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Integrated assessment of the natural purification capacity of tidal flat for persistent toxic substances and heavy metals in contaminated sediments

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ABSTRACT

Natural purification of pollutants is highly recognized as regulating ecosystem services; however, the purification capacity of tidal flats remains largely unknown and/or unquantified. A 60-day mesocosm transplant experiment was conducted in situ to assess the purification capacity of natural tidal flats. We adopted the advanced sediment quality triad approach, monitoring 10 endpoints, including chemical reduction, toxicity changes, and community recoveries. The results indicated that contaminated sediments rapidly recovered over time, particularly > 50% within a day, then slowly recovered up to ~ 70% in a given period (60 days). A significant early reduction of parent pollutants was evidenced across all treatments, primarily due to active bacterial decomposition. Notably, the presence of benthic fauna and vegetated halophytes in the treatments significantly enhanced the purification of pollutants in both efficacy and efficiency. A forecast linear modeling further suggested additive effects of biota on the natural purification of tidal flats, reducing a full recovery time from 500 to 300 days. Overall, the triad approach with machine learning practices successfully demonstrated quantitative insight into the integrated assessment of natural purification.

Synopsis: With the dynamical change of chemical, toxicological, and ecological components, bio-irrigation and phytoremediation promoted the recovery of contaminated sediments.

1. Introduction

Coastal pollution by persistent toxic substances (PTSs) and heavy metals (HMs) in sediments primarily originates from human activities associated with population growth and expansion of industrial and commercial developments. In particular, polycyclic aromatic

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hydrocarbons (PAHs), alkylphenols (APs), styrene oligomers (SOs), and HMs have been the main pollutants of the Yellow Sea for a long time (Hong et al., 2012). The fate of such PTSs and HMs in marine ecosystems depends significantly on their physicochemical characteristics. Their hydrophobicity and particle reactivity results in PTSs and HMs concentrations several orders of magnitude higher in sediments than in the overlying water. As such, sediment is often considered a sink for pollutants in aquatic environments (Bae et al., 2017) The ultimate fate of pollutant-containing sediments in a given environment is determined by diagenetic processes within the sediments and the physical stability of that particular environment. It is well documented that PTSs and HMs accumulated in sediments adversely impact benthic ecosystems such as the decrease of community of marine organisms in a variety of ways (Lee et al., 2017; Khim et al., 2018).

The role of tidal flats in natural purification has been highlighted primarily from the standpoint of hydrocarbon and nutrient turnover (Hu et al., 2006). Imbalance between PTSs and HMs load and natural purification processes in tidal flats could change the role of coastal sediments from their source to the purifier. Recent reconsideration of marine ecosystem services has garnered interest in the restoration or creation of tidal flats and wetlands as a countermeasure for land-driven coastal pollution (Kim et al., 2020b) The purification capacity of tidal flats has been described through various mesocosm experiments (Kim, 2013). However, since the purification mechanisms for PTSs and HMs in tidal flats are diverse and complicated, the specific contribution of the individual purification process remains poorly understood (Hasanudin et al., 2004).

Bio-irrigation (irrigation by macrofauna) and phytoremediation are important natural processes in tidal flats. For example, bio-irrigation leads to an increased exchange of solutes between porewater and the overlying water. One of the major effects of bio-irrigation is to extend the oxidized zone to a depth within the sediments (Fenchel, 1996) and to stimulate aerobic and subtoxic (i.e., metal reduction) decomposition processes (Banta et al., 1999; Hansen and Kristensen, 1998; Thamdrup et al., 1994);; Moreover, the activity of marine organisms in the sediments has been found to increase the dynamics of water column. The sediment particle resuspension by the movement of the organisms, ingestion, and biological sedimentation could increase, and some sediment particles may be resuspended and released to the outside due to tidal differences (Amato et al., 2016; Dong et al., 2017).

Vegetations have also been shown to play an important role in biogeochemistry in salt marshes through the active and/or passive circulation of elements. Active uptake of elements by vegetation may promote their immobilization in vegetation tissues, as seen in either natural or artificial wetlands through phytoremediation (Kadlec and Reddy, 2001). The key to such phytoremediation processes would be the activity of bacteria communities, which contributes to the overall purification function of tidal flats (Yagi and Terai, 2001). For example, the primary processes involved in the bacterial degradation of PTSs and HMs are biosorption, biomineralization, and co-metabolism, with biosorption being the primary degradation process. As interest grows in the use of tidal flats for purification, it is clear that further studies are needed to characterize better the relationship between biological responses and dynamics of PTSs and HMs.

Outdoor experimental systems, mesocosms have long been developed and become one representative research tool for studying environmental phenomena at small and/or large scales of both terrestrial and aquatic ecosystems. Mesocosm experiments commonly serve as physical models of ecosystems (Ahn and Mitsch, 2002) and provide researchers with a controlled and replicable framework that can be applied in various scientific disciplines (Sapozhnikova et al., 2009; Couto et al., 2011). However, mesocosm experiments have some inherent limitations to consider. Small-scale mesocosms may not be able to reflect the full ecological complexity of the reference ecosystem sufficiently (Ahn and Mitsch, 2002). In addition, mesocosms cannot always comprehensively simulate the complicated array of interactions occurring in natural ecosystems (Blake and Olin, 2022; Kim et al., 2020b; Pennington et al., 2009) To overcome these problems, a half-field mesocosm experiment was conducted, but it was insufficient to reproduce the natural environment in the field (Kim et al., 2022; Lee et al., 2019a). And also, the installation of in situ mesocosms in the marine environment has been difficult due to physical and national regulations. When conducting field-mesocosm experiments, the physical design should be simple without affecting the ecological response and environment. With national approval and consideration of these issues, we applied a newly designed 1.6-m-diameter mesocosm system in the present study, which closely mimicked large-scale, whole-field experiments. Further, we set it up in a natural tidal flat to reflect natural circumstances, which allowed us to expose systems to natural weather conditions, physical dynamics of tides and waves, and chemical processes associated with natural seawater.

Our recent mesocosm studies demonstrated the most appropriate biological methods for evaluating the natural capacity for PTSs and HMs purification in the recovery of soft-bottom benthic communities (Kim et al., 2020b; Lee et al., 2019a). In the present study, we employed the in situ mesocosm system and machine learning method to present and predict a quantitative evaluation of the natural purification capacity for contaminated sediment in tidal flats. Furthermore, the present study adopted an advanced sediment quality triad (SQT) approach, with multiple measures of chemical, toxicological, and ecological indicators, including 1) PTS (PAHs; APs; SOs) and HMs (Zn; As; Pb; Cr; Cd; Cu; Ni) concentrations, 2) aryl hydrocarbon receptor (AhR)-mediated potency for two sediment fractions, and 3) benthic community indices, such as bacterial operational taxonomic unit (OTU), microphytobenthos (MPB) cell counting, meiofauna abundance, and sediment chlorophyll-a (Chl-*a*). To analyze the characteristics of purification status and the benthic community, we used a self-organizing map (SOM; also referred to as a Kohonen map or topology-preserving feature map), which is an artificial neural network-based machine learning method. The present study provides an integrated insight into habitat type-dependent remediation efficiency in the context of the natural purification capacity of tidal flats for PTSs and HMs.

2. Materials and methods

2.1. Sample preparation and in situ mesocosm setting

Contaminated sediment cores from Lake Sihwa near an industrial area in Incheon, Yellow Sea, Korea, were collected for use in mesocosm transplantation (Fig. 1). While transplanting the sediments to the mesocosm, the concentrations of PTSs, such as PAHs, APs, and SOs, in the sediments, were estimated as 2450 ng g^{-1}, 2150 ng g^{-1}, and 550 ng g^{-1}, respectively. In the case of the concentrations of HMs, such as Zn, Cd, Cu, Pb, As, Cr, and Ni, were 229.3 mg kg^{-1}, 1.3 mg kg^{-1}, 114.9 mg kg^{-1}, 78.1 mg kg^{-1}, 20.3 mg kg^{-1}, 176.5 mg kg^{-1}, and 206.7 mg kg^{-1}. The contaminated sediments were then transplanted to the Bong-Am tidal flat, Masan, at a height of 12–15 cm above the in-situ sediment bed. Plastic tubs with a diameter of 1.6 m were used as the base of each mesocosm structure, covered by 1.7 × 1.7 m^2 mesh to prevent the introduction of macrofauna. With reference to previous studies, the shape of mesocosm was designed to better reflect the marine ecosystem (Petersen et al., 2009). Three valves at the lowest, middle, and highest seawater levels were installed on each mesocosm so that seawater could only flow through the valves. In situ mesocosms were designed to simulate four habitat types: bare tidal flat (BT), bare tidal flat + macrofauna (BTm), salt marsh (SM), and salt marsh + macrofauna (SMm). To reflect a natural marine ecosystem, the simulated habitats were treated with native organisms of *Cyclina sinensis, Macrophthalmus japonicus*, and *Phragmites australis*. Sampling was conducted 11 times: 0 h, 2 h, 6 h, 12 h, 1 d, 2 d, 3 d, 7 d, 14 d, 30 d, and 60 d after the installation of the mesocosm. The natural purification capacity for PTSs and HMs in each habitat type was evaluated by adopting a SQT approach that

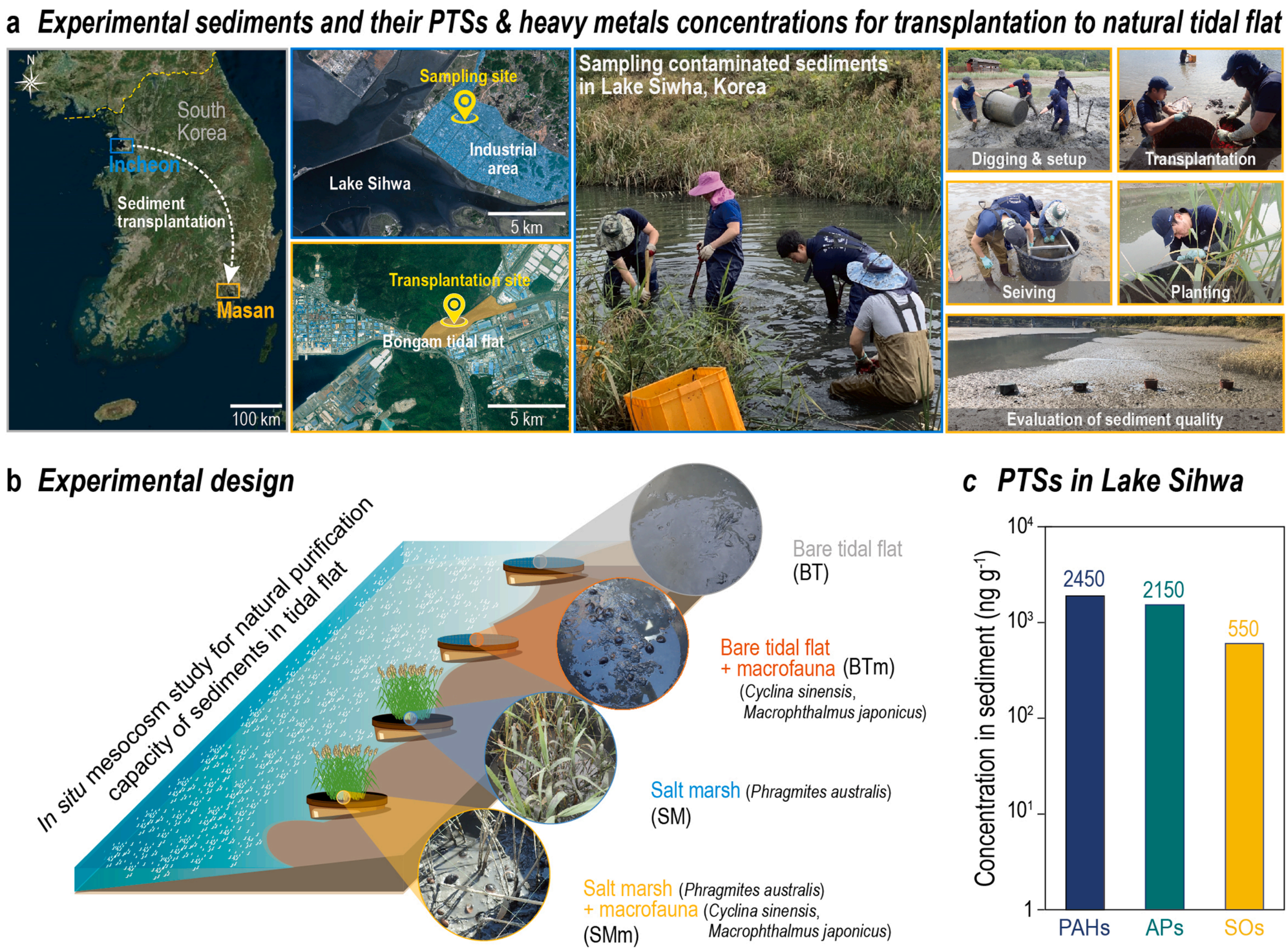

Fig. 1. Schematic of physical and biological remediation techniques, experimental conditions, and sampling design of this study.

evaluated multiple endpoints of chemistry, toxicology, and ecology. Details of the experimental conditions, descriptions of each habitat type, and sampling details are presented in Fig. S1 and Table S1 of the Supplementary materials.

2.2. Chemical assessment

We targeted various PTSs such as PAHs, APs, SOs, and HMs for determining to evaluate the natural purification capacity of tidal flats. These substances are widespread in the Earth's environment and are known to accumulate in living organisms and cause adverse effects by their persistent characteristics (Hong et al., 2016; Kwon and Moon, 2019; Hwang et al., 2021). Concentrations of target PTSs in sediment were measured during the mesocosm experiments, as chemical endpoints, following the methods of previous studies, with minor modifications (Hong et al., 2022; Yoon et al., 2019). Targeted chemicals and instrumental conditions are presented in Tables S2, S3, and S4, respectively. The ranges of method detection limits (MDLs) were 0.27–0.85 ng g^{-1} for PAHs, 0.09–0.97 ng g^{-1} for APs, and 0.30–0.94 ng g^{-1} for SOs (Table S4). Mean recoveries of the four surrogate standards (acenaphthene-d_{10}, phenanthrene-d_{10}, chrysene-d_{12}, and perylene-d_{12}) were generally acceptable: 77.9%–111.3%. Detailed analysis for the HMs was given in the method section of the Supplementary.

2.3. Toxicological assessment

H4IIE-luciferase bioassay was used to test AhR activity to the residual PTSs in the sediments, as toxicological endpoints (Lee et al., 2020). Two sedimentary fractions (F2: mid-polar and F3: polar) were collected and substituted with dimethyl sulfoxide, then concentrated to 1 mL (~ 10 mg contaminated sediment equivalent mL^{-1}) for toxicity testing. After 4 and 72 h of exposure, luciferase assays were performed. The results were calculated using a microplate reading luminometer to calculate mean luciferase luminescence (RLUs) (PerkinElmer, Waltham, MA). Detailed information is shown in the method section of the Supplementary.

2.4. Ecological assessment

Various ecological endpoints were assessed during the mesocosm experiments to evaluate the natural purification of contaminated sediments in the tidal flat. A set of ecological endpoints, including bacterial OTUs (Lee et al., 2019b), MPB cells (Bae et al., 2021), meiofauna individuals (Kim et al., 2020a), and Chl-*a* (Kwon et al., 2020), was simultaneously analyzed, which integrates overall community responses and changes of the benthic organisms in accordance with the reduction of sediment PTSs. Throughout the experimental duration, we conducted a total of 11 samplings for bacteria, meiofauna, and Chl-*a* assessment, while MPB cells were specifically collected three times (D + 0, D + 30, and D + 60). Additional details can be found in the method section of the Supplementary.

2.5. Self-organizing map (SOM)

A self-organizing map (SOM) was used to classify the sample characteristics in each habitat type during the 60 d experimental period. SOMs are powerful and effective tools for creating multidimensional maps from complex data and approximating the probability density function of the input data in a more comprehensive fashion (Kohonen, 1990). The SOM as an ordination method was applied to summarize the variability of data. Thus, the sampling days in each treatment could be arranged on the reduced dimensions so that these arrangements visually summarize the temporal variability of their biological and environmental features.

In the SOM network, the output layer consisted of computation nodes (j) in low dimension. Assuming the input data consisted of four parameters, the value for a given parameter, i, was expressed as a vector of x_i and fed into the input layer of the SOM. In the network, each computation node of j is connected to each node of i for the input layer. The connectivity is represented as the weight, $w_{ij}(t)$, between input and output nodes, which adaptively changes with each iteration of calculation, t, until the convergence is reached. Initially, the weight is randomly assigned to a small value. Each neuron of the network computes the summed distance of $d_j(t)$ between the weight [$w_{ji}(t)$] and the vector (x_i) at the output node of j using an equation below (Eom et al., 2014):

$$d_j(t) = \sum_{i=0}^{N-1} \left(x_i - w_{ji}(t)\right)^2 \quad (1)$$

The input data matrix consists of seven behavior parameters (concentration of PTSs, index of HMs, AhR-mediated potency, bacterial OTU, number of diatom species, abundance of meiofauna, and Chl-*a*) as variables and 44 movement segments (11 sampling times × 4 habitat type treatments) as sample units (Fig. S2). Detailed algorithms could be referred to Zurada (1992) and Chon et al. (1996). The SOM algorithm was implemented using the R package (aweSOM) introducing interactive plots and making analysis of the SOM easier (Boelaert et al., 2022). The detailed R scripts are presented in the Supplementary materials.

2.6. K-means Clustering via principal component analysis (PCA)

Benthic community data across meiofauna, MPB, and sediment bacteria was analyzed to determine the relationship between sedimentary chemical concentrations and community changes (dominant species) over time by combining the benefits of PCA and K-means clustering (Ding and He, 2004). Following PCA analysis, the result is passed to unsupervised clustering using K-means clustering due to K-means' ability to handle outliers. The optimal number of clusters, k, was the value that generated the greatest reduction in the within-cluster sum of squares across a range of k values. Our supervised classification for the dataset is then constructed using logistic regression.

2.7. Forecast model for predicting sediment quality

To predict the future status of sedimentary quality beyond the 60 days of the experimental period, we used the forecast models. This solution would be to propose a mathematical model as a theory, with some free parameters, to fit our data and estimate the values of free parameters. Machine learning is a method that relies on a large dataset with a certain level of complexity, while the latter method is an optimization technique that offers more control over the limitations of a small dataset. Having a proposed theory allows for the interpretation of results and working with less data .

Due to the small number of sediment samples (11 samples) as above mentioned among the limitations of the project, our evaluation does not include any test samples and thus is only for showing the theoretical estimation of trends in SQT values beyond the duration of experiments. Considering that the temporal steps for the SQT values vary from a few hours at the beginning of experiments to 30 days at the final step, we plotted the temporal change of SQT values in a logarithmic scale and proposed the theoretical models with three free parameters. To fit the data using these models, we used the least squares minimization method. For the values of chemistry, we used a simple linear model with two free parameters, α and β (Levenberg, 1944; Newville et al., 2016):

$$\text{Theory}_{\text{Chem}} = \alpha + \beta \ln(hour + 1) \quad (2)$$

The recovery of toxicological values was estimated using the following model:

$$\text{Theory}_{\text{Tox}} = \alpha + \beta \tanh[\gamma \ln(hour)] \quad (3)$$

Finally, the ecological values were fitted using the following model with three parameters, α, β, and γ:

$$\text{Theory}_{\text{Ecol}} = 0.25 + \alpha \ln(hour) + \beta \ln(hour)^{\gamma} \quad (4)$$

We also provide the averages of these fits and the estimated change of SQT values for 4 experimental groups in Fig. 4.

2.8. Statistical analysis

Data analyses were carried out using IBM SPSS software (version 23.0; SPSS Inc., Chicago, IL, USA). The difference in recovery and reduction rate among each treatment was analyzed by Analysis of covariance (ANCOVA). For comparisons of changes between parent and metabolic compounds were conducted based on Tukey's test and paired-*t*-test. In all statistical analyses, p values less than 0.05 were considered to be statistically significant. Principal component analysis (PCA) to visualize the similarity between selected endpoints.

3. Results and discussion

3.1. Chemical, toxicological, and biological responses in the natural purification process of a tidal flat

The average initial (0 h) concentrations of PAHs, APs, and SOs in the contaminated sediments were 2450 ng g^{-1}, 2150 ng g^{-1}, and 550 ng g^{-1}, respectively (Fig. 2 and Table S5). All PTS concentrations decreased sharply within one day of sediment transplant. After the first day, changes to PTS concentrations were dependent upon habitat type: those without macrofauna (BT and SM) decreased slowly, while those with macrofauna (BTm and SMm) decreased more rapidly and with more fluctuation. A contamination index (mCd) for HMs was calculated using Zn, Cd, Pb, Cr, and Cu (Fig. S3) (Kowalska et al., 2018). All habitat types showed a moderate level of contamination (contamination index between 2 and 4) at the initial stage (0 h). The contamination index decreased across the 60 day study period to a very low level (<1.5) in all habitat types except BT.

The rapid decrease in the concentration of PTSs and HMs in the sediments at the initial stage can be attributed to the environmental conditions at the installation site of the mesocosm. Bio-irrigation processes can cause the resuspension of PTSs and HMs adsorbed on organic matter, moving them to the sediment–water interface. At this time, the release of suspended particles to the outside becomes easier, which may reduce the overall concentration of PTSs and HMs (Amato et al., 2016). Additionally, the relatively lower concentration of PTSs and HMs in both the water column and suspended particulate matter at study area, as compared to the transplanted sediment, might have led to dilution effects upon the initial seawater influx (Table S6). The characteristics of the Bong-am tidal flat, categorized as an inland landform, are susceptible to varied influences from freshwater inflow (e.g., street runoff and treatment plant effluents), significantly affecting the physical and chemical properties of both water and sediment layers (e.g., street runoff and effluent of a treatment plant) (Muzuka, 2008; Rezaie-Boroon et al., 2013; Winkels et al., 1998). Moreover, degradation of PTSs by bioavailability has been known to be an important contributor to PTSs reduction (Granberg and Selck, 2007; Olguín and Sánchez-Galván, 2012; Wang et al., 2019). However this study did not measure the concentration of PTSs and HMs in living organisms (benthic fauna and vegetation), and then their decrease due to bioaccumulation cannot be

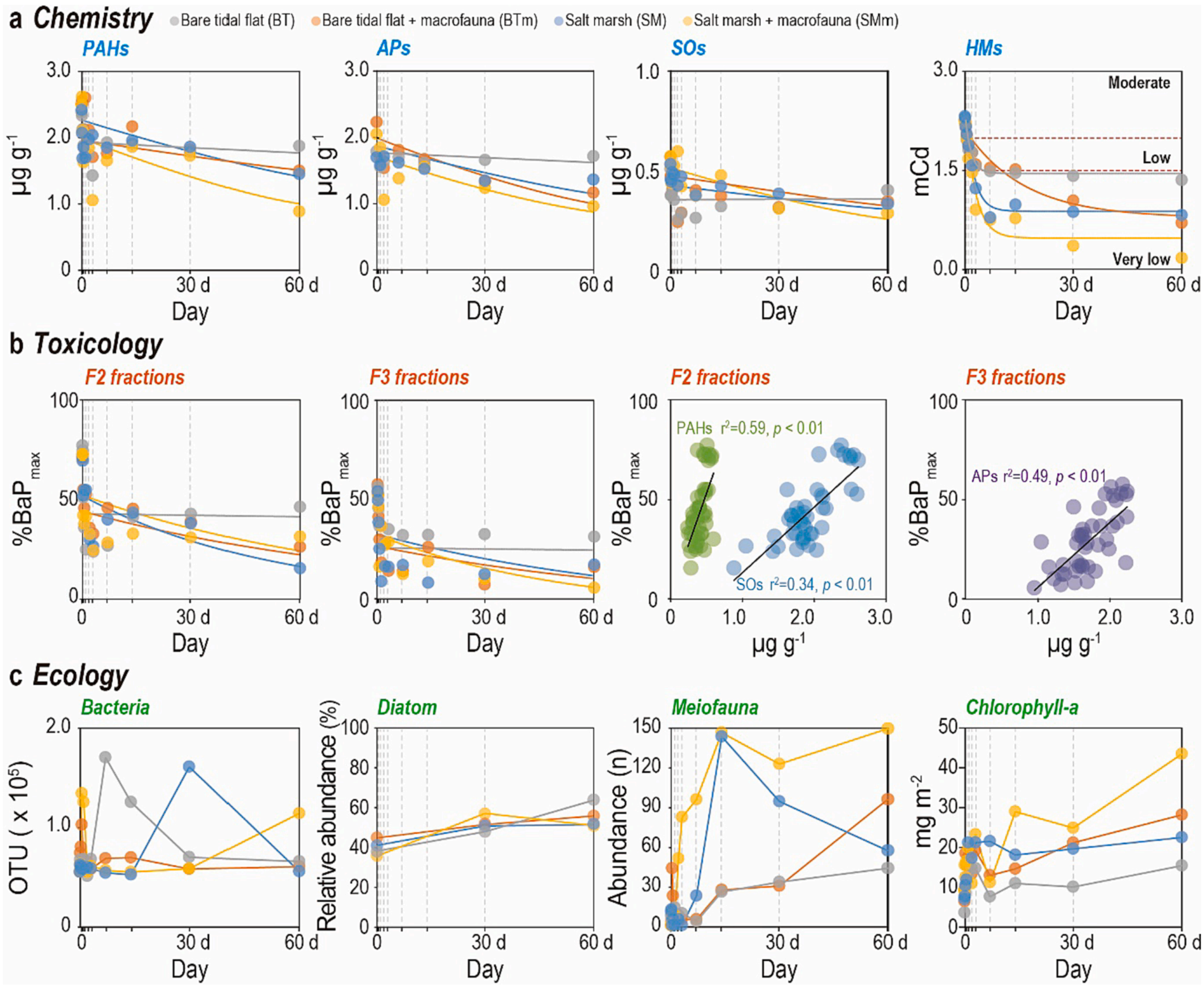

Fig. 2. Chemical- and toxicological reduction rates, and ecological recovery rates after implementation of each treatment.

explained.

In unperturbed organic carbon–enriched sediment, heterotrophic bacterial metabolism rapidly depletes oxygen, limiting its availability to surface sediment (Montgomery et al., 2008). The presence of macrofauna drives bio-irrigation, which leads to sediment mixing and drastic reoxygenation-driven changes to the composition of electron acceptors deep within the sediment, promoting PTSs degradation. Macrofauna can also enhance PTSs degradation or absorption through their various secreted substances, which are a significant source of organic nutrients and inorganic compounds. Vegetations also have a more direct effect on contaminant levels via phytoextraction, which concentrates PTSs and HMs from the environment into their tissues (Shrestha et al., 2019). In particular, phytoextraction and/or phytoremediation were considered promising tools for the bioremediation of polluted environments. For example, Milner and Kochian (2008) demonstrating the Zn, Cd, and Ni model between vegetations and HMs, suggested there were crucial physiological steps of HMs detoxification during the phytoextraction. These are (i) increased HMs ion uptake across the root cell plasma membrane, (ii) decreased HMs ion sequestration in root vacuoles, (iii) increased HMs ion loading into the xylem for transport to shoots, (iv) increased HMs ion influx across the leaf cell plasma membrane, and (v) sequestration in the leaf vacuole. Due to these processes, each HM ion could be accumulated selectively in the vegetation. In the previous study, *P. australis* was evaluated as a promised hyper-accumulator and bio-remediator for Zn, Cd, and Ni (Cicero-Fernández et al., 2017).

Among silica gel fractions from the sediments, greater AhR-mediated potencies were observed in the F2 and F3 fractions at the initial stage (0 h) (Fig. 2). AhR-mediated potency decreased across the 60 day study period, and its variation was consistent with the decrease in PTS concentrations (Fig. 2 and Table S7). After the sediment transplantation, AhR-mediated potency decreased until 3 d and increased from 7 d in F2 fractions. The decrease in AhR-mediated potency of F2 fractions was most dramatic in the habitat types that included macrofauna. The F2 fraction showed a faster reduction in potency than the F3 fraction. Most AhR-mediated responses appeared to be caused by aromatic compounds such as PAHs associated with the residual PTSs in sediments. The target PTSs in each fraction correlated strongly with AhR-mediated potency (PAHs, SOs, and APs showed R^2 = 0.59, 0.34, and 0.49, respectively), indicating that PAHs contributed the most to AhR-mediated potency. The results of the present study were consistent with previous findings that showed that AhR agonists in the fractions were degraded or transformed into less-toxic chemicals during the purification of contaminated sediments (Lee et al., 2018). In particular, the rapid and sharp reduction in potential toxicity indicated the activity of organisms accelerated more degradation of the contributing PTSs in F2 fractions (high molecular weight PAHs and styrene trimers) and F3 fractions (APs) in habitat types containing macrofauna and vegetations (Ohyama et al., 2001; Billiard et al., 2006; Acir and Guenther, 2018).

There were also dynamic changes in bacterial OTU during the study period (Fig. 2 and Table S8). An OTU value greater than 0.5×10^5 was generally detected in all habitat types but surged at different times depending on the type of habitat. In BTm and SMm, the OTU increased more than 2 to 3 times during the period of 6 h d^{-1}. In BT and SM, the OTU increased rapidly after 3 d. The number of species of diatom and the abundance of meiofauna were greater at 60 d than at 0 h. A marked increase in the abundance of meiofauna and the highest value of Chl-*a* was also observed in the SMm habitat. In BTm and SMm, the amount of Chl-*a* more than doubled by the end of the 60 d study period, suggesting that the contaminated sediments were gradually remediated to create an environment favorable for benthic organisms to live (Table S8).

Bio-irrigation has been linked to dramatic changes in both the composition and metabolic activity of the sedimentary bacterial assemblage. Macrofaunal burrows harbor unique populations of bacteria that can mineralize PTSs more rapidly than those from adjacent nonborrowed sediment (Swanberg, 1991). The influence of benthic macrofauna not only influences the biomass, spatial distribution, production rate, and feeding activity (i.e., grazing pressure) of MPB but also on organic matter mineralization rates and nutrient cycling through sediment bio-irrigation (D'Hondt et al., 2018; Thrush et al., 2006). In addition, two species of *C. sinensis* and *M. japonicus* may have a positive effect on MPB photosynthesis and growth by enhancing the remineralization processes and nutrient fluxes across the benthic interface (Sandwell et al., 2009; Lohrer et al., 2010; Donadi et al., 2013; Volkenborn et al., 2016; Hope et al., 2020). Overall, bioturbation processes (i.e., both sediment reworking and bio-irrigation) increase the incorporation and regeneration rates of organic carbon within the benthic compartment. Thus, the processes stimulate nutrient flux across the sediment–water interface where primary microbenthic producers can assimilate them for photosynthesis under optimal conditions (Eriksson et al., 2017).

3.2. *Self-organizing map (SOM) analysis for the evaluation of habitat recovery using machine learning of time-series clustering*

A SOM as a comprehensive analysis was built to categorize the recovery status of each habitat from chemical, toxicological, and ecological data. Considering the number of samples and unexplained variance, four super-classes (groups) were chosen (Fig. S4a). The clustering results of SOM prototypes into super-classes elicited 0.23 of the average silhouette width (S_i), indicating the significant cohesion of a given cell to the cluster that it belongs to (Fig. S4b). In particular, cells C1, C2, and C5 in Group I showed the highest S_i value ($S_i = 0.32$), which meant that they held the longest distance (weakest cohesion or less relationship) to other groups. A spatial gradient was observed for all of the sampling data (Fig. 3a); Group I (10 total) with BT^{0h}, BT^{2h}, SM^{0h}, SM^{2h}, BTm^{0h}, BTm^{2h}, BTm^{6h}, BTm^{1d}, SMm^{0h}, and SMm^{2h}; Group II (13 total) with BT^{6h}, BT^{12}h, SM^{6h}, SM^{12h}, SM^{1d}, SM^{2d}, SM^{3d}, BTm^{12h}, BTm^{7d}, SMm^{6h}, SMm^{12h}, SMm^{1d}, and SMm^{2d}; Group III (11 total) with BT^{1d}, BT^{2d}, BT^{3d}, BT^{7d}, BT^{14d}, BT^{30d}, BT^{60d}, SM^{30d}, BTm^{2d}, BTm^{3d}, and BTm^{14d}; Group IV (10 total) with SM^{7d}, SM^{14d}, SM^{60d}, BTm^{30d}, BTm^{60d}, SMm^{3d}, SMm^{7d}, SMm^{14d}, SMm^{30d}, and SMm^{60d}. This result, along with sampling time/date and various other results (Fig. 2), suggests that PTSs, HMs, and their toxicities decreased over time, and the benthic community recovered.

Each group had characteristics that showed varying recovery features in chemistry (reduction rate of PTSs and HMs), toxicology (reduction rate of AhR-mediated potency), and ecology (increase of benthic community health) from the initial stage (0 h) (Fig. 3b). The common characteristics of Group I are high concentration and potential toxicity of PTSs and low level of ecological restoration. In contrast, Group IV showed 40% reduction in PTSs and HMs, 54% reduction in toxicity, and 2.9 times increase in the benthic community, which indicated the greatest natural purification capacity as compared with the other groups. According to this observation, each group could be considered to represent a different degree of remediation from PTSs and HMs contamination set by cross-related chemical, toxicological, and ecological data (Group I for least remediation and Group IV for greatest). It should be noted that each habitat type moved across groups and followed a different route of remediation (Fig. 3c). All habitats except BT achieved the same state as Group IV by day 60. Interestingly, BTm data moved back and forward between groups as time elapsed, and its transition period was shorter than other groups, reflecting the vigorous remediation process occurring in that habitat. Both SM and SMm habitats recovered more quickly than non-saltmarsh habitats, and SMm followed the most effective route and reached Group IV after only three days.

There are various methods for evaluating the health of a benthic ecosystem (Pearce et al., 2014). SQT is an integrated analysis for evaluating marine ecosystem health associated with recovery from contamination (Lee et al., 2018). In this study using novel classifications and groupings, machine learning offered a useful tool to characterize aspects of sediment quality (Pearce et al., 2014). To this end, we presented an approach for identifying a reasonable number of classes that

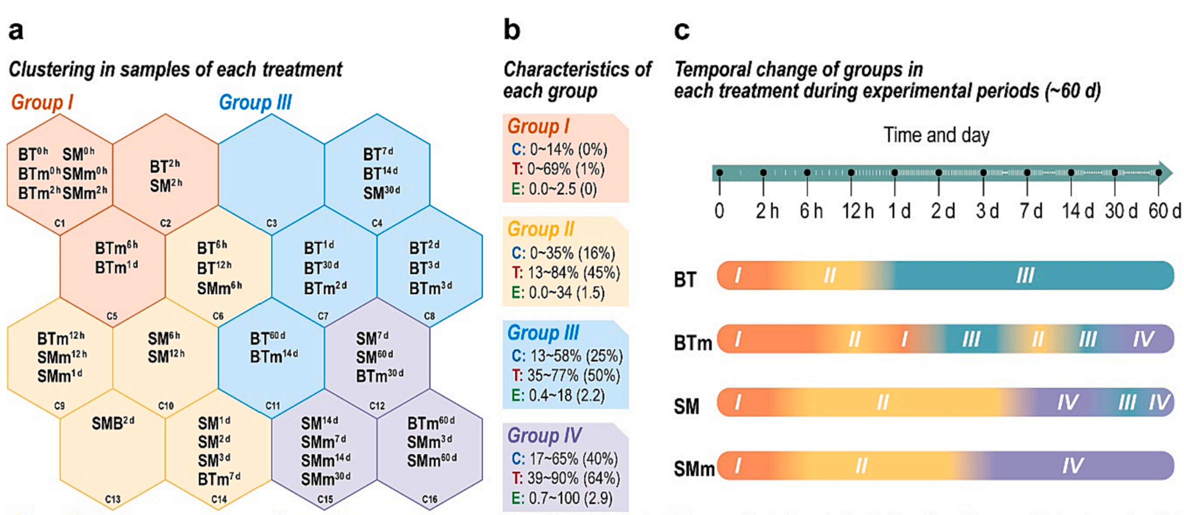

Fig. 3. A novel, machine learning-based approach for determining sediment quality and evaluating the purification capacity of tidal flats for contaminated sediments. **(a)** Self-organizing map showing treatments of each sediment quality group. **(b)** Sediment quality triad characteristics of each group. **(c)** Change of characteristics in each group across the treatment period.

emphasized aspects of classification representativeness and considered potential statistical power using a SOM (Zurada, 1992). This strategy found four super-classes to be a reasonable grouping for the data. The results from the approach of SQT via SOM could help to understand the kinds of complex interclass relationships that would be useful for evaluating the recovery of marine ecosystems requiring larger classification systems.

3.3. *Effect of macrofauna (C. Sinensis and m. japonicus) and salt marsh (P. australis) on the change of benthic community*

To investigate the effects of macrofauna and vegetations in variation in a benthic community, habitats with similar characteristics were categorized into four groups in terms of the presence and absence of macrofauna and vegetations ('w/ macrofauna': BTm + SMm, 'w/o macrofauna': BT + Sm, 'w/ vegetation': SM + SMm, and 'w/o vegetation': BT + BTm). The multivariate variability of z-standardized data was visualized using axes of the two principal components (PC1 and PC2) and accounted for 75.9% (41.6% and 34.3% for axes 1 and 2, respectively) and 79.2% (48.7% and 30.5% for axes 1 and 2, respectively) of the variation (Fig. S5a and b).

As reflected in the plot, four habitat type groups were formed according to the sampling periods on the PCA diagram (A1–A4 and B1–B4) (Fig. 4). Group A1 showed a significant correlation with Nematoda and *Paralia sulcate*, *Chloroflexi*, *Acidobacteriota*, *Desulfobacterota*, *Firmicutes*, and *Verrucomicrobia* in bacteria, and Polychaeta and Amphipoda in meiofauna, were the most closely related groups (Fig. 4a). Nematoda, *Thalassinonema nitzschioides*, *Pastescribacteria*, *Actinobacteriota*, and *Thermotogota* showed a strong correlation with group A3, and the intermediate sampling dates showed lower contamination in w/ macrofauna group. Finally, group A4 was correlated with Copepoda, *Navicula perminuta*, *Cyclotella cf. stylorum*, and *Campilobacterota*. Each habitat group moved between the clusters as time elapsed, and its route depended on the presence or absence of macrofauna in the habitats (w/ macrofauna vs. w/o macrofauna). In particular, the 'w/ macrofauna' group showed more frequent transitions in the benthic community structure. Fig. 4b also showed that four groups (B1–B4) were identified with a similar benthic community to group A (Fig. 4b). Group B1

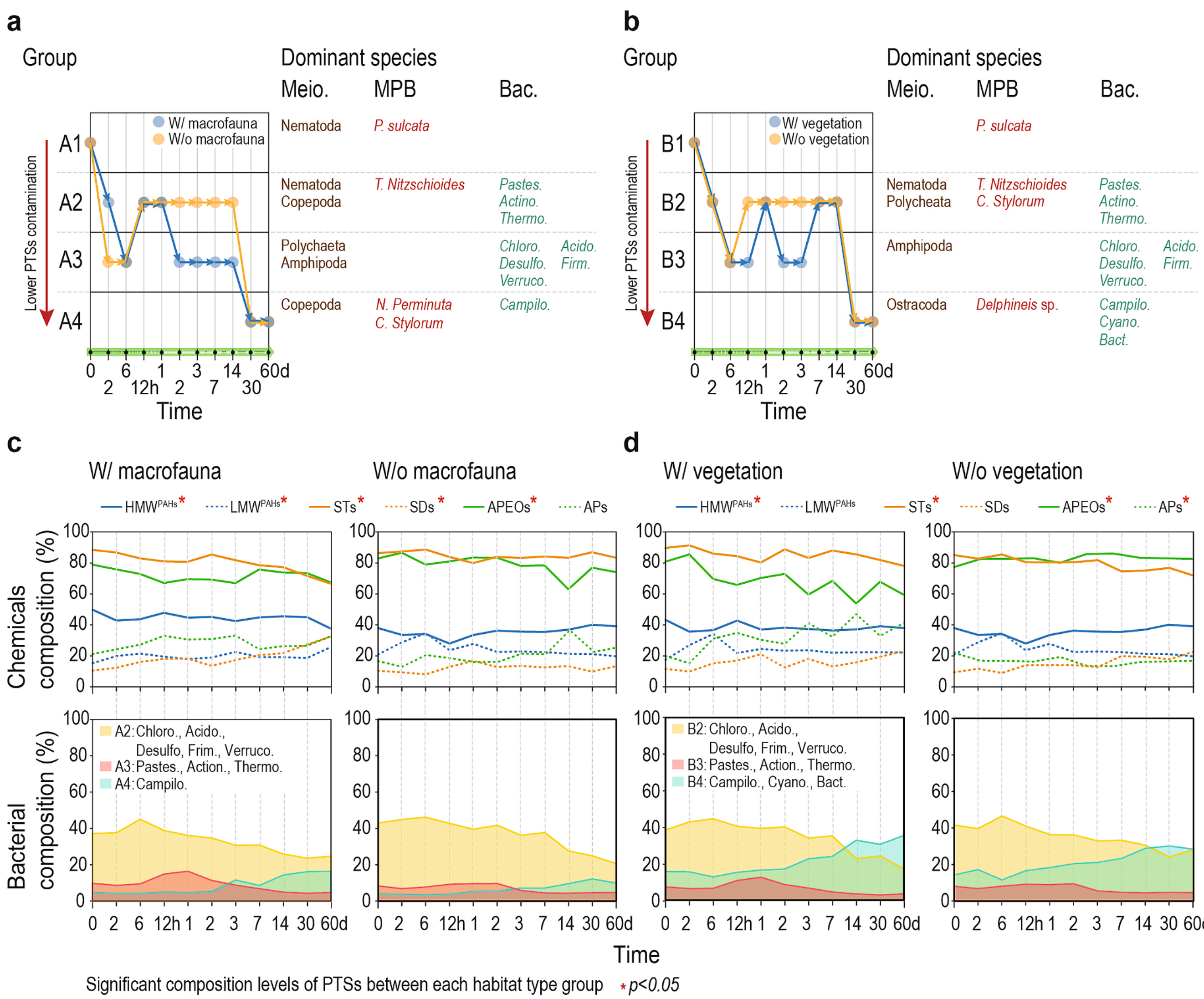

Fig. 4. Change in the benthic community over time under each unique PTSs and HMs composition in each habitat group. **(a–b)** Changes in benthic community group (bacteria, MPB, and meiofauna) based on the results of the principal component analysis in each habitat group. **(c–d)** Changes in PTS composition according to the dominant bacteria in each habitat group. (High molecular weight, HMW and low molecular weight, LMW) (MPB: microphytobenthos including *P. sulcata*: *Paralia sulcate*; *T. nitzschioides*: *Thalassinonema nitzschioides*; *N. perminuta*: *Navicula perminuta*; *C. stylorum*: *Cyclotella* cf. *stylorum*) (Bacteria: Pastes.: *Pastescibacteria*; Actino.: *Actinobacteriota*; Thermo.: *Thermotogota*; Chloro.: *Chloroflexi*; Acido.: *Acidobacteriota*; *Desulfobacterota*: Desulfo.; Firm.: *Firmicutes*; Campilo.: *Campilobacterota*; Cyano: *Cyanobacteria*; Bact.: *Bacteroidota*).

suggested *P. sulcate* as a dominant diatom at the initial stage (0 h). Group B2 was dominant in w/o vegetation, with a similar community as A2 but without Polychaeta. During most of the study period, Nematoda was strongly dominant in the 'w/o vegetation' habitat group (cluster B3). Interestingly, group B4 contained Ostracoda, *Delphineis* sp. *Cyanobacteria*, and *Bacteroidota* were not dominant in 'w/ macrofauna' or the 'w/o macrofauna' habitat groups.

Macrofauna either stabilize or destabilize the sediment by facilitating or inhibiting effects on shoreline succession (Reise, 2002). The present study provides experimental evidence that macrofauna has the potential to change the ecology of a habitat in the depositional coastal environment. Change in the dominant benthic organisms (meiofauna: Nematoda → Copepod; MPB: *P. sulcata* → *N. Perminuta* and *C.* stylorum; bacteria: *Chloroflexi* → *Campilobacterota*) reflected that bio-irrigation by macrofauna could enhance biodegradation of native PTSs and promote the emergence of several benthic communities through improved sediment quality (McQuoid and Nordberg, 2003; Saros and Anderson, 2015; Drira et al., 2018; Chen et al., 2019; Speirs et al., 2019; Wang et al., 2022). The influence of vegetation transplantation on the benthic community promoted a change in bacterial communities and made a slight difference in meiofauna communities between w/o vegetation and w/ vegetation groups. On day 30, the habitats with vegetation seemed to rapidly acquire diatom communities of similar composition and diversity as those present on day 60, indicating a relatively rapid recovery of MPB communities at the level of major taxonomic groups. Furthermore, the appearance of various meiofauna species occurred earlier in the contaminated sediments (Fig. S6). An except for the early appearance of *Chloroflexi* in the w/ vegetation group, the benthic community changed similarly to that of the w/o vegetation group. The structural heterogeneity in tidal flats might accelerate the change in MPB and bacterial communities, without any notable effect from halophyte seeding (Tolhurst et al., 2020).

3.4. *Correlation between the composition of PTSs and bacterial community in each habitat type group*

Among all the bacteria detected in the samples (Fig. S7a), we analyzed bacterial order, which accounted for over 1% of the community, and compared the relationship between PTS components (parent and metabolite compounds) and bacteria to the results of each clustered group (Fig. 4c). In group w/ macrofauna, the parent compounds (including high molecular weight PAHs, STs, and alkylphenol ethoxylates (OPEOs (octylphenol ethoxylate) + NPEOs (nonylphenol ethoxylate)) decreased over the experimental period, while the metabolites (low molecular weight, LMW; SDs, and APs) tended to increase. In group w/o macrofauna, except for APs the change in the composition of the other two PTSs was not evident. The A1 group was observed to have a positive correlation with LMW, SDs, and OPEOs + NPEOs, and A2 was associated with an increase in SD ($p < 0.05$). In particular, A3 showed a positive correlation with APs ($p < 0.05$) (Fig. S8). In the case of w/o macrofauna, there was no clear relationship between bacterial community and compound composition. Considering the period of rapid change from A1 to A3, with the bio-irrigation effect from the macrofauna, it was confirmed that PTSs in the contaminated sediment showed rapid changes in the order of PAHs, SOs, and APs due to the change of the bacterial community.

For the w/o vegetation group, three bacterial groups changed according to the composition of PTSs across the study period. When comparing the compound compositions of w/ and w/o vegetation groups, there was a marked decrease in APs and an increase in OPEOs + NPEOs (Fig. 4d). Among the bacterial groups, B1 showed significantly increased levels of metabolites such as LMW and SD, and APs in group SO ($p < 0.05$) (Fig. S8). The correlations between chemical compositions and other bacterial groups were similar to those in w/ macrofauna. Meanwhile, the w/o vegetation group showed a similar result to the w/o macrofauna group, such that B3, including *Campilobacterota*, *Cyanobacteria*, and *Bacteroidota,* was only positively correlated with the metabolites (LMW, SD, and APs) ($p < 0.05$).

3.5. *Temporal change of SQT values to evaluate natural purification of contaminated sediments and the effect of bio-irrigation and phytoremediation in tidal flats*

The SQT approach was employed to identify how bio-irrigation and phytoremediation mitigate the impact of PTS and HMs contaminations on the chemical, toxicological, and ecological features of the sediments. To evaluate the effectiveness of various methods of physical and biological remediation, ratios of ten target endpoints to negative control values (reference sediment) were determined (Fig. 5). SQTs of purification rates were compared for the habitat groups in terms of bio-irrigation (w/ macrofauna vs. w/o macrofauna) and phytoremediation (w/ vegetation vs. w/o vegetation), and revealed an overall improvement of SQTs in all groups. Furthermore, the regression model predicting the change in SQT value for each habitat was able to predict the change trend of each recovery value.

The w/ macrofauna group had an average recovery of 71.2% due to superior restoration in all areas (chemistry: 11.0%p, toxicology: 14.1%p, ecology: 30.0%p) as compared to the w/o macrofauna group (average recovery: 52.8%). The forecast model based on the linear regression proposed that for chemistry values to show a gradual recovery compared to those of toxicology and ecology. Full recovery of the toxicology component in w/o macrofauna was predicted to take over 60 days, which was later than that of w/ macrofauna (30 days). Better purification of the w/ macrofauna group can be attributed to bio-irrigation by *Cyclina sinensis* and *Macrophthalmus japonicus*. In the case of ecology, w/ macrofauna showed a more rapid recovery rate than w/o macrofauna, and 100% recovery was predicted before 100 days. The w/ vegetation group had an average of 68. % recovery, which is higher than the w/o vegetation group's 55.0%. The results suggested that phytoremediation would enhance the final ecological, toxicological, and chemical recoveries by an additional 17.6%p, 14.1%p, and 10.3%p, respectively, as compared to the w/o vegetation group. Unlike chemistry, the pattern of recovery in toxicology and ecology is the result of differences in biological reactions according to the effect of PTSs and HMs. Biochemical and ecological responses are more diverse and complex than chemical changes (concentrations of PTS and HMs), and the degradation of a highly toxic parent chemical dramatically increases the recovery rate in toxicology and ecology after a certain period of time, changing the recovery pattern (Yim et al., 2017).

Changes in physicochemical properties by the activity of organisms in tidal flats provide many positive attributes toward the purification of PTSs and HMs. First, sediment porosity, percolation rates, and horizontal hydrologic conductivity rates, which are created by the activity of macrofauna or vegetation roots, control the rate of oxygen delivery to subsurface layers. Second, macrofauna and vegetation in tidal flats provide an enriched culture zone for microbes involved in degradation (Olsen et al., 1998; Macek et al., 2000; Meharg and Cairney, 2000). As interest in constructed wetlands grows, many marine organisms, including shrimps, crabs, and mussels, can be considered eco-friendly biological remediators. Currently, vegetation is the most commonly used biological element in constructed wetlands and plays important roles in PTS and HMs purification, both directly and indirectly, by stimulating bacterial composition changes. Indeed, constructed wetlands with combined vegetation and macrofauna tend to show greater efficiency in removing organic waste (Sachaniya et al., 2019).

4. Conclusion

The present study is the first to provide a comprehensive assessment of bio-irrigation and phytoremediation effects in a tidal flat through the implementation of an in situ mesocosm and artificial neural network-based clustering machine learning method. The findings included

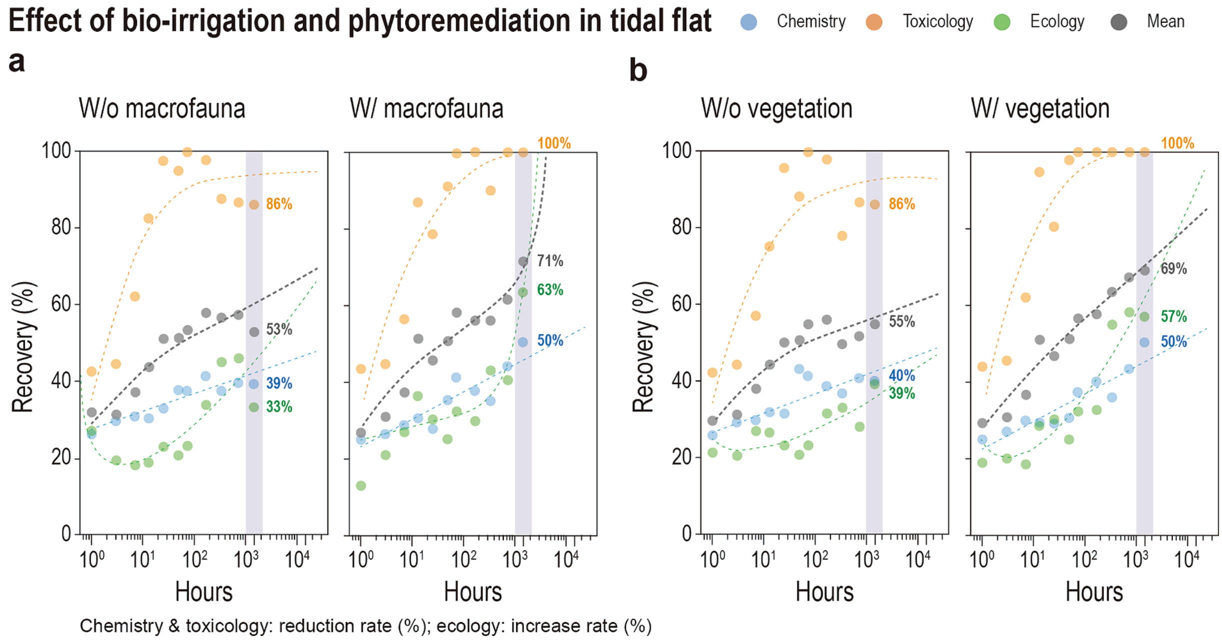

Fig. 5. Temporal change of SQT values based on integrated approaches with chemistry, toxicology, and ecology by using forecast model. The integrated approaches included: 1) PTSs and HMs reduction (concentration), 2) toxicology attenuation (%BaP_{max} from F2 and F3), and 3) benthic ecosystem recovery (bacterial community, MPB cell, meiofaunal individual, and chl-*a*).

reduced concentrations of PTSs and HMs, and AhR-mediated potencies, and the recovery of benthic communities. Specifically, the concentrations of target PTSs and HMs, and their associated toxicities decreased significantly as benthic community increased over the 60 d study period. Greater reductions in PTS and HMs concentrations were observed in the habitat types that contained macrofauna and vegetation as compared to those without. Because this study conducted chemical, toxicological, and biological evaluations following the recovery of contaminated sediments, there is the limitation of not estimating bioavailability through bioaccumulation and phytoextraction. This substantial remediation of PTS and HMs contamination was presumably due to bio-irrigation and phytoremediation effects. Bio-irrigation from macrofauna drove the change in the benthic community and hydrocarbon degradation by bacterial succession. However, further research is needed to determine the detailed contribution of physical, chemical, and biological factors to the decreased concentration of PTSs and HMs over the 60 day experimental period.

Both bio-irrigation and phytoremediation caused the bacterial community to utilize the parent hydrocarbon compounds first, which caused the rapid reduction of AhR-mediated potency in contaminated sediments. Through integrated SQT analysis, bio-irrigation resulted in more recovery gains of 11.0%p chemically, 14.1%p toxicologically, and 30.0%p ecologically, and phytoremediation showed more recovery gains of 10.3%p chemically, 14.1%p toxicologically, and 17.6%p ecologically. Altogether, the results of the present study document the natural purification capacity in tidal flats through an integrated analysis of environmental pollutants and associated biological responses in severely contaminated sediments. Such the study of quantification of the capacity can contribute to further valuing the Yellow Sea, which has many tidal flats.

CRediT authorship contribution statement

Taewoo Kim: Writing – original draft, Visualization, Formal analysis, Data curation, Conceptualization. **Changkeun Lee:** Writing – review & editing, Validation. **Inha Kwon:** Investigation, Formal analysis. **Junghyun Lee:** Investigation, Formal analysis. **Shin Yeong Park:** Investigation, Formal analysis. **Dong-U Kim:** Investigation, Formal analysis. **Jongmin Lee:** Investigation, Formal analysis. **Gayoung Jin:** Investigation, Formal analysis. **Mehdi Yousefzadeh:** Formal analysis, Data curation. **Hanna Bae:** Investigation, Formal analysis. **Yeonjae Yoo:** Formal analysis. **Jae-Jin Kim:** Writing – review & editing. **Junsung Noh:** Writing – review & editing. **Seongjin Hong:** Writing – review & editing, Formal analysis. **Bong-Oh Kwon:** Writing – review & editing. **Won Keun Chang:** Writing – review & editing, Project administration. **Gap Soo Chang:** Writing – original draft, Visualization, Conceptualization. **Jong Seong Khim:** Writing – review & editing, Visualization, 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.

Data availability

The data that has been used is confidential.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2024.108534.

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