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RESEARCH PAPER

Presence and Risk Assessment of Legacy Organochlorine Pesticides Residue in Water and Sediments from Lake Kariba: Implications for Multiple Uses of the Lake

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Abstract

There has been a consistent rise in the utilization of resources from Lake Kariba, Zambia over the years. Monitoring the health of the aquatic ecosystem is crucial to assess the potential impacts on public health. This study presents the first assessment of the levels and risks associated with organochlorine pesticide (OCP) residues in the surface water and sediment of Lake Kariba in Zambia. Four sediments and twenty-two surface water samples were collected from twelve sites downstream in April 2024. Modified QuEChERS and solid phase extraction (SPE) were used to extract the sediment and water samples, respectively, and GC-μECD detector was used for analysis. A total of fourteen OCPs were detected in the water and twelve in sediment samples with total concentrations ranging from 0.022 to 3.119 ng mL^{-1} and 30.22–112.7 ng g^{-1} respectively. Dichlorodiphenyltrichloroethane (DDT) metabolites were most common in sediment samples, whereas hexachlorocyclohexane (HCH) isomers were most common in water samples. According to the risk assessment study, children's life average daily doses (LADD) of endrin and δ-HCH were marginally higher than the USEPA upper limit of 10^{-6}, suggesting that these OCPs may pose a lifetime risk of cancer. In conclusion, the results of this study underscore the importance of consistently monitoring OCP levels in the Lake to detect trends over time. Additionally, they offer insights into the possible health effects of the identified compounds, assisting policymakers in implementing OCP reduction strategies in line with the objectives of the Stockholm Convention.

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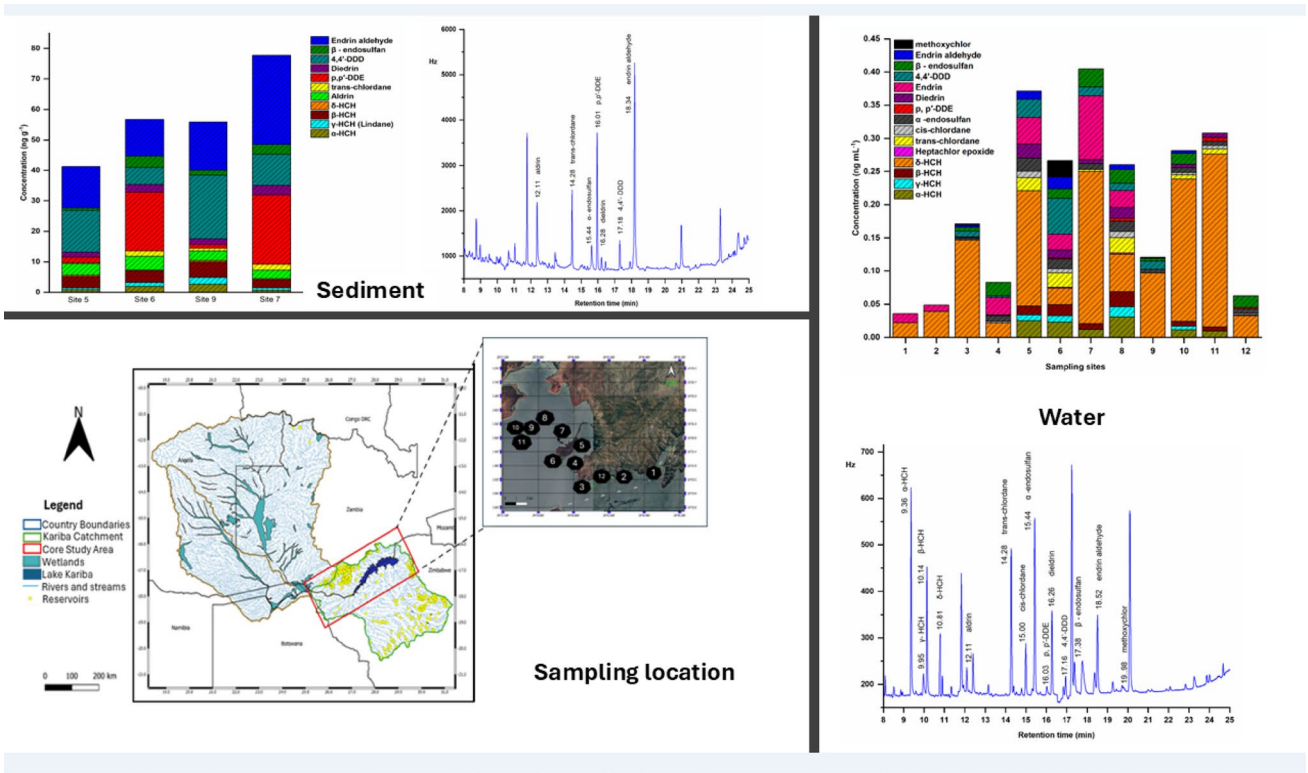
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Graphical Abstract



Keywords Organochlorine pesticides · QuEChERS, Lake Kariba, Risk assessment · Average daily dose · Life average daily dose

Introduction

The extensive application of organochlorine pesticides (OCPs) in the 1950s left lasting impacts on the environment, a topic of global concern. Known as persistent organic pollutants (POPs), OCPs gained popularity for their low cost, effectiveness, and durability, finding extensive applications as pesticides and insecticides (Kai et al. 2025). However, exposure to this group of compounds has been linked to reproductive abnormalities, behavioural disorders and cancer (Asefa et al. 2024). This is because they have capacity to interfere with the actions of specific hormones, enzymes, growth factors, and neurotransmitters (Ahmad et al. 2024; de Souza Pomacena et al. 2025) .Due to the aforementioned ecological and human health risks of OCPs, most developed countries banned their production and usage during the 1970s and 1980s (Hu et al. 2024). Although many developing nations did not yet have these prohibitions, regulation and restriction was enforced with the establishment of the Stockholm Convention (SC) in 2001 (UNEP 2023; Akinrinade et al. 2024).Under this convention, member states agreed to address the dangers associated with POPs with an aim of reducing, preventing, and ultimately eradicating them from the environment (Nekouei Esfahani et al. 2012; UNEP 2023).

Zambia like other member countries, is a signatory to the SC, which came into effect in 2006. To meet the provisions of the convention, Zambia developed its National Implementation Plan (NIP) in 2007 for the first twelve POPs, including OCPs. According to this NIP, Zambia does not produce any POP pesticides domestically. Furthermore, a nationwide inventory survey conducted in 2004 indicated that the country was utilizing approximately 145 metric tons of POPs pesticides each year prior to the mid-1980s (UNEP 2007). It was further noted with concern that chlordane and DDT are still in use. DDT is utilized in public health programs like Indoor Residue Spray (IRS) to reduce malaria transmitting vectors, while chlordane is used for termite control in the construction industry (Bouwman 2004; Chanda et al. 2011; Shanungu 2018). Infact, according to (Gilbreath and Steinemann 2000), Zambia imports about 1,400 tons of various agricultural pesticides annually, with a small fraction of these imports containing DDT. Most of these pesticides are utilized in agriculture primarily for pest

management (Asefa et al. 2024), thereby enhancing the overall yield of agricultural products. Agricultural activities remain the sole source of income for many families in Zambia, amounting to more than 75% of the population (Gilbreath and Steinemann 2000). However, most farmers lack knowledge about the dangers of pesticides, which results in careless use that can raise risk and prolong the chemicals' presence in the environment.

Owing to the mode of application and chemical nature, pesticides are among the various pollutants encountered in aquatic environments (Ogola et al. 2024). The flow of these contaminants into aquatic environments either from runoff or effluent discharges is and has been a cause for significant concern in numerous nations for years (Nasrabadi et al. 2011). Once released, aquatic systems like seas, dams or lakes frequently serve as temporary or permanent repositories for any possible pollutants (PS 2015).The octanol-water partition coefficient (log Kow) and hydrophobicity of organic substances affect their distribution in lipid-rich environments like sediments and aquatic environments like surface water (Gobas and Zhang 2024).The majority of OCPs are drawn out of the water column, adsorbed onto particulate matter, and accumulated in sediments due to their high log Kow values. This makes the sediment component of the aquatic system serve as a secondary source of pollution (Lee et al. 2001; Ogola et al. 2024). Therefore, sediments can serve as indicators of changes occurring in the water column and reflect the effects of human activities on both air and water and play a critical role in environmental monitoring (Mohammadi et al. 2024).

Lake Kariba is located on the southern boundary of Zambia and Zimbabwe. It has been recognised as one of the biggest manmade Lakes that has experienced significant industrial, agricultural, and economic growth over the years. Having been created primarily for hydropower generation in the late 1950's, there has been a steady increase in the usage of Lake Kariba resources. Large aquaculture firms currently operate in the lake, in addition to many agricultural operations that take place in its vicinity. In addition to endangering the freshwater ecosystem and disrupting biodiversity, this exposes the lake to foreign substances including pesticides (Mwakalapa et al. 2018; Simukoko et al. 2023).Despite the possibility of organic contaminants being directly released into the lake, the amount and makeup of OCPs in the lake's surface water have not yet been determined. Generally, previous research on Lake Kariba's environmental health focused mostly on fish, which reflect the biota of the ecosystem component. Citeable studies in this line include one conducted by (Berg et al. 1992) in 1992 and more recently is a study conducted by (Simukoko et al. 2023) in Lake Kariba, Zambia. Berg et al. (1992) reported presence of 1900 ng g^{-1} and 5000 ng g^{-1} of DDT in Redbreast Tilapia *(Tilapia rendalli)* and predatory tigerfish *(Hydrocynus forskahlii)* respectively. On the other hand, results from Simukoko et al. (2023) indicated that wild tilapia had higher levels of DDTs ranging between 93 ng g^{-1} and 81 ng g^{-1} as compared to farmed tilapia. Based on a comparison of these two available studies, it can be generally concluded that DDT levels in the biota component of the lake's ecosystem have largely decreased during the past three decades. Regarding the lake's sediment however, no additional information on the distribution of OCPs in this ecosystem component is currently known, except for a limited study by Zaranyika et al. (1994). In this study conducted more than three decades ago, the researchers reported elevated levels of OCP residues in sediment samples of selected river bays in Lake Kariba, Zimbabwe. The mean values reported were as follows: DDT metabolites (5.62–117.7 ng g^{-1}), aldrin (0.017- 4.10 ng g^{-1}), endosulfan's (1.91–16.5 ng g^{-1}) and dieldrin (2.27–37.1 ng g^{-1}). Other than these cited reports, the current investigation comes in handy to evaluate the current levels of OCPs in sediments and surface water in Lake Kariba, Zambia.

Based on the available data within the study area, it is evident that there has not been proper sustained environmental monitoring to ascertain trends on the emerging pollutants or on the legacy pesticides. The fact that OCPs were detected recently in fish from the same lake while these compounds were banned many years ago, suggests that they are still circulating within the ecosystem of the lake and its catchment. Currently, the lake is home to various commercial fish farming activities and provides water for daily usage to neighbouring communities. Because the water and fish raised in the lake are exposed to these chemicals, it poses a public health risk as both the local community and other people across the country may be exposed through oral consumption. This study therefore aimed to evaluate levels of OCPs in surface water and sediment of Lake Kariba, Zambia, as well as their health risk assessment. Notably, as far as we know, this is the first assessment of OCPs in surface water and sediments of Lake Kariba from Zambia's perspective. Hence, this monitoring data will form a basis upon which intervention decisions are made to safeguard public health.

Materials and Methods

Chemicals and Reagents

Organochlorine pesticide mixture standard of concentration 200 µg mL^{-1} was obtained from Merk (Johannesburg, South Africa). The standard mixture consisted of twenty OCPs with an inclusion of 2,4,5,6-tetrachloro-*m*-xylene and decachlorobiphenyl as surrogates. A stock solution at 1.0 µg

mL^{-1} was prepared in *n*-hexane (HPLC grade) and stored in a refrigerator (4 °C). The stock solution was suitably diluted to provide standard solutions with all the analytes under investigation at different concentrations ranging from 0.01 to 300 ng mL^{-1}. During the development and validation of the analytical method, the prepared working standards were employed to plot calibration curves and for spiking in recovery studies. Methanol, *n*-hexane (HPLC grade), and nitric acid were obtained from Merck (Johannesburg, South Africa). In both blank extractions and spiking experiments, water purified by milli-Q SP treatment was utilized as reagent water. WHATMAN grade 1 filter papers (11 µm) and Affinisep prefilter glassfiber (1 µm) were obtained from Anatech (Johannesburg, South Africa). Sep-Pak cartridges for SPE packed with 1000 mg of C_{18} sorbent were supplied by Microsep (Johannesburg, South Africa).

Table 1 Sampling locations co-ordinates and description

Site	Sample	Latitude	Longitude	Description of Sites/activity
1	Water	S 16° 32.127’	E 028° 44.927’	Near the Lake Kariba dam wall
2	Water	S 16° 32.587’	E 028° 43.419	Near Manchichi, lodging and recreation
3	Water	S 16° 32.838’	E 028° 41.562’	Near fresh view, hotel for accommodation
4	Water	S 16° 31.916’	E 028° 40.888	Near the Lake fishing camps
5	Water/sediment	S 16° 31.313’	E 028° 40.924’	Near small scale fish cages, fish farming activities
6	Water/Sediment	S 16° 31.734’	E 028° 39.956’	Site near Banana Island-South, beach & fishing
7	Water/sediment	S 16° 30.674’	E 028° 39.953’	Site near Banana Island-West, beach & fishing
8	Water	S 16° 30.259’	E 028° 39.256’	Near Yalelo fish cages, commercial fish farming,
9	Water/sediment	S 16° 32.511’	E 028° 42.372’	Near government/Marine offices
10	Water	S 16° 32.511’	E 028° 42.372’	Near Zamfresh fish cages, commercial fish farming
11	Water	S 16° 32.2818’	E 28° 42.3018’	Near Kariba Inn Harbour, Kapenta boat’s location
12	Water	S 16° 32.2806’	E 28° 42.3516’	Near Kariba Inn Harbour, cruise boat’s location

Study Area

Lake Kariba is a man-made reservoir which sits on Zambia’s southern border with Zimbabwe (16° 28′ to 18° 04′S; 26° 40′ to 29° 03′ E) (Table 1). The Lake’s surface area is 5580 km^2, its maximum width is 32 km, and its maximum reservoir length is 280 km (Tumbare 2008). The Lake Kariba catchment is surrounded by five republican states which include Zambia, Zimbabwe, Angola, Namibia, and Botswana (Fig. 1). The lake’s drainage flows eastward, with multiple rivers entering it from both the Zimbabwean and Zambian sides. On the Zambian side of the Lake, the three localities (Siavonga, Sinazongwe and Gwembe) are home to major human endeavours such as commercial fishing, coal mining, and agricultural pursuits like crop cultivation and cow breeding. The majority of fish farming in the Siavonga district involves raising tilapia in cages on the lake. Twelve different sampling sites were identified for collecting the samples as shown in (Fig. 1).

Sample Collection and Processing

The surface water and sediment samples were collected in April 2024 from selected sites of Lake Kariba, Zambia (Fig. 1). The water samples were collected from all twelve locations, while sediment samples were collected from only four of the selected locations. This was due to the lake’s extensive rocky terrain which does not allow for the collection of sediments in all the identified sampling sites (Tumbare 2008). For water samples, one-liter polypropylene bottles were cleaned with ultrapure water in the laboratory and later on-site with the water sample from the lake. The water was collected by filling the container to the brim, ensuring that there was no headspace. Water was sampled from between 0 and 40 cm below the lake’s surface. Prior to being extracted, the collected surface water was carried to the laboratory in coolers filled with ice and kept in the dark at 0–4 °C (Golfinopoulos et al. 2003; Adeyemi et al. 2011). A stainless-steel grab sampler was used to collect sediment samples from four locations downstream. Sediment samples were well mixed and stored frozen (-20 °C) in cleaned glass bottles wrapped in aluminum foil and taken to the laboratory. Before extraction, the sediments were freeze dried and sieved through a 212 µm steel mechanical shaker.

Extraction and Clean-up of Water Samples

Prior to the extraction, 10% methanol modifier was added to 1 L water samples, followed by shaking for 10 s, and left to stand for 30 min. The water samples were then filtered using WHATMAN grade 1 filter paper (11 µm) to remove suspended large particles and later with Affinisep prefilter

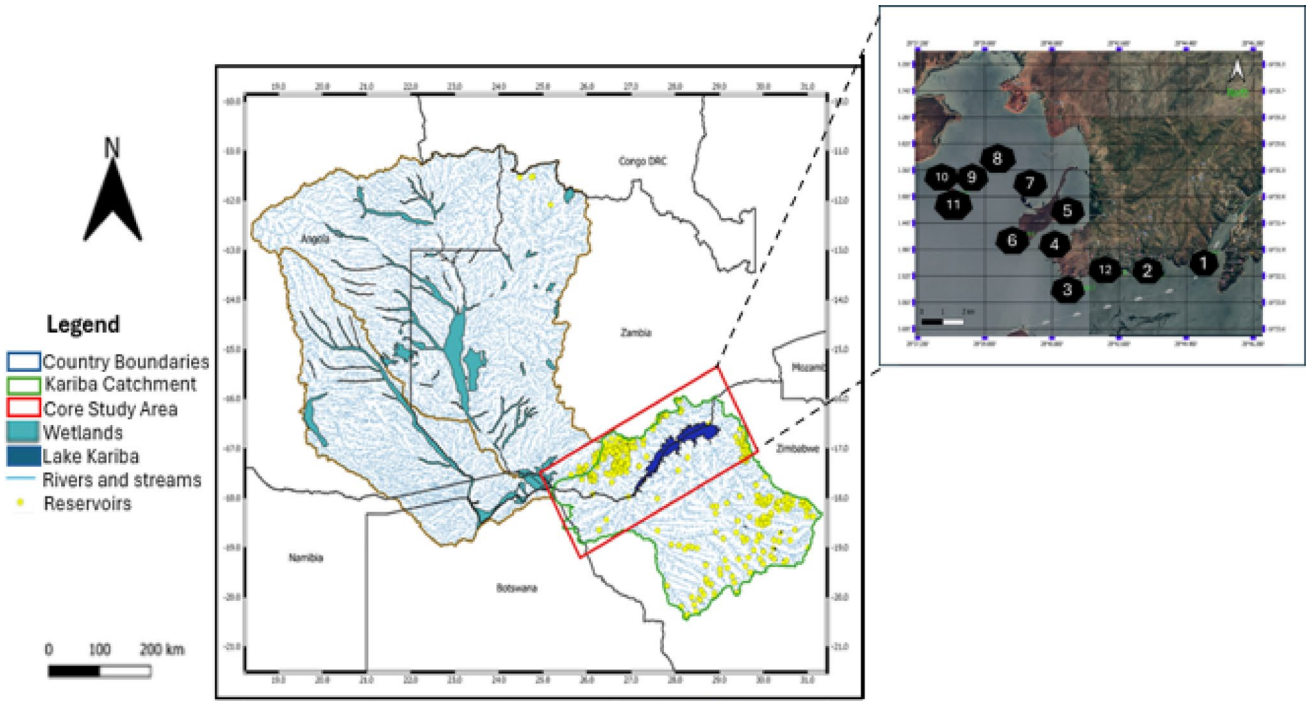


Fig. 1 Map of sampling point locations on Lake Kariba

glassfiber (1 µm) to eliminate the remaining smaller suspensions. The extraction was performed with SPE cartridges containing 1000 mg reversed phase octadecyl (C_{18} bonded silica) sorbent. The SPE technique offers many benefits including increased consistency as well as recoveries and lower solvent consumption. Briefly, 10 mL of methanol and 5 mL ultrapure water was used to prepare and equilibrate the cartridge to start the SPE procedure. Between loading with sample solution, the column was not permitted to dry. A mild vacuum was applied at a rate of roughly 10 mL min^{-1} to allow the 300 mL of water sample to penetrate through the cartridge. Following sample extraction, SPE cartridges were vacuum-dried for 15 min.5 mL hexane was used to elute the analytes, and a mild stream of nitrogen gas was used to evaporate the extract to 1 mL. The concentrated eluate was then collected into 2.0 mL GC vials and injected in GC-µECD for analysis. Each experiment was repeated three times.

Extraction of Sediment Samples

QuEChERS extraction technique was used to extract dried sediment samples as described by Ben Salem et al., (2016) with some modifications. This method was chosen because it offers efficient recoveries, shorter analysis time and requires less sample handling. Additionally, the method involves low solvent consumption, works with very volatile substances, and may be applied to any type of solid matrix. Briefly, 5.0 g of sediment was weighed and placed into an empty 15 mL polypropylene tube followed by addition of 4 mL ultrapure water and shaking by hand for 1 min. 20 mL extraction solvent of hexane/acetone solution (1:1) and extraction salts (1.5 g NaCl, 6.0 g $MgSO_4$, 0.750 g disodium citrate and 1.5 g trisodium citrate) were then added to the tube followed by shaking by vortex for 5 min. The sample was then centrifuged for 3 min at 2500 rpm and the supernatant was collected and placed into a clean centrifuge tube. For clean-up 900 mg of $MgSO_4$ and 150 mg PSA was added to the collected supernatant and mixture was vortexed for 3 min. The mixture was later centrifuged for 4 min at 3000 rpm and supernatant collected, dried by gentle nitrogen stream and transferred to GC vial for GC-µECD analysis.

Instrumental Analysis

Analysis of the OCPs was conducted using an Agilent 7890 A gas chromatograph (Agilent Technologies, Palo Alto, CA, USA), equipped with a split/splitless injector and a ^{63}Ni electron-capture detection (ECD) system operating at 320 °C. The make-up gas for the detector was nitrogen at 99.99% purity. HP-5 phenyl methyl siloxane 5% capillary column of (30 m x 320 µm i.d x 0.25 µm film thickness) was used with nitrogen at 20 mL min^{-1} as carrier gas. The injector was operated in splitless mode at 250 °C. The oven temperature program was initially set to 100 °C for 1 min, then increased to 180 °C at 20 °C min^{-1}, then raised to 270 °C at 5 °C min^{-1} and finally to 320 °C at 20 °C min^{-1} and held for 2 min. The total run time was 26 min. The injection

volume used was 1 µL and each analysis was carried out in triplicate.

Agilent ChemStation software (Agilent Technologies) was used for data collection and analysis. Comparing the measured retention times of a sample to those of established standards allowed for the identification of the pesticide residue. The external standard approach, which plots calibration curves for each analyte using its peak regions versus concentrations was used for quantitative analysis of the OCP residues. All measurements were performed within the detector's linear range. To determine levels of analytes present, the peak regions whose retention durations match the standards were extrapolated on their matching calibration curves. Calibration standards solutions were prepared from 0.01 to 300 ng mL^{-1} and used to calibrate the instrument.

Quality Assurance and Quality Control

In this work, a number of quality assurance procedures were regularly employed, such as running blanks with every sample set and doing triplicate analyses of the samples. Every solvent utilized in the analysis was of analytical grade and they were analysed to look for any compounds that might interfere. The target compounds were not found in the blanks for any extraction process. Prior to analysis, the recovery rate of the extraction techniques was assessed and used adjust the target compounds quantitative values. In water samples, the SPE process was optimized and validated by spiking 100 mL milli-Q water at 8.0 ng mL^{-1} using OCP pure standard mixture and extracted according to the procedure outlined in Sect. 2.4. For sediment samples,5.0 g of sediment samples was fortified with 100 µL of 1000 ng g^{-1} of mixed standard of OCPs to generate concentration of 20 ng g^{-1}. After fortification, the samples were left overnight and subjected to extraction as described in Sect. 2.5.

Statistical Analysis

One-way analysis of variance (ANOVA), regression analysis, mean, and associated standard deviation are among the statistical analyses used in the paper. Microsoft Excel was used to do a descriptive analysis of the data, maintaining significance at 0.05. Percentage (%) recoveries, a limit of detection (LOD) and linearity were established in order to verify the analytical technique.

Risk Assessment

Using the USEPA recommended guideline as described by Ge et al. (2014) and Yahaya et al. (2017), the health risk evaluation of OCPs after oral water intake of the lake surface water was carried out. Risk assessment in this study was evaluated through the calculation of average daily intake using lifetime average daily dose (LADD, $mg^{-1}kg^{-1}day^{-1}$) for carcinogenic effects and average daily dose (ADD, $mg^{-1}kg^{-1}day^{-1}$) for toxic effects. ADD and LADD were evaluated using Eq. (1), in accordance with standard methods set by (ECETOC 2016).

$$\text{ADD or LADD} = \text{C} \times \text{FI} \times \frac{(\text{IR} \times \text{EF} \times \text{ED})}{(\text{BW} \times \text{AT})}, \quad (1)$$

In this case, C represents the mean concentration of analyte (OCPs) (mg L^{-1}); ADD intake exposure level (mg^{-1} kg^{-1} day^{-1}). FI is the fraction ingested (an absolute number in the range 0–1) which is considered as 0.98 according to Megahed et al. (2015) and Yahaya et al. (2017). IR stands for the daily water intake rates (ECETOC 2016) which are 0.3 L Day^{-1} for children between ages of 0 to 6 years, 1 L for age group 7–17 years, and 1.4 L Day^{-1} for adults. EF is the exposure frequency assumed as 365 days $year^{-1}$. ED is the exposure duration, which is determined taking into account the age group, 0–6 years = 6; Age 7–17 years = 11; Adult = 3.BW is the average body weight of 30 Kg (0–6), 46 Kg (7–17), and 70 Kg for adults (Megahed et al. 2015). AT is period over which exposure to averaged expressed in days. For ADD, the AT value are 2190 days, 4015 days and 10,950 days for 0–6 years,7–17 years and adults respectively. Whereas in LADD (for carcinogens) the AT value is based on lifetime exposure of 70 years, equivalent to 25,550 in days.

Further risk analysis was conducted by evaluating the hazard quotient (HQ), which is the ratio of exposure to an appropriate reference dose such as average daily intake coupled with specific toxic and carcinogenic parameters. In this instance, the reference daily dose (RfD) was used to evaluate the HQ for non- carcinogenic risk Eq. (2).The RfD values were sourced from the United States Environmental Protection Agency-Integrated Risk Information System (USEPA-IRIS 2019).On the other hand, cancer slope factors were applied to compute cancer risk (CR), Eq. (3). The following equations were used for calculation of the risk assessment:

$$\text{Hazard Quotient } \left(\text{HQ}_{\text{Non-carcinogenic}}\right) = \frac{\text{ADD}}{\text{RfD}}, \quad (2)$$

$$\text{Cancer risk } (\text{CR}) = \text{ LADD x CSF} \quad (3)$$

RfD is the reference dose and CSF (0.07) is cancer slope factor, and all expressed in ($mg^{-1}kg^{-1}day^{-1}$). Values of HQ for contaminants under 1 are considered safe whereas cancer risk values greater than 1×10^{-6} indicate highest risk to cancer and values equal to 1×10^{-3} require protective

measures (USEPA - IRIS 2014; USEPA 2020).Additionally, ADD > 10^{-4} indicates the maximum lifetime cancer risk and LADD > 10^{-6} indicates the highest risk of cancer but LADD = 10^{-3} indicates that protective measures are required to prevent contact with polluted water.

Results and Discussion

Quality Assurance

Table 2 summarises the various analytical control measures conducted. Of the 20 targeted OCPs, 18 were linear from 0.01 to 50 ng mL^{-1} range, except for endosulfan sulfate and endrin ketone whose linear range was found to be 40–300 ng mL^{-1}. This was due to low peak intensity hence they could not be detected at lower concentrations. The calibration curves for the 18 OCPs showed good linearity with R^2 values in the range of 0.990–0.999, which were within the acceptable range of $R^2 \geq 0.990$ (Pihlström et al. 2017; Yahaya et al. 2017). Instrument detection limits (IDLs) were obtained from linear regression equations of the calibration curves of the OCP standards (He et al. 2024), and they varied from 0.23 ng mL^{-1} (dieldrin) to 18.34 ng mL^{-1} (endosulfan sulfate). To optimize and validate the extraction procedure in water, recoveries of OCP compounds were determined from 100 mL of ultrapure milli-Q water spiked with 80 µL of 10 mg L^{-1} pure OCP standard mixture. The average recoveries ($n = 3$) of OCPs in water ranged from 75.94 to 116.67% except for aldrin with 61.04% recovery, whereas surrogate recoveries were between 61.92 and 72.14% shown in (Table 2). These recoveries were all in acceptable levels according to analytical guidelines, except for endrin ketone and endosulfan sulfate which were not recovered at this spiking concentration of 8.0 ng mL^{-1}. The QuEChERS extraction for sediments was validated by fortifying 5.0 g of sediment samples with 100 µL of 1000 ng g^{-1} of mixed standard of OCPs to generate concentration of 20 ng g^{-1}. The average recoveries were found to range from 74.15 to 120.59% except for heptachlor, endosulfan sulfate and endrin ketone which were not recovered.

OCP Levels in Water and Sediment Samples

Table 3 lists the total concentrations of OCPs (∑OCPs) in the water and sediment samples from selected sites of Lake

Table 2 Retention time (t_r), correlation coefficient(R^2), linear range, instrument limit of detection and recoveries of OCPs in sediment and water

					Sediment		Water	
Compound	t_r (min)	R^2	Linear Range (ng mL^{-1})	LOD	Recovery (%)	R.S.D %	Recovery (%)	R.S.D (%)
Surrogate [1]	8.52	0.999	0.05–50	0.37	72.14	4.9	78.75	4.5
α-HCH	9.36	0.999	0.05–50	1.66	107.69	5.5	74.49	4.2
γ-HCH (Lindane)	9.95	0.999	1.00–50	0.44	114.15	5.8	111.43	7.6
β-HCH	10.14	0.999	0.05–50	1.13	111.91	7.8	79.58	5.2
δ-HCH	10.76	0.999	1.00–50	1.24	87.90	3.6	120.59	4.1
Heptachlor	11.34	0.990	2.0–50	6.32	88.26	8.9	NR	NR
Aldrin	12.11	0.999	0.10–50	1.97	61.04	7.8	90.03	4.7
Heptachlor epoxide	13.16	0.999	0.01–50	0.26	104.44	7.8	77.4	7.2
trans-chlordane	14.28	0.999	0.05–50	0.47	106.26	3.5	91.35	2.5
cis-chlordane	15.00	0.999	0.05–50	0.44	82.05	12.8	82.33	4.6
α -endosulfan	15.44	0.999	0.05–50	0.43	99.53	6.9	86.00	1.6
p, p′-DDE	16.03	0.999	0.10–50	0.53	82.05	12.8	78.74	1.5
Dieldrin	16.28	0.999	0.05–50	0.23	89.22	7.5	93.40	7.4
Endrin	16.96	0.990	0.50–50	1.87	104.66	7.8	99.56	0.9
4,4'-DDD	17.26	0.999	0.05–50	0.72	116.67	5.8	112.45	3.3
β - endosulfan	17.38	0.999	1.0–50	1.08	98.21	6.5	80.27	3.5
p, p'-DDT	17.77	0.999	1.0–50	1.73	108.30	6.7	74.15	3.2
Endrin aldehyde	18.52	0.999	0.5–50	0.56	75.94	2.9	106.32	4.4
Endosulfan sulfate	18.64	0.996	40–300	18.34	NR	NR	NR	NR
methoxychlor	20.1	0.999	0.5–50	1.81	100.23	11	93.37	5.5
Endrin Ketone	20.43	0.997	50–300	17.28	NR	NR	NR	NR
Surrogate[2]	25.26	0.996	0.1–50	2.63	61.92	5.6	106.2	0.9

Surrogate[1] is 2,4,5,6-Tetrachloro-m-xylene; Surrogate[2] is Decachlorobiphenyl

RSD relative standard deviation ($n = 3$)

LOD Limit of detection

NR No Recovery

Table 3 Concentrations OCPs in surface water and sediment samples of lake Kariba

		Water (ng mL^{-1})			Sediment (ng g^{-1})		
Analyte	logkow	Range	Mean	SD	Range	Mean	SD
α-HCH	3.80	nd − 0.111	0.013	2.36E-04	0.701–2.659	1.522	0.033
γ-HCH (Lindane)	3.76	nd − 0.040	0.01	7.07E-04	2.158–3.754	1.932	0.269
β-HCH	3.36	nd − 0.075	0.012	1.19E-03	0.589–5.148	3.08	0.184
δ-HCH	4.14	0.022–1.839	0.111	3.17E-03	0.341–0.449	0.389	0.04
∑HCHs		0.022–2.065	0.146	5.30E-3	3.789–12.01	6.923	0.526
Heptachlor	5.20	< LOD	< LOD	< LOD	nd	nd	nd
Aldrin	6.50	< LOD	< LOD	< LOD	2.801–4.526	3.559	0.26
Heptachlor epoxide	5.44	nd − 0.006	0.001	1.04E-04	nd	nd	nd
trans-chlordane	5.73	nd – 0.023	0.01	2.45E-04	0.813–15.14	7.97	0.74
cis-chlordane	5.54	nd-0.134	0.005	3.58E-04	nd	nd	nd
α-endosulfan	5.54	nd − 0.167	0.008	1.76E-04	0.308–1.945	1.208	0.063
Dieldrin	8.78	nd − 0.073	0.007	2.43E-04	1.604–3.237	2.324	0.093
Endrin	5.60	nd	nd	nd	nd	nd	nd
β-endosulfan	4.8	nd – 0.135	0.012	5.90E-02	0.813–3.136	1.924	0.14
Endrin aldehyde		nd − 0.339	0.034	3.58E-03	12.10–29.29	17.72	1.023
Methoxychlor	5.08	nd − 0.027	0.009	1.79E-04	nd	nd	nd
p, p′-DDE	6.51	nd − 0.026	0.005	2.54E-04	6.750–22.64	12.88	1.29
4,4'-DDD	6.02	nd − 0.151	0.014	2.73E-04	1.243–20.78	11.27	0.546
p, p'-DDT	6.91	< LOD	< LOD	< LOD	nd	nd	nd
∑DDTs		0.00–0.177	0.019	5.27E-04	7.993–43.42	24.15	1.836
∑OCPs		0.022–3.119	0.251	7.01E-02	30.22–112.7	58.86	2.865
DDEs/∑DDTs			0.263			0.53	
DDDs/∑DDTs			0.737			0.47	

∑HCHs: sum of α-, β-, γ- and δ -HCHs

∑*DDTs* Sum of p.p′-DDE, 4,4′-DDD

nd not detected

<*LOD* below limit of detection

SD standard error of mean ($n = 3$)

Kariba. A total of fourteen OCPs were identified in the surface water samples. The total OCP ($\sum_{14}$OCPs) concentration ranged from 0.022 to 3.119 ng mL^{-1} with an average concentration of 0.257 ng mL^{-1} .The compounds that were found included HCH isomers (α-HCH, β-HCH, γ-HCH, δ-HCH), heptachlor epoxide, CHLs (trans- and cis-chlordane), ENDOs (α- and β-endosulfan), dieldrin, endrin aldehyde, methoxychlor and DDT metabolites dichlorodiphenyldichloroethylene (p, p'-DDE) and dichlorodiphenyldichloroethane (4,4'-DDD) (Fig. 2a). The mean concentrations of most abundant OCPs in the study area descended in the order of δ-HCH > endrin aldehyde > DDT metabolites > the rest of the OCPs (Fig. 3a).Among the targeted OCPs, the HCH isomers exhibited high concentration with a total concentration ranging from 0.022 to 2.065 ng mL^{-1}, with δ-HCH being the most abundant accounting for 76% of total HCH concentration. The concentration of endrin aldehyde varied from nd − 0.339 ng mL^{-1} with a mean concentration of 0.034 ng mL^{-1}. According to Kılınç and Yenisoy (Kılınç and Yenisoy 2025), endrin has a tendency of transforming into its metabolites endrin aldehyde and endrin ketone thus, the detection of endrin aldehyde signifies a renewal source of endrin. On the other hand, the DDT metabolites were found to have total concentration ranging from nd − 0.177 ng mL^{-1} and a mean concentration of 0.019 ng mL^{-1}. As reported by Kurek et al. (Kurek et al. 2025), DDT can break down into DDE and DDD through environmental processes including sun radiation and biological metabolism supporting their detection.

The summarised results for the OCP concentrations in sediments are presented in (Table 3).The total concentrations of $\sum_{12}$OCPs ranged from 30.22–112.7 ng g^{-1} with a mean value of 58.86 ng g^{-1}.The DDT metabolites were the most prevalent in sediment samples with total concentrations ranging from 7.99–43.42 ng g^{-1} and a mean value of 24.15 ng g^{-1}.The mean concentrations decreased in the order DDTs > endrin aldehyde > trans chlordane > the rest of OCPs (Fig. 3b). For DDTs, p,p'-DDE was detected at the highest level with total concentration ranging from 6.75–22.64 ng g^{-1} followed by 4,4'-DDD (1.24–20.78 ng g^{-1}). p, p'-DDE and 4,4'-DDD constituted 53% and 47% of total DDTs (tDDTs) in sediments, respectively. A metabolite of endrin, which is endrin aldehyde was detected with total concentration ranging from 12.10–29.29 ng g^{-1} and mean

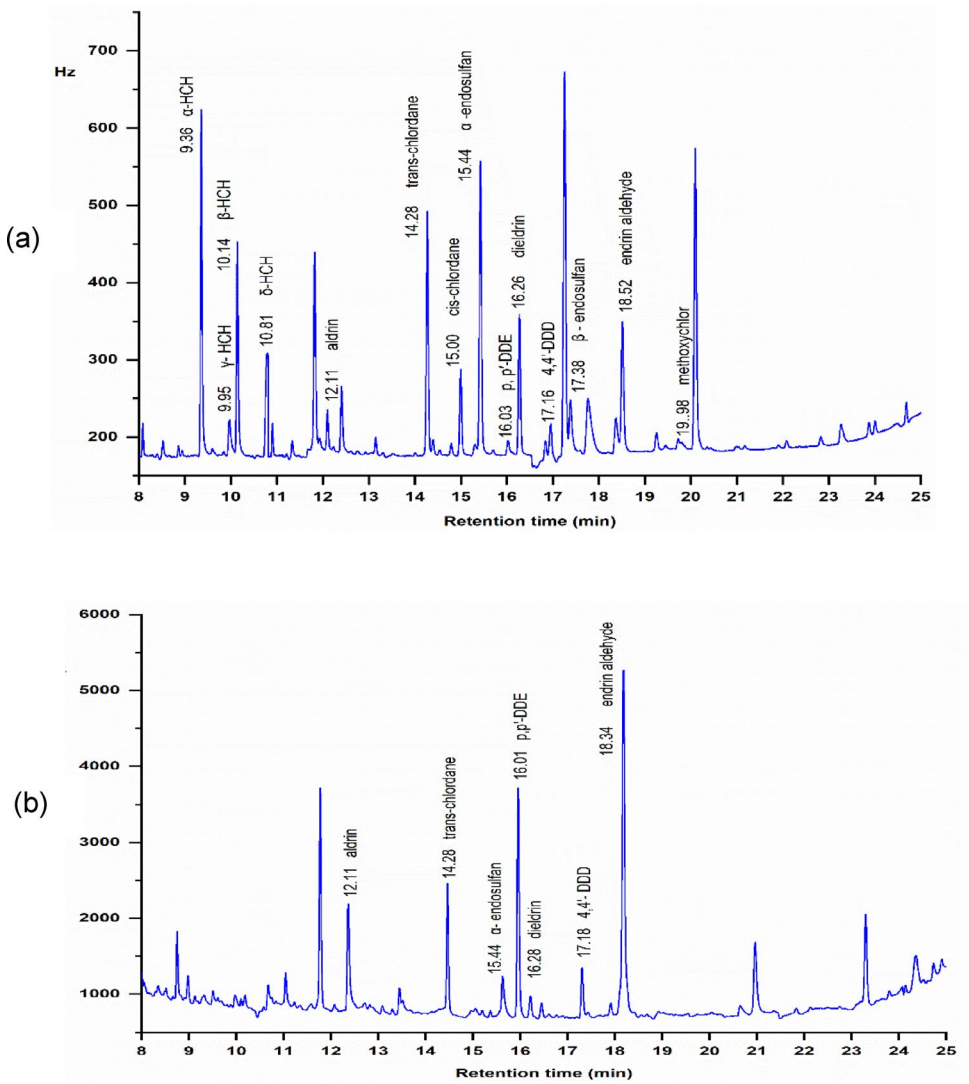


Fig. 2 GC-ECD chromatogram of (**a**) extracted water sample (**b**) sediment sample

value of 17.72 ng g^{-1}.Trans chlordane was detected with concentrations ranging from 0.813 to 15.14 ng g^{-1} with a mean concentration of 7.970 ng g^{-1} .The total concentration of HCH isomers ranged from 3.79–12.01 ng g^{-1} with a mean value of 6.92 ng g^{-1}. The mean concentrations of the HCH isomers were in the order: β-HCH (3.080 ng g^{-1}) > γ- HCH (1.932 ng g^{-1}) > α-HCH (1.522 ng g^{-1}) > δ-HCH (0.389 ng g^{-1}) and β-HCH isomer constituted about 44% of total HCHs in sediments. Aldrin was detected with total concentration of 2.801–4.526 ng g^{-1}, and mean value of 3.559 ng g^{-1}.

log Kow were obtained from (Shen and Wania 2005).

Distribution of OCPs in Surface Water and Sediments

Figure 3a and b illustrates the distribution of OCP residues in the surface water and sediment samples respectively. The analysed surface water samples showed presence of α-, β-, γ-, and δ-HCH, heptachlor epoxide, cis- and trans-chlordane, α- and β-endosulfan, dieldrin, endrin aldehyde, methoxychlor and DDT metabolites (p, p'-DDE and 4,4'-DDD). Overall, OCPs showed the highest concentration in site 7 which is location on west of banana island on the Lake Kariba (Fig. 3a). This was followed by site 5 and 6 which are locations near small scale fish cages and south of banana island respectively. The possible explanation for this is that the banana island is located on the lakes eastern shore in the lower Zambezi region which is heavily reliant on agriculture. Because hexachlorocyclohexane has a wide range of applications as insecticides on crops and animals, it is frequently used to control agricultural pests. As a result, it can find its way in the lakes ecosystem via surface runoff from surrounding farmlands. Furthermore, it was noted that the HCH isomers were the most predominant OCPs detected in the water samples. This observation is consistent with the fact that HCHs have the lowest octanol-water partition coefficient (log Kow) values among the target OCPs in this

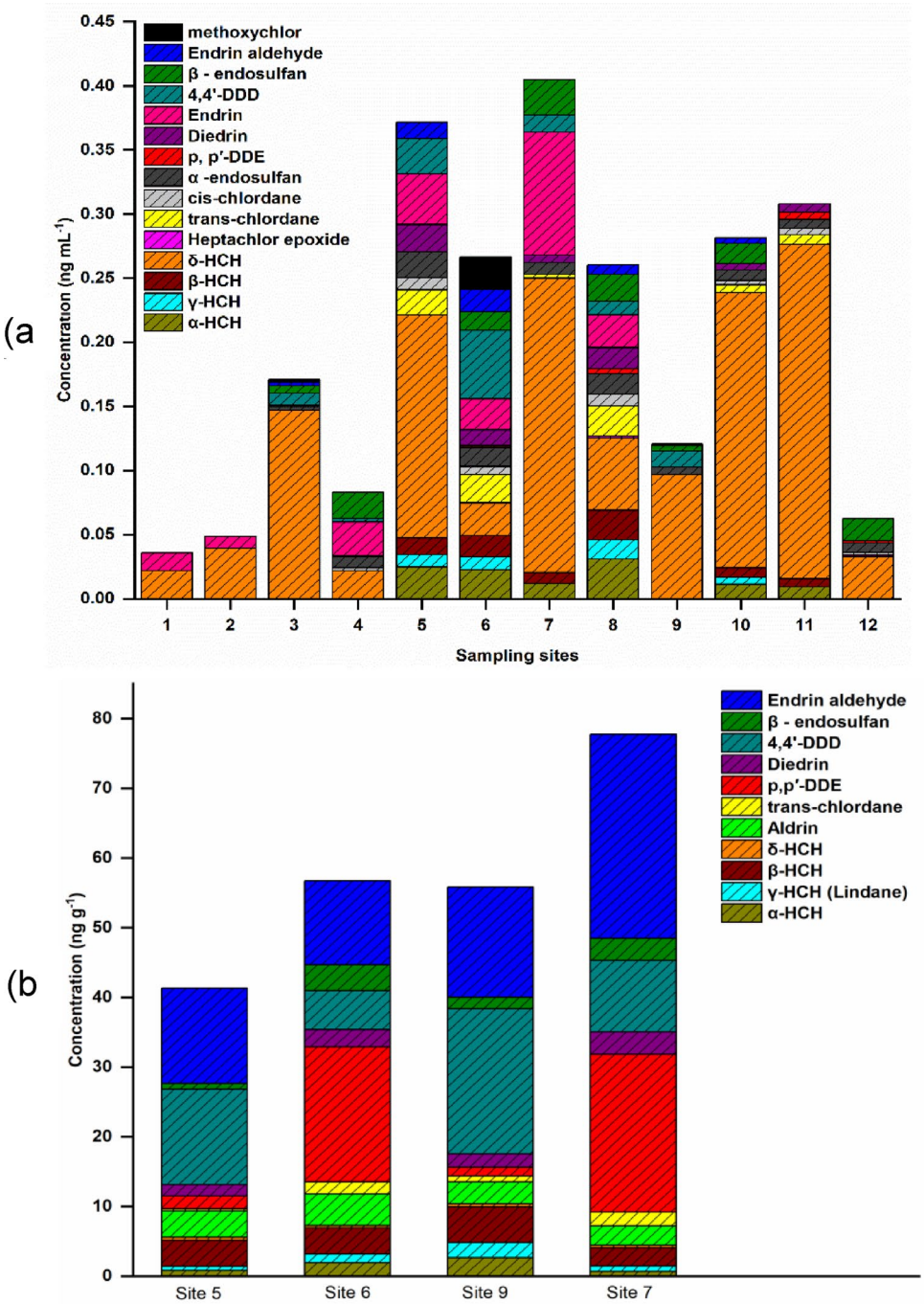


Fig. 3 Concentrations of OCP in (**a**) water (ng mL^{-1}) and (**b**) sediment (ng g^{-1}) from sampling sites

investigation (log Kow ranging from 3.72 to 4.14) (Seung-Kyu 2020).The distribution of a chemical at equilibrium between the organic (octanol) and aqueous (water) phases is indicated by the log Kow, a measure of hydrophobicity. This coefficient can be used to predict how organic compounds, like pesticides, would behave in the environment. The likelihood that a chemical will partition into organic phases increases with log Kow. The HCH isomers are therefore partitioned to the water phase as stated since they have the lowest log Kow.

Moreover, the elevated HCH concentrations in water found in this investigation support prior research demonstrating that the HCH isomer pollution in aquatic ecosystems is a significant global issue (Vijgen et al. 2011).The two major sources of environmental HCH exposure is either by direct application of lindane or technical HCH (Li et al. 2006).According to Hu et al., (Hu et al. 2011), lindane contains 99% γ-HCH, while technical grade HCH has roughly 60–70% α-HCH, 5–12% β-HCH, and 10–15% γ-HCH. Changes in the environmental composition of HCH isomers

indicate the source of pollution (Doong et al. 2002). In this instance, the current study clearly showed that δ-HCH was more prevalent than the α-, β-, and γ-HCH isomers. δ-HCH was the most common isomer, accounting for an average of 73.7% of all HCHs. It was followed by α-HCH (12.2%), β-HCH (7.7%), and γ-HCH (6.4%) (Table 3).From compositional variations, there is a notable increase in percentage of δ-HCH from (5% −12%) in technical grade HCH (Hu et al. 2011) to 73.7% detected in current investigation, an indication that its accumulating in the environment. However, there is a decrease in percentage of α-HCH (60–70%) and γ-HCH (10–15%) in technical grade HCH (Hu et al. 2011) to 12.2% and 6.4% respectively, as reported in this study, indicating the likelihood of their transformation (Li et al. 2006).

Furthermore, an examination of the ratio of α-HCH to γ-HCH can be used to identify the principal source of HCH contamination and ascertain whether the source is technical HCH or lindane (Doong et al. 2002). If the α-HCH/γ-HCH ratio is between 4 and 7, it would be technical HCH; if it is nearly zero, it would be for lindane (Hu et al. 2011). The ratio of α-HCH/γ-HCH in the water samples in this investigation ranges from 2.0 to 2.7, suggesting that the majority of the HCH residue in the area under study comes from recent lindane usage. According to reports by Joint ,1997 (Joint 1997), Zambian agriculture uses lindane, which is traded as gammalin-20, to manage pests. This current result is in contrast to previous reports from the Buffalo River in South Africa (Yahaya et al. 2017), the Densu River basin in Ghana (Kuranchie-Mensah et al. 2012), and the River Wuchan in China (Zhang et al. 2002). However, the findings of this study are similar to a report from the Northern Greek rivers (Golfinopoulos et al. 2003) and the underground rivers in Chongqing, Southwest China (Hu et al. 2011).

DDT metabolites on the other hand were found in very low concentrations in the water samples analysed (Table 3). Due to high octanol-water coefficient values (log Kow ranging from 6.02 to 6.91), DDT and its metabolites tend to be partitioned away from the water column. The ratio between DDT and its metabolites (DDD and DDE) can be used to evaluate whether the DDT inputs are recent or not. If the calculated ratios are much lower than unity, then recent input occurred in the area (Ahmed et al. 2012). In the current study, the calculated DDEs/tDDTs and DDDs/DDTs ratios (Table 3) were much lower than unity, suggesting that the DDTs found in water samples mainly came from recent inputs of DDT in the area. The fact that DDT is still in use especially for malaria prevention in Zambia aligns with this observation.

The distribution of OCP residues found in sediments taken from the four sampling sites along Lake Kariba is displayed in (Fig. 3b). The results in sediments revealed significantly higher concentrations of OCPs than in water samples, with total values ranging from 30.22 to 112.7 ng g^{-1} .This observation aligns with poor water solubilities (from 700 mg L^{-1} to 21,300 mg L^{-1}) and high octanol/water partition coefficients (log Kow) values (3.9–6.2) of OCPs (Ahmed et al. 2012).This results in OCPs to exhibit a strong affinity for suspended particles and eventually settle in sediments. It was further noted that the greatest levels of OCPs was found in sediments collected from site 7 which is location on west of banana island on the Lake Kariba. Additionally, higher OCP levels were observed at sites 5 and 6 which are locations near small scale fish cages and south of banana island respectively. These locations are in the lower Zambezi region, which is impacted by agricultural pollution from pesticides, as was previously mentioned.

The predominant OCPs in sediments were found to be the DDT metabolites. This observation is consistent with the fact that DDT and its metabolites have very low water solubility and high octanol-water partition coefficient with log Kow > 5 making them exhibit high affinity for suspended matter and eventually partitioned to sediments in the ecosystem. Further evaluation of whether the DDT inputs are recent or not was conducted by computing the ratios between tDDTs and its metabolites namely, DDEs and DDTs. The calculated DDEs/tDDTs and DDDs/tDDT ratios in sediment were found to be 0.53 and 0.47 respectively (Table 3). According to (Ahmed et al. 2012), since the ratios were less than unity, it suggests that the DDT deposits are recent. Similarly, elevated levels of endrin aldehyde, a metabolite of endrin was found with total concentration ranging from 12.10 to 29.29 ng g^{-1} and mean value of 17.72 ng g^{-1}.This can be attributed to possible use of endrin as insecticide in agriculture and can find its way in lakes ecosystem, owing to the fact the community around the lake is heavily reliant on agriculture. Trans chlordane was equally found in elevated levels with mean concentrations of 7.970 ng g^{-1} and total concentrations ranging from 0.813 to 15.14 ng g^{-1}. The detection of DDT and chlordane compliments the revelations in the 2004 national inventory of Zambia (UNEP 2007). Due to their continued use, DDT and chlordane were identified in this report as POPs of concern in Zambia. It was observed that while DDT is only used for IRS to prevent malaria, chlordane is still used to control termites in the construction sector and on plantations. It was further revealed that Zambia used a total of 29,615 Kg of DDT for IRIS from 2000 to 2004, with a projected increase in quantity to 56.280 Kg in the 2006/2007 procurement period (UNEP 2007).

Risk Assessment

Health Risk Assessment

Health risks expected when drinking water contaminated with OCPs for adults and children were evaluated using the USEPA exposure factors handbook. Table 4 displays the average cancer risk, LADD, and ADD determined for all age groups. The cancer risk values for all age groups were found to be lower than the maximum limit of 10^{-6} as set by USEPA measures (USEPA - IRIS 2014; USEPA 2020), indicating no cancer risk. The calculated ADD for all compounds considering both children and adults were below the acceptable limit except the ADD for δ-HCH in children which was found to be higher than the limit of 1×10^{-4} an indication of maximum lifetime cancer risk. Risk evaluated using life average daily dose (LAAD) for all the detected compounds were below the set limit 1×10^{-6} except for δ-HCH in all age groups and Endrin in children aged between 7 and 17 yrs. The LADD values for δ-HCH and endrin which were found to be higher than the set limit suggest the highest risk of cancer for these age groups. This implies that children's exposure to these compounds over time may pose health risks, and it is advised that they avoid contact. Children's lower weight and increased hand-to-mouth activities may be the cause of this elevated risk. Figure 4 provides a summary of the HQs of the contaminants found in the lake through drinking water and were found to be less than 1 as an indication of low risk.

Public Health Implications

The study provides reasonable evidence that OCPs are still present in the ecosystem of Lake Kariba. Even if they are found in small quantities, their detection is a danger to the environment and human health. This necessitates remedial measures because the lake is currently the site of significant aquaculture operations and a supply of drinking water for nearby populations. As previously mentioned, eating or being exposed to these chemicals through water or fish raised in the lake poses a number of health hazards to the general public, including cancer, behavioural issues, and reproductive abnormalities among others. Based on this data, there is a need for policy makers to prioritise legislative and management framework to deal with the reduction in POPs release to the environment. Updating national POP inventories in accordance with SC regulations would also provide information on the nation's progress in managing POPs. The findings also show that a robust method for monitoring organochlorine pesticides in aquatic ecosystems is desperately needed (Arar et al. 2025), particularly in areas where commercial fish farming is practiced. The current Zambia fisheries act of 2011 (The Fisheries Act 2011) and the National Fisheries and Aquaculture Policy (NFAP) of 2023 (MFL 2023) are both silent in this aspect. Likewise, the local community should be made aware of the proper usage of the lake's water and educated to keep kids away from it because of their lower body weight and increased risk from hand-to-mouth activities.

Comparative Analysis with Other Studies

This study is the first to evaluate the amount and presence of OCPs in the sediment and surface waters of Lake Kariba, Zambia and essentially lacks data for comparison. However, prior research on the lake from Zimbabwe's perspective has been conducted, including a study on fish by (Berg et al. 1992) and one on sediments by (Zaranyika et al. 1994), both of which were carried out more than thirty years ago. On

Table 4 Individual average daily dose (ADD), life average daily dose (LADD), and cancer risk of OCPs

	Maximum	ADD x 10^{-6}			LADD x 10^{-8}		Cancer
OCPs	Conc. (μg L^{-1})	0 to 6 yrs	7 to 17 yrs	Adult	0 to 6 yrs	7 to 17 yrs	Risk x 10^{-16}
α-HCH	0.111	1.09	2.36	0.22	9.32	37.16	6.53
γ-HCH (Lindane)	0.040	0.39	0.85	0.08	3.36	13.39	2.36
β-HCH	0.075	0.73	1.60	0.15	6.30	25.11	4.40
δ-HCH	1.839	18.02	39.18	3.60	154.48	615.67	108.13
Heptachlor epoxide	0.006	0.06	0.13	0.01	0.50	2.01	0.34
trans-chlordane	0.088	0.84	1.87	0.17	7.39	28.46	5.06
cis-chlordane	0.041	0.40	0.87	0.08	3.44	13.73	2.43
α-endosulfan	0.119	1.17	2.54	0.23	10.00	39.84	6.41
p, p'-DDE	0.026	0.13	0.55	0.05	2.18	8.70	0.76
Diedrin	0.073	0.71	1.56	0.14	6.13	24.44	4.28
Endrin	0.339	3.32	7.22	0.66	28.48	113.49	19.91
4,4'-DDD	0.151	1.48	3.22	0.30	12.68	50.55	8.86
β-endosulfan	0.135	1.32	2.88	0.26	11.34	45.20	7.94
endrin aldehyde	0.049	0.49	1.04	0.10	4.12	16.40	2.90
methoxychlor	0.028	0.27	0.60	0.05	2.35	9.37	1.62

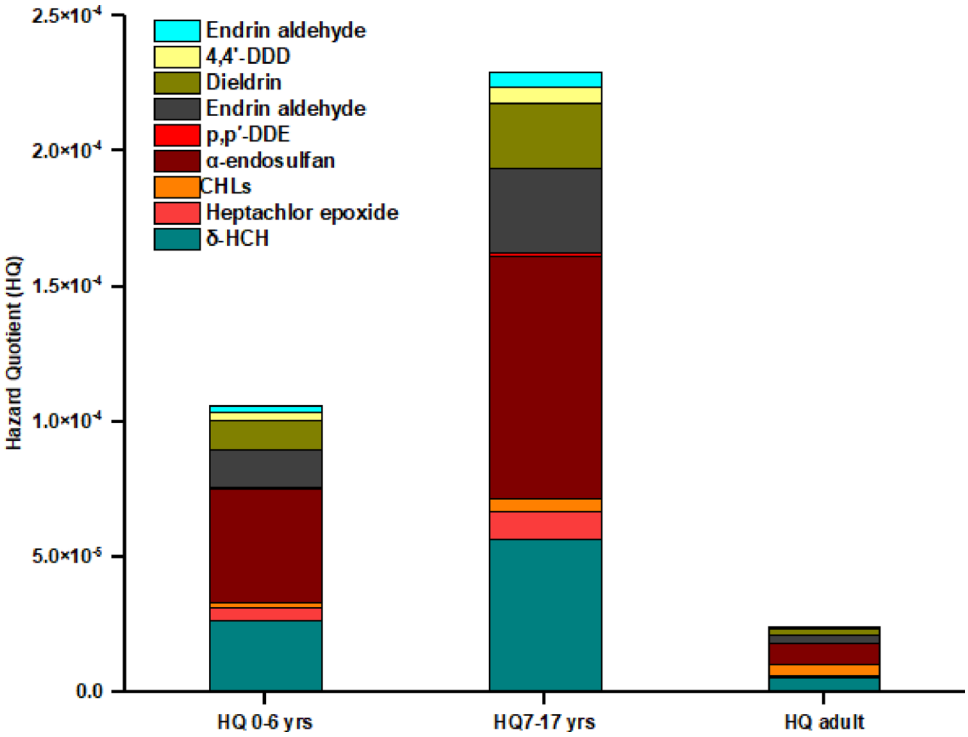


Fig. 4 Hazard Quotient (HQ) of OCPs for age 0–6, 7–17 years, and adults

Table 5 Comparison of the mean concentration of OCPs in water (ng mL^{-1}) and sediment (ng g^{-1}) from lake Kariba with other studies

Matrix	Location/ Reference	∑HCHs	Aldrin	∑DDTs	Chlordane's	Endosulfan's	Dieldrin	endrin aldehyde
Water	Lake Victoria, Tanzania (Henry and Kishimba 2003)	nd-200		nd-1600				
	Lagos Lagoon, Nigeria (Adeyemi et al. 2011)	0.005–0.910			0.006–0.95	0.015–0.996	0.015–0.996	
	Okavango Delta, Botswana (Mmualefe et al. 2009)			7.7	3.2			
	Lake Volta, Ghana (Gbeddy et al. 2015)	<LOQ – 0.669		<LOQ – 0.031	<LOQ-0.009	<LOQ-0.076	<LOQ-0.031	
	uMngeni River, South Africa (Birungi et al. 2018)	0.50–1.97		1.21–2.32				0.88–3.48
	Lake Kariba, Zambia (this study)	0.146	<LOD	0.019	0.015	0.020	0.007	nd -0.339
Sediment	Lake Victoria, Tanzania (Henry and Kishimba 2003)	1.32×10^5		6.0×10^5				
	Lake Bosomtwi, Ghana (Darko et al. 2008)			12.75		9.683 ± 1.76	0.072 ± 0.02	
	Lake Manzala, Egypt (Barakat et al. 2012)	nd-3.42		0.2–7.25	nd-2.35		nd − 0.03	
	Lake Victoria, Uganda (Wasswa et al. 2011)	4.24 ± 3.83			0.45 ± 0.23	6.00 ± 4.36	3.803 ± 55	
	Lake Kariba, Zimbabwe (Zaranyika et al. 1994)		0.017–4.10	1.243–49.14		1.91–16.5	2.27–37.1	
	Lake Kariba, Zambia (this study)	6.923	3.559	24.15	7.970 ng g^{-1}	3.13	2.32	17.72

Zambian side of Lake Kariba, a notable study includes one on fish conducted by (Simukoko et al. 2023).

However, a comparative analysis of the findings was done against studies from other aquatic bodies within Africa and is depicted in (Table 5). In the current study, 14 OCPs were identified in the lake's surface waters with total concentrations ranging from 0.022 to 3.119 ng mL^{-1} and mean value of 0.257 ng mL^{-1}.The most prevalent OCPs in decreasing order of mean concentrations included HCHs isomers (0.146 ng mL^{-1}), endrin aldehyde (0.034 ng mL^{-1}),

DDT metabolites (0.019 ng mL^{-1}), endosulfan's (0.016 ng mL^{-1}). When compared to results from Lake Victoria in Tanzania (Henry and Kishimba 2003), Lake Volta in Ghana (Gbeddy et al. 2015), Lagos Lagoon in Nigeria (Adeyemi et al. 2011), Okavango Delta in Botswana (Mbongwe et al. 2003) and the uMngeni River in South Africa (Birungi et al. 2018), the mean concentrations of OCPs in the water samples from this investigation were generally lower.

The total concentration of OCP residues in sediments in the current investigation ranged from 30.22–112.7 ng g^{-1}, with an average of 58.86 ng g^{-1} .The individual mean concentrations of detected OCPs in sediments followed the decreasing order: DDT metabolites (24.15 ng g^{-1}), endrin aldehyde (17.72 ng g^{-1}), trans chlordane (7.970 ng g^{-1}), HCH isomers (6.923 ng g^{-1}), aldrin (3.56 ng g^{-1}), endosulfan's (3.13 ng g^{-1}) dieldrin (2.32 ng g^{-1}). Comparatively, the concentrations in this investigation were largely consistent with the findings of Zaranyika et al., (Zaranyika et al. 1994) in 1992 on the same lake but on the Zimbabwe side. However, the DDT levels found in this study were lower than those reported by Zaranyika on the same lake over thirty years ago. Given that DDT is prohibited and its use is restricted in many nations, this may be an indication that its occurrence in the environment is declining. In further comparison, results from Lake Victoria, Tanzania (Henry and Kishimba 2003) showed higher levels of HCHs and DDTs than those found in this study, but lower levels than those found in Lake Manzala, Egypt (Barakat et al. 2012) and Lake Victoria, Uganda (Wasswa et al. 2011).Trans chlordane in sediment from this study was found to be higher than that reported from Lake Manzala Egypt (Barakat et al. 2012) and Lake Victoria Uganda (Wasswa et al. 2011).

Conclusion

The current investigation has revealed the current state of the levels of OCPs in water and sediment samples from Lake Kariba, Zambia. Out of twenty targeted pesticides, fourteen OCPs were detected. HCHs, endrin aldehyde and DDT metabolites were commonly detected in both water and sediments. While endrin, endosulfan sulfate, and endrin ketone were not found in any of the twelve sites, heptachlor and aldrin were below the detection limits. A comparative analysis with other studies revealed that the concentration of these class of compounds in both water and sediments was generally lower and, in certain circumstances, equivalent. Legacy use of these pesticides could be the primary sources of contamination with some instances of recent deposits from DDT and chlordane which are still in use. According to the findings of the risk assessment, OCPs found in water would not be harmful to human health, but long-term usage of Lake Kariba water could have carcinogenic consequences from δ-HCH from oral ingestion. However, given that the Lake is a host of numerous aquaculture activities across the nation, the finding of OCPs in the water samples even in trace amounts, warrants further research and monitoring. It is therefore recommended that existing policies should have clear focus on continuous monitoring of OCP levels and regulation of the use of certain pesticides and insecticides to reduce the negative effect on the environment.

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Data Availability The data supporting the findings of this manuscript are available from the corresponding author upon reasonable request.

Declarations

Conflict of interest The authors declare no conflict of interest.

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