

Study on Heavy Metals (As, Cr, Cd & Pb) and its Human Health Risk Assessment in Commercially Important Finfishes and Shell Fishes Collected from Chennai Coast, India

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ABSTRACT

Fish, shrimp, crab, and mollusks are significant providers of proteins, vitamins, minerals, polyunsaturated fats and polysaccharides and form the chief source of food to a peninsular country like India. Fish food production and Human health can be affected by heavy metal contamination. Heavy metals can be significant carcinogenic agents based on their toxic effects and bioaccumulation. The present study aims to analyse the concentrations of four heavy metals (As, Cr, Cd and Pb) in seawater and their bioaccumulation in commercially important fishes (*Nemipterus japonicus*, *Rastrelliger kanagurta*), Crab (*Portunus sanguinolentus*) and Shrimp (*Penaeus monodon*, *Penaeus indicus*). The samples were collected from Pulicat, Ennore, Pattinapakkam, Kovalam and Thiruvannamiyur regions of the Chennai coast and analysed using atomic absorption spectrophotometer and the results were interpreted with the USEPA human health risk assessment. The hierarchy of the concentration of heavy metals in the Crude oil is in the order Ni> As> Cr> Pb> Cd>Cu, seawater is in the order Pd> Ar> Cr> Cd. The species studied showed the highest accumulation of heavy metal in the liver and lowest in muscle tissues. The highest BCF concentration was recorded for Cr and lowest for Pb and BCF of muscles of all species showed lowered accumulation rate. THQ values of As, Cd and Pb in *P. sanguinolentus* from Pattinapakkam, Ennore and Kovalam, As, Cd and Pb in *R. kanagurta* from Ennore and Pattinapakkam were above 1. The values stipulate that prolonged consumption of these two species from the Ennore, Kovalam and Pattinapakkam coast may be potentially hazardous to human health. Cancer risk values for Cd in all five species studied was higher than the acceptable range in all the studied locations. Cr in the species *N. japonicus*, *R. kanagurta* from Ennore, Pulicat and Thiruvannamiyur and in *P. sanguinolentus* from Ennore and Kovalam, *P. indicus* and *P. monodon* from Pulicat and Pattinapakkam were close to the cancer risk value range, which indicates that the local consumers are at carcinogenic risk associated with Cd and Cr posed from consumption of fish and crustaceans' species. Application of effective preventive measures and appropriate strategies to minimize heavy metal pollution can be identified and developed by studies on the earlier and current incidents and adopted or integrated into coastal management policies.

Key words : Heavy metals, Bioaccumulation, Health risk assessment, Chennai coast.

Introduction

Fish, shrimp, crab, and mollusks are significant providers of proteins (amino acids), vitamins (Vitamin-B complex, Vitamin-D, A), minerals (iron, selenium, iodine, zinc), polyunsaturated fats (omega-3 fatty

Inbaraj R.M. - Since deceased

acids), and polysaccharides (chitosan) (Tilami and Sampels 2018; Dale *et al.*, 2019; Inanli *et al.*, 2020; Nurdiani *et al.*, 2020). These nutrients are necessary for human health and are helpful in the treatment of heart disease, cognitive difficulties, and muscle problems (Hosomi *et al.*, 2012; Jayasekara *et al.*, 2020). Fish food contributes about 20% of the average per capita intake of about 3.2 billion people (FAO, 2018). The annual per capita fish consumption for the total population in India is 5-6 kg and for fish-eating population it is 8-9 kg (Salim, 2016). India contributes about 4.7% of global marine water capture production and ranks first in Inland water captures produce (FAO, 2022). Contamination with metals or any hazardous substance in the fish produce can affect the country's economy and health of the consuming population.

Heavy metals entering the ecosystem through pesticides, fertilizers, agricultural waste, urban mining, domestic and factory sewage waste, effluents from industrial activities related to the development of human society are major source of metal contamination (Hauton *et al.*, 2017; Vardhan *et al.*, 2019; Wang *et al.*, 2019; Kortei *et al.*, 2020; Ozden *et al.*, 2020; Zhang *et al.*, 2020). Another significant known source of heavy metal contamination is spillage of crude oil from the vessels (Kankara *et al.*, 2016). Metals thus discharged into the ecosystem have an impact on the environment due to their long persistence, accumulative behaviour and toxicity (Hossain *et al.*, 2022). Though influenced by pH, temperature, alkalinity, pollutant type, geological position and feeding behaviour, habitat, age and sex of the species (Jonathan *et al.*, 2015; Aytakin *et al.*, 2019), aquatic organisms living in the contaminated environment accumulate trace metals and transfer them to the organisms in the upper level in the hierarchy of the food chain. The quantity of heavy metals accumulated in organisms varies based on the organisms' ecological and physiological characteristics, as well as the source of the metal in the environment (Barone *et al.*, 2018). Toxic metals, through their influence on mutational processes and gene flow, can change genetic diversity and species population numbers (Rumisha *et al.*, 2017).

Finfish and shellfish are considered important bioindicator organisms as they are in the upper hierarchy of the food chain and consumed by humans and have a high tendency to accumulate metals. Metals released into aquatic bodies build up due to the poor waste management and improper disposal

practices and pose a significant health danger to consumers, particularly in underdeveloped countries (Chen *et al.*, 2011; Hossain *et al.*, 2022). Heavy metal pollution has increased in recent years and has become a worldwide concern.

The exploding human-population and the reduction in terrestrial resources have resulted in increased consumption of sea foods. As the marine environment is extensively contaminated due to heavy metal deposition, increased consumption of seafood may raise concerns about human health issues, despite its advantages. (Dehghani *et al.*, 2021; Lloret *et al.*, 2016).

The growing interest of the people in fish and fish products in recent years makes this study significant. Many local and international monitoring programmes have been established to monitor the quality of fish for human consumption and the health of the aquatic ecosystem, as diet is the main route of exposure to heavy metals in humans, hence the risk assessment of these metals is essential (Hossain *et al.*, 2022). Heavy metals can be carcinogenic or non-carcinogenic based on their toxic effects and bioaccumulation. USEPA had suggested human health risk assessment associated with fish consumption which includes (Oral reference dose (RfD) value, Target hazard quotient (THQ), Target Cancer Risk (TR), Hazard Index (HI).

The present study aims to analyse the heavy metals present in the oil sludge and the concentrations of heavy metals [Arsenic (As), Chromium (Cr), Cadmium (Cd) and Lead (Pb)] in seawater and their bioaccumulation in commercially important fin fishes (*Nemipterus japonicus*, *Rastrelliger kanagurta*), and shellfishes (*Portunus sanguinolentus*) (*Penaeus indicus*, *Penaeus monodon*) collected from the coastal regions of Chennai.

Materials and Method

Study Area

The coast of Chennai, extending from Pulicat in the north to Kovalam in the south is selected to carry out the present study. This study area is significant as its coastal waters are highly polluted with the effluents from thermal power plants and petroleum, paint, pharmaceutical, leather, automobile and chemical industries which are potential sources of heavy metal contamination. The Cooum, Adyar and Kosasthalaiyar rivers which run across the city

carry these effluents directly into the sea (Kankara *et al.*, 2016; Santhiya *et al.*, 2011). Besides these, the probability of heavy metal pollution through oil spills is high as the city hosts India's second busiest container port, which receives all types of cargo and container vessels. The massive oil spill incident due to the collision between two cargo ships BW maple and Indian vessel MT Kanchipuram at Ennore port which released about 75 metric tons of hazardous oil into the sea (Han *et al.*, 2018) that covered about 30 km of shoreline area from Ennore port to Thiruvannmiyur beach was considered for the selection of the study area. The study area is orientated between 13°N and 12°N which covers approximately equal distances on either side from the point of Chennai oil spill which includes places of historic importance, tourist beaches and coastal habitats like estuaries, lagoons, etc. The five sampling locations along the coast for the present study are; Pulicat (13°25'41.2"N 80°18'33.4"E), Ennore (13°10'54.0"N 80°18'58.6"E), Pattinapakkam (13°02'19.1"N 80°16'53.3"E), Thiruvannmiyur (12°58'32.3"N 80°16'03.7"E) and Kovalam (12°47'28.5"N 80°15'07.4"E) (shown in Fig. 1.)

Sample collection and analysis

One-time crude oil samples were collected manually by scooping from floating oil (surface spill) into clean polyethylene bags from Ennore (13°10'54.0"N 80°18'58.6"E). Samples of deposited oil sludge were

collected in clean polyethylene bags from sandy beaches of Ennore, Thiruvannmiyur, (12°58'32.3"N 80°16'03.7"E) and Kovalam (12°47'28.5"N 80°15'07.4"E). The samples were analyzed using the Atomic Absorption Spectrophotometer method for the presence of heavy metals. The crude oil samples were analysed and the heavy metals present are quantified. The metals As, Cr, Cd and Pb are identified for the present study based on their abundance in the crude oil samples and their relative toxicity.

Seawater samples were collected seasonally from the five locations of the present study during Winter, Summer, Pre-monsoon and Monsoon. The bottles were rinsed with seawater of corresponding locations just before the collection of water samples to ensure minimal or zero alteration of the concentration of heavy metals. Water samples collected were brought to the laboratory in an icebox and filtered. The samples were then tested for quantifying the concentrations of the four heavy metals (As, Cr, Cd and Pb) to analyze the level of contamination.

Five specimens of similar size of each candidate species (fishes (*N. japonicus*, *R. kanagurta*), Crab (*P. sanguinolentus*) and Shrimps (*P. indicus*, *P. monodon*)) (Fig. 2.) were collected from all five locations from the fishermen when the fishing boat arrived at the shore, in clean zip-lock pouches kept in icebox and transported to the laboratory. Morphometric measures and ecological characteristics of the samples were recorded.

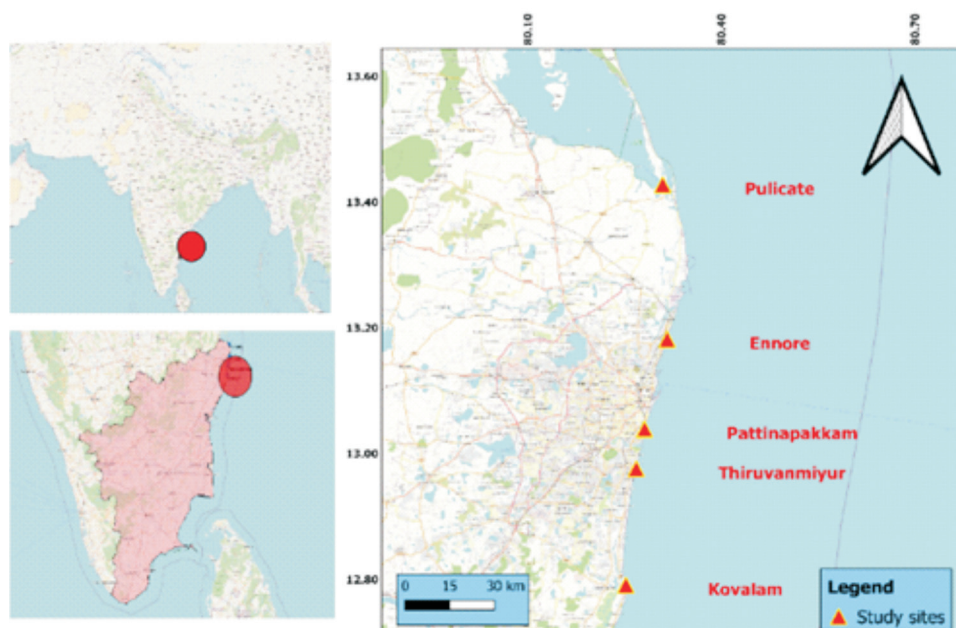


Fig. 1. Map showing the 5 different sample collection location of the present study

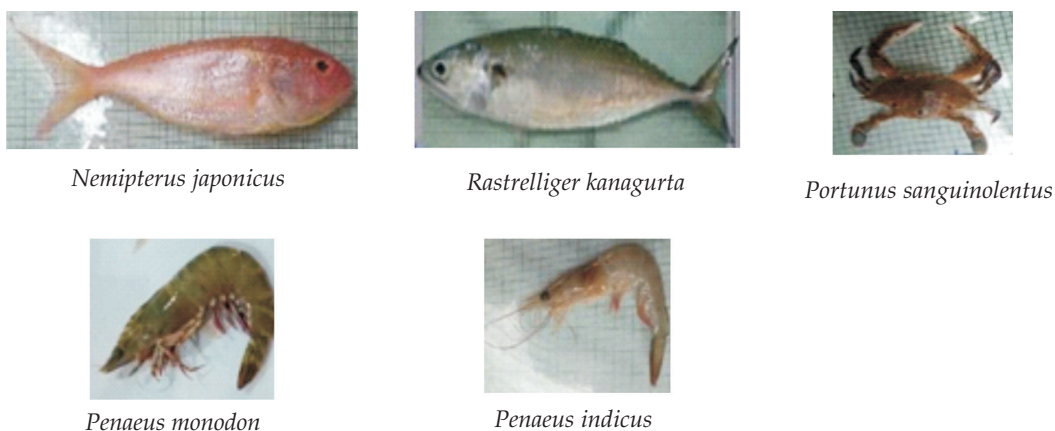


Fig. 2. Species collected for present analysis

Specimens were washed with distilled water and different organs of fish (liver, intestine, gills, skin, gonads, muscle), Crab (Spawn, gills, muscles) and shrimps (muscle, carapace) were dissected out separately on a clean table with a sterile knife and blade. Pooled samples of the organs were taken to obtain sufficient quantity of tissue in each sample and preserved in -20°C after labeling. Each preserved sample was lyophilized, powdered in a clean mortar and pestle and further stored at -20°C until the analysis of heavy metal. The lyophilised samples were digested in digestion flasks with perchloric acid, nitric acid and sulphuric acid in the ratio of 1:5:1 on a hot plate until the tissues were dissolved completely and subjected to quantitative analysis of the heavy metals using a calibrated atomic absorption spectrophotometer with recovery rates above 90%. The values obtained were used for following analytical studies to draw inferences.

Bioconcentration Factor (BCFs)

BCF is the ratio of concentration of a heavy metal in an aquatic organism to its concentration in the water (Zhang *et al.*, 2018; Maurya *et al.*, 2019). Where BCF is represented as l/Kg dry weight as concentration in biota is mg/kg dry weight and concentration in water is mg/l.

Estimated Daily Intake (EDI) of metals

$$\text{EDI} = \frac{C \times \text{IR}}{\text{BW}}$$

Where IR is the average food ingestion rate (55.8 g/day for adults) and (52.5 g/day for children) (USEPA, 2008; Tong *et al.*, 2016; Vu *et al.*, 2017;

Zhang *et al.*, 2018), considering 79% moisture content 4.8 is the conversion factor to convert from dry to wet weight (Maurya *et al.*, 2019), C is the mean concentration of heavy metals in crustacean and fish (mg/ kg dry weight), BW is the average body weight which is 60 kg for an adult and 30 kg for a child (Zhang *et al.*, 2018).

Target Hazard Quotient (THQ)

THQ is the non-carcinogenic risk level estimated for consumption of heavy metal and it is calculated as per the Screening values for target analyses table set aside by USEPA region III risk-based concentration table suggested by USEPA, 2011 (Baki *et al.*, 2018).

$$\text{THQ} = \frac{\text{EF} \times \text{ED} \times \text{IR} \times \text{Cf} \times \text{C} \times 10^{-3}}{\text{BW} \times \text{ATn} \times \text{RfD}}$$

Where EF is the exposure frequency (365 days/year), ED is the exposure Duration (here we consider 60 years for adults to estimate non-carcinogen risk level), IR is the average food ingestion rate (55.8 g/day for adults) (Tong *et al.*, 2016; Zhang *et al.*, 2018), considering 79% moisture content 4.8 is the conversion factor to convert from dry to wet weight (Maurya *et al.*, 2019), C is the mean concentration of heavy metals in crustacean and fish (mg/ kg dry weight), BW is the average body weight which is 60 kg for an adult and 30 kg for a child (Zhang *et al.*, 2018). ATn is average exposure duration to non-carcinogen pollutant ($\text{ED} \times 365 \text{ days/year}$) (ATn for adults = 21900 days), RfD is the oral reference dose for the non-carcinogen risk level of heavy metal exposure (mg/kg /day, 3×10^{-4} for As, 1×10^{-3} for Cd, 3×10^{-3} for Cr, 4×10^{-3} for Pb) according to USEPA (2000, 2016).

Target Cancer Risk (TR)

TR is used to estimate carcinogen risk level of consumption of heavy metal and it is calculated according to the Screening values for target analyses table provided by USEPA region III risk- based concentration table suggested by USEPA, 2011(Maurya *et al.*, 2019).

$$TR = \frac{EF \times ED \times IR \times Cf \times C \times CSF}{BW \times ATn}$$

Where C is the concentration of metal, CSF is Cancer slope factor for oral consumptions of metals (mg/kg bw/day) and CSF values for As, Cd, Cr and Pb is recommended by USEPA to determine the TR values of each metal in fish sample.

Hazard Index (HI)

HI= THQ (As) + THQ (Cd) + THQ (Cr) + THQ (Pd)
HI is the sum of the HQs. If $H < 1$ it is safe for human consumption $H > 1$ is considered hazardous according to USEPA, 2011.

Results and Discussion

Crude oil

Crude oil is a mixture of Hydrocarbons, Sulphur, Nitrogen and Metals (Yasin, 2013). Heavy metals present in the crude oil samples collected from the study area were analysed for the metals. Concentration of metals are found in the order $Cu < Cd < Pb < Cr < As < Ni$ and Nickel was found to be in largest quantity (10.2 mg/kg), followed by Arsenic (5 mg/kg) and Copper was found to be in least quantity (0.55 mg/kg) in the Crude oil samples. (Table 1 & Fig. 3.). Four metals: Lead (Pb), Arsenic (As), Chromium (Cr) and Cadmium (Cd) were chosen based on their abundance in the crude oil samples collected and their significance in relation to their carcinogenic effects noted from the review of

Table 1. Concentration of Heavy metals found in Oil Sludge

Heavy metals	Quantity present (mg/kg)
Arsenic	5
Cadmium	1.25
Chromium	3.4
Copper	0.55
Lead	2.5
Nickel	10.2

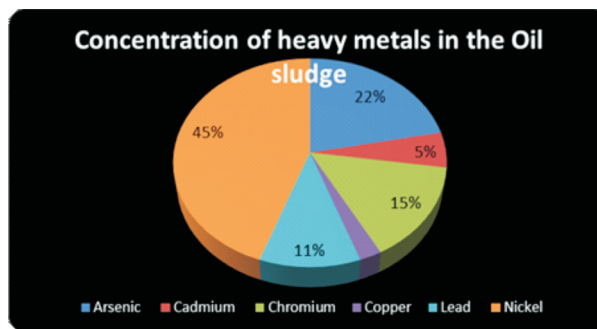


Fig. 3. Percentage of heavy metals present in the Oil sludge

literature. The measurements of metals that are present in the crude oil would largely represent the metals in the seawater.

Heavy metals in Water samples

Seawater samples collected from all the five locations during four seasons were analysed for the presence of selected heavy metals to establish the seasonal mean concentrations (Table 2), heavy metals present in the water samples represented in the decreasing order of contamination, $Pb > As > Cr > Cd$. The concentration of lead is found to be higher than all the four metals studied, in all the locations throughout all the seasons. As concentration found to be more in winter, concentration of Pb and Cr is seen high during Monsoon and Cd during summer. The highest concentration of Lead (23.88 mg/l) is found in Kovalam and the lowest (11.75 mg/l) in Pattinapakkam. The highest value of Arsenic (5.5 mg/l) was detected in Pattinapakkam and the lowest value (0.35 mg/l) in Thiruvanniyur, The highest level of cadmium (4.34 mg/l) is found in Ennore and the lowest (0.89 mg/l) in Thiruvanniyur. The highest concentration of Chromium (5.04 mg/l) is found in Kovalam and the lowest (0.78 mg/l) in Ennore. Detection of high concentration of metals during monsoon could be due to the runaway water from the industries near the sampling sites. Of all the 5 study locations, seawater from Kovalam, Pattinapakkam and Ennore coastal areas contained high concentrations of metals which shows high degree of metal contamination in these areas. This necessarily calls for the need for appropriately treating industrial effluents before being discharged into the sea.

Morphometric study

Morphometric measures of five species (*N. japonicus*,

Table 2. The concentration of heavy metals in the water samples

Seasons	Stations	Heavy metals			
		(concentration in mg/L)			
		Ar	Cd	Cr	Pb
Winter	1	5.02±0.04	0.05±0.02	0.32±0.02	12.56±0.03
	2	2.55±0.02	1.76±0.02	1.56±0.02	16.81±0.01
	3	5.5±0.01	2.2±0.02	1.55±0.02	15.51±0.01
	4	0.35±0.02	0.89±0.01	1.25±0.02	20.9±0.01
	5	4.89±0.02	3.34±0.02	2.12±0.02	20.94±0.02
Summer	1	3.31±0.01	2.75±0.03	1.26±0.02	13.23±0.02
	2	2.89±0.02	2.34±0.03	3.09±0.02	13.89±0.01
	3	3.56±0.02	1.56±0.02	0.8±0.01	17.41±0.01
	4	2.99±0.01	3.43±0.02	3.19±0.02	18.51±0.01
	5	5.15±0.02	3.56±0.03	4.11±0.01	20.51±0.01
Pre Monsoon	1	2.60±0.03	3.24±0.01	0.98±0.02	17.09±0.01
	2	2.78±0.01	1.89±0.01	0.78±0.01	13.21±0.01
	3	1.94±0.02	2.35±0.02	1.98±0.02	14.55±0.01
	4	2.13±0.02	3.11±0.01	2.45±0.02	19.23±0.02
	5	3.05±0.01	3.94±0.03	3.54±0.01	20.95±0.02
Monsoon	1	1.67±0.02	2.53±0.01	4.07±0.02	13.26±0.02
	2	2.55±0.02	2.78±0.02	2.65±0.01	12.7±0.01
	3	2.5±0.02	0.94±0.01	1.85±0.03	11.75±0.03
	4	0.94±0.01	2.03±0.01	1.56±0.02	15.5±0.01
	5	3.94±0.02	3.94±0.02	5.04±0.01	23.88±0.02

Legend: Stations – 1- Pulicut, 2- Ennore, 3- Pattinapakkam, 4- Thiruvannamiyur, 5- Kovalam.

R. kanagurta, *P. sanguinolentus*, *P. indicus*, *P. monodon*) collected for the study are represented in Table 3.

Heavy metals in fish and crustacean

Accumulated metal concentration in fish and crusta-

ceans are good ecological indicators as it reflects the metal contamination in their environment. The concentration in various organs (liver, intestine, gills, skin, gonads, and muscle) of *N. japonicus* and *R. kanagurta*, (Spawn, Gills and Muscle) of *P.*

Table 3. Morphometric measures and habitat of samples collected

Scientific Name	Common name	Inhabit stratum and distribution range	Feeding behaviour	Climate zone, depth	No. of samples	Length (cm) Mean(± SD)	Weight (g)
<i>Nemipterus japonicus</i>	Japanese Threadfin bream	Benthic	Carnivores	Tropical; depth range 5 - 80 m	25(5*5)	21.5±3.4	133.5±26.8
<i>Rastrelliger kanagurta</i>	Indian mackerel	Pelagic-neritic	Carnivores	Tropical; depth range 20 - 90 m	25(5*5)	27.6±4.5	187.7±43.7
<i>Portunus sanguinolentus</i>	Three spotted swimming crab	Benthic	Carnivores	depth of 30 m	25(5*5)	12.06±1.4	128.7±31.6
<i>Penaeus monodon</i>	Tiger prawn	Adult - Benthic	Omnivorous	Indo-West Pacific depths from 0 to 110 m.	25(5*5)	17.3±7.2 carapace (rostrum–telson)	41.3±7.2
<i>Penaeus indicus</i>	Indian prawn	Adult - Benthic	Omnivorous	Indo-West Pacific found at depths of 2 to 90 m.	25(5*5)	12.1±2.5 carapace (rostrum–telson)	14.3±5.5

sanguinolentus and (Carapace and muscle) of *P.indicus* and *P. monodon* is listed in the Tables 4, 5 and 6. The distribution of the heavy metal concentrations in the tissues of *N. japonicus*, *P. indicus*, *P.monodon* is in the order Pb> Cr> Cd > As, *R.kanagurta* is in the order Pb> As>Cr > Cd and

Table 4. Average concentration ($\mu\text{g/g}$ dry weight) of heavy metals (As, Cd, Cr & Pb) in tissues of Fish Species collected along the Chennai coast.

Metal	tissues	<i>N. japonicus</i>	<i>R. kanagurta</i>
As	Liver	3.63±0.8	6.79±2.2
	Gonad	3.04±1.4	5.41±1.7
	Gills	3.02±1.2	3.15±1.6
	Intestine	3.28±1.2	6.05±1.2
	Skin	2.31±0.7	4.13±0.8
Cd	Muscles	0.03±0.02	0.07±0.03
	Liver	3.89±0.7	5.26±1.7
	Gonad	3.13±0.4	4.48±2.5
	Gills	3.98±1.2	4.99±1.5
	Intestine	3.39±0.6	5.42±1.0
Cr	Skin	2.81±0.7	4.43±0.8
	Muscles	0.12±0.1	0.87±1.1
	Liver	5.72±1.3	5.92±2.4
	Gonad	5.29±1.2	5.49±2.4
	Gills	5.74±0.7	5.67±1.8
Pb	Intestine	5.18±1.3	5.68±2.1
	Skin	4.42±1.2	4.90±2.0
	Muscles	0.39±0.2	0.35±0.2
	Liver	15.55±3.1	16.95±4.8
	Gonad	14.84±2.4	18.00±3.6
	Gills	16.66±4.1	14.21±3.0
	Intestine	17.28±3.4	14.97±3.8
	Skin	9.44±2.4	15.11±3.6
	Muscles	0.59±0.1	1.72±1.8

Table 5. Average concentration ($\mu\text{g/g}$ dry weight) of heavy metals (As, Cd, Cr & Pb) in tissues of *P.sanguinolentus* collected along the Chennai coast.

Metals	Tissue	concentration
As	Spawn	6.06±1.8
	Gills	5.55±1.3
Cd	Muscle	0.87±1.4
	Spawn	4.96±2.2
Cr	Gills	4.42±2.4
	Muscle	1.89±2.8
Pb	Spawn	6.66±3.1
	Gills	6.01±2.9
	Muscle	1.64±2.7
	Spawn	16.44±6.2
	Gills	16.75±5
	Muscle	4.18±6.2

Table 6. Average concentration ($\mu\text{g/g}$ dry weight) of heavy metals (As, Cd, Cr & Pb) in muscle tissues of Shrimp species collected along the Chennai coast

Metals	As	Cd	Cr	Pb
<i>P. indicus</i>	0.02±0.01	0.15±0.05	0.36±0.2	0.62±0.2
<i>P. monodon</i>	0.02±0.03	0.14±0.05	0.37±0.2	0.69±0.09

P.sanguinolentus is in the order Pb> Cr > As> Cd. Tolerance level of heavy metals (As, Cr, Cd and Pb) in fish and Crustaceans for Human consumption represented by various organizations is represented in Table 7.

Table 7. Recommended values of heavy metals (As, Cr, Cd and Pb) in fish and Crustaceans for Human consumption.

Standard levels of metals	As	Cd	Cr	Pb
Tolerance level of heavy metals in fishes muscles (mg/kg) for consumption				
WHO-1989		1		2
FAO-2003		0.05		0.2
USEPA		2000		284
EC-2014		0.5		0.3
FSSAI 2015	76	0.3-0.5	12	0.3
Tolerance level of heavy metals in crustaceans muscles (mg/kg) for consumption				
EU-2006	5	0.5	0.5	0.5
USEPA, 2000				
RfDo(mg/kg-day)	0.0003	0.001	0.003	0.004

The mean concentration of Arsenic in *N. japonicus* ranged from (0.03±0.02- 3.63±0.8 $\mu\text{g/g}$) and in *R. kanagurta* the concentration ranged between (0.07 ±0.03- 6.79±2.2 $\mu\text{g/g}$). As accumulation in the tissues of *N. japonicus* is in the order (liver> intestine> gonads> gills> skin> muscle) and in *R. kanagurta* is in the order (liver> intestine> gonads> skin>gills> muscle) and recorded highest in the liver tissue and lowest in the muscle tissue of both the species. A higher range of As concentration 0.8-13 mg/kg was reported in three fish species (*Tenualosa ilisha*, *Pampus argentius*, *Cynoglossus lingua*) in the coastal area of Bangladesh (Raknuzzaman *et al.*, 2016). The results obtained in the present study show that the concentration of Arsenic is much higher than the value permissible by (USEPA, 2000), i.e. 1.2 $\mu\text{g/g}$, while it is well within the permissible range by FSSAI, 2015, i.e. 76 ppm. As concentration in *P.sanguinolentus* ranges between (0.87±1.4- 6.06±1.8

µg/g) and accumulation pattern in the tissues of *P.sanguinolentus* is in the order (spawn, > gills> muscle). The concentration of As in muscle tissue of *P.indicus* and *P.monodon* is (0.02±0.01 and 0.02±0.03 µg/g). The range of 0.3-2.5 mg/kg of Arsenic was reported in *P.indicus* in coastal areas of Bangladesh (Raknuzzaman *et al.*, 2016). European Union (EU), 2006 permitted 5µg/g of As in Crustaceans. Thus the As concentration in the gills and spawn of *P.sanguinolentus* exceeded the EU permissible limit and in the muscle tissue of *P. sanguinolentus*, *P.indicus* and *P. monodon* is well within the range. Arsenic ingestion has been linked to a multiple health problems and inorganic arsenic can have long-term consequences like cancer of skin, liver, gall bladders and lungs and damages to nerves system, respiratory tract and cardiovascular system (Baki *et al.*, 2018; Mandal and Suzuki, 2002) and acute cases causes Vomiting, anaemia and liver damages (Ahmeed *et al.*, 2015). The concentration of Cadmium was recorded highest in the spawn of *P. sanguinolentus* (4.96±2.2 µg/g), in fish the highest is recorded in gills of *R. kanagurta* (5.42±1.0 µg/g). The mean concentration of Cd ranged from (0.12±0.1-3.98±1.2 µg/g) in *N. japonicus* and (0.87 ±1.1-5.42±1.0 µg/g) in *R. kanagurta*. Accumulation pattern of Cd in the tissues of *N. japonicus* is in the order (gills>liver> intestine> gonads> skin> muscle) and in *R. kanagurta* is in the order (intestine>liver> gills>gonads> skin> muscle). Similar to the study, Rejomon *et al.*, 2010, reported Cd levels of 3.25-6.38 mg/kg in 5 fish species *Nemipterus japonicus*, *Lates calcarifer*, *Caranx melampygus*, *Rastrelliger kanagurta*, and *Cyanoglossus macrostomus* from coastal regions of Kochi and Mangalore, of which highest concentration is observed in *Nemipterus japonicus* and *Rastrelliger*. Similarly, Baki *et al.*, 2018 reported cadmium values 14.09-1.53 mg/kg, in few marine fishes, the highest value observed in *H. nigresceus*. The maximum cadmium level permissible in fish by FSSAI, 2015 is 0.3 ppm. The mean Cd concentration observed in *P. sanguinolentus* is (1.89 ±2.8-4.96±2.2 µg/g); (0.15± 0.05 and 0.14±0.05µg/g) in *P.indicus* and *P.monodon*, Raknuzzaman, 2016 reported (0.02-0.12mg/kg) of Cadmium in *P. indicus*. The values reported in this study are considerably higher than the value allowed by the European community legislation (EC, 2001), i.e. only up to 0.05 mg/kg ww in crustaceans. Cd even in the consumable muscle tissue of fish and crustaceans are higher than the FSSAI recommended values. Cadmium is capable of

producing chronic health effects even at low concentrations (Friberg *et al.*, 1971) and long-term exposure to cadmium can cause softening of bones, kidney failure, prostate cancer (Vannoort and Thomson, 2006; Gray *et al.*, 2005). The mean Chromium concentration ranged from (0.39 ±0.2 to 5.74±0.7 µg/g) in *N. japonicus*, (0.35 ±0.2 to 5.92±2.4 µg/g) in *R. kanagurta*, (1.64 ±2.7 to 6.66±3.1 µg/g) in *P. sanguinolentus* which is very close to the permissible concentration (12 ppm) as per FSSAI, 2015. Cr accumulation in the tissues of *N. japonicus* is in the order (gills>liver>gonads>intestine> skin> muscle), in *R. kanagurta* is in the order (liver> intestine> gills>gonads> skin> muscle) and (spawn> gills > muscle) in *P. sanguinolentus*. The highest load of Cr is observed (6.66±3.1 µg/g) in the spawn of *P. sanguinolentus* followed by liver tissue (5.92±2.4 µg/g) of *R. kanagurta* and gills (5.74 0.7 µg/g) of *N. japonicus*. Baki *et al.*, 2018 reported Cr levels of 1.87-0.1909mg/kg in fish, Raknuzzaman *et al.*, 2016 reported Cd levels of 0.15-2.2mg/kg ww. in fish and 1.1- 3.2mg/kg in *P. indicus*. The observed concentration of Cr in *P. indicus* and *P. monodon* ranged between (0.36± 0.2 and 0.37±0.2 µg/g). Chronic exposure of Cr causes adverse health conditions including allergic responses, anaemia, burns and blisters, particularly in the stomach and small intestine, and damage to sperm and the male reproductive system (Hossini *et al.*, 2022). The gonads of *R. kanagurta* collected from Pattinapakkam showed highest concentration of Lead. The mean Pb concentration ranged from (1.72±1.8 to 18±3.6 µg/g) in *N. japonicus* and (0.59 ±0.1- 17.28±3.4 µg/g) in *R.kanagurta*. Accumulation pattern of Pb in the tissues of *N. japonicus* is in the order (intestine> gills>liver> gonads> skin> muscle) and in *R. kanagurta* is in the order (gonads> liver> skin>intestine> gills> muscle). The observed concentration of Pb is higher than the permissible concentration (0.3 ppm) of Pb as per FSSAI (2015). Baki *et al.*, 2018 studied 8 mostly consumed fish sp (*L. fasciatus*, *R. kanagurta*, *H. nigrescens*, *P. cuneatus*, *P. annularis*, *S. rubrum*, *T. jarbua* and *L. atkinson*, in Saint Martin Island, Bangladesh of which Pb in fish is found to be 0.1209 - 8.92 mg/kg ww. In a similar study Rejomon *et al.*, 2010, also reported 0.45- 0.56 ppm of which highest is reported in *Nemipterus japonicus* and *Rastrelliger kanagurta* in Kochi and in Mangalore 0.23- 0.37ppm highest in *Rastrelliger kanagurta*. The mean Pb concentration observed in *P. sanguinolentus* is (4.18 ±6.2 to 16.44±6.2 µg/g); (0.62± 0.2 and 0.69±0.09 µg/g) in *P. indicus* and *P.monodon*,

EU, 2006 permissible limit Pb in crustaceans for consumption is 0.5mg/kg, in the present study muscle tissues of crustaceans showed values higher than the EU, 2006 permissible limit. Raknuzzaman *et al.*, 2016 reported values of 0.1- 0.38 in *P.indicus*. Lead is a non-essential element, high levels of lead are known to cause blood pressure and cardiovascular disease, nervous system, kidney and reproductive system disorders in adults and cognitive developments in children (Baki *et al.*, 2018; Wang *et al.*, 2019).

The high concentration of metal in fish is observed in liver, gills and intestine, this could be due to the reason that livers are the primary organ for detoxification (Brusle and Andadon, 1996) and gills are the first organs to be impacted by environmental changes while being in constant contact with

water (Ahmed *et al.*, 2015; Kumar *et al.*, 2014). In crab, all the metals except As in muscle tissues exceeded FSSAI recommended value for consumption for crustaceans (FSSAI, 2015). High loads of metals observed in Crabs owes to its benthic habitat, feeding on detritus and benthic prey, exposure to direct contact with sediments which leads to accumulating more heavy metal (Zhao *et al.*, 2012). Different species showed different accumulation patterns (Fig. 4-1 to 4.5), the differences in the concentration of heavy metals may be articulated to its different physiological and biochemical characters like absorptions, excretion and detoxification by the organisms (Bryan, 1971). Of all the studied metals Pb is found to be more readily accumulated and found in high concentration than the FSSAI, 2015 recom-

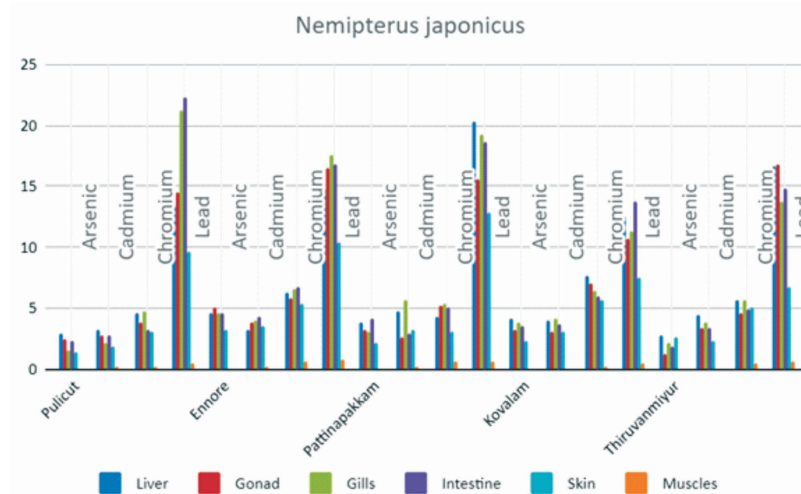


Fig. 4.1. Concentration of Heavy metals in tissues of *N. japonicas*

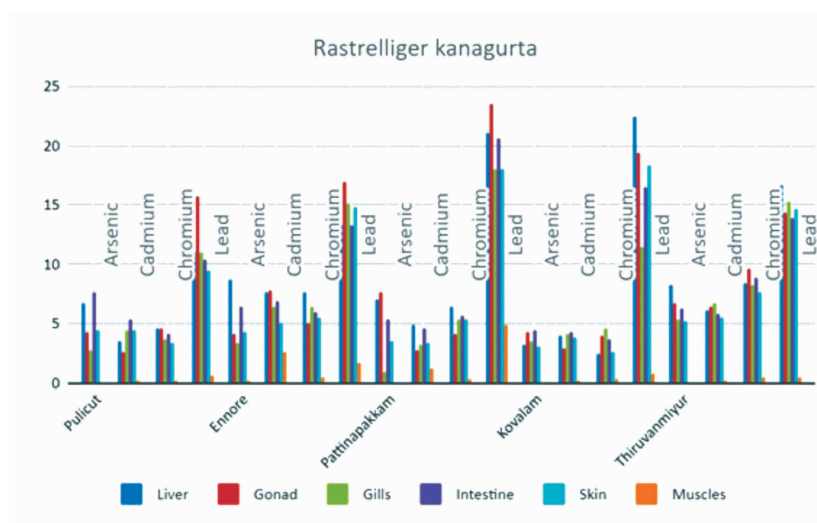


Fig. 4.2. Concentration of Heavy metals in tissues of *R.kanagurta*

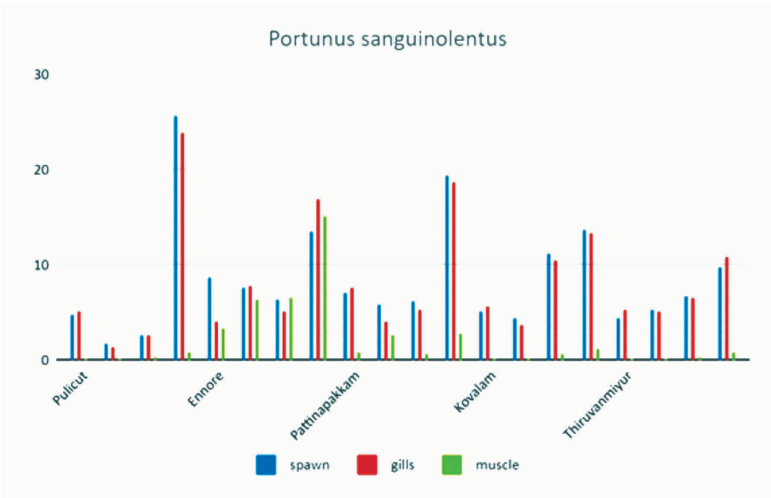


Fig. 4.3. Concentration of Heavy metals in tissues of *P. sanguinolentus*

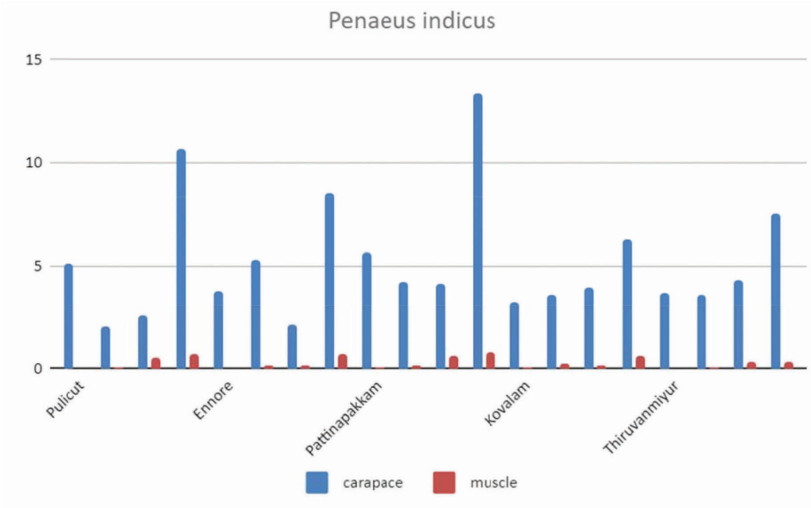


Fig. 4.4. Concentration of Heavy metals in tissues of *Penaeus indicus*

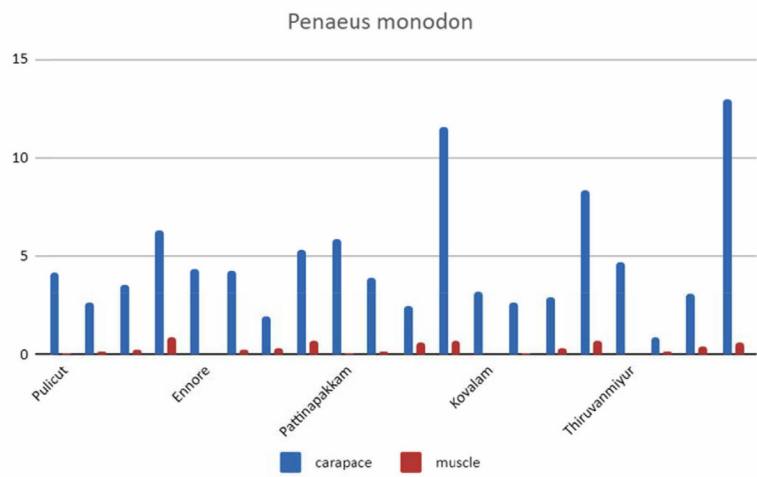


Fig. 4.5. Concentration of Heavy metals in tissues of *Penaeus monodon*

mended value for consumption. This could be attributed to the tendency of the metal to accumulate. The possible sources of Pb in the environment include gasoline, boats, batteries, paint and book printing industries (Wani *et al.*, 2015).

Bioconcentration Factor (BCFs)

In the present study heavy metal concentration in different tissues of various organs (liver, intestine, gills, skin, gonads, and muscle) of fish and crustaceans showed bioaccumulation of heavy metals Fig (5.1 to 5.4). The highest BCF concentration was recorded for Cr and lowest for Pb and the results depicted a decreasing trend of $Cr > Cd > As > Pb$. The liver of fish showed higher BCF, followed by gonads, intestine, gills and skin. BCF of muscle was recorded much lower than that of the other tissues. Accumulation of As is found more in liver, Cd and Cr in Intestine and gills and Pb more in intestine. In crab, BCF was in the order Spawn > gills > muscle. The BCF of muscle of shrimps was comparatively lower than that of the other species studied.

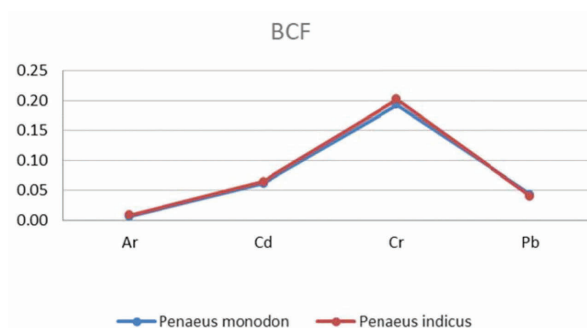


Fig. 5.4. Bio- concentration factor of heavy metals (As, Cd, Cr, Pb) L/kg in muscle tissues of *P. monodon* & *P. indicus*

Health risk assessment of the trace metals in fish and crustaceans

Humans are exposed to toxic metals predominantly through diet. Fish forms the chief source of food to a peninsular country like India and dispenses exposure to toxic metals as fish and crustaceans have a strong potential to accumulate metals (Aytakin *et al.*, 2019). Accumulation of heavy metals in fish could

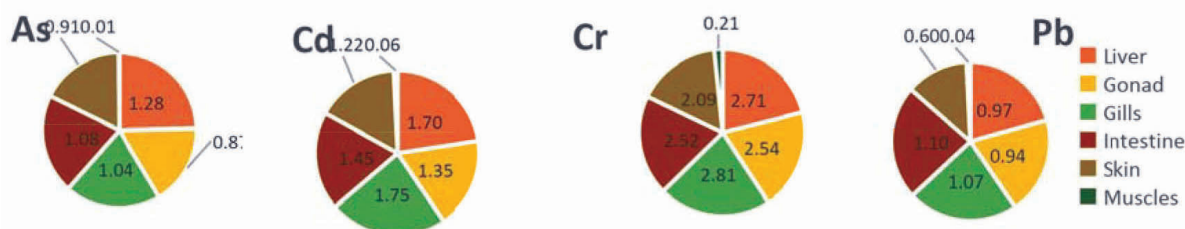


Fig. 5.1. Bio- concentration factor of heavy metals (As, Cd, Cr, Pb) L/kg in different tissues of *N. japonicus*

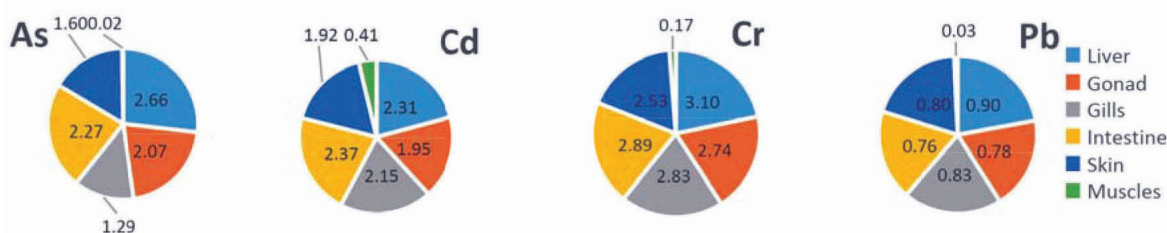


Fig. 5.2. Bio- concentration factor of heavy metals (As, Cd, Cr, Pb) L/kg in different tissues of *R. kanagurta*

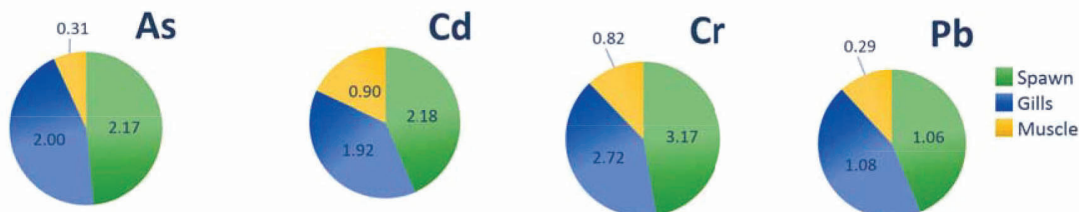


Fig. 5.3. Bio- concentration factor of heavy metals (As, Cd, Cr, Pb) L/kg in different tissues of *P. sanguinolentus*.

affect the health conditions of consumers on consuming fish on a daily basis (Maurya *et al.*, 2019). Therefore, it is important to calculate Health risk assessment for fishes and crustaceans from polluted coastal areas. Muscle tissues of fish and Crustaceans are explored more than other organs because it is the part consumed by humans (Aytekin *et al.*, 2019).

The Estimated Daily Intake (EDI) determines the toxicity of heavy metal in humans. The daily intake of As, Cd, Cr and Pb through the muscle tissue of the studied species were examined (Table 8) and compared with the recommended values to check whether the consumption of fish and crustaceans contaminated with metal from Chennai coasts were safe for human consumption. The highest recorded EDI values are in the order Pb > Cd > Cr > As with their values 4.53, 2.42, 0.59 and 0.10 mg/day/person respectively. The toxicity risk is considered low if the derived EDI values are equal to or less than the RfD values (Baki *et al.*, 2018). The higher EDI values than that recommended by USEPA denote that there is a higher possibility of health impact associated with the consumption of heavy metals from the contaminated sites.

Target Hazard Quotient (THQ) and Hazard Index (HI) estimated for the consumption of fish and crustaceans for all the four metals studied are represented in Table 9. The THQ and HI proposed by USEPA has been considered as one of the suitable

indicators for assessing the risk of consuming the metals through contaminated seafood. (Li *et al.*, 2013; USEPA, 2015). If the THQ-average of each metal is less than 1, it is suggested that people would not experience significant health risk through consumption of fish (Yi *et al.*, 2017). The highest THQ average was observed for Cd in *P. sanguinolentus* was from Ennore and Pattinapakkam, *R.kanagurta* from Pattinapakkam followed by Pb in *P. sanguinolentus* from Pattinapakkam, Ennore and Kovalam, *R.kanagurta* from Ennore and Pattinapakkam and As in *R.kanagurta* from Ennore and Pattinapakkam. THQ values of As, Cd and Pb in *P. sanguinolentus* from Pattinapakkam, Ennore and Kovalam, As, Cd and Pb in *R. kanagurta* from Ennore and Pattinapakkam were above 1. The values stipulate that prolonged consumption of these two species from the Ennore, Kovalam and Pattinapakkam coast may be potentially hazardous to human health. THQ values of *N. japonicus*, *P. indicus* and *P. monodon* were below the threshold limit and thus indicate no significant risk to human health. HI was above 1 for all the species studied, which indicates that the consumption of seafoods from these coastal areas are harmful to human health. Though there is no potential non carcinogenic health hazard through consuming single metal in these species, continuous excessive consumption may result in a certain degree of adverse health ef-

Table 8. EDI of Fish and Crustaceans collected along the Chennai coasts.

		Pulicut	Ennore	Pattinapakkam	Kovalam	Thiruvanniyur
N.japonicus	As	0.03	0.06	0.04	0.02	0.02
	Cd	0.14	0.15	0.20	0.04	0.07
	Cr	0.20	0.57	0.52	0.59	0.39
	Pb	0.47	0.68	0.54	0.44	0.61
R. kanagurta	As	0.05	0.10	0.08	0.05	0.04
	Cd	0.17	2.37	1.22	0.16	0.11
	Cr	0.12	0.39	0.36	0.26	0.49
	Pb	0.63	1.64	4.53	0.67	0.51
P.sanguinolentus	As	0.04	0.06	0.07	0.05	0.06
	Cd	0.08	0.95	2.42	0.12	0.18
	Cr	0.20	0.59	0.45	0.57	0.25
	Pb	0.83	3.51	2.61	1.13	0.76
P.monodon	As	0.01	0.02	0.04	0.03	0.02
	Cd	0.10	0.18	0.15	0.20	0.08
	Cr	0.52	0.13	0.55	0.15	0.34
	Pb	0.69	0.61	0.73	0.55	0.31
P.indicus	As	0.02	0.01	0.05	0.01	0.01
	Cd	0.11	0.20	0.10	0.08	0.16
	Cr	0.20	0.33	0.57	0.33	0.34
	Pb	0.77	0.60	0.67	0.61	0.54

Table 9. Target hazard quotient (THQ) of Fish and Crustaceans collected along the Chennai coasts.

Species		Pulicat	Ennore	Pattinapakkam	Kovalam	Thiruvanmiyur
<i>N. japonicus</i>	As	0.4464	0.8928	0.5952	0.2976	0.2976
	Cd	0.6696	0.7142	0.9821	0.1786	0.3125
	Cr	0.3274	0.9077	0.8333	0.9374	0.6250
	Pb	0.5692	0.8147	0.6473	0.5245	0.7366
<i>R. kanagurta</i>	As	0.7440	1.6368	1.3392	0.7440	0.5952
	Cd	0.8035	11.3832	5.8478	0.7589	0.5357
	Cr	0.1934	0.6250	0.5803	0.4166	0.7886
	Pb	0.7589	1.9642	5.4349	0.8035	0.6138
<i>P. sanguinolentus</i>	As	0.5952	0.8928	1.0416	0.7440	0.8928
	Cd	0.4018	4.5533	11.6064	0.5803	0.8482
	Cr	0.3274	0.9374	0.7142	0.9077	0.4018
	Pb	0.9932	4.2073	3.1360	1.3504	0.9151
<i>P. monodon</i>	As	0.1488	0.2976	0.5952	0.4464	0.2976
	Cd	0.4910	0.8482	0.7142	0.9821	0.4018
	Cr	0.8333	0.2083	0.8779	0.2381	0.5506
	Pb	0.8258	0.7366	0.8705	0.6584	0.3683
<i>P. indicus</i>	As	0.2976	0.1488	0.7440	0.1488	0.1488
	Cd	0.5357	0.9374	0.4910	0.4018	0.7589
	Cr	0.3125	0.5357	0.9077	0.5208	0.5506
	Pb	0.9263	0.7142	0.8035	0.7366	0.6473

Table 10. Target Risk Factor (TR) of Fish and Crustaceans collected along the Chennai coasts.

Species		Pulicat	Ennore	Pattinapakkam	Kovalam	Thiruvanmiyur
<i>N. japonicus</i>	As	1.72E-04	3.44E-04	2.30E-04	1.15E-04	1.15E-04
	Cd	8.61E-03	9.18E-03	1.26E-02	2.30E-03	4.02E-03
	Cr	4.21E-04	1.17E-03	1.07E-03	1.21E-03	8.04E-04
	Pb	1.66E-05	2.37E-05	1.89E-05	1.53E-05	2.15E-05
<i>R. kanagurta</i>	As	2.87E-04	6.31E-04	5.17E-04	2.87E-04	2.30E-04
	Cd	1.03E-02	1.46E-01	7.52E-02	9.76E-03	6.89E-03
	Cr	2.49E-04	8.04E-04	7.46E-04	5.36E-04	1.01E-03
	Pb	2.21E-05	5.72E-05	1.58E-04	2.34E-05	1.79E-05
<i>P. sanguinolentus</i>	As	2.30E-04	3.44E-04	4.02E-04	2.87E-04	3.44E-04
	Cd	5.17E-03	5.85E-02	1.49E-01	1.49E-01	1.09E-02
	Cr	4.21E-04	1.21E-03	9.18E-04	1.17E-03	5.17E-04
	Pb	2.89E-05	1.23E-04	9.14E-05	3.94E-05	2.67E-05
<i>P. monodon</i>	As	5.74E-05	1.15E-04	2.30E-04	1.72E-04	1.15E-04
	Cd	6.31E-03	1.09E-02	9.18E-03	1.26E-02	5.17E-03
	Cr	1.07E-03	2.68E-04	1.13E-03	3.06E-04	7.08E-04
	Pb	2.41E-05	2.15E-05	2.54E-05	1.92E-05	1.07E-05
<i>P. indicus</i>	As	1.15E-04	5.74E-05	2.87E-04	5.74E-05	5.74E-05
	Cd	6.89E-03	1.21E-02	6.31E-03	5.17E-03	9.76E-03
	Cr	4.02E-04	6.89E-04	1.17E-03	6.70E-04	7.08E-04
	Pb	2.70E-05	2.08E-05	2.34E-05	2.15E-05	1.89E-05

fects among local people.

The lifetime Target Cancer risk exposure to As, Cd, Cr and Pb through consumption of sea foods is listed in the Table 10. Cancer risks lying between 10^{-4} and 10^{-6} are considered acceptable range, the cancer risks are higher when it is above 10^{-4} , cancer risk below 10^{-6} are negligible (Varol *et al.*, 2017; USEPA, 2000). TR values of As and Pb are within acceptable dose range in all the species studied. Cd showed higher risk values than other metals. Cancer risk values for Cd in all five species studied (*Nemipterus japonicus*, *Rastrelliger kanagurta*, *Portunus sanguinolentus*, *Penaeus indicus* and *Penaeus monodon*) was higher than the acceptable range in all the studied locations. Cr in the species *N. japonicus*, *R. kanagurta* from Ennore, Pulicat and Thiruvannamiyur and in *P. sanguinolentus* from Ennore and Kovalam, *P. indicus* and *P. monodon* from Pulicat and Pattinapakkam is also close to the cancer risk value range, which indicates that the local consumers are at carcinogenic risk associated with Cd and Cr posed from consumption of fish and crustaceans' species.

Deleterious effects of Arsenic, Cadmium, Chromium and lead on human health even at a marginal concentration (Ahmed *et al.*, 2015; Zhang *et al.*, 2018) are widely known. Differences in metal accumulation in different species could be due to differences in their ecological niche, physiology, habitat and feeding. Invertebrates accumulate heavy metals in their body organs in different concentration, this is due to various factors like their feeding habitat, stratum inhabited, sexual maturity, age, metabolism, absorption, excretion and efficiency of the invertebrate in regulating and detoxification of the inculcated heavy metals (Bryan, 1971; Eisler, 1981; Baki *et al.*, 2018). Besides all the above, food source, skin, and breathing contribute more to heavy metal bioaccumulation and bio transmission in shrimps, crab (Baki *et al.*, 2018).

Conclusion

The present study is highly significant as the study area is a center of various human activities, has ports, harbours, industries of various kinds, places of historic importance, tourist beaches, etc. and has coastal habitats like estuaries, lagoons, etc. The Nearshore ecosystem is one of the most important ecosystems on the earth, providing many ecological services, such as food supply and human activity

space (Arikibe and Prasad, 2020). The rapid economic development and increased human activities in coastal areas has led to more significant impact on marine bio environment, especially the coastal areas, which are the main sink of anthropogenic pollutants, and the environment is facing greater pressure (Gao *et al.*, 2014; Zhang and Chen, 2019). Heavy metals in the fish and crustaceans' tissues widely varied among species and within the tissues of species. Bioaccumulation is more in the liver and gonads followed by gills, skin, intestine and muscle. The study revealed that all the five species studied pose carcinogenic risk from Cd and Cr consumption. These results can be used to provide baseline information of human health risk associated with consumption of fish. This tendency of bioaccumulation may be inferred as the possibility of higher absorption in higher concentration, influenced by various other factors. This risk factor highlights the need to minimize the heavy metal pollution caused by various identified sources such as industries, fertilizers factories, paint factories, dye factories, etc. Many studies across the world have reported high concentration of heavy metals in seafoods and water. Application of effective preventive measures and appropriate strategies to minimize heavy metal pollution can be identified, developed and adopted or integrated into coastal management practices. This study depicts a high risk factor which is close to the threshold value, in consumption of fish due to bioaccumulation of heavy metals in the environment. It necessarily calls for periodical monitoring of levels of heavy metals and updating the standards for food exports and imports as heavy metals contaminated foods are related to carcinogenic health effects.

Conflict of Interest - None

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