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Taxon-toxicity study of fish to typical transition metals: Most sensitive species are edible fish ☆

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ABSTRACT

Transition metals are widespread in aquatic environments and can be harmful when concentrations exceed thresholds. Especially for fish, an important component of the human diet, low concentrations of transition metals will directly affect their well-being. Different taxa are protected by unified water quality criteria (WQC) thresholds, which rarely consider the ecological status and economic value of different species. There is therefore an urgent need to study taxon-specific sensitivity. The present study established the species sensitivity distributions of nine typical transition metals (Cr, Mn, Fe, Ni, Cu, Zn, Ag, Cd and Hg) for protecting freshwater and seawater fish based on non-parametric kernel density estimation methods, and then derived their acute and chronic HC5-values. The results showed that Ag and Hg have the highest acute toxic potency to fish in freshwater as well as seawater. Compared with marine fish, freshwater fish were more tolerant to acute exposure to Cr, Fe, Ni and Zn, whilst being more sensitive to Ag and Cd. Moreover, edible fish are more sensitive than other fish to these metals in both freshwater and seawater, encouraging more protection of economically valuable fish that may potentially affect human health. The study provides a strong reference for future research on taxon-specific WQC for transition metals.

1. Introduction

Transition metals are widespread in aquatic environments and most of them are micro-nutrients which play a key role in the cell metabolism of living organisms (Festa and Thiele 2011). Transition metals can seriously affect the species diversity and distribution of aquatic organisms when concentrations exceed threshold values (Friberg et al., 1979). The aquatic environment is in general seriously polluted by transition metals such as Cr, Ni, Cu, Zn, Cd and Hg, which originate from industrial discharge and mining (Wang et al., 2011). It has been reported that the ecological risks of transition metals are generally higher in China than in other countries (Fu et al., 2016). In addition, the mining amounts of Cd, Pb, Cu, Hg and Zn in China have doubled in the recent decades (U.S.DOI and U.S.GS, 2008, 2020), a trend which is expected to continue and which will lead to higher risks in the future. Moreover, transition metals are difficult to remove by self-purification of water, and their bioaccumulation in aquatic organisms will subsequently cause serious harm to human health (Barron 2003). Therefore, it is urgent and essential to study the toxicology of transition metals to aquatic organisms and to derive water quality criteria (WQC) for transition metals.

WQC is the maximum acceptable threshold value for chemical substances or environmental parameters for protecting specific functions of water bodies and organisms from adverse effects under certain environmental conditions (U.S.EPA, 1985). At present, unified WQCs of transition metals are derived with the aim of protecting most species potentially present in the aquatic environment. However, the sensitivity of different taxa to different transition metals differs and while it is impossible to protect all

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species possibly present in an ecosystem, it is well possible that sometimes WQC are non-protective for specific taxa or even groups of taxa. For example, the acceptable hazardous concentration of Ag for protection of 95% of all fish species (HC5) (2.82 μg/L) was about 20% lower than the HC5 for all organisms (3.47 μg/L) (Wang et al., 2017b), while the HC5 for Hg of all organisms (2.14 μg/L) was 4.5 times higher than the HC5 of crustaceans (0.38 μg/L) (Wang et al., 2015b). With the aim of protecting specific taxa or even specific individual species it is very important to compare taxon-specific sensitivity with the WQC of transition metals to make sure that sensitive taxa and biodiversity are sufficiently protected by HC5-values derived for transition metals.

The assessment of the toxicity of transition metals to aquatic organisms commonly focuses on fish, crustaceans and amphibians (Bernhoft 2012; Flemming and Trevors 1989). Among these species, experimental research on fish toxicity has been carried out in depth. Low concentrations of transition metals were found to interfere with the endocrine system, the reproductive system, and the immune system of fish, to reduce the hatching rate of juvenile fish, to increase the deformity rate, to cause cancer, teratogenicity and mutations, and to even directly affect the normal reproduction and survival of fish (Bakshi and Panigrahi 2018; Sfakianakis et al., 2015), which will seriously endanger the ecological balance. In addition, the ecological and economic value of many kinds of fish is high. Fish represent for instance an important component of the human diet with the global consumption of *Oncorhynchus mykiss* reaching 950,000 tons in 2019 (FAO 2020). A comparative study on the sensitivity of fish species to different transition metals assists in the improved protection of the ecologically and economically valuable fish, and can be used as an example for assessment of taxon-specific sensitivity towards transition metals and/or other pollutants.

The approach of species sensitivity distribution (SSD) is widely used to derive the HC5 and to assess environmental risks in ecosystems (Wu et al., 2012). We previously reported that compared with parametric density estimations, non-parametric kernel density estimation (NPKDE) is capable of deriving better fitting SSDs with more robustness and more accurate prediction of adverse effects (Wang et al., 2015b). More importantly, the NPKDE method yields the true cumulative density function (CDF) of all species for some metals. This solves the problem of lack of a single, universally applicable parametric distribution to be appropriate for all toxicity data of pollutants (Wang et al., 2017a).

Hence, in the present study we investigated the differences between acute and chronic taxon-specific sensitivity of fish to nine transition metals (Cr, Mn, Fe, Ni, Cu, Zn, Ag, Cd and Hg) by using the SSD approach based on the NPKDE. Moreover, we compared and analyzed the HC5-values of freshwater and seawater fish, and we finally discussed the most suited indicator species for taxon-specific sensitivity of fish.

2. Materials and methods

2.1. Toxicity data sets

The present study selected all historical studies up to 31st, December 2019 that have reported acute and chronic toxicity data of nine metals (Cr, Mn, Fe, Ni, Cu, Zn, Ag, Cd and Hg) for freshwater and marine fish species from the ECOTOX database developed by the United States Environmental Protection Agency (USEPA) (http://cfpub.epa.gov/ecotox/) and China National Knowledge Infrastructure database (CNKI) (http://www/cnki/net/). Accuracy and reliability of data were evaluated by use of standard methods (Klimisch et al., 1997). The screening standards of toxicity data used in the study were based on the strict rules as detailed in *Text S1*. The lethal concentration affecting 50% of individuals (LC50) or the effective concentration affecting 50% of individuals (EC50) were selected as the acute toxicity endpoint (Wang et al., 2017a), while the no observed effect concentration (NOEC) or the lowest observed effect concentration (LOEC) were selected as the chronic toxicity endpoint (Monti et al., 2018). The species mean acute/chronic value (SMAV/SMCV) was calculated by use of geometric means when more than one toxicity data for the same species were available (Stephen et al., 1985). Matlab 2019b software (Mathworks, Natick, MA, USA) was used to carry out the SSD modelling and to perform goodness-of-fit testing. Chronic HC5-values for marine fish were not derived, since there was insufficient information on chronic effects on sub-lethal endpoints to construct chronic SSDs for seawater fish.

2.2. Hardness correction

The toxic potency of heavy metals to fish decreases with increasing pH, water hardness or DOC concentration (Cusimano et al., 1986; Liao et al., 2019). WQC documents issued by USEPA and Ministry of Ecology and Environment of China clearly require hardness correction for acute and chronic toxicity data of freshwater aquatic organisms (State Key Laboratory of Environmental Criteria and Risk Assessment, 2017; U.S.EPA, 1985). This study used the hardness correction method reported in Chinese WQC documents (State Key Laboratory of Environmental Criteria and Risk Assessment, 2017). The hardness of acute and chronic toxicity data was corrected to 150 mg/L $CaCO_3$ based on the range of hardness values (Tables S1−S2).

2.3. Modeling of NPKDE-SSDs and HC5 derivation

Statistical analysis and SSDs were constructed based on our previous study (Wang et al., 2015a), in which NPKED-SSDs were developed based on Gauss kernel function estimation. We have proven that a NPKDE-SSD better reflects the overall trend of changes in species toxicity data than parametric models, whereas a NPKDE-SSD also better reflects the internal structural characteristics of toxicity data. A NPKDE-SSD also yields better fits, more robustness and better predictions than parametric models for sensitive species. This makes the calculated HC5 more reliable (Wang et al., 2015a; 2017a). Similar to the use of HC5-values as the basis for derivation of WQC for all species (U.S.EPA, 2005; Van Straalen and Denneman 1989), we used the underlying concept to calculate the concentration at which 95% of all fish species is protected, which is also the basis of taxon-specific WQC. The Kolmogorov-Smirnov (K−S) test, root mean square errors (RMSE) and error sum of squares (SSE) were utilized to examine the goodness-of-fit for establishing SSDs, for which the model is considered sufficient when the P_{ks} value greater than 0.05, and RMSE and SSE are relatively small.

3. Results and discussion

3.1. Developing SSDs based on NPKED

In freshwater, there was a total of 328 species for which a total of 3296 acute toxicity data were generated for Fe (7), Mn (8), Ag (13), Hg (22), Ni (27), Cr (35), Cd (46), Cu (103) and Zn (67) (Table S1). A total of 249 chronic toxicity data were retrieved for 29 freshwater species for Zn (8), Cd (10) and Cu (11) (Table S2). In seawater, there was a total of 243 species with 1232 acute toxicity data for Fe (6), Ni (11), Ag (15), Cr (23), Hg (28), Zn (43), Cd (50) and Cu (65) (Table S3). The reported concentration ranges across the various studies are relatively large, and the number and range of acute toxicity data for freshwater and seawater fish were much larger than those of the chronic toxicity data. The toxicity studies of transition metals on freshwater fish focused on Cu, Zn, Ag and Cd, accounting for more than 85% of all freshwater toxicity data, while the studies on marine fish focused on Cu, Zn, Cd, and Hg, accounting for nearly 80% of all seawater toxicity data. The underlying reason for this observation might be that Cu, Zn, Ag, Cd and Hg are IB and IIB group elements, which more easily form stable complexes with biological macromolecules in water than the other transition elements.

After logarithmic transformation and hardness correction (Table S4), the acute and chronic NPKDE-SSDs of transition metals to freshwater and seawater fish were established (Table 1). The P_{ks} values of the K−S test of the NPKED-SSD models were all greater than 0.05 and values of RMSE and SSE were small. This shows that the NPKED-SSD models for these metals developed here are all good-fitting, highly accurate and suited for use in hazard simulation.

3.2. Derivation of acute and chronic HC5-values for freshwater and seawater fish

The sequence order of acute HC5-values for nine transition metals to freshwater fish was Mn > Cr > Fe > Ni > Zn > Hg > Cu > Cd > Ag (Fig. 1A and B). Obviously, Ag has the strongest acute toxic potency to freshwater fish among these nine transition metals. The toxicity of Ag to fish in water mainly originates from the free silver ions, which can precipitate or solidify on the gill surface of fish to prevent the exchange of oxygen and carbon dioxide, which quickly turns the fish asphyxiated (Ma et al., 2015). On the other hand, the toxic potencies of Fe and Mn to freshwater fish were low. These elements have similar chemical properties and can be metallic co-factors of many enzymes, which could improve the efficiency of enzymatic reactions and have important effects on for instance their role as electron carrier in redox reactions and in the promotion of protein enzyme synthesis in organisms (Crossgrove and Zheng 2004; Czernel et al., 2016). The acute HC5-values collected differed significantly from the results on our research on all taxa (Wang et al., 2015a; 2017a) as the previous studies showed that Cd, Hg and Cr (VI) were more toxic than Ag (the acute HC5 to all taxa of Cd, Hg, Cr (VI) and Ag is 3.39, 2.14, 0.07, and 3.47 μg/L, respectively). In addition to the influence of sample size and hardness correction, the mechanisms of toxicity of different organisms differ a lot, which suggests that there is an urgent need to study taxon-dependent sensitivity. The sequence of the chronic HC5-values for Zn, Cu and Cd to freshwater fish was the same as the sequence of the acute HC5-values (Fig. 1C and D).

Unlike freshwater fish, Hg has the strongest acute toxic potency to seawater fish among eight transition metals (Fig. 1E and F). Hence, Hg is more harmful in seawater. The findings by Denton et al.(Denton and Burdon-jones 1986) showed that the ranking of metal toxicity to juvenile glass perch (marine fish) was Hg > Cu > Cd = Zn > Ni > Pb is consistent with our results. However, marine fish is not properly protected at present, as is for instance clear for Zn. Zn can accumulate in the viscera of fish and will damage the visceral tissues by changing the structure of non-enzymatic organic molecular ligands when the concentration exceeds chronic thresholds (Vardy et al., 2011). The acute HC5-value derived for all aquatic organisms in seawater (Wheeler et al., 2002) and the marine WQC recommended by USEPA (U.S.EPA, 2017) are 435 μg/L and 90 μg/L, respectively, which are much higher than the HC5 value of 40 μg/L derived for fish in the present study.

3.3. Determination of sensitive species

A search of the original data on acute toxicity showed that the sensitive species near the 5% cumulative probability for transition metals to freshwater and seawater are mainly edible fish. In freshwater, the species most sensitive to acute exposure to Mn, Fe, Ag and Cu are *Ptychocheilus oregonensis*, *Pimephales promelas*, and *Cyprinus carpio*、*Pseudorasbora parva*, respectively, all of which belong to the family of cyprinids (cypriniformes, subclass Actinopterygii) which is the largest family in the subclass actinopterygii, containing many economically important and edible fishes such as four major Chinese carps (black carp, grass carp, silver carp and bighead carp). In addition, the most sensitive species to short term exposure to Cr, Ni, Cu and Ag are the freshwater fishes *Salmo salar*, *Ambloplites rupestris*, *Pseudorasbora parva* and *Cyprinus carpio*, whilst the most sensitive species in case of exposure of marine fish to Cu, Ag and Hg are *Apocryptes bato*, *Paralichthys dentatus* and *Pagrus major*. These species are all edible fish. Actually, there is a new record of an estimated annual supply of 20.5 kg per capita for human fish consumption in 2018, in which more than 30% of consumed freshwater fish is filter-feeding inland-water finfish (mostly silver carp and bighead carp) (FAO 2020). Therefore, it is necessary to pay attention to preventing the bioaccumulation and harm to human health when metal assimilates in biota and tends to biomagnify in the food chain.

The freshwater fish species that are most sensitive to chronic exposure to Cu, Zn and Cd are *Pimephales promelas*, *Jordanella floridae* and *Salmo trutta*. *Pimephales promelas* and *Jordanella floridae* are standard model organisms recommended by USEPA for chronic toxicity testing of freshwater fish (Call et al., 1983; Norberg and Mount 1985), whereas *Salmo trutta* is an edible fish that is produced in high quantities (FAO 2020). These three sensitive species are widely distributed internationally, while the ecological risks for these fish species are relatively high since the concentrations of three heavy metals in many rivers exceed the thresholds (He et al., 2019).

Notably, *Oryzias melastigma*, a new marine model organism, is a common sensitive species of seawater fish to Cr and Zn exposure. In addition, it was found that the LC50 values of Hg and Cd to *Oryzias melastigma* were around HC10 and HC20, which means that *Oryzias melastigma* could be selected as an acute sensitive indicator species for heavy metals to seawater fish in the future.

3.4. Comparison of HC5-values

Comparing the obtained HC5-values (Table 1), it was found that freshwater fish are much more tolerant to Cr, Fe, Zn and Hg than marine fish. The acute HC5-values of Fe, Zn, Hg and Cr for freshwater fish were respectively more than 21, 12, 4 and 3 times the corresponding values for seawater fish. The HC5-values of Cu and Ni to freshwater and seawater fish are similar. Finally, the acute HC5-values of Ag and Cd for seawater are more than 7 and 28 times the values for freshwater fish, which illustrates that these two metals are much more toxic to seawater fish. The differences in sensitivity to these transition metals between freshwater and marine fish may be caused by following three factors: (1) the free ions of transition metals in freshwater are more abundant and have different free forms, which may be helpful to enhance the tolerance of freshwater organisms to heavy metals (Phillips and Rainbow 1993); (2) the kinds of freshwater and marine fish species differ a lot, which might modify the absorption of transition metals in fish; (3) the different environmental conditions between freshwater and seawater, such as salinity, hardness, temperature, pH, etc., can lead to differences of the toxicity values (Wang et al., 2014). Moreover, it was found that Cd is highly toxic to both freshwater and seawater

Table 1
Overview of the acute and chronic NPKDE-SSDs and current WQC recommended by USEPA of transition metals for freshwater and seawater fish. The overview includes the parameters describing the distribution of the data and the results of the goodness-of-fit evaluation.

	Metals	Equations	Bandwidth	P_{K-S}	RMSE	SSE	HC5 (μg/L)	Current WQC (μg/L, USEPA)
Freshwater-Acute	Mn	$f(x) = \frac{1}{3.01}\sum_{i=1}^{8}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.376})^2}{2}}$	0.376	0.71	0.064	0.033	24682	/
	Cr	$f(x) = \frac{1}{8.30}\sum_{i=1}^{35}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.237})^2}{2}}$	0.237	0.71	0.038	0.051	12216	570 (Cr(III)) 16(Cr(V))
	Fe	$f(x) = \frac{1}{2.34}\sum_{i=1}^{7}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.334})^2}{2}}$	0.334	0.94	0.063	0.028	8054	/
	Ni	$f(x) = \frac{1}{8.51}\sum_{i=1}^{27}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.315})^2}{2}}$	0.315	0.95	0.029	0.022	5225	470
	Zn	$f(x) = \frac{1}{23.18}\sum_{i=1}^{67}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.346})^2}{2}}$	0.346	0.82	0.029	0.057	504	120
	Hg	$f(x) = \frac{1}{4.25}\sum_{i=1}^{22}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.193})^2}{2}}$	0.193	0.77	0.036	0.029	76.87	1.4
	Cu	$f(x) = \frac{1}{26.27}\sum_{i=1}^{103}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.255})^2}{2}}$	0.255	0.76	0.018	0.035	47.52	using the Biotic Ligand Model
	Cd	$f(x) = \frac{1}{19.6}\sum_{i=1}^{46}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.426})^2}{2}}$	0.426	0.88	0.029	0.038	15.09	1.8
	Ag	$f(x) = \frac{1}{2.42}\sum_{i=1}^{13}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.186})^2}{2}}$	0.186	0.87	0.049	0.032	4.95	3.2
Freshwater-Chronic	Cd	$f(x) = \frac{1}{2.03}\sum_{i=1}^{8}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.254})^2}{2}}$	0.254	0.80	0.066	0.044	1.03	0.72
	Cu	$f(x) = \frac{1}{1.58}\sum_{i=1}^{10}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.158})^2}{2}}$	0.158	0.93	0.038	0.015	1.33	using the Biotic Ligand Model
	Zn	$f(x) = \frac{1}{3.71}\sum_{i=1}^{11}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.337})^2}{2}}$	0.337	0.81	0.047	0.017	34.09	120
Seawater-Acute	Ni	$f(x) = \frac{1}{2.94}\sum_{i=1}^{11}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.267})^2}{2}}$	0.267	0.98	0.044	0.021	4356	74
	Cr	$f(x) = \frac{1}{5.45}\sum_{i=1}^{23}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.237})^2}{2}}$	0.237	0.89	0.046	0.049	3535	1100(Cr(V))
	Cd	$f(x) = \frac{1}{13.45}\sum_{i=1}^{50}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.269})^2}{2}}$	0.269	0.94	0.024	0.028	433	33
	Fe	$f(x) = \frac{1}{3.81}\sum_{i=1}^{6}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.635})^2}{2}}$	0.635	0.93	0.037	0.0083	374	/
	Cu	$f(x) = \frac{1}{20.74}\sum_{i=1}^{65}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.319})^2}{2}}$	0.319	0.95	0.02	0.027	41.82	4.8
	Zn	$f(x) = \frac{1}{16.34}\sum_{i=1}^{43}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.380})^2}{2}}$	0.380	0.92	0.026	0.03	39.66	90
	Ag	$f(x) = \frac{1}{2.94}\sum_{i=1}^{15}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.196})^2}{2}}$	0.196	0.99	0.029	0.012	34.68	1.9
	Hg	$f(x) = \frac{1}{12.24}\sum_{i=1}^{28}\frac{1}{\sqrt{2\pi}}e^{\frac{-(\frac{x-x_i}{0.437})^2}{2}}$	0.437	0.86	0.044	0.055	18.04	1.8

Note.
Current WQC USEPA recommended are collected from USEPA website (https://www.epa.gov/wqc).

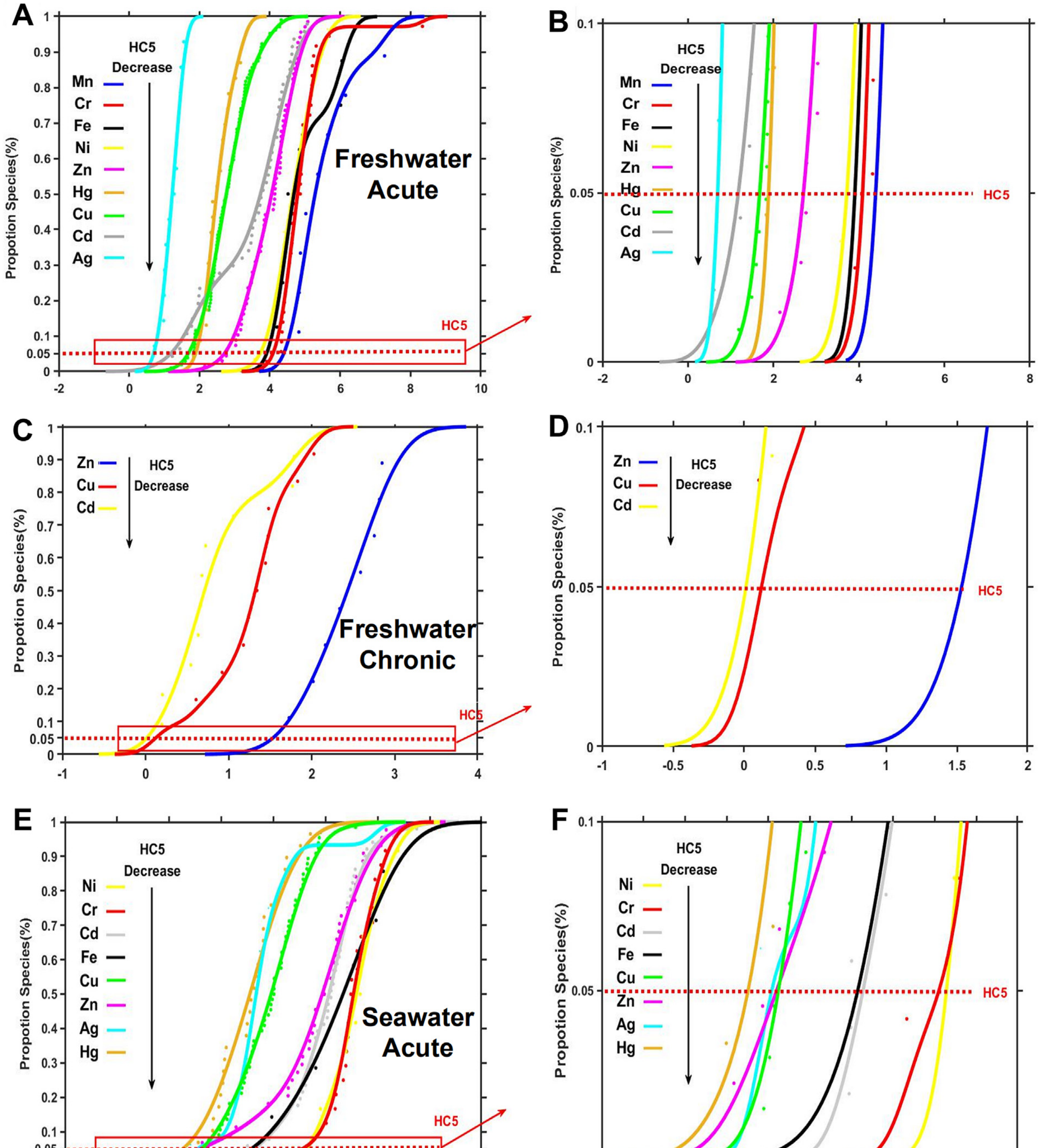

Fig. 1. NPKED-SSD models of typical transition metals to freshwater and seawater fish. (A) Acute SSDs to freshwater fish for Mn (●), Cr (●), Fe (●), Ni(●), Zn (●), Hg (●), Cu(●), Cd (●) and Ag (●). (B) HC5 values derived based on SSDs for 5% of the species. (C) Chronic SSDs to freshwater fish for Zn (●), Cu(●) and Cd (●). (D) HC5 values derived based on SSDs for 5% of proportion species. (E) Acute SSDs for seawater fish for Ni(●), Cr (●), Cd (●), Fe (●), Cu(●), Zn (●), Ag (●) and Hg (●). (F) HC5 values derived based on SSDs for 5% of proportion species.

fish, in which the acute-WQC, chronic-WQC and acute seawater WQC recommended by USEPA are 1.8, 0.72 and 33 μg/L, respectively (U.S.EPA, 2017). One reason for this difference in sensitivity might be that upon exposure to high concentrations of Cd^{2+}, the normal physiological, biochemical and metabolic processes of both freshwater and seawater fish will be affected, including e.g. rapid reduction of mitochondrial activity of the gills, and reduction of the self-regulation ability of the osmotic pressure in fish (Torre et al., 2000).

Most of derived acute HC5-values of both freshwater and seawater fish in the present study appear to an order of magnitude or higher than current WQC values recommended by USEPA (Table 1), which indicates that although our results considered the hardness correction, both freshwater and seawater fish can be protected by use of the current WQC values USEPA recommended. However, the derived chronic HC5-value of freshwater fish and acute HC5-value of seawater fish to Zn are both lower than current WQC values recommended by USEPA (Table 1), which suggests that both freshwater and seawater fish are not well protected for Zn. It may be caused by two reasons: (1) our results obtained are performed at the condition of corrected hardness (150 mg/L $CaCO_3$), which the hardness level of transition metals is not specified or corresponds to a hardness of 100 mg/L $CaCO_3$ (only Cr(III), Cd and Ni) in the WQC USEPA recommended; (2) the HC5-values derived in the present study also used representative fish in aquatic ecosystems of China, such as four major Chinese carps, of which sensitivity are different from fishes to be protected in North America such as cold-water fishes of the family *Salmonidae*. In addition, at the hardness of 150 mg/L $CaCO_3$, the acute and chronic HC5 of Cd to freshwater fish (15.09 and 1.03 μg/L) is also higher than acute and chronic WQC value of Cd recommended by Ministry of Ecology and Environment of China (12.94 and 0.5836 μg/L) (Ministry of Ecology and Environment of China, 2020), respectively, for which edible fish including *Salmo trutta* could be protected better.

3.5. Impotency and limitations

This study is the first that deals with taxon-specific sensitivity of freshwater and marine fish. The HC5-values generated in the present study could be useful for judging the toxic potency of transition metals to fish. More importantly, the present results demonstrated that edible fish are commonly the most sensitive fish species, which suggests that it is urgently needed to establish taxon-specific thresholds in order to specifically protect economically important fish species. Meanwhile, different toxic potency of transition metals to freshwater and marine fish suggested that site-specific WQC is indeed needed for future risk assessment due to the difference of water environments and species in different areas.

As a follow-up on this study, it is needed to focus on four areas of research. First, we need more toxicity data for seawater fish since additional data will make the derived HC5-values for marine fish more accurate. Second, it is needed to check for outliers in small sample sizes as such outliers will induce fitting errors when using the NPKED-SSD models. Third, dividing the different valence of metals, such as Cr(III) and Cr(VI), should be considered in SSD modeling in order to improve the accuracy of model prediction when data is enough. Finally, underlying data collection and screening should be strict in many areas, such as tested species, number of species, and the conditions of the tests, since it is critical to the accuracy and consistency of the model. Nevertheless, the present study points out the urgency and necessity of the protection of edible fish, and provides reference for the future protection of fish from pollution by transition metals.

4. Conclusion

The present study used NPKDE-SSD approach to investigate the acute and chronic taxon-specific sensitivity of freshwater and marine fish for nine transition metals (Cr, Mn, Fe, Ni, Cu, Zn, Ag, Cd and Hg). The results showed that the sequence order of acute HC5-values for nine transition metals to freshwater fish was Mn > Cr > Fe > Ni > Zn > Hg > Cu > Cd > Ag, and to marine fish was Ni > Cr > Cd > Fe > Cu > Zn > Ag > Hg. The sequence of chronic HC5-values for Zn, Cu and Cd to freshwater fish was the same as that of the acute HC5-values. In addition, over 50% sensitive species to these metals in both freshwater and seawater are edible fish, encouraging more protection of economically valuable fish that may potentially affect human health. Moreover, both freshwater and seawater fish are not well protected for Zn under current WQC guideline recommended by USEPA. The study provides a strong reference for future research on taxon-specific WQC for transition metals.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117154.

Credit author statement

Ying Wang: Conceptualization, Writing – original draft, Methodology. Linhui Cui: Data curation, Methodology, Formal analysis. Chenglian Feng: Visualization, Investigation., Zhaomin Dong: Software, Validation. Wenhong Fan: Supervision, Writing-Reviewing and Editing. Willie J. G. M. Peijnenburg: Writing-Reviewing and Editing.

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