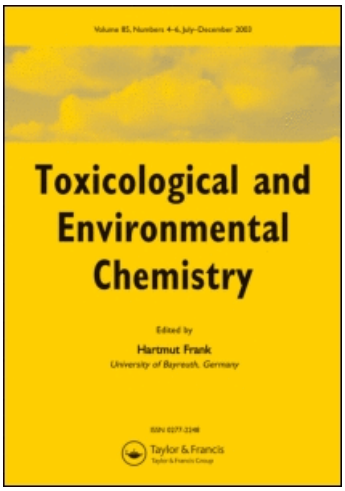

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M. W. Barasa [a]; J. O. Lalah [b]; S. O. Wandiga [c]
[a] Faculty of Science and Engineering, Department of Chemistry, Western University College of Science and Technology, Kakamega, Kenya [b] Faculty of Science, Department of Chemistry, Maseno University, Maseno, Kenya [c] Faculty of Science, Department of Chemistry, University of Nairobi, Nairobi, Kenya

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Seasonal variability of persistent organochlorine pesticide residues in marine fish along the Indian Ocean coast of Kenya

M.W. Barasa[a], J.O. Lalah[b]* and S.O. Wandiga[c]

[a]*Faculty of Science and Engineering, Department of Chemistry, Western University College of Science and Technology, P.O. Box 190, Kakamega, Kenya;* [b]*Faculty of Science, Department of Chemistry, Maseno University, P.O. Box 333, Maseno, Kenya;* [c]*Faculty of Science, Department of Chemistry, University of Nairobi, P.O. Box 30197, Nairobi, Kenya*

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Organochlorine pesticide residues were analyzed in samples of fish taken from four different locations along the Indian Ocean coast of Kenya. Considering all the four sampling sites in the two sampling seasons, the ranges of concentrations of residues detected in the fish samples (in $\mu g\,kg^{-1}$ wet weight) were: lindane 16.1 (in sample from Mombasa) – 1445 (Sabaki), aldrin 1.55 (Kilifi) – 323 (Kilifi), dieldrin 4.81 (Ramisi) – 109 (Sabaki), endosulfan 5.91 (Mombasa) – 54.6 (Sabaki), p,p′-DDT 9.11 (Mombasa) – 29.3 (Kilifi), p,p′-DDE 1.94 (Kilifi) – 97.5 (Sabaki) and p,p′-DDD 1.68 (Mombasa) – 98.9 (Kilifi). The concentration ranges obtained in fish in this study indicate that Mombasa Old Town was least contaminated by organochlorine residues as higher residues were detected near Malindi, at Funzi Lazy lagoon and at Kilifi creek which were near the confluences of Rivers Sabaki, Ramisi and Goshi at the Indian Ocean coast, indicating that the sources of the residues in fish the Indian Ocean coast were through discharge from the rivers. Seasonal variation in concentration of residues detected in fish samples was clear, with higher residue levels being recorded in the rainy season in May compared with those detected in the dry season in January. This seasonal variation was particularly observable in p,p′-DDT residues which were only detected in samples taken in the rainy season but none in the dry season in all the sampling sites. The high DDT/DDE and aldrin/dieldrin concentration ratios also indicated that the sources of these residues were recent. This conclusion was also supported by high BCF values of lindane in fish sampled in Sabaki and of aldrin sampled from Kilifi and Ramisi. Overall, the concentration ranges of some of the residues such as DDT, DDE, and DDD were comparable with those reported earlier for fish samples from Lake Kariba in Zimbabwe but concentration ranges of lindane and aldrin were much higher than those reported in fish from Lake Kariba. The concentration ranges of the residues of dieldrin, lindane and DDE were comparable with those obtained in fish from Tana River in Kenya but the other residues of aldrin, endosulfan, DDD, and DDT were much higher than those detected in fish from River Tana.

Keywords: organochlorines residues; marine fish; seasonal variation

Introduction

The issue of environmental fate of pesticides has received more attention in Kenya recently due to international residue requirements in export products, food and drinking water

*Corresponding author. Email: josephlalah57@yahoo.com

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(Lalah et al. 2002; Wandiga, Lalah, and Kaigwara 2002). Owing to increase in illegal usage, ecological and health implications, monitoring of residues in the aquatic compartments including sediments as the main sinks should be given high priority. Due to the recent government ban following the Stockholm Convention on the use of some of the organochlorines such as DDT, chlordane, endrin, dibromochloropropane, chlordimeform, heptachlor and toxaphene (Lalah et al. 2003) it is expected that the concentration levels of organochlorine residues may be lower in the coastal water at this time. Lindane, aldrin and its metabolite, dieldrin, are still allowed for use restrictively for seed dressing and termite control and for mosquito control, respectively (Lalah et al. 2003). Due to the rapid population increase (currently at 2.9% p.a) in Kenya, there is increased need for chemical pesticide use in both agriculture, veterinary and public health vector control. The annual amount of imported pesticides has now doubled from 2645 metric tonnes in 1985 (PCPB 1996; Lalah et al. 2002). If the current food policy is to be implemented to achieve the goal of self-sufficiency then even larger amounts of pesticides will have to be applied in the near future (Mathews 1979; Hassall 1990; Barasa 1999). Because of their persistence in food chain and toxicity to aquatic organisms, there is need to monitor the concentrations of organochlorine pesticide residues in aquatic environments in Kenya to provide baseline data useful for regulation. When provided with baseline data on residue levels of pesticides in fish, the government authorities can also be prompted to intensify the regulation of the use of the banned pesticides.

The Indian Ocean coastal environment of Kenya contains critical terrestrial and aquatic habitats, which comprise unique ecosystems, and which support a rich biological diversity and valuable assortment of natural resources which may be adversely affected by pesticide residues transported from inland waterways into coastal waters via rivers and estuaries through flooding, deposition, and rainfall (UNEP 1998). Marine fish are regarded as good bio-indicator species for evaluating pesticide exposure toxicity levels in marine environments and their susceptibility is depicted by reduction in species richness (Matin 2002).

This study was undertaken to analyze the concentrations of organochlorine pesticide residues in different fish species samples obtained from different locations along the Indian Ocean coast of Kenya to determine the distribution and seasonal variation of the residue concentration levels within sites. This tropical coastline extends over a distance of 600 km and lies between latitudes 5°4′N and 4°4′S and between longitudes 33°50′W and 41°45′E and receives major rivers of Kenya including Tana, Sabaki, Goshi, and Ramisi which transverse through rich agricultural lands and settlement areas where pesticides are used in large amounts (UNEP 1998).

Materials and methods

Chemicals

High purity (99%) standards p,p′-DDT, p,p′-DDE, p,p′-DDD, endosulfan, aldrin, dieldrin and γ-BHC (lindane) were obtained from Aldrich Chemical Company Inc., USA. Extraction solvents i.e. *n*-hexane, acetone and methanol were obtained from a local supplier (Kobian, Nairobi, Kenya). Proanalys grade anhydrous sodium sulphate, certified A.C.S acetonitrile and analytical grade *n*-hexane were obtained from J.T. Baker Inc., USA. Florisil (60–100 mesh) for column chromatography was purchased from Fisher Scientific, for use in column clean-up procedures. Whatman No. 42 filter papers were obtained from Whatman International Ltd and were used in Soxhlet extraction as

substitutes for extraction thimbles and for filtration purposes. White spot nitrogen, from East Africa Oxygen, Nairobi, Kenya, was used as a purging agent. All glassware used was thoroughly cleaned to eliminate electron-capturing contaminants which can interfere with detection and analysis.

Sampling

The location of the area on which the study was conducted lies within latitudes 5°4′N and 4°4′S and within longitudes 33°50′W and 41°45′E (Figure A1). There were four sampling sites along the coast; namely, Funzi Lazy Lagoon next to the confluence of Ramisi River (Ramisi sample), Mombasa Old Town (Mombasa sample), Kilifi Creek at the Confluence of Goshi River (Kilifi sample) and at the Confluence of Sabaki River near Malindi Town (Sabaki sample). Fish samples were taken twice in mid January and early May in 2001 corresponding to two seasons: the hot, dry north–east monsoon and the warm and wet long rains period, respectively. Thirty six specimen samples of fresh, small fishes of different species including *Sardines* (*Sardinella fimbrita*), *Penaeus* sp., black pomfret (*Apolectus niger*, silver cap (*Pampus argenteus)* were sampled from lots brought from fishermen at each site on the shores of the Indian Ocean and wrapped in pre-cleaned aluminium foil, placed in polythene bags and preserved in a deep freezer prior to analysis.

Sample preparation

Sub-samples of 30 g whole fish homogenate prepared by pooling nine samples of the sampled fish species were weighed in triplicates and each portion transferred to a mortar. Some 20 g of anhydrous sodium sulphate was added and the mixture thoroughly crushed in a Pestle and Mortar to ensure good homogeneity. The homogenate was then quantitatively transferred to a filter paper and sealed and were Soxhlet-extracted for 3 h at the rate of 6 cycles h^{-1}, using 100 mL of a solvent system comprising 85% hexane, 10% acetone, and 5% deionized water. The extracts were subjected to clean-up procedures in glass-wool-plugged miniature columns of height 10 cm and 2 cm in diameter containing 4 g of florisil, topped with a 2 cm layer of anhydrous sodium sulphate. The extract was reduced to small aliquot on a rotary evaporator and made up to 1 mL hexane and applied to the florisil column which was subsequently eluted with first 10 mL hexane, then 10 mL of 1% acetone in hexane, and finally with 10 mL of 2% acetone in hexane. The eluates were combined and solvent completely expelled on a water bath at 50°C using N_2 gas as a purging aid. Each sample extract was reconstituted to 0.5 mL of analytical grade *n*-hexane prior to GLC analysis.

In separate experiments, portions of fish homogenate were pulverized to obtain 1.5 g of powder which was heated in an oven at 110°C for 24 h. The loss in weight was attributed to the moisture content of the sample. The crucible plus sample were transferred and combusted in a muffle furnace at 550°C for 3 h. The difference in weight was taken as the organic matter of fish. A gravimetric method was used to determine the fat content of extracted samples, which enabled calculation of percent fat content.

GLC analysis of pesticide residues

Identification and quantification of organochlorine pesticide residues in the 36 fish samples were done using a Perkin-Elmer model 8500 Gas Chromatograph (GC) equipped

with a ^{63}Ni-ECD and a Perkin-Elmer model GP-100 graphics printer. White Spot Nitrogen flowing at a rate of 2 mL min^{-1} was used as carrier gas. The stationary phase was an SE-54 capillary column of dimension 30 m × 0.25 mm i.d × 0.25 μm film. Column and oven temperatures were set at 200 and 260°C, respectively, using a programmed ramp rate of 4°C min^{-1} with a 17 min hold at 260°C. Detector and injector temperatures were set at 350 and 270°C, respectively. An attenuation of 128 and Chart speed of 5 mm min^{-1} were used. A 1 μL solution of each of the reconstituted samples and a blank extract were injected, separately, into the GC. For every five injections made, the GC programmed conditions were calibrated by injecting 1 μL solution of the standard chlorinated pesticide mixture (CPM). Quantification was achieved by comparing the peak areas of sample injections with those of the standards analyzed under the same conditions. The results from triplicate samples were compared and their means calculated. The limit of detection (LOD) was determined using prepared standard solutions of 0.01–0.10 ppm and injecting 1 μL (10–100 pcg) solution into the SE-54 column. Recovery studies were performed by spiking standard fish samples with 0.05 ppm of the multi-residue pesticide standard, to estimate the losses of pesticide residues during the clean-up and concentration procedures.

Results and discussion

The limit of detection (LOD) of the pesticide standards gave an indication of the sensitivity of the ECD detector. However, minimum levels of pesticide residues from marine fish samples which could be reliably detected and quantified could be influenced by the purity of solvents as well as by the efficiency of the clean-up procedures but this effect was nullified by a parallel run of the blank. Mean percent recoveries of over 83% were obtained the marine fish samples analyzed (Table 1). These recoveries were taken as good enough and therefore no adjustments in concentrations of organochlorine pesticide residues in the current study were made. Fish characteristics were analyzed and the results are given in Table 2. In the marine environment, moisture content in fish accounts for about three-quarters of its mass on wet weight basis, whereas the organic and lipid contents cater for the remaining weight. Our results were comparable with the results obtained by Nahn et al. (1998) at the Red River estuary in Vietnam. Similarly, Munga (1985) found 3.62% lipid content of fish from the Tana River while Everaarts et al. (1996) obtained 2.09% lipid content from Formosa Bay adjacent to the confluence of Sabaki River, along the Indian Ocean coast of Kenya (Munga 1985; Everaarts et al. 1996; Nahn et al. 1998). Generally, the marine fish from the four sampling stations under this study

Table 1. Mean percent recovery of the amounts of residuesin spiked samples.

Pesticide	Spiked (μg kg^{-1})	Recovered (μg kg^{-1})	Mean % recovery
Aldrin	10.00	9.23	92.3
Lindane	10.00	9.85	98.5
p,p′-DDT	15.00	12.9	86.2
p,p′-DDE	15.00	13.0	86.5
p,p′-DDD	15.00	13.1	87.5
Endosulfan	10.00	8.44	84.4
Dieldrin	10.00	8.8	87.9

Note: $n = 3$.

showed similar trends in composition of moisture, organic and lipid parameters. All the fishes analyzed were tropical, migratory and well distributed along the coastal areas of Kenya at depths ranging from zero up to 210 meters, and the species analyzed, their habitats and diets are given in Table 2. Although the species were different among the sites, it should be notable that our objective was to determine the concentrations of the residues in each species and to compare seasonal variations within each site.

Nine samples of fish species from each of the four sites per season were analyzed for organochlorine residues. All the results are expressed in $\mu g\,kg^{-1}$. The following main factors were considered important for discussion of the results of this investigation: the seasonal variation of organochlorine residues along the Kenyan coastal marine environment; and the distribution of these contaminants in marine samples from different sampling stations. Different fish species were taken as biomarkers of pollution levels. Concentrations of seven chlorinated pesticides measured in the dry seasons (January 2001) and in the rainy season (May 2001) at four different stations are shown in Table 3.

Table 4 summarizes the highest and lowest concentration levels of residues detected in different species in different sites and the potential sources of the detected contaminants. Lindane residues showed the highest mean concentration of $833\,\mu g\,kg^{-1}$ lipid content in *S. fimbriata* samples during the rainy season. These concentrations suggested a point source of contamination for lindane since other residues had relatively lower amounts ranging from $12.8\,\mu g\,kg^{-1}$ for p,p′-DDE to $63.4\,\mu g\,kg^{-1}$ lipid for p,p′-DDD. Four residues were absent in fish samples during the dry season but present in the rainy season. However, three residues were present in both seasons indicating their high frequency of use around Sabaki River and its environment, and also high persistence of these residues in lipophilic tissues of fish from that region. *Penaeus* sp. samples from Kilifi Creek showed no detectable quantities of p,p′-DDE and dieldrin in both seasons. Only lindane and aldrin were present during the dry season suggesting low levels of contamination of fish in that region in the dry spell. The high levels of contamination were that of aldrin with a mean concentration of $323\,\mu g\,kg^{-1}$ lipid in the rainy season. Aldrin was also present at

Table 2. Characteristics of the fish samples.

Station	Sabaki	Kilifi	Mombasa	Ramisi
Fish species	*S. fimbriata* (Sardine)	*Penaeus* sp.	*A. niger* (black pomfret)	*P. argenteus* (Silver pomfret)
Mean weight (g)	27.0	77.0	62.7	54.7
Mean length (cm)	19.2	18.5	18.5	15.3
Moisture content (%)	73.4	79.5	74.3	75.7
Organic matter content (%)	19.4	18.5	21.5	18.9
Lipid content (%)	3.9	4.4	3.3	3.7
Maximum size (cm)	19.5	30	75	60
Maximum age (years)	1.9–3.2	–	–	–
Depth found (m)	0–50	0–27	15–105	5–210
Temperature (°C)	Tropical	Tropical	Tropical	Subtropical
Diet	Plants, zooplankton	Omnivorous sediments	Plants, zoobenthos Zooplanktons	Zoobenthos Zooplanktons
Resilience	High: minimum population doubles in <15 months	High:	High:	Medium: minimum population doubles in 1.4–4.4 years

14.5 $\mu g\,kg^{-1}$ in the fish samples in the dry season, indicating the possibility of its frequent application in Kilifi region. Members of the DDT family did not show significant contamination in fish in Kilifi and p,p′-DDD and p,p′-DDT were present only during the rainy season. All the organochlorine residues under investigation in *A. niger* samples from Mombasa Old Town, except for p,p′-DDT which was only present during the rainy season, were detected in both seasons giving a strong indication of contamination of fish from that region. Lindane was present in the highest concentrations during the dry season

Table 3. Mean concentrations of organochlorine pesticide residues ($\mu g\,kg^{-1}$) in fish sampled in dry (January) and wet (May) seasons.

Residue analyzed	Sabaki	Kilifi	Mombasa	Ramisi
Aldrin (dry)	BDL	14.5 ± 1.82	2.44 ± 0.01	2.02 ± 0.08
Aldrin (wet)	29.0 ± 4.52	323 ± 25.5	4.14 ± 0.51	201.0 ± 13.2
Dieldrin (dry)	58.8 ± 9.21	BDL	7.11 ± 0.91	4.81 ± 0.05
Dieldrin (wet)	27.9 ± 6.77	BDL	13.3 ± 1.99	62.0 ± 4.55
Endosulfan (dry)	BDL	BDL	9.99 ± 2.11	BDL
Endosulfan (wet)	40.2 ± 6.55	22.9 ± 3.31	13.8 ± 1.22	10.4 ± 0.35
Lindane (dry)	48.6 ± 5.92	26.5 ± 1.22	93.8 ± 6.77	BDL
Lindane (wet)	833.0 ± 23.1	BDL	20.8 ± 4.22	281.0 ± 11.1
p,p′-DDE (dry)	BDL	BDL	7.34 ± 1.21	BDL
p,p′-DDE (wet)	1.28 ± 1.25	BDL	12.2 ± 3.22	15.7 ± 1.22
p,p′-DDD (dry)	85.8 ± 11.2	BDL	54.2 ± 9.21	3.32 ± 0.88
p,p′-DDD (wet)	63.4 ± 15.2	99.0 ± 13.5	8.59 ± 1.09	31.1 ± 6.11
p,p′-DDT (dry)	BDL	BDL	BDL	BDL
p,p′-DDT (wet)	71.9 ± 2.31	21.8 ± 2.71	10.4 ± 1.54	17.9 ± 2.57

Notes: (dry) – dry season; (wet) – wet season; BDL – below detection limit; SD – standard deviation; $n = 3$.

Table 4. Highest and lowest concentrations of organochlorine pesticide residues in fish sampled in dry and wet seasons.

Station	Sabaki	Kilifi	Mombasa	Ramisi
Highest conc (dry)	p,p′-DDD (86)	Lindane (26)	Lindane (94)	Dieldrin (4.8)
Highest conc (wet)	Lindane (830)	Aldrin (320)	Lindane (21)	Lindane (280)
Lowest conc (dry)	Aldrin (BDL) Endosulfan (BDL) p,p′-DDE (BDL) p,p′-DDT (BDL)	Dieldrin (BDL) Endosulsan (BDL) p,p′-DDE (BDL) p,p′-DDD (BDL) p,p′-DDT (BDL)	p,p′-DDT(BDL)	Endosulsan (BDL) Lindane (BDL) p,p′-DDE (BDL) p,p′-DDT (BDL)
Lowest conc (wet)	p,p′-DDE (1.4)	Lindane (BDL) Dieldrin (BDL) p,p′-DDE (BDL)	Aldrin (4.1)	Endosulfan (BDL)
Potential sources	Agricultural	Agricultural	Public health	Agricultural

Notes: Conc: mean concentration in $\mu g\,kg^{-1}$ in parenthesis; (dry) – dry season; (wet) – wet season; BDL: below detection limit.

followed by p,p′-DDD. It is probable that the use of DDT for public health in the harbor resulted in contamination of fish in this region namely in form of the anaerobic metabolite DDD especially in dry seasons. Most residues were present in *A. niger* samples probably due to their application in the domestic, industrial and agricultural sectors around the town. As found in Mombasa, lindane had the highest mean residue concentration in *P. argenteus* from Ramisi River followed closely by aldrin during the rainy season. Lindane, endosulfan, p,p′-DDE, and p,p′-DDT were absent in fish samples during the dry season. However, aldrin, dieldrin and p,p′-DDD were detectable in both seasons. Aldrin had higher concentrations than its metabolite dieldrin in both seasons.

Other coastal areas of the world have shown similar seasonal patterns in organochlorine pesticide residue concentrations. For instance, carp fish from the coastal region North of Vietnam, with a subtropical climate, had relatively higher occurrences of these compounds in rainy season than in the dry season with concentrations ranging between 0.23 to 120 (wet season) and 0.15 (dry season) $\mu g\,kg^{-1}$ lipid content, respectively (Nahn et al. 1998). However, chlorinated hydrocarbons in shellfish from the Northern Adriatic Sea did not show a clear seasonal pattern although higher PCB contents were found for stations influenced by discharges from River Po. Values for DDTs and PCBs were between 2.1 and 18.3 and 3.2 and 12.1 $\mu g\,kg^{-1}$ lipid, respectively, in any season (Najdek and Bazulic 1988). Data on distribution of organochlorine residues shown in Table 3 and in Figure 1(a) and 1(b) revealed that the samples from confluence of Sabaki River had lindane mean concentration of 612.02 $\mu g\,kg^{-1}$ lipid content with highest mean residue concentration as well as frequency of occurrence (80%) in *S. fimbriata* samples analyzed. Similar high values of lindane have been reported in Suruga Bay, Japan and could suggest that there is little metabolism and excretion of lindane in fish (Lee et al. 1997). Aldrin, dieldrin and endosulfan residues were at comparable concentration levels of 41.8, 48.5, and 40.2 $\mu g\,kg^{-1}$ lipid content, respectively. Among the metabolites of DDT, the concentrations of p,p′-DDD (70.8 $\mu g\,kg^{-1}$ lipid) were the highest and were approximately 4 times of p,p′-DDT and 2 times that of p,p′-DDE. The lowest mean concentrations in fish samples from Sabaki site were those of p,p′-DDT (17.9 $\mu g\,kg^{-1}$ lipid) with the least frequency of occurrence of 40% of all the analyzed species.

As shown in Table 5, at the Kilifi Creek, the major residue detected in *S. fimbriata* was aldrin (91.7 $\mu g\,kg^{-1}$ lipid) but p,p′-DDE showed the lowest mean concentration of 1.94 $\mu g\,kg^{-1}$ lipid. Other members of the DDT family were present in high amounts: p,p′-DDT (21.8 $\mu g\,kg^{-1}$) and p,p′-DDD (51.9 $\mu g\,kg^{-1}$) lipid content. Of the pesticide residues analyzed in *Apolectus niger* from Mombasa site, lindane (57.3 $\mu g\,kg^{-1}$ lipid) was present in the highest amounts, and the frequency of occurrence at 80%. Levels of aldrin (3.29 $\mu g\,kg^{-1}$ lipid) were slightly above their detection limits while its metabolite dieldrin was approximately three times more, suggesting decreased uptake by fish. p,p′-DDD (23.8 $\mu g\,kg^{-1}$ lipid) was the highest at about twice the concentrations of other DDT family compounds. Lindane at concentration levels of 281 $\mu g\,kg^{-1}$ lipid were detected in the highest mean amounts in *P. argenteus* from Ramisi site followed by aldrin (102 $\mu g\,kg^{-1}$ lipid) which was two times more than its metabolite dieldrin. But the concentrations of endosulfan at 10.4 $\mu g\,kg^{-1}$ lipid were the lowest while members of the DDT family had relatively similar amounts although p,p′-DDD (23.8 $\mu g\,kg^{-1}$ lipid) was slightly higher.

Persistent organochlorines have high lipophilicity. Therefore, relatively high amounts of chlorinated compounds present in fish samples were not astonishing since fishes have high extractable lipid content. From all the sampling stations except at Kilifi Creek, lindane was present in highest concentrations suggesting high rates of continued usage of

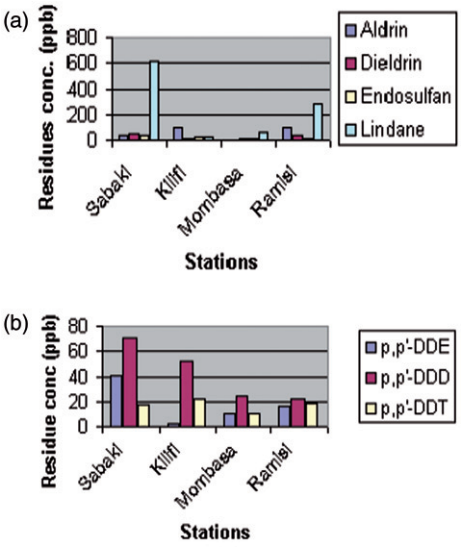

Figure 1. (a) The distribution of mean annual residue concentrations in fish from the four sites. (b) The distribution of mean annual residue concentrations in fish from the four sites.

Table 5. Ranges of concentrations of organochlorine pesticide residues in fish samples (in $\mu g\,kg^{-1}$) from different sites in the two seasons.

Residue analysed	Sabaki	Kilifi	Mombasa	Ramisi
Aldrin	11.3–67	1.6–323	1.2–7.0	2.0–201
Dieldrin	9.1–109	8.7–9.8	7.1–15	4.8–95
Endosulfan	26–55	23–23.1	5.9–20	10.4–11.1
Lindane	49–1445	26.5–27.6	16–145	42–750
p,p′-DDE	9.4–97	1.9–2.1	2.7–22	5.3–26
p,p′-DDD	15–112	4.9–99	1.7–54	3.3–36
p,p′-DDT	13.6–22.2	14.3–29.3	9.1–11.6	10.0–26
Highest conc	Lindane	Aldrin	Lindane	Aldrin
Lowest conc	Dieldrin	Aldrin	Aldrin	Aldrin

Note: Mean concentration ranges of residues in fish for the two seasons. Higher concentrations were detected in fish samples taken during the wet season; conc: mean concentration.

technical BHC formulations for soil treatment, foliage application on fruit and nut trees, vegetables, ornamentals, timber and wood protection. The bioaccumulation factors (Table 6) were calculated based on the mean concentration of pesticide residues in fish and mean concentration in seawater (based on unreported mean water concentrations of the same residues detected in water samples taken from the same sampling sites during the same period) and indicate high concentrations of aldrin in Kilifi and Ramisi which would suggest recent discharge as the potential source in these areas as the concentration of dieldrin (its metabolite) was relatively much lower. Similarly high BCF of lindane in fish from Sabaki indicates recent contamination. Transportation of pollutants to the ocean waters and subsequent bioaccumulation in marine fish samples could have occurred.

Table 6. The bioaccumulation factors (BCF) in fish samples from different sampling sites.

Residue	Sabaki	Kilifi	Mombasa	Ramisi
Aldrin	106	4774	61	4064
Dieldrin	187	48	21	294
Endosulfan	239	92	28	71
Lindane	2531	51	BDL	BDL
p,p′-DDE	188	7	55	244
p,p′-DDD	339	288	317	360
p,p′-DDT	101	56	BDL	87

Notes: The BCF is obtained by dividing the mean Concentration by mean concentration in the sea water; BDL: below detection limit in water.

In this study, concentrations of p,p′-DDT were less than its metabolite p,p′-DDD. Moreover, higher concentrations of p,p′-DDD than those of p,p′-DDE suggest anaerobic transformation of parent p,p′-DDT. Since p,p′-DDE is less toxic than p,p′-DDT, the transformation of the parent compound probably represents a good protective mechanism in fish. However, it is now accepted that p,p′-DDE may have sublethal impacts on reproduction in rats and mammals. The reported contamination of organochlorines in Suruga Bay (Japan) coastal fish are comparable with levels obtained in this study, especially those of lindane at Kilifi and Ramisi. In Suruga Bay, DDT's and HCH's concentrations ranged from 80 to 1700 $\mu g\,kg^{-1}$ and 1.0 to 250 $\mu g\,kg^{-1}$ lipid, respectively (Lee et al. 1997).

Fish from Meghna–Dhogoda River estuary contained relatively higher mean levels of residues than in the current study: p,p′-DDT and p,p′-DDD were at concentrations of 1280 and 1370 $\mu g\,kg^{-1}$ lipid, respectively. Lindane levels were 26 $\mu g\,kg^{-1}$ lipid while quantities of aldrin and dieldrin were not detectable (Matin et al. 1997). All the chlorinated hydrocarbons which were analyzed in this study were banned in Kenya in 1986 except aldrin and dieldrin, whose use is restricted to the control of termites in the building industry (PCPB 1992; Lalah et al. 2003). Their presence in marine environment (fish) indicates possible continued use although illegally according to the Kenya Government policy. This has been attributed to their availability at low costs and their efficacy (Wandiga 1995). However, considering the range of concentrations of residues obtained (Table 5), there is no danger to human consumption of fish from this region because all the residues (except for lindane in one sample taken in Ramisi and another one sample from Sabaki in the rainy season) were all below the maximum allowable concentrations in fish edible parts according to FDA EPA. The maximum allowable limits of concentrations in fish edible parts are: 300 $\mu g\,kg^{-1}$ (for aldrin and dieldrin), 500 $\mu g\,kg^{-1}$ (for DDT, DDE, and DDD) and 300 $\mu g\,kg^{-1}$ (for lindane in shell fish and frog legs) (Wandiga, Lalah, and Kaigwara 2002).

Ratios of DDT/DDE and aldrin/dieldrin in biota are often used as indicators of recent DDT and Aldrin inputs into the environment, or as measures of efficient degradation, with high ratios (if >1) indicating recent input (Gonzelaz et al. 2003). The hypothesis of recent input was applied to the ratios of pesticide residues in this study (Figure 2). Aldrin/dieldrin and DDT/DDE residue ratios were very significant (>11) in fish samples from the Kilifi Creek suggestive of recent aldrin and DDT current use. The fish species *S. fimbriata* had both high ratios of 11.0 which confirmed the hypothesis. Furthermore, an increasing

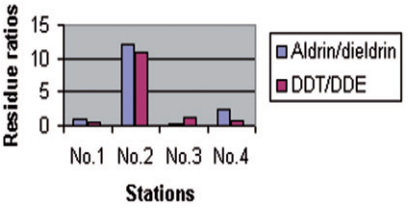

Figure 2. Some residue ratios for samples from the four sampling sites.
Notes: No. 1 = Sabaki; No. 2 = Sabaki; No. 3 = Mombasa; No. 4 = Ramisi.

Table 7. Mean concentrations of residues obtained in fish samples in this study compared with those reported earlier in other studies.

	Aldrin	Dieldrin	Endosulfan	Lindane	DDE	DDD	DDT
Sabaki[a]	41.8	48.5	40.2	612	41.1	70.8	17.9
Kilifi[a]	91.7	8.69	22.9	26.5	1.94	51.9	21.8
Mombasa[a]	3.29	11.2	12.0	57.3	9.77	23.8	10.4
Ramisi[a]	102	43.1	10.4	281	15.7	21.8	17.9
Tana[b]	<0.007	109.4	<0.042	131.2	140.4	<0.009	<0.003
Kariba[c]	0.30	nr	nr	0.6	14.7	2.10	4.30

Notes: nr: not reported.
[a]Present study.
[b]Residues in *Tilapia zilli* in Tana River (Lalah et al. 2003).
[c]Pesidues in *Tilapia rendalli* in Kasese Bay, Lake Kariba, Zimbabwe (Everaarts, Wasreen, and Hillebrand 1991).

proportion of a metabolite in relation to the parent compound reflects a decreasing exposure to new sources of pesticide pollution and vice versa.

Small fishes of different species develop common facultative mechanisms. They also have little fat reserves unlike big fishes, hence are less prone to adverse effects of some lipophilic pesticides (Munga 1985). Some of the resilience factors for the different species that were sampled and their adult sizes are given in Table 2. In this study, lindane was established at highest levels in all stations which ranged from 26.5 ppb $\mu g\,kg^{-1}$ (Kilifi Creek) to 612 $\mu g\,kg^{-1}$ lipid (confluence of Sabaki River). The presence of a high proportion of p,p′-DDD and p,p′-DDE in relation to total DDT burden in coastal fishes indicated that either the migratory fish species, could have, perhaps, converted DDT rapidly its metabolites because of high metabolic body rates or those areas along the coast were not threatened by new inputs of pesticides. Dieldrin and p,p′-DDD were present in the fish species from all the four sampling station except at Kilifi site in both seasons. Of the DDT family, p,p′-DDD gave a similar trend as dieldrin which could reliably be considered as an indication of fish contaminant exposure to organochlorines along the coast of Kenya. Finally, residue levels of pesticides in fish samples were highest during the rainy seasons indicating increased transportation of contaminated materials by water from upland areas into the marine environment. The values of concentrations obtained in this study were also comparable with those reported in earlier studies in Africa as shown in Table 7 (Berg, Kiibus, and Kautsky 1992; Lalah et al. 2003). With regard to human health, the concentrations of pesticide residues were below the maximum admissible concentration (MAC). For instance, the Food and Agriculture (FAO) and World Health Organization

(WHO) MAC-value for dieldrin and total DDT in fish is 500 and 2000–3500 ng g^{-1} of lipid, respectively (Everaarts, Wasreen, and Hillebrand 1991). However, low levels of several pesticide residues detected in various fishes along the coast of Kenya may probably pose chronic effects on the marine fauna and flora through bioaccumulation.

Conclusions

The concentration ranges obtained in fish in this study indicate that Mombasa Old Town was least contaminated by organochlorine residues as higher residues were detected near Malindi, at Funzi Lazy lagoon and at Kilifi creek which were near the confluences of Rivers Sabaki, Ramisi and Goshi at the Indian Ocean coast, indicating that the sources of the residues in the Indian Ocean coast were through discharge from the rivers. Seasonal variation in concentration of residues detected in fish samples was clear, with higher residue levels being recorded in the rainy season in May compared with those detected in the dry season in January. This seasonal variation was particularly observable in p,p′-DDT residues which were only detected in samples taken in the rainy season but none in the dry season in all the sampling sites. The high DDT/DDE and aldrin/dieldrin concentration ratios also indicated that the sources of these residues were recent. This conclusion was also supported by high BCF values of lindane in fish sampled in Sabaki and of aldrin sampled from Kilifi and Ramisi. Overall, the concentration ranges of some of the residues such as DDT, DDE and DDD were comparable with those reported earlier for fish samples from Lake Kariba in Zimbabwe but concentration ranges of lindane and aldrin were much higher than those reported in fish from Lake Kariba. The concentration ranges of the residues of dieldrin, lindane and DDE were comparable with those obtained in fish from Tana River in Kenya but the other residues aldrin, endosulfan, DDD, and DDT were much higher than those detected in fish from River Tana.

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Appendix

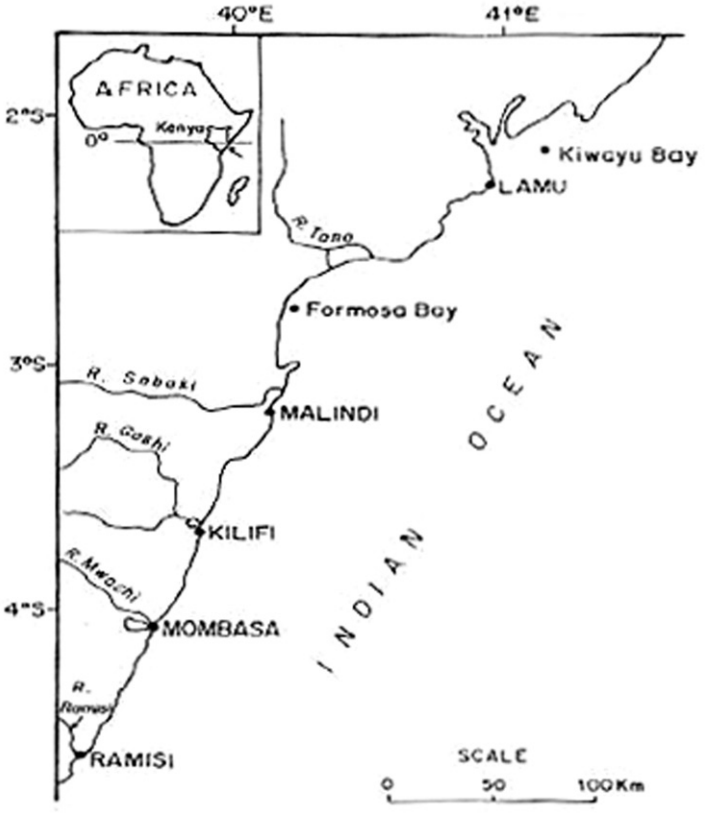

Figure A1. The Indian Ocean coastal area where sampling was done.