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Ecological risk assessment to marine organisms induced by heavy metals in China's coastal waters

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Abstract

China's coastal environment has been heavily affected by the loading of terrestrial pollutants in recent decades, and quantitative risk assessment is urgently needed to assess the ecological risks of China's coastal environment. We assessed the ecological risks induced by five heavy metals (including Cu, Zn, Pb, Hg and As) in China's coastal waters for three groups of marine organisms (including crustacean, fish and mollusc) based on data obtained from a nationwide unified coastal environment monitoring program consisting of 301 sampling sites. The results show that higher heavy metal concentrations occurred more frequently in the Bohai Sea and in the estuaries of major sea-going rivers. The ecological risks decreased in the following order: Bohai Sea > Yellow Sea > South China Sea > East China Sea. There was generally low ecological risk, but certain hotspots existed near Tianjin and Jinzhou, which had relatively high ecological risks caused by Cu and Zn.

Keywords

Ecological risk assessment; heavy metals; marine organisms; China's costal water; species sensitivity distributions

1. Introduction

In recent decades, China's coastal water environment has faced serious contamination due to continuous pollutant loadings from the terrestrial system (Du et al., 2013; Liu et al., 2013; State Oceanic Administration, China, 2015, 2016). Generally, these pollutant loadings can be divided into two major categories: nutrient elements (i.e., nitrogen and phosphorus) and toxic pollutants (i.e., heavy metals and organic pollutants) (Li et al., 2014; Ministry of Environmental Protection, China, 2015; Selman et al., 2015; Tong et al., 2015). Unlike nutrients, heavy metals in water are especially of concern for marine organisms and seafood consumers due to their toxicity, bio-concentration and non-degradation (Morillo et al., 2002; Loska and Wiechuła, 2003; Xing et al., 2013). In 2015, the riverine discharges of heavy metals into seas were reported to be ~21,000 tons, including 3,318 tons of Cu, 16,243 tons of

Zn, 858 tons of Pb, 49 tons of Hg and 3,188 tons of As (State Oceanic Administration, China, 2015). In addition to the riverine discharges, the contribution from direct discharges by coastal cities is also significant. For instance, up to ~18 tons of Pb was directly discharged into China's seas in 2015 (Ministry of Environmental Protection, China, 2015).

These heavy metals in coastal waters could have seriously negative impacts on living organisms. Kim et al. (2003) reported that in the coastal waters of Southeast Korea, the decreased genetic diversity of organisms occurred more frequently in the regions polluted by heavy metals. Johnston et al. (2009) estimated that an ~30%-50% reduction of species richness was caused by marine contaminants, especially heavy metals. In addition to heavy metals with high acute toxicity (i.e., Pb and Hg), less-toxic heavy metals in water could pose even more serious ecological risk concerns for aquatic organisms (Fu et al., 2017). Fu et al. (2016, 2017) reported that the ecological risks to freshwater organisms caused by Cu and Zn were much higher than those of other heavy metals. The greatest probabilities of ecological risks to aquatic species were ~100% for Cu and ~50% for Zn, whereas the risks caused by other toxic heavy metals (<0.1%) could almost be neglected (Fu et al., 2016, 2017).

Ecological risk assessment (ERA) is used to estimate the possibility of the potential ecological effects if aquatic organisms are exposed to one or several environmental stressors (Solomon et al., 1996; Steen et al., 1999). Usually, the ecological risk is expressed as the percentage of the affected organisms relative to all of the organisms belonging to the same or similar groups of species. The US Environmental Protection Agency established a complete database (ECOTOX database) based on the EC_{50} (median effect concentration) and LC_{50} (median lethal concentration) values of various aquatic organisms in fresh or sea water if exposed to different concentrations and species of heavy metals. To apply the toxicological data to develop favourable and reasonable environmental standards for the entire ecosystem or to assess the hazards of pollutants at the ecological level, interspecies correlation estimation (ICE) models and a species sensitivity distributions (SSDs) method has been proposed and widely applied during the past decades (Traas et al., 2002; Wheeler et al., 2002; Azevedo et al., 2015; Zheng et al., 2015). The ICE mode and SSDs were used to assess ecological risks when they were first introduced in the 1970s (Kooijman, 1987). These were used to build statistical distribution models on the basis of the varying sensitivities of species to different pollutants and to provide assessments of a certain region's ecological risks (Wheeler et al., 2002). Lenwood et al. (1998) applied SSDs to calculate the concentrations of Cu and Ca that could protect 90% of the species in the freshwater and saltwater systems of the Chesapeake Bay of the United States. The 90% protected concentrations of Cu were 8.3 mg/L in freshwater and 6.3 mg/L in saltwater and those of Ca were 5.1 mg/L and 31.4 mg/L, respectively. The derived value could be applied as a reference value for the development of environmental quality standards. Van Sprang et al. (2004) estimated that the no-effect concentration for aquatic organisms based on dissolved Zn was 34.2 mg/L in Dutch surface water and estimated that there were only very limited ecological risks. Mu et al. (2014) integrated quantitative ion character-activity relationships and SSDs to derive the saltwater quality criteria for 34 different metals or metalloids. Even when considering the uncertainty due to the application of different correlation models and variances in the toxicity data between different species (He et al., 2017), ICE and SSDs are still important tools to assess the ERA caused by heavy metals to organisms.

Although marine organisms are facing increasing pressure from terrestrial pollutant loadings to China's coastal waters, the ecological risks to these organisms have never been previously assessed by a unified dataset. In this study, we compared the heavy metal concentrations (including Cu, Zn, Pb, Hg and As) in China's four adjacent seas (including the Bohai Sea, Yellow Sea, East China Sea and South China Sea) based on a nationwide dataset obtained from 301 sampling sites in 2015. To establish the linkage between aquatic heavy metal concentrations and the ecological risks of marine organisms, the ICE and SSDs methods based on the US EPA ECOTOX database were applied to estimate the potential ecological risks for crustacean, fish and mollusc in coastal waters. The coastal regions near the estuaries of major sea-going rivers were addressed in this study. This study attempts to describe the current status of heavy metal contamination in China's coastal waters and to pinpoint the major heavy metals, regions and sensitive aquatic species with high ecological risks, which is important for future policy-making regarding the control of heavy metal pollution and the protection of coastal environments.

2. Methods and materials

2.1 Concentrations of heavy metals in China's coastal waters

The dataset applied in this study was derived from the coastal water quality monitoring program, which was conducted by the Ministry of Environmental Protection, China, in 2015 (see Table S1). The dataset includes the concentrations of five major inorganic heavy metals, including Cu, Zn, Pb, Hg and As, from a total of 301 monitoring sites distributed in four seas (35 sites in the Bohai Sea, 68 sites in the Yellow Sea, 95 sites in the East China Sea and 103 sites in the South China Sea as shown in Figure S1). The water sampling locations were usually set ~30-50 km away from coastlines to avoid direct interferences from human activities. All of the field samplings were conducted during the period between August and October in 2015 and based on the guidelines of "Specifications on Spot Location of Monitoring Sites Related to Coastal Area Environment (HJ730-2014)". Approximately 500 mL of filtered mixed water samples (~1 meter below sea surface, ~10 meters below the sea surface and ~2 meters above the seabed, filtered by 0.45 μm Millipore filter) were collected. HNO_3 (guaranteed reagent, GR) was added to make the pH of the water lower than 2 before sample analysis. The detection limits for Cu, Zn, Pb, Hg and As were 0.100, 1.550, 0.150, 0.001 and 0.200 μg/L, respectively (HJ730-2014). During data analysis, for sampling sites with measured values that were lower than the detection limit, their values were assumed to be half of the detection limits.

2.2 Ecological risk assessment

As recommended by previous studies (Newman et al., 1995, 2000; Zheng et al., 2015, 2016), we used the EC_{50} or LC_{50} values from laboratory tests as an effective indicator of marine organisms exposed to heavy metals. The EC_{50} or LC_{50} data of the selected heavy metals were obtained from the US EPA ECOTOX database (US EPA, 2017). In this study, we selected crustacean, fish and mollusc as the target marine organisms, which are the most common marine organisms in China's coastal waters; they also have good quantity and available ecotoxicity data. The following principles were applied during the selection of ecotoxicity data (Durda and Preziosi, 2000; Liu et al., 2011; Zheng et al., 2015): I. The ecotoxicity data for the organisms were obtained in a sea water environment. II. The forms of heavy metals exposed to organisms in the laboratory test were inorganic, and organic forms (i.e., methyl mercury) were excluded in the data screening due to a lack of monitoring data. III. Eight or more previously reported toxicity data points were required to construct ICE models and SSD curves for one group of species. For instance, the SSD curve for As to mollusc is not applicable in this study due to the limited reported ecotoxicity data. The ecotoxicity data for different groups of marine organisms are provided in Table S2. The specific process of calculating SSDs and its uncertainty was performed by the SSD generator (US EPA, 2016).

By using the data of organisms belonging to similar groups of species from laboratory tests, SSD curves could assist in the interpretation of site data for stressor identification and risk assessment by relating them to the proportions of species expected to be affected at the prescribed concentrations, which is used to represent the ecological risks to marine organisms. SSDs could model the variations in the sensitivity of different groups of species to a certain stressor (i.e., heavy metals). For instance, HC_5 (5% Hazardous Concentration) represents the exposure concentration, which causes 5% of the target organisms (i.e., fish and mollusc) to experience negative impacts, which are predefined by different endpoints, including reduced mobility and fertility. Several interspecies correlation models have been applied to describe the distribution of toxicological data when building SSD curves. Different selections of the fitting models (i.e., log-normal distribution, log-logistic model and bootstrap regression) could cause variances in the estimation of ecological risks to marine organisms (Newman et al., 2000; Durda & Preziosi, 2000; Wheeler et al., 2002). Among these models, the log-normal model was recommended by US EPA for the assessment of the ecological risks of heavy metals (US EPA, 2016). The SSD curves for different marine organisms and heavy metals are provided in Figure S2. Based on the SSD curves and monitoring data in the coastal waters, we calculated the ecological risks and evaluated the proportions of marine organisms expected to be affected at prescribed concentrations for crustacean, fish and mollusc, respectively. To describe the general risk to all organisms (regardless of species), three groups of marine organisms (crustacean, fish and mollusc) were also mixed to construct the SSD curves and estimate the general risks induced by a single heavy metal to all organisms. In addition to the risks induced by a single heavy metal, the following equation was applied to estimate the effects of multiple substances, i.e., the potential affected fraction (msPAF) of marine organisms by the five heavy metals (Kong et al., 2011; Hu et al., 2012; Jiang et al., 2012; He et al., 2014):

$$\text{msPAF} = 1-(1-P_{\text{Cu}})\times(1-P_{\text{Zn}})\times(1-P_{\text{Pb}})\times(1-P_{\text{Hg}})\times(1-P_{\text{As}})$$

Where P_{Cu}, P_{Zn}, P_{Pb}, P_{Hg} and P_{As} are the ecological risks for organisms under the exposure of Cu, Zn, Pb, Hg and As in the seas, respectively.

3. Results

3.1 Concentrations of heavy metals in China's coastal waters

Figure 1 shows the concentrations of selected heavy metals in 301 sampling sites distributed in China's four seas in 2015. Unlike serious nutrient pollution (i.e., nitrogen and phosphorus) (Li et al., 2014; Selman et al., 2015), heavy metal concentrations in China's coastal waters were relatively low for the majority of the sampling sites, and higher measured values usually occurred in the Bohai Sea. For Cu, ~95% of the sampling sites had concentrations of lower than 5 μg/L (Figure 1), which was the Grade I limit by China's sea water quality standard (see Table S3). The average measured Cu concentrations in the Bohai Sea, Yellow Sea, East China Sea and South China Sea were 3.7±2.5, 1.6±1.5, 0.8±0.5 and 1.2±1.0 μg/L, respectively. For Zn, most of the sampling sites had concentrations of lower than 20 μg/L, except for some sites in the Bohai Sea, where Zn concentrations ranged from 5.6 to 49.6 μg/L and ~60% of the monitored values exceeded 20 μg/L. The lowest Zn concentrations were observed in the East China Sea, with a range of 1.6-15.8 μg/L. For Pb, ~50% of the sites in the Bohai Sea had concentrations of higher than 1 μg/L, which is the Grade I limit based on China's sea water standard (Table S3). For Hg, the difference in the spatial distributions among the four seas was not significant, and ~98% of monitored values were lower than 0.05 μg/L, which was the Grade I limit based on China's sea water standard (Tong et al., 2017). The As concentrations in the four seas were far below the Grade I limit based on China's sea water standard (i.e., 0.8±1.1 μg/L in the Bohai Sea, 1.1±0.9 μg/L in the Yellow Sea, 1.8±0.7 μg/L in the East China Sea, and 1.0±0.9 μg/L in the South China Sea).

Spatial variances in heavy metal concentrations along the coastal regions were observed (Figure 1), and the higher concentrations usually occurred at the estuaries of the major sea-going rivers (i.e., the Yangtze and Pearl Rivers). In coastal waters, the concentrations of heavy metals largely depended on terrestrial pollutant loadings, water volume and the water exchange rate with the open ocean (Lin et al., 2016; Sharples et al., 2016). It has been reported that the residence time of riverine water on the continental shelf around China could last from months to years before the water is completely exchanged with the open ocean (Lin et al., 2016; Sharples et al., 2016), and limited exchange with the open ocean could prolong the residence time of heavy metals in China's coastal waters. The main reason for the poor exchange capacity of the Bohai Sea is its semi-enclosed bay. Also, within the same sea, relatively higher concentrations usually occurred at the estuaries of the major sea-going rivers. For instance, the Cu and As concentrations at the estuaries of the Pearl River were higher than those at the other monitoring sites in the South China Sea (Figure 1). This fact could probably be explained by huge riverine pollutant loadings to the sea. For example, sea-going rivers were believed to be important Hg contributors to coastal waters (Liu et al., 2016). Among all of the sources (e.g., riverine discharges, coastal erosion), riverine Hg discharge was dominant and occupied ~70% of the terrestrial Hg loadings (Liu et al., 2016). In 2015, riverine discharges of heavy metals into the seas were reported to be ~21,000 tons (3,318 tons of Cu, 16,243 tons of Zn, 858 tons of Pb, 49 tons of Hg and 3,188 tons of As) (State Oceanic Administration, China, 2015).

3.2 Interspecies correlation estimation based on the ECOTOX database

The parameters used to obtain the ICE values for different marine organisms under the exposure of the selected heavy metals were calculated based on the ECOTOX database, and the HC_5 value for different organisms was also calculated. As shown in Figure S2, if exposed to the same concentration levels (<1000 μg/L) of heavy metals, the hazardous levels of crustacean, fish and mollusc for each pollutant ranked as: Hg > Cu > As > Zn > Pb, Hg > Cu > Pb > Zn > As, and Hg > Cu > Zn > Pb, respectively. This point could also be reflected in the HC_5 value. For instance, the HC_5 values for Hg to marine organisms (1.3 μg/L for crustacean, 29.8 μg/L for fish and 9.0 μg/L for mollusc, respectively) were generally lower than those of the other heavy metals (Table 1). The corresponding HC_5 values for three groups of marine organisms by Cu were 19.5(9.4-40.5) μg/L, 48.4(29.1-80.3) μg/L, and 9.2(4.4-19.2) μg/L, respectively, whereas for Zn, they were 75.8(44.8-128.2) μg/L, 116.2(27.4-493.8) μg/L, and 202.6(66.7-615.8) μg/L, respectively.

4. Discussion

First, we evaluated the geographic differences in the general ecological risks for all organisms (regardless of species types) induced by Cu, Zn, Pb, Hg and As in China's four adjacent seas. Among the four seas, the Bohai Sea had the highest ecological risk of all selected heavy metals except As, and the lowest values usually occurred in the South China Sea, except for some sites at the estuary of the Pearl River (shown in

Figure 2). The risk induced by Cu to all of the selected organisms in the Bohai Sea was highest among the four seas, but the potential risk did not exceed 2.1% (Figure 2). For the other three seas, the potential ecological risks caused by Cu were all lower than 1.0%. The ecological risks induced by Zn were slightly lower than those induced by Cu in China's coastal waters, and the lowest values were observed in the East China Sea. The ecological risks caused by Pb in the four seas decreased in the following order: Bohai Sea > Yellow Sea > South China Sea > East China Sea (Figure 2). Although marine organisms were more sensitive to Hg (i.e., lower HC_5 values compared with other heavy metals, as shown in Table 1), the higher potential ecological risks were usually caused by Cu and Zn (Figure 2) due to their higher concentrations in coastal seawater. The coastal regions with the highest potential ecological risks for each heavy metal are shown in Figure 3. Due to their higher concentrations, the major river estuaries (i.e., the Pearl and Yangtze Rivers) and the Bohai Sea were hotspots in terms of ecological risks. For instance, the ecological risk of Cu at the monitoring sites near Tianjin City in the Bohai Sea could be higher than 2%.

Second, we evaluated the single ecological risks for each group of species induced by Cu, Zn, Pb, Hg and As in China's four adjacent seas. For different organism types, the heavy metals with the highest ecological risks were almost the same. For crustacean, the heavy metal with the highest ecological risk was Zn, and the ecological risk caused by Cu was just slightly lower than that of Zn. For fish and mollusc, the heavy metals were Zn and Cu, respectively (Figure 4). Significant variances in ecological risks could be observed among different seas. Due to the higher concentrations of heavy metals, the organisms in the Bohai Sea usually had the highest ecological risks, especially for the sampling sites near Jinzhou, Tianjin and Tangshan (Figure S3) where the potential ecological risk of Cu to mollusc could be even higher than 5%, which should draw the attention of local environmental authorities. The ecological risks caused by Pb varied significantly among different groups of organisms, e.g., fish could have higher risk levels, while the risks to mollusc and crustacean could be neglected. The risks caused by Hg and As in the four seas were low due to their lowest concentrations in the water (Figure 4).

It must be noted that benthic invertebrates may mostly ingest heavy metals in the sediments (Lin et al., 2013; Guo et al., 2016); however, due to limited data, the risk assessment conducted in this study only evaluated the risks due to exposure in the water column. Guo et al. (2016) also noted that a significant increase in Hg levels was observed with increasing trophic levels of fish, and fish were found to have a high bioaccumulation factor for Hg. Therefore, the low risk of Hg due to exposure in the water column does not mean that the overall risk for exposure to Hg is low. Rather, the majority of Hg in fish potentially accumulated through the water and food web (Tong et al., 2017), and fish with high trophic levels should receive special attention.

Third, we evaluated the msPAF induced by all heavy metals to all organisms in China's coastal waters (Figure 5). In most of the risk distributions, the risk levels in the four seas decreased in the order of Bohai Sea > Yellow Sea > South China Sea > East China Sea. Additionally, for the ecological risks of all heavy metals to all organisms, higher levels usually occurred at the estuaries of major rivers flowing into seas. All of the calculated msPAFs values were lower than 5%, which was a safe value for aquatic organisms (Aldenberg and Slob, 1993; Newman et al., 2000). For the msPAF values of the five major heavy metals, the Bohai Sea was the highest among the four seas, where ~25% of the sites were above the risk of 2.5% and the highest risk in the Bohai Sea exceeded 3.0%, such as at the sampling sites in Jinzhou, Tangshan and Tianjin. The msPAF in the Yellow Sea was the second highest, and was much lower than that in the Bohai Sea. Almost all sites had a risk of less than 1.0%, and only one site in Dalian, Liaoning Province, had a relatively high risk of 3.5%. From the risk distribution in the Yellow Sea, we could see that all of the sampling sites in Dalian increased the average risk probability level of this area. For the South

China Sea, the percentage of the monitoring sites with msPAS values lower than 0.5% was ~77%.

5. Conclusions

The Bohai Sea contains the highest concentrations of Cu, Zn, Hg and Pb among the four Chinese adjacent seas, whereas the East China Sea contains the highest concentration of As. The river estuaries, such as the Yangtze River Estuary and the Pearl River Estuary, usually have higher concentrations of heavy metals compared with other regions.

Due to the high concentrations of heavy metals, Bohai Sea also poses the highest ecological risks for crustacean, fish and mollusc among the four Chinese adjacent seas. Most of the sites showed the affected proportion to be below 2.0%, and all sites had an affected proportion below 5.0%.

In general, the ecological risk level is low in all Chinese adjacent seas based on the current evaluation. However, hotspot areas may still exist, such as industrial cities along the Bohai Sea and the Pearl River Delta. Certain monitoring sites near Tianjin City and Jinzhou in the Bohai Sea had high ecological risks by Cu (>2% of the total) and Zn (>2.2% of the total), which should draw the attention of environmental authorities in the future.

The joint ecological risk posed by the five heavy metals to the three target organisms in coastal waters is relatively low, and the risk decreases in the following order: Bohai Sea > Yellow Sea > South China Sea > East China Sea. For crustacean, the heavy metal causing the highest ecological risk was Zn, and the ecological risk caused by Cu was slightly lower than that of Zn. For fish and mollusc, their corresponding heavy metals were Zn and Cu, respectively.

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Figure captions

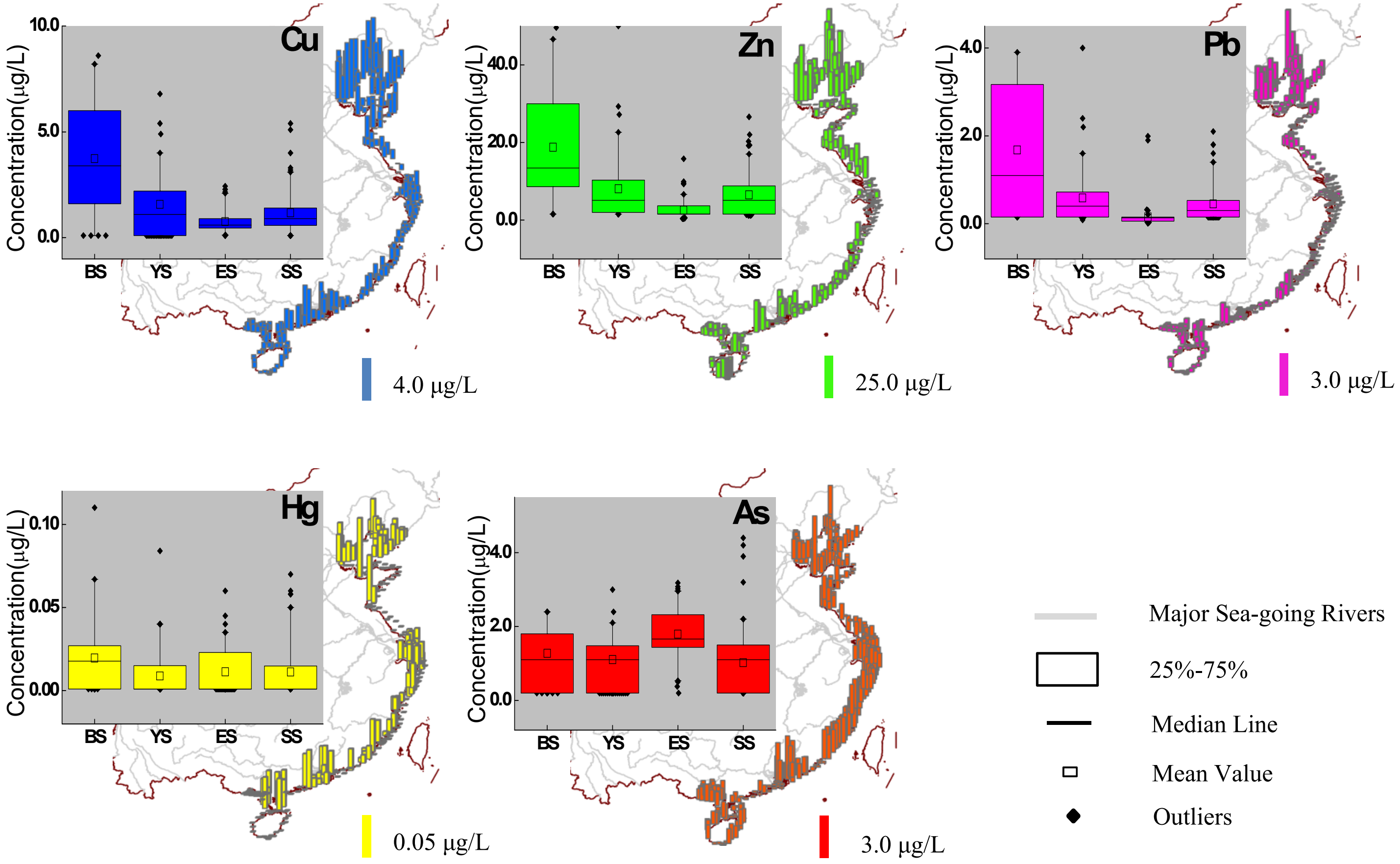
Figure 1. Concentrations of Cu, Zn, Pb, Hg and As in China’s coastal waters

Figure 2. Ecological risks faced by all of the selected organisms induced by heavy metals

Figure 3. Coastal regions with the highest potential ecological risks

Figure 4. Ecological risks to crustacean, fish and mollusc by Cu, Zn, Pb, Hg and As

Figure 5. Combined ecological risks of multiple substance in China’s coastal waters



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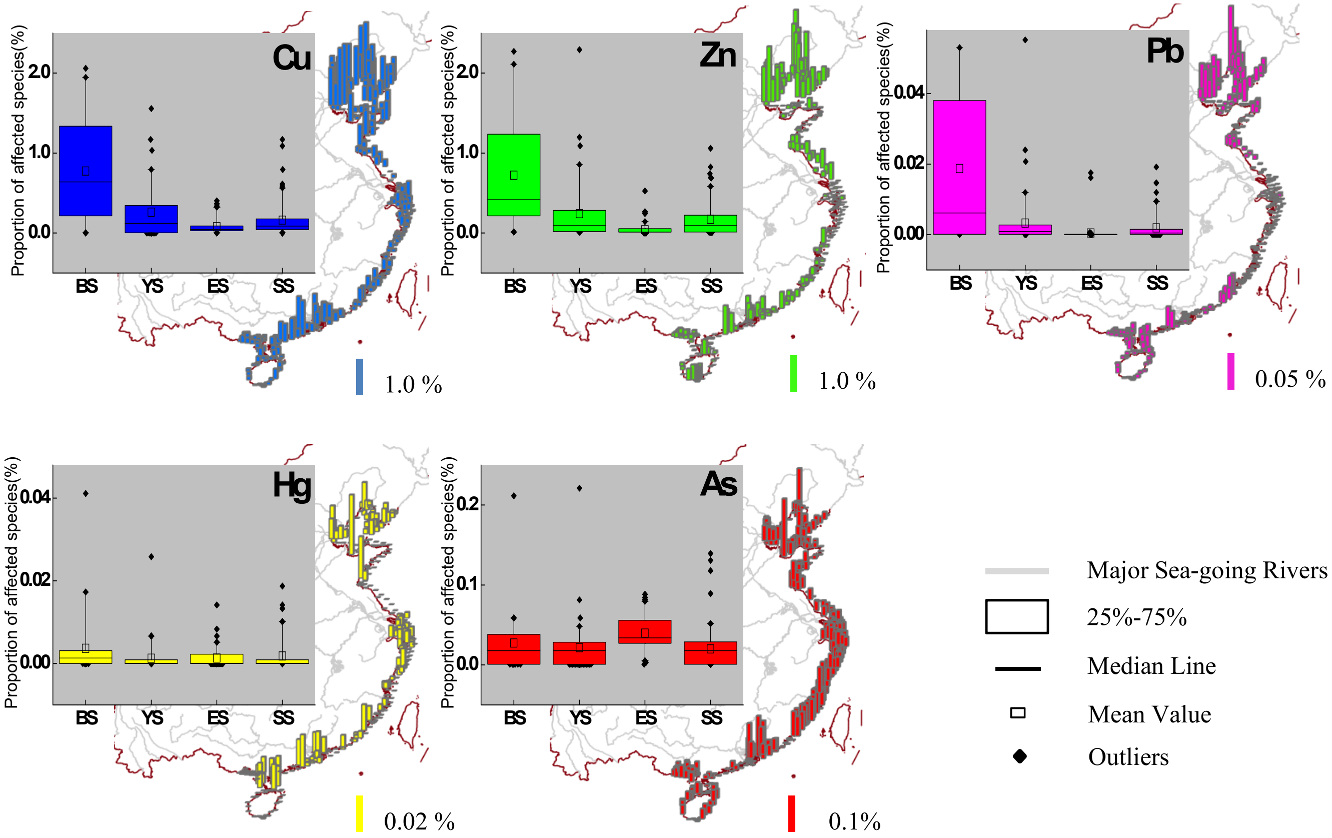
(BS: Bohai Sea; YS: Yellow Sea; ES: East China Sea; SS: South China Sea)



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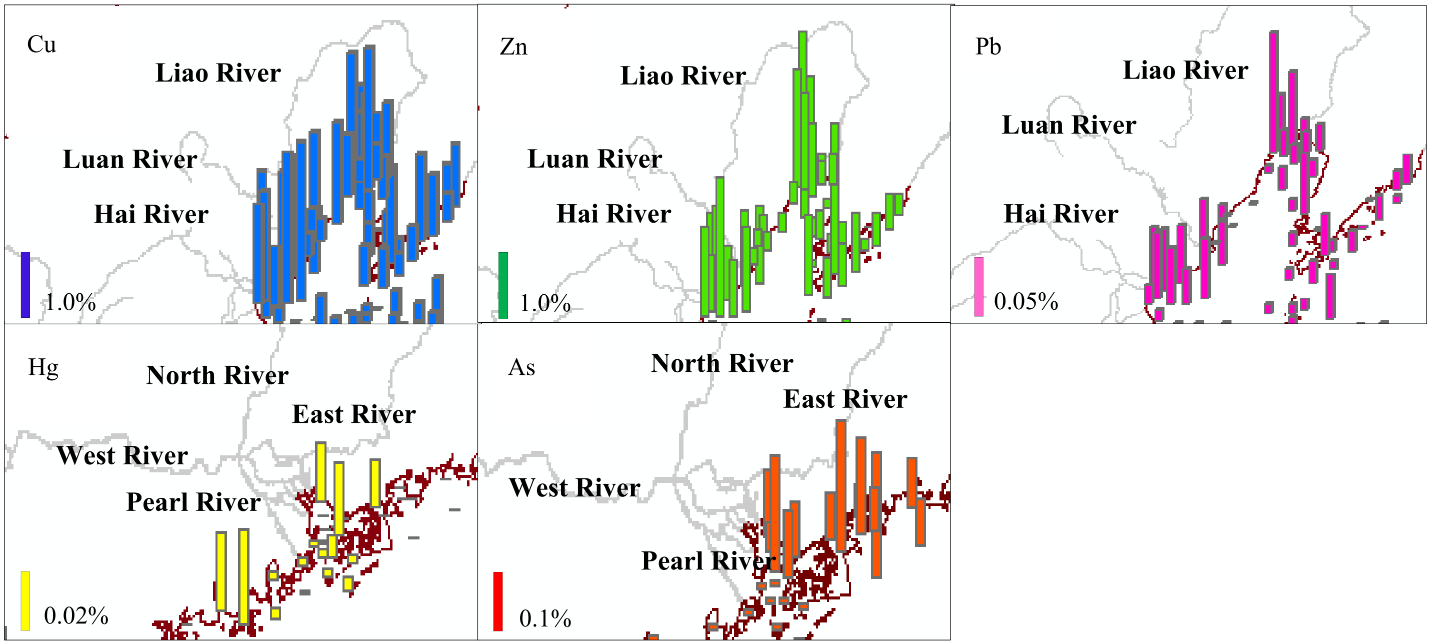

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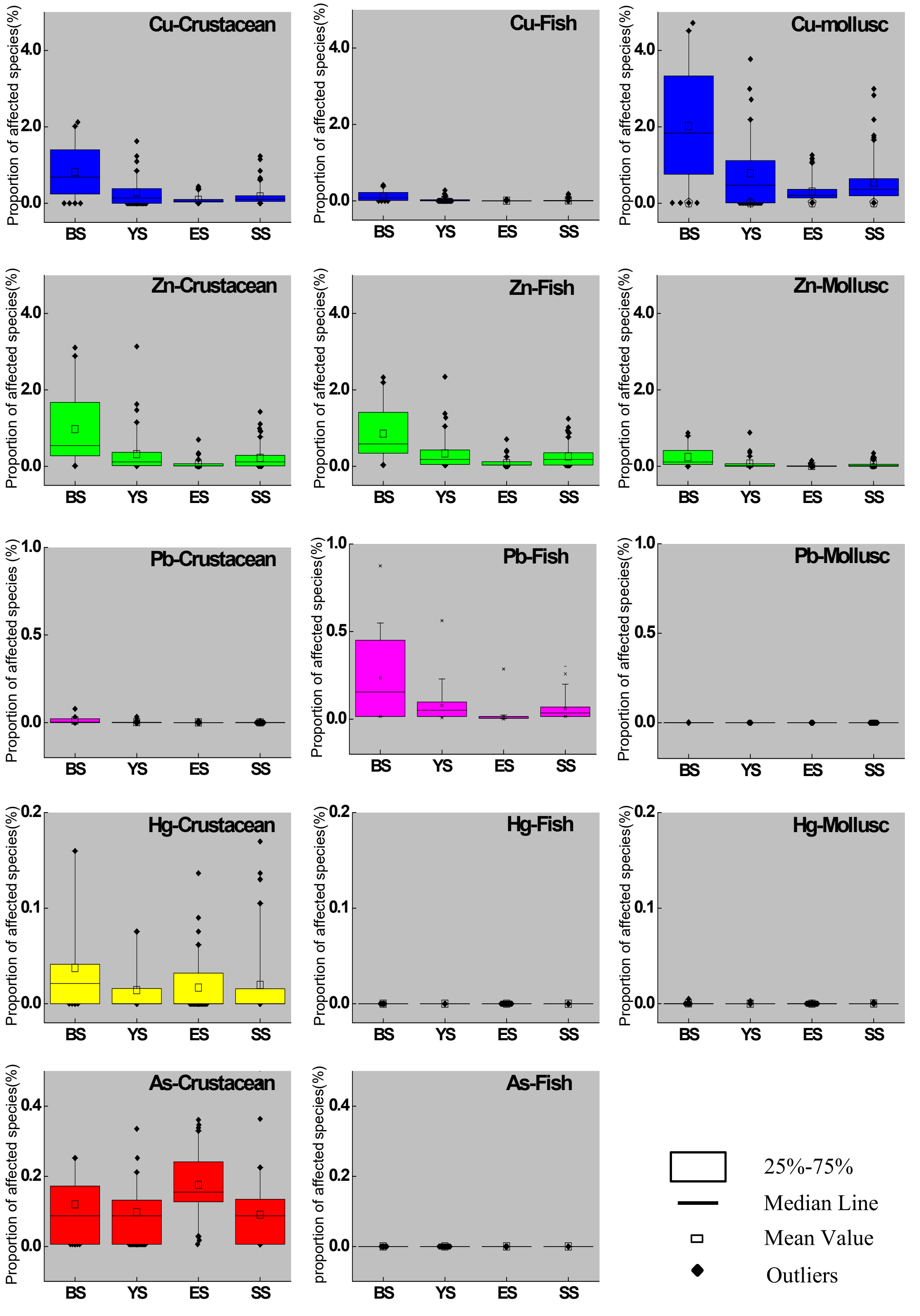

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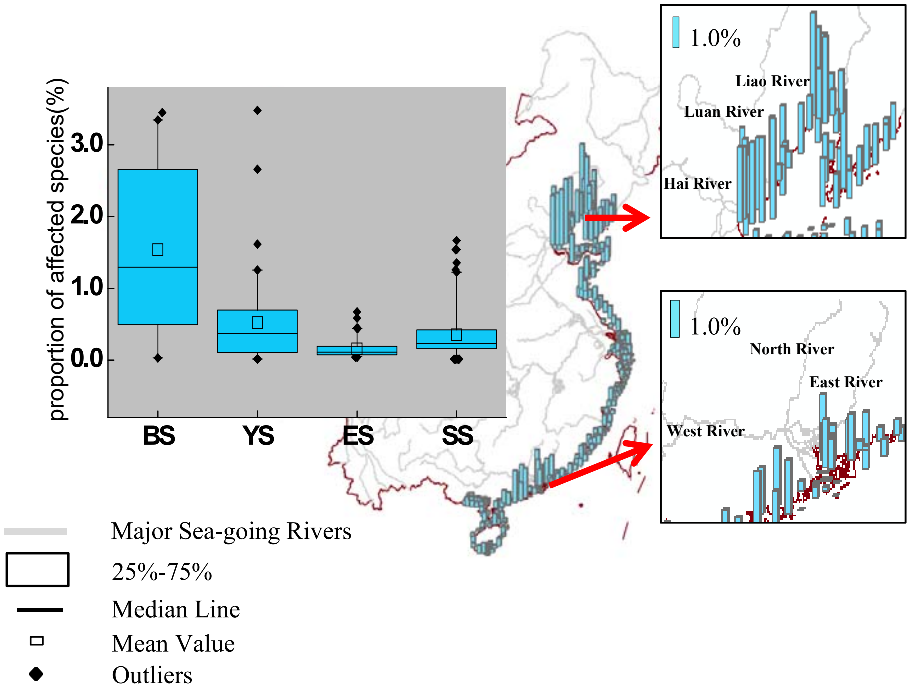

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