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FOCUSED REVIEW

Research Progress of Mercury Bioaccumulation in the Aquatic Food Chain, China: A Review

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Abstract
Research on mercury (Hg) in aquatic ecosystems in China has focused mainly on fish, with little research on the base of the food chain and Hg bioaccumulation mechanisms. This paper summarizes research progress pertaining to the characteristics, current status, and trends of Hg accumulation in the aquatic food chain in China, analyzes the effects of human activities on the transmission and accumulation of Hg in aquatic food chains, and assesses their risks to human and ecosystem health. A comparison of fish samples in China between 2000 and 2018 indicates that their total Hg content remains at relatively safe levels. However, because current information is generally insufficient to confirm how anthropogenic activities affect transformation and bioaccumulation in the aqueous environment, Hg isotope studies should be a focus of research on aquatic food webs. Additionally, more attention should be paid to Hg transport and bioaccumulation in the basic food chain by focusing on multi-contaminant joint exposure studies and establishing Hg bio-transport models.

Keywords Hg · Aquatic food chain · Bioaccumulation · China

Hg is one of the most hazardous pollutants in aquatic ecosystems, where it is readily transformed to methylmercury (MeHg), which bioaccumulates and biomagnifies in aquatic organisms. Elevated Hg concentrations in fish have been an environmental concern worldwide for decades, both in fish living near contaminated sites receiving direct Hg loads (Buzina et al. 1989; Mikac and Picer 1985; Puga et al. 2018) and also in fish living in environments where no direct sources exist, such as natural lakes (Håkanson et al. 1990; Huggett et al. 2001; Lindqvist et al. 1991; Watras and Frost 1989; Evans et al. 2005; Eagles-Smith et al. 2016), wetlands (Snodgrass et al. 2000; Gbogbo et al. 2017), oceans (Rolfhus and Fitzgerald 1995), and even new reservoirs (Bodaly et al. 1984; Hecky et al. 1991; Lucotte et al. 1999; Hylander et al. 2006) since the 1980s.

China is the world's largest producer, user, and emitter of Hg, as well as the world's largest producer and exporter of fish which contributed more than 60% of world production by quantity (FAO 2012). Through the past decades, thousands of hydropower reservoirs have been built in China. Meanwhile, aquaculture blooms and eutrophication have become common in rivers, lakes, reservoirs, and coastal zones (Li 2018). Environmental pollution and destruction of ecosystems have also made the accumulation of Hg in aquatic food chains more complex in China. Therefore, more attention has been paid to the bioaccumulation of Hg in fish. However, results show that most of the Hg concentrations in fish across the country, except for in Tibet and Qiandao Lake, do not exceed the limit of Hg content established by the World Health Organization (WHO < 0.5 mg/kg) (Pan et al. 2014; Yan et al. 2010; Yi et al. 2011) and that concentrations have even been very low in some Hg-contaminated areas in the last decades (Liu et al. 2012; Yan et al. 2010).

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Previous research has suggested that these low concentrations can be attributed to the use of aquaculture and overfishing, which lead to a short and simple food chain due to the biodilution effect and destruction of food chain integrity (He et al. 2008; Liu et al. 2012; Wang et al. 2011; Yao et al. 2011; Zhang 2009). Based on this context, this paper summarizes and analyzes the research progress pertaining to Hg accumulation in China, including the characteristics, current status, and trends of accumulation in the aquatic food chain. We aim to provide a summary of the research on Hg bioaccumulation in aquatic ecosystems in China and also discuss prospects for future research.

To examine the spatial changes of Hg in fish, we established a national database of Hg in both marine and freshwater fish in China covering up to 7197 fish samples collected from 35 different administrative 73 sites. Based on these extensive data collection and meta-analysis, we then created a national spatial distribution maps of fish Hg levels (Fig. 1), which illustrated all fish Hg data from 2000 to 2018 from different basins. All fish Hg data are presented on a wet weight (ww) basis.

Research Progress

In the past three decades, research on Hg in aquatic ecosystems in China has focused mainly on the accumulation of Hg in fish (Liu et al. 2014b; Razavi et al. 2014b; Shao et al. 2011), but there has been little research on the base of the food chain, such as on the benthic and pelagic community or biofilms and aquatic plants, and more attention has been given to current human health risks than to ecosystem health risks. Additionally, the research on the accumulation of Hg in fish has focused mainly on the following points: (1) health risk assessment of edible aquatic products; (2) research on the effects of human activities on the biological transport and accumulation of Hg in aquatic food chains; and (3) evaluating bioaccumulation processes of Hg using stable isotopes of carbon, nitrogen, and Hg. These three points are discussed in detail in the remainder of this section.

Because fish is the main dietary source of high-quality protein for humans, health risk assessment of fish products has been given high priority. Investigations initiated in the late 1980 s in the northern-tier states of the United States,

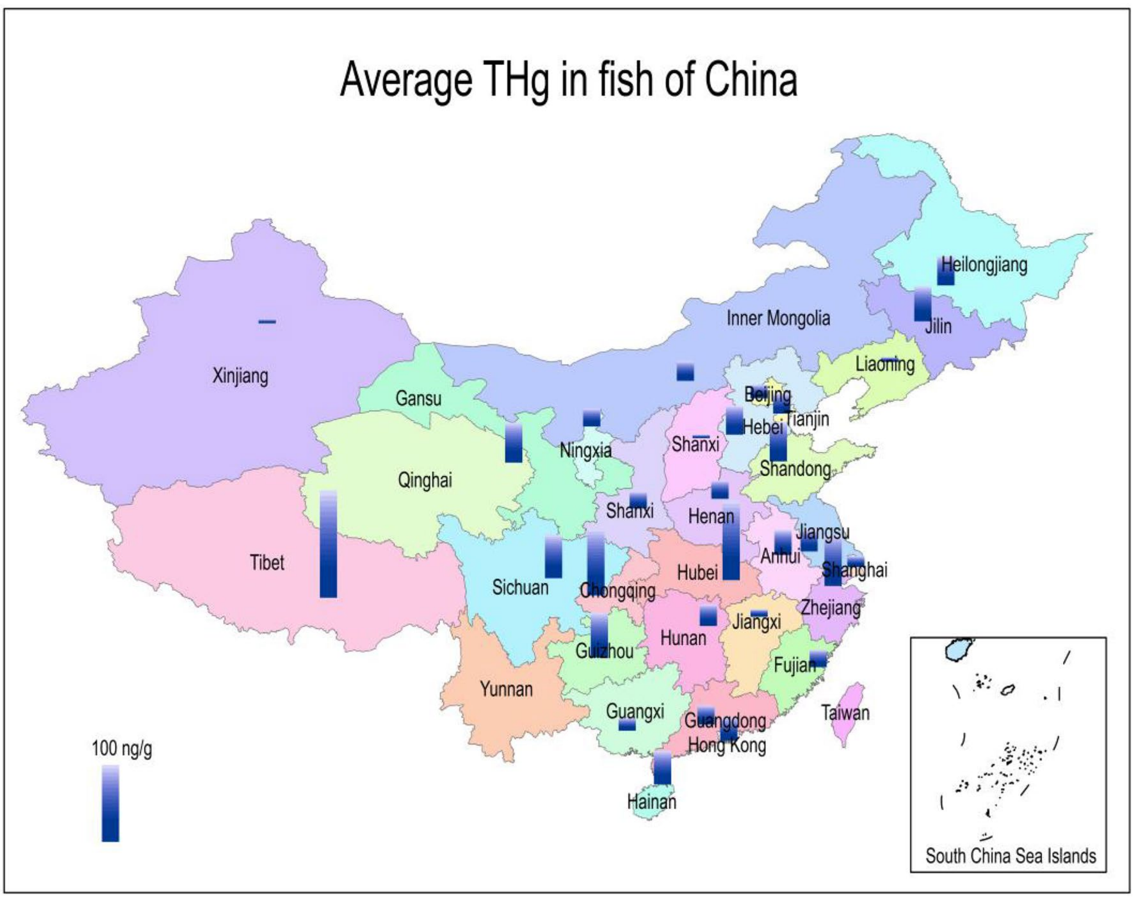

Fig. 1 Map of average THg in fish in China during 2000 to 2018 (See Supplemental information for References in S1-R5)

Canada, and Nordic countries found that fish, mainly from oligotrophic lakes and often from very remote areas, commonly contained high levels of Hg. In the 1990s, elevated concentration levels of Hg were found in fish in some new reservoirs (Hecky et al. 1991; Schetagne et al. 2000). However, because China is a major producer of fish products, it is highly important to consider the status of Hg content in fish products in China.

As a whole, health risk assessments of Hg exposure from edible aquatic products have focused mainly on fish from coastal areas (Cheng and Hu 2012), Hg-contaminated areas (Zhang et al. 2014) and some wild fish from pristine areas (Zhang et al. 2014). Figure 1 summarizes the average THg content in fish from different regions and provinces in China. The results show that the THg in fish is within the average value range of 8.5–274.7.0 ng/g (w/w) with the highest value of 1218 ng/g (ww) (Liu et al. 2014a; Zhang et al. 2014). Generally, MeHg in fish increases with size, weight, age, trophic position and feeding habits (herbivorous < omnivorous < carnivorous). According to references, the concentration of MeHg is very low, only about 49.1 ng/g (ww) with a range from 8.0 to 680.0 ng/g (ww) (Wang et al. 2011; Liu et al. 2012; Feng et al. 2018; Xu et al. 2018). Therefore, the MeHg contents of most fish don't exceed the national limit recommended by the Standardization Administration of China (500 ng/g ww) (GB2762-2012 2013), which indicates that THg in fish in China remains at a relatively safe level or a potential health risk by PDI/PTWI (probable daily intake-PDI, provisional tolerable weekly intake-PTWI) assessment (Cheung et al. 2008; Li et al. 2013; Razavi et al. 2014a, b; Zhang et al. 2018a, b). Nevertheless, marine fish on the consumer market are primarily carnivores, and may contain higher concentrations of Hg just like fishes from populated areas. Therefore, the potential health risks from Hg in fish, it is necessary to develop consumption guidelines for pregnant or nursing women and young children with diets heavy in marine fish in the coastal areas.

Human activities have had considerable effects on the accumulation and transmission of Hg in aquatic food chains. In the past three decades, the impacts of reservoir construction, aquaculture, pollution of water environments, and eutrophication have been of particular concern in China. This aspect of research began receiving more attention as a result of work in reservoirs in the Wujiang watershed since the 2000s. Subsequently, research has been conducted in many aquatic systems, including reservoirs (Li et al. 2015; Razavi et al. 2014a; Wang et al. 2014a, b; Yan et al. 2010), lakes (Yang et al. 2011; Zhu et al. 2012a, b; Zeng 2017), rivers (Shao et al. 2016; Zhu et al. 2012a, b; Zhu et al. 2012a; Zeng 2017), aquaculture farms (Gao et al. 2011; Liang et al. 2016; Shao et al. 2011; Xu and Wang 2017; Zhang et al. 2018a, b), estuaries (Liu et al. 2018a, b, c; Yin et al. 2016), and other coastal areas (Liu et al. 2014a; Pan et al. 2014; Qiu and Wang 2016; Wang et al. 2005) across the country.

The results show that overfishing, aquaculture, eutrophication, and environmental pollution have caused great changes in aquatic food webs. Most results have supported the following conclusions: (1) Overfishing, eutrophication, and aquaculture lead to shorter food chains, lower ages of fish, faster growth rates of fish (Liang et al. 2016), and low bioaccumulation factors (BMFs), about 0.06–0.08 for the correlation slope between $\log_{10}$[Hg] and δ^{15}N‰ (Li et al. 2015; Liu et al. 2012; Pan et al. 2014; Razavi et al. 2014a, b). (2) The number of large carnivorous fish has declined, and the Hg concentration in fish no longer increases with trophic level, size, and age (Feng et al. 2018; Yan et al. 2010). (3) There has been no significant increase in Hg concentration in the fish at the beginning of reservoir construction due to the low organic matter content in flooded soil (Larssen 2010; Li et al. 2013, 2015, 2017; Yu et al. 2013). (4) Hg concentration in fish depends on their feeding habits (Li et al. 2009; Liang et al. 2016; Zhou and Wong 2000), and the proportion of MeHg to THg in fish and other organisms is low, mostly in the range of 30%–60%, due to the low% MeHg in water and sediment (Feng et al. 2018; Li et al. 2018; Liu et al. 2012; Zhu et al. 2012a). The ratio of MeHg to THg in some maricultured-fish is up to 66%–81% (Liang et al. 2011; Liu et al. 2018b). Additionally, in some aquatic systems, such as in Tibet (Zhang et al. 2014) and Qiandao Lake (Razavi et al. 2014a), both Hg-contaminated areas (Xu and Wang 2017), a small part of fish have higher Hg concentration for the possible reason that they feed commercial food pellets with high Hg concentration or are wild fish with higher age. This contamination probably occurs at high trophic position and with low growth rate.

Anthropogenic activities have greatly altered the sources and biogeochemical processes of Hg; however, their impact on the transformation and bioaccumulation of Hg in the aqueous environment remains less clear (Fitzgerald et al. 2007). The use of stable isotopes to solve biogeochemical problems in ecosystem analysis is increasing rapidly because stable isotope data can contribute to both source–sink (tracer) and process information (Peterson and Fry 1987). Therefore, stable isotopes are often used to trace the source, process, and fate of pollutants or other matters in air, water, sediment, soil and organisms. For instance, the elements C, N, and Hg all have more than one isotope, and the isotopic compositions of natural materials can be measured with great precision with a mass spectrometer. Isotopic compositions change in predictable ways as elements cycle through the biosphere.

The stable isotopes of nitrogen (expressed as δ^{15}N) and carbon (δ^{13}C) have been used to estimate trophic positions of consumers and carbon flows to consumers in food webs (Liu et al. 2012; Liu et al. 2018a, b, c; Post 2002; Zhang et al. 2014). However, it has been found that phytoplankton, as the base of the basic food chain, sometimes have much higher $\delta^{15}N$ values than fish, which suggests that the discharge of nitrogen-containing wastewater might have an effect on accurately identifying the nutrient position of organisms in the food chain using only nitrogen isotopes.

Stable isotopic compositions of Hg in aquatic organisms have been used to trace the source and transport of Hg in the environment. The results of Yin et al. showed variations in mass-independent fractionation ($\Delta^{199}Hg$: $+0.05 \pm 0.10$‰ to $+0.59 \pm 0.30$‰) with a $\Delta^{199}Hg/\Delta^{201}Hg$ of ~ 1.26, suggesting that aqueous MeHg underwent photodegradation prior to incorporation into the food chain. The significant differences of $\Delta^{199}Hg$ values among different feeding habit fish indicated that it may have incorporated MeHg with various degrees of photo-demethylation (Yin et al. 2016). Xu and Wang (2017) tracked Hg exposure from different food sources in marine-caged fish in southern China using $\delta^{202}Hg$ and $\Delta^{199}Hg$ values. According to the slope of $\Delta^{202}Hg/\Delta^{199}Hg$ in fish, the study of Hua et al. indicated the mass independent fractionation in fish from the Xiaolangdi Reservoir subject to the photodegradation of monomethylmercury (Hua et al. 2016). In addition, his results inferred the MeHg source of fish by the correlation of MeHg concentrations and % MeHg in the fish with $\delta^{202}Hg$ and $\Delta^{199}Hg$. The results show that Hg isotopes, especially the Hg-MIF (mass-independent isotope fractionation) can be a powerful tool for revealing the exposure pathways and geochemical behaviors of MeHg in aquatic food webs (Hua et al. 2016; Liu et al. 2018a; Wang et al. 2013; Xu and Wang 2017; Xu et al. 2016; Yin et al. 2016). However, this field remains in the development stage and requires further research due to the uncertainty of some theories and methods.

Areas of Limited Research

Compared with research on fish, research on the base of the food chain in China is relatively weak because risk to human health is not directly involved. The following provides a summary of recent research progress on bioaccumulation of Hg in plankton and benthic organisms in China.

Study of the concentration of Hg in phytoplankton in China is at a moderate level compared with such studies worldwide, and studies in China are still being carried out mostly in Guizhou (Table 1). As revealed by these studies, the high Hg content of phytoplankton in Guizhou may be because the high Hg geological background value. Because the natural THg is higher, the Hg content in phytoplankton is also higher. Correspondingly, phytoplankton Hg content in Daya Bay in Guangdong is low.

Zooplankton is one of the largest aquatic communities in lakes and reservoirs and is also one of the important food sources for aquatic organisms. The important position of zooplankton in the aquatic food chain has led to study of its concentration of heavy metals, including Hg. Hg pollutants in water can be understood in the contexts of the food chain and the aquatic environment. However, there have been few studies on the concentrations of heavy metals, including Hg, in zooplankton in China. Guizhou, located in southwest China, is within one of the three major Hg mineralization zones in the Pacific Rim. The region has abundant Hg resources, mining of Hg, and a high natural Hg geological

Table 1 THg concentrations in phytoplankton (64–112 μm) in different areas from selected published data

Country	Geographical area	Concentration (ng/g dw)	References
Brazil	Tapajos River	66 (n = 8)	(Roulet et al. 2000)
Canada	La Grande Hydroelectric Complex	87 ± 20 (n = 13)	(Schetagne et al. 2000)
Colombia	Grande Marsh, Cauca River Basin	520 ± 30 (n = 9)	(Marrugo-Negrete et al. 2008)
India	Husain Sagar Lake	43 (n = 5)	(Prahalad and Seenayya 1988)
China	Baihua Reservoir, Guizhou	0–227 (n = 21)	(Li et al. 2014; Deng 2016)
	Caohai Wetland, Guizhou	135 (n = 17)	(Zeng 2017)
	Guangdong, Daya Bay	54 (n = 4)	(Lin et al. 2013)
	Taihu Lake, Jiangsu	33.0 ± 11.0 (n = 6)	(Wang et al. 2012)
		680–1050 (n = 102)	(Wang et al. 2015)
	Xiaoguan Reservoir, Guizhou	24.43–42.34 (n = 24)	(Deng 2016)
	Hongfeng Reservoir, Guizhou	21.63–79.48 (n = 20)	(Deng 2016)
	River, Hunan	4.0–15.0 (n = 5)	(Lu et al. 2016)

background, which makes Guizhou one of the regions with serious Hg pollution. Therefore, the few systematic studies currently available on the concentration of heavy metals in zooplankton have been concentrated in Guizhou and a few other locations (Li et al. 2014; Zeng 2017). According to the available data, the median value of Hg in zooplankton is relatively lower than at other sites in the world (Table 2).

The study of Hg content in benthic organisms in China is still very limited with large research gaps, and published literature on benthic THg data are available in only about nine provinces (Fig. 2, S1-R5). China has a vast marine area and abundant inland water resources, so many types of aquatic products are output. If high Hg concentrations exist in these edible aquatic products, it will cause great harm to humans. Therefore, the research objects of Hg enrichment in benthic animals in China are mostly listed aquatic products such as shrimp, crab, and shellfish, and the research sites are mostly coastal provinces such as Guangdong and Jiangsu.

Table 2 THg in zooplankton (112–500 µm) in different areas from selected published data

Country	Geographical area	Concentration (ng/g dw)	References
Canada	Northern Quebec	75–310 (n = 19)	(Kainz and Lucotte 2002)
	La Grande Hydroelectric Complex	132 ± 10 (n = 13)	(Schetagne et al. 2000)
	Canadian lakes	25–377 (n = 3)	(Li 2018)
Colombia	Grande Marsh, Cauca River Basin	940 ± 50 (n = 9)	(Schetagne et al. 2000)
India	Husain Sagar Lake	68 (n = 5)	(Prahalad and Seenayya 1988)
USA	Vermont and New Hampshire lakes	16–335 (n = 20)	(Kamman et al. 2004)
USA–Canada	Lake Superior	20–130 (n = 9)	(Lucotte et al. 1999)
China	Baihua Reservoir, Guizhou	186 (n = 16)	(Xu and Wang 2017)
	Caohai Wetland, Guizhou	181.96 (n = 17)	(Zeng 2017)
	Reservoirs and wetlands, Guizhou	152.59 (n = 30)	(Long et al. 2018)
	Taiwan, Okinawa Trough	80–1090 (n = 8)	(Xu 2014)

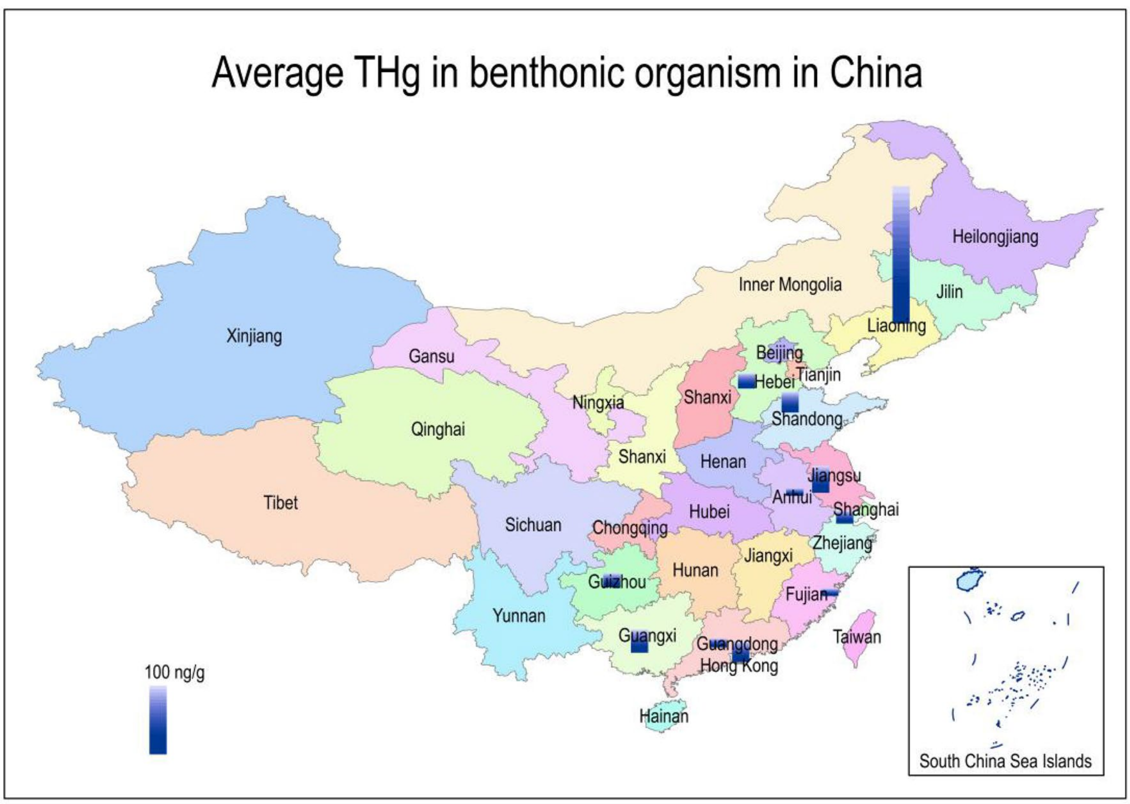

Fig. 2 Average THg in benthonic organisms in China

The province with the highest Hg content in benthic animals has levels as high as 393.92 ng/g at Deer Island, northern Yellow Sea (Lin et al. 2013). Provinces with lower concentrations are located mostly in East China: Shandong, about 21.24 ng/g (Cui et al. 2011; Zhang and Zhang 2014), Fujian, about 22.5 ng/g (Lin et al. 2013), and Guangdong (Qiu and Wang 2016). A comparison of national and international limits for Hg in marine products shows that the Hg in benthos is lower than the national Hg content limit. In addition, crab has higher Hg content than other benthos (Fan et al. 2012).

As mentioned above, studies on Hg bioaccumulation in the basic food chain have been carried out only in a few areas. Meanwhile, there are some problems for sampling methods of base food chain samples. For instance, those 64–112 μm phytoplankton and 112–500 μm zooplankton may be too large to be fed by these fishes in higher trophic position. Therefore, this could affect us to draw accurate conclusions on Hg transport and bioaccumulation in the food chain. Additionally, the study on Hg bioaccumulation mechanisms in food chain is very lack till now. For instance, it is still unclear what factors affect the transformation and accumulation efficiency of Hg in the food chain. What is the bioaccumulation and metabolic mechanism of Hg in different fish species? There are few studies to investigate these similar questions which is in vivo Hg methylation and demethylation in freshwater tilapia. Results indicated the inter-organ transportation of MeHg from liver toward muscle and net methylation in vivo (Wang et al. 2013).

Perspectives on Future Research

As mentioned above, most of the research has focused on the bioaccumulation of Hg in the aquatic environment. But there are only a few detailed studies on the mechanism of Hg bioaccumulation. Additionally, although many studies on the bioaccumulation of Hg in benthic organisms and plankton have already been conducted in the world, the research published in this field in China is quite lacking, with fewer detailed studies of mechanism (Wang et al. 2013, 2017).

In recent years, the ecosystem environment in China has improved with implementation of environmental protection policies such as no-fishing policies and policies pertaining to the elimination of eutrophication of water bodies, which should be conducive to the restoration of ecosystems and the intact food chain. However, this would increase Hg concentration level in fish in the future. Recent international studies have shown that the Hg levels in fish are increasing in some areas. Therefore, we need to pay more attention to the transport and bioaccumulation of Hg in the basic food chain, such as in benthic organisms and plankton, and also pay continuous attention to ecosystem change for an extended period of time. Secondly, multi-pollution exposure is more toxic than single-contamination exposure for aquatic organisms. Therefore, we should pay attention to multi-contaminant joint exposure study of the food chain in some polluted areas and also conduct research on the mechanism of joint exposure through laboratory simulation, which protects both human health and ecological health. Additionally, establishment and application of a biological transport model of Hg in China's water environment is needed.

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