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Total and methyl mercury in the water, sediment, and fishes of Vembanad, a tropical backwater system in India

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Abstract Mercury contamination in the water bodies of developing countries is a serious concern due to its toxicity, persistence, and bioaccumulation. Vembanad, a tropical backwater lake situated at the southwest coast of India, is the largest Ramsar site in southern India. The lake supports thousands of people directly and indirectly through its resources and ecosystem services. It is highly polluted with toxic pollutants such as heavy metals, as it receives effluent discharges from Kerala's major industrial zone. In the present study, water, pore water, sediment, and fish samples collected from Vembanad Lake were analysed for total mercury (THg) and methyl mercury (MHg) contents. The maximum concentrations of THg and MHg in surface water samples were31.8 and 0.21 ng/L, respectively, and those in bottom water samples were 206 and 1.22 ng/L, respectively. Maximum concentration of THg in surface sediment was observed during monsoon season (2850 ng/g) followed by that in the pre-monsoon season (2730 ng/g) and the post-monsoon season (2140 ng/g). The highest sediment concentration of MHg (202.02 ng/g) was obtained during monsoon season. The spatial variation in the mercury contamination clearly indicates that the industrial discharge into the Periyar River is a major reason for pollution in the lake. The mercury pollution was found to be much higher in Vembanad Lake than in other wetlands in India. The bioaccumulation was high in carnivorous fishes, followed by benthic carnivores. The THg limit in fish for human consumption (0.5 mg/kg dry wt.) was exceeded for all fish species, except for *Glossogobius guiris* and *Synaptura orientalis*. The concentration of THg was five times higher in *Megalops cyprinoides* and four times higher in *Gazza minuta.* Significant variation was observed among species with different habits and habitats. Overall, risk assessment factors showed that the mercury levels in the edible fishes of Vembanad Lake can pose serious health impacts to the human population.

Keywords Heavy metal · Bioaccumulation · Sediment pollution

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Introduction

Mercury is an extremely toxic trace metal, naturally occurring in air, water, and soil (Lin et al. 2007; Li et al. 2009) and is one of the most studied pollutants (Geen Ruiz et al. 2005). Because of its transboundary nature, mercury is depositing in remote places where no specific sources present (Azevedo-Silva et al. 2016; Kang et al. 2016), with burdens in sediments and other non-biological materials estimated to have increased up to five times pre-human levels, primarily as a result of anthropogenic activities (Daga et al. 2016; Ramasamy et al. 2012; Brim et al. 1994). Earlier studies estimated a global natural mercury emission of 1800–5800 tons/year (Li et al. 2009). The global anthropogenic mercury emission to the atmosphere in 2000 was 2190

tons (Pacyna et al. 2006) and 54% of that was contributed by Asian countries (Li et al. 2009). Therefore, a number of studies have been undertaken on mercury contamination (Kang et al. 2016; Yin et al. 2016; Mason et al. 2006; Bhattacharya et al. 2014; Subramanian et al. 2003), speciation (Gabriel and Williamson 2004), bioavailability (Boszke et al. 2007; Bower et al. 2008; Davis et al. 1997), and bioaccumulation (Lawson and Mason 1998; Sunderland 2007) in various ecosystems.

Earlier studies estimated a global natural mercury emission of 1920.6 tons/year (Pacyna and Pacyna 2002; Wilson et al. 2010). Asian countries are the highest emitter (two thirds of the total emission) of anthropogenic mercury into the atmosphere (Pacyna et al. 2011). India is one among the top three emitters of mercury. According to Ryzhkov et al. (2011) mercury deposition has been significantly increased during the period of 1995–2005 in East and South Asia at 26 and 8% respectively.

The major sink of mercury in aquatic ecosystems is sediments. In sediments, it can stay for a long period and be transformed to more toxic, water-soluble organic forms (mainly methyl mercury) through biotic or abiotic processes (Tomiyasu et al. 2000). Mercury is very hazardous to aquatic ecosystems, especially for brackish water systems because of their high biological productivity (Geen Ruiz et al. 2005). Methyl mercury (MHg) accounts for the major portion of total mercury (THg) accumulated in fish (Lawrence and Mason 2001), posing a threat to human health because of its large half-life for elimination (UNEP 2002; Wiener and Spry 1996) and its biomagnification capability (Mieiro et al. 2009; Cizdziel et al. 2003). Mercury can also cause significant health impacts on fishes and other wildlife (Wiener et al. 2003; Scheuhammer et al. 2007).

Tropical regions (except the Florida Everglades and the Amazon basin) are less studied compared to temperate regions (Watras et al. 1994; Bowles et al. 2001). Therefore, it is very important to study the mercury pollution in developing countries such as India, which has become one of the hotspots of mercury pollution (Sharma 2003). There was little data in the United Nations Environment Program's (UNEP) report (2002, 2008, and 2013) on mercury monitoring from India, which has been the largest user of mercury in processes and products releasing large amounts of mercury into the environment (Omana and Mahesh 2008; Sharma 2003; Down to Earth 2003; BSCB 2003).

Very few studies have been carried out on mercury pollution along the coastal regions of India (Mohan et al. 2014; Ramasamy et al. 2012; Omana and Mahesh 2008; Ram et al. 2003; Krishnamoorthy and Nambi 1999; Ouseph 1992; Mahajan and Srinivasan 1988; Srinivasan and Mahajan 1989). Studies on contamination of mercury in the lakes and rivers of India are equally meagre (Das et al. 2015; Karunasagar et al. 2006; Mohan and Omana 2004; Agarwal et al. 2007). The rise in mercury resistant bacteria (MRB) also indicates increased mercury pollution in these areas (Ramaiah and De 2003). Vembanad backwater, situated on the southern part of the western coast of India, is highly polluted due to industrial effluents, insecticide usage in agricultural fields, urban sewage, and soil erosion caused by the deforestation in the highland areas. More than 60% of industries in Kerala, including the chlor-alkali industry, are situated on the banks of the Vembanad backwaters (Ouseph 1996). Mercury cell process was used in the chlor-alkali industry and has been releasing three times of the national average. However, the industry decommissioned the mercury cell process and adopted membrane filter technology in 2004. Earlier studies (Li et al. 2009; Karunasagar et al. 2006; Lindeström 2001) report that even after elimination of such sources, mercury remains in the environmental matrices and may take a long period to return to background concentration levels.

In this context, it is very important to study the present state of mercury pollution in the Vembanad backwaters, particularly since a large number of people for their livelihood are using it. There are about 20,000 people engaged in fishing, with an annual catch of 72,000 tons. Furthermore, Vembanad is a Ramsar site. The present study is therefore significant in terms of the knowledge acquired on the distribution patterns of and factors influencing mercury concentrations in the water and sediment. The present study also investigates the accumulation of mercury in selected edible fishes of this backwater, and analyses the potential hazards of contaminated fish to the human population living around Vembanad.

Materials and methods

Study area

Vembanad Lake, one of three Ramsar sites in Kerala, is a brackish, humid tropical wetland ecosystem having an area of 151,250 ha (Fig. 1), and is of extraordinary importance for its hydrological function, its biodiversity,

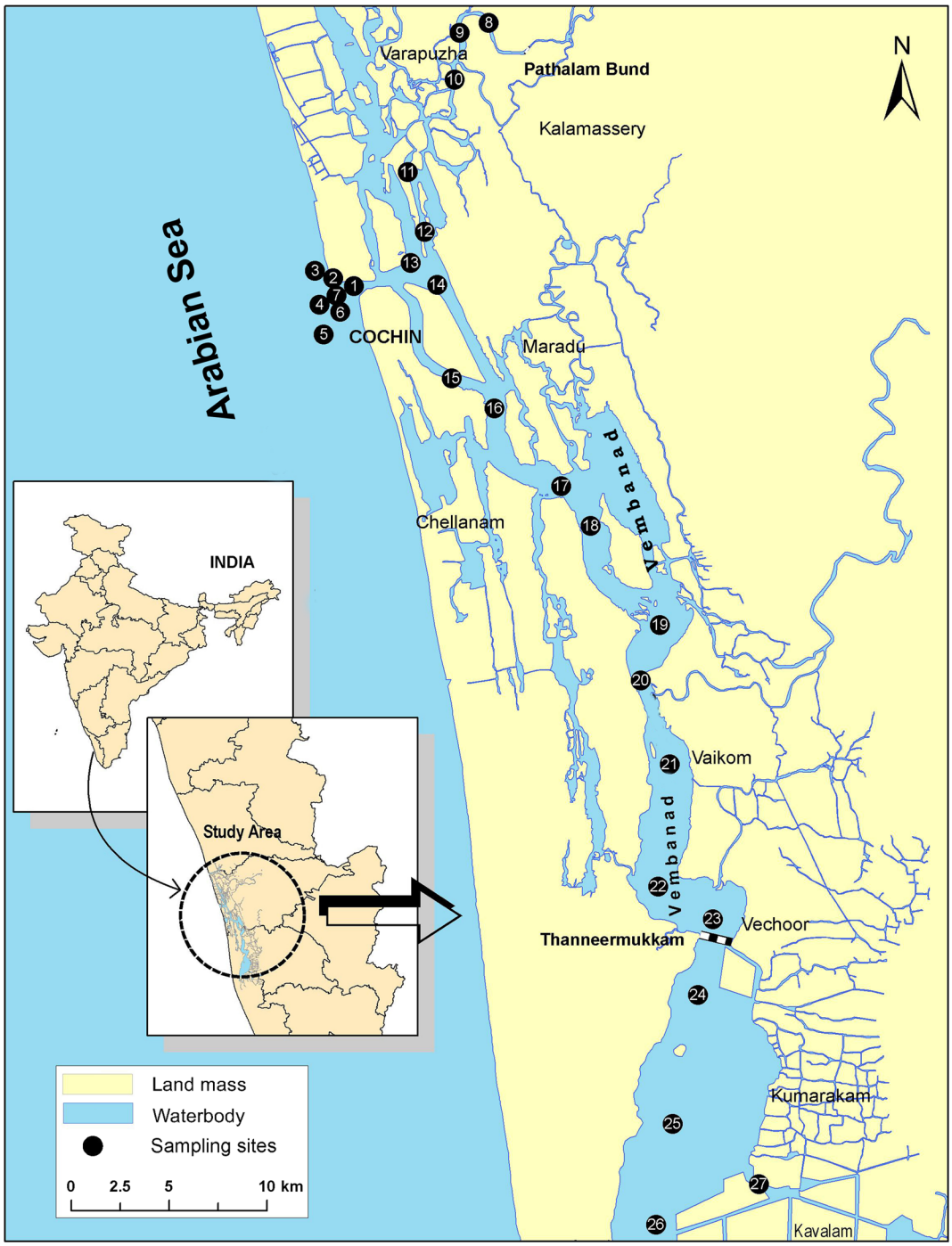

Fig. 1 Location map showing sampling sites in the Vembanad backwater system

and its support of fish populations. The lake is connected to the Arabian Sea through Cochin estuary (Jayakumar 2002). The physical settings of Vembanad wetland has been well established by the earlier works of authors and other studies (Mohan et al. 2014; Joseph and Ouseph 2010; Ramasamy et al. 2012; Mohan and Omana 2007). Six major rivers are contributing fresh water into this system. Hence, Vembanad wetland is

highly influenced by the fresh water input during the monsoon season as any other wetland systems in south west coast of India (Babu et al. 2010).

Reagents

All samples, reagents, and standards were prepared using ultrapure water (18.2 MΩ). Working standards were prepared from 1000-ppm standard stock solution of mercury (Merck, Germany). Stannous chloride ($SnCl_2$) and sodium borohydride ($NaBH_4$) were used as the reducing agents and were prepared daily. All glassware was cleaned according to the USEPA method 1631 RE (2002).

Instruments

A Cold Vapour Atomic Fluorescence Spectrometer (CVAFS- Model III, Brooks Rand, USA) was used for the determination of Hg concentration in all samples. Hg-free N_2 was used as a purging gas, while high-purity A\ argon was used as a carrier gas. Cold vapour of elemental mercury formed during aqueous reduction with $SnCl_2$/$NaBH_4$. The resulting Hg vapour was then purged with nitrogen and trapped in a gold trap. After 20 min of purging, the gold trap was moved to the trap desorption module, where the amalgamated mercury was thermally desorbed into the carrier gas stream and detected in the CVAFS detector.

An ultrasonic processor with 130 W power and 20 kHz frequency (Cole-Parmer Instruments, USA) was used for ultrasound-assisted extraction. Organic forms of mercury were converted to inorganic form using UV irradiation in a photochemical reactor (Scientific Aids and Instruments Corporation, India).

Sample collection and preservation

Water and sediment samples (250 g) were collected from 30 locations of the Vembanad Lake, near-shore areas and adjoining river systems during the post-monsoon, pre-monsoon, and monsoon seasons (Fig. 1). Surface and bottom water samples were collected and preserved as per standard procedures for analysis of total mercury and methyl mercury (Parker and Bloom 2005; USEPA 2001, 2002). Sediment samples were collected using a grab sampler, transferred to plastic bags, and kept in a deep freezer at 0 °C.

Fish samples were collected from the study area using traditional fishing methods, such as Chinese dip-nets, with the help of local fishermen. Three specimens of each species were captured for the analysis. The fish samples were sealed in polyurethane bags and kept in a freezer at 0 °C. The length and weight of each fish were measured and were sealed in polythene bags and kept in freeze until analysis. The collected fish species form a major part of the human diet for populations living on the banks of Vembanad Lake.

THg and MHg in water

Total mercury (THg) was determined with CVAFS following bromine monochloride (BrCl) oxidation (USEPA–Method 1631, revision E, 2002).Organic mercury was extracted from water samples by selective extraction with dichloromethane (CH_2Cl_2) and HCl followed by UV irradiation (Logar et al. 2001 and Karunasagar et al. 2006). After the selective extraction, the organic layer was evaporated and mercury was extracted back into water. One portion of the solution was analysed for inorganic mercury. The second portion undergone UV irradiation in a photochemical reactor after acidification with 2%HCl for converting organic to inorganic forms of mercury. The UV irradiated samples were reduced with $NaBH_4$ and detected inorganic mercury with CVAFS. The difference between inorganic and total mercury from the split sample was taken as the methyl mercury as the other forms of organic mercury are less stable, (Karunasagar et al. 2006).

THg and MHg in surface sediments

Sediment samples for THg analysis were dried at 40 °C. The dried samples were subsequently ground in an agate mortar and sieved to <63 μm fraction. Samples for THg analysis (0.1–0.25 g) were subjected to hot re-fluxing HNO_3/H_2SO_4 digestion followed by bromine monochloride (BrCl) oxidation (USEPA 2001).

Alkali-leaching solvent extraction method (25% solution KOH-methanol), proposed by Liang et al. (1996) and later modified by Karunasagar et al. (2006), was used for the extraction of methyl mercury (MHg) in sediments. The results were presented in wet weight basis. After the solvent extraction, MHg was determined by the difference of total and inorganic Hg, as described above for water samples.

THg and MHg in fish

The fish samples were defrosted and rinsed with deionized water. Muscle tissue was extracted with stainless-steel instruments. Each tissue sample was dried at 50 °C to a constant weight. Hot re-fluxing with HNO_3/H_2SO_4 digestion was followed by bromine chloride oxidation (USEPA 2001) for the extraction of mercury. MHg was extracted by rapid ultrasonic extraction (Krishna et al. 2005) followed by UV irradiation (Karunasagar et al. 2006). Extracted solutions were then analysed in the way described for THg and MHg in water samples.

Quality assurance

In order to check procedure validity, Certified Reference Materials (from the European Reference Materials of the Institute of Reference Materials and Measurements) ERM CC580 (Estuarine sediment) and ERM CE464 (Tuna fish) were used (Table 1). Replicates of the samples were analysed regularly. The detection limit of Hg was 0.08 ng/L. Pearson's correlation (SPSS-11) was used for establishing the relationship between background Hg concentrations and the concentrations in fish.

Results and discussion

THg in water

The minimum and maximum concentrations of THg in surface water samples of post-monsoon were 5 and 31.8 ng/L, respectively (Table 2). During the pre-monsoon and monsoon seasons, THg varied from 6.7 to 28.4 and 8.3 to 31.5 ng/L, respectively. In the case of bottom water, the THg concentration was observed to be the maximum during the post-monsoon season (206 ng/L) and the minimum during the pre-monsoon season (2.4 ng/L).

The mean THg of surface and bottom water samples was calculated and graphically represented (Fig. 2). The mean concentration of THg in surface water samples from the different seasons did not show much variation (16.5–17.0 ng/L).Seasonal variation was observed, however, in bottom water THg concentrations (21.6–44.4 ng/L). The maximum mean THg concentration occurred in the bottom water collected during the post-monsoon season (44.4 ng/L). The maximum mean THg content was observed in the post-monsoon season (206 ng/L) whereas during the pre-monsoon and monsoon seasons, the mean THg concentrations varied between 43.7 and 58.1 ng/L. The minimum water concentration (2.4 ng/L) was observed during the pre-monsoon season. THg was high in northern parts of the lake and near-shore areas.

The Vembanad backwater system is being influenced by six major rivers of Kerala originated from the southern Western Ghats. The system is fully fresh water dominated with high rate of flow during the monsoon time. These rivers may carry a huge amount of surface runoff water from the catchment area. High fresh water flow may influence the sedimentary environment too as the depth of the lake is shallow and most of the places are 1.5 to 4 m deep except water ways where it is varied from 8 to 10 m. The high bottom water concentration obtained in near-shore regions during the post-monsoon season might be due to the dissolution of mercury from dredged sediments and to particulate matter carried by rivers during the end of the monsoon season. That the highest concentrations occur mainly in the bottom waters of the lower reaches of the Periyar River indicates dissolution of mercury (inorganic or organic) from bottom sediments. Furthermore, volatilization of Hg^{2+} due to photo-reduction can also happen during the pre-monsoon season, which would lower surface water concentrations.

MHg in water

The results showed that MHg content in water samples of all seasons was very low (Table 3). MHg was observed only at a few sites. The maximum MHg concentration in surface water observed during the post-

Table 1 Values obtained for certified reference materials

CRM	Matrix	Metal	Certified value (mg/kg)	Obtained value (mg/kg)	Recovery %
ERM CC580	Estuarine sediment	Total Hg	132 ± 3	129.45 ± 2.38	98.07
ERM CE464	Tuna fish	Total Hg	5.24 ± 0.10	5.17 ± 0.32	98.66

Table 2 THg (ng/L) in water samples of Vembanad Lake during various seasons

Sample no.	Post-monsoon		Pre-monsoon		Monsoon	
	Surface	Bottom	Surface	Bottom	Surface	Bottom
1	11.2	206.0	10.6	28.4	26.4	28.3
2	24.0	63.0	10.4	20.1	22.3	31.4
3	26.8	115.0	14.5	32.6	12.7	16.8
4	19.4	53.7	10.0	16.3	14.6	16.4
5	19.4	55.9	16.8	22.4	20.4	29.5
6	31.8	84.4	28.2	29.4	22.8	27.5
7	27.4	69.7	26.4	34.7	17.6	21.2
8	21.1	41.6	21.2	33.8	31.5	44.1
9	19.6	32.7	17.2	22.8	16.1	41.5
10	nd	23.8	20.5	29.4	21.6	31.2
11	nd	18.9	28.4	30.8	12.5	46.4
12	nd	39.8	24.6	32.9	23.8	27.9
13	16.0	27.1	nd	20.4	26.5	29.5
14	21.3	76.4	26.1	43.7	23.7	58.1
15	8.3	18.8	20.2	21.5	20.7	20.9
16	7.2	12.7	nd	10.4	16.1	26.5
17	13.3	18.6	10.6	19.3	14.3	17.9
18	18.2	22.5	10.5	19.8	9.4	21.8
19	17.6	68.8	8.4	17.6	10.7	11.4
20	16.5	47.7	10.0	11.2	15.4	17.3
21	16.2	66.7	6.7	10.2	12.3	17.6
22	18.4	22.2	16.1	17.3	16.1	7.8
23	13.7	22.2	14.0	17.46	22.7	10.2
24	12.1	21.9	14.7	16.8	10.1	12.6
25	nd	7.7	nd	2.4	14.0	22.0
26	5.0	10.1	15.1	18.1	13.3	11.4
27	7.7	18.9	17.1	17.8	8.3	14.7
28	13	21.5	8.5	10.7	14.0	14.3
29	6.8	16.2	13.8	14.3	9.6	21.2
30	20.6	27.2	23.7	24.6	9.0	11.7

nd not detectable

monsoon season was 0.18 ng/L, while in the pre-monsoon and monsoon seasons, it was 0.21 ng/L. Among bottom water samples, the highest concentrations were observed in post-monsoon samples (1.22 ng/L). The minimum MHg concentration in bottom water varied from 0.1 ng/L (monsoon) to 0.3 ng/L (pre-monsoon).

As in the case of THg, variation in MHg concentration along different sites was observed only for bottom water (0.22–0.49 ng/L) and very little difference was observed in the surface water (0.13–0.16 ng/L). The maximum mean MHg was observed during the post-monsoon season (0.49 ng/L).

The mean values of surface and bottom water MHg showed that high concentrations were obtained during the post-monsoon season (Fig. 3). However, at the lower reaches of the Periyar River, post-monsoon samples showed a lower concentration than the other seasons. The sites in the southern part of Vembanad Lake showed maximum concentrations during the post-monsoon season, while in the pre-monsoon season MHg was below detection level.

The Lake Kodaikanal, which was polluted with the effluent from a thermometer factory, showed a higher concentration (34.24–57.36 ng/L) than the Vembanad (0.11–0.52 ng/L) (Karunasagar et al. 2006). The Cochin near-shore region contained a much lower THg than the Kalpakam near-shore (20,420 ng/L) on the southwestern coast of India (Satpathy et al. 2008). The results obtained by Kannan et al. (1998) showed that the concentration of methyl mercury accounted for 0.03–52% of the total mercury. In the case of Kodaikanal Lake, around 10% of the total mercury accounted for MHg, where the total mercury concentration of 356–465 ng/L and methyl mercury concentration of 50 ng/L were observed (Karunasagar et al. 2006). In this study, MHg content was 1.09% of THg.

THg and MHg in surface sediment

THg and MHg in sediments during various seasons are given in Figs. 4 and 5. 13.3% of the samples are below the detection limit for THg. The maximum concentration of THg during the post-monsoon period was observed at Site 9 (2140 ng/g), and the mean THg concentration obtained during this season was 387.49 ng/g (Fig. 4). The THg varied between 382 and 2140 ng/g along the northern limb of Cochin estuary (sites 8–13), whereas the southern part of Vembanad Lake (south of Thanneermukkom bund—sites 27–30) had very low concentrations (17–74.5 ng/g).

THg concentration was ranged from 47 ng/g (site 29) to 2730 ng/g (site 11) with a mean concentration of 755.72 ng/g during pre-monsoon (Fig. 4). Thirty percent of the samples were below detection limit. THg at the confluence point of Periyar River with Cochin estuary (sites 8–13) ranged from 550 to2730ng/g. Maximum THg obtained during monsoon season were 2850 ng/g

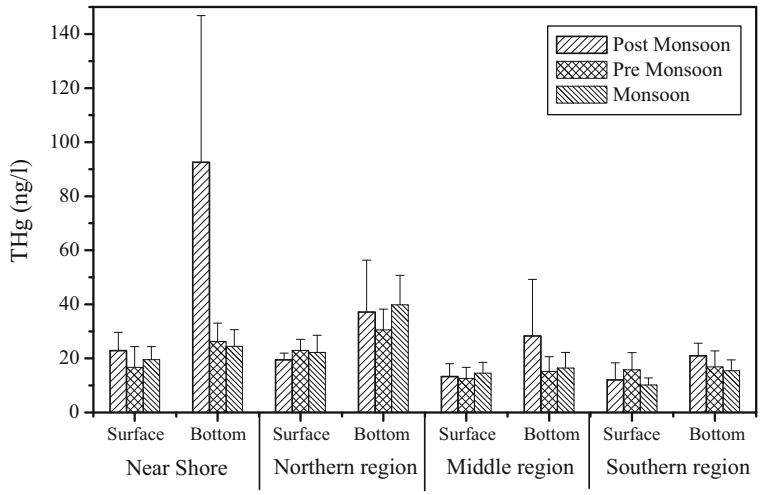

Fig. 2 Seasonal variation of THg in the water of Vembanad Lake and near-shore areas

(site 8) and the mean THg observed was 507.07 ng/g. The concentration was found higher at the near-shore regions.

Very few (13.3%) samples had detectable MHg, and the concentration varied from 0.38 to 9 ng/g during the post-monsoon season. The mean MHg was 6.35 ng/g during the pre-monsoon season. Seventy-seven percent of samples were below detection limit for MHg. Maximum MHg, obtained during monsoon season was 202.02 ng/g (site 11) with a mean of 22.57 ng/g. The mean concentration for THg obtained in this study was higher than the background concentration (200 ng/g) suggested by earlier studies (Craig 1986; Lindqvist et al. 1984).

The sediments of northern limb of the lake have high organic mercury content (sites 8–13) (Fig. 5). All other samples except few in this region and central part of the Lake were below the background concentration (<2 ng/g) as suggested by Davis et al. (1997). The distribution pattern of total mercury in the lake sediment showed high concentration at sites8–15. All of these points are located near the industrial area downstream to the bar mouth. The industrial effluent discharge into the river is a major source of this pollution. Slightly higher concentrations of THg were also recorded from the middle portion of the lake (sites 22 to 25). This may be due to the contribution of effluents from a public sector newsprint-manufacturing unit, which discharges its effluents to the Muvattupuzha River. High concentrations of THg in the comparatively unpolluted southern region of the lake are also an example of indiscriminate disposal of municipal solid wastes from the developing regions of central Kerala. Moreover, the unique water current pattern existing in the lake prevents the mixing and distribution of the Periyar River water from the northern limb to the southern portion of the lake, which is in turn nourished by the Muvattupuzha River. Since the flow from the Periyar River is directly discharged into the sea, the industrial pollution in the lake is confined to a small portion of the northern limb. However, the human population and the fishing activity in the area is very high, posing great risk to human beings even though the actual polluted area is small.

Comparison with Indian sites

Studies on mercury contamination in Indian aquatic ecosystems are rare, and hence, a comparison of THg concentrations in sediments with the concentrations reported for other lakes, rivers, and estuaries in India was very difficult. Still, the comparison (Table 4) indicated that the mean THg content in Vembanad Lake was higher than that in the Kodaikanal Lake (freshwater). The latter one was polluted with the effluent from a thermometer factory. However, the organic mercury (MHg) content was higher in Kodaikanal Lake. Not many studies were reported methyl mercury concentration in the aquatic systems of India except Karunasagar et al. (2006). Pristine lake like Berijam and Kukkal showed low THg content than the Vembanad.

THg content in Cochin estuary (<0.001–1.88 μg/g) observed in the present study is higher than the values obtained by Ouseph (1992) for the same region. This might be due to the release of industrial

Table 3 MHg (ng/L) in water samples of Vembanadu Lake during various seasons

Sample no.	Post-monsoon		Pre-monsoon		Monsoon	
	S	B	S	B	S	B
1	nd	1.22	nd	0.18	0.14	0.23
2	nd	0.50	0.12	0.16	0.18	0.28
3	0.10	0.78	0.16	0.22	nd	0.16
4	nd	nd	nd	nd	nd	nd
5	0.12	0.46	nd	nd	0.14	0.21
6	0.13	0.82	0.21	0.25	nd	nd
7	nd	nd	nd	0.27	nd	nd
8	nd	0.40	0.17	0.24	0.21	0.24
9	nd	0.28	0.12	0.22	nd	0.43
10	nd	0.18	0.19	0.30	0.11	0.27
11	nd	nd	nd	0.26	nd	0.29
12	nd	0.34	nd	0.25	nd	0.29
13	nd	nd	nd	nd	nd	0.18
14	nd	0.67	nd	nd	nd	0.19
15	nd	nd	nd	nd	nd	nd
16	nd	nd	nd	nd	nd	nd
17	nd	nd	nd	nd	nd	0.11
18	nd	nd	nd	nd	nd	0.12
19	nd	0.46	nd	nd	nd	nd
20	nd	0.28	nd	nd	nd	nd
21	nd	0.32	nd	nd	nd	nd
22	nd	nd	nd	nd	nd	nd
23	nd	0.18	nd	nd	nd	nd
24	nd	nd	nd	nd	nd	0.10
25	nd	nd	nd	nd	nd	0.26
26	nd	nd	nd	nd	nd	nd
27	nd	nd	nd	nd	nd	nd
28	nd	0.11	nd	nd	nd	nd
29	nd	nd	nd	nd	nd	0.20
30	nd	nd	nd	nd	nd	nd

nd not detectable

mercury as discussed earlier. However, the sediments of Cochin estuary have shown lower concentrations compared to other estuarine systems of India (Table 4). THg concentration of 0.38–2.85 μg/g as observed in the present study in the industrial region of study area was less compared with the earlier study (5.5–11.5 μg/g) carried out in the same region by Ouseph (1992). Also, he has demonstrated that the upstream regions of industrial area got very low values (0.04–0.05 μg/g) when compared with the lower reaches of Periyar river (Table 4).

Smith et al. (1996) suggested threshold effect level (TEL) (0.13 μg/g) and probable effect level (PEL) (0.70 μg/g) for total mercury in sediments. The mean concentration observed for THg in Vembanad Lake (0.395 μg/g) was between the observed TEL and PEL and this could be associated with adverse biological effects. In the present study, 16% of the sampling sites had Hg content higher than PEL while those of rest of the sites fell between TEL and PEL; hence, adverse effects on biota are expected to occur frequently.

The mercury concentration in sediments was high in the northern part of the lake, which may be the remnants of the once deposited mercury from the chlor-alkali industry. Even though the discharge of mercury has been ceased, the remnant of the mercury remains in the environment for a long time (Alonso et al. 2000; Karunasagar et al. 2006).

THg in pore water

Seasonal THg variation of pore water samples was analysed and the results were graphically represented (Fig. 6). During the post-monsoon season, site 12 showed a maximum concentration of 85.2 ng/L whereas site 29 was observed with the lowest value of 12.9 ng/L. Maximum THg in the pore water samples of the pre-monsoon season was observed at Site 13 (47.1 ng/L). The minimum concentration during this period was 7.4 ng/L. THg content of pore water samples varied from 0.075 to 22.96 ng/L. A larger number of samples had detectable THg content during pre-monsoon (84%) followed by post-monsoon (60%) and monsoon (53.4%).

THg in pore water samples was high during the post-monsoon season, except for the samples from the southern part of the lake and in near-shore areas where it was high during the pre-monsoon season (Fig. 6). The THg content in pore water samples from near-shore areas was detected only during the pre-monsoon season. During the monsoon season, mercury concentration was very low. High pore water mercury concentrations were observed in the northern part of the Cochin estuary followed by the central part of the lake (Fig. 6). During monsoon season, THg in pore water (THg_{PW}) was not detected in the southern part of the lake. The Muvattupuzha River samples also did not have detectable THg_{PW} content during any of the seasons.

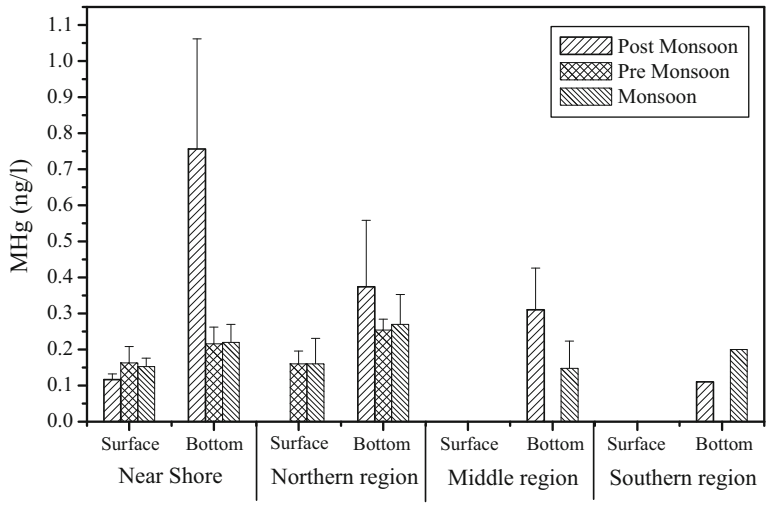

Fig. 3 Seasonal variation of MHg in the water samples of Vembanad Lake and near-shore areas

THg and MHg in fish muscle

The length and weight of collected fish were measured (Table 5). Previous studies indicate that in non-tropical environments, seasonal variations have no significant effect on mercury bioaccumulation. According to Lewis and Chancy (2007), natural, physical, chemical, and biological factors of habitat, along with life stage, life mode, and feeding category are important factors in bioaccumulation of Hg. The fish samples were studied for total and organic mercury accumulation in the muscle tissue. Samples were analysed for each fish and the mean values are given in Table 6. The THg was calculated based on a dry weight (d.wt.) basis whereas MHg was based on a wet weight (w.wt.) basis. The fish were classified according to feeding habitat as benthic carnivorous, carnivorous (feeding fishes and zooplanktons), or omnivorous. Of the two benthic carnivorous fishes sampled, *Arius arius* showed higher THg (0.977 mg/kg) and MHg content (0.648 mg/kg *w wt.*) than *Synaptura orientalis* (Table 6). Of the carnivorous fishes, the maximum THg concentrations were observed in the muscles of *Caranx affinis* (3 mg/kg), and the minimum in *Glossogobius giuris* (0.118 mg/kg). Among the carnivorous fishes, MHg was not detectable in *Thryssa mystax* and varied from 0.46 mg/kg w.wt (*Gazza minuta*) to 2.26 mg/kg w.wt. (*C. affinis*) (Table 6). *Cynoglossus semifasciatus* showed the maximum THg (2.85 mg/kg

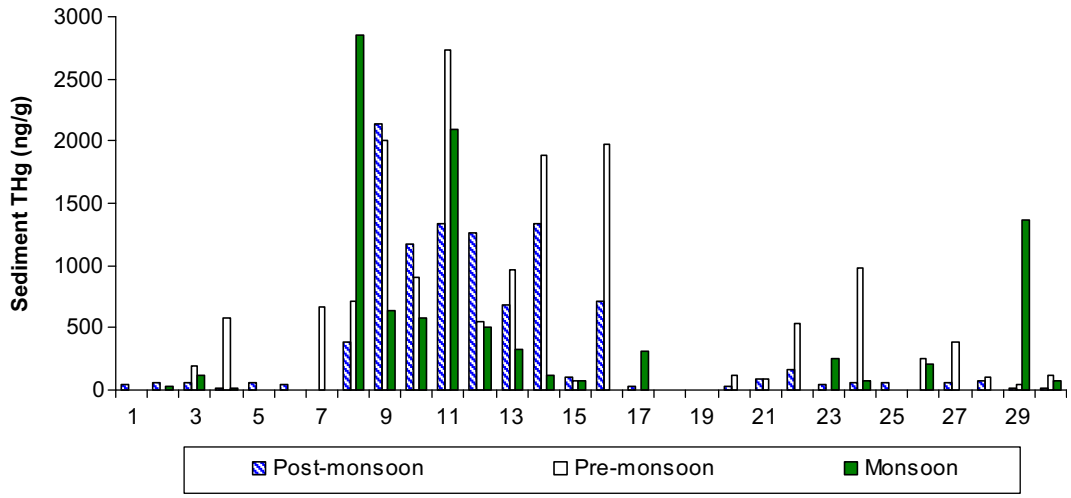

Fig. 4 THg in sediments during different seasons

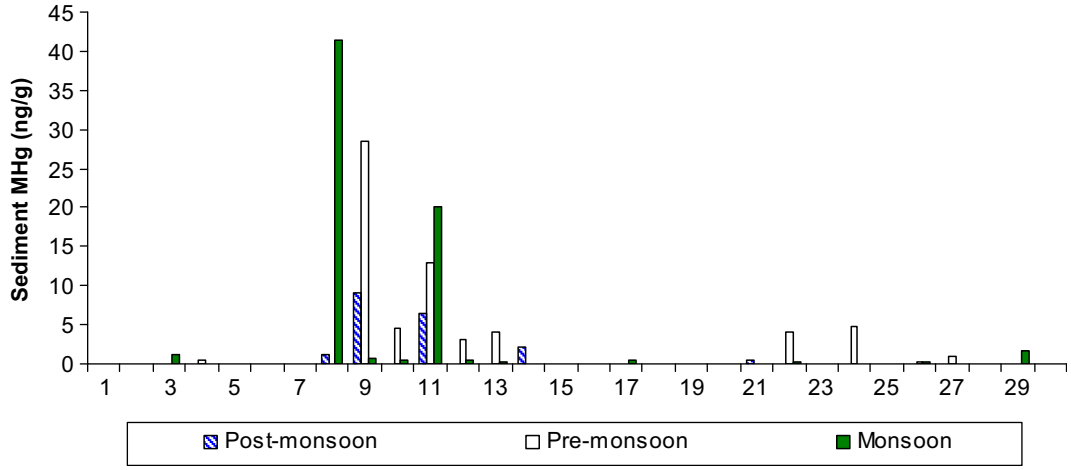

Fig. 5 MHg in sediments during different seasons

d.wt.) and MHg (1.743 mg/kg w.wt.) content compared to other omnivorous fishes. Neither form of mercury was detectable in the muscles of *Parambasis dayi*.

As per the standards, total mercury in fish meat for human consumption should be below 0.5 mg/kg dry wt., but in the current study, this value was exceeded for all fishes except *Glossogobius guiris* and *S. orientalis*. The concentration of THg was measured at five times higher than the safe consumption level in *Megalops cyprinoides* and four times higher in *G. minuta.*

THg and MHg content in omnivorous, benthic carnivorous, and carnivorous fishes were graphically plotted (Fig. 7) to understand the variation in accumulation of mercury in relation with their diet. Higher THg and MHg content was observed in carnivorous fishes, followed by omnivorous and benthic carnivorous fishes.

Table 4 Comparison of the mercury concentration in surficial sediments of Vembanadu Lake with different Lakes, estuaries and rivers in India

Study area	THg (μg/g) dry.wt.	MHg(ng/g) wet.wt.	Reference
Vembanadu backwater	0.395 ± 0.53	3.69 ± 13.67	Present study
Kodaikanal Lake	0.276–0.35	15.58–24.63[a]	Karunasagar et al. 2006
Kukkal lake	0.085–0.091	1.27–1.68[a]	Karunasagar et al. 2006
Berijam Lake	0.226–0.189	7.38–8.27[a]	Karunasagar et al. 2006
Cochin estuary	<0.001–1.88	<1–2.25	Present study
Cochin estuary	0.12–1.10	NA	Ouseph 1992
Beypore estuary, Kerala	0.05–2.0	NA	Nair 1994
Amba estuary	0.05–2.66	NA	Ram et al. 2003
Tambraparni estuary	43	NA	Roy et al. 2004
Industrially polluted area of Cochin estuary (confluence area of the Periyar river)	0.38–2.85	<1–202.2	Present study
Industrially polluted area (Lower stretch of the Periyar river)	5.5–11.5	NA	Ouseph 1992
Periyar river at upstream of industrial area	0.04–0.05	NA	Ouseph 1992
River Yamuna	0.81(0.01–2.59)	NA	Subramanian et al. 2003
River Ganga at Varanasi	0.067	NA	Sinha et al. 2007

NA not analysed

[a] Dry weight

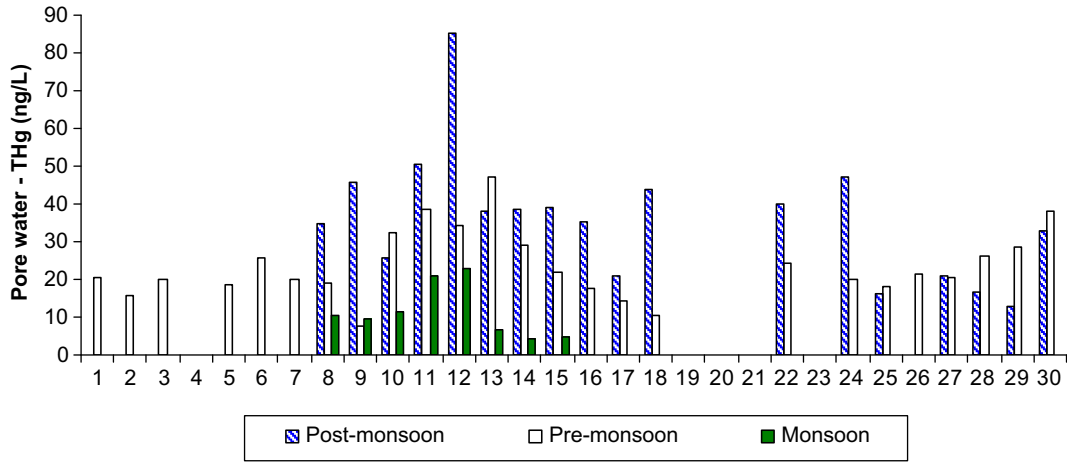

Fig. 6 Seasonal variation of total mercury in pore water samples

The percentage of MHg to THg was calculated on a dry weight basis (Fig. 8), showing the efficiency of carnivorous fishes in accumulating more toxic organic mercury through their diet. The MHg accounts for 16.68 to 90.25% of THg and varied between species (Fig. 8). A higher percentage of MHg was obtained for pelagic carnivorous fishes (*C. affinis* and *Stolephorus commersonnii*), carnivorous bottom feeder fishes (*A. arius*, *C. semifasciatus* and *Platycephalus indicus*), and benthopelagic carnivorous fishes (*Lutjanus johnii*). High concentrations of mercury in the omnivorous and carnivorous fishes obtained in the current study can be compared with earlier reports (Durrieu et al. 2005). The concentration of mercury in carnivorous fishes was

Table 5 Length and weight (mean) of the fish samples collected

Fish	Length (cm)	Weight (g)
Omnivore		
Parambasis dayi	6.4	8.12
Liza macrolepis	17.35 ± 0.21	55.75 ± 0.21
Cynoglossus semifasciatus	20.95 ± 0.21	55.65 ± 1.48
Etroplus suratensis	16.97 ± 3.04	159.33 ± 95.97
Gerres setifer	13.70 ± 3.32	51.64 ± 22.99
Oreochromis mossambica	14.40 ± 0.85	42.10 ± 13.72
Scatophagus argus	6.9	11.7
Benthic carnivore		
Gazza minuta	7.5	8.28
Synaptura orientalis	19.5	177.9
Arius arius	21.98 ± 3.81	102.87 ± 74.59
Platycephalus indicus	27	115.9
Megalops cyprinoides	33.8	375.2
Lutjanus johnii	21.10 ± 2.10	77.07 ± 19.94
Glossogobius giuris	18.5	68.5
Carnivore		
Caranx affinis	8.2	6.17
Stolephorus commersonnii	10.2	8.61
Thryssa mystax	8.68	13.6

Table 6 THg and MHg in muscle tissues of fishes (mg/kg)

Sl no.	Scientific name	THg (d.wt.)	MHg (w.wt.)
1	*Arius arius*	0.977	0.648
2	*Parambasis dayi*	nd	Nd
3	*Liza macrolepis*	1.860	0.629
4	*Gazza minuta*	1.97	0.46
5	*Cynoglossus semifasciatus*	2.850	1.743
6	*Synaptura orientalis*	0.3150	0.082
7	*Platycephalus indicus*	0.6540	0.384
8	*Etroplus suratensis*	0.956	0.481
9	*Megalops cyprinoides*	2.75	0.944
10	*Gerres setifer*	1.023	0.500
11	*Oreochromis mossambica*	0.708	0.154
12	*Glossogobius giuris*	0.1180	0.048
13	*Lutjanus johnii*	0.465	0.268
14	*Scatophagus argus*	1.41	0.19
15	*Caranx affinis*	3	2.26
16	*Stolephorus commersonnii*	1.89	1.28
17	*Thryssa mystax*	1.36	Nd

nd not detectable

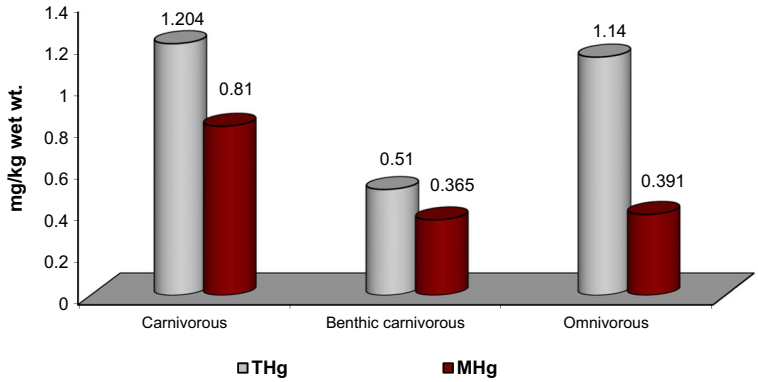

Fig. 7 Variation of THg and MHg content in fishes of different feeding habitat

found to be exceeding the permissible limits in several studies (Durrieu et al. 2005).

The THg accumulation in the muscle tissue of carnivorous fishes in other regions of the world showed a similar pattern. Benthic carnivores of the present study showed slightly lower values, however (Table 7). A higher concentration of THg was observed for omnivorous fishes when compared to others (Table 8). The total mean concentration for Vembanad backwater fish was higher than those reported in earlier works, except for those at a Northeast Brazilian estuary (Table 8). The proportion of MHg in THg for carnivorous (max. 90.25%) and benthic carnivorous (max. 80.22%) fishes were found to be less than those reported from around the world (Table 7). Kehrig et al. (2009) observed 7–49% of MHg in THg content for benthic carnivorous fishes of Guanabara Bay, while the present study showed comparatively higher values (31.46–80.22%). Omnivorous fishes of other regions of the world, however, showed a lower percentage of MHg content than in the present study (Table 8). For instance, Agarwal et al. (2007) observed only 14% of THg present as MHg in bottom feeder fishes of the Gomti river (Table 8).

The relationship between THg and MHg accumulation and fish size was statistically tested (ANOVA) for various fish species. Significant difference was observed for THg and MHg in all the tested fishes, which were *A. arius* ($F = 11.195$, $p < 0.05$), *Etroplussuratensis* ($F = 19.064$ and 19.178, $p < 0.05$), and *Gerres setifer* ($F = 19.38$ and 19.79, $p < 0.05$). These results are in agreement with the principle of MHg bioaccumulation in organisms of aquatic food webs, which says that Hg concentrations increase with increasing size (Kainz et al. 2006; Barbosa et al. 2003). The trophic level of the organism based on the feeding habit is also an important factor for this variation as *A. arius* and others are benthic carnivore and ominivore respectively (Azevedo-Silva et al. 2016). Both positive and negative correlation between body size (age) and mercury bioaccumulation have been reported. Dorea et al. 2006 found no significant relationship between fish size and mercury bioaccumulation from fishes of the RioNegro. Lima et al. (2000) reported a negative correlation between fish size and bioaccumulation of Hg in fishes collected from Santarem, Brazil. Furthermore, the correlation depends on the sampling size and availability of Hg at the

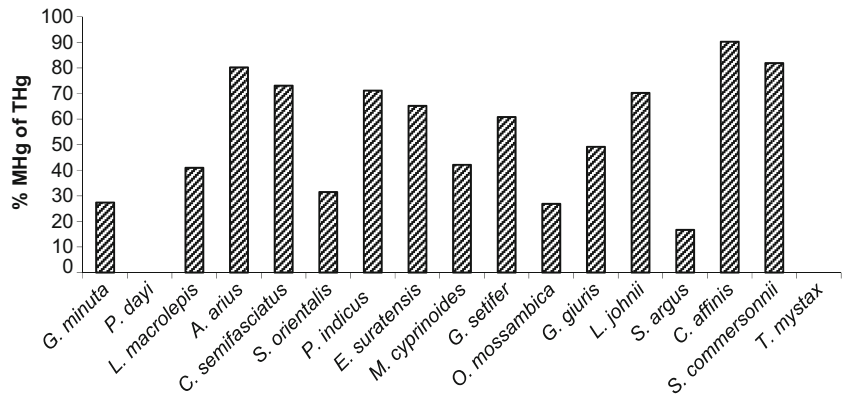

Fig. 8 Percentage of MHg of THg in the fishes

Table 7 Comparison of the THg and MHg concentrations in muscle tissues of carnivorous fishes from different regions of world with the present study

Fish	Area	THg (ppm) wet.wt.	MHg	Reference
Carnivorous	Vembanadu backwater	1.06 (1.34[a])	0.51 (27.35–90.25%)	Present study
Piscivorous	Madeira river	0.33–2.33	73–98%	Bourgoin et al. 2000
Piscivorous	Mobile-Alabama	0.53	0.44 (83%)	Bonzongo and Lyons 2004
Piscivorous	Brackish water lagoons of Suriname	0.03–0.04	NA	Mol et al. 2001
Piscivorous	Alvarado Lagoon	0.07–0.35	NA	Guentzel et al. 2007
Piscivorous	Rio Negro, Brazil	0.015–5.44	NA	Barbosa et al. 2003
Pelagic carnivorous	Guanabara Bay	3.4[b]	90–98%	Kehrig et al. 2009
Carnivorous	Guanabara Bay	0.063–0.556	0.065–0.561 (98%)	Kehrig et al. 2001
Carnivorous	Columbia-river	0.101–0.996 (0.371)	NA	Marrugo-Negrete et al. 2008
Benthic carnivorous	Vembanadu backwater	1.10 (1.38[a])	0.62 (31.46–80.22%)	Present study
Benthic carnivorous	Guanabara Bay	7.7[b]	7–49%	Kehrig et al. 2009
Benthic carnivorous	Guanabara Bay	2.9[b]	83–108%	Kehrig et al. 2009
Cat fish (benthic carnivorous)	Biscayne bay	1.58[a]	1.96[a]	Kannan et al. 1998
Cat fish (benthic carnivorous)	Florida bay	2.64[a]	1.68[a]	Kannan et al. 1998
Cat fish (benthic carnivorous)	Tampa Bay	2.09[a]	1.7[a]	Kannan et al. 1998

NA not analysed

[a] Dry wt basis

[b] nmol/g dry wt.

sampling station (Alonso et al. 2000). The species that showed positive correlation with size in the current study prefer strictly estuarine or fresh water habitats, where the probability of exposure to mercury from the lake is high.

Correlation was also carried out for mercury concentration in carnivorous, benthic carnivorous and omnivorous fishes with that in water and sediment (Table 9). THg and MHg in omnivorous fishes were positively correlated with sediment MHg and sediment THg. Other types of fishes did not show any positive correlation with THg and MHg in sediment, indicating that most of the mercury transport was through the food web. The feeding habit of omnivorous fishes (detritus feeding) exposes them to the sediment THg and MHg concentration. A significant positive correlation for THg and MHg content between carnivorous and benthic carnivorous fishes shows that their route of exposure is entirely different from omnivorous fishes. This was also confirmed by the negative correlation obtained for omnivorous fishes compared with carnivorous and benthic carnivorous fishes (Table 9). MHg of benthic carnivorous fishes and THg of omnivorous fishes are negatively correlated with water THg and MHg. The positive correlation ofMHg in omnivorous fishes with THg and MHg in water shows the presence of active diffusion of MHg and THg from sediments of the lake. The feeding habits are reported to be the most influencing factor in determining the bioaccumulation of mercury in fishes (Evans et al. 2000; Wiener et al. 2006). More studies are needed to confirm the role of mercury methylation rate in this part of the world.

THg content in fishes was negatively correlated with sediment THg (Table 10). A positive but not significant correlation was obtained between MHg in fishes and MHg in sediment (Table 10). The sediment showed a positive correlation ($p = 0.01$) between THg and MHg.

Bioaccumulation factor

Bioaccumulation factors were calculated using wet mass data as follows:

$$\mathrm{BAF} = \mathrm{MHg\ in\ the\ organism}\left(\mu g/g\right) / \mathrm{mean\ MHg\ in\ the\ water\ column}\left(\mu g/g\right).$$

A mean water column MHg concentration of 0.1388 ng/L was used in these calculations (Bowles et al. 2001). Bioaccumulation factors were calculated

Table 8 Comparison of the THg and MHg concentrations (ppm, wet.wt.) in muscle tissues of non-carnivorous and general fishes from different regions of world with the present study

Fish	Area	THg	MHg	Reference
Omnivorous	Vembanadu backwaters	1.12 (1.47[a])	0.47 (16.68–65.19%)	Present study
Omnivrous and mud feeding	Madeira river	0.02–0.19	NA	Bourgoin et al. 2000
Omnivorous	Rio Negro, Brazil	0.005–0.778	NA	Barbosa et al. 2003
Omnivorous	Guanabara Bay	4.5[b]	7–39%	Kehrig et al. 2009
Omnivorous and planktivorous	Guanabara Bay	5.3 [b]	2.7–23.8%	Kehrig et al. 2009
Herbivorous	Rio Negro, Brazil	0.002–0.186	NA	Barbosa et al. 2003
Non carnivorus	Columbia-river	0.035–0.436 (0.155)	NA	Marrugo-Negrete et al. 2008
Bottom feeder	Gomti, Lucknow, Uttar Pradesh	0.45	0.062 (14%)	Agarwal et al. 2007
Planktivorous	Gomti, Lucknow, Uttar Pradesh	0.18	0.180 (100%)	Agarwal et al. 2007
Fish	Vembanadu backwater	1.053 (1.36[a])	**NA**	**Present study**
Fish	Gomti	0.067–0.277	NA	Agarwal et al. 2007
Fish	Gulf of Mexico	0.933[a]	NA	Lewis and Chancy 2007
Fish	Bahia Blanca estuary	0.09–.12	NA	De Marco et al. 2006
Fish	Savanah river	0.009–1.2	NA	Paller et al. 2004
Fish	Northeast Brazilian estuary	501.0 ± 247.5[a] 125.3	NA	Costa et al. 2009

NA not analysed

[a] Dry wt basis

[b] nmol/g dry wt.

based on the ratio of MHg concentrations in fish muscle tissues to that measured in water. Bioaccumulation factor (BAF) values were presented as log values (Table 11). Among the fish, a maximum BAF value was obtained for pelagic carnivorous species *C. affinis* (7.21) and a minimum was obtained for benthopelagic carnivorous fish *G. giuris* (5.54). Both maximum and minimum log BAF values were observed for carnivorous fish. Among the omnivorous fish species, *C. semifasciatus* showed the maximum BAF (7.10). In the benthic carnivorous

Table 9 Correlation of mercury in carnivorous, benthic carnivorous and omnivorous fishes with that in water and sediment

	STHg	SMHg	WTHg	WMHg	CTHg	CMHg	BTHg	BMHg	OTHg	OMHg
STHg	1									
SMHg	0.720**	1								
WTHg	−0.23	0.12	1							
WMHg	−0.21	0.02	0.89**	1						
CTHg	−0.150	−0.084	0.286	0.180	1					
CMHg	−0.214	−0.554	0.098	0.031	0.824*	1				
BTHg	−0.666	−0.697	0.246	0.212	1.00**	0.929*	1			
BMHg	−0.593	−0.748	−0.102	−0.021	0.929*	1.00**	0.929*	1		
OTHg	0.808	0.213	−0.283	−0.731	−0.054	−0.140	−0.054	−0.140	1	
OMHg	−0.279	0.354	0.847	0.683	0.376	−0.212	0.376	−0.212	−0.055	1

CTHg THg in carnivorous, *CMHg* MHg in carnivorous, *BTHg* THg in benthic carnivorous, *BMHg* MHg in benthic carnivorous, *OTHg* THg in omnivorous, *OMHg* MHg in omnivorous

**$p = 0.01$; *$p = 0.05$, significant correlation

Table 10 Correlation of mercury in fishes with water and sediments

	Sediment		Water		Fish	
	STHg	SMHg	WTHg	WMHg	FTHg	FMHg
STHg	1.00					
SMHg	0.82**	1.00				
WTHg	−0.23	0.12	1.00			
WMHg	−0.21	0.02	0.89**	1.00		
FTHg	−0.09	0.13	0.15	0.33	1.00	
FMHg	0.19	0.36	−0.06	0.01	0.85**	1.00

FTHg THg in fish *FMHg* MHg in fish

**$p = 0.01$; *$p = 0.05$, significant correlation

group, the maximum BAF was observed for *A. arius* (6.67). Carnivorous (6.54) and omnivorous (6.51) fishes had almost same BAF values as the benthic carnivorous fishes (6.22). This further indicates that bioaccumulation was mainly through food. The BAF values obtained for carnivorous fishes were comparable with the result obtained by Bowles et al. (2001). He observed log BAF values of 5.84 to 7.25 for carnivorous fishes, while the values obtained in the present study were only slightly less (5.54–7.21). The omnivorous fishes also had similar values (6.045–7.099) as compared with the work done in Lake Murray (6.08–7.09) (Bowles et al. 2001).

Conclusions

The total and methyl mercury content in the water and sediments of Vembanad back water indicated that the mercury contamination is greatest the northern part of the system and is comparable to mercury contamination reported for other regions. The mercury content showed seasonal variation as the system is influenced by the fresh water input from six major rivers. The sediment mercury content is higher than that in water, and they do not positively correlate with each other. More than 16% of the samples have Hg content higher than the PEL. The high mercury content in the pore and bottom water indicates the formation of bioavailable forms of mercury in the sediments. The concentrations of MHg in sediments indicate varying physico-chemical conditions favouring the chemical transformation of mercury in the lake, which is further confirmed by the results obtained from the fish muscle analysis. The MHg content ranged from 16.68 to 90.25% of THg in fish muscle tissues. The statistical analysis showed that mercury accumulation is related to the feeding habits of the fish. High MHg concentrations were found in carnivorous fishes.

Frequent consumption of mercury contaminated muscle tissues can lead to higher accumulation in humans. More than 90% of the THg was in its most toxic organic form in fish, which increases concerns

Table 11 Bioaccumulation factors (BAFs) for fish species from Vembanad Lake

Fish type	Ecosystem component	MHg (μg/g)	Log BAF
	Water	13.88×10^{-8}	
Carnivorous	*Gazza minuta*	0.46	6.52
	Platycephalus indicus	0.384	6.44
	Glossogobius giuris	0.048	5.54
	Megalops cyprinoides	0.944	6.83
	Lutjanus johnii	0.268	6.29
	Caranx affinis	2.26	7.21
	Stolephorus commersonnii	1.28	6.97
Benthic carnivorous	*Synaptura orientalis*	0.082	5.77
	Arius arius	0.648	6.67
Omnivorous	*Cynoglossus semifasciatus*	1.743	7.10
	Liza macrolepis	0.629	6.66
	Etroplus suratensis	0.481	6.54
	Gerres setifer	0.500	6.56
	Oreochromis mossambica	0.154	6.05
	Scatophagus argus	0.19	6.14

about THg concentration in the common edible fishes observed in the present study.

Because fish is a staple food for humans, the accumulation of metals exceeding permissible limits is a serious health concern. The present study highlighted the mercury bioaccumulation potential in fish, which constitute an important group of animals in this brackish water ecosystem. Since the high concentration of mercury in the backwaters is attributed to anthropogenic origins, adequate strategies are to be adopted in order to control the content so that the possible health hazards to different life forms—including humans—can be prevented.

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