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Diet influence on mercury bioaccumulation as revealed by polyunsaturated fatty acids in zoobenthos from two contrasting environments: Chinese reservoirs and Swedish lakes

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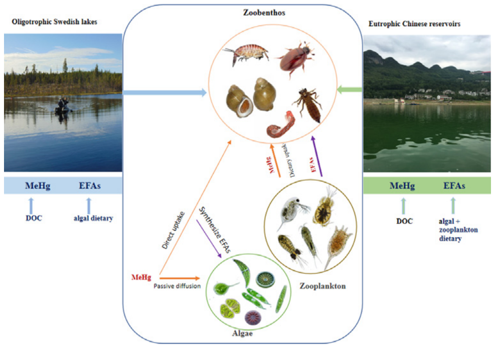
HIGHLIGHTS

- Comparing Hg and PUFA bioaccumulation of zoobenthos in two aquatic ecosystems
- DOC concentration of water influence Hg of zoobenthos in Swedish lakes.
- Plankton dietary increases MeHg content of zoobenthos in Chinese reservoirs.
- EFA contents in zoobenthos associated with both composition and density of algae
- Abiotic factors influence EFA accumulation of zoobenthos in Chinese reservoirs.

GRAPHICAL ABSTRACT



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ABSTRACT

The bioaccumulation of mercury (Hg) and polyunsaturated fatty acids (PUFA) in zoobenthos varies across aquatic food webs. In this field study, contents of total Hg (THg), methylmercury (MeHg) and PUFA were investigated in zoobenthos of Chinese reservoirs and Swedish lakes, with contrasting environmental characteristics and algal diet sources, which can result in difference of Hg and PUFA in zoobenthos from these two habits. Using PUFA as dietary biomarkers of algae in zoobenthos, we evaluated effects of environmental factors and algal diet sources on the accumulation of THg, MeHg, and the highly required PUFA eicosapentaenoic (EPA, 20: 5n-3) and docosahexaenoic acid (DHA, 22:6n-3) in zoobenthos. Average THg and MeHg in zoobenthos were higher in Chinese reservoirs than Swedish lakes ($p < 0.05$). Average EPA content of zoobenthos was similar in these two habitats ($p > 0.05$), yet average DHA content of zoobenthos was higher in Chinese reservoirs than Swedish lakes ($p < 0.05$). Total Hg and MeHg contents of zoobenthos in Swedish lakes were predicted by environmental factors; e.g., negatively with pH and positively with dissolved organic carbon (DOC) concentrations, yet had no significant relationship with the algal dietary. In Chinese reservoirs, however, no environmental factor correlated well with THg contents in zoobenthos, and only DOC concentrations showed positive correlation with MeHg contents in zoobenthos. Besides, the algal dietary was also positively correlated with MeHg contents in zoobenthos. EPA and DHA contents of zoobenthos in Swedish lakes primarily associated with algal diet. By contrast, in Chinese reservoirs, EPA and DHA contents of zoobenthos were affected by both environmental factors and algal diet.

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1. Introduction

Fish provide important dietary nutrients, such as essential proteins and lipids, to humans. In particular, fish are generally rich in polyunsaturated fatty acids (PUFA), nutrients which are beneficial to human health, especially the omega-3 (n-3) long-chain (LC) PUFA eicosapentaenoic (EPA, 20:5n-3) and docosahexaenoic acid (DHA, 22:6n-3) (Pavlicevic et al., 2014; Rhee et al., 2017). EPA and DHA are associated with lower indices of cardiovascular diseases, and support cognitive functions (Zhang et al., 2019). However, fish is also a major contributor to the potent neurotoxin mercury (Hg) exposure to humans (Vieira et al., 2020). Methyl Hg (MeHg) impairs the central and peripheral nervous systems, and can produce harmful effects on the cardiovascular system and kidneys (Duan et al., 2016; Hu et al., 2017; Rosa-Silva et al., 2020). As results of dietary uptake, both Hg and PUFA in fish are conveyed from the base of aquatic food webs mainly consisted of primary producer e.g. phytoplankton and consumers e.g. zoobenthos. It is necessary to realize that Hg and PUFA in zoobenthos are also co-obtained from their diet (Parrish et al., 2000), but influence consumers differently. PUFA are important building blocks of cell membrane and essential for metabolic activity. In particular, the n-3 LC-PUFA EPA and DHA are referred to as essential fatty acids due to their vital role in supporting survival, somatic growth and reproductive success of organisms. In contrast to the physiologically important nutritional function of PUFA, Hg does not serve any beneficial physiological purpose in organisms. Thus, there is a need to investigate the bioaccumulation of PUFA and MeHg in consumers simultaneously.

Trophic transfer patterns of Hg and PUFA in food chains can be affected by both abiotic (water chemistry) and biotic factors (dietary type) (Jing et al., 2020; Razavi et al., 2014). There is evidence that the Hg content in organisms is often related to abiotic factors, such as Hg influx from wetlands and forest harvest, pH, and DOC concentration of water (Bishop et al., 2020; Broadley et al., 2019; Clayden et al., 2013; De Wit et al., 2014). Environmental change can also result in changes of fatty acids in aquatic organisms. For example, water contamination (Mahboob et al., 2019), lake eutrophication and brownification can downgrade the availability and transfer of PUFA that are required for the development of aquatic consumers, such as fish. Despite this, the primary pathway of Hg and PUFA accumulation is through dietary for organisms occupying higher trophic positions (Hall et al., 1997; Arslan et al., 2008; Kainz and Fisk, 2009). In aquatic systems, algae, which are the main primary producers, play a key role in determining PUFA and Hg availability. Hg bioconcentrates in phytoplankton from which it is transferred to higher trophic levels. Several studies reported that enhanced algal biomass decreases MeHg biomagnification (Charles T et al., 2007; Chen and Folt, 2005; Clayden et al., 2013). In addition, most PUFA in aquatic food webs are synthesized by phytoplankton and subsequently transferred to organisms at higher trophic levels. However, the fatty acid composition can differ considerably among phytoplankton taxa. For example, diatoms and Cryptophytes synthesize the essential EPA and DHA; whereas Chlorophyta and Cyanophyta mainly contain linoleic acid (LIN) and alpha-linolenic acid (ALA), which are essential precursors for EPA and DHA (Kainz and Fisk, 2009). Therefore, besides being essential for physiological functions of organisms, PUFA are also indicators of algal diet sources and transferred along aquatic food chains (Parrish et al., 2000). It is realized that the phytoplankton community structure can be influenced by the physical and chemical characteristics of the environment, including acidity/alkalinity, DOC, trophic status as well as light and temperature conditions of lakes which all respond to environmental forcing, including anthropogenic pressures (Maileht et al., 2013; Razavi et al., 2014; Strandberg et al., 2016). Due to environmental characteristics and dietary structure variation between habitats, Hg and PUFA bioaccumulation in aquatic systems exhibits spatial variability (Hussain et al., 2019).

Two largely contrasting aquatic habitat types have caught our attention; freshwater reservoirs in China and natural lakes in Sweden. Chinese reservoirs, where frequent aquaculture activities can be seen, are generally eutrophic and alkaline (Feng et al., 2018a, 2018b; Yan et al., 2013; Li et al., 2008; Shen et al., 2013; Razavi et al., 2014; Zeng et al., 2017), whereas lakes in Sweden are mostly oligotrophic, acidic and humic (Åkerblom et al., 2017; Bravo et al., 2018). These ecosystems differ considerably in the Hg and PUFA bioaccumulation of fish (Jing et al., 2020; Lau et al., 2012; Liang et al., 2017; Wu et al., 2018). Increasing nutrient concentrations in reservoirs are widely seen as an important regulator of low Hg and PUFA contents in fish from Chinese reservoirs because high nutrient levels (trophic status) shift the taxonomic composition of primary producers to species with low PUFA (e.g., cyanobacteria); Moreover, MeHg can be more highly sequestered by higher algal biomass (algal bloom dilution) (Noh et al., 2017; Pickhardt et al., 2002), which leads to lower MeHg concentrations in organisms at the base as well as at higher trophic levels along the food chain. It should be noted that farmed fish in Chinese reservoirs mainly feed on artificial feed in addition to natural feed, which reduces the trophic steps in the corresponding food webs (Feng et al., 2018a). In contrast, the primary production is much lower in oligotrophic lakes, such as those of Sweden with low pH, and high DOC concentrations. In such lakes, different phytoplankton taxa that are frequently dominated by diatoms and cryptophytes which supply relatively large amounts of EPA and DHA to consumers (Wu et al., 2019). On the other hand, low pH and high DOC concentrations can increase the MeHg bioavailability in organisms (Broadley et al., 2019; Jardine et al., 2013). Thus, these two contrasting aquatic ecosystems function differently with regards to energetics and ecotoxicology. This implies divergences in the trophic transfer and retention of potentially toxic Hg as well as highly required PUFA.

As an intermediate trophic link, benthic macroinvertebrates (zoobenthos) are major food sources for fish and play key conduits in aquatic food chains, but also convey contaminants from the base of food webs to fish (Vander Zanden and Vadeboncoeur, 2002). In fact, compared to fish, zoobenthos rely more on phytoplankton as food (Liu, 2006). As a result, the bioaccumulation of Hg and PUFA in zoobenthos is susceptible to changes in environmental conditions and phytoplankton composition, while hardly know about the effect of the anthropogenic activity of providing artificial feed to fish. Unfortunately, food web studies of Hg and PUFA accumulation often focus on higher trophic levels, and studies of Hg and PUFA transfer at the lower trophic levels of freshwater food webs are scarce.

Our goal is to quantify and compare the effects of abiotic (water chemistry) and biotic factors (dietary algae) factors on Hg and PUFA bioaccumulation in zoobenthos across two different ecosystem types: Chinese reservoirs and Swedish lakes. Here, we first compare the bioaccumulation patterns of Hg and PUFA in zoobenthos from these two freshwater ecosystems. The contents of THg, MeHg and PUFA of zoobenthos in Chinese reservoirs and Swedish lakes were examined. In addition, relationships between Hg, PUFA and water chemistry were investigated to discern potential sources of the variation of Hg and PUFA contents in zoobenthos. We hypothesize that: 1) higher abundance of diatoms and cryptophytes as algal diet contribute to higher contents of EPA and DHA and Hg in zoobenthos of Swedish lakes compared to Chinese reservoirs; 2) low pH and high DOC concentrations of Swedish lakes will increase Hg content and, due to shading effects of DOC, EPA and DHA contents in zoobenthos will be reduced, 3) eutrophication in Chinese reservoirs decrease Hg in zoobenthos due to algal bio-dilution, and enhance the biomass of Chlorophyta and Cyanophyta, which reduces EPA and DHA contents in zoobenthos.

1.1. Study sites and sample collection

During the summer of 2016, sampling was conducted in four reservoirs from Guizhou, Southwest China and from six lakes in Sweden, respectively. Sampling and Hg analysis in Chinese reservoirs and Swedish lakes were conducted in Institute of Geochemistry, Chinese Academy of

Sciences and Swedish University of Agricultural Sciences, respectively. The two laboratories adopted basically the same sampling and analysis methods. Fatty acids analysis was conducted in Lunz am See, Austria. The labs in this study participate in the Sino-Swedish Mercury Management Research Framework (SMaRef).

1.1.1. Sampling in Chinese reservoirs

The four Chinese reservoirs belong to the Wujiang River, which is one of the largest tributaries of the Yangtze River with a subtropical climate (Fig. 1). Before flooding, there were agriculture farmlands distributed along the valleys. These reservoirs are meso- to eutrophic (Feng et al., 2018b). The eutrophic Wujiangdu Reservoir (WJD) (27°19′10″N, 106°45′39″E) is the only reservoir remained with aquaculture now, characterized by the high-density of net caged fish production. Baihua Reservoir (BH) (26°39′49″N, 106°30′40E″) and Hongfeng Reservoir (HF) (26°29′30″N, 106°26′16″E) are drinking water sources for the capital city of Guizhou with about 10 years history of aquaculture, but the aquaculture has ceased over the past 10 years. Hongjiadu Reservoir (HJD) (26°54′44″N, 105°51′32″E) has no history of aquaculture.

Water sampling was conducted in the open water zone of reservoirs. Water samples collected for chemistry analysis using 1-L Niskin sampler from different depths of 0.5 m, 10 m below the water surface and 0.5 m away from the bottom sediment (Yan et al., 2013). Zoobenthos samples were collected from littoral sites of reservoirs (0–1 m depth). Zoobenthos samples were hand-picked by screening the sediment through a 500-μm sieve. After being rinsed with pure water, the zoobenthos samples were kept cold (in cooling box) and sorted live in the lab by taxa within 24 h of collection. We classified zoobenthos by their feeding habits (Flannagan, 1979). *Bellamya quadrata* was classified as herbivorous; *Chironornidae* was classified as detritivorous; *Crucian fry* and *Decapoda* were classified as omnivorous; *Odonata*, *Aquarlus elongatus*, *Hemiptera*, *Crustacea*, *Coleoptera*, *Tubellaria* and *Hydrophilidae* were classified as carnivorous. Obviously, the categorization for feeding type are not clear-cut borderlines as they also depend on the food available. Nevertheless, we believe that our grouping provides a first approximation to reality.

Potential food sources for zoobenthos including phytoplankton, zooplankton, artificial fish baits and sediment were also sampled in Chinese reservoirs. Phytoplankton (64–112 μm) and zooplankton (112–500 μm) in the four reservoirs were collected vertically from the open water using 64-μm and 112-μm plankton nets. Plankton samples were transferred to polypropylene vials and placed in liquid nitrogen immediately. Artificial fish food were purchased from the local fisherman in the WJD Reservoir. The upper surface sediment in the four reservoirs was collected, and were placed in centrifuge tubes, capped and sealed with parafilm.

All samples were stored at −80 °C before freeze-drying. These freeze-dried samples were homogenized by taxa for further analysis.

1.1.2. Sampling in Swedish lakes

Swedish lakes are much less influenced by human activities compared to the Chinese reservoirs. As shown in Fig. 1, five lakes are located in the Västerbotten region, northern Sweden (64.12 to 64.25° N, 18.76 to 18.80° E) and close to each other. While Erken lake (ER, 59°8′N, 18°6′E) is further south, in eastern Sweden within the Stockholm region. Zoobenthos were retained on nets with different mesh size (500 μm and 250 μm) by kick-sampling method (kicking littoral substrate for a few times while placing the mesh net in the direction of flow) in shallow stony and/or sandy habitats (0–1 m depth) by Wu et al. (2019). *Chironmidae* was classified as detritivorous; *Diptera*, *Megaloptera*, *Trichoptera*, *Hemiptera* and *Gastropoda* were classified as herbivorous; *Amphipoda*, *Clitellata* and *Bivalvia* were classified as omnivorous; *Odonata* and *Coleoptera* were classified as carnivorous. Swedish samples were treated the same as for Chinese samples.

1.2. Sample analysis

The detailed information of analysis for samples from Swedish lakes has been provided by Wu et al. (2019). Briefly, total Hg (THg) and MeHg concentrations in water were determined by cold vapor atomic fluorescence spectrometry (CVAFS), Method 1631 (USEPA, 2002) and chromatography (GC)-CVAFS, Method 1630 (USEPA, 2001), respectively. The method has a detection limit of 0.05 ng/L and a quantitation limit of 0.1 ng/L. THg concentrations of zoobenthos were analyzed with thermal decomposition Mercury Analyzer (Milestone Srl, Italy), Method 7473 (SW-846) (USEPA, 1998), or solid-sampling thermal decomposition

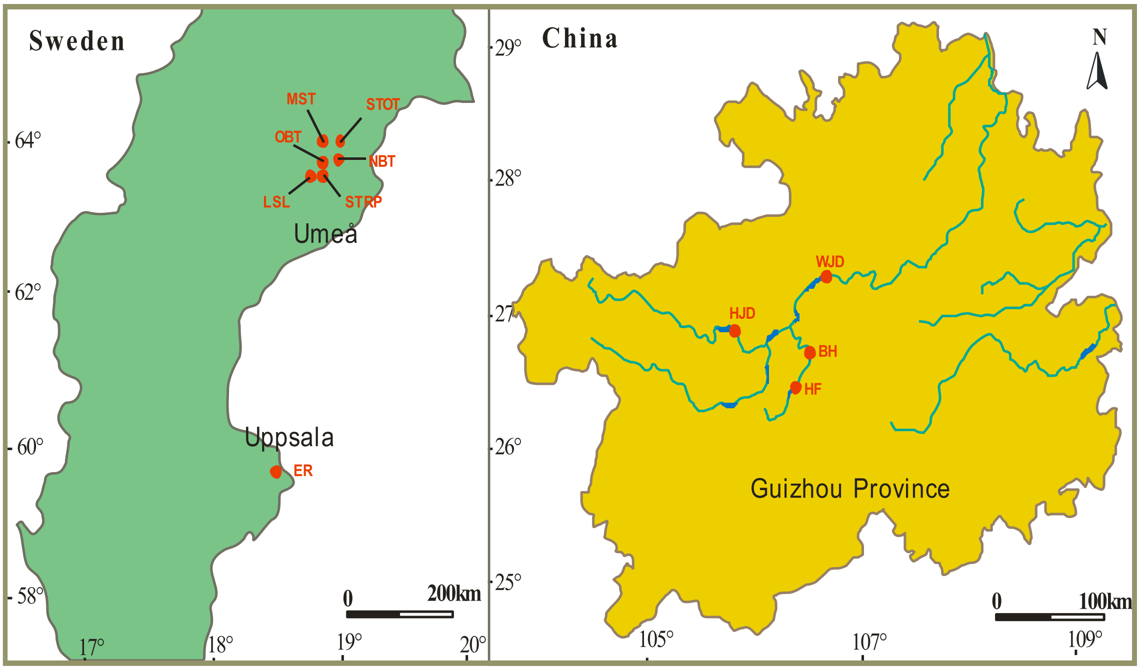

Fig. 1. Map of study sites in Swedish lakes and Chinese reservoirs.

amalgamation atomic absorption spectrometry (TDA AAS). The equipment's method detection limit was documented as low as 0.1 ng. The precision of measurement from replicate analyses was >80%. Samples, blanks, certified reference material CRMs (DORM-2, TORT-2 and MESS-3) were run at least once per every 10 samples to assess analysis accuracy. Recoveries for CRMs were within 100 ± 20%, while blanks were <10% of the lowest sample [THg].The MeHg concentrations in zoobenthos were analyzed with a Tekran® 2700 Methyl Mercury Auto-Analysis System (Model 2700; Tekran Instrument Corporation.). The equipment is equipped with atomic fluorescence detection (following EPA Method 1630). The MeHg method detection limit was 0.072 ng/g. Sample replicates and CRMs (IAEA-086, DORM-3, and TORT-3 with) were run at least once per every 15 samples to assess analysis accuracy (method relative percent difference <35%).

Total nitrogen (TN), total phosphorus (TP), dissolved organic carbon (DOC), and chlorophyll *a* (Chl-a) and pH in lake water samples were analyzed at the Geochemical Lab at Department of Aquatic Sciences and Assessment (Swedish University of Agricultural Sciences, Uppsala) according to accredited practices and analytical methods accredited (Swedish Board for Accreditation/Conformity Assessment; SWEDAC).

Total Hg concentrations in water sampled in Chinese reservoirs were determined by CVAFS Method 1631 (USEPA, 2002). The samples were oxidized with 0.5% BrCl, 0.2% v/v $NH_2OH \cdot HCl$ and $SnCl_2$ were added for reduction. Hg^0 was pre-concentrated into a gold trap with an aspirator (Yan et al., 2003). The MeHg in water was determined using GC-CVAFS method (USEPA, 2001), followed by ethylation, purging, and trapping onto Tenax traps. Total Hg and MeHg concentrations in water were detected by GC-CVAFS (Brooks Rand Model III, Seattle, USA) (He et al., 2004; Horvat et al., 1993). The detection limit for THg and MeHg in water was 0.02 and 0.009 ng/L, respectively.

THg and MeHg contents in zoobenthos, phytoplankton and zooplankton from Chinese reservoirs were determined by GC- CVAFS (Brooks Rand Model III, Seattle, USA) after digested. For THg analyses, the samples were first digested in 10 mL HNO_3:H_2SO_4 ratio of 7:3 (v/v) at 95 °C for 3 h, and was then diluted with 0.5 mL BrCl and deionized water to 25 mL after cooling the solution. The samples for MeHg analyses were digested in 5 mL KOH solution at 75 °C for 3 h and then diluted to 25 mL with deionized water (Yan et al., 2008). Blank spikes (5%) and duplicates (>10% of samples) were taken regularly throughout each sampling process.THg in sediment were analyzed with a DMA-80 Total Mercury Analyzer (Milestone Srl, Italy) employing USEPA method 7473 (USEPA, 1998). MeHg in sediment was determined using HNO_3 leaching/CH_2Cl_2 extraction, ethylation, trapping on Tenax trap, isothermal GC separation, and CVAFS detection method (He et al., 2004). The detailed analysis procedure is presented in Section 1 of SI.

TORT-3 from National Research Council of Canada was used as the CRM for biota samples. Recovery of THg and MeHg in the CRM Tort-3 were 93–114% and 101%–106%, respectively. The detection limits for THg and MeHg in biota samples were 0.013 ng/g (3σ) and 0.002 ng/g (3σ), respectively. ERM-CC580 was used as CRM for sediment. Recovery of THg and MeHg in the ERM-CC580 were 90–108% and 114–118%, respectively. Limits of determination for THg and MeHg in sediment were 0.02 ng/g (3σ) and 0.003 ng/g (3σ), respectively. THg and MeHg contents (ng/g) in dry weight were converted to wet weight (w.w.) using the water content.

Total lipids and fatty acids analyses of zoobenthos in Swedish lakes and Chinese reservoirs were conducted at the inter-university center for ecosystem research Wasser Cluster Lunz (Lunz am See, Austria). Lipids were extracted in a chloroform-methanol mixture (2:1 v/v), following the methods described elsewhere (Heissenberger et al., 2010) and their fatty acids (FA) were trans-esterified to fatty acid methyl esters (FAME) (Jing et al., 2020) and subsequently analyzed by a gas-chromatograph (Thermo Scientific™ TRACE ™) using a Capillary Column (SP-2560 Supelco™, 100 m, 0.25 mm i.d., 0.2 μm film thickness; Sigma-Aldrich, Bellefonte, PA, USA).

For water chemistry analysis, three replicates were collected for each water sample from four Chinese reservoirs. Analysis of TN, TP and Chl-a were performed using Spectrophotometry method (Li and Han, 2007). Water temperature (T), dissolved oxygen (DO), and pH were measured on site with a water quality meter (SX751). DOC was measured as total organic carbon (TOC) with a TOC analyzer (Elementar, High TOC 2). All the analyses of these parameters were performed following the procedures outlined in the Chinese Standard Methods for Water Quality Analysis (GB-3838-2002, 2002). For the analysis of DOC, water was filtered through a Whatman (GF/F) glass-fiber C-free filter. DOC was measured as TOC and determined by high temperature catalytic oxidation with a TOC analyzer (Elementar, High TOC 2).

1.3. Statistical analysis

The species composition of zoobenthos from Chinese reservoirs and Swedish lakes were compared. Since the data were non-normal distribution, comparisons of Hg and PUFA contents in zoobenthos from Chinese reservoirs and Swedish lakes were conducted by analysis of non-parametric Kruskal-Wallis test. The difference was considered statistically significant at *p*-value <0.05. Corrgram function of corrgram package in R Programming Language (R version 3.6.2) was employed to calculate correlation of Hg and PUFA contents of zoobenthos with abiotic variables (water chemistry) and biotic variables (algal dietary) and display the results graphically. The abiotic variables included in corrgram were TN, TP, Chla, DOC, pH, THg and MeHg concentrations in water; the biotic variables in corrgram were contents of algal fatty acids biomarkers, including PUFA, LIN, ALA, ARA, EPA and DHA. Correlation coefficients (r) and significance probabilities (p) were computed according to Spearman correlation analysis.

2. Results

2.1. Study sites characteristics

The surface area of our studied Chinese reservoirs were more than 1000 times larger than the Swedish lakes, and the catchment area of the Chinese reservoirs were more than 200 times larger than the Swedish lakes, except the ER Lake was comparable in water depth to BH Reservoir and in surface area to WJD Reservoir. But the lake area ratios of Chinese reservoirs and Swedish lakes were similar ($p > 0.05$). The Chinese reservoirs were generally mesotrophic and neutral and slightly alkaline (pH: 7.8–8.3), except WJD which was eutrophic. By contrast, the Swedish lakes were generally oligotrophic and ranged from acidic to neutral (pH: 4.9–7.7), yet ER Lake was mesotrophic and slightly alkaline (pH: 7.7). The TN:TP ratio in Chinese reservoirs (0.4–2.3) was higher than the Swedish lakes (0.02–0.08). Average DOC concentrations of water in the Swedish lakes (19 ± 6.9 μg/L) were almost 6 times higher than of the Chinese reservoirs (2.5 ± 1.7 μg/L). The aqueous THg and MeHg concentrations in the Chinese reservoirs (THg: 3.2 ± 2.4 ng/L, MeHg: 0.4 ± 0.6 ng/L) and the Swedish lakes (THg: 4.6 ± 4.8, MeHg: 0.2 ± 0.1 ng/L) were similar ($p > 0.05$) (Table 1).

In the Chinese reservoirs, the BH Reservoir had the highest Chl-a and DOC concentrations. The HF and HJD Reservoirs had the highest TN and TP concentrations, respectively (Table 1). The aqueous THg and MeHg concentrations of the BH (THg: 4.5 ng/L, MeHg: 0.14 ng/L) and HF Reservoirs (THg: 4.6 ng/L, MeHg: 0.18 ng/L) were higher than that of the WJD (THg: 1.4 ng/L, MeHg: 0.12 ng/L) and HJD Reservoirs (THg: 1.1 ng/L, MeHg: 0.04 ng/L) (Table 1). In the Swedish lakes, mesotrophic ER Lake had the highest TN, TP concentrations but the lowest aqueous Hg concentrations. NBT Lake had the highest DOC concentration. STRT Lake had the highest aqueous Hg concentration (Table 1).

Table 1
Physical and chemical characteristics of Chinese reservoirs and Swedish lakes.

Lake name	Water depth (m)	Surface area (km^2)	Catchment area (km^2)	Lake area ratio	Trophic status	Chla (μg/L)	TN (μg/L)	TP (μg/L)	DOC (mg/L)	pH	THg_w (ng/L)	$MeHg_w$ (ng/L)
Chinese reservoirs												
WJD	150	21.4	2.8×10^4	7.6×10^{-4}	Eutrophic	9.3	200	436	1.1	8.3	1.4	0.12
BH	10.5	14.5	1.9×10^3	7.6×10^{-3}	Mesotrophic	12	50	135	4.8	7.9	4.5	0.14
HF	20	57.2	1.6×10^3	3.6×10^{-2}	Mesotrophic	3.6	300	130	2.8	8	4.6	0.18
HJD	105	80.5	1.0×10^4	8.1×10^{-3}	Mesotrophic	5.2	190	492	1.5	7.8	1.1	0.04
Swedish lakes												
OBT	4	4.8×10^{-2}	2.8	1.7×10^{-2}	Oligotrophic	1.5	16	510	27	4.9	7.1	0.29
STOT	2.7	3.9×10^{-2}	2.92	1.3×10^{-2}	Oligotrophic	2.5	12	469	20	5.3	3.8	0.16
STRT	3.8	3.1×10^{-2}	11.8	2.6×10^{-3}	Oligotrophic	34	24	547	25	5.2	11	0.45
NBT	6	3.2×10^{-2}	32.5	1.0×10^{-3}	Oligotrophic	1.5	17	432	23	5	6.4	0.28
MST	5.3	1.8×10^{-2}	0.14	1.3	Oligotrophic	2.4	17	423	10	6.5	1.2	0.14
ER	9	24	141	0.2	Mesotrophic	8	60	712	11	7.9	0.3	0.06

2.2. Zoobenthos

2.2.1. Zoobenthos species composition

Taxonomic composition of zoobenthos differed remarkably between the Swedish lakes and Chinese reservoirs. In order to make the comparison feasible and plausible, we categorized zoobenthos from these two habitats into four groups according to their feeding habits - detritivorous, herbivorous, omnivorous and carnivorous (Table S1).

The functional feeding types as well as taxonomic composition of zoobenthos showed significant difference between Swedish lakes and Chinese reservoirs (Fig. 2 and Table S1). In the Swedish lakes, the most abundant zoobenthos was herbivorous (66 ± 37%), followed by carnivorous (28 ± 36%). Particularly, zoobenthos were dominated by herbivorous in OBT, NBT, MST and ER Lakes, while carnivorous zoobenthos dominated in STOT and STRT Lakes.

On the contrary, the highest average biomass proportion was carnivorous zoobenthos (47 ± 47%) and followed by herbivorous (31 ± 46%) in the Chinese reservoirs. Most zoobenthos were carnivorous in HF and HJD Reservoirs. While zoobenthos was exclusively herbivorous in WJD Reservoir. However, zoobenthos was mainly omnivorous in BH Reservoir.

2.2.2. Algal diet composition of zoobenthos

The relative PUFA content of zoobenthos in the Chinese reservoirs (35.5 ± 12.7%) and Swedish lakes (39.6 ± 8.7%) did not significantly differ from each other ($p > 0.05$) (Fig. 3; Table S1). Among these dietary PUFA, the DHA content (%) in zoobenthos of Chinese reservoirs (3.9 ± 3.1%) was 6 times higher than of Swedish lakes (0.6 ± 0.5%), while EPA of zoobenthos in Swedish lakes (11 ± 4.2%) was almost 2 times higher compared to the Chinese reservoirs (6.6 ± 5.2%). There were no significant differences in LIN, ALA and ARA in zoobenthos from Chinese reservoirs and Swedish lakes ($p > 0.05$) (Fig. 3).

In the Chinese reservoirs, zoobenthos in the WJD and HF Reservoirs had higher LIN and ALA contents than EPA and DHA. Contrastingly, zooenthos in BH and HJD Reservoirs had higher EPA and DHA contents than LIN and ALA. EPA and DHA of zoobenthos in the BH and HJD Reservoirs were significantly higher than that in the WJD and HF Reservoirs ($p < 0.05$). In the Swedish lakes, except for MST Lake, LIN + ALA and EPA + DHA of zoobenthos did not show significant difference. Among the PUFA, DHA occupied the lowest proportion (<1%) in zoobenthos, and only LIN of zoobenthos varied across the 6 lakes ($p < 0.05$) (Fig. 3).

As to the variation of PUFA composition in zoobenthos with different feeding habits, in Chinese reservoirs, except for HF Reservoir, omnivorous and carnivorous zoobenthos had higher EPA + DHA than LIN + ALA, which is in contrast to herbivorous and detritivorous zoobenthos. Besides, omnivorous and carnivorous zoobenthos had higher EPA + DHA than herbivorous and detritivorous zoobenthos. By contrast, in Swedish lakes, except for omnivorous zoobenthos in ER Lake and phytophagous zoobenthos in MST Lake, there was no significant difference of LIN + ALA and EPA + DHA in zoobenthos with different feeding habits. Omnivorous zoobenthos in ER Lake had higher EPA + DHA than LIN + ALA, while herbivorous zoobenthos in MST Lake had the opposite distribution (Table S2).

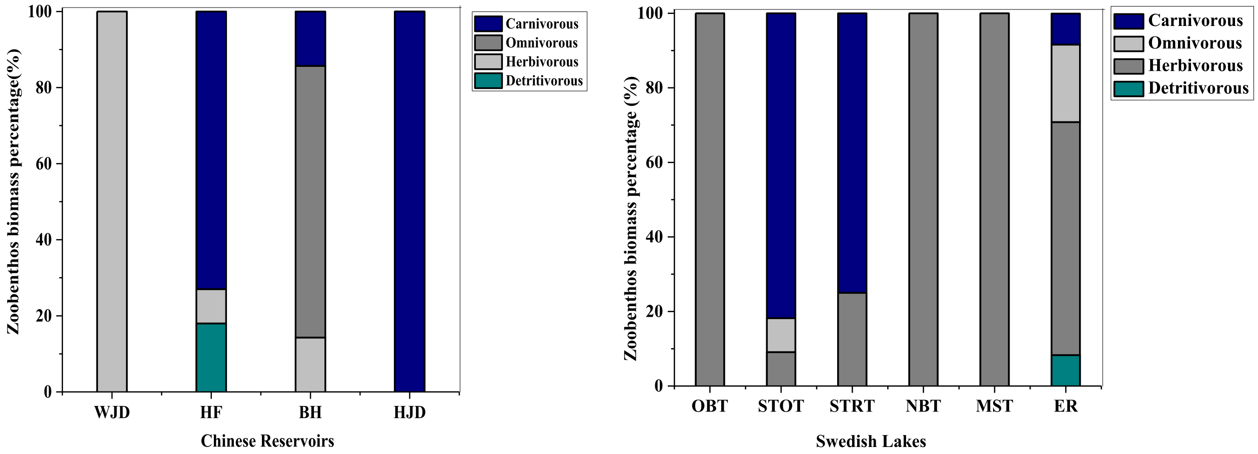

Fig. 2. Zoobenthos taxonomic composition in Chinese reservoirs and Swedish lakes. *Note*: Relative abundance of zoobenthos feeding groups based on number of taxa collected from investigated reservoirs or lakes. WJD: Wujiangdu Reservoir; HF: Hongfeng Reservoir; BH: Baihua Reservoir; HJD: Hongjiadu Reservoir. OBT: Övre Björntjärn Lake; STOT: Stortjärn Lake; STRT: Struptjärn Lake; NBT: Nedre Björntjärn Lake; MST: Mångstenstjärn Lake; ER: Erken Lake.

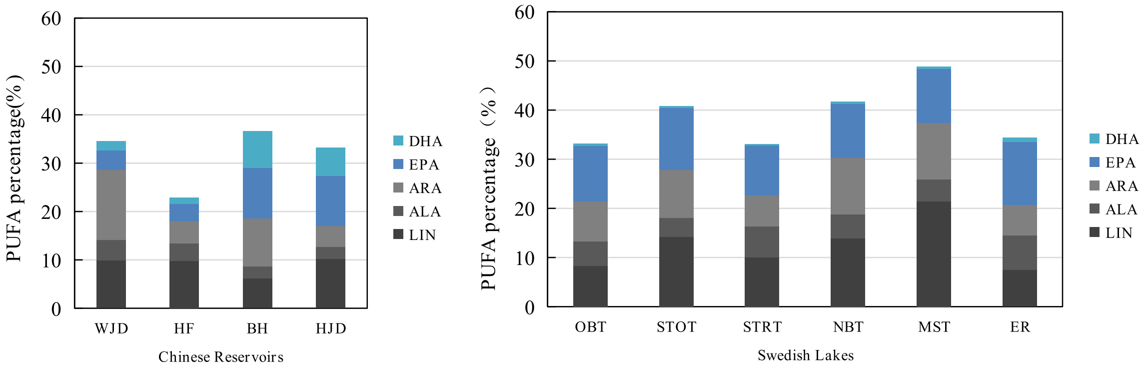

Fig. 3. Algal dietary fatty acids biomarkers percentages of zoobenthos in different Chinese reservoirs and Swedish lakes. *Note*: LIN: linoleic acid (18:2n-6), ALA: alpha-linolenic acid (18:3n-3), ARA: arachidonic acid (C20:4n-6), EPA: eicosapentaenoic (20:5n-3), DHA: docosahexaenoic acid (22:6n-3).

2.3. Mercury in zoobenthos

As shown in Fig. 4, THg and MeHg contents of zoobenthos in the Chinese reservoirs were significantly higher than Swedish lakes ($p < 0.05$). The average THg contents of zoobenthos in Chinese reservoirs ($n = 22$, 269 ± 185 ng/g d.w.) was almost 2 times that in Swedish lakes ($n = 60$, 141 ± 134 ng/g d.w.). The average MeHg contents of zoobenthos in Chinese reservoirs was of 39 ± 126 ng/g d.w. While in the Swedish lakes, MeHg was only detectable in 25% zoobenthos, with an average value of 99.9 ± 109.4 ng/g d.w.

Although there was no significant differences of THg and MeHg contents of zoobenthos in the Chinese reservoirs ($p > 0.05$), average THg content of zoobenthos in eutrophic WJD Reservoir ($n = 3$, 366 ± 22 ng/g d.w.) was almost 2 times that in mesotrophic HF Reservoir ($n = 9$, 195 ± 108 ng/g d.w.), and average MeHg content of zoobenthos in mesotrophic BH Reservoir ($n = 7$, 235 ± 164 ng/g d.w.) was about 10 times that in eutrophic WJD Reservoir (n = 3, 29 ± 4.0 ng/g d.w.). THg and MeHg contents of zoobenthos varied significantly across Swedish lakes ($p < 0.05$). Average THg and MeHg contents were highest in oligotrophic STRT lake ($n = 17$, THg: 279 ± 113 ng/g d.w., MeHg: 226 ± 63 ng/g d.w.), and lowest in mesotrophic ER lake ($n = 24$, THg: 50 ± 41 ng/g d.w., MeHg: 7.5 ± 4.2 ng/g d.w.).

In terms of feeding habits, THg and MeHg contents in zoobenthos from the Chinese reservoirs were generally higher in omnivorous and carnivorous and lower in phytophagous ones ($p < 0.05$). While THg (41.3 ng/g d.w.) and MeHg (1.6 ng/g d.w.) contents of carnivorous zoobenthos in BH Reservoir were lowest, THg (366 ± 22 ng/g d.w.) content of phytophagous zoobenthos in eutrophic WJD was higher than that in mesotrophic reservoirs. Similar to the Chinese reservoirs, THg and MeHg contents were generally higher in carnivorous and lower in phytophagous and detritivorous zoobenthos in the mesotrophic or oligotrophic Swedish lakes ($p < 0.05$) (Table S3).

2.4. Polyunsaturated fatty acids in zoobenthos

The PUFA and EPA + DHA contents in zoobenthos from the Chinese reservoirs were 0.05–35 mg/g d.w. and 0.02–18 mg/g d.w., respectively, and from the Swedish lakes 0.03–86 mg/g d.w. and 0.01–18 mg/g d.w., respectively (Fig. 5). The PUFA contents of zoobenthos in the Swedish lakes (n = 60, 17 ± 17 mg/g d.w.) and Chinese reservoirs (n = 22, 13 ± 10 mg/g d.w.) were similar ($p > 0.05$), and so were the EPA + DHA contents of zoobenthos in the Chinese reservoirs (n = 22, 4.7 ± 4.7 mg/g d.w.) and Swedish lakes (n = 60, 5.1 ± 4.9 mg/g d.w.) ($p > 0.05$). However, DHA of zoobenthos in the Chinese reservoirs (1.7 ± 2.5 mg/g d.w.) was 8 times higher than the Swedish lakes (0.2 ± 0.3 mg/g d.w.) ($p < 0.01$). By contrast, LIN, ALA, ARA and EPA of zoobenthos in the Chinese reservoirs (4.6 ± 5.2, 1.6 ± 1.7, 2.4 ± 1.7 and 3.1 ± 3.0, respectively) and the Swedish lakes (6.5 ± 6.2, 4.4 ± 7.0, 2.7 ± 2.2 and 5.1 ± 4.7, respectively) were similar ($p > 0.05$).

DHA of zoobenthos varied significantly among the Chinese reservoirs ($p < 0.05$), while there was no significant difference in bulk and individual PUFA contents. The highest EPA + DHA and DHA contents of zoobenthos were in the BH Reservoir (n = 7, 3.1 ± 2.9 and 4.3 ± 3.1 mg/g d.w., respectively), 5 and 20 times that the lowest contents in WJD Reservoir (n = 3, 0.6 ± 0.3 and 0.2 ± 0.1 mg/g d.w., respectively). However, there is no significant difference of EPA and DHA contents among the six Swedish lakes ($p > 0.05$).

In the mesotrophic Chinese reservoirs, EPA + DHA contents of zoobenthos were lower in herbivorous zoobenthos, while higher in omnivorous and carnivorous zoobenthos. The similar patterns were also observed in the oligotrophic Swedish lakes (Table S1).

2.5. Correlation between abiotic factors and algal dietary with mercury and PUFA contents in zoobenthos

The correlogram function was performed to investigate the potential impact on THg, MeHg and EPA, DHA contents in zoobenthos. Figs. 6 and 7 were correlograms between the explanatory variables (including water chemistry and algal dietary fatty acids biomarkers) and THg and MeHg contents in zoobenthos from Chinese reservoirs and Swedish lakes, respectively. The correlograms showed two ways to graphically display the correlation matrix using the panel.shade () and panel.pie () functions. As to the cells in lower triangle position, blue and a slash from bottom left to top right of a cell indicate that the two variables in the cell are positively correlated. In turn, red and a slash from top left to bottom right indicate a negative correlation. The darker the color and the higher the saturation, the greater the correlation among variables. The upper triangle cells display the same information in a pie chart. The function of color is the same as above, but the correlation strength is displayed by the size of the filled pie chart block. The pie chart filled in the clockwise direction means a positive correlation, while a pie chart counterclockwise means negative correlation. In these figures, to pool variables with similar correlation patterns together, the rows and columns of the matrix have been reordered (using the principal component method).

According to Fig. 6, in the Chinese reservoirs, THg, MeHg contents of zoobenthos didn't show close association with other variables. THg contents of zoobenthos had little correlation with other variables. MeHg content of zoobenthos was only found positively correlated with the DOC concentrations of water ($r = 0.42$, $p < 0.05$) and the DHA contents of zoobenthos ($r = 0.53$, $p = 0.01$). By contrast, EPA and DHA contents of zoobenthos correlated with both abiotic and biotic variables. EPA contents of zoobenthos showed greater correlations with algal diet

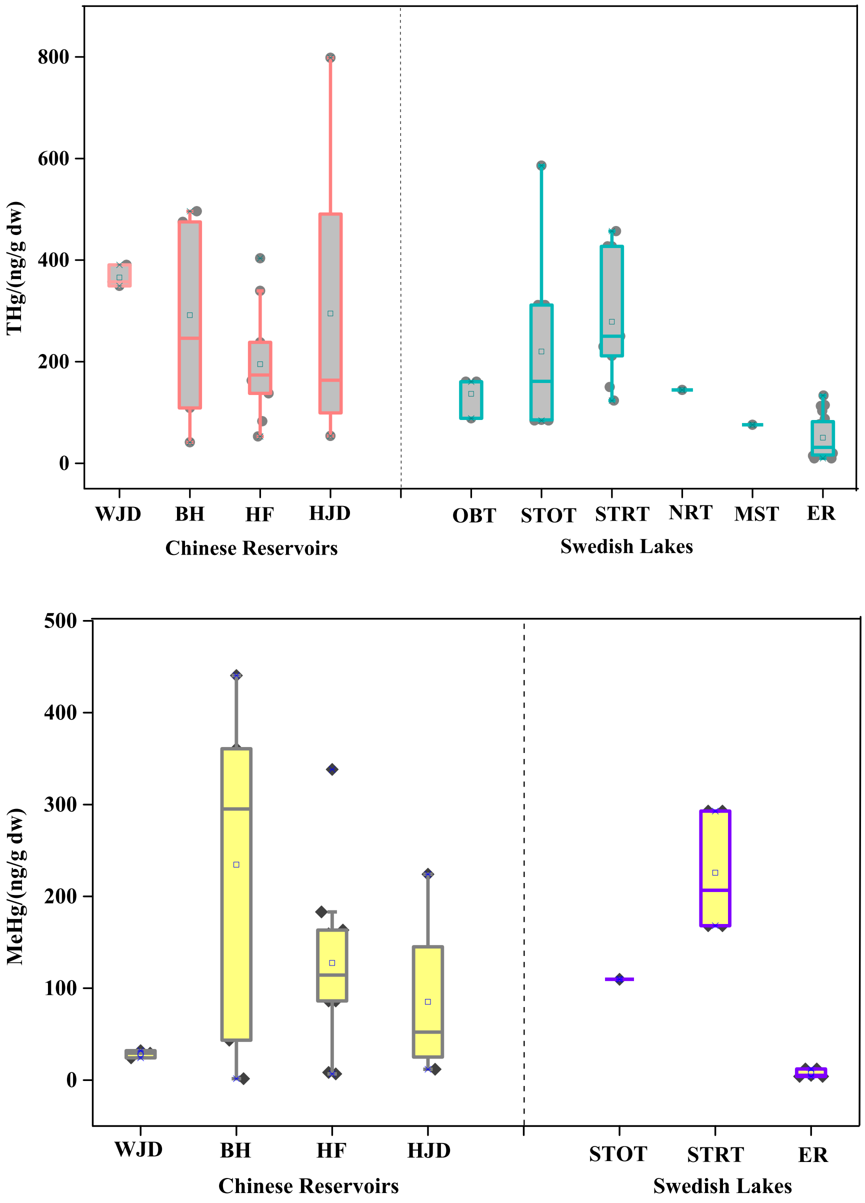

Fig. 4. THg and MeHg contents of zoobenthos in different Chinese reservoirs and Swedish lakes. *Note*: WJD: Wujiangdu Reservoir; HF: Hongfeng Reservoir; BH: Baihua Reservoir; HJD: Hongjiadu Reservoir. OBT: Övre Björntjärn Lake; STOT: Stortjärn Lake; STRT: Struptjärn Lake; NBT: Nedre Björntjärn Lake; MST: Mångstenstjärn Lake; ER: Erken Lake.

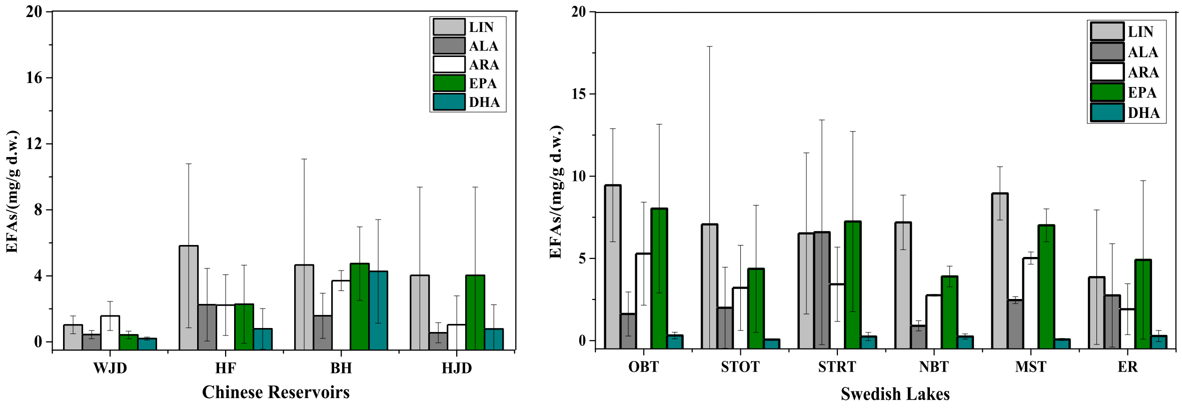

Fig. 5. Essential fatty acids (EFA) contents of zoobenthos in Chinese reservoirs and Swedish lakes. *Note*: WJD: Wujiangdu Reservoir; HF: Hongfeng Reservoir; BH: Baihua Reservoir; HJD: Hongjiadu Reservoir. OBT: Övre Björntjärn Lake; STOT: Stortjärn Lake; STRT: Struptjärn Lake; NBT: Nedre Björntjärn Lake; MST: Mångstenstjärn Lake; ER: Erken Lake. LIN: linoleic acid (18:2n-6), ALA: alpha-linolenic acid (18:3n-3), ARA: arachidonic acid (C20:4n-6), EPA: eicosapentaenoic (20:5n-3), DHA: docosahexaenoic acid (22:6n-3).

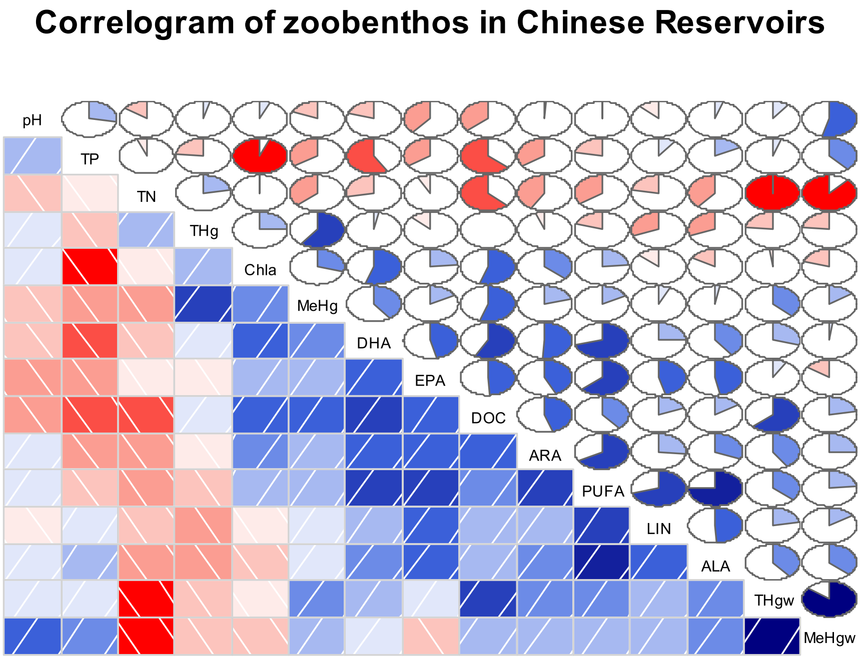

Fig. 6. Correlogram illustrating the correlations among water chemistry variables, PUFA, LIN, ALA, ARA, EPA, DHA, THg and MeHg contents in zoobenthos from Chinese reservoirs. *Note*: Chla: Chlorophyll-a, T: Temperature, TN: Total nitrogen, TP: Total phosphorous, DOC: Dissolved organic carbon, THg_w: THg concentrtions in water, $MeHg_w$: MeHg concentrations in water. PUFA: algal dietary fatty acids biomarker, LIN: linoleic acid (18:2n-6), ALA: alpha-linolenic acid (18:3n-3), ARA: arachidonic acid (C20:4n-6), EPA: eicosapentaenoic (20:5n-3), DHA: docosahexaenoic acid (22:6n-3).

fatty acids biomarkers than abiotic factors. Specifically, EPA contents of zoobenthos positively related to PUFA ($r = 0.76$, $p < 0.01$), LIN ($r = 0.59$, $p < 0.01$), ALA ($r = 0.55$, $p < 0.01$), ARA ($r = 0.55$, $p < 0.01$) contents, and aqueous DOC concentrations ($r = 0.43$, $p < 0.05$). DHA contents in zoobenthos showed positive correlations with aqueous DOC ($r = 0.68$, $p < 0.01$) and Chla concentrations ($r = 0.41$, $p < 0.05$), and negative correlation with TP concentration ($r = -0.46$, $p < 0.05$). DHA contents also showed positive correlation with PUFA ($r = 0.70$, $p < 0.01$), ALA ($r = 0.54$, $p < 0.01$) and ARA ($r = 0.54$, $p < 0.01$) contents.

In the Swedish lakes, THg and MeHg contents in zoobenthos were more related with abiotic factors (Fig. 7). The THg content in

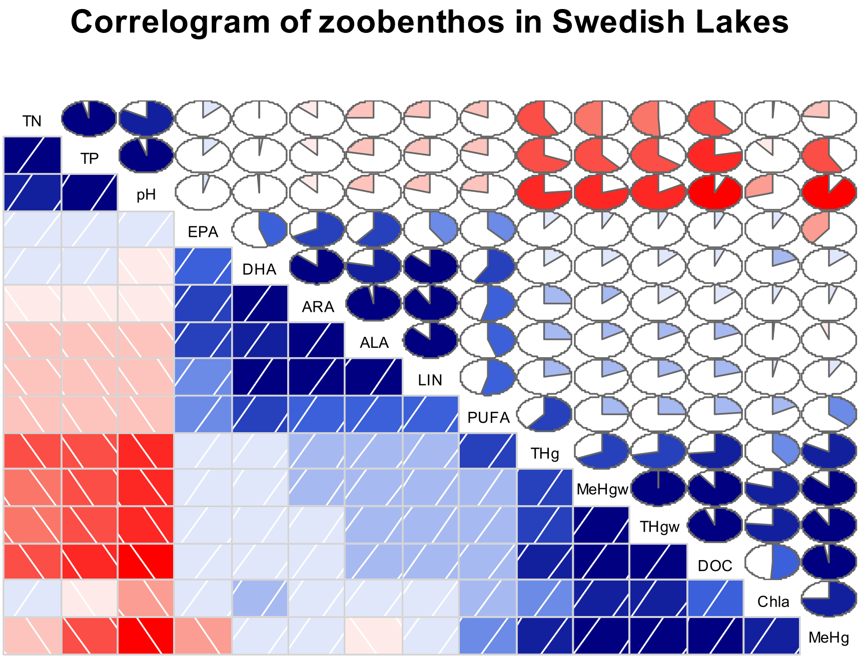

Fig. 7. Correlogram illustrating the correlations among water chemistry variables, PUFA, LIN, ALA, ARA, EPA, DHA, THg and MeHg contents in zoobenthos from from Swedish lakes. *Note*: Chla: Chlorophyll-a, T: Temperature, TN: Total nitrogen, TP: Total phosphorous, DOC: Dissolved organic carbon, THg_w: THg concentrtions in water, $MeHg_w$: MeHg concentrations in water. PUFA: algal dietary fatty acids biomarker, LIN: linoleic acid (18:2n-6), ALA: alpha-linolenic acid (18:3n-3), ARA: arachidonic acid (C20:4n-6), EPA: eicosapentaenoic (20:5n-3), DHA: docosahexaenoic acid (22:6n-3).

zoobenthos positively correlated with aqueous THg, MeHg and DOC concentrations, while negatively with pH ($r = -0.73$, $p < 0.01$), TP ($r = -0.55$, $p < 0.01$) and TN ($r = -0.35$, $p < 0.01$) concentrations. MeHg contents of zoobenthos were positively correlated to aqueous THg ($r = 0.89$, $p < 0.01$), MeHg ($r = 0.89$, $p < 0.01$), DOC ($r = 0.86$, $p < 0.01$) and temperature ($r = 0.82$, $p < 0.01$), while were negatively correlated to the pH of water ($r = -0.89$, $p < 0.01$) and TP ($r = -0.58$, $p < 0.05$). As to diet fatty acids biomarkers, only THg contents showed positive correlations with PUFA ($r = 0.58$, $p < 0.01$) content.

The EPA and DHA contents of zoobenthos showed little correlation with abiotic variables, but had significantly positive relationship with PUFA profiles of zoobenthos. Specially, EPA positively correlated with PUFA ($r = 0.38$, $p < 0.01$), LIN ($r = 0.41$, $p < 0.01$), ALA ($r = 0.46$, $p < 0.01$) and ARA ($r = 0.67$, $p < 0.01$). DHA positively correlated with PUFA ($r = 0.66$, $p < 0.01$), LIN ($r = 0.73$, $p < 0.01$), ALA ($r = 0.60$, $p < 0.01$) and ARA ($r = 0.75$, $p < 0.01$).

3. Discussion

It is eco-toxicologically important to compare the bioaccumulation of Hg and PUFA of zoobenthos in two contrasting ecosystems. Contrary to our hypothesis, the study showed that the content of potentially toxic Hg of zoobenthos was generally higher in Chinese reservoirs than Swedish lakes, nutritious EPA + DHA contents of zoobenthos were similar in Chinese reservoirs and Swedish lakes, while DHA contents of zoobenthos were significantly higher in Chinese reservoirs. It was found that eutrophication of Chinese Reservoirs didn't results in less Hg and PUFA accumulated in zoobenthos through enhancing algal biomass and altering phytoplankton composition, which was inconsistent with the effect of eutrophication on fish reported in previous studies (Liu et al., 2012; Wang et al., 2012). Moreover, we examined this unexpected variation of Hg and PUFA in zoobenthos from these two ecosystems was mainly attributed to their algal dietary.

3.1. Effects of abiotic factors on mercury and EFA variations in zoobenthos

It has been widely suggested that Hg bioaccumulation in aquatic food chains can be affected by abiotic factors, including the eutrophic status (Razavi et al., 2015), aqueous Hg, sulphate deposition (Braaten et al., 2020), DOC (Rennie et al., 2005; Wu et al., 2019) and pH (Jardine et al., 2013). Numerous previous findings have argued that pH and DOC concentrations are important indicators of MeHg bioavailability in aquatic food webs, since Hg in zoobenthos is often negatively correlated with pH and positively correlated with DOC (Rennie et al., 2005). In line with the meta-analysis conducted by Wu et al. (2019), our analysis revealed that high DOC and low pH correlated with the elevated Hg content in zoobenthos of the Swedish lakes (Fig. 7). The average Hg content of zoobenthos was lowest in ER Lake, which is the only neutral lake with lower DOC concentrations among our investigated Swedish lakes. Nonetheless, the MeHg content of herbivorous zoobenthos was higher in ER, implying that Hg accumulation of zoobenthos in Swedish lakes was also affected by their feeding habits. The prevailing view about the effect of eutrophication on Hg dynamics is that it will lead to lower Hg concentrations in aquatic organisms (Razavi et al., 2015). But Hg contents of zoobenthos in eutrophic WJD Reservoir were not significantly lower than in other mesotrophic reservoirs, especially for THg contents. In fact, no eutrophication factor was found correlated to Hg contents of zoobenthos in Chinese reservoirs. DOC concentrations were apparently the only important abiotic factor for Hg contents in zoobenthos in both the Chinese Reservoirs and Swedish lakes. Compared to zoobenthos in Swedish lakes, DOC concentrations increased with increasing MeHg contents of zoobenthos in Chinese reservoirs, yet showed little effect on the THg contents of zoobenthos in Chinese reservoirs, which may partially explain that zoobenthos in the BH Reservoir with highest DOC concentration had higher MeHg contents but similar THg contents compared to that in the WJD Reservoir with lowest DOC concentration. We suggest that dietary algal Hg might contribute more to the Hg variation in zoobenthos than the water chemistry.

Contrary to THg and MeHg, no correlation was found between EPA and DHA contents in zoobenthos and abiotic variables in the Swedish lakes, suggesting that EPA and DHA in zoobenthos were weakly influenced by environmental factors, and that is why EPA and DHA in zoobenthos varied little across sampling sites. In the Chinese reservoirs, though, EPA and DHA in zoobenthos were positively correlated with DOC. Besides, DHA also positively correlated to Chl-a, which is an indicator of algal biomass, thus zoobenthos in the BH Reservoir with the highest DOC and Chl-a concentrations contained the highest DHA and EPA contents. Chl-a is an indicator of algal density, higher algal density can increase DHA and EPA transferred in benthic food web. On the other hand, DOC concentrations in the Chinese reservoirs was low (≤4.8 mg/L), thus the light extinction by colored DOC could not suppress the algal biomass, and contrastly, a moderate increase in DOC may stimulate biomass production of zoobenthos (Seekell et al., 2015), which might increase the retention of PUFA in zoobenthos. It should be noted that DOC also positively correlated with MeHg in zoobenthos. The DOC might also indirectly influence the MeHg accumulation of zoobenthos changing the biomass production of zoobenthos. Based on the relationships between abiotic factors and algal diet biomarkers (Figs. 6 and 7), we propose that the potential effect of environmental conditions on Hg bioaccumulation could be explored by fatty acids as diet biomarkers.

3.2. Effects of algae on mercury and PUFA variation in zoobenthos

Both the quality and quantity of algae can impact Hg and PUFA conveyed to organisms (Chen and Folt, 2005; Guo et al., 2016; Guo et al., 2020; Kainz and Mazumder, 2005). In aquatic systems, not all algal species produce the same PUFA: important EPA and DHA are predominantly synthesized by diatoms and dinoflagellates, respectively, while LIN and ALA are produced by Chlorophyta and Cyanophyta (Bell and Tocher, 2009). Neither phytoplankton species composition in four Chinese reservoirs eutrophic reservoir nor in six Swedish lakes can be exactly the same, thus PUFA composition and contents supplied for consumers would varied across the Chinese reservoirs or the Swedish lakes (Jing et al., 2020; Wu et al., 2019). In addition, zoobenthos has species-specific physiological retention of PUFA obtained from diet (Martinz, 2011; Willem et al., 2000), thus together resulted in the difference of PUFA concentration and composition of zoobenthos among the Chinese reservoirs or the Swedish lakes. The PUFA composition and contents in zoobenthos indicated that omnivorous and carnivorous zoobenthos obtained more diatoms and dinoflagellates comparing to herbivorous and detritivorous zoobenthos in most of the Chinese reservoirs and Swedish lakes.

Notably, both Hg and EPA + DHA contents of zoobenthos were higher in omnivorous and carnivorous and lower in herbivorous and detritivorous zoobenthos, implication of trophic magnification of both Hg and EPA + DHA contents.

The significantly higher dinoflagellate in algal diet for zoobenthos in the Chinese reservoirs than the Swedish lakes prevail in our comparison. The positive correlations among EFA and its precursors indicated that the retention of precursors of EFA are favorable to accumulation of EPA and DHA in zoobenthos from the Chinese reservoirs and Swedish lakes. Despite the eutrophication of Chinese reservoirs, zoobenthos in the Chinese reservoirs contained similar contents of EPA and DHA to those in Swedish lakes. Surprisingly, zoobenthos from the Chinese reservoirs contained significantly higher DHA content than those from the Swedish lakes. We speculate that although eutrophication could lower the proportion of high-quality diatoms and dinoflagellates in the Chinese reservoirs, the phytoplankton community still provide sufficient EPA and DHA for zoobenthos. In fact, similar cases are also found in the mesotrophic ER Lake, where zoobenthos had EPA and DHA contents similar to those from oligotrophic lakes. Thereby, both phytoplankton composition and density influence the EPA and DHA transferred to zoobenthos.

In the Swedish lakes, the THg contents positively correlated with PUFA contents in zoobenthos. Moreover, THg contents in both phyto- and zooplankton also positively correlated PUFA contents (Wu et al., 2019), implying that algal diet conveys THg to consumers. This facilitates THg accumulation at the base of the food chain and thus increases THg transferred to zoobenthos. However, in the Chinese reservoirs, the positive relationship was only observed between MeHg and the dinoflagellate biomarker DHA in zoobenthos suggest that dinoflagellates are transferring MeHg to these consumers, thus the higher DHA contents in BH Reservoir than WJD Reservoir can also contribute to the significantly higher MeHg of zoobenthos in BH than in WJD Reservoir. By contrast, algal diet showed no significant relationship with THg contents in zoobenthos, and the high THg contents of zoobenthos in WJD might contributed by other diet sources such as terrestrial or bacterial dietary. Actually, the potential food sources for zoobenthos in the Chinese reservoirs included phytoplankton, zooplankton, particulate artificial fish baits and sediment (Liu, 2006). Besides phytoplankton, copepods also contain DHA, while artificial fish baits were characterized by high LIN content, and there low EPA and DHA contents in the sediment (Fig. S1). Thereby, we infer that the high DHA contents of zoobenthos in Chinese reservoirs were primarily derived from algae and possibly copepods (Fig. S1). Moreover, phyto- and zooplankton contained higher THg and MeHg contents than artificial fish baits (Fig. S2). We hence argue that phyto- and zooplankton were the main dietary MeHg sources of zoobenthos in the Chinese reservoirs. While due to zooplankton predominantly feeds on phytoplankton, MeHg and PUFA in zooplankton mainly obtained from phytoplankton. Therefore, we concluded that phytoplankton was the fundamental source of MeHg in zoobenthos.

We argue that the role of diet in Hg accumulation of zoobenthos varied in different habitats, and is likely related to their dominant zoobenthos species and environmental conditions, especially DOC characteristics.

3.3. Comparison with variation of mercury and PUFA in fish

The variations of potentially toxic Hg and physiologically required PUFA of zoobenthos from the Chinese reservoirs and Swedish lakes were quite different from those in fish. Previous studies found that prevailing eutrophication of water in China can reduce Hg and PUFA bioaccumulation in fish (Liu et al., 2012; Wang et al., 2012). However, we assume that aquaculture was another reason for low fish Hg level in Chinese reservoirs (Jing et al., 2020). Unlike the Swedish omnivorous fish that mainly rely on zoobenthos for food, cultured omnivorous Chinese fish mainly feed on artificial fish feeds rather than zoobenthos, where the high supply of artificial fish feeds with lower Hg content enhanced the somatic growth rate of cultured fish, and thus diluted the Hg bioaccumulation in fish (Jing et al., 2020; Liu et al., 2012). By contrast, zoobenthos with higher Hg content would increase Hg in fish. Moreover, artificial fish feed contained high LIN rather than EPA and DHA (Fig. S1), which might reduce EPA and DHA intake by fish from diet. Compared to fish, Hg content and the PUFA profile of zoobenthos at lower trophic levels are more susceptible to environmental and autochthonous diet influences. Consequently, we conclude that eutrophication didn't lower Hg and EPA + DHA contents of zoobenthos in Chinese reservoirs, which is different from the previous study conclusions of biomass dilution in fish Hg due to the impact of eutrophication (Razavi et al., 2014). This was due to zoobenthos feeding less on artificial feeds, together with relatively shorter survival time and slower growth rates than fish. Thus there is no significant algal or growth dilution effect on Hg bioaccumulation in zoobenthos. On the other hand, relying on plankton instead of artificial feeds is more beneficial to zoobenthos as seen by enhanced EFA. Fortunately, in recent years, the eutrophication of Chinese reservoirs has been improving (i.e. eutrophication is declining) due to the implementation of the government environmental policies (Guo et al., 2015). The biomass of high-quality primary producers (diatom and dinoflagellate) may thus increase, and this would elevate the EPA + DHA content conveyed to consumers in Chinese reservoirs. Accordingly, we could foresee that EPA and DHA contents of wild fish in Chinese reservoirs will be further improved, if reservoir aquaculture activities would be completely banned. Yet it is difficult to predict a long-term trend for Hg bioaccumulation of wild fish in Chinese reservoirs. In either way, wild fish mainly feed on plankton and zoobenthos, unlike the cultured fish feeding that is almost exclusively feed on artificial fish baits. This diet structure based on algal food in the Chinese reservoirs and similar freshwaters might result in higher Hg accumulated in wild fish than cultured fish, since plankton and zoobenthos generally had higher Hg contents than artificial fish baits. However, the higher food quality (higher EPA + DHA contents) of zoobenthos and plankton compared to artificial fish baits might counteract the dietary Hg risk from fish (Schultz et al., 2012).

4. Conclusions

Our work is a very first study to compare the potentially toxic Hg and physiologically required PUFA contents of zoobenthos in two contrasting freshwater systems: Chinese reservoirs and Swedish lakes. Average THg and MeHg contents of zoobenthos in the Chinese reservoirs were higher than those in the Swedish lakes. The average EPA + DHA contents of zoobenthos were similar in the two habitats, yet DHA contents of zoobenthos were significantly higher in the Chinese reservoirs. Algal diet was associated with Hg and PUFA contents of zoobenthos in both the Chinese reservoirs and the Swedish lakes. By contrast, abiotic factors only affect total Hg accumulation of zoobenthos in the Swedish lakes, while in the Chinese reservoirs, DOC concentration correlated with MeHg and EPA, as well as DHA contents of zoobenthos. We argue that our study is conducive to assessing the dietary quality of aquatic food webs as well as developing strategies to alleviate Hg and enhance PUFA accumulations in zoobenthos in such freshwater food webs. Further research is recommended to elucidate the explicit mechanism of Hg uptake and metabolism processes in zoobenthos.

CRediT authorship contribution statement

Min Jing: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft. **Dan Lin:** Methodology, Investigation. **Pianpian Wu:** Conceptualization, Methodology, Investigation, Resources, Writing – review & editing. **Martin J. Kainz:** Conceptualization, Investigation, Writing – review & editing. **Kevin Bishop:** Conceptualization, Writing – review & editing, Funding acquisition. **Haiyu Yan:** Conceptualization, Visualization, Writing – review & editing, Supervision, Funding acquisition. **Qiuhua Li:** Resources, Writing – review & editing. **Xinbin Feng:** Writing – review & editing, Funding acquisition.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.146410.

References

Åkerblom, S., Negm, N., Wu, P., Bishop, K., Ahrens, L., 2017. Variation and accumulation patterns of poly- and perfluoroalkyl substances (PFAS) in European perch (*Perca fluviatilis*) across a gradient of pristine Swedish lakes. Sci. Total Environ. 599-600, 1685–1692.

Arslan, M., Rinchard, J., Dabrowski, K., Portella, M.C., 2008. Effects of different dietary lipid sources on the survival, growth, and fatty acid composition of South American *Catfish, Pseudoplatystoma fasciatum, Surubim, Juveniles*. J. World Aquacult. Soc. 39, 51–61.

Bell, M.V. and Tocher, D.R. 2009. Lipids in Aquatic Ecosystems. Kainz, M., Brett, M.T. and Arts, M.T. (eds), pp. 211–236, Springer New York, New York, NY.

Bishop, K., Shanley, J.B., Riscassi, A., de Wit, H.A., Eklöf, K., Meng, B., Mitchell, C., Osterwalder, S., Schuster, P.F., Webster, J., Zhu, W., 2020. Recent advances in understanding and measurement of mercury in the environment: terrestrial Hg cycling. Sci. Total Environ. 721, 137647.

Braaten, H.F.V., Lindholm, M., de Wit, H.A., 2020. Five decades of declining methylmercury concentrations in boreal foodwebs suggest pivotal role for sulphate deposition. Sci. Total Environ. 714, 136774.

Bravo, A.G., Peura, S., Buck, M., Ahmed, O., Mateos-Rivera, A., Herrero Ortega, S., Schaefer, J.K., Bouchet, S., Tolu, J., Björn, E., Bertilsson, S., 2018. Methanogens and iron-reducing bacteria: the overlooked members of mercury-methylating microbial communities in boreal lakes. Appl. Environ. Microbiol. 84, e01774-18.

Broadley, H.J., Cottingham, K.L., Baer, N.A., Weathers, K.C., Ewing, H.A., Chaves-Ulloa, R., Chickering, J., Wilson, A.M., Shrestha, J., Chen, C.Y., 2019. Factors affecting MeHg bioaccumulation in stream biota: the role of dissolved organic carbon and diet. Ecotoxicology 28, 949–963.

Charles T, D., Young-Ji, H.A.N., Celia Y, C., David C, E., Kathleen Fallon, L., Thomas M, H., Neil C, K., Ronald K, M., 2007. Mercury contamination in forest and freshwater ecosystems in the Northeastern United States. BioScience 57, 17–28.

Chen, C.Y., Folt, C.L., 2005. High plankton densities reduce mercury biomagnification. Environmental Science & Technology 39, 115–121.

Clayden, M.G., Kidd, K.A., Wyn, B., Kirk, J.L., Muir, D.C.G., O'Driscoll, N.J., 2013. Mercury biomagnification through food webs is affected by physical and chemical characteristics of lakes. Environmental Science & Technology 47, 12047–12053.

De Wit, H.A., Granhus, A., Lindholm, M., Kainz, M.J., Lin, Y., Braaten, H.F.V., Blaszczak, J., 2014. Forest harvest effects on mercury in streams and biota in Norwegian boreal catchments. For. Ecol. Manag. 324, 52–63.

Duan, J., Hu, H., Li, Q., Jiang, L., Zou, Y., Wang, Y., Sun, Z., 2016. Combined toxicity of silica nanoparticles and methylmercury on cardiovascular system in zebrafish (*Danio rerio*) embryos. Environ. Toxicol. Pharmacol. 44, 120–127.

Feng, X., Meng, B., Yan, H., Fu, X., Yao, H. and Shang, L. 2018a. Biogeochemical Cycle of Mercury in Reservoir Systems in Wujiang River Basin, Southwest China. Feng, X., Meng, B., Yan, H., Fu, X., Yao, H. and Shang, L. (eds), pp. 339–389, Springer Singapore, Singapore.

Feng, X., Meng, B., Yan, H., Fu, X., Yao, H. and Shang, L. 2018b. Biogeochemical Cycle of Mercury in Reservoir Systems in Wujiang River Basin, Southwest China. Feng, X., Meng, B., Yan, H., Fu, X., Yao, H. and Shang, L. (eds), pp. 95–199, Springer Singapore, Singapore.

Flannagan, John F., 1979. An introduction to the aquatic insects of North America. J. Fish. Res. Board Can. 36, 231.

GB-3838-2002 2002. Environmental Quality Standards for Surface Water. Ministry of Environmental Protection of the People's Republic of China. ICS 13.060 Z 50 (in Chinese).

Guo, Y., Zhao, Y.Z., Long, S.X., Li, Yulin, Zhang, B., 2015. Study on water environment characteristics in the stage of governance effect of lake eutrophication: taking drinking water sources of Hongfeng lake in Guiyang as example. Environmental Pollution and Prevention 37, 55–68.

Guo, F., Kainz, M.J., Sheldon, F., Bunn, S.E., 2016. The importance of high-quality algal food sources in stream food webs – current status and future perspectives. Freshw. Biol. 61, 815–831.

Guo, F., Lee, S.Y., Kainz, M.J., Brett, M.T., 2020. Fatty acids as dietary biomarkers in mangrove ecosystems: current status and future perspective. Sci. Total Environ. 739, 139907.

Hall, B.D., Bodaly, R.A., Fudge, R.J.P., Rudd, J.W.M., Rosenberg, D.M., 1997. Food as the dominant pathway of methylmercury uptake by fish. Water Air Soil Pollut. 100, 13–24.

He, T., Feng, X., Dai, Q., Qiu, G., Shang, L., Jiang, H., 2004. Determination of methyl mercury in sediments and soils by gc-cvafs after aqueous phase ethylation. Geology-Geochemistry 32, 83–86.

Heissenberger, M., Watzke, J., Kainz, M.J., 2010. Effect of nutrition on fatty acid profiles of riverine, lacustrine, and aquaculture-raised salmonids of pre-alpine habitats. Hydrobiologia 650, 243–254.

Horvat, M., Liang, L., Bloom, N.S., 1993. Comparison of distillation with other current isolation methods for the determination of methyl mercury compounds in low level environmental samples: part II. Water. Anal. Chim. Acta 282, 153–168.

Hu, H., Shi, Y., Zhang, Y., Wu, J., Asweto, C.O., Feng, L., Yang, X., Duan, J., Sun, Z., 2017. Comprehensive gene and microRNA expression profiling on cardiovascular system in zebrafish co-exposured of SiNPs and MeHg. Sci. Total Environ. 607–608, 795–805.

Hussain, B., Sultana, T., Sultana, S., Al-Ghanim, K.A., Al-Misned, F., Mahboob, S., 2019. Influence of habitat degradation on the fatty acid profiles of fish, microalgae, and zoobenthos in a river ecosystem. Process Saf. Environ. Prot. 123, 24–32.

Jardine, T.D., Kidd, K.A., O' Driscoll, N., 2013. Food web analysis reveals effects of pH on mercury bioaccumulation at multiple trophic levels in streams. Aquat. Toxicol. 132–133, 46–52.

Jing, M., Lin, D., Wu, P., Kainz, M.J., Bishop, K., Yan, H., Wang, R., Wang, Q., Li, Q., 2020. Effect of aquaculture on mercury and polyunsaturated fatty acids in fishes from reservoirs in Southwest China. Environ. Pollut. 257, 113543.

Kainz, M.J. and Fisk, A.T. 2009. Lipids in Aquatic Ecosystems. Kainz, M., Brett, M.T. and Arts, M.T. (eds), pp. 93–114, Springer New York, New York, NY.

Kainz, M., Mazumder, A., 2005. Effect of algal and bacterial diet on methyl mercury concentrations in zooplankton. Environmental Science & Technology 39, 1666–1672.

Lau, D.C.P., Vrede, T., Pickova, J., Goedkoop, W., 2012. Fatty acid composition of consumers in boreal lakes – variation across species, space and time. Freshw. Biol. 57, 24–38.

Li, Q., Han, B., 2007. Dynamics and structure of phytoplankton community in spring in a southern subtropical pumped-water reservoir. Journal of Tropical & Subtropical Botany 15, 294–300.

Li, L., Li, Y., Biswas, D.K., Nian, Y., Jiang, G., 2008. Potential of constructed wetlands in treating the eutrophic water: evidence from Taihu Lake of China. Bioresour. Technol. 99 (6), 1656–1663.

Liang, P., Feng, X., You, Q., Gao, X., Xu, J., Wong, H., Peter, C., Wu, C., 2017. The effects of aquaculture on mercury distribution, changing speciation, and bioaccumulation in a reservoir ecosystem. Environ. Sci. Pollut. Res. 24, 25923–25932.

Liu, Xueqin, 2006. Food Composition and Food Webs of Zoobenthos in Yangze Lakes. Doctoral dissertation. Institute of Hydrobiology. Chinese Academy of Sciences.

Liu, B., Yan, H., Wang, C., Li, Q., Guédron, S., Spangenberg, J.E., Feng, X., Dominik, J., 2012. Insights into low fish mercury bioaccumulation in a mercury-contaminated reservoir, Guizhou, China. Environ. Pollut. 160, 109–117.

Mahboob, S., Al-Ghanim, K.A., Al-Misned, F., Shahid, T., Sultana, S., Sultan, T., Hussain, B., Ahmed, Z., 2019. Impact of water pollution on trophic transfer of fatty acids in fish, microalgae, and zoobenthos in the food web of a freshwater ecosystem. Biomolecules 9, 231.

Maileht, K., Nõges, T., Nõges, P., Ott, I., Mischke, U., Carvalho, L., Dudley, B., 2013. Water colour, phosphorus and alkalinity are the major determinants of the dominant phytoplankton species in European lakes. Hydrobiologia 704, 115–126.

Martinz, Mariella, 2011. Effect of Aquaculture on Essential Nutrients and Potential Contaminants in Benthic Organisms and Fish of Pre-alpine Streams. Diploma thesis. University of Vienna.

Noh, S., Kim, C.-K., Kim, Y., Lee, J.-H., Han, S., 2017. Assessing correlations between monomethylmercury accumulation in fish and trophic states of artificial temperate reservoirs. Sci. Total Environ. 580, 912–919.

Parrish, C.C., Abrajano, T.A., Budge, S.M., Helleur, R.J., Hudson, E.D., Pulchan, K. and Ramos, C. (2000) Marine Chemistry. Wangersky, P.J. (ed), pp. 193–223, Springer Berlin Heidelberg, Berlin, Heidelberg.

Pavlicevic, N., Baltic, M., Dimitrijevic, M., Karabasil, N., Grbic, S., 2014. Polyunsaturated fatty acids in the fish meat and their significance for human health. Tehnologija Mesa 55, 1–7.

Pickhardt, P.C., Folt, C.L., Chen, C.Y., Klaue, B., Blum, J.D., 2002. Algal blooms reduce the uptake of toxic methylmercury in freshwater food webs. Proc. Natl. Acad. Sci. 99, 4419.

Razavi, N.R., Arts, M.T., Qu, M., Jin, B., Ren, W., Wang, Y., Campbell, L.M., 2014. Effect of eutrophication on mercury, selenium, and essential fatty acids in Bighead Carp (*Hypophthalmichthys nobilis*) from reservoirs of eastern China. Sci. Total Environ. 499, 36–46.

Razavi, N.R., Qu, M., Chen, D., Zhong, Y., Ren, W., Wang, Y., Campbell, L.M., 2015. Effect of eutrophication on mercury (Hg) dynamics in subtropical reservoirs from a high Hg deposition ecoregion. Limnol. Oceanogr. 60, 386–401.

Rennie, M., Collins, N., Purchase, C., Tremblay, A., 2005. Predictive models of benthic invertebrate methylmercury in Ontario and Quebec lakes. Can. J. Fish. Aquat. Sci. 62, 2770–2783.

Rhee, J.J., Kim, E., Buring, J.E., Kurth, T., 2017. Fish consumption, omega-3 fatty acids, and risk of cardiovascular disease. Am. J. Prev. Med. 52, 10–19.

Rosa-Silva, H.T.d., Panzenhagen, A.C., Espitia-Pérez, P., Teixeira, A.A., Roitman, A., Almeida, R.F., Heimfarth, L., Moreira, J.C.F., 2020. Effects of foetal and breastfeeding exposure to methylmercury (MeHg) and retinol palmitate (vitamin A) in rats: redox parameters and susceptibility to DNA damage in liver. Mutation Research/Genetic Toxicology and Environmental Mutagenesis 858–860, 503239.

Schultz, S., Vallant, B., Kainz, M.J., 2012. Preferential feeding on high quality diets decreases methyl mercury of farm-raised common carp (*Cyprinus carpio L.*). Aquaculture 338-341, 105–110.

Seekell, D., Lapierre, J.F., Ask, J.B., Ann, K., Deininger, A., Rodriguez, P., Karlsson, Jan, 2015. The influence of dissolved organic carbon on primary production in northern lakes: influence of DOC on production. Limnol. Oceanogr. 60, 1276–1285.

Shen, Z., Niu J., Wang Y., Wang H. and Zhao X. 2013. Eutrophication and Distribution of Nitrogen and Phosphorus. Distribution and Transformation of Nutrients and Eutrophication in Large-scale Lakes and Reservoirs: The Three Gorges Reservoir. Shen Z., Niu J., Wang Y., Wang H. and Zhao X. Berlin, Heidelberg, Springer Berlin Heidelberg: 17–42.

Strandberg, U., Palviainen, M., Eronen, A., Piirainen, S., Laurén, A., Akkanen, J., Kankaala, P., 2016. Spatial variability of mercury and polyunsaturated fatty acids in the European perch (*Perca fluviatilis*) – implications for risk-benefit analyses of fish consumption. Environ. Pollut. 219, 305–314.

USEPA, 1998. Method 7473 (SW-846): Mercury in Solids and Solutions by Thermal Decomposition, Amalgamation, and Atomic Absorption Spectrophotometry U.S. Environmental Protection Agency.

USEPA 2001. Method 1630: Methyl mercury in Water by Distillation, Aqueous Ethylation, Purge and Trap, and CVAFS. U.S. EPA-821-R-01-020. Washington, DC Environmental Protection Agency.

USEPA, 2002. Method 1631, Reversion E: Mercury in Water by Oxidation, Purge and Trap, and Cold Vapor Atomic Fluorescence Spectrometry.

Vander Zanden, M.J., Vadeboncoeur, Y., 2002. Fishes as integrators of benthic and pelagic food webs in lakes. Ecology 83, 2152–2161.

Vieira, H.C., Rendón-von Osten, J., Soares, A.M.V.M., Morgado, F., Abreu, S.N., 2020. Contribution of commercial fish species to human mercury exposure: an evaluation near the Mid-Atlantic Ridge. J. Food Compos. Anal. 103688.

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

Willem, G., Lars, S., Gunnel, A., Merike, B., 2000. Fatty acids in profundal benthic invertebrates and their major food resources in lake Erken, Sweden: seasonal variation and trophic indications. Canadian Journal of Fisheries & Aquatic Sciences 57, 2267–2279.

Wu, P., Bishop, K., von Brömssen, C., Eklöf, K., Futter, M., Hultberg, H., Martin, J., Åkerblom, S., 2018. Does forest harvest increase the mercury concentrations in fish? Evidence from Swedish lakes. Sci. Total Environ. 622–623, 1353–1362.

Wu, P., Kainz, M., Åkerblom, S., Bravo, A.G., Sonesten, L., Branfireun, B., Deininger, A., Bergström, A.-K., Bishop, K., 2019. Terrestrial diet influences mercury bioaccumulation in zooplankton and macroinvertebrates in lakes with differing dissolved organic carbon concentrations. Sci. Total Environ. 669, 821–832.

Yan, H., Feng, X., Shang, L., Tang, S. and Qiu, G. 2003. Speciation analysis of ultra trace levels of mercury in natural waters. Journal of Instrumental Analysis 22, 10–13. (Chinese with a English abstract).

Yan, H., Feng, X., Liu, T., Shang, L. and Li, Z. 2008. Present situation of fish mercury pollution in heavily mercury-contaminated Baihua reservoir in Guizhou. Chinese Journal of Ecology 27, 1357–1361. (Chinese with a English abstract).

Yan, H., Li, Q., Meng, B., Wang, C., Feng, X., He, T., Dominik, J., 2013. Spatial distribution and methylation of mercury in a eutrophic reservoir heavily contaminated by mercury in Southwest China. Appl. Geochem. 33, 182–190.

Zeng, Q., Liu, Y., Zhao, H., Sun, M., Li, X., 2017. Comparison of models for predicting the changes in phytoplankton community composition in the receiving water system of an inter-basin water transfer project. Environ. Pollut. 223, 676–684.

Zhang, T.T., Xu, J., Wang, Y.M., Xue, C.H., 2019. Health benefits of dietary marine DHA/EPA-enriched glycerophospholipids. Prog. Lipid Res. 75, 100997.