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Organochlorine Pesticide Residues in Tana and Sabaki Rivers in Kenya

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The use of organochlorine pesticides in Kenya has contributed signicantly in both agricultural development and in public health vector control. With the rapid increase in population and the increasing demand for agricultural and veterinary production the use of some of these chemical pesticides will continue to increase. The annual importation of pesticides in 1998 was 7606 tonnes, which included 1814 tonnes insecticides/acaricides, 1408 herbicides and 4225 fungicides (Anonymous, 1999). In keeping with global trends, some of the organochlorines have now been replaced with organophosphates and carbamates which are less persistent in the environment (table 1).

Due to their chemical stability, high lipophilicity and long residual life times in the aquatic environment, organochlorine pesticide residues bioaccumulate in aquatic food chains (Geyer et al 1982, Cecilia et al1992). Residues from agricultural activities and from industrial and domestic effluent are often transported through canals and rivers into lakes and seas. Some are also transported into the aquatic environment from the point of application by wind and rain. Consequently, residues of DDT, DDE, DDD, endosulfan, lindane, aldrin and dieldrin have been detected in water, sediment, fish, molluscs and other aquatic animals in coastal and inland waters in many countries (Bidleman et al 1990, Larsson et al 1992, Ragendran et al 1992, Silva-Herrera and Zapata-Perez 1993, Tanabe et al 1993, Hong et al 1995, Sericano et al 1995, Roots and Zitko 2001). Some of the significant ecological toxic effects of organochlorine pesticide residues have been expressed in their ability to disrupt the endocrine functions in fish and birds which interferes with their growth and development, as has been demonstrated by the DDT group (Hickey and Anderson 1968, Cooke and Bell 1976, Gralla et al 1977, O'Brien and Hilton 1978).

Recent monitoring conducted in Kenya have shown presence of lindane, aldrin, endosulfan, p,p'-DDE, dieldrin, p,p'-DDD, and p,p'-DDT residues in seawater, seaweeds, sediment, fish samples sampled from different sites along the Indian Ocean Coast of Kenya (Everaarts et al 1996, Barasa 1998). The concentration levels of these residues were slightly lower in comparison to those reported in samples from other tropical coastal waters. In freshwaters, Koeman et al 1972 reported low levels of DDT residues (from <0.001 to 0.064 mg/g) in birds and fish

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from Lake Nakuru. Greichus et al 1978 found slightly higher concentration levels of DDE, DDD, lindane and dieldrin in fish from the same lake. DDT and its metabolite residues have also been reported in the Kenyan inland lakes of Baringo, Naivasha, Nakuru and Victoria (Foxall 1983). Munga (1985) also reported presence of significant levels of DDT, DDE, DDD and endosulfan in River Tana fish, believed to have originated from aerial spraying of cotton and rice fields in the area around Mwea and the coastal region near the Tana River.

Other organochlorine residue analysis work of significance in Kenya has been reported by Omoebe (1986) who found residues of organochlorine and organophosphate residues (mainly dioxathion) in cow milk and meat products in Athi River and Ngong areas where meat and milk are main staple diet. Kanja (1988) also reported low levels of DDT in human milk, chicken eggs and other food sources. Evidently, these monitoring activities have been very limited especially in fresh water environments and particularly in view of the importance of drinking water quality and residue limits in fish products. In this paper, the results of recent monitoring of organochlorine pesticide residues in two rivers in Kenya are reported.

MATERIALS AND METHODS

High purity (99%) p,p'-DDT, p,p'-DDE, p,p'-DDD, endrin, aldrin, endosulfan, dieldrin and lindane (γ-HCH) were obtained from Aldrich, USA. Acetone and n-hexane were triply distilled before use. Anhydrous sodium sulphate, Na_2SO_4, and florisil (60-100 mesh) for separation and clean-up were activated at 130 °C and cooled before use. Activated charcoal from Zeta Chemicals, Nairobi, was used for removal of lipid and colour from plant and fish tissue extracts. All the glassware were thoroughly cleaned to eliminate all electron-capturing contaminants that would interfere with GC detection according to standard procedures (UNEP 1982).

Samples of water, fish, sediment, and waterweeds were collected in January and July 1998 and in January and July1999 from the Tana and Sabaki rivers (see Fig 1). Water samples were collected in 1-litre brown glass bottles with screw caps. The sample bottles were lowered to fill at knee height, placed in an icebox and transported to the laboratory in Nairobi for analysis. Sediment samples were collected using a large scoop to a depth of 2 cm of undisturbed layer. The samples were wrapped in aluminium foil and transported in an icebox to the laboratory. Fish samples were obtained from fishermen along the rivers and were identified by the Kenya Marine and Fisheries Research Institute (KMFRI), Mombasa. Waterweeds were harvested along the rivers with a penknife.

Thirty (30) g sediment samples were weighed, in triplicate, mixed separately with 20 g anhydrous sodium sulphate and homogenized in a mortar with pestle. The homogenized samples were placed in thimbles and extracted in a Soxhlet extractor for 3 hours with 130 ml mixture of n-hexane and acetone (10:3, volume). Fish and waterweed samples were homogenized and extracted following

the same procedure. The Soxhlet extracts were concentrated in a rotary evaporator to about 2 ml at 50 °C. The water samples (500 ml, each) were partitioned with n-hexane in a 1-litre separatory funnel, shaking with 100 ml n-hexane for 15 min., then repeating with 80 ml and 60 ml n-hexane, respectively. The organic extracts were pooled and concentrated to 2 ml in a rotary evaporator.

Separation and clean-up of sample extracts were performed using florisil in small glass columns. The glass columns were plugged with glass wool at the bottom end, 4 g florisil was added then 2 g anhydrous sodium sulphate added on top. For waterweed and fish sample extracts, a spatula of activated charcoal was also added at the top of the column for decolourising the plant pigments and fish lipids, respectively. The extract (2 ml) was poured on the top of the column and eluted with 10 ml n-hexane, then with 10 ml n-hexane/acetone (95:5, volume) and then finally with 10 ml acetone/n-hexane (10:90, volume). The eluates were pooled and reduced to dryness in a rotary evaporator under nitrogen. Recovery efficiency was determined by spiking with known amounts of the standards and going through a similar extraction and clean-up procedure before GC analysis. Solvent background residue concentration levels and pesticide standard residue detection limits were also determined.

Samples were analysed in a Hewlett Packard GC Model 5890 equipped with an N-electron capture detector. GC conditions: SE30 packed capillary column (60 m × 0.319 mm × 0.25 μm), carrier gas: N_2 at a flow rate of 0.84 ml/min; injection volume: 1 μl; attenuation of the integrator was 0, chart speed 0.5 cm/min, injector and detector temperatures were 250 °C and 310 °C, respectively. The initial column temperature was 180 °C programmed at a rate of 5 °C/min to 225 °C and held for 10 minutes before increasing (at 20 °C/minute) to and holding at 280 °C for 20 minutes.

The organochlorine residue components were identified by comparing their retention times with those of the standards and quantified by extrapolation of corresponding sample peak areas with those from standard curves prepared for each pesticide standard. Small variations in retention times and response factors of each compound during the experiments were corrected for by obtaining fresh chromatograms of the standard mixture after every nine injections.

Standard solutions of concentrations ranging from 0.01 to 1 ppm were prepared for each pesticide standard and 1 μl was injected into the GC. Peak areas of standard solutions were plotted against their concentrations. A line of best fit was drawn through the points and the limits of detection were taken at 3 times the detector noise level.

RESULTS AND DISCUSSION

These two rivers were chosen in this study because of their central location within a catchment with large scale agricultural activities likely to influence residue contamination in aquatic environment. River Tana passes through areas with a

known history of organochlorine pesticide use. Some of the main agricultural activities include the Mwea Cotton and Rice farming. In this farming region earlier application of pesticides including DDT and endosulfan was practised by aerial spraying especially in the 1980's (Munga 1985). River Sabaki drains from Kiambu District, one of the most densely populated areas in Kenya with extensive agricultural activities including horticulture and animal husbandry which rely heavily on agrochemical application. The two rivers then flow through the relatively drier coastal province and discharge into the Indian Ocean (Fig 1). Samples (water, waterweed and sediment) were taken upstream but near their estuaries discharging into the Indian Ocean coast.

The organochlorine pesticides are no longer extensively used in Kenya. Some of them have now been completely banned or are under restricted use while awaiting their fate. These are shown in Table 1 (Table 1). Of the residues that were analysed, DDT, DDE, lindane and aldrin were predominantly detected reflecting their widespread recent use such as lindane and aldrin and their environmental persistence (such as DDT). DDT has been very popular in the recent past for malaria vector control because of its cheap cost and pesticidal efficacy. However, it was under restricted use between 1992 and 1997 when it was finally completely eliminated officially by the government. In our samples, DDT was detected mainly in its metabolite form of DDE.

Table 1. Some common organochlorine pesticides currently banned or under restricted use in Kenya

Banned pesticide:	**Previous use:**
DDT	Insecticide
Dibromochloropropane (DBCP)	Soil fumigant
Chlordimeform	Acaricide/insecticide
Chlordane	Insecticide
Heptachlor	Insecticide
Toxaphene	Acaricide
Endrin	insecticide
Restricted pesticides:	**Permitted use:**
Lindane	Seed dressing and termite control
Aldrin	In public health (mosquito control)
Dieldrin	In public health (mosquito control)

Note: The data was obtained from the Pest Control Products Board of Kenya, Ministry of Agriculture, Nairobi.

The recovery efficiencies were high for all pesticides in various matrices, ranging from 80% (fish muscle), 80.8% (waterweeds), 82.6% (sediment) to 88.1% (water). The detection limits of the pesticide standards were: aldrin (0.007 ng), endosulfan (0.042 ng), dieldrin (0.006 ng), endrin (0.004 ng), DDT (0.003 ng), DDE (0.036 ng), DDD (0.009 ng) and lindane (0.004 ng). The Sabaki River

sediment had pH 7.6, 4.7% organic carbon, 20.3% moisture, 42.5% silt, 30.2% clay and a silty clay texture.

Table 2. Organochlorine pesticide residue concentrations in River Tana (mean±s.d.).

residues	Water µg/L	Fish[1] ng/g lipid	Fish[2] ng/g lipid
Aldrin	0.037±0.006	<0.007	<0.007
Endosulfan	<0.042	<0.042	<0.042
Dieldrin	<0.006	109.375±1.125	1.750±0.018
Endrin	0.484±0.022	<0.004	<0.004
DDT	0.026±0.027	<0.003	<0.003
DDE	0.090±0.001	140.625±1.688	2.250±0.027
DDD	0.025±0.022	<0.009	<0.009
lindane	0.058±0.019	131.25±1.500	2.100±0.024

Note: Fish[1] and water samples were taken in July 1998. Fish[2] samples were taken in January 1998. The s.d. denotes 'standard deviation', n=6. The fish sampled were juvenile *Tilapia zilli*, which had mean weight (83.14 g), mean length (19.5 cm) and mean lipid content (0.32% in lateral muscle).

Table 3. Organochlorine pesticide residues detected in samples from River Sabaki (ng/g wet weight, mean ± sd).

water	residue	waterweed	sediment[1]	sediment[2]
0.018±0.007	Aldrin	11.900±0.007	5.100±0.001	108.511
0.050±+0.001	Endosulfan	0.867±0.012	<0.042	<0.042
<0.006	Dieldrin	<0.006	<0.006	<0.006
<0.004	Endrin	0.933±0.006	<0.004	<0.004
<0.003	DDT	<0.003	<0.003	<0.003
0.026±0.035	DDE	5.200±0.092	<0.036	-
<0.009	DDD	<0.009	<0.009	<0.009
0.058±0.025	lindane	5.367±0.024	6.933±0.025	-

Note: waterweeds and sediment[2] were sampled in January 1998; water and sediment[1] were sampled in January 1999.

Lindane has been used in Kenya extensively since 1949 for both agricultural and public health vector control purposes. Now its use is restricted to termite control and in seed dressing only. Despite this restriction, the residue data showed that it was widely distributed in waterweeds, with high concentrations (5.36 ng/g) in samples from Sabaki River (Table 3). The weeds from Sabaki River did not show any DDT. At the time of sampling DDT was already banned but we would still have expected minor or illegal applications especially for malaria vector control and for spraying against tse tse fly in the area north of Malindi near the sampling area. Waterweeds from R. Sabaki showed highest DDE concentration of 5.2 ng/g January samples. This indicates the possibility of accumulating DDT residues by

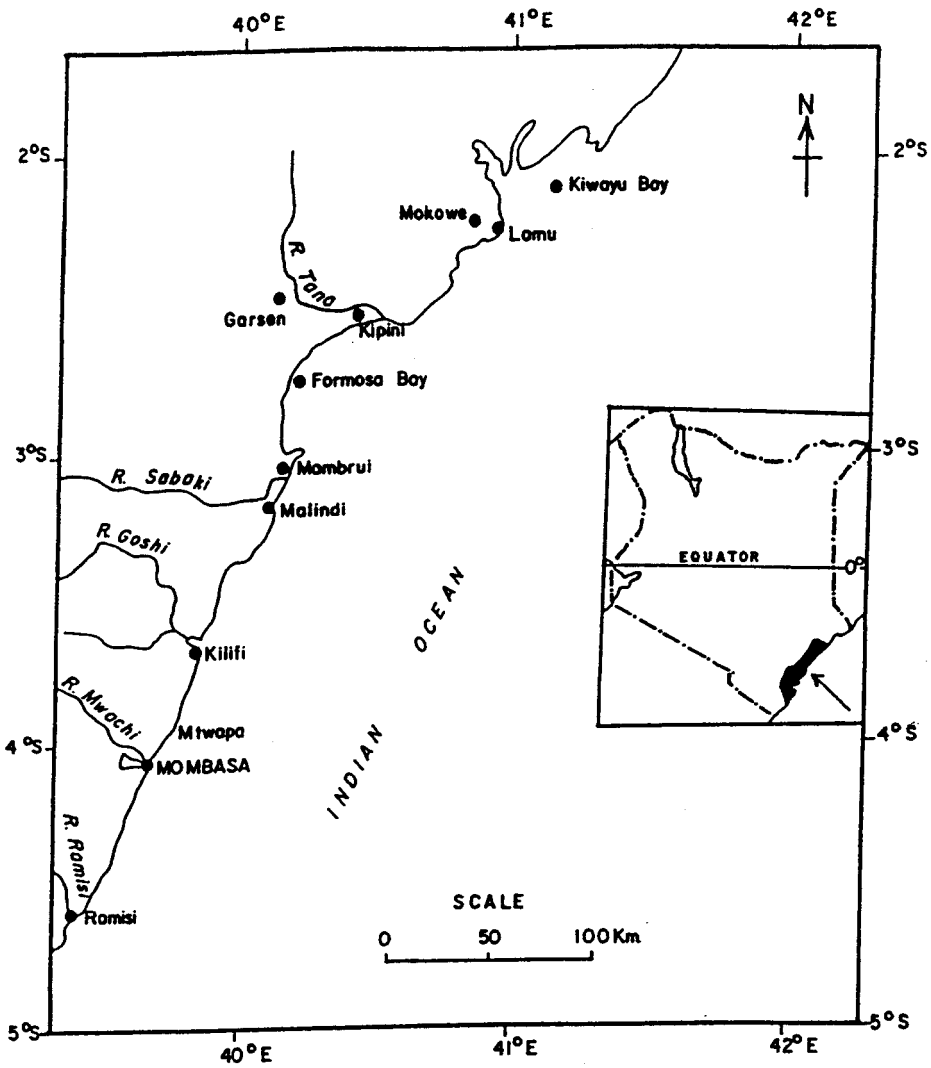

Figure 1. The Indian Ocean coastal area of Kenya showing the Tana and Sabaki rivers.

plant uptake. Aldrin is used mainly for mosquito control and termite control. Highest concentration level of this contaminant (11.9 ng/g) was recorded in River Sabaki in the January waterweed samples (Table 3). During sampling, there was a lot of El Nino rain in the first half of 1998 resulting in flooding in River Sabaki basin which may have increased stagnant water pools and therefore the need for increased pesticide application.

The solubility of organochlorine pesticides is very low and therefore low concentrations are expected in contaminated waters. The residue concentrations of

lindane, DDE, endosulfan, DDT, DDD and aldrin were low in water samples from both rivers. Their concentration levels were lower than the EC/WHO drinking water residue concentration limit of 0.1 ppb. However, the concentration level of endrin in water sample from River Tana of 0.484 µg/l was surprisingly high (Table 2). This may indicate illegal use of this pesticide despite the ban.

Juvenile *Tilapia zilli* fish were sampled and their lateral muscle analysed for the residues. Immature fish tend to have higher fat content in their muscle. Although the residue concentration levels in river water were low, we expected higher concentration levels in fish due to bioaccumulation. Lindane, DDE, Aldrin, DDT were detected but in low concentrations in fish muscle (the edible portion) in this study (Table 2). This may indicate that more metabolite residues could be accumulated in the stomach, intestines and liver which were not analysed in this study. However, the residue concentration levels were lower that the FDA EPA maximum allowable limits for edible portions of 0.3 ppm (aldrin and dieldrin), 0.5 ppm (DDT, DDE and DDD) and 0.3 ppm (lindane in shell fish and frog legs) (Wandiga et al 2001).

Significant residue concentration levels were detected for lindane, DDE, endosulfan, dieldrin but low levels of aldrin. Endosulfan and dieldrin are used for controlling banana weevils, beans and mango pests in this area. Typically, lindane is used widely s technical BHC formulation for soil treatment, foliage application on fruit, nut, vegetables, ornamental and for timber in this catchment area. Low levels of dieldrin and endosulfan detected in the sediment may have been as a result flooding caused by El Nino rains experienced during the sampling period. The highest residue concentration level of 2.067 ng/g in January in Sabaki River for aldrin indicate recent input since the concentration of dieldrin was low (Table 3).

Freshwater resources worldwide have received attention with regard to toxicity due to their importance as sources drinking water supplies. Even though the concentration levels of residues detected may sometimes be low, chronic exposure can have adverse potential ecological consequences. Using the Toxicity Identification Evaluation Method developed by EPA, mutagenic effects have been reported in rivers in Japan with highest activities in the fall (Hosokawa et al 1995). The rivers in their study receive discharge from sewage treatment works and urban runoff. Hosokawa et al 1995 reported the presence of pesticide residues (mainly organophosphate insecticides and herbicides in rivers receiving discharge from agricultural activities and sewage treatment works although no acute toxicity was detected with *Daphnia magna*. The concentrations were elevated after flooding. In Egypt, El-Kabbany et al (2000) reported detection of 16 organochlorines (most<1 µg/g) in waters samples from a river flowing through agricultural land and urban areas although at concentration levels considered to be low. They also reported residue concentration ranges of 2.3-280.7 µg/L (ΣDDT), 13.2-86.2 µg/L (BHC) and identified total DDT and heptachlor as some of the predominant pesticide residue contaminants in fish from the river Nile.

Some of the persistent OC's such as DDT have also been detected in sediment and fish samples in countries where its use has been discontinued for a long time. This can be due to aerial deposition or illegal use. Using sediment cores, Barra et al (2001) detected highest concentration of residues (0.64-1.4 ng/g dry weight) in the 1993-1996 river sediment core eight years after DDT ban. Miadis (1998) analysed 69 natural water samples (including 44 of drinking water) from different regions in Greece and found lindane and its isomer α-BHC most frequent although with high levels of heptachlor (0.05-0.1μ/L) and α -BHC (0.01-0.05 μg/L) which were well below the EU maximum acceptable levels for surface waters (Barasa 1998, EEC/80/6778).

It is expected that organochlorine pesticides would be less persistent in tropical aquatic environments due to persistently high temperatures and intense solar radiation intensity which accelerate their degradation and volatilisation rates (Wandiga 1996). The major route of transfer of OC's in tropical areas is through volatilisation due to their high vapour pressures. In paddy areas, for example, up to 99.6 % lindane was reported to be removed into the atmosphere and only 0.1% drained in to the sea (Rajendran et al 1992). Degradation of these pesticides also occurs in river water which can account for the low concentrations detected especially in high intensity agricultural areas where concentration levels of residues would be expected to be high (Eichelberg and Lichtenberg 1971, El-Kabbany et al 2000). In river water, heptachlor is completely degraded to equal concentrations of 1-hydroxy chlordane and heptachlor epoxide metabolites in four weeks, both isomers of endosulfan completely to endulfan alcohol in two weeks, whereas aldrin degrades to its epoxide (80%) in eight weeks (Eichelberg and Lichtenberg 1971). Although the concentration levels of organochlorine pesticide residues found in our study were low and do not represent any health risk, continuous monitoring of residues needs to be done, especially covering other rivers and lakes located in strategic agricultural and urban areas that may be threatened by contaminant discharge. Since chronic exposure even at even low concentrations can affect natural communities such as zooplanktons in aquatic ecosystems, such monitoring should include biological studies

According to literature, the OC concentration in sediment, water, weeds and fish in relatively less polluted areas are in the ng/g range while in polluted areas the concentration levels are in the ng/g-μg/g range. Therefore the concentration levels of organochlorine residues in our samples were low showing that these two rivers are currently non-polluted with respect to the organochlorines.

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