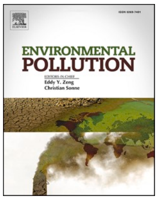
Environmental Pollution 309 (2022) 119791

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol



Microcystin pollution in lakes and reservoirs: A nationwide meta-analysis and assessment in China☆

Huimin Wei[a,c,1], Yunlu Jia[b,1], Zhi Wang[a,*]

[a] *Key Laboratory for Environment and Disaster Monitoring and Evaluation of Hubei, Innovation Academy for Precision Measurement Science and Technology, Chinese Academy of Sciences, Wuhan, 430077, China*
[b] *Institute of Hydrobiology, Chinese Academy of Sciences, Wuhan, 430072, China*
[c] *University of Chinese Academy of Sciences, Beijing, 100049, China*

ARTICLE INFO

Keywords:
Microcystins
China
Aquatic organisms
Ecological risks
Health risks

ABSTRACT

The frequent occurrence of microcystins (MCs) has caused a series of water security issues worldwide. Although MC pollution in natural waters of China has been reported, a systematic analysis of the risk of MCs in Chinese lakes and reservoirs is still lacking. In this study, the distribution, trend, and risk of MCs in Chinese lakes and reservoirs were comprehensively revealed through meta-analysis for the first time. The results showed that MC pollution occurrence in numerous lakes and reservoirs have been reported, with MC pollution being distributed in the waters of 15 provinces in China. For lakes, the maximum mean total MC (TMC) and dissolved MC (DMC) concentrations occurred in Lake Dianchi (23.06 μg/L) and Lake Taihu (1.00 μg/L), respectively. For reservoirs, the maximum mean TMC and DMC concentrations were detected in Guanting (4.31 μg/L) and Yanghe reservoirs (0.98 μg/L), respectively. The TMC concentrations in lakes were significantly higher than those in the reservoirs ($p < 0.05$), but no difference was observed in the DMC between the two water bodies ($p > 0.05$). Correlation analysis showed that the total phosphorus concentrations, pH, transparency, chlorophyll *a*, and dissolved oxygen were significantly related to the DMC in lakes and reservoirs. The ecological risks of DMC in Chinese lakes and reservoirs were generally at low levels, but high or moderate ecological risks of TMC had occurred in several waters, which were not negligible. Direct drinking water and consumption of aquatic products in several MC-polluted lakes and reservoirs may pose human health risks. This study systematically analyzed the pollution and risk of MCs in lakes and reservoirs nationwide in China and pointed out the need for further MC research and management in waters.

1. Introduction

The frequent occurrence of harmful cyanobacterial blooms (CyanoHABs) in eutrophic water has been a worldwide problem (Chen et al., 2006). Numerous bloom-forming cyanobacteria can produce a wide variety of cyanotoxins, threatening the aquatic ecosystem and drinking water quality. Microcystins (MCs), a class of potent hepatotoxins with cyclic peptides and tumor promoters, are among the most common cyanotoxins (Zhang et al., 2009). A large number of documents have reported the direct toxicological effects of MCs on plants, animals, and microbial communities in aquatic ecosystems (Cao et al., 2017; Jia et al., 2018; Wang et al., 2017). Meanwhile, MCs can threaten human health through drinking water and the food chain (Pan et al., 2021). The World Health Organization (WHO) provisional guideline for microcystin-LR (MCLR) in drinking water is 1 μg/L, which is based on a daily water intake of 2 L by a 60 kg adult (Hrudey et al., 1999).

Lakes and reservoirs are important components of drinking water sources. However, given the intensification of human activities and global warming, CyanoHABs in lakes and reservoirs showed a gradually increasing trend in recent decades. A recent publication reported that the frequency of CyanoHABs has increased in global inland waters during the past decades (Ho et al., 2019; Hou et al., 2022). MCs are frequently detected in lakes and reservoirs worldwide, e.g., Lake Erie in the United States, Lake Los Padres in Argentina, Lake Victoria in Africa,

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☆ This paper has been recommended for acceptance by Sarah Harmon.

* Corresponding author.

E-mail address: zwang@apm.ac.cn (Z. Wang).

[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.envpol.2022.119791
Received 2 May 2022; Received in revised form 18 June 2022; Accepted 12 July 2022
Available online 15 July 2022

Lake Suwa in Japan, and polar lakes in the southwestern part of Greenland in the Arctic Circle (Jessica et al., 2016; Belisle et al., 2016; Miles et al., 2013; Xie et al., 2007; Ame et al., 2010). In addition, in recent years, poisoning incidents and drinking water crises caused by CyanoHABs appears normalized (Qin et al., 2010; Wang et al., 2021). Therefore, it is necessary to accurately understand the situation of MCs pollution and reinforce risk assessment.

In China, massive studies have reported the pollution of MCs in lakes, e.g., Lake Taihu (Li et al., 2017), Lake Chaohu (Yang et al., 2016), Lake Dianchi (Wu et al., 2014), Lake Poyang (Zhang et al., 2015). Lakes and reservoirs in China are more seriously polluted by CyanoHABs and MCs compared with sparsely populated countries (Peng et al., 2010). In addition, the eutrophication of water bodies in China has attracted considerable attention. Researchers conducted surveys on 277 lakes scattered across the country and observed that 69.5% of the lakes surveyed suffered from eutrophication (Wen et al., 2019), suggesting a high risk of CyanoHABs and MCs in freshwaters in China. Subsequently, the nationwide assessment of CyanoHABs and MCs calls for systematic investigation, review, and summary of current situations.

In this study, we reviewed and meta-analyzed the available data of MC pollution in lakes and reservoirs in China over the past 20 years. The specific objectives were to (1) evaluate the occurrence of MC pollution in waters of lakes and reservoirs, (2) examine the bioaccumulation of MCs in typical aquatic organisms, (3) clarify the main environmental parameters that may affect the pollution and distribution of MCs, and (4) assess the ecological and health risks of MCs in major lakes and reservoirs in China.

2. Materials and methods

2.1. Data source and analysis

We used search keywords ("microcystins," "MCs," or "cyanobacteria toxins") and ("lakes" or "reservoirs," or "bioaccumulation") in the title, keyword, or abstract to select articles that were published between 2000 and 2021 in Web of Science and China National Knowledge Infrastructure to obtain data for the review and meta-analysis. More than 1000 articles were found via keyword search. Then, documents not related to the aims of this study were manually removed. Based on the criteria, 86 articles were finally selected for further analysis. The information extracted from articles included the study site, environmental parameter, the type and concentration of MCs, MC accumulation. The data were directly extracted from figures, tables, and supplementary materials of the articles. For data in certain figures, WebPlotDigitalizer software was used (Burda et al., 2017). Notably, not all selected articles recorded all the information we needed. However, all the data were extracted into a new database pool, and different subsets of the database were used for various statistical analyses.

The data selection according to the following principles: (1) there are available MC data in the published documents from the results or their supporting information; (2) MCs was detected by internationally recognized methods, i.e., high-performance liquid chromatography (HPLC), mass spectra (MS) and enzyme-linked immunosorbent assay (ELISA) (Massey et al., 2020). All available MC data in each lake or reservoir are included in the meta-analysis, and these MC data are mainly from the summer and autumn seasons. There are more than 200 kinds of MC isomers reported, only few literatures have determined multiple MC isomers simultaneously, so that it is difficult to evaluate the pollution and risk of all MC isomers. In fact, the current documents mainly focused on the investigation of total toxins, and MCLR, -YR, –RR isomers (Table S8). Therefore, dissolved MC (DMC, the sum of all dissolved MC isomers) and total MC (TMC, the sum of DMC and intracellular MC) were selected as the indicators to evaluate the pollution and risks of MCs. MCLR is considered to be one of the most widely distributed and most toxic MC isomers (Zegura, 2016), and only MCLR has sufficient toxicological data. Therefore, in this paper, MC risks were assessed according to the toxic data of MCLR.

Correlation analysis was used to clarify the relationship between MCs and environmental factors. The calculations and visualization were completed by R soft 4.1.2, Microsoft Office 2019, and Origin 2018.

2.2. Risk assessment

2.2.1. Ecological risk assessment

Optimized risk quotient (RQ_f) was used to evaluate the ecological risk of MCs in lakes and reservoirs (Zhou et al., 2019). This evaluation method considers the detection frequency and large temporal and spatial changes of pollutants and can accurately reflect the ecological risk of pollutants under the natural scenario. The RQ_f value was calculated using the following equations (Zhou et al., 2019):

$$RQ_f = RQ \times F = (MEC/PNEC) \times F \quad (1)$$

$$F = NO_1/NO_2 \quad (2)$$

Where PNEC means predicted no-effect concentration; MEC means measured environmental concentration; The RQ_f means the risk quotient (RQ) after considering the frequency of MECs exceeding PNEC; The RQ represents the ratio of MEC and PNEC; F is the frequency of MECs exceeding PNEC; NO_1 means the number of samples with higher concentrations than PNECs; NO_2 represents the total number of samples. The PNEC of MCLR was 5.73 μg/L, which was obtained from a species sensitivity distribution (SSD) model; the toxicity data used in the SSD model were mainly screened from the USEPA ECOTOX database (http://cfpub.epa.gov/ecotox/) (Niu et al., 2018). Difference risk levels were classified as follows: high environmental risk ($RQ_f \geq 1$); moderate environmental risk ($1 > RQ_f \geq 0.1$); small-scale adverse effect (endurable) ($0.1 > RQ_f \geq 0.01$); limited effect (negligible) ($0.01 > RQ_f > 0$); no risk (safe) ($RQ_f = 0$). Compared with other types of MCs, MCLR had a relatively sufficient toxicological data and high toxicity, and MCs were equivalent to MCLR for risk assessment in the present study.

2.2.2. Human health risk assessment

Hazard quotients (HQ) based on the were was used to evaluate the health risk of MCs to human beings. The HQ value was calculated using the following equations:

$$HQ = CDI/RfD \quad (3)$$

$$CDI = (C \times IR)/BW \quad (4)$$

Where CDI (, μg/kg body weight) is the chronic daily intake of MCs through ingestion per unit body weight, RfD (, μg/(kg•day)) is the reference dose for the MCs through oral exposure only. The RfD is represented by the acceptable daily intake, but its value is currently available for MCLR only (0.04 μg/(kg•day)) according to the WHO. The C (μg/L) is the concentration of MCLR equivalents in waters, IR is the ingestion rate (2 L/day for **drinking water** and 300 g/day for food), and BW is the body weight (60 kg for an adult and 10 kg for children) (Wan et al., 2020). For better remind the health risk of MC, we used the same amount of drinking water and food as adults in the ecological risk assessment of children. Difference risk levels were classified as follows: high ($HQ > 1$), moderate ($0.1 \leq HQ < 1$), and low ($HQ < 0.1$) health risks.

3. Results and discussion

3.1. Occurrence and distribution of MCs in water

From the selected 86 articles, MCs have been reported in 59 lakes and 37 reservoirs with a 100% detected rate in the investigated lakes and 84% detected rate in the investigated reservoirs, respectively (Fig. 1 and Table S1). These lakes and reservoirs are distributed in 15 provinces of China (Table S2), which are mainly located in the southeast region of

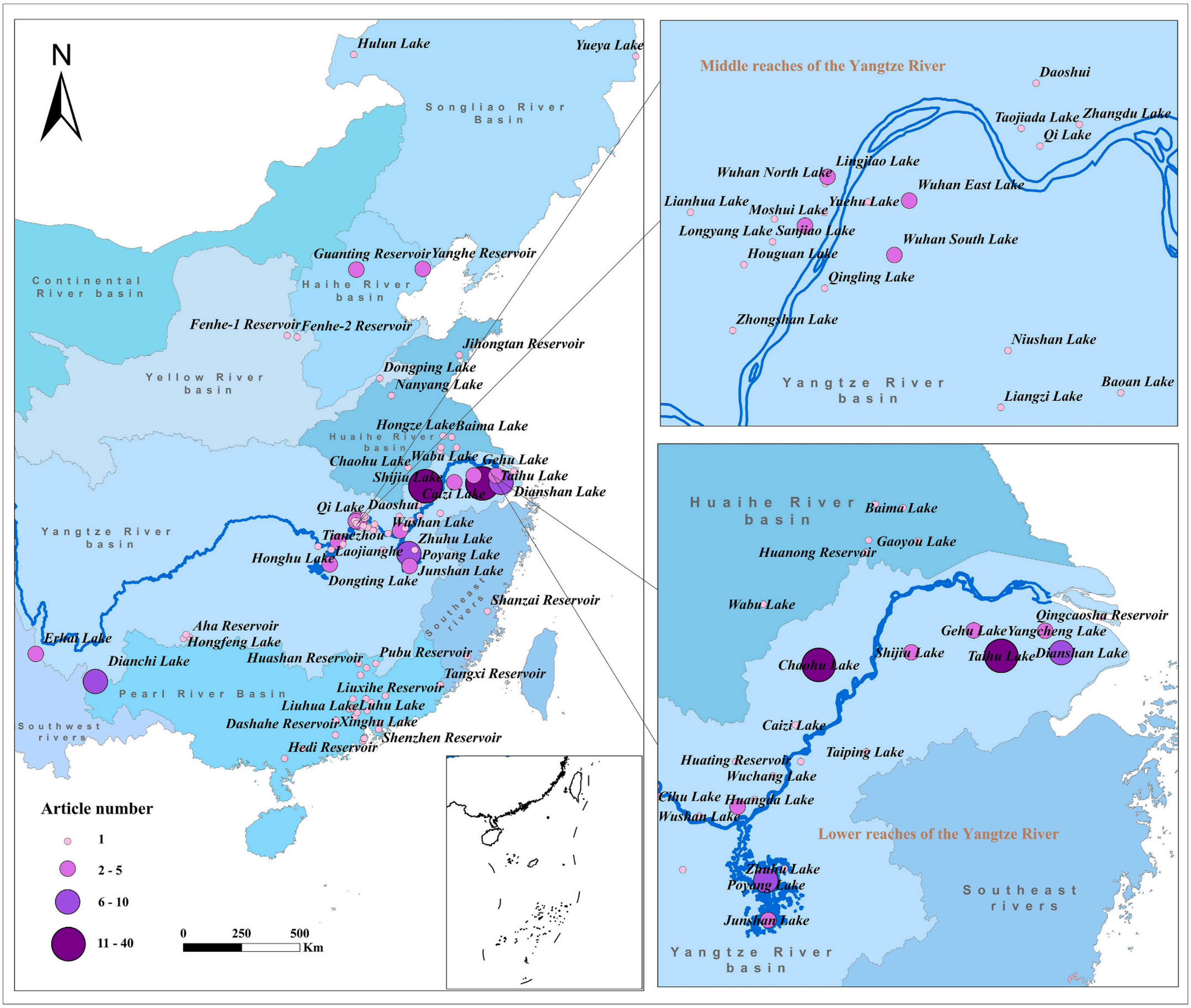


Fig. 1. Geographic location of MC-contaminated lakes and reservoirs in China. The point size represents the number of articles that published MC pollution in lakes and reservoirs.

the country (Fig. 1). In these regions, intensive anthropogenic activity led to eutrophication and the frequent occurrence of CyanoHABs in lakes and reservoirs (Huo et al., 2021). Lake Taihu (reported 33 times, n = 33) had been reported the most, followed by Lake Chaohu (n = 11), Lake Dianchi (n = 9), and Lake Poyang (n = 8). Different forms of MCs in the selected articles were systematically analyzed. The results showed that the ratio of DMC to TMC ranged from 1% to 36%, and the percentage in most water bodies was less than 20% (Table S1), suggesting that most of the MCs exist in intact cyanobacterial cells as intracellular MC (IMC) forms.

3.1.1. MC distribution

Among the 59 lakes with MC pollution, 52 lakes were located in the Yangtze River Basin (n = 47) and Huaihe River basin (n = 5), especially in the middle and downstream reaches of the Yangtze River Basin (n = 45) (Fig. 1), in part because these areas feature by a large number of lakes and extensive anthropogenic activities, contributing to the regional eutrophication of waters (Qin, 2002). A recent survey also showed that more than 90% of the shallow lakes in this area are eutrophic and hypereutrophic (Wan et al., 2020). Based on our data collection, 22 of the 59 lakes have sufficient TMC data. To describe the results conveniently, we classified the MC pollution degree based on the limit value (1 μg/L) stipulated by the WHO. The first type is high-pollution lakes, with the mean TMC concentration greater than 1 μg/L, e.g., Lake Dianchi, Lake Taihu, Lake Chaohu, and Lake Hongfeng. The mean TMC concentration of Lake Dianchi reached 23.06 μg/L. The second type is low-pollution lakes, e.g., Lake Dianshan, Lake Poyang, and Lake Erhai, with the mean TMC concentration lower than 1 μg/L (Fig. 2a). A total of 59% of the 22 lakes exceeded the WHO guideline. For DMC, only the mean DMC concentration in Lake Taihu reached the WHO guideline, and the highest DMC concentration in 39% of lakes, e.g., Lake Chaohu, Lake Dianchi, and Lake Poyang, exceeded this guideline (Fig. 2b).

MC contamination had been reported in 37 reservoirs, and 21 reservoirs contained sufficient TMC or DMC data for further analysis. Three reservoirs, i.e., Guanting Reservoir, Yanghe Reservoir, and Shanzi Reservoir, had a high pollution with mean TMC concentrations. The other reservoirs had low pollution levels with mean TMC concentrations below 1 μg/L (Fig. 2c). The mean concentrations of DMC in all the reported reservoirs were below this value (Fig. 2d). However, the highest

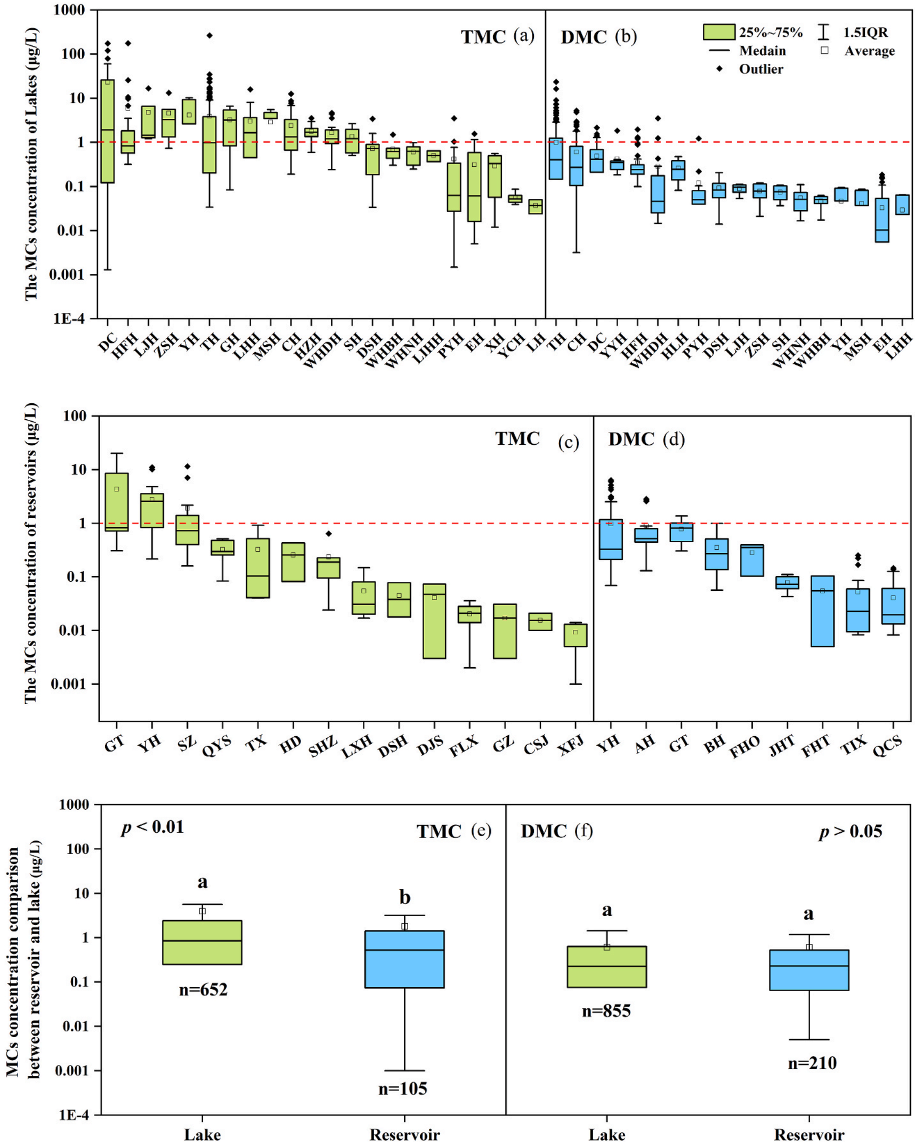


Fig. 2. Concentrations of TMC (a) and DMC (b) in main lakes and concentrations of TMC (c) and DMC (d) in main reservoirs. Comparison of TMC (e) and DMC (f) concentrations between lakes and reservoirs. The abbreviations of lakes and reservoirs are as follows: DC, Dianchi; HFH, Hongfeng; LJH, Lingjiao; ZSH, Zhongshan; YH, Yuehu; HZH, Hongze; TH, Taihu; GH, Gehu; LHH, Lianhua; MSH, Moshui; CH, Chaohu; WHDH, Wuhan Donghu; SH, Shahu; DSH, Dianshan; WHBH, Wuhan Beihu; WHNH, Wuhan Nanhu; LIHH, Liuhua; PYH, Poyang; EH, Erhai; XH, Xinghu; YC, Yangcheng; LH, Luhu; YYH, Yueya; HLH, Hulun. GT, Guanting; YH, Yanghe; SZ, Shanzi; QYS, Qiyeshi; TX, Tangxi; HD, Hedi; SHZ, Shenzhen; LXH, Liuxihe; DSH, Dashahe; DJS, Dajingshan; FLX, Feilaixia; GZ, Gaozhou; CSJ, Chishijing; XFJ, Xinfengjiang; AH, Aha; BH, Baihua; FHO, Fenhe-1; JHT, Jihongtan; FHT, Fenhe-2; TIX, Tingxi; QCS, Qingcaosha. The full names of the above abbreviations apply to Fig. 6 and 7.

DMC concentrations in Yanghe Reservoir, Guanting reservoir, and Aha Reservoir were higher than 1 μg/L. These reservoirs suffered from increased eutrophication by frequent CyanoHABs during the past decades because a large number of industrial and agricultural sewage flowed into the reservoir watershed (Li et al., 2010; Wang et al., 2017; Shen et al., 2011).

By comparison, the TMC concentration in lakes was significantly higher than that in reservoirs (Fig. 2e). This phenomenon was probably due to the following reasons: (1) In general, lakes are naturally formed low-lying areas, which are easy to be inputted with large amounts of exogenous pollutants from the basin surrounding areas through the inflow of water flow, resulting in water eutrophication. Meanwhile, the lakes investigated in previous studies are mainly shallow-water lakes, which are vulnerable to climate and hydrological regime changes and have a high efficient nutrient cycling effect (Qin, 2020). This condition will lead to frequent CyanoHABs and high TMC concentrations. (2) Reservoirs are mostly located in remote urban areas with less population. Their higher water depth and less pollution input will result in the generally lower intensity and frequency of CyanoHABs compared with those of shallow lakes (Han, 2010; Qin, 2020). Interestingly, our results showed no significant difference between lakes and reservoirs in DMC concentrations (Fig. 2f). On one hand, most of the cyanobacteria toxins mainly exist in intact cyanobacteria cells, resulting in the low DMC concentrations. On the other hand, it may be that the MCs are easily to be diluted, adsorbed, photodegradation and biodegradation in the water environment (Schmidt et al., 2014; Kurtz et al., 2021; Lin et al., 2017), which reduced the occurrence frequencies of high concentrations of DMC. In addition, the environmental fate of MCs in natural waters is a pending problem in the current research.

It is worth noting that our data are a comprehensive comparison of total toxins determined by ELISA and some specific isomers determined by HPLC and MS. In fact, in our meta-analysis, more than 70% MC data were determined by HPLC and MS (Table S8). There were more than 200 kinds of MC isomers probably existed in waters, but current HPLC and MS determination usually only select 1–5 isomers as target MCs (Table S8). Therefore, the MC pollution level of lake and reservoirs may be underestimated.

3.1.2. *Temporal trend of MCs*

Over the past several decades, the Chinese government has stepped up efforts to control CyanoHABs (Wang et al., 2012). Therefore, whether the concentration of MCs in lakes has been alleviated in the past several years must be determined. To answer this question, we analyzed the interannual variation trends of TMCs and DMCs during the outbreak season of CyanoHABs in Lake Taihu and Lake Chaohu because only these lakes have sufficient data. The results showed that the TMC and DMC concentrations in Lake Taihu increased from 2005 to 2015 (Fig. 3a and b, respectively). This MC variation trend was consistent with that of the scope and frequency of CyanoHAB in Lake Taihu during this period (Qin et al., 2021). Similarly, the TMC and DMC concentrations in Lake Chaohu showed a growing trend from 2002 to 2012 (Fig. 3c and d, respectively). This trend was similar with the duration, frequency, percentage of coverage area of CyanoHABs in Chaohu Lake from 2002 to 2012 (Tang et al., 2017). We do not think that the increase in the concentrations of MCs was because of the more MC isomers were determined simultaneously. In fact, according to our literature survey data, the MC isomers investigated by scholars in different periods have not changed obviously (Table S8).

Interestingly, compared with 2012, the concentration of DMC in Lake Chaohu decreased in 2017. To cope with the continuous high-temperature weather and improve the water quality of Lake Chaohu, Anhui province implemented the project of water diversion from Yangtze River to supply Lake Chaohu in 2017, with a total inflow of 2.69×10^8 m^3. Although the project cannot fundamentally improve the water quality of Lake Chaohu and inhibit the outbreak of CyanoHABs, the dilution of Lake Chaohu by river water may reduce the concentration of MCs (Gao et al., 2018). In addition, *Pseudoanabaena mucicola* and *Aulacoseira Granata* had formed dominant species in several sampling points in Lake Chaohu (Zhu et al., 2018). Notably, not all species of *Microcystis* sp. can produce MCs, which should also probably explain the descent of MC concentration.

3.1.3. Correlations between MC concentration and environment factors

Environmental factors were extensively reported as regulators for MC production and release in toxigenic cyanobacteria (Wan et al., 2020; Wu et al., 2006; Li et al., 2017). The relationship between MC concentration and environmental factors has been studied in water bodies all over the world, and most studies usually obtained contradictory results. The research on environmental and MC data were selected for further correlation analysis (Fig. 4). The results showed that the content of ammonia nitrogen (NH_4^+-N) was significantly positively correlated with TMC concentration ($p < 0.01$), whereas pH and transparency were significantly negatively correlated with TMC concentration ($p < 0.01$). The total phosphorus (TP) concentrations, pH, and chlorophyll *a* (Chl *a*) contents were significantly positively correlated with DMC concentration, whereas dissolved oxygen (DO) and transparency were significantly negatively correlated with the DMC concentration ($p < 0.01$).

CyanoHABs are related to massive nitrogen (N) and phosphorus (P) inputs (Xu et al., 2021). A whole-lake fertilization showed that under high P loads, large CyanoHABs can develop rapidly without anthropogenic nitrogen inputs (Molot et al., 2021). Several studies suggested the decrease in the TN concentration of Lake Taihu since 2006 but not the concentration of TP and MCs (Qin et al., 2021; Wan et al., 2020). This finding indicates that the P concentration may play a leading role in the change in CyanoHABs and can explain the significant positive correlation of TP with DMC. Meanwhile, the results showed that NH_4^+-N has a significant positive correlation with TMC concentration. This result may be due to the rapid turnover of NH_4^+ in the aquatic environment, which may strongly support the persistence of CyanoHABs and promote the increase in phytoplankton biomass (Wu et al., 2020; Xue et al., 2021). Cyanobacteria can live using regenerated NH_4^+ to sustain the bloom, and internal NH_4^+ regeneration exceeds the external N loading to the lake by two times (Hampel et al., 2018).

A large number of cyanobacteria are bound to increase the content of chlorophyll in water, thus enhancing photosynthesis and reducing carbon dioxide content, which will increase pH. Meanwhile, pH can also affect the growth of cyanobacteria, the dissolution and metabolite release of cyanobacteria cells, and the adsorption of MC on suspended particles (Liu et al., 2008; Unrein et al., 2010; Qian et al., 2014). In addition, the proliferation of cyanobacteria will reduce the transparency of water and consume DO in water. High pH value and low DO can promote the mineralization and degradation of nutrients in sediments into dissolved forms, and the resuspension of sediments can release a large amount of soluble P to the overlying water (Christophoridis and Fytianos, 2006; Niemist et al., 2011; Ding et al., 2018; Zhu et al., 2020). The result of a microcosm system that simulates the decomposition processes of CyanoHAB showed that the feedback between CyanoHABs formation and environmental factors of freshwater lakes is bidirectional (Yan et al., 2017).

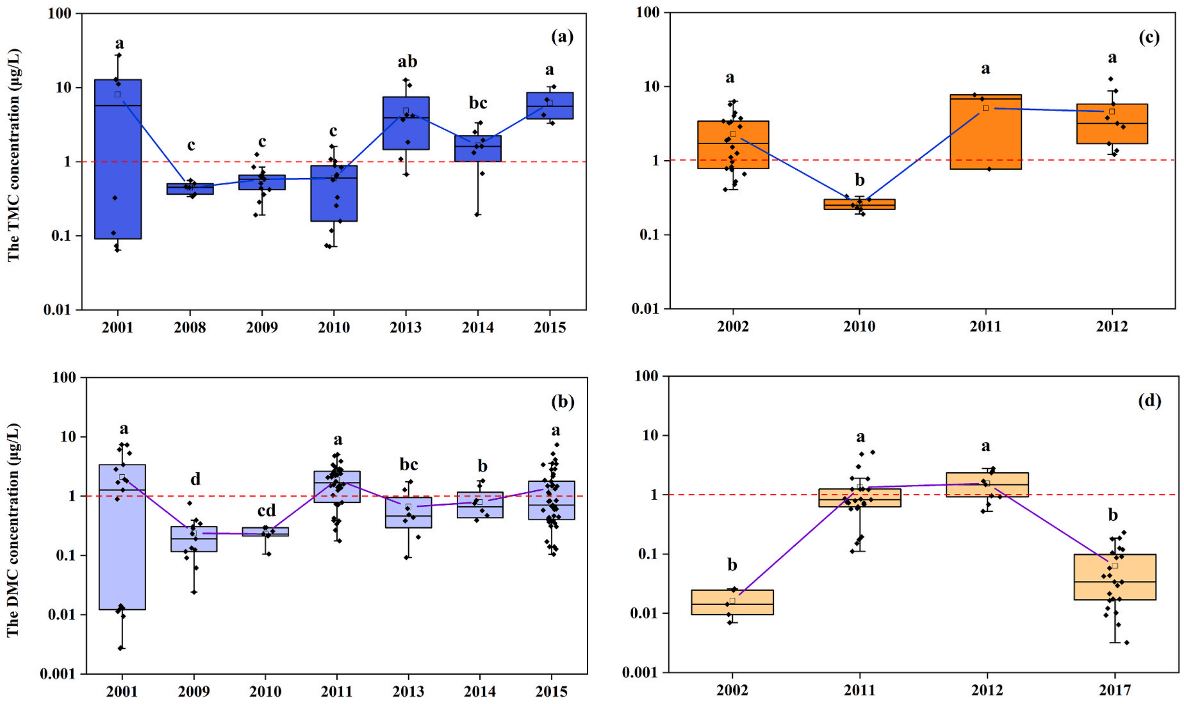
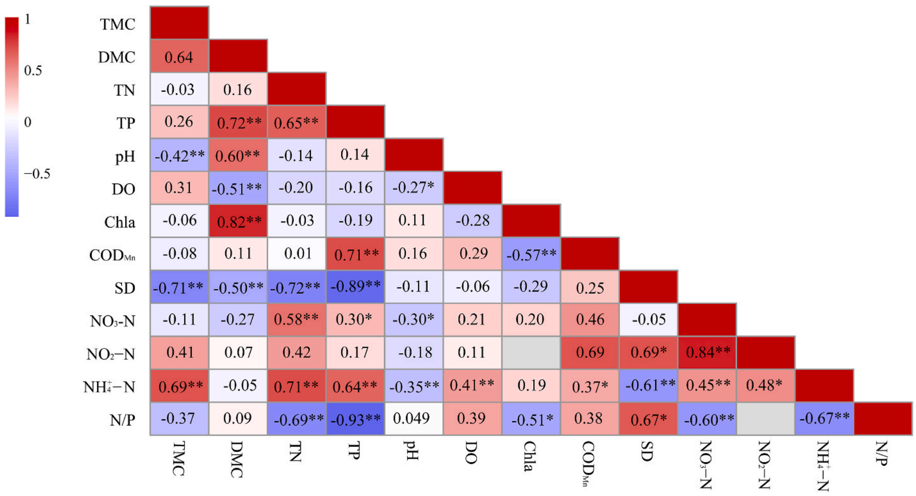


Fig. 3. Time variation of TMC and DMC concentrations in Lake Taihu (a and b) and Lake Chaohu (c and d).



	TMC	DMC	TN	TP	pH	DO	Chla	COD_{Mn}	SD	NO_3–N	NO_2–N	NH_4^+–N	N/P
TMC													
DMC	0.64												
TN	-0.03	0.16											
TP	0.26	0.72**	0.65**										
pH	-0.42**	0.60**	-0.14	0.14									
DO	0.31	-0.51**	-0.20	-0.16	-0.27*								
Chla	-0.06	0.82**	-0.03	-0.19	0.11	-0.28							
COD_{Mn}	-0.08	0.11	0.01	0.71**	0.16	0.29	-0.57**						
SD	-0.71**	-0.50**	-0.72**	-0.89**	-0.11	-0.06	-0.29	0.25					
NO_3-N	-0.11	-0.27	0.58**	0.30*	-0.30*	0.21	0.20	0.46	-0.05				
NO_2–N	0.41	0.07	0.42	0.17	-0.18	0.11		0.69	0.69*	0.84**			
NH_4^+–N	0.69**	-0.05	0.71**	0.64**	-0.35**	0.41**	0.19	0.37*	-0.61**	0.45**	0.48*		
N/P	-0.37	0.09	-0.69**	-0.93**	0.049	0.39	-0.51*	0.38	0.67*	-0.60**		-0.67**	

Fig. 4. Spearman correlations between MC concentrations and environmental factors. TMC: total microcystin; DMC: dissolved microcystin; TN: total nitrogen; TP: total phosphorus; DO: dissolved oxygen; Chl *a*: chlorophyll *a*; COD_{Mn}: permanganate index; SD: secchi depth; NO_3-: Nitrate nitrogen; NO_2-: Nitrite nitrogen; NH_4^+-N: ammonia nitrogen; N/P: nitrogen/phosphorus. * represent $p < 0.05$, ** represent $p < 0.01$.

3.2. MC pollution in aquatic organisms

The MC accumulations of 22 aquatic species (including fish and mollusks) were summarized from the previous data on Lake Taihu (Jia et al., 2014), Lake Chaohu (Xie et al., 2005), Lake Dianchi (Zhang et al., 2012), and Lake Poyang (Xu et al., 2003). The results showed that MCs were detected in numerous organs of aquatic organisms, including hepatopancreas, muscle, gonad, gill, blood, digestive system (i.e., gut content, intestine, intestinal wall, gallbladder, digestive tracts, bile, and stomach), and visceral mass (i.e., kidney, spleen, abdomen, and heart) (Zhang et al., 2012; Song et al., 2007; Chen et al., 2016; Xie et al., 2005; Chen and Xie, 2005a, 2005b; Zhang et al., 2009).

MC accumulation in different tissues were compared (Fig. 5a–e). For 13 fish species, the results showed that the high-to-low mean MC

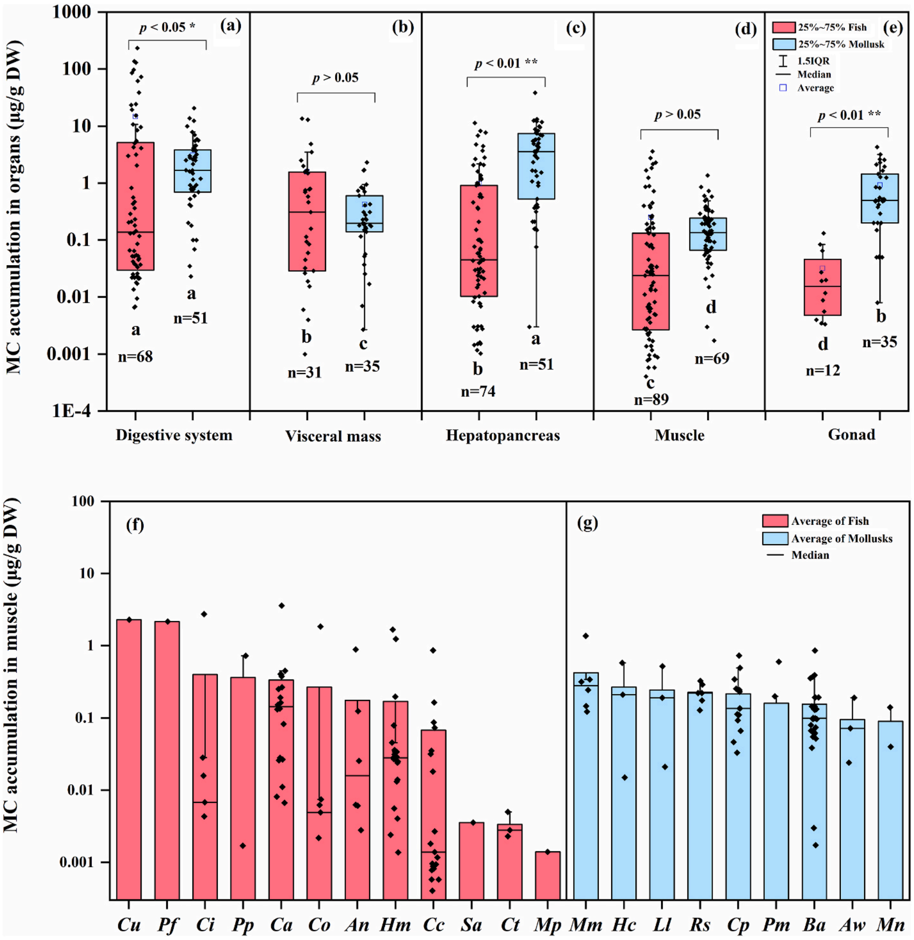


Fig. 5. MC accumulation in different organs of fish and mollusks and comparison of MC accumulation between fish and mollusk in the digestive system (a), visceral mass (b), hepatopancreas (c), muscle (d), and gonad (e). MC accumulation in the muscle of different fish (f) and mollusks species (g). The abbreviations of fish and mollusks are as follows: *Cu, Culter erythropterus; Pf, Pseudobagrus fulvidraco; Ci, Culter ilishaeformis; Pp, Parabramis pekinensis; Ca, Carassius auratus; Co, Coilia ectenes; An, Aristicbthys nobilis; Hm, Hypophthalmichthys molitrix; Cc, Cyprinus carpio; Sa, Silurus asotus; Ct, Ctenopharyngodon idellus; Mp, Mylopharyngodon piceus; Mm, Margarya melanioides; Hc, Hyriopsis cumingii; Ll, Lamprotula leai; Rs, Radix swinhoei; Cp, Cristaria plicata; Pm, Palaemon modestus; Ba, Bellamya aeruginosa; Aw, Anodonta woodiana; Mn, Macrobrachium nipponensis.* The full names of the above abbreviations apply to Fig. 8.

accumulations were observed in the digestive system (14.814 μg/g dry weight (DW)) > visceral mass (1.587 μg/g DW) > hepatopancreas (0.971 μg/g DW) > muscles (0.257 μg/g DW) > gonads (0.032 μg/g DW). For 9 species of mollusks, the mean MC accumulations from high to low values were noticed in the hepatopancreas (4.906 μg/g DW) > digestive system (3.083 μg/g DW) > gonads (0.931 μg/g DW) > visceral mass (0.423 μg/g DW) > muscles (0.202 μg/g DW) (Table S3). Furthermore, MC accumulations in the similar tissues between fish and mollusks were compared (Fig. 5a–e). The results showed that the MC accumulation in the digestive system of fish was significantly higher than that of the mollusk. The MC accumulations in the hepatopancreas and gonads of mollusks were significantly higher than those in the fish. No significant difference was observed in the MC accumulation of visceral mass and muscle between fish and mollusks.

The digestive system had the highest MC accumulation in fish. Several studies showed that the diet of phytoplanktivorous fishes in Lake Taihu comprises 90% *Microcystis* sp.; omnivorous and carnivorous fishes can also accumulate MCs in their gut by ingesting toxic cyanobacterial cells, detritus, and benthic invertebrates (Jia et al., 2014; Zhang et al., 2009). Studies on MC accumulations in various fishes with different feeding habits from Lake Taihu and Lake Chaohu also showed that the MC content in the gut was the highest in phytoplanktivorous fish, followed by that in omnivorous fish, and the lowest value was observed in carnivorous fish (Zhang et al., 2009; Xie et al., 2005; Peng et al., 2010). The MC accumulation in the digestive system of mollusks was second to hepatopancreas because the latter are the target organ of MCs (Chen and Xie, 2010). The hepatopancreas accounted for 95% of the MCs acquired by mollusks, resulting from the ingestion of toxic cyanobacteria. Other studies had proven the positive correlation between MC accumulation in the digestive tract and MC accumulation in the hepatopancreas of mollusks (Chen and Xie, 2005a; Chen and Xie, 2010; Zhang et al., 2012).

In addition, we observed a large MC accumulation in the kidney of fish, with values higher than hepatopancreas, which is also the MC target organ in fish (Song and Chen, 2009). Once MCs are absorbed by the intestinal wall, they can be transported rapidly through blood circulation and spread to various organs or tissues (Chen et al., 2006). Hepatopancreas and kidneys are highly hemoperfusion organs, which may be the reason for the high accumulation of MCs in the fish kidney (Chen et al., 2006); however, the specific mechanism needs to be further studied.

The accumulation of MC in muscles was frequently reported because muscle is the main edible part of fish. The mean MC accumulations in muscles of 12 species of fish were compared, and the results showed that *Culter erythropterus* and *Pseudobagrus fulvidraco* had high MC accumulations, *Silurus asotus*, *Ctenopharyngodon idellus,* and *Mylopharyngodon piceus* had low MC accumulations, and the other species had a medium MC accumulation (Fig. 5f). Given that the results did not show the evident food dependence of fish MC accumulation, this study cannot determine the kind of feeding fish with a higher MC bioaccumulation. This dilemma may be due to, on the one hand, the collection of the investigated fish from different environments and the inconsistent concentrations of MCs around them. On the other hand, different fish may have varied detoxification abilities (Zhang et al., 2009). The MC accumulations in the foot or muscle of mollusk were also compared, and no significant difference was observed in MC accumulation in different mollusk species (Fig. 5g).

3.3. Risk assessment

3.3.1. Ecological risk assessment

Considering the potential harmful effects of MCs on aquatic ecosystems, the ecological risk of MCs was evaluated (Fig. 6 and Table S4). For TMC, when using the mean reported concentration to estimate the ecological risk, only Lake Dianchi had a high ecological risk, and the mean value of RQ_f in Lake Dianchi was 1.87. However, when estimating the ecological risk at the highest reported concentration, the results showed that the ecological risk of Lake Dianchi, Lake Hongfeng, Lake Taihu, and Guanting Reservoir can reach a high level, and the highest RQ_f value of these water bodies were 14.16, 3.35, 6.28, and 1.02, respectively. The other lakes or reservoirs were at moderate risk levels (the highest $RQ_f < 1$). For DMC, Lake Taihu and YangHe reservoir had a negligible risk, with the mean DMC concentrations being used to estimate the ecological risk. However, when estimating the ecological risk by the highest concentrations, Lake Taihu (the highest $RQ_f = 0.082$) and YangHe reservoir (the highest $RQ_f = 0.025$) reached endurable risk.

Although our results illustrated that the ecological risk of DMC in natural water bodies in China is at low levels to a large extent, aquatic animals in lakes or reservoirs often feed on or passively ingest planktonic algae. Therefore, in order to protect the health of aquatic ecosystem to the greatest extent, we suggest to use the TMCs in water to assess the ecological risk of MCs in water.

3.3.2. Health risk assessment

The health risk of MCs to human is mainly caused by human drinking water containing MC. Generally, human do not directly drink natural water with CyanoHABs. However, the filtration technology in the process of drinking water production can only remove the undamaged cyanobacteria cells, and flocculation, disinfection and other measures may even destroy the cyanobacteria cells, resulting in the increase of DMC in the water. Therefore, in order to make the evaluation results more scientific and accurate, DMC and TMC are both used to evaluate the health risk of MC in drinking water.

For adults, all lakes and reservoirs were at moderate or low health risk (HQ < 1) when using average DMC concentrations to assess the health risk for drinking water (Fig. 7a; Table S5). When the highest reported DMC concentrations were used to assess the health risk, the results showed that the high health risk (HQ > 1) of MCs for drinking water existed in 7 lakes and 3 reservoirs, i.e., Lake Taihu (HQ = 19.63), Lake Chaohu (HQ = 6.35), Lake Dianchi (HQ = 1.8), Lake Yueya (HQ = 1.53), Lake Hongfeng (HQ = 1.63), Wuhan Lake Donghu (HQ = 2.93),

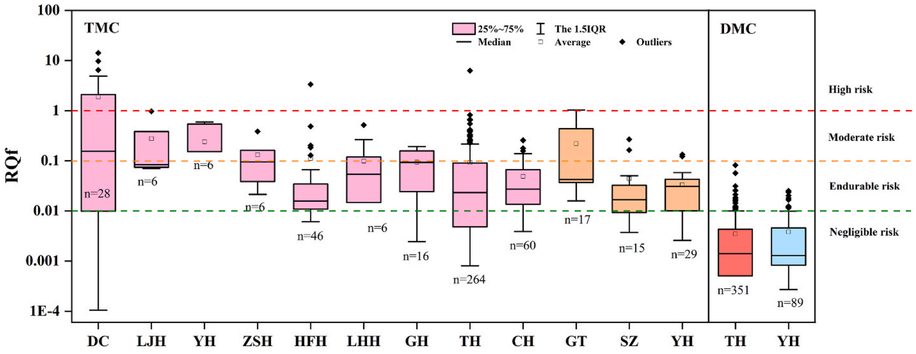


Fig. 6. Ecological risk assessment results (RQ_f) of TMC and DMC concentrations of lakes and reservoirs. Only RQf values exceeding 0 are shown.

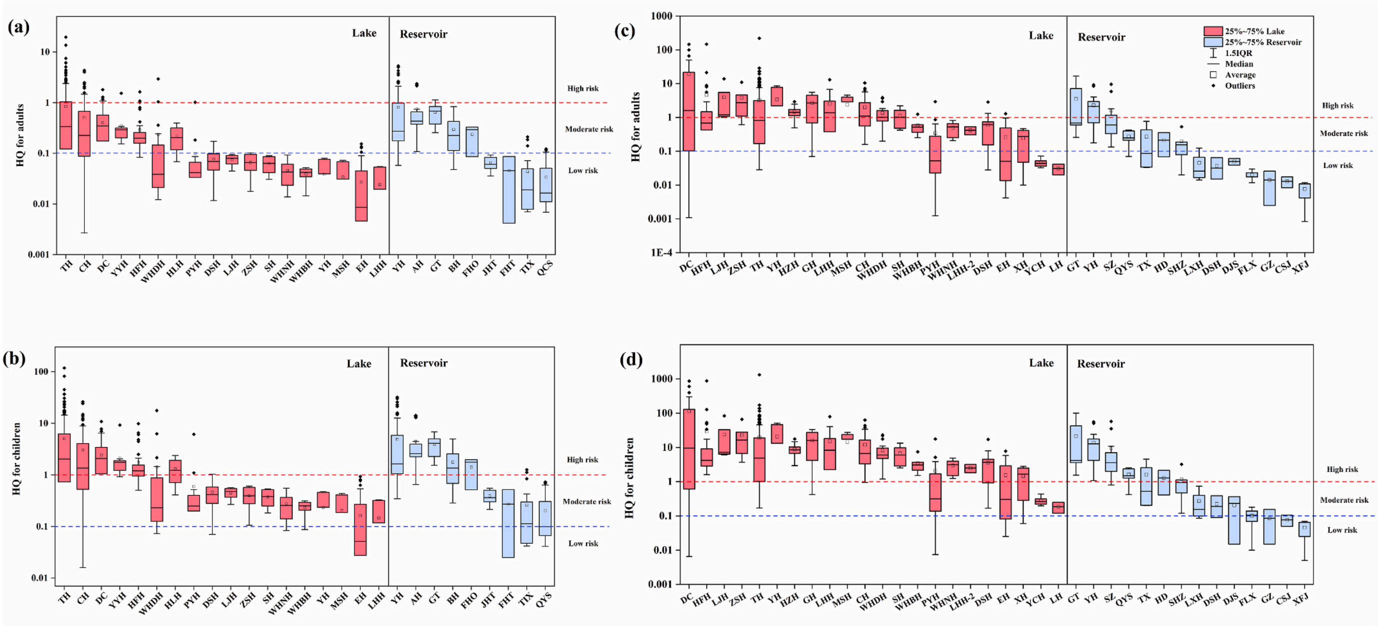


Fig. 7. Human health risk assessment of DMC to adults (a) and children (b) in lakes and reservoirs, human health risk assessment of TMC to adults (c) and children (d) in lakes and reservoirs.

Lake Poyang (HQ = 1.02), Yanghe reservoir (HQ = 5.37), Aha reservoir (HQ = 2.33), and Guanting reservoir (HQ = 1.14). Lake Hulun, Lake Dianshan, Lake Zhongshan, Lake Erhai, Fenhe-1 reservoir, Baihua reservoir, Tingxi reservoir, and Qingcaosha reservoir can reach a moderate health risk ($0.1 \leq HQ < 1$), whereas the other lakes and reservoirs have low health risks. However, for children, when using the mean DMC concentrations to assess the health risk, the high health risk of MCs for drinking water existed in 7 lakes and 5 reservoirs, and other lakes and reservoirs had moderate health risks (Table S5). Moreover, 9 lakes and 6 reservoirs had a high health risk when assessed by the highest DMC concentrations. Meanwhile, other lakes and reservoirs showed moderate health risks (Fig. 7b).

For adults, 13 lakes and 3 reservoirs were at high health risk ($HQ > 1$) and 7 lakes and 4 reservoirs were at moderate health risk when using mean TMC concentrations to evaluate the health risk for drinking water (Fig. 7c; Table S6). Once the highest reported TMC concentrations were used to assess the health risk, the results indicated the high health risk ($HQ > 1$) of MCs for drinking water existed in 17 lakes and 3 reservoirs, and only 5 lakes at moderate or low health risk. However, for children, when using the mean TMC concentrations to assess, the low health risk of MCs for drinking water only existed in 3 reservoirs, and other lakes and reservoirs had moderate or high health risks. In addition, only one reservoir had a low health risk when evaluated by the highest TMC concentrations, and other lakes and reservoirs showed moderate or high health risks (Fig. 7d).

In China, lakes and reservoirs are the most and the best centralized drinking water sources, especially in the eastern developed areas, serving 51.0% of the population. Therefore, understanding the MC pollution risk in lakes and reservoirs is critically important for protecting people's lives and health (Zhang et al., 2022). Our results showed that in several lakes and reservoirs with CyanoHABs, despite the physical filtration of water, the human health risk cannot be ignored. In the drinking water treatment process, preventing the damage of cyanobacteria cells is necessary because it can result in increased DMC and aggravate health risks. When considering the natural water body with CyanoHABs as the water source, more focus is needed on the MC removal technology. The environmental behavior of MCs in the water supply system also needs research attention. This health risk assessment model only discusses the human health risk of MCs in the drinking water source. Lakes, reservoirs, and ponds are often used as drinking water sources for large terrestrial animals. Given the lack of a water treatment system, large terrestrial animals often drink water containing toxic cyanobacteria directly, posing a great threat to them. The main reason reported for the numerous wild elephant deaths in Africa is the probable drinking of large volumes of natural water containing MCs (Wang et al., 2021).

The entry of MCs in the human body through the food chain is another important pathway to threaten human health. Our study evaluated the health risks of MCs in aquatic products consumed by humans. The data were mainly from 21 aquatic organisms in Lake Taihu, Lake Chaohu, Lake Poyang, and Lake Dianchi (Jia et al., 2014; Zhang et al., 2012; Xie et al., 2005; Xu et al., 2003). A coefficient of 5 was used to convert muscle DW to muscle wet weight (Chen et al., 2006). The health risk via eating muscle of aquatic organisms was evaluated with the highest reported MC accumulation. The results showed that for adults, all mollusks and almost all fish (except *Ctenopharyngodon idellus*, *Silurus asotus*, and *Mylopharyngodon piceus*) are at a high health risk. In particular, the HQ values of *Culter erythropterus*, *Pseudobagrus fulvidraco*, and *Carassius auratus* in Lake Chaohu reached 57.4, 54.3, and 90.2, respectively. For children, the results showed that all aquatic organisms that had been reported from the above four lakes were at moderate or high health risk (Fig. 8 and Table S7).

Most of the MC accumulation data in this study were derived from the period of CyanoHABs and high MC concentration. Aquatic organisms are affected by the MC concentration in water and thus have a high MC accumulation, which will result in high health risks. Moreover, the fishing season is from September to December (Peng et al., 2010). Although CyanoHABs rarely occur in winter, autumn is still a high-incidence season of CyanoHABs. Thus, the aquatic products consumed by humans may present high health risks. In lakes with frequent CyanoHABs, the government should put forward effective management strategies to reduce or avoid potential health risks.

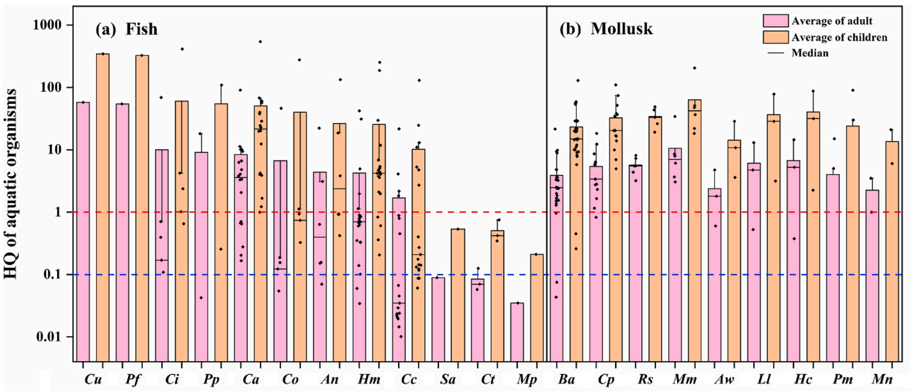


Fig. 8. Human health risk assessment results (HQ) of MC accumulation in muscle/foot of fishes and mollusks through direct consumption for adults and children.

4. Implications for management and research

4.1. MC routine monitoring is inevitable for lake and reservoir management

The national investigation of MCs distribution in Chinese lakes and reservoirs revealed that MC risks the request of inspection in these waterbodies. In China, lakes and reservoirs often serve not only as a source of drinking water but also aquaculture, irrigation, and receiving sewage, which will lead to overloading of nutrients and frequent occurrence of CyanoHABs. At present, MCs only include the unconventional indicators of drinking water, and the monitoring scope only includes water sources to protect human health. However, our research showed that MC accumulation in aquatic organisms also has health risks to humans. Although standard methods for the determination of MC accumulation in aquatic products are available, no specific monitoring and evaluation plan can be used for MC health risks in aquatic products in China. Therefore, further legislation is still needed to reduce MC health risks in aquatic products. Most of the lakes and reservoirs currently reported are mainly large and medium-sized water bodies. Numerous small water bodies, e.g., urban lakes and landscape ponds, are beyond the monitoring scope. These water bodies are also closely related to urban life. Finally, although the government has included MCLR into the legislation standard, the current monitoring of MCs in water is mostly used for scientific research. This monitoring lacks time continuity and regularity, which are not conducive to the accurate understanding of MC pollution in water. Hence, a strict routine monitoring program should be implemented especially during the period of CyanoHABs. In addition, in order to effectively and accurately monitor MCs in the environment, it is necessary to strengthen the scientific research and develop novel and easy methods for measuring trace MCs in different environmental matrices.

4.2. MC removal measures should be integrated into the controlling strategies for CyanoHABs

Nutrient control is one of the most effective measures to mitigate CyanoHABs. However, this measure is unachievable in the short term due to exogenous nutrient input reduction being constrained by the demand for economic development. The endogenous nutrient release also contributes to the difficulty of eutrophication control in lakes and reservoirs of China. In addition, toxigenic cyanobacteria may gain advantages upon the non-toxic strains by several biological traits, such as MC production. Thus, MCs will exist for a long time. Given this condition, the ecological and health risks of MCs must be comprehensively considered and MC removal technology be developed to avoid risks. MCs mostly exist in healthy cyanobacteria cells in the form of IMC, which means that when cyanobacteria cell rupture, IMC will be released to the DMC. Our study showed that the risk of DMC was significantly lower than that of TMC. This finding indicated that in the water treatment process, if the algae are not treated properly, toxins may be released, and the direct ecological and health risks may increase. Therefore, MC removal measures must be incorporated into the control strategy of CyanoHABs. First, the research on the mechanism and influencing factors of MC production and release should be strengthened. Second, the risk assessment of MC removal measures must be strengthened to avoid secondary contamination.

5. Conclusion

MC pollution in lakes and reservoirs of China was investigated by meta-analysis. The results showed that MC pollution is widespread in lakes and reservoirs of China. A total of 59 lakes and 37 reservoirs, which are located in 15 provinces, have been reported with microcystin pollution. The TMC pollution level in lakes is significantly higher than that in reservoirs, but there was no different in DMC between these two water bodies. Correlation analysis showed that phosphorus rather than nitrogen may play a leading role in the dynamics of DMC concentration. Due to the limited investigated and the lack of unified standards, the current data did not show that fish MC accumulation was obvious food dependence. Ecological risk of DMC in natural water bodies in China was at a low level to a large extent. However, in order to protect the aquatic ecosystem to a greater extent, it may be more meaningful to use TMC as the assessment indicator of MC ecological risk, because aquatic animals are threatened by both IMCs and DMCs, simultaneously. Direct drinking water and eating aquatic products in several MC polluted lakes and reservoirs may pose human health risk, especially to children. Further works are needed to strengthen the MC routine monitoring and control not only in lakes and reservoirs but also in small water bodies.

Author statement

Huimin Wei and Zhi Wang designed the study. Huimin Wei, Yunlu Jia, and Zhi Wang wrote the initial draft of the manuscript. All authors read, modified, and approved the final manuscript.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

This work was supported by the China National Key R&D Program (Grant No. 2019YFD0900604), the National Natural Science Foundation of China (NSFC: 41601545), Key Project of Hubei Province Natural Science Foundation (2020CFA110) and Youth Innovation Promotion Association, Chinese Academy of Sciences (2018369).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119791.

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