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**Review article** 

# Occurrence of organochlorine pesticide residues in biological and environmental matrices in Africa: A two-decade review

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#### ABSTRACT

The widespread use of organochlorine pesticides (OCPs), essentially for the control of insects and the cultivation of food crops, has led to the pollution of ecosystems. Despite being banned several years ago in the developed world, extensive use remains ongoing on the African continent. This review summarizes the occurrence, distributions, sources, and trends of OCPs in seven environmental matrices (atmosphere, water, sediments, soils, biota, human fluids and food products) in Africa. Findings in this review revealed that  $\alpha$ -HCH,  $\beta$ -HCH dichlorodiphenyltrichloroethane (DDTs), and endosulfans were the most persistent OCP residues in the African environment, particularly DDTs in breast milk samples occurring in levels above the WHO stipulated limits, thus indicating a call for concern. Also, there was paucity of data available on OCP concentrations in ambient air. Future research efforts should prioritize testing these pollutants in the atmosphere, especially in countries where they are used more frequently. While most POP analysis studies used gas chromatography coupled to electron capture detector or mass spectrometer, it is recommended that further studies should use more sensitive analytical techniques such as gas chromatography with tandem mass spectrometry (GC-MS/MS), or gas chromatography coupled to high-resolution mass spectrometry (GC-HRMS). These instruments allow for the detection of secondary and tertiary metabolites, especially those found in water, biota and food products, which are critical vectors of OCPs to human and animal bodies. Training of farmers and other domestic users on the handling of pesticides is proposed.

#### 1. Introduction

The adverse effects of persistent organic pollutants (POPs) in the ecosystem have been one of the major subjects of discussion all over the world. These chemicals are toxic and because of their ability to be transported long distance from the place of use and release either by water or wind, they tend to affect biota and humans in diverse ways (Buccini, 2003; EPA, 2009). Due to their ubiquitous nature, lipophilic properties, and persistence in the environment, they tend to accumulate in tissues of living organisms (Afful and Anim, 2010; Olisah et al., 2019a). Many of these compounds have been classified as endocrine disruptors, because of their tendency to reduce the efficiency of thyroid hormones and to affect the neurobehavioral development and reproductive systems in animals and humans (ATSDR, 2000). Carcinogenic cases of these compounds have been reported (El-Shahawi et al., 2010). Initially, 12 POPs classified as "Dirty Dozen" were identified as endocrine

disruptors (UNEP, 2001). Chemicals such as organochlorine pesticides (OCPs) in this group, are used for pest control because of their versatility (Olisah et al., 2019b). These include insecticides such as aldrin, dieldrin, endrin, chlordane, dichlorodiphenyltrichloroethane (DDT), mirex, toxaphene, and heptachlor; fungicides - hexachlorobenzene (HCB) and industrial chemicals -polychlorinated biphenyls (PCBs). Other chemicals in this group that are unintentionally produced are polychlorinated dibenzo-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs). Nine others including industrial chemicals (pentabromodiphenyl ether, lindane, octabromodiphenyl ether, chlordecone, α  $-hexachlorocyclohexane, \beta-hexachlorocyclohexane, perfluorooctane$ sulfonate, hexabromobiphenyl and pentachlorobenzene), classified as "Nasty Nine" were added during the meeting held in May 2009 under the treaty of the Stockholm convention (UNEP, 2009).

Population growth has led to increased use of pesticides for crop cultivation. Research have shown that pesticides are used to cultivate

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more than one quarter of farm products (Liu et al., 2002). A decrease in production of cereals (32%), vegetables (54%) and fruits (78%) has been reported without the use of pesticides (Cai, 2008). Apart from agriculture, the importance of pesticides in the health sector cannot be overemphasised. According to World Health Organisation (WHO), indoor residual spraying of pesticides and the use of nets treated with insecticides are the two most important measures in which humans are being protected from mosquitos and other insects capable of causing illness. This has been found in many countries in the world mostly in malaria-endemic regions (World Health Organization, 2016). Despite the benefits of using pesticides, the risk it poses to humans and animals cannot be overlooked. The improper handling of these chemicals by farmers and other end users have been responsible for the high level of pesticides found in the atmosphere, water bodies, and soils, hence posing threat to human health and the ecosystem at large. Various problems resulting from pesticides found (especially the organochlorine types) in the environment have been reported in Asia, America, Europe and Africa (Hoh and Hites, 2004; Gunnell et al., 2007; Ali et al., 2013; Elibariki and Maguta, 2017). These include DDTs, HCHs, endosulfans, HCB, Drins (aldrin, endrin, and dieldrin). As of 2009, most African countries continued usage of OCPs even though they were banned (UNEP, 2009). Most of these pesticides get into the ecosystem through runoff from agricultural farmlands, illegal disposal of waste, and by-products from chlorine combustion processes (Afful and Anim, 2010). Extremely high levels of these pollutants have been detected by researchers in Africa and other parts of the world in various matrices, including biota, sediment, soil, water and food products (Kishimba et al., 2004; Bempah et al., 2012; Kolani et al., 2016).

In the last two decades (1996–2016), several studies have been conducted to analyse environmental matrices in Africa for the presence of OCPs, although many were not documented. Based on published literature, our current knowledge of the persistence of OCPs in the African environment remains scarce (Williamson et al., 2008; Elibariki and Maguta, 2017). This article therefore summarises the levels of OCPs in environmental and biological matrices in African environment in the last two decades (1996–2016).

#### 1.1. Pesticide use in Africa

The pesticides used in Africa from 1990 to 2016 were estimated to be 69,355.36 tons active ingredient (a.i), accounting for about 2.1% world total pesticide usage. This value is low compared to that of Asia, Americas and Europe which were reported to be 1,727,322.77 ton a.i, 956,040.22 tons a.i and 455,159.85 tons a.i respectively (http:// www.fao.org/faostat/en/#data/RP/visualize). Pesticides are majorly used for large scale farming especially in the cultivation of cash crops such as cocoa, oil palm, and vegetables (Matthews et al., 2003). Their use in small scale farming was found to be almost insignificant (Nsibande and McGeoch, 1999; Ebenebe et al., 2001). Their use for large scale commercial farming pose a greater risk to the ecosystem in Africa, and studies indicate a positive correlation with poor pesticide handling (Sibanda et al., 2000; Vaissayre and Cauquil, 2000). Example of such practices include over dosage, incorrect use of equipment for pesticide application, leakage from storage tanks. This inappropriate use leads to soil, water and air contamination which in turn pose risks to humans and livestock (Conway, 2013). Some extreme cases include lack of adequate personal protective equipment, tongue tasting to verify the originality or concentration of the pesticides, the sprinkling of pesticides directly to the germinated crop and mixing of pesticides with bare hands (Sibanda et al., 2000; Dinham, 2003). Reports from the pesticides policy initiatives for various countries in Africa indicated that pesticides usage in Africa is largely influenced by various biological, economic and climatic factors (Mudimu et al., 1995; Ngowi, 2002). Hence, the levels and trends of usage vary from one country to another.

## 2. Occurrence of OCPs in Africa

#### 2.1. Atmosphere

OCPs are volatile compounds that are largely found in the atmosphere after volatilising from contaminated soils and water. Very few reports have been documented on the evaluation of these contaminants in air in Africa, even though the atmosphere is considered a medium, in which they travel long distance. Karlsson et al. (2000) quantified HCHs, DDTs, chlordane and endosufan in air collected twice a week in 1997 and 1998 using polyurethane foam from Senga Bay and Laurentian Great Lake Malawi area. Samples were analysed with gas chromatograph - electron capture detector (GC-ECD). The analyses revealed that the mean concentrations of ΣDDTs, ΣHCHs, dieldrin, endrin and mirex in Senga bay were  $26 \pm 31 \text{ pg/m}^3$ ,  $40 \pm 43 \text{ pg/m}^3$ ,  $80 \pm 80 \text{ pg/m}^3$ ,  $1.0 \pm 1.1 \text{ pg/m}^3$ , and  $0.15 \pm 0.44 \text{ pg/m}^3$  respectively. In general, OCP residues ranged from 31 to 257 pg/m<sup>3</sup> in Senga Bay while Laurentian Great Lake recorded a value ranging from 24 to 40  $pg/m^3$ . From the results, the authors inferred that excessive application of pesticides in tropical regions might be responsible for their occurrence in the atmosphere. Batterman et al. (2008) monitored three sample sites for OCPs. Two sites were industrial areas located in the southern part of Durban while the other was a residential area in the northern part. Sampling was done between the periods of 2004–2005. Air samples were collected using polyurethane foam plugs (75 mm) and analysis was done using GC-MS. p,p'-DDT and p,p'-DDD were detected at the 3 sampling locations with a mean concentration of  $42 \pm 27 \text{ pg/m}^3$  and  $12 \pm 11 \text{ pg/m}^3$  respectively. These values were considered to be relatively high. Other contaminants that were quantified include dieldrin, HCB aldrin, lindane, and chlordane. The authors inferred that chlordane and lindane emanated from regional or global sources as well as local sources. Data from this monitoring also revealed that this study is one of the most detailed in evaluating airborne pesticides in Africa.

### 2.2. Water

The aquatic systems harbour appreciable amount of OCPs through agricultural run-off, leachates from contaminated soils, spray drift from pesticides application on food crops, and careless disposal of pesticide containers. Such processes have led researchers to evaluate the occurrence of pesticides in various aquatic systems in South Africa especially in Lourens River, Hex River and Pongolo floodplain where endosulfan, DDTs and other pesticides were reported in significant concentrations (Bollmohr et al., 2007; Dabrowski et al., 2002a; Dabrowski et al., 2002b; Dalvie et al., 2003; McGregor, 1999; Naudé et al., 1998; Quinn et al., 2009; Schulz et al., 2001; Schulz, 2001a; Schulz and Peall, 2001; Thiere and Schulz, 2004). Table 1 shows the concentrations of some residues of OCP in water resources from different African countries.

Reports on OCPs from Ghana showed lindane and endosulfan sulphate as the most persistent pesticides found in over 70% of the total water samples (n = 50) collected around a farming community. Approximately 68% of waters recorded the presence of  $\alpha$ -endosulfan with an average concentration of 62.3 ng/L. Other pesticides such as heptachlor epoxide, HCB and DDE were not detected (ND) (Ntow, 2001). In contrast, a similar study conducted in Ghana showed that *p*,*p*'-DDE and *p*,*p*'-DDT were the most prevalent, occurring in 82% and 78% of water samples from Lake Bosumtwi (Darko et al., 2008).

Surface and ground waters from the Eastern Cape province in South Africa collected during June and December 2002 were analysed for OCPs (BHC isomers, metabolites of DDTs, Drins, chlordanes, endosulfan, HCB and heptachlor) by Fatoki and Awofolu (2004). These compounds were determined using GC-ECD. In this study, the OCPs detected were lower than the maximum stipulated limit of European Union of 0.1  $\mu$ g/L (Council Directives, 2008). However, an elevated level of HCB and  $\delta$ -BHC was detected in water from Buffalo River; an important freshwater resource for domestic use by its host communities in the province (Fatoki

Table 1. Mean concentration ( $\pm$ standard deviation) and ranges of OCPs in water samples from Africa ( $\mu$ g/L).

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Location	Country	ΣHCHs	ΣDDTs	Lindane	ΣChlordane	Endosulfans	Dieldrin	HCBs	References
Hex River	South Africa					ND-1.79			McGregor (1999)
Hex River	South Africa					BDL-26.3			London et al. (2000)
El-Haram, Giza	Egypt					3.8-290.0		20.7-86.2	El-Kabbany et al. (2000)
Akumadan	Ghana			$\textbf{9.5} \pm \textbf{5.2}$		124.5		<100	Ntow (2001)
Lourens River	South Africa					0.05-2.9			Schulz (2001b)
Lourens River	South Africa					$0.13 \pm 2.9$			Schulz (2001a)
Lourens River	South Africa					0.03-0.16			Schulz et al. (2001)
Lourens River	South Africa					0.20			Schulz and Peall (2001)
Lourens River	South Africa					ND-0.3			Dabrowski et al. (2002b)
Lourens River	South Africa					0.01-0.35			Dabrowski et al. (2002a)
Lake Victoria	Tanzania	ND - 200	ND -1600						Henry and Kishimba (2003)
Eastern Cape	South Africa		BDL-0.26		BDL-0.12	BDL-0.1	BDL-0.1		Fatoki and Awofolu (2003)
Banjul and Dakar	Senegal and Gambia	<loq -="" 0.120<="" td=""><td><loq- 0.231<="" td=""><td></td><td></td><td>ND - 0.026</td><td>ND - 0.0008</td><td></td><td>Manirakiza et al. (2003)</td></loq-></td></loq>	<loq- 0.231<="" td=""><td></td><td></td><td>ND - 0.026</td><td>ND - 0.0008</td><td></td><td>Manirakiza et al. (2003)</td></loq->			ND - 0.026	ND - 0.0008		Manirakiza et al. (2003)
Hex River	South Africa					$0.83 \pm 1.0$			Dalvie et al. (2003)
Grabouw dam	South Africa					$3.16\pm3.5$			Dalvie et al. (2003)
KwaZulu Natal	South Africa	BDL-0.002	-						Sereda and Meinhardt (2003)
Tana and Sabaki	Kenya		$5.2\pm0.09$	$5.3\pm0.024$					Lalah et al. (2003)
East London	South Africa		0.0055-0.04		0.0206	0.018	0.0057	0.015-0.16	Fatoki and Awofolu (2004)
Port Elizabeth River	South Africa		0.0055-0.06		0.0206	0.018	0.0057	0.01-0.07	Fatoki and Awofolu (2004)
Buffalo River	South Africa		0.0055-0.07		0.0206	0.018	0.0057	0.01-0.15	Fatoki and Awofolu (2004)
Thyume River	South Africa		0.0055-0.04		0.0206	0.018	0.0055	0.0077-0.08	Fatoki and Awofolu (2004)
Southern Lake Victoria Basin	Tanzania	BDL-5.7	BDL-33				BDL-2.5		Kishimba et al. (2004)
KwaZulu Natal	South Africa		0.0002-0.161						Sereda and Meinhardt (2005)
Lake Volta	Ghana			0.008		0.083			Ntow (2005)
Lake Bosumtwi	Ghana		0.073	0.071		0.064			Darko et al. (2008)
Lourens	South Africa		0.01			0.03			Bollmohr et al. (2007)
Juskei River	South Africa	0.63–196	1.32-3086			3.82–967			Sibali et al. (2008)
Lagos Lagoon	Nigeria		0.005-0.910		0.006-0.950	0.015-0.996	0.015-0.996	0.015-0.774	Adeyemi et al. (2011)
Kilimanjaro	Tanzania	BDL-0.0552	BDL-0.218		-	BDL-0.012	BDL-0.048		Hellar-Kihampa (2011)
Vaal River Region A	South Africa	$80.8 \pm 9.8$	$261\pm67.3$		$2.5\pm0.9$			$8.5\pm2.2$	Wepener et al. (2011)
Vaal River Region B	South Africa	$70.1\pm2.8$	$227\pm56$		$1.1\pm0.5$			$135\pm89$	Wepener et al. (2011)
Vaal River Region C	South Africa	$10.2\pm0.9$	$160\pm47$		BDL			$257\pm204$	Wepener et al. (2011)
Vaal River Region D	South Africa	$156\pm29.6$	$468 \pm 147$		$2.7\pm0.7$			$588 \pm 443$	Wepener et al. (2011)
Densu River Basin	Ghana	0.02-1.07	ND - 0.02		ND - 0.12	ND - 0.26	ND - 0.02		Kuranchie-Mensah et al. (201
Ondo State	Nigeria		ND-0.11				ND-1.51		Okoya et al. (2013)
Weruweru	Tanzania			BDL-3.66	BDL-0.81	BDL-12.7	-		Mohamed et al. (2014)
Lake Volta	Ghana	<loq -="" 0.669<="" td=""><td><loq -="" 0.031<="" td=""><td></td><td><loq -="" 0.009<="" td=""><td><loq -0.076<="" td=""><td><loq -="" 0.031<="" td=""><td></td><td>Gbeddy et al. (2014)</td></loq></td></loq></td></loq></td></loq></td></loq>	<loq -="" 0.031<="" td=""><td></td><td><loq -="" 0.009<="" td=""><td><loq -0.076<="" td=""><td><loq -="" 0.031<="" td=""><td></td><td>Gbeddy et al. (2014)</td></loq></td></loq></td></loq></td></loq>		<loq -="" 0.009<="" td=""><td><loq -0.076<="" td=""><td><loq -="" 0.031<="" td=""><td></td><td>Gbeddy et al. (2014)</td></loq></td></loq></td></loq>	<loq -0.076<="" td=""><td><loq -="" 0.031<="" td=""><td></td><td>Gbeddy et al. (2014)</td></loq></td></loq>	<loq -="" 0.031<="" td=""><td></td><td>Gbeddy et al. (2014)</td></loq>		Gbeddy et al. (2014)
Ogbesse River	Nigeria	0.92	$0.12\pm0.06$		-	0.54	0.14		Lawrence et al. (2015)
		1**	2***			0.03**			

and Awofolu, 2004). Similar studies carried out in this province also detected high levels of  $\delta$ -BCH and heptachlor in runoff from agricultural farm land (Fatoki and Awofolu, 2003). Henry and Kishimba (2003) determined DDTs, HCHs, and endosulfan in surface waters from 9 districts in Southern Lake Victoria in Tanzania. In this study, endosulfans were not detected in all the samples while the concentrations of HCHs and DDTs were 200 µg/L and 1600 µg/L respectively. These level were higher than those reported for water bodies in Banjul and Dakar (Manirakiza et al., 2003). However, high levels of these residues were detected in the dry season compared to rainy season. This was due to the dilution in the latter season with a possible view that the kinetics of pollutants in a marine environment is controlled by advection (Hewitt, 2000). Similar seasonal variations and OCPs dominance were reported in water collected from 4 rivers in Kilimanjaro, Tanzania (Hellar-Kihampa, 2011).

Two rivers (Tana and Sabaki Rivers) largely influenced by agricultural activities in Kenya were sampled by Lalah et al. (2003) for OCP analysis. Samples were collected between 1998 and 1999 and instrumental analysis was done using GC-ECD. Despite the ban of pesticides in Kenya, notably high concentrations were detected. Among such include lindane (5.367  $\pm$  0.024 µg/g), DDE (5.200  $\pm$  0.092 µg/g) and aldrin  $(11.90 \pm 0.007 \,\mu\text{g/g})$  in water weed from River Sabaki. Their levels were found to be very low in all samples from rivers probably because of their hydrophobic properties. Water samples from Ingwavuma and Ubombo districts of KwaZulu Natal in South Africa collected between 2000 and 2001 were analysed for DDDs and DDEs. Sampling was planned to cover the entire duration of the malaria spraying season and instrumental analysis was conducted using GC-ECD. This study showed that over 40% of the samples in each location contained the target analytes, thus suggesting a possible source of contamination (Sereda and Meinhardt, 2005). Adeyemi et al. (2011) determined chlordane, heptachlor, methoxychlor, HCB, endosulfan, DDTs, and drins in water samples from Lagos Lagoon in Nigeria. The concentrations of all OCPs ranged up to 0.996  $\mu$ g/L, while the pesticides concentration in water collected from selected rivers around a cocoa producing area in Ondo State, Nigeria ranged from ND to 1.65 µg/L (Okoya et al., 2013).

α-Chlordane, endosulfan sulfate, p,p' –DDT, p,p' –DDE and lindane were analysed in water samples collected from Weruweru Subcatchment, Tanzania, in rainy and dry season of 2013 using GC-ECD (Mohamed et al., 2014). The sample sites were established based on details provided by key stakeholders and data from a 3DEM software algorithm. In most cases, pesticide residues in samples were below detection limits (BDL). Furthermore, endosulfan sulphate was the most dominant, detected in approximately 33% (in both seasons) of the total samples analysed at concentrations ranging between BDL and 12.7 µg/L. DDT metabolites levels were within the permissible limits stipulated by WHO (1 µg/L) (WHO, 2004), while lindane, detected in one of the site in dry season (3.66 µg/L), was above the WHO permissible limit (2 µg/L) (FAO/WHO, 2002). Other OCPs quantified, ranged from BDL to 45.7 µg/L in all samples. Findings from this study indicated that pesticide residues in the sample area emerged from agricultural run-off and deposits from agricultural soils. The findings summarised in this review indicates that DDTs and endosulfans were the most predominant in water samples from African environment occurring in 26.2% and 24.2% respectively (Figure 1).

## 2.3. Sediments and soils

Sediments, long recognised as a reservoir for pesticides, are considered one of the environmental matrices in the monitoring of pollutants (Bhattacharya et al., 2003). The heterogeneous organic and inorganic particles found in sediments make the accumulation of contaminants possible. OCPs get into sediments through runoff from farmlands, deposition from the atmosphere through precipitation, illegal dumping of used pesticide containers in water bodies and leachate from contaminated soil and glacier deposits (Sarkar et al., 1988). These account for the varying levels of endosulfans and DDTs reported in African soils and sediments as shown in Table 2.

Manirakiza et al. (2003) determined 21 OCPs comprising of alachlor. mirex, heptachlor, methoxychlor, aldrin, dieldrin, endrin, heptachlor epoxide, endosulfans, HCB, HCHs and DDT metabolites in 76 soil samples. Samples were collected in 9 locations from Banjul and Dakar in Gambia and Senegal respectively. GC-µECD was used to detect analytes with lower concentration and GC-MS for analytes with higher concentration. In general, low mean concentration of OCPs in soils were observed. Levels of SHCHs, Sendosulfans, mirex, dieldrin, methoxychlor and aldrin were detected below  $4 \mu g/kg dry$  weight with exception of the  $\Sigma$ HCHs (19.6 µg/kg)in SHG sampling site, dieldrin (12 µg/kg) in BHG site, and  $\Sigma$ endosulfan in Mbao (6.7 µg/kg) and SGH (6.0 µg/kg) sites. Heptachlor, heptachlorepoxides, and alachlor were present in low concentration, except soils from SHG sites with a heptachlorepoxides concentration of 9.4  $\mu$ g/kg whilst DDT and its metabolites had a concentration lower than 15 µg/kg in most of the sampling locations. In general, OCPs in this study were below that reported in Ghanaian sediments (Ntow, 2001), suggesting a very low pesticide usage in Senegal and Gambia.

Fatoki and Awofolu (2003) analysed 15 OCPs in sediments collected from Buffalo (BR), Swartkops (SKP), Thyume (TR) and Keiskamma (KR) Rivers in the Eastern Cape Province, South Africa between January and April 2002. Analysis was carried out using GC-ECD. 2,4'-DDT (trace) recorded the lowest concentration found in Buffalo River sediments while the highest was heptachlor (184 µg/kg). OCPs in the other locations recorded the following range of concentrations: 2,4'-DDT and dieldrin (trace levels) to γ-chlordane (16 µg/kg) in Keiskamma River sediments; 2,4'-DDT and dieldrin (trace levels) to 2,4'-DDD (19 µg/kg) in Thyume River sediments and 2,4'-DDT, 2,4'-DDE, dieldrin (trace levels) to β-BCH (30 µg/kg) in Swartkops River sediments. Levels of DDTs in sediments and water in this study were in line with the concentration of OCPs found in Jukskei River (Sibali et al., 2008). Probable contamination of ground water by OCPs in Vikuge Farm, Tanzania was reported when HCHs and DDTs were detected in soil samples collected from 20 cm and



Figure 1. Percentage occurrence of OCPs in water resources from locations in Africa.

C. Olisah et al.

Table 2. Mean concentration ( $\pm$ standard deviation) and ranges of OCPs in soil and sediment samples from African ( $\mu$ g/kg dry weight).

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Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane	Mirex	ΣChlordane	Endosulfans	Dieldrin	HCB	Reference
KwaZulu Natal	South Africa	Sediment		18.7–374							Naudé et al. (1998)
Mpumalanga	South Africa	Sediment		8.7–396							Naudé et al. (1998)
El-Haram, Giza	Egypt	Soil								ND-16.2	El-Kabbany et al. (2000)
Schulz et al., 2001	South Africa	Sediment						3.9–245			Schulz et al. (2001)
Akumadan	Ghana	Sediment		$0.46 \pm 0.24$	$\textbf{3.2}\pm\textbf{0.6}$			0.55			Ntow (2001)
Lourens River	South Africa	Suspended Particulate						ND -12082			Schulz (2001b)
Lourens River	South Africa	Sediment						9.70–273			Dabrowski et al. (2002a)
Lourens River	South Africa	Sediment						ND-34.75			Dabrowski et al. (2002b)
Lake Victoria	Tanzania	Soil	$5.9\times10^4$	$2  imes 10^4$							Henry and Kishimba (2003)
Lake Victoria	Tanzania	Sediment	$1\;32\times10^5$	$6.0 imes10^5$							Henry and Kishimba (2003)
Banjul and Dakar	Senegal and Gambia	Soil	0.7–19.6	1.5-71.4		ND- 0.7			ND-88.2	ND-23	Manirakiza et al. (2003)
KwaZulu Natal	South Africa	Sediment <sup>a</sup>		$\text{BDL-9.5}\times10^{-3}$				$9 imes 10^{-5}$ - $2.4 imes 10^{-3}$			Sereda and Meinhardt (2003)
Eastern Cape	South Africa	Sediment		BDL-109			BDL-117	BDL-72	BDL-60	BDL-177	Fatoki and Awofolu (2003)
Dar es Salaam coast	Tanzania	Sediment	BDL-2.7	6.4–51					BDL-48		Kishimba et al. (2004)
Lake Victoria	Tanzania	Sediment	BDL-131	BDL-705							Kishimba et al. (2004)
TPC-Moshi	Tanzania	Sediment	BDL-61	BDL-716					BDL-7.2		Kishimba et al. (2004)
Zanzibar	Tanzania	Sediment	BDL-106	BDL-20							Kishimba et al. (2004)
Lourens River	South Africa	Suspended Particulate						23–122			Thiere and Schulz (2004)
Lake Volta	Ghana	Sediment		61.3	2.3			0.74			Ntow (2005)
KwaZulu-Natal	South Africa	Sediment		$1  imes 10^{-3} - 13.7$							Sereda and Meinhardt (2005)
Oueme River	Republic of Benin	Sediment <sup>b</sup>	<0.1–61	<0.1-809				<0.1–164			Pazou et al. (2006)
Lourens River	South Africa	Sediment		BDL-16.4				0.43–12.7			Bollmohr et al. (2007)
Ogba River	Nigeria	Sediment	713	733							Ize-Iyamu et al. (2007)
Ovia River	Nigeria	Sediment	790	560							Ize-Iyamu et al. (2007)
Ikoro River	Nigeria	Sediment	600								Ize-Iyamu et al. (2007)
Upper Awash	Ethiopia	Soil		<0.7–230				<1.2-56000	ND-2.4		Westbom et al. (2008)
Lake Bosomtwi	Ghana	Sediment		12.75	$6.755 \pm 1.15$			$9.683 \pm 1.76$	$0.072\pm0.02$		Darko et al. (2008)
Juskei River	South Africa	Sediment	0.23-12221	4.62-22914				25.3-11462			Sibali et al. (2008)
Owvian River Warri	Nigeria	Sediment	11.48								Lawrence et al. (2009)
Ekakpamre River Warri	Nigeria	Sediment	4.82								Lawrence et al. (2009)
Old Korogwe	Tanzania	Soil		BDL-2.0 $ imes$ 10 <sup>6</sup>							Kihampa and Mato (2009)
Vikuge Farm	Tanzania	Soil	ND-6.3 $\times$ 10 <sup>7</sup>	ND-2.8 $\times$ 10 <sup>8</sup>							Kishimba and Mihale (2009)
Vaal River	South Africa	Soil		3.53				0.09		4.77	Quinn et al. (2009)
Vaal River	South Africa	Sediment		1.65				0.05		0.12	Quinn et al. (2009)
Ngarenanyuki	Tanzania	Soil		2270	213			240		0.12	Kihampa et al. (2010)
Lake Victoria	Uganda	Sediment	$2.80\pm2.00$	$4.24 \pm 3.83$	210		$0.45 \pm 0.23$	6.00 ± 4.36	$3.80 \pm 3.55$		Wasswa et al. (2011)
Calabar River	Nigeria	Sediment	10	970			0.45 ± 0.25	0.00 ± 4.30	$3.00 \pm 3.00$		Sojinu et al. (2011)
Bakassi River	Nigeria	Sediment	20	850							Sojinu et al. (2011)
Imo River	Nigeria	Sediment	10	930							Sojinu et al. (2011)
Oginni River	Nigeria	Sediment	70	290							Sojinu et al. (2011)
Densu River Basin	Ghana	Sediment	0.04-1.04	0.04-1.67			0.15-3.29	0.01–14.21	0.03–0.47		Kuranchie-Mensah et al. (2012)
Lake Maryut		Sediment	0.04-1.04 ND-2.20	0.04-1.67		ND – 2.79	0.15-3.29 ND-19.67	ND-2.84	0.03-0.47 ND-0.85	- 0.25–10.49	Barakat et al. (2012a)
Lake Maryut Lake Manzala	Egypt Egypt	Sediment	ND-2.20 ND-3.42	0.07-105.6		ND – 2.79 ND – 0.37		ND-2.04	ND-0.85 ND-0.03	0.25-10.49 ND - 0.57	Barakat et al. (2012a) Barakat et al. (2012b)

(continued on next page)

С.	Olisah	et	al.	
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Table 2 (continued)											
Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane N	Mirex 2Ch	ΣChlordane Endosulfans	ndosulfans	Dieldrin	HCB	Reference
Ondo State	Nigeria	Sediment		ND-57400		ND	ND-81320 N	ND-7040	10-8820	ND- 10910	Okoya et al. (2013)
Lake Qarun	Egypt	Sediment	0.13-100.6	ND-5.88		0.2	0.25 - 15.9		ND-3.41	ND-41.2	Barakat et al. (2013)
Arusha and Mbeye region	Tanzania	Soil	$0.4{-}4.2\times10^7$	$2.4 extrm{}5.4 imes10^{6}$			2	ND-310	$0.042.4\times10^4$		Mahugija (2013)
Tarkwa Bay Lagos Lagoon	Nigeria	Sediment		4.5-227	0.5-45.7	0.6	0.6-80.5		0.6-48.8	9.2–176	Williams (2013)
Nyando River	Kenya	Soil					e	$30.26\pm2.09$			Abong'o et al. (2015)
Lake Volta	Ghana	Sediment	2.3	1.23		0.43		1.34	0.82		Gbeddy et al. (2014)
Ogbesse River	Nigeria	Sediment	5210	850			ß	540	940		Lawrence et al. (2015)
Illushi River	Nigeria	Sediment	4890	026							Ogbeide et al. (2016)
Ogbese River	Nigeria	Sediment	5220	850							Ogbeide et al. (2016)
Owan River	Nigeria	Sediment	4900	290							Ogbeide et al. (2016)
Agboi Creek, Lagos Lagoon	Nigeria	Sediment	2090	139							Ogbeide et al. (2016)
ERL	Sediment guideline			3	NG	0.5			0.02		MacDonald et al. (2000)
ERM	Sediment guideline			350	DN	9			8		MacDonald et al. (2000)
TEL	Sediment guideline			NG	0.32	2.26	5		0.71		CCME (1999)
PEL	Sediment guideline			NG	0.99	4.79	6		4.30		CCME (1999)
ISQG	Sediment guideline			DN	0.94	4.5			2.85		CCME (1999)
ERL - effects low range; ERM - effects median range; TEL - threshold effect level; PEL-probable effect level; ISQG - Interim fresh water sediment quality guidelines, NG - no guidelines. ND – not detected, <sup>a</sup> wet weight, <sup>b</sup> $\mu$ g/kg organic matter.	RM - effects median weight, <sup>b</sup> μg/kg orga	range; TEL - threshol nic matter.	d effect level; P	EL-probable effe	ct level; ISQG - Iı	aterim fresh v	vater sedim	ent quality guidelin	les, NG - no guid	elines.	

Heliyon 6 (2020) e03518

50 cm depths (Kishimba and Mihale, 2009). Alarming levels of ΣDDTs [(2.8 × 10<sup>8</sup> µg/kg dry weight (dw)] and ΣHCHs (6.3 × 10<sup>7</sup> µg/kg dw) in soils were detected. These concentrations were said to be higher than the DDTs contained in commercial formulations. Their high concentrations was attributed to the illegal dumping and storage of obsolete pesticides around the study area since 1986, thus, raising concerns on the health status of people living in the area over the period of reference. These levels were higher than a similar study reported for sediments and soils from 9 districts in Tanzania where the concentrations of DDTs and HCHs were  $6 \times 10^5$  µg/kg dry mass (dm) and  $1.32 \times 10^5$  µg/kg dm respectively for sediments and  $2 \times 10^4$  µg/kg dm and  $5.9 \times 10^4$  µg/kg dm respectively for soils (Henry and Kishimba, 2003). High level of OCPs were also reported in a previous study carried out in soils and sediments collected around Vikuge farm (Elfvendahl et al., 2004; Kishimba et al., 2004) and Old Korogwe (Kihampa et al., 2010).

Kishimba et al. (2004) investigated the level of pesticides in soils and sediments collected from sugar plantations located in Arusha Chini, Kilimanjaro region, Dar es Salaam region, and Mahonda-Makoba Basin in Zanzibar, Tanzania. The targeted contaminants were analysed with GC-ECD/NPD. A lower concentration of pesticides was observed generally in all the sample matrices. For DDTs, the total concentration in soil and sediment samples were up to 700  $\mu$ g/kg and 500  $\mu$ g/kg, respectively whereas  $\Sigma$ HCHs recorded 132  $\mu$ g/kg and 60  $\mu$ g/kg in that order. Results of evaluation of OCPs in environmental compartments from Tanzania shows that these contaminants are still persistent in designated regions in the country despite the ban.

Pazou et al. (2006) determined 20 pesticides, including octachlorostyrene in 37 sediments collected from 9 locations from Oueme River in the Republic of Benin. OCPs in sediments ranged from BDL to  $526 \mu g/kg$  organic matter. DDTs and HCHs dominated the OCP profile in sediments. Other contaminants such as dieldrin and endosulfans were present in significant concentrations. Endosulfans and HCHs were mostly detected in cotton culture areas, while locations that had maize growing farms were largely contaminated with DDTs. In general, most sites recorded pesticide concentrations above the sediment quality guidelines.

Soils from 15 sites in Uwiro village (Arusha City) in Tanzania were randomly sampled in 2009 and analysed for lindane, *p*,*p*'-DDE, *p*,*p*'-DDD, and endosulfan (Kihampa et al., 2010). Samples were collected at a depth of 0-15 cm, and analysis were done by gas chromatographic technique (GC-ECD). Results from the analysis showed that endosulfan sulphate recorded a mean concentration of 240 µg/kg dw and was the most predominant. Such high occurrence (74% in 11 soil samples) was also reported in soil samples collected from a cotton growing area in Mali by Dem et al. (2007). This indicated that endosulfan sulfate (sold as thionex) which was banned in Tanzania several years ago was still in use by a large number of local farmers as at the time of the study. Metabolites of DDT (p, p'-DDE and p,p'-DDD) were detected in 40% and 47% of soil samples with mean concentrations of 148.2 µg/kg dw and 154 µg/kg dw respectively. Thus indicating the persistence of these OCPs in Tanzania despite being banned since 1983 (Wu et al., 2008). In this study, lindane recorded the lowest occurrence of all the pesticides in the samples (33%) with a mean concentration of 212  $\mu$ g/kg dw.

Westbom et al. (2008) determined 13 classes of OCPs in soils collected from Upper Awash State Farms, Ethiopia. Quantification and qualification was done using GC- $\mu$ ECD and GC-MS respectively. In this study, aldrin, dieldrin, endrin and heptachlor were not detectable in all samples while HCHs were detected in a very low concentration in few soils. A significant amount of DDTs and endosulfans were detected in soils while the sum total of these two classes of OCP gave up to 230  $\mu$ g/kg dw and 56000  $\mu$ g/kg dw respectively, signifying a potential threat to the ecosystems and the local community around the study area.

Darko et al. (2008) determined lindane, endosulfan, drins, p,p'-DDE and p,p'-DDT in sediment samples from Ghana using micro-capillary GC-ECD. These OCPs were found in all the sediments (n = 50) analysed and lindane was detected in 68% of the sediments with a total mean concentration of 6.755 ng/g. This is in agreement with a study carried



Figure 2. Percentage distribution of OCPs in soil and sediment from African environment.

out by Ntow (2005) whose total mean concentration of lindane in sediments from Lake Volta in Ghana was 6.56 µg/kg. Endosulfan (9.883  $\mu g/kg$ ) and *p*,*p*'-DDE (8.342  $\mu g/kg$ ) recorded the highest mean concentrations, occurring in 42% and 98% respectively of the total sediments analysed. The mean concentration of dieldrin (0.072 µg/kg) was lower than that of the parent compound – aldrin (0.065  $\mu$ g/kg), indicating that the occurrence of the former in the sediment was not as a result of decomposition of the latter, but of the independent application of both contaminants as suggested by the authors. In general, the concentrations of OCPs in the study were in line with results obtained from two major lakes in Africa; Lake Tanganyika and Lake Victoria (Manirakiza et al., 2002; Kasozi et al., 2006). Endosulfan, aldrin, and dieldrin concentrations ranged from 0.82 to 5.62 µg/kg, 0.22-15.96 µg/kg, and 0.94-7.18  $\mu$ g/kg, respectively in sediments (n = 117) collected from a depth of 0–20 cm from Lake Victoria in Uganda. More so, ΣDDTs concentrations ranged from 0.01 to 4.02  $\mu$ g/kg while  $\gamma$ -HCH, and chlordane ranged from 0.05 to 5.48 µg/kg (Wasswa et al., 2011).

The extent of degradation of pesticide residues buried a few years ago was investigated in soil samples collected in 2009 from various depths (0–220 cm) at different sites (Tengeru, Akheri, ACU maize farm, Mbocu, and Mbimba) in Tanzania using high-resolution GC-MS (Mahugija, 2013). Of all metabolites of DDTs, *4*, *4'*-DDT was the most predominant with a detection frequency of 100% and concentrations ranging from 9 × 10<sup>5</sup> to 4.8 × 10<sup>6</sup> µg/kg dw in soils from Tengeru. The low ratio of (DDE + DDD)/DDT (<1) in most of the sites suggested a little or no occurrence of degradation whilst the high ratio of degradation (>1) in few samples from Mbocu, Tengeru and ACU maize signified the transformation of DDT residues to other compounds. Similarly, HCHs were detected in all the soil samples with concentrations ranging from  $1.4 \times 10^2$  to  $4.2 \times 10^7$ µg/kg dw. Low ratios of  $\alpha$ -HCH/ $\gamma$ -HCH in most soils suggested lindane as the main cause of HCHs contamination while high ratios in few samples were related to the degradation of  $\gamma$ -HCH to  $\alpha$ -HCH. Endosulfan-1 was the most consistent of all endosulfans detected in soil samples from Mbocu, Tengeru, and Akheri vegetable gardens, suggesting an insignificant degradation process. The isomers of chlordane (*cis*-chlordane & *trans*-chlordane) and heptachlor (*cis*-heptachloroepoxide & *trans*-heptachloroepoxide) were detected in samples from Akheri and Mbimba at levels that suggested ineffective degradation. Furthermore, a strong positive correlation existed among compounds from each study site, indicating a common source. The results in general showed a slow rate of degradation with OCPs concentration decreasing with an increase in depth of burial points.

Sediments from 34 sites from Lake Qarun in Egypt were analysed using GC-MS (Barakat et al., 2013). The relative level of pesticides in the samples were observed in the following order: endrin > HCHs > chlordane > aldrin > endosulfans. A significant difference occurred in the spatial variation of contaminants among the locations, which could be as a result of the rate of degradation, the texture of sediments within the locations as well as different input sources strength. Other causes of variations can be linked to desorption and preferential sedimentation (Iwata et al., 1993; Martinez et al., 2010). Except HCHs, the levels of OCPs in sediment samples collected from the north-eastern and eastern regions close to the urban areas of the Lake showed higher concentrations of the analytes. This suggests that the urban area (Faiyum city) was responsible for contamination of the Lake with OCP residues. Comparatively, it was discovered that concentration of OCPs in sediments from the study locations were lower than a similar study carried out few years in the coastal areas of Egypt (Barakat et al., 2012a, 2012b).

In Nigeria, water sediments collected from a cocoa producing area showed high concentrations of *cis*-chlordane,  $\alpha$ -endosulfan, *p.p*'-DDE and



Figure 3. Mean concentration ( $\mu$ g/kg) of OCPs in black bass and red swamp crayfish (Gitahi et al., 2002).

C. Olisah et al.

Table 3. Mean concentration (±star	ndard deviation) and ranges	of OCPs in biota samples fro	m African (µg/kg).

8

Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane	Toxaphene	Mirex	ΣChlordane	Endosulfans	Dieldrin	HCB	References
Lake Kariba	Zambia	Fish <sup>a</sup>		7–70								Berg and Kautsky (1997)
Gamasa	Egypt	Fish <sup>a</sup>		12.5			3.0		2.0			Abd-Allah et al. (1998)
Abu-Quir	Egypt	Fish <sup>a</sup>		7.8			1.4		3.6			Abd-Allah et al. (1998)
Demiatta	Egypt	Fish <sup>a</sup>		7.1			1.6			1.3		Abd-Allah et al. (1998)
Gamasa	Egypt	Bivalve Donax <sup>a</sup>		38.1							0.9	Abd-Allah et al. (1998)
Abu-Quir	Egypt	Bivalve Donax <sup>a</sup>		22.6			2.2		1.1		2.1	Abd-Allah et al. (1998)
Demiatta	Egypt	Bivalve Donax <sup>a</sup>		19.9			2.3		2.8		1.2	Abd-Allah et al. (1998)
Cross cape	Namibia	Fish <sup>a</sup>	5–19	11–1115		10–97		3–159		4–36		Vetter et al. (1999)
Lake Tanganyika	Burundi	Fish <sup>b</sup>	21.2-288.2	68.3–909.1			ND-54.8		ND-36.1	ND-10.2	2.1–19.5	Manirakiza et al. (2002)
Lake Naivasha	Kenya	Red swamp cray fish <sup>a</sup>		9.2	100.5				21.6	34.6		Gitahi et al. (2002)
Lake Naivasha	Kenya	Black bass <sup>a</sup>		78.6	2				2	2		Gitahi et al. (2002)
Dar es Salaam City	Tanzania	Fish <sup>b</sup>		6.6–53.0	BDL-31					BDL-2.7		Mwevura et al. (2002)
Southern Lake Victoria	Tanzania	Fish <sup>a</sup>		ND-30					20–200			Henry and Kishimba (2006)
Vaal River	South Africa	Egret eggs <sup>a</sup>	0.82	24			0.68	0.8			0.78	Bouwman et al. (2008)
Vaal River	South Africa	African dartar eggs <sup>a</sup>	99	260			2	8.8			4.1	Bouwman et al. (2008)
Vaal River	South Africa	Reed cormorant eggs <sup>a</sup>	3.4	300			1.5	2.4			1.7	Bouwman et al. (2008)
Vaal River	South Africa	Ibis eggs <sup>a</sup>	2.3	68			0.3	22			0.9	Bouwman et al. (2008)
Vaal River	South Africa	Plover eggs <sup>a</sup>	4.2	23			0.56	0.63			1.2	Bouwman et al. (2008)
Vaal River	South Africa	Grebe eggs <sup>a</sup>	0.8	46			0.49	0.29			0.88	Bouwman et al. (2008)
Vaal River	South Africa	White fronted plover eggs <sup>a</sup>	1.7	43			0.32	0.52			0.96	Bouwman et al. (2008)
Western Cape	South Africa	Kelp gull eggs <sup>a</sup>	1.3	88			0.81	0.77			5.2	Bouwman et al. (2008)
Lake Bosumtwi	Ghana	Fish <sup>a</sup>		8.877	$0.126\pm0.11$				$0.713 \pm 0.94$	$0.035\pm0.42$		Darko et al. (2008)
Sharkia Province	Egypt	Animal tissue carcasses before cooking <sup>a</sup>		$\textbf{96.17} \pm \textbf{7.12}$	$10.28\pm0.29$	$0.66\pm0.11$				$\textbf{8.55}\pm\textbf{0.36}$	$0.26\pm0.04$	(Sallam and Mohammed, 2008
Sharkia Province	Egypt	Animal tissue carcasses after cooking <sup>a</sup>		$\textbf{57.31} \pm \textbf{4.84}$	$\textbf{4.63} \pm \textbf{0.053}$	$0.38\pm0.08$				$5.98 \pm 0.23$	$1.50\pm0.03$	(Sallam and Mohammed, 200
Crocodile River	South Africa	Fish <sup>a</sup>							10.0-30.0			Ansara-Ross et al. (2008)
KwaZulu Natal	South Africa	Fish <sup>b</sup>		5403-5537								McHugh et al. (2011)
Volta Lake	Ghana	Fish <sup>a</sup>	4.03-13.04	7.96-38.05				4.55-36.62			0.34-1.21	Kuranchie-mensah et al. (201
Lake Ziway	Ethiopia	Fish <sup>a</sup>	0.29-5.10	0.9-61.9				0.17-4.00				Nakayama and Ishizuka (2014
Lake Awassa	Ethiopia	Fish <sup>a</sup>		1.65-409.6				0.85-3.56	ND-42.5			Rosseland et al. (2014)
Roodeplat Dam	South Africa	Fish <sup>a</sup>	$\textbf{288.75} \pm \textbf{123.2}$	$150.9 \pm 87.6$	$179.0\pm119.1$				$\textbf{244.7} \pm \textbf{123.6}$	$\textbf{239.8} \pm \textbf{85.2}$		Barnhoorn et al. (2015)
Rietvlei dam	South Africa	Fish <sup>a</sup>		$336\pm263$	$55\pm7$				$86\pm26$	$77\pm36$		Barnhoorn et al. (2015)
Harbeespoort dam	South Africa	Fish <sup>a</sup>	$79 \pm 17$	$131\pm25$	$71\pm18$							Barnhoorn et al. (2015)
Lake Volta	Ghana	Fish <sup>a</sup>	12.62-41.62	1.26-10.74				0.49-1.02	2.13-3.11	0.10-1.29		Gbeddy et al. (2014)
Ogbesse River	Nigeria	Fish <sup>a</sup>	2610-4200	40-80	130–660				430–1040	80–530		Lawrence et al. (2015)
European commissic	on maximum res	sidue limits <sup>c</sup>		1000	20				100	200		

dieldrin with value range of 30–6990  $\mu$ g/kg, 30–6990  $\mu$ g/kg, 80–19040  $\mu$ g/kg and 10–7620  $\mu$ g/kg respectively, thus, suggesting pesticide contamination from agricultural activities (Okoya et al., 2013). Ogbeide et al. (2016) detected significant levels of ΣHCHs and ΣDDTs in sediments from 3 water bodies (Illushi, Ogbese, and Owan River) in Niger Delta, Nigeria using GC-ECD. The average concentration of **SHCHs** at Illushi, Ogbesse and Owan River were 4089  $\mu$ g/kg, 4080  $\mu$ g/kg and 4900  $\mu$ g/kg dw respectively, while  $\Sigma$ DDTs were 970  $\mu$ g/kg, 1160  $\mu$ g/kg and 930  $\mu$ g/kg dw in that order. These concentrations were higher than those reported for rivers in Niger Delta region and Lagos Lagoon, Nigeria where a widespread distribution of OCPs in sediment was reported (Ize-Iyamu et al., 2007; Lawrence et al., 2009; Williams, 2013). From the data generated from this review, it can be deduced that the OCPs concentration in soils and sediments from Africa were in the following order:  $\Sigma DDTs > \Sigma HCHs >$  endosulfans > dieldrins > HCB >  $\Sigma chlordane$  > lindane > mirex (Figure 2).

#### 2.4. Biota

Pesticides enter into biota through the accumulation of contaminated dust on dermal piercing, inhaling polluted air consuming contaminated food and drinking contaminated water (Winter, 1992; El-Shahawi et al., 2010). EPA (2009) reported that fish are major reservoirs of these pollutants and their concentrations in fish tissues are indicators of bioaccumulation. Significant concentration of OCPs has been detected in fish tissues from aquatic systems in Africa (Kuranchie-mensah et al., 2011; Lawrence et al., 2015). Table 3 describes the concentration of various OCP residues in biotas from different African countries.

Fur seals (*Arctocephalus pusillus pusillus*) blubber from Cape Cross/ Namibia were analysed for toxaphene, dieldrin, chlordane, DDTs and HCHs using GC-MS in electron capture negative and electron impact ionization modes (Vetter et al., 1999). *p*,*p*'-DDT, *p*,*p*'-DDD and *p*,*p*'-DDE were detected in all the samples with concentrations ranging from 3 to 73 µg/kg, 2–59 µg/kg and 6–1030 µg/kg respectively. *p*,*p*'-DDT recorded 80% dominance of the total DDT burden. Interestingly,  $\alpha$ -HCH was below the detection limit in all samples. This was contrary to its level in other species of seals (Luckas et al., 1990). Toxaphene and dieldrin were detected in significant concentrations in all the samples analysed with a concentration ranges of 10–97 µg/kg and 4–36 µg/kg respectively, while chlordane ranged from 3 to159 µg/kg.

Abd-Allah et al. (1998) determined pesticide residues in fish and bivalve *Donax sp* collected from Abu-Quir, Demiatta and Gamasa in Egypt. Results from the samples analysed using capillary GLC showed that p,p'-DDE dominated the DDT profiles in all locations, occurring in a range of 2.0–4.1 µg/kg and 3.3–18.0 µg/kg in fish and bivalve, respectively. The endrin concentrations in fish from Gamasa and Abu-Quir were 0.6 µg/kg and 0.2 µg/kg, respectively, while that of bivalve ranged from ND to 1.5 µg/kg in that order. Heptachlor was not detected in fish samples from Abu-Quir and Demiatta and bivalve from Gamasa and

Abu-Quir. The concentration of Toxaphene (9.7  $\mu$ g/kg) in bivalve from Demiatta was the highest amongst all pesticides.

Gitahi et al. (2002) determined 7 OCPs (p,p'-DDT, o,p'-DDT, p,p'-DDE, lindane, dieldrin,  $\beta$ -endosulfan, and aldrin) in red swamp crayfish (*Procambarus clarkii*) and black bass (*Micropterus salmoides*) collected from Lake Naivasha in Kenya. OCP average concentrations ranged from 16.1 µg/kg (p,p'-DDE) to 100.5 µg/kg (lindane) in black bass and 1.4 µg/kg (p, p' -DDE) to 4.6 µg/kg (o,p' -DDT) in crayfish as shown in Figure 3. In general, the level of OCPs in black bass were higher compared to red swamp crayfish.

Manirakiza et al. (2002) determined 21 classes of OCPs using GC- $\mu$ ECD in 7 fish species (*Boulengerochromis microlepis*, n = 2; *Chrysichthys sianenna*, n = 1; *Oreochromis niloticus*, n = 10; *Stolothrissa tanganikae*, n = 15; *Limnothrissa miodon*, n = 1; *Lates angustifrons*, n = 1, and *Lates stappersii*, n = 1) from certain locations in Burundi. HCB was present in all the fish species with the highest concentration of  $19.5 \pm 3.7 \ \mu g/kg$  fat occurring in *Boulengerochromis microlepis*. Aldrin,  $\alpha$ -endosulfan and endrin were present in 42% of all the samples followed by dieldrin and  $\beta$ -endosulfan which recorded 28% individually. Furthermore, DDTs dominated the total OCPs in all samples. Their concentrations were lower than the European Union permissible level of 1000  $\mu g/kg$  fat. However, the positive correlation that existed between HCHs and DDTs in this study (R<sup>2</sup> = 0.833) may be attributed to the similar manner of bio-accumulation of both contaminants.

Varying levels of DDT metabolites and endosulfan were detected in fish samples collected in July 1999 from 9 districts around Southern Lake Victoria, Tanzania (Henry and Kishimba, 2006). Isomers of endosulfan (a and  $\beta$ ) were more dominant in a wide range of samples, having a ratio ranging from 0.5 to 10. The concentration of total endosulfan in each sample ranged from 400 to 42000  $\mu$ g/kg lipid weight (lw), this is equivalent to 20–200  $\mu$ g/kg wet weight (ww) while a high level of DDT and its metabolites were also recorded in most fish samples, suggesting a recent exposure of fish to DDT contamination. Similarly, DDT and its metabolites were the prevailing OCPs when Pazou et al. (2006) studied the occurrence of 24 OCPs in fish samples from Oueme River catchment in the Republic of Benin. Fish samples were collected from 6 locations around the catchment of Oueme River namely: Kpassa, Atchakpa-Beri, Toue, Bonou, Lowe and Houdedo areas in 2003 and instrumental analysis was carried out using GC-ECD. The results of analysis showed that  $\alpha$ -endosulfan recorded the highest concentration of 57 µg/kg, constituting 68% of all OCPs determined in fish from Lowe area. The total OCPs concentration ranged from BDL to 1364 µg/kg lipid in all fish samples in Oueme River basin, suggesting a low intake of pesticide by people consuming fish from this water body and thus suggesting a little or no risk of exposure to these contaminants on consumption of fish from this location. More so, p,p'-DDT had the highest bioaccumulation factor ranging from 5.7 to 37 gOM/g lw followed by o,p'- DDD, p,p'-DDE,  $\Sigma$  endosulfan and *o*,*p*'-DDT with values ranging from 0.9 to 22 gOM/g lw, 1.0-26 g OM/g lw, 0.6 to 11 gOM/g lipid and 0.13 to 4.2 gOM/g lw



Figure 4. Concentration ( $\mu$ g/kg, mean  $\pm$  SD) of OCPs in fish samples from Lake Bosumtwi collected from 2006 to 2008 (Darko et al., 2008).

<b>Table 4.</b> Mean concentration ( $\pm$ standard deviation) and ranges of OCPs in milk and serum samples from Africa ( $\mu$ g/kg).

Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane	Endosulfans	Dieldrin	HCB	Reference
Cairo	Egypt	Human serum <sup>a</sup>	$17.6\pm46.8$	$67 \pm 109.7$				$0.5\pm0.6$	Soliman et al. (1997)
Nairobi	Kenya	Human milk <sup>b</sup>	96	4–6321	2–134		4–273		Kinyamu et al. (1998)
Siphofaneni	Swaziland	Human milk <sup>b</sup>		ND-1930					Okonkwo et al. (1999)
Akumadan	Ghana	Human blood <sup>b</sup>		$380 \pm 120$				$30\pm10$	Ntow (2001)
Akumadan	Ghana	Human milk <sup>b</sup>		$490\pm230$				$40\pm20$	Ntow (2001)
Jozini	South Africa	Human milk <sup>a</sup> (µg/L)		5.87-1537					Bouwman et al. (2006)
Mkuzi	South Africa	Human milk <sup>a</sup> (µg/L)		3.52-154.85					Bouwman et al. (2006)
Kwaliweni	South Africa	Human milk <sup>a</sup> (µg/L)		2.12-141.79					Bouwman et al. (2006)
Ashanti region and Kassena Nankana District	Ghana	Human milk <sup>b</sup>	$46.4\pm5.5$	$73.3\pm7.0$			$122.8\pm24.8$	$\textbf{4.9}\pm\textbf{0.3}$	Ntow et al. (2008)
Ashanti region and Kassena Nankana District	Ghana	Human serum <sup>b</sup>	$7.3\pm7.0$	$9.1\pm1.3$			$127.0\pm27.2$	$\textbf{5.3} \pm \textbf{1.9}$	Ntow et al. (2008)
Thohoyandou area	South Africa	Human milk <sup>b</sup>		ND-1930					Okonkwo et al. (2008)
Siphofaneni	Swaziland	Human milk <sup>a</sup>		10-8870					
Western cape	South Africa	Human blood <sup>a</sup> (µg/L)				$544.1 \pm 82.1$			Dalvie et al. (2009)
Tunisia	Tunisia	Human milk <sup>b</sup>	ND-189	ND-1100			NA-31		Ennaceur and Driss (2013
Asendabo	Ethiopia	Human milk <sup>b</sup>		17170					Gebremichael et al. (2013
Jimma	Ethiopia	Human milk <sup>b</sup>		14460					Gebremichael et al. (2013
Serbo	Ethiopia	Human milk <sup>b</sup>		6420					Gebremichael et al. (2013
Asendabo	Ethiopia	Cow milk <sup>b</sup>		269					Gebremichael et al. (2013
Jimma	Ethiopia	Cow milk <sup>b</sup>		477					Gebremichael et al. (2013
Serbo	Ethiopia	Cow milk <sup>b</sup>		421					Gebremichael et al. (2013
Elfaw	Sudan	Human blood <sup>a</sup> (ng/mL)	ND-7	56-123			4-43		Elbashir et al. (2015)
Elmanagil	Sudan	Human blood <sup>a</sup> (ng/mL)	2–21	30–187			ND-43		Elbashir et al. (2015)
ElHasahisa	Sudan	Human blood <sup>a</sup> (ng/mL)	3–92	17-380			8–58		Elbashir et al. (2015)
Wad Medani	Sudan	Human blood <sup>a</sup> (ng/mL)	ND-88	69–618			5–18		Elbashir et al. (2015)
Kinana	Sudan	Human blood <sup>a</sup> (ng/mL)	ND-22	21–167			16-82		Elbashir et al. (2015)
Gunaid	Sudan	Human blood <sup>a</sup> (ng/mL)	ND-17	61–13			10–23		Elbashir et al. (2015)
World health organisation perm	issible limit			20					FAO/WHO (2005)

ND - not detected; <sup>a</sup> wet weight; <sup>b</sup> fat/lipid weight.

Table 5. Mean concentration ( $\pm$ standard deviation) and ranges of OCPs in vegetables, fruits and food products from Africa ( $\mu$ g/kg).

11

Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane	Endosulfans	Dieldrin	Reference
Arusha region	Tanzania	Beans <sup>b</sup>		BDL- 99	BDL- 60	BDL-29	ND-24	Rusibamayila et al. (1998)
Arusha region	Tanzania	Maize <sup>b</sup>		BDL-105	BDL-402	BDL-80	ND-23	Rusibamayila et al. (1998)
Arusha region	Tanzania	Wheat <sup>b</sup>		BDL-19	BDL-70	BDL-208	ND-19	Rusibamayila et al. (1998)
South West	Nigeria	Sweet orange <sup>a</sup>	1.2-3.4	1.0-8.5				Adeyeye and Osibanjo (1999)
South West	Nigeria	Tangerine <sup>a</sup>	1.5-4.2	15–5.6				Adeyeye and Osibanjo (1999)
South West	Nigeria	Guava <sup>a</sup>	1.5-8.7	5.3-47.2				Adeyeye and Osibanjo (1999)
South West	Nigeria	Kolanut <sup>a</sup>	2.1-10.2					Adeyeye and Osibanjo (1999)
South West	Nigeria	Banana <sup>a</sup>	1.5-21.4	2.0-5.6				Adeyeye and Osibanjo (1999)
South West	Nigeria	Plantain <sup>a</sup>	1.2-3.3	1.5-3.8				Adeyeye and Osibanjo (1999)
South West	Nigeria	Pawpaw <sup>a</sup>	1.1–3.6	2.0–16				Adeyeye and Osibanjo (1999)
South West	Nigeria	Pineapple <sup>a</sup>	1.5-4.2	2.0-4.5				Adeyeye and Osibanjo (1999)
South West	Nigeria	Bitter Kola <sup>a</sup>	30–321	2.8-17.6				Adeyeye and Osibanjo (1999)
South West	Nigeria	Amaranth <sup>a</sup>	1.4–17.4	1.0-50				Adeyeye and Osibanjo (1999)
South West	Nigeria	Water leaf <sup>a</sup>	1.6-7.2	1.0-33				Adeyeye and Osibanjo (1999)
South West	Nigeria	Leafy vegetables <sup>a</sup>	1.0-24.2	1.2-130				Adeyeye and Osibanjo (1999)
South West	Nigeria	Egg plant <sup>a</sup>	4.0-24.2	1.2-6.4				Adeyeye and Osibanjo (1999)
South West	Nigeria	Red pepper <sup>a</sup>	1.5–14.4	5.1-66.6				Adeyeye and Osibanjo (1999)
South West	Nigeria	Tomato <sup>a</sup>	2.1-7.1	12.2-102				Adeyeye and Osibanjo (1999)
South West	Nigeria	Okro <sup>a</sup>	1.4-12.1	1.0-8.9				Adeyeye and Osibanjo (1999)
South West	Nigeria	Onion <sup>a</sup>	1.5-8.4					Adeyeye and Osibanjo (1999)
Banjul and Dakar	Sene-Gambia	Sweet potato <sup>a</sup>	0.4–0.6	ND-0.13				Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Egg plant <sup>a</sup>	ND-0.3	ND-5.1		ND-1.2	ND-2.19	Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Cabbage <sup>a</sup>	ND-1.1	ND-5.03		ND – 2.1	ND – 1.77	Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Tomato <sup>a</sup>	ND-0.9	ND-1.05		ND-0.14		Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Radish <sup>a</sup>	0.3	1.44		0.33	0.49	Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Sweet pepper <sup>a</sup>	ND-0.1	ND-0.16		ND-0.14		Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Turnip <sup>a</sup>	ND-0.3	0.07-2.39		ND-0.66	ND-0.74	Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Onion <sup>a</sup>	1.7	4.53		2.68		Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Red pepper <sup>a</sup>		2.89				Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Lettuce <sup>a</sup>	ND-0.3	0.06-0.45				Manirakiza et al. (2003)
Banjul and Dakar	Sene-Gambia	Carrot <sup>a</sup>	ND-0.03	0.12-3.14				Manirakiza et al. (2003)
Dar es Salaam City	Tanzania	Spinach <sup>a</sup>	0.08	2.89				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Cassava leaves <sup>a</sup>	0.13	0.4				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Stiff porridge <sup>a</sup>	0.06	0.3				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Rice <sup>a</sup>	0.08	1.7				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Cooked banana <sup>a</sup>		2.08				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Beans <sup>a</sup>	0.04	0.13				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Pigeon peas <sup>a</sup>		7.6				Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Fish variety <sup>a</sup>	0.05					Ndengerio-Ndossi and Cram (200
Dar es Salaam City	Tanzania	Beef meat <sup>a</sup>	0.14	0.76				Ndengerio-Ndossi and Cram (200
	Tanzania	Eucalyptus sp leaves <sup>a</sup>	15	818				Marco and Kishimba (2006)
Vikuge farm		· · ·						
Vikuge farm Vikuge farm	Tanzania	Cassava leaves <sup>a</sup>	ND-2	7-425				Marco and Kishimba (2006)

(continued on next page)

Table 5 (	continued)
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Location	Country	Matrix	ΣHCHs	ΣDDTs	Lindane	Endosulfans	Dieldrin	Reference
Vikuge farm	Tanzania	Plum leaves <sup>a</sup>		4				Marco and Kishimba (2006)
Vikuge farm	Tanzania	Cashew leaves <sup>a</sup>		16				Marco and Kishimba (2006)
Kumasi	Ghana	Beef fat <sup>a</sup>		663.7	$4.04\pm3.49$	$21.35\pm3.85$	$5.23 \pm 2.76$	Darko and Acquaah (2007)
Kumasi	Ghana	Beef <sup>a</sup>		61.7	$2.07 \pm 1.31$	$1.88\pm0.42$	$5.92\pm0.05$	Darko and Acquaah (2007)
Buoho	Ghana	Beef fat <sup>a</sup>		435.7	$1.79\pm0.38$	$2.28 \pm 1.74$	$6.01\pm5.14$	Darko and Acquaah (2007)
Buoho	Ghana	Beef <sup>a</sup>		16.7	$0.60\pm0.38$	$0.59\pm0.38$	$11.48 \pm 4.98$	Darko and Acquaah (2007)
Aboaba	Ghana	Local cheese <sup>a</sup>		163.1		0.57-9.06	0.88-30.49	Darko and Acquaah (2008)
Гаfo	Ghana	Local cheese <sup>a</sup>		438.7	BDL-4.41	0.14-8.21	1.41-30.5	Darko and Acquaah (2008)
Asawasi	Ghana	Local cheese <sup>a</sup>		73.7		1.29-4.88	1.21-3.94	Darko and Acquaah (2008)
Ayeduasi	Ghana	Yoghurt <sup>a</sup>		10.6		0.02-0.12	BDL-0.34	Darko and Acquaah (2008)
KNUST	Ghana	Yoghurt <sup>a</sup>		4.58	0.01-0.05	BDL-0.34	0.01-0.03	Darko and Acquaah (2008)
K-Poly	Ghana	Yoghurt <sup>a</sup>		8.07	BDL-0.01	0.01-0.14	BDL-0.03	Darko and Acquaah (2008)
Accra	Ghana	Pawpaw <sup>a</sup>	100	100				Bempah and Donkor (2011)
Accra	Ghana	Tomato <sup>a</sup>	10	20	20	30		Bempah and Donkor (2011)
Accra	Ghana	Imported apple <sup>a</sup>	100		60			Bempah and Donkor (2011)
Accra	Ghana	Pawpaw <sup>a</sup>	BDL-60	BDL-30	20-40	BDL-30		Bempah et al. (2011)
Accra	Ghana	Tomato <sup>a</sup>	10–20	BDL-10	BDL-20			Bempah et al. (2011)
Accra	Ghana	Imported apple <sup>a</sup>	10–20	BDL-90	BDL-20			Bempah et al. (2011)
Gelemso	Ethiopia	Khat <sup>a</sup>		673.5–1372				Daba et al. (2011)
BadaBuna	Ethiopia	Khat <sup>a</sup>		ND				Daba et al. (2011)
Aseno	Ethiopia	Khat <sup>a</sup>		256.5-1223.8				Daba et al. (2011)
Accra	Ghana	Carrot <sup>a</sup>		156	220			Bempah et al. (2012)
Accra	Ghana	Cabbage <sup>a</sup>		402	196			Bempah et al. (2012)
Accra	Ghana	Lettuce <sup>a</sup>		186	24			Bempah et al. (2012)
Accra	Ghana	Tomato <sup>a</sup>		571	27		6.0	Bempah et al. (2012)
Olomoro	Nigeria	Higher plants <sup>b</sup>	$116.09\pm95.12$	$\textbf{6.40} \pm \textbf{7.81}$		30.2	$1.66 \pm 1.91$	Sojinu et al. (2012)
Oginni	Nigeria	Higher plants <sup>b</sup>	$78.29 \pm 86.54$	$1.58\pm0.04$		7.69	0.14	Sojinu et al. (2012)
Uzere	Nigeria	Higher plants <sup>b</sup>	$70.82\pm96.35$	$2.73 \pm 2.37$		6.47	0.11	Sojinu et al. (2012)
rri and Calabar	Nigeria	Higher plants <sup>b</sup>	$55.55\pm22.54$	$2.65 \pm 1.37$		17.2	$\textbf{0.58} \pm \textbf{0.46}$	Sojinu et al. (2012)
	Togo	Tomato <sup>a</sup>	BDL-11.307	BDL- 0.78			BDL-0.194	Kolani et al. (2016)
	Togo	Cabbage <sup>a</sup>	BDL-93.83	BDL-1.518			BDL-0.086	Kolani et al. (2016)
	Togo	Lettuce <sup>a</sup>	0.150-2.634	BDL-1.236			BDL-0.010	Kolani et al. (2016)

respectively. Fish samples collected from 8 locations in the coastal area of Dar es Salaam City showed that p,p'-DDE and p,p'-DDD dominated the DDT profile, occurring in all the samples with a concentration range of 3.1–53.0 µg/kg lw while dieldrin and  $\delta$ -HCH ranged from BDL to 2.7 µg/kg lw and BDL to 1.0 µg/kg lw respectively (Mwevura et al., 2002).

Cormorant, darter, cattle egret, and ibis eggs from South Africa sampled between November 2004 and March 2005 were analysed for 14 OCPs using a high-resolution GC-ECD (Bouwman et al., 2008). The concentration of HCB,  $\Sigma$ chlordanes,  $\Sigma$ DDTs,  $\Sigma$ HCHs and p,p' –DDE in cattle egret eggs from Barberspan and Vaal River were 0.78 µg/kg ww, 0.80 µg/kg ww, 24 µg/kg ww, 0.82 µg/kg ww, and 24 µg/kg ww respectively. These values were lower compared to that of African darter, Reed Cormorant, and African sacred ibis. Mirex which is an unregistered OCP residue in South Africa was also detected in all the samples with the highest concentration of 2.0 µg/kg ww recorded in African darter eggs from Vaal River. The authors suggested that the occurrence of mirex in the South African environment may be attributed to its usage in various products as a flame retardant. It was recommended by the authors that monitoring of organic pollutants in the terrestrial and aquatic environment can be done using cattle egret and African darter. The residual contents of DDTs, HCHs,  $\delta$ -HCH, aldrin endrin, dieldrin, HCB, toxaphene, and chlordane in the muscles, liver and kidney from carcasses of 30 Camel, 30 Cattle and 30 Sheep, slaughtered in Sharkia Province, Egypt were analysed by Sallam and Mohammed (2008). Sampling was carried out between July and September 2005 and targeted OCPs were determined using GC-ECD. The results showed that over 30% of the carcasses contained dieldrin aldrin, DDTs, lindane and HCHs while HCB, chlordane and toxaphene where only detected in 10% of the carcasses. Levels of all OCPs detected where reduced by more than 20% when samples were subjected to heat (1.5 h of boiling). Lindane recorded the highest reduction rate of 58.6 % in meat muscle. Results from this study revealed that OCPs in all samples analysed from the 3 tissues were below their respective maximum limits stipulated by local or international organisations.

In another study, *p*,*p*'-DDE and *p*,*p*'-DDT were the predominant pesticides with mean concentrations of 5.232 µg/kg and 3.645 µg/kg respectively, when 50 fish samples, collected randomly from Lake Bosumtwi, Ghana were analysed for OCPs (Darko et al., 2008). Other OCPs recorded an average concentration of 0.126 µg/kg for lindane, 0.035 µg/kg for dieldrin, 0.713 µg/kg for endosulfan, and 0.018 µg/kg for aldrin. This indicates that the level of contaminants were in the order: p,p'-DDE > p,p'-DDT > endosulfan > lindane > dieldrin > aldrin (Figure 4).

The possible effect of DDTs on the health status of Tigerfish (*Hydrocynus vittatus*) was also evaluated in fish samples collected from a low risk malaria area in KwaZulu Natal, South Africa. The location was chosen because of the usage of DDT for vector control in the area. Samples (n = 45) were collected in 2009 and analysed using GC-MS while other analytes were estimated using histological based techniques. The total DDTs analysed ranged from 5404 to 5537  $\mu$ g/kg lw (McHugh et al., 2011). Levels of DDT were also evaluated in fish samples from Vaal River (Wepener et al., 2011) and it was deduced by the authors that the high level of OCPs found in fish tissues were as a result of commercial farming activities taking place within the area.

Twenty *Clarias gariepinus* samples were collected from 3 Dams in Gauteng South Africa for OCPs analysis (Barnhoorn et al., 2015). Sampling was done in the summer months of 2007 while instrumental analysis was carried out using GC-MS. OCPs were detected in over 15% (n = 3) of the total fish samples from Roodeplat dam (RDPD). *p*,*p*'-DDE, endosulfan, dieldrin, and lindane were the most predominant detected in the fish tissues from RDPD with a concentration range of 77.0–350 µg/kg wet mass (wm), 141.4–516 µg/kg wm, 128.5–314.5 µg/kg wm, and 108.4–514.8 µg/kg wm, respectively. *p*,*p*'- DDT, methoxychlor, and heptachlor were only detected in one of the fish sample while other

samples revealed the presence of endrin aldehyde, aldrin,  $\alpha$ -HCH, and  $\delta$ -HCH. A detectable level of DDTs, endosulfans, heptachlor, endrin, and δ-HCH were observed in fish tissues from Rietvlei dam (RVD). p,p'-DDE (n = 10) and endosulfan II (n = 9) were the most prevalent contaminants while *p*,*p*'- DDE ranged from 72.4 to 426  $\mu$ g/kg wm. Only *p*,*p*'- DDE,  $\beta$  – HCH and  $\delta$ -HCH were detected in fish tissues from Hartbeespoort Dam (HBPD) with values ranging from 101.9 to 161 µg/kg wm, 60.7-111.5  $\mu$ g/kg wm, and 52.4–95.9  $\mu$ g/kg wm, respectively. From the results, the health risk associated with fish consumption from RDPD was mainly from dieldrin and  $\gamma$ -lindane while the cancer and non-cancer risk for RVD was below that of RDPD. This was due to the levels of DDTs, heptachlor, dieldrin and d-HCH whose cancer risk values were higher than the threshold value of  $10^{-6}$ . In view of the findings from the reviewed articles, consuming fish from Cross Cape (Namibia), Oueme River (Republic of Benin), Ogbesse River (Nigeria) and Roodeplat Dam (South Africa) may pose serious health risk since DDTs were found in concentrations above the FAO stipulated limits (FAO, 1983). Apart from the carcinogenic risk, these chemicals have been proven to have the potential of causing diabetes (Cox et al., 2007).

#### 2.5. Blood and milk

There are possibilities for the occurrence of OCPs in human fluids due to their lipophilic nature. Various ways have been reported as possible routes in which these pollutants get into humans and animals fluids (El-Shahawi et al., 2010). Not much works have been done on evaluating this contaminants in human fluids in Africa. OCPs level in serum and milk of humans and animals are shown in Table 4. In a study carried out by Kinyamu et al. (1998), 9 OCPs were detected in 216 samples of breast milk collected from nursing mothers in a Hospital in Nairobi, Kenya. *p*, *p*'-DDE was the highest residue detected, occurring in 99.5% of the total milk samples collected while DDT and its metabolites ranged from 4 to 6321 µg/kg milk fat in all milk samples. Dieldrin was detected at a mean concentration of 19 µg/kg milk fat while  $\alpha$ -HCH,  $\beta$ -HCH, and lindane were detected at average concentrations of 13 µg/kg, 83 µg/kg and 19 µg/kg milk fat respectively.

Breast milk (n = 190) and serum (n = 115) of farmers from Ghana sampled in 2005/2006 were analysed for DDTs, HCHs, HCB, and dieldrin using GC-ECD and GC-MS (Ntow et al., 2008). Eighty-eight percent and 75% of the breast milk and serum samples respectively, were positive for DDTs. Of all the isomers of DDT, DDE had the highest mean concentration (lw) of 7.1  $\mu$ g/kg in serum and 44.8  $\mu$ g/kg in breast milk while the mean sum of the HCH isomers were 7.3  $\mu$ g/kg lw in serum and 46.4  $\mu$ g/kg in breast milk. In this study, there was a positive correlation between OCPs and the age grade of the milk donors. Furthermore, the accumulated DDTs and HCHs in breast milk in some farmers were above the daily amount of DDT recommended by Health Canada.

Okonkwo et al. (2008) analysed DDTs (using GC-MS) in human milk collected from breastfeeding mothers within the age bracket of 19 and 40 years residing in Limpopo province, South Africa. About 5–50 mL of breast milk samples were collected from 30 mothers in 10 villages (Lufule, Mangondi, Dumasi, Budeli, Tshikhudini, Tshilungoma, Gondeni, Makhuva, and Thohoyandou) within the province. Questionnaires were issued to each mother to collect data of their diet, frequency of DDT application, number of children and age. DDT and its metabolites from the study locations ranged from ND to 1930  $\mu$ g/kg fat weight, the occurrence of DDTs in the study area were in the following order:  $\Sigma DDE > \Sigma DDD > \Sigma DDT$ . The level of DDTs were lower than those present in breast milk from lactating mothers in Siphofaneni, Swaziland (10–6470  $\mu$ g/kg; Okonkwo et al., 1999).

Bouwman et al. (2006) carried out similar studies and detected a higher level of  $\Sigma$ DDTs (4797 µg/kg milk fat) in human breast milk from women in Jozini in KwaZulu Natal province of South Africa compared to those in Kwaliweni (799 µg/kg milk fat). This was 15 times lower than a



Figure 5. Concentration ( $\mu g/kg$ , mean  $\pm$  SD) of OCPs in vegetables collected from Togo (Kolani et al., 2016).

study carried out 14 years ago in the same location (Bouwman et al., 1992).

A comparative study of the occurrence of pesticides in blood of residents performing first post spraying exercise and those who were occupationally exposed to endosulfan on a farm around the Valley Hex River in the Western Cape Province of South Africa was carried out by Dalvie et al. (2009). Analysis of endosulfan in blood was done using GC-ECD. This was performed 24 h before the first spraying season and a day after application. Information from farmers on the method of application, use of personal protective equipment, medical history, and conditions of living were taken during the testing period. Also, the job-exposure matrix was calculated using a method developed by London and Myers (1998). The baseline concentration of endosulfan of all workers had a mean concentration of 524.2  $\pm$  49.5  $\mu g/L$  while an increase of this contaminant of a better part of workers involved in post-spraying was observed (mean concentration - 544.1  $\pm$  82.1  $\mu$ g/L). The correlations between gender, drinking of alcohol, smoking and levels of endosulfan after spraying were insignificant. At the same time, there was a positive correlation between age and endosulfan concentrations in workers after spraying. This was also the case when age was correlated with high levels of DDTs,  $\beta$ -HCH and HCB found in human serum collected from colorectal cancer patients in Egypt during March and April 1996 (Soliman et al., 1997).

Gebremichael et al. (2013) studied OCPs in breast milk from mothers and cows residing in 3 towns in Addis Ababa. These locations were selected based on the frequent use of DDTs for mosquito control in the area. Milk samples from 101 mothers were collected in 2010, composited together for each location and analysed using GC-µECD. The average concentration of  $\Sigma$ DDTs in human milk from the 3 locations ranged from 6420 to 17170 µg/kg milk fat. In this study, the mean level of DDT estimated to be ingested by infant from mothers was 62.17  $\mu$ g/kg. This value is said to be approximately 3 times higher than the recommended daily intake limit stipulated by WHO/FOA (20 µg/kg of body weight) and consequently a call for concern on the health status of children feeding on breast milk from mothers in the study area. Moreover, the mean concentrations of  $\Sigma$ DDTs in cow milk from the 3 locations ranged from 269 to 477 µg/kg lipid. The estimated daily intake (EDI) was calculated using 60 kg body weight and a cow milk consumption of 350 mL per person. It was deduced that the EDI of DDT were below the FAO/WHO recommended limit.

Most of the studies from the reviewed articles showed that DDTs were found above the stipulated limit of FAO/WHO. The rapid transformation of *p*,*p*'-DDT to *p*,*p*'-DDE may be responsible for the high concentration in human fluid since this congener has a long-lasting time in the body (Jaga and Dharmani, 2003). Infact, high ratio of *p*,*p*'-DDT to *p*,*p*'-DDE in breast milk was reported in countries where the use of DDTs have been stopped several years ago (Gebremichael et al., 2013). The high concentration in the reviewed studies may be attributed to improper handling of pesticides during farming or indoor applications of DDTs for vector control, which may result to the contamination of food stuffs, kitchen utensils and other domestic items that could easily have direct contact with the mouth.

#### 2.6. Vegetables, fruits and food products

The use of pesticides for the control of vectors during cultivation of various agricultural produce is responsible for their varying levels in vegetables, fruits and food products in Africa (Bempah and Donkor, 2011; Darko and Acquaah, 2008; Manirakiza et al., 2003; Sojinu et al., 2012) as shown in Table 5. High levels of DDTs, endosulfan, lindane and dieldrin were detected in beans samples collected from Arusha city in Tanzania in 1992 in concentrations ranging from BDL to 99 µg/kg, BDL to 29 µg/kg, BDL to 60 µg/kg and BDL to 24 µg/kg respectively. These compounds were also found in maize and wheat samples analysed. Detected levels were above FAO/WHO permissible limits, posing a significant health risk on its consumption (Rusibamavila et al., 1998). This was disputed few years later when Ndengerio-Ndossi and Cram (2005) determined the levels of pesticides in table-ready foods (spinach, cassava leaves, stiff porridge, rice, cooking banana, beans, pigeon peas, fish varieties and beef meat) commonly eaten in Tanzania. Samples were collected from Tandale, Buguruni and Temeke in Tanzania and sample sizes were determined using the following equation:

$$n = \frac{N\sigma^2}{\left(\frac{N\delta^2}{4} + \sigma^2\right)}$$

Where n describes the desired sample size, N is the total population,  $\sigma$  is the standard deviation and  $\delta$  is the error margin (Manly, 1992). Instrumental analysis was done by GC-ECD. *p*,*p*'-DDE was the dominant OCPs in all samples followed by  $\delta$ -HCH and *p*,*p*'-DDT. The highest mean concentration of pesticides (2.89 µg/kg) was found in 72.7% of total spinach samples analysed, followed by rice (1.70 µg/kg), beef meat (0.76 µg/kg), stiff porridge (0.30 µg/kg) and beans (0.13 µg/kg) detected in 50%, 42.1%, 8.6% and 6.7% samples respectively.

Data from this study was below the WHO/FAO Codex Alimentarius daily acceptable intake limits thus revealing that the concentration of OCPs in all food samples were not high enough to pose any significant health threat to humans. With this, the authors refuted the claims that pesticides were responsible for thousands of death in Tanzania annually.

The pattern and distribution of 11 pesticides from an obsolete storage site at Vikuge farms in Tanzania were monitored using the leaves and roots of *Manihot esculenta* (cassava), the leaves of *Eucalyptus sp, Anacardium occidentale* (cashew) and *Prunus domestica* (plum) (Marco and Kishimba, 2006). Choice of plant parts was based on the solubility of OCPs in the epicuticular wax found in plants (Reischl et al., 1989). Samples were collected in September 2002 from 5 locations around the point source and analysis was done using GC-ECD. A wide distribution pattern of DDTs was observed in all matrices of samples, accounting for the 100% detection of *p*,*p*'-DDT and *p*,*p*'-DDD. Furthermore, leaves of *Eucalyptus sp*, collected from the point source, recorded the highest mean concentration of DDTs (818 ng/g) and HCHs (9 ng/g), these levels were higher than those detected in leaves of *M. esculenta* and *P.domestica*. Meanwhile, level of DDTs in cassava roots found 4 km away from the point source were higher than the FAO/WHO limit of 200 ng/g. In addition, the ratio of DDE/DDT was very low in all samples, signifying that the presence of non-degraded DDTs. A strong positive correlation existed between concentration of  $\Sigma$ DDTs and  $\Sigma$ HCHs in all samples (p < 0.01) indicating a common source. The findings revealed that pesticide levels decreased in samples collected in points far from the point source.

Raw meat samples from 2 abattoirs in Ghana (Kumasi and Buoho), collected between 2004 and 2006 were analysed for OCPs using GC-ECD (Darko and Acquaah, 2007). Beef fat samples from Kumasi and Buoho showed a higher concentration of DDTs compared to other OCPs with values ranging from 254.26 to 905.10 µg/kg and 37-844.28 µg/kg respectively. The maximum values of  $\Sigma$ DDTs in beef fat from these two abattoirs were reported to be almost twice the value of WHO limit of 500  $\mu$ g/kg. Also, the mean concentration of dieldrin (6.01  $\pm$  5.14  $\mu$ g/kg) in beef fat from Buoho was slightly higher than the limit stipulated by WHO  $(6.00 \ \mu g/kg)$  while other contaminants were all detected in concentrations that were slightly lower than their respective WHO recommended limits. In general, beef fat samples accounted for higher OCPs compared to beef in both abattoirs thereby making evident the lipophilic nature of chlorinated hydrocarbon. Furthermore, the authors inferred that the occurrence of OCPs in these samples may be attributed to the presence of pesticides in the Cattle's feeds or the application of pesticides on the body of Cattles for the control of parasites.

Similarly, GC-ECD was used to quantify OCPs in food crops purchased from supermarkets and open markets in Ghana in 2007 and 2008 (Bempah and Donkor, 2011). Samples of tomato (n = 110), pawpaw (n = 110) and apples (n = 110) were analysed for 16 OCP residues. Samples of pawpaw from Agbogbloshie, a popular open market dominated by rural farmers in Accra, showed a high average concentration of pesticides, notably  $\delta$ -HCH (60  $\mu$ g/kg), indicating the application of pesticides to food crops by farmers in rural communities. This was lower than the mean concentration of lindane reported in Nigeria food crops (Adeyeye and Osibanjo, 1999). Also,  $\delta$ -HCH were detected in tomato samples collected from all locations. AT the same time, heptachlor epoxide, DDTs,  $\gamma$ -chlordane, alpha endosulfan and methoxychlor were only present in samples collected from the open market. The average concentration of heptachlor (20  $\mu$ g/kg),  $\gamma$ -HCH (20  $\mu$ g/kg), heptachlor epoxide (40  $\mu$ g/kg) and  $p_{p}$ '-DDT (10 µg/kg) were below that found in tomato samples from a vegetable farming community in Akumadan, Ghana (Ntow, 2001). Also, the mean concentration of endrin aldehyde, endrin ketone, p,p'-DDE and *p,p*'-DDT in the imported apples were 110  $\mu$ g/kg, 100  $\mu$ g/kg, 10  $\mu$ g/kg and 90 µg/kg respectively. These values were far lower than those reported in food crops in Shanghai, China (Nakata et al., 2002). In general, the study revealed that the OCPs detected in these food crops were lower than their respective stipulated Codex Alimentarius Commission limits (FAO/WHO, 1998).

Likewise, the quantification of pesticide residues present in 240 vegetable samples (carrot, cabbage, lettuce, and tomato) collected from different market centres in Ghana were carried out by Bempah et al. (2012). DDT and its metabolites dominated the total OCPs found in vegetables from supermarkets with the highest concentration being *o*, *p*-DDT (239 µg/kg) in tomato. Vegetables which had the highest mean concentration of pesticides in samples collected from supermarkets, roadside grocery stores and open market were in the following order: tomato > cabbage > carrot > lettuce, cabbage > tomato > lettuce > carrot and tomato > lettuce > cabbage > carrot respectively. However, only 31.48% of vegetables had pesticide residues above the European commission maximum residue levels.

A study conducted by Daba et al. (2011) showed high concentration of p.p'-DDT in khat samples from 3 study areas in Ethiopia, Galemso,

Asena and Bada Buna. Amongst the three locations, Galemso and Asena had levels of p,p'-DDT ranging from 141 to 973 µg/kg and 194–990 µg/kg respectively, while the level of aldrin was below the limit of quantification. The authors inferred that the high level of p.p'-DDT might be a contributing factor to pathological diseases that exist among khat users.

Essumang et al. (2013) determined pesticides in 500 okra samples close to a watermelon farm that uses these contaminants for its cultivation process in Ghana. The OCPs' mean concentration in okra samples ranged from 3.10 to 7.60 µg/kg. These values were below the WHO/-FOA recommended residue limits. Although not existing in levels that pose a threat to life, their presence is of great concern, and pre-emptive measures must be taken to prevent further contamination. The authors inferred that since the application of pesticides affects neighbouring crops, crop mixing might not be advisable. In a recent study, Kolani et al. (2016) monitored the level of 20 pesticides in 3 types of vegetables (lettuce, cabbage, and tomato). These vegetables were chosen because of their dominance in diet among the Togolese. Analysis of OCPs was done using GC-ECD. From the results, **ZHCHs** recorded the highest pesticides with a mean concentration of 5.847 µg/kg while  $\Sigma$ heptachlors was the lowest with an average concentration of 0.012  $\mu$ g/kg. In general, the occurrence of OCPs in all the vegetables were in the following order:  $\Sigma DDTs > \Sigma chlordanes > \Sigma drins > \Sigma heptachlors$ (Figure 5).

# 3. Discussions

Environmental samples from Vikuge Farm in Tanzania was adjudged to be the most polluted from the reviewed articles due to the accumulation of OCPs resulting from illegal dumping and storage of obsolete pesticides since 1986, thus suggesting the need for clean-up of pollutants from the area. Several countries in Africa lack information on the occurrence of these contaminants in their environment, hence making it difficult to ascertain the pollution load in the Africa ecosystem. Over 90 % of the studies employed GC-MS and GC-ECD for analyses of OCP residues. This might be due to a lack of collaboration between researchers in the African continent and other researchers in developed countries. Hence, there is need for the use of more sensitive analytical tools such as gas chromatograph/tandem mass spectrometer (GC-MS/MS), gas chromatograph/high-resolution mass spectrometer (GC-HRMS) in order to detect their secondary and tertiary metabolites, especially in water, biota, and food products since these are major media through which these pollutants get into humans and animals.

Also, most studies have been restricted to the analysis of OCPs in fish. Other aquatic animals such as Crab, Mussels, Periwinkles and Shrimps which are choice foods in Africa should attract further studies to thoroughly monitor the levels of bioaccumulation of these pesticides in aquatic systems and the ecosystem at large. There was limited data on the occurrence of OCPs in ambient air and tissues of animals and humans. There is, therefore, the need for more studies on these matrices to investigate the potential risk they pose on the health status of humans and animals and also to evaluate the levels of these chemicals in human fluids especially in countries where they are used more often. More studies have been reported on the occurrence of OCPs in Southern and Western Africa compared to other parts of the continent probably due to challenges such as lack of facilities and infrastructures as well as lack of political will and priorities of governments in such countries. Other problems may be related to lack of environmental awareness of this pollutants, the incompetence to analyse POPs among researchers, the convenience during sampling (most researchers tends to have scientific goals other than providing a geographic distribution of OCPs in the environment; therefore, sampling close to home is more convenient). To this end, the need for government and academic partnership is proposed to enable proper documentation of the occurrence, distribution, sources and health impact of OCPs (and indeed other pollutants) in their respective countries.

Generally, studies have revealed that the main challenges facing pesticide usage include inadequate enforcement of policies to checkmate the importation of these chemicals, levity in measures to stop their usage, improper pesticide handling by farmers and local users, and non-existence of training programs on the use of pesticide. These practices have led to a high concentration in environmental and biological matrices. Hence, it is advised that breastfeeding of infants by mothers living in locations where the usage of DDTs and other pollutants is rampant should be discouraged since this is an obvious medium in which these pollutants get into infants and also more models should be developed to carry out health risk assessment in infants. Furthermore, training of farmers and other domestic users on pesticides handling is highly recommended. In addition to the above-stated recommendation, the following further perspective on OCPs monitoring in the African environment should be considered.

- There is a paramount need for an organised monitoring plan in the African environment, with respect to the geographical distribution of this pollutant class. This plan should be designed to cover remote and rural areas in the continent, especially where these pollutant class has been used extensively in the past.
- Concerning DDTs, some reports have shown that this chemical is intentionally used to fight critical vectors, especially in malariaendemic areas in Africa. Appropriate monitoring schemes should be established and well documented. DDT will be present in the environment for decade's therefore long term monitoring is germane in securing the environment.
- Further studies in the African environment should be designed to monitor its level in the ecosystem components not only on a single matrix (sediment or water) as noticed in majority of the reviewed literature. This will provide an in-depth understanding of the mobility of this pollutant class in the African environment.
- Data from the reviewed articles revealed that most researchers focus their study on "hot-spots". This practice should be continued, and more sampling sites should be established, bearing in mind that these chemicals have the ability to transport long-range, and besides they take several years to phase out from the environment. We, therefore, recommend inventories of pesticides contamination in the African environment as well as statistical sampling design to capture newly established sites and hot-spots.

## 4. Conclusions

Data from published articles on the occurrence of OCPs in air, biota, water soil, sediment, vegetables, and food products in Africa environment reveals that OCPs are widely distributed in different environmental compartments in Africa, especially HCHs, DDTs, and endosulfans. Such occurrences might be due to the historical application of this pollutant class for an extended period in the region. Despite being banned several years ago, many individual OCP is still being used for various functions. From the reviewed literature, it is difficult to tell whether or not the concentration of OCPs has decreased or increased in the African environment over the years since its level of occurrence appears to be fluctuating. The current review revealed that DDTs were detected in over 90% of the biota and breast milk studied at a relatively high concentration with some samples occurring at levels above the recommended daily human intake stipulated by WHO. Such high concentrations may be attributed to runoff from agricultural farmlands, accumulated levels in the food chain and frequent use in malaria dominated areas. More so, DDTs, endosulfans, and HCHs were among the prevalent pesticides detected in water, soils, sediments, vegetation and food products, probably a consequence of their use for the control of vectors and the cultivation of food crops.

## Declarations

#### Author contribution statement

All authors listed have significantly contributed to the development and the writing of this article.

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#### Competing interest statement

The authors declare no conflict of interest.

#### Additional information

No additional information is available for this paper.

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#### C. Olisah et al.

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