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# Spatial variation of heavy metals in Tenualosa ilisha muscle: A case study from the lower Gangetic delta and coastal West Bengal

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**Abstract:** We analyzed the concentrations of zinc, copper, lead and cadmium in the muscle of Hilsha fish (*Tenualosa ilisha*) in the lower stretch of the River Ganga and coastal West Bengal during April, 2015 using a Perkin-Elmer Sciex ELAN 5000 ICP mass spectrometer and expressed as ppm dry weight. The concentrations ranged from 11.37 ±0.93– 76.18 ±1.09 for Zn, 9.09 ±0.44– 21.44 ±0.89 for Cu, BDL – 1.85 ±0.50 for Pb and BDL – 0.04 ±0.01 for Cd in ppm dry wt. Heavy metals in the muscle of the investigated fish species were compared with the permissible (safety) levels for human consumption. All the selected heavy metals except Pb (in station 2) were below the permissible level confirming the species mostly suitable for human consumption.

**Keywords:** Heavy metals, *Tenualosa ilisha*, River Ganga, Coastal West Bengal, ICP mass spectrometer

## INTRODUCTION

Pollution of environment represents a major problem in both developed and underdeveloped countries. Among the different categories of pollutants persistent environmental contaminants are the worst in context to human health. Emissions of harmful substances have negative effects on the natural environment and human health (Gadzala-Kopciuch *et al*., 2004). When the consequences of environmental pollution become visible, it is often too late to prevent and chronic toxic effects, impossible to notice at the initial stage of the process, may manifest themselves after many years (Alloway and Ayres, 1998). That is the main reason why it is imperative to conduct periodic pollution monitoring of aquatic environments. Heavy metals are stable and persistent environmental contaminants of aquatic environments. They occur in the environment both as a result of natural processes and as pollutants from human activities (Garcia-Montelongo *et al.,* 1994; Jordao *et al*., 2002). Some metals like Zn and Cu, which are required for metabolic activity in organisms, lie in the narrow "window" between their essentiality and toxicity. Other heavy metals like Pb and Cd may exhibit extreme toxicity even at low levels under certain conditions, thus necessitating regular monitoring of sensitive aquatic environments (Cohen *et al.,* 2001; Fergusson, 1990; Peerzada *et al*., 1990). From an environmental point of view, coastal zones can be considered as the geographic space of interaction between terrestrial and marine ecosystems that is of great importance for the survival of a large variety of plants, animals and marine species (Castro *et al*., 1999). The coastal zone receives a large amount of metal pollution from agricultural and industrial activity (Usero *et al.,* 2005). Adverse anthropogenic effects on the coastal environment include eutrophication, heavy metals, organic and microbial pollution and oil spills (Boudouresque and Verlaque, 2002). Heavy metals have the tendency to accumulate in the various aquatic animals, and the accumulation depends upon the intake and the elimination from the body (Karadede, *et al.*, 2004). Marine fishes exposed to these heavy metals have been consumed as sea foods and hence are a connecting pathway for the transfer of toxic heavy metals in human beings. Fishes are major part of the human diet because, it has high protein content, low saturated fat and also contains omega fatty acids known to support good health (Dural, *et al*., 2007; Ikem and Egiebor, 2005).

The Gangetic delta, at the apex of Bay of Bengal is recognized as one of the most diversified and productive ecosystems of the Tropics. The deltaic lobe is unique for its wilderness, mangrove gene pool and tiger habitat. However due to intense industrial activities in the upstream zone, and several anthropogenic factors, the western part of the deltaic complex is exposed to pollution from domestic sewage and industrial effluents leading to serious impacts on biota (Mitra and Choudhury, 1992). The presence of Haldia port-cum-industrial complex in the downstream region of the River Ganga (also known as the Hooghly River) has accelerated the pollution problem with a much greater dimension (Mitra, 1998). The organic and inorganic wastes released from these industries and urban units contain substantial concentrations of heavy metals. The central part of the delta (encompassing the surroundings of Matla River) is relatively less stressful in terms of industrial discharge. Due to siltation of the Bidyadhari channel the area does not receive any water supply from the Hooghly River in the western sector and is therefore tide-fed in nature receiving the tidal flux from the Bay of Bengal (average salinity = ~32 psu).

Hisha fish (*Tenualosa ilisha*) is one of the major dollar earning items of Indian sub-continent. The fish is mainly planktivorous and schools in coastal waters. It shows anadromous habit *i.e*., it ascends up the River Ganga for around 50 – 150 km to spawn during the south-west monsoons (June to September) and also in January to March. Hence, it is caught mostly during monsoon and early summer every year (Hora and Nair, 1940). It is very much popular for it’s delicacy. People living in the eastern sector of India specially in West Bengal, Bihar, Assam, Odisha and Jharkhand are been able to consume lot of this species due to it’s availability in these area thoughout the year. For this reason it is very essential to monitor the pollution status of the fish muscle time to time. Various studies have already been undertaken worldwide on the contamination of different fish species to determine their heavy metal concentration (Carvalho, *et al*., 2005; Sivaperumal, *et al*., 2007; Raja, *et al.*, 2009; Yilmaz, 2009; Ahmed and Nain, 2008; Nawal, 2008; Nath and Banerjee, 2012) but little information is available pertaining to the accumulation of heavy metals in coastal biota especially coastal fishes collected earlier from the North Eastern Bay of Bengal, India (Mitra, 2000; Bhattacharya, *et al*., 2001 & 2006). The present paper aims to focus on the concentrations of selective heavy metals (Zn, Cu, Pb and Cd) in the muscle tissue of Hilsha fish (*Tenualosa ilisha)* collected from four stations distributed in two sectors (western and central Indian Sundarbans) of the lower Gangetic region thereby asses if it is safe to consume at all.

## MATERIALS AND METHODS

### Description of the study site

Two sampling sites were selected each in the western and central sectors of lower Gangetic delta and coastal West Bengal at the apex of the Bay of Bengal. The western sector of the study area receives the snowmelt water of mighty Himalayan

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glaciers after being regulated through several barrages on the way. The central sector on the other hand, is fully deprived from such supply due to heavy siltation and clogging of the Bidyadhari channel since the late 15$^{th}$ century (Chaudhuri and Choudhury, 1994). The western sector also receives wastes and effluents of complex nature from multifarious industries concentrated mainly in the upstream zone. On this background four sampling stations (two each in western and central sectors) were selected (Table 1 and Figure 1) to analyze the concentrations of heavy metals in the selected ilish fish inhabiting the zone.

**Table 1:** Sampling stations with coordinates and salient features

| **Station** | **Coordinates** | **Salient Features** |
|---|---|---|
| Digha (Stn.1) | 21° 37′ 17.4" N 87° 31′ 36.5" E | Situated at the confluence of the River Hooghly and the Bay of Bengal in the south-western sector of Indian Sundarbans. The station is an important tourism spot and fish landing zone in the northeast coast of India. |
| Haldia Island (Stn.2) | 22° 01′ 18.3" N 88° 03′ 11.4" E | It is located in the Hooghly estuary in the western sector of the lower Gangetic delta and is the industrial HUB of the maritime state of West Bengal in India. |
| Gosaba (Stn. 3) | 22° 15′ 45" N 88° 39′ 46" E | Located in the Matla Riverine stretch in the central sector of Indian Sundarbans. |
| Annpur in Satjelia Island (Stn. 4) | 22° 11′ 52" N 88° 50′ 43" E | Located in the central sector of Indian Sundarbans. Noted for its wilderness and mangrove diversity; selected as our control zone. |

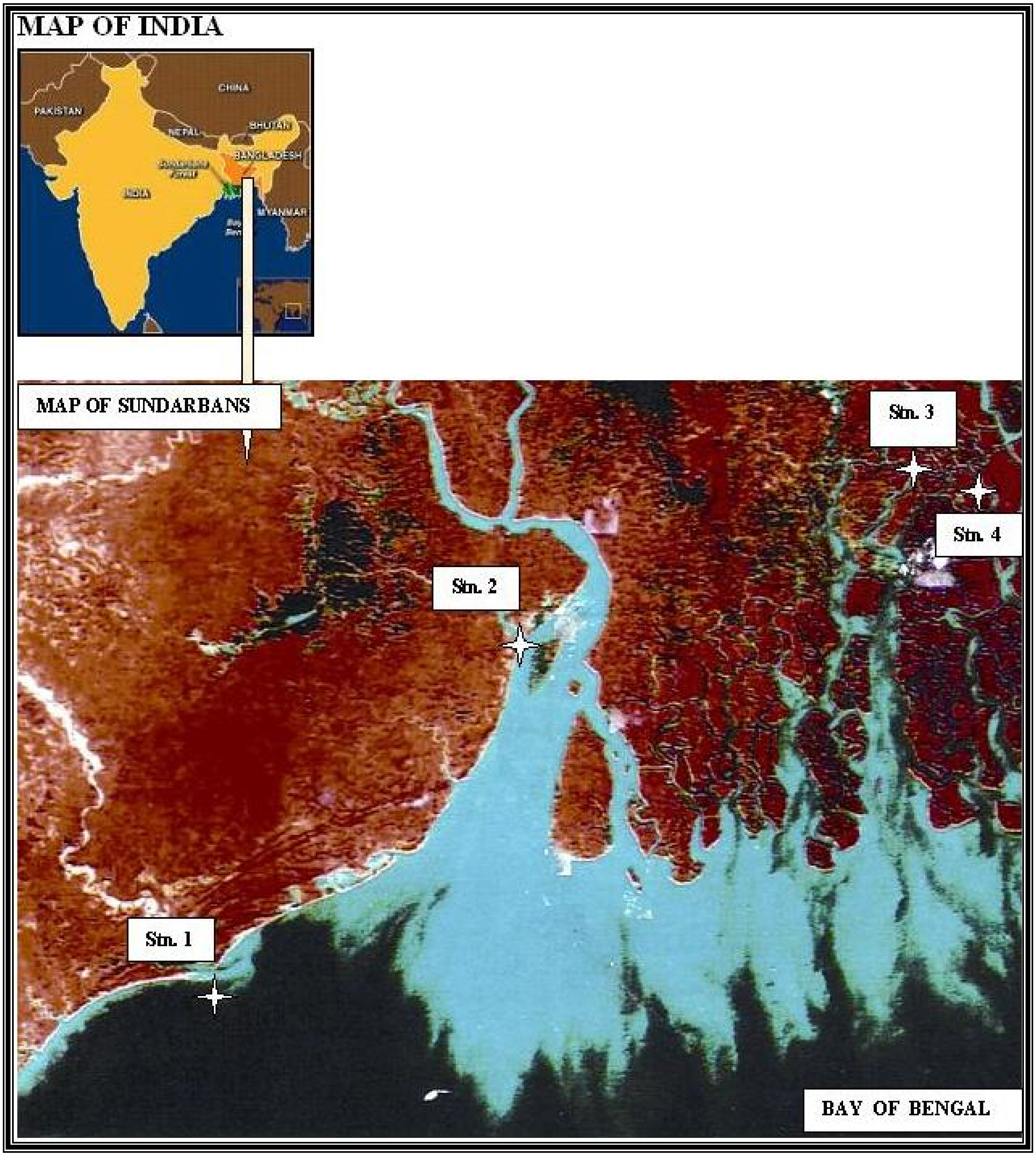


**Figure 1:** Location of sampling stations

**Sampling of Specimen**

Matured fish samples (*Tenualosa ilisha)* were collected from the local fisherman from the selected stations (Table 1) during 10th April to 25th April, 2015. The collected samples were stored in a container, preserved in crushed ice, and brought to the laboratory for further analysis. Similar sized specimens of the species were sorted out for analyzing the metal level in the muscle.

**Metal Analysis**

**Inductively Coupled Plasma – Mass Spectrometry (ICP-MS)** is now - a - day accepted as a fast, reliable means of multi-elemental analysis for a wide variety of sample types (Date and Gray, 1988). A **Perkin-Elmer Sciex ELAN 5000 ICP** mass spectrometer was used for the present analysis. A standard torch for this instrument was used with an outer argon gas flow rate of 15 L/min and an intermediate gas flow of 0.9 L/min. The applied power was 1.0 kW. The ion settings were standard settings recommended, when a conventional nebulizer/spray was used with a liquid sample uptake rate of 1.0 mL/min. A Moulinex Super Crousty microwave oven of 2450 MHz frequency magnetron and 1100 Watt maximum power Polytetrafluoroethylene (PTFE) reactor of 115 ml volume, 1 cm wall thickness with hermetic screw caps, were used for the digestion of the collected biological samples. All reagents used were of high purity available and of analytical reagent grade. High purity water was obtained with a Barnstead Nanopure II water-purification system. All glasswares were soaked in 10% (v/v) nitric acid for 24 h and washed with deionised water prior to use.

The analyses were carried out on composite samples of 10 specimens of the species having uniform size. This is a measure to reduce possible variations in metal concentrations due to size and age. 20 mg composite sample from each species were weighed and successively treated with 4 ml aqua regia, 1.5 mL HF and 3 ml $H_2O_2$ in a hermetically sealed PIFE reactor, inside a microwave oven, at power levels between 330-550 Watt, for 12 min to obtain a clear solution. The use of microwave-assisted digestion appears to be very relevant for sample dissolution, especially because it is very fast (Nadkarni, 1984; Matusiewicz and Sturgeon, 1989; De la Guardia, 1990). After digestion, 4 ml $H_2BO_3$ was added and kept in a hot water bath for 10 min, diluted with distilled water to make up the volume to 50 ml. Taking distilled water in place of biological samples and following all the treatment steps described above the blank process was prepared. The final volume was made up to 50 ml. Finally, the samples and process blank solutions were analyzed by ICP-MS. All analyses were done in triplicate and the results were expressed with standard deviation.

The accuracy and precision of our results were checked by analyzing standard reference material (SRM, Dorm-2). The results indicated good agreement between the certified and the analytical values (Table 2).

**Table 2:** Concentrations of metals found in Standard Reference Material DORM-2 (dogfish muscle) from the National Research Council, Canada (all data as means ± standard errors, in ppm dry wt)

| **Value** | **Zn** | **Cu** | **Pb** | **Cd** |
|---|---|---|---|---|
| Certified | 26.8 | 2.34 | 0.065 | 0.043 |
| SE | 2.41 | 0.18 | 0.009 | 0.005 |
| Observed* | 23.9 | 2.29 | 0.060 | 0.040 |
| SE | 1.99 | 0.17 | 0.006 | 0.006 |
| Recovery (%) | 89.2 | 97.8 | 92.3 | 93.0 |

*Each value is the average of 5 determinations

## RESULTS

In *Tenualosa ilisha*, heavy metals accumulated as per the order Zn > Cu > Pb > Cd. In the present study we considered marketable sized fish (approximately 500 gm in weight). The maximum level of heavy metals was found at Stn. 2 and minimum at Stn. 4 (Table 3). The concentrations of selected heavy metals in the muscle ranged from 11.37 ±0.93– 76.18 ±1.09 for Zn, 9.09 ±0.44– 21.44 ±0.89 for Cu, BDL – 1.85 ±0.50 for Pb and BDL – 0.04 ±0.01 for Cd in ppm dry wt. in the study area (Table 3).

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**Table 3:** Heavy metal concentrations (in ppm dry wt.) in Hilsha fish (*Tenualosa ilisha)* muscles

| *Heavy metal* | Stn. 1 | Stn. 2 | Stn. 3 | Stn. 4 | WHO (1989) level in food |
|---|---|---|---|---|---|
| *Zn* | 39.13 ±1.22 | 76.18 ±1.09 | 16.48 ±1.04 | 11.37 ±0.93 | 100 |
| *Cu* | 19.69 ±1.51 | 21.44 ±0.89 | 15.70 ±0.59 | 9.09 ±0.44 | 30 |
| *Pb* | BDL | 1.85 ±0.50 | BDL | BDL | 0.05 |
| *Cd* | 0.04 ±0.01 | BDL | BDL | BDL | 0.05 |

## DISCUSSION

Heavy metal contamination of the environment has been occurring for centuries, but its extent has increased markedly in the last fifty years due to technological developments and increased consumer use of materials containing these metals. Pollution by heavy metals is a serious problem due to their toxicity and ability to accumulate in the biota (Islam and Tanaka, 2004). There is still a general concern about the impact of metals in the aquatic environment (Grosell and Brix, 2005). Heavy metals have contaminated the aquatic environment in the present century due to intense industrialization and urbanization. The Gangetic delta and coastal zone of West Bengal is no exception to this usual trend. The rapid industrialization and urbanization of the city of Kolkata (formerly known as Calcutta), Howrah and the newly emerging Haldia complex in the maritime state of West Bengal has caused considerable ecological imbalance in the adjacent coastal zone (Mitra and Choudhury, 1992; Mitra, 1998). The Hooghly estuary, situated on the western sector of the Gangetic delta receives drainage from these adjacent cities, which have sewage outlets into the estuarine system. The chain of factories and industries situated on the western bank of the Hooghly estuary is a major cause behind the gradual transformation of this beautiful ecotone into stinking cesspools of the megapolis (Mitra and Choudhury, 1992). The lower part of the estuary has multifarious industries such as paper, textiles, chemicals, pharmaceuticals, plastic, shellac, food, leather, jute, tyres and cycle rims (UNEP, 1982). In addition to industrial discharges, proliferation of tourism units has also contaminated the environment to a great extent particularly around Haldia (station 2). These units are point sources of heavy metals in the estuarine and coastal waters. Due to toxic nature of certain heavy metals, these chemical constituents interfere with the ecology of a particular environment and on entering into the food chain they cause potential health hazards, mainly to human beings. The present study is important not only from the safety point of view of human health, but also from the quality point of view as this species has high export value.

Of the four metals studied, Zn and Cu are essential elements while Pb and Cd are non-essential elements for most of the living organisms (Radojevic and Bashkin, 1999). The concentrations of Zn and Cu in the species were relatively higher, compared to the concentration of other metals in the same sample. It can be explained because these metals (Zn and Cu) are essential elements required by animals for metabolic process. Carbonell and Tarazona (1994) concluded that different tissues of aquatic animals provide and/or synthesize non-exchangeable binding sites resulting in different accumulation levels.

The primary sources of Zn in the present geographical locale are the galvanization units, paint manufacturing units and pharmaceutical processes, which are mainly concentrated in the Haldia industrial sector (Station 2). Reports of high concentration of Zn were also highlighted in the same environment by earlier workers (Mitra and Choudhury, 1992; Mitra and Choudhury, 1993; Mitra, 1998).

The main sources of Cu in the coastal waters are antifouling paints (Goldberg, 1975), particular type of algaecides used in different aquaculture farms, paint manufacturing units, pipe line corrosion and oil sludges (32 to 120 ppm). Ship bottom paint has been found to produce very high concentration of Cu is sea water and sediment in harbours of Great Britain and southern California (Bellinger and Benham, 1978; Young *et al*., 1979). In the present study area, the major source of Cu is the antifouling paints used for conditioning fishing vessels and trawlers apart from industrial discharges (that is predominant around station 1). Station 1 (Digha) is not only the site for tourism and beach recreational activities, but it is also a major fish landing station in northeast coast of India, where large number of fishing vessels and trawlers contaminate the water with Cu. This is the reason why Cu was detected in considerable quantity in the fish muscle of

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station 1 (although below the safety level). Traces of Cu in the samples of stations 3 and 4 (which is totally an industry-free zone) may also be related to use of antifouling paints to keep the fishing vessels and passenger boats free from biofoulers.

Pb is a toxic heavy metal, which finds its way in coastal waters through the discharge of industrial waste waters, such as from painting, dyeing, battery manufacturing units and oil refineries *etc*. Antifouling paints used to prevent growth of marine organisms at the bottom of the boats and trawlers also contain lead as an important component. These paints are designed to constantly leach toxic metals into the water to kill organisms that may attach to bottom of the boats, which ultimately is transported to the sediment and aquatic compartments. Lead also enters the oceans and coastal waters both from terrestrial sources and atmosphere and the atmospheric input of lead aerosols can be substantial. Station 2 is exposed to all these activities being proximal to the highly urbanized city of Kolkata, Howrah and adjacent to the newly emerging Haldia port - cum - industrial complex.

The main sources of Cd in the present geographical locale are electroplating, manufacturing of Cd alloys, production of Ni-Cd batteries and wielding. Cadmium may accumulate in humans from food chain magnification and may induce kidney dysfunction, skeletal damage, and reproductive deficiencies (Commission of the European communities, 2001).

The levels of Zn and Cu were found much below the permissible limit (as prescribed by WHO, 1989) in all the selected stations. No trace of Pb and Cd was recorded in the fish muscle from stations 3 and 4, which are located almost in industry-free zone surrounded by mangrove vegetation although, in station 2 the heavy metal Pb was found slightly above the safety/permissible (Table 3). Mass awareness, frequent monitoring and implementing laws to curb the situation at point sources in and around station 2 (Haldia industrial complex) is a must-to-do approach to prevent the probability of bioaccumulation by organisms belonging to upper trophic level of the coastal and estuarine food-chain.

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