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Organochlorine Pesticides and Polychlorinated Biphenyls in Fish from Lake Awassa in the Ethiopian Rift Valley: Human Health Risks

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Abstracts Dietary intake of fish containing organic contaminants poses a potential threat to human health. In the present work, an assessment has been carried out to look at the human health risk associated with consumption of fish contaminated with organochlorine pesticides (OCPs) and polychlorinated biphenyles (PCBs) in certain fish species collected from Lake Hawassa, Ethiopia. The health risk assessment was made by comparing the concentrations of OCPs and PCBs in fish muscle tissues with reference doses given in the USEPA guidelines. Dichlorodiphenyltrichloroethanes (DDTs), endosulfans, PCBs and chloridanes were identified in fish species collected from Lake Hawassa. The most predominant pesticides were DDTs, with mean concentrations of ΣDDT ranging from 19 to 56 ngg^{-1} wet weights. The highest concentrations of DDTs were found in *Barbus intermedius,* representing the highest trophic level. PCBs, DDT and endosulfan concentrations found in *B. intermedius* exceeded the reference dose for children between the ages of 0–1 year (with hazard index of above 1.0). Therefore, consumption of fish from a high trophic level (e.g. *B. intermedius*) from Lake Hawassa may pose a special health risk to children.

Keywords Fish consumption · Health risk · Lake Hawassa · Trophic position · DDT · PCB

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Humans have benefited from the use of organochlorine pesticides (OCPs) and polychlorinated biphenyles (PCBs) as these chemicals are known for their contribution in increasing agricultural yield and controlling vectors, as well as having industrial benefits (Brinkman 1989). Unfortunately, exposure to these chemicals has become a matter of environmental and health concern, particularly in countries where environmental safety regulations are not strictly implemented, and users' knowledge of safe handling procedures is inadequate (Jones and de Voogt 1999; WHO 1989). Accordingly, several studies, regular surveys, and monitoring programmes of such chemical residues in fish coupled with risk assessments have been carried out in a number of countries (Binelli and Provini 2003; Fianko et al. 2011; Polder et al. 2010; Sun et al. 2006; Xuemei et al. 2008). Although Ethiopians are traditionally meat eaters, food habits have been shifting in favour of fish in communities where there is regular and sufficient supply; and in such communities, annual fish consumption can exceed the normal rate (FAO 2011). Consequently, consumption of contaminated fish may be an important route of human exposure to OCPs and PCBs (Sun et al. 2006) in these communities. Therefore, there is a pressing need for monitoring and controlling these chemicals in fish used for consumption. Due to the intensive agricultural and deforestation activities in the catchments of the Ethiopian Rift Valley Lakes (Zinabu 2002; Zinabu and Elias 1989), there

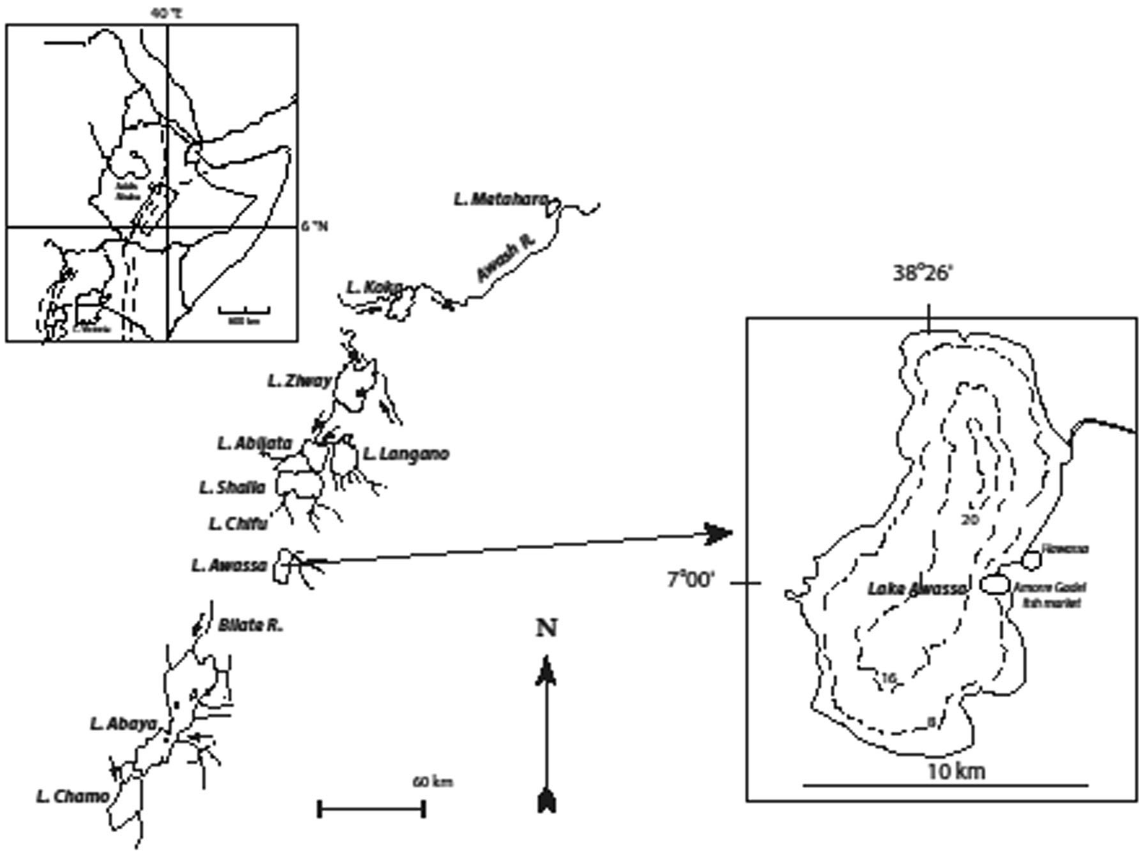

Fig. 1 Map of the study area

is a risk of chemical pollution in these lakes. Run-off and erosion from the surrounding catchment could possibly transport chemicals sorbed to soil particles to the lake, where chemicals may be desorbed and become bioavailable. Hence, the chemicals become a public health concern due to the subsequent uptake and bioaccumulation in fish. However, the level and the extent of contamination of OCPs and PCBs in fish species at various trophic levels in these lakes have not been studied. The objective of this study was therefore, to assess possible health risks of OCPs and selected PCBs in relation to consumption of contaminated fish from different trophic positions in Lake Hawassa – one of the Ethiopian Rift Valley Lakes.

Study Area

The study area, Lake Hawassa (7°01′52.05″N and 38°25′19.41″E) is located in the south-eastern part of Ethiopia, at 1,680 m above sea level (Fig. 1). The lake has an average surface area of 90 km^2 and a maximum depth of 22 m (Vallet-Coulomb et al. 2001). The fish species of the lake sampled for this study include the Nile tilapia (*Oreochromis niloticus* Linnaeus, 1758), the African sharptooth catfish (*Clarias gariepinus* Burchell, 1822) and African big barb (*Barbus intermedius* Rüppell, 1835).

Materials and Methods

Fish sampling was carried out between February and April 2008, partly by purchasing fish from the local fishermen upon landing, and partly by own gillnetting, using Nordic gill nets with mesh sizes from 5 to 45 mm (bar mesh) (Appelberg et al. 1995). A total of 64 fish samples (39 females and 25 males) from the three fish species (*O. niloticus, C. gariepinus,* and *B. intermedius*) were collected.

Stomach content analysis was conducted using 143 stomach samples at Hawassa University, Hawassa, Ethiopia. Large items were identified visually, and smaller items were identified under a dissecting microscope. For microscopic food item like algae, the stomach contents from each fish were first diluted with water to a known volume, mixed thoroughly, a drop of it was placed on a microscopic slide and identified, and finally the mean volume percentage of food items was calculated. For large food items: volume of the food items was determined by the amount of water each prey category displaced in volumetric glassware, and mean volume percentage of food items was calculated. The relative importance and contribution of each food item in the diet of each fish species was determined using the frequency of occurrence method and the relative (%) composition by volume (volumetric analysis) (Hyslop 1980).

Stable N and C isotope analyses were carried out at the Environmental Chemistry Section, Department of Plant and Environmental Sciences, UMB, using Isotope Ratio-Mass Spectrometry IRMS (IRMS). Homogenized and freeze-dried muscle samples were subjected to combustion in a Flash Elemental Analyzer (EA) and stable isotopes of nitrogen (^{15}N and ^{14}N) and carbon (^{13}C and ^{12}C) were analyzed as described in Desta et al. (2007). The isotopic ratios ($^{15}N/^{14}N$, $^{13}C/^{12}C$) were expressed in delta-values as follows:

$$\delta^{15}\text{N and } \delta^{13}\text{C}(‰) = [(\text{R}_{\text{Sample}}/\text{R}_{\text{Standard}})] \times 1{,}000$$

where, R = $^{15}N/^{14}N$ for $\delta^{15}N$ or R = $^{13}C/^{12}C$ for $\delta^{13}C$.

The 21 OCPs and selected PCBs (PCB-28, PCB-52, PCB-101, PCB-118, PCB-138, PCB-153, and PCB-180) were analysed at the laboratory of the Norwegian Institute for Agricultural and Environmental Research (Bioforsk), Chemistry and Pesticide Section, Norway. The fish tissue samples were extracted with acetonitrile as described in (Norli et al. 2011). The results from the analysis of the standard were all within the certified range. Estimation of the limit of quantification (LOQ) and limit of detection (LOD) were made by spiking *O. niloticus* with DDTs. The samples were analysed using gas chromatography coupled with mass spectrometry (GC–MS). A rough estimate of LOQ was performed on acetonitrile extracts of tilapia and salmon spiked at 5 ng/g using the signal/noise (S/N) function in ChemStation. Analysis of a standard reference material shows acceptable results for most of the targeted OCPs and PCBs and the LOQ values are in the range 1–3 ng/g.

The reference dose (RfD), drawn from the USEPA potential health risk assessment guidelines was used to assess the health risk posed by OCPs and PCBs exposure (USEPA 1996). The following assumptions were made:

1. Hypothetical body weight of 10 kg for children in age-class 0–1 years, 30 kg for children in age-class 1–11 years, and 70 kg for adults (USEPA 1996).
2. Maximum absorption rate of 100 % and a bioavailability rate of 100 % (USEPA 1996).
3. Fish consumption rate in Ethiopia, estimated to be 0.027 kg day^{-1} (FAO 2011).

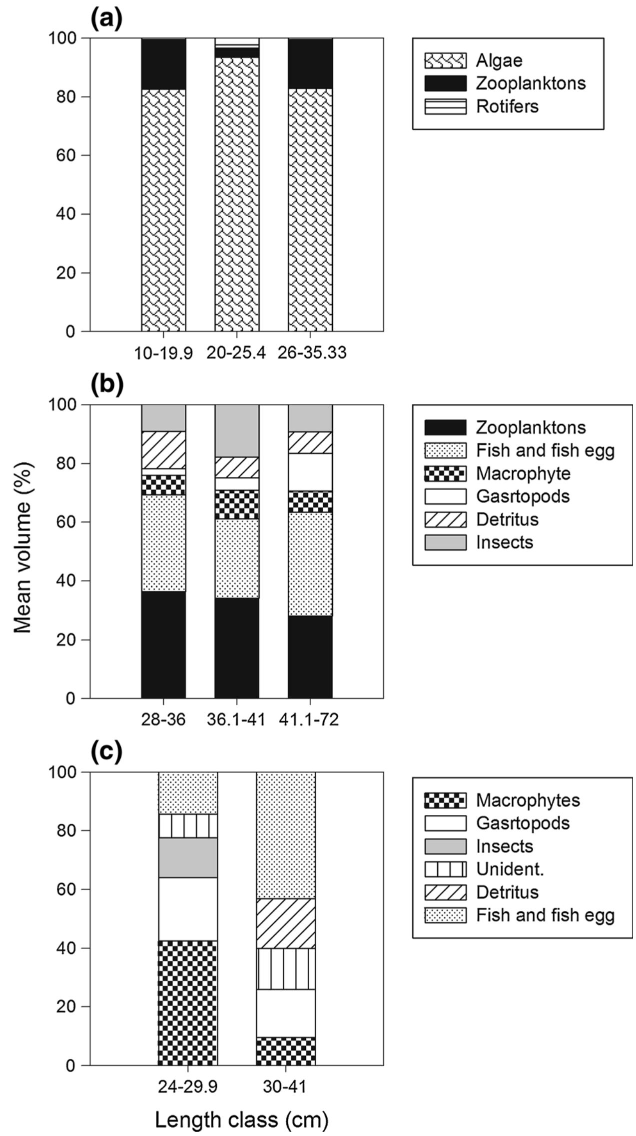

Fig. 2 Relative distribution (mean volume, %) of food items in fish stomach from different length-classes of **a** *O. niloticus,* **b** *C. gariepinus,* and **c** *B. intermedius,* sampled in Lake Hawassa

Consumption of contaminants in food was calculated based on its concentration in the fish, and on an estimate of the fish consumption rates. The lifetime exposure dose (LED) (mg kg^{-1} day^{-1}) was obtained, and the hazard indices (HI) for each age class were estimated.

$$\text{LED} = \frac{\text{Residue concentrations in fish tissue sample (mg/kg)} \times \text{ fish consumption rate (kg/day)}}{\text{body weight (kg)}}$$

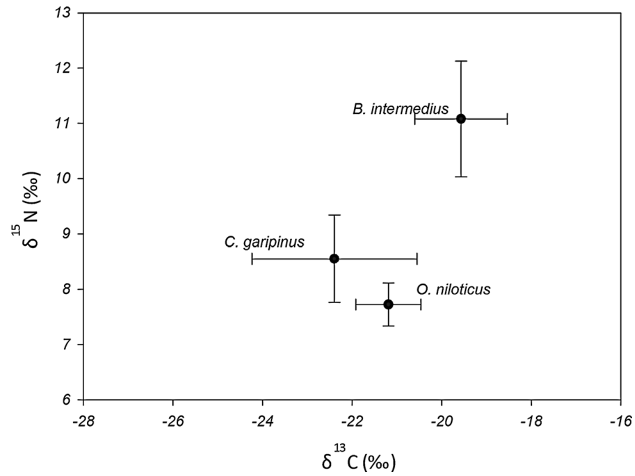

Fig. 3 Relative trophic position of the three fish species, *O. niloticus, C. gariepinus, and B. intermedius*, sampled in 2008 from Lake Hawassa, values are based on the mean of the stable isotope ratios of nitrogen (δ^{15}N, ‰) and carbon (δ^{13}C, ‰). *Horizontal* and *vertical lines* indicate the SDs of the mean values

$$HI = \frac{\text{Estimated dose}}{\text{Reference dose}}$$

Results from this study were compared with the oral RfD values in mg kg bw^{-1} day^{-1}, obtained from USEPA's Integrated Risk Information System.

Results and Discussion

Paired gut content and stomach content analyses showed that there are at least 2 trophic levels in the studied fish community of the Ethiopian Rift Valley Lake Hawassa (Figs. 2, 3). A difference of 5.5 between the maximum and the minimum signature of stable isotope of nitrogen also confirmed this. According to the present study, *B. intermedius* clearly occupied the highest trophic level in Lake Hawassa as also confirmed by the signature of stable isotope of nitrogen. This species is omnivorous in Lake Hawassa, with large individuals having fish in their diet. Previous studies, in Lake Hawassa (Desta et al. 2006), in Lake Victoria, Uganda (Corbet 1961), and in Lake Kinneret, Israel (Spataru et al. 1987) have also shown that the larger individuals of *B. intermedius* have included fish in their diet. As reported by Bootsma et al. (1996) and Hecky and Hesslein (1995), aquatic organisms such as fish having a benthic-based food source tend to have a more positive δ^{13}C signal than organisms (e.g. fish) utilizing a pelagic-based food source. The fish species (from Lake Hawassa) included in this study had overlapping habitat use. Moreover, the signatures of δ^{13}C in general indicate that they had a wide range of carbon sources that extend from benthic to pelagic-based food sources. However, *B. intermedius* in Lake Hawassa exhibited relatively more enriched δ^{13}C than the other fish species, suggesting that *B. intermedius* obtained a large proportion of its prey from benthic sources.

The predominant pesticide in fish from Lake Hawassa, was the DDT; with concentrations much higher (a factor of 10) than the concentration of chlordane and PCBs, and at least two times higher than endosulfan in all fish species (Table 1). Reports from other African lakes, for instance in Lake Malawi (Kidd et al. 2000) and in previous studies in the Ethiopian Rift Valley Lakes Koka and Ziway (Deribe et al. 2011; 2013) also indicated much higher levels of DDT in aquatic organisms compared to other POPs. Such high concentrations of DDTs may be attributed to the intensive and continuous use of DDT in vector control in Africa.

The positive relationship between the log transformed concentration of ΣDDT and δ^{15}N in the fish species from Lake Hawassa indicates that DDT is biomagnified in the

Table 1 Mean values and SDs of concentrations of ΣOCPs and ΣPCB in ngg^{-1} ww, with minimum and maximum values, number of samples (N) and frequency of detection (percent), in the three fish species *O. niloticus, C. gariepinus, and B. intermedius*, from Lake Hawassa, all sampled in 2008

Variable	Spp	N	Percent	Mean	StDev	Minimum	Maximum
ΣDDTs	*O. niloticus*	20	100	19.16	28.91	1.65	102.5
	C. gariepinus	21	100	28.52	26.82	5.41	100.11
	B. intermedius	23	100	56	86.30	13	409.6
ΣPCBs	*O. niloticus*	20	10	2.36	2.80	0.38	4.33
	C. gariepinus	21	42.9	1.04	1.15	0.35	3.95
	B. intermedius	23	56.5	2.75	7.44	0.35	27.47
Σchlordane	*O. niloticus*	20	5	0.85	–	0.85	0.85
	C. gariepinus	21	4.8	1.21	–	1.21	1.21
	B. intermedius	23	8.7	2.15	1.99	0.75	3.56
Σendosulfan	*O. niloticus*	20	0	–	–	–	–
	C. gariepinus	21	9.5	3.67	0.40	3.39	3.96
	B. intermedius	23	13.0	25.90	17.4	7.8	42.5

Table 2 Estimated dose values and hazard indices of ΣOCP and ΣPCB exposures in fish at different trophic levels (*O. niloticus*, *C. gariepinus* and *B. intermedius)* sampled in L. Hawassa

OCs and PCBs	Fish species	RfD (μg kg^{-1} day^{-1}) (USEPA 1996)	Estimated dose (μg kg^{-1} day^{-1})			Hazard index		
			0–1 years	1–11 years	Adult	0–1 year	1–11 year	Adult
ΣDDT	*O. niloticus*	0.5	0.281	0.094	0.040	0.561	0.187	0.080
	C. gariepinus	0.5	0.274	0.091	0.039	0.548	0.183	0.078
	B. intermedius	0.5	1.121	0.374	0.160	**2.243**	0.748	0.320
ΣPCB	*O. niloticus*	0.02	0.012	0.004	0.002	0.593	0.198	0.085
	C. gariepinus	0.02	0.011	0.004	0.002	0.541	0.180	0.077
	B. intermedius	0.02	0.075	0.025	0.011	**3.761**	**1.254**	0.537
ΣChloridane	*O. niloticus*	0.06	0.002	0.001	0.000	0.039	0.013	0.006
	C. gariepinus	0.06	0.003	0.001	0.000	0.055	0.018	0.008
	B. intermedius	0.06	0.010	0.003	0.001	0.162	0.054	0.023
ΣEndosulfan	*O. niloticus*	0.05	–	–	–	–	–	–
	C. gariepinus	0.05	0.011	0.004	0.002	0.217	0.072	0.031
	B. intermedius	0.05	0.116	0.039	0.017	**2.327**	0.776	0.332

Hypothetical body weight of 10 kg for children in age-class 0–1 years, 30 kg for children in age-class 1–11 years, and 70 kg for adults

Numbers in bold indicate hazard indices greater than one

food web ($p < 0.05$). However, the biomagnification rate of DDT in this lake is lower than what has been reported from other localities, such as the Southern Beaufort-Chukchi Seas, in the Arctic (Hoekstra et al. 2003), sub-arctic lakes in Yukon Territory (Kidd et al. 1998), marine food web from south-eastern Norway (Russ et al. 1999), Mekong Delta, South Vietnam (Ikemoto et al. 2008), as well as Lake Malawi, East Africa (Kidd et al. 2000). The highest concentration of DDTs were detected in the piscivorous *B. intermedius*, this is probably due to the biomagnification of DDT along the food chain. This finding is similar to results obtained by Zhou et al. (2007) in piscivorous fish species from Qiantang River, East China, and in the piscivorous Arctic charr (*Salvelinus alpines*) in Lake Arresjøen, Svalbard (Rognerud et al. 2002). The other persistent contaminants, endosulfan, chlordane and PCB, did not seem to biomagnify in Lake Hawassa. Only a few specimens contained detectable concentrations of these contaminants, and due to the low concentrations, a possible biomagnification is difficult to detect. Variation in the accumulation of POPs in different fish species is attributed to trophic position, age, and fat content, and therefore; consumption of old fish, with more fat content, and from high trophic levels, may expose consumers to possible health hazard. The highest estimated dose is found in *B. intermedius* which is found at the highest trophic level of the studied species. The fact that the mercury concentrations in most individuals of *B. intermedius* in Lake Hawassa were found to exceed the WHO and EU trade level (Desta et al. 2006) points to the high risk of multiple stressors, especially for consumers who use this species from Lake Hawassa as part of their food intake. The same may apply for Ethiopians in general or other populations on the African continent, who use this species for food.

Comparison between estimated dose and the reference dose shows that children are the most vulnerable population sub-group (Table 2). Feeding *B. intermedius* to children would consequently expose them to systemic toxicity. Since the estimated dose of ΣPCB for adults also would exceed the reference dose (with hazard index above 1), consumption of *B. intermedius* also poses a health risk for an adult. However, consumption of the other fish species (*O. niloticus* and *C. garipienus*) does not pose a direct hazard to human health, based on the current findings. In the Hawassa area, the species, *O. niloticus* and *C. garipienus* are more preferred for consumption than *B. intermedius,* probably because of the high infection rate with tape worms encountered in *B. intermedius* which makes this fish species less preferable to be eaten by the local people (personal communication). However, this information needs further verification. Moreover, *B. intermedius* also contains much more intramuscular bones than the other species, making it less attractive for consumption (e.g., the restaurants in Hawassa town – Desta et al. 2007). However, the fish fillet is still prepared and served as soup by the local fishermen families. For those who eat fish more or less daily and much more than the estimated mean Ethiopian daily consumption (0.027 kg day^{-1}), the OCP and PCB intake will thus be considerably higher (FAO 2011). Therefore, children from the fishermen families may be the most exposed and vulnerable group among the local people.

Conclusion

People that live around Lake Hawassa, and regularly consume fish from the lake, are exposed to DDTs and PCBs when eating large fish positioned at higher trophic level. The results of this study suggest that consumption of *B. intermedius* from Lake Hawassa may expose the consumers for systemic toxicity, especially in young children who are considered to be the most vulnerable population sub-group. Although the estimated dose of DDT in *O. niloticus* and *C. gariepinus* is below the current reference dose, the presence of DDT residue in all samples indicates that any consumption of fish from the lake in the future will require continuous dietary exposure assessment. Moreover, interactions and combined effects among contaminants should be taken into account in evaluation of the health risk posed by OCPs and PCBs exposure more precisely.

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