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# Seasonal investigation of heavy metals in marine fishes captured from the Bay of Bengal and the implications for human health risk assessment

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ABSTRACT

To investigate the seasonal contamination levels and to evaluate the potential human health risks, ten heavy metals (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Se, and Zn) were measured in ten different marine fish species from the Bay of Bengal during four seasons. The metal concentrations varied significantly ($p < 0.05$) among the fish species with maximum and minimum accumulation of Zn (46.47 μg/g) and Cd (0.25 μg/g), respectively. The fishes captured during summer accumulated a higher amount of metals relative to other seasons, which was attributed to a higher influx of agricultural waste, sewage and sludge by heavy rainfall and floods. According to estimated daily intake (EDI), target hazard quotient (THQ), total target hazard quotient (TTHQ), and the permissible safety limits prescribed by various agencies, consumption of the examined fish species should be considered as safe for human health. However, the estimation of carcinogenic risk (CR > $10^{-5}$) due to exposure to arsenic indicated that consumers remain at risk of cancer.



## 1. Introduction

Global aquatic environments are being heavily polluted by various contaminants, such as heavy metals (Rahman, Molla, Saha, & Rahman, 2012; and reference therein), polycyclic aromatic hydrocarbons (PAHs) (Y. Li et al., 2015; and reference therein), polychlorinated biphenyls (PCBs) (Hosseini et al., 2008; Zhang, Li, Wang, Zhang, Zhang, Zhang, et al., 2014 and reference therein), with rapid economic development and population growth. Pollution from the heavy metals (e.g., Cd, Cr, Pb etc.) has become a serious issue due to their stable, persistent, and nonbiodegradable nature (Saha & Zaman, 2013). Although the metals are naturally occurring constituents in the environment, their concentrations can be exacerbated by anthropogenic activities such as rapid industrialization and urbanization, massive land use changes and associated enhanced terrestrial runoff (Rahman et al., 2012). Another important source of anthropogenic pollution around Chittagong coastal area in Bangladesh is ship breaking activities began since 1969. Various refuse and disposable materials are being discharged and spilled from scrapped ships, and often get mix with the beach soil and sea water. Under certain conditions, the heavy metals can be bioaccumulated in aquatic organisms (e.g., fish) from the surrounding environments and/or bioamplified to hazardous levels via dietary exposure. Subsequently, these contaminants migrate to the human body through diet and results in various adverse health effects such as impaired kidney function, poor reproductive capacity, liver damage, skin and bladder cancer, and even death (Wei, Zhang, Zhang, Tu, & Luo, 2014). However, there are some essential heavy metals, such as Fe, Zn, and Cu, which are required for both aquatic organisms and human body. But prolonged exposure to the excess level of these metals could be toxic. On the contrary, there are some nonessential toxic metals, such as As, Cd, and Pb, with no known potential benefits for a human being (Wei et al., 2014).

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Food consumption is the main pathway (accounting for > 90%) for human exposure to heavy metals compared to other routes such as inhalation and dermal contact. Among various foodstuffs, fish is widely consumed and a main source of nutrition in many coastal communities. It contributes to a healthy diet, providing high-value amino acids, nutrients and essential omega-3 fatty acids. Although fish is highly nutritious, their higher consumption rate can have significant deleterious effects on human health because of bioaccumulated toxic metals beyond the safe limits. To date, the balance between benefits and risks has been poorly understood. However, fishes are often treated as the most suitable bioindicator in aquatic ecosystem since they occupy high trophic level (Rahman et al., 2012). Many researchers have made efforts to determine the heavy metal concentrations in fish to investigate the eco-environments and potential health risks to the consumers (Ahmed et al., 2015; Copat et al., 2013; P.; Li, Zhang, Xie, Liu, Liang, Ren, et al., 2015; Mendil, Demirci, Tuzen, & Soylak, 2010; Saha & Zaman, 2013; Wei et al., 2014).

In recent years, the quantification of risk has become important due to the fact that exceedance of recommended levels of contaminants prescribed by various regulatory bodies does not always represent a risk for the human health. The target hazard quotient (THQ) and/or total target hazard quotient (TTHQ) set by US Environmental Protection Agency (USEPA, 2000) are commonly used to evaluate the potential non-carcinogenic health risks associated with variety of metals through fish consumption (Copat et al., 2013; Saha & Zaman, 2013). The United States Environmental Protection Agency also provided cancer slope factor for arsenic, to determine the carcinogenic risk (CR) over a lifetime exposure to arsenic.

Bangladesh is one of the coastal countries of the Bay of Bengal, and 37 to 38 million people are living in the coastal zone. The country is located between 20°34′ to 26°38′ north latitude and 88°01′ to 92°42′ east longitude. Fishery resources play a vital role in the economy of Bangladesh (4.57% of GDP in 2008–09) and it is the second most important source of foreign exchange earnings (5.71%). The national and international demand of Bangladeshi fishes is increasing gradually. Recently, CBI (CBI, 2012) reported that in the period of 2002–2011, import of marine fish by the European Union (EU) has been increased from US$ 150 m to US$ 360 m. Thus, heavy metal concentrations in the seafood deserves attention and continuous investigation not only for their potential ecological impacts but for health risks of the population in Bangladesh and other importer countries.

The objectives of this study are (i) to report on the seasonal fluctuation of heavy metal concentrations (As, Cd, Co, Cr, Cu, Mn, Ni, Pb, Se, and Zn) in various fish species captured from Chittagong coastal zone, and (ii) to quantify the non-carcinogenic and carcinogenic human health risks associated with these metals.

## 2. Materials and methods

### 2.1. Sample collection and preservation

Ten fish species of *Lates calcarifer* (Bhetki), *Pangasius pangasius* (Pangas), *Polynemus indicus* (Lakhua), *Ilisha megaloptera* (Choukya), *Arius cruciger* (Rita), *Pampus chinensis* (Rupchanda), *Setipinna phasa* (Phasa), *Scomberomorus guttatum* (Maitya), *Cirrhina reba* (Beta) and *Arius arius* (Kata mach) were procured from fishermen while they were fishing nearshore sites at Cox's Bazar, Chittagong (Fig. 1) during spring, summer, autumn and winter. Samples were collected in triplicate and in total, 120 individuals (i.e., 30 samples in each season) representing 10 different species were collected. Immediately after collection, fishes were washed with fresh water to remove the mud or other fouling substances, and were wrapped in polyethylene bags to transport into the analytical chemistry laboratory of Institute of Nuclear Science and Technology, Bangladesh Atomic Energy Commission (BAEC). After transportation to the laboratory, the muscle tissue of each sample was removed and chopped into pieces with the aid of a steam cleaned stainless steel knife. The samples were then air dried to remove the extra water and subsequently, ovendried to a constant weight. Finally, the dried samples were ground, sieved, and stored in clean and dry airtight plastic vials inside desiccators for succeeding uses.

### 2.2. Sample digestion

For metal analysis, 0.5 g of each powdered sample was digested with 2.5 ml of conc. $H_2SO_4$ and 4.0 ml of conc. $HNO_3$. When initial vigorous reaction subsided, the mixture was heated slowly on an oil bath by the addition of 3/4 drops of $H_2O_2$. This step was repeated till the solution became clear. Subsequently, the solution was heated for additional 20 min at about 150 °C and allowed to cool to room temperature. The digested fish sample was then diluted to a total volume of 50 ml with double distilled water. Thereafter, the diluted solution was filtered and stored in 50-ml polypropylene tubes.

### 2.3. Analytical methods and quality control

Concentration of Cd, Co, Cr, Cu, Mn, Ni, Pb, and Zn in the digested solutions was analyzed by atomic absorption spectrophotometer (AAS) (Model AA-6800, Shimadzu Corporation, Japan) using air-acetylene flame with digital read out system. However, hydride vapor generators (HVG) were used along with the flame AAS (FAAS) systems to determine As and Se concentrations. The instrument calibration standards were made by diluting standards (1000 mg/l) supplied by Wako Pure Chemical Industry Ltd., Japan.

The accuracy of the method was evaluated by analyzing blanks and a certified reference material MA-A-2 [fish-flesh standard from International Atomic Energy Agency (IAEA), Vienna] by the same procedure used for fish samples. Mean recoveries of the analyzed metals were between 95 and 104%, indicating a good agreement between certified and measured values (Table S1).

### 2.4. Statistical analysis

Analysis of variance (ANOVA) test was conducted to investigate the effect of different fish species and seasons on the variation in metal concentrations. In order to quantify the seasonal effects on the variability of metals, regression analyses were performed using dummy variables for the seasons (winter as reference). In all cases, the level of significance was set at a 95% (i.e., $\alpha = 0.05$). The data were analyzed statistically using IBM-SPSS Statistics (Version 21) for windows (IBM, USA).

### 2.5. Human health risk assessment

#### 2.5.1. Estimated daily intake (EDI)

The estimated daily intake (EDI) for each analyzed heavy metal was calculated in the following way (Saha & Zaman, 2013):

$$EDI = \frac{E_F \times E_D \times F_{IR} \times C_f \times C_m}{W_{AB} \times T_A} \times 10^{-3} \quad (1)$$

Where $E_F$ is the exposure frequency (365 days/year). $E_D$ is the exposure duration, equivalent to an average life time of the Bangladeshi (i.e., 60 years). $F_{IR}$ is the ingestion rate (g/person/day) of fish tissue. The fish production in Bangladesh was unable to keep patch with high population growth and as a result, per capita