Probabilistic ecological risk assessment of copper in Chinese offshore marine environments from 2005 to 2012

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ABSTRACT

The objective of the present study was to conduct a probabilistic assessment of risk posed by copper found in the coastal marine environment of China from 2005 to 2012. This was achieved by applying a tiered ecological risk assessment (ERA) approach for characterization of risks of concentrations of copper from nationwide marine water monitoring program. The results show that from 2005 to 2012 the overall trend of hazard quotients (HQs) in the coastal marine environment of China the proportion of locations that exceed a HQ of 1.0 decreased from 64% in 2005 to 31% in 2012. While this indicates an overall improvement of the environment, there still have potential ecological risks in the most of the area, especially for the major gulfs of Liaodong and Bohai Bays and Yellow River Estuary. In addition, probabilities of exceeding the toxicity threshold for 5% of species were 27.6%, 5.4%, 4.9%, 0.8%, 0.4%, 1.0%, 1.8% and 0.12% annually between 2005 and 2012, respectively.

1. Introduction

Contamination of the environment with metals is widespread with air, soils, water, and sediments being affected. This is particularly true in Asia where rapid development of industry and agriculture has mobilized certain metals into the environment, especially in China (Cheng, 2003; Wang et al., 2010). The contamination in the environment caused by the metals is a matter of concern due to abundance and persistence, and subsequent potential of bioaccumulation as well as toxicity (Barlas et al., 2005).

Among the metals, copper (Cu) is a micronutrient, required for aquatic life present in all natural waters and sediment, that when present in excess amounts can also elicit toxic effects. Because it is an essential element, some species, such as phytoplankton have developed mechanisms to accumulate sufficient amounts of Cu from dilute concentrations in the environment to maintain health (Barlas et al., 2005). Natural fluxes of Cu have led to adaptation by many organisms, which have influenced aquatic species composition, diversity and abundance of populations. Invertebrates and fish possess elaborate homeostatic mechanisms that can maintain internal concentrations of Cu within narrow limits while external concentrations fluctuate over several orders of magnitude. Fish utilize proteins called metallothioneins that are involved with sequestration, transport and metabolism of Cu.

Although Cu is a minor nutrient for both plants and animals, it can become toxic to aquatic life at concentrations approximately 10–50 times greater than the required concentration (Hall et al., 1998). Thus, concentrations of Cu in natural environments, and its biological availability, are important and the optimal range is

relatively small and Cu is an excellent example of the potential effects of defined by the subsidy-stress hypothesis. Naturally occurring concentrations of Cu have been reported to range from 0.03 to 0.23 µg/L in surface seawaters and from 0.20 to 30 µg/L in freshwater systems (Flemming and Trevors, 1989). Thus, concentrations of Cu in locations receiving inputs influenced by activities of humans can vary anywhere from levels that approach natural background to 100 µg/L or greater (Hem, 1985; Nriagu, 1979) and have in some cases been reported to be in the 200,000 µg/L range in mining areas (Davis and Ashenberg, 1989). Mining, leather and leather products, fabricated metal products, and electric equipment are a few of the industries with Cu-bearing discharges that contribute inputs of Cu to surface waters (Biggs and D'Anna, 2012; Brooks and Waldock, 2009). In recent years potential ecological risk from exposure to Cu has become an issue worldwide (Hall and Anderson, 1999; Pekey et al., 2004; Schuler et al., 2008). A specific area of concern is the loading of Cu into the marine environment from the use of Cu-based antifouling paints on both recreational and commercial watercraft (Jones and Bolam, 2007).

The coastal zone of China comprises an area of more than three million square kilometers, a coastline of more than 18,000 km, stretching across tropical, subtropical and temperate zones. More than 70% of large Chinese cities are located on the coastal plane and coastal development in China is important to the national economy, accounting for 55% of its gross domestic productivity (GDP) (Wang, 2003). However, the continuing increase in population, coupled with economic growth, rapid urbanization and development of infrastructure have resulted in conflicts among multiple groups, especially among terrestrially and marine-based industrial sectors (Yu, 1994). To balance anthropogenic activities and ecosystem health and environmental protection, a comprehensive management scheme is urgently required in the coastal zone on a sustainable basis (Cao and Wong, 2007). Therefore, the deleterious effects and ecological risk of metals, including Cu in estuarine and coastal ecosystems have raised concern.

The most rudimentary approach to assessing the potential risks of chemicals is to compare measured or predicted exposure concentrations, with concentrations associated with thresholds for adverse effects, expressed as the predicted no-observed effect concentrations (PNECs), derived from dose-response relationships. The simplest comparison is a single-point estimate, usually expressed as the hazard quotient (HQ), which is the measured or estimated environmental concentration divided by the toxicant reference value (Jin et al., 2012; Solomon et al., 2000; Wang et al., 2002). Because of its simplicity and effectiveness, the HQ approach is widely used; its use is only appropriate for conservative screening-level risk assessment and for the early stages or tiers of assessment of potential effect. Because potential risk represents a likelihood or probability of occurrence, it cannot be established from point estimates, such as the HQ (Bartell et al., 2000; SETAC, 1994). Probabilistic ecological risk assessments (PERAs), which qualify and quantify ecological risks through exposure and effect probability distributions, are regarded as an improvement on the HQ approach and, thus, recommended for later tiers in the ecological risk assessment (ERA) process (Solomon et al., 1996). Because PERAs can better describe the likelihood of exceeding the effect thresholds and the risk of adverse effects (Solomon and Sibley, 2002), this approach has been adopted by a number of researchers (Giesy et al., 1999; Hall et al., 2000; Jin et al., 2014; Zolezzi et al., 2005). To obtain more reliable results for decision-making processes on site-specific contamination, some researchers and scientific institutions have suggested the use of a tiered approach, ranging from simple deterministic methods to probabilistic methods, for risk characterization, (Jin et al., 2012; Wang et al., 2009; Zolezzi et al., 2005). The objective of the present

study was to undertake an ERA which would provide a realistic assessment of the risk posed by Cu found in offshore areas throughout China during the period 2005–2012. This was achieved by applying a tiered ERA approach consisting of several probabilistic options, concentrations of Cu from nationwide marine water monitoring program, and toxicity data for marine species from published literature.

2. Materials and methods

2.1. Exposure concentration of Cu

Concentrations of Cu in the water column of seawater were collected from four sea regions of China, including the Yellow Sea, Bohai Sea, East China Sea and South China Seas and in particular bays along the coast of China from 2005 to 2012 (Fig. 1). The total number of locations was approximately 300 each year with fixed sample site, with 49, 54, 95 and 103 for Yellow Sea, Bohai Sea, East China Sea and South China Seas, respectively. Sample collection and analysis were following national standards (MEP, 2008; Xu et al., 2007). For statistical analyses, to be conservative and error on the side of caution, for samples for which concentrations were less than the method detection limits (MDLs) were replaced with a surrogate value equal to half the MDL. The distribution of exposure data in each of sea regions will be tested for normality as raw or log-transformed data (concentrations greater than the MDL) by use of the Kolmogorov–Smirnov test.

2.2. Toxicity data collection and SSD generation

Data on toxic potency of Cu towards marine organisms were obtained from the USEPA water quality criteria documents for Cu (USEPA, 2007), an AQUIRE literature search and a recent risk assessment for Cu in Chesapeake Bay (Hall et al., 1997) and stratified by tropic group. An initial analysis of toxicity data showed that toxicity values generated from measured and nominal values (when similar studies could be compared) gave similar results. Therefore, properly screened data from both measured and nominal studies were used for the analysis. Additional toxicity data for Cu were collected from existing toxicity databases, such as the ECOTOX database, http://cfpub.epa.gov/ecotox/, those published in the open, peer-reviewed literature, and government documents following the principles of accuracy, relevance and reliability (Caldwell et al., 2008; Klimisch et al., 1997). Since data for chronic toxicity were limited, it was not possible to generate a complete species sensitivity distribution (SSD) for saltwater species. Consideration was given to calculating the FACR based on all ACRs within a factor of 10 (Chapman et al., 1998). Therefore, this study focused on acute toxicity data and an assessment factor of 3.23 was used as the acute to chronic ratio for Cu in saltwater to construct joint probability curve (JPC) (USEPA, 2003). For acute toxicity data, selected measurement endpoints were the median lethal concentration (LC50) or median effect concentration (EC50) based on immobility for animals and biomass or growth for plants. When there were multiple toxicity data for a species, they were summarized into a single value by using the geometric mean (Geomean) as recommended by Ecological Committee on FIFRA Assessment Methods (ECOFRAM; http://www.epa.gov/oppefed1/ecorisk/aquareport.pdf). Outlier(s) as detected by application of Grubb's test (Grubbs, 1969) were excluded. Toxicity data were then ranked in ascending order, and centiles were assigned from the formula $i/(N+1)$, where i is the rank of the datum in ascending order and N is the total number of data points (Wheeler et al., 2002). Datasets must contain a minimum of eight data points from at least three different taxonomic groups to be
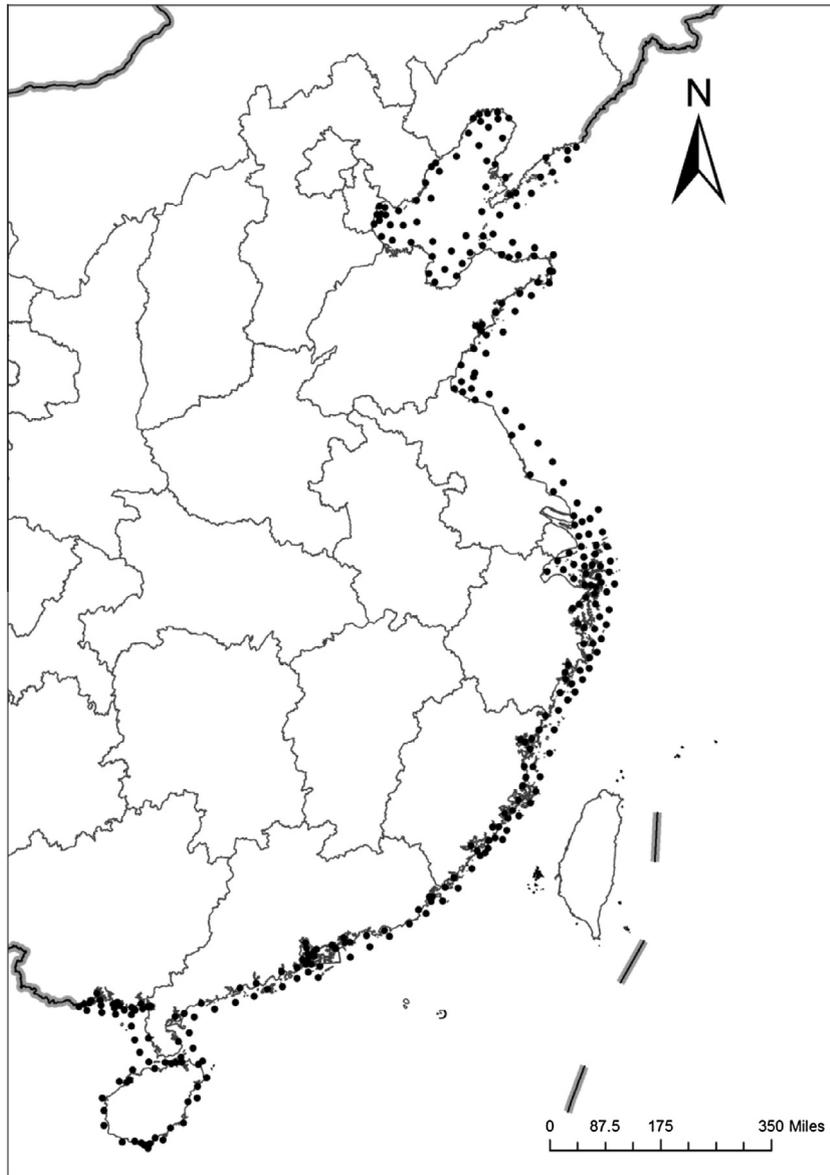
Fig. 1. Spatial distribution map of copper in Chinese offshore marine environments.

considered suitable for construction of SSDs. In addition, a taxon-specific SSD was constructed and taxon-specific species sensitivities were compared through a comprehensive meta-analysis of taxon-specific 10% hazardous concentrations (HC_{10}) and their 95% confidence interval (95% CI).

2.3. Ecological risk assessment of Cu

Level 1 deterministic HQs were calculated by dividing measured environmental concentrations (MECs) with PNECs. For comparison, two MEC values were used for HQ calculations, the mean, which was calculated by both excluding and replacing the values less than the MDL, and the maximum concentrations of Cu, within an area, respectively.

$$HQ = MECs/PNECs$$

Level 2 utilized both exposure and effect distributions. The probabilistic ecological risk assessment (PERA) was performed by using the Probabilistic Risk Assessment Tool (PRAT) (Solomon et al., 2000, 1996). In PERA, estimation of risk is described as being proportional to the degree of overlap of the distributions, and one method of displaying risk is through the use of joint probability curves (JPCs), which describes the probability of a particular set of exposure conditions occurring relative to the number of taxa that would be affected (Jin et al., 2012; Zolezzi et al., 2005). The *x*-axis of the JPC represents the intensity of effects, while the *y*-axis represents their probability. Each point on the curve represents both the probability that the chosen proportion of species will be affected and the frequency with which that magnitude of effect would be exceeded. The closer the JPC is to the axes, the less the probability of adverse effects.

3. Results and discussion

3.1. Exposure assessment

The distribution of concentrations of Cu in Chinese offshore marine waters was tested for normality as either the raw, untransformed data or the log-transformed data (concentrations greater than the MDL) by use of the Kolmogorov–Smirnov test. Since all

data were log-normally distributed, the log-transformed data were used for all further characterizations. Cumulative probabilities of concentrations of Cu were calculated (Fig. 2). Concentrations of Cu in marine waters offshore of China ranged from 0.1 to 43.2 µg/L. Concentrations of Cu were highest in 2005 which showed in the rightmost of the distribution, with mean of 2.87 µg/L. Concentrations of Cu decreased annually from 2005 to 2008, but then changed little from 2008 to 2011. Concentrations of Cu were least in 2012 when they ranged from 0.3 to 9.9 µg/L among locations with a mean of 1.73 µg/L. The highest concentrations of Cu were observed in Liaodong, the Yellow River Estuary and Bohai Bays with averaged of 10.16 µg/L, 8.81 µg/L and 4.59 µg/L, respectively (Fig. 3). Concentrations of Cu in Hangzhou Bay and Beibu Bays, the Yellow and Pearl River Estuaries decreased from 2005 to 2012. Concentrations of Cu in sea waters off the coast of China were similar to those of marine water near Europe (Brooks and Waldock, 2009; Hall and Anderson, 1999). Typical copper concentrations in marine coastal waters are found in the range 0.5–3 µg/L (Jones and Bolam, 2005, 2007).

The quality standard for Cu is 10 µg/L for class II seawater in China (GB3097-1997). Approximately 98% of concentrations of Cu in coastal waters were less than the class II seawater quality standard (Fig. 2), with only 0.7–2.3% of locations exceeding the standard from 2005 to 2012. When the USEPA revised its water quality standards regulations in 2007 a criterion maximum concentration (CMC) and criterion continuous concentration (CCC) for Cu were set at 3.2 µg/L and 1.9 µg/L, respectively (USEPA, 2007). In 2005, 31% of concentrations of Cu in Chinese coastal waters exceeded the CMC, while 64% exceeded than the CCC. In 2012, 14% of concentrations still exceeded the CMC, while 31% of measured concentrations of Cu exceeded the CCC (Fig. 2).

3.2. Effect assessment

A total of 232 values for acute toxicity of Cu, based on lethality and effects on behavior, were collected for marine species, including 35 algae, 84 crustaceans, 51 molluscs, 49 fish and 14 Oligochaeta. Acute toxicity values ranged from 6 µg/L for the bivalve *Ostrea* sp., to 346,700 µg/L for the crab *Scylla serrata*.

A taxon-specific species sensitivity distribution (SSD) for marine species was developed (Fig. 4). Marine algae are the most sensitive to Cu (SSD curve on the far left), fish are relatively tolerant (SSD curve at the far right). This also can be obtained by ANCOVA analysis, which have the same slope ($p > 0.05$), but with different y- intercept ($p < 0.05$). The SSD for crustaceans on the left side of molluscs SSD, indicates that crustaceans are more sensitive than molluscs. By comparing SSD and hazardous concentration for 10% of species (HC_{10}) values (ANOVA) the decreasing order of sensitivity of marine taxa to Cu was: algae > crustaceans > mollusks > oligochaeta > fish ($p < 0.05$; Table 1).

Bioavailability and toxicity of copper to aquatic organisms depends on the total concentration of copper and its speciation (Hung et al., 1990). Copper toxicity to aquatic biota is related primarily to the dissolved cupric ion (Cu^{2+}) and possibly to some hydroxyl complexes. Soluble copper is largely complexed with carbonate, amino acids, or humic substances (Hung et al., 1990). In freshwater algae, movement of copper into cells occurs mainly by physical transport; the plasmalemma is the initial site of copper binding. Copper on the plasmalemma increases its permeability, as shown by the leakage of potassium and other ions from copper-treated cells and entry of copper into intracellular sites (Stokes, 1979). In aquatic invertebrates, copper causes gill damage at high concentrations, and in fishes it interferes with osmoregulation (Hodson et al., 1979). Elevated concentrations of copper interfere with oxygen transport and energy metabolism; tissue hypoxia is the cause of death and is associated with reductions in the
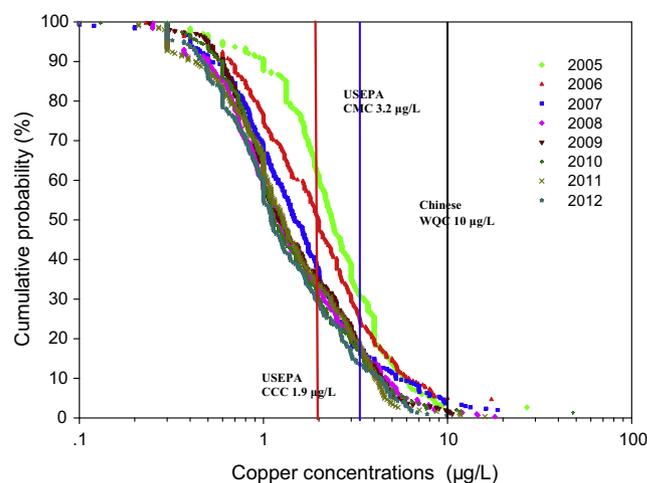

Fig. 2. The cumulative probabilities of exposure distribution of copper in Chinese offshore marine environments from 2005 to 2012.

activities of regulatory enzymes of ATP-synthesizing pathways (Hansen et al., 1992). In crabs, the gills are a major target organ of copper toxicity; waterborne copper decreases hemocyanin-oxygen affinity (Truchot, 1975). In fishes, the gill surface's low affinity for metal allows greater entry of the metal to the intracellular compartment. Once there, more complex binding sites are present. Binding to these ligands causes one or more of the following toxic mechanisms: (1) blocking of the essential biological functional groups of biomolecules; (2) displacing the essential metal ion in molecules; or (3) modifying the active conformation of biomolecules. These mechanisms may account for the specific inhibition of ion transport from ionic copper (Cu^{2+}) exposure (Reid and McDonald, 1991). Generally, taxon-specific sensitivities differences to copper are mainly attributed to the differences of copper-related the physicochemical and biochemical mechanisms among different freshwater and/or marine taxa.

Assessment of risks of effects of Cu on the ecosystem was achieved by applying a tiered ERA approach, consisting of both simple, deterministic methods to assess hazard and more complex probabilistic methods to characterize risks. Deterministic HQs were calculated by dividing mean, measured environmental concentrations (MECs) of Cu with PNECs. The PNECs used was the USEPA CCC of 1.9 µg/L for Cu (USEPA, 2003, 2007). Since information on chronic toxicity was limited, it was not possible to generate SSDs for effects of Cu on saltwater species Therefore, an acute to chronic ratio of 3.23 for Cu in saltwater (USEPA, 2003) was used to estimate the threshold for chronic toxicity that could be used in the joint probability curve (JPC) and the USEPA CCC of 1.9 µg/L for Cu was used for calculation of the chronic HQ.

3.3. Probabilistic ecological risk assessment

When mean concentrations of Cu from a national program to monitor marine waters, were compared to the PNEC of 1.9 µg Cu/l, HQs of Cu in Chinese offshore exceeded 1.0 in all major areas studied (Table 2). In the period of 2005 to 2012, greatest hazard was indicated for Liaodong and Bohai Bays and the estuary of the Yellow, with HQ exceeding 1.0 during all periods. The HQ for Liaodong Bay was 6.0 in 2010, while that of Bohai Bay was 3.2 in 2006. The HQ for Cu in the Yellow River Estuary decreased from 4.6 to 1.4 between 2005 and 2012. In the period 2005 to 2012, HQs exceeding 1.0 were observed most of the time for Jiaozhou Bay, and estuaries of the Minjiang and Pearl Rivers. There have a low risk in Yangtze River estuary, Hangzhou Bay and Beibu Bay
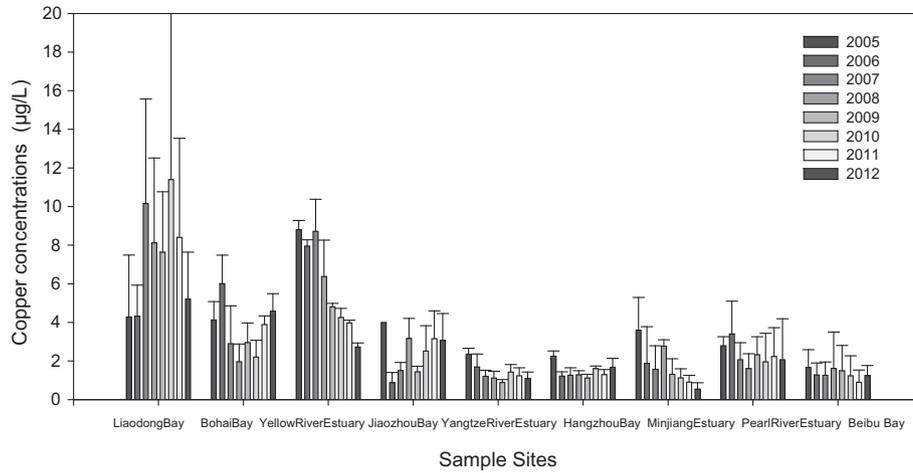
Fig. 3. The exposure concentrations of copper in Chinese major gulf from 2005 to 2012, expressed as mean ± standard deviation.
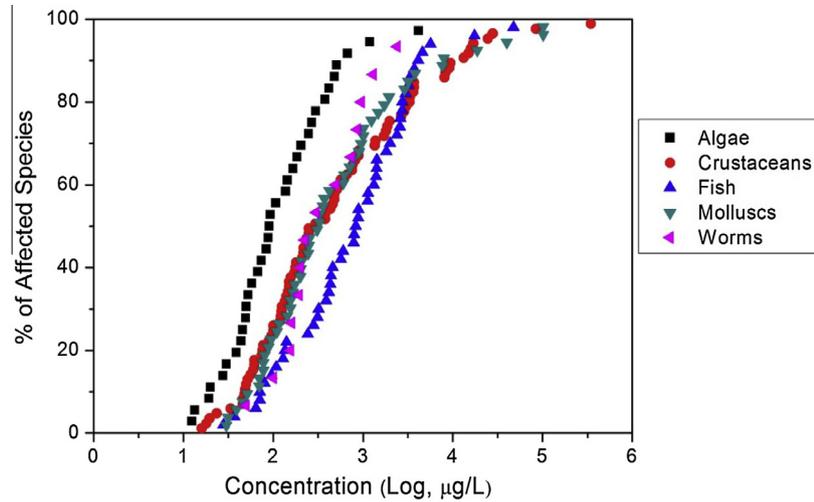

Fig. 4. Marine taxon-specific species sensitivity distribution (SSD). Symbols for taxonomic groups are given in the right key.

Table 1

Marine taxon-specific 10% hazardous concentration (HC_{10}) and its 95% confidence interval (95% CI; µg/L) from the best fit model which must pass both Shapiro–Francia and Anderson–Darling tests, and have the minimum root mean square (Min-*RMS*). *N* indicates sample size.

Taxa	HC_{10} (95% CI; µg/L)	*N*	Best fit model	Min-*RMS*
Algae	25 (22, 28)[a]	35	Fisher–Tippett	0.0005
Crustaceans	40 (35, 45)	84	Fisher–Tippett	0.0005
Molluscs	55 (51, 59)	51	Fisher–Tippett	0.0003
Worms	71 (52, 98)	14	Log-normal	0.0029
Fish	89 (79, 101)	49	Weibull	0.0007

[a] One-way analysis of variance (ANOVA), $\alpha = 0.05$.

during 2005 to 2012, with HQ lower than 1 in most of the time, except for Yangtze River estuary and Hangzhou Bay in 2005 (Table 2).

The proportion of locations in Chinese offshore areas with HQs > 1.0 decreased from 64% in 2005 to 31% in 2012. In the meantime, the proportion of locations with negligible risk (HQ ⩽ 0.3) increased from 5.1% in 2005 to 16.3% in 2012. However, there were still some locations where potential ecological risks were indicated. The HQs of remaining locations ranged from 0.3 to 1.0, which indicated a small potential for adverse effects (Fig. 5).

Joint probability curves (JPCs) resulting from direct comparison of exceedance probability function (EXF, calculated as 100 – cumulative distribution function (CDF)) and SSD offer a representation of the overall risk (Fig. 6). The results of the JPC analysis indicate that Cu posed a potential risk on aquatic organisms' and reduced year by year. Overall probabilities, of exceeding the threshold for lethality to 5% of the species, across all locations, were 27.6%, 5.4%, 4.9%, 0.8%, 0.4%, 1.0%, 1.8% and 0.12% in 2005 to 2012, respectively.

Methods ranging from deterministic to probabilistic methods are usually recommended for ecological risk assessment, with the probabilistic approach serving to refine the risk estimated after more conservative lower tier approaches. Results of probabilistic ERAs can provide useful information for risk managers and decision makers. However, hazard quotients, especially the single-value estimates, still have the advantage of readily identifying chemicals that have a likelihood of producing effects on species. The single-value estimate method is useful as a screening tool that can help to focus risk assessments. The initial estimations found in the first tier of the present study, which predicted relatively large HQs, suggested that a generalized potential risk for the whole area could not be excluded. The higher tier approaches allowed the estimation of the proportional risk of measured Cu concentrations to marine species in Chinese seawater.

Table 2
The HQs of copper in Chinese major gulf from 2005 to 2012.

| Regions	HQs							
	2005	2006	2007	2008	2009	2010	2011	2012
Liaodong Bay	2.25 ± 1.69	2.27 ± 0.85	5.35 ± 2.85	4.27 ± 2.32	4.02 ± 1.65	6.0 ± 8.10	4.42 ± 2.71	2.74 ± 1.28
Bohai Bay	2.17 ± 0.50	3.16 ± 0.77	1.53 ± 1.03	1.04 ± 0.48	1.56 ± 0.53	1.16 ± 0.46	2.05 ± 0.24	2.42 ± 0.47
Yellow River Estuary	4.64 ± 0.25	4.18 ± 0.17	4.59 ± 0.87	3.36 ± 0.99	2.53 ± 0.09	2.24 ± 0.25	2.09 ± 0.08	1.44 ± 0.11
Jiaozhou Bay	2.10 ± 0	0.46 ± 0.28	0.79 ± 0.22	1.67 ± 0.55	0.76 ± 0.15	1.33 ± 0.69	1.66 ± 0.76	1.62 ± 0.73
Yangtze River estuary	1.24 ± 0.16	0.89 ± 0.35	0.64 ± 0.16	0.59 ± 0.18	0.47 ± 0.08	0.75 ± 0.21	0.65 ± 0.22	0.57 ± 0.18
Hangzhou Bay	1.18 ± 0.14	0.64 ± 0.12	0.67 ± 0.20	0.68 ± 0.11	0.59 ± 0.10	0.85 ± 0.07	0.68 ± 0.14	0.88 ± 0.24
Minjiang Estuary	1.89 ± 0.89	0.99 ± 1.0	0.83 ± 0.64	1.46 ± 0.17	0.69 ± 0.43	0.59 ± 0.25	0.48 ± 0.18	0.29 ± 0.17
Pearl River Estuary	1.47 ± 0.25	1.79 ± 0.89	1.09 ± 0.46	0.85 ± 0.41	1.23 ± 0.49	1.03 ± 0.78	1.18 ± 0.78	1.09 ± 1.12
Beibu Bay	0.88 ± 0.48	0.67 ± 0.33	0.66 ± 0.36	0.85 ± 0.99	0.79 ± 0.69	0.65 ± 0.54	0.47 ± 0.33	0.66 ± 0.27
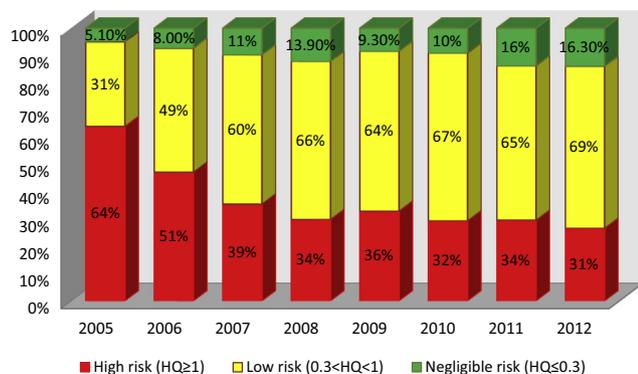
Fig. 5. The overall trend of HQs in Chinese offshore marine environments from 2005 to 2012.

3.4. Uncertainty analysis

Uncertainty in ERA is inevitable even when higher-tier methods are used. Sources of uncertainty in the ERA include: the variability of Cu concentrations within a single body of water, the ecological relevance of the toxicity data, and the risk characterization model. In particular, information on spatial and temporal variation in Cu concentrations was limited, especially for a nationwide monitoring. To describe exposures more completely, further information needs to be collected to describe concentrations of Cu at various spatial and temporal scales. In the present study, acute toxicity data and an assessment factor of 3.23 was used to construct joint
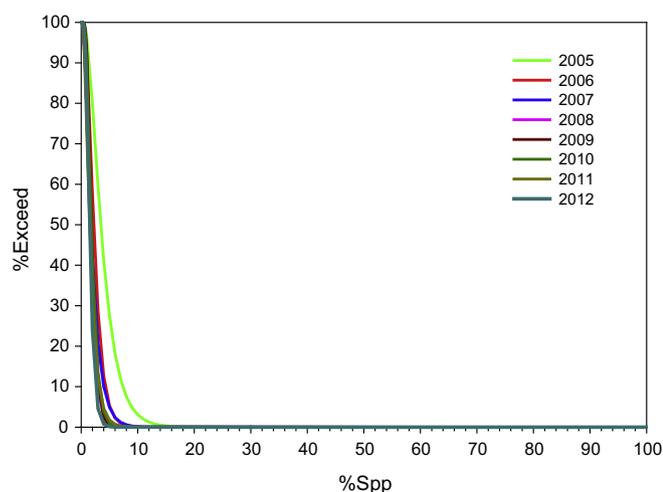
probability curve (JPC) due to chronic toxicity data were very limited, especially for marine species. Chronic toxicities, such as no observed effect concentrations (NOECs), are preferable to derive toxicity threshold in ERA to better mimic real-world scenarios of long-term exposure (Pennington, 2003), but there is a lack of available chronic data, especially for local species for site-specific ERAs. So, chronic toxicity data for various pollutants for site-specific species are critically needed in order to produce more accurate ecological risk assessments. In addition, the chemistry of copper affects its distribution, availability and ultimately its potential toxicity to aquatic biota. The most common oxidation states of copper are Cu(I) and Cu(II). Cu(I) is found primarily in anaerobic environments and is readily oxidized to Cu(II) which is the most environmentally relevant form. "Free" hydrated Cu(II) is generally considered to be the most toxic form to aquatic organisms. In seawater systems, copper speciation is dominated by naturally occurring organic ligands (e.g., humic acid), the major component of dissolved organic carbon responsible for binding copper. The bioavailable form of soluble Cu(II) is further reduced by inorganic speciation. Therefore, the importance of speciation and changes in copper chemistry, as reflected by the low percentage of Cu(II) in low particulate load estuarine laboratory water, that must be considered when predicting the ecological risk of copper in the environment based on dissolved copper concentrations (Hall and Anderson, 1999). Also, the lack of consensus on ERA methods precluded effective comparison among calculated risk levels by different risk assessors. Standard methods should be developed and employed by researchers and public agencies to evaluate ecological risk (Wang et al., 2009).

4. Conclusions

Concentrations of Cu in Chinese offshore marine waters ranged from 0.1 to 43.2 µg/L. Acute toxicity values ranged from 6 µg/L for the bivalve *Ostrea* sp., to 346,700 µg/L for the crab *Scylla serrata*. By examining the SSD and HC$_{10}$ values the sensitivity of marine taxa to Cu follows as: algae > crustaceans > mollusks > oligochaeta > fish ($p < 0.05$). Risks posed by Cu to ecological systems in Chinese offshore areas declined from 2005 to 2012. While the overall potential for adverse effects, as indicated by both HQ and JPC decreased, there was still potential for adverse effects due to Cu in some areas. The greatest risk of adverse effects was observed in Liaodong and Bohai Bays and the Yellow River Estuary, with lesser risk indicated for Jiaozhou Bay, and the estuaries of the Minjiang Pearl Rivers. The risk of effects of Cu on the ecosystems of the Yangtze River estuary, and Hangzhou and Beibu Bays was less.

Acknowledgements

Fig. 6. Joint probability curves for ecological risk of copper in Chinese offshore marine environments from 2005 to 2012.

Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.marpolbul.2015.03.005.

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Species Scientific Name	Species Common Name	Species Group	Conc_geometric
Algae	Algae	Algae, Moss, Fungi	58.00871847
Amphidinium carterae	Dinoflagellate	Algae, Moss, Fungi	138.539
Bacillariophyta	Diatom Division	Algae, Moss, Fungi	74.2040865
Ceratocorys horrida	Dinoflagellate	Algae, Moss, Fungi	166
Chlamydomonas bullosa	Green Algae	Algae, Moss, Fungi	49.569
Chlorella autotrophica	Green Algae	Algae, Moss, Fungi	19.21656556
Chlorella protothecoides	Green Algae	Algae, Moss, Fungi	13.3455
Chlorella pyrenoidosa	Green Algae	Algae, Moss, Fungi	509.12
Desmarestia menziesii	Algae	Algae, Moss, Fungi	420.8316853
Dunaliella minuta	Green Algae	Algae, Moss, Fungi	481.0735
Dunaliella salina	Green Algae	Algae, Moss, Fungi	377.487
Dunaliella tertiolecta	Green Algae	Algae, Moss, Fungi	4179.305298
Entomoneis punctulata	Algae	Algae, Moss, Fungi	12.43784547
Gonyaulax polyedra	Dinoflagellate	Algae, Moss, Fungi	82.54651795
Gracilaria tenuistipitata	Red Algae	Algae, Moss, Fungi	88.01117368
Gymnodinium splendens	Dinoflagellate	Algae, Moss, Fungi	52.7988623
Hormosira banksii	Neptune'S Necklace	Algae, Moss, Fungi	144.6319914
Isochrysis galbana	Haptophyte	Algae, Moss, Fungi	34.4647281
Isochrysis sp.	Haptophyte	Algae, Moss, Fungi	270
Lingulodinium polyedrum	Dinoflagellate	Algae, Moss, Fungi	90
Macrocystis pyrifera	Giant Kelp	Algae, Moss, Fungi	83.69240027
Nannochloris atomus	Microalgae	Algae, Moss, Fungi	27.61682182
Nitzschia closterium	Diatom	Algae, Moss, Fungi	19.94819928
Nitzschia paleacea	Algae	Algae, Moss, Fungi	107.2380529
Phaeodactylum tricornutum	Diatom	Algae, Moss, Fungi	69.62267312
Pseudokirchneriella subcapitata	Green Algae	Algae, Moss, Fungi	38.7655
Pyrocystis noctiluca	Dinoflagellate	Algae, Moss, Fungi	185
Rhodomonas salina	Marine Microalga	Algae, Moss, Fungi	30
Skeletonema costatum	Diatom	Algae, Moss, Fungi	247.845
Synechocystis aquatilis	Blue-Green Algae	Algae, Moss, Fungi	674.5368782
Tetraselmis sp.	Green Flagellate	Algae, Moss, Fungi	203.7154879
Tetraselmis suecica	Prasinophyte	Algae, Moss, Fungi	1190

Thalassiosira pseudonana	Diatom	Algae, Moss, Fungi	198.521447
Ulva fasciata	Sea Lettuce	Algae, Moss, Fungi	44
Ulva pertusa	Green Algae	Algae, Moss, Fungi	50.19960159