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Copper and zinc, but not other priority toxic metals, pose risks to native aquatic species in a large urban lake in Eastern China[☆]

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ABSTRACT

Over the past 20 years, global production of copper (Cu) and zinc (Zn) rank in the top three compared to other metals such as Pb, Cd, Cr, Ni, As and Hg. However, due to the potential for exposure and toxicity to humans, more attention of environmental pollution was paid to other metals such as Cd and Hg. Aquatic organisms are sensitive to Cu and Zn. Even though internal concentrations of these required elements are homeostatically controlled, toxic effects can occur at the fish gill surface. In this work, concentrations in surface waters and toxic effects of Cu, Zn, Ni, Cr, Pb, Cd, As, Hg were determined and risk of various metals in Tai Lake, China were evaluated using both risk quotients and joint probability distributions. Two transition metals, Cu and Zn posed the greatest risks to aquatic organisms while measured concentrations of other metals were less than thresholds for adverse effects. Approximately 99.9% and 50.7% of the aquatic organisms were predicted to be affected by Cu and Zn in surface water of Tai Lake respectively. Our results highlight ecological risks of Cu and Zn in water of a typical, large, urban lake in Eastern China, which was ignored in the past.

1. Introduction

Results of recent studies have shown that concentration of Cu and Zn in many species of organisms were greater than those of Cr, Pb and Cd in Tai Lake, a large, shallow, urban lake of southeastern China (Yu et al., 2012a). Although other metals such as cadmium (Cd) and mercury (Hg) were thought to pose greater risks to humans, high levels of Cu and Zn in Tai Lake aroused concern especially for their ecological risk. During the past 20 years, annual production of Cu and Zn in both China and the world ranked in the top three metals compared to other metals such as Pb, Cd, Cr, Ni, As and Hg (Fig. 1). Zn and Cu are released to surface waters due to activities of humans as both point- and nonpoint-source. Production of Cu and Zn in China accounted for 8.4% and 7.9% of the world's production, respectively. Thus, considerable exposures to Cu and Zn were predicted for surface water, not only in China, but also other regions of the world.

Cu and Zn are nutritionally essential elements, concentrations of which are homeostatically regulated in tissues and thus their toxic effects on humans are relatively less in comparison to other more potent nonessential metals. However, just as too little of these required elements results in a deficiency, too much of these elements can be toxic when exposures exceeded required concentrations especially for aquatic organisms. Aquatic organisms are generally more sensitive to Cu and Zn in water than are humans. For instance, fishes and crustaceans were 10–100 times more sensitive to Cu than mammals (Förstner et al., 1979; Flemming and Trevors, 1989; Wright and Welbourn, 2002). This is generally interpreted as liver and kidney of mammals being part of a generally more mature detoxification system than other organisms (Wright and Welbourn, 2002). Accordingly, some countries have enacted more stringent standards to protect aquatic organisms from effects of Cu and Zn than for protection of health of humans from exposure via drinking water. For instance, Chinese standards for waters containing economically valuable fisheries, for which water quality criteria (WQC) are 10 and 100 μg/L, for Cu and Zn, respectively. These values

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are factors of 100 and 10 less than that for drinking water, for which standards are 1000 μg/L both Cu and Zn (Table 1). Chronic water quality reference values of Cu and Zn provided by USEPA for protection of freshwater aquatic life are 9 μg Cu/L and 120 μg Zn/L, are also significantly less than that for protection of human health, which are 1300 μg Cu/L and 7400 μg Zn/L (USEPA, 1985, 2002).

With increasing emissions to water, and sensitivity of aquatic species, Cu and Zn in aquatic environments might pose greater potential risks to aquatic organisms. Although predecessors have conducted research on environmental behavior of Cu and Zn, ecological risks of nutritionally essential metals Cu and Zn, in water of lakes were still largely unknown (Hough et al., 2004; Yu et al., 2012a). Previous studies have looked at ecological risk of metals in sediments of freshwater ecosystems in China but without considering region distribution of aquatic organisms (Niu et al., 2010; Song, 2014). Here an assessment of risk of metals to native, aquatic organisms was conducted in Tai Lake, China. To aid regulators and scientists about whether focus their concerns and efforts on the ecological risk of Cu and Zn in freshwater lake of Eastern China, this assessment focused on the ecological risk of Cu and Zn on a range of different species of aquatic organisms in Tai Lake.

2. Materials and methods

2.1. Site description

Tai Lake, China's third largest freshwater lake, is located along downstream reaches of the Yangtze River (Yangtze Delta), in the southeast of China. Tai Lake covers a water area of 2338 km^2 with average water depth of 1.89 m. The regional climate of Tai Lake is subtropical monsoon, with a long-term mean, annual temperature of 15–17 °C (Yu et al., 2013). Tai Lake region is one of China's most economically developed regions, with a large human population and dense industrial development. The human population in the region surrounding Tai Lake accounts for approximately 3% of the population China, but 12% of the gross domestic product (GDP). Tai Lake provides drinking water for more than 2 million people, and sustains one of China's most important fisheries for crabs, carp, and eels. With rapid economic development over the last 20 years, large amounts of pollutants discharged into Tai Lake, the quality of its water has decreased (Guo, 2007; Qiao et al., 2006; Stone, 2011).

2.2. Quantification of metals exposure in surface water of Tai Lake

Samples of surface water were collected from 40 locations in 7 districts throughout Tai Lake in September 2010 and July 2011. The sampling sites included Zhushan Lake (Locations 1, 17, 18, 19, 20, 40), Meiliang Bay (Locations 21, 22, 23, 24, 25), West Coast (Locations 2, 3, 4, 5, 6, 39), Gong Lake (Locations 26, 27, 28, 29, 30), South Lake (Locations 7, 8, 9, 10, 11, 12), East Coast (Locations 31, 32, 33, 34, 35) and Center area of Lake (Locations 13, 14, 15, 16, 36, 37, 38) (Fig. 2). Samples of water were collected from each location. To avoid metals contamination of water samples during sampling, all sampling and filtration equipment used were plastic, acid-soaked and rinsed with Milli-Q water in advance. During sampling, surface water sample was collected by one acid-cleaned plastic bottle, and then was filtered by disposable 0.45-μm membrane filter head and plastic syringe in the field. Filtered samples of water were stored in another acid-cleaned plastic bottle, which was acidified to 1% v/v with ultrapure HCl, and immediately placed in ice-packed coolers and transported to the laboratory where they were stored at 2 °C until analysis.

Concentrations of dissolved Cr, Cd, Pb, Zn, Ni, Cu, As and Hg in water were determined using Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7500, Agilent, United States (US)). Concentrations of dissolved As and Hg in water were determined using Hydride Generation-Atomic Fluorescence Spectrometer (HG-AFS, PSA-10.055, Millennium Excalibur System, United Kingdom (UK)). Operating conditions of the ICP-MS and AFS instruments were optimized and all calibration curves demonstrated good

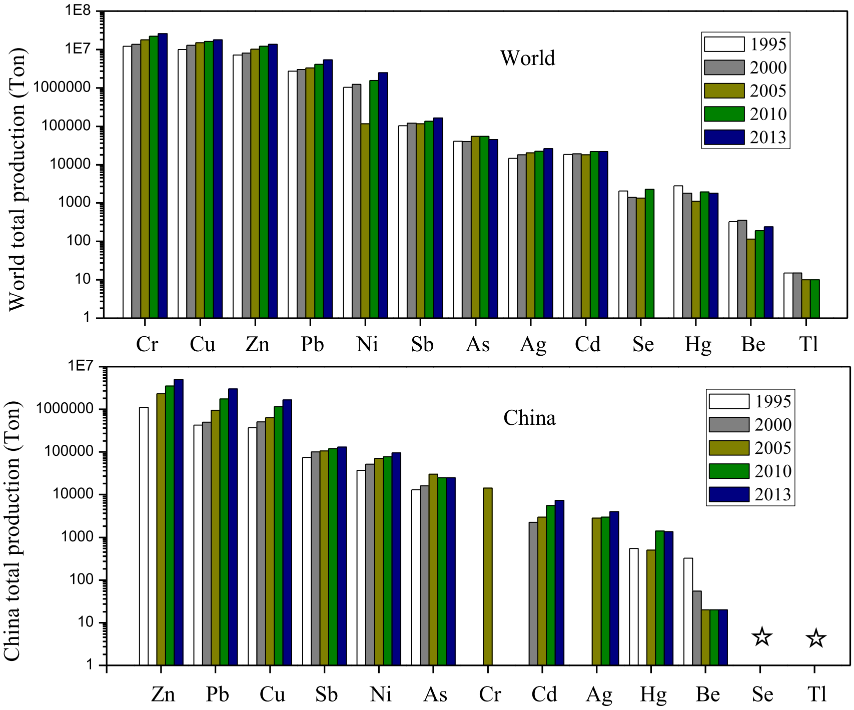

Fig. 1. World and Chinese annual production of metals (1995–2013) (The data of world refer to (USGS, 1995–2013); The data of China refer to (Chen et al., 2005; USGS, 1995–2013; Wu et al., 2013; Yuan et al., 2012); ☆ refer to no reference data).

Table 1
Concentrations of eight metals in water of Tai Lake and respective Chinese water quality standards (μg/L) (Administration of Environmental Protection of China, 1989, 2002; Ministry of Health of China, 2006).

	Concentration	Range	Standard of surface water			Fisheries water quality standards	Drinking water quality standards
			Igrade	IIgrade	III grade		
Zn	70	18–1246	50	1000	1000	100	1000
Cu	19	2.4–171	10	1000	1000	10	1000
Cr	40	32–76	10	50	50	100	50
Pb	17	9.9–30	10	10	50	50	10
Ni	20	17–31	–	–	–	50	20
As	4.5	0.67–12	50	50	50	50	10
Cd	0.85	0.76–1.1	1	5	5	5	5
Hg[a]	4.8	0.7–246	50	50	100	500	1000

[a] Units of Hg concentrations expressed in ng/L.

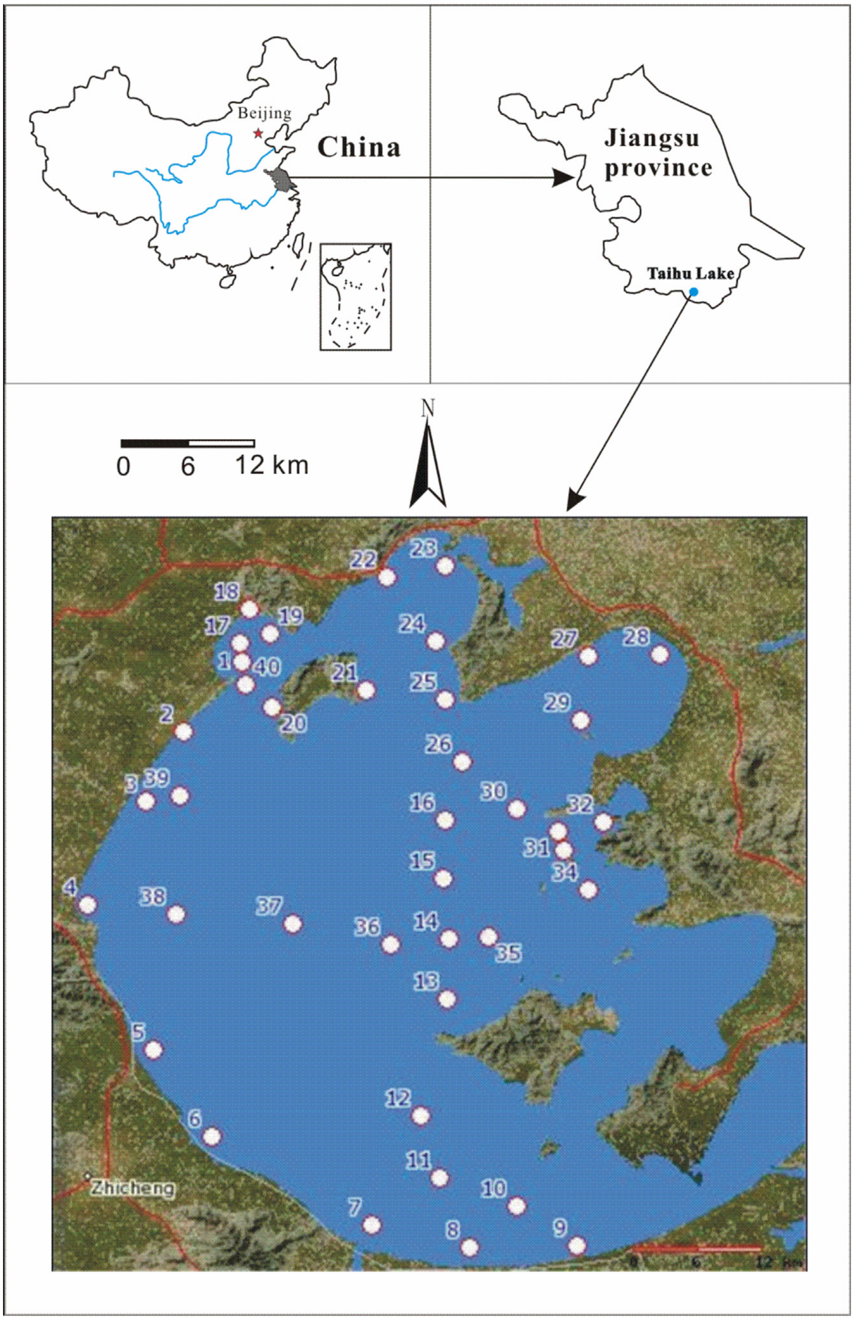

Fig. 2. Map of the study area with sampling locations.

linearity ($r > 0.999$). Quality control included the analyses of method blanks, blank spikes and matrix spikes. All method blanks contained less of each metal than the corresponding element detection limits (<100 ng/L for all metals). The percentage of recoveries of spikes in samples ranged from 89 to 106% for metals in all samples. Cumulative probability functions were constructed by fitting concentrations of each metal. Concentrations of metals measured in Tai Lake were used to develop exposure concentration distribution (ECD) curves.

2.3. *Acquisition of toxicity data for metals*

Toxicity data for metals were acquired based on collection, analysis and screening of the data from the US Environmental Protection Agency (USEPA) Toxicity Database (http://cfpub.epa.gov/ecotox/) and the China National Knowledge Infrastructure (CNKI) Database (http://www.cnki.net/). No Observed Effect Concentration (NOEC) of chronic toxicity data were collected as toxicity data. Compared with acute toxicity data, chronic toxicity data can better characterize the long-term toxic effects of toxic substances on organisms and thus, are more suitable for assessment of potential effects of long-term exposure to metals in aquatic environment. If NOEC data were insufficient to construct species sensitivity distributions (SSDs), Lowest Observed Effect Concentration (LOEC) or Maximum Acceptable Toxicant Concentration (MATC) were converted ($\mathrm{NOEC} = \mathrm{LOEC}/2$ or 10% EC10, $\mathrm{NOEC} = \mathrm{MATC}/\sqrt{2}$) (EU, 2003).

The ultimate adopted toxicity data for metals were screened based on the following criteria: First, tested organisms had to be present in Tai Lake; Second, duration of exposure was longer than 4 days in fresh water. NOEC data for toxic potencies of eight metals on existing aquatic species in Tai Lake were used to develop SSD curves. Table S2—Table S9 (see Supporting Information (SI)) provides the chronic toxicity data, representative species as well as data sources for each metal on aquatic organisms native to Tai Lake. While lognormal, log-logistic, Weibull and Gauss fitting functions can be used to construct SSD curves (Chen et al., 2015; Wheeler et al., 2002; Wu et al., 2010), in this work, data were fit with the log-logistic function.

2.4. *Ecological risk assessment methods of metals in Tai Lake*

It was recognized that deterministic methods are the primary tool for lower tiers of risk assessment, but might result in upwardly biased estimates of hazard resulting from use of upper bound input variables. While probabilistic methods constitute one of several approaches that may be used for higher tier assessments, which may lessen or avoid such problem of deterministic methods (Hope, 2006; USEPA, 2004). In order to obtain more realistic estimates of exposure and effect of metals in water, both deterministic method (risk quotients) and probabilistic method (joint probability) were used to assess ecological risk in this study. Based on constructed environmental ECD and SSD curves, risk quotients were obtained by comparing $C_{0.9}$ exposure (ECD curve at 90% concentration) and $C_{0.1}$ toxicity (SSD curve at 10% concentration). Risk quotients >1 suggest potential risks, whereas risk quotients <1 suggest no risks to aquatic communities (Solomon et al., 1996). The joint probability method is a semi-probabilistic approach in which probabilities of exposure are compared to a point estimate of effect, which is based on the probability of a species being affected. The joint probability distribution was used to determine risk of metals to aquatic organisms in Tai Lake (Giesy et al., 1999; Solomon et al., 2000; Solomon and Giesy, 2001). Finally, confidence limits were calculated to give the probability that ten (10%) percent of species would be affected with a 95% confidence interval around that estimate.

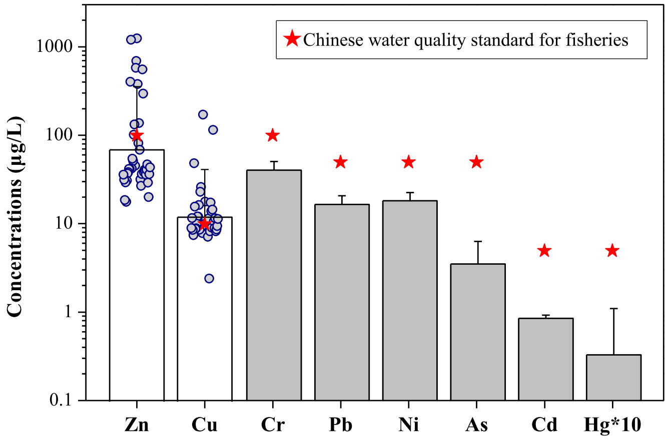

Fig. 3. Concentrations of eight metals in water of Tai Lake and comparison with Chinese water quality standards for fishes.

2.5. *Statistical analyses*

Origin 8. 0 were used to construct ECD and SSD curve of metals. The statistical package SPSS for windows 16.0 (SPSS Inc., Chicago, Illinois, USA) was used for data analyses. Normality of NOEC data was confirmed by the Kolmogorov-Smirnov test.

3. Result and discussion

3.1. *Distributions of concentrations of metals in water of Tai Lake*

Chromium, Cd, Pb, Zn, Ni, Cu, As and Hg were all detected in water of Tai Lake (Table 1). Zinc and Cr exhibited greatest concentrations among the eight metals studied. Concentrations of Cu were comparable to concentrations of Pb and Ni. Mean concentrations of Cu and Zn in water were 19 (2.4–171) μg/L and 71 (18–1246) μg/L, respectively. Concentrations of Cu and Zn in 45% and 28% of samples of water exceeded corresponding Chinese water quality standards (WQS) for protection of fisheries, but the other six metals did not exceed their respective WQSs (Table 1 and Fig. 3).

Orders of metal concentrations in water of Tai Lake were roughly consistent with production of metals over the past two decades in China (Figs. 1 and 3). Similarly, greatest concentrations of Zn, Cr and Ni were also reported by other studies for water (Yu et al., 2012b) and sediments of Tai Lake (Bing et al., 2011; Jin et al., 2010; Liu et al., 2011; Zhang et al., 2012) (SI, Table S11). Concentrations of metals in water of Tai Lake were greater than those in three other major, freshwater lakes in China, including Chaohu (Tong et al., 2006), Dongting (Yang et al., 2008) and Poyang (Hu et al., 2012) (Table S1).

Overall, greater concentrations of metals in water were found in western and northern areas of Tai Lake, where industry and agriculture are more intense (Fig. S1). Previous results have indicated that western Tai Lake contributed about 80% of pollutants into Tai Lake (Chi and Zhu, 2005; Ma et al., 2010).

Table 2
Risk quotients for metals in Tai Lake.

	Cu	Zn	Ni	Cr	Pb	Cd	As	Hg
$C_{0.1\mathrm{toxicity}}$ (μg/L)	0.17	20	14	31	15	0.95	22	2.3
$C_{0.9\ \mathrm{exposure}}$ (μg/L)	17	193	24	48	21	0.94	8.9	0.14
Risk quotients	101	9.5	1.7	1.6	1.4	0.99	0.40	0.06

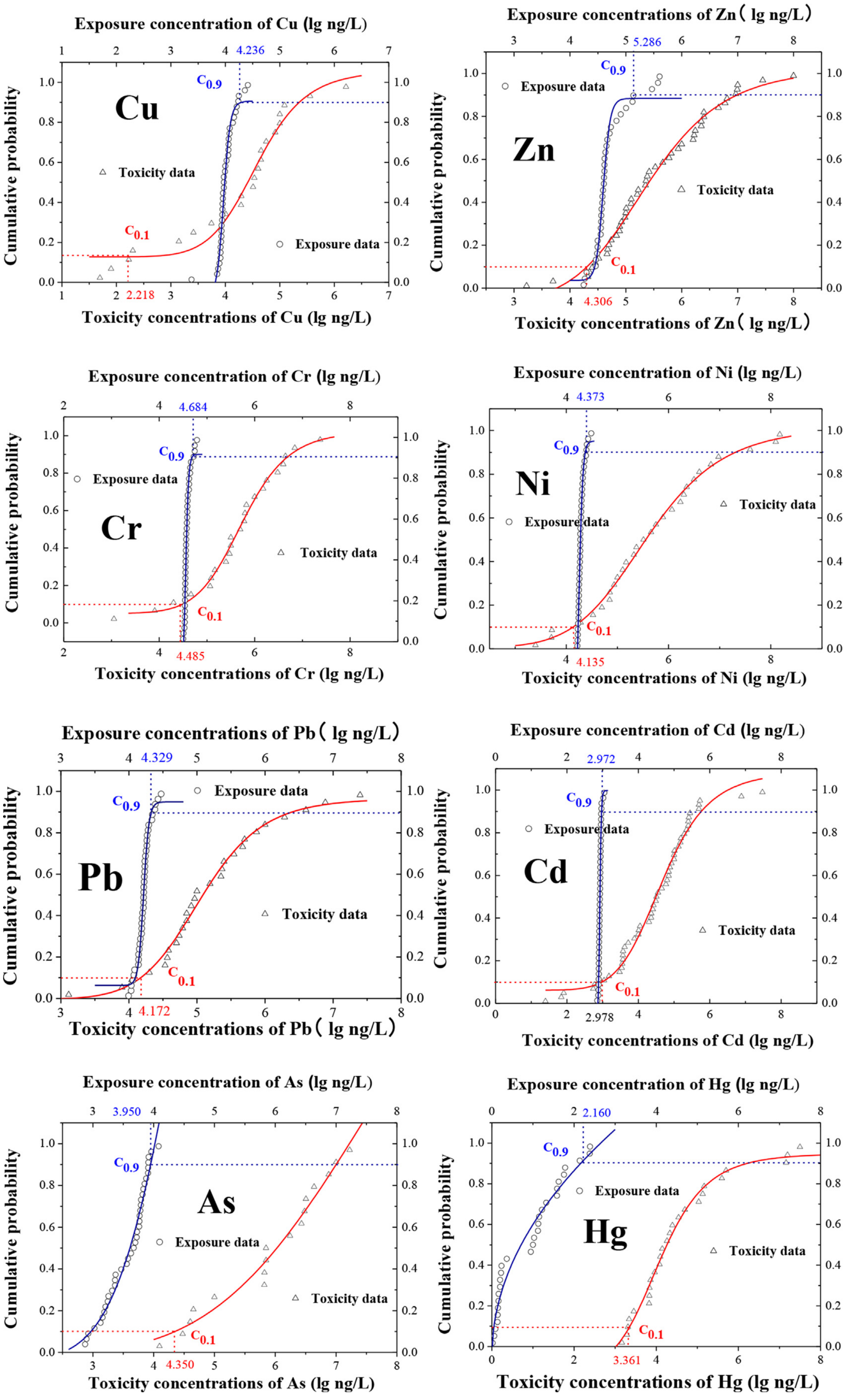

Fig. 4. ECD and SSD curves for metals in surface water of Tai Lake.

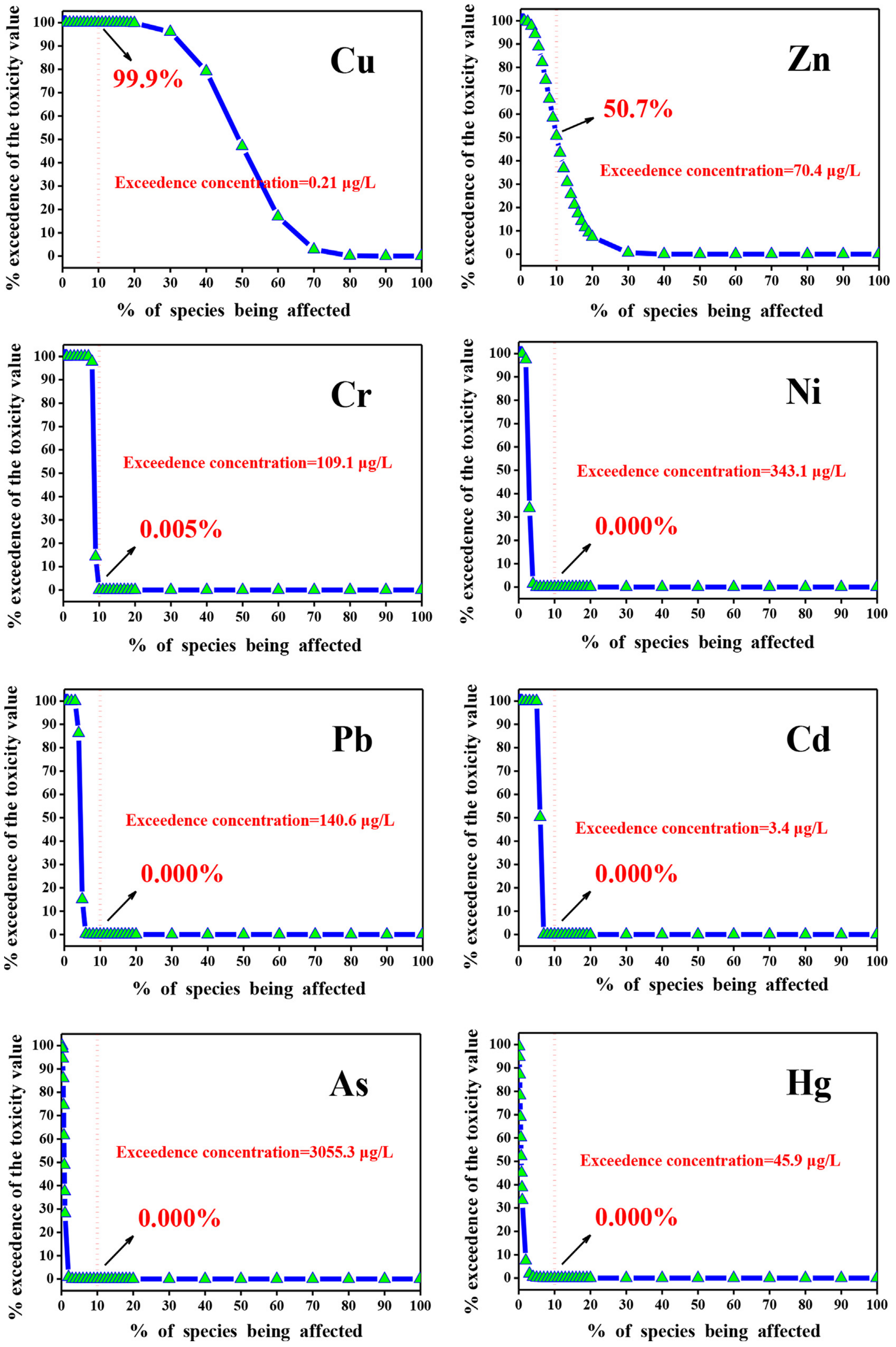

Fig. 5. Joint probability curves of metals in surface water of Tai Lake.

3.2. Species sensitivity and toxic effects of metals in Tai Lake

Aquatic species in Tai Lake include Chordata, Mollusca, Arthropoda, Annelida, phytoplankton, aquatic plants and other biological categories, including more than 400 species and 146 families (Su, 2011; Su et al., 2011). Results of the Kolmogorov-

Smirnov test of log-transformed NOEC data ($p > 0.05$) confirmed that log-transformed data were sufficiently described by the normal probability function. Coefficients of determination (R^2) of ECD and SSD curves were both near 1.0. Coefficients of Reduced Chi-Square tests and Residual Sum of Squares were also small. These results indicated that the curves were well fitted. Thus, it was deemed appropriate to use these data to assess risks of metals to aquatic organisms.

Concentrations to affect 10% of Species ($C_{0.1\text{toxicity}}$) of individual metals, determined by use of SSDs, increased in the following order of most to least potent: Cu < Cd < Hg < Ni < Pb < Zn < As < Cr (Table 2). Although As can cause adverse effects on humans at environmentally relevant concentrations, it was less potent toward aquatic organisms. In fact, differences in potencies of Cu, Zn between humans and aquatic organisms were as much as several orders of magnitude. When sensitive species of fishes and amphibians were exposed to 5–10 μg Cu/L in drinking water, resulted in teratogenicity or death of embryos (Beck et al., 2002). In the crustacean, water flea, activity of enzyme chitinase was reduced by 20%–40% when exposed to 100–500 μg Zn/L, and resulted in lethality when exposed to 1000 μg Zn/L (Poynton et al., 2007).

3.3. Ecological risk of metals in Tai Lake

Based on ECD and SSD curves (Fig. 4), risk quotients of metals in Tai Lake were obtained (Table 2). Risk quotients for concentrations of Cu, Zn, Ni, Cr, Pb in Tai Lake were greater than 1.0 (Cu: 101, Zn: 9.5, Ni: 1.7, Cr: 1.6, Pb: 1.4). In particular Cu and Zn in water exhibited the greatest ecological risk to aquatic species of Tai Lake. These results were consistent with results of the joint probability method (Fig. 5), which indicated the greatest probabilities of ecological risk were 99.9% for Cu and 50.7% for Zn in surface water of Tai Lake. Other metals exhibited lesser probabilities of ecological risk with probabilities ranging from 0.000% to 0.005% in surface waters of Tai Lake (Fig. 5). Exceedence concentrations (i.e. a concentration would affect 10% of species) were 0.21, 70, 109, 343, 141, 3.4, 3055 and 46 μg/L for Cu, Zn, Cr, Ni, Pb, Cd, As and Hg, respectively (Fig. 5). Mean concentrations of Cu (19 μg/L) and Zn (70 μg/L) in Tai Lake exceeded or nearly exceeded corresponding threshold concentrations.

Recently a study using risk-ranking of metals identified Cu, Al, Zn and Ni as metals that posed the greatest threat to freshwater organisms in the UK (Donnachie et al., 2014). In order to provide an unbiased view of what a metal's potential threat to the environment might be, Donnachie et al. (2014) devoted tremendous effort to take toxicity data straight from the literature regardless of tested species and has allowed all possible effects to be considered. In contrast, our study focused on those tested organisms present in Tai Lake and screened with chronic toxicity data. Similarly, both of our results suggested that Cu and Zn posed greatest risk to freshwater organisms. In freshwater ecosystems of other regions of the world ambient concentrations of both Cu and Zn have been predicted to have adverse effects on aquatic organisms. For instance, an integrated risk assessment on a national scale performed in The Netherlands, indicated that concentrations of Cu and Zn in soils result in risks in surface water. Critical limits of Cu and Zn for impacts on aquatic organisms in surface water were exceeded throughout The Netherlands (De Vries et al., 2008).

These results stress the need for harmonization of water policy to reduce the load in surface water to meet targets for protection of aquatic organisms. China had suggested five metals including Pb, As, Cd, Cr and Hg as priority pollutants for control. Because Cu and Zn were not deemed to be toxicologically relevant for protection of health of humans, Cu and Zn were not listed as priority pollutants and thus, although China developed stringent standards for concentrations Cu and Zn in water for protection of fisheries, controlling limits of metals in water of Lakes were not implemented as part of the "Chinese water quality standard for fisheries" (Administration of Environmental Protection of China, 1989) and a grade I standard of "Chinese Surface Water Quality Standards" (10 and 50 μg/L for Cu and Zn respectively were available), they were generally implemented according to grade II and III standards (1000 μg/L for both Cu and Zn) (Administration of Environmental Protection of China, 2002). This management strategy is adequate for protection of human health, but would not be considered sufficiently protective to guarantee safety of concentrations of Cu and Zn for aquatic species. More stringent standards for emissions might ultimately need to be made to reduce ecological risks posed by Cu and Zn in the aquatic environment.

3.4. Uncertainty analysis

Under natural conditions, changes in pH, organic carbon, hardness or particulate matter of water can alter bioavailability and toxicity of metals such as Cu. Therefore, in order to assess the most realistic exposure and risk of metals, we measured dissolved metals in water as the exposure concentrations (USEPA, 1985, 2007). Nevertheless, in most cases bioavailable fractions of metals account only for a proportion of the total.

Results obtained by use of the joint probability method allowed estimation of proportional risk of measured metal concentrations to aquatic species in Tai Lake. These results can provide useful information for risk managers and decision makers. However, it should be noted that neither probability of overlap between distributions of exposure and toxicology data, nor joint probability curves are very accurate as quantitative predictors of risk by themselves. Rather, they provide assessors and managers of risks with information on relative risk. The joint probability method gives the probability of exceeding thresholds for any probability of effect, which is more flexible and allows risk assessors to look at a range of scenarios for levels of protection desired.

4. Summary

In the present study we found that, in comparison to other metals, Cu and Zn in water posed the greatest ecological risk to aquatic species in waters of Tai Lake in Eastern China. The greatest probabilities of ecological risk were 99.9% for Cu and 50.7% for Zn in surface water of Tai Lake. Other metals exhibited lesser probabilities of ecological risk with probabilities ranging from 0.000% to 0.005% in waters of Tai Lake. Studies are needed to determine whether dose–response relationships established for Cu and Zn for aquatic organisms is valid in Tai Lake and other lakes of China.

Acknowledgements

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://

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Supporting Information:

Copper and Zinc, but not Other Priority Toxic Metals, Pose Risks to Native Aquatic Species in a Large Urban Lake in Eastern China

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Table S1. Concentrations of metals in the five major freshwater lakes of China.

Lake	Zn	Cu	Cr	Pb	Ni	As	Cd	Hg (μg/L)	References
Tai	70 (18-1246)	19 (2.4-171)	40 (32-76)	17 (9.9-30)	20 (17-31)	4.5 (0.7-12)	0.85 (0.8-1.1)	4.8 (0.7-246)	This study
Tai								5.2 (2.3-10)	[1]
Chao	19(7.5-47)	6.0(4.5-7.7)	7.1(2.8-11)	NA	NA	NA	0.8		[2]
Dongting	6.0(5.0-9.9)	1.5 (0.9-2.3)	NA	1.5(1.1-2.0)	0.6(0.5-0.6)	1.2(0.8-1.8)	0.1(0.1-0.2)		[3]
Poyang	16 (5.9-30)	4.7 (3.1-6.2)	4.4(2.5-6.0)	4.4(2.7-6.0)					[4]
Hongze	NA	NA	NA	NA	NA	NA	NA		

Table S2. Chronic toxicities of zinc (Zn) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	15	1.7-10000	*Pseudokirchneriella subcapitata*, *Oocystis submarina*, *Surirella ovata*, etc.	[5-27]
Aquatic plants	7	160-28117	*Potamogeton natans*, *Lemna minor*, *Myriophyllum sibiricum*, etc.	[28-35]
Fish	11	33-9859	*Cyprinus carpio*, *Cottus bairdi*, *Carassius auratus*, etc.	[36-55]
Crustaceans	9	25-2500	*Ceriodaphnia dubia*, *Atyaephyra desmarestii*, *Echinogammarus meridionalis*, etc.	[6; 38; 39; 56-76]
Molluscs	4	47-224	*Mytilus galloprovinciaLis*, *Perna viridis*, *Potamopyrgus jenkinsi*, etc.	[77-87]
Rotifers	1	2500	*Brachionus plicatilis*	[88]

Table S3. Chronic toxicities of copper (Cu) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	4	0.08-100	*Diatoma tenue var. elongatum*, *Chlamydomonas reinhardtii*, *Pseudokirchneriella subcapitata*, etc.	[89-101]
Aquatic plants	6	0.05-360	*Lemna minor*, *Nymphoides pelatum*, *Ceratophyllum demersum*, etc.	[32; 102-104]
Fish	5	0.2-1639	*Cyprinus carpio*, *Anguilla anguilla*, *Fundulus heteroclitus*, etc.	[105-112]
Crustaceans	4	10-40	*Palaemonetes pugio*, *Gammarus pulex*, *Daphnia magna*, etc.	[56; 113-125]
Insects	1	83	*Chironomus riparius*	[126-130]
Molluscs	1	5.6	*Mytilus galloprovincialis*	[85]
Annelid	1	0.17	*LumbricuLus variegatus*	[131]

Table S4. Chronic toxicities of chromium (Cr) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	5	8.0-670	*Glenodinium halli*, *Chlorella vulgaris*, *Scenedesmus quadricauda* , etc.	[132]
Aquatic plants	4	122-1000	*Ipomoea aquatica*, *Lemna minor*, *Azolla pinnata* , etc.	[33; 133]
Fish	11	1.1-23515	*Channa punctat*, *Lepidocephalichthys guntea*, *Cyprinus carpio*, *Fundulus heteroclitus*, *Danio rerio* , etc.	[134]
Crustaceans	2	20-250	*Daphnia magna*, *Palaemonetes pugio* , etc.	[135; 136]
Rotifers	1	1788	*Brachionus calyciflorus*	[137]

Table S5. Chronic toxicities of lead (Pb) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	8	50-500	*Microcystis aeruginosa*, *Oocystis submarina*, *Scenedesmus quadricauda*, etc.	[12; 13; 16; 18; 138-140]
Aquatic plants	5	155-25000	*Azolla pinnata*, *Eichhornia crassipes*, *Typha latifolia*, etc.	[12; 28; 33; 34; 141-144]
Fish	6	1.3-470	*Cyprinus carpio*, *Epinephelus coioides*, *Danio rerio*, etc.	[43; 145; 146]
Crustaceans	4	7.9-1936	*Palaemonetes pugio*, *Gammarus pulex*, *Daphnia magna*, etc.	[48; 53; 60; 66; 68; 69; 73; 117; 147-152]
Molluscs	3	34-750	*Mytilus edulis*, *Biomphalaria glabrata*, *Anodonta imbecillis*, etc.	[80; 153-157]
Insects	1	82	*Chironomus plumosus*	[158; 159]

Table S6. Chronic toxicities of nickel (Ni) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	11	2.5-4000	*Glenodinium halli*, *Microcystis aeruginosa*, *Oocystis submarina*, *Scenedesmus quadricauda*	[11-13; 16; 19; 25; 138; 140; 160-162]
Aquatic plants	3	148-324	*Lemna minor*, *Ipomoea aquatica*, *Lemna aequinoctiales* , etc.	[33; 34; 144; 163; 164]
Fish	10	5.1-150000	*Cyprinus carpio*, *Leuciscus cephalus*, *Lepomis macrochirus*, *Oncorhynchus mykiss*, *Danio rerio* , etc.	[43; 48; 165-171]
Molluscs	2	455-2000	*Dreissena polymorpha*, *Goniastrea aspera*, etc.	[172; 173]
Insects	1	777	*Chironomus plumosus*	[174]
Crustaceans	2	127-210	*Daphnia magna*, *Americamysis bahia*	[60; 175-177]

Table S7. Chronic toxicities of arsenic (As) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	4	30-10000	*Microcystis aeruginosa*, *Scenedesmus quadricauda*, *Chlorella vulgaris*, etc.	[13; 138; 178-180]
Aquatic plants	2	707-3020	*Lemna minor*, *Azolla pinnata*	[29; 141]
Fish	7	13-16773	*Cyprinus carpio*, *Fundulus heteroclitus*, *Danio rerio*, *Jordanella floridae*, etc.	[181-188]
Crustaceans	3	100-3192	*Daphnia magna*, *Tigriopus japonicus*, *Americamysis bahia*	[60; 64; 189-191]
Molluscs	1	45	*Biomphalaria glabrata*	[156]

Table S8. Chronic toxicities of cadmium (Cd) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	10	0.025-100	*Anabaena sp.*, *Scenedesmus subspicatus*, *Staurastrum cristatum*, *Cyclotella meneghiniana* , etc.	[16]
Aquatic plants	8	10-7687	*Lemna minor*, *Azolla pinnata*, *Marsilea minuta* , etc.	[28; 33]
Fish	13	0.59-259	*Gobius niger*, *Cyprinus carpio*, *Cottus bairdi*, *Cyprinodon variegatus* , etc.	[53; 134]
Molluscs	5	0.082-132	*Perna perna*, *Elliptio complanata*, *Corbicula manilensis*, *Radix plicatula* , etc.	[192; 193]
Crustaceans	8	3.5-63	*Mesocyclops hyalinus*, *Moina macrocopa*, *Ceriodaphnia dubia*, *Atyaephyra desmarestii*, *Echinogammarus meridionalis* , etc.	[124; 136]
Insects	1	4.2	*Chironomus riparius*	[194]
Rotifers	3	26-30000	*Brachionus calyciflorus*, *Brachionus plicatilis*, *Brachionus rubens* , etc.	[195]

Table S9. Chronic toxicities of mercury (Hg) to aquatic organisms in Tai Lake.

Aquatic species	Number of species	Range of toxicity data (μg/L)	Representative species	Data sources
Algae	4	6.9-35	*Gymnodinium splendens*, *Chlorella vulgaris*, *Scenedesmus quadricauda*, etc.	[196]
Aquatic plants	3	135-14550	*Lemna aequinoctiales*, *Azolla pinnata*, *Enhalus acoroides*	[33; 141; 197; 198]
Fish	11	1.5-31498	*Danio rerio*, *Leuciscus cephalu*, *Pomatoschistus microps*, etc.	[171; 199-206]
Crustaceans	2	2.9-9.2	*Ceriodaphnia dubia*, *Daphnia magna*	[207; 208]
Molluscs	4	12-108	*Mytilus edulis*, *Perna viridis*, *Anodonta imbecillis*, etc.	[209-211]
Rotifers	2	13-22	*Brachionus calyciflorus*, *Brachionus plicatilis*	[212-214]

Table S10. Curve fitting parameters.

Metals	Items	Fitting parameters of cumulative distribution curve				Determination coefficient	Coefficients of Reduced Chi	Residual Sum of Squares
		A_1	A_2	x_0	p	R^2	Reduced Chi-Sqr	Residual Sum of Squares
Cd	Toxicity	0.06	1.1	4.7	7.2	0.97	1.0E-03	0.06
	Exposure	-0.08	0.99	2.9	134	0.99	8.2E-04	0.03
Cr	Toxicity	0.05	1.0	5.7	12	0.99	7.4E-04	0.01
	Exposure	-0.21	0.92	4.6	152	0.98	2.0E-03	0.05
Ni	Toxicity	0.01	1.0	5.6	7.5	0.99	4.4E-04	0.01
	Exposure	-0.38	0.95	4.3	116	0.99	4.2E-04	0.02
Cu	Toxicity	0.13	1.1	4.6	10	0.97	2.9E-03	0.05
	Exposure	-0.08	0.9	4.0	55	0.97	2.6E-03	0.09
Zn	Toxicity	-0.06	1.0	5.4	7.8	0.99	9.7E-04	0.04
	Exposure	0.04	0.88	4.6	74	0.98	1.8E-03	0.05
Pb	Toxicity	0	0.97	5.1	11	0.99	8.4E-04	0.02
	Exposure	0.06	0.95	4.2	107	0.99	7.6E-04	0.03
As	Toxicity	-0.02	2.9	8.2	5.1	0.97	2.7E-03	0.04
	Exposure	-0.09	7.9	5.4	6.3	0.98	1.9E-03	0.07
Hg	Toxicity	-0.11	0.95	4.1	7.2	0.99	1.2E-03	0.03
	Exposure	-0.05	3.7E+03	4.3E+07	0.49	0.95	9.5E-01	0.10

Table S11. Concentrations of metals in sediments of Tai Lake.

Zn	Cr	Ni	Pb	Cu	As	Cd	References
104	81.3	35	32	27		0.54	[215]
313 (255-385)	72(71-73)		59(53-65)	56 (43-74)		5.8(2.6-13)	[216]
64 (62-66)	67 (62-73)	34(31-37)	16 (14-19)	18.6(18.5-18.6)			[217]
					10 (6.5-14)		[218]

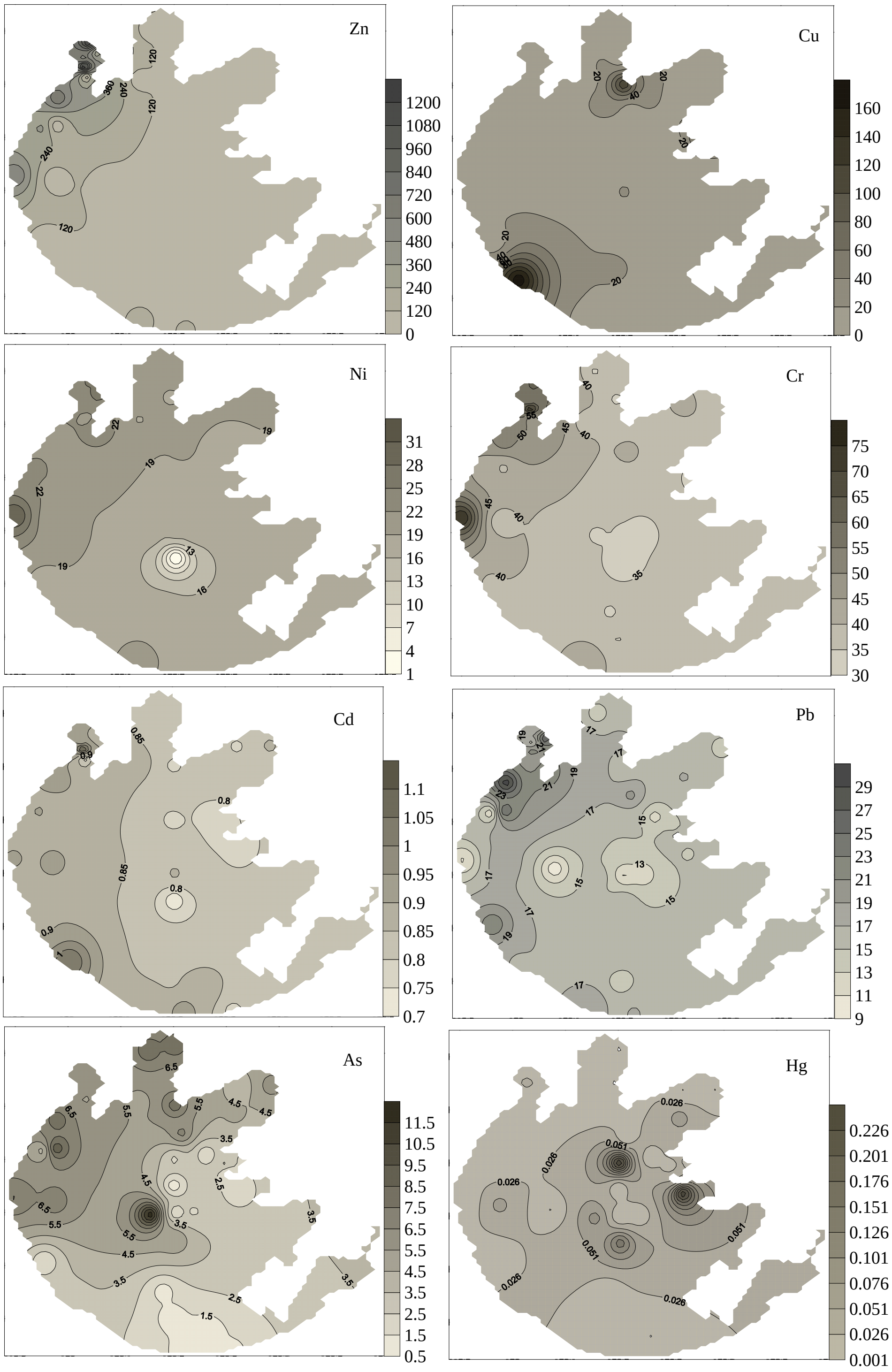

Figure S1. Spatial distribution of metals in Tai Lake.

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