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## RESEARCH ARTICLE

**Heavy metal distribution in core sediments of lower Gadilam River, Cuddalore district, Tamil Nadu, India.**

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Due to problems caused by their toxicity, persistence in the environment and bioaccumulation, heavy metals are among serious pollutants. This study was about the vertical distribution of heavy metals in sediments of lower Gadilam River in Cuddalore district, Tamil Nadu-India. Along the river, core sediment samples have been collected from three locations: River mouth, Estuary and Fresh water area; and following heavy metals have been analysed by using AAS (Atomic Absorption Spectroscopy) method: Fe, Mn, Cr, Cu, Ni, Co, Pb, Cd and Zn. Except Pb, Ni, Cr and Cd with concentration less than 5mg/Kg, all other remaining metals have higher concentration values. The relative decreasing order of all analysed metals, in terms of their average concentrations, is the following:  
Fe>Zn>Mn>Cu>Co>Pb>Ni>Cr>Cd.

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**Introduction:-**

There is natural occurrence of heavy metals in the earth's crust and their environmental contents may also vary within different regions conducting to the spatial variations of background concentrations. The properties of the metal and some environmental factors' influences are the ones which mostly govern the distribution of metals in environment. Heavy metals can enter the environment either by natural process and anthropogenic mean from different sources like: natural weathering of earth's crust, soil erosion, urban runoff, mining, sewage effluents, industrial discharge, air pollution fallout, pesticides, disease control agents used in agriculture (Simone Morais et al., 2012) etc....

Many metals associate with particulates and hence adsorbed or co-precipitated with sulphide, oxyhydroxides, carbonates and clay minerals. Therefore, sediments agglomerate contaminants and may store metals for long time in the environment. Sediment residing organisms may be exposed to metals via ingestion of sediment particles, uptake of interstitial waters and via the food chain (Spencer and MacLeod, 2002). Due to problems caused by their toxicity, persistence in the environment and bioaccumulation, heavy metals are among serious pollutants. All over the world, heavy metal contamination has really become a serious issue especially in marine ecosystems and this is mainly due to the discharge of heavy metals into the aquatic systems as result of different developments such as mines and rapid industrial developments, ship maintenance, agricultural and urban development over the past century (Feng et al., 2012).

The interest of this study is that it scientifically shows qualitative and quantitative results of different heavy metal pollutants that have vertically accumulated in core sediments along Gadilamriver and hence this is a step to the prevention of people from being contaminated by the identified pollutants.

In additional, by being aware of different metal pollutants present in sediments along Gadilam river and their suspected anthropogenic sources, concerned people could take measures regarding the treatment of wastes before their discharge to environment. Besides that, to understand the source, distribution and levels of heavy metals in

sediments is helpful for environmental management and ease the controlling of associated water quality which is mostly based on the sediment quality assessment by sediment quality guides (SQGs).

### Study area:-

The present study area (Gadilam river) lies between  $79^{\circ}40'$  and  $79^{\circ}50'$  East longitude; and between  $11^{\circ}41'$  and  $11^{\circ}47'$  North latitude. It lies in SOI toposheet No. 58M/2 of survey of India (Fig.1). This study area covers an area about  $181.315\text{Km}^2$ , the Gadilam River originates near Sankarapuram and passes through the Cuddalore and Villupuram districts towards Bay of Bengal at Cuddalore. The Gadilamriver flows through the town of Cuddalore and separates the Old Town from Thirupadiripuliyur. During the monsoon season, this river is generally flooded, raises the water table and feed tanks on its basin. The district geology setting ranges in age from the oldest Archaean rocks to recent sediments. The study area is enclosed by Quaternary formations consisting of marine, sandstone with clay, fluvial flood plains and fluvio-marine (Fig.2). Generally, the Quaternary landforms of East coast of India represent the features of emergence characteristics, while that of west coast are mostly dominated by features of submergence characteristics. In this study, three locations have been selected and core samples have been collected from such locations as shown on the Fig.1.

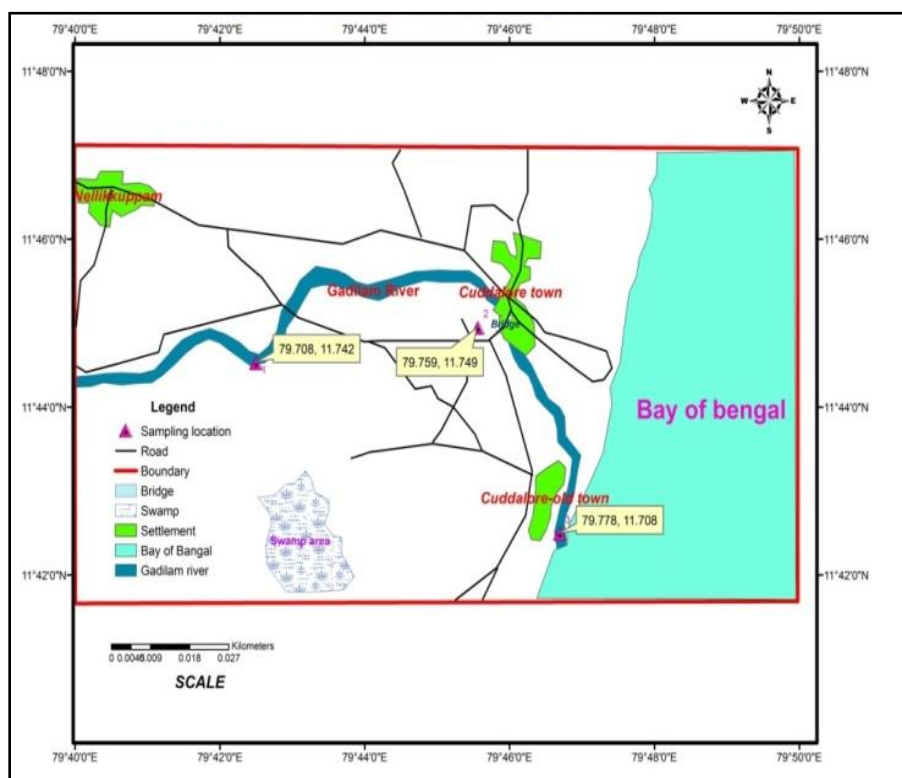
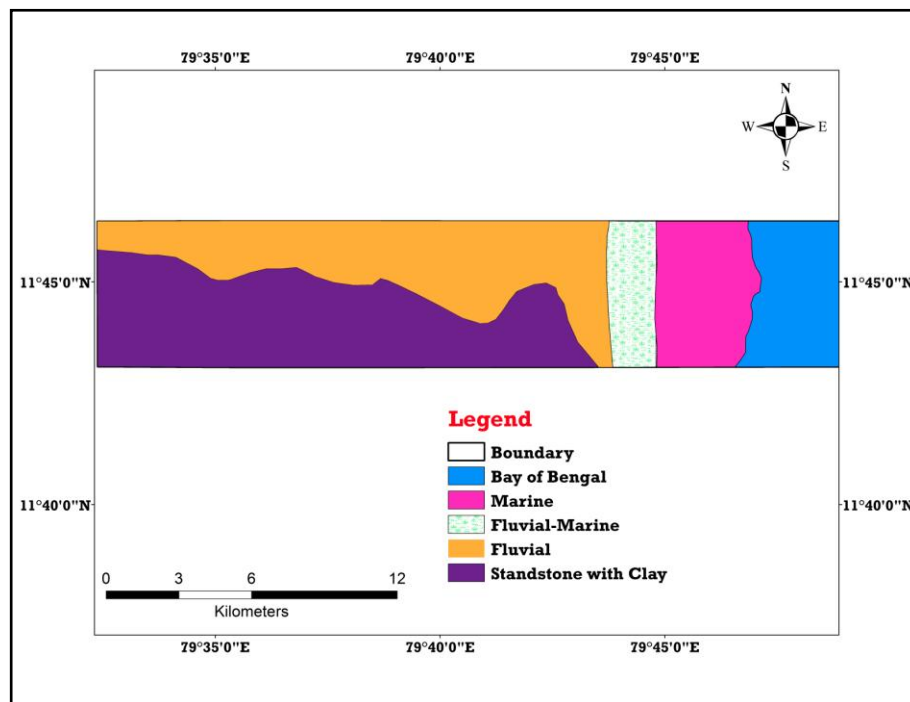


Fig.1 Study area and sampling location map



**Fig.2 Geology map of study area**

### Methodology:-

The specific objective of this study is the assessment of metals Fe, Mn, Cr, Cu, Ni, Co, Pb, Cd and Zn in core sediments of lower Gadilamriver, Cuddalore district. Flame AAS was used for estimating Cu, Fe, Zn, Mn, Co, Ni and Cr while Graphite Furnace (GF) AAS was employed for analysing Cd and Pb. Core sediments were collected from three different locations using PVC pipes of 1m. In laboratory each location core sample has been subdivided into sub-samples of same interval from top to bottom and nearly thirteen core sediment samples have been prepared and analysed (Table 1). The particular samples were dried well at 60°C. About 1gm of the sample was digested for a period of 30 minutes in a beaker with 6N HCl. The digested samples were filtered through G3 crucible and made up to 100ml with double distilled water in a standard flask. The digested filtered samples were fed in AAS (Atomic Absorption Spectrometer).

**Table 1: Location and sample codes**

S. No.	Location	Sample Id.	Core length in (cm)
1.	River mouth (Location – I)	C <sub>1</sub> S <sub>1</sub> (T)	0-20cm
2.		C <sub>1</sub> S <sub>2</sub>	20-40cm
3.		C <sub>1</sub> S <sub>3</sub>	40-60cm
4.		C <sub>1</sub> S <sub>4</sub>	60-80cm
5.		C <sub>1</sub> S <sub>5</sub> (B)	80-100cm(bottom)
6.	Estuary (Location –II)	C <sub>2</sub> S <sub>1</sub> (T):”	0-20cm
7.		C <sub>2</sub> S <sub>2</sub>	20 to 40cm
8.		C <sub>2</sub> S <sub>3</sub>	40 to 60cm
9.		C <sub>2</sub> S <sub>4</sub> (B)	60 to 80cm(bottom)
10.	Fresh Water (Thiruvandhipuram) (Location-III)	C <sub>3</sub> S <sub>1</sub> (T)”	0-20cm
11.		C <sub>3</sub> S <sub>2</sub>	20 to 40cm
12.		C <sub>3</sub> S <sub>3</sub>	40 to 60cm
13.		C <sub>3</sub> S <sub>4</sub> (B)	60 to 80cm(bottom)

Metal ions were analysed by AAS method using a series of solution over the range 2-10mg/l. The absorbance was measured at respective wavelength of metal ions by using the corresponding Hollow Cathode Lamps. Average value of 10 replicates was taken for each determination. Calibration graphs were prepared by diluting the stock solution of

respective metal ions of 100mg/l. The concentration in the sample aliquot was established by referring to the calibration graph.

### Results and discussion:-

Sediments are components of the aquatic ecosystems which are ecologically sensitive and they are also reservoir of the contaminants taking a considerable part in maintaining the trophic status for any water reservoir (Singh et al., 2005). With respect to physicochemical conditions, sediments can be both the source and the sink for heavy metals as well as for nutrients. Thus, in addition to being considered as contaminant carriers, sediments can also potentially be secondary sources of contaminants in aquatic ecosystems. So, the heavy metal pollution in any aquatic area can be studied through analysis of associated sediments.

Analysed core sediment samples and obtained results are shown on Table 1 and Table 2.

**Table 2: Heavy metal results**

Sample code ↓		Concentration of elements (in core sediments) in mg/Kg								
		Cu	Fe	Zn	Pb	Cd	Mn	Co	Ni	Cr
C <sub>1</sub> S <sub>1</sub> (T)	River mouth	14.11	573.12	38.60	3.06	1.70	32.09	14.49	3.06	3.04
C <sub>1</sub> S <sub>2</sub>		12.48	575.48	40.16	3.61	1.61	33.43	11.38	3.59	3.63
C <sub>1</sub> S <sub>3</sub>		10.93	579.35	43.61	4.01	1.42	35.43	12.27	4.02	4.03
C <sub>1</sub> S <sub>4</sub>		9.27	562.21	45.32	4.08	1.02	32.18	11.18	4.06	4.07
C <sub>1</sub> S <sub>5</sub> (B)		8.42	549.51	40.92	3.15	0.99	30.05	13.80	3.16	3.17
C <sub>2</sub> S <sub>1</sub> (T)	Estuary	11.37	619.11	38.19	4.15	1.01	29.60	10.86	4.13	4.14
C <sub>2</sub> S <sub>2</sub>		12.85	651.38	43.28	4.10	1.20	18.29	10.66	4.11	4.09
C <sub>2</sub> S <sub>3</sub>		13.95	491.96	50.88	3.09	1.67	13.28	10.50	3.07	3.06
C <sub>2</sub> S <sub>4</sub> (B)		14.18	398.11	52.20	3.04	1.89	11.96	9.80	3.05	3.02
C <sub>3</sub> S <sub>1</sub> (T)	Fresh water	7.91	628.00	50.89	4.09	0.78	49.13	15.83	4.06	4.08
C <sub>3</sub> S <sub>2</sub>		15.48	598.12	45.42	4.06	1.41	36.46	10.14	4.07	4.04
C <sub>3</sub> S <sub>3</sub>		49.08	508.14	37.29	3.03	1.50	30.02	12.09	3.04	3.01
C <sub>3</sub> S <sub>4</sub> (B)		51.13	530.11	35.48	3.01	1.93	28.14	10.86	2.99	2.08
<b>Minimum</b>		7.91	398.11	35.48	3.01	0.78	11.96	9.80	2.99	2.08
<b>Maximum</b>		51.13	651.38	52.20	4.15	1.93	49.13	14.49	4.13	4.14
<b>Average</b>		17.78	558.82	43.25	3.58	1.39	29.24	11.84	3.57	3.50

### Copper:-

At the river mouth, copper has been found to be decreasingly distributed as the depth increases. It varies from 8.42mg/Kg (bottom) to 14.11mg/Kg (top). This means that the accumulation rate of copper is nowadays higher than days ago. In both location II and III, copper has been found to be increasingly distributed as the depth increases. It increases from 11.37mg/Kg (top) to 14.11 mg/Kg (bottom) for location II, and from 7.91mg/kg (top) to 51.13mg/Kg (bottom) for location III. For both these locations, there is higher accumulation of copper to greater depth because of water infiltrating the sediments downward along with accumulated copper.

This higher accumulation rate may mainly be due to increased use of fertilizer in Cuddalore district. In fact, cereal crops grown on Cu-deficient soils are occasionally treated with Cu as an addition in form of fertilizer to the soil. Other sources are copper containing compounds used in electroplating industries such as cupric acetate and cupric sulphate and paint industries as cuprous oxide, ceramics and glass industries as cupric acetate, cupric and cuprous oxides used as pigments and for making glazes which may have been discharged. The variation in concentration of copper is also due to high alkalinity of water, when most of the copper is precipitated.

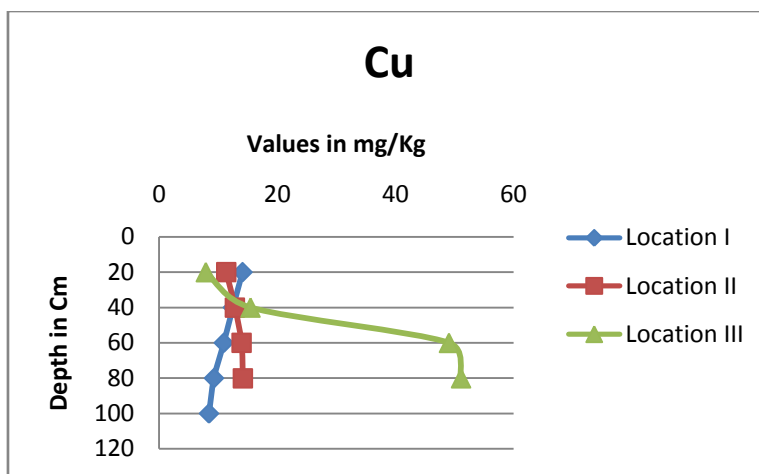


Figure 3: Vertical distribution of Cu

#### Iron:-

The accumulation of iron is generally decreasing from top to bottom for all three locations. In location I, it is decreasing from 573.12mg/Kg (top) to 549.51mg/Kg (bottom); for location II it is decreasing from 619.11mg/Kg (top) to 398.11mg/Kg (bottom), whereas for location III it is decreasing from 628mg/Kg (top) to 530.11mg/Kg (bottom). These high values of iron accumulation in sediments may be due to its high abundance within the earth crust. Most often, Iron attains significantly higher concentrations in sediments and water than other trace metals because it is naturally very prevalent in all fresh water environments (Silambarasan et al., 2012).

Iron has different roles such as biological as well as ecological ones but high concentration in fresh waters and in sediments has long been considered a problem. At a given extent, Iron plays an important role as a constituent of enzymes like catalase and cytochromes, as a constituent of oxygen carrying proteins like myoglobin and haemoglobin. In addition, iron is an essential nutrient for algae and other aquatic organisms in sediments and fresh water. Though the iron has such mentioned roles, its high concentration in sediments causes pollution and highly polluted sediments will also in turn induce water pollution. In the study area, the higher iron accumulation may be from some anthropogenic source of urban runoff, agricultural and municipal wastage (Dhanakumar et al., 2013). However, contamination of anthropogenic origin cannot be excluded, since the Fe content is not only attributed to weathering of soil and rocks but also to various activities of mining and use of high fertilizers for agricultural products (Shrivastava et al., 2011). The mining of iron rich ores has been the degradation cause of many river ecosystems (Vuori et al., 1995). Excess of iron also influences the presence of bacteria (iron reducing) in fresh water.

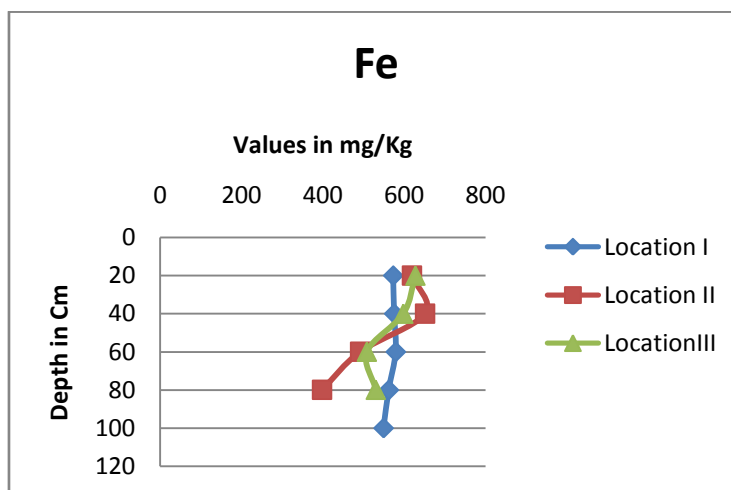
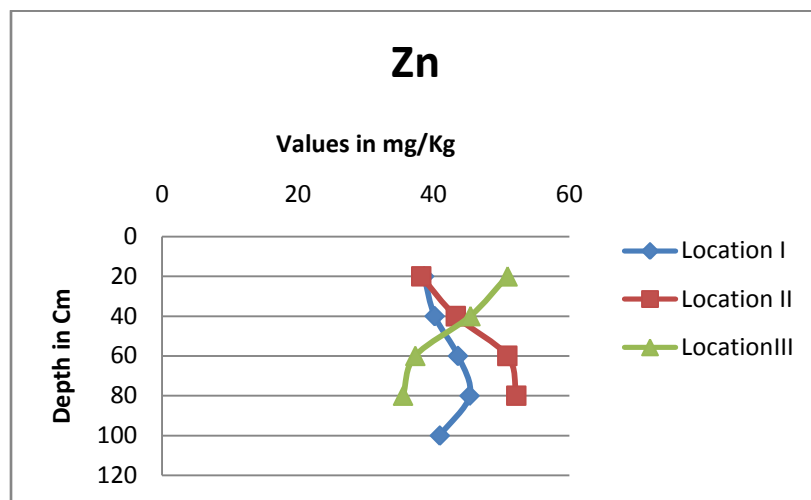


Figure 4: Vertical distribution of Fe

**Zinc:-**

Generally, there is accumulation of Zinc increases with depth for the location I and II even though there is a slight decrease at the depth 80cm to the bottom of location I. It increases from 38.60mg/Kg (top) to 40.92mg/Kg (bottom) for location I and from 38.19mg/Kg (top) to 52.20mg/Kg (bottom) for location II. But it decreases with depth for location III, where it varies from 50.89mg/Kg (top) to 35.48mg/Kg (bottom). This environmental pollution due to Zinc, along Gadilam River, may be due to municipal refuse, automobiles and agricultural use of pesticides and fungicides containing  $ZnSO_4$ . This metal can be naturally introduced into water by erosion of minerals from rocks and soil; however zinc ores are slightly soluble in water.

Zinc is normally found in nature, but at a limited concentration, it is a very important micronutrient in humans and only at very high concentration it may cause harmful effects. The zinc absorption in the study area may be emitted from different sources through discharges of many commercial industries during smelting (metal processing) and mining activities, urban runoff, and municipal sewages (Shah et al., 2005, Shrivastava et al., 2011).



**Figure 5: Vertical distribution of Zn**

**Lead:-**

The vertical distribution of lead in all analysed sediment samples shows the accumulation of lead to be less than 5mg/Kg. In location I, lead values increase at shallow depth and then decreases before reaching the bottom, whereas in both location II and III the values are in decreasing order from top to bottom. The constant changes in values differ from 3.06mg/Kg (top) to 3.15mg/Kg (bottom) for location I, then from 4.15mg/Kg (top) to 3.04mg/Kg (bottom) for location II and from 4.09mg/Kg (top) to 3.01mg/Kg (bottom) for location III.

Lead is among the most contaminant, when lead is present in significant quantity in environment, it constitutes a source of pollution and causes threat to environment, human, animal and aquatic lives. Lead accumulated in sediments may also be carried to interacting water and then bio-accumulated to aquatic organisms including fish and finally enters the food chain. Lead as  $Pb^{2+}$  ion has a large affinity for the thio (-SH) and phosphate ion ( $PO_4^{3-}$ ) containing enzymes, ligands and biomolecules, thereby, inhibiting the biosynthesis of home units, affecting membrane permeability of liver, kidney and brain cells (Okoye et al., 2010).

Even though the lead accumulation is very less in the environment, it may arrive to surroundings from old lead piping in the water distribution system, lead jointed waste pipes and PVC pipes (as an impurity), old paint pigments (as oxides, carbonates) and from fuel wastes which might have transported with wastewater and water run-off and then accumulated within analysed sediments. The overall concentration of Lead in the study area is higher than other Indian River concentration.

The profile of the Pb indicates that it did not have only one source; furthermore higher concentration of Pb in the study area is by the contamination through various industrial effluents and local anthropogenic inputs.

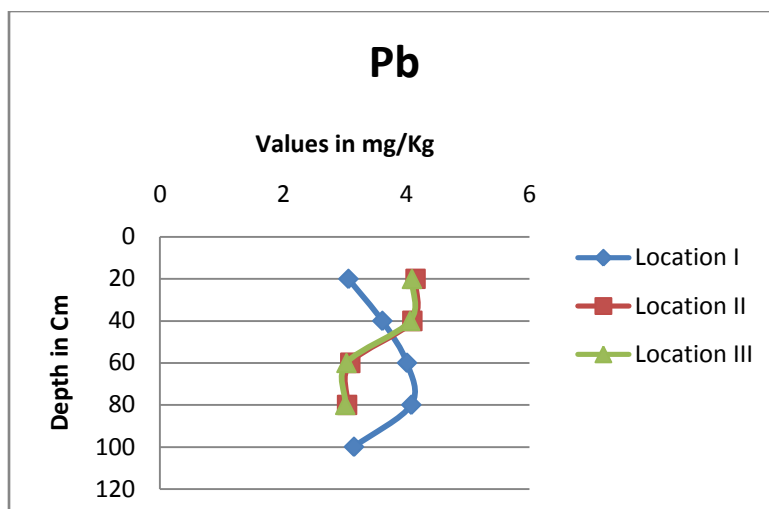


Figure 6: Vertical distribution of Pb

#### Cadmium:-

Cadmium is a crystalline non-essential metal and its static state in the natural environment is Cd. It is among the most dangerous pollutants because of its high-potential toxic effects. Cadmium is very toxic and the primary use of water with high Cd content could cause cancer and renal disease to consumers (Satheeshkumar et al., 2011). The high concentration of cadmium may be due to input of sewage discharge of plastic pipes or industrial metal constituting a possible source of Cd in water. Like other metals in solution, cadmium has tendency of being absorbed by suspended particles and bottom sediments. Due to this reason, cadmium levels may be below the detectable limit in the water phase even in polluted rivers. In the analysed samples the accumulation of Cadmium decreases with increase of depth for location I; where it varies from 1.70mg/Kg (top) to 0.99mg/Kg (bottom); whereas its accumulation increases with increase of depth for both location II and III and it varies from 1.01 mg/Kg (top) to 1.89mg/Kg (bottom) for location II and from 0.78 mg/Kg (top) to 1.93mg/Kg (bottom) for location III. This accumulation of Cd may be due to input of sewage of discharge from Ni-Cd batteries, PVC stabilizers and pigments used in plastic (main use), glass, paint industries and ceramics in Cuddalore district. The higher fluctuation of metals in analysed samples is attributed to some reason of the other anthropogenic and industrial sources.

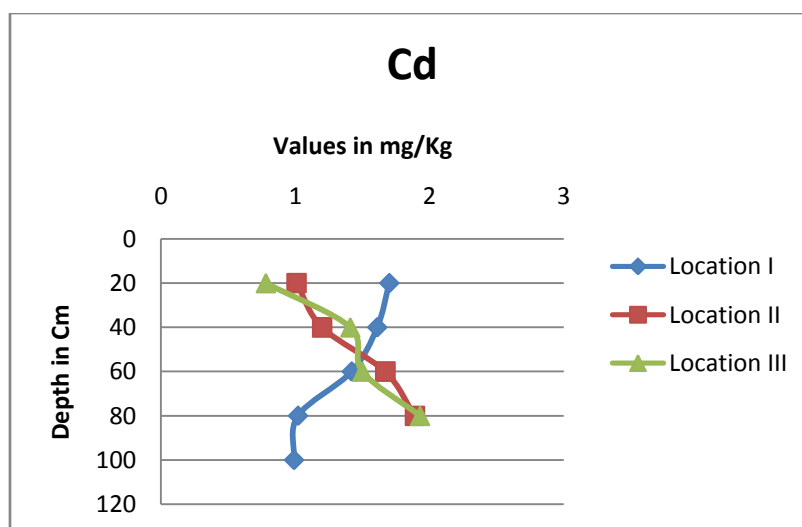
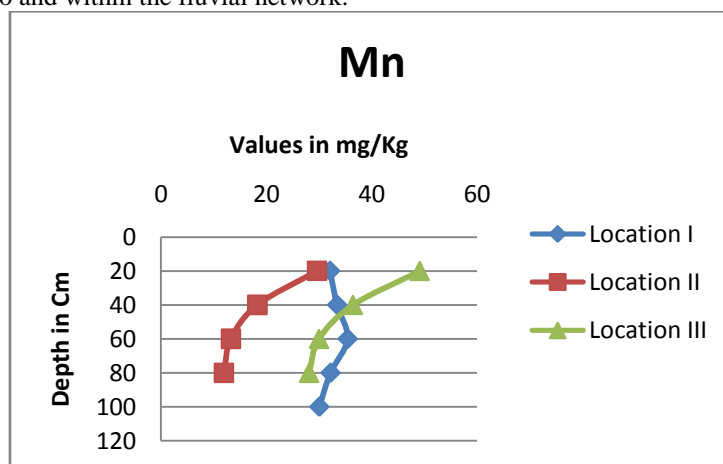


Figure 7: Vertical distribution of Cd

**Manganese:-**

Manganese occurs essentially as a soluble ion and the percentage in the residual fraction is low. In general the contents are relatively higher and manganese does not present a danger to human health, nor for the environment. Accumulation of manganese varies from 32.09mg/Kg (top) to 30.05 mg/Kg (bottom) for location I , then from 29.60mg/Kg (top) to 11.96mg/Kg (bottom) for location II and from 49.13 mg/Kg (top) to 28.14 mg/Kg (bottom) for location III. It is important to take into account the significant amount of metal influence from stream flow regime in this kind of geomorphological hydro graphic condition. During each rainy season, finer sediments are moved downstream in the drainage network, representing a major contribution of metals in the main stream river. The results show that the transport of pollutants and their dispersion within fluvial environments are led by different interrelated factors which are difficult to be controlled over time and need information about the processes which govern the transport into and within the fluvial network.



**Figure 8: Vertical distribution of Mn**

**Cobalt:-**

Cobalt is present in sediments, since it is rapidly taken from solution by sorption and co-precipitation in most oxidising, near-neutral or alkaline stream water as the dissolved Fe and Mn are precipitated as secondary oxides where hydrous Mn oxides have a particularly strong sorption affinity for cobalt. There are different anthropogenic sources of cobalt like fertilizers, special steels, coal combustion, and iron, silver and lead mining and processing (Reimann and de Caritat, 1998), although, apart from these few cases, anthropogenic anomalies in drainage are rarely recorded. Environmental pollution problems from Cobalt are generally less significant than those associated with some other heavy metals (Cole and Carson, 1981).

Regarding the analysed samples, obtained results indicate that for all three locations, cobalt concentration decreases with increasing the depth. It decreases from 14.49 mg/Kg (top) to 13.80 mg/Kg (bottom) for location I, from 10.86 mg/Kg (top) to 9.80 mg/Kg (bottom) for location II, and from 15.83 mg/Kg (top) to 10.86 mg/Kg (bottom) for location III. In an estuary, cobalt will be more accumulated in the more oxidized layer of sediments and drift landward (Andree et al., 1986). Therefore, this may be common to all sediments, the reason for the variation in concentration of these analysed samples, where Co generally decreases from oxidized layer (top layer) to reduced layer (bottom layer).



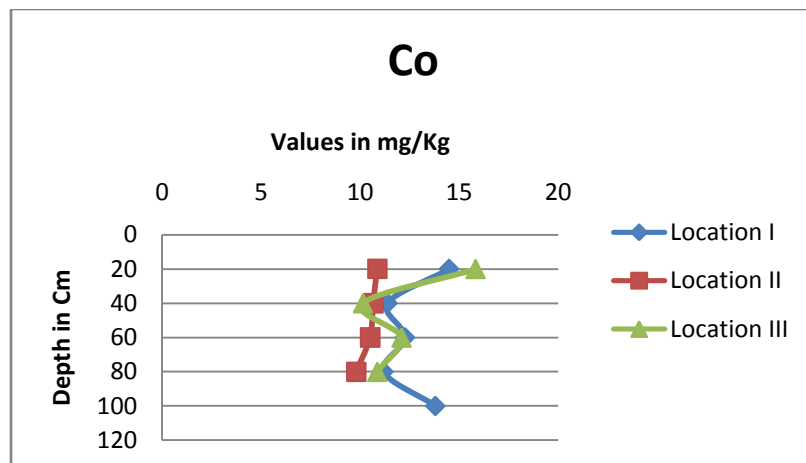


Figure 9: Vertical distribution of Co

#### Nickel:-

Nickel is a prevalent trace metal and occurs in water, soil, air as well as in the biosphere. In the Earth's crust, the average percentage of Ni is about 0.008%. Nickel can enter the aquatic environment through different natural processes like surface run-off, natural erosion of soils and rocks as well as through anthropogenic sources by discharge of municipal and industrial waste for instance. In rivers, nickel is mostly moved as a precipitate which coats on particles and associated with some organic matters.

Main sources of nickel into the ambient air may be attributed to different processes like the incineration of sewage and waste sludge, burning of oil and coal for power or heat generation, mining and primary production of nickel, electroplating, steel manufacturing and some miscellaneous sources such as manufacturing of cement. Finally, nickel emitted from those different industrial processes as well as other sources reaches waste water and sediments too.

In this study area the following representation is made as in all three locations, obtained values of Ni concentration are less than 5mg/kg. In brief, it varies from 3.06 mg/kg (top) to 3.16 mg/kg (bottom) for location I, from 4.13 mg/kg (top) to 3.05 mg/kg (bottom) for location II, and from 4.06 mg/kg (top) to 2.99 mg/kg (bottom) for location III. The presence of nickel in this area of study may be caused by its removal from the atmosphere, surface run-off, discharge of municipal sewage & waste and also flowing natural erosion of both soils and rocks.

Nickel is a moderately toxic element, and the volatile compound like nickel tetra carbonyl  $\text{Ni}(\text{CO})_4$ , which is used for the extraction of the element, is poisonous. Ni is not generally recognized to cause any health issues on human being, but long term exposure was found to be the cause of skin irritation, heart, liver damage and decreased body weight (Tiwana, 2005).

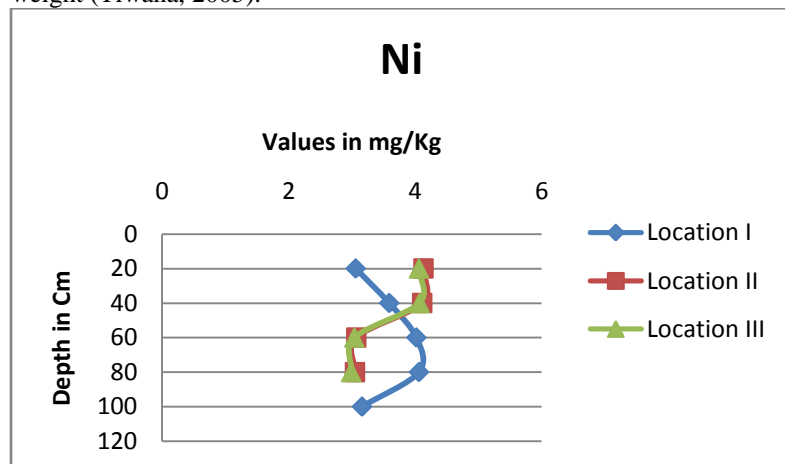


Figure 10: Vertical distribution of Ni

### Chromium:-

Cr is an essential trace element that can be toxic to aquatic biota at elevated concentrations and it can also be bio-accumulated and then enters the food chain. Aerial deposition and surface runoff may be main processes by which chromium can enter aquatic systems and its subsequent association with some particulate matter leads to its deposition in bed sediments. Therefore, sediments are main route of exposure to Cr for aquatic organisms since lots of varieties of organisms live in contact with bed sediments. Chromium VI is more toxic compared to chromium III though the long term exposure to chromium III can cause allergic skin reaction and cancer (Agency for Toxic substances and Diseases Registry, 2000). In general, it has been found that at higher temperatures Cr is more toxic and its compounds also cause cancer in humans (Lokhande, 2011).

In the analyzed samples, the concentration of chromium has been found to be less than 5mg/Kg. It varies from 3.04mg/Kg (top) to 3.17mg/Kg (bottom) for location I, from 4.14mg/Kg (top) to 3.02mg/Kg (bottom) for location II, and from 4.08mg/Kg (top) to 2.08mg/Kg (bottom) for location III. The presence of chromium in this area may be originated from cleaning products, washing powders, paints and pigments used by different people in Cuddalore district.

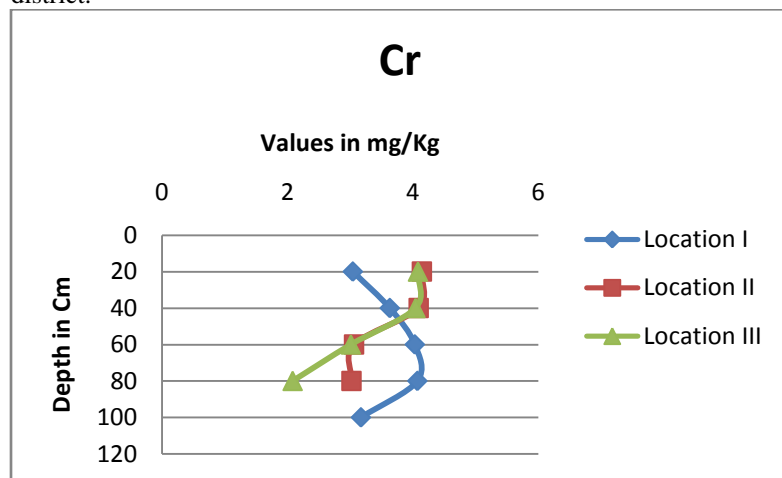


Figure 11: Vertical distribution of Cr

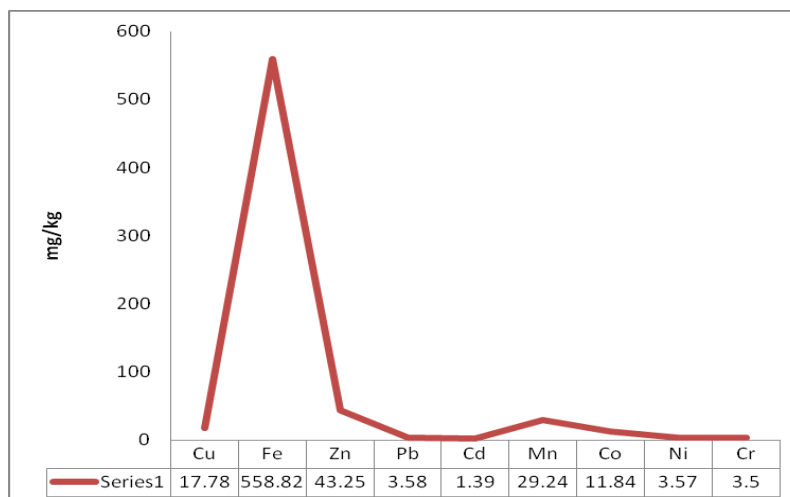
### Comparison of average concentrations:-

General average concentrations of all analyzed heavy metals are given as follow:

Table 3: Average values of distributed heavy metals

S. No.	Element	Avg. Conc. (in mg/Kg)
1.	Cu	17.78
2.	Fe	558.82
3.	Zn	43.25
4.	Pb	3.58
5.	Cd	1.39
6.	Mn	29.24
7.	Co	11.84
8.	Ni	3.57
9.	Cr	3.5

As shown on the figure below as well as by the table 3; the relative decreasing order of all analyzed metals, in terms of their average concentrations, is the following: **Fe>Zn>Mn>Cu>Co>Pb>Ni>Cr>Cd**. This observed highest value of iron accumulation in sediments may be attributed to its usual high content within the earth crust. Iron is prevalent in all environments of fresh water and most often reaches distinguishably higher concentrations in water and sediments than other trace metals.



**Figure 12: Comparison of general average concentrations of all analyzed heavy metals**

### Conclusion:-

The aim of this study was the assessment of “Fe, Mn, Cr, Cu, Ni, Co, Pb, Cd and Zn in core sediments of lower Gadilam River, Cuddalore district”. Regarding their vertical distribution in chosen three sampling locations (River mouth, Estuary and Fresh water), some metals like copper are of higher concentration at greater depth due to water infiltration and some others like Cobalt are of higher concentration at top layer because of their preference of oxidised layers. The relative decreasing order of all analyzed metals, in terms of their average concentrations, is the following: Fe>Zn>Mn>Cu>Co>Pb>Ni>Cr>Cd. Different sources of accumulated heavy metals along the Gadilam river may mainly be by use of fertilizer containing some metals like copper, natural high abundance of some other elements like Iron in the earth crust, Municipal refuse, automobiles, agricultural use of pesticides and fungicides containing ZnSO<sub>4</sub>, discharge from Ni-Cd batteries, PVC stabilizers and pigments used in plastics, ceramics, glass and paints used by different people in Cuddalore district.

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