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Original Research Article

Decreasing mercury levels in consumer fish over the three decades of increasing mercury emissions in China

Hua Zhang [a,b,*], Wenxiong Wang [c], Chejen Lin [d], Xinbin Feng [a], Jianbo Shi [e,f], Guibin Jiang [e], Thorjørn Larssen [b]

[a] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China
[b] Norwegian Institute for Water Research, Økernveien 94, 0579 Oslo, Norway
[c] School of Energy and Environment and State Key Laboratory of Marine Pollution, City University of Hong Kong, Kowloon, Hong Kong, 999077, China
[d] Center for Advances in Water and Air Quality, Lamar University, Beaumont, TX, USA
[e] State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, China
[f] State Environmental Protection Key Laboratory of Source Apportionment and Control of Aquatic Pollution, School of Environmental Studies, China University of Geosciences, Wuhan, 430074, China

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ABSTRACT

Fish consumption is the primary dietary route of human exposure to methylmercury. It has been well documented that elevated mercury concentration in fish in North America and Europe is linked to anthropogenic mercury emissions. China is the world's largest producer, consumer, and emitter of mercury, as well as the world's largest commercial fish producer and consumer. Although mercury pollution in fish in China is currently receiving much attention worldwide, its status remains largely unknown. Here, we conducted a meta-analysis on total mercury concentrations in marine and freshwater fish samples, covering 35,464 samples collected in China over the past 30 years. It is found that, opposite to the increasing emission and documented mercury contamination events, mercury levels in fish have gradually decreased in China over the past 30 years. The results were in sharp contrast to those found in North America and Europe. The mercury concentrations in fish were significantly anticorrelated with the fish catch and fish aquaculture and were inverse to trophic levels. Overfishing and the short lifecycle of aquaculture fish, both reducing the trophic level and the duration of mercury accumulation, were the most likely causes leading to the decline of mercury concentrations found in fish in China.

1. Introduction

Mercury (Hg) pollution is of great international concern because of its unique properties such as global transport, persistence, bioaccumulation, high toxicity, and its negative effects on food safety [1,2]. Besides natural processes, human activities have discharged a large amount of Hg into the earth's supergene environments. It was estimated that in merely 100 years (1850–1950), the cumulative Hg emission by countries and regions from North America and Europe exceeded 200,000 Mg [3], which led to Hg contents in fish in thousands of freshwater lakes in these countries and regions exceeding limits advised for human consumption [4]. According to summarized studies by Streets et al. [3], Tian et al. [5], and Wu et al. [6], China's atmospheric cumulative Hg emissions were approximately 16,000 Mg from 1850 to 2014, accounting for only roughly 6% of the global cumulative Hg emissions (far lower than the contribution of >50% from North America and Europe in the same period). It is noteworthy that these Hg emissions in China mainly occurred after China's reform and opening-up since the 1980s.

Fish are considered to be the key receptors of long-range transboundary air pollution (LRTAP) of Hg [4]. Because of the prevalent Hg emissions from coal combustion, nonferrous metal smelting (approximately 80% of the total emission), and other anthropogenic sources (such as cement production, Hg mining, biofuel combustion, etc.), China is now considered to be one of the most severe airborne mercury-polluted regions worldwide, where the potential Hg pollution of fish has increasingly attracted great public attention, especially after the signing and entry into force of the Minamata Convention in 2017 [5–8]. The atmospheric Hg levels (1.6–34 ng/m^3) and Hg deposition fluxes (6.0–152

* Corresponding author. State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, 550081, China. zhanghua@mail.gyig.ac.cn (H. Zhang).

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μg/(m^2·y)) in China are generally 2–10 times higher than those in North America and Europe [5,6]. In recent years, China has actively carried out intensive air pollution control measures (e.g., ultralow emission renovations of coal-fired power plants, elimination of small coal combustion boilers in industrial activities and domestic heating), which has substantially decreased the anthropogenic Hg emissions (e.g., 5.5% per year) [7]. However, the superimposition of over 200 years of historical Hg emissions by countries and regions in Europe and North America and 40 years of Hg emissions in China has inevitably led to relatively elevated Hg concentrations in the environmental media in China. According to the limited reports available, a considerable part of the average Hg concentrations in the freshwater bodies (0.001–0.69 μg/L), marine waters bodies (0.001–1.1 μg/L), freshwater sediments (0.002–0.88 mg/kg, dry weight–dw), and marine sediments (0.027–0.9 mg/kg dw) in China exceeded the Class I Chinese National Standards (0.05 μg/L and 0.2 mg/kg for inland waters and marine sediments, respectively) (Supplementary Tables S6–S7). It must be emphasized here that studies on Hg pollution in Chinese waters have not been very systematic in general. Most studies on waters have focused on hotspot areas with significant pollution occurrence [8] and tended to represent those environmental samples in polluted sites, but to some extent still could reflect the severe Hg contamination in China.

According to the Bulletin of Marine Environmental Quality of China [9], the total amount of Hg pollutants directly discharged into the coastal seawaters in 2020 was approximately 382.2 kg. Heavily Hg-contaminated marine sediments have also been reported at multiple coastal locations in China, with the highest Hg level up to 17 mg/kg, far exceeding the Chinese National Standard of 0.2 mg/kg for marine sediments (Supplementary Table S7). Elevated Hg levels were also extensively reported at certain locations in China, such as estuaries and large cities (Supplementary Table S6), which are much higher than the concentrations reported for similar environmental settings in the United States (US) and Europe (<0.004 μg/L) [3,9].

One particular concern on Hg (especially the methylated Hg, MeHg) in the aquatic environment is its trophic transfer and biomagnification in fish [10]. The consumption of fish contaminated by MeHg is recognized as the primary route of human Hg exposure [1,7]. MeHg levels in fish exceeding the reference limits for human consumption have been reported for fish produced in the remote aquatic ecosystems of North America and Europe [4,11]. Increased MeHg exposure due to the increasing contribution of fish to the global dietary protein intake is a severe human health issue. Globally, fish provides over 2.9 billion people with approximately 20% of their average per capita animal protein intake [12]. In 2010, for example, fish accounted for 16.7% of the global intake of animal protein and 6.5% of all protein consumed. The quantity of fish caught in China was approximately 4 million Mg in the 1980s, 9 million Mg in the 1990s, and 11 million Mg in the 2000s, with the majority (90%) from China's coastal waters (The Food and Agriculture Organization of the United Nations, FAO; http://www.fao.org/fishery). China is now the world's largest commercial fish producer and consumer, with a total annual fish production of up to 34 million Mg in 2010, accounting for >30% of the total global production [12].

Despite the widespread Hg contamination events reported in China, a comprehensive, integrated national survey on Hg concentrations in fish remains absent. Given the extensive geographical range of China, the synthesis of existing datasets remains a challenge, and the present state of Hg contamination in fish in China is poorly understood. A longstanding question is whether the large Hg emission quantity in China has a direct impact on Hg levels in fish. In the present study, we address this very important question through a meta-analysis of data covering over 35,000 fish samples collected comprehensively from the entire aquatic environment in China over the past three decades. In addition, the temporal and spatial variations of Hg concentrations found in these fish samples were analyzed in an attempt to reveal the underlying causes of Hg contamination status in fish of China.

2. Materials and methods

We first established a national database of total Hg concentrations from marine and freshwater fish samples collected in China (N = 35,464; Supplementary Tables S1–S3). These datasets, dating back to the measurements made in 1980, were obtained from 34 different administrative regions. The data and the temporal variation analyses were presented statistically and spatially on maps. We hypothesized that Hg concentrations in fish are closely related to anthropogenic Hg emissions and fishery activities. Data of historical fishery, trophic diversity, and fish body size were also analyzed to establish their relationships with the Hg concentration trends in fish.

In addition to peer-reviewed literature indexed in the global database, China Knowledge Resource Integrated Database (CNKI, www.cnki.net), and the VIP Periodical Resource Integration Services platform (VIP, http://lib.cqvip.com) (the CNKI and VIP include most papers published in China's scientific journals and PhD/master's theses) were also studied. To obtain data with higher analytical quality, we only analyzed the data collected after 1980, when more reliable quantification methods and quality control measures for Hg analysis had been developed. For comparison, total Hg concentration data reported for muscle tissue of fish samples in 1980–2014 were selected. In total, 203 publications (75 in English, 127 in Chinese, and 1 in Japanese) were utilized in the preparation of the final database. The data on Hg levels in fish muscles, number of fish species/samples, sampling year, study location, fish type, feeding habits, and Hg analysis methods were compiled from the available original literature, covering a total of 35,464 fish samples. The statistical analyses were based on Hg data from different sets of fish. We mainly focused on the trend of the entire fish Hg level in the whole country rather than specific basins or areas. All data were combined, instead of being separated by different species, to reduce the uncertainty of analysis and ensure enough samples. All fish Hg data was presented on a wet weight (ww) basis. The aquaculture and catch quantity of marine fish, freshwater fish, and diadromous fish in China were obtained from the FAO. The changes in fish trophic level indices and mean maximum fish body length data in the coastal marine ecosystem in China were available from the Sea Around Us Project (http://www.seaaroundus.org). All statistical analyses (e.g., regression analyses, ANOVA, etc.) were performed using IBM SPSS Statistics 21. A *P*-value less than 0.05 is considered to be significant.

3. Results and discussion

3.1. Temporal variations of fish Hg in China

To our surprise, Hg levels in fish samples collected in China showed a significant decreasing trend from 1980 to 2014 (Fig. 1A), despite a continuous increase in atmospheric Hg release during the same period (Fig. 1B). The mean Hg concentration in fish decreased remarkably by 70% from 1980 to 2014 ($r^2 = 0.79$, $P < 0.01$). Using the 5-year running weighted averages, the Hg levels in fish decreased from 0.196 mg/kg (N = 2153) during 1980–1985 to 0.054 mg/kg (N = 4198) during 2011–2014. The average 5-year rate of decrease was 0.024 mg/kg. It should be noted that the decreasing trend was significant for freshwater fish samples ($r^2 = 0.67$; $P < 0.01$) but not for marine fish samples ($P > 0.05$). However, as mentioned earlier, we mainly focused on the trend of all fish samples. All data in the present study were combined and analyzed, instead of being separated by fish species, to reduce the uncertainty of analysis and ensure enough samples, unless otherwise stated.

During the same period, anthropogenic emissions of Hg into the atmosphere continuously increased in China. The increase has been particularly drastic since 2002, with the annual emission growth rate as high as 10%, resulting in approximately 500–800 Mg anthropogenic Hg emitted in the last few years [5,6]. The average 5-year Hg emission in the past three decades increased by 70 Mg (Fig. 1B). The Hg emission trend

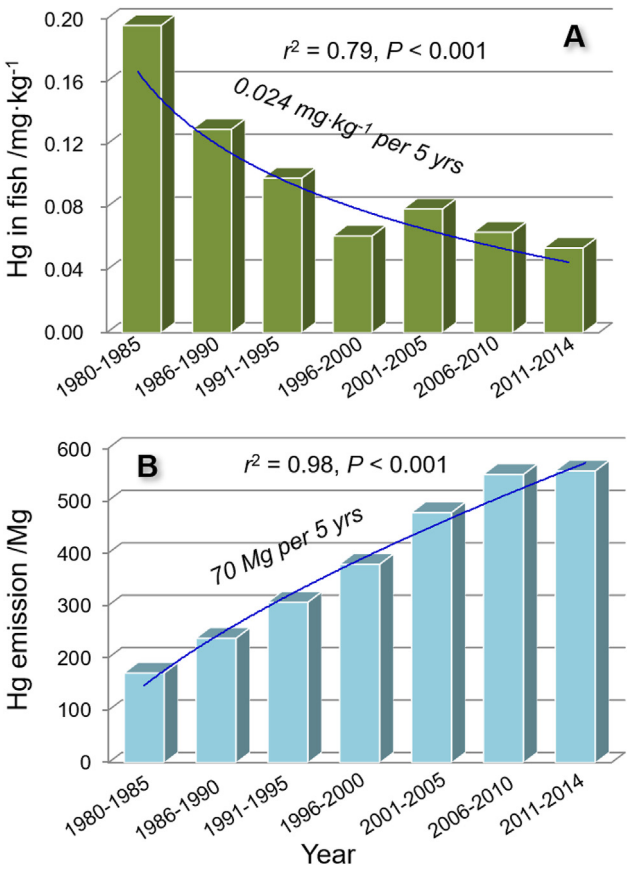

Fig. 1. Temporal change of fish Hg compared with Hg emission in China from 1980 to 2014. (A) The weighted averages of total Hg levels in fish samples (mg/kg; ww) in China (as the fish sampling time in some original documents is multi-year data, the data were summarized with every 5 yrs as a time span); (B) Anthropogenic Hg emission in China (data were originated from Wu et al., 2016 [6]) .

in China is different from those in North America and Europe, where atmospheric Hg emissions have declined since 1990 due to the co-benefits of Hg emission reduction brought by the implementation of air pollution control measures [13–15].

Previous studies in both United States and Europe showed that elevated Hg concentrations in fish were closely linked to Hg emissions as a result of anthropogenic activities [1,13–15]. However, Hg levels in fish samples in China gradually decreased by over 70% from 1980 to 2014, which was in strong contrast with the increasing Hg emissions in China in recent decades (Fig. 1).

Hg concentrations in fish samples collected in China were generally low between 1980 and 2014, when the weighted averages of Hg in fish in China were 0.063 mg/kg and 0.114 mg/kg, respectively, for marine fish (with the number of species >370) and freshwater fish (with the number of species >230) (Supplementary Tables S3–S4). These averages were usually 2–12 times lower than those recorded in North America and Europe (commonly averaged at 0.200–0.800 mg/kg) [4,13–16]. Indeed, despite reports of a declining trend of fish Hg levels in Boreal and Sub-arctic Fennoscandia [4], several studies have observed that the Hg levels in fish in North America and Europe appear to be increasing recently, although the local Hg releases have declined over the last few decades [13–16].

3.2. Spatial distributions of fish Hg in China

The spatial distribution of recent fish Hg levels in China from 1980 to 2014 is presented in Fig. 2, which did not share the same variation pattern as the spatial distribution characteristics of atmospheric Hg emissions in China (Supplementary Fig. S1). The weighted average of Hg concentrations in fish in China from 2001 to 2014 (0.066 mg/kg; N = 19,726) was significantly (P < 0.001) lower than that from 1980 to 2000 (0.118 mg/kg; N = 15,738) (Supplementary Table S3). The Hg levels in both periods were well below the Chinese National Standard (0.500 mg/kg), suggesting that the high Hg release in China in the past few decades did not result in a direct bearing on Hg in fish.

The weighted average of Hg levels in freshwater fish from 34 administrative regions from 1980 to 2014 showed greater spatial variability from 0.008 to 0.181 mg/kg (Fig. 2). The fish Hg concentrations were higher in inland regions (e.g., Tibet, Xinjiang, Qinghai, and Shaanxi provinces) and lower in the southeastern regions adjacent to coastal regions (e.g., Hunan, Jiangxi, Anhui, and Henan provinces, which are the main areas for aquatic products; Fig. 2). This difference might be related to the longer lifespan of fish due to less fish consumption by residents in inland regions (less than 0.700 kg/yr per capita; thereby less fishing pressure) than that in these southeastern regions adjacent to coastal regions (5–18 kg/yr per capita) (Fig. 2). For example, many Tibetans do not eat fish because of their religious beliefs, and elevated Hg levels as high as 1.220 mg/kg were observed from local wild fish samples (Supplementary Table S3), which may be due to the longer Hg enrichment process in fish (with a longer lifespan reaching 10–30 years) than that in other regions in China (usually with an average lifespan of 1–3 years; Supplementary Table S5). However, even the highest Hg levels in individual fish samples in inland areas (e.g., the Huaihe Basin, the Taihu Basin, and the Zhejiang-Fujian-Guangdong Area) were generally well below 0.500 mg/kg of the Chinese National Standard.

The spatial distribution of all fish Hg data from 1980 to 2014 for the four Chinese coastal waters, i.e., the Bohai Sea, the Yellow Sea, the East China Sea, and the South China Sea; and rivers/lakes/reservoirs in different main basins, i.e., the Yangtze River Basin, the Yellow River Basin, the Songliao River Basin, the Huaihe Basin, the Haihe Basin, the Taihu Lake Basin, the Pearl River Basin, and the rest areas, are presented in detail in Supplementary Figs. S4–S10. The Hg levels in fish samples from the four Chinese coastal waters from 1980 to 2014 were comparatively low, with a weighted mean of 0.115 mg/kg in the Yellow Sea, 0.072 mg/kg in the South China Sea, 0.068 mg/kg in the East China Sea, and 0.053 mg/kg in the Bohai Sea (Fig. 2).

It should be noted that, although a vast majority of studies reported low fish total Hg concentrations (commonly far below 0.500 mg/kg, the Chinese National Standard limit for Hg in fish), elevated Hg levels were reported for marine fish samples collected in Jinzhou from the Bohai Sea (0.330 mg/kg), Zhoushan, Zhejiang from the East China Sea (0.260 mg/kg), and Hong Kong from the South China Sea (0.270 mg/kg), with concentrations in predatory fish samples reaching up to 0.510–0.660 mg/kg (Supplementary Table S3).

Elevated Hg levels were also found in selected freshwater fish samples from the Second Songhua River (0.270 mg/kg) and Liaoning coastal sites (0.400 mg/kg) of the Songliao Basin, Qingtongxia (0.310 mg/kg) of the Yellow River Basin, Wanshan Hg mining area (0.700 mg/kg) of the Yangtze River Basin (Supplementary Figs. S5–S8) and remote rivers/reservoirs in Tibet (0.017–1.218 mg/kg; Supplementary Table S3). Interestingly, low fish Hg (<0.100 mg/kg) concentrations were consistently observed from heavily Hg-contaminated sites such as Baihua Reservoirs, where averaged Hg levels in sediments were elevated up to 3–13 mg/kg (Supplementary Table S3).

3.3. Potential mechanisms for Hg anomaly in China

Bioaccumulation of Hg in fish is complicated by Hg speciation in the aquatic environment and the trophic level of fish. Dietary exposure is considered one of the dominant sources of Hg accumulation. Enrichment of Hg while fish grow older and biomass dilution caused by growing phytoplankton biomass are two predominant characteristics of Hg bioaccumulation in fish. As demonstrated in recent studies, the two

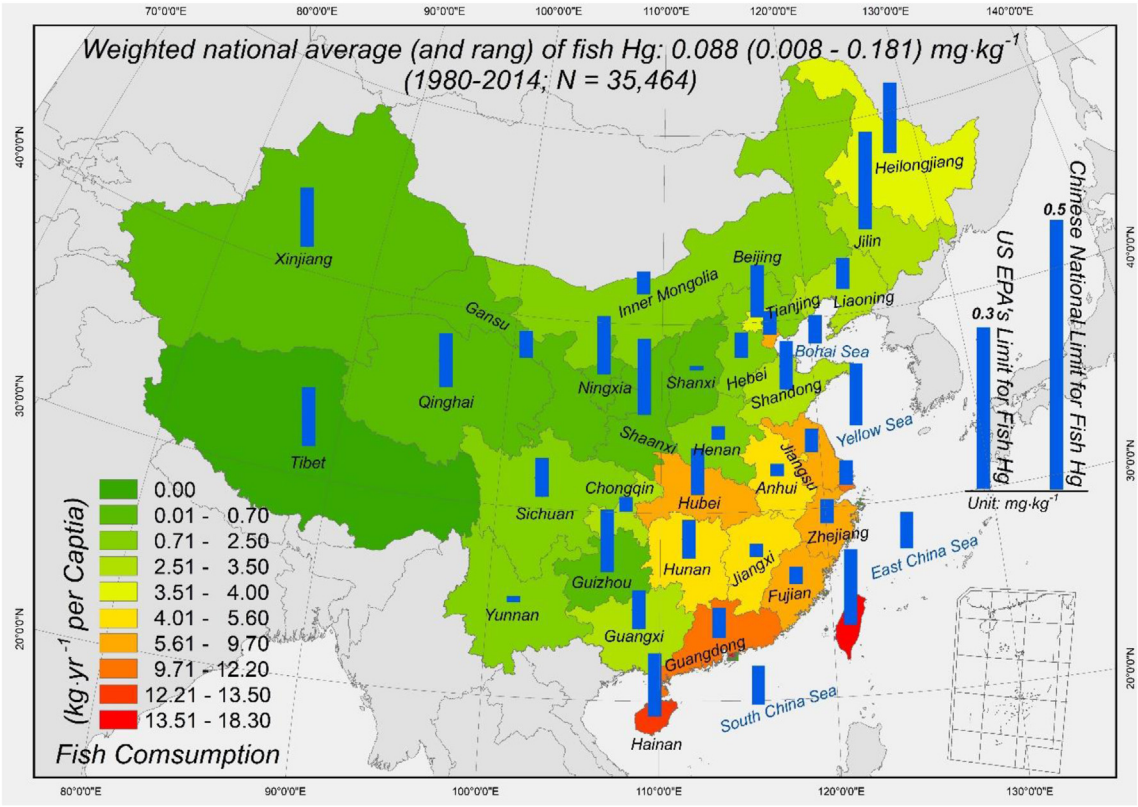

Fig. 2. Spatial variation of total Hg concentrations in fish in China from 1980 to 2014 (mg/kg; blue bar) and the corresponding fish consumption rate of general populations (indicated by different background colors). More detailed information is provided in Supplementary Table S2 of the Supplementary Material.

characteristics significantly affect Hg bioaccumulation, interplayed by fish growth [10]. Such trophic physiology may therefore overwhelm the increasing Hg input into the aquatic environments, resulting in a Hg bioaccumulation lower than expected in freshwater and marine fish. Hg accumulation in fish is closely related to fish size and trophic status. It is hypothesized that the identified decrease in Hg accumulation over the past 30 years in China was caused by trophic level changes.

Long-term aggressive overfishing has damaged the ecological balance of aquatic ecosystems in China [8,12,17,18]. Fig. 3A shows that both aquaculture fish production (class 1–3) and wild fish catch (class 4–6) increased dramatically since 1950 (wild fish catch was 0.79 million Mg in 1950 and peaked at 11 million Mg in 1998). After 2000, the wild fish catch remained relatively stable. In contrast, aquaculture fish production continued to increase due to population growth. The aggressive overfishing activities have caused the depletion of inshore fishery resources in China. This is evidenced by the declined growth of wild fish catch from 6.2% during 1950–1998 to 0.15% during 1999–2010. Overfishing also eliminated larger/older and predatory fish populations and decreased fish trophic level, body size (Fig. 3B), and lifespan. Take the decrease in the trophic level index and body size of marine fish since 1978 as an example (Fig. 3B). The fish trophic level in Chinese coastal waters has reduced from 3.6–3.9 during 1950–1978 to 3.3–3.6 after 1978. Similarly, corresponding fish body length decreased from 90–130 cm during 1950–1978 to 70–90 cm after 1978 (Fig. 3B). Such results were consistent with the long-term ecosystem surveys in the Bohai Sea from 1959 to 2008 showing that large-sized species with high economic value had been replaced by the short-lived, low-trophic-level planktivorous pelagic species since the 1980s [18]. The biomass of fishery resources in the Bohai Sea declined continuously from 423.6 kg/(haul·h) in 1959 to 164.6 kg/(haul·h) in 1982, 37.7 kg/(haul·h) in 1993, and less than 8 kg/(haul·h) during 1998–2008 [18]. The natural lifespan of fish is typically 2–20 years [19]. However, most wild marine and freshwater fish caught in China have a lifespan of 1–3 years, except in remote ecosystems (Fig. 4). Because of the shortened lifespan, low trophic level, and reduced body size, most commercial fish in China are consumed before they have lived sufficiently long for bioaccumulation of Hg. It should be mentioned here that overfishing is being regulated in China to preserve the balance of the aquatic ecosystems, which could offset its co-benefit on fish Hg control.

Furthermore, farming of freshwater and marine fish has dramatically increased in China over the past three decades due to increasing demand for fish and depleting wild fishery resources (Fig. 3A). Farmed fish accounted for >60% of total fish production in 2010 (Fig. 3A). The feed of farmed fish primarily contains crops (e.g., corn, wheat) with very low levels of Hg, and therefore, effectively reduces Hg biomagnification and concentrations in farmed fish [7]. An examination of our database (2001–2014) showed that the Hg levels in farmed freshwater fish (0.049 mg/kg) were much lower than those in wild fish (0.084 mg/kg), although the Hg levels in farmed marine fish (0.078 mg/kg) were comparable to those in the wild marine catch (0.067 mg/kg).

Besides, the number of water bodies classified as eutrophic has increased dramatically in China during the past three decades [17]. The incidence and affected areas of red tides (harmful algae blooms) in the coastal waters of China have increased dramatically since the 1980s (Fig. 3C). Long-term monitoring in Bohai Bay indicated that the biomass of zooplankton and phytoplankton displayed an increasing trend from 1959 to 2006 [18]. Approximately 90.8% of lakes and reservoirs in China in 2020 were classified as mesotrophic (61.8%) and mild/moderate/severe eutrophication (29%), and in summer, up to 45,330 km^2 of coastal areas were classified as eutrophic [12,17]. Eutrophic

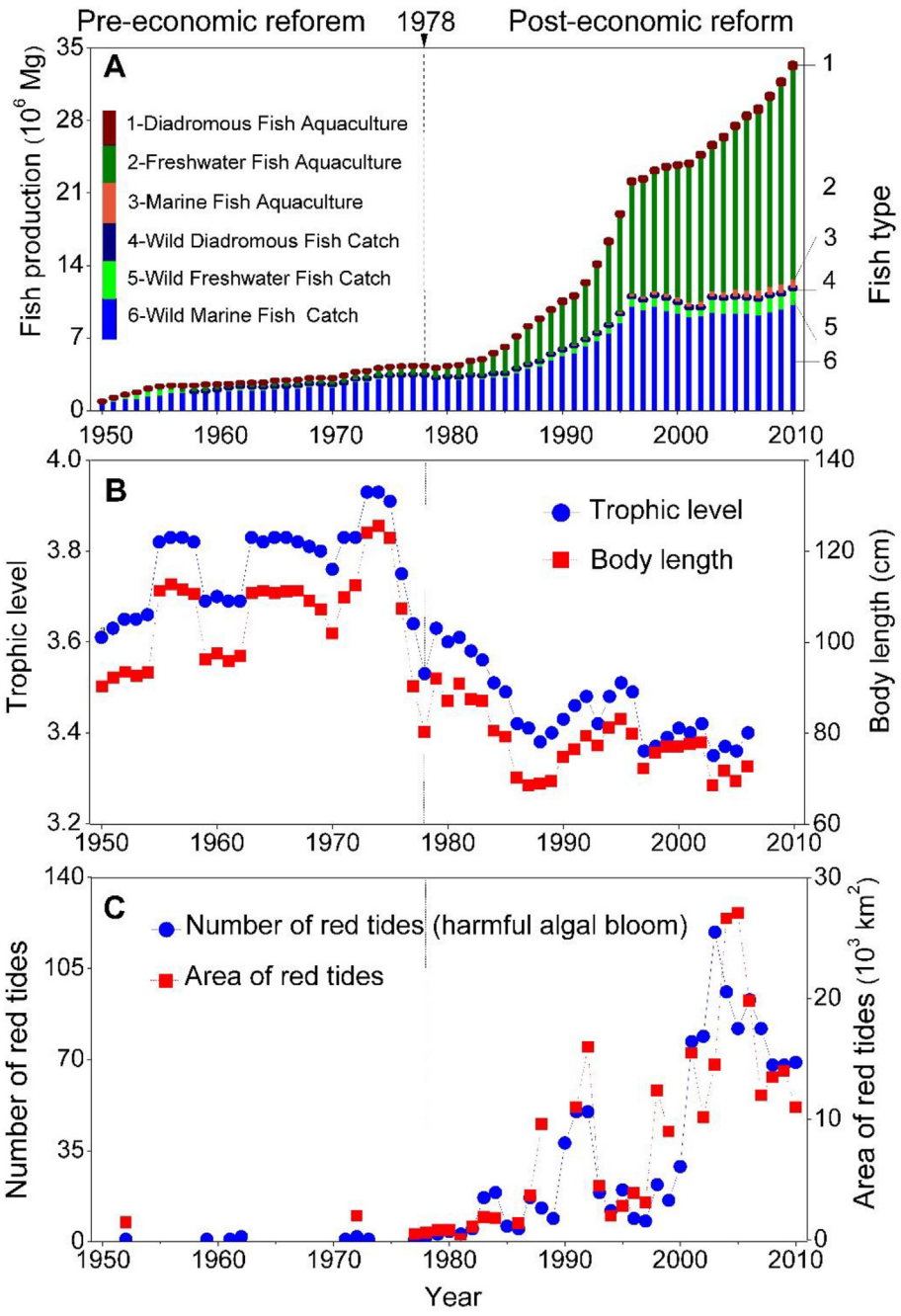

Fig. 3. Historical trends of trophic diversity, fish body size, red tides (harmful algae blooms) in coastal ecosystems, and fish production in China. Aquaculture and catch data of marine fish, freshwater fish, and diadromous fish (A) were developed by the Food and agriculture organization of the United Nations (http://www.fao.org/fishery). The change in fish trophic level indices (the trophic level of primary producers set as 1) and mean body length data (B) were developed by the Sea Around Us Project (http://www.seaaroundus.org). Annual occurrence and area of major coastal algae blooms (red tides) data (C) were extracted from the Marine Bulletins of the State Oceanic Administration of China (http://www.soa.gov.cn/zwgk/hygb) and references [20].

environments may enable fish to grow faster at limited trophic levels, restricting Hg concentration in the fish through biomass dilution. In addition, nutrient enrichment can diminish Hg bioaccumulation in phytoplankton and zooplankton, thus reducing Hg levels in fish [10].

The decreased fish trophic level leading to reduced fish size and decreased Hg concentrations in fish over time in China is supported by the data collected in this study. For example, Hg concentrations in freshwater and marine fish were significantly and negatively correlated with the fish catch ($r^2 = 0.90$, $P < 0.01$) and fish aquaculture ($r^2 = 0.74$, $P < 0.05$; Supplementary Fig. S11) from 1980 to 2010. The Hg concentrations in fish were also significantly correlated with the trophic level ($r^2 = 0.72$, $P < 0.05$) but anticorrelated with the incidence (and area) of red tides ($r^2 = 0.34$–0.52, $P < 0.05$). A multiple regression analysis, including the potential factors, revealed that fish catch ($Catch_F$) is the only variable significantly controlling the Hg concentration in fish (Hg_F), with the ability to predict the value of approximately 90% of the observed Hg in fish ("$Hg_F = -0.002Catch_F + 0.223$"; adjusted $R^2 = 0.88$, $P < 0.01$). These results demonstrated that the decreasing fish Hg levels over the past 30 years in China could be explained by the lower trophic levels, a reduced body size, and a shorter lifespan largely driven by the increasing fishing pressure, pervasive eutrophication, and extensive fish aquaculture. Of these factors, overfishing may be the most critical.

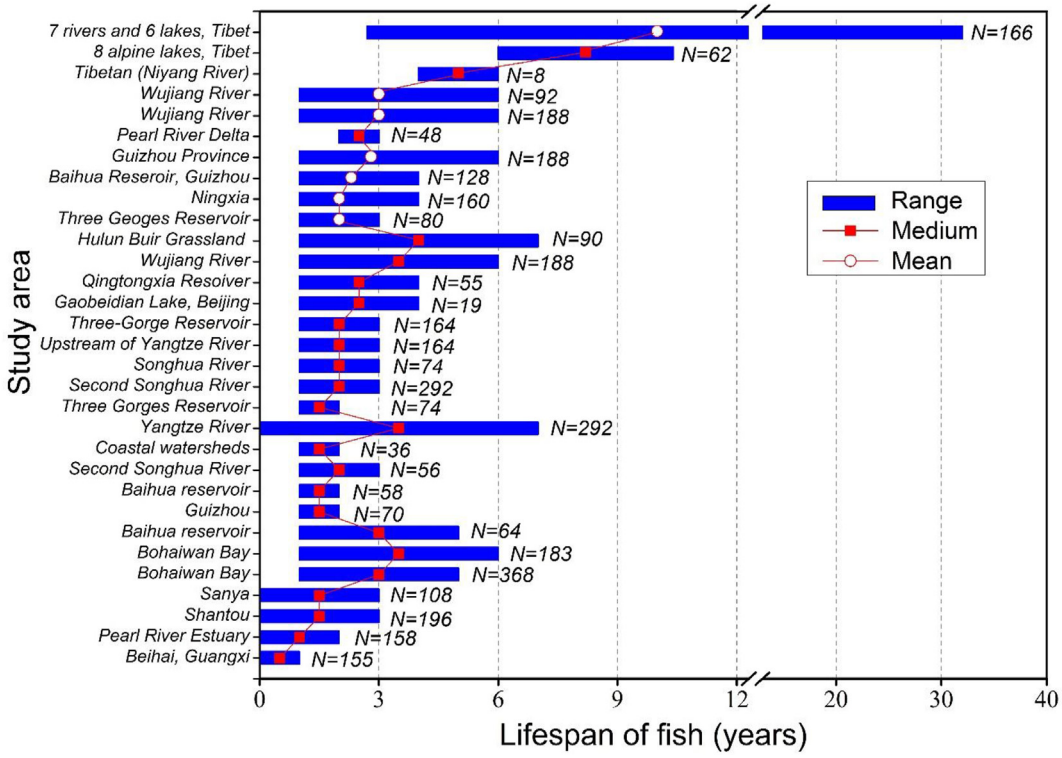

Fig. 4. Lifespans of fish from samples collected at different locations in China (median value was used when the mean value was not available). Detailed information and references are provided in Supplementary Table S5 of the Supplementary Material.

4. Conclusion and implication

From the perspective of an overall trend, our study reveals, for the first time, that Hg concentrations in fish decreased over time in China over the past three decades and exhibited a sharp contrasting trend to the increasing Hg emissions. The meta-analysis in this study clearly shows that unexpectedly low Hg accumulation in fish was associated with the lower trophic levels, a reduced body size, and a shorter lifespan largely driven by increasing fishing pressure, pervasive eutrophication, and extensive fish aquaculture.

This finding indicates the profound and complex impacts of the social system on the eco-environment system and has important implications for the implementation of the Minamata Convention on mercury in China and elsewhere. For instance, in the coming few decades, it could be expected that although China's anthropogenic Hg emission will decline continuingly and greatly, with the substantial progress made by China vigorously promoting the construction of ecological civilization (e.g., overfishing has been strictly regulated in recent years, and the balance of aquatic ecosystem has been restored accordingly), which may offset its co-benefit on fish Hg control (i.e., with the extension of fish food chain level and lifespan, China's fish Hg content might increase significantly). However, it is optimistic that given China's recent fish Hg has been far lower than the relevant standard limits recommended by the World Health Organization, China is expected to find a new balance between the fish Hg control and the ecological protection so as to avoid the challenges of fish Hg pollution prevention and treatment faced by European and American countries.

Declaration of competing interests

The authors have declared no conflicts of interest.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.eehl.2022.04.002.

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