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Levels and distribution of pesticide residues in soil and sediments in Eastern Lake Tanganyika environs

John A.M. MAHUGIJA[1*], Lutamyo NAMBELA[2] and Aviti J. MMOCHI[3]

[1]*Chemistry Department, University of Dar es Salaam, P.O. Box 35061 Dar es Salaam, Tanzania.*
[2]*College of Engineering and Technology, University of Dar es Salaam, P.O. Box 35131 Dar es Salaam, Tanzania.*
[3]*Institute of Marine Sciences, University of Dar es Salaam, P.O. Box 668 Zanzibar, Tanzania.*
[*]*Corresponding author; E-mail: mahugija@udsm.ac.tz; johnmahugija@yahoo.com; Tel.: + 255-222410038*

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ABSTRACT

The aim of this study was to investigate the types, levels and distribution of pesticide residues in Eastern Lake Tanganyika basin in Tanzania. Water, soil and sediments samples were collected from various sites in Kigoma region. Analyses of cleaned sample extracts were performed using gas chromatography-electron capture detection (GC-ECD) and gas chromatography-mass spectrometry (GC-MS). Six pesticide residues and metabolites were detected, namely, *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDE, *p,p′*-DDD, diazinon and chlorpyrifos. DDT, DDD and DDE were the predominantly detected compounds in all of the samples. Diazinon and chlorpyrifos were detected in soil samples only. The highest concentrations of total DDT in sediments and soil ranged from 10.02 to 116 µg/kg dry weight (dw) and 7.5 to 564.2 µg/kg dw, respectively. Chlorpyrifos and diazinon had concentrations up to 36 and 184 µg/kg dw, respectively. The concentrations of DDT residues were greater in soil samples than in sediments. The highest concentrations of DDT residues were detected in soil samples. The ratios of (*p,p′*-DDE + *p,p′*-DDD)/*p,p′*-DDT indicated recent inputs in most samples. The study reveals that there were recent uses of DDT, diazinon and chlorpyrifos in the region. Continued use of DDT indicates risks and may result into serious environmental problems. The area therefore needs serious environmental monitoring.

Keywords: Pesticide, Lake, soil, sediments, Tanzania.

INTRODUCTION

The improvements in the agricultural sector in African countries such as Tanzania have led to the increased uses of pesticides. As a result, the importation of pesticides has been increasing annually (Ngowi, 2002). During the 1970s and 1980s, pesticides used in Tanzania were mostly organochlorines. Due to their environmental and health effects (e.g. persistence, toxicity and bioaccumulation) most of the organochlorine pesticides were banned in the 1990s and replaced by organophosphorus pesticides, pyrethroids and carbamates which are readily biodegradable and less persistent in the environment (Rwetabula, 2007). In African

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countries, the farmers' practices in the use of the pesticides are not predictable due to lack of proper training or knowledge. Consequently, the non-observance of good agricultural practices, especially the abusive use of agricultural inputs such as pesticides are frequently observed on farms (Agboyi et al., 2015). Farmers may use any pesticides even if they are banned or not recommended for particular uses provided they are available for other applications and they function well on the control of pests or if they are cheap. Under these conditions, there are high risks of environmental pollution related to farmers' practices on indiscriminate use of pesticides. The increased use of various types of pesticides in agriculture can cause serious environmental contamination leading to high levels of their residues in environmental matrices such as soil and sediments. Pesticides have been linked with many health effects to organisms such as humans and birds. The effects caused by pesticides vary from acute health effects (diarrhoea, skin and eye irritation, headache, etc.) to chronic health effects such as cancer, mutation, immunosuppression, reproductive defects and endocrine disruption. The pesticides have serious implications to the environment due to persistence, bioaccumulation, long-range transport and destruction of non-targeted organisms (Aktar et al., 2009).

The Lake Tanganyika catchment has human settlements from small villages to capital cities which host a variety of human activities including farming, fishing, livestock keeping (husbandry) and tourism. Tobacco, coffee, cotton and horticultural crops, the major pesticide consumer crops, are among the cash crops grown in Lake Tanganyika basin in Kigoma region. Pesticides are widely used in the control of pests in the crops such as tobacco, coffee, and maize. Some studies on pesticide residues have been conducted in Lake Tanganyika (Foxall et al., 2000; Manirakiza et al., 2002). However, to the best of our knowledge, there is no study conducted on agricultural pesticide residues in soil and sediments in Lake Tanganyika basin in Tanzania. Therefore, this study was conducted to assess the concentrations of the pesticide residues in soil and sediments to gauge the level of contamination in the area.

MATERIALS AND METHODS

Study area and sample collection

Lake Tanganyika, the second deepest lake in the world is about 676 km long, with width varying from 50 to 80 km, maximum depth of 1470 m and average depth of 570 m. It is located in Africa's Western Great Rift Valley, between the latitudes 03˚20' and 08˚48' South and the longitudes 29˚03' and 31˚12' East. The major rivers and streams that flow into this lake include Malagarasi River and Luiche River that enter the lake from the eastern side. Malagarasi River is the second largest river in Tanzania (Foxall et al., 2000).

Soil and sediment samples were collected in Kigoma region in Tanzania. The soil samples were collected along the Malagarasi River at Malagarasi Bridge station and Ilagala (from tomato fields near the river). Other soil samples were collected from tobacco fields at Mganza (Nguruka area), from tomato fields at Luiche and Forodhani stations as well as from coffee farms at Kalinzi. The sediment samples were collected from Lake Tanganyika at Forodhani station, Malagarasi River from two stations (Malagarasi Bridge and Ilagala), Luiche River and Lake Nyamagoma station. Lake Nyamagoma discharges its water into Malagarasi River. The samples were collected in April 2012 (rainy season) and June 2012 (dry season). Figure 1 shows the locations of the sampling sites. A total of 62 samples (31 soil samples and 31 sediment samples were collected. Sampling was done applying the procedures described by Marco and Kishimba (2007). The soil and sediment and samples were collected using clean spoons, enveloped in aluminium foil and kept frozen at –18 °C until extraction.

Extraction and clean-up

Extraction and clean-up were performed at the Chemistry Department, Dhaka University in Bangladesh. The soil and sediment samples were extracted by QuEChERS (Quick, Easy, Cheap, Effective, Rugged, and Safe) method that involved homogenization of the sample, soaking the

sample with acetonitrile for a shake extraction followed by dispersive solid phase extraction clean-up using $MgSO_4$ and PSA (Primary Secondary Amine) adsorbents to remove water and interferences from the sample extracts (Lehotay, 2004). Two subsamples (5 g each) were taken simultaneously. One subsample was placed in a pre-weighed petri dish and dried at 105 °C to constant weight in order to determine the dry weight of the sample. Another subsample was placed in a 45-mLTeflon centrifuge tube and acetonitrile (10 mL) was added and the content was vigorously shaken by an inverter for 1 min, then anhydrous $MgSO_4$ (6 g) and NaCl (1.5 g) were added. The mixture was again vigorously shaken by an inverter for 1 min followed by centrifugation (4000 rpm for 5 min). The supernatant (5 mL) was drawn using a pipette and kept into a round bottomed flask. The solvent was evaporated and replaced with n-hexane (2 mL) for clean-up. The sample extract (2 mL) was placed in a centrifuge test tube containing PSA (150 mg) and anhydrous $MgSO_4$ (950 mg). The mixture was vigorously shaken by an inverter for 1 min then centrifuged (4000 rpm for 5 min). The extract was filtered through a 5-mL column containing a 0.45 µm filter into a vial for GC analysis and the final volume of extract was adjusted to 2 mL.

Analytical quality assurance

The glassware and tools used were cleaned by detergent and tap water, rinsed with distilled water, dried and rinsed with acetone before use. All the reagents and solvents used were of analytical grade and high purity. A 50 mL aliquot of each solvent was concentrated to 2 mL and analysed to check the contamination from the solvents used. Three blanks in each sample matrix were prepared and analysed using the same procedures as for the samples. No significant peaks were observed in their chromatograms. The following reference standards of above 95% purity were used: *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDD, *p,p′*-DDE, diazinon, chlorpyrifos, cypermethrin and fenvalerate (Dr. Ehrenstorfer, Germany). Methods were validated by spiking blank soil and sediment samples with standards at concentrations of 0.05, 0.0625, 0.1, 0.125, 0.25 and 0.5 µg/mL for each of the compounds. The mean percentage recoveries ± SD (n = 6) ranged from 88 ± 8% to 101.3 ± 19.7% for soil and sediments. The recoveries were within the acceptable range of 70-120% (Hill, 2000). Standards of concentrations ranging from 0.000125 to 1.0 µg/mL were injected to determine the limits of detection. The detection limits in soil and sediment samples were 0.03 µg/kg for organochlorines and 0.1 µg/kg for organophosphorus and pyrethroid pesticides.

Sample analysis

A GC-2010 Shimadzu gas chromatograph equipped with ^{63}Ni Electron Capture Detector (ECD) and non-polar (HP-5ms) capillary column of dimensions 30 m x 0.25 mm i.d. x 0.25 µm film thickness was used for the analyses at Chemistry Department, Dhaka University, Bangladesh. Nitrogen was used as both a carrier and make up gas at a flow rate of 23.7 ml/min. The temperature programme was 120 °C held for 2 min, 10 °C/min to 270 °C, held for 1 min, 2 °C/min to 290 °C held for 3 min. The injector and detector temperatures were 220 °C and 290 °C, respectively. The GC was operated in a splitless model and for each injection, the volume injected was 1 µL. Samples were injected in duplicate. The standard mixture was injected at the beginning and after every six samples. Confirmatory analyses were performed using gas chromatography-mass spectrometry (GC-MS) at Chemistry Department, University of Dar es Salaam. Identification of analytes was done by comparing the retention times and mass spectra of the sample analytes to those of external reference standard solutions run at the same conditions with the samples. Quantification was carried out by linear integration of the standards and sample data based on peak areas.

Statistical analysis

Statistical analyses were performed using SPSS software 19.0. The mean concentrations of pesticide residues were compared using *t*-test. The correlations in the concentrations of the related compounds were computed using Pearson's *r* coefficient.

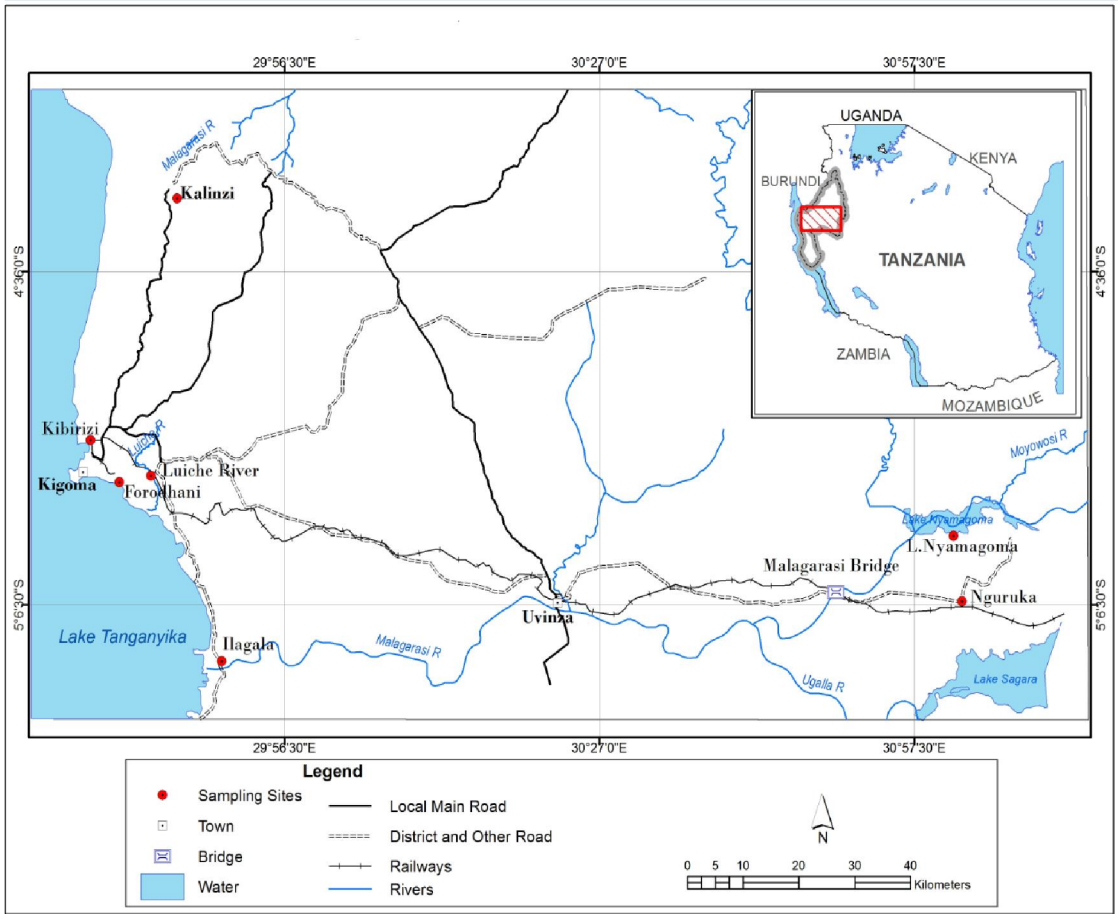

Figure 1: A map showing the study area and sampling sites.

RESULTS

Pesticide residues in soil samples

The pesticide residues detected in soil samples were *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDD, *p,p′*-DDE, diazinon and chlorpyrifos. Cypermethrin and fenvalerate were not detected in all the soil samples. The concentrations of the pesticide residues detected in the soil samples in dry weights (dw) are presented in Table 1 and Table 2. The dominant compounds detected in all the soil samples were *p,p′*-DDT, *o,p′*-DDT, *p,p′*-DDD, and *p,p′*-DDE. The highest concentrations of total DDT in soil samples varied from 7.5 to 564.2 µg/kg and the mean concentrations of total DDT ranged from 10.51 to 278 µg/kg. The highest concentrations of DDT residues were found in samples from tobacco fields at Mganza Nguruka. The concentrations of diazinon and chlorpyrifos detected in soil samples are shown in Table 2. Chlorpyrifos and diazinon were detected in samples collected from coffee, vegetable and tomato farms. The highest concentration of diazinon was 184 µg/kg in samples collected from tomato and vegetable fields at Ilagala and the highest concentration of chlorpyrifos was 36 µg/kg dw in samples from coffee fields at Kalinzi.

Pesticide residues in sediment samples

The pesticide residues detected in sediment samples were mainly DDT residues. Diazinon, chlorpyrifos, cypermethrin and fenvalerate were not detected in any sediment sample. The mean concentrations of the

pesticide residues detected in sediment samples are shown in Table 3. The concentrations of total DDT in sediment samples ranged from 10.02 to 116 µg/kg. The highest total DDT concentration was obtained in sediment samples from Lake Nyamagoma, while the lowest concentration was found in samples from L. Tanganyika at Forodhani.

Distribution of pesticide residues in the samples and sampling periods

The distributions of DDT residues in the samples are summarised in Figure 2 using their mean concentrations in the sample matrices collected at the same sites and same periods. The figure shows the distribution of total DDT in soil and sediment samples. The distributions of the pesticide residues (total DDT, diazinon and chlorpyrifos) in the samples between the sampling periods are summarised in Figure 3 using the mean total concentrations of the pesticide residues. The figure represents the sites from which the samples were collected during both periods.

Table 1: Concentrations of DDT residues in soil samples (µg/kg dw).

Sampling Sites	SP	Range/ Mean	*p,p′*- DDE	*p,p′*- DDD	*o,p′*- DDT	*p,p′*- DDT	ΣDDT	(DDE+DDD)/ DDT
Malagarasi	Apr	Max	20.0	18.0	40.0	57.0	134.2	0.67
		Min	11.0	14.44	16.3	54.4	96.04	0.47
		Mean, n = 2	15.4	16.12	28.1	55.6	115.22	0.57
	Jun	Max	3.0	3.0	7.32	9.0	22.0	0.80
		Min	2.0	1.21	3.4	4.0	10.3	0.67
		Mean, n = 3	2.4	2.0	5.0	5.53	15.0	0.80
Ilagala	Apr	Max	1.12	3.0	5.0	6.0	15.0	0.70
		Min	0.9	2.7	4.6	5.5	13.7	0.65
		Mean, n = 2	1.0	2.9	4.8	5.8	14.4	0.68
	Jun	Max	2.5	3.01	5.5	6.2	17.2	1.12
		Min	2.23	2.23	3.53	4.0	12.0	0.90
		Mean, n = 2	2.4	2.0	4.5	5.0	14.0	0.88
Kalinzi	Apr	Max	79.0	22.32	36.2	57.0	194.2	1.78
		Min	9.5	9.01	14.2	23.53	56.2	0.80
		Mean, n = 5	27.0	15.4	25.6	44.4	112.3	0.95
	Jun	Max	2.5	1.63	4.4	5.11	14.0	0.90
		Min	1.84	0.83	2.0	3.0	7.5	0.81
		Mean, n = 3	2.13	1.3	3.11	4.0	10.51	0.86
Luiche	Apr	Max	10.63	10.8	45.64	59.6	127	0.70
		Min	3.01	2.6	7.2	8.0	20.81	0.36
		Mean, n = 3	6.71	7.0	24.5	33.72	72.0	0.41
	Jun	Max	6.2	10.0	10.31	17.52	44.0	0.94
		Min	1.41	1.4	3.0	3.0	8.33	0.92
		Mean, n = 2	4.0	6.0	5.0	10.2	24.6	0.98
Forodhani	Apr	Max	4.2	4.5	10.5	12.2	29.2	1.40
		Min	2.02	2.1	3.83	4.5	14.62	0.53
		Mean, n = 3	3.0	3.0	6.2	7.23	19.43	0.83
	Jun	Max	6.62	2.41	6.4	8.61	24.0	1.27
		Min	2.24	2.0	3.0	3.33	10.24	1.05
		Mean, n = 3	5.0	2.2	4.1	5.4	16.4	1.33
Mganza Nguruka	Apr	Max	14.33	35.4	190	324.4	564.2	0.37
		Min	10.3	13.0	45.1	62.2	130.3	0.15
		Mean, n = 3	12.0	20.5	93.5	152	278	0.21

SP = sampling period, Min = minimum, Max = maximum, Apr = April, Jun = June

Table 2: Concentrations of diazinon and chlorpyrifos in soil samples (µg/kg dw).

Sampling Sites	Concentrations in April samples			Concentrations in June samples		
	Range, Mean	Diazinon	Chlorpyrifos	Range, Mean	Diazinon	Chlorpyrifos
Malagarasi	Max	nd	Nd	Max	nd	0.4
	Min	nd	Nd	Min	nd	nd
	Mean, n = 2	nd	Nd	Mean, n = 3	nd	0.12
Ilagala	Max	184	5.4	Max	142	0.5
	Min	179	4.9	Min	83.0	nd
	Mean, n = 2	181.5	5.2	Mean, n = 2	112.5	0.23
Kalinzi	Max	25.0	4.3	Max	2.62	36.0
	Min	5.0	Nd	Min	nd	17.1
	Mean, n = 5	16.0	1.4	Mean, n = 3	1.0	23.51
Luiche	Max	3.0	0.52	Max	56.0	1.0
	Min	nd	Nd	Min	14.0	nd
	Mean, n = 3	1.51	0.3	Mean, n = 2	35.0	0.33
Forodhani	Max	84.0	22.1	Max	6.02	1.53
	Min	nd	9.0	Min	nd	nd
	Mean, n = 3	28.0	13.1	Mean, n = 3	2.01	1.0
Mganza Nguruka	Max, Min, Mean, n = 3	nd	nd	na	na	na

nd = not detected (below detection limit), na = not analysed

Table 3: Mean concentrations of DDT and metabolites in sediment samples (µg/kg dw).

Sampling Sites/Points	Sample period	*p,p′*-DDE	*p,p′*-DDD	*o,p′*-DDT	*p,p′*-DDT	ΣDDT	(DDE+DDD)/DDT ratio
L. Nyamagoma (2 points, n = 6)	April	17.0	21.0	38.4	39.10	116.0	0.97
		4.0	4.5	9.41	11.03	29.00	0.77
Malagarasi (2 points, n = 7)	April	2.0	1.0	4.30	5.33	12.63	0.56
	June	2.0	1.2	1.62	6.10	10.92	0.52
		2.0	2.3	2.0	5.0	11.30	0.86
Ilagala (n = 6)	April	3.0	3.0	7.12	8.13	21.30	0.74
	June	3.0	1.43	3.0	3.01	10.40	1.47
Luiche (n = 6)	April	6.0	11.2	18.0	19.4	54.60	0.90
	June	2.0	1.52	3.0	5.40	12.00	0.65
Forodhani (n = 6)	April	3.1	2.23	7.4	9.04	22.00	0.60
	June	2.0	1.0	3.0	4.02	10.02	0.75

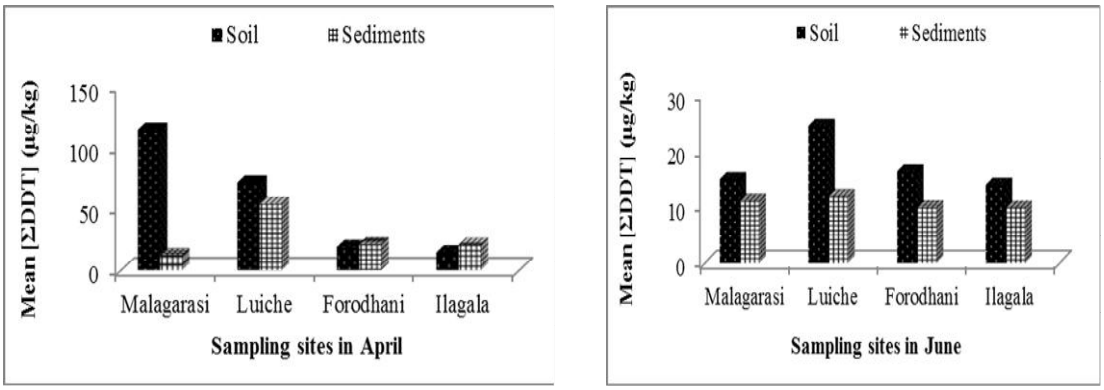

Figure 2: Distribution of DDT residues in soil and sediment samples.

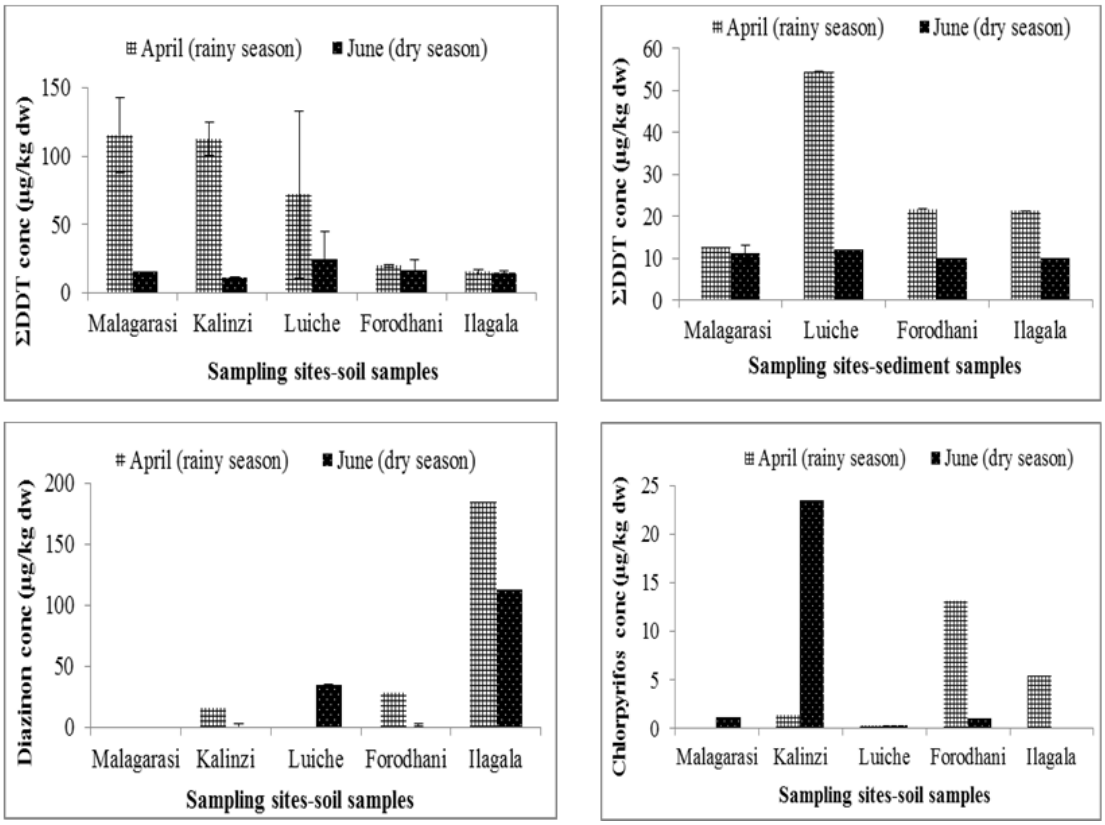

Figure 3: Distribution of the concentrations of total DDT, diazinon and chlorpyrifos in samples between sampling periods; Error bars indicate standard deviations.

DISCUSSION

Profiles of the pesticide residues

In all the soil and sediment samples, the concentrations of *p,p′*-DDT were higher than those of *o,p′*-DDT, indicating contamination by technical DDT (ATSDR, 2002). There were no significant differences between the concentrations of *p,p′*-DDD and *p,p′*-DDE in soil samples (t = 1.447, p = 0.155, 41 degrees of freedom) and in sediment samples (t = 0.590, p = 0.568, 10 degrees of freedom) from all the studied sites. This indicated neither anaerobic nor aerobic degradation was favoured over the other. DDD is a major metabolite of DDT resulting from anaerobic transformation while DDE results from aerobic transformation of DDT in the environment (ATSDR, 2002). The concentrations of *p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD correlated significantly with each other (soil: r = 0.84–0.94, $p < 0.05$, 30 degrees of freedom and sediments: r = 0.8–0.95, $p <$ 0.05, 11 degrees of freedom), which suggested common sources. The ratios of (*p,p′*-DDE + *p,p′*-DDD)/*p,p′*-DDT can be used to establish whether the contamination is due to recent input/application or historical/past use, because the levels of the parent compound (*p,p'*-DDT) in the environment decrease with time relative to its metabolites, *p,p′*-DDE and *p,p′*-DDD. A high ratio (> 1) indicates past use or old source of DDT which has been largely converted to DDE and DDD, while a low ratio (< 1) indicates recent use or exposure to the parent DDT or insignificant degradation (Jaga and Dharmani, 2003; Qiu et al., 2004; Fianko et al., 2013). The (*p,p′*- DDE + *p,p′*-DDD)/*p,p′*-DDT ratios were low (< 1) in most of the soil samples and in almost all sediments (Table 1 and Table 3) indicating recent use of DDT in nearby areas or old input of DDT without significant degradation. High (*p,p′*- DDE + *p,p′*-DDD)/*p,p′*-DDT ratios (> 1) were found in soil samples from Kalinzi, Forodhani and Ilagala and in sediment samples from Malagarasi River at Ilagala station, indicating past use or significant degradation. The sources of contamination might be due to applications in the coffee farms, tobacco farms and horticultural farms located in the vicinity of the sampling locations. The DDT contaminations in the rivers and lakes might be associated with the agricultural activities taking place along the rivers and lake areas. This may be due to illegal uses because DDT was banned for use in agriculture, but it is allowed for use in malaria control. Some of the DDT residues might be from environmental sources.

The detection of the organophosphorus pesticide residues in soil samples implied recent application since they are environmentally unstable. Recent applications in the agricultural fields located in the vicinity of the study areas could be the sources of contamination. These findings are similar to those observed in a study conducted in tomato fields in Owiro Estate, Tanzania where the detection of chlorpyrifos was related to the recent use (Kihampa et al., 2010).

Studies from other agricultural areas in Tanzania have shown the presence of higher levels of pesticide residues mainly oganochlorine pesticides in soil and sediments (Kishimba et al., 2004). For example, the study that was conducted on soil samples collected from cotton growing areas in the southern Lake Victoria basin revealed the presence of *p,p′*-DDT, *p,p′*-DDE and *p,p′*-DDD with the concentrations up to 12000, 9000 and 2000 µg/kg dw, respectively (Henry and Kishimba, 2003). Another study conducted in the sugarcane plantations reported concentrations of total DDT ranging from 22.6 ± 1.1 to 1452.6 ± 2.1 µg/kg dw (Hellar-Kihampa, 2011). The concentrations of the pesticide residues found in these studies were generally higher than those found in the present study. The concentrations of the DDT residues found in the sediment samples in this study were generally lower than those found in previous studies conducted in other parts of Tanzania such as Coastal region and Southern

Lake Victoria and its basin (Kishimba et al., 2004).

Studies on pesticide residues in other countries have found different trends for DDT residues in various samples that were contaminated due to environmental sources or application. For example, Fianko et al. (2013) found high ratios of DDE/DDT in fish samples (with a mean of 2.51) indicating old input of DDT and significant degradation. Similarly, the study by Sosan et al. (2015) found higher concentrations of *p,p′*-DDD and *p,p′*-DDE than the concentrations of *p,p′*-DDT in foodstuffs from markets in Ile-Ife, Nigeria indicating that the DDT had substantially metabolized into DDD and DDE.

Variations of pesticide residues between samples and sampling periods

The concentrations of DDT residues in soil samples were greater than those in sediments (Figure 2). The distributions of DDT residues in soil and sediments depend on, among other factors, the sources of contamination and their strong adsorption to the particles (Delle Site, 2001). However, the levels of the pesticide residues in sediments can be reduced due to dilution effects and washing away by water. The contamination patterns of the DDT residues were very similar to the findings in water samples from the rivers and lakes within the study area except that the concentrations of *p,p′*-DDD were higher than those of *p,p′*-DDE in almost all water samples indicating that anaerobic degradation pathway was more favoured than aerobic degradation (Mahugija and Nambela 2015).

The results indicated marked differences in the concentrations of the DDT residues between the sampling periods. The trends in the mean concentrations of total DDT in soil samples showed that the concentrations of DDT residues in soil were greater in April than June (Figure 3). This indicated that higher DDT contamination occurred during the rainy season than the dry season probably due to application and other sources in the vicinity of the study areas. Similarly, in all sites the concentrations of DDT residues detected in sediment samples in April were greater than those detected in June. This indicated that during the rainy period, more pesticide residues from fields and gardens were carried by water into the rivers and lakes than in the dry period. Moreover, during the dry season, sediments do not receive any significant pesticide residues from point sources because there is no runoff. The variations are generally similar to those found in water samples (Mahugija and Nambela 2015). For diazinon and chlorpyrifos there was no specific trend observed (Figure 3) suggesting very minor variations in the applications. This could also be due to similarities in applications or contamination patterns for both periods.

Conclusion

The level of contamination in Eastern Lake Tanganyika basin was generally moderate to high. The findings indicate risks and concerns for public health and the environment because of the fresh inputs or applications indicated especially of the persistent compounds (DDT and metabolites) that can undergo bioaccumulation and biomagnification in organisms. The regulatory authorities should check the pesticides used in the areas surrounding the basin.

COMPETING INTERESTS

The authors declare that they have no competing interests.

AUTHORS' CONTRIBUTIONS

LN and JAMM designed the methods, performed the field works, laboratory works and data analysis and drafted the manuscript. JAMM and AJM supervised the research. All authors read and approved the final manuscript.

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