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RESEARCH ARTICLE

Tissue-based assessment of hazard posed by mercury and selenium to wild fishes in two shallow Chinese lakes

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Abstract
Total (all forms of inorganic and organic) concentrations of mercury (Hg) and selenium (Se) were measured in dorsal muscle and eggs of wild fishes from two shallow lakes in China: Tai Lake (Ch: *Taihu*; TL) and Baiyangdian Lake (BYDL). Hazard quotients (HQs) were calculated by dividing concentrations of Se or Hg in muscle or eggs of fishes by threshold concentrations for effects expressed as tissue residue toxicity reference values (TR-TRVs). Concentrations of Hg in whole bodies of fishes were estimated by concentrations in muscle. Based on concentrations of Hg in whole body, HQs for fishes in TL and BYDL were less than 1.0, which suggests little to moderate potential for effects on these fishes and unaccepted adverse effects of Hg are unexpected for adult fishes. HQs of Se in muscle of common carp from TL were closed to 1.0, and 27% of HQs based on concentrations of Hg in eggs of fishes from BYDL exceeded 1.0. Potential hazard due to Hg on common carp in TL and reproductive effects of Se on fishes from BYDL exhibited need for concern. Ratios of molar concentrations of Se to Hg were greater than 1.0. Thus, there might be some protective effects of Se on effects of Hg on fishes in TL and BYDL.

Keywords Tai Lake · Baiyangdian Lake · Asia · Tissue residue approach · Hazard assessment · Hg · Se · Antagonism

Introduction

Contamination of the environment by mercury (Hg) has drawn extensive, global attention because of long-range atmospheric transport, bioaccumulation through the food chain and toxic potency of Hg (Wang et al. 2012a, b; Zhang et al. 2013). Mercury can be released from natural sources and/or activities of humans and can enter aquatic ecosystems, where it can be accumulated by organisms to concentrations sufficient to cause adverse effects on those organisms or predators, including humans that eat them. Because of potential for biomagnification of Hg in food webs, most of the concern has been for potential of adverse effects on humans that eat fish and other piscivorous wildlife (Duvall and Barron 2000; Moore et al. 1999; Rumbold et al. 2008; Rumbold 2005; Weech et al. 2006; Zhang et al. 2013). However, results of laboratory and eco-epidemiological studies have indicated that environmentally relevant concentrations of Hg in fishes have potential to affect survival, growth, and reproduction (Scheuhammer et al. 2007). For example, significant negative correlations between concentrations of androgens in blood plasma and concentrations of Hg in tissues were observed in white sturgeon (*Acipenser transmontanus*) from the lower Columbia River (Webb et al. 2006). Methylmercury in the diet of adult fathead minnows (*Pimephales promelas*) during oogenesis is the primary vector of exposure of embryos (Hammerschmidt and Sandheinrich 2005). Moreover, total (all forms of inorganic and organic) concentrations of mercury (0.07 μg/g to 0.10 μg/g wet mass, wm) in eggs of rainbow

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trout (*Oncorhynchus mykiss*) were directly proportional to mortality of embryos (Birge et al. 1979). Concentrations of Hg in some wild fishes have exceeded reported thresholds for toxicity. Thus, it was deemed appropriate to investigate hazards posed by Hg to wild populations of fishes.

Selenium (Se) is unique in that it is a required element so that fish can be deficient, but greater concentrations can be toxic. Thus, there are upper and lower limits of optimal concentrations of Se in both tissues and diets. For fishes, dietary concentrations of 0.1–0.5 μg/g dry mass (dm) are optimal; however, concentrations 7- to 30-fold greater than optimal nutritional concentrations can be hazardous to fishes and other wildlife (Hodson and Hilton 1983; Lemly 1993a; Muscatello et al. 2006). Selenium can be bioaccumulated through aquatic food webs and adversely affect reproduction and have teratogenic effects (Holm et al. 2005; Lemly 1993b; Muscatello et al. 2006; Rudolph et al. 2008). Results of previous studies have shown that Se can be protective against bioaccumulation and toxicity of Hg (Belzile et al. 2006; Dang and Wang 2011; Sørmo et al. 2011). Thus, interactions between Se and Hg in tissues should be considered in assessments of hazards of Hg to aquatic organisms.

The tissue residue approach is useful to investigate adverse effects of contaminants on organisms because it obviates the need to predict the critical body burden accumulated from surrounding media or diet and is recommended to be used in deriving environmental quality criteria and assessing hazards of bioaccumulative chemicals (Beckvar et al. 2005; Meador 2015; Sappington et al. 2011). Because Hg and Se are both considered to be bioaccumulative and dietary intake is the main exposure pathway for fishes and other higher trophic level organisms, tissue residue concentrations in fishes can be used to assess hazards of internal doses to organisms (Jarvinen and Ankley 1999).

Tai Lake (TL) is the second largest freshwater lake in China (Guo et al. 2012), and Baiyangdian Lake (BYDL) is the largest freshwater body on the North Plain of China. Both areas receive contaminants from atmospheric deposition, and sewage or industrial effluents and agricultural runoff, thus they are affected by pollution by various metals (Chen et al. 2008; Shen et al. 2005; Wenchuan et al. 2001; Yang et al. 1996; Zhao et al. 2012). Concentrations of Hg in aquatic organisms from these two lakes have been previously reported (Chen et al. 2008; Wang et al. 2012c), and concentrations of Hg in TL are greater than what is considered to be background from natural sources (Hu et al. 2014). Concentrations of Hg in fishes from BYDL exceeded critical thresholds and were considered to be potentially hazardous to local humans and wildlife (Chen et al. 2008). Concentrations of Se are only available for icefish (*Protosalanx hyalocranius*) from TL (Yang et al. 2009). Hazards of Hg and Se in tissues of fishes from these two lakes were still unclear. To address these concerns, concentrations of Hg and Se in fishes were monitored and tissue-based hazard assessments were performed for Hg and Se including consideration of the antagonistic protective effects of Se on toxic potency of Hg.

Materials and methods

Collection of fishes

Fish were captured from various locations in TL and BYDL (Fig. 1), by local commercial fishermen with the use of various types of nets. Samples of fish muscle ($n = 46$) were collected from five fishes, including catfish (*Silurus asotus*), common carp (*Cyprinus carpio*), bighead carp (*Aristichthys nobilis*), spotted steed (*Hemibarbus maculatus*), and topmouth culter (*Erythroculter ilishaeformis*). Individuals were collected from the Meiliang Bay, TL in 2011. Samples of muscle ($n = 41$) from six fishes, including bighead carp (*Aristichthys nobilis*), common carp (*Cyprinus carpio*), Chinese perch (*Siniperca chuatsi*), topmouth culter (*Erythroculter ilishaeformis*), catfish (*Silurus asotus*), and crucian carp (*Carassius cuvieri*), were collected from East TL. Samples of whole body of weather loach (*Misgurnus anguillicaudatus*) and fish muscle ($n = 63$) from four species including catfish (*Silurus asotus*), silver carp (*Hypophthalmichthys molitrix*), yellow catfish (*Pelteobagrus fulvidraco*), and crucian carp (*Carassius cuvieri*) were collected from BYDL in 2012, and samples of eggs ($n = 53$) were stripped from the same female fishes, except oriental weather fish (*Misgurnus anguillicaudatus*). Before collecting tissues, lengths of fish were measured. Upon collection, samples were stored in polyethylene bags, kept on ice, and transported immediately to the laboratory where they were stored at − 20 °C. Frozen fish samples were thawed and rinsed individually with deionized water to remove possible metal contaminants. Samples were freeze dried for 48 h, and then ground into powders and stored at − 5 °C until analysis.

Quantifications of total concentrations of Hg and Se

Total concentrations of Hg and Se were measured. Samples were digested by use of microwave-accelerated digestion followed by hydride generation-atomic fluorescence spectrometry, following described methods for Hg (Fu et al. 2010) and Se (MOH 2010), respectively. Due to insufficient masses of tissues, only 14 samples of crucian carp from BYDL were measured for Se. Precision and accuracy of analytical methods were determined and monitored using method blanks, blank spikes, blind duplicates, and certified reference materials (CRMs) including TORT-2 (lobster hepatopancreas) from the National Research Council, Canada, GBW10050 (GSB-28) (giant river prawn) and GBW10020 (citrus leaf). Relative percentage differences for duplicate samples were

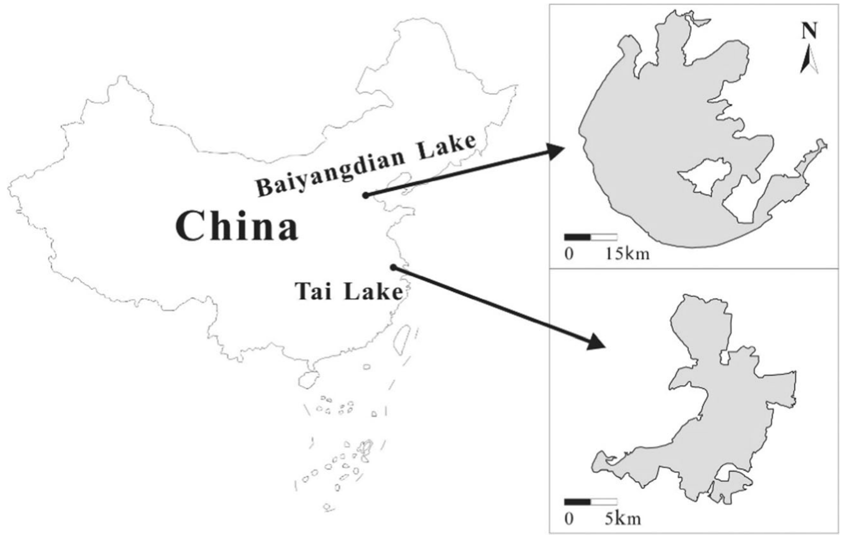

Fig. 1 Map of the study areas, including Tai Lake and Baiyangdian Lake

less than 9%. Recoveries (measured value/certified value × 100%) for total Hg and Se in CRMs were in the range of 90–112% and 92–107%, respectively.

Because toxicity reference values (TRVs) for mercury used in the present study were expressed as whole-body concentrations, concentrations of Hg in whole bodies were predicted from those in muscle. The predictive relationship was derived based on analyses of 210 fish of various sizes, representing 13 species (Peterson et al. 2005) (Eq. 1).

$$C_{\text{whole-body}} = 10^{(-0.2712+0.9005\times 1\ \text{g}[\text{Hg}_{\text{muscle}}])} \quad (1)$$

Hazard analysis

The hazard quotient (HQ) method was used to conduct a screening-level assessment of hazards of Hg and Se in tissues of fishes inhabiting TL and BYDL. The method used is consistent with methods for assessments of hazards to aquatic organisms (USEPA 1998). The HQ was calculated for each species (Eq. 2).

$$\text{HQ} = \text{TRC}/\text{TRV} \quad (2)$$

where TRC is the concentration of Hg or Se in whole fish or eggs, and the TRV is the toxicity reference value used as the hazard-screening benchmark. If values of HQ exceed 1.0, the endpoint used for assessment would be expected to be observed. Values of $0.1 \leq \text{HQ} < 1$ indicate a moderate hazard, and values of $0.01 \leq \text{HQ} < 0.1$ indicate little hazard. Values of $\text{HQ} < 0.01$ indicate negligible or *de minimis* hazard (Sánchez-Bayo et al. 2002).

To assess hazards of Hg to fishes, HQs were calculated by dividing concentrations of Hg in whole fish, by a TRV of 0.2 μg/g wm in whole bodies of fishes (Beckvar et al. 2005). To assess hazards of Hg in eggs of fishes, a TRV of 0.02 μg/g wm in eggs of fishes was used, which is calculated using a ranking approach (Beckvar et al. 2005). The TRV based on concentrations of Hg in whole bodies of fishes was converted to dry mass by assuming 80% moisture, then expressing values as 1.0 μg/g dm. A mean of 63% moisture was calculated for eggs based on data in the present study so the threshold effect concentration of 0.02 μg/g wm in fish egg was converted to 0.054 μg/g dm.

Concentrations of 11.3 μg/g dm in fish muscle and 15.1 μg/g dm in fish egg were used as the TRVs of Se, and these TRVs are available in US Environmental Protection Agency's criteria document (USEPA 2016). The TRVs were calculated based on reproductive chronic effects of Se in fishes. Once concentrations of Se exceed these values in muscle or egg, it is assumed and thus predicted that adverse effects will occur.

Statistical analyses

All data of concentrations were tested for normality by use of the Shapiro-Wilk test. The independent sample *t* test or Kolmogorov–Smirnov test were performed to test differences between concentrations Hg or Se in fishes from the Meiliang Bay and East TL or TL and BYDL. Statistical comparisons among concentrations of Hg or Se in different feeding guilds were performed using the ANOVA. Nonparametric Kruskal–Wallis test were used if model assumptions were not met. Statistical significance was accepted at $p < 0.05$ for all tests. All statistical tests were performed using IBM SPSS Statistics 19 (IBM Corporation, USA). All figures were created using Sigmaplot 12.5 (Systat Software, Inc., USA).

Results and discussion

Accumulation of Hg and Se in fishes

Because no significant differences were observed in total concentrations of Hg or Se between the Meiliang Bay and East TL ($p = 0.577$ for Hg, and $p = 0.066$ for Se), data from these two regions were combined in the analyses below. Characteristics of samples of fishes and concentrations of Hg and Se are listed in Table 1. Mean concentrations of Hg in muscle of fishes from TL and BYDL were 0.12 (0.00019–1.4) and 0.22 (0.034–0.60) μg/g dm, respectively. Concentrations of Hg in muscle of fishes from TL observed in the present study were comparable to those reported previously (Wang et al. 2012c), while concentrations of Hg in fishes from BYDL were less than those reported previously (Chen et al. 2008), but significantly greater than those from TL ($p < 0.05$).

Total concentrations of Hg in fishes of various feeding guilds were calculated (Fig. 2). There were significant differences between the concentrations of Hg in the carnivorous fishes and filter feeders or omnivorous fishes from TL ($p < 0.05$), while no difference was observed between filter feeders and omnivorous fishes ($p = 0.163$). Significant differences were also observed between concentrations of Hg in the carnivorous fishes and filter feeders or omnivorous fishes in BYDL ($p < 0.05$), but no differences were observed between filter feeders and omnivorous fishes ($p = 0.565$).

Mean concentrations of Se in muscle of fishes from TL and BYDL were 3.3 (range 0.19–12) and 1.6 (range 0.96–3.4) μg/g dm, respectively. Concentrations of Se in muscle of fishes from TL were significantly greater than those in BYDL ($p < 0.05$). There were significant differences between total concentrations of Se in carnivorous fishes or filter feeders and omnivorous fishes from TL (Fig. 3, $p < 0.05$), and no difference were observed between carnivorous fishes and filter feeders ($p = 0.808$). There were also significant differences between concentrations of Se in carnivores or omnivores and filter feeders from BYDL ($p < 0.05$) by no differences were observed between carnivores and omnivores ($p = 1.0$). Concentrations of Se measured in fishes during the present study were greater than those in prey of little egrets (*Egretta garzetta*) from TL and Poyang Lake (1.3 and 1.4 μg/g dm) (Zhang et al. 2006), fishes from the Chongming wetland (0.03–1.34 μg/g dm), Ya-Er Lake (0.39–1.5 μg/g dm) (Jin et al. 2006), reservoirs of eastern China (0.12 to 0.28 μg/g dm), Nam Co Lake, and Lhasa River (Yang et al. 2007), but less than concentrations in icefish (*Protosalanx hyalocranius*) from TL (5.4 μg/g dm) (Yang et al. 2009) and fishes from Yamdro Lake (Yang et al. 2007). The concentrations of Se observed in this study were less than those in fishes from the Republican River Basin of Colorado, Nebraska, and Kansas (1.14–24.3 μg/g dm) (May et al. 2001), Solomon River Basin (9.5 μg/g dm) (May et al. 2008), and a mountaintop removal coal mining-impacted stream in West Virginia, USA (6.3 and 8.8 μg/g dm for creek chub and green sunfish fillets, respectively).

Table 1 Characteristics of fish samples and concentrations (μg/g dm; mean, min–max) of Hg and Se in fishes collected from Tai and Baiyangdian Lakes

Fish species	Number of sample	Feeding habit	Body length (cm)	Total Hg		Total Se
			(Mean, min–max)	Muscle tissue	Whole body[b]	Muscle tissue
Tai Lake						
Catfish	9	C	35, 31–44	0.27, 0.14–0.56	0.16, 0.089–0.32	3.5, 3.0–4.1
Common carp	25	O	29, 20–40	0.037, 0.00019–0.092	0.027, 0.00024–0.063	6.8, 3.7–12.4
Bighead carp	19	F	40, 33–43	0.057, 0.013–0.16	0.040, 0.011–0.10	1.8, 0.2–2.4
Spotted steed	6	C	23, 21–24	0.20, 0.079–0.32	0.12, 0.055–0.19	1.6, 1.2–2.4
Topmouth culter	19	C	33, 24–40	0.20, 0.064–1.38	0.12, 0.045–0.72	1.0, 0.6–2.1
Chinese perch	7	C	26, 23–33	0.12, 0.081–0.21	0.078, 0.056–0.13	4.1, 3.4–4.5
Crucian carp	2	O	22, 22–22	0.047, 0.043–0.051	0.034, 0.032–0.037	0.6, 0.6–0.7
Baiyangdian Lake						
Weather loach	2	O	16, 15–17	0.064, 0.034–0.093	0.044, 0.026–0.063	3.0, 2.8–3.2
Catfish	2	C	26, 25–27	0.52, 0.43–0.60	0.30, 0.25–0.34	1.3, 1.3–1.3
Silver carp	10	F	27, 20–37	0.13, 0.053–0.48	0.082, 0.038–0.28	1.2, 1.0–1.4
Yellow catfish	10	C	17, 14–22	0.28, 0.10–0.41	0.17, 0.067–0.24	1.9, 1.3–3.4
Crucian carp[a]	39	O	14, 13–19	0.19, 0.088–0.37	0.12, 0.060–0.22	1.6, 1.1–2.2[a]

C, carnivore; *O*, omnivore; *F*, filter feeder

[a] The number of sample measured for Se was 14, and the body length of fish ranged 13–19 cm with a mean value of 15 cm

[b] The whole body concentrations of Hg or Se were converted from muscle concentrations with equations

Previously, there were few data available for total concentrations of Hg or Se eggs of fishes in China. Mean total concentrations of Hg and Se in eggs of fishes from BYDL were 0.047 (range 0.011–0.15) and 2.6 μg/g dm (range 1.4–4.1), respectively (Fig. 4). These concentrations were comparable to concentrations in muscle of fishes. Concentrations of both Hg and Se in eggs of carnivorous fishes, such as yellow catfish and catfish, were greater than those in eggs of omnivorous fishes or filter feeder. Compared with total concentrations of Hg in eggs of fishes from other regions, concentrations of Hg measured in the current study were less than concentrations determined in eggs of walleye from North America (range 5 to 762 ng/g dm) (Johnston et al. 2001) and those in eggs of fishes from the Mississippi River (range < 44 to 220 ng/g dm) (May et al. 2009) and eggs of female largemouth bass collected from six waterbodies in North Carolina, USA (range 0.1 to 1.0 μg/g dm) (Sackett et al. 2013). Concentrations of Se in eggs of fishes from BYDL were less than those in gonads of fishes where Se contamination or adverse effects occurred in the field (Brandt et al. 2017; Herrmann et al. 2016).

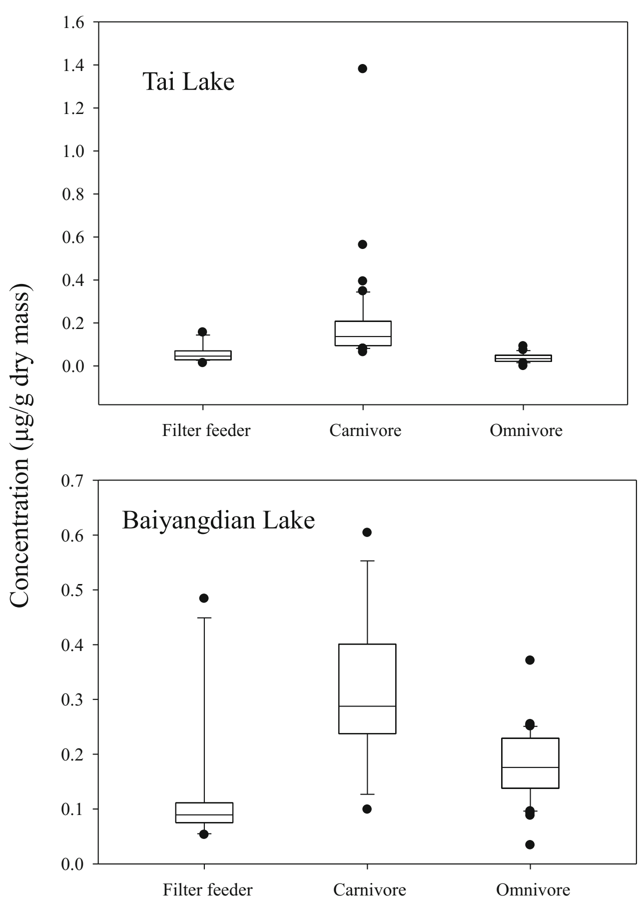

Fig. 2 Box and whisker plots of total concentrations of mercury (Hg) in various feeding guilds of fishes. Box edges represent the upper and lower quantile with median value shown in the middle of the box. Whiskers indicate 1.5 times the interquartile range of the data. Points falling outside the range of whiskers are shown as dots

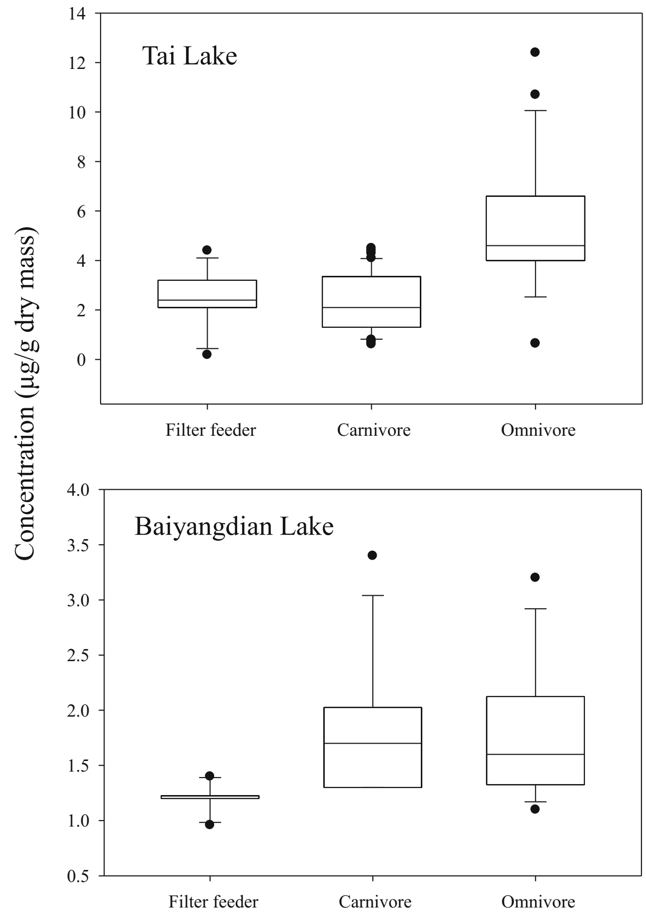

Fig. 3 Box and whisker plots of total concentrations of Se among feeding guilds

Screening level characterization of hazard

Hazard based on Hg and Se in whole bodies or muscles of fishes

Values of HQs suggested that hazards posed by concentrations of Hg in whole bodies of fishes from TL and BYDL are small (Fig. 5a). The mean HQ for fishes from TL was 0.07 (range from 0.0002 to 0.7), indicating little hazard. HQs for Hg in whole bodies of fishes from BYDL ranged from 0.03 to 0.3 with a mean of 0.1. Approximately 9% and 14% of HQs exceeded 0.1 for fishes from TL and BYDL, respectively. These results suggested moderate hazard of Hg toward fishes in these two lakes. Concentrations of Hg in whole fish that are less than the threshold value of 0.1 μg/g dm are not expected to have adversely effects on survival, growth, or reproduction of juvenile or adult fish (Beckvar et al. 2005). Therefore, exposures of fishes to Hg in neither TL nor BYDL currently pose unacceptable hazards. Relatively greater HQs were observed for carnivorous fishes, such as catfish, spotted steed, top mouth culter and Chinese perch in TL, and catfish and yellow catfish in BYDL. Fishes at higher trophic levels exhibited greater hazard caused by Hg. This can be explained, in part, by biomagnification of Hg. Carnivorous and omnivorous fishes exhibited greater hazard for Hg than did filter feeding fishes. Previously, when hazard of Hg to four piscivorous fishes in the North American Great Lakes were assessed, fishes at 3 to 18% of sites exhibited hazard for adverse effects on reproduction and survival (Sandheinrich et al. 2011). A screening level probabilistic assessment indicated that piscivorous wildlife feeding in the south-central region of the Everglades are at relatively great risk of effects of Hg (Duvall and Barron 2000).

Based on concentrations of Se in muscle of fishes, the mean HQ of Se from TL was 0.3, with a range of 0.02 to 1.1, while the mean HQ for BYDL was 0.1, with a range of 0.1 to 0.3. Rates of HQs exceeding 0.1 were almost 100% at BYDL and 86% at TL. HQ of one sample of common carp from TL exceeded 1.0 and the mean HQ for common carp was greatest among species collected during the present study (Fig. 5b), which indicated moderate hazard of Se for these fishes. Longer durations of larger common carp might have resulted in adverse effects, including larval mortality, edema, or deformities due to exposure of Se in TL. In freshwater ecosystems, Se can be bioaccumulated and biomagnified through food webs, and greater concentrations of Se can have effects on reproduction of fishes (Linares-Casenave et al. 2015). In a recent study, concentrations of Se were surveyed in three North Carolina lakes, which had received effluent containing Se from coal-fired power plants, and in lakes with the greatest inputs of Se, 85% of muscle of fishes exceeded the TRV used in the present study, while 31% of concentrations of Se in ovary/egg exceeded the TRV based on concentrations in egg (Brandt et al. 2017). Concentrations of Se in these lakes were comparable to those measured during historic fish extirpation events in the USA, in which effects on fish were attributed to contamination in runoff from selenium-laden coal combustion residuals (Brandt et al. 2017).

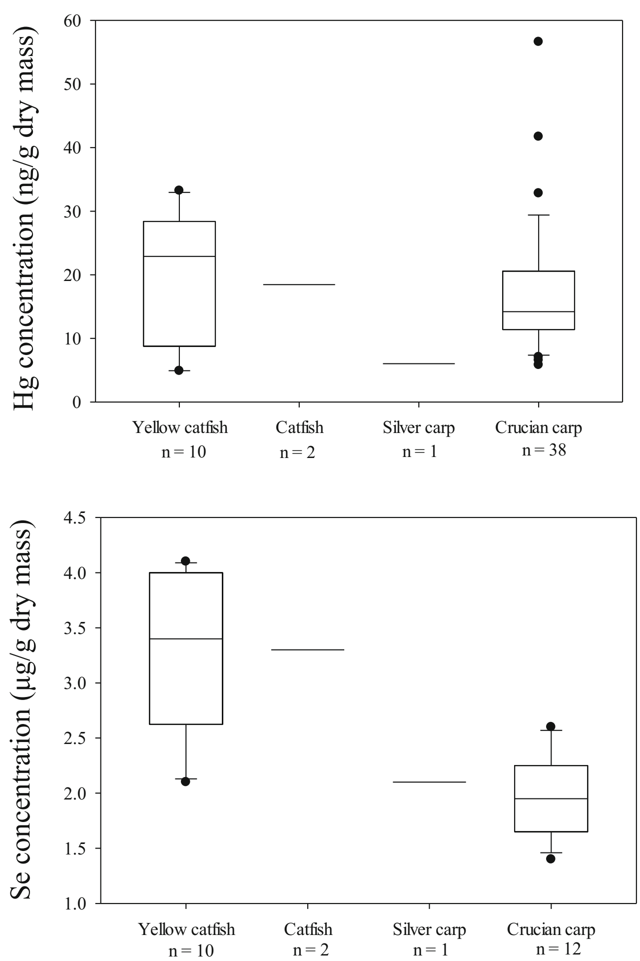

Fig. 4 Box and whisker plots of total concentrations of Hg and Se in eggs of fishes from Baiyangdian Lake. *n* represents the number of samples

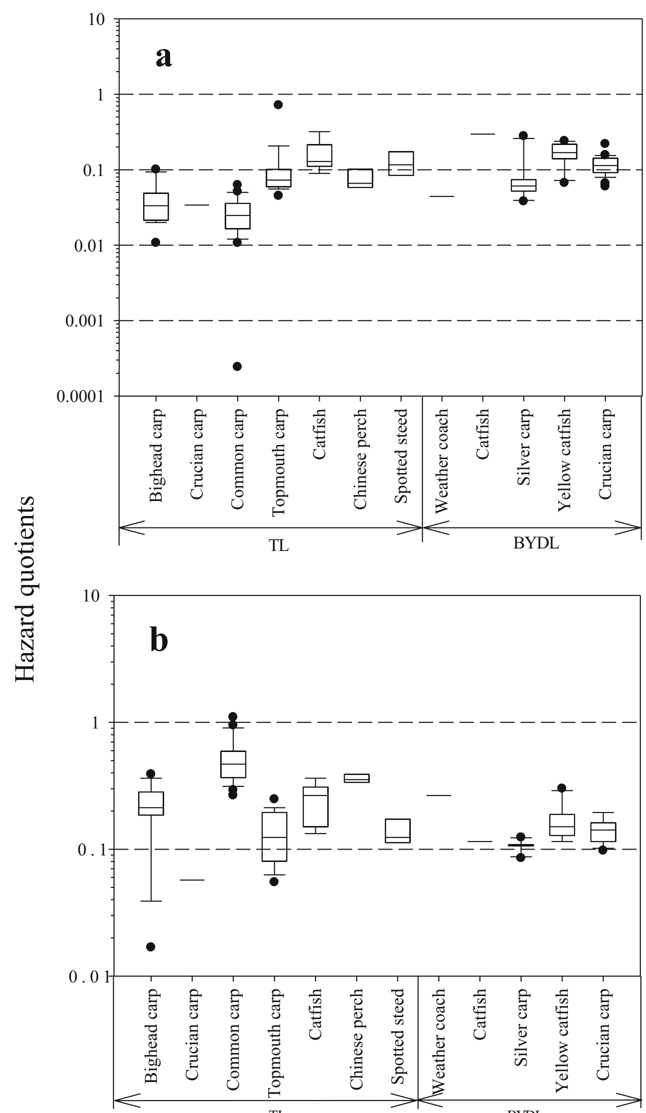

Fig. 5 Box and whisker plots of hazard to fishes from Hg (**a**) residue in whole fish and Se (**b**) in fish muscles in Tai Lake and Baiyangdian Lake

Hazard based on Hg and Se in fish egg

Maternal transfer of Se or Hg to eggs can result in adverse effects on populations of fishes. Therefore, eggs are appropriate tissue for evaluating whether concentrations of Se or Hg have potential to cause toxicity to larval fishes as a result of maternal transfer (DeForest et al. 1999, 2012; Stefansson et al. 2014). Based on concentrations of Hg in fishes, HQs for Hg in eggs of fishes from BYDL ranged from 0.2 to 2.8 with a mean of 0.9. Twenty seven percent of HQs exceeded 1.0 for fishes including yellow catfish and crucian carp, which would likely be adversely affected by the presence of Hg (Fig. 6). HQs for catfish and silver carp were near 1.0. The TRV of 0.02 μg/g wm, derived by the simple ranking approach, was the same concentration of Hg in the control treatment from a single experiment (Beckvar et al. 2005), and thus might not be representative and thus conclusions based on that TRV should be interpreted with caution. In a study of reproductive effects of methyl mercury (MeHg), on *Fundulus heteroclitus*, reduced fertilization success was observed at concentrations of 0.01 to 0.63 μg/g wm in egg, and alterations in sex ratios were also observed at concentrations of less than 0.01 μg/g wm in eggs (Matta Mary et al. 2009). In a laboratory experiment, in which adult Atlantic croaker (*Micropogonias undulatus*) was fed MeHg-contaminated food resulted in concentrations of MeHg ranging from 0.04 to 4.6 ng/g wm in eggs, which resulted in concentration-dependent effects on survival and behaviors of larvae (Alvarez Mdel et al. 2006). For walleye (*Stizostedion vitreum*) eggs collected from an environment in which the content of Hg was significantly greater than background, Clay Lake, Ontario, Canada, concentrations of Hg ranged from 588 to 1714 ng/g dm, which is greater than the TRV as well as greater than concentrations observed during the present study. While relatively great of mortality of eggs, which was about 80% with occurrence of deformities, was not significantly correlated with MeHg in eggs (Latif et al. 2001). This result suggested that factors other than Hg were more likely causes of the observed adverse effects on survival of eggs. Alternatively, the minimum threshold for adverse effects Hg might have been exceeded, but what was being observed was variation in sensitivities among individuals.

The HQs for Se in eggs of fishes ranged from 0.1 to 0.3 with a mean of 0.2, which indicated moderate hazard to reproduction of these fishes. Se deposited in eggs via maternal transfer has potential to cause mortality, edema, and deformities of larval fishes (DeForest et al. 2012). Concentrations of Se in eggs of northern pike (*Esox lucius*) collected from locations of greater and moderate contamination from a uranium milling operation in northern Saskatchewan, Canada, were 48 and 31 μg/g dm, respectively. There were significantly greater frequencies in deformities and edema in fry at these locations compared to rates for fry from females collected at an uncontaminated, reference location (Muscatello et al. 2006). For Dolly Varden char (*Salvelinus malma*) eggs collected from waterbodies in British Columbia, Canada, in a region where concentrations of Se resulting from mining were greater than background, there was a relationship between concentrations (5.4 to 66 μg/g dm) in eggs and rates of deformities of larvae. The threshold concentration of Se corresponding to a 10% greater frequency of deformities was 54 μg/g dm (Mcdonald et al. 2010). For cutthroat trout (*Oncorhynchus clarki lewisi*) collected from a site of active coal mining in British Columbia, a significant positive relationship between concentrations of Se (11.8 to 140.0 μg/g dm) in eggs and rates of mortality was observed, but no relationship between rates of deformities or edema and concentrations of Se in eggs was observed in the range of 11.8 and 20.6 μg/g dm (Rudolph et al. 2008). Based on this information, it was deemed that it was unlikely that the Se measured in eggs of fishes from BYDL would have adverse effects on fecundity.

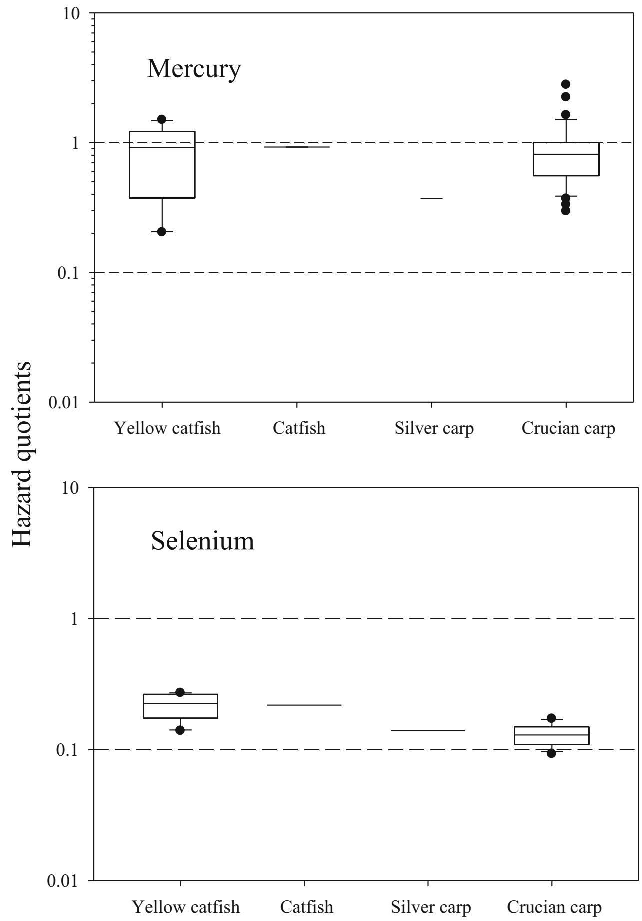

Fig. 6 Box and whisker plots of hazard posed by concentrations of Hg or Se in eggs of fishes in Baiyangdian Lake

Protection of Se against Hg

If Se is present in a 1:1 M ratio with Hg, it can protect fishes from toxic effects of Hg without causing toxicity due to the Se (Ganther et al. 1972; Peterson et al. 2009; Ralston et al. 2008). Ratios of Se to Hg based on molar concentrations in tissues that exceed 1.0 are largely protective from adverse effects of Hg to fish. Selenium might reduce availability of Hg to aquatic organisms in lakes, and limit the whole-body assimilation of Hg at lower levels of the aquatic food chain (Belzile et al. 2006). Concentrations of Hg in walleye were inversely proportional to concentrations of Se (Yang et al. 2010). Greater concentrations of Hg in fishes inhabiting a stream in the western USA were observed only when concentrations of Se in tissues were small (Peterson et al. 2009). In the present study, mean molar ratios of Se to Hg were 200, 32, and 210 for fish muscle in TL and BYDL, and eggs of fishes from BYDL, respectively. Inverse relationships between concentrations of Hg and Se were also observed in tissues of fishes from TL or BYDL (Fig. 7). When concentrations of Se in muscle of fishes from TL were greater than 60 μmol/kg, concentrations of Hg were small. Similar results were obtained for fishes from the Tapajos River, Brazilian Amazon, where greatest mean concentrations of Se and smallest mean concentrations of Hg were measured in herbivorous; the opposite trend was observed for piscivorous fishes from the same region (Sampaio da Silva et al. 2013). Thresholds of concentrations of Se in muscle of fishes were 80 μmol/kg (6.2 mg/kg dm) or greater; concentrations of Hg were generally small (Yang et al. 2010). Median Se:Hg molar ratios in a tropical estuary were 5 to 70 in the liver and muscle of four fishes (Kehrig et al. 2009). For stream fishes of the Western USA, 97.5% of individual fish had molar ratios greater than 1.0 (Peterson et al. 2009). Molar ratios of Se to Hg in muscle of fishes from rivers and lakes in Eastern Slovakia were in the range of 0.56 to 2.5 (Strapáč et al. 2012). An inverse correlation was also observed between the Se/Hg ratio and total concentrations of Hg in muscle of examined fishes. Greater molar ratios of Se to MeHg (range 69–9263) were available in freshwater organisms from lakes near metal smelters in Sudbury, Ontario (Belzile et al. 2006). Based on the molar ratios observed for fishes in this study, Se would be expected to mitigate accumulation of Hg and its adverse effects on fishes inhabiting TL and BYDL. Moreover, in both TL and BYDL, molar ratios of Se to Hg in muscle of carnivores were less than ratios for filter feeders or omnivores. This observation is consistent with results of several previous studies that piscivores had lesser concentrations of Se or molar ratios of Se to Hg than did nonpiscivores (Peterson et al. 2009; Sampaio da Silva et al. 2013). This perhaps is another reason that piscivores fish have great hazard from Hg exposure.

Uncertainties

Tissue-based concentrations of bioaccumulative contaminants could exceed the protective threshold even though dissolved surface water concentrations did not (Brandt et al. 2017). Tissue-based TRVs were prioritized over concentrations in environmental media because it better describes toxicity throughout food chain exposure of aquatic organisms, such as fishes (Brandt et al. 2017; USEPA 2016). In present study, tissue-based TRVs were used, and the results will be useful for understanding hazards posed by Hg and Se to fishes in TL and BYDL and guiding the toxicity risk assessment or pollution control in future. However, uncertainties in assessments of hazard that depend on measurement endpoints as well as durations and intensities of exposures and resources available, knowledge of the assessor, and method of hazard characterization need to be considered. In the present study, the tissue-based TRV was based primarily on concentrations of MeHg (Beckvar et al. 2005). However, data for concentrations of Hg in fishes (exposure) were expressed as total concentrations of Hg, including both organic and inorganic Hg and neutral and ionic forms. Comparing total concentrations of Hg directly with the TRV based on MeHg could result in an overestimate of the HQ, such that further investigation might be needed to quantify MeHg in fishes. A conservative assumption that vertebrates have > 50% of the THg as MeHg in muscle (Albers et al. 2007; Eisler 2000) indicates that this bias would be small. Since the greater bias caused by not accounting for the relative proportions of organic and inorganic Hg, minimization of this potential bias is further supported by the fact that most HQs were less than 1.0. Moreover, the TRV applied in this study does not consider the protective effects of the simultaneous presence of Se. TRVs available for effects of Hg on fishes are based on total body burdens and in this study exposure to Hg was estimated by use of concentrations in dorsal muscle and excluded viscera, including liver, in which concentrations of Se as well as Hg can be greater. Therefore, predictive models previously used in hazard assessment or aquatic life water quality criteria were used to convert the muscle-based concentrations of Hg into whole-body concentrations (Dillon et al. 2010; Sandheinrich et al. 2011; USEPA 2004). Because conversion from tissue to whole-body Se concentration can result in some uncertainty in the estimate, site data analysis should be conducted to develop specific conversion factors. A TRV of 0.02 μg/g wm was used to assess the hazard of Hg in eggs of fishes and this value was derived by using the simple ranking approach (Beckvar et al. 2005). In this method, only three data based on toxicity to egg, fry, and larvae of fishes were used and ranked from lesser to greater concentrations, and the least value of 0.02 μg/g wm was selected to be the TRV. The quotient method is conservative, and it only illustrates a preliminary estimation of hazard risk. Uncertainty can be lessened or at least described by assessing

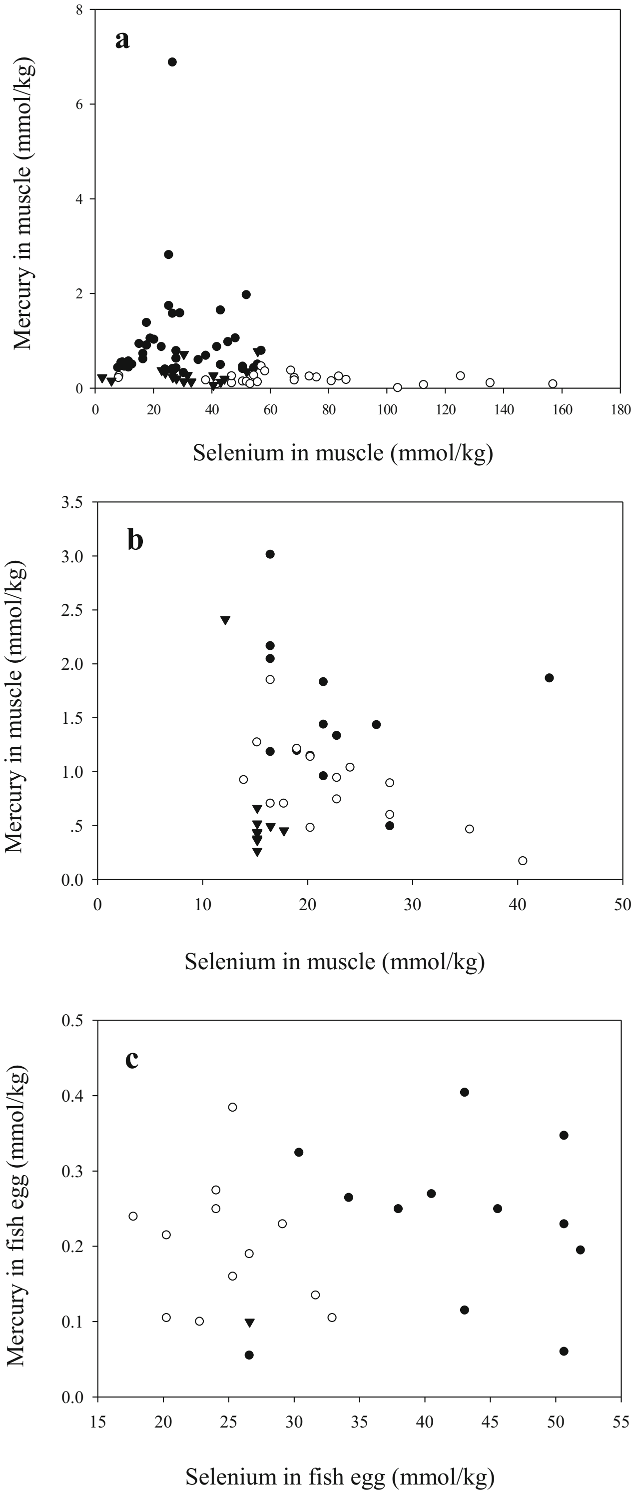

Fig. 7 Relationships between concentrations of Se and Hg muscle of fishes from Tai Lake (**a**) and Baiyangdian Lake (**b**) and egg from Baiyangdian Lake (**c**). (Triangles for filter feeder, open circles for omnivore, dots for carnivore)

risk instead of hazard, by use of probabilistic estimates of both hazard and exposure can be used more complete describe variations in these parameters.

Conclusions

Concentration of both Hg and Se in muscle of fishes inhabiting TL and BYDL in China were deemed to be minimal to moderate, based on whole body and egg concentrations for most fishes in TL and BYDL. Fishes in feeding guilds at higher trophic levels experienced greater hazard from Hg than did those at lower levels. The hazard of Hg in fish eggs and Se in common carp muscle need to be concerned. In addition, the molar ratios of Se to Hg were greater than 1.0 in TL and BYDL, such that fishes might have been protected from adverse effects of Hg on fishes from both TL and BYDL.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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References

Albers PH, Koterba MT, Rossmann R, Link WA, French JB, Bennett RS, Bauer WC (2007) Effects of methylmercury on reproduction in American kestrels. Environ Toxicol Chem 26:1856–1866

Alvarez Mdel C, Murphy CA, Rose KA, McCarthy ID, Fuiman LA (2006) Maternal body burdens of methylmercury impair survival skills of offspring in Atlantic croaker (Micropogonias undulatus). Aquat Toxicol 80:329–337

Beckvar N, Dillon TM, Read LB (2005) Approaches for linking whole-body fish tissue residues of mercury or DDT to biological effects thresholds. Environ Toxicol Chem 24:2094–2105

Belzile N, Chen YW, Gunn JM, Tong J, Alarie Y, Delonchamp T, Lang CY (2006) The effect of selenium on mercury assimilation by freshwater organisms. Can J Fish Aquat Sci 63:1–10

Birge W, Black J, Westerman A, Hudson J (1979): The effects of mercury on reproduction of fish and amphibians. The biogeochemistry of mercury in the environment. Elsevier/North Holland Biomedical Press, Amsterdam, 629–55

Brandt JE, Bernhardt ES, Dwyer GS, Di Giulio RT (2017) Selenium ecotoxicology in freshwater lakes receiving coal combustion residual effluents: a North Carolina example. Environ Sci Technol 51: 2418–2426

Chen C, Pickhardt P, Xu M, Folt C (2008) Mercury and arsenic bioaccumulation and eutrophication in Baiyangdian Lake, China. Water Air Soil Pollut 190:115–127

Dang F, Wang WX (2011) Antagonistic interaction of mercury and selenium in a marine fish is dependent on their chemical species. Environ Sci Technol 45:3116–3122

DeForest DK, Brix KV, Adams WJ (1999) Critical review of proposed residue-based selenium toxicity thresholds for freshwater fish. Hum Ecol Risk Assess 5:1187–1228

DeForest DK, Gilron G, Armstrong SA, Robertson EL (2012) Species sensitivity distribution evaluation for selenium in fish eggs: considerations for development of a Canadian tissue-based guideline. Integr Environ Assess Manag 8:6–12

Dillon T, Beckvar N, Kern J (2010) Residue-based mercury dose–response in fish: an analysis using lethality-equivalent test endpoints. Environ Toxicol Chem 29:2559–2565

Duvall SE, Barron MG (2000) A screening level probabilistic risk assessment of mercury in Florida Everglades food webs. Ecotoxicol Environ Saf 47:298–305

Eisler R, 2000: Handbook of chemical risk assessment: health hazards to humans, plants, and animals, volume 1: metals. Lewis Publishers, Boca Raton, FL, pp 313–409

Fu Z, Wu F, Amarasiriwardena D, Mo C, Liu B, Zhu J, Deng Q, Liao H (2010) Antimony, arsenic and mercury in the aquatic environment and fish in a large antimony mining area in Hunan, China. Sci Total Environ 408:3403–3410

Ganther HE, Goudie C, Sunde ML, Kopecky MJ, Wagner P, Oh S-H, Hoekstra WG (1972) Selenium: relation to decreased toxicity of methylmercury added to diets containing tuna. Science 175:1122–1124

Guo G, Wu F, He H, Zhang R, Li H (2012): Ecological risk assessment of organochlorine pesticides in surface waters of Lake Taihu, China. Hum Ecol Risk Assess

Hammerschmidt CR, Sandheinrich MB (2005) Maternal diet during oogenesis is the major source of methylmercury in fish embryos. Environ Sci Technol 39:3580–3584

Herrmann SJ, Nimmo DR, Carsella JS, Herrmann-Hoesing LM, Turner JA, Gregorich JM, Heuvel BDV, Nehring RB, Foutz HP (2016) Differential accumulation of mercury and selenium in brown trout tissues of a high-gradient urbanized stream in Colorado, USA. Arch Environ Contam Toxicol 70:204–218

Hodson PV, Hilton JW (1983) The nutritional requirements and toxicity to fish of dietary and waterborne selenium. Ecol Bull:335–340

Holm J, Palace V, Siwik P, Sterling G, Evans R, Baron C, Werner J, Wautier K (2005) Developmental effects of bioaccumulated selenium in eggs and larvae of two salmonid species. Environ Toxicol Chem 24:2373–2381

Hu L, Wang Y, Wang Q, Lu G, Xie Z, Zhang Z (2014): Distribution and ecological risk assessment of mercury in water, sediments and typical aquatic organisms from northern Taihu Lake. Journal of Agro-Environment Science 33, 1183–1188

Jarvinen AW, Ankley GT (1999) Linkage of effects to tissue residues: development of a comprehensive database for aquatic organisms exposed to inorganic and organic chemicals, SETAC Press

Jin L, Liang L, Jiang G, Xu Y (2006) Methylmercury, total mercury and total selenium in four common freshwater fish species from Ya-Er Lake, China. Environ Geochem Health 28:401–407

Johnston TA, Bodaly RA, Latif MA, Fudge RJP, Strange NE (2001) Intra- and interpopulation variability in maternal transfer of mercury to eggs of walleye (Stizostedion vitreum). Aquat Toxicol 52:73–85

Kehrig Hd ST, Palermo E, Baêta A, Castelo-Branco C, Malm O, Moreira I (2009) The relationships between mercury and selenium in plankton and fish from a tropical food web. Environ Sci Pollut Res 16:10–24

Latif MA, Bodaly RA, Johnston TA, Fudge RJP (2001) Effects of environmental and maternally derived methylmercury on the embryonic and larval stages of walleye (Stizostedion vitreum). Environ Pollut 111:139–148

Lemly AD (1993a) Guidelines for evaluating selenium data from aquatic monitoring and assessment studies. Environ Monit Assess 28:83–100

Lemly AD (1993b) Teratogenic effects of selenium in natural populations of fresh water fish. Ecotoxicol Environ Saf 26:181–204

Linares-Casenave J, Linville R, Eenennaam JPV, Muguet JB, Doroshov SI (2015) Selenium tissue burden compartmentalization in resident white sturgeon (Acipenser transmontanus) of the San Francisco Bay Delta estuary. Environ Toxicol Chem 34:152–160

Matta Mary B, Linse J, Cairncross C, Francendese L, Kocan Richard M (2009) Reproductive and transgenerational effects of methylmercury or Aroclor 1268 on Fundulus heteroclitus. Environ Toxicol Chem 20:327–335

May TW, Walther MJ, Petty JD, Fairchild JF, Lucero J, Delvaux M, Manring J, Armbruster M, Hartman D (2001) An evaluation of selenium concentrations in water, sediment, invertebrates, and fish from the Republican River Basin: 1997–1999. Environ Monit Assess 72:179–206

May TW, Fairchild JF, Petty JD, Walther MJ, Lucero J, Delvaux M, Manring J, Armbruster M (2008) An evaluation of selenium concentrations in water, sediment, invertebrates, and fish from the Solomon River Basin. Environ Monit Assess 137:213–232

May TW, Walther MJ, Brumbaugh WG, McKee MJ (2009) Concentrations of elements in whole-body fish, fish fillets, fish muscle plugs, and fish eggs from the 2008 Missouri Department of Conservation general contaminant monitoring program. Open-file report 2009–1278, U.S. Geological Survey, Reston, Virginia

Mcdonald BG, Debruyn AMH, Elphick JRF, Davies M, Bustard D, Chapman PM (2010) Developmental toxicity of selenium to Dolly Varden char (Salvelinus malma). Environ Toxicol Chem 29:2800–2805

Meador JP (2015) Tissue concentrations as the dose metric to assess potential toxic effects of metals in field-collected fish: copper and cadmium. Environ Toxicol Chem 34:1309–1319

MOH (2010) National Food Safety Standard, Determination of Selenium in Foods. GB5009.93–2010. Ministry of Health of the People's Republic of China, Beijing

Moore DR, Sample BE, Suter GW, Parkhurst BR, Teed RS (1999) A probabilistic risk assessment of the effects of methylmercury and PCBs on mink and kingfishers along East Fork Poplar Creek, Oak Ridge, Tennessee, USA. Environ Toxicol Chem 18:2941–2953

Muscatello JR, Bennett PM, Himbeault KT, Belknap AM, Janz DM (2006) Larval deformities associated with selenium accumulation in northern pike (Esox lucius) exposed to metal mining effluent. Environ Sci Technol 40:6506–6512

Peterson SA, Sickle JV, Hughes RM, Schacher JA, Echols SF (2005) A biopsy procedure for determining filet and predicting whole-fish mercury concentration. Arch Environ Con Tox 48:99–107

Peterson SA, Ralston NVC, Peck DV, John VS, J David R, Spate VL, J Steven M (2009) How might selenium moderate the toxic effects of mercury in stream fish of the western U.S.? Environ Sci Technol 43:3919–3925

Ralston NVC, Ralston CR, Blackwell Iii JL, Raymond LJ (2008) Dietary and tissue selenium in relation to methylmercury toxicity. NeuroToxicology 29:802–811

Rudolph BL, Andreller I, Kennedy CJ (2008) Reproductive success, early life stage development, and survival of westslope cutthroat trout (Oncorhynchus clarki lewisi) exposed to elevated selenium in an area of active coal mining. Environ Sci Technol 42:3109–3114

Rumbold DG (2005) A probabilistic risk assessment of the effects of methylmercury on great egrets and bald eagles foraging at a constructed wetland in South Florida relative to the Everglades. Hum Ecol Risk Assess 11:365–388

Rumbold D, Lange T, Axelrad D, Atkeson T (2008) Ecological risk of methylmercury in Everglades National Park, Florida, USA. Ecotoxicology 17:632–641

Sackett DK, Aday DD, Rice JA, Cope WG (2013) Maternally transferred mercury in wild largemouth bass, Micropterus salmoides. Environ Pollut 178:493–497

Sampaio da Silva D, Lucotte M, Paquet S, Brux G, Lemire M (2013) Inverse mercury and selenium concentration patterns between herbivorous and piscivorous fish in the Tapajos River, Brazilian Amazon. Ecotoxicol Environ Saf 97:17–25

Sánchez-Bayo F, Baskaran S, Kennedy IR (2002) Ecological relative risk (EcoRR): another approach for risk assessment of pesticides in agriculture. Agric Ecosyst Environ 91:37–57

Sandheinrich M, Bhavsar S, Bodaly RA, Drevnick P, Paul E (2011) Ecological risk of methylmercury to piscivorous fish of the Great Lakes region. Ecotoxicology 20:1577–1587

Sappington KG, Bridges TS, Bradbury SP, Erickson RJ, Hendriks AJ, Lanno RP, Meador JP, Mount DR, Salazar MH, Spry DJ (2011) Application of the tissue residue approach in ecological risk assessment. Integr Environ Assess Manag 7:116–140

Scheuhammer AM, Meyer MW, Sandheinrich MB, Murray MW (2007) Effects of environmental methylmercury on the health of wild birds, mammals, and fish. AMBIO: A Journal of the Human Environment 36:12–19

Shen G, Lu Y, Wang M, Sun Y (2005) Status and fuzzy comprehensive assessment of combined heavy metal and organo-chlorine pesticide pollution in the Taihu Lake region of China. J Environ Manag 76: 355–362

Sørmo EG, Ciesielski TM, Øverjordet IB, Lierhagen S, Eggen GS, Berg T, Jenssen BM (2011) Selenium moderates mercury toxicity in free-ranging freshwater fish. Environ Sci Technol 45:6561–6566

Stefansson ES, Heyes A, Rowe CL (2014) Tracing maternal transfer of methylmercury in the sheepshead minnow (Cyprinodon variegatus) with an enriched mercury stable isotope. Environ Sci Technol 48: 1957–1963

Strapáč I, Sokol J, D Ž BM (2012) Mercury and selenium concentrations in muscle tissue of different species of predatory freshwater fish and correlation between these elements. Food Additives & Contaminants Part B Surveillance 5:194–199

USEPA (1998) Guidelines for ecological risk assessment. EPA/630/R095/002F, U.S. Environmental Protection Agency, Risk Assessment Forum, Washington, DC

USEPA (2004) Aquatic life water quality criteria for selenium. U.S. Environmental Protection Agency, Office of Water, Washington, D.C.

USEPA (2016) Final criterion: aquatic life ambient water quality criterion for selenium - freshwater 2016, U.S. Environmental Protection Agency, Washington, D.C.

Wang J, Feng X, Anderson CWN, Wang H, Zheng L, Hu T (2012a) Implications of mercury speciation in thiosulfate treated plants. Environ Sci Technol 46:5361–5368

Wang J, Feng X, Anderson CWN, Xing Y, Shang L (2012b) Remediation of mercury contaminated sites – a review. J Hazard Mater 221–222:1–18

Wang S, Li B, Zhang M, Xing D, Jia Y, Wei C (2012c) Bioaccumulation and trophic transfer of mercury in a food web from a large, shallow, hypereutrophic lake (Lake Taihu) in China. Environ Sci Pollut Res 19:2820–2831

Webb MAH, Feist GW, Fitzpatrick MS, Foster EP, Schreck CB, Plumlee M, Wong C, Gundersen DT (2006) Mercury concentrations in gonad, liver, and muscle of white sturgeon Acipenser transmontanus in the lower Columbia River. Arch Environ Con Tox 50:443–451

Weech SA, Scheuhammer AM, Elliott JE (2006) Mercury exposure and reproduction in fish-eating birds breeding in the Pinchi Lake region, British Columbia, Canada. Environ Toxicol Chem 25:1433–1440

Wenchuan Q, Dickman M, Sumin W (2001): Multivariate analysis of heavy metal and nutrient concentrations in sediments of Taihu Lake, China. Hydrobiologia 450, 83–89

Yang W, Yang L, Zheng J (1996) Effect of metal pollution on the water quality in Taihu Lake. GeoJournal 40:197–200

Yang R, Yao T, Xu B, Jiang G, Xin X (2007) Accumulation features of organochlorine pesticides and heavy metals in fish from high mountain lakes and Lhasa River in the Tibetan Plateau. Environ Int 33: 151–156

Yang J, Xun XU, Liu HB (2009) Bioaccumulation of elements in icefish Protosalanx hyalocranius from the Taihu Lake and Hongze Lake. Oceanologia Et Limnologia Sinica 40:201–207

Yang D-Y, Xu Y, Chen Y-W, Belzile N (2010) Inverse relationships between selenium and mercury in tissues of young walleye (Stizosedion vitreum) from Canadian boreal lakes. Sci Total Environ 408:1676–1683

Zhang Y, Ruan L, Fasola M, Boncompagni E, Dong Y, Dai N, Gandini C, Orvini E, Ruiz X (2006) Little egrets (Egretta garzetta) and trace-metal contamination in wetlands of China. Environ Monit Assess 118:355–368

Zhang R, Wu F, Li H, Guo G, Feng C, Giesy JP, Chang H (2013) Toxicity reference values and tissue residue criteria for protecting avian wildlife exposed to methylmercury in China. Rev Environ Contam Toxicol 223:53–80

Zhao Y, Xia XH, Yang ZF, Wang F (2012) Assessment of water quality in Baiyangdian Lake using multivariate statistical techniques. Procedia Environ Sci 13:1213–1226