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Invisible threats from typical endocrine disrupting compounds in estuarine environments caused by continuing seawater incursion: *in-situ* evidence of bio-geochemical processes captured by diffusive gradients in thin films

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1 **Abstract:** Continued seawater incursion significantly affects the fate of pollutants in
2 coastal estuaries, yet understanding of the *in-situ* behavior of endocrine-disrupting
3 compounds (EDCs) in these areas remains limited. The distribution, transport and
4 microbial response of two model EDCs, bisphenol A (BPA) and nonylphenol (NP), in
5 three estuarine zones of slight (SZ), moderate (MZ) and complete (CZ) seawater
6 incursion were investigated *in-situ*. Results showed seawater incursion reshaped the
7 environmental gradients of the coastal estuaries on a spatial scale. Varying salinity
8 gradient and tidal hydrodynamic conditions altered the dependence of EDCs on organic
9 carbon, and promoted the release of accumulated EDCs from estuarine sediments
10 resulting in the lowest residues of BPA ($2.74 \pm 0.76 \mu\text{g/kg}$) and NP ($10.25 \pm 5.86 \mu\text{g/kg}$)
11 in the MZ. The resupply potential of BPA ($R = 0.171 \pm 0.058$) and NP ($R = 0.107 \pm$
12 0.015) from sediment to porewater was significantly higher in the SZ than in other
13 zones ($p < 0.001$), due to both higher contaminant accumulation in this zone and
14 inhibited resupply in MZ and CZ caused by seawater incursion. Furthermore, seawater
15 incursion significantly reduced the microbial community diversity in the CZ ($p < 0.001$),
16 being dominated by *Vibrio* ($67.00 \pm 1.13\%$), and accordingly weakened the ability to
17 transform organic matter in this region. Based on predicted sea level rise and the
18 transport characteristics of EDCs under increased seawater incursion, it is estimated
19 that the cumulative additional release of BPA and NP in the estuary will reach 1.8 and
20 1.5 tons by 2100, respectively. In order to mitigate the risk of additional estuarine EDCs
21 release due to seawater incursion, increasing vegetation cover, strict monitoring, and

climate policy interventions may be effective strategies.

Keywords: Seawater incursion, estuarine sediment, endocrine disrupting compounds, *In-situ*, microbial communities.

1. Introduction

Endocrine-disrupting compounds (EDCs) are examples of refractory organic micropollutants distributed widely in the environment and exhibiting a variety of complex and concerning characteristics (Yan et al., 2022). For example, EDCs can cause irreversible effects on the reproductive and development systems of animal life (Yuan et al., 2017a), and annual productivity loss due to EDC exposure is estimated to cost over US\$40 billion (Zeng et al., 2024). Bisphenol A (BPA) and nonylphenol (NP) are typical EDCs extensively used as raw materials in the production of consumer products such as surfactants, plastics and food containers (Xiao et al., 2023); both have been listed as priority pollutants by the European Union (EU) (Wang et al., 2016). The annual consumption of NP ranges between 25 and 50 kilotons in the EU (Chen et al., 2019), and global production for BPA increases from 2.8 million t·year⁻¹ in 2002 to more than 8 million t·year⁻¹ in 2024 (Paolella et al., 2024). As a result, large amounts of BPA and NP (BPA, in particular, more than 2,000 t·year⁻¹) entered the aquatic environment through wastewater discharge and rainfall runoff to waterbodies (Lee et al., 2020). Through these vectors, BPA and NP may spread over huge distances in the aqueous environment in either dissolved or solid forms due to their hydrophobic, persistent, and bio-accumulative characteristics (Du et al., 2025). Subsequently, these

compounds pose a serious threat to aquatic organisms. Long-term exposure to environmentally relevant concentrations of NP (1.0 µg/L) and BPA (0.1 µg/L) has been shown to impair the immune system of fish (Lee et al., 2013; Qiu et al., 2016). Moreover, some studies have confirmed that simultaneous exposure to BPA and NP may lead to additive or synergistic toxicity (Wu et al., 2011; Huang et al., 2017, 2018). Additionally, BPA and NP can significantly alter the microbial community structure in depositional environments and reduce microbial diversity (Yuan et al., 2017b; Zoppini et al., 2020). Under aerobic environments, they can readily involve in microbial metabolic processes, potentially influencing the succession of microbial communities in sediments (De Weert et al., 2010; Bradley et al., 2016; Zoppini et al., 2018). Currently, BPA and NP are considered to be the main EDC pollutants in the marine environment, and present a continued challenge to the viability of marine species such as dolphins, cetaceans and pelagic fish, etc. (Luo et al., 2024).

Coastal estuaries are key channels for material cycling between terrestrial and marine environments, and are amongst the most fragile and economically important ecosystems on the planet (Kirwan and Megonigal, 2013). Estuaries are also important reservoirs in the retention, storage, and release of pollutants into the marine environment (Yuan et al., 2017a). Once released into estuaries, BPA and NP tend to partition preferentially into organic matter in the sediment (Lee et al., 2013). This is particularly evident for NP, which is subject to significant accumulation in sediments due to its high hydrophobicity ($\text{Log } K_{ow} = 5.76$) and strong soil/sediment sorption

capacity ($\text{Log } K_{oc} = 4.48\text{--}5.6$) (Lee et al., 2013). Although BPA is only moderately hydrophobic ($\text{Log } K_{ow} = 3.32$), it still accumulates extensively in sediments. This may be attributed to the presence of two benzene rings in BPA, which promote strong $\pi\text{--}\pi$ interactions with organic matter and increase its affinity for sedimentary organic carbon pore sites (Sun et al., 2010). As a result, BPA also exhibits a high soil/sediment adsorption coefficient ($\text{Log } K_{oc} = 2.5\text{--}3.18$). In addition, the salting-out effects further enhance the sorption of BPA and NP on marine sediments, resulting in their accumulation rates in marine sediments that are least 1,000 times higher than in other environmental compartments (Safakhah et al., 2020). Thus, with the continuous anthropogenic use and discharge of BPA and NP (Chen et al., 2019; Lee et al., 2020; Xiao et al., 2023), estuaries as recipients of terrestrial pollution will accumulate significant amounts of these contaminants (Chen et al., 2019), ultimately becoming important sinks and sources of BPA and NP in their own right (Omar et al., 2017). Indeed, several studies have revealed that BPA and NP contamination levels in estuarine sediments is extremely high. For example, surface sediments of Thane Creek in India contained 35.8 $\mu\text{g/kg}$ and 537.8 $\mu\text{g/kg}$ BPA and NP, respectively (Tiware et al., 2016), and, in the Pearl River Estuary in China, concentrations of BPA and NP in sediments reached 13.2 $\mu\text{g/kg}$ and 20.8 $\mu\text{g/kg}$, respectively (Diao et al., 2017).

Additionally, as sea levels rise due to global warming, the problem of seawater extension into estuaries is becoming an increasingly important global concern (Tang et al., 2020). By 2100, coastal estuaries could lose up to 30% of their area, threatening

85 approximately US\$27 trillion of ecological economic activity ([Schuerch et al., 2018](#);
86 [Saintilan et al., 2022](#)). At the same time, seawater incursion will also alter
87 biogeochemical processes in estuarine sediments. When seawater incursion occurs,
88 sediments are frequently washed away, and strong tidal hydrodynamics result in
89 continuous re-release of legacy pollutants accumulated in estuarine sediments,
90 enhancing their mobility and bioavailability ([Fetters et al., 2016](#)). Furthermore, with
91 tidal pumping and advective transport of oxygenated seawater, the oxygenation
92 capacity of sediment is enhanced ([Huettel et al., 2018](#)) which enhances aerobic
93 microorganism activity in the sediment ([Anschutz et al., 2009](#)). Once seawater
94 incursion is completed, sediments may be permanently submerged by seawater, and the
95 evolution of microbial communities in the sedimentary aquifer will be achieved. Thus,
96 nitrogen reduction process (such as denitrification and anammox) will also increase
97 ([Xiong et al., 2023](#)). In this manner, the intensification of seawater incursion will affect
98 pollutants in estuarine zones to different degrees, hence they will exhibit different
99 environmental and geochemical behaviors. This can lead to a number of unforeseen
100 environmental risks and consequences. Although previous studies have focused on the
101 fate of some inorganic pollutants such as nitrogen, phosphorus and heavy metals in
102 estuaries under the influence of seawater incursion ([Chakraborty et al., 2019](#); [Zhang et](#)
103 [al., 2023a](#)). There is a lack of research on the migration and transformation behavior of
104 refractory organic pollutants such as EDCs in estuarine areas under the influence of
105 seawater incursion, especially at the sediment-water interface (SWI) in areas affected

by different degrees of seawater incursion. In addition, to the best of our knowledge, most studies still focus on investigating the concentration distribution of pollutants and assessing ecological risk by comparison them with some environmental quality standards (Chiriac et al., 2021; Zainuddin et al., 2023). However, fewer researches have been conducted on the prediction of pollutant releases fluxes at SWI under dynamically changing environmental conditions, even though this is directly related to the bioavailability of pollutants. The exploration of these problems is considered important for the ecological and environmental security of the estuary. Therefore, further analysis of the distribution, diffusion and transformation of EDCs in the estuarine environment under seawater incursion is of significant scientific value for understanding the comprehensive effects of seawater incursion and EDC input on estuarine ecosystems.

Sediments, unlike water bodies, are heterogeneous and more likely to host various biogeochemical processes at the microscale (Lin and Pan, 2023). Thus, small changes in the sedimentary environment can lead to significant changes in the form and concentration of pollutants in sediments, especially at the SWI (Liu et al., 2022a). A precise study of pollutant exchange mechanisms at the SWI will be helpful in understanding the source-sink relationship between sediments and estuarine waters under the influence of seawater incursion, and assessing the bioavailability of pollutants (Chen et al., 2022). However, recent studies on the pollution process and the release flux of pollutants at the SWI usually use active sampling methods (i.e., "*ex-situ*" methods) (Puttonen et al., 2024). These methods disturb the *in-situ* sediment

environment, disrupt the equilibrium of contaminant distribution in the sedimentary environment, and fail to capture the dynamic information on pollution under changing environmental conditions (Li et al., 2023a). These disadvantages can cause considerable analytical error, and result in major deviations between experimental observations and the real world situation, which in turn affects our understanding of pollution behavior and risk assessment (Gao et al., 2016). The Diffusive Gradients in Thin film technique (DGT) is an innovative passive sampling method, and can be used to determine the diffusion and exchange of pollutants at the SWI based on Fick's first Law (Arsic et al., 2018). More importantly, DGT can capture the labile fractions of pollutants (including the free fraction and the fraction that can be released from the environment medium), thus providing a more accurate representation of the bioavailability of pollutants than total concentration measurements (Liang et al., 2023). To date, DGT has been used to obtain information on the *in-situ* release of organic micropollutants such as pesticides and antibiotics in sediments (Chen et al., 2014; Li et al., 2021). Therefore, the DGT technique could help us obtain *in-situ* dynamic exchange processes and bioavailability characteristics of EDCs at the estuarine sediment-water interface (ESWI), especially under changing environmental conditions, e.g., as affected by seawater incursion.

In this study, active sampling and DGT were used in combination to investigate the *in-situ* fate of typical EDCs such as BPA and NP in the ESWI under the influence of seawater incursion. Sediment cores from slight (SZ), moderate (MZ) and complete (CZ)

estuarine zones of seawater incursion were collected. The interfacial distribution and diffusion of both BPA and NP, as well as the potential correlations between microbial communities and these two EDCs at the three different estuarine zones were analyzed. In addition, a release model of estuarine EDCs due to seawater incursion caused by sea level rise was also established. Thus, the main objectives of our study were as follows: 1) to clarify the impact of seawater incursion on the *in-situ* distribution of BPA and NP in estuarine sediments; 2) to investigate the impact of seawater incursion on the diffusion characteristics of BPA and NP in sediments; 3) to elucidate the influence of seawater incursion on microbial community succession in sediments and microbial responses to the fate of BPA and NP; and 4) to further assess the possible risk of EDC release from estuarine sediments due to sea level rise.

2. Materials and Methods

2.1 Sampling area, DGT field deployment, and sample collection

Frequent seawater incursion into estuaries due to global sea level rise has resulted in threats and losses to groundwater, agriculture, drinking water safety, and coastal bird habitats (Mondal et al., 2023; Lee et al., 2024; Su et al., 2024; van de Pol et al., 2024). The latest research also confirms that seawater incursion is a prominent problem in China's coastal estuarine areas (Wang et al., 2022). Amongst these, the Liaohe estuary in northern China, a semi-enclosed estuary suffering from relatively poor diffusion conditions, has experienced serious coastal vegetation degradation and soil salinization problems due to seawater incursion (Ma et al., 2019; Guo et al., 2022). Thus, the Liaohe

estuary was chosen to study the fate of organic pollutants in estuarine areas caused by seawater incursion. The sample collection and DGT deployment were carried out at Red Beach on the Liaohe estuary (Fig. S1 and S2; 120°44'6.67"E, 40°33'41.66"N). The discharge of runoff from the Liaohe estuary into the sea has long been regulated by the upstream sluice tidal gate, the average runoff is 1.2-1.3 billion·m³ monthly (Hu et al., 2023; Dong et al., 2024). Red Beach is located in the wetland reserve of Huludao City in China, with area 600 hectares, most of the area is bare, flat and tidal. Tidal action, which is common in coastal areas, plays a crucial role in facilitating the diffusion and transport of sediment particles, nutrients, and pollutants across the tidal flats (Gu et al., 2024). According to the (Liaoning Provincial Marine Ecological Early Warning Monitoring Bulletin (2023), 2024), the tide at Red Beach is in a semidiurnal state with a tidal range of 2–3 m. The average annual temperature in the region is 10°C and the average annual precipitation and evaporation are 600 mm and 1500 mm, respectively. Estuarine pollution mainly comes from terrestrial runoff from industrial and agricultural activities in the upper reaches of tributary rivers (Guo et al., 2022). Since Red Beach is located in a nature reserve there are no significant urban or industrial developments nearby, which avoids potential human disturbance. Hence, the sedimentary environment in Red Beach may be deemed as relatively stable and well preserved.

Based on the environmental conditions and seasonal characteristics of Red Beach, the sampling activities was selected to be conducted during the flat-water period from

late July to the end of August 2024. During this period, the upstream sluice was closed, causing the river flow to be generally less than 0.3 billion·m³ monthly, resulting in very low runoff. And seawater incursion typically occurred during low runoff periods, extending approximately 5-10 km upstream of the estuary (Hu et al., 2023; Dong et al., 2024). Furthermore, this period is characterized by moderate evaporation (average 600 mm) and relatively active microbiological activity (average temperature 19.5°C) (Huludao Environmental Quality Report, 2024), which facilitates sample collection, DGT placement and the conduct of studies on seawater incursion (Li et al., 2023b; Hlaing et al., 2024; Gao et al., 2025). In order to more objectively assess the impact of seawater incursion on the release of EDCs, three sampling campaigns were conducted, with each sampling event lasting one week. During each sampling period, samples were collected from three zones (SZ, MZ, and CZ) along the direction of seawater incursion to reveal the impact of the degree of seawater incursion. DGT was deployed simultaneously to represent the characteristics of slight, moderate and complete incursion (Fig. S1). According to field survey, under tidal influence, SZ and MZ were inundated by seawater for between 3 and 5 h, and 12 and 15 h per day, respectively, while CZ was permanently inundated by seawater. The distance between each sampling zone was about 500 m. Three sites were selected within each sampling zone to eliminate spatial heterogeneity, and the distance between these sites was about 30 m. Subsequently, one DGT sediment probe (DGT-HLB) for BPA and NP, and one DGT sediment probe (DGT-Chelex100/AgI) for iron (Fe), manganese (Mn), and dissolved

sulfur (S) were deployed *in-situ* at each site at ebb tide. Each type of DGT was deployed in three parallel replicates (Fig. S2). The details of the above DGT composition are described in Text S2. When deploying DGT probes, the tidal flats of SZ and MZ were exposed, and the water depths of the CZ was between 0.5 and 1.0 m. In the SZ and MZ, the entire window area of the DGT probes was slowly inserted vertically and directly into the sediment bed by 15 cm. In the CZ, the DGT probes were attached in the bottom of perforated plexiglass profiles (thickness: 3 cm, width: 3 cm, length: 120 cm). Then, in each site, six separate profiles with attached probes were slowly inserted into the sediment bed by 15 cm, and protected by six stone supports in each site (Fig. S3). All probes in the three zones were fully inserted for one week, experiencing a total of about 14 tidal cycles. At the last ebb tide the DGT probes were recovered, and were then washed with Milli-Q (MQ) water and placed into sealed bags. At the beginning and end of each sampling period, four sediment cores (~25 cm) adjacent to the DGT probes in each zone were collected from the three sites using a 10-cm diameter PVC sampling tube. No bioturbation was observed in all collected sediment cores, and the SWI of the obtained sediment cores was clear. An additional sediment sample was also collected at each station using a small shovel to determine the effect of matrix on extraction and concentration recoveries. Finally, the sediment cores, recovered DGT probes, and surface sediments were immediately stored in sample boxes with ice packs prior to transportation to the laboratory.

2.2 Sample treatment and analysis of physicochemical properties

In the laboratory, the sediment cores were carefully divided into 1-cm portions and centrifuged at 3,000 rpm for 40 min to obtain porewater. After centrifugation, the residual sediments were freeze-dried, ground, and sieved (100-mesh) for subsequent analysis. BPA and NP analyses were performed on three sediment cores collected at the beginning and end of each sampling event, and the mean average was used as result. The remaining sediment core was used for sediment environmental factor analysis. Total organic carbon (TOC) in sediment was measured using the combustion oxidation-titration method (Du et al., 2025). The salinity and pH values of each sediment slice were determined using a water quality tester (WTW GmbH, Germany). Before measurement, dry sediments were mixed with MQ water at a ratio of 1:5 for salinity and 1:2.5 for pH (Yu et al., 2022).

2.3 Extraction of BPA and NP from Samples

2.3.1 DGT

The details of DGT samples' extraction were described in our previous study (Du et al., 2024, 2025). Briefly, in the laboratory, the DGT gel was cut along the window area using a Teflon blade. For DGT-HLB, the binding gel was divided into 1 cm segments, and each segment was transferred to 10 mL glass centrifuge tube. Then, 8 mL of acetone was added to the tube, sonicated for 40 min, and centrifuged at 3500 rpm for 10 min to extract the target substances from the binding gel. The same procedure was repeated twice for the residual gel, then the extracts were combined and evaporated to dryness under a gentle flow of nitrogen gas. Subsequently, 1 mL of methanol was added to the

dry residue to redissolve the target compound. The internal standard (BPA-d16 and NP-d4) was introduced into the tube, vortexed, and then filtered using a 0.22- μ m PTFE filter (Aladdin, Shanghai, China) into 2 mL LC vials, which were stored at -20°C until LC-MS analysis. For DGT-Chelex100/AgI, the AgI diffusion gel and Chelex100 binding gel were together peeled from the probe. Next, the AgI gel was scanned using a flat-bed scanner (HP 3100) for analysis of dissolved S. The Chelex100 gel was sliced into 5 mm segments, and each segment was placed in a glass centrifuge tube containing 0.4 mL of 1 M HNO₃ for 24 hours to elute, then stored carefully prior to ICP-MS analysis.

2.3.2 Porewater

Porewater extraction methods were based on our previous research with some modifications, and procedures are described in Supporting Information (SI), [Fig. S4](#). Briefly, 1 mL of porewater was diluted to 150 mL using MQ water to reduce matrix concentration and minimize error. The diluted solution was filtered through a glass fiber membrane (47 mm, Aladdin, Shanghai, China) and then concentrated by solid-phase extraction (SPE) as described in the SI ([Text S3](#)). Each sample was tested in triplicate. The concentration of BPA and NP extracted from porewater is denoted as C_d .

2.3.3 Sediments

Sediment extraction methods were based on our previous research with some modifications, and procedures are described in the SI, [Fig. S4](#). Briefly, 1.0 g of sediment was transferred to a 10-mL glass tube with 8 mL acetone, sonicated for 40 min, and

centrifuged at 3500 rpm for 10 min to extract BPA and NP. This was repeated a second time, and the extracted supernatants combined and evaporated to 1-2 mL under a gentle stream of high-purity nitrogen, diluted to 150 mL with MQ water (acetone content <1%, V/V), prior to further processing. The diluted extracts were filtered (47 mm glass fiber membrane, Aladdin, Shanghai, China) and purified by SPE (Text S3). Each sample was tested in triplicate. The concentration of BPA and NP of the sediment is denoted C_s .

2.4 Diffusion analysis of BPA and NP in sediment

The binding layer in the DGT acts as a sink for compounds in the sediment porewater/water, where an induced flux from the sediment/water passes through the diffusive layer and is bound in the binding layer (Ji et al., 2022). Thus, the diffusion characteristics of pollutants at the SWI may be monitored *in situ* by DGT. The ratio R , an index reflecting depletion of sediment porewater concentration to the DGT interface, was introduced to characterize the magnitude of this diffusion and resupply. Typically, higher values of R indicate a stronger capacity of resupply from sediment to porewater (Du et al., 2025). And three scenarios of the resupply capability were determined based on the R values. (i) $R > 0.95$, the target substances are fully resupplied from sediment to porewater; (ii) $0.1 < R < 0.95$, the target substances are partially resupplied from sediment to porewater; (iii) $R < 0.1$, there is diffusion only, with very limited resupply from the sediment to porewater. Moreover, the values of R in sediments can be impacted by various environmental factors, such as the labile pool size of target substances and particle concentration (P_c , the ratio of the mass of total sediment solid particles to the

volume of total porewater) (Li et al., 2021). In this study, the diffusion process of BPA and NP in sediments was quantified using R as follows (Chen et al., 2014):

$$R = \frac{C_{DGT}}{C_d} \quad (1)$$

where C_{DGT} is the time-weighted average concentration of BPA and NP in DGT (Eq. 2):

$$C_{DGT} = \frac{M\Delta g}{DA t} \quad (2)$$

where M is the mass of BPA and NP bound in the DGT (Text S4), Δg is the thickness of the diffusion layer in mm, D is the diffusion coefficient of BPA and NP in $\text{cm}^2 \cdot \text{s}^{-1}$, A is the exposed area of DGT in cm^2 , and t is the deployment time, s.

2.5 Characterization of microbial community in sediment

To further evaluate the impact of seawater incursion on the microbial communities in sediments at different depths, the sediment cores were classified into four depth intervals, as referred to by a number of previous studies (Zhang et al., 2021; Gong et al., 2022; Wang et al., 2024a): Part 1 (0 ~ -4 cm), Part 2 (-5 ~ -8 cm), Part 3 (-9 ~ -12 cm), and Part 4 (-13 ~ -15 cm). Three replicate sediment samples (1.0 g, wet weight) were collected from each part and preserved at $-80\text{ }^{\circ}\text{C}$ prior to high-throughput sequencing. Genomic DNA was extracted from the sediment using OMEGA Soil DNA Kit (M5635-02) (Omega Bio-Tek, Norcross, GA, USA). PCR amplification of the bacterial 16S rRNA gene V3–V4 region was performed using the forward primer 338F (5'-ACTCCTACGGGAGGCAGCA-3'). Libraries were constructed using the Illumina TruSeq Nano DNA LT Library Prep Kit. The constructed libraries were quantified using a Quant-iT PicoGreen dsDNA Assay Kit, and high-throughput sequencing was

performed on an Illumina NovaSeq-6000. Taxonomic assignment of amplicon sequence variants (ASVs) was conducted using a Silva 138.1 database. The raw 16S rRNA gene reads were deposited on the NCBI Sequence Read Archive database with the accession number PRJNA1201922.

2.6 Construction of predictive models for BPA and NP release with sea level rise

A simple release prediction model of BPA and NP from sediment to seawater during the 2024-2100 period under different sea level rise scenarios was developed, by referring to previous methods with minor modifications (Stigebrandt et al., 2014; Lu et al., 2021; He et al., 2023). The derivation and details of model construction are described in SI (Text S5). The model was constructed to predict cumulative additional releases of BPA and NP from the perspective of the ESWI considering both the loss of coastal wetland area and the net release fluxes of BPA and NP. Briefly, (i) the DGT-labile concentration gradients of BPA and NP in the vicinity of the SWI was used to estimate the apparent diffusion flux of BPA and NP across the SWI. It was calculated as the sum of the diffusion flux of BPA and NP from the overlying water to the SWI, and from the sediments to the SWI; (ii) the function of additional estuarine area loss and seawater incursion time was determined by referring to previous studies (Mengel et al., 2016); (iii) the cumulative additional release over time was determined by multiplying the integral of the function of estuarine area and seawater incursion time with the release flux. Based on the above steps, Equation 3 was established to estimate the cumulative increase in BPA and NP over time starting from 2024 to the

corresponding year of different sea level rise scenarios under continuous seawater incursion:

$$P = [-D_w(\frac{\delta C_{DGT}}{\delta x_w})_{(x=0)} - \phi D_s(\frac{\delta C_{DGT}}{\delta x_s})_{(x=0)}] \cdot \int_{2024}^T f(T) \quad (3)$$

where P is the projected increase in release of BPA and NP over time in tones, D_w and D_s are the diffusion coefficients of BPA and NP ($\text{cm}^2 \cdot \text{s}^{-1}$), respectively, in the overlying water and sediment, $(\frac{\delta C_{DGT}}{\delta x_w})_{(x=0)}$ and $(\frac{\delta C_{DGT}}{\delta x_s})_{(x=0)}$ are the DGT-labile concentration gradients in the overlying water and sediment in $\text{ng} \cdot \text{mL}^{-1} \cdot \text{mm}^{-1}$, respectively. A distance of 1 cm from the SWI to the sediment was used to fit the gradient, ϕ is the porosity of the sediment, T is time of seawater incursion in years, and $f(T)$ is defined as the loss of coastal estuarine area as a function of time.

2.7 Data analysis and quality assurance

BPA and NP were analyzed by LC-MS with details in [Text S6](#). Briefly, they were identified using an Agilent 1290 Infinity/6460 LC/QQQ MS operated in negative multiple reaction monitoring (MRM) mode. LC separation was achieved with a Poroshell 120 EC-C18 (1000 bar, 3 mm×150 mm, 2.7 μm , Agilent). The mobile phase consisted of phase A (Milli-Q water with 0.025% ammonia) and phase B (methanol with 0.025% ammonia) with a flow rate of 0.4 mL/min. The gradient elution program was 20% B kept to 1 min, then increased to 100% B at 3 min and kept constant for 4 min. Followed by returning to the initial conditions within 1 min. Finally, the column was re-equilibrated for 2 min. The column temperature was kept at 40°C, and the injection volume was 20 μL . The MS parameters were given in [Table S4](#).

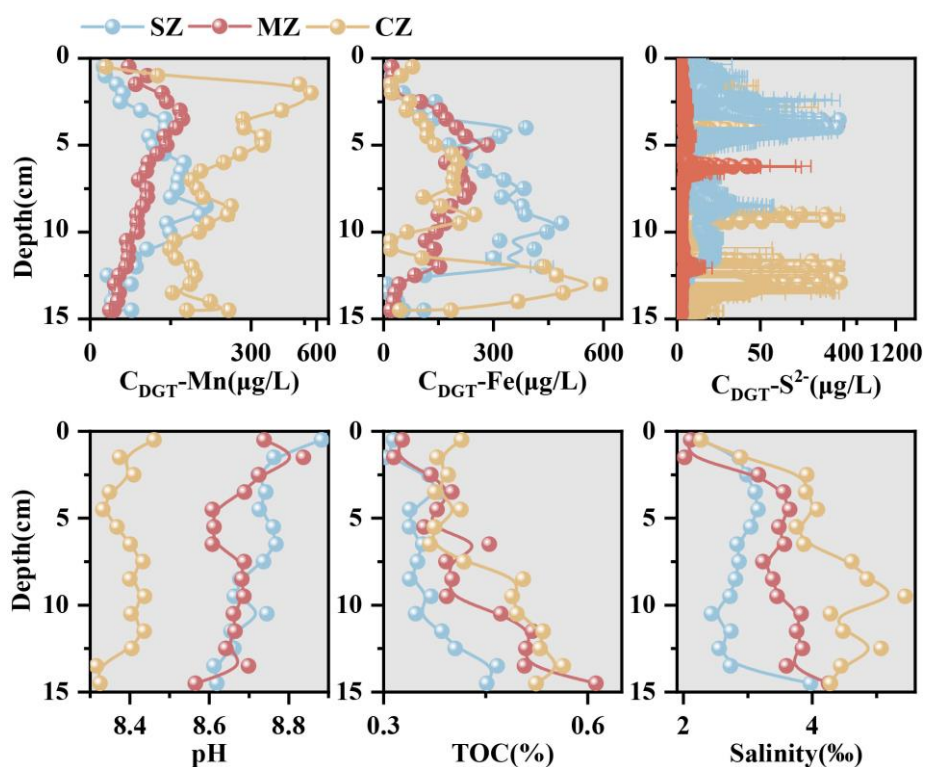
Fe and Mn in DGT were analyzed by ICP-MS (Optima8300, PerkinElmer). Dissolved S concentration was obtained through Computer-Imaging Densitometry (Text S7). Details of quality assurance/quality control are available in SI (Text S8). Variations in the profile distribution of BPA, NP, and environmental factors were plotted using OriginPro 2024. Data obtained from DGT and the active sampling method were checked for normality and homogeneity using Levene's test. Since the data did not satisfy a normal distribution and did not show homogeneous variances, the non-parametric Kruskal-Wallis test was used to check for significant differences (significance at $p < 0.05$) between BPA and NP profiles, and between SZ, MZ and CZ (Chen et al., 2024). Co-occurrence networks were constructed by the top 500 ASVs of SZ, MZ and CZ sediments using Cytoscape and Gephi (Chen et al., 2024).

3. Results

3.1 Physicochemical parameters of sediments in different zones

Fig.1 shows the variation of physicochemical parameters in the sediment profiles of SZ, MZ and CZ. DGT measurements reflect the labile fractions of elements in the sediment. The results confirm the lowest labile fractions of Mn, Fe and S^{2-} in the MZ (Mn: 96.33 ± 2.21 $\mu\text{g/L}$; Fe: 129.56 ± 2.57 $\mu\text{g/L}$; S^{2-} : 3.92 ± 3.19 mg/L). Typically, the accumulation of dissolved Fe, dissolved Mn and S in the sediments indicate a successive reduction of Fe(III), Mn(III/IV) and sulfate occurred with increasing oxygen depletion (Pan et al., 2021). The profiles of Fe, Mn and S^{2-} in the sediments indicated anoxic conditions in the SZ and CZ, and relatively aerobic conditions in the MZ. The

379 pH values in the SZ, MZ and CZ sediment profiles all decreased with increasing depth,
 380 ranging from 8.61 to 8.88, 8.57 to 8.84 and 8.32 to 8.46, respectively. TOC and salinity
 381 in the SZ, MZ and CZ sediment profiles all increased with increasing depth; TOC in
 382 the range of 0.34 - 0.52 %, 0.32 - 0.61 % and 0.37 - 0.56 %, respectively, and salinity
 383 in the range of 2.27 - 3.98 ‰, 2.02 - 4.26 ‰ and 2.28 - 5.45 ‰, respectively. In short,
 384 the extent of seawater incursion resulted in significantly different environmental factors
 385 in the SZ, MZ and CZ sediments (Fig. S5). Greater salinity (4.14 ± 0.77 ‰) and TOC
 386 (0.45 ± 0.07 %) accumulated in CZ sediments, whereas the pH of SZ sediments was
 387 higher (8.72 ± 0.07).



388
 389 **Fig. 1** Physicochemical parameters in the sediment profiles of SZ, MZ and CZ.

390 **3.2 Variations in BPA and NP concentration in sediment from different zones**

391 **Fig. 2** shows the profile variations of BPA and NP in the sediment, porewater and

392 DGT of the SZ, MZ and CZ. In the sediment profile, NP concentrations in the SZ, MZ
393 and CZ decreased with increasing depth, ranging from 44.44 to 11.45 $\mu\text{g/kg}$, 24.85 to
394 4.99 $\mu\text{g/kg}$, and 36.88 to 7.23 $\mu\text{g/kg}$, respectively. BPA concentrations fluctuated with
395 depth in the SZ, MZ and CZ, with average concentrations of $7.09 \pm 2.46 \mu\text{g/kg}$, $2.74 \pm$
396 $0.76 \mu\text{g/kg}$, and $3.35 \pm 0.81 \mu\text{g/kg}$, respectively. Overall, NP concentrations in sediment
397 were higher than BPA concentrations in all three zones, which was attributed to higher
398 hydrophobicity ($\log K_{ow}=5.76$) and dissociation constant ($pK_a=10.3$) of NP, which
399 tended to accumulate on solid particles ($K_d= 3.07 \sim 19.49 \text{ g/mL}$, [Table S2](#)). For the
400 porewater and DGT profiles, BPA and NP concentrations fluctuated with depth in the
401 SZ, MZ and CZ. In these three zones, the average DGT concentration of BPA was 0.45
402 $\pm 0.12 \mu\text{g/L}$, $0.11 \pm 0.03 \mu\text{g/L}$, and $0.14 \pm 0.04 \mu\text{g/L}$, respectively, and in porewater it
403 was $2.71 \pm 0.49 \mu\text{g/L}$, $1.50 \pm 0.29 \mu\text{g/L}$ and $1.81 \pm 0.28 \mu\text{g/L}$, respectively. The average
404 concentrations of NP in DGT were $0.23 \pm 0.03 \mu\text{g/L}$, $0.08 \pm 0.02 \mu\text{g/L}$, and 0.10 ± 0.02
405 $\mu\text{g/L}$, respectively, and in porewater $2.21 \pm 0.37 \mu\text{g/L}$, $1.48 \pm 0.33 \mu\text{g/L}$, and $1.73 \pm$
406 $0.30 \mu\text{g/L}$, respectively. It was worth noting that BPA concentrations in porewater and
407 DGT were slightly higher than those of NP. This may be attributed to the higher
408 accumulation of NP in sediments, which resulted in a larger pool of releasable NP for
409 diffusion from sediments into porewater, leading to a residual NP concentrations in
410 porewater close to that of BPA, similar to the results in other studies ([Zhang et al., 2009,](#)
411 [2011](#)). In addition, this result was further supported by the distribution coefficient (K_d),
412 which ranged from 3.07 to 19.48 g/mL for NP across the three zones, while for BPA,

the K_d values ranged from 0.71 to 4.53 g/mL, with an average value approximately 4.19 times lower than that of NP (Table S2).

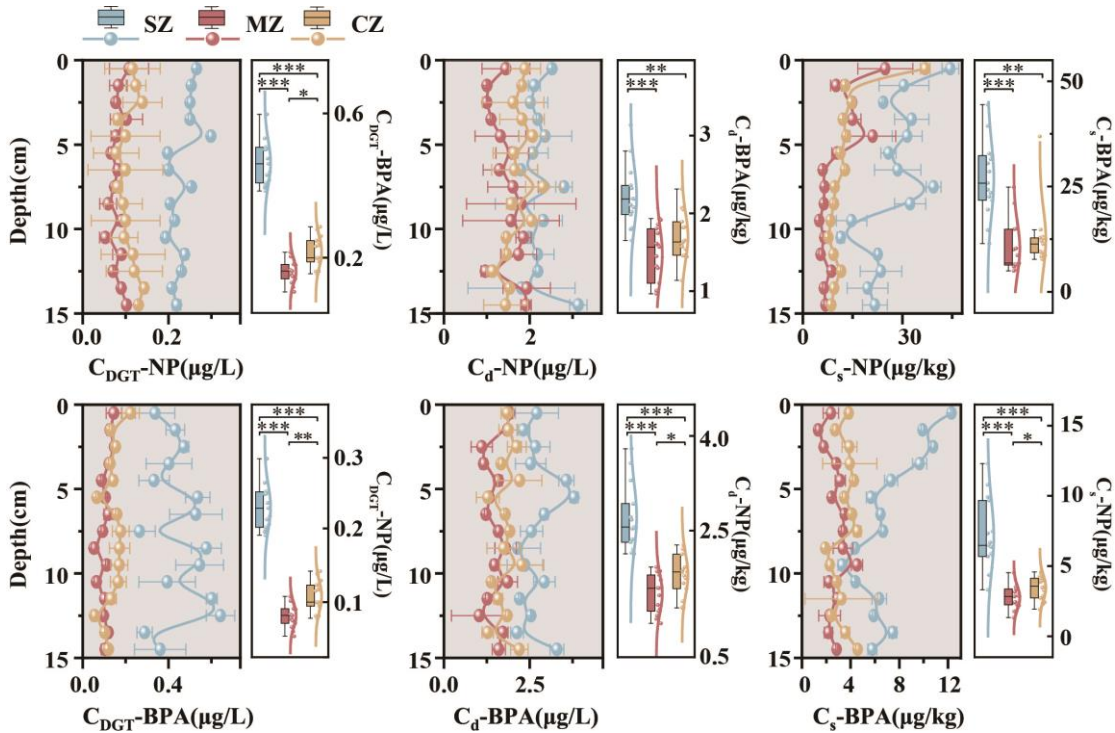


Fig.2 Variation of NP and BPA concentrations in DGT, porewater and sediment profiles of the SZ, MZ and CZ. Here, “*”, “**” and “***” indicate $p < 0.05$, 0.01 and 0.001, respectively.

3.3 BPA and NP diffusion in sediment with depth from different zones

Fig. S6 shows the profile variations for the R values with depth in the SZ, MZ and CZ. For BPA, the R values ranged from 0.093 to 0.272 in the SZ, 0.029 to 0.134 in the MZ, and 0.029 to 0.121 in the CZ, with mean values of 0.171 ± 0.058 , 0.075 ± 0.026 and 0.080 ± 0.024 , respectively. For NP, the R values ranged from 0.070 to 0.127 in the SZ, 0.028 to 0.092 in the MZ, and 0.035 to 0.106 in the CZ, with mean values of 0.107 ± 0.015 , 0.059 ± 0.018 and 0.062 ± 0.020 , respectively. The R for BPA was higher than NP in all three zones due to the higher hydrophilicity and lower pK_a (9.6) of BPA.

Furthermore, in the SZ, *R* values of BPA and NP were mostly greater than 0.1, indicating there was a partial resupply from sediment to porewater, but that this resupply was insufficient to maintain initial porewater concentrations. However, in the MZ and the CZ, *R* values of BPA and NP were mostly below 0.1, indicating limited resupply from sediment to porewater (Chen et al., 2014).

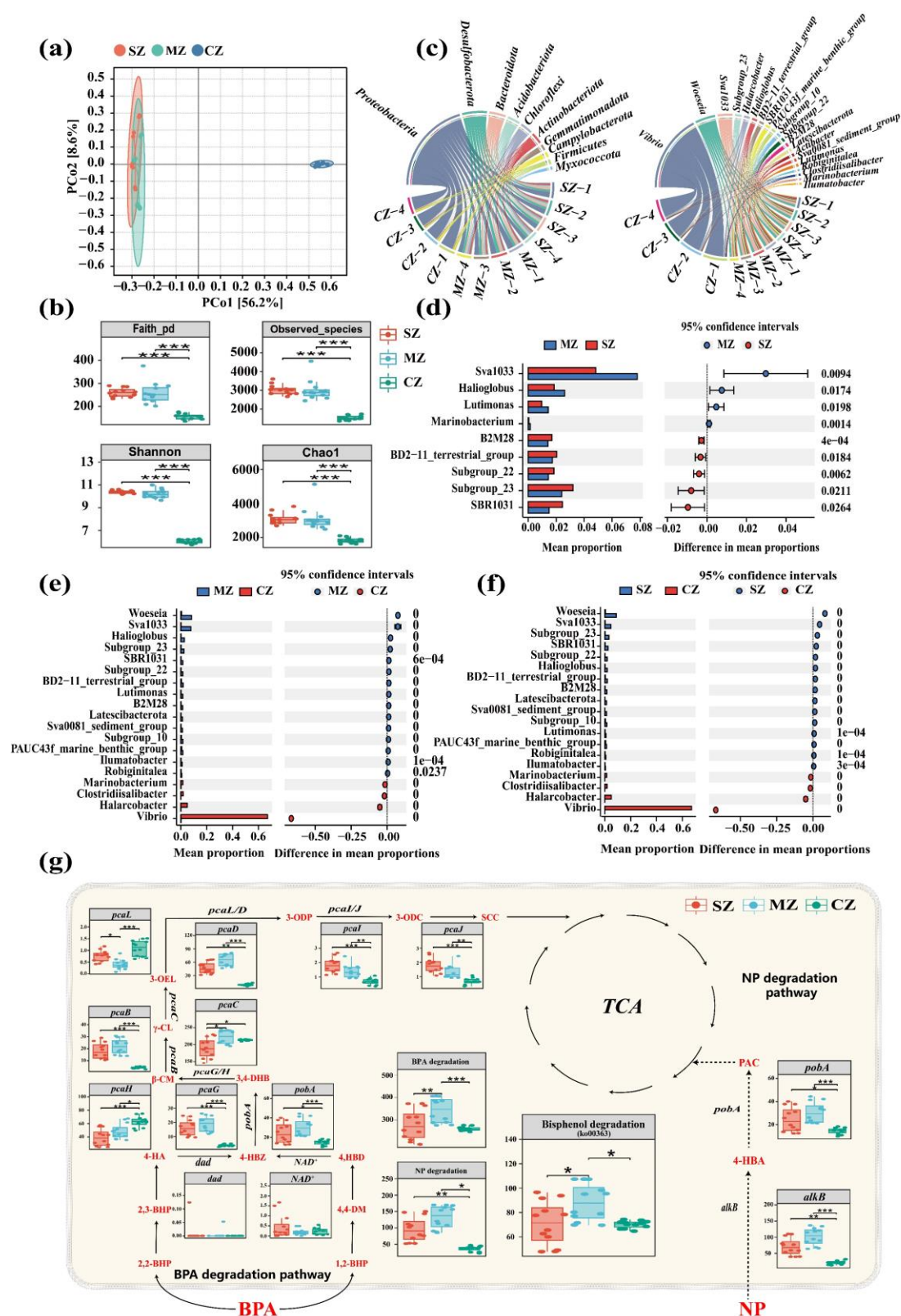
3.4 Differences in microbial communities of the sediments from the three zones

The bacterial community composition of the CZ was shown to be different to the SZ and the MZ based on the Bray-Curtis, the CZ samples were distinctly separated from the SZ and the MZ on the pCo1 (Fig. 3a). Similarly, the Shannon and Chao1 indices for the CZ were significantly lower than for the SZ and the MZ ($p < 0.001$), indicating the diversity of the microbial community of the CZ was the lowest (Fig. 3b). *Proteobacteria* were the most dominant phyla in all zones: SZ $28.68 \pm 3.35\%$, MZ $31.07 \pm 4.18\%$, and CZ $76.34 \pm 0.63\%$. *Desulfobacterota* and *Bacteroidota* were also the main phyla in the SZ: $16.68 \pm 1.56\%$, and $9.84 \pm 1.56\%$, respectively, and the MZ: $18.69 \pm 2.68\%$, and $11.81 \pm 2.48\%$, respectively (Fig. 3c). Additionally, among the top 20 genera in the three zones (Fig. 3c and Fig. S7), *Woeseia* ($8.84 \pm 0.16\%$) and *Subgroup 23* ($3.19 \pm 0.92\%$) showed the highest relative abundance in the SZ, *Sva1033* ($7.80 \pm 2.9\%$) showed the highest relative abundance in the MZ, and *Vibrio* ($67.00 \pm 0.79\%$) and *Halarcobacter* ($5.02 \pm 1.80\%$) showed the highest relative abundance in the CZ. Based on statistical analysis of metagenomic profiles (Fig. 3d-f), among the top 20 genera in these three zones, *B2M28*, *BD2-11_terrestrial_group*, *Subgroup 23* and

448 *SBR1031* were significantly higher in the SZ ($p<0.05$). *Sva1033*, *Halioglobus* and
449 *Lutimonas* were significantly higher in the MZ ($p<0.05$), and *Vibrio*, *Halarcobacter*,
450 *Clostridiisalibacter* and *Marinobacterium* were significantly higher in the CZ
451 ($p<0.001$). Fig. 3g shows the BPA and NP potential biotransformation pathways and the
452 related functional gene abundances based on the KEGG pathway database and previous
453 studies (Yu et al., 2019; Wu et al., 2022). The relative abundance of most functional
454 genes for both BPA and NP (such as *alkB* and *pobA* etc.) were significantly higher in
455 the MZ ($p<0.05$). Furthermore, the relative abundance of the bisphenol degradation
456 (ko00363) pathway was also significantly higher in the MZ ($p<0.05$).

457 Microbial interactions in the three zones was also examined by establishing co-
458 occurrence networks of the SZ (Fig. S8a), MZ (Fig. S8b) and CZ (Fig. S8c).
459 *Proteobacteria* were shown to be the main phyla in the SZ, MZ and CZ. The MZ
460 network captured more nodes (175), edges (788), and higher average clustering
461 coefficient (0.468) than either the SZ network (170, 782, and 0.395) or the CZ network
462 (171, 788, and 0.393), indicating higher species co-occurrence association frequencies
463 in the MZ. Additionally, the positive/negative edge ratios in the SZ and CZ networks
464 were lower than in the MZ network, suggesting stronger species competition in the
465 oxygen-limited SZ and CZ (Lei et al., 2023). In contrast to the SZ and CZ networks,
466 the MZ network showed higher modular structures (Modularity>0.4) indicating non-
467 random MZ network structures which were unlikely to appear due to chance (Zhao et
468 al., 2023). Additionally, 14 key ASVs were identified as “connectors” in the MZ

469 network, compared to 10 in the SZ and CZ networks.



470
 471 **Fig. 3** Differences in microbial communities in the different estuarine zones: (a) the
 472 PCoA plot based on the Bray-Curtis, (b) boxplots showing the bacterial alpha diversity

indices in sediments, (c) Chord maps of the top 10 phyla and top 20 genera, in which -
1, -2, -3 and -4 correspond to part 1, part 2, part 3, part 4 of the sediment profiles in the
SZ and the MZ as mentioned in section 2.5. Differential analysis of the top 20 genera
between (d) the SZ and the MZ, (e) the MZ and the CZ and (f) the SZ and the CZ by
STAMP (Statistical Analysis of Metagenomic Profiles), and (g) possible bio-
transformation pathways of BPA and NP (the abbreviations of intermediates involved
in the pathways are listed in [Table S3](#)). “*”, “**” and “***” indicate $p < 0.05$, 0.01 and
0.001, respectively.

4. Discussion

4.1 Seawater incursion reshapes the environmental spatial gradient in estuarine sediments

In SZ sediments (slight seawater incursion), the environmental conditions were relatively stable and anoxic with O₂ supplied exclusively by diffusion across the sediment surface ([Fig. 4](#)) ([Huettel et al., 2018](#)). Here, the O₂ supply was insufficient to support bioremediation and respiration of heterotrophic microorganisms, resulting in rapid anoxia in surface layer sediments of the SZ ([Meng et al., 2022](#)). Subsequently, the intense hypoxic respiration of microorganisms in the SZ led to the consumption of TOC as a carbon source ([van Erk et al., 2023](#)), resulting in the low observed TOC concentrations here. However, with the increase in seawater incursion, the environmental conditions changed significantly ([Fig. 1 and S4](#)). Tidal pumping and advection transport of oxygenated seawater enhanced gas exchange rates in MZ

494 sediments with moderate seawater incursion, increasing O₂ penetration and reducing
495 CO₂ accumulation (Huettel et al., 2018; Karthikeyan et al., 2020), shifting these
496 sediments to relatively oxic conditions. For CZ sediments i.e., permanently inundated
497 by seawater, the exchange between oxygenated seawater and porewater can only
498 influence the surface layer of sediments, resulting in anoxic conditions at the bottom
499 (Geng et al., 2022). Furthermore, and in contrast to the SZ, frequent oxygenated
500 seawater level fluctuations in the MZ and CZ induced oxidative stress from reactive
501 oxygen species, which inhibited microbial respiration and facilitated the accumulation
502 of TOC (van Erk et al., 2023). Hence, the TOC was significantly higher in the MZ and
503 the CZ than the SZ ($p<0.05$), and there was no statistical difference between the MZ
504 and the CZ ($p>0.28$) (Fig. S5). Salinity and pH, as the environmental tracers of seawater
505 incursion (Zhao et al., 2023), were also different amongst the three estuarine zones (Fig.
506 1). Since seawater incursion began at the bottom (He et al., 2021), the sediments at the
507 bottom of the estuarine SZ, MZ and CZ all had higher salinity and lower pH. As the
508 degree of seawater incursion increased, the pH in the sediments continued to decrease
509 and the salinity continued to increase. This resulted in the CZ, with complete seawater
510 incursion, showing the lowest pH ($p<0.001$) and the highest salinity ($p<0.05$) of all
511 three zones. In summary, seawater incursion changed the spatial gradient of the
512 dissolved oxygen environment, TOC ($p<0.05$), pH ($p<0.001$) and salinity ($p<0.05$)
513 significantly, and it will further drive changes in the distribution and diffusion BPA and
514 NP in sediments.

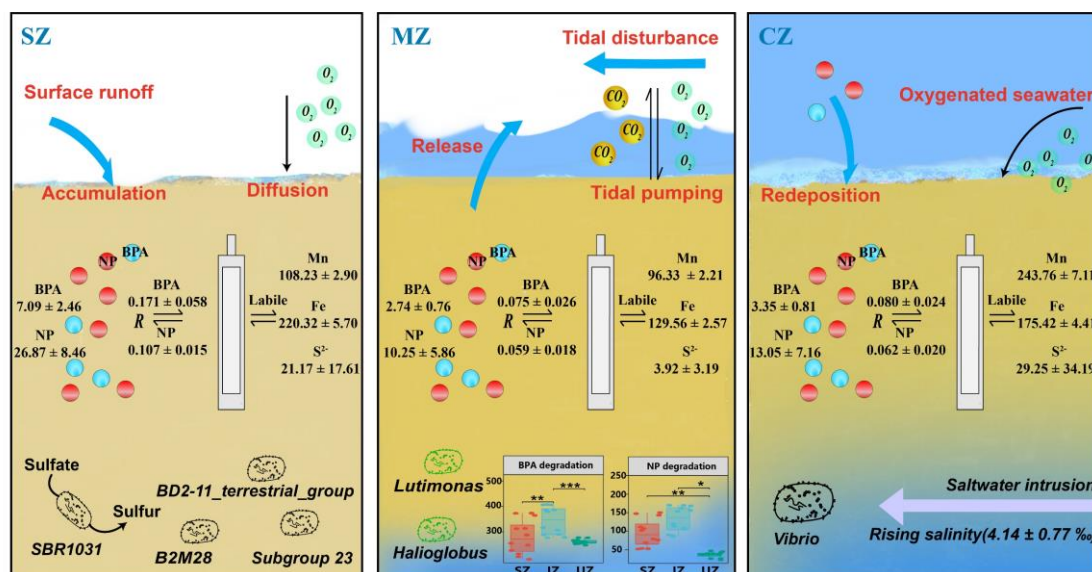


Fig. 4 Biogeochemical processes at the IESW in the different estuarine zones under seawater incursion. BPA and NP concentrations in μg/kg (leftmost in each figure); Mn and Fe concentrations in μg/L, S²⁻ concentrations in mg/L (rightmost in each figure).

4.2 Seawater incursion enhances the mobility of BPA and NP in sediments

Commonly, sediments with higher TOC content should have a stronger ability to trap pollutants (Xu et al., 2018), however seawater incursion alters this pattern, and the distribution of organic pollutants in sediments is dominated by hydrodynamic conditions (Ma et al., 2023) (Fig. 4). Frequent tidal action in the MZ facilitated material exchange between sediment and overlying seawater, resulting in the release of more BPA and NP into the seawater (Liu et al., 2022b). In addition, the oxygenated environment of the MZ, generated by tidal water level fluctuations, promoted the biotransformation of BPA and NP (Huettel et al., 2018; Karthikeyan et al., 2020; Liu et al., 2022b). Moreover, dilution by open seawater is an important factor influencing the concentrations and spatial distribution of pollutants (Cao et al., 2024). Seawater

incursion leads to a large influx of seawater, which further dilutes the concentrations of target substances (Van Ael et al., 2013; Wu et al., 2014; Wang et al., 2019a). As a result, the lowest concentrations of BPA and NP were observed in the MZ (Fig. 2). As for the CZ, with complete seawater inundation, the sedimentary environment has become relatively stable after undergoing a similar process to that of the MZ. Suspended sediment particles transported from the MZ by tidal events gradually settle in the CZ. BPA and NP still adsorbed on these suspended particles will accumulate in the CZ as these particles are deposited. However, due to the very slow deposition process (Álvarez-Iglesias et al., 2007), the concentration of BPA and NP in the CZ and the MZ sediments did not change much. Meanwhile, in the estuarine SZ, TOC had a great dominance of pollutants due to less influence from seawater incursion, which promoted the accumulation of BPA and NP in sediments. Additionally, the diffusion and distribution of BPA and NP in DGT and porewater are shown in Fig. 4. Higher TOC in the MZ and CZ sediments provided more adsorption sites for binding BPA and NP (Xu et al., 2018). Meanwhile, in the MZ and CZ, the “salting-out” effect induced by high salinity promoted the separation of BPA and NP from the water, combined with higher P_c (Table S2), both of them hindering the diffusion of these compounds. All of these factors significantly decreased resupply in the MZ and CZ, and resulted in the highest R in the SZ ($p<0.001$) (Fig. S9). DGT concentrations reflected the labile fractions of target substances in sediments and, similarly to porewater, they both depended on the resupply capacity and target substance concentrations from the sediments (Arsic et al.,

2018). Thus, the highest R and highest concentrations of BPA and NP in sediments ($p<0.01$) also determined their highest DGT ($p<0.001$) and porewater concentrations in SZ ($p<0.01$).

4.3 Microbial community differences and their potential correlation with BPA and NP under seawater incursion

The SZ sediments, with slight seawater coverage, were characterized by high relative abundances of *B2M28*, *BD2-11_terrestrial_group*, *Subgroup 23* and *SBR1031*. *B2M28* and *BD2-11_terrestrial_group* are typically found with high relative abundances in polluted areas (Quintero et al., 2022; Wang et al., 2024b), *Subgroup 23* is commonly found in anoxic environments (Walker et al., 2023), and *SBR1031* is a common sulfate-reducing bacteria (Zhang et al., 2023b) (Fig. 3). This information provides further evidence that the SZ with anoxic deposition environments (high S^{2-} concentrations) accumulated the highest levels of BPA and NP. Meanwhile, seawater incursion drove the evolution of microbial communities, and significantly changed them to adapt to the reshaped environmental conditions (Fig. 4). In the moderate seawater incursion sediments of the MZ, *Halioglobus* (with potential for contribution to hydrocarbon degradation (Suárez-Moo et al., 2020)) and *Lutimonas* (a strictly aerobic genus (Ma et al., 2015)) showed significantly higher relative abundance than in the SZ and CZ ($p<0.05$). Additionally, *Bacteroidota*, as one of the main phyla in the sediments of all three zones, which included heterotrophic bacteria capable of degrading organic compounds, also showed the highest relative abundance in the MZ (Savvichev et al.,

2023). These indicate that the MZ with oxic conditions seemed to exhibit stronger degradation of organic matter. This is consistent with the results shown in section 3.4 (Fig. 3g) i.e., that the most functional genes related to the biotransformation of BPA and NP were significantly higher in the MZ ($p < 0.05$). Seawater incursion created the oxic conditions of the MZ, which appeared to favor increased biotransformation of BPA and NP, and explains the observation that the lowest concentrations of BPA and NP were in this zone. Once seawater coverage was complete, the composition of the microbial community would be further modified due to the shift in environmental conditions. Salinity, which was highest in the CZ with full seawater incursion, has been shown to be a robust predictor for microbial community distribution in estuarine sediments. Previous studies have also shown that high salinity would inhibit the diversity and functioning of microbial communities, and result in the dominance of *Proteobacteria* (Zhang et al., 2017). Thus, the highest salinity observed in the CZ shaped the lowest diversity in the microbial community ($p < 0.001$), and the abnormal dominance of *Proteobacteria* ($76.34 \pm 0.63\%$) (Fig. 3). Moreover, the significantly higher relative abundance genera of *Vibrio*, *Clostridiisalibacter*, *Halarcobacter* and *Marinobacterium* in the CZ are commonly observed in high salinity marine sediments (Sun et al., 2015; Wang et al., 2019b; Baek et al., 2021; Romanenko et al., 2022). Taken together, these findings suggest that complete seawater incursion has severely affected the microbial composition of CZ sediments. Indeed, the microbial community structure in the CZ was unlikely to appear just by chance, and seems to be controlled by seawater incursion,

which corresponds with the higher observed modular structure (Modularity>0.4) of the microbial co-occurrence networks in the CZ (Fig. S8). Meanwhile, the relative abundance of biotransformation genes related to BPA and NP were significantly reduced ($p<0.05$), and may be attributed to the degradation of these pollutants typically performed under oxic conditions. Thus, the biodegradation of BPA and NP appears to cease in the CZ.

4.4 Evaluation of possible EDC release from estuarine sediments due to sea level rise

Coastal estuaries provide crucial ecological and economic functions, including carbon sequestration, support for fisheries and water quality improvement, creating economic services valued at up to US\$194,000 ha⁻¹y⁻¹ (Costanza et al., 2014). However, with global sea level rise, coastal wetlands will be affected, and the threat to aquatic organisms in the ocean will continue to increase due to the transportation and transformation of pollutants (Irrgang et al., 2022). Based on the median of three greenhouse gas concentration scenarios (RCP26, RCP45, and RCP85), sea level rise is projected to be 39.3 cm, 52.9 cm, and 84.6 cm by 2100 (Mengel et al., 2016). Further, under business-as-usual human adaptation scenarios i.e., population density of 5–20 people km⁻², coastal wetland areas would be lost by up to 8%, 12%, and 30% for RCP26, RCP45, and RCP85 by 2100, respectively (Schuerch et al., 2018). Consequently, under these three scenarios, the cumulative additional release of BPA and NP between 2024 and 2100 is projected to be 0.6 t, 0.8 t and 1.8 t, and 0.5 t, 0.7 t and 1.5 t, respectively,

from Red Beach, based on the established model (Fig. 5). Due to the environmental persistence and bioaccumulation of BPA and NP, they would be accumulated and concentrated in marine species, such as mollusks, crustaceans, mussels, and fish (Arditsoglou and Voutsas, 2012) leading to marked anomalies in growth and reproductive rates and migration. This raises concerns for the health and sustainability of estuarine ecosystems, requiring more attention from government and communities and demonstrating the need for advanced management strategies. Increasing vegetative cover in estuarine wetlands could be a viable management strategy since it could decrease sediment suspension under hydraulic disturbance, reducing the risk of pollutant mobilization (Reif et al., 2022). Meanwhile, more rigorous monitoring campaigns should be conducted to assess the mobility and risk of contaminants (Bavi et al., 2023). Additionally, there is a need to develop more stringent chemical management policies, and encourage the use of eco-friendly products to prevent the release of contaminants into coastal estuaries (Pastorino et al., 2024).

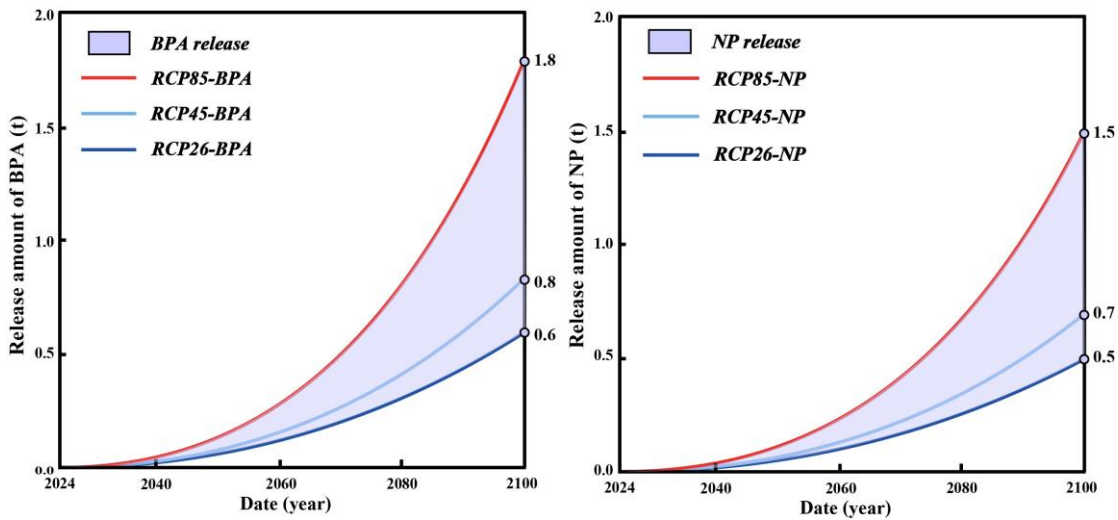


Fig. 5 Cumulative additional release predictions of (a) BPA and (b) NP under three

Representative Concentration Pathways (RCP26, RCP45 and RCP85) between 2024 and 2100.

5. Conclusions

This study reveals continuing seawater incursion has significantly altered geochemical processes and microbial communities of estuarine sediments, which in turn has shifted the distribution, diffusion and biotransformation of EDCs in the Red Beach wetland of the Liaohe estuary. Specifically, the intense tidal actions in the MZ with moderate seawater incursion, resulted in the release of BPA and NP from sediments to overlying seawater. Further, the oxic environment of MZ sediments caused by tidal actions enhanced the potential for BPA and NP biotransformation. These factors contributed to the lowest BPA and NP concentrations observed in our study. However, in the CZ, with complete seawater inundation hence highest observed salinity and lowest pH, the diversity of the microbial community was significantly reduced ($p < 0.001$) and dominated by *Proteobacteria* ($76.34 \pm 0.63\%$). The biodegradation of BPA and NP is significantly reduced and, indeed, tended to stop altogether. This facilitates the continued deposition and accumulation of BPA and NP from the water body into the CZ sediment, turning it into an important sink and source of EDCs. As for the SZ, with slight seawater incursion, the relatively stable anoxic sedimentary environment resulted in the lowest TOC concentrations, highest BPA and NP concentrations, and the strongest resupply capacity. The release of BPA and NP from the Red Beach wetlands could reach a maximum of 1.8 t and 1.5 t, respectively, by 2100

651 due to seawater incursion, calling for government attention.

652 However, there are still many shortcomings and limitations in this study. The next
653 study could expand the study area in the estuary and increase the sampling frequency
654 to further elucidate the spatial and temporal patterns of the influence of seawater
655 incursion on the fate of EDCs in the estuarine environment. In addition, simulation
656 experiments in laboratory by controlling environmental conditions such as salinity
657 gradients and tidal cycles should be carried out to provide a more valuable theoretical
658 basis for the migration and transformation of EDCs in estuarine sedimentary
659 environments under the influence of seawater incursion. Meanwhile, based on more
660 abundant field survey and *in situ* experimental data, the biogeochemical processes and
661 mechanisms of EDCs in estuarine environments under the influence of seawater
662 incursion should be further deduced with the help of artificial intelligence methods such
663 as machine learning. In addition, the impact of seawater incursion on the fate of other
664 emerging organic pollutants (e.g., antimicrobials, perfluorinated compounds, and tyre
665 leaching chemicals) should also be investigated. These studies will help to improve the
666 data and theories on the fate, transport, transformation and risk of emerging organic
667 pollutants in estuaries, and provide a technical basis for the development of coastal
668 ecological and economic restoration strategies.

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