Risk assessment for organochlorines and organophosphates pesticide residues in water and Sediments from lower Nyando/Sondu-Miriu river within Lake Victoria Basin, Kenya

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Abstract

The objective of this study was to survey and document pesticide residue levels in the lower Nyando/Sondu-Miriu catchment areas of Lake Victoria, Kenya, during the dry and rainy seasons of 2009. Water and sediment samples from the Nyando/Sondu-Miriu Basin were analysed for selected pesticide residues, using gas chromatography equipped with Ni^{63} and CP-SIL 8CB-15m and TSD detectors for organochlorine and organophosphorus pesticide residues, respectively. The findings indicated that banned organochlorines are still being used in the catchment. Dieldrin and p,p′-DDD were notably higher ($P < 0.05$) in concentrations than their metabolically formed analogues of aldrin and DDT, respectively. Notably, organophosphorus was below detection levels in water samples, whereas diazinon and malathion were at higher levels in sediment samples. The total residues of DDT, HCH, methoxychlor and endrin generally were below WHO drinking water limits of 2, 2, 20 and 0.01 μg L^{-1}, respectively, whereas aldrin and dieldrin were above the recommended values of 0.03 μg L^{-1}. Agricultural activities in the Lake Victoria Basin are influencing accumulation of the pesticide residues in the basin rivers and the lake. The study recommends creation of buffer zones around the natural water bodies to reduce the inflow of pesticides into water bodies. An integrated pest management approach that encourages reduced usage of chemical compounds also should be encouraged.

Key words

Lake Victoria, organochlorine, organophosphorus, residues.

INTRODUCTION

Lake Victoria is the source of the Nile River, providing domestic, industrial, irrigation and power-generation water to the Nile Basin countries of Kenya, Uganda, Tanzania, Sudan and Egypt. The lake has an estimated area of 68 800 km^2, with about 6% lying in Kenya, 43% in Uganda and 51% in Tanzania. It provides a productive lake fishery for these three East African countries, estimated to be 500 000 metric tonnes annually. The fishery is valued at US$ 600 million, with exports estimated to be US$ 271 million (LVFO 2001). The lake also provides employment in fish-processing factories and provides high protein food (fish) for local consumption. The lake enjoys a major tourist potential, contains a high fish species diversity, is an important research site, and modulates the regional climate. The lake basin has been designated an economic growth zone by the East African Community.

The use of chemical pesticides is still indispensable in the Lake Victoria Basin because of the humid and tropical environmental conditions, which are conducive to the

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development of myriad of pests, weeds and disease vectors. Further, the public health sector in the lake Basin also depends heavily on pesticides to control vector-borne diseases such as malaria, sleeping sickness, bilharzias and filariasis, using pesticide spray programs directed to controlling disease vectors such as mosquitoes, tsetse flies and water snails. Several contaminants from the Lake Victoria catchment, however, may escape from the drainage basin and are likely to compromise the quality of the lake water supporting the fishery industry and that used for domestic human consumption in the Nile Basin Countries. Pesticide use poses a great challenge to the country to develop satisfactory techniques combining optimal agricultural productivity and environmental protection.

Everaarts *et al.* (1996) analysed sediments from the Kenyan coast, finding α-BHC, β-BHC and dieldrin present in concentrations of 7.1–62.2, 7.3–53.2 and 37 ng g^{-1} of organic carbon, respectively. Dieldrin, DDT, β-BHC and heptachlor epoxide have been detected in the breast milk of Kenyan women (Wandiga & Mutere 1988). The main route of exposure to the respondents was attributed to oral intake through vegetables, beef, contaminated water and dairy milk containing the residues (Kanja *et al.* 1988; Barlas 1999; Barasa *et al.* 2007). DDT and other organochlorine pesticides have been shown to affect reproduction of various test animals (Hayes 1982; Mugachia *et al.* 1992; Gonzalez *et al.* 2003; Getenga *et al.* 2004).

Information on pesticide residues in the Lake Victoria catchment, and particularly the Nyando Basin, is currently fragmentary and inadequate. Thus, there is a need for data on the major contaminants in the Lake Victoria drainage system, to properly manage lake water quality, the Nile River, and sustainability of the Nile Basin ecosystem. To this end, the objective of this study was to determine contaminant levels, mainly organochlorines and organophosphorus, in the Nyando/Sondu-Miriu Basin and their respective catchment. The findings of this study were intended to provide vital information needed to develop informed catchment management plans to mitigate the impacts of micropollutants to aquatic organisms and humans in the Lake Victoria Basin.

MATERIALS AND METHODS

Pesticide use survey

An inventory of the types of pesticides used in the Nyando/Sondu-Miriu catchment was conducted with a structured questionnaire observation, and Focused Group Discussions (FGD) among large-scale and small-scale farmers, and stockist shops. More information was gathered from the offices of the Ministry of Agriculture, Livestock and Ministry of Health.

Field sampling

Sampling was conducted at seven selected sites at the Nyando/Sondu-Miriu Basin (Fig. 1) between January and March (Dry season) and October and December (Rainy season), 2009. The sampling focused on locations influenced by activities in nearby urban areas, agricultural areas and river mouths. Water samples were collected by a grab method into 2.5 L amber bottles, making several composite samples. The sediment was sampled with a pre-cleaned Eckman grab sampler and packed in a black polythene bag. The samples were kept on ice during the sampling trip, and later stored in a refrigerator at −4 °C after the sampling trip, and prior to extraction. Field sampling was conducted three times during the rainy season (October, November and December) and three times during the dry season (January, February and March).

Reagents

All solvents (n-hexane; acetone; dichloromethane; diethyl ether), as well as other analytical reagents used (anhydrous sodium chloride; Florisil), were of analytical grade quality, obtained from Dr EHRENSTORFER GmbH, (Augsburg, Germany) and double distilled to ensure purity. Florisil and individual pesticide standards were available at the Kenya Plant Health Inspectorate Services (KEPHIS) laboratories in Nairobi.

Sample extraction and clean-up

Sample extraction and clean-up were based on previously described protocols (NRI 1991). Two litres of each water sample was transferred into a separatory funnel, and the pH was measured. A 50 mL portion of 0.2 mol L^{-1} disodium hydrogen phosphate buffer was added to the sample, and the pH was adjusted to 7, utilizing drops of 0.1 N sodium hydroxide and HCl solutions. The neutralized sample was treated with 100 g sodium chloride to salt out the pesticides from the aqueous phase. A 60 mL aliquot of triple-distilled dichloromethane was added, and the sample was shaken for 2 min while releasing the pressure. The sample was allowed to settle for 30 min to enhance separation of the phases. The organic layer was collected in 250 mL Erlenmeyer flasks and stored at 4 °C in a refrigerator. The extractions were repeated twice, using 60 mL portions of dichloromethane. The organic phase was combined and cleaned by passing through a Florisil column topped with anhydrous sodium sulphate. Pesticide residues were sequentially eluted with 200 mL of 6, 15 and then 50% diethyl ether in hexane. The elutes

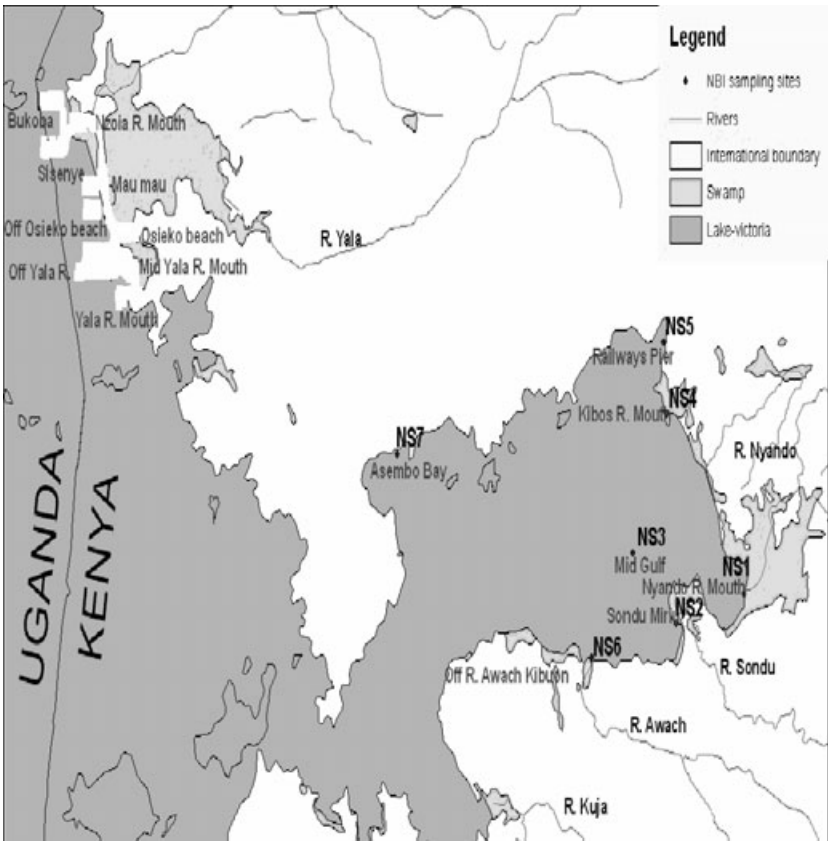

Fig. 1. Map of Lake Victoria catchment showing sampling sites in Kenyan part of lake (NS, sites in Nyando/Sondu-Miriu Basin).

were combined and concentrated to near dryness, using a rotary evaporator at 60 °C, and reconstituted in 5 mL HPLC hexane for GC analyses.

The sediment samples were allowed to thaw for 4 h in the laboratory prior to mixing. Triplicates of 25 g sediment samples were placed in 150 mL teflon vials, treated with 3.5 mL of 0.2 mol L^{-1} ammonium chloride and allowed to settle for 15 min. A 50 mL aliquot of a 1:1 mixture of hexane/acetone was added to each sample vial and extracted for 12 h on an orbital shaker (model SO1), whereas separation was achieved with centrifugation at 62.72 g for 30 min. The extracts were decanted into 250 mL Erlenmeyer flasks, covered with aluminium foil and kept in a refrigerator at 4 °C. The sediments were re-extracted twice with 15 mL of a 1:1 hexane/acetone mixture. The extracts were cleaned by passing through a Florisil column. The clean extracts were concentrated on a rotor evaporator to near dryness and reconstituted into 5 mL portions of HPLC grade hexane (NRI 1991).

Chromatographic analyses

Analysis of organochlorine pesticide residues was carried out with a Varian Chrompack CP-3800 GC at the Department of Chemistry, Masinde Muliro University, Kenya. The GC was equipped with Ni^{63} ECD and CP-SIL 8CB-15m, 0.25 mm (ID) and 0.25 μm film. The column temperature was programmed at 150 °C (1 min), changed at 4 °C min^{-1} to 200 °C (0 min) and 4.5 °C min^{-1} to 300 °C. The injector temperature was maintained at 250 and 300 °C for the detector, whereas a flow pressure of 30 psi of nitrogen gas was utilized as the carrier gas. A sample size of 1 μL, and a split ratio of 1:20, was used for all the samples, whereas the data processing was carried out using Star version 5.4; 5.5 (OAGi 9.2 and UNCEFACT Core Components, STAR organization 2007).

Organophosphorus pesticide residues in the field sample were analysed with a GC Varian Chrompack 3400 at the Department of Chemistry, Masinde Muliro University, Kenya. The instrument was equipped with TSD detector and column type of OV-1701 (30 m × 0.32 mm × 0.5 μm). The temperature was programmed at 90 °C (1 min), changed at 30 °C (0 min) and 4 °C min^{-1} to 250 °C. The injector temperature was set at 250 °C, while the detector temperature was at 300 °C. The carrier (N_2) gas flow rate of 5 mL min^{-1}, make up (H_2) at 25 mL $min^{-1,}$ and an airflow rate of 175 mL min^{-1} were applied. The sample analysis was carried out by injecting a 2 μL sample into the GC. Identification and quantification

were accomplished with an external standards method, whereas processing was made with a Star workstation version 5.5. External standard calibration was used to determine peak areas from the samples.

Quality control and quality assurance procedures included replicate sampling, extraction and analysis for all the samples. Extraction of the water samples also incorporated studies of spiked samples to determine the recovery rate of the utilized method. Pure distilled water samples were also incorporated as blanks. Together with external standards, these samples were used to determine the detection limit for each investigated pesticide. High recovery rates were obtained with the solvent–solvent extraction method. The average recovery rates for the analysed pesticides were 95.55% for α-HCH, 94.23% for β-HCH, 96.62% for γ-HCH, 97.53% for p,p′-DDT, 97.21% for o,p′-DDE, 98.32% for p,p-DDD, 88.62% for aldrin and 97.23% for dieldrin. These values represented good recoveries in relation to the recommended rate ranging between 70 and 120%.

The data obtained in this study were recorded in Microsoft EXCEL (Microsoft Corporation Mac OS X 2011, Spreadsheet Version 12) spreadsheets and subjected to analyses in STATISTICA (Statsoft Inc., 2010, version 8.0). Lack of significant variations for pesticide residues within stations ($P > 0.05$) resulted in pooling of data at a basin level. The pesticide ranges and means (±SE) were obtained for comparison to WHO/-NEMA recommended guidelines. Variations among pesticides and seasons were made with two-way ANOVA at $P < 0.05$.

RESULTS

Organochlorine residues. Some of the tested organochlorines were detected in water and sediment samples in varying concentrations both during the rainy and dry seasons. Organochlorine pesticide residues detected in water samples collected in the Nyando/Sondu-Miriu Basin during the rainy season ranged between 0.01 and 0.31 μg L^{-1}, being below the detection limit (BDL) – 0.14 μg L^{-1} during the dry season (Table 2). Residue levels of organochlorine pesticides detected in the sediments collected during the rainy season ranged between 0.03 and 69.55 μg kg^{-1} and 0.02–23.12 μg kg^{-1} in the sediments collected during the dry season (Table 2). Dieldrin constituted the highest residue ($P < 0.05$) detected during the rainy season in both water and sediment samples (Table 2). The dieldrin and p,p′-DDD concentrations were notably higher ($P < 0.05$) than those for aldrin and p,p′DDT, respectively, in most (98%) of the samples. The residue levels of aldrin dieldrin, heptachlor and heptachlor epoxide were both above the WHO/NEMA limits in drinking water of 0.03, 0.03, 0.03 and 0.02 μg L^{-1}, respectively, during the rainy season, whereas aldrin and dieldrin were above the WHO/NEMA-recommended values during the dry season (Table 2).

Organophosphorus residues: No organophosphorus pesticide residues were detected in water samples during either the rainy or dry season (Table 3). The residence concentrations detected in sediment samples collected during the rainy season ranged between BDL – 1.08 μg kg^{-1} and BDL – 0.56 μg kg^{-1} during the dry season (Table 3). Diazinon and dimethoate constituted the highest residues detected in sediment during the rainy and dry seasons, being below the NEMA limits in drinking water of 0.03 μg L^{-1} (Table 3).

Seasonal variations of pesticide residues

Analysis of seasonal variations of the residue levels across the two seasons indicated that samples collected from the Nyando/Sondu-Miriu Basin during the rainy season contained the highest ($P < 0.05$) quantity of pesticide residues in both the water and sediment samples, compared with the dry season (Figs 2,3, Table 4). The exception was water samples that did not contain any detectable levels of organophosphorus during either the rainy or dry season.

DISCUSSION

The most commonly used pesticides in the Nyando/Sondu-Miriu catchment were organophosphates, organochlorine, with very little amidine. The majority of the pesticides used are not registered in Kenya by the Pest Control Products Board (Pest Control Board 1992), meaning that these pesticides are considered relatively unsafe to human health and the environment.

All the organochlorine compounds analysed in this study were banned in Kenya in 1986 except for aldrin and dieldrin, whose use is restricted to controlling termites in the building industry (PCPB 1992). The present study, however, revealed high concentrations of these compounds in water and sediment samples in the Nyando/Sondu-Miriu Basin. Their continued use in the agricultural sector, although illegal in regard to Kenyan government policy, has been attributed to their availability at low costs, their low mammalian toxicity and their broad spectrum bioactivity over a long duration (Munga 1985; Kahunyo *et al.* 1986; Wandiga *et al.* 2002). Pesticide residues of hexachlorobenzene (HCB) in water

Table 1. Most commonly used pesticides in the Nyando catchment area during the study period

Agricultural farms			Small-scale shops in catchments		
Family	Trade name	Active ingredient	Family	Name	Active ingredient
Organophosphates	Diagran	Diazinon	Organophosphates	Diazole	Diazinon
	Basudin	Diazinon		Fenom	Diazinnon
	Diazole	Diazinon		Attain	Malathion
	Fenom	Diazinnon		Skanar Super	Malathion
	Perfekthion	Dimethoate		Roundup	Glyphosate Acid
	Attain	Malathion		Actelic Super	Primiphos-methyl
	Lysol	Malathion		Velpar	Hexazinone
	Roundup	Glyphosate Acid		Kelthane	Pirimiphos-methyl
	Actelic Super	Primiphos-methyl		Hostathion	Triazophos
	Skanar Super	Malathion		Folimat	Omethoate
	Kelthane	Pirimiphos-methyl		Nemacur	Fenamiphos
	Velpar	Hexazinone		Orthene	Acephate
	Hostathion	Triazophos		Gesapak Combi	Ametryn + Atrazine
	Folimat	Omethoate		Lysol	Malathion
	Nemacur	Fenamiphos	Organochlorines	Alanex	Alachlor
	Miratex	Bromadiolone		Pentac	Dienochlor
	Gesapak Combi	Ametryn + Atrazine		Akarint	Dicofol
	Dual gold	Pirimiphos-methyl		Mitigan	Dicofol
Organochlorines	Alanex	Alachlor		Chlortox	Chlordane
	Gamma BHC Lindane	Delta-HCH		Velsicol	Heptachlor
	Akarint	Dicofol		Nendrin	Endrin
	Pentac	Dienochlor		Murtano	Lindane
	Chlortox	Chlordane		Novadrin	Aldrin
	Velsicol	Heptachlor	Pyrethroids	Karate	Lambdacyhalothrin
	Novadrin	Aldrin		Brigade	Bifenthrin
	Murtano	Lindane		Bulldock	Beta-cyfluthrin
	Nendrin	Endrin		Folicur	Sulphur
Pyrethroids	Karate	Lambda-Cyhalothrin		Krismat	Lambdacyhalothrin
	Krismat	Lambdacyhalothrin	Amidine	Taktic	Amitraz
	Folicur	Sulphur		Triatix	Amitraz
	Bulldock	Niclosamide		Almatix	Amitraz
Amidine	Triatix	Amitraz		Dieldrex	Dieldrin
	Taktik	Amitraz	Triazoles	Sencor	Metribuzin
	Almatix	Amitraz			
	Dieldrex	Dieldrin			
Botanicals	Vectocid	Deltamethrin			

sampled from Nyando/Sondu-Miriu Basin were found to be below 2.0 μg L^{-1}. According to the WHO (1984) guideline for drinking water, the minimum contaminated level (MCL's) for γ-isomers of HCH (Lindane) is 4 μg L^{-1}. The WHO (1984) Minimal Risk Level (MRL) for DDT is 1.0 μg L^{-1}, and the DDT level detected in this study was below this limit. Thus, no unacceptable risks were found from DDT contamination in the Nyando/Sondu Miriu Basin. Aldrin and dieldrin, however, were slightly above the recommended values. The dieldrin and p,p′-DDD concentrations were notably higher than those of aldrin and p,p′DDT, respectively, in most (98%) of the samples. As the later are the degradation products of the former, this finding indicated possible transformation processes taking place for p,p′-DDT and aldrin previously used in the region.

Table 2. Mean concentration (±SE) of organochlorine pesticide residues in water and sediment samples from Nyando/Sondu-Miriu Basin during rainy and dry seasons

	Rainy season	Dry season	Rainy season	Dry season	Drinking water pesticide guidelines	
Pesticide	Water (μg L^{-1})	Water (μg L^{-1})	Sediment (μg kg^{-1})	Sediment (μg kg^{-1})	NEMA	WHO
α-HCH	0.03 ± 0.003	0.02 ± 0.006	0.91 ± 0.006	6.59 ± 0.012	—	2
β-HCH	0.01 ± 0.00	0.01 ± 0.000	0.52 ± 0.006	2.33 ± 0.006	—	2
γ-HCH	0.03 ± 0.003	0.03 ± 0.006	3.37 ± 0.012	23.12 ± 0.058	2	2
p,p′-DDT	0.09 ± 0.012	0.08 ± 0.012	1.62 ± 0.012	12.32 ± 0.058	2	2
o,p-DDE	0.09 ± 0.017	0.07 ± 0.006	10.53 ± 0.017	5.15 ± 0.017	—	—
p,p′-DDD	0.18 ± 0.035	0.14 ± 0.012	12.27 ± 0.012	18.91 ± 0.017	—	—
α-Endosulfan	0.13 ± 0.006	0.06 ± 0.006	5.93 ± 0.006	13.45 ± 0.017	0.01	—
Endosulfan sulphate	0.14 ± 0.017	0.07 ± 0.012	13.06 ± 0.023	6.72 ± 0.012	0.01	—
β-Endosulfan	0.20 ± 0.058	BDL	0.03 ± 0.006	0.02 ± 0.006	0.01	—
Aldrin	0.05 ± 0.012*†	0.08 ± 0.006*†	14.00 ± 0.577*†	11.44 ± 0.012*†	0.03	0.03
Dieldrin	0.31 ± 0.006*†	0.14 ± 0.012*†	69.55 ± 0.029*†	11.94 ± 0.029*†	0.03	0.03
Endrin	0.09 ± 0.012	0.04 ± 0.006	7.96 ± 0.017†	10.99 ± 0.058†	NR	0.01
Heptachlor	0.07 ± 0.012*†	0.03 ± 4.910	30.84 ± 0.023*†	16.71 ± 0.098	0.03	0.03
Heptachlor epoxide	0.11 ± 0.006*†	0.02 ± 0.006	6.31 ± 0.006*†	6.41 ± 0.006	0.02	0.03
Methoxychlor	0.14 ± 0.012	0.02 ± 0.006	4.41 ± 0.006	3.78 ± 0.012	20	20

*Above WHO limit guideline; †above NEMA limit guidelines; *†above both NEMA and WHO limit guideline. BDL; below the detection limit.

Table 3. Mean concentration (±SE) of organophosphorus pesticide residues in water and sediment samples from Nyando Basin during rainy and dry seasons

	Water (μg L^{-1})		Sediment (μg kg^{-1})		Drinking water Pesticide guidelines	
Pesticide	Rainy season	Dry season	Rainy season	Dry season	NEMA	WHO
Diazinon	BDL	BDL	1.08 ± 0.02	0.56 ± 0.01	0.03	—
Dimethoate	BDL	BDL	0.29 ± 0.02	0.02 ± 0.00	0.03	—
Malathion	BDL	BDL	BDL	BDL	0.01	—
Fenitrothion	BDL	BDL	BDL	BDL	0.01	—
Ethyl parathion	BDL	BDL	BDL	BDL	NR	—

*Above WHO limit guideline; †above NEMA limit guidelines; *†above both NEMA and WHO limit guidelines; NR, not regulated. BDL, below the detection limit.

The water samples in this study did not contain detectable levels of organophosphorus. Lack of organophosphates residues does not necessarily indicate a lack of impact of organophosphate compounds on the environment. They may still impact aquatic systems in the short term before they degrade to non-toxic products. While the fate of organophosphate compounds in the aquatic system is not clear, however, the most serious concern relating to organophosphates is that they are still suspected to have serious toxicological impacts on terrestrial wildlife and human beings. Pesticide residues in the sediment samples were higher than those detected in the water samples, suggesting that the sediments are pesticide sinks, rarely releasing back to the water column (ATSDR 2002; Gitahi *et al.* 2002; Abuodha & Hecky 2005). High diazinon and malathion levels in the sediment samples were attributed to current use of these compounds, as indicated in the baseline survey (Table 1).

The seasonal changes were observed to influence the pesticide concentrations in the field samples. The pesti-

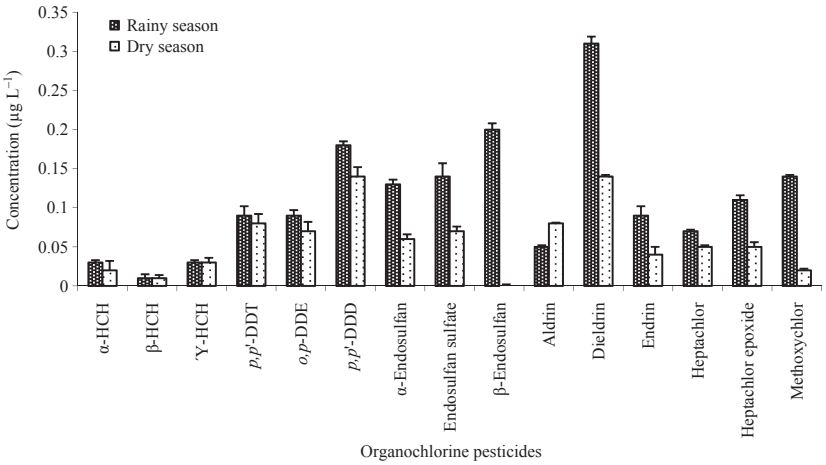

Fig. 2. Seasonal variation in organochlorine pesticide residue concentrations in water from Nyando/Sondu-Miriu Basin during study period.

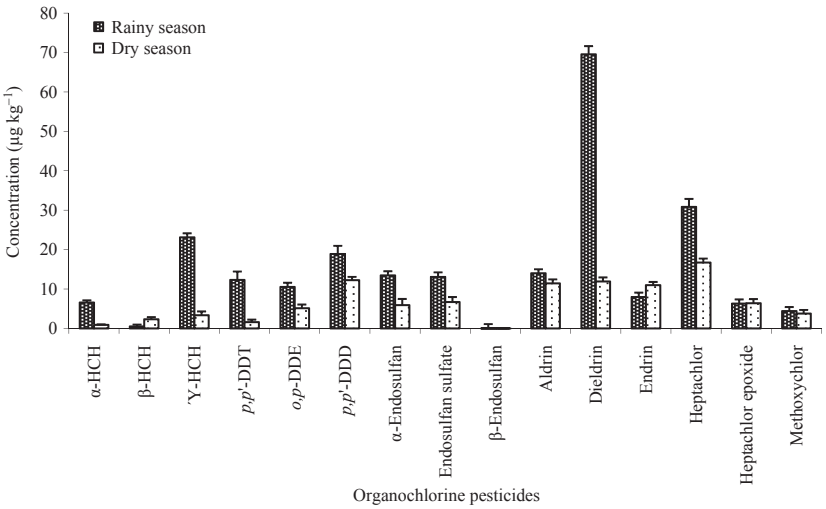

Fig. 3. Seasonal variation in organochlorine pesticide residue concentrations in sediment from Nyando/Sondu-Miriu Basin during study period.

Table 4. Seasonal variation in organophosphorus pesticide residue concentrations in sediment from Nyando/Sondu-Miriu Basin during study period

Pesticide	Rainy season	Dry season
Diazinon	1.08 ± 0.00	0.56 ± 0.01
Dimethoate	0.29 ± 0.02	0.02 ± 0.00
Malathion	BDL	BDL
Fenitrothion	BDL	BDL
Ethyl parathion	BDL	BDL

BDL, below the detection limit.

cides residues in various samples indicated a general trend of higher concentration levels during the rainy season than during the dry season. As the rainfall was the major factor influencing seasonal changes, this could imply that most residues were washed off agricultural fields by the stormwater into the surrounding rivers and the lake.

Concentrations of most organochlorines (96%) and organophosphates (100%) found in the Nyando/Sondu-Miriu basin are below the minimum levels and may not pose environmental and/or human health risks. There is an increasing need for constant monitoring of these compounds in the aquatic ecosystem, however, to ensure protection of aquatic food sources. There also is a need to

establish maximum permissible levels of toxic substances for protection of aquatic biota.

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