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Chemical-, site-, and taxa-dependent benthic community health in coastal areas of the Bohai Sea and northern Yellow Sea: A sediment quality triad approach

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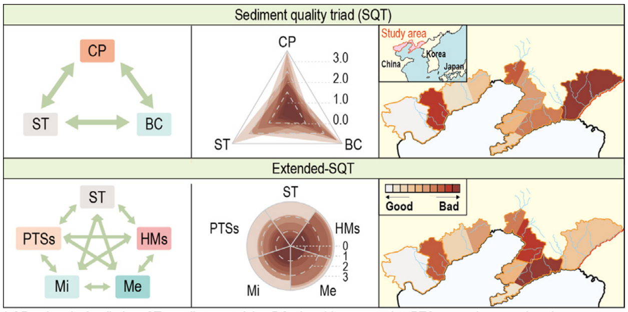
HIGHLIGHTS

- Extended-sediment quality triad was applied to enhance typical benthic assessment.
- Sediment toxicity might be over- or under-estimate chemical pollution and benthic health.
- Meiofaunal abundance would be a good ecological indicator of pollution by arsenic.
- Moderate to severe pollution evidenced in over half of the sampling locations.

GRAPHICAL ABSTRACT



* CP - chemical pollution; ST - sediment toxicity; BC - benthic community; PTSs - persistent toxic substances; HMs - heavy metals; Mi - microbiome; Me - meiofauna

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ABSTRACT

To investigate benthic ecological quality associated with coastal pollution in the Bohai Sea and northern Yellow Sea, a sediment quality triad (SQT) approach was applied. Chemical (six groups of persistent toxic substances (PTSs) and 8 metals and metalloids), toxicological (AhR-mediated potency), and ecological (bacterial and meiofaunal communities) elements were selected and used in an integrated sediment assessment. The benthic meiofaunal community was newly analyzed and used as an additional component of the infaunal community during the SQT. Concentrations of chemicals and potential toxicity in sediments both indicated moderate to severe pollution in the study area, characterized by site-specific and land-uses. In particular, As, DDTs, and bioassay-derived dioxin equivalents exceeded corresponding sediment quality guidelines at nearly all locations. Limited occurrences of meiofaunal taxa (mean = 5.2) and relatively low species diversity, mainly comprised of nematodes (75.3%) and copepods (14.6%), among locations was generally consistent with pollution. The benthic community was consistent with compound-specific responses to gradients of contamination, particularly for As. Densities of two taxa, Nematoda and Nemertea exhibited strong negative correlations with concentrations of As.

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Sediment quality assessment
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Compound-, site-, and taxa-specific variability of pollution of sediments was further supported by results of cluster analysis (CA) and nonmetric multidimensional scaling (NMDS). Finally, assessment integrating five benthic quality elements, including: 1) PTSs; 2) metals and metalloids; 3) sediment toxicity; 4) sediment microbiome; and 5) benthic meiofaunal community, explained contamination of sediments associated with land-uses, locality, or habitat. Status of the benthic community could not be explained by single component and their associations were not quantitative. Results of the integrated assessment, considering multiple benthic quality elements were useful to address overall quality of sediment, and were consistent with chemical-, species-, or site-dependent pollution of sediments in the Bohai and Yellow Seas.

1. Introduction

It is well known that anthropogenic pollutants such as persistent toxic substances (PTSs), metals, and metalloids exert various adverse effects on infauna inhabiting ecosystems. In particular, utility of benthic communities for assessment of sediment quality in coastal marine ecosystems has been documented (Warwick et al., 1990; Engle et al., 1994; Ryu et al., 2011; Lee et al., 2016; Bae et al., 2017). Benthic invertebrates are locally sedentary and thus are subjected to prospective toxicity of ambient water and sediments with respect to the localized pollution. Molluscs have received attention. For instance, Boyden (1974) studied relationships between the body sizes of six mollusk species and the concentrations of six metals (Cd, Cu, Fe, Ni, Pb, and Zn). Also, Navrot et al. (1970) proposed use of a limpet species for monitoring of coastal metal pollution.

While these earlier studies were more or less centered on physiological responses of various species, other studies have tried to understand community- or population-level responses to environmental pollutants. Succession of benthic macrofauna has been studied relative to organic pollution (Pearson and Rosenberg, 1978), infaunal benthic communities have been compared between areas with and without oil seepages (Spies and Davis, 1979). Comparisons have been made to determine if contamination of sediments changes structures of benthic communities (Rygg, 1986). More recently, the historical association between exposures of macrofaunal communities to metals alters their structure at either the population or community levels (Ryu et al., 2011). While, compared to those of macrofauna, less attention has been given to responses of benthic meiofaunal communities (Raffaelli and Mason, 1981; Raffaelli, 1982). Several former studies utilized ratios of numbers of nematodes to copepods (N/C ratio) for monitoring of marine pollution and use of such ratios has subsequently been refined and adopted by other researchers (Warwick, 1981; Moore, 1987; Moore and Bett, 1989; Sandulli and de Nicola-Giudici, 1990). It should be noted that most of these studies of meiofauna have focused on organic enrichment and a few studies have aimed to address the relationship between meiofaunal communities and chemical pollution, particularly for PTSs and metals (Lee et al., 2001; Liu et al., 2015).

While many previous studies were collective efforts to address ecological responses related to chemical pollutants, there have been other combined efforts to better understand status and trends in benthic communities by use of holistic approaches (Long et al., 2001; Khim and Hong, 2014). The so-called 'sediment quality triad (SQT)' was first proposed in the middle of 1980s (Long and Chapman, 1985). The "triad" was an approach that consisted of concentrations of chemicals, toxicity of sediments in bioassays, and structure of the benthic community, and applied as multiple lines of evidence. This approach has increasingly included other techniques such as environmental DNA (e-DNA) (Xie et al., 2017).

In the present study, an extended SQT approach was adopted to better assess pollution of sediment in coastal areas of the Bohai Sea and the northern Yellow Sea, with a focus on responses of meiofaunal communities by use of a multiple lines of evidence approach in which five sets of data on sediment qualities were compared. Target elements included: 1) PTSs; 2) metals and metalloids; 3) toxic potencies of sediments; 4) sediment-microbiome, based on e-DNA; and 5) structure of benthic meiofaunal community. Specific goals of this study were to: 1) determine overall health of the benthic invertebrate community; 2) identify priority chemicals and/or hot spots of concern; and 3) investigate and rank primary characteristics contributing to status of the benthic invertebrate communities.

2. Materials and methods

2.1. Study area

The area of interest in the present study spans seven districts along the coast of the northern Bohai Sea, China, which is adjacent to the marine ecosystem of the Yellow Sea (Fig. 1). This was part of a larger study of contaminants in water, soil, sediment, and biota of watersheds of the Bohai Sea and the northern Yellow Sea, which was conducted in 2008 to understand overall chemical pollution and ecosystem health. During the present study, additional data on assemblages of meiofauna and general sediment properties, such as total organic carbon (TOC), total nitrogen (TN), and stable carbon isotope ratio ($\delta^{13}C$) in sediment were measured in samples collected from 10 locations along coastal areas of the Bohai and northern Yellow Seas. Sampling locations were generally in regions of major rivers, either downstream of or in mouths of rivers and coastal areas, such as beaches or tidal flats of the Bohai Sea; Tangshan (TS5), Qinhuangdao (QH3), Huludao (HL1 and HL5), Panjin (PJ2), Yingkou (YK2), Dalian (DL1, DL4, and DL6), and Dandong (DD3). In order to provide comparable information on sampling locations, they are identified by the same identifiers (IDs) that were assigned during previous studies. Detailed descriptions of the study area and locations have been given previously (Naile et al., 2011).

2.2. Field sampling

Sediment samples were collected during 2008 from along the coastal areas of the Bohai Sea and the northern Yellow Sea and meiofaunal assemblages and general sediment properties were evaluated. Samples were collected using a plastic corer (i.d. = 15 cm) and stored on ice during transfer to the laboratory. Top 10 cm segments of sediments were utilized for quantifications of general sediment properties, to be used as supporting data for interpretation of data on assemblages of meiofauna observed in the top 4 cm segments of the cylindrical corer (i.d. = 2.2 cm), collected simultaneously from the same locations. Samples containing meiofauna were fixed with 3% formaldehyde and transferred to the laboratory for microscopic analyses.

2.3. TOC, TN, and $\delta^{13}C$ analyses

In the laboratory, TOC, TN, and $\delta^{13}C$ were measured by use of an Elemental Analyzer-Isotope Ratio Mass Spectrometry (IRMS, Elementar, Hanau, Germany) according to previously published methods (Rumolo et al., 2011). Samples of sediment for TOC and $\delta^{13}C$ measurements were treated with 1 M HCl to remove inorganic carbon prior to instrumental analysis. IAEA-CH3 and IAEA-CH6 were used as standards to calibrate the $\delta^{13}C$ value.

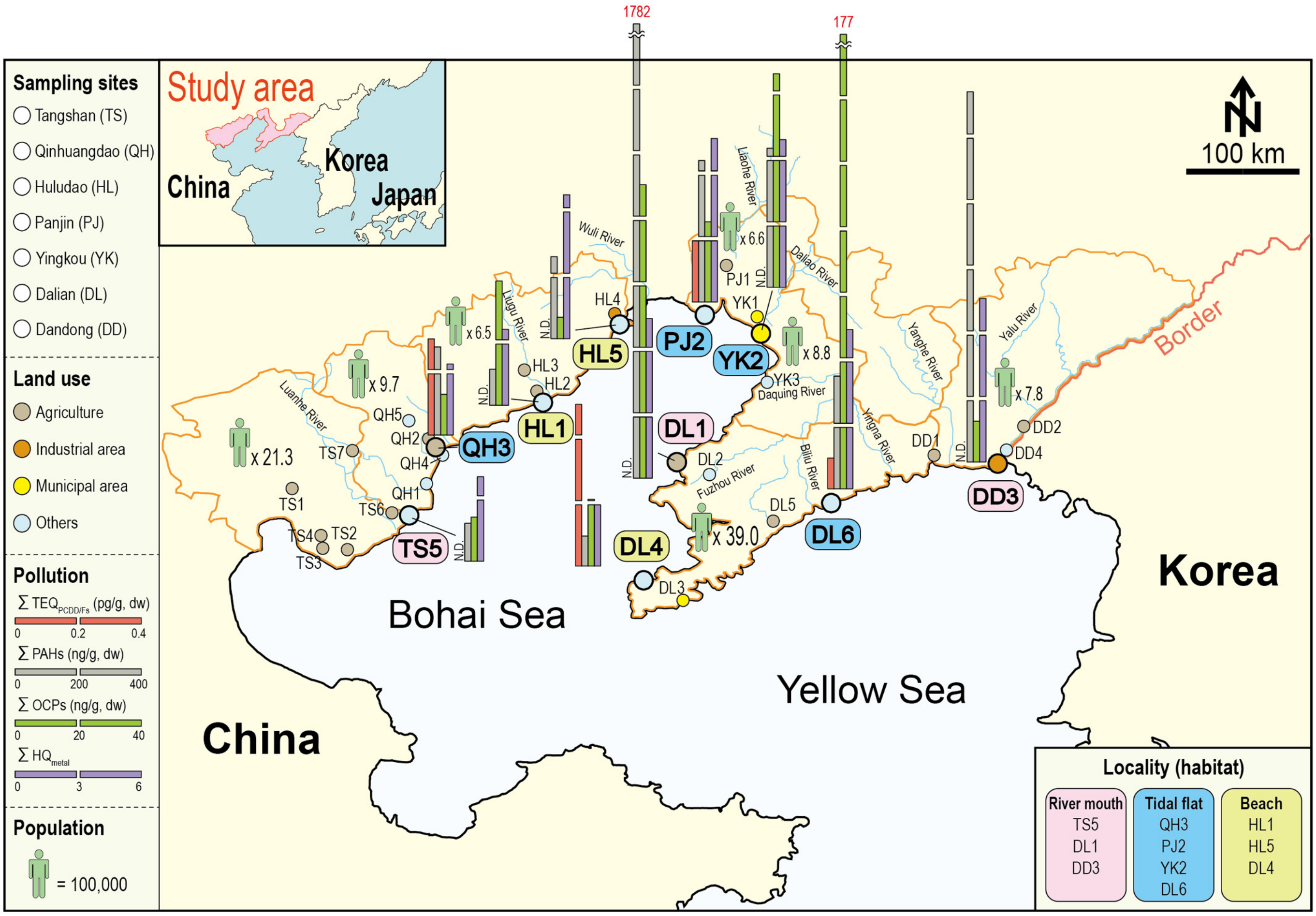


Fig. 1. Map showing the study area and sampling locations along the coastal areas of the Bohai Sea and the northern Yellow Sea. Ten sampling sites are depicted as open circles in different colors according to their land use types. The epithet of each site has also been highlighted in different colors according to the habitat type, i.e. river mouth, tidal flat, and beach. Concentrations of persistent toxic substances (PTSs) and metals and metalloids reported in the locations were provided to present the degree of chemical pollution. All chemicals are listed in Table 1 with basic statistics. Populations of the eight districts to which study sites belong were also given. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2.4. Meiofaunal community

Meiobenthic animals were first segregated from the formalin-fixed sediments by use of a 63 μm sieve. Each individual was subsequently sorted and preserved in 80% ethanol before identification based on morphologies observed under the microscope (Olympus SZ11 or Olympus BX60). Identifications of animals were made to the lowest practical taxonomic level of Nematoda, Copepoda, Sarcomastigophora, Polychaeta, Nemertea, Ostracoda, Bivalvia, Tardigrada, Halacaroidea, Gastrotricha, and Priapulida.

2.5. Data collection

In this study, in order to conduct an integrated sediment assessment by adopting the SQT, all data on contaminants, obtained from previous studies, were combined with data on the meiofaunal community. Concentrations of residues consisted of two major classes of environmental pollutants, viz., PTSs and metals and metalloids. PTSs included PCDD/Fs (Naile et al., 2011), PAHs (Jiao et al., 2012), and organochlorine pesticides (OCPs) such as DDTs, HCHs, and HCB (Hu et al., 2010); and perfluorinated chemicals (PFCs) (Wang et al., 2011), of which sources, concentrations, and distribution had been previously reported. Target metals and metalloids consisted of As, Cd, Cr, Cu, Hg, Ni, Pb, and Zn, quantified in coastal sediments (Luo et al., 2010, 2012).

Result of the H4IIE-*luc in vitro* bioassay, previously reported for the same locations (Hong et al., 2012), were utilized to integrate potencies of dioxin-like chemicals in organic extracts that act through activation of the aryl hydrocarbon receptor. Potencies of organic extracts of sediments are given as 2,3,7,8 tetrachlorodibenzo *p* dioxin equivalents (TCDD-EQ). The microbiome, consisting of bacteria and fungi in sediments was formerly assessed by use of e-DNA (Xie et al., 2017). Chao1 richness was calculated for bacteria and protists (Xie et al., 2017) and used for the index of sediment microbiome community health. Information on the benthic meiofaunal community used in the SQT is newly obtained as part of the present study. All abbreviations used in the present study are presented in Table S1 of the Supplementary materials. Sample preparation and determination of target elements reported previously are summarized in Table S2. In addition, concentrations of PTSs and metals and metalloids in sediments of the Bohai Bay are presented in Table S3.

2.6. Data analyses and statistics

Numbers and densities of observed taxa of meiofauna were collectively used for the index of meiofaunal community health. Of note, Copepoda was able to be identified to copepodid and nauplius and Harpacticoida (order of Copepoda) was identified to species level. Statistical analyses were performed by use of SPSS 23.0 (SPSS INC., Chicago, IL), PRIMER software (Clarke and Gorley, 2006), and SigmaPlot 13.0. Spearman rank correlation coefficients were used to address cross-associations of target characteristics; properties of sediments, concentrations of

PTSs, metals and metalloids, toxic potencies of sediments, and the meiofaunal community, except for scarcely occurring taxa at one or two locations were excluded. Cluster analysis (CA) and nonmetric multidimensional scaling (NMDS) were used to investigate relations between ecological data (bacterial and meiofaunal communities) and certain environmental factors. The similarity matrix for CA and NMDS was calculated using Bray-Curtis similarity (BCS) based on: alpha diversity index and evenness of Bacteroidetes and Proteobacteria; fourth-root transformed abundance data of six meiofauna (Nematoda, Copepoda, Sarcomastigophora, Polychaeta, and Ostracoda). Gastrotiricha, Tardigrada, Bivalvia, Halacaroidea, and Priapulida were excluded from the calculation of similarity because a few individuals ($n < 5$) occurred in only one or two locations among whole sites.

The integrated assessment used five characteristics of sediments, including concentrations of PTSs, metals and metalloids, toxic potencies, sediment-microbiome, and meiofauna data, and ratio to mean (RTM) values were calculated (Cesar et al., 2009). RTM scores were normalized to values of 0–3 for each characteristic then represented as five-axis plots to produce multiple circular sectors indicating contributions of each quality (Lee et al., 2018). This five-axis plotting in SQT assessment was first applied in the present study to visualize associations among sediment characteristics. It should be noted that the visualization of quality status in each region refers to the corresponding locations, which aids simple visual comparison only.

3. Results and discussion

3.1. General feature of chemical pollution

Concentrations of targeted PTSs and metals and metalloids, particularly elevated concentrations of some OCPs, indicated that the study area was moderately to severely polluted (Table 1). For example, among the six groups of PTSs, HCHs ($n = 10$) and DDTs ($n = 8$) exhibited greater concentrations compared to corresponding sediment quality guidelines (SQGs), such as threshold effects level (TEL) or probable effect level (PEL) at most of the locations (CCME, 1999). In particular, concentrations of HCHs exceeded the PEL (0.99 ng g^{-1} dw) at all locations, with a maximum of 150-fold at location DL6 (Table 1), thus HCHs were designated to be chemicals of priority concern in the area. Meantime, PAHs seemed to be another chemical group of concern because two locations (DL1 and DD3) contained PAHs (sum of 13 PAHs) exceeding the TEL (768 ng g^{-1} dw) (Table 1).

Among the eight metals and metalloids measured, concentrations of five elements, As, Cr, Cu, Ni, and Pb were greater than the corresponding SQGs, for more than one location. In particular, concentrations of As exceeded the TEL (7.24 ng g^{-1} dw) at 8 out of 10 locations. When compared to background concentrations of target metals, measured in dated sediment cores to a depth corresponding to 190 years ago, Cd, Pb, and Zn were the most enriched metals compared to others in more than half of the locations (Luo et al., 2010). Correlations between metals indicated common sources, for Cr, Cu, Ni, and Zn, all of which exhibited significant correlations to other metals and metalloids (≥3 elements).

Sites most severely contaminated by metals and metalloids were mostly situated in the inner areas of the Bohai Sea (HL5, PJ2, YK2, and DL1) and one location (DL6) in the northern Yellow Sea, which indicated point sources from nearby inland activities. It should be also noted that relatively greater concentrations of metals and metalloids were evidenced at DD3, which is located downstream on the Yalu River (Dandong), close to the mouth of the river, which indicated possible point sources from the upstream region (Fig. 1). The Tangshan district has the largest population, followed by Dalian, while Dandong had the smallest population (National Bureau of Statistics of China, 2010), thus population itself could not be the only factor influencing the degree of pollution in the study area. Overall, chemical pollution data suggested that distributions of sedimentary PTSs and metals were generally associated with surrounding inland activities in the study area. There were several hot spots and management of site-dependent priority chemicals of concern was suggested.

Table 1
Data structure analyzed in the present study with basic statistics (All abbreviations of specific target chemicals and raw data are presented in Tables S1 and S2 of the Supplementary materials, respectively).

Target elements	Target	Unit	Range	Mean	SQGs[a]	# >SQG[b]	References
			(Min.–max.)	(Mean)	(TEL or ERL/PEL or ERM)		
Sediment property	TOC	%	0.030–2.8	0.82			This study
	TN	%	0.020–0.21	0.090			
	$\delta^{13}C$	‰	−24.7–19.9	−22.4			
Persistent toxic substances	$TEQ_{PCDD/Fs}$[c]	pg g^{-1} dw	0.001–0.47	0.1	0.85/21.5	0	Naile et al., 2011
	PAHs[d]	ng g^{-1} dw	98–1800	550	768/7071	2	Jiao et al., 2012
	DDTs[e]		1.7–87	17	4.48/386	8	Hu et al., 2010
	HCHs[f]		2.6–150	24	0.32/0.99	10	
	HCB		0.10–18	2.5	20/24000	0	
	PFCs[g]		0.10–2.2	0.50			Wang et al., 2011
Metals and metalloids	As	mg kg^{-1} dw	6.0–12	9.0	7.2/41.6	8	Luo et al., 2010
	Cd		0.070–0.34	0.14	0.7/4.2	0	
	Cr		4.2–78	45	52.3/160	4	
	Cu		3.4–23	15	18.7/108	3	
	Ni		5.5–34	20	21/52	4	
	Pb		15–49	25	30.2/112	3	
	Zn		9.8–100	56	124/271	0	
	Hg	μg kg^{-1} dw	18–54	24	130/700	0	Luo et al., 2012
Sediment toxicity	TCDD-EQ	pg g^{-1} dw	0.90–9.5	4.1			Hong et al., 2012
Bacterial community	Richness	Number	612–11,302	4719			Xie et al., 2017
	Alpha-diversity		6.3–11.6	9.0			
Meiofaunal community	Number of taxa	Number	4–7	5			This study
	Density of taxa	Individual 10 cm^{-2}	11–1074	547			

[a] SQGs: sediment quality guidelines. Threshold effects level (TEL) and Probable effect level (PEL) values were used for $TEQ_{PCDD/Fs}$ PAHs, DDTs, HCHs, As, Cd, Cr, Cu, Pb, Zn, and Hg (CCME, 1999), effects range low (ERL) and effects range median (ERM) values were used for Ni (Long et al., 1995), and TEL and PEL values were used for HCB (Persaud et al., 1993).

[b] Number of sites exceeded the SQGs among ten sites.

[c] $TEQ_{PCDD/Fs}$ (Toxic equivalents of PCDD/Fs): Sum of TCDD, PeCDD, HxCDD, HpCDD, OCDD, TCDF, PeCDF, HxCDF, HpCDF, and OCDF concentrations.

[d] PAHs: Sum of Nap, Acl, Ace, Flu, Phe, Ant, Fl, Py, BaA, Chr, BbF, BkF, BaP, IcdP, DBahA, and BghiP concentrations.

[e] DDTs: Sum of *p,p′*-DDE, *p,p′*-DDD, *o,p′*-DDT, and *p,p′*-DDT concentrations.

[f] HCHs: Sum of α-HCH, β-HCH, γ-HCH, and δ-HCH concentrations.

[g] PFCs: Sum of PFBA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFDoA, PFOS, and PFDS concentrations.

3.2. Benthic environment and meiofaunal community structure

Samples of sediment, collected from 10 locations, in varying geological locality along the coastal areas of the Bohai and northern Yellow Seas, represented typical bottom conditions. For example, all samples collected from tidal flat areas (n = 4; QH3, PJ2, YK2, and DL6) exhibited greater concentrations of TOC and TN, compared to those of other samples (Fig. 2a), while samples collected from beach areas (n = 3; HL1, HL5, and DL4) consisted mainly of sand and contained the least concentrations of TOC (<0.1%) and TN (<0.05%). Carbon stable isotope signatures generally indicated that DL4 and DL6 locations are more influenced by marine organic materials, while YK2 was more influenced by terrestrial sources (Fig. 2a). Sediment property data generally exhibited a concentration gradient in the three representative habitats from inland-river to tidal flat and beach in terms of organic enrichment.

Meiofaunal species were identified to the lowest possible taxonomic rank e.g. subphylum, class, subclass, or species. A total of eleven meiofaunal groups viz. Nematoda, Copepoda, Sarcomastigophora,

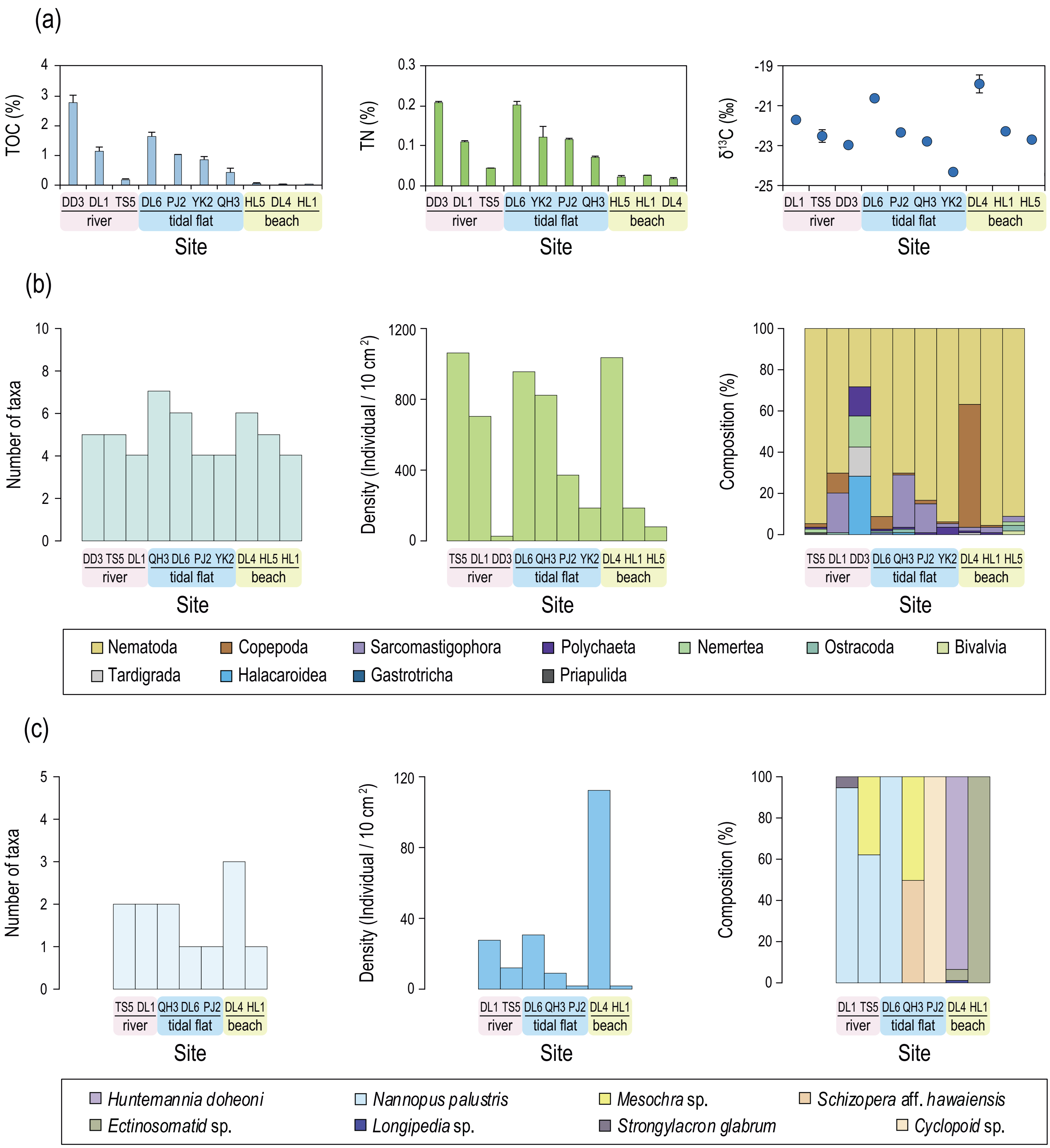


Fig. 2. Overview data of sediment properties and benthic community structure in the coastal areas of the Bohai Sea and the northern Yellow Sea; (a) total organic carbon (TOC), total nitrogen (TN), and stable carbon isotope ratio ($\delta^{13}C$), (b) benthic meiofaunal community, and (c) diversity of benthic Copepoda.

Polychaeta, Nemertea, Ostracoda, Bivalvia, Tardigrada, Halacaridae, Gastrotricha and Priapulida were observed and identified from sediments (Fig. 2b). Overall, in terms of distribution as well as species abundances, Nematoda was the dominant taxa followed by Copepoda and Sarcomastigophora (as based on density). It is notable that Nematoda comprised >80% of total meiofaunal composition from more than half of the samples (6 of 10 locations; TS5, HL1, HL5, PJ2, YK2, and DL6). Meanwhile, Gastrotricha occurred only in DL6 with two individuals and Priapulida occurred only in TS5 with five individuals, which exhibited a lack of meiozoobenthos diversity in the open coastal region. At present, occurrences of meiofauna did not seem to be related to geographical locality or habitat type.

Among the eleven meiofaunal taxa, harpacticoid copepods could be identified to the species level and the very taxonomic group species occurred in 7 out of 10 locations (Fig. 2c). However, the number of copepod species was small, mostly one or two except in DL4 where three species were found. DL4 also exhibited not only the greatest density of Copepoda with a dominance of *Huntemannia hoheonii*, but also showed the second greatest density of overall meiofaunal species. Furthermore, it was the only location where the numbers of individuals of Copepoda was greater than that of Nematoda (Fig. 2b).

3.3. Benthic community response to chemical pollution

There seemed to be relationships or associations between chemical pollution and structure of the meiofaunal community. Numbers of taxa and/or abundances were less in severely polluted locations (Figs. 1 and 2). Thus, potential pollutants that influence structures of meiofaunal communities were assessed. First, relationships between concentrations of individual target chemicals (Table 1) and abundances of benthic meiofauna were analyzed (Fig. 3). Target meiofaunal taxa included Nematoda, Copepoda, Sarcomastigophora, Nemertea, Ostracoda,

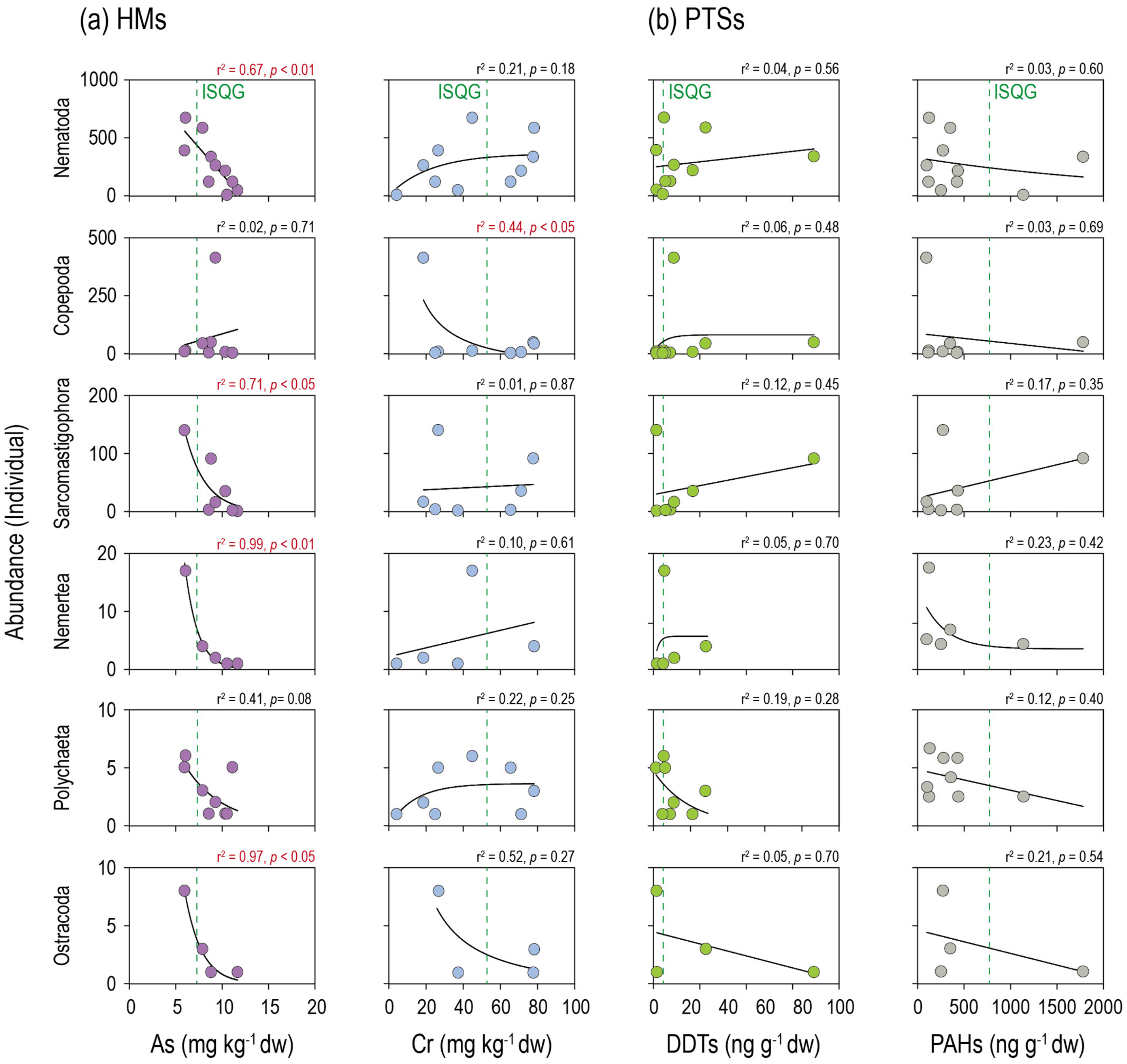


Fig. 3. Relationship between chemical pollution and meiofaunal abundances of six selected taxa (Nematoda, Copepoda, Sarcomastigophora, Nemertea, Polychaeta, and Ostracoda) in the coastal areas of the Bohai Sea and the northern Yellow Sea; (a) metals and metalloids: As and Cr and (b) persistent toxic substances (PTSs): DDTs and PAHs.

and Polychaeta. Four taxa of Gastrotricha, Tardigrada, Bivalvia, and Halacaroidea were excluded due to limited cross sampling locations. Results of that analysis indicated that the chemical pollutant most likely causing the observed effects was As, which exhibited significant, strong, negative correlations with five of the six target taxa, Nematoda, Sarcomastigophora, Ostracoda, Nemertea, and Polychaeta. In particular, numbers of Nemertea and Ostracoda exhibited negative correlations with concentrations of As with significant coefficients of determination ($r^2 > 0.9$). Relationships between the six taxa and other metals and metalloids were mostly weaker and not statistically significant, except for the association between Copepoda and Cu or Ni (Fig. S1). This result suggested that abundances of meiofauna can be a proxy of ecological index for estimating potential pollution by metals and metalloids.

In general, negative relationships with abundances of meiofauna were also observed for PTSs, but the associations were not as prominent as they were for metals (Figs. 3b and S2). For example, all meiofaunal taxa except Sacromastigophora were inversely proportional to concentrations of PAHs (Fig. 3b), with r^2 between 0.03 and 0.23 and *p* values of 0.40–0.69. Meanwhile, only Polychaeta and Ostracoda exhibited negative correlations with concentrations of DDTs, but those correlations were not statistically significant. Meiofaunal species did not seem to be affected by the gradient of PTSs such as PAHs or DDTs, although their concentrations exceeded the corresponding SQGs (Table 1). Overall, the spearman rank correlation analysis supported a conclusion that abundances of meiofauna exhibited relatively strong, negative associations with concentrations of metals and metalloids, but were insignificant for organic matters or PTSs (Table 2).

Results of previous studies have suggested that benthic meiofaunal communities and their biodiversity are related to effects of sedimentary pollution (Raffaelli and Mason, 1981; Raffaelli, 1982; Moore, 1987; Moore and Bett, 1989; Sutherland et al., 2007). Results of the present study revealed that lesser densities of Nematoda ($r = -0.83$, $p < 0.01$) and Nemertea ($r = -0.82$, $p < 0.01$) were observed in sediments more contaminated with As, and lesser densities of Copepoda ($r = 0.78$, $p < 0.05$) were found in locations affected by terrestrial organic matter showing lighter values of $\delta^{13}C$ (Table 2). Thus, benthic meiofauna was an indicator of benthic ecosystem health affected by chemical contaminations. However, the benthic meiofaunal communities were not fully explained by presence of sedimentary organic matter, PTSs, metals, or metalloids (i.e., not significant; *p* value > 0.05). This seems to be due to the influence of other environmental factors on meiofaunal community, such as free sulfide concentrations, redox potential, and grain size of sediment (Sutherland et al., 2007). Benthic meiofauna are good indicators to sedimentary pollution as a result of their small size (i.e., easy to sampling), sensitivity to toxic substances, short generation times, high reproduction rates, and direct benthic recruitment (Sutherland et al., 2007). Thus, it is expected that the meiofauna will be useful as an important target element for assessing benthic ecosystem health.

3.4. Integrated benthic quality assessment: extended-SQT

CA and NMDS were conducted to better understand the complex positive and/or negative relationships among environmental parameters, chemical pollutants, sediment toxicity, sediment-microbiome, and meiozoobenthos diversities in the study area (Fig. 4). The results of CA and NMDS showed that two groups were categorized in >85% of BCS indices, and communities of Nematoda, Copepoda, and Sarcomastigophora were distributed as function of concentrations of HCB, As, and Cd. Nematoda was the most abundant at less contaminated sites (TS5 and DL6 in Group 1) and also widely occurred in moderately contaminated sites (DL1, DL4, and PJ2 in Group 2). Copepoda generally occurred at locations near Dalian in Group 2, and were especially most abundant in DL4. The occurrence of Sarcomastigophora seemed to indicate that they were relatively less affected by pollutants, and in particular, showed a tendency that was opposite to that of Nematoda and Copepoda. Yet, there were no tendency for relation between bacterial communities and environmental variables in the CA and NMDS.

The site-specific abundance of meiofaunal communities in NMDS could be explained by site-specific contamination by certain chemicals such as greater As observed in sediments from HL5, YK2, and DD3, which was negatively associated with meiofaunal assemblage; Nematoda, Copepoda, and Sarcomastigophora (Fig. 4b). HL5, YK2, and DD3 were situated downstream in Rivers; Wuli River, Daliao River, and Yalu River, respectively. Thus, sediments seemed to be influenced by brackish conditions.

To further understand the overall status of the benthic habitat of the study area, the SQT approach was applied (Fig. 5). First, three components comprising chemical pollution (Fig. 5a), sediment toxicity (Fig. 5b), and benthic community structure (Fig. 5c) were determined based on RTM values of each component separately. Degree of contamination and/or ranking of the three components varied among locations, which affected dynamic and complex associations between and among components. Thus, it is reasonable to combine the above three components to estimate the overall status of sediment pollution, where a few hot spot locations were identified, QH3 and PJ2. QH3 has been identified as severely contaminated site by PTSs, particularly PFCs, while PJ2 contained elevated concentrations of metals and metalloids, for As, Cr, Cu, and Ni, exceeding the corresponding SQGs.

Finally, five-angular circular sectors representing the degree of quality in five benthic quality elements, comprising PTSs, metals and metalloids, sediment toxicity, sediment-microbiome, and meiofaunal community, were produced for each location, as extended-SQT. This was done to separate effects of PTSs and metals and metalloids in

Table 2

Spearman rank (r) correlations between meiofaunal community and sediment characteristics, persistent toxic substances (PTSs), metals and metalloids, and sediment toxicity (ST) in the coasts of Bohai Sea and northern Yellow Sea.

Meiofauna	Sediment property			PTSs				Metals and metalloids								ST
	TOC	$\delta^{13}C$	TN	$TEQ_{PCDD/Fs}$	OCPs	PAHs	PFCs	As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	TCDD-EQ
Sarcomastigophora	0.43	0.71	0.18	−0.5	0.14	0.39	**0.90**[*]	−0.75	−0.18	0.18	0.18	0.04	0.14	−0.32	−0.50	0.64
Nematoda	0.02	0.31	−0.07	0.20	0.35	−0.17	0.23	**−0.83**[**]	−0.43	0.49	−0.01	0.33	0.20	−0.55	−0.34	0.22
Gastrotricha	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Tardigrada	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Nemertea	−0.05	0.21	−0.05	–	0.67	−0.41	−0.87	**−0.98**[**]	−0.87	0.72	0.01	0.62	0.60	−0.87	−0.40	0.10
Bivalvia	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Polychaeta	−0.16	−0.27	−0.10	0.40	−0.03	−0.25	−0.15	−0.45	−0.10	0.32	−0.15	0.45	0.02	−0.57	−0.13	0.03
Halacaroidea	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–
Copepoda	0.16	**0.78**[*]	−0.26	0.8	0.01	−0.14	0.20	−0.24	−0.48	0.48	−0.24	0.38	−0.12	−0.02	−0.31	−0.21
Ostracoda	0.62	−0.67	0.62	−0.5	0.05	0.46	–	−0.82	−0.15	0.36	0.21	−0.15	0.36	0.21	0.21	0.82
Priapulida	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–	–

— mean not analyzed and $N \le 2$.

[*] $p < 0.05$.

[**] $p < 0.01$.

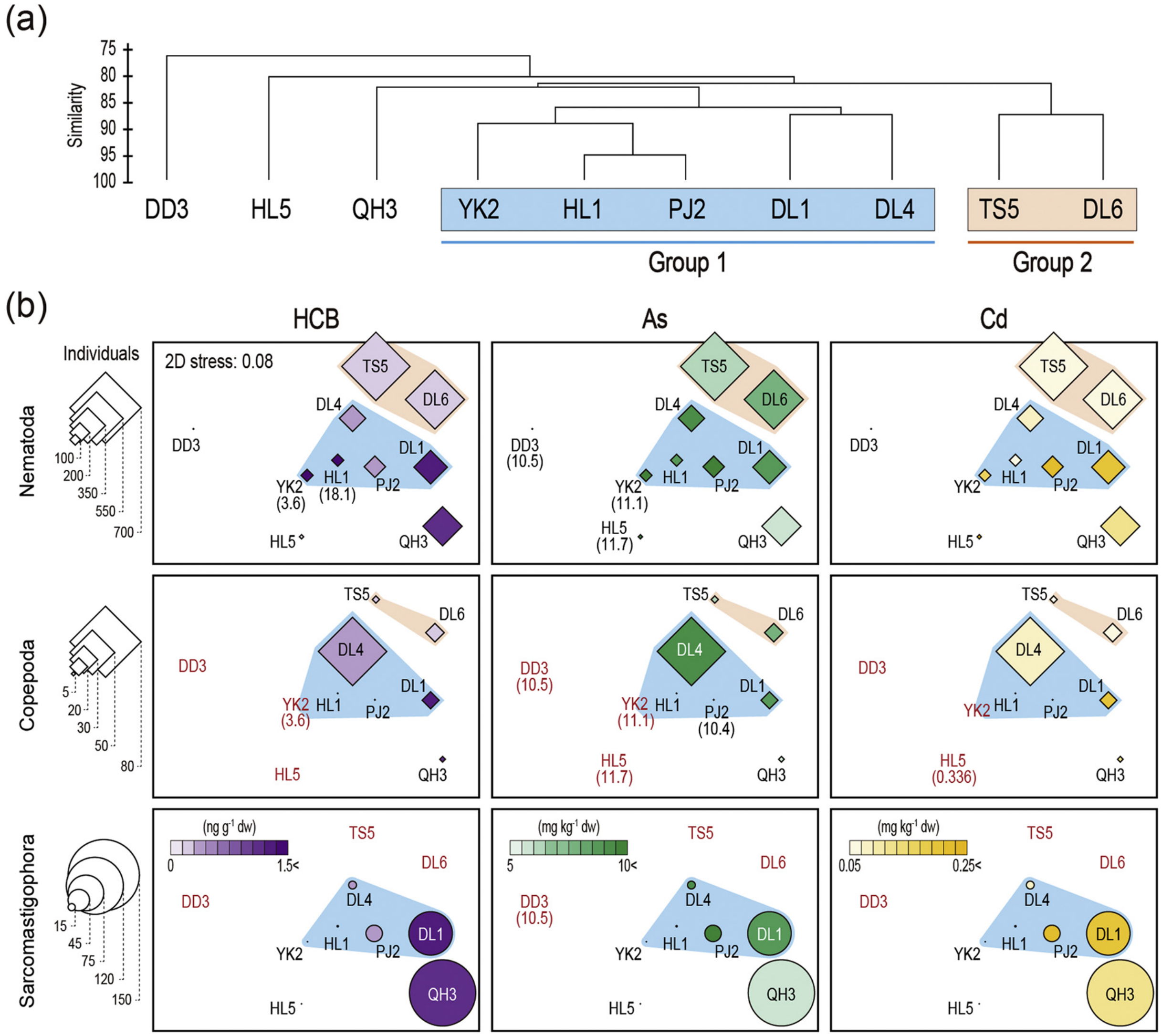


Fig. 4. Results of (a) a cluster analysis (CA) and (b) ordination plots of nonmetric multidimensional scaling (NMDS) from the metadata of sediment-microbiome and meiobenthic taxa in the coastal areas of the Bohai Sea and northern Yellow Sea. The concentrations of HCB, As, and Cd in coastal sediment were cross-compared with individuals of three taxa (Nematoda, Copepoda, and Sarcomastigophora) in NMDS plots. Locations: 1) in red indicated not detected individuals with each corresponding taxa, and 2) with noticeably high concentration of pollutants were indicated in parentheses. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

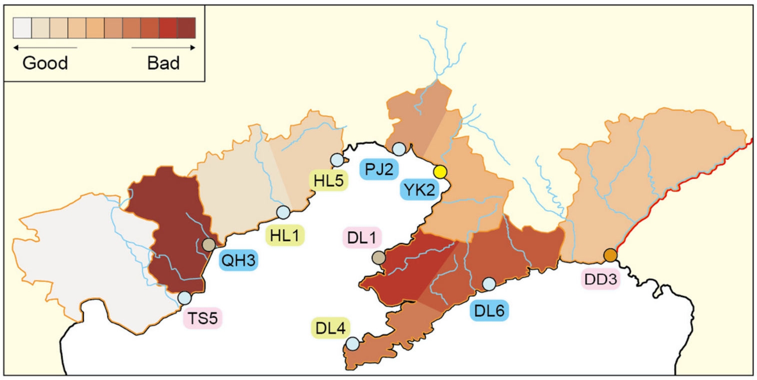
association to sediment toxicity and benthic community responses, which aids characterization of site-specific and/or chemical-specific pollution. The color of each circle represents the state of benthic habitat. This visualized image was found to have associations between five benthic quality elements. For example, sediment toxicity data seemed to be associated with benthic community responses, but less polluted locations generally exhibited lesser toxicity of sediments. Although the *in vitro* sediment toxicity utilized in the present study could not provide the overall toxicity of contamination of mixture compounds, the toxicity component could contribute as one extended triad component for comparisons (Khim and Hong, 2014).

More polluted locations, particularly the four most contaminated locations (QH3, PJ2, YK2, and HL5) contained elevated concentrations of either PTSs or metals and metalloids. Of note, among these, three locations (PJ2, YK2, and HL5) were located in the most inner areas of the Bohai Sea and another set of three locations (QH3, PJ2, and YK2) were situated in tidal flats. In general, severe pollution in this area was attributable to combined features of geological/oceanographic factors or bottom habitat conditions. Overall, the metals and metalloids were key contaminants and apparently negatively associated with the structure of meiofaunal communities (Fig. S3). Thus, special attention should be given for future monitoring and management, particularly in the inner areas of the Bohai Sea. Finally, it would be reasonable to assume that there were unknown and/or unmeasured chemical pollutant(s) that might have influenced responses of the benthic community (Khim and Hong, 2014). Altogether, the multiple lines of evidences approach is recommended to further address missing components of the SQT.

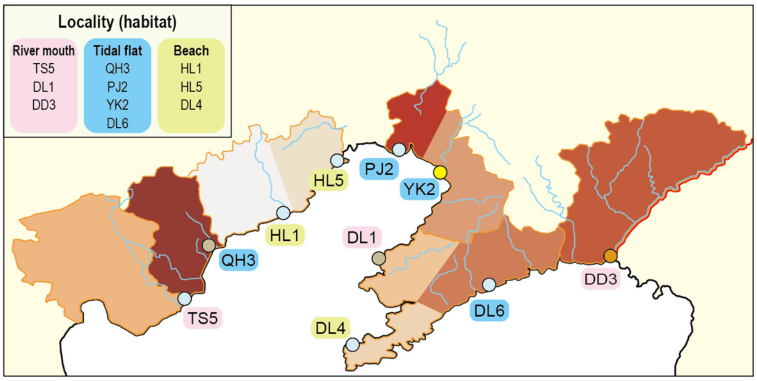
4. Conclusions

More than half of the locations were identified as moderately to severely contaminated sites with various PTSs and metals and metalloids, accordingly relatively low meiozoobenthos diversity was evidenced. Concentrations of PAHs and OCPs and As, Cr, Cu, Ni, and Zn in sediments exceeded their corresponding SQGs for two to ten locations of Bohai Bay and northern Yellow Sea. HCHs and As were the priority chemicals of concern in the study area, and particularly As seemed to greatly influence diversity and abundances of meiofauna. The extended SQT using five target benthic elements: 1) PTSs; 2) metals and metalloids; 3) sediment-toxicity; 4) sediment-microbiome; and 5) structure of the meiofaunal community, successfully demonstrated their strong and/or weak association towards integrated benthic quality assessment. In general, responses of the meiofaunal community and sediment toxicity were less sensitive to effects of metals and metalloids than was

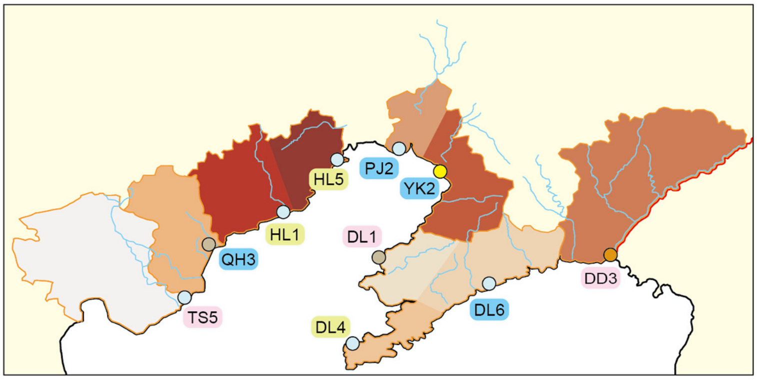
(a) Chemical Pollution (PTSs + HMs)



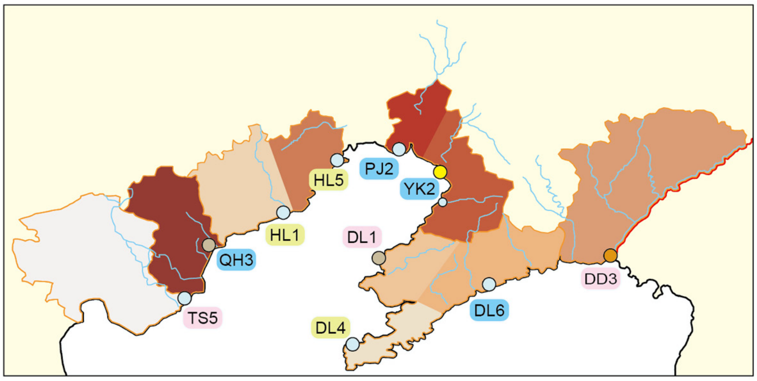
(b) Sediment Toxicity (ST)



(c) Benthic Community (Microbiome (Mi) + Meiofauna (Me))



(d) Combined result of (a), (b), & (c)



(e) Advanced-SQT (Five benthic quality objectives)

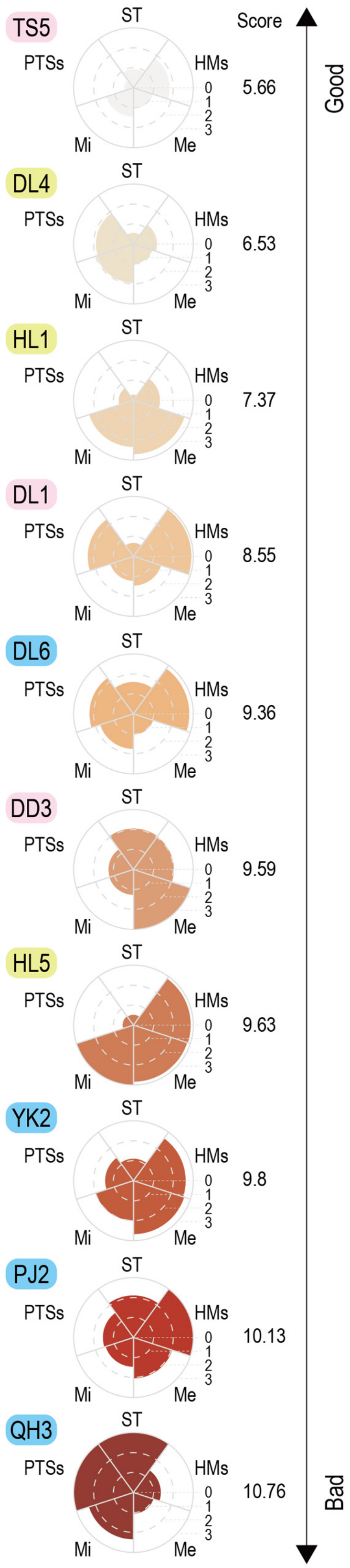


Fig. 5. Integrated benthic quality assessment in the coastal areas of the Bohai Sea and northern Yellow Sea by use of sediment quality triad (SQT) method. Sediment qualities depicted in terms of (a) chemical pollution, (b) sediment toxicity, (c) benthic community (sum of microbiome (Mi) and meiofauna (Me)), (d) combined result of (a), (b), and (c), and (e) extended-SQT, comprising five benthic quality elements, including: 1) PTSs; 2) metals and metalloids; 3) sediment toxicity; 4) sediment-microbiome; and 5) meiofaunal community given as 'five-angular circular sectors' for each location. The order given from low to high pollution score (sum of 5 RTM values) indicating degree of pollution.

structure of the benthic community. Overall, the present study emphasized the importance of adopting the multiple lines of evidence approach, particularly when assessing large coastal ecosystems with various land use activities and/or multiple sources of pollution, such as in the Yellow Sea environment.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.07.169.

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<Supplementary Materials>

Chemical-, site-, and taxa-dependent benthic community health in coastal areas of the Bohai Sea and northern Yellow Sea: A sediment quality triad approach

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Supplementary Tables

Table S1. List of abbreviation used in the present study.

Abbreviation	Description	Abbreviation	Description
Ace	Acenaphthene	PAHs	Polycyclic aromatic hydrocarbons
Acl	Acenaphthylene	PCA	Principle component analysis
Ant	Anthracene	PCDD/Fs	Polychlorinated dibenzo-*p*-dioxin and dibenzofuran
BaA	Benzo[*a*]anthracene	PeCDD	Pentachlorodibenzo-*p*-dioxin
BaP	Benzo[*a*]pyrene	PeCDF	Pentachlorodibenzofuran
BbF	Benzo[*b*]fluoranthene	PEL	Probable effect level
BghiP	Benzo[*g*,*h*,*i*]perylene	PFBA	Perfluorobutanoic acid
BkF	Benzo[*k*]fluoranthene	PFCs	Perfluorinated chemicals
CCME	Canadian Council of Ministers of the Environment	PFDA	Perfluorodecanoic acid
Chr	Chrysene	PFDoA	Perfluorododecanoic acid
DBahA	Dibenzo[*a*,*h*]anthracene	PFDS	Perfluorodecane sulfonate
DDD	Dichlorodiphenyldichloroethane	PFHpA	Perfluoroheptanoic acid
DDE	Dichlorodiphenyldichloroethylene	PFHxA	Perfluorohexanoic acid
DDT	Dichlorodiphenyltrichloroethane	PFNA	Perfluorononanoic acid
ERL	Effects range low	PFOA	Perfluorooctanoic acid
ERM	Effects range median	PFOS	Perfluorooctane sulfonic acid
Fl	Fluoranthene	PFUnA	Perfluoroundecanoic acid
Flu	Fluorene	Phe	Phenanthrene
HCB	Hexachlorobenzene	PTSs	Persistent toxic substances
HCHs	Hexachlorocyclohexane	Py	Pyrene
HpCDD	Heptachlorodibenzo-*p*-dioxin	RTM	Ratio to mean
HpCDF	Heptachlorodibenzofuran	SQG	Sediment quality guideline
HxCDD	Hexachlorodibenzo-*p*-dioxin	SQT	Sediment quality triad
HxCDF	Hexachlorodibenzofuran	TCDD	Tetrachlorodibenzo-p-dioxin
IAEA	International Atomic Energy Agency	TCDF	Tetrachlorodibenzofuran
IcdP	Indeno[*1*,*2*,*3*,-*c*,*d*]pyrene	TEL	Threshold effects level
Nap	Naphthalene	TEQ	Toxic Equivalent
NOAA	National Oceanic and Atmospheric Administration	TN	Total nitrogen
OCDD	Octachlorodibenzo-*p*-dioxin	TOC	Total organic carbon
OCDF	Octachlorodibenzofuran	$\delta^{13}C$	Carbon stable isotope ratio
OCPs	Organochlorine pesticides		

Table S2. Summary of sample preparation and determination of target elements (chemical, toxicological, and ecological) in sediment samples of the Bohai Bay, China, reported previously.

Elements	**Targets**	**No. of samples**	**Pretreatments (Extraction, isolation, & cleanup)**	**Analysis (Instrumental or bioanalytical)**	**References**
Sediment property	TOC & $\delta^{13}C$	10	· Removed inorganic carbon using 1M HCl	· Elemental analyzer (EA)-isotope ratio mass spectrometry (IRMS)	This study Xie et al., 2017
	TN	10	· No treatment	· Elemental analyzer (EA)	
Persistent toxic substances	17 PCDD/Fs		· Soxhlet extraction with hexane:DCM (1:1, v:v) · Removing elemental sulfur (activated Cu) · Multi-layer silica gel column clean up	· High resolution gas chromatography (HRGC)-high resolution mass spectrometer (HRMS) analysis	Naile et al., 2011
	16 PAHs	35	· Soxhlet extraction with DCM · Silica gel column clean-up · Elution with hexane:DCM (7:3, v:v)	· Gas chromatography (GC)-mass selective detector (MSD) analysis	Jiao et al., 2012
	9 OCPs	35	· Ultra-sonication extraction with hexane:DCM (1:1, v:v) · SPE cartridge clean-up (silica gel) · Elution with hexane:DCM (7:3, v:v)	· Gas chromatography (GC)-electron capture detection (ECD) analysis	Hu et al., 2010
	10 PFCs	35	· Extraction with 1% acetic acid solution · Sonication for 15 min at 60 °C · Centrifugation at 3000 rpm · SPE (Oasis HLB cartridge)	· High performance liquid chromatography (HPLC)-tandem mass spectrometer analysis	Wang et al., 2011
Metals and metalloids	As, Cd, Cr, Cu, Ni, Pb, & Zn	35	· Acid (HNO_3 and H_2O_2) digestion	· Inductively coupled plasma mass spectrometry (ICP-MS) analysis	Luo et al., 2010
	Hg	35	· Acid (3 N HNO_3) or hot alkali (25% NaOH) digestion	· Cold vapor atomic fluorescence spectrometry (CVAFS) analysis	Luo et al., 2012
Sediment toxicity	TCDD-EQ	35	· Soxhlet extraction (DCM) · Removing elemental sulfur (activated Cu) · Solvent exchange to DMSO	· H4IIE-luc bioassay · Luciferase assay (luminometer)	Hong et al., 2012
Benthic community	Bacteria	35	· DNA extracted with MoBio Power Soil DNA Kit · Protistan 18S rRNA genes were amplified by PCR (V9 primers) · Amplification conducted in a SureCycler 8800 Thermal Cycler	· Sequenced in the Ion Proton or Personal Genome Machine (PGM) sequencer	Xie et al., 2017

Abbreviations. DCM: dichloromethane; DMSO: dimethyl sulfoxide; DNA: Deoxyribonucleic acid; PCR: polymerase chain reaction; RNA: Ribonucleic acid; SPE: solid phase extraction.

Table S3. Concentrations of persistent toxic substances and metals and metalloides in sediments of the Bohai Bay reported previously.

Target chemicals^a	**Unit**	**TS5**	**QH3**	**HL1**	**HL5**	**PJ2**	**YK2**	**DL1**	**DL4**	**DL6**	**DD3**	**References**
PCDD/Fs												
2,3,7,8-TCDD	pg/g dw	^{b}										Naile et al., 2011
1,2,3,7,8-PeCDF												
1,2,3,4,7,8-HxCDD												
1,2,3,6,7,8-HxCDD												
1,2,3,7,8,9-HxCDD												
1,2,3,4,6,7,8-HpCDD									0.66			
OCDD			18			5.7			5.8	38		
2,3,7,8-TCDF									0.83			
1,2,3,7,8-PeCDF			0.65						0.50			
2,3,4,7,8-PeCDF			0.65						0.66			
1,2,3,4,7,8-HxCDF						0.36			0.83			
1,2,3,6,7,8-HxCDF												
2,3,4,6,7,8-HxCDF									0.50			
1,2,3,7,8,9-HxCDF												
1,2,3,4,6,7,8-HpCDF			4.3			0.71			1.7			
1,2,3,4,7,8,9-HpCDF												
OCDF			6.5			1.4			1.3			
∑PCDD/Fs			30			8.2			13	38		
∑TEQ$_{PCDD/Fs}$			0.33			0.060			0.47	0.11		
PAHs												
Nap	ng/g dw	2.5	3.7	3.4	2.9	12	8.0	49	3.6		20	Jiao et al., 2012
Acl		0.24	0.55	2.3	2.0	4.5	4.0	4.4	1.3	2.3	12	
Ace		0.74	2.1	5.8	3.4	19	3.1	16		29	7.9	
Flu		2.5	6.2	3.1	4.3	16	12	52	2.4	15	24	
Phe		30	30	8.4	29	48	56	190	8.6	56	110	
Ant		2.5	4.3	1.8	3.3	8.0	8.4	26	1.1	5.8	28	
Fl		12	27	7.6	25	46	47	200	8.0	33	130	
Py		15	26	6.0	34	45	50	160	8.2	38	110	
BaA		7.5	22	6.8	22	33	35	140	13	18	98	
Chr		5.7	25	4.3	18	31	32	140	2.6	19	83	
BbF		10	51	21	28	78	75	370	20	59	210	
BkF		1.6	6.9	4.4	6.8	5.8	5.0	34	2.1	2.6	35	
BaP		11	24	10	24	30	30	100	7.6	19	85	
IcdP		12	25	20	30	39	40	200	12	37	130	
DBahA		1.5	1.0	1.0	1.6	2.7	1.1	8.3	1.4	0.77	3.3	

BghiP		8.0	19	13	19	18	19	92	6.9	17	55	
∑PAHs		120	270	120	250	430	430	1800	98	350	1100	
OCPs												
α-HCH				1.5		0.26	3.4	0.23			0.21	Hu et al., 2010
β-HCH		8.2	12	8.5		2.7	52	1.6			7.4	
γ-HCH				0.60	4.9			0.52	9.2			
δ-HCH		0.30	0.24	1.2	0.27	0.21	0.28	0.30		150	0.23	
∑HCHs		8.5	12	12	5.2	3.2	55	2.7	9.2	150	7.9	
p, *p*′-DDE		0.49		1.3	0.44	0.65	3.7	4.9	10	2.2	0.30	
p, *p*′-DDD		2.6		1.1		4.0	0.60	8.9		12	1.6	
o, *p*′-DDT		0.51		0.81		3.3		13		3.4		
p, *p*′-DDT		2.2	1.7	6.1	1.4	13	2.3	59	0.99	11	3.3	
∑DDTs		5.9	1.7	9.2	1.9	21	6.6	86	11	28	5.2	
HCB			1.1	18		0.31	3.6	1.2	0.28		0.27	
PFCs												
PFOS	ng/g dw		1.6									Wang et al., 2011
PFOA						0.24						
PFHxA			0.43									
PFHpA												
PFNA			0.11			0.62	0.64					
PFDA												
PFUnA		0.21	0.12	0.42	0.26	0.23					0.24	
PFDoA												
PFDS												
PFBA												
∑PFCs		0.21	2.2	0.42	0.26	1.1	0.64				0.24	
Metals and metalloids												
As	mg/kg dw	6.1	6.0	8.6	12	10	11	8.8	9.3	7.9	11	Luo et al., 2010
Cd		0.070	0.13	0.070	0.34	0.21	0.16	0.20	0.070	0.070	0.15	
Cr		45	27	25	37	71	66	78	19	78	4.2	
Cu		6.2	8.2	6.6	12	19	16	23	3.4	22		
Ni		14	10	9.7	12	34	29	31	5.5	34		
Pb		15	17	17	48	27	23	29	16	30	34	
Zn		30	10	27	100	95	77	76	9.8	90		
Hg	µg/kg, dw	54	21	18	20	20	21	20	26	23	25	Luo et al., 2012

[a] Abbreviation of target chemicals are presented in Table S1.

[b] Blank: below detection limits.

Supplementary Figures

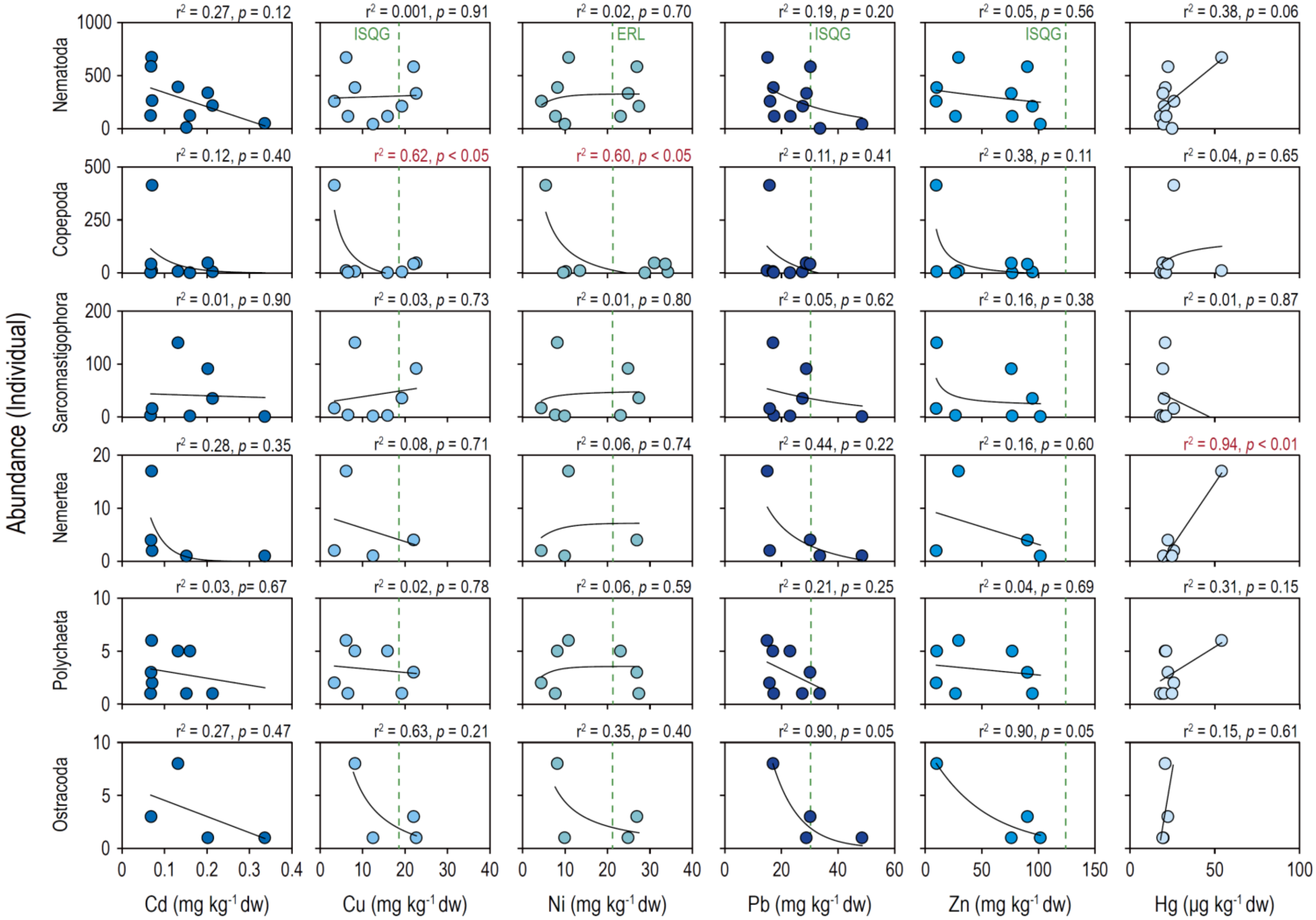

Fig. S1. Relationships between concentrations of heavy metals (Cd, Cu, Ni, Pb, Zn, and Hg) in sediments and abundances of six selected benthic meiofauna (Nematoda, Copepoda, Sarcomastigophora, Nemertea, Polychaeta, and Ostracoda) in the coastal areas of the Bohai Sea and the northern Yellow Sea.

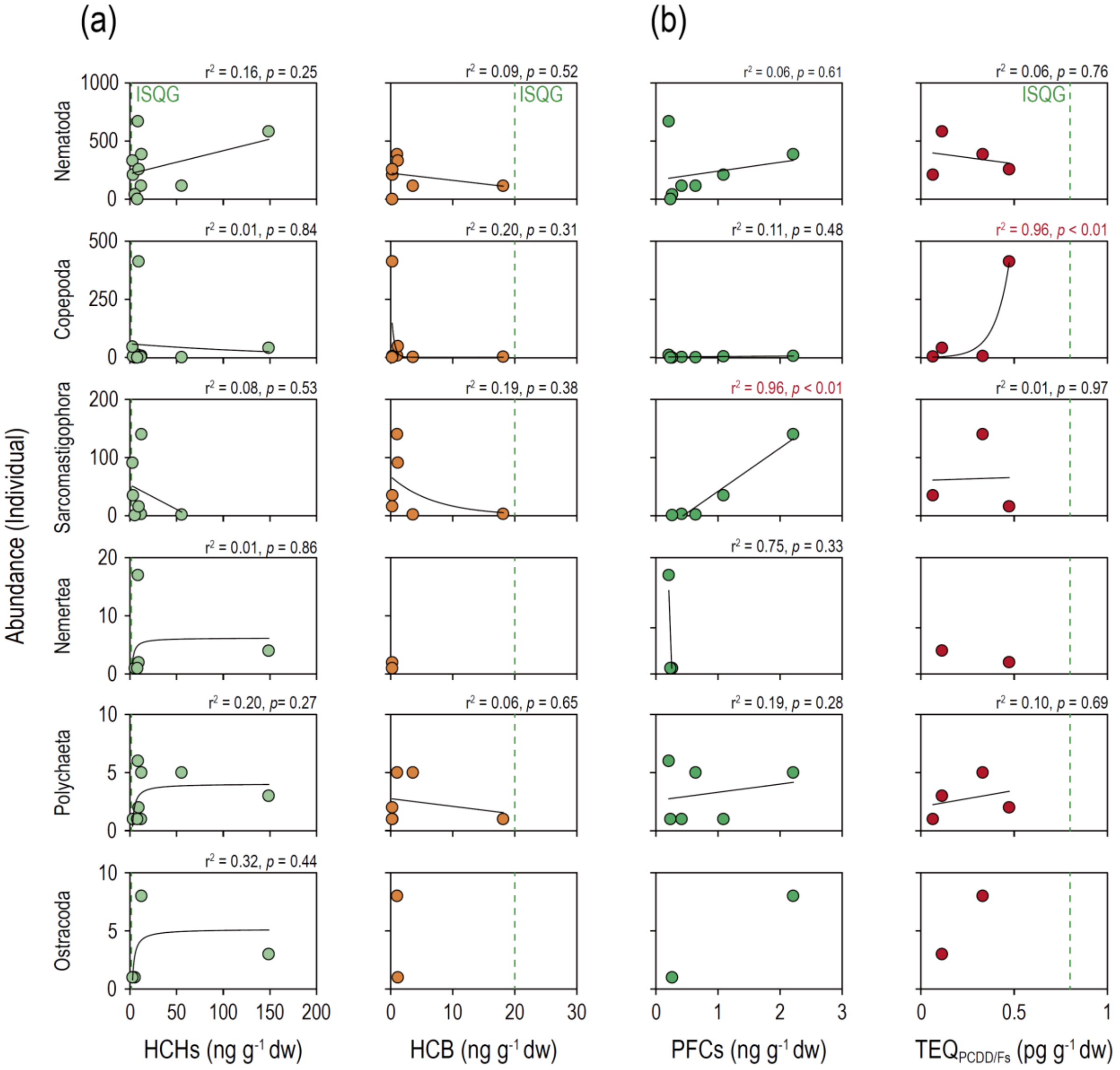

Fig. S2. Relationships between concentrations of trace organic chemicals (HCHs, HCB, PFCs, and $TEQ_{PCDD/Fs}$) and abundances of six selected benthic meiofauna (Nematoda, Copepoda, Sarcomastigophora, Nemertea, Polychaeta, and Ostracoda) in the coastal areas of the Bohai Sea and the northern Yellow Sea.

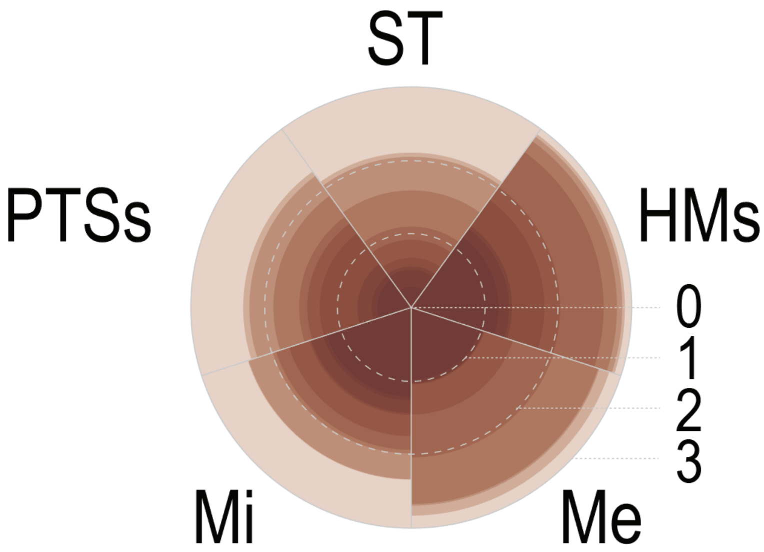

Fig. S3. Summary of results for advanced-sediment quality triad (SQT) assessments in coastal areas of the Bohai Sea and the northern Yellow Sea. The five-angular circular sectors obtained from 10 sites are overlaid. Darker color indicates the more overlapping the data from the each site.

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