



# Health Risk Assessment of Heavy Metals Through Six Common Spices of Mohanpur Upazila of Rajshahi District, Bangladesh

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## Abstract

Six types of spices samples (viz. Capsicum annum, Allium cepa, Curcuma longa, Zingiber officinale, Allium sativum and Coriandrum sativum) were collected from Mohanpur Upazila of Rajshahi district. For the determination of heavy metals (Pb, Cd, Cr and As) concentrations and human health risks, the spice samples were digested in a mixture of HNO<sub>3</sub> and H<sub>2</sub>O<sub>2</sub>. It is observed that the average concentrations of Pb, Cd, Cr and As are varied from 0.8504 mg/kg to 1.9694 mg/kg, 0.1522 mg/kg to 1.2173 mg/kg, 0.7172 mg/kg to 1.4760 mg/kg and 0.2285 mg/kg to 4.0157 mg/kg respectively. The highest concentration of As 4.0157 mg/kg were investigated in Green Chili. Results indicate that Pb, Cd, Cr and As content in all spice samples are within the International standards limit. The MPI (metal pollution index) value for Green chili, Onion, Turmeric, Ginger, Coriander and Garlic is generally low indicated heavy metal contamination and has no significant threat of negative impact on human health. THQ values of Pb, Cd, Cr and 66.67% of As in all spices were less than 1. About 66.67% of the HI values of the samples under study were less than 1. They indicate that the consumers will not experience potential health risks from the intake of individual metals through spices consumption. However, a continuous monitoring should be carried on.

**Keywords:** Heavy Metal; Spices; Health Risk; Oral Reference Doses

## Introduction

A spice is a dried seed, fruit, root, bark, or vegetable substance primarily used for flavouring, colouring or preserving food. Sometimes a spice is used to hide other flavours. Many spices have antimicrobial properties. This may explain why spices are more commonly used in warmer climates, which have more infectious diseases, and why use of spices is especially prominent in meats, which is particularly susceptible to spoiling [1]. Natural food spices such as pepper and mustard have been reported to contain significant quantities of some trace metals [2]. Spices have played an important role in the history of civilization, exploration and commerce as these had a universal acceptance as condiment and flavours in human diet as well as in treatment of ailments. There are evidence of plant derived aromatic compounds especially

spices being used by almost all ancient civilizations, the Indian, the Egyptian, the Babylonian, the Persian, the Jews, the Chinese, the Greek and the Roman [3]. Many common spices have outstanding antimicrobial effects. On the other hand, the process of preparation and handling can make them a source of food poisoning [4]. Moreover, in the last three decades, mainly because of their medicinal values, the use of spices has increased markedly in most regions of the world. Several researches have shown that heavy metals could be present in spices and the addition of contaminated spices to food may result in accumulation of these metals in human organs. Heavy metals above the permissible levels affect human health and may result in illness to human foetus, abortion and preterm labour, and mental retardation to children. Adults also may experience high blood pressure, fatigue kidney and neurological

disorder [5]. Due to the significant amount of spices consumed, it is important to know the toxic metal contents in these spices. Natural spices (ginger, garlic, onion and locust beans) and processed spices (curry, thyme, nutmeg and beef spicy) are therefore assessed for lead, cadmium, copper and iron contents and possible health risk. Therefore, the main objective of this study is to determine the level of selected metals and to evaluate the potential health risk of these metals through the consumption of spices (green chili, onion, turmeric, ginger, garlic and coriander) which are widely grown in Mohanpur Upazila of Rajshahi district.

## Materials and Methods

### Description of study area

Mohanpur is located at 24.5639°N 88.6500°E. It is 25 km away from Rajshahi district headquarter. It has 43984 households and a total area of 162.65 km<sup>2</sup>. The Postal Code is 6220. It is surrounded by Tanore Upazila of Rajshahi District and Manda Upazila of Naogaon District to the north, Bagmara Upazila to the east, Paba Upazila and Durgapur Upazila to the south, Tanore Upazila to the west. Mohanpur Thana was formed in 1917 and it was turned into an upazila in 1983. (Figure 1,2) (Table 1)



Figure 1: Study area of Mohanpur Upazila in Rajshahi District map.



Green Chili (*Capsicum annuum*)



Onion (*Allium cepa*)



Turmeric (*Curcuma longa*)



Ginger (*Zingiber officinale*)



Garlic (*Allium sativum*)



Coriander (*Coriandrum sativum*)

FIGURE 2: PHOTOGRAPH OF SPICES.

**Table 1:** Local names, English names, scientific names and family of analyzed spice samples.

Local name (Bangla)	English name	Scientific name	Family
Kacha Morich	Green Chili	Capsicum annuum	Solanaceae
Payaj	Onion	Allium cepa	Amaryllidaceae
Holud	Turmeric	Curcuma longa	Zingiberaceae
Aadah	Ginger	Zingiber officinale	Zingiberaceae
Roshun	Garlic	Allium sativum	Amaryllidaceae
Dhoniah	Coriander	Coriandrum sativum	Apiaceae

## Collection and preparation of spices samples

Six (6) different species samples Figure 1 were collected from the field around the Mohanpur Upazila area. A detailed description (English name, scientific name, local name, types of spices etc.) has been presented in Table 1. The samples were thoroughly cleaned first under tap water to remove dust, dirt, possible parasites or the eggs and then washed again with double distilled water (DDW). Non-edible parts were removed according to common household practices and the edible portion was chopped into small pieces. The small pieces of samples were dried in sun. Then it dried an air-dried oven at 90-95°C until a constant weight was obtained. The dried samples were crushed in a grind machine and passed through a 85 mesh sieve. The samples were homogenized by grinding and stored in desiccators in the dark until analysis, usually with the same week [6].

## Analysis of spices sample

An investigation is conducted on the status of toxic heavy elements in some rain water samples by using Atomic Absorption Spectrophotometric (AAS) technique. For each digestion, 1 gm of dried power sample was taken into the appropriate digestion vessels. 10 ml of 69% w/w HNO<sub>3</sub> and 2 ml of 30% w/w H<sub>2</sub>O<sub>2</sub> were added with sample into digestion vessels (250ml) and the samples were heated continuously at a temperature slowly raised up to 80-100°C until a clear solution was appeared. After cooling the digest solution, it were centrifuged for 2 min at 30 rpm and filtered with filter paper (Whatman no.42). Final volume was made in a 50 ml volumetric flask with double distilled water. Lastly, the digested samples were analyzed by using an atomic absorption spectrophotometer (AAS). Concentration of Pb, Cd, Cr and As in the filtrate of digested spice samples were estimated by using atomic absorption spectrophotometer (GTA 120-AA240Z, Varian, Australia and SpectrAA 220 AAS, Varian, Australia). The instrument was fitted with specific lamp of particular metal. The instrument was calibrated using manually prepared standard solution of respective heavy metals as well as drift blanks. Standard stock solutions of 1000 ppm for all the metals were obtained from Kanto Chemical Co. Inc, Tokyo, Japan. These solutions were diluted for desired concentrations to calibrate the instrument.

## Data analysis

**Metal pollution index:** The MPI (metal pollution index) was calculated based on the total heavy metal concentration in all spices

tested. This index was calculated by taking the geometrical mean of all metal concentrations in the spices [7].

$$MPI = (Cf1 \times Cf2 \times \dots \times Cfn)^{1/n} \text{-----(1)}$$

Where Cfn = concentration of metal in the spice samples.

**Health risk assessment of spices consumption:** The potential health risks of heavy metal consumption through spices were assessed using the estimated daily intake (EDI), target hazard quotient (THQ), and Hazard Index (HI). The EDI value is determined by the element concentrations in spices, the amount consumed daily, and body weight. EDI values of the analyzed metals were estimated based on Eq. (2). [8]

$$EDI = (C_{\text{metal}} \times IR) / BW \text{.....(2)}$$

Where, EDI is estimated daily intake and Cmetal (mg/kg) is an average weighted heavy metal content in spices, IR (ingestion rate) is the average daily consumption of spices (gram/day person), BW is the average body weight (Kg) [9]. The average IR of spices for adult is 10 g/day/person of dry weight which is similar to the literature. The average body weight for adult was 60.0 kg [10].

THQ is used to assess the non-carcinogenic risks of long-term exposure to contaminants in vegetables, and the calculations were done using Eq (3).

$$THQ = EDI / RfD \text{.....(3)}$$

Where, RfD is reference dose values for each metals of interest (mg/kg day<sub>-1</sub>). The RfD values for Pb, Cd, Cr and As were 0.004, 0.001, 0.003 and 0.0003 mg/kg per day, respectively [11,12].

The HI has been developed to estimate the overall non-carcinogenic risk to human health through exposure of more than one pollutant. As shown in Eq. (4), HI is the total of the hazard quotients all heavy metals in spices.

$$HI = THQ(Pb) + THQ(Cd) + THQ(Cr) + THQ(As) \text{ ..... (4)}$$

If the values of THQ/HI ≥ 1 indicate that the population will pose potential adverse health effects, while if THQ/HI < 1, the population is unlikely to experience obvious adverse effect [13-15].

## Results and Discussion

### Metal concentrations

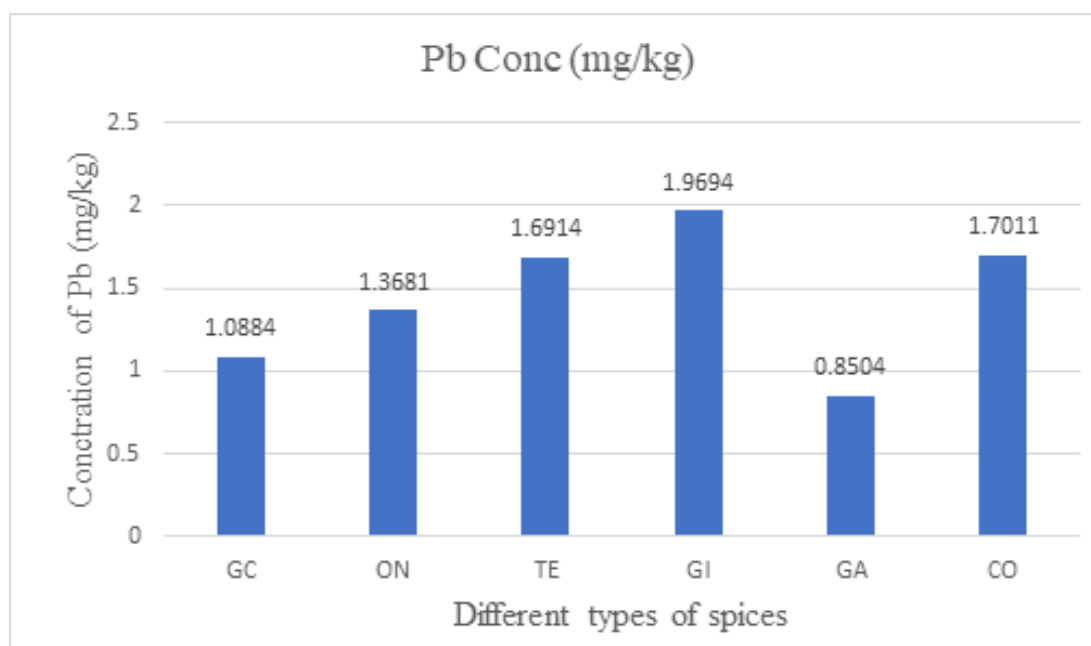
**Lead (Pb):** Total lead concentrations in all the spice samples studied through AAS are shown in Table 2. The concentrations are

reported as milligrams of total lead per kilogram of edible parts of spice (dry wt. basis). From the results, it has been seen that the concentrations of lead were varied from 0.8504 mg/kg to 1.9694 mg/kg. However, the sequence for Pb concentration in the studied spices was found as Garlic < Green chili < Onion < Turmeric < Coriander < Ginger respectively Figure 3. The maximum permissible limit of lead in foodstuffs is 7.2mg/kg (dry wt. basis) [16]. From the present investigation, it is observed that the concentration of lead is lower than the toxic level. K Gleason et al reported that in Munshiganj District of Bangladesh, lead concentrations as high as 483 ppm in many turmeric samples [17]. Again, according to the FDA report PRAN's turmeric powder in plastic packages and plastic jars contained lead ranging from 28 ppm to 53 ppm [18]. Recently,

TIME reported that, researchers in Boston tested 15 spices and food products imported from Indian and found a quarter of the samples contained more than 1 microgram of lead per gram [19]. CG Lin et al. found twenty-two of 86 spices and foodstuff products contained > 1 µg/g lead [for these 22 samples, mean: 2.6 µg/g [95% confidence interval: 1.9-3.3]; maximum: 7.6 µg/g] [20,21]. About 25% of the food items, including spices such as cardamom, fenugreek and chili powder, contained more than 1 microgram of lead per gram of product. M P Senanayake et al. observed spices sold in Sri Lanka contain lead below the US Food and Drug Administration's action level of 0.5 µg/g [22]. So, it is concluded that lead content in all the spice samples collected from Mohanpur Upazila are within normal limit. These are safe for human consumption. (Table 2)

**Table 2:** Concentration of heavy metals in spice sample from Mohanpur Upazila.

Sample ID	Heavy metal concentration in spices samples (mg/kg)			
	Pb	Cd	Cr	As
Green Chili	1.0884	1.2173	0.9184	4.0157
Onion	1.3681	0.2996	0.7172	0.2285
Turmeric	1.6914	0.1522	0.808	0.2862
Ginger	1.9694	0.3204	1.476	2.99
Garlic	0.8504	0.1854	1.2265	0.5903
Coriander	1.7011	0.237	0.8131	0.3457



**Figure 3:** Concentration of lead (Pb) in mg/kg for different spices.

**Cadmium (Cd):** Cadmium is a highly toxic metal that accumulates in the kidneys and liver and can cause a variety of serious diseases even at low concentrations [23-25]. This study reveals that the total Cadmium (Cd) concentration in the studied spices samples remain in the range of 0.1522-1.2173 mg/kg respectively Table 2. However, the sequence for Cd concentration in the studied spics was found as Turmeric < Garlic < Coriander < Onion < Ginger < Green chili respectively Figure 4. The maximum permissible concentration of

cadmium in all foods of Bangladesh is 6 mg/kg (dry wt. basis) [26]. Abou-Arab et al. investigated 303 samples, which represent 20 different types of spices and medicinal plants that were collected from Egypt and found maximum levels of Cd 2.44 µg/g [27]. Again, Turkey Divrikli et al. obtained the Cd level 0.2-2.7 µg g<sup>-1</sup> among eleven different spices and herbal plants collected western Anatolia [25]. O D Adeyolanu et al. identified that the levels of cadmium contents in the basil leaf are lower than other parts; there could be



accumulation of this metal through long-term consumption of the leaf [28]. M Ziyaina et al. investigated cadmium (Cd) concentration in selected spices commonly consumed in Libya and found Cd concentrations for *C. frutescens*, *C. longa* and *P. nigrum* were 0.32

$\pm 0.04$ ,  $0.36 \pm 0.09$  and  $0.35 \pm 0.07$  mg/kg respectively [29]. The result we obtained from the present analysis indicates that all the spice samples are within the permissible limit in terms of cadmium and they are safe for human consumption.

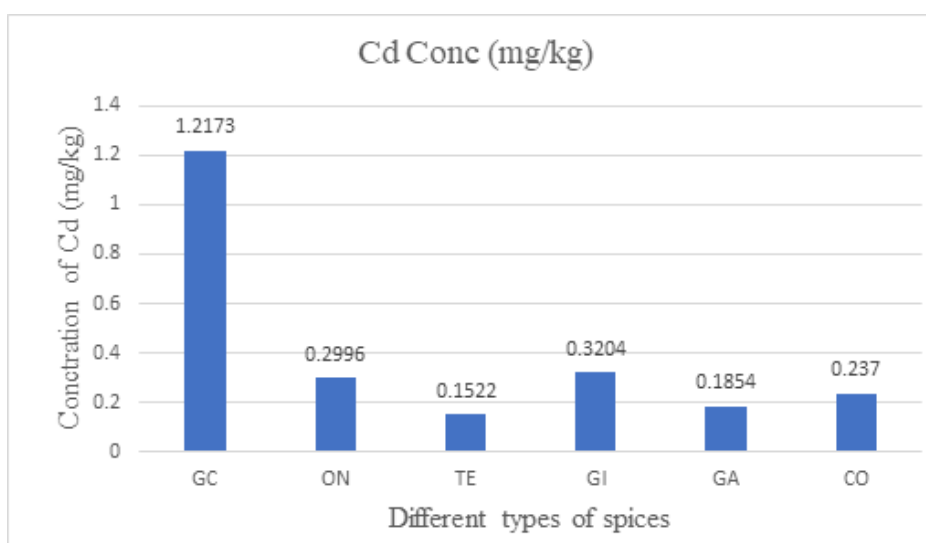


Figure 4: Concentration of cadmium (Cd) in mg/kg for different spices.

**Chromium (Cr):** Chromium is a toxic heavy metal which plays important role for physiological and biological functions of the human body when it is below tolerable limits [30]. However, its deficiency causes hyperglycemia and elevated body fat; and high Cr concentration damages kidneys, liver, and blood cells through oxidation reactions [24]. From the results of Table 2, it is seen that the concentrations of chromium in spics samples were varied from 0.7172-1.4760 mg/kg which were bellow than the maximum permissible limit except two samples (Ginger and Garlic). However, the sequence for Cr concentration in the studied spices was found as Onion < Turmeric < Coriander < Green chili < Garlic < Ginger respectively Figure 5. Maximum permitted concentration of chromium in foods is 1 mg/kg recommended by International/

National standards for heavy metals in food [31]. PFA safe limit of chromium for vegetable is 20 mg/kg [32,33]. The chromium content in vegetables and fruits are 0.16679 mg/kg to 1.92625 mg/kg and 0.513 to 2.5021 mg/kg that are comparable with the values reported by N Saha et al. and E Garcia et al. determined the presence of chromium in a total of 72 samples of 17 different spices and aromatic herbs which are consumed in the Spanish diet and in the Mediterranean diet [34,35] In Egyptian spices and medicinal plants Abou-Arab et al. determined maximum levels of Cr was 33.75  $\mu\text{g/g}$  among a total of 303 samples, which represent 20 different types of spices and medicinal plants [27]. Divrikli et al. found the range of Cr from 0.1 to 9.7  $\mu\text{g/g}$  in eleven different spice and herbal plant species from western Anatolia, Turkey [25].

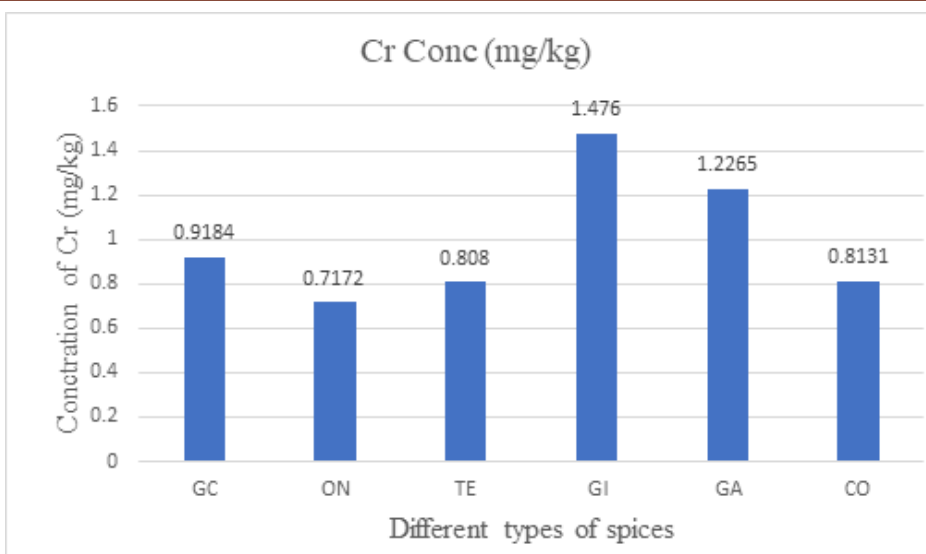


Figure 5: Concentration of chromium (Cr) in mg/kg for different spices.

**Arsenic (As):** From the results of Table 2, it is seen that the concentrations of arsenic in spics samples were varied from 0.5903-4.0157 mg/kg. The recommended and tolerance limits of arsenic in foodstuffs, water and soil are prescribed as 1.0 mg/kg, 0.05 mg/kg and 20 mg/kg respectively [36]. However, the sequence for As concentration in the studied spices was found as Onion < Turmeric < Coriander < Garlic < Ginger < Green chili respectively Figure 6.

In the present investigation we found that, green chili and ginger possess higher arsenic content than permissible limit. Finally, we can say that the samples of onion, turmeric, garlic and coriander have permissible limit of arsenic for human consumption and green chili and ginger are highly toxic for human consumption. (Figure 3-6)

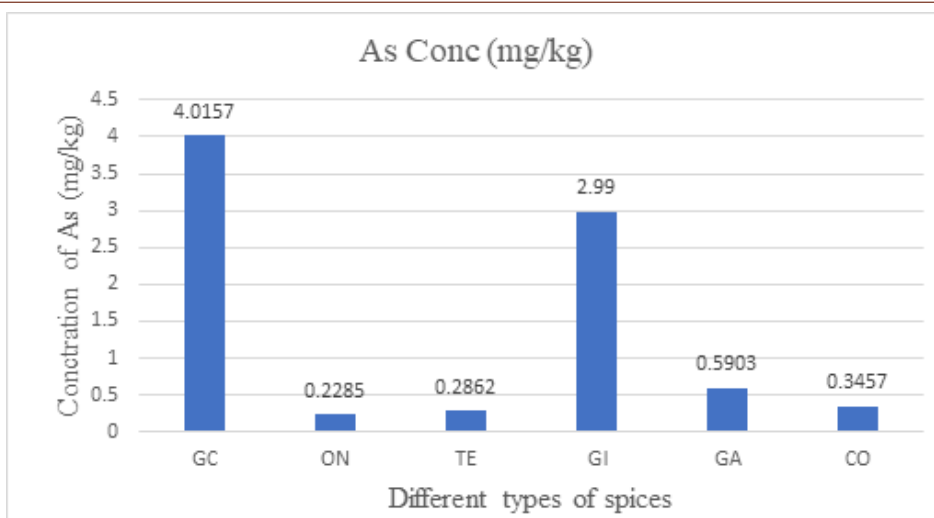


Figure 6: Concentration of arsenic (As) in mg/kg for different spices.

**Metal pollution index (MPI)**

Calculate metal pollution index (MPI) in different spices samples as shown in Figure 7 followed decreasing sequence of Green chili > Ginger > Garlic > Coriander > Onion > Turmeric respectively. Mahtab et al. (2021) calculate metal pollution index (MPI) in different foodstuffs from Ruppur Nuclear Power plant site in Pabna district, Bangladesh and found the sequence of Bitter gourd (BG) > Pawpaw (PP) > Ceylon Spinach (CS) > Red Amaranth (RA) > Guava

(GU) > Mango (MN) > Lady’s Finger (LF) > Brinjal (BR) > Jackfruit (JAC) > Banana (BA) respectively [37]. Jolly et al. (2013) calculate metal pollution index (MPI) in different vegetables from Ruppur Nuclear Power plant site in Pabna district, Bangladesh and found the sequence of BGL > WS > LF > SP > TM > BN > BA > LR > WG > AM > SG > BG > CA > BR > RA > PL > PP > CF > TO respectively [38]. However, from results of MPI, it is obvious that, these spices may cause human health risk due to the higher accumulation of heavy metals.

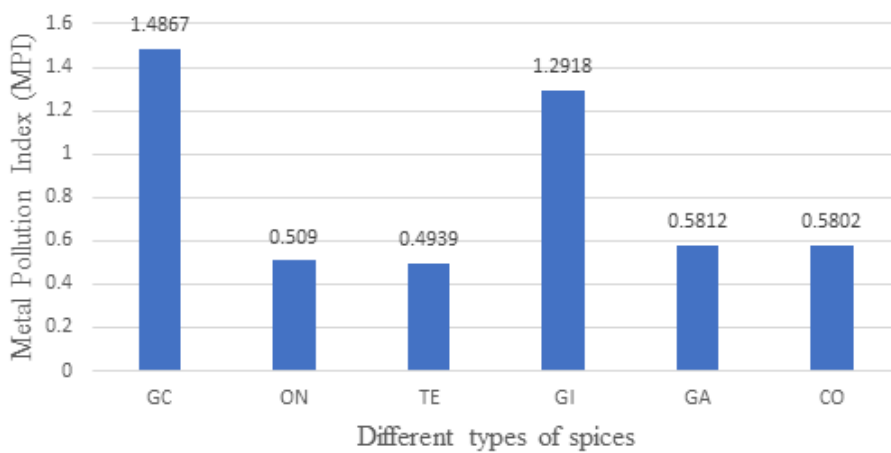


Figure 7: Metal pollution index of spices samples from Mohanpur Upazila.

## Health risk assessment

As shown in Table 2, the EDI values of Pb Cd, Cr and As were calculated to be 0.000181-0.000328, 0.0000253-0.000202, 0.000134-0.000246 and 0.0000380-0.0000983 respectively. M Tefera et. al. found the EDI values of Cr, Cu and Zn were calculated to be 0.0002-0.0041, 0.0018-0.0270 and 0.0018-0.0090, respectively [8]. The THQ values were ranged from 0.0354-0.0820 for Pb, 0.0253-0.2028 for Cd, 0.0328-0.0820 for Cr and 0.1269-0.2.2306 for As. The THQ values in all spices were less than one except As in two spices

samples (Green Chili and Ginger), indicating that consumption of spices do not impose a potential health risk from As. M Tefera et. al. found The THQ values were ranged from 0.008\_1.3805 for Cr, 0.0499-0.1453 for Cu and 0.0062-0.0301 for Zn [8]. It can be seen that 100 % of the HI values due to the consumption of spices were less than one. In general, serious chronic health impact has been suggested when the value of HI is > 10. Therefore, THQ and HI values suggest that the exposed population is assumed to be safe. (Table 3) (Figure 7)

**Table 3:** EDI, THQ and HI values of heavy metals in different spices from Mohanpur Upazila.

Spice Name	Pb		Cd		Cr		As		HI
	EDI	THQ	EDI	THQ	EDI	THQ	EDI	THQ	
Green Chili	0.000181	0.0453	0.000202	0.2028	0.000153	0.051	0.0000669	2.2306	0.4411
Onion	0.000282	0.057	0.0000499	0.0499	0.000195	0.0398	0.000038	0.1269	0.2736
Turmeric	0.000281	0.0704	0.0000253	0.0253	0.000134	0.0448	0.0000477	0.159	0.2995
Ginger	0.000328	0.082	0.0000534	0.0534	0.000246	0.082	0.0000498	1.66	0.4407
Garlic	0.000141	0.0354	0.0000309	0.0309	0.000204	0.0681	0.0000983	0.3276	0.462
Coriander	0.000283	0.0708	0.0000395	0.0395	0.000135	0.0451	0.0000576	0.192	0.3474

## Conclusion

The purpose of this study was to look into the concentrations of various metals in spices. Metals analysis in spices was performed using the optimized wet digestion method. On the basis of the present results it can be concluded that the spices collected from Mohanpur upazila area of Rajshahi district are safe to eat except Green chili, Ginger and some of Garlic. The possible reason of high chromium and arsenic content in spices may be soil nature, irrigated ground water quality, fertilizer and pesticide quality. The difference in the content of metals in each spice may be attributed the difference in soil types and agricultural inputs. The MPI (metal pollution index) value for Green chili, Onion, Turmeric, Ginger, Coriander and Garlic is generally low to spices analyzed. The results of the present study indicate that As is the only metal in two spices (Green chili and Ginger) slightly contributes non-carcinogenic risk from the point of view of THQ. In general, the findings of the study show low health risk of the metals Pb, Cd and Cr through consumption of these spices. Finally, it is suggested that constant monitoring of heavy metals is needed on all food commodities in order to evaluate health risks from toxic heavy metal and protect the consumer.

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## Conflict of Interest

Author declare no conflict of interest

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