Received: 20 April 2021 | Revised: 7 October 2022 | Accepted: 19 December 2022

DOI: 10.1111/lre.12424

Lakes & Reservoirs
Science, Policy and Management for Sustainable Use
WILEY

ORIGINAL ARTICLE

Organochlorine pesticide residues in water, sediments and Nile tilapia (*Oreochromis niloticus*) of Lake Nakuru, Kenya and implications for its fishery

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Funding information
Orange Knowledge Programme, the Netherlands (NUFFIC)

Abstract

The present study was conducted to assess the levels of selected organochlorine pesticide (OCP) residues in water, sediments and Nile tilapia, as well as the potential health risks to humans who consume the fish of Lake Nakuru. Nine composite water and nine sediment samples, as well as 30 fish specimens, were collected from five representative sampling sites within Lake Nakuru. The OCPs targeted in the present study included p,p′-Dichlorodiphenyl dichloroethane (p,p′-DDD), p,p′-dichlorodiphenyldichloroethylene (p,p′-DDE), p,p′-dichlorodiphenyltrichloroethane (p,p′-DDT), heptachlor, heptachlor epoxide, α endosulfan (1), β endosulfan (2), endosulfan sulphate, endrin, lindane, aldrin, dieldrin, methoxychlor and hexachlorocyclohexanes (alpha; beta; gamma; delta). Except for aldrin, all 16 OCPs targeted in the present study were detected. The OCP residue levels ranged from below detection limit (BDL) to 7.44 ± 0.66 μg/L, BDL to 6.39 ± 1.10 μg/kg ww and BDL to 319.74 ± 66.94 μg/kg ww in water, sediment and fish samples respectively. Some OCP concentrations exhibited significant differences across the sampling sites ($p < .05$). The residue level of endosulfan was above the European Union standard, whereas the target hazard quotient of heptachlor epoxide was above 1, therefore being unsafe for human consumption. The present study results indicate that most of organochlorine pesticides though banned in Kenya are still detected in the environment, posing potential long-term health hazards to humans. Accordingly, environmental monitoring programme and mitigation strategies of reducing pollutant inputs into the lake is recommended, as well as an immediate ban on harvesting and consumption of fish from Lake Nakuru.

KEYWORDS
aquatic pollution, fishery safety, Lake Nakuru, *Oreochromis niloticus*, organochlorine pesticide residues

1 | INTRODUCTION

According to the world pollution problems report, one-third of agricultural crops are produced with the use of pesticides (Blacksmith Institute, 2012; Gitahi et al., 2002; Lincer et al., 1981). Most pesticides have been used in crop pest and disease vector control, therefore having greatly benefitted mankind globally. However, it is estimated that 4.6 million tons of 500 different types of pesticides are applied on crops annually, but that only about 1 percent is effectively utilized (Aktar et al., 2009; Jayaraj et al., 2016; PE GC, 2016). As a result, the biggest percentage of the applied pesticides end up in different environmental compartments, causing pollution and posing an increasing threat to the environment and ecosystems. Pesticides have been listed among the pollutants

requiring more study (Gitahi et al., 2002; Kanangire et al., 2016; Manda et al., 2017). Accordingly, because of the health hazards associated with pesticide pollution, the United Nations Food and Agricultural Organization (FAO), in collaboration with the World Health Organization, developed permissible limits of pesticide residues in food, drinking water, export products such as fish, horticultural products and fruits (FAO/WHO, 1994).

Agriculture accounts for about 24% of Kenya's GDP, with an estimated 70% of its rural population working either directly or indirectly in the sector (Birch, 2018; Nyaundi et al., 2019). The sector is highly dependent on the use of pesticides, fertilizers and acaricides to sustain productivity (Abong'o et al., 2014; Musa et al., 2011; Saoke, 2005). As of 2017, the pesticide application rate in Kenya was estimated to be about 0.25 kg/ha, being third after South Africa (2.16 kg/ha) and Zimbabwe (0.53 kg/ha) in sub-Saharan Africa (Roser, 2019). Pesticide use in Kenya commenced in 1946 with the prominent use of DDT (Dichlorodiphenyltrichloroethane) for mosquito control, especially in the Lake Victoria basin (Abong'o, 2009; Getenga et al., 2004). The undesirable features of organochlorine pesticides have since led many countries around the world, both developed and developing, to ban them. The United States of America (USA), for example, banned their use in 1972, with more countries following thereafter (Madadi, 2017; Teklit, 2016). Kenya banned and/or restricted the use of hexachlorocyclohexane isomers, DDT, heptachlor and endrin in 1986, aldrin and dieldrin in 2004, while lindane was restricted for use only in seed dressing in 2004 (PCPB, 2008).

Organochlorine pesticide pollution in Kenyan aquatic environments has been observed by a number of researchers (Gitahi et al., 2002; Madadi, 2017; Nzeve et al., 2018; Tenai et al., 2016). These studies confirm pollutants are of concern in many Kenyan aquatic ecosystems, requiring monitoring at least from time to time. Despite their ban, organochlorine pesticide residues in sediments, soil, water and biota have continually been reported by various researchers (Abong'o et al., 2015; Olutona et al., 2014; Omwenga et al., 2016). These different researchers attributed the continuous detection of pesticide residues either to its illegal use or its persistence in the environment since the time they were banned (Getenga et al., 2004; Lalah et al., 2003).

No studies have yet been undertaken on the status of pesticide pollution in the different lake matrices of Lake Nakuru for the past 15 years, meriting a need for continuous monitoring of these pollutants to establish their levels in the environment and to undertake necessary actions. Further, there was a need to undertake a study of the safety of harvested fish from Lake Nakuru regarding organochlorine pesticide (OCP) residues that was subsequently consumed or supplied to local markets, as a means of determining whether or not their levels were within the maximum permissible limits deemed safe for human consumption. Accordingly, the main objective of the present study was to determine the content of selected OCP residues (DDT and its metabolites, heptachlor, heptachlor epoxide, α-endosulfan (1), β-endosulfan (2), endosulfan sulphate, endrin, lindane, aldrin, dieldrin, methoxychlor, hexachlorocyclohexanes [alpha, beta, gamma, delta]) in water, sediments and Nile tilapia (*Oreochromis niloticus*) of Lake Nakuru, which comprise the most abundant species of fish in landed catches.

2 | MATERIALS AND METHODS

2.1 | Study site

Lake Nakuru is a shallow endorheic lake located in the Eastern arm of the Rift Valley. It is a regular-shaped basin with gentle slopes and a nearly flat bottom (Leichtfried & Shivoga, 1995). It is located in Nakuru County, south of the town of Nakuru, approximately 157 km from the Kenyan capital of Nairobi. The lake has a surface area of 70 m^2 and a total catchment area of 1800 m^2 (Figure 1). It lies at an altitude of 1759-m above sea level, at latitude 0°23′S and longitude 36°7′E within Lake Nakuru National Park (Iradukunda et al., 2020). The lake catchment is bordered by Menengai crater in the North. Between the lake and the crater is the town of Nakuru, Bahati highlands in the Northeast, Mau escarpment in the West, Eburru crater to the South and grasslands between Nakuru and Elementeita in the East (Odada et al., 2004). Prior to its recent limnological changes, Lake Nakuru was famous for providing habitat to one of the largest population of lesser flamingos.

2.2 | Water and sediment sampling

A total of nine composite water and nine sediment samples were collected from the Njoro River mouth, the sewage discharge point and a Mid-lake point, following APHA (2012) Standard Methods. Two and a half litres of water sample was collected in amber glass bottles from each sampling site, being labelled and treated with 1 g of mercuric chloride to dehydrate microorganisms that could degrade the pesticides. Following the International Atomic Energy Agency (IAEA, 2003) standard methods, approximately 300-g of a sediment sample was scooped from each sampling site using a stainless Ekman grab sampler and packed in labelled sterile plastic containers. The water and sediment samples were stored on ice in the field and during transportation to the Analytical Chemistry and Food Safety Laboratory at the Directorate of Veterinary Services (DVS), Kabete for temporary storage before analysis at the end of the entire sampling period.

2.3 | Fish sampling

A total of 30 Nile tilapia (*O. niloticus*) specimens were collected with the assistance of the Kenya Marine and Fisheries Research Institute (KMFRI) research team of Naivasha by setting gill nets (2.5-in. mesh size) around sunset, retrieving them the following morning from the Lake Nakuru sampling sites. Ten fish samples from the Njoro River mouth, eleven from Fisher's Point, five from the sewage discharge point and four from Nyati. The fish were placed in a cool box and

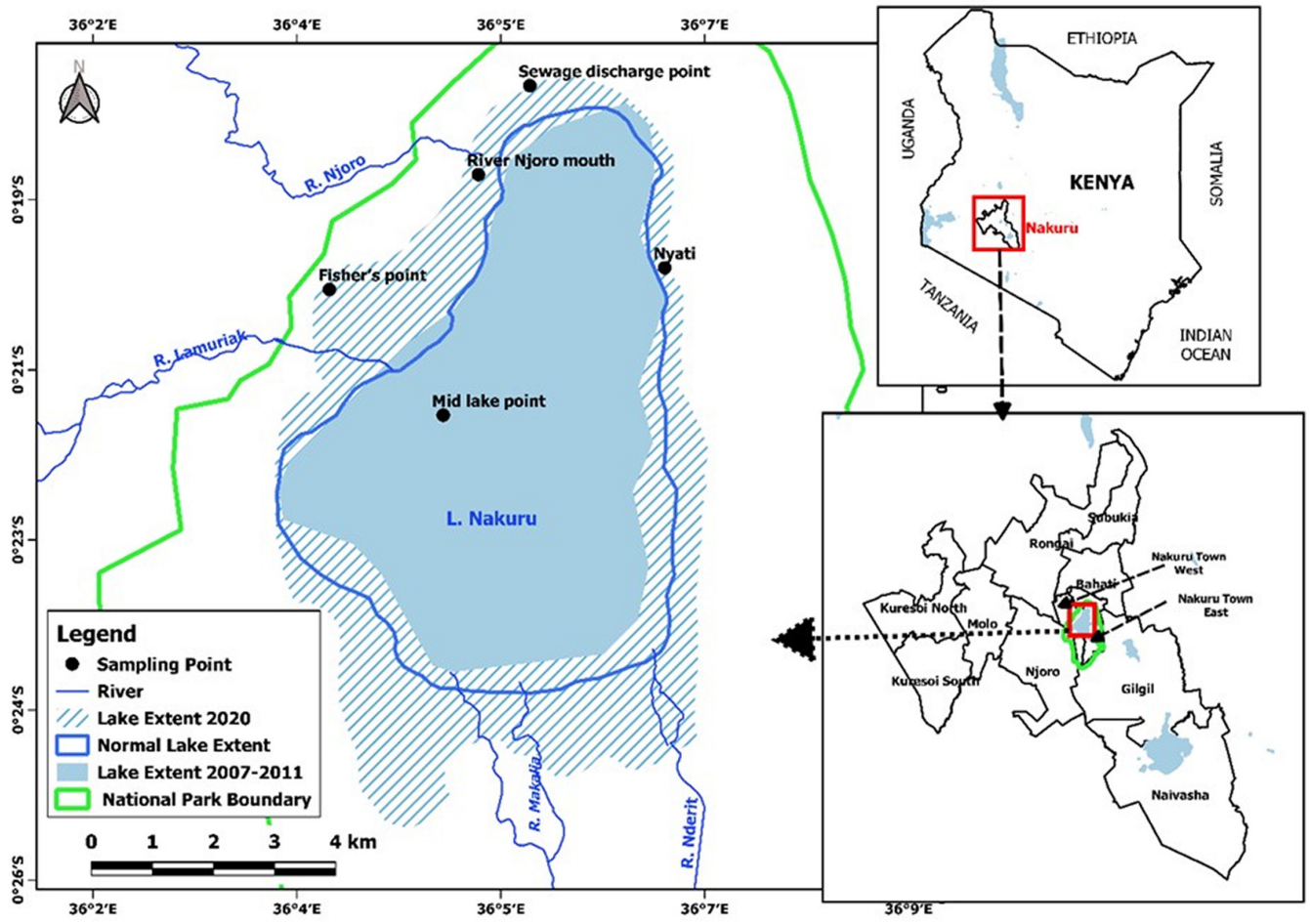

FIGURE 1 A map of Lake Nakuru showing the location of sampling points.

transported to the Department of Biological Sciences, Egerton University for tissue extraction. The fish skin was removed from the dorsal part in the laboratory and muscle tissue was extracted with a ceramic knife. The extracted muscle tissue was placed inside plastic vials and transported to the Analytical Chemistry and Food Safety Laboratory, DVS, Kabete, Kenya and stored at a temperature of −20°C prior to OCP residue analysis.

2.4 | Sample extraction and clean up

Water samples were analysed following the EN 15662 Method. A total of 250 ml of water sample was transferred into a 1-L separatory funnel, followed by the addition of 50-mls of dichloromethane and 5-g of anhydrous sodium sulphate to aid in salting out the organochlorine pesticides (OCPs) from the aqueous phase. The solution was shaken vigorously for 2 min (with venting intervals) and the extracts being collected. The extraction process was repeated using 50 ml of triply distilled dichloromethane (DCM). The combined extracts were eluted into conical flasks followed by drying, using anhydrous sodium sulphate. The extracts were concentrated in the LABCONCO rotary evaporator at a temperature of 70°C. Twenty-five millilitres of isooctane was added into the concentrated sample and sonicated for proper mixing. The mixture was filtered into a glass vial ready for GC-MC analysis.

The sediment samples were thawed for about 6 h before extraction, following the EN 15662 Method. Fifteen g of homogenized sediment sample was weighed into a 50-ml centrifuge tube, with 15 ml of an extraction solvent in a 1:99 ratio of acetic acid and acetonitrile added to the sample. The sample was capped and shaken for 1 min, after which 10-g of an extraction salt (sodium acetate) was added, followed by vigorous shaking for 1 min, and then centrifuged at 1957 *g* for 5 min at 4°C. A 6-ml aliquot of upper acetonitrile layer was transferred into a 15-ml centrifuge tube containing a clean-up salt (150-mg $MgSO_4$, 25-mg PSA and 25-mg C18E). The mixture was capped and shaken for 1 min, after which it was centrifuged at 1957 *g* for 5 min. One millilitre of the extract was transferred into another tube and dried with a nitrogen flow at 40°C. This was followed by re-dissolving the filtrate with 1 ml of isooctane and filtering it through a nylon syringe filter (0.22-μm size). The filtrate was then transferred into a 10-ml glass vial with cap and stored in the refrigerator at a temperature of 4°C prior to GC–MS analysis.

The fish samples were thawed for about 6 h before extraction. Extraction was done following the EN 15662 Method. Ten g of

homogenized fish sample was weighed into a 50 ml centrifuge tube, 10 ml of an extraction solvent (acetonitrile) being added. The sample was capped and shaken for 1 min, after which an extraction salt (6 g of $MgSO_4$; 1.5-g NaOAc) was added, and the mixture was shaken vigorously for 1 min, followed by centrifuging it at 1957 x g for 5 min at 4°C. A 6 ml aliquot of the upper acetonitrile layer was transferred into a 15-ml centrifuge tube containing 1200 mg $MgSO_4$, 400 mg PSA, 400 mg C18E. The mixture was capped and shaken for 1 min, followed by centrifuging it at 1957 *g* for 5 min. One millilitre of the extract was transferred into another tube containing a clean-up salt and dried via nitrogen flow at 40°C, followed by redissolving it with 1 ml of isooctane and filtering it through a nylon syringe filter (0.22 µm size). The filtrate was then transferred into a 10 ml glass vial with cap and stored in the refrigerator at a temperature of 4°C prior to GC–MS analysis.

2.5 | Quantification and analysis of samples

A Shimadzu Gas Chromatography Mass Spectrophotometer (GC–MS/MS) TQ8040 was used to analyse the OCPs in the water, sediment and fish samples. The injection port temperature was maintained at 250°C. Nitrogen was used as the carrier gas and make-up gas at a constant flow rate of 30 ml/min. The sample injection volume was 1 µl with a pulsed split less injection mode. Standard calibration curves were prepared from pesticide standard stock solutions containing the OCPs. Sample analysis was carried out by injecting a 1 µl sample size into the GC in split less mode. The resulting chromatograms were analysed for OCPs following an external standard method. Retention times of sample peaks were matched with those of the reference standards to identify the specific congeners, while standard calibration curves were used for quantitative analysis. The column was a high-performance capillary column, HP5 (5% phenyl methyl siloxane) with dimensions of 30 m length, 0.25 mm internal diameter and 0.25 µm film thickness. The temperature limit was −60°C to 325/350°C.

Quality control and quality assurance were ensured by having several blanks, replicate extractions and analyses for all samples, together with the analysis of reference materials (WDNR, 1996). Field blanks and method blanks were also incorporated to check contamination during sampling, transportation and laboratory preparation procedures (UNEP, 1993). Recovery tests were carried out using the reference pesticide standards to determine the methodology performance (EC, 2000). Pure distilled water samples were also incorporated as blanks and used together with external standards to determine the detection limit of each OCP being investigated.

2.6 | Health risk assessment

The obtained OCP concentrations in the analysed samples were compared with the recommended reference standards of the World Health Organization and European Union deemed safe for public health. Target Hazard Quotients (THQ) for OCP concentrations were conducted, following the USEPA (2012) procedure to evaluate the human health risk of consuming OCP-contaminated fish. The THQ was defined as the ratio between the possible exposure to a substance and the reference dose. It was computed according to the USEPA (2012), as follows:

$$\mathrm{THQ} = \frac{\mathrm{EFr} \times \mathrm{EDr} \times \mathrm{IRFa} \times \mathrm{C}}{\mathrm{RfDo} \times \mathrm{BW} \times \mathrm{AT}} \quad (1)$$

where THQ > 1 = indicates risk for non-carcinogens; EFr = exposure frequency (350 days/year); EDr = exposure duration (30 years) because some of the adverse effects are experienced only after a prolonged exposure to OCPs; IRFa = fish consumption per day (0.0123 kg/day) because per capita fish consumption is 4.5 kg/year in Kenya (KMFRI, 2017); C = pollutant concentration in edible part of fish (mg/kg wet weight [ww]); RfDo = reference dose, oral (mg/kg per day), according to the updated 2017 Regional Screening Level (RSL) in the fish ingestion table (USEPA, 2017); BWa = body weight of an adult male (63.9-kg) and female (61.8-kg) for Kenya (WorldData, n.d.); and AT = averaging time for non-carcinogens (365 days/year).

2.7 | Data analysis

All collected data were stored in Microsoft Excel. Statistical tests were done using the R statistical software version 3.6.3. The OCP concentrations in the analysed samples were presented as means with standard deviation (mean ± standard deviation). One-way analysis of variance was used to test the differences in mean concentrations of the selected pollutants in the different lake matrices (water, sediments and fish) across sampling points. All statistical tests were performed at a significance level (α) of 0.05.

3 | RESULTS

3.1 | OCPs recoveries and limits of detection

The mean recoveries for the 16 OCPs ranged from 80% to 108%, being within the acceptable range of 70% and 110%. Accordingly, the results were not corrected (EC, 2000). The recoveries for the pesticide residues and limits of detection for the OCPs in water, sediments and fish samples analysed are summarized in Table 1.

3.2 | Organochlorine pesticide concentrations in water

With the exception of aldrin, all 16 targeted OCPs in the present study were detected in all the examined water samples (Figure 2). The cyclodienes, including dieldrin, endrin, heptachlor, methoxychlor and endosulfan exhibited the highest concentrations, followed by DDT and its metabolites., while the hexachlorocyclohexane

TABLE 1 Percentage recoveries of pesticide residues and limits of detection in water, sediment and fish samples (mean ± SD; $n = 3$)

Organochlorine pesticide	Percentage recoveries			LOD (µg/L)
	Water	Sediment	Fish	
Alpha-HCH	107.92 ± 2.21	100.98 ± 1.12	92.84 ± 1.03	0.0011
Beta-HCH	100.72 ± 1.09	102.98 ± 3.17	98.87 ± 1.06	0.0016
Gamma-HCH	108.67 ± 0.56	93.00 ± 1.36	90.20 ± 1.01	0.0016
Delta-HCH	100.86 ± 2.01	90.51 ± 2.13	81.75 ± 2.19	0.0014
p,p′-DDD	102.29 ± 4.25	87.50 ± 2.70	84.92 ± 1.21	0.0016
p,p′-DDE	102.97 ± 1.11	94.07 ± 2.22	90.25 ± 0.87	0.0018
p,p′-DDT	104.83 ± 3.32	102.05 ± 3.44	100.31 ± 1.05	0.0017
Endosulfan 1	92.58 ± 4.31	88.84 ± 1.60	98.81 ± 2.03	0.0011
Endosulfan 2	86.75 ± 3.22	92.58 ± 5.65	80.86 ± 1.63	0.0015
Endosulfan sulphate	95.28 ± 2.25	98.03 ± 1.00	83.38 ± 2.48	0.0021
Aldrin	105.62 ± 3.05	96.89 ± 4.11	83.72 ± 1.54	0.0036
Endrin	103.09 ± 2.64	88.01 ± 2.96	85.68 ± 2.08	0.0022
Dieldrin	101.96 ± 2.96	97.55 ± 3.77	88.04 ± 1.08	0.0031
Heptachlor	94.29 ± 4.12	83.70 ± 3.55	97.67 ± 2.96	0.0011
Heptachlor epoxide	81.22 ± 2.41	89.06 ± 4.58	80.75 ± 1.11	0.0011
Methoxychlor	86.17 ± 3.16	84.23 ± 3.49	80.77 ± 2.06	0.0016

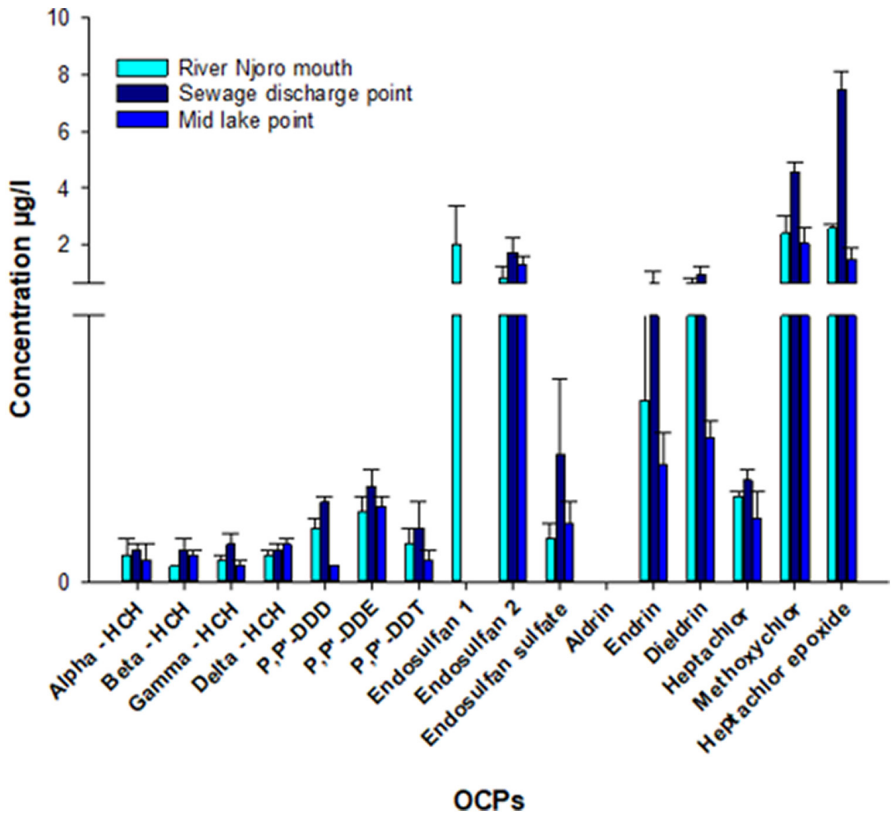

FIGURE 2 Average concentrations of organochlorine pesticides in water.

(HCH) isomers exhibited the lowest levels in the water samples. Although the HCH isomer concentrations were low, the frequencies of their detection in all analysed water samples were very high (100% for alpha & beta HCH; 77% gamma-HCH; 67% for delta-HCH) (Table A1). There were no significant differences in the mean concentration of some organochlorine residues in water samples across the sampling sites ($p > .05$), whereas significant differences were observed for delta-HCH ([one-way ANOVA, $F_{2,6} = 9.15$, $p = .015$], DDD [$F_{2,6} = 52.31$, $p = .0002$], DDT [$F_{2,6} = 14.2$, $p = .005$], endosulfan 1 [$F_{2,6} = 7.36$, $p = .02$] and heptachlor epoxide [$F_{2,6} = 78.89$, $p = 4.92 \times 10^{-5}$]). A post hoc evaluation using a Tukey HSD test indicated significant differences in mean residue levels across sampling

sites for some OCPs in water (Table A2). The concentration of almost all OCPs in the water samples was below the World Health Organization (WHO), Environmental Protection Agency (EPA) and Australian guidelines for OCPs in water, except for heptachlor epoxide (Table 2).

3.3 | Organochlorine pesticide concentrations in sediments

All 16 OCPs were detected in all the sediment samples examined in the present study, except for aldrin, although their concentrations were generally low (Figure 3). The OCP residues in sediments were higher than in the water samples, and there were no significant differences in the mean concentration of some organochlorine residues in sediment samples across sites ($p > .05$), whereas significant differences were observed for DDE ([one-way ANOVA, $F_{2,6} = 178.9$, $p = 4.49 \times 10^{-6}$], endosulfan sulphate [$F_{2,6} = 5.28$, $p = .047$] and dieldrin [$F_{2,6} = 25.67$, $p = .00115$]). A post hoc evaluation using a Tukey HSD test indicated significant differences in the mean residue levels across the sampling sites for some OCPs in the sediments (Table A3). The mean OCP residue concentrations observed in the present study were compared with different sediment quality guidelines, with all being less than LEL (lowest effect level in sediment), SEL (severe effect level in sediment), TEC (threshold effect concentration in sediment) and PEL (probable effect level) values of sedimentary rocks. The exception was heptachlor epoxide, whose concentration exceeded TEC and PEL (Table 3).

TABLE 2 Average organochlorine pesticide residue concentrations for water samples and comparison with WHO maximum permissible levels (WHO, 2008), EPA natural potable water limits and Australia water quality standards (IUPAC, 2003; $n = 9$)

Pesticide	Concentration (μg/L)	WHO	EPA	Australia
Alpha HCH	0.05	–	–	–
Beta HCH	0.05	–	–	–
Delta HCH	0.05	–	–	–
Gamma HCH	0.06	20	0.2	0.5
p,p′-DDD	0.09	–	–	–
p,p′-DDE	0.15	–	7	–
p,p′-DDT	0.10	50	200	60
Endosulfan 1	1.75	20	–	–
Endosulfan 2	1.45	20	–	–
Endosulfan sulphate	0.16	–	–	–
Aldrin	ND	10	–	10
Endrin	0.41	2	2	–
Dieldrin	0.47	0.3	–	–
Methoxychlor	2.27	20	40	20
Heptachlor	0.15	0.3	0.4	0.5
Heptachlor epoxide	3.68	0.3	0.2	0.5

Note: (–), value not provided; WHO, EPA and Australian guideline values were converted from mg/L to μg/L for purposes of comparison.

3.4 | Organochlorine pesticide concentrations in fish samples

As was the case with the water and sediment samples, all 16 OCPs were observed in the fish samples, except for aldrin (Figure 4). There were no significant differences in the mean concentrations of some organochlorine residues in fish samples across sampling sites ($p > .05$), whereas significant differences were observed for Alpha-HCH ([one-way ANOVA, $F_{3,26} = 4.27$, $p = .01$], DDT [$F_{3,26} = 13.11$, $p = 2.08 \times 10^{-5}$], endosulfan 2 [$F_{3,26} = 7.10$, $p = .001$] and methoxychlor [$F_{3,26} = 6.50$, $p = .002$]). A post hoc evaluation using a Tukey HSD test did indicate significant differences in the mean residue levels across sampling sites for some OCPs (Table A4).

3.5 | Organochlorine pesticide pollution and safety of Nile tilapia from Lake Nakuru

Some OCP residue concentrations observed in the fish samples were compared to the maximum residue limits (MRLs) provided by different organizations (Table 4). The target hazard quotients for organochlorine pesticides were also computed. Heptachlor epoxide exhibited the highest THQ for both male and female human fish consumers, also being the only OCP with a THQ value exceeding 1, based on USEPA limits (2017). A slightly higher THQ was observed in female than in male human fish consumers for all OCPs (Table 5).

4 | DISCUSSION

4.1 | Organochlorine pesticide concentrations in water

The present study results obtained for OCP concentrations in water could indicate that the cyclodienes have been recently used in the Lake Nakuru catchment. Aldrin was banned for use in Kenya in 2004 (PCPB, 2008). And its absence in the water samples could indicate it was no longer being used in the Lake Nakuru catchment during the sampling period. Although all hexachlorocyclohexane (HCH) isomers were banned in Kenya, they were unfortunately detected in the present study. DDT was banned in Kenya for use in agriculture in 1986, but restricted in disease vector control (Teklit, 2016; Wandiga et al., 2002), stemming from its undesirable environmental effects, an example being detection of its residues in foodstuffs more so of animal origin (Jayaraj et al., 2016; Musa et al., 2011; Saoke, 2005). The slightly high dieldrin levels in the water samples could be attributable by their recent use, rather than to the conversion of aldrin

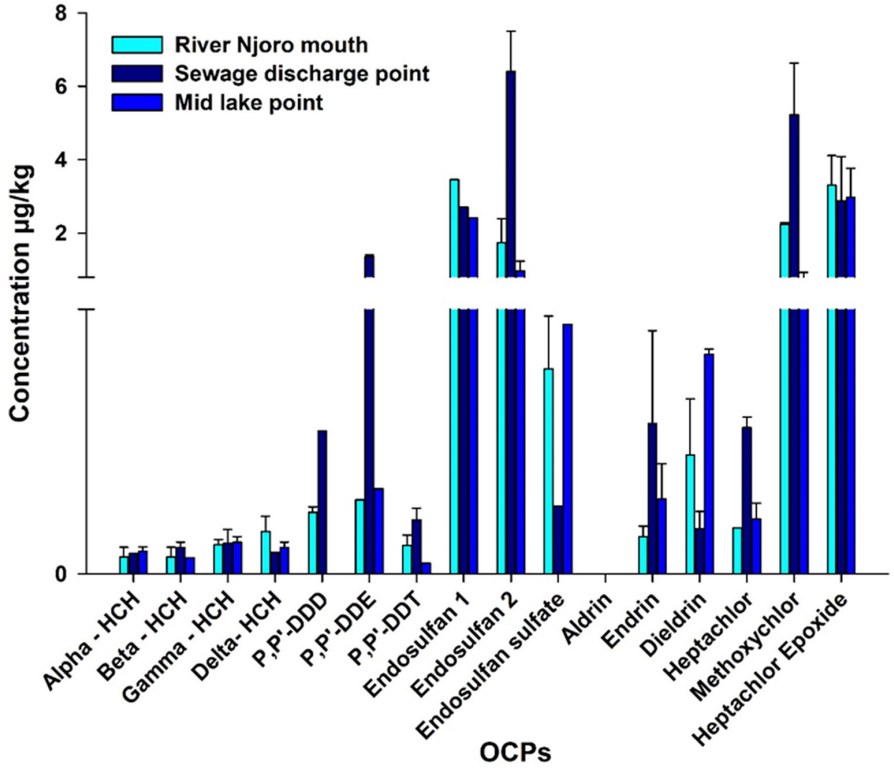

FIGURE 3 Average concentrations of organochlorine pesticides in sediments.

TABLE 3 Concentrations of organochlorine pesticide residues in sediment samples from Lake Nakuru and comparison with different sediment quality guidelines ($n = 9$)

Pesticide	Sediment concentration (µg/kg)	LEL	SEL	TEC	PEL
Alpha HCH	0.04	6	100	–	–
Beta HCH	0.04	5	210	5	–
Delta HCH	0.06	3	120	3	
Gamma HCH	0.06	3	10	2.37	1.38
p,p′-DDD	0.13	8	60	4.88	8.51
p,p′-DDE	0.55	5	190	3.16	6.75
p,p′-DDT	0.09	8	710	4.16	4.77
Endosulfan 1	2.86	–	–	–	–
Endosulfan 2	3.04	–	–	–	–
Endosulfan sulphate	0.33	–	–	–	–
Aldrin	–	2	80	2	
Endrin	0.17	3	1300	2.22	62.4
Dieldrin	0.24	2	910	1.9	6.67
Methoxychlor	2.74	–	–	–	–
Heptachlor	0.16	–	–	–	–
Heptachlor epoxide	3.05	5	50	2.47	2.74

Note: (–), value not provided; sediment quality guidelines are given in µg/kg (Buchman, 2008; Doyle et al., 2003; Persaud et al., 1993).

Abbreviations: LEL, lowest effect level in sediment; PEL, probable effect levels; SEL, severe effect level in sediment; TEC, threshold effect concentration in sediment.

to dieldrin by sunlight and bacteria, based on the observation that aldrin was not detected in any examined samples from the lake (ATSDR, 2002; Mugambi et al., 1989). The detection of banned OCPs in the present study, although in low concentrations, could nevertheless be attributable to its continued illegal use (Lalah et al., 2003; Musa et al., 2011).

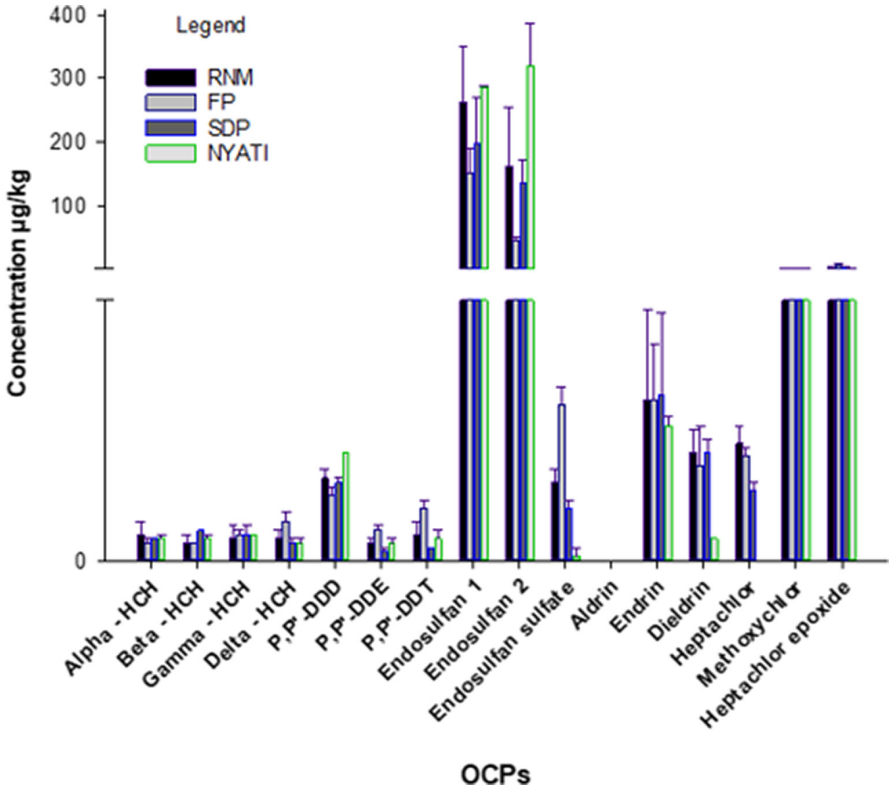

FIGURE 4 Average concentrations of organochlorine pesticides in fish samples (FP, fishers' point; RNM, River Njoro mouth; SDP, sewage discharge point).

TABLE 4 Comparison of organochlorine pesticide residue levels obtained in fish with maximum residue limits (MRLs) reported by different organizations ($n = 30$)

Pesticide	Concentration of OCP in fish (µg/kg ww)	MRLs (µg/kg)	Reference source
Gamma-HCH (Lindane)	0.06	200	CAC (2009)
p,p′-DDE	0.04	5000	FDA (2001)
p,p′-DDT	0.06	5000	FDA (2001)
Endosulfan	168.27	100	EU (2011b, 2011a)
Aldrin	ND	300	FDA (2001)
Endrin	0.33	200	CAC (2009)
Dieldrin	0.20	300	FDA (2001)
Heptachlor	0.22	200	CAC (2009)
Heptachlor epoxide	3.12	200	FAO/WHO (1983)

Note: Maximum residue levels were converted from ppm to µg/kg for comparison purposes.

TABLE 5 Target hazard quotients (THQs) for Lake Nakuru adult male and female human fish consumers for organochlorine pesticide residues

Pesticide	THQ-males	THQ-females
Alpha-HCH	3.41 E-05	3.58 E-05
Beta-HCH	1.41 E-05	2.62 E-05
Gamma-HCH	1.09 E-03	1.15 E-03
p,p′-DDD	2.91 E-05	3.06 E-05
p,p′-DDT	1.64 E-03	1.72 E-03
Endosulfan 1	1.63 E-01	1.71 E-01
Endosulfan 2	1.43 E-01	1.50 E-01
Endosulfan sulphate	1.27 E-04	1.34 E-04
Endrin	6.00 E-03	6.31 E-03
Dieldrin	2.18 E-02	2.29 E-02
Heptachlor	2.40 E-03	2.52 E-03
Methoxychlor	1.33 E-03	1.40 E-03
Heptachlor epoxide	1.31	1.38

Except for the work of Mavura and Wangila (2003), earlier studies of OCPs in Lake Nakuru have not examined their concentrations in the lake water. All their analysed OCP residues in the water samples, including DDT, DDE, DDD, gamma-HCH, heptachlor and aldrin, exhibited higher concentrations compared to the results of the present study. Wandiga et al. (2002) reported higher OCP residue concentrations in water samples (0.50–9.03 µg/L) along the Kenyan Indian Ocean coast, except for endosulfan 1, whose value was lower than that observed in the present study.

The OCP concentrations obtained in the water samples of the present study (BDL – 7.44 µg/L) were generally lower than that reported by Osoro et al. (2016) in water samples of Rusinga Island, Lake Victoria during the short rain season (BDL – 15.53 µg/L). The OCP residue levels of the present study also were higher than those reported by Nyaundi et al. (2019) for the Kuja River catchment in Kenya (BDL – 2.20 µg/L), by Madadi (2017) in water samples from the Kargi area in Marsabit County, Kenya (BDL – 3.37 µg/L) and Ondiere (2016) from the Lake Elementeita drainage basin (BDL

– 0.16 μg/L). The present study results also were comparable to those obtained by Njogu (2011) for Lake Naivasha (0.006–6.76 μg/L). Getenga et al. (2004) observed high OCP levels in water samples (BDL – 1240 μg/L) obtained from the Nyando River drainage system of Lake Victoria.

4.2 | Organochlorine pesticide concentrations in sediments

The observed trend of contaminants in the three lake matrices in the present study is consistent with the findings of Wandiga et al. (2002), who reported the residue concentration trend as fish > sediments > water. Compared to other studies, Mavura and Wangila (2003) observed higher OCP residue levels in Lake Nakuru sediments than those obtained in the present study. Bettinetti et al. (2011) also reported higher residue levels of DDT and its metabolites in sediments of another saline Rift Valley lake (Lake Bogoria) than observed for the present study, although the residue concentration trend of DDE > DDD > DDT was similar to those observed in the current study, variations that might be attributable to sampling season and analytical method differences, noting their studies were conducted in the dry seasons, in contrast to the short rains that occurred during the conduct of the present study. Further, the OCP residue concentrations (BDL – 6.39 μg/kg) observed in the current study were lower than that reported by Wandiga et al. (2002) (0.58–59 μg/kg) in sediment samples from the Kenyan Indian Ocean coast.

In other studies, Osoro et al. (2016) reported higher residue concentrations (BDL – 24.84 μg/kg) during the wet season in Rusinga Island, Lake Victoria, than observed in the current study. Similarly, Wasswa et al. (2011) reported higher OCP residue levels (BDL – 15.96 μg/kg) in sediments in the Ugandan side of Lake Victoria. Lalah et al. (2003) also reported higher sediment concentrations in the Sabaki River, with residue levels of BDL – 108.5 μg/kg observed for 1998 and BDL – 6.93 μg/kg for 1999, with aldrin and lindane contributing significantly to the observed levels.

There was no clear trend of OCP residues among the sampling sites, although sediments from the sewage discharge point and Njoro River mouth exhibited relatively high OCP residues, indicating their significant contribution to the contaminant load to the lake. According to Mavura and Wangila (2003), extensive farming and intense application of pesticides were observed in the Njoro River headwaters region. The slightly higher sediment concentration of p,p′-DDE, compared to p,p′-DDT, could be attributable to the fact that DDE is a degradation product of DDT in the environment (Kanja et al., 1986; Njogu, 2011).

The heptachlor epoxide concentration was high compared to heptachlor, possibly attributable to degradation. It has been reported (Lalah et al., 2003; Madadi, 2017) that heptachlor can be degraded into equal portions of heptachlor epoxide and 1-hydroxy chlordane within 4 weeks. Further, heptachlor has a half-life of approximately 6 months in water and sediment, whereas its derivative breaks down in about three to 7 years (Howard, 1991). Thus, the presence of heptachlor pesticide residue observed in the present study could be attributed to its current use at the time of sampling, as well as bioaccumulation, noting it was banned in Kenya in 1986 (Abong'o et al., 2014; Botaro et al., 2011; PCPB, 2008).

The observed low Lindane (γ-HCH) concentration in sediments could be attributed to its reduced input into the lake following its restricted use to only seed dressing. The relatively high concentrations of endosulfan, endrin, heptachlor and methoxychlor initially used as insecticides could probably be attributed to their continued illegal use in Lake Nakuru catchment (Getenga et al., 2004; Mutuku et al., 2014).

4.3 | Organochlorine pesticide concentrations in fish samples

The trend from highest to lowest of the residue levels in the OCP groups was the same in all the matrices considered (cyclodienes > DDT and its metabolites > hexachlorocyclohexanes), indicating that some, if not all, OCPs in the cyclodiene group are still being used in the Lake Nakuru catchment, while the other groups (hexachlorocyclohexanes; DDT & its metabolites) are phasing out.

Endosulfan 1 and 2 residue levels were generally high in the fish samples, irrespective of their sampling site, when compared to other OCPs. These high residue levels could be attributed to the frequent use of endosulfan in the Lake Nakuru catchment, as well as bioaccumulation effects. Endosulfan is of environmental importance because of its apparent persistence and toxicity to many non-target organisms, including fish (Gitahi et al., 2002; Nyaundi et al., 2019). The heptachlor residue concentration was generally lower than that of heptachlor epoxide, regardless of the sampling site., possibly attributable to the breakdown of heptachlor into heptachlor epoxide by microorganisms. Although heptachlor epoxide is a degradation product, it is as toxic and persistent as its parent compound (Manda et al., 2017; Murty, 1986).

Compared to other studies, the present study indicated lower pp' DDT, pp' DDE and dieldrin levels than previously reported for Tilapia grahami (*Alcalicus grahami*) of Lake Nakuru (Koeman et al., 1972). Further, Kairu (1999) also reported higher alpha-HCH, lindane, beta-HCH and DDE residue levels in Tilapia grahami (*A. grahami*) from the same lake, compared to the present study, although they did not detect aldrin in the fish samples. Further, contrary to the findings of the current study, dieldrin was not detected. However, similar findings of very low residue levels in Tilapia grahami (*A. grahami*) in Lake Nakuru were also reported by Greichus et al. (1978). Elsewhere, Mwevura et al. (2002) reported relatively high OCP frequencies during the wet season, observing frequencies of p,p′-DDT (81%), p,p′-DDE (100%), dieldrin (100%) and γ-HCH (6%) in fish samples from the coastal area of Dar es Salaam, Tanzania.

4.4 | Organochlorine pesticide pollution and safety of Nile tilapia from Lake Nakuru

All OCP residue levels were below the MRLs, except for endosulfan, the latter exceeding the EU acceptable limit. Several studies

have indicated that endosulfan, either alone or in combination with other pesticides, may bind to oestrogen receptors, thereby perturbing the endocrine system (WHO, 2008). Although the calculated THQs for 15 OCP residue levels in all the examined fish were below 1, their long-term impact may be of significance because of their high biomagnification potential through the food chain.

5 | CONCLUSIONS

Organochlorine pesticide residues were detected in low concentrations in all the matrices considered in the present study, except for aldrin, which was not detected. The findings of the current study indicate Nile tilapia taken from Lake Nakuru is not safe for human consumption, being limited by elevated levels of endosulfan and THQs exceeding 1 for an OCP (heptachlor epoxide). Accordingly, it is recommended that policymakers should implement an environmental monitoring programme and mitigation strategies for reducing pollutant inputs into the lake, thereby managing its water quality status, which suggests serious implications to its aquatic life. Further, since the results of the present study indicate Lake Nakuru fish is not safe for human consumption, an immediate ban on fish harvesting and consumption from Lake Nakuru is recommended.

ACKNOWLEDGEMENTS

The authors thank the Orange Knowledge Program (OKP), The Netherlands, for funding this study through a Masters Fellowship awarded to Ms. Mary Florence Nantongo. Special thanks also are due to Egerton University for providing laboratory space, the Joint Limnology and Wetland Management (LWM) programme for providing field equipment and the Kenya Wildlife Service (KWS) for granting unlimited access into the Lake Nakuru National Park that enabled us to reach all the intended study sites. We also thank the laboratory technicians (Ms Sheridan Wanjiku; Mr Shadrack Karanja) for their indispensable assistance with necessary logistics regarding the organochlorine pesticide residue analyses conducted at the Analytical Chemistry and Food Safety Laboratory at Directorate of Veterinary Services, Kabete. Finally, we are indebted to the KMFRI team for assisting us in obtaining fish samples from Lake Nakuru, as well as the National Commission for Science, Technology and Innovation, Kenya, for granting us research clearance permit number NACOSTI/P/20/6476.

FUNDING INFORMATION

This research work was supported by the Orange Knowledge Programme (OKP), the Netherlands.

CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest.

DATA AVAILABILITY STATEMENT

Data are available on request from the corresponding author.

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How to cite this article: Nantongo, M. F., Edebe, J., Otachi, E. O., & Kipkemboi, J. (2023). Organochlorine pesticide residues in water, sediments and Nile tilapia (*Oreochromis niloticus*) of Lake Nakuru, Kenya and implications for its fishery. *Lakes & Reservoirs: Research & Management*, *28*, e12424. https://doi.org/10.1111/lre.12424

APPENDIX 1

TABLE A1 Percentage detection frequency of organochlorine pesticides in water, sediment, and fish samples analysed in present study

OCP	Detection frequency (%)		
	Water ($n = 9$)	Sediment ($n = 9$)	Fish ($n = 30$)
Alpha-HCH	100	66.7	70
Beta-HCH	100	66.7	70
Gamma-HCH	77.8	66.7	76.7
Delta-HCH	66.7	44.4	60
p,p′-DDD	44.4	44.4	40
p,p′-DDE	77.8	77.8	73.3
p,p′-DDT	88.9	55.6	76.7
Endosulfan 1	22.2	33.3	53.3
Endosulfan 2	66.7	66.7	76.7
Endosulfan sulphate	77.8	44.4	43.3
Endrin	100	88.9	86.7
Dieldrin	100	77.8	63.3
Heptachlor	66.7	55.6	40
Heptachlor epoxide	66.7	88.9	86.7
Methoxychlor	100	100	100

TABLE A2 Tukey HSD summary table showing adjusted *p*-values indicating significant differences in OCP residue concentrations in water across sampling sites ($p < .05$)

OCP	Sites showing differences in residue levels	*p*-Values
Delta-HCH	Njoro River mouth and mid lake point	.01
DDD	Njoro River mouth and mid lake	.003
	Njoro River mouth and sewage discharge point	.01
	Sewage discharge point and mid lake	.00
DDT	Njoro River mouth and sewage discharge point	.015
	Sewage discharge point and mid lake	.006
Endosulfan 1	Njoro River mouth and mid lake point	.037
	Njoro mouth and sewage discharge point	.037
Heptachlor epoxide	Njoro River mouth and sewage discharge point	.00
	Sewage discharge point and mid lake	.00

TABLE A3 Tukey HSD summary table showing adjusted *p*-values indicating significant differences in organochlorine pesticides residue concentrations in sediment samples across sampling sites ($p < .05$)

OCP	Sites showing differences in residue levels	*p*-Values
DDE	Njoro River mouth and sewage discharge point	.00
	Sewage discharge point and mid lake	.00
Dieldrin	Njoro River mouth and mid lake	.03
	Njoro River mouth and sewage discharge point	.02
	Sewage discharge point and mid lake	.00
Endosulfan sulphate	Sewage discharge point and mid lake	.04

TABLE A4 Tukey HSD summary table showing adjusted *p*-values indicating significant differences in organochlorine pesticides residue concentrations in fish samples across sites ($p < .05$)

OCP	Sites showing differences in residue levels	*p*-Values
Alpha-HCH	Njoro River mouth and fishers' point	.02
DDT	Nyati and fishers' point	.006
	Njoro River mouth and fishers' point	.00
	Sewage discharge point and fishers' point	.00
Endosulfan 2	Nyati and fishers' point	.002
	Njoro River mouth and fishers' point	.01
Methoxychlor	Nyati and fishers' point	.01
	Sewage discharge point and fishers' point	.01
	Njoro River mouth and nyati	.03