

## **Distribution of heavy metals and total hydrocarbons in coastal waters and sediments of Cross River State South Eastern Nigeria**

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**ABSTRACT** - The mean concentrations and distributions of ten heavy metals (Cr, Cd, Ni, V, Co, Pb, Zn, Fe, Mn, Cu) and hydrocarbon levels found in the coastal waters and sediments of Cross River State are herein reported. The results indicate background levels to high contamination found in industrial areas. The most abundant metal was Fe; with a value of  $0.79 \pm 0.14 \text{ mg/l}$  in water and  $2227.5 \pm 1062.2 \text{ mg/kg}$  in sediments. The study also showed that coastal waters in the proximity of the South Atlantic contained higher concentrations of metals than inland waters. The spatial distribution of metals in sediments followed a similar sequence with Cu or Cd being the least available metals and Fe the most dominant metal at all locations. Total hydrocarbon concentrations ranged from undetectable to  $263.2 \text{ mg/l}$  in water and 190 to  $758.4 \text{ mg/kg}$  in sediment. The enhanced levels of Cr, Zn, Mn, and Cu and THC in most locations monitored point to contamination of the coastal waters by industrial and sewage inputs.

*Key Words: Heavy metals, hydrocarbons, water, sediment.*

### **INTRODUCTION**

There has been a growing concern world wide that near shore marine ecosystems are being affected by man made pollutants leading to environmental stress (Clark et al; 1997; Michel and Zengel, 1998; Asuquo, 1999a). For instance, a single oil spill in Puerto Rico resulted in the contamination of extensive areas, impacting both on biotic and abiotic resources along more than 48km of shore, killing thousands of marine organisms. (Mignucci-Giannoni, 1999). Furthermore, the decomposition of dead wild life aggravated the situation by generating obnoxious odour and exerting high BOD.

There has been concern exemplified in Cross River State and Nigeria as a nation whose coasts have been besieged by incessant oil spills in recent times. For instance, a major oil spill occurred in January, 1998, and brought these concerns to forefront and gave impetus for the government investigating team to study the impact of the oil spill on coastline, estuaries and fluvial systems. This study had confirmed the fears of oil spill impact on the marine ecosystem (Mobil Report, 1998).

Besides oil spills and other petroleum related activities, other human activities affecting the coastline are municipal drainage, sand mining and dredging, quarrying and agro-industrial ventures in addition to atmospheric fall outs. All these introduce large quantities of heavy metal into coastal waters (UNEP, 1992; Wagner, 1974). River borne metals tend to sediment before eventual transportation into open sea (Nicolaidou and Nott, 1998; UNEP, 1992).

Generally, water along the coasts (including the estuaries) contain a variety of marine resources namely fishes, shell fish and seaweeds (Gregory *et al*; 1996; Muse *et al.*, 1999) which attract fishing activities. These fishery resources are exploited by the riverine communities of Cross River and Akwa Ibom States of Nigeria, not only for local consumption but also for income generation to meet other domestic needs.

Additionally, estuaries and coastal ecosystems are very delicate because they serve as nursery grounds for most species in the sea (Weyl, 1970; Giddings, 1973; Wagner, 1974; UNEP 1992). The concern here is the contamination of water resources by potentially toxic chemicals (such as heavy metals and hydrocarbon) (THC). Pollution problems may include toxicity to aquatic plants and animals. Uptake of these toxins in the tissues of organisms may create health hazards to human and predators that consume them (Smith *et al*; 1996).

## **STUDY AREA**

The Cross River Coast is a low lying environment, ranging from the fine sandy beach at Abana/Atabong (in Bakassi Peninsula), and stretching upstream to lower Cross River, Calabar, Great Kwa River and the mud(silty clay) sediments at Ilang and Ikoneto (Fig.1). The area covered lies between Latitude. 4°30' and 5°15'N, and Longitude. 6° 00' and 8°10'E. The entire coastline here was divided into five (5) areas: Calabar Municipality (CM), Calabar South (CS), Bakassi (BA), Odukpani (OD) and Akpabuyo (AK) during the study. Most of the areas up to the Cross River estuary are covered by mangrove swamp forest.

The estuaries area are influenced by semi diurnal tides that go upstream to about 80km. Influx of pollutants into the coastal waters are enhanced by the two principal ocean currents: north-ward rising Benguela Current, and eastward flowing Guinea Current. These currents flow through offshore areas where crude oil prospecting and production are located. Accidental oil spills in this zone are transported by prevailing winds and waves into Cross River estuary, the associated mangrove swamps, tidal flats, creeks and adjoining tributaries. These are the main sources of hydrocarbon and heavy metals in this coastal region. The effects of oil activities are aggravated by the fact that the shoreline is low energy, shallow water environment and highly vulnerable and sensitive to the frequent oil spills in the area. The zone is also one of the busiest fishing grounds in the coast of West Africa.

This study, therefore, was to assess the level of heavy metals and hydrocarbon in water and sediments from the Cross River coastal water with the view to determining the pollution index, if any, and recommending abatement measures.

## **MATERIALS AND METHODS**

Sampling of surface sediments was carried out by the use of hand driven sediment auger. Composite samples were collected in labeled low-density polyethylene (LDPE) bags from each location and taken in ice cool boxes to the laboratory for analysis. Samples were air dried overnight, pulverized and sieved through 2mm mesh size (Loring and Rantala, 1992). The physical and chemical properties of sediments were determined as reported by Asuquo *et al.*, (1998) involving the measurement of soil pH, using WTW pH meter, electrical conductivity using conductivity meter (Hach, Model 4460), exchangeable cations and organic carbon contents of the sediments.

Total Hydrocarbon (THC) was determined by extracting the sediment samples with redistilled n-hexane and measuring the THC content colorimetrically at 430nm using Hach spectrophotometer, Model DR/3000. Heavy metal contents were extracted from the sediment samples by acid digestion using a mixture of nitric and perchloric acid (Barnhisel and Bertsel, 1982). The metal concentration in the extracts were read on Atomic Absorption Spectrometer (AAS), Hewlett Packard, Model 2380.

## **WATER SAMPLING AND ANALYSIS**

A total of 37 water samples were collected from the estuaries and surrounding rivers for chemical analysis. Water temperature, pH, Dissolved Oxygen (DO), and electrical conductivities were measured *insitu*, using Oxyguard Handy MK II electronic meter (sensitivity  $\pm 1.0\%$ ) for DO and temperature, WTW-pH electronic meter (sensitivity  $\pm 0.1\%$ ) for pH and WTW LF - 90 conductivity meter ( $\pm 1.0\%$ ) for conductivity respectively. Water samples for physicochemical analysis were collected in one liter high density polyethylene (HDPE) bottles and stored at about 4°C prior to analysis within 5 days (UNESCO/WHO, 1984).

THC (Mg/L in water was determined by colorimetric method after extracting 250ml in two batches with 100ml n-hexane. The extract was concentrated by careful evaporation on regulated water bath. The contents were measured at 430nm using DR/300, Hach Spectrophotometer.

**Heavy metals (mg/L):** The content of each metal in water was determined from the samples after due calibration runs using appropriate salts of the metals investigated (Cr, Cd, Zn, V, Ni Pb, Co, Mn, Cu and Fe). The metals were analysed for using AAS, Hewlett Packard Model 2380 (Precision  $\pm 0.01\%$ ) (Forsterner and Wittman, 1979).

**RESULTS AND DISCUSSION:**

**TABLE 1: Mean levels heavy metals from Cross River Coastal Waters**

Location	Cr	Cd	Ni	V	Co	Pb	Zn	Fe	Mn	Cu
CM	-	-	-	-	-	0.06	-	0.30	-	-
AK	-	0.03	0.60	0.01	0.04	0.07	-	0.79	-	-
CS	-	0.02	-	0.03	0.03	0.08	0.30	1.20	0.43	0.0
OD	-	0.01	-	0.01	-	-	-	-	-	-
BA	-	0.06	0.20	0.10	0.10	0.44	0.44	-	0.25	-

- CM = Calabar Municipality coastal waters (Brackish marine)
- CS = Calabar South coastal waters (Brackish marine)
- BA = Bakassi coastal waters (Marine)
- AK = Akpabuyo coastal waters (Fresh water)
- OD = Odukpani coastal water (Fresh water)

**TABLE 2: Heavy metal levels in sediment from Cross River system**

Location	Cr	Cd	Ni	V	Co	Pb	Zn	Fe
AK1	20.0	2.2	2.0	4.4	3.0	30.0	1.9	2169
AK2	30.0	2.2	9.0	4.8	4.5	22.5	24.0	1046
AK3	10.0	0.0	0.0	0.42	0.0	0.0	22.1	1704
AK4	20.0	4.4	15.0	4.4	9.0	37.5	20.8	2760
AK5	10.0	0.0	3.0	2.4	0.0	0.0	9.2	1602
AK6	10.0	0.0	3.0	3.2	0.0	10.0	8.7	795
AK7	0.0	0.0	3.0	3.0	0.0	0.0	11.1	1053
BA1	0.0	0.0	0.0	0.0	0.0	7.5	8.3	1476
BA2	20.0	1.6	9.0	4.0	6.0	7.5	21.7	2979
CM1	70.0	7.8	3.1	2.8	1.1	5.3	60.0	946
CM2	68.0	1.4	3.8	5.1	0.8	1.6	45.0	751
CM3	59.0	8.2	1.2	3.2	1.2	2.5	52.0	814
CM4	60.0	4.3	4.0	2.6	0.7	0.8	38.0	526

**Distribution of heavy metal**

CS1	0.0	0.0	3.0	3.2	0.0	0.0	42.0	2217
CS2	0.0	1.62	12.0	3.2	6.0	1.5	48.8	2976
CS3	30.0	1.1	6.0	4.0	1.5	0.0	26.4	2724
CS4	10.0	0.0	0.0	4.4	0.0	7.5	3.8	144
CS5	20.0	1.1	6.0	4.4	3.0	11.3	38.4	3024
CS6	10.0	0.0	0.0	0.0	0.0	0.0	2.3	1761
CS7	20.0	0.0	6.0	4.4	3.0	11.3	38.4	3024
CS8	10.0	0.54	3.0	4.4	3.0	7.5	12.0	1695
CS9	0.0	0.0	3.0	4.0	0.0	0.0	4.5	1338
CS10	20.0	1.2	3	3.2	0.0	0.0	16.5	1887
OD1	20.0	1.6	9.0	3.2	9.0	7.5	20.8	2889
OD2	20.0	1.2	3.0	2.0	3.0	0.0	10.6	2508
OD3	40.0	6.5	27.0	6.0	12.0	6.0	24.6	2616
OD4	10.0	0.54	3.0	2.4	0.0	3.8	10.1	1926
OD5	40.0	1.1	21.0	4.8	9.0	15	33.7	3123
OD6	10.0	0.0	3.0	3.2	0.0	0.0	3.0	1438
OD7	10.0	0.0	0.0	0.42	0.0	7.5	2.8	999
OD8	21.4	2.2	11.0	3.1	8.3	18.8	15.1	2157

**Table 3: Mean levels of THC in water and sediments of Cross River**

	Location	Water	Sediment
AK1	Ife Beach	2.48	93.2
AK2	Idunda Beach	3.16	112.4
AK3	Atimbo Beach	2.34	116.4
AK4	Idabo Beach	9.28	456.0
AK5	Esuk Mba	1.88	130.4
AK6	Inua Esighi Beach	3.24	190.0
AK7	Ikang Beach	2.28	377.2
BA1	Abana Beach	263.2	242.8
BA2	Atabong Beach	331.8	758.4
CM1	Ohutung Beach	115.2	288.4

*Asuquo, Ogrl and Bassey*

CM2	Esuk Utan	0.0	0.0
CM3	Edibe Beach	0.0	0.0
CM4	Kasuk Beach	3.2	222.4
CS1	Duke Town Beach	4.9	161.6
CS2	Ala Nao Iyok	0.84	149.6
CS3	Abitu Beach	1.48	139.6
CS4	Ofuo Beach	2.28	43.94
CS5	Akoni Esuk Utan	1.86	0.0
CS6	Esuk Effiom Beach	2.1	0.0
CS7	Obufa Esuk Orok	1.52	22.4
OD1	Isong Inyang	1.76	260.64
OD2	Ikoneto Beach	1.68	234.4
OD3	Mbiabo Esin Ufot	1.40	162.8
OD4	Atan Onoyom	1.58	(922.6)*
OD5	Idim Ndom Eki	26.72	(671.6 x10)
OD6	Adiabo Okurikang	27.82	0.0
OD7	Mkpara Otop Ekpo	14.7	326.8
OD8	Creek Town Beach	94.2	0.0
OD9	Inua Akpa Okovong	0.0	122.0

\*Concentrations in parenthesis are seepages from nearby untapped reservoir

**Table 4: Mean levels of physicochemical parameters of water from Cross system**

Area	Temp °C	DO MI	%Sat. DO	pH	Turbidity NTU	TSS mg/l	Pt Co	E
AK	30.7±1.1	52±1.0	70.6±13.8	162±14.2	192.4±1.65	192.4±1.65	508.6±2.96	3
BA	32.4±2.0	6.3±0.6	90±13.2	8.2±1.2	167.3	240.0	402±33.7	3
CM	29.9±1.1	3.5±0.2	67.5±1.29	6.1±1.0	62±80	4.5±31.5	412±39.5	4
CS	31.3±1.9	3.3±1.2	44.3±17.3	6.4±0.9	79.7	96.3	354±28.9	7
OD	32.1±1.1	4.9±0.1	65±1.4	6.4±0.3	21.8±17	20.4±15.8	45.5±94.2	9

### ***Distribution of heavy metal***

The mean levels of heavy metals in water of the study area are shown in Table 1. The levels of heavy metals in sediments are given in Table 2. The concentration of THC in both water and sediments are given in Table 3. The spatial variations of THC in water and sediments are illustrated in Fig.2, while the distribution of heavy metals in sediments are shown in Fig.3. The mean physiochemical characteristics of water analyzed are represented in Table 4.

Heavy metal levels were generally very low in water. Values ranged from undetectable for all metals to 1.1 mg/l for Mn, 2.1 mg/l for Pb and 2.8mg/l for Fe in the estuarine waters of Calabar South and Akpabuyo areas. The distribution pattern followed the sequence:

$Cr < V < Cd < Co \leq Cu < Ni < Zn < Mn < Pb < Fe.$

The concentration of heavy metals in sediments rather than water should reflect the extent to which the coastal areas have been impacted by industrialization. Pristine coastal sediments should have natural or background metal level depending on the soil or rock type of the area which can influence the metal concentration. High levels of heavy metals in most sediments have been attributed to industrialization (Ntekim et al., 1993) caused by anthropogenic inputs. The dynamics, variability and availability of heavy metals depend solely on the sediment and morphological characteristics. This is particularly true for mangrove swamp sediments which are composed primarily of silt/mud fractions capable of absorbing and retaining pollutants.

Higher level ( $2227.5 \pm 1062.8$  mg/kg) of Fe and ( $1503 \pm 136$  mg/kg of Mn) for BA area were obtained in such sediments. Sandy sediments on the other hand, are porous and the mobility of heavy metals are not retarded due to large pore spaces between the sand particles. Hence, the heavy metals levels were low for the riverine areas such as Zn:  $13 \pm 9.1$  at AK. And  $11.64 \pm 2.6$  at OD areas than  $15 \pm 9.5$  mg/kg at BA and  $17.1 \pm 18.0$  mg/kg at CS located at near shore areas.

Spatial variations of heavy metals in sediments measured were as follows: Cr was highest ( $64.3 \pm 5.6$  mg/kg) at CM area, Cd ( $5.4 \pm 3.1$  mg/kg) at CM area, Ni ( $10.5 \pm 2.5$  mg/kg) at AK area, V ( $3.4 \pm 1.2$  mg/kg) at CM area, Co ( $5.5 \pm 7.5$  mg/kg) at Ak area, Zn ( $48.8 \pm 9.4$  mg/kg) at CM area, Fe ( $2227.5 \pm 1062.8$  mg/kg) at BA area, Mn ( $150.0 \pm 13.6$  mg/kg) at BA area and Cu ( $2000.3 \pm 111.7$  mg/kg) at CM area.

The Calabar Municipality area was the most contaminated zone with metals such as Cr, Cd, V, Zn and Cu. The presence of these metals above background levels are indicative of inputs from industrial and domestic sewage from the metropolis. Ni and V availability (highest 10.5 and 3.4 mg/kg respectively) suggest contamination from petrogenic sources (NRC, 1985) as the THC levels were equally high in these areas.

The levels of THC in the surface water ranged from 0.84 - 331.8 mg/l with noticeable impacts on the environment in most of the areas studied. Very high levels were obtained at near shore location - above 10mg/l) recommended limit (DPR, 1991). Such areas include Atabong (331.8mg/l), Abana (263.3 mg/l) all in Bakassi Peninsula and Obutong beach (115.2mg/l) in Calabar Municipality Area (Table 2, Fig.

1). As expected, the heavy metals and THC concentration were higher in se than the water samples.

Such marked levels of THC would generate much concern as the serious environmental implications (Giddings, 1973; NRC, 1985, Clark, et al. It is widely reported that polyaromatic hydrocarbons (PAHs) such as benzo(a)pyrene, anthracene and anthrene present in crude oils are carcinogenic (NRC Clark, et al., 1997). Many marine and estuarine fish and shell fishes are known to accumulate PAHs (the main components of THC) from water and food (Clark 1997) The ingestion of hydrocarbon - contaminated sea food predisposes one to increased risk of human cancer and associated physiological problems (Giddings 1973; Mitchell, 1974; Clark, et al; 1997).

The low levels of DO (<1.4mg/l) measured in some areas confirm the impact of high level of hydrocarbon (as DO is used for biodegradation of HC) low values of DO as observed in the study are indicative of an impacted environment by HC (Giddings, 1973; USEPA, 1976). High level of HC, especially floating oil, impairs respiration of aquatic organisms and hinders bio-productivity (Tait, 1976). Persistent HC pollution especially in low energy environment as in the study area directly affect the bio-diversity of organisms (Clark et al., 1997). This was observed during this study as dwindling fish - catch was widely complained by fisherman in the area. This was supported by relatively low abundance of fish and shell fish indicated by fecundity studies (Mobil Report, 1998) which implies low spawning activity. The report also indicated declining phyto-plankton flora species abundance and diversity.

The self cleansing potential of water may facilitate the process of re-oxidation such that the retention time of pollutants and higher levels are only noticed through their interference in human activities (Asuquo, et al; 1997).

The levels of THC in sediments were remarkably high, especially for Atabong West (7584 mg/kg) and Idim Udom Eki (6,716 mg/kg). These unusually high levels in these two locations may have been caused by seepage from underground. At Idim Udom Eki, a location far removed from industrial activities, was one of the most prominent sources of THC contamination in the entire area, it was strongly suggested that there must be some seepage from the river bed. Suggestion was made for a petroleum oil exploration in the areas especially for Idim Udom Eki. The Bakassi area is also undergoing oil exploration and production activities. Underground seepage was confirmed at Atabong West where it was observed that a borehole was rendered useless due to high level contamination with THC (in taste and visual oil film on the surface of water from the borehole water).

The mean concentration of heavy metal in water samples determined (Table 1) are all within the DPR/FEPA Limit except for Cd and Pb whose permissible limits are 0.01 and 0.05 mg/l respectively (DPR guidelines).

Generally, heavy metals were more prevalent and higher in the marine than the fluvial areas. For instance Bakassi and Calabar South that are quite nearer the marine and hence marine environment showed higher levels than Calabar Municipality and Odukpani areas that are higher up the fluvial areas (Table 1, Fig. 2). Though

#### *Distribution of heavy metal*

metals such as Cr were absent from some of the areas, the relatively higher level of heavy metals in marine areas have been linked up with crude oil pollution that is prevalent in the area. The occurrence of metal indicators of petroleum origin, such as Ni and V in water and sediment samples suggest that the areas may have received much impact from oil spill as reflected the high concentration of THC in these areas. Heavy metal analysis of fresh tar balls collected from the area (Atabong West) gave results that are consistent with previous analysis confirm the source of crude from off shore wells of the areas (Asuquo *et al*; 1995 Mobil Report, 1998). This confirm the source of THC pollution here to be mostly crude oil from offshore wells. Tar balls may present "finger print" for positive identification of the origin of THC and heavy metals in marine and fluvial water (NRC, 1985).

#### **CONCLUSION**

From the foregoing it could be concluded that the waters and sediments of the Cross River Coast of Nigeria are highly polluted with THC, with levels in water higher than the recommended limit by the DPR, and very high in sediments. The heavy metal levels, though high in sediments, are generally below the permissible levels in waters.

To enhance pollution abatement in this fragile ecosystem, it is recommended that anthropogenic activities, especially those of the petroleum industry which is perceived as the major source of pollutants here should be controlled. This could be achieved through stringent legislation and regulations (such as emission standards) and effective monitoring of the area for improved environmental quality control. The discharge of untreated domestic and municipal effluent up-stream of the estuaries should also be controlled. These and other measures are necessary to curb the excess environmental degradation of this coastal and estuarine environment that serve one of the major sources of seafood along the coast of West Africa.

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***Distribution of heavy metal***

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FIG. 1: MAP OF CROSS RIVER SYSTEM SHOWING SAMPLING LOCATIONS

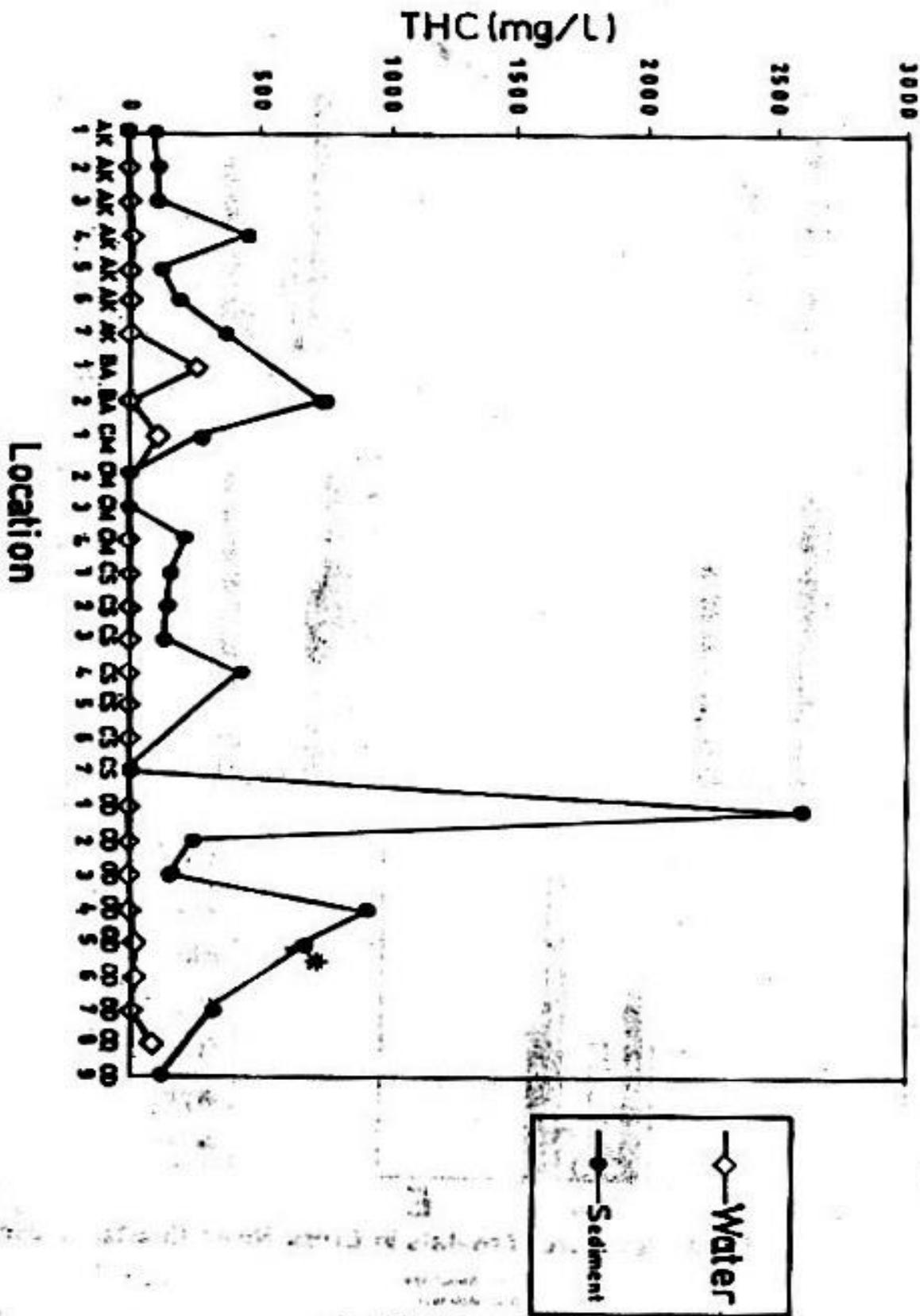
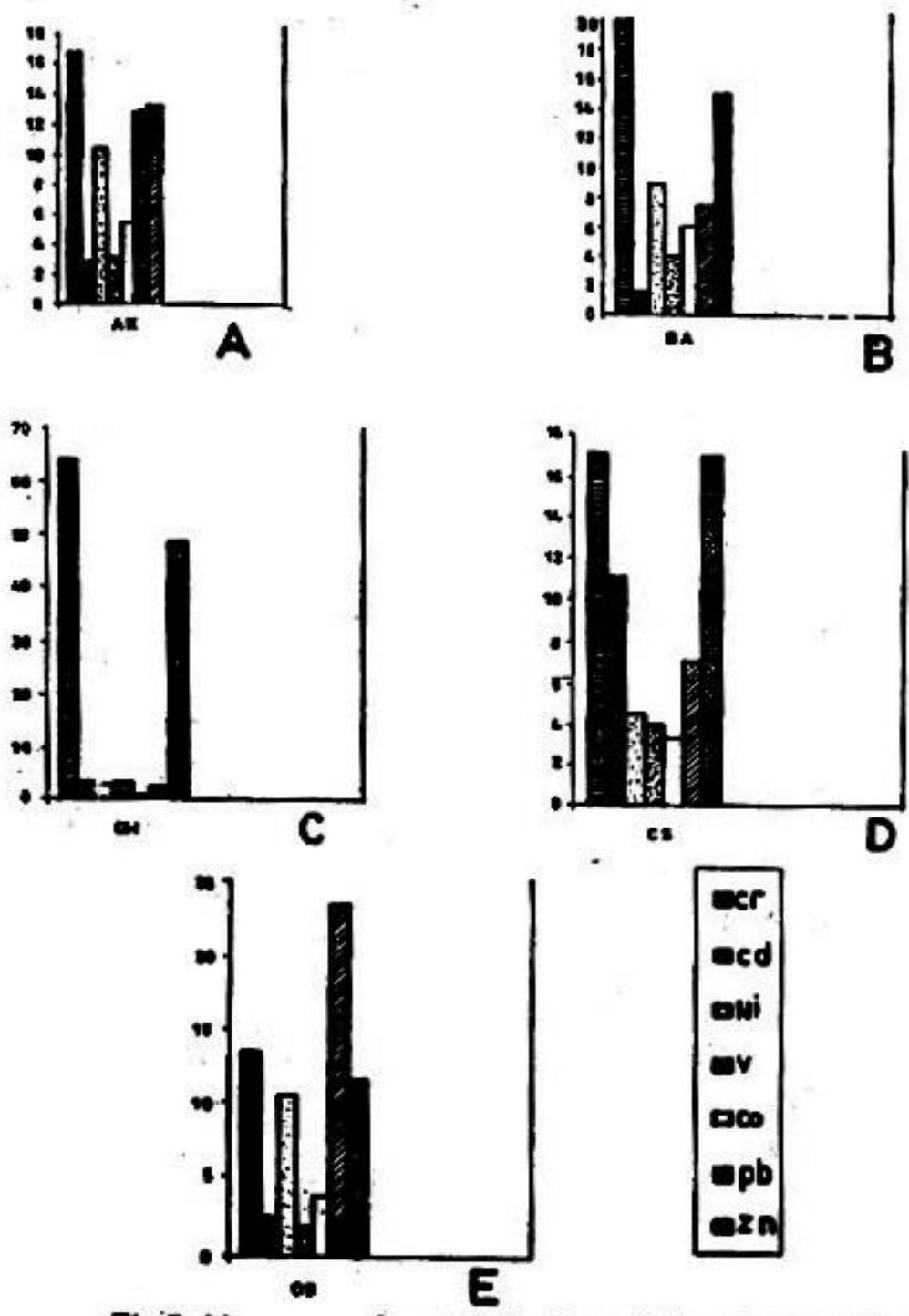


Fig 2 Spatial variation of THC in water and sediment in Cross River Coastal Waters

(Scale: OD 5 = Value = 10)



**Fig 3 Mean conc:of metals in Cross River Coastal Sediments**

- ▲ - Akpa
- - Bakassi
- ▨ - Calabar Municipality
- ▩ - Calabar State
- - Calabar