et al., 2014; Turcotte et al., 2011). SOs derived from the high-temperature decomposition of polystyrene, are associated with reproductive and genetic toxicity risks, while APs, such as nonylphenols and octylphenols, disrupt endocrine functions and are commonly linked to urban wastewater (Amamiya et al., 2019; Chen and Yen, 2013). Metal(loid)s, though naturally occurring in the Earth's crust, accumulate in marine organisms and contribute to long-term toxicity (Khan et al., 2013; Kotze et al., 1999).

Despite global efforts to reduce PTS concentrations, levels exceeding ecological safety thresholds remain a critical issue (Khim and Hong, 2014; Liu et al., 2011; Lorgeoux et al., 2016; Yoon et al., 2023). As a result, regular monitoring and ecological risk assessments of PTSs in marine sediments are crucial for the sustainable management of coastal ecosystems. Ecological risk assessments provide valuable insights into how complex mixtures of pollutants impact marine organisms over time, informing environmental policy and management strategies (Shea and Thorsen, 2012). However, contaminants often exist in complex mixtures, and assessments across organisms are challenging, making it difficult to identify key toxic contaminants. To address these challenges, methods such as the positive matrix factorization (PMF) model have been widely adopted to monitor and trace sources of contamination. The PMF model enables effective source apportionment by analyzing the concentrations and compositions of pollutants, such as PAHs and metal(loid)s, without requiring direct source profiles (Li et al., 2019; Yu et al., 2015). Risk quotient (RQ) calculations further facilitate ecological risk assessments by evaluating the relative risks posed by individual PTSs to marine organisms. Integrating advanced monitoring techniques with source identification and ecological risk analyses strengthens our understanding of pollution dynamics, thereby supporting effective conservation strategies for Gyeonggi Bay.

This study aimed to investigate the distributions, sources, and ecological risks of PAHs, A-PAHs, SOs, APs, and metal(loid)s in Gyeonggi Bay sediments. Specifically, the objectives were to: i) quantify the concentrations of PTSs in sediments, ii) identify potential contamination sources using the PMF model, and iii) evaluate the ecological risks of sedimentary PTSs to marine organisms through risk quotient analysis. The findings of this study will provide critical insights into the contamination profiles of Gyeonggi Bay, highlighting the interplay between human activities and ecological risks, and contribute essential baseline data for future environmental monitoring and management strategies.

2. Materials and methods

2.1. Study area, sampling, and sample preparations

Eighty-nine sediment samples were collected from Incheon (IS1–IS4, ID, IN) and Ansan (AS), covering both source sites ($n = 7$) and bay sites ($n = 82$) (Fig. 1). Sampling was carried out between November 19 and 24, 2018. Surface sediments (0–2 cm) were collected using a Van Veen grab, and samples were immediately transferred into pre-cleaned 125 mL glass jars. These jars were then stored at −20 °C to preserve the samples until laboratory analysis. Mud content was measured by rapid partial analysis using wet sieving (Buchanan, 1984), and grain size was measured using a MasterSizer 3000 (Malvern Panalytical Ltd., UK) following the removal of organic matter using diluted hydrogen peroxide (H_2O_2). Before the analysis, all sediment samples were freeze-dried, sieved with a 2-mm mesh, and thoroughly homogenized through grinding to ensure uniformity for the subsequent chemical analysis.

2.2. Organic carbon, nitrogen, and stable isotopes analyses

The organic carbon (OC) content and stable isotope ratios of carbon ($\delta^{13}C$) were analyzed following the method described by Yoon et al. (2020). Sediment samples were acidified with 1 M hydrochloric acid to remove inorganic carbon, then rinsed three times with deionized water and freeze-dried. The total nitrogen (TN) content and stable isotope ratios of nitrogen ($\delta^{15}N$) were analyzed without acidification. OC, TN, and their stable isotope ratios were measured using an Elemental Analyzer-Isotope Ratio Mass Spectrometer (EA-IRMS) (Elementar, Gmbh, and Hanau). The stable isotopic compositions were expressed in δ notation (Eq. (1)):

$$\delta^{13}\text{C or } \delta^{15}\text{N } (‰) = \left[\left(\frac{R_{\text{sample}}}{R_{\text{reference}}}\right) - 1\right] \times 1000 \tag{1}$$

where R_{sample} and $R_{\text{reference}}$ are the ratio of $^{13}C/^{12}C$ and $^{15}N/^{14}N$ for each sample and reference material, respectively. Vienna Peedee Belemnite (VPDB) and atmospheric N_2 were used as the reference materials for carbon and nitrogen isotopes, respectively. Analytical errors were estimated to be 0.05 ‰ for $\delta^{13}C$ and 0.1 ‰ for $\delta^{15}N$, based on International Atomic Energy Agency (IAEA) working standards (CH-6 for $\delta^{13}C$ and N-1 for $\delta^{15}N$, IAEA, Vienna, Austria).

2.3. Chemical analyses

The concentrations of PAHs, A-PAHs, SOs, APs, metals (Cd, Cr, Cu, Hg, Li, Ni, Pb, and Zn), and metalloid (As) were measured following the methods of previous studies with minor modifications (Hong et al., 2022; Lee et al., 2023; Liu et al., 2020a; Yoon et al., 2021). Standard materials of PAHs were obtained from Chem Service (West Chester, PA), SOs from Wako Pure Chemical Ind. (Osaka, Japan) and Hayashi Pure Chemical Ind. (Osaka, Japan), and APs from Sigma-Aldrich. A-PAH isomers with the same molar mass display highly similar fragmentation patterns. Due to the unavailability of standard materials for individual A-PAHs, the total concentration of each methyl substituent of PAHs (comprising 15 A-PAHs) was semi-quantified using 16 model compounds (Table S1) (Hong et al., 2012; Lee et al., 2023). Detailed information about the target chemicals, instrumental conditions, method detection limits (MDLs), and recovery rates is provided in Tables S1, S2, and S3. The ranges of MDLs were 0.270–1.07 ng g^{-1} dw for PAHs, 0.280–0.940 ng g^{-1} dw for SOs, and 0.0771–3.58 ng g^{-1} dw for APs (Table S1). Recoveries for five surrogate standards (acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, perylene-d_{12}, and bisphenol A-d_{16}), and standard reference material (MESS-4) were generally acceptable: mean = 92.1 % and 96.7 %, respectively (Table S3). The detailed analytical methods for the PTSs are provided in the Methods section of the Supplementary Material.

2.4. Contamination factors and enrichment factor

The pollution load index (PLI) and geoaccumulation index (I_{geo}) are integrated, and single contamination factors are related to the background but are not adjusted for variable sediment size and content. The PLI is the value obtained by integrating the divided concentration of each element, as introduced by Tomlinson et al. (1980). Eq. (2)

$$PLI = \sqrt[n]{\left(\frac{M_1}{B_1}\right) \times \left(\frac{M_2}{B_2}\right) \times \cdots \times \left(\frac{M_n}{B_n}\right)} \tag{2}$$

where M_n is the measured environmental concentration (MEC) of element n, and B_n is the background concentration in Korea coastal sediments (Woo et al., 2019). The MECs of Cu and Zn were corrected by Li for indicator substances (MOF, 2018).

The I_{geo}, introduced by Müller (1969) is calculated by the following Eq. (3):

$$I_{geo} = log_2\left[\frac{M_n}{1.5 \times B_n}\right] \tag{3}$$

The contamination level is classified according to the intensity ranging from <0, 0–1, 1–2, 2–3, 3–4, 4–5, and >5 can be interpreted as ‘practically unpolluted’, ‘unpolluted to moderately polluted’, ‘moderately polluted’, ‘moderately to strongly polluted’, ‘strongly polluted’, ‘strongly to extremely polluted’ and ‘extremely polluted’, respectively.

The enrichment factor (EF) is a method to evaluate anthropogenic activities on elements related to background, adjusted for variable sediment size and contents, and calculated by using conservative elements with Eq. (4):

$$EF = \frac{\left(\frac{M_i}{C_i}\right)_{sample}}{\left(\frac{M_i}{C_i}\right)_{background}} \tag{4}$$

where $(M_i/C_i)_{\text{sample}}$ is the ratio of the measured concentrations of each element to the conservative element, Li, and $(M_i/C_i)_{\text{background}}$ is the ratio of the background concentrations in Korean coastal sediments (Loring, 1990; Woo et al., 2019; Zhou et al., 2019). The enrichment levels are classified as <2, 2–5, 5–20, 20–40, and >40, which can be interpreted as ‘Deficiency to minimal enrichment’, ‘Moderated enrichment’, ‘Significant enrichment’, ‘Very high enrichment’ and ‘Extremely high enrichment’, respectively (Sutherland, 2000).

2.5. Positive matrix factorization receptor model

The EPA PMF model (Ver. 5.0) from the U.S. Environmental Protection Agency (USEPA) was used to estimate the sources of 15 PAHs and eight metal(loid)s. The PMF model is a generic factorization method used to quantify the contribution of source compositions (Larsen and Baker, 2003; Norris et al., 2019). The contributions and compositions of the factors were defined by minimizing the objective function Q (Eq. (5)):

$$Q = \sum_{i=1}^{n}\sum_{j=1}^{m}\left[\frac{X_{ij} - \sum_{k=1}^{p} g_{ik} f_{kj}}{u_{ij}}\right]^2 \tag{5}$$

where x_{ij} *is* the concentration of species j in the sample i, u_{ij} is the uncertainty of x_{ij}, and p is the number of factors. The relative contribution of each factor k is g_{ik}, and f_{kj} is the species profile of each source. Uncertainties (Unc) for each PAH and metal(loid)s were calculated using Eqs. (6) and (7).

$$Unc = \frac{5}{6} \times MDL \tag{6}$$

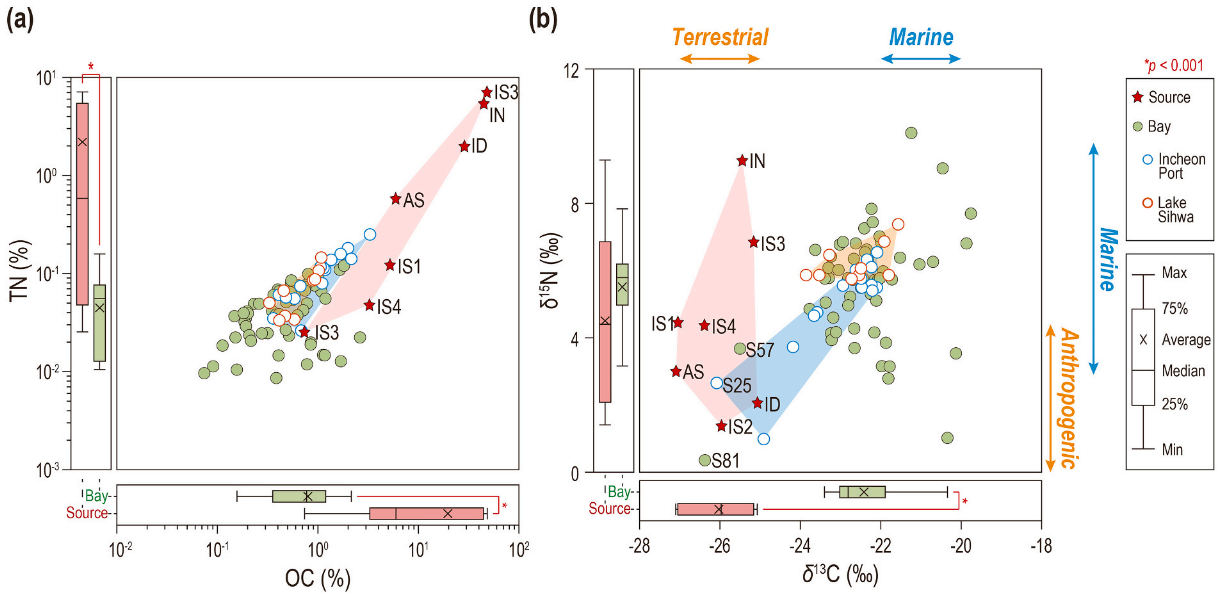


Fig. 2. Scatter plots and box plots for (a) organic carbon content (OC) and total nitrogen content (TN), and (b) δ^{13}C and δ^{15}N values collected from the source and bay sites of Gyeonggi Bay. Organic carbon and nitrogen are expressed as percentages.

$$\text{Unc} = \sqrt{(\textit{Error fraction} \times \textit{concentration})^2 + (0.5 \times \textit{MDL})^2} \tag{7}$$

When the concentration of PTSs or metal(loid)s was less than that of the MDL, Eq. (6) was used, and Eq. (7) was used when the concentration was higher than that of the MDL. The *Error Fraction* was calculated using the standard deviation of compound *j*. PMF modeling was performed following the previously described method (Kim et al., 2021; Lee et al., 2023). The base model was run 100 times for the best solution. The Q values with 2–5 factors were calculated, and the number of factors was determined by the largest decrease in the $Q_{\text{True}}/Q_{\text{Exp}}$ quotient value (Crilley et al., 2017). To test random errors and investigate rotational ambiguity, Displacement (DISP) and Bootstrap (BS) were applied, with the DISP output being 0, indicating no errors.

2.6. Ecological risk assessment

For ecological risk assessment, the risk quotient (RQ) values of PTSs and metal(loid)s in the sediments were calculated. RQ was calculated by dividing the predicted no-effect concentration (PNEC) by MEC (Pintado-Herrera et al., 2017) (Eq. (8)):

$$\text{RQ (risk quotient)} = \frac{\text{MEC}}{\text{PNEC}} \div 1000 \tag{8}$$

To calculate the PNEC, acute and chronic toxicity values for aquatic organisms were required. However, experimental data for all target compounds were limited. Therefore, the PNEC values for three trophic levels were derived using ECOSAR and ECOTOX, with calculations conducted using EnviroTox (Table S4) (Minguez et al., 2016; Kienzler et al., 2019; Sanderson et al., 2003; Lee et al., 2023). The toxicity of the chemicals to aquatic organisms was calculated using Eqs. (9) and (10) to estimate the in the sediments (Hu et al., 2021).

$$PNEC_{sediment} = PNEC_{water} \times \text{K}_d \tag{9}$$

$$K_d = K_{oc} \times f_{oc} \tag{10}$$

where $PNEC_{\text{water}}$ is the toxicity value for aquatic organisms, K_{oc} is the organic carbon-water partition coefficient calculated by Estimation Program Interface (EPI) Suite, and f_{oc} is OC content in each sediment sample. The K_{d} is the sediment-water partition coefficient proposed by Allison and Allsion (2005) was used for the metal(loid)s. The potential ecological risk was determined based on RQ values >1, indicating an elevated level of ecological risk. The total risk quotient ($\sum$RQ) is determined by summing the RQ values for each metal, under the assumption that their toxic effects are additive or approximately additive (Liu et al., 2020b; Selak et al., 2022; Zhang et al., 2024).

2.7. Statistical analysis

SPSS (version 24.0; SPSS Inc., Chicago, IL, USA) was used to verify the relationship between physicochemical properties and PTS concentrations. Principal Component Analysis (PCA) was applied with varimax rotation to enhance interpretability. Additionally, Bartlett's test and the Kaiser-Meyer-Olkin (KMO) test were conducted. The *p*-value of Bartlett's test below 0.001 and the KMO index above 0.7 indicated that the correlation matrix was appropriate for PCA analysis.

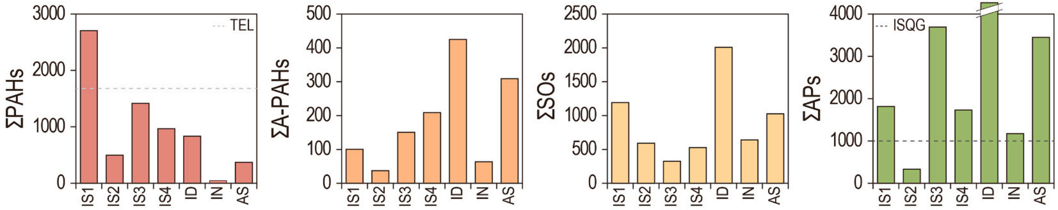
3. Results and discussion

3.1. Variations in organic carbon and nitrogen in Gyeonggi Bay sediments

The OC and TN contents of the sediment samples from Gyeonggi Bay varied significantly by site (Fig. 2a). The average OC and TN contents at the source sites (OC = 19.6 ± 19.2 %; TN = 2.19 ± 2.69 %) were notably higher than those at the bay sites (OC = 0.72 ± 0.58 %; TN = 0.060 ± 0.043 %). A significant positive correlation between OC and TN contents was observed ($p < 0.001$, $r^2 = 0.95$). The highest OC and TN contents were detected at source sites IS3 and IN, which were collected from wastewater treatment sludge (WWTS) in Incheon. Among the bay sites, Incheon Port (S25–S39) exhibited relatively higher OC and TN contents compared to other bay sites. Elevated OC and TN levels in these sediments indicate substantial influences from terrestrial and anthropogenic sources (Gao et al., 2012).

The C/N ratio, which was used to determine the origin of organic matter, ranged from 4.00 to 132 (mean: 17.6) (Table S4). The average C/N ratio was 25.6 ± 21.2 at the source sites and 16.9 ± 22.1 at the bay sites. Higher C/N ratios could reflect anthropogenic influences, including the widespread use of organic chemicals, which alter the C/N ratios of organic matter beyond natural terrestrial ranges. Even when excluding terrestrial sources, anthropogenic activities, such as the extensive application of synthetic organic substances, may contribute to

(a) Concentration of source sites (ng g^{-1} dw)



(b) Concentration of bay sites (ng g^{-1} dw)

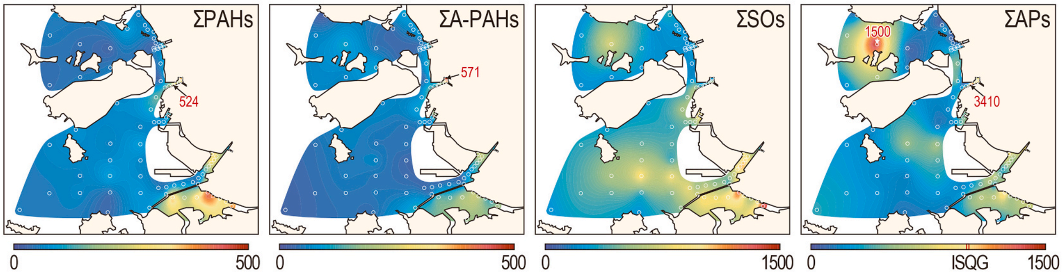


Fig. 3. Total concentrations and distributions of 15 PAHs, 15 A-PAHs, 10 SOs, and 6 APs in (a) source and (b) bay sites from the surface sediments of Gyeonggi Bay.

elevated C/N ratios (Gao et al., 2012).

The δ^{13}C values in sediments ranged from −27.1 to −19.8 ‰ (mean: −22.8 ‰), and δ^{15}N values ranged from 0.360 to 10.1 ‰ (mean: 5.44 ‰) (Fig. 2b). Depleted δ^{13}C values were observed at source sites and several bay sites near land, such as Incheon Port (S25), Daebu Island (S57), and Yeongheung Island (S81). On average, δ^{13}C values at source sites (−26.0 ± 0.787 ‰) were significantly more depleted compared to bay sites (−22.5 ± 1.13 ‰). Previous studies reported δ^{13}C values ranging from −27 to −25 ‰ for terrestrial organic matter and −22 to −20 ‰ to marine organic matter (Peters et al., 1978). Thus, sedimentary organic matter in Gyeonggi Bay primarily originates from terrestrial sources, with its influence diminishing as it approaches the open ocean.

Mean δ^{15}N values at the source sites (4.50 ± 2.57 ‰) were generally more depleted than those at the bay sites, although the difference was not statistically significant. δ15N values ranging from −0.9 to 4.4 ‰ suggest anthropogenic organic matter, while values between 2.9 and 9.8 ‰ indicate marine-origin organic matter (Liénart et al., 2017). Except for the two WWTS at the source sites IS3 and IN, sediment at the source sites showed more depleted δ^{15}N values than that at the bay sites. Overall, the distribution of sedimentary organic matter in Gyeonggi Bay highlights notable inputs of anthropogenic and terrestrial organic matter, particularly concentrated in Incheon Port and Lake Sihwa.

3.2. *Distributions and compositions of PTS in Gyeonggi Bay*

The distribution of PTS in Gyeonggi Bay varied widely across sites (Fig. 3), with significant positive correlations between PTS concentrations, OC content, and mud content ($p < 0.05$) (Fig. S1).

3.2.1. *PAHs*

The total concentration of the 15 PAHs (∑PAHs) ranged from 22.0 to 2710 ng g^{-1} dw (mean: 189 ng g^{-1} dw) (Fig. 3 and Table S5). Source sites exhibited significantly higher concentrations of ∑PAHs (977 ± 817 ng g^{-1} dw) compared to bay sites (122 ± 115 ng g^{-1} dw) (p = 0.001) (Fig. S2). This demonstrates the significant influence of anthropogenic activities, particularly industrial and wastewater discharges, on PAH accumulation. The highest concentration was observed at site IS1 (2710 ng g^{-1} dw), largely attributed to municipal and industrial wastewater discharge from nearby complexes and wastewater treatment plants, which are known to be significant sources of PAHs pollution (Choi et al., 2023; Syafiuddin and Boopathy, 2021). At site IS3, the second highest concentration (1420 ng g^{-1} dw) was linked to WWTS in Incheon, with the highest OC and TN contents in the study area.

Spatially, PAHs concentrations were highest in coastal areas like site S29 (524 ng g^{-1} dw) in Incheon Port (170 ± 125 ng g^{-1} dw), followed by site S63 (449 ng g^{-1} dw) in Lake Sihwa (322 ± 103 ng g^{-1} dw). Incheon Port, an industrial hub, houses crude oil refineries, petroleum product manufacturing, and other industrial activities, which significantly contribute to PAH emissions. (Abdel-Shafy and Mansour, 2016; Gupte et al., 2016; Güzel et al., 2022; Kim et al., 2024; Srogi, 2007). Similarly, the Sihwa industrial complexes, including machinery, electronics, steel, and petrochemical manufacturing, have released various chemicals into inland creeks, deteriorating the Lake Sihwa environment over the years (Lee et al., 2014; Yoo et al., 2006). Overall, PAH concentrations were higher at source sites (except for IN) and coastal areas, decreasing toward the outer sites. These observations suggest that coastal regions with industrial and human activity concentrations are hotspots for PAH pollution. (Dai et al., 2022; Liu et al., 2018; Neumann et al., 2015). The distribution pattern of PAHs aligns closely with that of organic matter, further indicating significant inputs from anthropogenic activities.

The PAHs composition showed dominance of high molecular weight (HMW) PAHs (4–6 rings) at most sites, indicative of combustion-related sources like vehicle emissions and fossil fuel combustion. Heavier PAHs are more likely to be absorbed onto particles, whereas lighter PAHs predominantly exist in the gas phase (Ravindra et al., 2008). This pattern supported that combustion sources contribute significantly to PAHs pollution in Gyeonggi Bay. While PAH concentrations are high at certain sites, such as IS1 (1684 ng g^{-1} dw), which is the only site where the concentration exceeds the threshold effect level (TEL) (Macdonald et al., 1996), particularly near industrial sources, the current levels in Gyeonggi Bay are unlikely to pose immediate risks to the coastal benthic environment. However, the long-term ecological impacts must still be considered. Continued monitoring and risk assessments are crucial to understand the long-term effects of PAH pollution on ecosystem health.

3.2.2. *A-PAHs*

The concentrations of the 15 A-PAHs (∑A-PAHs) ranged from 11.6 to 572 ng g^{-1} dw (mean: 102 ng g^{-1} dw) (Fig. 3 and Table S6). Unlike PAHs, the highest concentration of ∑A-PAHs was detected at site S25, a bay site located in the inner region of Incheon Port rather than a source site. The average of ∑A-PAHs at source sites (185 ± 130 ng g^{-1} dw) was greater than at bay sites (95.1 ± 81.8 ng g^{-1} dw) (Fig. S2). However, within the bay sites, Incheon Port showed a wide distribution ranging from 49.8 to 572 ng g^{-1} dw (mean: 154 ng g^{-1} dw). The second highest

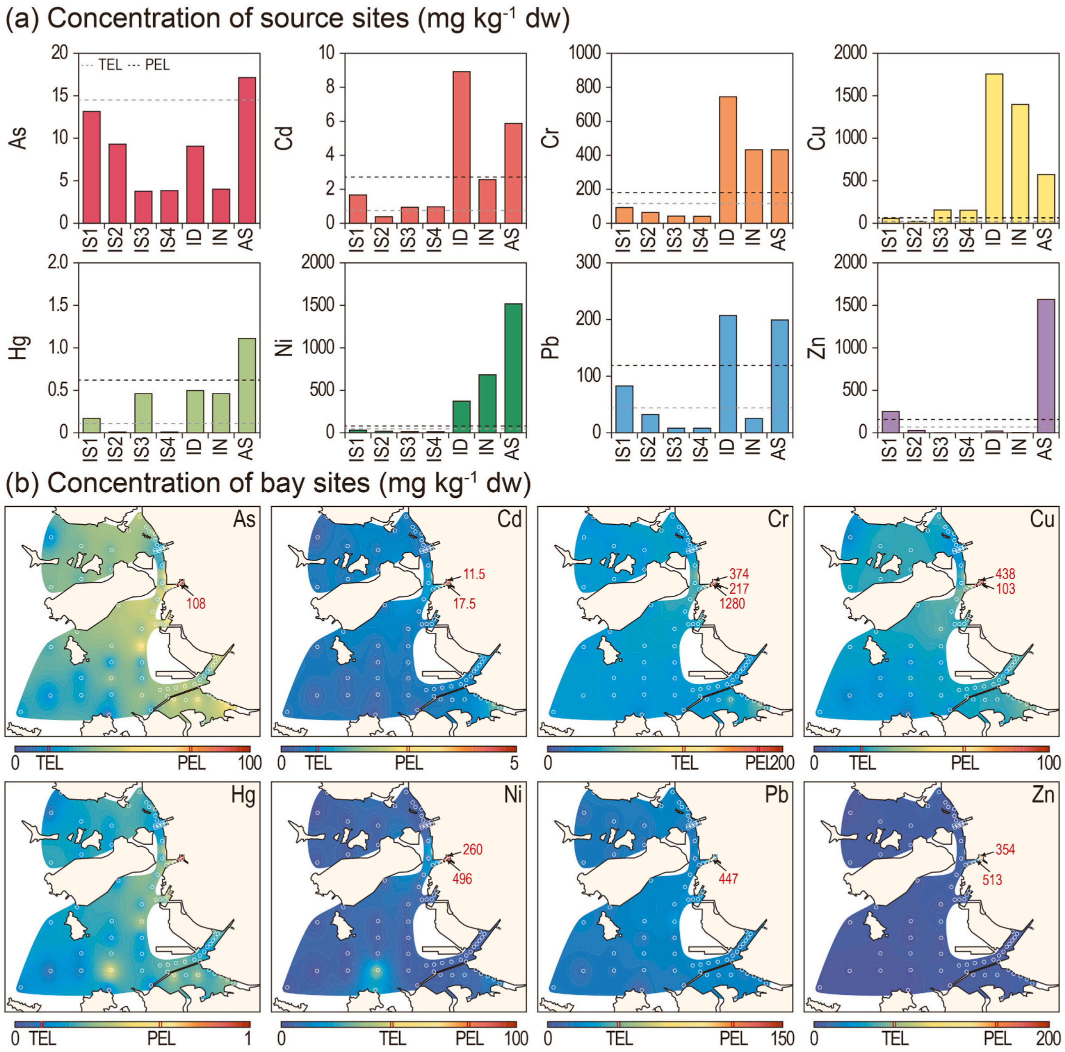


Fig. 4. Total concentrations and distributions of eight metal(loid)s (As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn) in (a) source and (b) bay sites from the surface sediments of Gyeonggi Bay.

concentration was recorded at ID, a source site situated in Incheon Port. Conversely, the lowest concentrations of ∑A-PAHs were found in the outer areas of Gyeonggi Bay (S82 and S18) and the Gyeongin Ara Waterway (S6 and S8), likely due to higher particle adsorption and reduced direct river inputs in these regions. These patterns indicate that ∑A-PAH concentrations in Gyeonggi Bay are primarily influenced by the proximity to industrial sources, with higher concentrations found closer to industrial and wastewater discharge areas like Incheon Port. At the same time, the lower concentrations in the outer regions suggest that decreased industrial influence and the higher particle adsorption capacity of these areas result in lower accumulation of A-PAHs in the sediments.

3.2.3. SOs

The concentrations of 10 SOs (∑SOs) in the sediments ranged 139–2010 ng g^{-1} dw (mean: 542 ng g^{-1} dw). The average ∑SOs concentrations at source sites (901 ± 529 ng g^{-1} dw) were significantly higher than those at the bay sites (511 ± 292 ng g^{-1} dw) ($p = 0.027$) (Fig. 3, S2 and Table S7). The highest concentration was detected at ID, near a plastic manufacturing and petrochemical plant, indicating that industrial activities are key contributors to SOs pollution. At bay sites, elevated ∑SOs in Lake Sihwa and offshore areas likely due to riverine runoff and plastic waste from fishery and aquaculture activities (Amamiya et al., 2019; Kwon et al., 2015; Tian et al., 2020). This suggests that plastic-based materials continue to contribute to SOs inputs in Gyeonggi Bay.

Despite site-specific variations, the relative compositions of styrene oligomers showed consistent patterns, with ST1 (mean 47.8 %) being dominant, followed by SD4 (mean: 16.3 %). The higher abundance of styrene dimers (SDs) at sites with high SOs concentrations suggests recent inputs from plastic-related industries. Compared to SDs, styrene trimers (STs) exhibit lower water solubility, volatility, and slower leaching rates, allowing them to persist longer in sediments (Tian et al., 2020). These findings highlight the ongoing contribution of plastic waste to SOs contamination and emphasize the need for further research on the long-term ecological impacts of these substances in marine environments.

3.2.4. APs

The concentration of six APs (∑APs) ranged from 34.8 to 21,500 ng g^{-1} dw (mean: 757 ng g^{-1} dw) (Fig. 3 and Table S8). Source sites had significantly higher ∑APs (4820 ± 6910 ng g^{-1} dw) compared to bay sites (410 ± 423 ng g^{-1} dw) ($p < 0.001$) (Fig. S2) with the Interim Sediment Quality Guideline (ISQG) for sediment quality exceeded at most source sites (CCME, 1999). The highest concentration was found at site ID, which is located near the Gajwa wastewater treatment plant, highlighting the impact of industrial and municipal wastewater discharges on APs pollution. At bay sites, the distribution patterns of APs

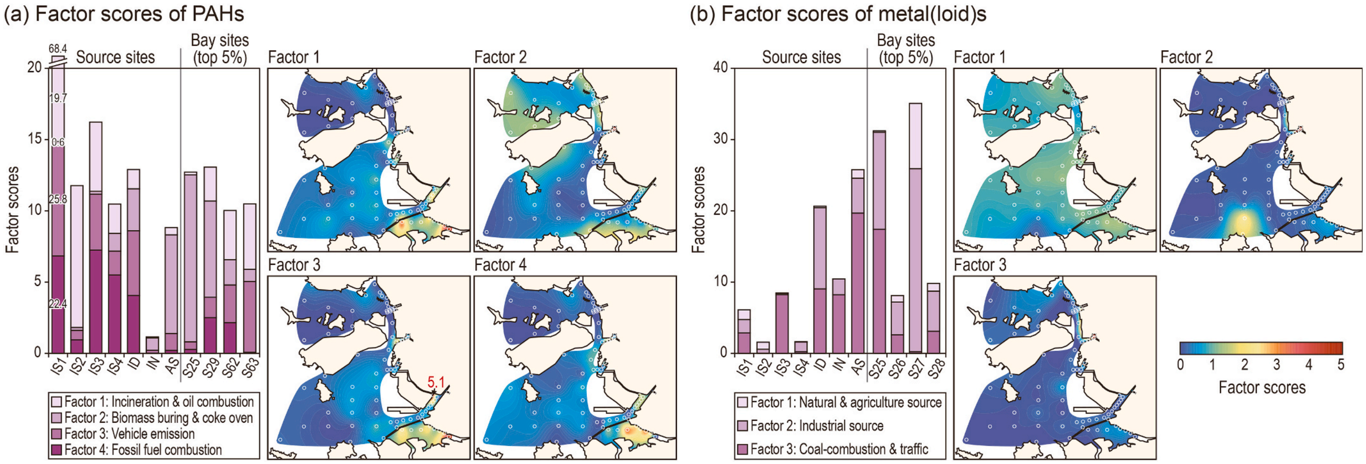


Fig. 5. Factor scores of (a) PAHs and (b) metal(loid)s in source and bay sites from surface sediments of Gyeonggi Bay. The source sites and top 5 % of the bay sites were represented using a bar graph, and the bay sites were visualized with a contour graph.

significantly correlated with styrene oligomers (SOs) ($r = 0.516$, $p < 0.001$), indicating that APs are primarily introduced into the marine environments through sewage treatment plants and waste discharge (Li et al., 2015; Soares et al., 2008; Ying et al., 2002; Ying, 2006). At most sites, NPs and their ethoxylates were more prevalent than other APs, accounting for over 64.6 % of the total APs. The high proportion of NPEOs suggests a continued influx of nonylphenol derivatives, which may exacerbate ecological risks due to their potential to bioaccumulate and disrupt endocrine functions in marine organisms. These findings underscore the need for stricter wastewater management and monitoring to address the long-term ecological risks posed by APs.

3.2.5. *Metals and metalloid*

Seven metals (Cd, Cr, Cu, Hg, Ni, Pb, and Zn) and one metalloid (As) varied across the study area (Fig. 4 and Table S9). The average PLI at the source sites were 6.3 ± 6.8 significantly higher than at the bay sites (1.4 ± 1.9) ($p = 0.047$) (Fig. S2). For example, As, Cd, Cr, and Ni concentrations exceeded the sediment quality guidelines (TEL and PEL) at several source sites, suggesting the persistence of metal contamination in these areas (Fig. 4a) (Woo et al., 2019). The average of concentrations of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn were 12.3 ± 10.9, 0.619 ± 2.33, 86.0 ± 140, 17.2 ± 48.2, 0.040 ± 0.107, 79.7 ± 125, 35.0 ± 48.1, and 145 ± 373 mg kg^{-1} dw in bay sites, respectively. As (19.3 ± 23.7 mg kg^{-1} dw), Cr (190 ± 302 mg kg^{-1} dw), Ni (79.7 ± 125 mg kg^{-1} dw), and Pb (71.2 ± 105 mg kg^{-1} dw) concentrations at the bay sites were higher than those at the source sites, along with the high concentrations observed at Incheon Port. Previous studies have identified Incheon Port as a hotspot for metal contamination and persistent organic pollutants, likely due to the proximity of a steel mill and other industrial activities (Choi et al., 2012). The I_{geo} and EF values further indicated pollution in specific sites, particularly in Incheon Port, where the contamination was significant. The high EF values at Incheon Port suggest that industrial activities, including smelting and metal processing, contribute substantially to the metal contamination in the region. The lower EF values at most sites, excluding source areas and Incheon Port, indicate that pollution in these regions is minimal. These findings underscore the importance of monitoring industrial hotspots, particularly in areas like Incheon Port, where industrial activity may exacerbate metal pollution.

3.3. *Source identification*

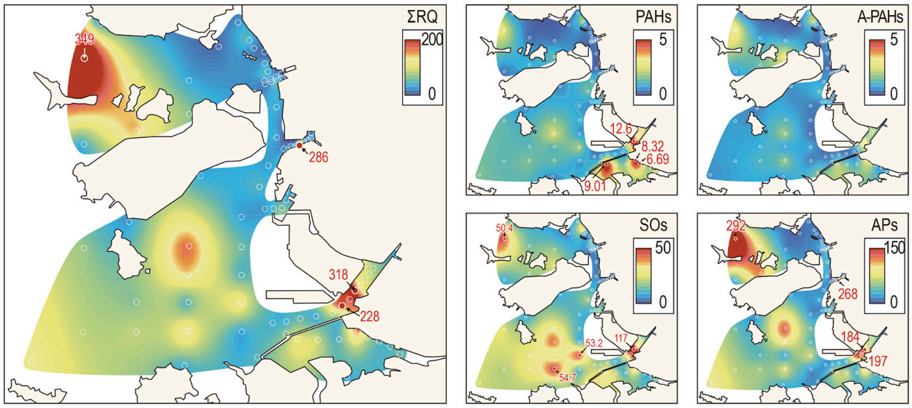
To identify the sources of organic pollutants in the sediments, the ratios between specific compounds were used (An et al., 2020; Çelebi et al., 2024; Hong et al., 2016; Kim et al., 2021; Tobiszewski and Namieśnik, 2012; Yoon et al., 2020). PAHs sources were estimated using anthracene (Ant), phenanthrene (Phe), fluoranthene (Fl), pyrene (Py), benzo[*a*]anthracene (BaA), chrysene (Chr), Indeno[1,2,3-*c*,*d*]pyrene (IcdP) and benzo[*g*,*h*,*i*]perylene (BghiP) (Fig. S5a and b). The ratios of these compounds, such as Ant/(Ant + Phe), Fl/(Fl + Py), BaA/(BaA + Chr), and IcdP/(IcdP + BghiP), varied across the sites, indicating diverse sources of contamination. Except for a few sites, most of the sites in Gyeonggi Bay appeared to be influenced by combustion sources, with A-PAHs typically indicating petrogenic origins due to their lower thermodynamic stability compared to parent PAHs (Saha et al., 2009). At the source sites, the relative compositions of PAHs and A-PAHs showed a decreasing pattern, indicative of a combustion origin, and a bell-shaped distribution, characteristic of petroleum origin (Hong et al., 2012) (Fig. S6). The pattern of A-PAHs, distributed according to alkylation through weathering, showed relatively low weathering in this study (Lee et al., 2023).

The sources of SOs were also analyzed using ratios of 2,4-diphenyl-1-butene (SD3) and 2,4,6-triphenyl-1-hexene (ST1) (Tian et al., 2020). The ratios ranged from 0.3 to 242.2 (mean: 21.5), with most sites (80.7 %) showing higher values, indicating aged expanded polystyrene (EPS) as a significant contributor. However, some inner and outer sites showed low ST1/SD3 values, suggesting fresh inputs and the ongoing flow of SOs. APs, as the degradation of APEOs, showed fresh inputs at most sites, with NPs/(NP1EOs + NP2EOs) ratios indicating a fresh input of APs (Fig. S5d) (Isobe et al., 2001). The Korean government banned the use of NPs for all domestic applications in 2007, for industrial applications such as paint and ink in 2010, and for industrial applications in 2016 (Choi et al., 2011; Jeon et al., 2017). However, the results of APs ratio values indicated a fresh influx of sources.

A PMF model was applied to identify the sources of PAHs and metal (loid)s in Gyeonggi Bay (Fig. 5). For PAHs, a 4-factor solution was most reliable, with Factor 1, accounting for 15.3 % of the total variance, linked to industrial sources, particularly waste incineration and combustion of lubricating oil, suggesting contamination from these activities (Daisey et al., 1986; Ravindra et al., 2008; Yang et al., 1998). High scores of Factor 1 were found at sites IS1 and IS2, located near the landfill area and Lake Sihwa in bay sites, indicating PAH inputs from industrial complexes transported to bay sites. Factor 2, primarily associated with biomass burning and coke ovens, was predominant in Incheon Port (Khalili et al., 1995; Lin et al., 2013; McGrath et al., 2001; Singh et al., 2013; Yunker et al., 2002), where a steel mill contributed significantly. Furthermore, the burning of biomass was the main source of the relatively high factor scores of Factor 2 in northern Gyeonggi Bay (Ren et al., 2021; Singh et al., 2013). Factor 3, related to vehicle emissions (Bzdusek et al., 2004), was found in the inner sites of the Gyeonggi Bay. Factor 4 contributing 41.1 %, was associated with fossil fuel

(a) Total risk quotient index (ΣRQ) of organic pollutants in source sites

(b) Risk quotient index of organic pollutants in bay sites



(c) Total risk quotient index (ΣRQ) of metal(loid)s in source sites

(d) Risk quotient index of metal(loid)s in bay sites

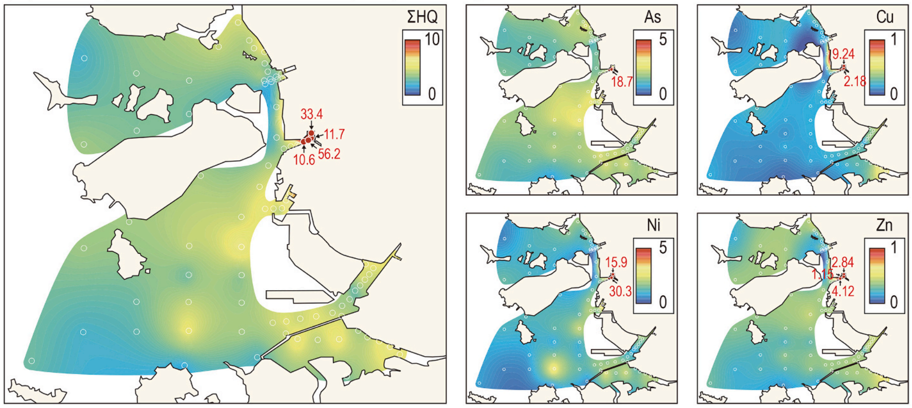


Fig. 6. Total and relative contributions of risk quotient (RQ) values for PTSs in (a) source and (b) bay sites from the surface sediments of Gyeonggi Bay.

combustion (Kavouras et al., 2001; Yang et al., 2013) and dominated in the source and inner sites. Thus, the influx of PAHs from pollutants affected the entire study area.

For metal(loid)s, the 3-factor solution was shown to have the most reliable results for pollutants (Figs. 5b and S7b). Factor 1 accounted for 58.7 %, predominant with As (90.2 %), Cr (78.9 %), Pb (75.2 %), and Ni (69.7 %). Factor 1 was widely distributed, with smelting of Cr, Pb, and Ni contributing to high score at Incheon Port, while As, Cr, and Ni were also linked to natural sources like rock weathering and erosion (Fei et al., 2019, 2020; Zhang et al., 2016). Therefore, Factor 1 was considered to convey both natural and agricultural sources. Factor 2 contributed 8.4 %, characterized by high concentrations of Cd (83.9 %), generally introduced through gas emissions generated from various industrial activities (Hu et al., 2021; Liu et al., 2018). The I_{geo} value of Cd was enriched in source sites and Incheon Port in bay sites where many industrial activities, such as oil production, occur nearby, and this trend was similar in Factor 2 (Fig. S4). Therefore, Cd contamination can be identified as an industrial source. Factor 3, related to Hg, Cu, and Zn, reflected mixed sources, including coal combustion and vehicle emissions. The main sources of Hg are complex and generally considered to be a mixed source of multiple contaminants. The accumulation of Hg is mostly related to atmospheric deposition due to coal combustion (Zhao et al., 2019), and Cu and Zn are considered important indicator pollutants for automobile exhaust gases and brake pad wear and are known to be easily deposited (Guan et al., 2018). Similar to Factor 2, it tended to be high in Incheon Port, which appears to be a result of the use of coal in industrial complexes.

PCA was also applied, with the first two components explaining 56.8 % of the total variance, with PC1 and PC2 accounting for 30.0 % and 26.8 %, respectively (Fig. S8). PC1 showed a strong positive correlation with heavy metals, including Cd, Cu, Pb, and As, indicating that these variables contributed similarly to the variance in this component. Most of the source sites were clearly differentiated from the other locations, exhibiting stronger associations with organic pollutants, except for As. The Incheon Port sites, including S25, S26, S27, and S28, which are characterized by elevated levels of heavy metal contamination, demonstrated strong correlations with metal(loid)s.

Table 1
Mini-review of PAHs, SOs, NPs, and OPs concentrations in sediments of Gyeonggi Bay in comparison to previous studies [Min.–Max. (Mean)].

Sampling regions	Type	Sampling year	# of site	Chemical compounds (ng g^{-1} dw)				Reference
				PAHs	SOs	NPs	OPs	
1. Gyeonggi Bay	Bay	1995	48	9.00–228 (64.4)	–[a]	–	–	Kim et al., 1997
		2000–2001	23	36.4–1480 (175)	–	–	–	Yim et al., 2007
		2003	25	7.00–278 (76.7)	–	3.00–63.0 (14.4)	0.100–1.40 (0.304)	Hong et al., 2009
		2008.5	7	–	–	14.4–96.9 (46.6)	–	Hong et al., 2010
		2008.8	7	–	–	6.50–36.9 (16.1)	–	Hong et al., 2010
		2015	4	29.0–62.0 (42.5)	20.0–35.0 (25.0)	–	–	Hong et al., 2016
		2015–2016	10	–	(57.4)	–	–	Tian et al., 2020
		2018	58	22.0–354 (78.3)	172–1180 (446)	29.9–689 (208)	4.98–812 (125)	This study
2. Incheon Port	Bay	1995	19	13.0–1430 (272)	–	–	–	Kim et al., 1997
		2003	8	6.00–648 (218)	–	5.00–1070 (160)	0.100–15.4 (2.43)	Hong et al., 2009
		2006–2007	1	–	–	(51.7)	–	Choi et al., 2009
		2018	15	66.3–524 (170)	139–968 (518)	69.4–2630 (386)	25.5–780 (186)	This study
3. Lake Sihwa	Lake	1998	8	(18.9)	132–324 (217)	(410)[c]		Lee et al., 2017
		1998	11	–	–	20.2–1820 (616)	4.69–50.5 (17.9)	Khim et al., 1999
		2000	12	–	–	11.0–624 (235)	–	Li et al., 2004a
		2000–2001	11	10.1–116 (47.4)	–	–	–	Yim et al., 2007
		2002	10	36.1–161 (98.3)	–	–	–	Imran et al., 2005
		2002.1	10	–	–	10.4–5050 (826)	–	Li et al., 2004b
		2002.10	10	–	–	15.6–2510 (772)	–	Li et al., 2004b
		2006–2007	5	–	–	103–702 (353)	–	Choi et al., 2009
		2008.5	7	–	–	23.5–352 (159)	–	Hong et al., 2010
		2008.8	7	–	–	<D.L.[b]–1050 (210)	–	Hong et al., 2010
		2015	11	25.0–79.0 (45.3)	10.0–70.0 (33.5)	–	–	Hong et al., 2016
		2015	5	(25.1)	10.1–62.6 (34.2)	(3.00)[c]		Lee et al., 2017
		2015–2016	10	–	(97.1)	–	–	Tian et al., 2020
		2018	9	117–449 (322)	464–1450 (914)	82.5–639 (355)	74.6–483 (284)	This study

[a] Not analyzed.
[b] Below detection limit.
[c] Concentrations of alkylphenols (APs).

3.4. Ecological risk assessments

The potential ecological risks of PTSs in the sediments of Gyeonggi Bay were evaluated by the PNEC values. At source sites, metal(loid)s represented the majority of the composition, contributing, on average, 49.1 % to the total RQ ($\sum$RQ), followed by APs (39.3 %), SOs (6.87 %), PAHs (4.45 %), and A-PAHs (0.337 %) (Fig. 6a and c). $\sum$RQ was particularly high in the AS, ID, and IN areas, where severe metal(loid) pollution was observed, with notable hot spots in IS2, IS4, IS1, and IS3 in descending order of contamination.

In contrast, at the bay sites, APs accounted for the highest proportion (56.6 %), followed by SOs (26.9 %), metal(loid)s (12.3 %), PAHs (2.60 %), and A-PAHs (1.58 %) (Fig. 6b and d). These findings suggest that, although metal(loid)s remain a major concern, organic pollutants, particularly APs, are more prevalent in coastal areas impacted by urban runoff, wastewater, and industrial discharges. Notably, NPs and NPEOs contributed significantly to the overall ecological risk, with RQ values exceeding 1 at an average of 82.9 % of the sites (excluding t-OP2EO, 32.9 %) (Fig. S9). Elevated levels of NP2EOs were particularly evident in the northern offshore areas of Gyeonggi Bay and the coastal region of Siheung. Similarly, high ecological risks were observed for SOs such as SD1 and ST1, with 97.6 % and 89.0 % of bay sites exceeding the threshold values, respectively. The substantial proportions of APs and SOs in these regions underscore the growing impact of plastic-derived pollutants. The chemical structures of these substances likely play a crucial role in their high RQ values, as their low volatility and high persistence in the environment increase their potential for bioaccumulation and toxicity.

The observed ecological risks also highlighted a concerning pattern in the coastal areas of Incheon Port. Elevated concentrations of metal(loid)s in the port, attributed to emissions from shipping and industrial complexes like steel mills, were dominated by As, Ni, and Cu (Kim et al., 2014; Mutlu et al., 2012). Among the PTSs analyzed in this study, APs and metal(loid)s were identified as major ecological risk factors in the sediments of Gyeonggi Bay. In addition, the spatial trends show inconsistencies across PTSs. Organic pollutants such as APs and SOs are less volatile and predominantly bind to organic matter, accumulating in sediments (Ying et al., 2002; Montuori et al., 2016; Kwon and Moon, 2019). In contrast to most organic pollutants, inorganic pollutants dissolve in seawater in various forms and can be distributed widely, with higher concentrations in industrial areas like ports. Additionally, metals are not usually removed from the aquatic ecosystems through natural process, further contributing to their persistence in the environment (Gautam et al., 2016). This persistence, combined with their bioaccumulation and magnification through the food chain, can lead to significant impacts on higher trophic levels, posing long-term risks to ecosystems and organisms at the top of the food web.

While the major sources of these pollutants vary, notably high RQ values observed in both source and coastal areas emphasize the severity of pollution. The ecological impact of PTSs is influenced by their physicochemical properties, and certain substances can pose disproportionately high ecological risks even at low concentrations. These results underscore the need for targeted management strategies to mitigate the impacts of these pollutants in Gyeonggi Bay.

3.5. Comparison of PTSs concentrations with previous studies

The concentrations of PTSs in the sediments obtained from this study were compared with those of previously reported studies in Gyeonggi Bay (Ahn et al., 1995; Choi et al., 2009; Hong et al., 2009; Hong et al., 2010; Hong et al., 2016; Imran et al., 2005; Khim et al., 1999; Kim et al., 1997; Kim et al., 2011; Lee et al., 2017; Lee et al., 1998; Li et al., 2004a; Li et al., 2004b; Na and Park, 2012; Ryu et al., 2011; Tian et al., 2020; Yim et al., 2007) (Tables 1 and 2). To ensure consistency, PTS concentrations were expressed as dry weight and categorized by region: “Gyeonggi Bay” (S1–S24, S40–S57, S67–S82), “Incheon Port” (S25–S39), and the inner “Lake Sihwa” (S58–S66).

Globally, studies have shown that the concentrations of persistent toxic substances (PTS) in Gyeonggi Bay are generally higher compared to many other regions worldwide (Table S11). This is likely attributed to the high degree of industrialization, urbanization, and shipping activities prevalent in the region. Such findings underscore the unique environmental pressures faced by the Gyeonggi Bay ecosystem, which are more pronounced compared to other global hotspots.

Since the early 2000s, PAH emissions through the atmosphere have been regulated, but in Gyeonggi Bay, no significant changes have been observed compared to previous studies. In contrast, a decreasing trend was observed in Incheon Port (Kim et al., 1997; Hong et al., 2009), whereas Lake Sihwa exhibited relatively higher PAH concentrations (Yim et al., 2007; Imran et al., 2005; Hong et al., 2016). Unlike other PTSs, few studies have been conducted on the SO concentrations in Gyeonggi Bay. The concentrations of SOs in this study were higher than those reported previously (Hong et al., 2019; Lee et al., 2017).

AP concentrations, mainly represented by NPs and OPs, were either comparable to or higher than previously reported levels in Gyeonggi Bay and Incheon Port (Hong et al., 2009; Hong et al., 2010; Choi et al., 2009). A notable decreasing trend for NPs was observed in Lake Sihwa (Li et al., 2004b). For metal(loid)s, limited comparable data exist, particularly for Lake Sihwa. Arsenic (As) levels exhibited an increasing trend, while Cd, Cr, Ni, and Pb concentrations were stable or lower. Zn levels showed a clear decline, whereas Incheon Port exhibited increasing concentrations of Cd, Cr, Ni, and Pb.

4. Conclusions

This study provides critical insights into the distribution, sources, and ecological risks of persistent toxic substances (PTSs) in Gyeonggi Bay sediments, offering a comprehensive understanding of contamination dynamics. By analyzing 54 PTSs across 89 sediment samples, distinct spatial distribution patterns were identified, particularly in coastal and industrial areas like Incheon Port and Lake Sihwa. These regions exhibited significantly higher concentrations of both organic pollutants (e.g., alkylphenols and steroidal estrogens) and metal(loid)s, highlighting the substantial impact of industrial discharges and urban runoff. The ecological risk assessment revealed that organic pollutants are predominantly elevated in areas affected by urban wastewater, posing potential threats through endocrine disruption in aquatic organisms. Conversely, metal(loid)s were more concentrated in industrial zones, indicating persistent contamination sources that may lead to bioaccumulation and biomagnification within the food web. Despite ongoing government regulations, certain hotspots still show elevated PTS levels, emphasizing the need for more stringent monitoring and management strategies.

Furthermore, the spatial variability of PTS distributions, influenced by their chemical properties and environmental behaviors, complicates broad-scale remediation efforts. This underscores the importance of targeted, site-specific approaches to effectively address pollution. Overall, the study emphasizes the necessity of continued monitoring of PTSs in marine sediments and suggests that enhanced regulatory measures and focused management efforts are crucial to mitigate the long-term ecological impacts of these pollutants in Gyeonggi Bay.

CRediT authorship contribution statement

Hyunseo Song: Writing – original draft, Visualization, Formal analysis, Data curation. **Taewoo Kim:** Writing – original draft, Visualization, Investigation, Formal analysis, Data curation, Conceptualization. **Junghyun Lee:** Writing – review & editing, Visualization, Investigation. **Seo Joon Yoon:** Investigation, Formal analysis. **Beomgi Kim:** Investigation. **Youngnam Kim:** Formal analysis. **Seongjin Hong:** Formal analysis, Data curation, Conceptualization. **Jong Seong Khim:** Writing – review & editing, Supervision, Project administration, Conceptualization.

Table 2

Mini-review of metal(loid)s concentrations in sediments of Gyeonggi Bay in comparison to previous studies [Min.–Max. (Mean)].

Sampling regions	Type	Sampling year	# of site	Chemical compounds (mg kg^{-1} dw)								Reference
				As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	
1. Gyeonggi Bay	Bay	1995	65	–[a]	–	23.0–82.0 (61.7)	1.00–29.0 (12.6)	–	10.0–36.0 (22.2)	–	31–117 (71.5)	Lee et al., 1998
		2005	8	3.85–8.65 (6.64)	0.0500–0.360 (0.150)	25.2–78.7 (56.7)	8.34–45.8 (21.6)	0.00977–0.0891 (0.0428)	9.16–31.1 (21.9)	23.7–33.7 (29.6)	34.3–148 (86.6)	Kim et al., 2011
		2008–2009	4	(6.97)	(0.430)	(63.2)	(56.1)	–	(28.2)	(40.7)	(69.4)	Na and Park, 2012
		2018	58	3.66–17.2 (10.6)	0.0200–1.82 (0.158)	8.83–110 (58.8)	2.15–27.8 (9.19)	0.00170–0.101 (0.0209)	6.62–19.9 (51.2)	17.4–33.0 (25.8)	9.80–56.7 (38.9)	This study
2. Incheon Port	Bay	1995	19	–	–	35.0–363 (96.6)	2.00–515 (68.7)	–	10.0–68 (27.4)	–	39.0–544 (160)	Lee et al., 1998
		1996	7	–	0.330–0.420 (0.357)	64.0–229 (98.6)	25.0–269 (79.6)	–	48.0–83.0 (58.9)	27.0–62.0 (35.7)	98.0–348 (163)	Ryu et al., 2011
		2018	15	8.40–108 (19.3)	0.100–17.5 (2.59)	45.1–1280 (190)	8.35–438 (48.8)	0.0132–0.969 (0.121)	11.8–495 (79.7)	25.2–447 (71.2)	31.4–514 (108)	This study
3. Lake Sihwa	Lake	1992	3	–	0.530–1.42 (1.08)	68.0–109 (91.7)	70.0–218 (154)	–	144–173 (161)	33.0–58.0 (47.3)	82.0–155 (116)	Ahn et al., 1995
		2018	9	8.94–14.28 (11.6)	0.0900–0.610 (0.310)	50.3–108 (86.9)	7.71–31.3 (16.6)	0.00900–0.0499 (0.0296)	11.9–43.3 (29.2)	20.8–49.7 (33.7)	30.4–65.5 (51.2)	This study

[a] Not analyzed.

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.marpolbul.2025.117652.

Data availability

Data will be made available on request.

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