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Historical trends of styrene oligomers in core sediments from the intertidal zone of the Yellow Sea and Bohai Seas

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ABSTRACT

Historical trends of styrene oligomers (SOs) in sediments from intertidal zones of the Yellow and Bohai Seas were investigated for the first time. In South Korea, SO concentrations (4.82–12.0 ng g^{-1} organic carbon (OC)) peaked during the 1990s and declined thereafter, reflecting the indirect effects of enhanced environmental regulations. In contrast, Chinese coastal regions exhibited relatively stable SO levels (1.48–20.5 ng g^{-1}OC) until the 1990s, followed by significant increases after the early 2000s. Consistently low styrene dimer-to-trimer ratios observed in Chinese sediments since 2000 indicate continuous fresh inputs of SOs. Furthermore, increasing SO concentrations and fluxes in China significantly correlated with rising national polystyrene production, suggesting the potential for continued future increases without effective controls. Conversely, the persistent decline of SO concentrations observed in surface sediments from Korean waters over the past two decades highlights the effectiveness of environmental policy interventions.

1. Introduction

Global consumption of plastics has increased dramatically, raising significant environmental concerns due to their persistence, degradation, and the release of hazardous substances (Verma et al., 2016). Among various plastics, polystyrene (PS) is one of the most widely used materials, extensively employed across multiple industrial sectors including packaging, construction, automotive manufacturing, electronics, agriculture, and household products (Plastics Europe, 2024). In 2024, global PS production accounted for approximately 5.2 % (21.5 million metric tons) of total plastic production, with Asia contributing nearly 57 % (MRCL, 2014).

PS is of particular environmental concern due to its potential to adsorb and transport hazardous pollutants, such as hexabromocyclododecanes (HBCDs) and polycyclic aromatic hydrocarbons (PAHs). In addition to PS, styrene oligomers (SOs)—which are generated as byproducts during polymerization (Mayo, 1968) and degradation of PS (Kwon et al., 2014)—have recently emerged as widespread and persistent contaminants. SOs pose considerable ecological risks due to their estrogenic or estrogen-like activities, which may disrupt endocrine functions in wildlife (Ohyama et al., 2001). They were initially detected in seawater and sand samples collected from beaches in Japan and South Korea, and subsequent studies have confirmed their global distribution and persistence in diverse environmental compartments (Saido et al., 2014; Kwon et al., 2015). Furthermore, elevated concentrations of SOs have been reported in surface sediments from creeks, rivers, estuaries, coastal waters, and offshore regions surrounding South Korea (Hong et al., 2016; Lee et al., 2018; An et al., 2020; Kim et al., 2021).

While the sources and environmental behavior of SOs have been explored to some extent, their long-term temporal trends and patterns of sedimentary accumulation remain poorly understood. Sediment cores serve as vital environmental archives, preserving chronological records of anthropogenic contamination and providing key insights into historical environmental conditions. Analyzing sediment layers enables the reconstruction of pollution histories and facilitates the identification of temporal associations between contaminant deposition, industrial production, usage patterns, and regulatory actions (Liu et al., 2012). The integration of radionuclide dating with vertical contaminant profiling

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further enhances temporal resolution, allowing for more accurate assessments of long-term contamination trends and the effectiveness of environmental policies (Zhang et al., 2013). Accordingly, sediment core analysis represents a fundamental tool for comprehensively understanding historical pollution dynamics.

The Yellow and Bohai Seas, semi-enclosed marine ecosystems shared by South Korea and China, are particularly vulnerable to environmental degradation driven by rapid urbanization and industrial expansion. Numerous rivers discharge pollutant loads from industrial complexes and densely populated urban centers into these coastal waters, posing substantial risks to ecosystem integrity (Meng et al., 2017; Kim et al., 2020; Kim et al., 2024). Although recent studies have characterized the spatial distributions and potential sources of various contaminants in the region, long-term trend analyses of persistent toxic substances (PTSs), including SOs, remain scarce. Consequently, comprehensive assessments of historical contamination and evaluations of policy effectiveness across these ecologically interconnected marine systems are urgently required.

This study aims to investigate historical contamination trends of SOs in the intertidal zones of South Korea and China. The specific objectives were to: (1) assess the vertical deposition patterns and estimate mass inventories of SOs; (2) reconstruct historical SO contamination using sediment core chronologies; and (3) explore potential correlations between SO contamination trends and national polystyrene production. Additionally, the study integrates existing datasets to evaluate temporal variations in SO contamination across diverse marine environments. The findings are expected to provide critical insights into the long-term effectiveness of past pollution control efforts and offer essential baseline data for guiding future environmental management strategies in the Yellow and Bohai Seas.

2. Materials and methods

2.1. Study area and sampling station

Eleven sediment cores were collected from intertidal zones across seven provinces bordering the Yellow and Bohai Seas in South Korea and China during 2018 (Fig. 1). Sampling stations were strategically selected based on previously published research (Yoon et al., 2020) to represent diverse land-use categories: industrial, agricultural, municipal, and aquacultural. Additional considerations included concentrations of SOs in surface sediment, sediment stability, and grain-size characteristics to minimize variability (Fig. S1). Sediment cores were obtained using square stainless-steel cores with inner dimensions of 30 × 100 mm and a length of 600 mm. The cores were immediately sealed to prevent contamination and transported to the laboratory. In the laboratory, cores were sectioned into 5-cm intervals, freeze-dried, homogenized, and sieved through a 2-mm mesh to remove coarse debris and biological

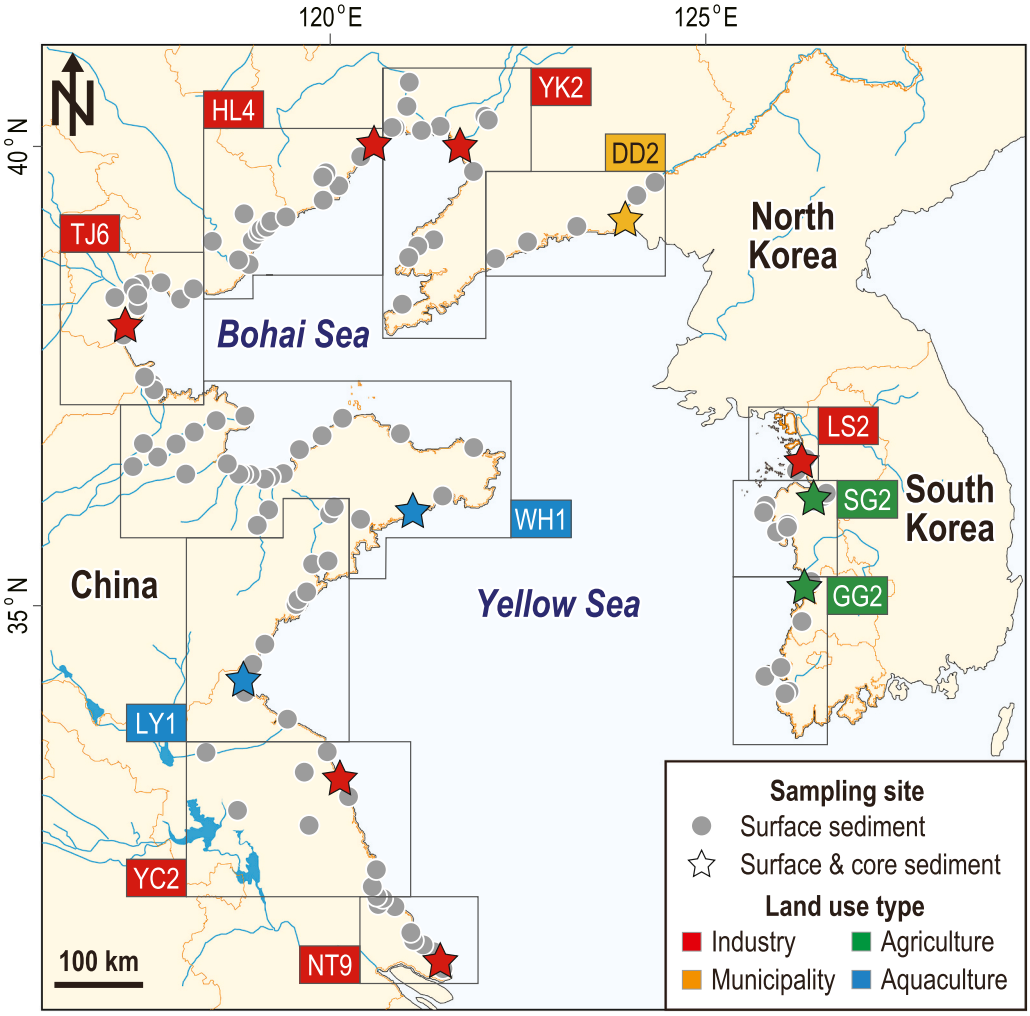

Fig. 1. Map showing sediment sampling stations from intertidal zones of the Yellow and Bohai Seas. Sediment cores collected in this study are marked with stars. The base map and land-use classifications are adapted from Yoon et al. (2020) and previous studies. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

materials, ensuring consistency across analyses.

2.2. Quantification of SOs

Extraction and quantification of SOs were performed following established protocols adapted from Hong et al. (2016). Approximately 10 g of freeze-dried sediment was extracted with 350 mL of dichloromethane (DCM, Burdick & Jackson, Muskegon, MI) using a Soxhlet extraction for 16 h. Prior to extraction, surrogate standards were added to assess analytical recoveries and potential matrix effects. Following extraction, solvents were concentrated via rotary evaporation and exchanged with hexane (Burdick & Jackson). Elemental sulfur was removed by treatment with activated copper powder (Sigma-Aldrich, St. Louis, MO, USA). Extracts were further purified using columns packed with 8 g of activated silica gel (70–230 mesh, Sigma Aldrich) and eluted with 60 mL of hexane:DCM mixture (80:20, v/v). Eluents were subsequently concentrated to approximately 1 mL under nitrogen, transferred to GC vials, and spiked with internal standards before analysis. Quantification of SOs was performed using gas chromatography (Agilent 7890B, Agilent Technologies, Santa Clara, CA, USA) coupled with a 5977B mass selective detector (MSD; Agilent Technologies). Detailed information on the target compounds, method detection limits (MDLs), recovery rates, and instrumental conditions is provided in Tables S1 and S2.

2.3. Calculation of mass inventory, date, and flux

Mass inventory (I) of SOs in intertidal sediments of the Yellow and Bohai Seas was calculated using Eq. (1):

$$I = C_x \times A_i \times p_x \times d \quad (1)$$

Where C_x is the concentration of SOs at sediment depth x (ng g^{-1}), A_i represents the regional intertidal area (km^2), p_x is the sediment bulk density (g cm^{-3}), and d is sediment depth (m). Intertidal area values were obtained from geographic surveys (Yim et al., 2018), and sediment bulk density data were sourced from previous studies (Yoon et al., 2023). Sediment depth was uniformly set at 0.5 m to ensure comparability. To reconstruct historical SO deposition, sediment cores were dated using the constant flux constant sedimentation (CFCS) model based on the radionuclide ^{210}Pb. Reliable chronological data were available for four representative stations (TJ6, DD2, YC2 in China; SG2 in South Korea). Sediment accumulation rates (SARs), derived from ^{210}Pb dating, ranged from 0.27 to 1.28 cm yr^{-1}, allowing historical reconstruction back to approximately the 1950s (Yoon et al., 2023). The deposition flux (F) of SOs was subsequently calculated using Eq. (2):

$$F = C_x \times p_x \times S \quad (2)$$

where S represents sediment accumulation rates (cm y^{-1}). Historical flux data provided insights into temporal contamination trends over approximately eight decades.

2.4. Data analyses

Sedimentary concentrations of SOs were expressed both as organic carbon-normalized (ng g^{-1} OC) values to account for variability in sediment organic matter content and original dry weight concentrations (ng g^{-1} dw) to facilitate direct comparison with prior studies. Statistical analyses were conducted using SPSS software (version 25.0). Due to non-parametric data distributions, Kruskal-Wallis tests with Bonferroni corrections were applied to identify significant concentration differences among land-use types (industrial, agricultural, municipal, and aquacultural). Principal Component Analysis (PCA) and hierarchical cluster analysis based on Euclidean distances were employed to evaluate distribution patterns of SO contamination. To account for limited data points and non-normal distributions, Spearman's rank correlation was used to evaluate the relationship between SO concentrations and national polystyrene production, while the Mann–Kendall trend test was applied to assess monotonic temporal trends. Additionally, sedimentary SO concentrations were categorized based on NOAA (1991) and Daskalakis and O'Connor (1995) guidelines. Concentrations exceeding the 85th percentile were classified as 'High', and those exceeding five times this value were defined as '5-x-High', enabling comparative evaluations and providing regional criteria for sediment contamination.

3. Results and discussion

3.1. Vertical distribution of SOs and mass inventory

SOs were detected in all sediment core samples, exhibiting irregular vertical distribution patterns that varied by both region and depth (Fig. 2). In South Korea, concentrations of SOs ranged from 2.47 to 17.0 ng g^{-1} OC (mean: 8.38 ng g^{-1} OC), whereas concentrations in China spanned from 1.38 to 118 ng g^{-1} OC (mean: 9.12 ng g^{-1} OC). The highest mean concentration was observed at station YK2 (22.6 ng g^{-1} OC), followed by YC2 (16.6 ng g^{-1} OC), HL4 (12.3 ng g^{-1} OC), and LS2 (9.65 ng g^{-1} OC). Elevated SO levels were strongly associated with industrial land-use types, consistent with previous studies that identified industrial activities as major sources of SO contamination in heavily industrialized cities, such as Siheung, Masan, and Ulsan in South Korea (Hong et al., 2016; Lee et al., 2018; An et al., 2020).

Beyond industrial zones, relatively high SO concentrations were also observed at agricultural sites (SG2 and GG2), likely due to the widespread use and disposal of polystyrene products in farming practices (Puoci et al., 2008). In contrast, lower concentrations were generally found at aquacultural and municipal stations, reflecting more limited usage of polystyrene in these sectors. Principal component analysis (PCA) further supported these observations, statistically distinguishing industrial and agricultural stations from municipal and aquacultural sites based on significantly higher SO concentrations (Kruskal-Wallis test with Bonferroni correction, $p < 0.05$; Fig. S2 and S3), thereby highlighting the dominant influence of industrial and agricultural sources on SO contamination.

Mass inventories of SOs in intertidal sediments also exhibited considerable regional variation across the Yellow and Bohai Seas, ranging from 0.51 to 10.4 tons (mean: 3.03 T) (Fig. 2). The highest inventory was recorded at YK2 (10.4 T, Bohai Sea), followed by YC2 (6.80 T) and GG2 (5.53 T). Despite its relatively small intertidal area, YK2 exhibited the largest inventory, suggesting the presence of intense localized pollution. In contrast, although YC2 and GG2 spanned larger intertidal areas, their inventories accounted for only 65.7 % and 53.4 % of the YK2 value, respectively. These results imply the existence of strong point sources near YK2, aligning with previous findings of elevated levels of other persistent toxic substances (e.g., PAHs, Dichlorodiphenyltrichloroethane (DDT), Polychlorinated Biphenyls (PCBs), Hexachlorobenzene (HCB)) in the Yingkou region (Zhang et al., 2009; Wang et al., 2020).

Among all study regions, the Bohai Sea accounted for 39.5 % of the total SO mass inventory, despite representing only ~24.0 % of the total intertidal area examined. In comparison, the Yellow Sea along the Chinese and Korean coasts contributed 32.7 % and 27.8 %, respectively. The disproportionately high SO inventory in the Bohai Sea suggests more severe historical contamination, likely associated with greater regional use and emissions of polystyrene. This finding is consistent with previous reports of elevated PAH inventories in the same region (Yoon et al., 2023). Furthermore, the higher SO burden along the Chinese coast underscores the influence of national differences in polystyrene consumption and management practices between China and Korea.

3.2. Historical trends of SOs

Historical trends of SO concentrations differed markedly between

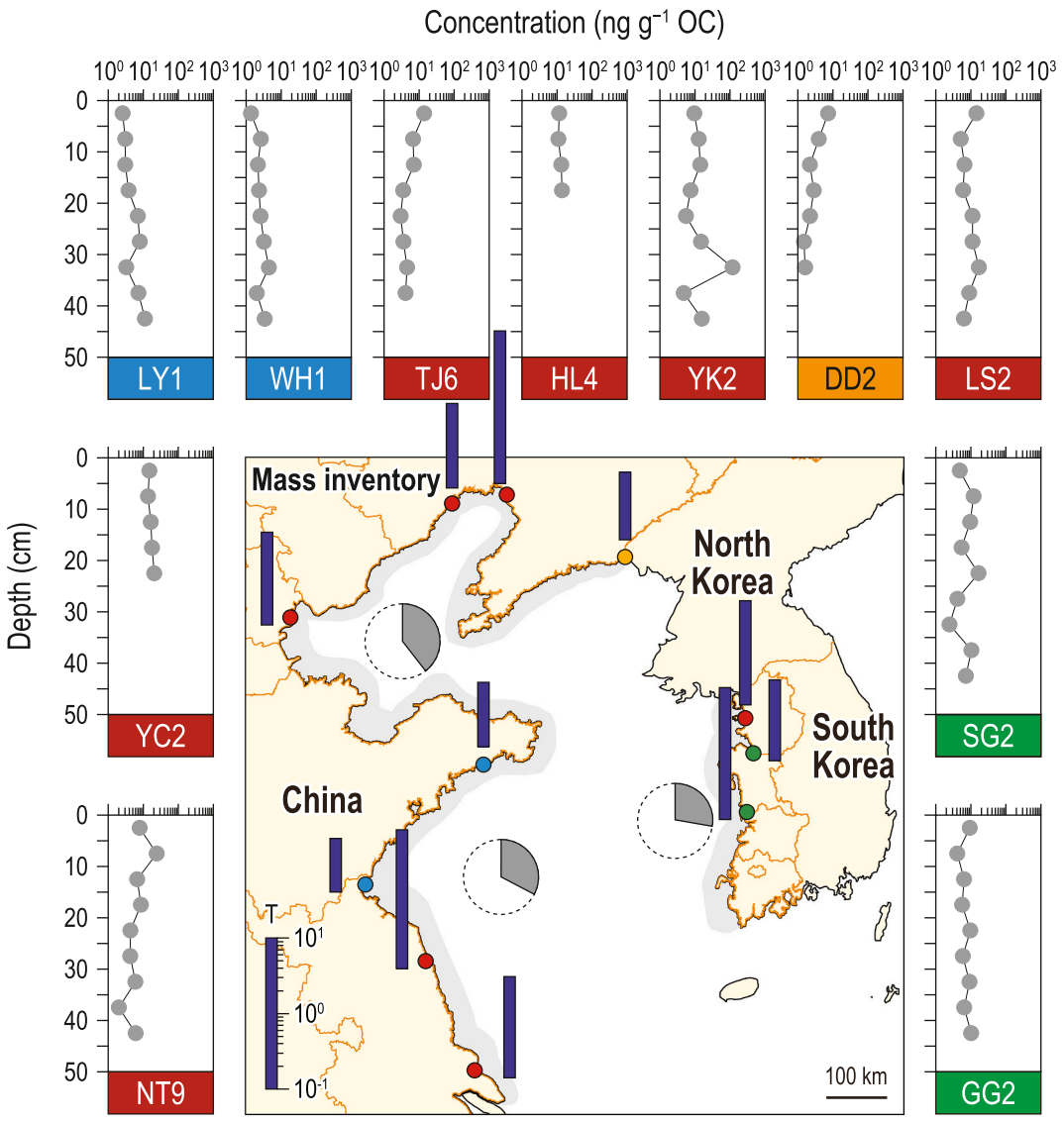

Fig. 2. Vertical distributions and mass inventories of SOs in intertidal sediments from the Yellow and Bohai Seas. Pie charts indicate the relative contributions (%) of each region (Yellow Sea of Korea, Yellow Sea of China, and Bohai Sea) to the total SO mass inventory. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

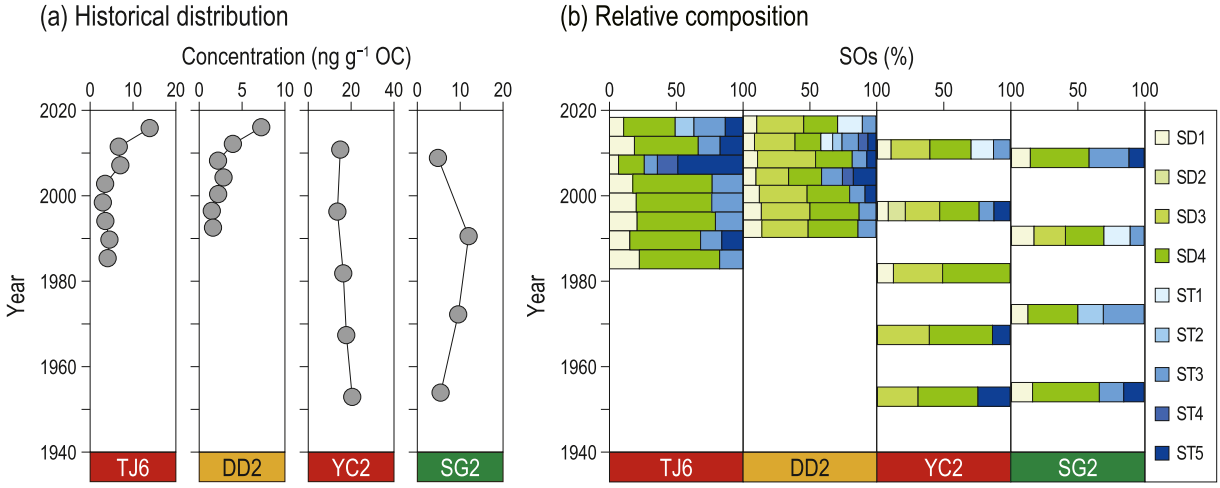

Fig. 3. (a) Historical trends and (b) relative compositional profiles of SOs in dated sediment cores from the Yellow and Bohai Seas. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

South Korea and China (Fig. 3a). In South Korea (station SG2), SO concentrations progressively increased from the 1950s, peaking around the 1990s coinciding with rapid industrialization. Since the early 2000s, concentrations have steadily declined, likely reflecting the indirect but positive impacts of strengthened environmental regulations (Kim et al., 2020). Notably, this trend contrasts with that of PAHs—pollutants unintentionally generated in the same region—which continued to increase after the early 2000s (Yoon et al., 2023). This divergence suggests that SO, unlike PAHs, have been indirectly controlled through reductions in source emissions associated with policy interventions. Similar declining patterns have been observed for other regulated contaminants, such as PCBs and alkylphenols (APs) at SG2 (Yoon et al., 2025). However, the decline in SO concentrations lagged approximately two decades behind these directly regulated substances, underscoring the indirect nature of regulation for SOs, which have not been explicitly targeted by environmental policy. Moreover, recent observations of stagnant or slightly decreasing microplastic levels in sediment cores from the same estuary in the Korean Yellow Sea (Park et al., 2023) support this conclusion. These findings suggest that the observed reductions in SO concentrations are more likely due to decreased source inputs rather than post-depositional processes such as sediment dilution.

Sedimentary SO concentrations at Chinese stations (TJ6, DD2, YC2) remained relatively stable or slightly increasing until the 1990s, but showed significant increases from the early 2000s onward. This trend suggests that environmental policies implemented after China's Reform and Opening-up (1978) and those introduced in the early 2000s were of limited effectiveness in curbing SO emissions (Ma et al., 2020). The rising concentrations of SOs parallel those observed for other persistent pollutants, including PAHs, PCBs, APs, and heavy metals (Yu et al., 2020; Yoon et al., 2023), indicating ongoing emissions likely driven by inadequate regulatory enforcement and sustained industrial activity. In contrast, certain southern regions of China, such as the Pearl River Delta and Dachan Bay, have experienced declines in metal concentrations since 2010, attributed to targeted local interventions including industrial relocation and emission control measures (Yang et al., 2020; Shi et al., 2022). These regional contrasts highlight the uneven implementation and effectiveness of environmental regulations across China, suggesting that current frameworks may be insufficient to mitigate SO contamination at the national scale. Complementary evidence from Hangzhou Bay, located adjacent to the Yellow Sea, further supports this conclusion. Sediment cores from this region show continuously increasing microplastic concentrations from the 1980s to the present (Li et al., 2020). Given that polystyrene—one of the main components of microplastics—is the parent material of SOs (Lin et al., 2021), this trend implies sustained or even growing SO inputs into Chinese coastal environments.

Compositional analyses of SOs revealed relatively consistent patterns across sediment samples from both Korea and China (Fig. 3b). Styrene dimer (SD) 3 was detected in all samples (100 %), followed by SD1 (91.7 %), ST3 (87.5 %), and ST5 (54.2 %), along with other minor components. Over approximately 70 years, SDs consistently dominated over styrene trimers (STs) in both countries, although temporal and regional variations were evident. Minimal compositional changes were observed at stations TJ6 and DD2, whereas substantial shifts occurred at YC2 and SG2, suggesting differences in local industrial practices or the introduction of new SO sources in recent decades (Fig. S4).

Notably, the proportion of SDs relative to STs decreased in recent sediments, particularly after the year 2000. This declining SD/ST ratio strongly indicates recent and ongoing SO inputs (Tian et al., 2020). Previous studies have shown that STs tend to dominate over SDs during low-temperature degradation (Kimukai et al., 2020) and fast pyrolysis of polystyrene (Zhou et al., 2016). Thus, the lower SD/ST ratios observed in sediments from Tianjin and Dandong likely reflect continuous influxes of fresh SOs, potentially originating from nearby chemical industries manufacturing expandable polystyrene (EPS) and polystyrene resins. Overall, the compositional and temporal trends indicate that SO contamination remains active in specific coastal areas. While historical patterns differ between Korea and China, the recent shifts in SO composition underscore the need for ongoing monitoring, particularly in regions experiencing increasing concentrations and fresh pollutant inputs.

3.3. Implications for the rise of polystyrene production

Historical trends and fluxes of SOs closely mirrored national PS production patterns in both South Korea and China (Fig. 4). In South Korea, total annual PS production peaked in the 1990s and has since remained relatively stable with minor fluctuations (Fig. S5). This production stagnation, combined with strengthened environmental regulations introduced in the early 2000s, likely contributed to the subsequent decline in sedimentary SO concentrations (Kim et al., 2020). In contrast, China's PS production has expanded rapidly since the 1970s, with a sharp acceleration after the early 2000s. The average annual growth rate has reached approximately 15 % (Jiang et al., 2020). This sustained expansion strongly corresponded with rising SO concentrations in estuarine sediments from stations such as TJ6, DD2, and YC2. Statically significant correlations were observed between annual SO concentrations and PS production at TJ6 ($r = 0.80, p < 0.05$) and DD2 ($r = 0.84, p < 0.05$), indicating that intensified PS manufacturing activities directly contributed to increased SO inputs into marine environments.

Sediment flux analyses of TJ6 and DD2 further supported these trends, revealing significant increases in SO accumulation rates ($r = 0.75–0.85$, $p < 0.05$). In addition, SO flux at station YC2 also exhibited a perfect positive monotonic correlation with national polystyrene production (Spearman's $\rho = 1.00, p < 0.01$), indicating a consistent increase in flux corresponding to rising PS production. Interestingly, the rate of increase in SO fluxes was even steeper than that observed in concentration data, suggesting continuous and accelerating accumulation of SOs in sediments of the Yellow and Bohai Seas. These findings align with similar trends observed for other industrial contaminants, including PAHs, PCBs, and brominated flame retardants such as HBCDs, particularly in rapidly industrializing coastal areas like Tianjin (Wu et al., 2019; Zhu et al., 2017). Zhu et al. (2017) reported substantial increases (12.9–41.6 %) in sedimentary HBCD concentrations near expanded styrene production facilities in Tianjin between 2010 and 2015, demonstrating a direct linkage between intensified plastic production and elevated environmental pollutant levels.

Tianjin is expected to host the world's largest EPS production facility. This development suggests that SO contamination in the region may persist or even intensify, underscoring the need for continuous environmental monitoring. Supporting this concern, the accumulation of plastic debris on beaches along the East China Sea increased steadily between 2016 and 2021, reflecting ongoing environmental pollution driven by rising plastic production, growing consumption, and inadequate waste management systems (Hao and Jiang, 2023). Furthermore, sediment core analyses from coastal regions of the East China Sea have revealed a continuous increase in microplastic concentrations since the 1960s, with polystyrene accounting for approximately 23.8 % of total microplastic particles (Lin et al., 2021). These findings provide direct evidence of long-term and increasing SO contamination in marine sediments.

Collectively, the observed relationships among escalating PS production, plastic debris generation, and SO contamination highlight an ongoing and significant environmental burden. These findings underscore the urgent need for proactive pollution control strategies, the implementation of more effective regulatory frameworks, and strengthened international cooperation to mitigate future SO contamination in coastal ecosystems.

3.4. Recent trends of SOs in sediment

Recently measured SO concentrations in surface sediments from

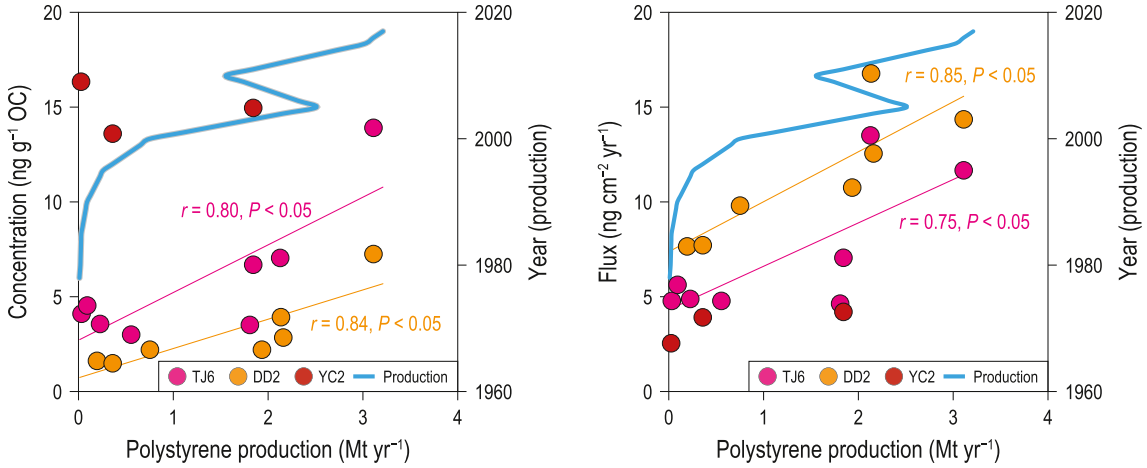

Fig. 4. Relationships between SO concentrations and fluxes in dated sediments and polystyrene production in China. The blue line represents annual polystyrene production data. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

various freshwater and marine environments in South Korea and China were compared to contextualize current contamination levels within broader geographical and historical perspectives (Fig. 5) (Hong et al., 2016; Lee et al., 2017; Yoon et al., 2017; Lee et al., 2018; Cha et al., 2019; Hong et al., 2019; Kim et al., 2019; An et al., 2020; Kim et al., 2020; Lee et al., 2020; Tian et al., 2020; Yoon et al., 2020; Kim et al., 2021; Gwak et al., 2022; An et al., 2023; Kim et al., 2024). Concentrations of SOs were significantly higher in freshwater sediments (e.g., creeks, rivers, and lakes) than in marine sediments (e.g., bays, estuaries, and offshore areas), indicating that terrestrial activities are major sources of SOs (An et al., 2020; Gwak et al., 2022).

According to threshold effect concentration (TEC)-based regional guidelines for freshwater environments, "High" and "5-x-High" levels are defined as 830 ng g^{-1} dw and 4150 ng g^{-1} dw, respectively (Yoon et al., 2020). Among the surveyed freshwater sites, only sediments from the heavily industrialized city of Ulsan exceeded the 5-x-High threshold, clearly suggesting industrial emissions as the dominant source of SO contamination (An et al., 2023). In contrast, marine sediments generally

Country	Region	Area	Sampling year	# of sites	References
		Freshwater			
Korea	Ulsan	Creeks	2017	2	An et al., 2020
	Ulsan	River	2017	7	An et al., 2023
	Ulsan	Creeks	2017	4	Kim et al., 2019
	Ansan	Creeks	2013	10	Hong et al., 2019
	Ansan	Creeks	2015	7	Tian et al., 2020
	Ansan	Creeks	2015	8	Cha et al., 2019
	Ansan	Creeks	2015	6	Hong et al., 2016
	Ansan	Creeks	2015	6	Lee et al., 2017
	Ansan	Lake	1998	8	Lee et al., 2017
	Pohang	Creeks	2018	1	Gwak et al., 2022
	Ansan	Lake	2015	10	Tian et al., 2020
	Ulsan	River	2017	2	An et al., 2020
	Ulsan	Creeks	2020	4	An et al., 2023
	Masan	Creeks	2016	2	Lee et al., 2020
	Ansan	Lake	2015	11	Hong et al., 2016
China	Yellow Sea	River & Lake	2018	73	Yoon et al., 2020
Korea	Yellow Sea	Lake	2010 – 2018	41	Kim et al., 2020
		Seawater			
Korea	Masan	Bay	1998	7	Lee et al., 2018
	National	Offshore	2017 – 2018	34	Kim et al., 2021
	Masan	Bay	2014	7	Lee et al., 2018
	Pohang	Bay	2018	5	Gwak et al., 2022
	Ansan	Bay	2015	9	Tian et al., 2020
	Ulsan	Bay	2017	5	An et al., 2020
	Ansan	Bay	2015	4	Hong et al., 2016
China	Yellow Sea	Estuary	2018	32	Yoon et al., 2020
Korea	Yellow Sea	Estuary	2010 – 2018	45	Kim et al., 2020
	Gunsan	Estuary	2014	58	Yoon et al., 2017
	Yellow Sea	Estuary	2018	14	Yoon et al., 2020
	Yellow Sea	Offshore	2021	35	Kim et al., 2024

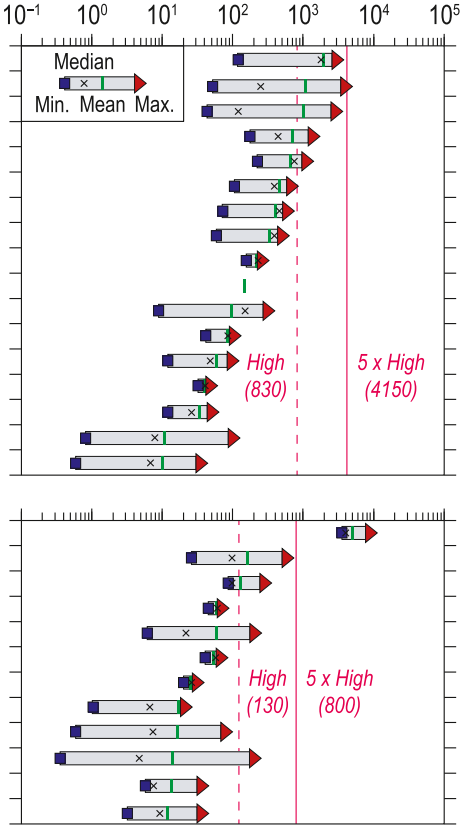

Fig. 5. Comparative summary of reported SO concentrations in surface sediments from freshwater environments (creeks, rivers, and lakes) and marine environments (estuaries, bays, and offshore areas).

showed lower SO concentrations. The corresponding "High" and "5-x-High" thresholds for marine sediments are 130 ng g^{-1} dw and 800 ng g^{-1} dw, respectively. Notably, concentrations exceeding the 5-x-High threshold were detected only in Masan Bay, a heavily industrialized coastal area in southern South Korea (Lee et al., 2018). These comparative finding highlight terrestrial and industrial activities as major contributors to marine SO contamination. Similar trends were observed in Tokyo Bay, where elevated SO inputs were linked to increased terrestrial runoff following precipitation events (Amamiya et al., 2019). Furthermore, studies from the broader Yellow Sea region—encompassing industrial, agricultural, municipal, and aquacultural areas—consistently reported lower SO concentrations compared to sites directly impacted by industrial activities. This further supports the conclusion that localized industrial emissions are a primary driver of SO contamination in both freshwater and marine systems (Kim et al., 2020; Yoon et al., 2020).

Temporal analyses of recent SO concentrations across various aquatic environments in South Korea (creeks, rivers, lakes, estuaries, bays) revealed an overall decreasing tendency from the mid-1990s through 2020, with ecosystem-specific patterns of decline (Fig. 6). While the temporal trends were not statistically significant in individual environments ($\tau = -0.40$–0.00, $p > 0.05$), it still suggests a system-wide reduction in SO contamination. These pronounced declines likely reflect the effective implementation of rigorous environmental regulations targeting industrial emissions and pollution sources since the 1990s in South Korea (Kim et al., 2020; Lee et al., 2018). Nevertheless, recent studies indicated increasing microplastic accumulation in sediments from industrial and aquacultural areas heavily utilizing EPS, raising concerns about the potential for ongoing SO contamination (Eo et al., 2023).

In contrast, comprehensive assessments of recent SO trends in China remain limited due to the lack of contemporary surface sediment data. While sediment core analyses indicate significant increases in SO concentrations since the early 2000s (e.g., TJ6 and DD2), the scarcity of recent surface sediment monitoring hampers conclusive evaluation of current contamination patterns across broader coastal regions. These findings highlight the urgent need for more systematic and sustained monitoring efforts to accurately track ongoing and emerging SO contamination dynamics in China. Overall, recent sedimentary trends of SOs underscore complex interactions between industrial growth, environmental policy effectiveness, and pollutant inputs into aquatic ecosystems. Continuous monitoring, coupled with adaptive and enforceable policy frameworks, remains essential for mitigating SO contamination, safeguarding marine ecosystem integrity, and protecting public health from persistent organic pollutants.

4. Conclusions

To the best of our knowledge, this is the first study to comprehensively assess the vertical distribution, historical trends, and fluxes of SOs in the intertidal sediments of the Yellow and Bohai Seas. Our findings identify industrial activity as the primary source of SO contamination, with clear temporal differences between South Korea and China from the 1950s to the 2010s. These contrasting trends reflect the differential effectiveness of environmental regulations in each country. In China, SO concentrations steadily increased in line with the rapid expansion of

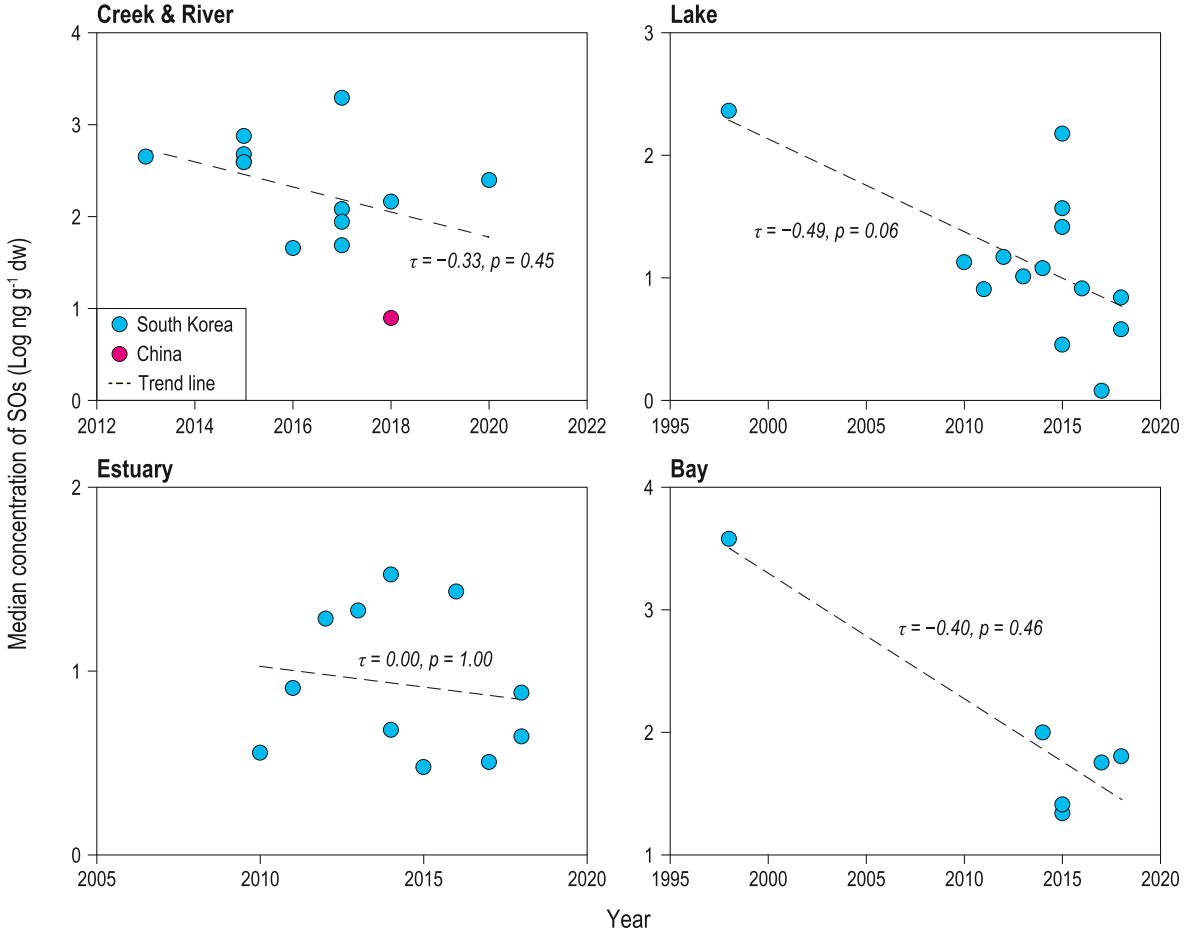

Fig. 6. Temporal variations of SO concentrations in surface sediments collected from creeks and rivers, lakes, estuaries, and bays in South Korea and China. Linear trend lines were fitted to visualize temporal changes in SO concentrations. Statistical significance was assessed separately using the Mann–Kendall trend test.

polystyrene production, whereas concentrations in South Korea declined markedly after the 2000s, likely due to strengthened policy measures. However, limitations in sediment dating resolution and data availability constrain our understanding of temporal variation and individual SO compound composition. Advancing compositional analyses will be crucial for accurately tracing contamination sources and understanding their environmental fate. Overall, this study provides critical historical baseline data to inform future environmental management and support the development of effective regulatory frameworks to address SO contamination.

CRediT authorship contribution statement

Seo Joon Yoon: Writing – original draft, Visualization, Formal analysis, Data curation, Conceptualization. **Junghyun Lee:** Writing – review & editing, Visualization, Investigation, Data curation, Conceptualization. **Youngnam Kim:** Formal analysis, Data curation. **Wenyou Hu:** Project administration, Investigation. **Tieyu Wang:** Project administration, Investigation. **Seongjin Hong:** Visualization, Formal analysis, Data curation. **Jong Seong Khim:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

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.2025.118252.

Data availability

Data will be made available on request.

References

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

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

An, S.-A., Lee, J., Cha, J., Gwak, J., Kim, M., Hur, J., Hong, S., Khim, J.S., 2023. Characterization of microalgal toxicants in the sediments from an industrial area: application of advanced effect-directed analysis with multiple endpoint bioassays. Environ. Int. 173, 107833.

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

Daskalakis, K.D., O'Connor, T.P., 1995. Distribution of chemical concentrations in US coastal and estuaine sediment. Mar. Environ. Res. 40, 381–398.

Eo, S., Hong, S.H., Cho, Y., Song, Y.K., Han, G.M., Shim, W.J., 2023. Spatial distribution and historical trend of microplastic pollution in sediments from enclosed bays of South Korea. Mar. Pollut. Bull. 193, 115121.

Gwak, J., Cha, J., Lee, J., Kim, Y., An, S.-A., Lee, S., Moon, H.-B., Hur, J., Giesy, J.P., Hong, S., 2022. Effect-directed identification of novel aryl hydrocarbon receptor-active aromatic compounds in coastal sediments collected from a highly industrialized area. Sci. Total Environ. 803, 149969.

Hao, H., Jiang, C., 2023. The path of transboundary marine plastic waste management in China, Japan, and South Korea from the perspective of the blue economy. Front. Mar. Sci. 9, 1075667.

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

Hong, S., Lee, Y., Yoon, S.J., Lee, J., Kang, S., Won, E.-J., Hur, J., Khim, J.S., Shin, K.-H., 2019. Carbon and nitrogen stable isotope signatures linked to anthropogenic toxic substances pollution in a highly industrialized area of South Korea. Mar. Pollut. Bull. 144, 152–159.

Jiang, X., Wang, T., Jiang, M., Xu, M., Yu, Y., Guo, B., Chen, D., Hu, S., Jiang, J., Zhang, Y., 2020. Assessment of plastic stocks and flows in China: 1978-2017. Resour. Conserv. Recycl. 161, 104969.

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

Kim, S., Hong, S., Lee, J., Kim, T., Yoon, S.J., Lee, J., Choi, K., Kwon, B.-O., Giesy, J.P., Khim, J.S., 2020. Long-term trends of persistent toxic substances and potential toxicities in sediments along the west coast of South Korea. Mar. Pollut. Bull. 151, 110821.

Kim, Y., Hong, S., Lee, J., Yoon, S.J., An, Y., Kim, M.-S., Jeong, H.-D., Khim, J.S., 2021. Spatial distribution and source identification of traditional and emerging persistent toxic substances in the offshore sediment of South Korea. Sci. Total Environ. 789, 147996.

Kim, Y., Lee, Y., Lee, C.-E., Jeong, H., Ra, K., Choi, D., Hong, S., 2024. Compound-and element-specific accumulation characteristics of persistent toxic substances and metals in sediments of the Yellow Sea. J. Hazard. Mater. 476, 134926.

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

Kimukai, H., Kodera, Y., Koizumi, K., Okada, M., Yamada, K., Hiaki, T., Saido, K., 2020. Low temperature decomposition of polystyrene. Appl. Sci. 10, 5100.

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

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

Lee, J., Hong, S., Kwon, B.-O., Cha, S.A., Jeong, H.-D., Chang, W.K., Ryu, J., Giesy, J.P., Khim, J.S., 2018. Integrated assessment of persistent toxic substances in sediments from Masan Bay, South Korea: comparison between 1998 and 2014. Environ. Pollut. 238, 317–325.

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

Li, J., Huang, W., Xu, Y., Jin, A., Zhang, D., Zhang, C., 2020. Microplastics in sediment cores as indicators of temporal trends in microplastic pollution in Andong salt marsh, Hangzhou Bay, China. Reg. Stud. Mar. Sci. 35, 101149.

Lin, J., Xu, X.-M., Yue, B.-Y., Xu, X.-P., Liu, J.-Z., Zhu, Q., Wang, J.-H., 2021. Multidecadal records of microplastic accumulation in the coastal sediments of the East China Sea. Chemosphere 270, 128658.

Liu, L.-Y., Wang, J.-Z., Wei, G.-L., Guan, Y.-F., Wong, C.S., Zeng, E.Y., 2012. Sediment records of polycyclic aromatic hydrocarbons (PAHs) in the continental shelf of China: implications for evolving anthropogenic impacts. Environ. Sci. Technol. 46, 6497–6504.

Ma, X., Wan, H., Zhou, J., Luo, D., Huang, T., Yang, H., Huang, C., 2020. Sediment record of polycyclic aromatic hydrocarbons in Dianchi lake, southwest China: influence of energy structure changes and economic development. Chemosphere 248, 126015.

Mayo, F.R., 1968. The dimerization of styrene. J. Am. Chem. Soc. 90, 1289–1295.

Meng, J., Hong, S., Wang, T., Li, Q., Yoon, S.J., Lu, Y., Giesy, J.P., Khim, J.S., 2017. Traditional and new POPs in environments along the Bohai and Yellow Seas: an overview of China and South Korea. Chemosphere 169, 503–515.

Merchant Research & Consulting Ltd (MRCL), 2014. Polystyrene in the global economy: production capacity and market insights.

NOAA, National Status and Trends Program, 1991. Second summary of data on chemical contaminants in sediment from the national status and trends program. National Oceanic and Atmospheric Administration. NOAA Tech. Memo. NOS ORCA (Formerly OMA) 59, NOAA Office of Oceanography and Marine Assessment, Rockville, MD..

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

Park, J., Hong, S., Shim, W.J., Khim, J.S., Park, J., 2023. Distribution, compositional characteristics, and historical pollution records of microplastics in tidal flats of South Korea. Mar. Pollut. Bull. 189, 114741.

Plastics Europe, 2024. Plastics - the facts 2024.

Puoci, F., Iemma, F., Spizzirri, U.G., Cirillo, G., Curcio, M., Picci, N., 2008. Polymer in agriculture: a review. Am. J. Agric. Biol. Sci. 3, 299–314.

Saido, K., Koizumi, K., Sato, H., Ogawa, N., Kwon, B.G., Chung, S.-Y., Kusui, T., Nishimura, M., Kodera, Y., 2014. New analytical method for the determination of styrene oligomers formed from polystyrene decomposition and its application at the coastlines of the North-West Pacific Ocean. Sci. Total Environ. 473, 490–495.

Shi, C., He, H., Xia, Z., Gan, H., Xue, Q., Cui, Z., Chen, J., 2022. Heavy metals and Pb isotopes in a marine sediment core record environmental changes and anthropogenic activities in the Pearl River Delta over a century. Sci. Total Environ. 814, 151934.
Tian, Z., Kim, S.-K., Hyun, J.-H., 2020. Environmental distribution of styrene oligomers (SOs) coupled with their source characteristics: tracing the origin of SOs in the environment. J. Hazard. Mater. 398, 122968.
Verma, R., Vinoda, K., Papireddy, M., Gowda, A., 2016. Toxic pollutants from plastic waste-a review. Procedia Environ. Sci. 35, 701–708.
Wang, P., Zhang, L., Lin, X., Yan, J., Zhang, P., Zhao, B., Zhang, C., Yu, Y., 2020. Spatial distribution, control factors and sources of heavy metal in the surface sediments of Fudu Estuary waters, East Liaodong Bay, China. Mar. Pollut. Bull. 156, 111279.
Wu, Z., Lin, T., Li, A., Zhou, S., He, H., Guo, J., Hu, L., Li, Y., Guo, Z., 2019. Sedimentary records of polychlorinated biphenyls in the East China marginal seas and Great Lakes: significance of recent rise of emissions in China and environmental implications. Environ. Pollut. 254, 112972.
Yang, G., Song, Z., Sun, X., Chen, C., Ke, S., Zhang, J., 2020. Heavy metals of sediment cores in Dachan Bay and their responses to human activities. Mar. Pollut. Bull. 150, 110764.
Yim, J., Kwon, B.-O., Nam, J., Hwang, J.H., Choi, K., Khim, J.S., 2018. Analysis of forty years long changes in coastal land use and land cover of the Yellow Sea: the gains or losses in ecosystem services. Environ. Pollut. 241, 74–84.
Yoon, S.J., Hong, S., Kwon, B.-O., Ryu, J., Lee, C.-H., Nam, J., Khim, J.S., 2017. Distributions of persistent organic contaminants in sediments and their potential impact on macrobenthic faunal community of the Geum River estuary and Saemangeum coast, Korea. Chemosphere 173, 216–226.
Yoon, S.J., Hong, S., Kim, S., Lee, J., Kim, T., Kim, B., Kwon, B.-O., Zhou, Y., Shi, B., Liu, P., 2020. Large-scale monitoring and ecological risk assessment of persistent toxic substances in riverine, estuarine, and coastal sediments of the Yellow and Bohai seas. Environ. Int. 137, 105517.
Yoon, S.J., Hong, S., Lee, J., Lee, J., Kim, Y., Lee, M.J., Ryu, J., Choi, K., Kwon, B.-O., Hu, W., Wang, T., Khim, J.S., 2023. Historical trends of traditional, emerging, and halogenated polycyclic aromatic hydrocarbons recorded in core sediments from the coastal areas of the Yellow and Bohai seas. Environ. Int. 178, 108037.
Yoon, S.J., Lee, J., Kim, Y., Kwon, B.-O., Hu, W., Wang, T., Hong, S., Khim, J.S., 2025. Historical trends of polychlorinated biphenyls and alkylphenols recorded in core sediments from the intertidal areas of the Yellow Sea and Bohai Sea. Mar. Pollut. Bull. 216, 118043.
Yu, X., Zhang, Z., Feng, A., Gu, D., Zhang, R., Xia, P., Yan, W., Zhou, X., 2020. Recent history of metal contamination in the Fangcheng Bay (Beibu Gulf, South China) utilizing spatially-distributed sediment cores: responding to local urbanization and industrialization. Mar. Pollut. Bull. 158, 111418.
Zhang, P., Song, J., Yuan, H., 2009. Persistent organic pollutant residues in the sediments and mollusks from the Bohai Sea coastal areas, North China: an overview. Environ. Int. 35, 632–646.
Zhang, R., Zhang, F., Zhang, T.-C., 2013. Sedimentary records of PAHs in a sediment core from tidal flat of Haizhou Bay, China. Sci. Total Environ. 450, 280–288.
Zhou, J., Qiao, Y., Wang, W., Leng, E., Huang, J., Yu, Y., Xu, M., 2016. Formation of styrene monomer, dimer and trimer in the primary volatiles produced from polystyrene pyrolysis in a wire-mesh reactor. Fuel 182, 333–339.
Zhu, H., Zhang, K., Sun, H., Wang, F., Yao, Y., 2017. Spatial and temporal distributions of hexabromocyclododecanes in the vicinity of an expanded polystyrene material manufacturing plant in Tianjin. China. Environ. Pollut. 222, 338–347.