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ORIGINAL PAPER

Distributions of total mercury and methylmercury and regulating factors in lake water and surface sediment in the cold-arid Wuliangsuhai Lake region

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Abstract This study aimed to understand the occurrence of mercury in the water environment of typical cold and arid lakes and the regulating environmental factors. Water and surface sediment samples were collected from July to August, 2022 in the Wuliangsuhai Lake region for the analysis of total mercury (THg) and total methylmercury (TMeHg). Lake water THg and TMeHg ranged between 19.20 ~ 668.10 and 0.10 ~ 11.40 ng/L, respectively, exceeding China's environmental quality standards and contents of other lakes and reservoirs in China and other areas. Surface sediments showed lower mean THg and TMeHg of 261.85 and 0.18 μg/kg, respectively, with the former significantly exceeding the background value of Inner Mongolia and unpolluted natural lakes but lower than those of lakes affected by human factors, such as aquaculture. Sediments showed relatively low methylation and TMeHg (0.01–0.21%) concentrations. Correlation analysis identified salinity, total dissolved solids, conductivity, and redox potential as important factors affecting mercury speciation in water, whereas those in surface sediments were organic matter, pH, and total iron content. This study conducted preliminary research on the different species of Hg in Wuliangsuhai Lake water environment, which can provide scientific evidence for the specific treatment of Hg pollution in agriculture, or industry and other related fields. Our results suggest that upstream and downstream regulatory agencies should strengthen the regulation of agricultural and industrial production, moderately reduce human activities, and reduce the use of mercury-rich substances such as pesticides.

Keyword Distribution · Total mercury · Methylmercury · Surface sediments · Water body · Cold-arid

Introduction

Mercury (Hg) is characterized by persistence, high toxicity, and biological enrichment (Guo et al., 2018). It is also the only heavy metal existing in a liquid state at room temperature and under constant pressure (Long, 2020) and can be transported over long distances in the atmosphere (Seelen et al., 2021). These unique physicochemical properties of Hg have contributed to its status as a global pollutant (Liu et al., 2022a; b). Mercury toxicity within the ecological

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environment is related to its structure and morphology. Methylmercury (MeHg) is the most toxic among the many mercury compounds. Unlike inorganic mercury and other inactive mercury species, methylmercury is fat-soluble and human exposure to methylmercury through its movement through the blood–brain barrier can cause permanent damage to the central nervous system (Todorova et al., 2014). Mercury in the water environment gradually accumulates up the food chain (Guimares et al., 1998), ultimately resulting in the risk of mercury exposure in humans. Past studies have shown that biotic and abiotic processes in the water environment convert inorganic mercury into methylmercury, with most of these processes occurring in the sediment surface layer (Regnell et al., 2014; Sun et al., 2018; Wang et al., 2016; Yu et al., 2021). Wetland ecosystems are generally characterized by an anaerobic environment, rich nutrient salts, and high microbial activity, providing conditions suitable for mercury methylation. Therefore, there is a significant need to evaluate the risks of different types of mercury to environmental health, with particular attention to the migration and transformation of different forms of mercury regulated by environmental factors. There is also value in exploring the biogeochemical cycle of mercury in wetland ecosystems.

Wuliangsuhai Lake is the largest shallow lake in the Yellow River Basin, the eighth-largest lake in China, and the largest wetland at its latitude on Earth (Sun, 2019). It is a typical cold-arid region lake, with the lake region characterized by a dry summer and long, cold winter. Therefore, research into the ecological services of Wuliangsuhai Lake can guide further understanding of cold-arid region lakes. The lake region also forms a key link between the Hetao Irrigation Area and the Yellow River. However, the lake receives large yearly volumes of farmland irrigation return flow, domestic sewage, and industrial wastewater (Zhang et al., 2022). The unique ecological environment of the lake is conducive to the migration and transformation of mercury. Contemporary studies on mercury and methylmercury in wetland ecosystems in China have mainly focused on southwest China, particularly on the Three Gorges Basin (Guo et al., 2018; Sun et al., 2018; Wang et al., 2014; Zhang et al., 2014) and some coastal wetlands (Zheng et al., 2017; Long et al., 2019). In contrast relatively few studies have focused on the mechanisms regulating mercury in wetland ecosystems in the northern cold and arid regions. Therefore, the present study adopted Wuliangsuhai Lake as a study area as representative of a cold-arid lake region and aimed to explore the distributions of total mercury and methylmercury in different media and regulating factors. The results of the present study can guide the prevention and control of mercury in water ecosystems in cold and arid regions in northern China.

Materials and methods

Study area

Wuliangsuhai Lake is in Wulateqianqi County, Bayannaoer City, Inner Mongolia (40°36’–41°03’ N, 108°43’–108°57’ E) and represents one of a few lakes of large grassland desert and semi-desert areas on Earth. The lake is a typical shallow cold-arid region lake which is of great significance to the ecological security of the Yellow River Basin. The lake has an elongated shape extending in a northeast direction and had a total area of 341 km^2 in 2020. Currently, the volume of the lake is maintained between 400 million–500 million m^3. The lake falls in the northern cold and arid region with a typical mid-temperate continental monsoon climate characterized by low rainfall and high evaporation with clear seasonality. The ice thaw period starts in late March to early April, with the ice-free period extending from then to November and the ice period lasting about five months.

The present study established 20 sampling points in accordance with the lake and wetland survey standards for China and the characteristics of Wuliangsuhai Lake (Fig. 1). Of these sampling sites, 5 and 15 were in the phragmites area and water area, respectively, to obtain samples that are fully representative of the range of ecological characteristics of different regions of Wuliangsuhai Lake.

Collection of samples

Collection of water samples

Sampling was conducted from July to August 2022. Surface water samples were collected using a Niskin (5L) sampler. Water samples were collected in duplicate at each sampling point, with one stored in a 500 mL borosilicate glass bottle and the other

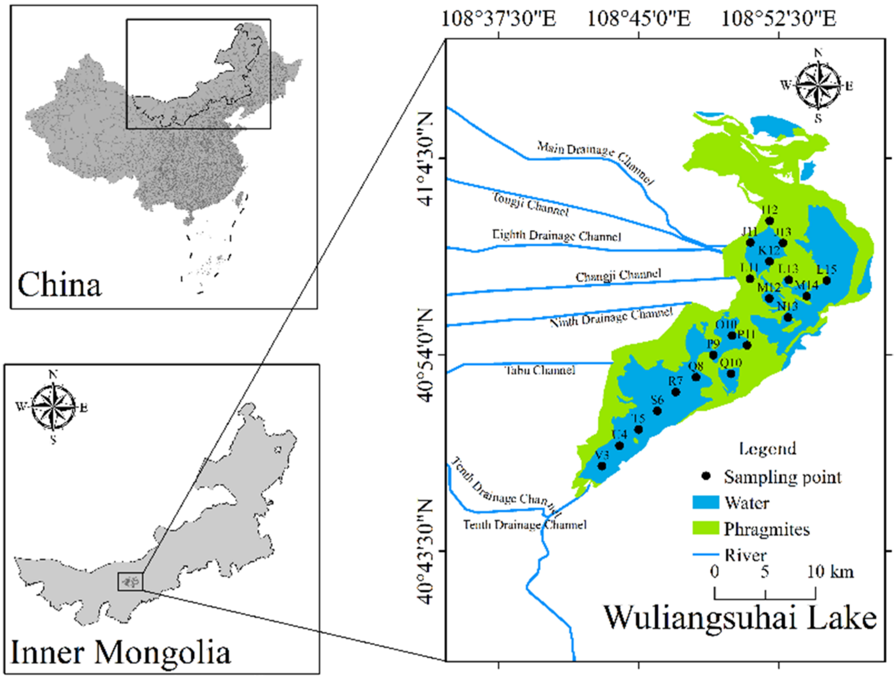

Fig. 1 Wuliangsuhai Lake study area and sampling points

stored in a 500 mL polytetrafluoroethylene (Teflon) bottle. To prevent volatilization of mercury, after the water samples collected, the polytetrafluoroethylene bottles were filled with two bags and stored in a dark refrigerator at 4 °C. Sample bottles were washed three times with the sample source water before sampling. Electronic grade (MOS) 1% HCl was added to the water samples stored in the borosilicate glass bottles on site, and these samples were used to determine THg. Electronic grade (MOS) 0.4% HCl was added to the samples stored in polypropylene glass bottles on site, and these samples were used for the determination of total methylmercury. The samples were stored at 4 ℃ in an incubator and transported back to the laboratory. Sample processing was completed within two weeks of collection. During sampling, water temperature, pH, dissolved oxygen (DO), total dissolved solids (TDS), and redox potential (ORP) were measured on-site (Yan et al., 2003).

Collection of surface sediment samples

Surface sediment samples were collected using a grab collector. After ensuring all sediment had settled, overlying water was carefully removed, and surface sediment within the 0–5 cm layer was collected. Surface sediment samples were collected in triplicate at each sampling site. The sediment samples were stored in 250 mL polyethylene bottles. Sample bottles were placed in labeled double-layer plastic bag packaging with labels indicating the sampling time, location, and sample number. Samples were refrigerated and transported to the laboratory for processing. At the laboratory, samples were dried using a vacuum freeze-drying instrument. After drying, each sample was ground through a 100-mesh sieve and stored in a brown glass bottle. Sediment samples were used to determine total mercury (THg) and total methylmercury (TMeHg).

Sample analysis

Analysis of THg and TMeHg in water

The THg of lake water samples was determined following the United States Environmental Protection Agency (USEPA) method 1631. Under this method, 0.125 mL BrCl (0.2 N) was added to 25 mL of the lake water sample and allowed to react for 12 h, after which 0.0625 mL $NH_2OH{\cdot}HCl$ (30% w/v) was added to remove residual BrCl. Then, 0.125 mL $SnCl_2$ (20% w/v) was added to convert all Hg^{2+} to Hg^0. Generated Hg^0 was then detected using a MERX automated modular Hg system (Brooks Rand Labs, Seattle, WA, USA).

Lake water sample TMeHg was determined following the USEPA method 1630. Under this method, 45 mL of the lake water sample was distilled at 125 ± 3 ℃ under the addition of 90 ± 10 mL/min N_2 for 3–4 h, thereby reducing the sample to 35. Then, 65 mL de-ionized (DI) water was added, and the mixture was reacted with 50 μL $NaBEt_4$(1% w/v) for 15 min. The sample was then purged with 200 mL/min N_2 for 15 min to trap generated methyl ethyl mercury on a Tenax-TA trap (35/60 mesh, Supelco, Bellefonte, PA, USA). The methyl ethyl mercury on the Tenax trap was then thermally desorbed at 200 ℃, separated using an OV-3 column at 70 ℃, decomposed to Hg^0 at 800 ℃, and finally detected using a Model III AFS (Brooks Rand Lab., Seattle, WA, USA) (United States Environmental Protection Agency, 2001).

Analysis of THg and TMeHg in the surface sediment

THg of sediment samples was determined using a modified USEPA 7474 method. Under the method, ± 0.2 g freeze-dried sediment sample was placed in an ampoule bottle, to which a mixture of concentrated 2 mL HNO_3 + 1 mL ultrapure water was added and allowed to stand for 20 min. The mixture was then sealed with a flame gun and placed in an autoclave (105 °C) for digestion for one h. After digestion, 200 μL of the solution was added to a chromatographic bottle, and the solution was reduced with 20% $SnCl_2$ and quantified using cold vapor atomic fluorescence spectroscopy (CVAFS) (Ma et al., 2019; United States Environmental Protection Agency, 2001).

Sediment TMeHg was determined using the USEPA 1630 method. Under the method, 0.5 g freeze-dried sediment was placed in a 50-mL centrifuge tube. A mixture of 5 mL KBr/$H2SO_4$ (18%, m/V) and 1 mL $CuSO_4$ (1 mol/L) solution was added to the sample. The mixture was then shaken at 200 rpm for 12 h in a constant temperature shaker at 25 °C. Then, 10 mL dichloromethane was added and the solution was shaken at 200 rpm for 1 h to extract TMeHg. Subsequently, 10 mL dichloromethane was added to extract TMeHg. Next, 2 mL dichloromethane and 35 mL ultrapure water was added into a centrifuge tube, and the solution was heated in water bath at 45 °C with nitrogen purging until no visible solvent was left. The distillate was then pH adjusted and ethylated using $NaBEt_4$. Finally, the samples were quantified using gas chromatography (GC)-CVAFS (United States Environmental Protection Agency, 2001).

Quality control

Borosilicate glass bottles used to store samples were soaked beforehand in a 30% volume ratio nitric acid solution for 24 h, following which they were heated at 500 ℃ for 40 min. After cooling, the bottles were coated with three layers of polyethylene film on an ultra-clean test table. Quality control of sample analysis was ensured through the use of parallel experiments, standard materials, and blank controls. Three blank samples were established for each batch of samples, and relevant tests were conducted using the samples to ensure no contamination in the experiment. Test results showed that the blank values were 1 ~ 4% of the measured sample value. Of each sample batch, 20% of samples were processed in triplicate for calculating the mean and relative standard deviation. In addition, of each sample batch, 20% of samples were randomly selected for spike recovery, with the measured spike recovery rate between 82 ~ 112%. The standard substances for THg in water, methyl mercury in water, THg in surface sediment, and methylmercury in surface sediment were GSBZ50016-90, GBW (E) 083364, GBW07405 (GSS-5), and ERMCC580, respectively.

Risk assessment guidelines

Concentrations of THg, TMeHg and other physical–chemical properties in water bodies refer to national standards in China (GB3838-2002). Sediment quality guidelines (SQGs) have been applied to interpret chemical data in the context of potential biological effects in sediments according to its toxicity potential. The SQGs developed by the Canadian Environmental Agency present the TEL (threshold effect level) and PEL (probable effect level). Under the SQG method, the Threshold Effect Limit represents the critical effect concentration, whereas the Probable Effect Limit (PEL) represents the possible effect concentration. The occurrence of a heavy metal within the environment at a concentration below the TEL is unlikely to have an ecotoxic effect. The occurrence of the heavy metal at a concentration between the TEL and PEL can result in an occasional ecotoxic effect. Finally, the occurrence of the heavy metal at a concentration exceeding the PEL is likely to result in an ecotoxic effect. The TEL and PEL for Hg are 174.00 μg/kg and 486.00 μg/kg, respectively.

Data statistics and analysis

Data were summarized using Microsoft Excel 2020. Sampling point plots were generated using Arcgis10.7 software. The present study applied Spearman correlation analysis to identify factors influencing mercury chemical speciation in water and sediments. Spearman correlation analysis was performed in SPSS25. All plots were generated in Origin2021.

Results and discussion

Environmental parameters of Wuliangsuhai Lake water

On-site measurement of water temperature (T), pH, dissolved oxygen (DO), conductivity (EC), REDOX potential (ORP), salinity (SAL), and total dissolved solid (TDS) of Wuliangsuhai Lake water was conducted; whereas, chlorophyll-a (chl-a) was analyzed in the laboratory (Table 1). As shown in Table 1, Wuliangsuhai Lake is weakly alkaline. Measured ORP indicated that lake water provided a good oxidative environment in summer due to a higher water temperature of between 22.71 ℃ and 25.84 ℃. This higher water temperature may favor mercury methylation in water bodies. High salinity, conductivity, and TDS during summer indicated seasonal salinization. Moderate levels of chl-a were measured in lake water, whereas high microbial activity may promote mercury methylation.

Environmental parameters of surface sediment of Wuliangsuhai Lake

Table 2 shows a summary of measured environmental parameters of surface sediment of Wuliangsuhai Lake. Since China has not established national or industrial standards related to sediments, and SQG is mainly used to evaluate toxicological indicators of sediments, the current study selected the specification of land quality geochemical assessment (DZ / T0295--2016) for reference. As shown in Table 2, pH of the surface sediment of the lake area was variable, ranging between 7.28 and 7.77, indicating an overall weak alkalinity. TN, TP, and organic matter OM in sediments ranged between 0.68 ~ 3.50 g/kg, 0.18 ~ 1.21 g/kg, and 8.17 ~ 85.67 g/kg, respectively, falling within the secondary standard of the soil nutrient classification of China (DZ / T0295- -2016). The process of mercury methylation may be facilitated by abundant nutrients. The coefficients of variation of total nitrogen (TN), total organic carbon (TOC), NH_4^+-N, and OM all exceeded 35%, indicating their high variability across space. Moderate levels of EC, TP, and total iron (TFe) were noted. These results indicated that certain anthropogenic factors affect the physical and chemical properties of surface sediments. The distribution of nutrients in the surface sediments of other lakes in China is as follows. In Chaohu Lake (Yang et al., 2020a), the surface sediment TN content ranged between 0.37 and 5.23 g/kg with an average of 1.46 g/kg, the surface sediment TP content ranged

Table 1 A summary of measured water quality variables of Wuliangsuhai Lake

Physicochemical indices	pH	DO (mg/L)	T (℃)	ORP (mv)	EC (μs/cm)	Salinity (mg/L)	Chl-a (μg/L)	TDS (mg/L)
Maximum	9.33	12.11	27.20	201.41	3698.20	2.08	14.29	2554.51
Minimum	8.83	1.43	21.72	116.44	1330.30	0.57	1.87	877.54
Mean ± standard deviation	9.08 ± 0.16	6.91 ± 2.27	23.91 ± 1.30	152.32 ± 30.11	2291.30 ± 758.40	1.21 ± 0.45	7.03 ± 3.24	1533.53 ± 528.90
Median	9.03	6.85	23.85	135.15	2190.50	1.16	7.31	1462.25
Variation	1.76%	32.84%	5.45%	19.77%	33.10%	37.14%	46.04%	34.49%
GB5749-2022	6.5 ~ 8.5	< 2	—	> 400	—	< 1000	—	1000

Abbreviations: *DO* dissolved oxygen, *T* temperature, *ORP* redox potential, *Chl-a* chlorophyll-a from 0.34 to 1.75 g/kg with an average of 0.90 g/kg, and the surface sediment NH_4^+-N content ranged from 0.06 to 0.54 g/kg with average 0.10 g/kg. Compared with Baiyang lake which is also a shallow lake (Wang et al., 2022), the ranges of pH, ORP, and EC values were in the range of 6.99 ~ 8.21, − 211.00 ~ 81.00 mV, and 403.00 ~ 837.00 µs/cm, respectively. TN, TP, and TOC in Baiyangdian Lake were in the range of 0.38 to 4.91 g/kg, 0.51 to 1.23 g/kg, and 2.81 to 70.30 g/kg. Compared with other lakes in China, the surface sediments of Wuliangsuhai Lake were higher TN content and lower TP content, and the contents of other nutrients such as TOC are like those of other lakes. The analysis of water ORP data showed that Wuliangsuhai Lake is in a strong oxidation state during the study period. It would stimulate the release of nutrients in the surface sediments and increase its content. Besides, the oxidation and reduction of sediments have a great influence on the level distribution of nutrients. Meanwhile, the presence of phragmites would affect the sedimentation rate of nutrients in the water, caused significant variabilities in nutrient contents in different regions. From the perspective of spatial distribution, the areas with high contents of nutrients were mainly concentrated near channel and drainage. These areas were greatly affected by farmland wastewater, industrial effluents, and domestic sewage. Water bodies carried many nutrient salts into the lake, migrated with the current and concentrated in the center of the lake. Therefore, the distribution of nutrients changed greatly.

Table 2 A summary of measured surface sediment quality variables of Wuliangsuhai Lake

Physicochemical indices	pH	TOC (g/kg)	TN (g/kg)	EC (µs/cm)	TP (g/kg)	TFe (mg/kg)	NH_4^+-N (mg/kg)	OM (g/kg)
Maximum	7.77	44.32	3.52	1969.20	1.21	39,800.00	44.39	85.67
Minimum	7.28	5.84	0.68	505.30	0.18	15,322.00	2.79	8.17
Mean ± standard deviation	7.50 ± 0.13	18.80 ± 11.91	1.90 ± 0.82	1169.10 ± 368.50	0.54 ± 0.18	24,869.00 ± 7839.12	19.57 ± 10.36	33.91 ± 20.35
Median	7.47	12.98	1.94	1065.51	0.52	23,333.00	17.25	29.89
Variation	1.79%	63.36%	43.36%	32.64%	33.26%	29.51%	52.94%	60.03%

Abbreviations: *TOC* total organic carbon, *EC* electrical conductivity, *TP* total phosphorus, *OM* organic matter

Characteristics of THg and TMeHg in lake water

The concentration of THg in Wuliang Lake ranged between 19.20 ~ 668.10 ng/L (mean of 192.15 ng/L) whereas that of TMeHg ranged between 0.01 ~ 11.40 ng/L (mean of 1.08 ng/L). The rate of methylation ranged between 0.05 and 9.67%. The average concentration of THg in Wuliangsuhai Lake water exceeded the surface water environmental quality (GB3838-2002) standard. The THg of 14 sampling points exceeded the class V standard, indicating higher mercury pollution of water. The background concentration of THg in water bodies is accepted to be below 5.00 ng/L (Ma et al., 2019), with any value above this generally considered to be related to the contents of particulate matter (PM) and OM (Bi et al., 2006). Past related studies have shown seasonality in

Table 3 Comparisons of analyzed total mercury (THg) and total methylmercury (TMeHg) values of surface water quality of Wuliangsuhai Lake with those of other regions

Study area	THg (ng /L)			TMeHg (ng/L)			References
	Maximum	Minimum	Mean	Maximum	Minimum	Mean	
Yaer Lake	48.60	6.20	16.80	3.62	0.22	1.10	(Chen et al., 2021)
Taihu Lake	83.00	6.80	28.00	0.81	0.04	0.14	(Wang et al., 2012)
Dahong Lake Reservoir	8.59	1.50	4.33	0.62	0.24	0.33	(Guo et al., 2018)
Nansi Lake	6.83	2.95	4.51	0.25	0.16	0.22	(Yang et al., 2020b)
Sanmenxia Reservoir	9.65	1.65	4.05	0.36	0.06	0.14	(Cheng et al., 2017)
Xiaolangdi Reservoir	3.30	0.84	1.72	–	–	–	(Cheng et al., 2015)
Daya Bay	45.00	12.00	23.00	–	–	0.06	(Tao et al., 2016; Liu et al., 2022a, 2022b)
Jiaozhou Bay	9.87	2.47	4.80	0.83	0.08	0.44	(Mao et al., 2020)
Wuliangsuhai Lake	668.10	19.20	192.15	11.40	0.10	1.08	This study
Lake Zapotlan, Mexico	10.70	0.90	3.00	0.12	0.01	0.03	(Malczyk & Branfireun, 2015)
Massachusetts Lake, USA	6.56	0.56	2.47	0.20	0.04	0.10	(Wang & Obrist, 2021)
The Great Salt Lake, USA	55.00	24.00	29.00	25.50	1.10	7.40	(Johnson et al., 2015)
The Vembanad Lake, India	27.60	4.67	15.39	0.30	0.01	0.09	(Mohan et al., 2021)

– Indicates that the relevant data are not queried or are not listed in the paper

PM of Wuliangsuhai Lake water, with a yearly peak in July (Geng et al., 2021). Therefore, the higher THg concentration in Wuliangsuhai Lake may be related to the external input of PM. The THg concentration of Wuliangsuhai Lake water was relatively high compared to those of natural water bodies in China and globally (Table 3). The average concentration of THg of Wuliangsuhai Lake water far exceeded those of comparable lakes in China, such as the Taihu and Nansi lakes in East China, the Yaer Lake in Central China, and Dahong Lake in Southwest China. The average concentration of THg of Wuliangsuhai Lake water also exceeded those of coastal areas in China, such as Jiaozhou Bay and Daya Bay. In addition, the THg concentration of Wuliangsuhai Lake water far exceeded those of some reservoirs such as the Xiaolangdi and Sanmenxia reservoirs in the Yellow River Basin. The THg concentration of Wuliangsuhai Lake water also exceeded those of water bodies internationally, including Lake Zapotlan in Mexico, Salt Lake in Massachusetts, and the Great Salt Lake in Utah, and Lake Vembanad in India.

The average concentration of TMeHg in Wuliangsuhai Lake water was 1.08 ng/L, far exceeding that of unpolluted natural water bodies of 0.01 ~ 0.10 ng/L (Shang et al., 2004). Around 15% of sampling sites showed a concentration of TMeHg that exceeded the limit for surface water of 1.00 ng/L. The concentration of TMeHg in Wuliangsuhai Lake water exceeded those of other lakes in China, including the Taihu, Dahong, and Nansi lakes; whereas, it was comparable to that of Yaer Lake in central China. The concentration of TMeHg in Wuliangsuhai Lake water also far exceeded that of reservoirs in China, such as the Sanmenxia and Xiaolangdi reservoirs in the Yellow River Basin. The TMeHg concentration of Wuliangsuhai Lake water also generally exceeded that of representative bays along the southeast coast of China, such as Daya Bay and Jiaozhou Bay. The concentration of TMeHg of Wuliangsuhai Lake water also far exceeded those of some lakes globally, such as Lake Zapotlan, Lake Massachusetts, and Lake Vembanad; whereas, it was less than that of the Great Salt Lake in Utah, which is also a high-altitude inland lake.

As shown in Fig. 2, higher concentrations of THg in the water of Wuliangsuhai Lake occurred close to sampling points U4 and V3 at the estuary, and at L11, L13, and O10. The concentration of THg in lake water generally increased from north to south. The spatial distribution of TMeHg in water was like that of THg, with the maximum at T5 in the open water area. The highest methylation rate also occurred at this sampling point, whereas the high contents of salinity and TDS at this site supported mercury

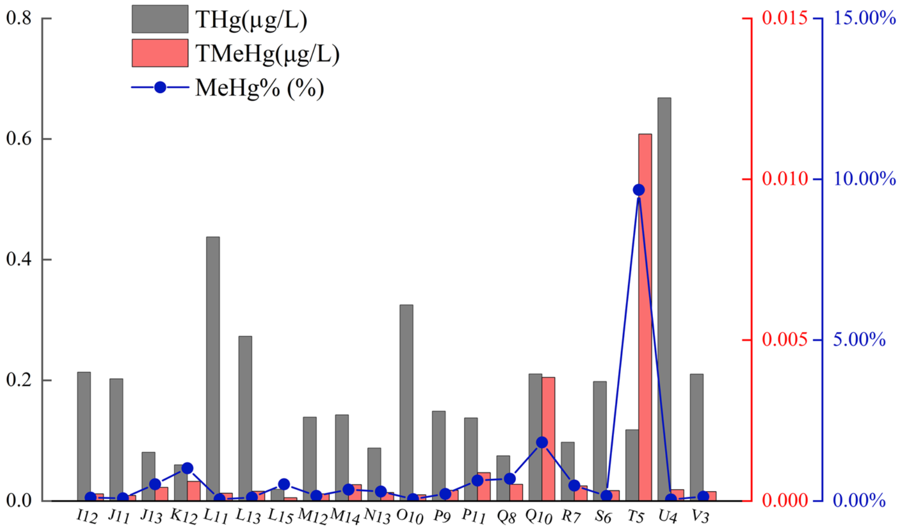

Fig. 2 Spatial distributions of different forms of mercury in Wuliangsuhai Lake water

Table 4 Comparison of total mercury (THg) and total methylmercury (TMeHg) values in Wuliangsuhai Lake surface sediments with those in other regions

Study area	THg (μg/kg)			TMeHg (μg/kg)			Reference
	Maximum	Minimum	Mean	Maximum	Minimum	Mean	
Yaer Lake,	2766.00	44.00	547.00	2.60	–	0.80	(Chen et al., 2021)
Taihu Lake	470.00	12.00	77.00	0.96	0.20	–	(Wang et al., 2012)
Dahong Lake	–	–	200.00	–	–	0.33	(Guo et al., 2018)
Nansi Lake	88.10	17.32	38.15	1.01	0.12	0.36	(Yang et al., 2020a, 2020b)
Sanmenxia Reservoir	95.82	55.89	80.06	0.74	0.27	0.52	(Cheng et al., 2017)
Xiaolangdi Reservoir	172.52	95.66	122.28	0.39	0.18	0.28	(Cheng et al., 2015)
Daya Bay	92.00	31.00	46.00	–	–	0.13	(Tao et al., 2016; Liu et al., 2022a, b)
Jiaozhou bay	145.16	24.60	53.11	1.10	0.23	0.63	(Mao et al., 2020)
Wuliangsuhai Lake	480.10	110.00	261.85	0.49	0.02	0.18	This study
Lake Zapotlan, Mexico	862.72	14.59	235.40	0.03	–	0.02	(Malczyk & Branfireun, 2015)
Massachusetts Lake, USA	–	–	23.00	–	–	0.90	(Wang & Obrist, 2021)
The Great Salt Lake, USA	260.00	90.00	122.00	0.85	0.11	0.44	(Johnson et al., 2015)
The Vembanad Lake, India	2054.33	0.36	406.67	73.81	0.12	5.95	(Mohan et al., 2021)

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methylation in water. Secondly, the concentration of THg and level of methylation in the Q10 phragmites area far exceeded those measured at other sampling points. The anaerobic conditions at each sampling point facilitated mercury methylation in water. These results suggested that MeHg in water is mainly a result of *in situ* methylation of mercury.

Characteristics of THg and TMeHg in surface sediments

Table 4 provides a summary of concentrations of different forms of mercury in the surface sediment of the lake. THg content of the lake sediment ranged from110.00 to 480.10 μg/kg (average of 261.85 μg/kg). Lake sediments TMeHg ranged from 0.02 to 0.49 μg/kg (average of 0.18 μg/kg), whereas the rate

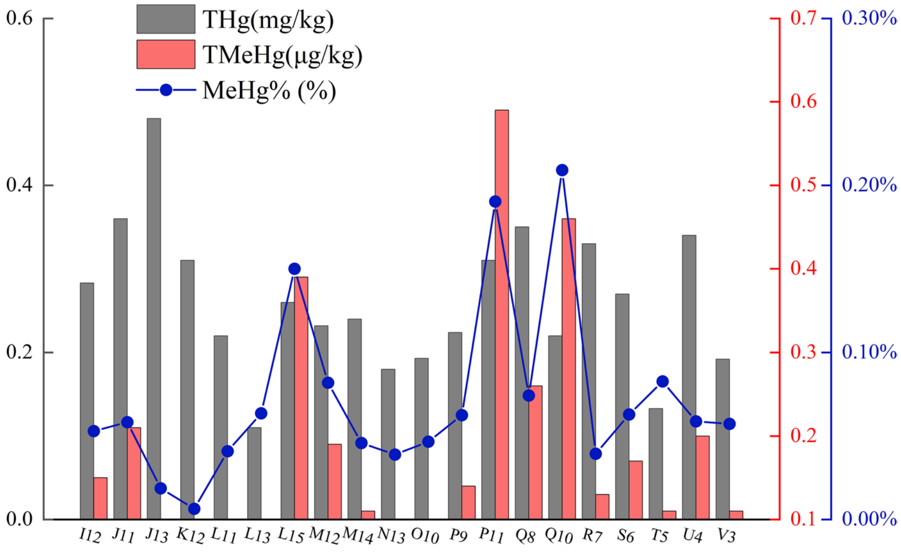

Fig. 3 Spatial distributions of different chemical species of mercury in Wuliangsuhai Lake surface sediments

of methylation ranged from 0.01 to 0.21%. Total mercury of surface sediment of Wuliangsuhai Lake far exceeded the background value of the drainage sediment in Inner Mongolia (18.00 μg/kg) (Liu et al., 2020). Since China has not yet established a national lake sediment quality standard, the current study selected the sediment quality benchmarks (SQGs) for evaluating the potential biological toxicity of sediment heavy metals in lakes, rivers, and other water bodies (MacDonald et al., 2000). These SQGs were used to assess the potential harm of THg to the environment and organisms in the Wuliangsuhai Lake sediment. The average THg values of 85% of sediment sampling sites of Wuliangsuhai Lake were between the TEL and PEL, with only that at site J13 close to the PEL. This result indicates that Hg pollution of surface sediments in Wuliangsuhai Lake can possibly result in certain ecotoxic effects and harm to the water environment. According to the above results, THg of surface sediment of Wuliangsuhai Lake is less than that of Yaer Lake in central China with a developed aquaculture industry and close to those of Dahong and Nansi lakes in southwest and East China, respectively. However, the THg of surface sediment of Wuliangsuhai Lake exceeded those of other water body categories, such as reservoirs and bays. At a global level, THg of the surface sediment of Wuliangsuhai Lake exceeded that of Lake Zapotlan, Lake Massachusetts, and the Great Salt Lake in Utah, and was less than that of Lake Vembanad in the chemical center of southern India.

The mean TMeHg and rate of methylation of surface sediments of Wuliangsuhai Lake calculated to be 0.18 μg/kg and 0.07%, respectively are typically used as a measure of the conversion efficiency of MeHg in sediments (Liu et al., 2020). This conversion efficiency is generally less than 0.50% in natural waters (Guo et al., 2018). Therefore, biological activity of inorganic mercury in the Wuliangsuhai Lake sediment may be low. The maximum and minimum TMeHg of surface sediment were measured at sites P11 and K12, respectively. P11 is in the phragmites area, and a previous study showed that the decomposition of phragmites leads to an increase in OM content in the aquatic environment (Wei et al., 2016). The high OM content and anaerobic environment (water DO at P11 was 1.43 mg/L) of Wuliangsuhai Lake provides an environment suitable for sulfate- and iron-reducing bacteria, thereby facilitating mercury methylation (Ma et al., 2017; Regnell et al., 2014; Sun et al., 2018) and increasing TMeHg. The TMeHg of Wuliangsuhai Lake was lower than those of some other lakes in China, such as Yaer, Nansi, and Taihu lakes, lower than that of Sanmenxia Reservoir, Xiaolangdi Reservoir, and Jiaozhou Bay in the Yellow River Basin, and lower than that of Jiaozhou Bay in the South China Sea. However, the average TMeHg of Wuliangsuhai Lake exceeded that of Big Honghu Lake in Southwest China. At a global level, TMeHg of surface sediment of Wuliangsuhai Lake far exceeded that of Lake Zapotlan, was comparable to that of Massachusetts

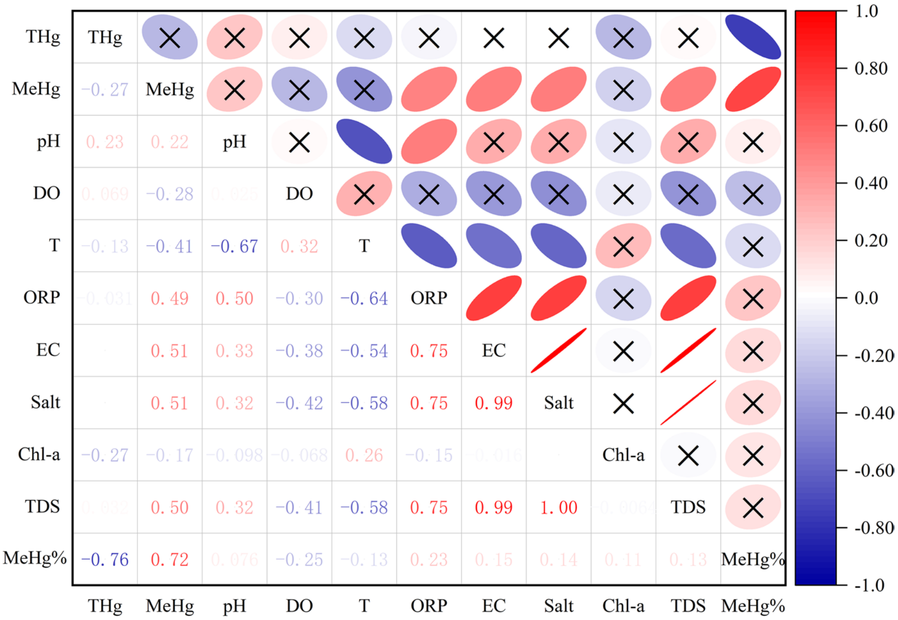

Fig. 4 Correlations between environmental factors and different mercury chemical species in water of Wuliangsuhai Lake

Salt Lake, and was less than that of the Great Salt Lake in Utah and Vembanad Lake in India, with the latter greatly affected by the chemical industry.

As shown in Fig. 3, there were clear differences in the spatial distribution of THg in the surface sediment of Wuliangsuhai Lake, with a coefficient of variation of 32.30%. The maximum THg of surface sediment was found at site J13 in the phragmites area, with high levels also found at sites J11 and Q8 close to the lake inflow and at sites I12, K12, and P9 near the phragmites area. These results showed that THg of surface sediment generally increased from the center of the lake to the north and south. The spatial distribution of surface sediment TMeHg in the lake differed from that of THg, with TMeHg decreasing from the central lake region to the south and north. Some exceptions to this trend were TMeHg at sites P11 in the largest phragmites area and at sites Q10 and L15 close to the phragmites area. The sites P11, Q10, and L15 showed high rates of methylation, whereas they had above-average OM concentrations of 81.43 g/kg, 49.09 g/kg, and 45.08 g/kg, respectively. The distribution of mercury in the surface sediments of Wuliangsuhai Lake appeared to be influenced by OM, consistent with the results of previous studies (Long et al., 2020; Mao et al., 2020; Liu et al., 2022a, b). Therefore, the biochemical effects of the phragmites and the distribution of OM may be important sediment factors affecting mercury methylation.

Environmental factors regulating chemical speciation of mercury in water

As shown in Fig. 4, the results showed lower relationships between environmental factors and THg, indicating the possibility of exogenous input. This result, in combination with the findings of previous studies which showed that atmospheric deposition contributes as much as 46.40% of THg in water, indicated that most oxidized Hg in the atmosphere will generate soluble bivalent Hg ions in water (Yin et al., 2014). These results showed that THg in water is greatly affected by inputs of Hg from the atmosphere and drainage. The lack of a significant correlation between THg and TMeHg ($P > 0.05$) indicated differences in the sources and transformation of inorganic mercury and methylmercury among the different water bodies of the Wuliangsuhai Lake area and that *in situ* methylation in lake water is not the main source of MeHg in the Wuliangsuhai Lake area. TMeHg showed significantly positive correlations with ORP, EC, salinity, and TDS. Previous studies (Mao et al., 2020) have shown that Cl^- can affect the bioavailability of methylated bacteria in water, which in turn affects the formation and accumulation of methylmercury. Monitoring of conventional physical and chemical indicators in the Wuliangsuhai Lake indicated high levels of salinity and TDS in summer as well as a positive correlation between these two

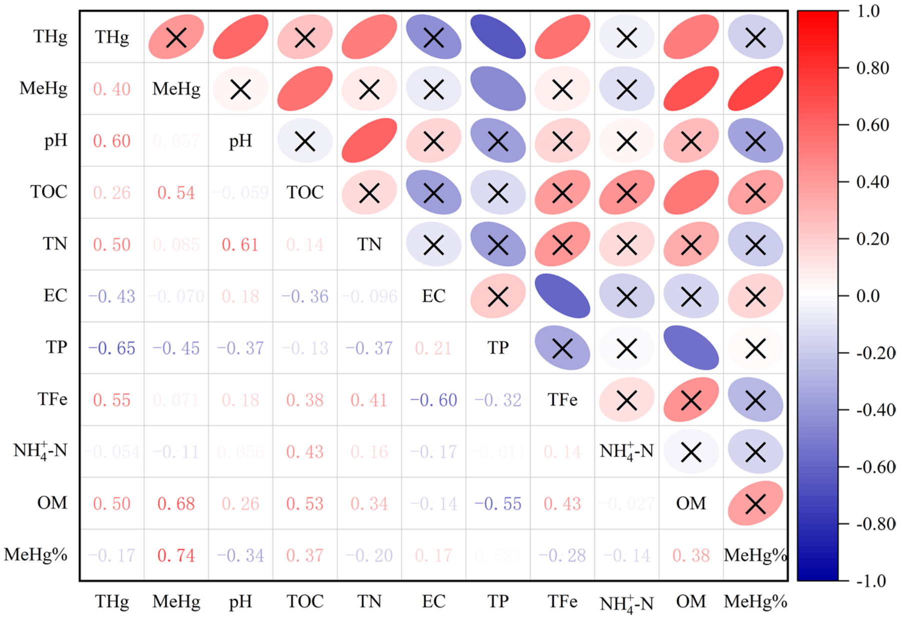

Fig. 5 Relationships between environmental factors and chemical speciation of mercury in surface sediments of Wuliangsuhai Lake

variables. This result indicated that Cl^- is the soluble substance in water contributing to this relationship. A high Cl^- content may affect the levels of TMeHg. Although TMeHg was significantly correlated with ORP, summer oxidization was not conducive to mercury methylation. EC was significantly correlated with both MeHg and ORP, indicating that MeHg and ORP were greatly affected by EC. These results indicated that the potential for mercury methylation in water is greatly influenced by the concentration of soluble ions in the water.

Factors affecting mercury chemical speciation in sediment

Figure 5 shows the results of Spearman correlation analysis to identify environmental factors affecting the chemical speciation of mercury in surface sediment. The results showed that THg in surface sediment had significantly negative correlations with pH, TN, TFe, OM, and TP ($P<0.05$), which indicates that oxidation of TFe may result in the precipitation of THg (Cesário et al., 2017). pH is an important factor affecting mercury methylation in a wetland ecosystem as this factor can indirectly affect the morphology of mercury by influencing microbial activity (Duan et al., 2021) and acidic conditions (low pH) often result in the stimulation of mercury methylation in sediments (Zhao et al., 2016). The weak alkalinity of surface sediment of Wuliangsuhai Lake hinders mercury methylation, leading to higher THg. TMeHg showed significant positive correlations with OM and TOC ($P<0.05$). OM can affect the distribution of sediment methylmercury by affecting redox conditions, and the decomposition of OM will promote the consumption of surface oxygen and the reduction of sulfur in the sediment (Azaroff et al., 2019). OM molecules also serve as a source of energy and nutrients within the stimulation of methylation groups, such as sulfate-reducing bacteria (SRB) (Lei et al., 2019), leading to the conversion of inorganic mercury to MeHg. TOC can accelerate aerobic decomposition of OM, resulting in reduced sediment surface DO, thus enhancing methylation, consistent with findings of previous related studies (Chen et al., 2021; Liu et al., 2015; Wang et al., 2012). These results indicated that mercury methylation in the surface sediment of Wuliangsuhai Lake was greatly affected by sources of carbon.

Conclusions

The average concentrations of THg and TMeHg in the water bodies of the Wuliangsuhai Lake area were 192.15 ng/L and 1.08 ng/L, respectively, with the former exceeding that of other natural water bodies in other regions and the Chinese surface water environmental quality standard (0.10 μg/L). The concentration of TMeHg in the water of Wuliangsuhai Lake exceeded MeHg in unpolluted natural water bodies and the Chinese surface water environmental quality standard (1.00 ng/L). MeHg of Wuliangsuhai Lake water exceeded that of natural water bodies and reservoirs both in China and globally. The results indicated relatively serious mercury pollution of Wuliangsuhai Lake water.

Average surface sediment THg of Wuliangsuhai Lake was 261.85 μg/kg, far exceeding the background concentration of drainage sediment in Inner Mongolia and at a moderate level in comparison with natural domestic water. SQG showed that surface sediment THg of Wuliangsuhai Lake was at an ecotoxic level and potentially harmful to the water environment. Average surface sediment TMeHg of Wuliangsuhai Lake was 0.18 μg/kg, lower than those of other natural water bodies in China and globally. This result could be attributed to low levels of Hg methylation in surface sediments and lower bioactivity of inorganic Hg.

Environmental factors showed weak relationships with THg in water of Wuliangsuhai Lake. Therefore, the high concentration of THg could mainly be attributed to external inputs, such as atmospheric deposition. TMeHg in water showed significant positively correlations with ORP, EC, salinity, and TDS, and was greatly affected by soluble ions. There was no significant correlation between THg and TMeHg and the two chemical species were affected by different geochemical processes in water.

pH, TN, TP, TFe, and OM were shown to be the environmental factors significantly impacting on surface sediment THg of Wuliangsuhai Lake, whereas TOC, TP, and OM were the main factors affecting the surface sediment TMeHg. OM significantly affected methylation in surface sediments. This study preliminarily investigated the distribution of different species of Hg in the water and sediment of Wuliangsuhai Lake, indicating that the pollution level of Hg in Wuliangsuhai Lake is high, and the distribution of Hg is affected by multiple environmental factors. Our results could summarize some useful experience for exploring the geochemical cycle of Hg in typical lakes in cold and arid areas. Meanwhile, there also provide appropriate data support for upstream and downstream environmental protection in the future. It could contribute to the formulation of upstream and downstream environmental protection policies, guiding agricultural and industrial safety production, guaranteeing the health level of regional population, and maintaining the coordinated development of man and nature.

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Author contributions All authors contributed to the study conception and design. ZC was involved in the methodology, software, writing–original draft preparation, writing–reviewing and editing; XS and SZ provided the research ideas and experimental equipment; JL was involved in the methodology, software, writing–original draft preparation, writing–reviewing and editing; ZT, HZ, XG, and YW completed the experimental operation and the arrangement and processing of experimental data.

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Declarations

Conflict of interest The authors have no relevant financial or non-financial interests to disclose.

Ethics approval We assured that this study did not involve any ethical experiments.

Consent to participate All authors were participated in this work.

Consent to publish All authors give the consent for the publication of identifiable details, which can include figures, tables, and details within the text (all authors declare that they have no objection) to be published in this Journal.

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