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# 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.

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#### 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).

http://dx.doi.org/10.1016/j.marpolbul.2015.03.005 0025-326X/© 2015 Elsevier Ltd. All rights reserved. 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

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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  $\mu$ g/L in surface seawaters and from 0.20 to 30  $\mu$ g/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; Pekev 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, (lin 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

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