



**UNIVERSITI PUTRA MALAYSIA**

**DISTRIBUTION OF SOME HEAVY METALS IN KELANG RIVER  
ESTUARY**

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DISTRIBUTION OF SOME HEAVY METALS IN KELANG RIVER  
ESTUARY

by

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DISTRIBUTION OF SOME HEAVY METALS IN KELANG RIVER ESTUARY

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## ABSTRACT

The distribution of copper, lead, mercury and zinc in the Kelang River estuary were studied. Six stations were selected for sampling. Two of these stations were in the freshwater regime while the rest were in the estuarine zone. The dissolved and total copper, lead and zinc concentrations in the water column were determined by flame atomic absorption spectrophotometer after chelation and extraction with solvent. The total manganese and iron concentration in water as well as the extractable copper, lead and zinc levels in the sediment were studied. The total mercury content in the water column, sediment and fishes was determined by cold-vapour atomic absorption spectrophotometric method. The heavy metal contents in six species of fishes, namely Plotosus anguillaris, Arius thalassinus, Dasyatis zugei, Gastrophysus lunaris, Setipinna taty and Johnius carutta were studied. Some physico-chemical parameters of the water column such as salinity, dissolved oxygen content, water velocity and pH were determined at different depths and stations.

In the estuarine water, the overall dissolved copper, lead and zinc means and ranges were  $5.7 \mu\text{g l}^{-1}$  (range: 0.5 to  $32.0 \mu\text{g l}^{-1}$ ),  $2.7 \mu\text{g l}^{-1}$  (range: 0.3 to 11.8



$\mu\text{g l}^{-1}$ ) and  $16.9 \mu\text{g l}^{-1}$  (range: 0.9 to  $175.0 \mu\text{g l}^{-1}$ ) respectively, while for the overall total copper, lead, mercury and zinc, the values were  $20.8 \mu\text{g l}^{-1}$  (range: 1.4 to  $333.5 \mu\text{g l}^{-1}$ ),  $9.4 \mu\text{g l}^{-1}$  (range: 0.5 to  $52.0 \mu\text{g l}^{-1}$ ),  $1.5 \mu\text{g l}^{-1}$  (range: 0.0 to  $8.5 \mu\text{g l}^{-1}$ ) and  $51.9 \mu\text{g l}^{-1}$  (range: 2.4 to  $420 \mu\text{g l}^{-1}$ ) respectively.

For the estuarine sediment, the overall mercury mean and range was  $0.32 \mu\text{g g}^{-1}$  (range: 0.08 to  $1.30 \mu\text{g g}^{-1}$ ). The overall extractable copper, lead, and zinc means and ranges were  $1.88 \mu\text{g l}^{-1}$  (range: 0.01 to  $5.70 \mu\text{g g}^{-1}$ ),  $0.52 \mu\text{g g}^{-1}$  (range: 0.01 to  $1.75 \mu\text{g g}^{-1}$ ) and  $6.10 \mu\text{g g}^{-1}$  (range: 0.35 to  $15.50 \mu\text{g g}^{-1}$ ) respectively.

The copper, lead, mercury and zinc mean ranges in the fish were 0.19 to  $0.71 \text{mg Kg}^{-1}$ , 0.08 to  $0.35 \text{mg Kg}^{-1}$ , 0.08 to  $0.39 \text{mg Kg}^{-1}$  and 4.83 to  $13.80 \text{mg Kg}^{-1}$  respectively.

The results clearly show there was mercury pollution in water and sediment, and the mercury content in the fish was much higher than that reported from other Malaysian coastal waters. The dissolved copper, lead and zinc concentrations were comparable to that of unpolluted estuaries but the concentrations of copper, lead and zinc in the water were much higher. The copper, lead and zinc contents in fish were below the maximum permissible levels allowed in food. Distinct relationships between the heavy metal contents and weights of fish were observed.



## I INTRODUCTION

Estuaries are important features of coastal ecosystem and act as transitional zones between fresh and saline waters. Pritchard (1967) defined estuary as " a semi-enclosed coastal body of water which has a free connection with the open sea and within which sea water is measurably diluted with fresh water derived from land drainage".

The hydrodynamic characteristics of an estuary are usually complex and unpredictable. This is due to the oscillation of salinity, hydrographical and sedimentological characteristics prevalent in this zone. The nature and volume of the terrestrial and riverine water input into the estuary can vary daily.

Estuaries are usually bordered by marsh wetlands or mangrove swamps which are abundant in nutrients. Being a rich source of nourishment, the estuaries are usually very productive and are rich fishery nursery grounds.

The estuaries being sheltered, usually form fine ports, and has helped to attract trade and industry to such areas over a period of time. These industries often discharge pollutants such as heavy metals into the riverine system. The estuaries act as receptacles for such pollutants and may affect the highly vulnerable aquatic resources. The pollutants may also pose hazards to human health if contaminated fish or shellfish, inhabiting such region are consumed. This was evident as shown by the outbreak of man-made epidemic Minamatta Bay disease in Japan, which was caused by consumption of mercury-contaminated fish and shellfish.

This incident brought about world-wide public concern and anxiety not only for mercury but also for



other non-degradable and persistent heavy metals which may be accumulated in the food chains and ultimately consumed by human beings.

In Malaysia, although some studies have been done on the distribution of heavy metals in Malaysian rivers such as Kelang River, no study has been conducted on the distribution and behaviour of heavy metals in Malaysian estuaries.

Chan et al. (1978) had studied the level of eleven heavy metals in Kelang River and reported elevated levels of arsenic and mercury at certain parts of the river. The mercury content in fish in Kelang River estuary was studied by Chia and Tong (1981).

There is a need to study the distribution of heavy metals in local estuaries especially the Kelang River estuary where the Kelang River is polluted. This study was therefore initiated: (i) to examine the heavy metals (copper, lead, mercury, zinc, iron, and manganese) concentrations in dissolved and particulate matter in the water column and "extractable" heavy metals in sediment; (ii) to ascertain whether the fish inhabiting this region is contaminated with heavy metals; and (iii) to compare the levels of heavy metals in water, sediment and fish with the values that have been reported in literature by various workers from different parts of the world.



## II LITERATURE REVIEW

### A) General

A heavy metal is usually considered to be an element having a density greater than  $5 \text{ gcm}^{-3}$  and an atomic number of more than 20 (Forstner and Wittmann, 1979). The heavy metals can be divided into two categories, namely essential and non-essential elements. Essential elements are elements that are abundant in biota, with no or minor variation within species. On their removal or depletion, there is appearance of reversal deficiency symptoms which are induced by distinct biochemical defects.

According to Schroeder and Darrow (1973), copper and zinc are essential elements while lead and mercury are non-essential and toxic elements. Copper catalyses various oxidation-reduction reactions while zinc is involved in hydration-dehydration and hydrolysis reactions (Wood, 1975).

The non-essential toxic elements can interfere with cellular biochemical reactions usually by interacting at prime binding sites such as sulphhydryl groups in the enzyme systems. These toxic elements also compete with essential metals as enzymatic co-factors (Friberg, 1977). Essential elements can become toxic if the supply exceeds the optimum nutritional value.

A number of significant incidents were reported on accidental consumption of heavy metals through food. An episode which attracted world-wide attention from scientific and epidemiological point of view was the Minamatta Bay Incident (Saha and McKinlay, 1975).

The Minamatta Bay Incident has been described as neuropathia arising from the consumption of large quantities



of fish and shellfish having high methylmercury content i.e. 27 to 102 ppm, dry weight (Takizawa, 1979; Saha and McKinlay, 1975). The first epidemic which occurred in 1953 to 1960 claimed 121 patients with 46 deaths. The second epidemic in Niigata (1964 - 1965) claimed 5 deaths with initially 30 persons suffering from the disease (D'Itri and D'Itri, 1977).

The other incident which increased the awareness of hazards on consuming contaminated aquatic food was when high content mercury was reported in aquatic food from Sweden which later led to the ban of the contaminated food. Similar high values of mercury were found in fish from Lake Clair and Lake Erie (Canada) in 1970.

Cadmium poisoning which was initially observed in 1974, was reported in Jintsu River, Japan, where 44 patients suffered from cadmium poisoning. This unusual disease which results in skeletal deformities was later called "Itai-Itai" disease. It has been estimated that at least 100 deaths occurred due to this disease until the end of 1965 (Forstner and Wittmann, 1979).

These incidents illustrate how aquatic food chains can effectively accumulate heavy metals from aquatic environment and, resulting in contaminated food for consumers higher in the food chain. The effect can be enhanced in highly productive regions such as the estuaries, where there may be elevated heavy metals content from various sources.

## B) Brief account on individual heavy metal

### i) Mercury

Mercury (Quicksilver) is a non-essential, ubiquitous and highly toxic substance. It occurs in three oxidation states, namely



a) Metallic mercury ( $\text{Hg}^{\circ}$ ) which is elementary, insoluble and most reduced form of mercury. It is a liquid at ordinary temperature but tends to vapourize. It is non-polar and capable of transversing membrane barriers. (WHO, 1976; Carty and Malone, 1979).

b) Mercurous ( $\text{Hg}^{+}$ ) is an unstable, reduced ionic form. The mercury (I) compounds are formed from the dimeric cations. Mercury (I) readily oxidized to mercury(II) by treatment of oxidizing agents. This form does not form covalent bonds with other elements (Krenkel, 1974).

c) Mercuric ( $\text{Hg}^{2+}$ ) is a stable form which has a tendency to form covalent bonds (Krenkel, 1974). The mercury (II) halides have characteristics properties of covalent compounds such as low boiling point and high solubility in organic solvent relative to water. The solubility of  $\text{Hg(II)}$  in water is low ( $0.02$  to  $0.03 \text{ mg l}^{-1}$ ) at  $20^{\circ}\text{C}$  (Krenkel, 1974).

Speciation of mercury: The principal forms of mercury existing in nature depend not only upon the redox potential and the pH of the solution, but also the nature of the anions and chemical species present which are capable of forming strong bonds.

In an aquatic system, the higher valency mercury species predominate at more positive redox potential. At pH 5 to 9 and below Eh 0.45 mV, the principal forms of mercury species found in aerobic or anerobic system are  $\text{Hg}^{\circ}$  and  $\text{HgS}$  (cinnabar) (Gavis and Ferguson, 1972). Based on Hem's thermodynamic model,  $\text{HgCl}_2$  and  $\text{Hg(OH)}_2$  are predominant species in a simple aerobic aquatic system. At low pH,  $\text{HgCl}_2$  complex is predicted to be predominant while at higher pH,  $\text{Hg(OH)}_2$  is predominant. (Gavis and Ferguson, 1972).

In a moderate oxidizing condition, undissociated

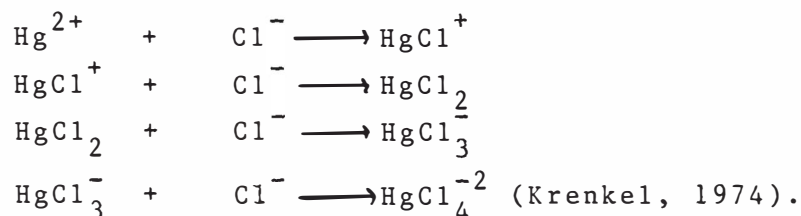




mercury is the predominant species and had a solubility of  $25 \mu\text{gl}^{-1}$ .

In a very reducing environment, the predominant species predicted to be present are  $\text{Hg}^\circ$ ,  $\text{HgS}_2^{2-}$  and  $\text{Hg}(\text{SH})_2^{2-}$ . In the presence of sulphide, mercury precipitates as insoluble stable  $\text{HgS}$  but a pH above 8.5 and in aerated solution, it may go into solution as polysulphides (Gavis and Ferguson, 1972; Benes and Havlik, 1979).

$\text{Hg}(\text{II})$  reacts with chloride ions, which are present in sea water, forming covalent complex ions like



The above reactions are important in studying the behaviour of  $\text{Hg}(\text{II})$  in an estuarine ecosystem. In oxygenated highly saline waters, the  $\text{Hg}$  solubility may be enhanced by the formation of uncharged  $\text{HgCl}_2$  or anionic complexes such as  $\text{HgCl}_3^-$ ,  $\text{HgCl}_4^{2-}$  and  $\text{HgOCl}^\circ$  whose concentrations depend on the chloride concentration and pH.

Organometallic Mercury Derivatives: Organometallic mercury (II) compounds are usually prevalent in aqueous environment and they constitute bulk of mercury transferred along the food web. The common organometallic mercury derivatives are mono- and di- organomercury (II) compounds. Mono-organic mercuric salts,  $\text{RHgX}$ , resembles the corresponding inorganic mercuric salts in their properties and reactions (Krenkel, 1974).

Methylmercury forms extremely stable complexes with anionic sulphhydryl or thiol groups which can lead to impairment of sulphhydryl-dependent processes, thus leading



to its toxicity (Carty and Malone, 1979). Methylmercury is very mobile in animal's body (Carty and Malone, 1979).

The formation of methylmercury compounds in the estuarine zone depends on the pH and chloride concentration. At low chloride concentration,  $\text{CH}_3\text{Hg}^+$  predominates while at a higher chloride concentration,  $\text{CH}_3\text{HgCl}$  predominates.

Methylation of Mercury: In an aqueous environment, the different mercury species which are present are ultimately converted to methyl mercury by process of methylation. Jensen and Jernelov (1969) reported that microorganisms are able to produce mono- and dimethyl mercury in aquatic sediment. The methylation of inorganic results in the release, upward movement and subsequent cycling of volatile mercury form (Aston et al., 1973).

The efficiency of methylation depends on microbial activity, the concentration and bioavailability of mercury which in turn depends on redox potential, pH, presence of sulphide and other chelating agents (Beijer and Jernelov, 1979).

The most favourable conditions for methylation is the oxidizing-aerobic regions of sediment where the pH range is 6 to 8 while the Eh is in the range of -0.2 to 0.4V (Andren and Nriagu, 1979).

The end products of methylation are determined by the pH. At neutral or acidic condition, monomethyl mercury predominates while in basic environment, dimethyl mercury predominates (Beijer and Jernelov, 1979).

In a highly reducing environment, where sulphide is present, the inorganic mercury is immobilised as mercuric sulphide. Thus, there is drastic decrease in methylation as sulphide begins to form (Fagerstrom and Jernelov, 1974;



Gavis and Ferguson, 1972).

### ii) Copper

Copper exhibits mono- and divalency. It forms simple water-soluble cation salts. It also forms strong complexes with sulphur and nitrogen-containing ligands.

In seawater, the free  $\text{Cu}^{2+}(\text{aq})$  is only a small fraction (about 1%) of the total dissolved copper (Boyle, 1979). The major species found in the seawater are  $\text{Cu}(\text{OH})\text{Cl}$  and  $\text{Cu}(\text{OH})_2^{\circ}$  (Boyle, 1979).

In the interstitial waters of estuarine sediment, cuprous sulphide ( $\text{Cu}_2\text{S}$ ) predominates at low pE but on increase of pE, cupric sulphide ( $\text{CuS}$ ) is the more stable form. As the pE increases to above -4.3, the stable phase is again cuprous sulphide (Leckie, 1979).

### iii) Lead

Lead is stimulating and oncogenic element and it is not essential for mammals (Venugopal and Luckey, 1978). It has been considered to be the largest contaminant in the environment (Schroeder and Darrow, 1973).

Lead resembles the divalent alkaline Group metals in its chemical behaviour more than its own Group, IVA. It differs from the Group IIA metals in its poor solubility of its salts such as hydroxides, sulphates and halides (Forstner and Wittmann, 1979).

Lead exhibits the valencies of +2 and +4 with valency +2 being more stable. The valency +4 is exhibited by organometallic lead compounds.

Majority of the lead ions in natural waters are associated with suspended and colloidal matter. In oxy-



generated sea water, lead oxide seems to be the most stable form of solid lead (Chow, 1978). In sea water having pH between 7 to 9, lead mainly complexes with  $\text{CO}_3^{2-}$  and to a lesser extent with  $\text{Cl}^-$ . At pH values nearer to 7, both  $\text{PbCO}_3^\circ$  and  $\text{PbCl}^+$  species are present in equal amounts but as the pH increases, the predominant specie is  $\text{PbCO}_3^\circ$  (Chow, 1978).

In an anerobic reducing environment, most of the dissolved lead precipitates as insoluble lead sulphide ( $\text{PbS}$ ).

#### iv) Zinc

Zinc is one of the most ubiquitous of all the essential trace elements. It is the 7th most abundant element on earth with concentration of 65 ppm in the earth crust. It exhibits a valency of +2 and forms stable, water soluble salts but at a higher pH, it forms insoluble  $\text{Zn(OH)}_2$ .

Zinc readily forms strong complexes with chloride, cysteine, fulvic acid and sulphide. It adsorbs strongly onto ferric hydroxide, silica, alumina, manganese dioxide and clay material (Young et al., 1980).

In sea water, zinc can exist in dissolved form (as free ions or dissolved complexes), solid precipitate form or adsorbed onto particulates. The predominant species present in sea water are  $\text{Zn}^{2+}$ ,  $\text{ZnOH}^+$ ,  $\text{ZnCO}_3$ ,  $\text{ZnSO}_4$  and  $\text{ZnCl}^+$ . In a reducing environment, bisulphides or polysulphides complexes are the predominant forms of zinc.

#### C) Studies done on distribution of heavy metals in estuaries

The heavy metal pollution in the estuaries throughout the world have received much attention. In British estuaries, numerous studies have been conducted on the



distribution and behaviour of heavy metals. Burton and Leatherland (1971) had examined the mercury concentration in the river and sea waters around Southampton and English Channel, which varied between 0.014 to 0.021  $\mu\text{gl}^{-1}$ . Abdullah and Royle (1974) analysed the dissolved and particulate trace elements in the Bristol Channel which included the Severn estuary and Swansea Bay. Clifton and Vivian (1975) studied the mercury contents in the sediment of Swansea Bay (South Wales Coast) where a chloro-alkali plant released small amount of mercury into the bay. They found high correlation between the mercury content, organic matter and mud fraction. But they concluded that the bay was not polluted as the typical mercury range was from 5 to 10  $\mu\text{gl}^{-1}$ .

The mercury content in Mersey estuarine sediment was studied by Craig and Morton (1976). They reported a mean value of 2.23  $\mu\text{gg}^{-1}$  mercury in the sediment. This estuary received domestic sewage and industrial wastes. Jaffe and Walters (1977) reported high trace element concentrations in the intertidal sediment of the Humber estuary. Later, Bartlett et al. (1978) examined the level of mercury and methylmercury in polluted British estuarine sediment which included the River Mersey estuary and Firth of Clyde area in Scotland. The total mercury level in sediment ranged from 0.4 to 44  $\mu\text{gg}^{-1}$ . Bellinger and Benham (1978) studied the level of copper, lead, tin and zinc in sediment of the enclosed dock-basins at Liverpool, Tilbury and Manchester.

Elderfield et al. (1979) studied the zinc levels in Conway River and its estuary. The distribution of eleven metals in the highly developed estuaries of Tyne, Tees and Humber were done by Taylor (1979). These rivers drain into the North Sea. Taylor (1979) observed that geology of an area was a more important factor than the industrial input in deciding the heavy metal contents in sediment.



Millward and Herbert (1981) analysed the iron, manganese and mercury contents in the Plym River estuary which receives both the industrial and domestic wastes. The mercury concentration in the sediment ranged from 0.18 to 2.61  $\mu\text{gg}^{-1}$ . Rae and Aston (1981) studied the level of mercury in Mersey and Wyre estuaries. Wyre estuary received two inputs of contaminated effluents. One was the chloroalkali factory and the other localised mercury contamination. The sediment at Mersey and Wyre contained 0.1 to 8.8  $\mu\text{gg}^{-1}$  and 0.1 to 29  $\mu\text{gg}^{-1}$  mercury respectively.

Other studies on European estuaries were on the River Rhine estuary. Duinker et al. (1974) studied the behaviour of copper, zinc, iron and manganese in the Dutch Walden Sea. Duinker and Nolting (1977, 1978) again studied the dissolved and particulate trace metals in the Rhine estuary and the behaviour of mixing, removal and mobilization of trace elements in this estuary. Duinker and Nolting (1977) measured the concentration of dissolved copper, zinc, cadmium in Southern Bight of the North Sea and compared them with the levels in the coastal regions under the influence of the Rhine and Scheldt rivers.

In America, Andren and Harriss (1973) studied the level of methylmercury in the estuarine sediment of Mobile Bay, Mississippi River and Everglades. The mercury content in the sediment ranged from 80 to 600  $\text{ngg}^{-1}$ . Later, Lindberg and Harriss (1974) reported a mercury level of 0.22 to 1.62 ppm range in the interstitial waters and sediment of Florida Everglades and Mobile Bay estuary. Fitzgerald and Lyons (1973) compared the organic and inorganic mercury in the coastal waters and adjacent waters in the vicinity of Long Island Sound. Waldhaeur et al. (1978) studied copper and lead levels in heavily urbanised and industrialised Raritan River and Arthur Kill estuary which drains into Raritan and Lower New York Bay. The concentration of copper and lead was 65  $\mu\text{gl}^{-1}$  and 13.9  $\mu\text{gl}^{-1}$  respectively. Bowers and Yeats (1978) examined the distribution and behaviour of iron, manganese, cobalt, nickel, copper and zinc in Upper St.



Lawrence River estuary. Menon et al. (1979) studied the trace elements released from the estuarine sediment of South Mosquito Lagoon near Kennedy Space Centre. Meyerson et al. (1981) analysed the bottom estuarine sediment of polluted Newark Bay, New Jersey for zinc, lead, cadmium and mercury. The mercury concentration was in the 0.3 to 31 ppm range.

Some heavy metals in the coastal rivers and estuaries around the Bay of Bengal were studied by Braganca and Sanzgiry (1980). The dissolved iron in sea water was reported to be in 6.3 to 131.5  $\mu\text{g l}^{-1}$  range. The average concentration of dissolved copper was 4.9  $\mu\text{g l}^{-1}$  and dissolved zinc ranged from 2.4 to 20.0  $\mu\text{g l}^{-1}$ .

In Australia, Bloom and Ayling (1977) conducted a comprehensive study of cadmium, chromium, copper, iron, lead, manganese, mercury, nickel and zinc in filtered water, suspended matter, sediment, fish, airborne particulates and sewage in Derwent River estuary. Derwent River is one of the most polluted rivers affected by metallurgical wastes, raw and partly treated sewage effluents.

Not much work has been reported on the heavy metal content in aquatic biota from estuarine region. Portmann (1972) assessed the presence of certain heavy metals like cadmium, copper, lead, mercury and zinc in coastal water fishes in vicinity of England and Wales. Topping (1973) studied copper, zinc, cadmium and lead contents in four commercial fish species collected from Scottish waters which included a heavily polluted estuary, Firth of Clyde, and relatively unpolluted regions of Scotland like Orkney and Shetland.

Wharfe and Van Den Broek (1977) had analysed zinc, mercury, copper, lead and cadmium in macro-invertebrates and fish from Medway estuary, England; while Zauke (1977)

