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**Heavy Metals and Pesticides in Marine Sediment,
Seawater, and Seaplants Along the
Kenya-Mombasa Coastline**

**Mildred P. Oyugi,¹ Sumesh C. Chhabra,^{1,*} Wilson M. Njue,¹
and Anthony M. Kinyua²**

¹Chemistry Department, Kenyatta University, Nairobi, Kenya

²Institute of Nuclear Science, University of Nairobi, Nairobi, Kenya

ABSTRACT

Marine sediment, seawater and several species of seaplants along the Kenya-Mombasa coastal region were analyzed to determine the levels of heavy metals by AAS and EDXRF and organochlorine pesticides (OCPs) by GLC-ECD. In marine sediments, there were high levels of manganese (1100 µg/L) at Vanga, compared to other study sites. In seawater, the heavy metals were detected in levels higher than those considered as natural in the ocean and the range was 0.01–0.30 mg/L. OCPs were detected in marine sediments and in seaplants. The trend of concentration of these pollutants in ascending order in seawater, marine sediment and seaplants was observed.

Key Words: Environmental samples; Marine samples; Trace elements; AXIL EDXRF; Kenya-Mombasa coastline; Sediment; Seawater; Seaplant.

INTRODUCTION

The increase in public awareness and concern about the state of the environment has been prompted by ever-growing evidence on the extent to which pollution has

*Correspondence: Sumesh C. Chhabra, Chemistry Department, Kenyatta University, P.O. Box 43844, Nairobi, Kenya; E-mail: scchhabra@avu.org.



caused severe environmental degradation. The introduction of harmful substances into the environment has many adverse effects on human health, agricultural productivity and natural resources. Pollution though, is an inevitable part of human activity, it is important to appreciate its extent, causes, the substances involved, their biological^[1] and environmental^[2] effects and methods of controlling and rectifying it. There is a growing concern over pollution of our Kenyan marine ecosystem tagged to the impact of industrial growth, interference with natural processes and disposal of wastes.

There have been several reported research works and data on the state of marine pollution from various parts of the world like Australia,^[3] Pacific ocean^[4] and India^[5] and also other pollution related studies worldwide such as in Pakistan.^[6] This kind of finding, however, has yet not been reported for the Kenyan coastal ecosystem.

The Kenyan coastline extends for about 480 km between 1°40'S and 4°40'S bordering Somalia in the North and Tanzania in the South and has five districts bordering it. Mombasa is the largest urban centre, the principle seaport serving several land locked east and central African countries. It experiences a bi-modal rainfall pattern and is quite warm throughout the year. The human population in Mombasa District is approximately 653,000, second largest in Kenya after Nairobi.^[7] As a result, the coastal area and resources are under pressure from the rapid population growth and increasing resource exploitation. Economic activities in the coastal area include tourism, industries (more than 65) and subsistence agriculture. Marine pollution has the potential to damage the tourism industry while other industrial activities such as shipping and agriculture may promote marine pollution.

The heavy metals and organochlorine pesticides can find their way into the marine flora, seawater and sediments.^[8] They are toxic and can be incorporated into biological processes in the ocean.^[9] It is essential to have knowledge of the contamination levels and the associated levels of risk to man, the marine ecosystem and to identify these polluting agents and then find the possible remedies.

The objectives of the present work were to determine the concentration levels of heavy metals and organochlorine pesticides in marine sediment, seawater and seaplants, and to study the trend of the marine pollutants along the coast of Mombasa Island. Marine samples of sediments, seawater and seaplants along the coast of Mombasa Island were analyzed for the presence of heavy metals by atomic absorption spectrophotometry (AAS) and energy dispersive X-ray fluorescence (EDXRF), while organochlorine pesticides (OCPs) were analyzed by gas liquid chromatography-electron capture detector (GLC-ECD).

MATERIALS AND METHODS

Sampling and Sample Preparation

The ten sampling locations were specifically at the English Point, Nyali Bridge, Kenya Meat Commission (KMC), Makupa Creek, Port Reitz, Vanga, Ramisi, Diani Beach, Mtwapa Creek and Marine Park (Fig. 1).



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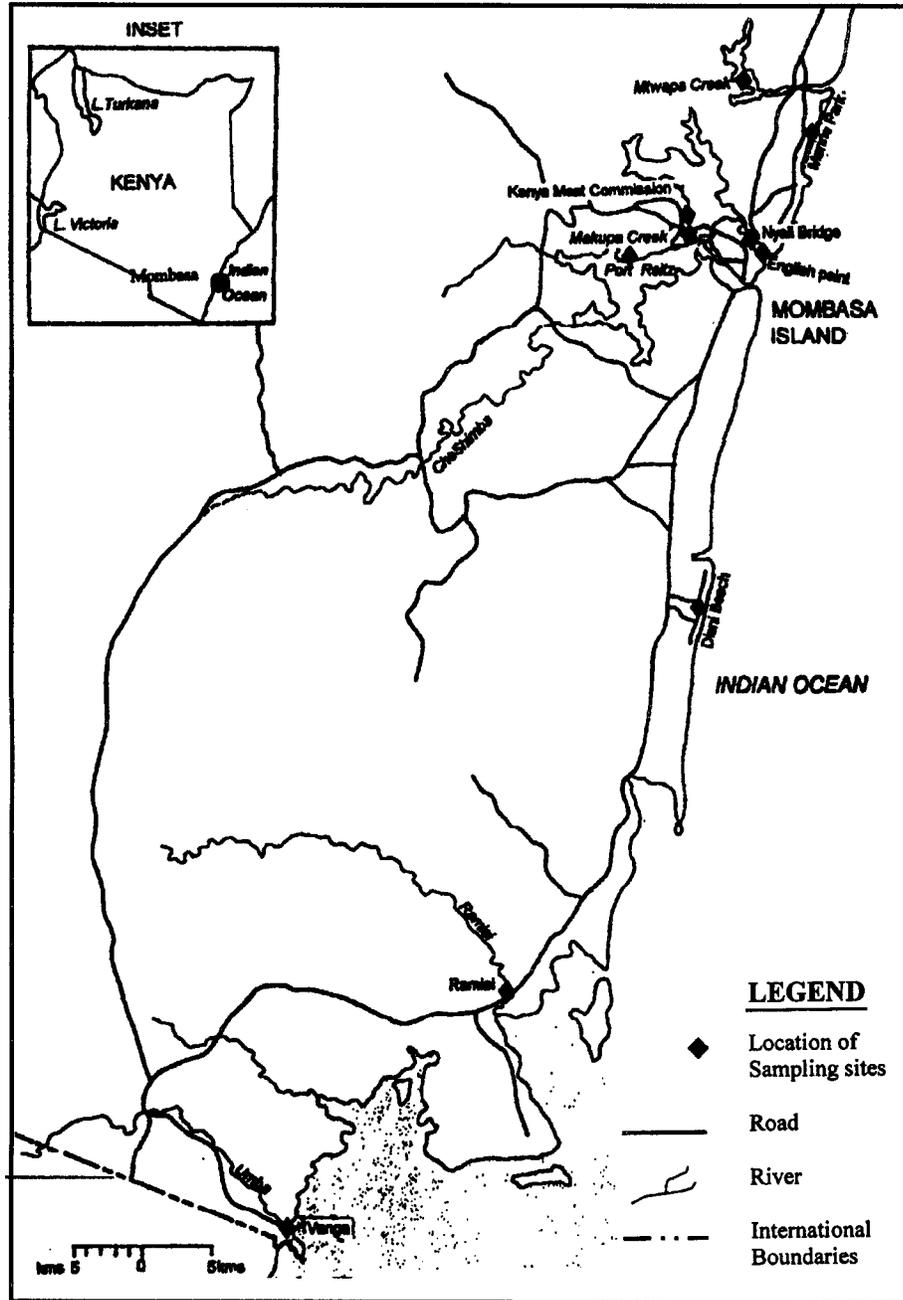


Figure 1. A map of Kenyan coastal line showing sampling sites.



Sampling of seawater, seaplants, and marine sediments was done at low tide ocean current. The vicinity of the sampling zone within a sampling site covered a range of at least 5 m radius from the shoreline at randomly chosen spots.

Seawater samples for heavy metal analysis were collected directly into clean plastic bottles from five random sampling points by submerging and opening the sampler bottle beneath the water surface. The samples were immediately acidified with 1 mL analar concentrated nitric acid (HNO_3) to pH of 3.5.^[10] The samples were then preserved at a low temperature of 4°C before further treatment and analysis by EDXRF.

The above samples (100 mL) were digested with a mixture (30 mL) of HNO_3 and HCl (3:1) in a 500 mL digestion flask at 100°C until the volume was reduced to about 30 mL. The solution was cooled, added HCl (10 mL) and double distilled water to make the standard solution. The pH was adjusted between 4 and 5 by exposing the samples to ammonia solution and nitric acid vapors before preconcentration for 15 min using sodium diethyldithiocarbamate trihydrate.^[11] The samples were then filtered through a millipore filter paper (0.45 μm).

Marine sediments were collected using a sediment sampler (corer) up to a depth of about 5 cm beneath the seabed and placed into clean plastic bags. The samples were preserved at temperature of 4°C before preparation and analysis by AAS and EDXRF.

The sediment samples were air dried, then oven dried at 60°C, ground and homogenized.

For EDXRF analysis, the samples were mixed with maize starch (33.3%) as a binder and made into pellets using a hydraulic pelletizer.^[12]

For AAS analysis, each sample (0.5 g) was heated with HNO_3 (2 mL) in a Teflon beaker to dryness at 80°C, added acid mixture (5 mL) of $\text{HNO}_3/\text{HClO}_4/\text{HF}$ (15:1:5) and further evaporated to dryness. Added 2 drops of H_2O_2 and the mixture was cooled to room temperature. The contents were then transformed to a volumetric flask (25 mL) and made to volume with HCl (0.1 M).^[13]

Seaplants were manually sampled either by uprooting or collected from those drifted to the shoreline and packed under water in clean polythene bags. Excess water was drained and then transported to the laboratory at a temperature of 4°C. The species were identified at an arboretum at Kenya Marine and Fisheries Research Institute, Mombasa. For AAS analysis, the seaplants samples were oven dried at 60°C. The dried sample (0.5 g) in a Teflon crucible was wetted with a few drops of double distilled water, mixed with concentrated HNO_3 (5 mL) and evaporated to dryness into dry salts. The salts were then dissolved in HNO_3 (5 mL), added HClO_4 (5 mL) and the solution boiled to remove HClO_4 fumes. The solution was made to 100 mL with double distilled water.^[14]

Seaplants and marine sediments for GLC-ECD analysis were collected and packed in aluminium foil to avoid contamination. Excess water was drained and then packed and transported to the laboratories at a temperature of 4°C. Wet sediments and plant samples were homogenised with sodium sulphate separately in the ratio 1:3 and left to dry overnight. One hundred and fifty milliliters hexane was added to each dried sample and pesticide extraction in soxhlet apparatus was carried out for 8 h at 70°C. This was heated to concentrate to 100 mL at 30°C under reduced pressure before analysis.

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The samples of seawater were extracted using hexane and dichloromethane sequentially; the combined extracts were concentrated at 30°C under reduced pressure and passed through a florisil chromatographic column before analysis.^[15]

Instrumentation and Analysis**AAS**

The instrument used for analysis of heavy metals (Cu, Pb, Mn, and Zn) was an atomic absorption spectrometer (AAS), Model 901 GBC (Scientific Equipment) with an air/acetylene flame temperature of 230°C. Details of the instrument, conditions adopted for estimation, standards and calibration for the heavy metals in samples of sediments and seaplants were adopted as described in detail by Gruenwedel and Whitaker.^[16] Other instrumental parameters were as given in Table 1.

EDXRF

Determination of heavy metals was done in triplicate by exposing millipore filters and pellets to a cadmium source (¹⁰⁹Cd) for 3000 s and 1000 s, respectively. The detector used was a semi-conductor detector lithium drifted silicon Si(Li). The spectral data were collected using a personal computer based Canberra S-100 multi-channel analyzer and analyzed by the software packages AXIL^[17] for spectrum fitting and quantitative X-ray analysis of environmental samples and QXAS^[18] for the evaluation of elemental concentrations. Matrix corrections were carried out using the emission–transmission method.^[19] Five pellets of about 50 mg/cm² area mass were prepared from each sample for EDXRF analysis using a die set 2.5 cm in diameter, while five replicates of about 1 g of the samples were directly analyzed. These samples were digested by PTFE pressure vessels for EDXRF analyses. The intensities of the characteristic radiation from a sample is the basis of quantification for an element *i* and is given by the equation:

$$I_i = G_o K_i (\rho_i d) \left[\frac{1 - e^{(-\rho d)}}{\rho d} \right]$$

where

Table 1. AAS instrumental parameters.

Operating parameters	Cu	Pb	Mn	Zn
Wavelength (nm)	324.7	217.0	279.5	313.0
Slit width (mm)	0.2	0.3	0.2	0.2
Lamp current (mA)	5.0	4.0	5.0	4.0
Sensitivity (mg/L)	0.04	0.015	0.04	0.01
Detection limit (µg/L)	5	20	5	3



- I_i = Fluorescence intensity (counts/s)
 G_o = Geometric constant (counts/s)
 K_i = Relative excitation detection efficiency (cm²/g)
 d = Thickness of material (cm)
 ρ = Density of the sample (g/cm³)
 ρ_i = Partial density of element i within the sample (g/cm³)
 a = Combined absorption coefficient for primary and fluorescent X-rays in the sample

GLC-ECD

The GLC-ECD model used was of Varian 3400 with an integrator Model Varian 4400. The operating parameters for analysis of OCPs by GLC-ECD included initial column temperature (100°C), final column temperature (220°C), injector temperature (230°C), detector temperature (300°C), detector type (electron capture), column type (capillary mega bore), carrier gas (nitrogen), carrier gas flow rate (5 mL/min), make-up gas flow rate (25 mL/min), run time (15 min), chart speed (1 cm/min), detection limit (0.01), sample size (2 µL) and mixed standard conc. (0.01 µg/g). The concentration of the triplicate measurements was calculated^[20] using the following equation:

$$C_x = \frac{C_{\text{std}} \times A_s \times D_f \times V_f}{A_{\text{std}} \times V_s}$$

where C_x is the concentration of the sample (µg/g), C_{std} is the concentration of mixed standards (µg/g), A_s is the peak area for sample (cm²), D_f is the dilution factor, V_f is the final volume (µL), A_{std} is the peak area for mixed standards (cm²) and V_s is the volume of the sample (µL).

RESULTS AND DISCUSSION

The results obtained in the present study are tabulated (Tables 2–5). The heavy metals in marine sediments were analyzed by AAS and EDXRF (Table 2). In analysis by AAS, Pb was recorded only at Makupa Creek and Vanga, Cu and Zn were recorded highest in marine sediment from Diani Beach an active tourist beach site and Makupa Creek a famous dumping site.

For seaplants (Table 3), not all species breed in all sites, hence, it was not possible to obtain similar species at all the sampling sites. These were analyzed by AAS only. All species (except *Ulva reticulata* from Makupa Creek) were detected to have some Pb. The highest recorded level was in the *Chaetomopha* sp. from Makupa Creek. This may be attributed to the dumping activities and by virtue of this being the main causeway. Cu was detected highest in the *Glacilaria* sp. from Diani Beach. This could be due to long time accumulation since Cu was not detected in most plants. At the Nyalı Bridge, Zn was recorded with the highest level in *Myrica* sp. that may be a Zn tolerant seaplant. A lot of traffic at Nyalı Bridge might have contributed to the Mn levels in the *Sargassum* sp.



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Table 2. Mean concentration ($\mu\text{g/g}$) of heavy metals by AAS and EDXRF in marine sediment.

Site	Pb		Cu		Zn		Mn	
	AAS	EDXRF	AAS	EDXRF	AAS	EDXRF	AAS	EDXRF
English Point	nd	nd	22 \pm 0.02	25 \pm 1.07	28 \pm 0.11	31 \pm 1.21	nd	198 \pm 2.01
Nyali Bridge	nd	nd	21 \pm 1.11	23 \pm 1.03	48 \pm 1.01	51 \pm 0.01	17 \pm 1.87	169 \pm 1.12
KMC	nd	nd	20 \pm 1.09	24 \pm 0.02	88 \pm 0.02	85 \pm 0.01	220 \pm 3.01	221 \pm 1.22
Makupa Creek	80 \pm 0.10	97 \pm 0.02	43 \pm 0.03	47 \pm 3.00	348 \pm 2.01	354 \pm 2.04	239 \pm 1.01	247 \pm 1.06
Port Reitz	nd	nd	25 \pm 0.02	27 \pm 0.01	90 \pm 1.10	93 \pm 1.24	nd	463 \pm 3.02
Vanga	50 \pm 0.02	52 \pm 0.02	26 \pm 2.01	27 \pm 1.22	77 \pm 3.00	90 \pm 1.17	1100 \pm 2.13	1300 \pm 0.03
Ramisi	nd	10 \pm 0.02	27 \pm 1.43	27 \pm 0.04	72 \pm 0.02	75 \pm 1.02	723 \pm 1.79	724 \pm 0.01
Diani Beach	nd	nd	50 \pm 0.08	52 \pm 3.22	60 \pm 0.12	66 \pm 0.14	nd	116 \pm 0.04
Mtwapa Creek	nd	nd	32 \pm 1.22	32 \pm 2.11	70 \pm 1.04	72 \pm 0.98	nd	362 \pm 1.08
Marine Park	nd	nd	19 \pm 0.02	25 \pm 1.09	38 \pm 0.05	43 \pm 0.75	648 \pm 1.88	784 \pm 1.02

**Table 3.** Mean concentration ($\mu\text{g/g}$) of heavy metals by AAS in seaplants.

Site/Plant species	Pb	Cu	Zn	Mn
Port Reitz				
<i>Glacilaria</i> sp.	8 ± 0.48	nd	15 ± 0.94	8 ± 0.01
<i>Ulva reticulata</i>	7 ± 0.39	1 ± 0.01	29 ± 0.83	35 ± 1.02
Makupa Creek				
<i>Chaetomorpha</i> sp.	67 ± 0.01	10 ± 0.09	76 ± 0.10	100 ± 2.11
<i>Sargassum</i> sp.	14 ± 0.13	2 ± 1.15	23 ± 1.13	56 ± 1.46
<i>U. reticulata</i>	nd	3 ± 0.44	12 ± 1.11	nd
Diani Beach				
<i>U. reticulata</i>	8 ± 0.50	7 ± 0.03	27 ± 0.02	nd
<i>Sargassum</i> sp.	13 ± 0.52	nd	23 ± 0.07	40 ± 1.6
<i>U. fasciata</i>	13 ± 1.04	5 ± 0.01	22 ± 1.35	16 ± 2.0
<i>Glacilaria</i> sp.	13 ± 0.53	45 ± 0.03	8 ± 0.42	8 ± 0.01
<i>Thalassalendron</i> <i>ciliatum</i>	13 ± 0.08	nd	8 ± 1.01	10 ± 0.18
<i>Padini gymnospora</i>	13 ± 1.01	12 ± 0.02	8 ± 0.06	nd
Nyali Bridge				
<i>T. ciliatum</i>	6 ± 0.50	36 ± 1.07	60 ± 0.11	nd
<i>U. reticulata</i>	13 ± 1.20	1 ± 1.23	8 ± 1.24	nd
<i>Sargassum</i> sp.	9 ± 0.55	nd	4 ± 0.01	189 ± 3.44
<i>U. fasciata</i>	11 ± 1.01	nd	18 ± 0.08	23 ± 1.92
<i>Myrica</i> sp.	11 ± 0.65	nd	139 ± 0.43	nd
<i>Glacilaria</i> sp.	15 ± 0.11	9 ± 0.02	8 ± 1.23	12 ± 0.01
<i>Hypnea</i> sp.	12 ± 0.01	nd	10 ± 1.01	31 ± 1.03
Marine Park				
<i>U. reticulata</i>	10 ± 0.09	nd	34 ± 0.07	79 ± 2.02
<i>S. isoetifolium</i>	10 ± 0.02	nd	nd	116 ± 2.93
<i>U. fasciata</i>	14 ± 1.30	nd	nd	nd
<i>Sargassum</i> sp.	9 ± 0.01	nd	8 ± 0.01	58 ± 3.24

Table 4. Mean concentration (mg/L) of heavy metals by EDXRF in seawater.

Site	Pb	Cu	Zn	Mn
English Point	0.02 ± 0.01	0.02 ± 0.01	0.05 ± 0.01	0.14 ± 0.03
Nyali Bridge	0.12 ± 0.02	0.02 ± 0.01	0.06 ± 0.01	0.03 ± 0.01
KMC	0.03 ± 0.01	nd	0.06 ± 0.02	0.06 ± 0.03
Makupa Creek	0.06 ± 0.02	nd	0.10 ± 0.02	0.05 ± 0.01
Port Reitz	nd	nd	0.04 ± 0.02	0.03 ± 0.83
Vanga	0.03 ± 0.02	0.05 ± 0.02	0.07 ± 0.01	0.08 ± 0.02
Ramisi	nd	0.02 ± 0.01	0.06 ± 0.36	0.30 ± 0.02
Diani Beach	0.04 ± 0.02	0.02 ± 0.01	0.06 ± 0.02	nd
Mtwapa Creek	nd	0.02 ± 0.01	0.05 ± 0.02	0.03 ± 0.40
Marine Park	0.02 ± 0.01	0.02 ± 0.01	0.03 ± 0.01	0.02 ± 0.01



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Table 5. Mean concentration of organochlorine pesticides in sediments and seaplants by GLC-ECD.

Site/Sample	Pesticide conc. ($\mu\text{g/g}$)
English Point	
Marine sediment—BHC	0.014 ± 0.02
<i>Amphiroa</i> —BHC	2.571 ± 0.01
Nyali Bridge	
Marine sediment—BHC	0.025 ± 0.01
Marine sediment—Endosulfan	0.322 ± 0.01
<i>Sargassum</i> —Aldrin	0.076 ± 0.01
KMC	
Marine sediment—BHC	0.032 ± 0.02
Makupa Creek	
Marine sediment—pesticides	nd
Port Reitz	
Marine sediment—Aldrin	0.134 ± 0.02
<i>Sargassum</i> —BHC	0.193 ± 0.01
Vanga	
Marine sediment—pesticides	nd
<i>Padini</i> —Aldrin	0.087 ± 0.02
Ramisi	
Marine sediment—pesticides	nd
<i>Padini</i> —BHC	0.094 ± 0.01
Diani Beach	
Marine sediment—pesticides	nd
Mtwapa Creek	
Marine sediment—pesticides	nd
<i>Thalasalendron</i> —BHC	0.045 ± 0.01
Marine Park	
Marine sediment—pesticides	nd

Pb, Cu, Zn, and Mn were detected in seawater samples from all the sites by EDXRF (Table 4). Compared with the studies^[3] taken near a lead smelter in Australia, the concentrations of Pb and Zn in this study fell within the ranges obtained in the former study. This implies that our marine ecosystem is polluted, as it would be expected that these values should be lower than the former study since there is no lead smelter at the Kenyan coast. With respect to the natural concentrations of Cu, Zn, Pb, and Mn, the seawater at the Mombasa coastline portrays evidence of marine pollution since the levels were found to be higher than the natural levels.^[11] Although, seawater is not directly used for drinking purposes, its pollution may affect the food chain. The levels of Pb, Cu, Zn, and Mn were compared to those given as proposed standards by Park^[21] as a guideline for the safety of the seawater. The concentration of Pb in seawater at the Nyali Bridge and Makupa Creek and that of Mn at the English Point, KMC, Vanga and Ramisi show evidence of marine pollution since these are higher than the proposed value of 0.05 mg/L.

Analysis of OCPs in marine sediment indicated BHC in most of the samples (Table 5) in particular at the English Point, Nyali Bridge, and KMC. Aldrin and endosulfan were detected only at Port Reitz and Nyali Bridge, respectively.



In seaplants, BHC was recorded highest in *Amphiroa* sp. at the English Point. Other species in which BHC was detected include *Sargassum*, *Padini* and *Thalasalendron* sp. from Port Reitz, Ramisi and Mtwapa Creek, respectively. Aldrin was recorded in *Sargassum* sp. from Nyali Bridge and *Padini* from Vanga. Except for the mentioned above, all other OCP's in the seaplants were below the detection limit of the GLC (0.001 µg/g).

Seawater from all sites did not show any levels of OCPs. These levels, if any, may have been much lower than the detection limit for the pesticides analyzed.

The level of BHC in the *Amphiroa* sp. from the English Point was higher than that in the sediment from the same site. This may be due to higher absorption and accumulation of pesticide in plants than in sediment. However, for BHC recorded in plant species from Port Reitz, Ramisi and Mtwapa Creek, the corresponding sediment samples had levels of pesticides lower than the detection limit. This may have been due to drifting of these species to the sample collection sites. Hence, absorption of the pesticide may not have been from the sampling site. Aldrin is reported in sediment from Port Reitz unlike from the seaplants in the region. These seaplants may, therefore, have been drifted to this site. This also applies to the seaplants from Vanga, Ramisi, Nyali Bridge and Mtwapa Creek.

Quality assessment and control of heavy metal analytical techniques (AAS and EDXRF) was achieved by correlating results of both techniques. The trends of concentrations of the heavy metals were generally higher for EDXRF than for AAS. However, a two-tailed student *t*-test carried out on the two sets of results at a 95% confidence limit implied that there was no significant difference between the

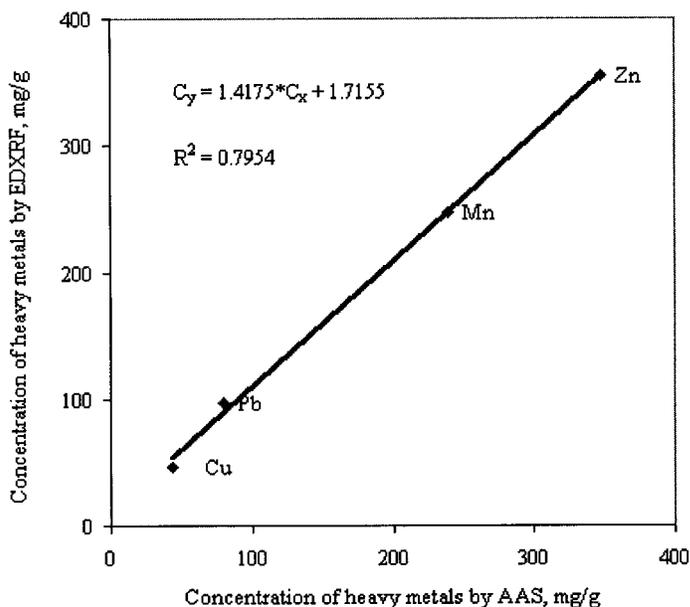


Figure 2. Correlation of the concentration of heavy metals determined in marine sediment samples at Makupa Creek.



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two methods. Figure 2 shows correlation of heavy metals by both methods in marine sediment from Makupa Creek. This was also done for the other sites and the r -values showed strong positive correlations. For organochlorine pesticides analysis, quality assessment and control was achieved by analyzing standards as reference materials under similar conditions as for the samples.

The results obtained in this study may be attributed to characteristic activities at the sampling sites that may promote marine pollution. These include accumulation of the heavy metals, dumping activities and heavy traffic at the Makupa causeway and Nyali Bridge. External effect of pollution could be from the River Uмба which discharges into the Indian Ocean at Vanga. Agriculture though subsistence with application of fertilizers is a contributing factor to marine pollution by both heavy metals and distant transport of OCP's. The influence of the agricultural show ground near the English point, the oceanic mixing processes, wastes from the slaughterhouse at the KMC contribute positively to pollution of the Kenya-Mombasa coastline. There are about 65 industries ranging from fishing, slaughter, milk, grain millers, bakeries, iron, steel, oil refineries, textiles, and breweries among others. These may have an adverse effect on the status of the marine ecosystem.

CONCLUSION

The results of the present study provide evidence of pollution of the Kenya-Mombasa coastline. The distribution of these pollutants can be described as random as explained by the physical influence of the oceanic motions of many kinds like current, waves, inputs from surrounding areas and turbulent mixing processes. The expected general trend^[5,22,23] of higher concentration of heavy metals and OCPs in seawater, marine sediment and seaplants is in increasing order from the same site and was observed in most of the sampling sites.

The management of ocean and coastal environment may not be an easy matter, due to contaminants from multiple sources. It is necessary to have thorough waste management programs as a strategy to minimize marine pollution since prevention is better than cure. Industries do have a moral, legal and economic responsibility to consider waste treatment as an integral part of their production expense and may, therefore, assist in reducing marine pollution. For example, by giving permission before wastes are discharged to the sea and by implementing techniques that reduce toxic levels of pollutants to non-toxic levels. The results of this work can be used to generate heavy metal profile for marine pollution monitoring and also cover other forms of pesticides such as the organophosphates so as to fully establish extend of marine pollution at the Kenyan coast.

ABBREVIATIONS

AAS, atomic absorption spectrophotometer; AXIL, analysis of X-ray spectra by iterative least squares fitting; EDXRF, energy dispersive X-ray fluorescence; GLC-ECD, gas liquid chromatography-electron capture detector; KMC, Kenya Meat



Commission; nd, not detected; OCPs, organochlorine pesticides; PTFE, polytetrafluoroethylene; QXAS, quantitative X-ray analysis system; sp., species.

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