



Lead Exposure of Four Biologically Important Common Branded and Non-branded Spices: Relative Analysis and Health Implication

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Abstract

Technological advances and socioeconomic development of Bangladesh are leading to the best choice between branded and non-branded powdered spices. But toxic metal issues can be another factor to elect them. So lead (Pb) exposure and its consequence on the human body for these two categories of four common spices have been ascertained here. A validated laboratory method using hot plate digestion and flame atomic absorption spectrometry (FAAS) has been used for that. The obtained mean concentration of Pb in branded and non-branded spice powders followed the decreasing order of turmeric > coriander > red chili > cumin powder and turmeric > red chili > cumin > coriander powder, respectively, where, in any analysis mode, the non-branded items are potentially ahead of Pb exposure. The market-based mean concentration between these two categories was not a significant difference (*t*-test). Approximately 90% of the spice samples have shown within the maximum allowable limit by Bangladesh Standard. The sum of estimated daily intake indicated that most of the city market (67%) for non-branded items and some branded (33%) samples have crossed the maximum tolerable daily intake (> 0.21 mg/kg). So, the human health risk assumption for the inhabitant of studied areas suggests there is no significant level of risk for them due to the consumption of spices regarding non-carcinogenic risk factor (THQ and total THQ) and carcinogenic risk factor (CR). But the development of a quality control process for loose spice powders as well as toxic metal screening from raw materials to finish products urgently needed and continuous monitoring with identifying the source of toxic metal is highly recommended.

Keywords Lead exposure · Estimated daily intake · Powdered spice · Branded and non-branded · Health risk

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Introduction

Spices are storehouses of plenty of natural bioactive compounds that are so much important to human beings in different ways. The most essential bioactive compounds in spices are flavonoids, carotenoids, alkaloids, proteins, lipids, fatty acids, phenols, vitamins, minerals, etc. [1, 2]. These molecules are biologically significant to the human body as an antioxidant, anti-inflammatory, antimicrobial, anticancer, anti-metabolic, anti-arthritis, anti-asthma, anti-diabetic, anti-obesity, cardio, and liver toxicity protection action [1, 3, 4]. For example, curcumin is the most imperative bioactive compound in turmeric powder and has all of these modes of action in the human body. So far curcumin is considered as GRAS (generally accepted as safe) referred by USFDA. So it has enormous use in the treatment of several diseases even carcinoma [5, 6]. However, spices have shown an outbreaking effect against the SARS-CoV-2 virus

due to their distinctive properties to reduce the activity of that virus or boost the immunity of the human body [6–8], since COVID-19 has been considered a respiratory disease, which has mainly affected the respiratory system, called direct damage to the lung, that produces acute viral pneumonia. Gradually it affects other organs like the nervous system, kidneys, and cardiovascular system, [7, 9]. But the bioactive compounds of major spices have shown significant molecular binding with spike protein and a key enzyme called main protease (Mpro key role in facilitating viral replication and transcription) of SARS-CoV-2. This action of spices has been shown potentially higher than normally used drugs [7, 10]. In addition, the bioactive molecules of spices have inhibited greatly coronavirus replication and cysteine protease, 3CLpro, in vitro antiviral activity (3CLpro is called viral 3-chymotrypsin-like cysteine protease). This has potentially reduced respiratory distress owing to the COVID-19 virus [7, 11, 12]. Consequently, Reported on considering 163 spice-consumed countries, the demise was the lower number for higher spice-consuming countries, as well as the retrieval rate, which was a high number for those countries in case of novel coronavirus [7]. So spices have been used since ancient times as traditional medicine called ethnomedicine as well as pharmaceutical drug development [13, 14]. Several reports and WHO have claimed that around 80% of the world population depends on spices as a primary medicine to cure [15–19]. People, particularly in the South Asian region, frequently are spending spices like cumin, coriander, black pepper, red pepper, turmeric, ginger, and garlic in daily food preparation, most of them in dry powder form [20].

In contrast, these notable biologically active compounds containing plant-like spices are being contaminated through different outsiders, such as organic and inorganic pollutants that are also bioactive adversely regarding the human body. The contamination is happening due to the utmost deterioration in ecological and environmental quality as a consequence of rapid urbanization [21]. Undesirable changes in ecological and environmental conditions cause a hazardous impact on plants, animals, and humans. However, the cruelty of the ecological and environmental impact and its harmful effects on human health depend on the nature and concentration of the pollutants [22, 23]. Several sets of environmental contaminants like carbon-based and inorganic substances have specific properties, e.g., harmfulness, persistence, and bioaccumulation, also called persistent toxic pollutants. The pollutants have been remaining in the ecosystem for a long time with non- or low biodegradability and are translocations to the food chain [22–25]. These can create severe impacts and threats on a global scale, not only to the environmental components (water, air, soil) but also to human health. The most significant variety of those pollutants is toxic heavy metals [22, 24, 26]. Heavy metals can easily

accumulate in soils, water, and biological sections of plants and animals. The accumulated or transferred heavy metals enter the food chain, finally disturbing the usual functions of the human body [22, 24, 27, 28]. A foremost way of exposure to heavy metals is the consumption of contaminated food worldwide, and one out of ten individuals becomes affected [24]. It has been found that heavy metal exposure is increasing the risk of cardiovascular, neurological, renal diseases, digestive infections, cancer, and more than 200 risks of acute and chronic diseases [24, 29, 30]. For instance, in Bangladesh, the cancer-related demise rate was 7.5% in 2005 and will reach up to 13% by 2030, which is almost twice the rate of 2005. Also, it is projected to reach 28.4 million cases of cancer worldwide in 2040, which is a 47% upsurge from 2020 [31]. Several studies have shown that carcinogenic lead (Pb) in the human body comes mainly from food items, and approximately 50% originates from plants [32]. Pb is one of the most common heavy metal contaminants in the total food chain and shares about 10% of total pollution led by heavy metals. Pb has no biological importance in plant life, even though it can generate morphological, physiological, and biochemical problems [32, 33]. However, Pb can easily accumulate in plants through the soil and roots. Most importantly, Pb can survive up to 2000 years on Earth and is harmful to plants in several ways [32].

Bangladesh is a densely populated country, with approximately 165 million people consuming a vast amount of spices every day. People use spices in cooking processes to impart aroma, color, and taste of food and sometimes mask unwanted smells. It is also cherished for its coloring as a preservative and fumigant [34, 35]. The spice market of Bangladesh is developing to branded (quality controlled in large industrial processing) with packaged form since users are showing unwillingness to go for non-branded (traditional systems of home or quality uncontrolled small industry processing spices) unpackaged form. It is assumed that females are more conscious about their future nowadays and people are more serious about the quality of products, and hygienic spices as well as busy urban life. This is supportive of the acceptance of branded spice powder in mass people gradually. The transformation of homemade or quality uncontrolled process non-branded to branded powdered spices is evolving by approximately 15% a year [36]. In a recent study, 42% of users chose only branded spice powder, and 58% chose non-branded spice powder [36]. It was found that on average, 90.70% of people think and believed that the spice powders were not pure in condition for both forms. They assumed the spice powder was made by mixing low-grade raw spice with high grade followed by brick dust and sand [37]. So consumers judge that cancer, chronic nephritis and high blood pressure such life-threatening diseases that come from the consumption of contaminated spice powder [37]. Finally, consumers are considering the spices which

assure purity and deliver authentic taste and color; they may be branded or non-branded. Therefore, the researchers conducted a study on powdered spices of branded and non-branded categories to determine the comparative contamination and health implication due to Pb exposure.

Materials and Methods

Sample Collection and Coding

The most communal powdered spices (turmeric, red pepper, cumin, coriander) from different branded (packet) and non-branded (loose) spices were collected directly from three city markets, e.g., Newmarket, Kawran Bazar, and Mirpur (denoted A, B, and C, respectively) in Dhaka City arbitrarily. Three branded (denoted 1B, 2B, and 3B) and three non-branded (denoted 1N, 2N, and 3N) spice samples in each market were collected randomly for analysis. A 500-g packet of each branded and non-branded spice sample was collected from the selected markets. The collected sample were coded as A1B, A2B, A3B, B1B, B2B, B3B, C1B, C2B, and C3B for every spices in branded category and A1N, A2N, A3N, B1N, B2N, B3N, C1N, C2N, and C3N for every spices in non-branded category. Also, the market-based mean value is expressed as AMB, BMB, and CMB for brand items and AMN, BMN, and CMN for non-brand items. A total of 72 spice samples were collected that were directly used in different households and hotels interested in food preparation. The traditional pictures of spices have presented in Fig. 1.

Sample Treatment Prior to Analysis

Analytical grade chemicals and reagents were kept organized from official local suppliers. For digestion purposes, 69% Normapur nitric acid (HNO_3) from VWR Chemical made by French and 30% (w/w) hydrogen peroxide (H_2O_2) from Scharlab made by Spain were preserved. The glassware was soaked in 3 M HNO_3 overnight then washed and rinsed with deionized water to minimize the chances of interference. Ultrapure deionized water (18 Ω) was used throughout the study. In this process, four separate batches of four different spices were treated. Each batch was maintained with more than one blank, spice sample, duplicate for each spice sample and spike sample (11–12% of batch total). Samples were taken exactly 1.0 g, and then poured 10 mL of conc. HNO_3 . The samples were placed in a hot plate (at 110 °C) until the sample volume was much reduced and more viscous fluid was observed. The samples were kept out of the hot plate to cool, and then 1 mL 30% H_2O_2 was added and again kept in the hotplate (at 110 °C) to ensure a clear solution. After that, each solution was filtered into a vial and diluted to 10 g with ultrapure-deionized water.

Instrument Optimization

A Varian AA240 FS atomic absorption spectrometer was prepared for the determination of Pb bestowing to batch by batch. The atomic absorption spectrometer contained manual sampling tools, and the atomic signal was measured for Pb in integrated mode. To obtain the highest sensitivity, the experimental conditions employed with the AAS instrument

Fig. 1 (a) Cumin (*Cuminum cyminum*) powder, (b) turmeric (*Curcuma longa*) powder, (c) red chili pepper (*Nahuatl chilli*) powder, (d) coriander (*Coriandrum sativum*) powder



were initially optimized. Working range standard solutions of Pb were prepared for each batch analysis by serial dilution of 1000 mg/kg. A standard stock solution of Pb was prepared using 1% (w/w) Suprapur nitric acid (Merck, Darmstadt, Germany). Calibration curves were constructed for every time of analysis by plotting the absorbance of the element versus the element concentration. The constructed calibration curve was subsequently utilized to determine the concentration of Pb in the spice samples. The characteristic concentrations in all cases were below 0.060 mg/L, and the r values were over 0.9990. The results were expressed as mg/kg or ppm of actual weight.

Quality Control and Quality Assurance Program

The environmental conditions were well-kept standard, and all experimental areas were cleaned every day for sample preparation for analysis. All obligatory precautions were taken into account to confirm the accuracy and consistency of the results of trace element analysis. All the equipment was under calibration coverage, and the analysis records were documented. All the chemicals were certified and international grade. The digestion was performed under a fume hood with a covered watch glass to remove contamination. The instrument was properly optimized, and the lamp was set properly. Each analysis was maintained with a blank subtracted. To confirm and ensure the preciseness of the data of analyzed samples, each spice sample was prepared in duplicate then both were analyzed in the triplicate run. The sample mean between the original and duplicate one was calculated using the machine-provided triple run mean data. Furthermore, each batch analysis was maintained with a spike recovery (11–15% of the total sample) performance evaluation for the accuracy of the analytical method. All spiked samples were prepared along with the collected sample preparation. The calculated percent recoveries for the trace metal analysis in spike samples were within the acceptable 90–110% recovery range. Also verify the null hypothesis of the t -test (at $p=0.05$) of spiked data that were significant, which validates the results of this study. Internal quality control (IQC) charts were also constructed to monitor the elongation of method validation, continuously checking the accuracy and precision of analytical data day to day in the laboratory.

Method Validation Process

In this study, method validation process was completed prior analysis using the standard reference materials (SRM) of BCR-060 (aquatic plant) produced by the Joint Research Centre, European Commission, and NIST-1547 (peach leaves) produced by the National Institute of Standards and Technology, USA. In this process, the agreement of

analytical results with reference values of SRM was measured. The analytical result's standard deviation, accuracy, precision, linearity of the calibration curve, significance test, and extended uncertainty were calculated for the best optimal method described in sections "Sample Treatment Prior to Analysis" and "Instrument Optimization." The standard deviation, total standard uncertainty, combined uncertainty, and expanded uncertainty (EU) of the analyzed method are calculated through standard guidelines [38]. For the linearity checking of the calibration curve, different concentration ranges and best-fitted regression line were considered, where r^2 value was very close to 1. To find out the limit of detection (LOD) and limit of quantification (LOQ), the equations $LOD=yB+3sB$ and $LOQ=yB+10sB$ were taken into account, where yB represent calibration blank concentration and sB represent the standard deviation of the blank [38]. The accuracy of the method measures the biasness of the measurements. The accuracy of the results of SRM is evaluated by the following equation [39]:

$$U_{score} = \frac{|X_{lab} - X_{ref}|}{\sqrt{\mu_{lab}^2 - \sigma_{ref}^2}} \quad (1)$$

where X_{lab} , μ_{lab} , X_{ref} , and σ_{ref} represent mean laboratory results, standard deviation, a reference value, and standard deviation, respectively. The U_{score} below 1 complies the agreement of reference value with the analyzed data, but over 1 represents unsatisfactory. For the evaluation of the precision of the analytical results, the relative standard deviation (RSD) was used to justify the random error of the data, and systematic error was evaluated by the null hypothesis of t -test at the $p=0.05$ level using the equation, $t = \frac{(\bar{x}-\mu)\sqrt{n}}{s}$ where \bar{x} is the sample mean, μ reference mean value, n sample size, and s sample standard deviation [38].

Statistical Analysis

The mean concentration of Pb and market-based mean categories of samples were analyzed using SPSS 29.0 (independent sample t -test) based on a completely randomized design. All other figures were also designed by using it.

Calculations

Concentration of Lead in Spice Samples

The concentration of lead in spice samples can be calculated by using the formula:

$$C = \frac{A \times W}{M} \quad (2)$$

where A = concentration of heavy metal in the digested solution, W = final weight after digestion, and M = weight of the sample.

Human Health Risk Assessment

The human health risk assessment by exposure to lead was estimated with the analysis of estimated daily intake (EDI), target hazard quotient (THQ), and carcinogenic risk (CR).

The EDIs for the analyzed metals were calculated by using the formula:

$$EDI = \frac{FIR \times MC}{BW} \quad (3)$$

where FIR is the food ingestion rate (g/person/day), which is 10.5 g for red chili, 5.25 g for turmeric, 3.5 for coriander, and 3.5 g for cumin for Bangladeshi people [40]; MC = mean concentration of lead in spice samples (mg/kg, actual weight basis); and BW = average body weight (adult 70 kg).

THQ is typically used to estimate the potential non-carcinogenic risk of pollutants. It is the ratio of EDI and reference dose (RfD, mg/kg-BW/day) that is expressed by:

$$THQ = \frac{Efr \times Ed \times EDI}{RfD \times T} \times 10^{-3} \quad (4)$$

The RfDs are based on 0.004 BW/day for Pb. Efr is the exposure frequency, which is 365 days/year; Ed is the exposure duration, which is 60 years; and T is the average time exposure, which is 365×60 [17]. If the THQ value is lower than 1, the exposed population should not experience any adverse health hazard. Conversely, if the THQ value is 1, there is a potential health risk, and the probability of health risk will increase with increasing THQ value.

Cancer risks due to lifetime exposure to Pb through the consumption of the selected spice sample were measured using the equation:

$$CR = \frac{Efr \times ED \times CSF \times EDI}{T} \times 10^{-3} \quad (5)$$

where CSF is the carcinogenic slope factor of 0.0085 mg/kg/day for Pb. EDI is the estimated daily intake of Pb. Acceptable risk levels for carcinogens range from 10^{-4} (risk of developing cancer over a human lifetime is 1 in 10,000) to

10^{-6} (risk of developing cancer over a human lifetime is 1 in 1,000,000) [41].

Results and Discussion

Method Validation and Internal Quality Control Results

To achieve the quality results from applied methods and process of spice sample analysis, method validation and quality control processes were performed as per ISO 17025:2017 guidelines. During the method validation process, BCR-060 (aquatic plant) and NIST-1547 (peach leaves) have been used as SRM. The method validation results were verified statistically (Eq. 1). The results are shown below Table 1.

The SRM results using the laboratory-developed method were found excellent agreement with recommended values (recovery 102% for both cases). The accuracy of the analysis method was verified with U_{score} and was found significant results (< 1). To verify the precision of the method, go through RSD for random error check and t -test for systematic error check, RSDs and t values in 95% confidence interval were found within the standard limit. The results of the used methods were confirmed fit for purpose in laboratory conditions. Finally, the same method has been applied for Pb content measurement in spice samples. During the analysis of spice samples, spike samples (11–15% of the total sample) and internal quality control standards (10% of the total analysis) of different sources have been taken for the quality control process. The recovery status has provided here in Table 2.

Valuation and Comparison of Pb in Branded and Non-branded Spices

The measured concentration of Pb for branded and non-branded spices samples (Fig. 2) from three popular city markets of the megacity, Dhaka shows that a considerable amount of Pb is found in all categories of samples. The mean value of Pb concentration in the branded spices followed the decreasing sequence of turmeric powder > coriander powder > cumin powder > red chili pepper powder. On the other side, non-branded items showed a decreasing order of

Table 1 Comparisons of laboratory observed data with the recommended values of SRMs: BCR-060 and NIST-1547

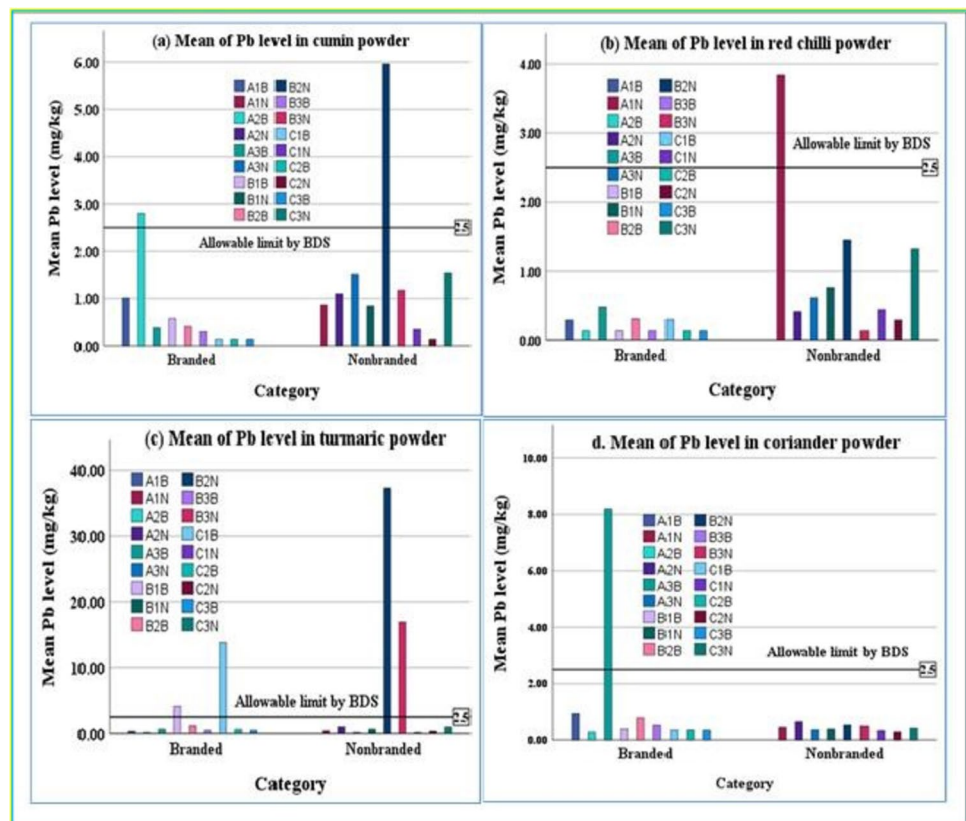
SRM	Recommended value of Pb (mg/kg)	Laboratory mean concentration of Pb (mg/Kg)	Recovery (%)	RSD (%)	EU (%)	U_{score}	Method detection limit (mg/kg)	t value ($p=0.05$)	t_{crit} for $n=17$ ($p=0.05$) [38]
BCR-060	64 ± 4	65.04 ± 2.23	102	2.42	4.63	0.23	0.27	1.96	2.11
NIST-1547	0.869 ± 0.018	0.89 ± 0.064	102.4	7.2	4.63	0.31		1.37	2.11

Table 2 Evaluation of recovery performance of added value with laboratory estimated value

Quality control samples	Added/true concentration (mg/kg)	Mean concentration found (mg/kg)	Recovery (%)	SD	RSD (%)	<i>t</i> value	<i>n</i>	<i>t</i> critical at <i>P</i> =0.05
Spike sample	0.200	0.189	94.5	0.017	9.01	1.76	8	2.36
IQC sample	0.500	0.503	100.6	0.0125	2.48	0.623	8	2.36

The quality control sample has shown excellent recovery performance (94.5% for spike samples, 100.6% for IQC samples) with a very good agreement of *t* value at 95% confidence interval which validated the analyzed data.

Fig. 2 The mean of Pb concentration variations for different spice samples (a) for cumin powder, (b) for red chili powder, (c) for turmeric powder, (d) for coriander powder

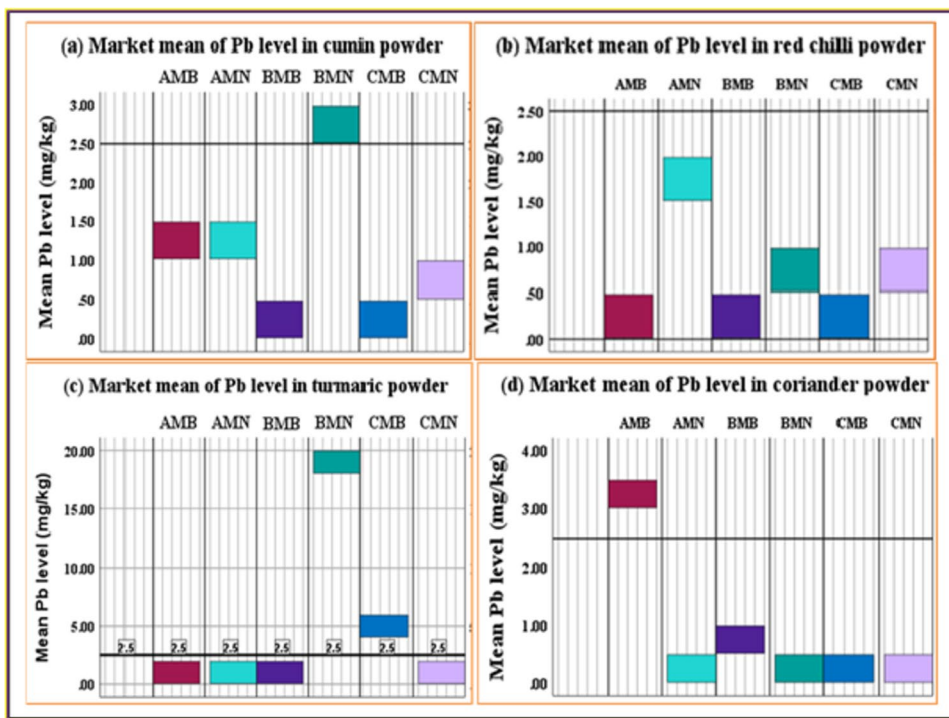


turmeric powder > cumin powder > red chili powder > coriander powder on average. The estimated average value of Pb concentration for cumin powder, red chili powder, and turmeric powder were higher for non-branded spice than for branded items except coriander. In addition, the most important finding is that some of the value of turmeric powder was unexpectedly too high than others in both the category. The concerned authority of Bangladesh (Bangladesh Standard, BDS) has set the allowable limit of spice as 2.5 mg/kg [42]. It was found that a minimum of one of the samples in each type of spice crossed this limit. The Pb concentration was found higher than this limit around 11% of samples of cumin powder followed by 5.5%, 22%, and 5.5% for red chili powder, turmeric powder, and coriander powder, respectively. Among the samples of the higher amount of Pb (> 2.5 mg/kg) in spices, branded samples are responsible for 50% of

the total and another 50% of the sample for the non-branded category.

However, the exposure of Pb in spice samples are also evaluated based on the market mean (Fig. 3) with branded and non-branded categories. The mean concentration for the branded category was higher for coriander powder at market A that also crossed the BDS limit, but other spice samples for branded and the non-branded categories were within the limit in that market. In the case of market B, all the mean of branded spices was within the BDS limit but cumin powder and turmeric powder of the non-branded category overlapped the border. The mean concentration of Pb at market C was also affected only for branded turmeric powder, whereas all other market's mean of brand and non-brand spices was below the borderline. So, the turmeric powders

Fig. 3 Market mean of Pb concentration variations for different spice samples (a) for cumin powder, (b) for red chili powder, (c) for turmeric powder, (d) for coriander powder. The line at 2.5 mg/kg indicated the allowable limit by BDS



of the non-branded category are more significant regarding Pb exposure in terms of others.

Randomly designed independent samples *t*-test shows that the significant variances are kept in different samples but not attained any significant difference in market-based categorical means of Pb exposure. Levene’s test indicated that at a 95% confidence level the data of cumin powder was not significantly varied. But the other spices data showed significant variance at a 95% confidence level. The *t* significant for one-sided or two-sided, both assumed that all

categorical means were not significantly different from each other except red chili for the one-sided test at a 95% confidence level which indicates the source of toxic metals may come from the same source Table 3.

The obtained results indicate that the turmeric powder was the most adulterated by Pb and the most inconsistent for Pb concentration as branded and non-branded items. The higher level of Pb in turmeric powder has also aligned with the report [43]. Besides, coriander powder was shown more consistent in this regard followed by red pepper powder

Table 3 Statistical analysis (independent samples *t*-test) of spice data according to different categorical means

Tests	Spices	Category of means			
		Market A branded with non-branded	Market B branded with non-branded	Market C branded with non-branded	All market Branded with non-branded
Levene’s test for equality of variance (<i>p</i> value)	Cumin powder	> 0.05	> 0.05	> 0.05	> 0.05
	Red chili powder	< 0.05	< 0.05	< 0.05	< 0.05
	Turmeric powder	< 0.05	< 0.05	< 0.05	< 0.05
	Coriander powder	< 0.05	< 0.05	< 0.05	< 0.05
<i>t</i> -test for equality of means (<i>p</i> value) one sided	Cumin powder	> 0.05	> 0.05	> 0.05	> 0.05
	Red chili powder	< 0.05	< 0.05	< 0.05	< 0.05
	Turmeric powder	> 0.05	> 0.05	> 0.05	> 0.05
	Coriander powder	> 0.05	> 0.05	> 0.05	> 0.05
<i>t</i> -test for equality of means (<i>p</i> value) two sided	Cumin powder	> 0.05	> 0.05	> 0.05	> 0.05
	Red chili powder	> 0.05	> 0.05	> 0.05	> 0.05
	Turmeric powder	> 0.05	> 0.05	> 0.05	> 0.05
	Coriander powder	> 0.05	> 0.05	> 0.05	> 0.05

and cumin powder. According to the WHO guidelines, the maximum allowable limit of Pb in common medicinal herbs/spices was mentioned as 10 mg/kg [44]. Almost all values in our study were shown beneath this limit except three and out of that two for non-branded turmeric powder, and one for branded items. According to BDS, 8 out of 72 samples were crossed over this limit and out of these 4 for branded and 4 for non-branded items. In Bangladesh, only a few spices are produced, and most of the demand is fulfilled by importing raw spice seed collection methods from different sources [20]. Again, the branded companies are not well developed in screening the toxic metals situation in raw materials to the finished product but it has some quality control process. On the other hand, unbranded loose spice powders in the market have no quality control process in some cases intentionally added some colored compound to it [37]. The Pb content in spices mainly originates from cultivation soil, irrigation water, fertilizer/pesticides, and cultivation region. Moreover, it can also affect from transportation and manufacturing of raw spices to powdered spices. The branded and non-branded items of spices are manufactured from almost the same raw materials but the manufacturing process can be considered controlled and uncontrolled, though it is difficult

to say any particular source for Pb exposure in spices, especially turmeric powder. So, it is highly recommended to continuously monitor the possible source of Pb in spice in terms of market-based data. Depending on their source of collection, transportation, processing methods, and environment, some of the spice samples have unexpectedly been contaminated and have shown high Pb levels in particular samples. Otherwise, the spices available in Dhaka City from different suppliers as branded or non-branded were shown to be moderately safe zones in terms of Pb level considering the maximum allowable limit of four common spices for food preparation.

Association of This Study with Other Countries

The present study is also compared, the Pb content in spices, with other countries' data which are published in different journals at different times in Asia, Africa, the Middle East, and Europe (Table 4).

The observation of this study is aligned with other countries regarding the findings of toxic metals in common spices. The Pb levels in spice in most of the countries were below 10 mg/kg which level has been guided by WHO.

Table 4 The minimum, maximum value, and final comments reported in the literature regarding Pb in common spices and medicinal herbs for different countries

Country	Minimum value (mg/kg)	Maximum value (mg/kg)	Final comments
This study	<0.27	37.29 ± 7.46	Very inconsistent Pb level especially for turmeric powder
Ethiopia [34]	ND	-	Not experienced potential risk
Eritrea [35]	1.58	12	Consumption controlled or addition controlled
Nigeria [45]	2.7	4.717	Not be exposed to any risk
UAE [46]	1.0	23.52	64% samples exceed the safe limit
Sri Lanka [47]	0.246	0.248	No potential risk
Bangladesh [40]	0.32	1.09	No potential risk
Italy [23]	0.164	2.934	Need attention considering consumer health
India [48]	3.3	4.59	Not be exposed to any risk
Ghana [18, 49]	0.6	115.3	Concerned about the lead level
Egypt [50, 51]	2.3	13.9	Not be exposed to any risk
Malaysia [52]	1.54	8.94	Not be exposed to any risk
South Korea [43]	0.039	0.972	Metal levels vary and should be monitor
Pakistan [53][53][54] [59] [63]	0.55	50	Not be exposed to any risk
Iraq [55]	1.25	14.6	Not be exposed to any risk
Libya [56]	0.96	1.05	Not be exposed to any risk
Iran [57]	0.72	6.04	Investigation needed for health risk
Saudi Arabia [58][58]	trace	14.3	Not be exposed to any risk
Polish [60]	0.14	5.43	Not be exposed to any risk
Serbia [61]	0.11	0.4	Not be exposed to any risk
Turkey [62][62]	0.001	18.0	Consideration of saving the field otherwise poses a risk
Romania [64]	0.04	1.28	Exceed some samples to EU standards and need constant Monitoring
Poland [65]	0.005	5.68	No probability of the appearance of an undesirable effect

Only a few countries have reported higher this value of Pb levels in some spices. This was perfectly matched with our study. Among the countries, around 60% countries reported they have not found any risk. Other countries mentioned a very anxious situation over toxic metal contamination in such useful spices, recommended continuous monitoring, and needed to control the consumption of spices in some cases. Due to the huge application of spices as herbal/traditional medicine as well as food preparation and consumption, spices contaminated by toxic metal is not clearly safe in level, which is a great concern all over the world. Due to this concern, Codex Committee on Spices and Culinary Herbs (CCSCH) has arranged the sixth session of global gatherings regarding spices this year [66].

Health Risk Assessment due to Pb Exposure to Collected Spice Samples

Evaluation of Estimated Daily Intake (EDI)

The study also estimated the dietary exposure of Pb through consumption of the most common spices used in daily food preparation for the adult people who live in the study area in Bangladesh Table 5. The estimated daily intakes of Pb were evaluated according to the mean concentration of each sample and the respective consumption rate [40].

It observed that due to the consumption of spices for selected areas, EDI of Pb for branded items found only around 5.5% samples higher compared with MTDI, whereas about 14% of samples shown higher than MTDI for non-branded samples. On average, Pb consumed most through turmeric powder in both the categories and lowest for cumin powder in the branded category and coriander powder in the non-branded category. The total EDI of the studied metal through the consumption of different spices was observed according to the studied market. The market-based total EDI of Pb for branded spices showed a significant total in almost all the markets. Among the 18 categories of spices, a minimum of 9 categories were crossed MTDI where non-branded categories were most adulterated. So, the inhabitants of market B who were taking non-branded spices consumed a significant amount of Pb. In Bangladesh, any kind of dish is generally made using these four items of spices so the total EDI was particularly important to consider. For this reason, these spices might pose a risk to consumers in the study area of Bangladesh. Since the total EDI of the studied metals was higher than the MTDI in several categories, periodic surveillance will be necessary to set up regulatory norms for the dietary intake of spices, especially for non-branded items, because Bangladeshi people are taking a high amount of spice compared to other countries and are consuming non-branded items more than branded ones [36]. It was reported

Table 5 Estimated daily intake of Pb due to consumption of the prescribed amount of spices in the study area

Sample ID	Cumin powder	Red pepper powder	Turmeric powder	Coriander powder	Total EDI
A1B	0.050	0.044	0.028	0.047	0.169
A2B	0.140	0.020	0.010	0.014	0.184
A3B	0.019	0.072	0.047	<i>0.410</i>	<i>0.547</i>
B1B	0.029	0.020	0.309	0.020	<i>0.378</i>
B2B	0.021	0.047	0.089	0.039	0.195
B3B	0.015	0.020	0.035	0.027	0.096
C1B	0.007	0.045	<i>1.037</i>	0.018	<i>1.107</i>
C2B	0.007	0.020	0.045	0.018	0.090
C3B	0.007	0.020	0.035	0.018	0.079
Average	0.033	0.034	0.181	0.068	<i>0.316</i>
A1N	0.043	<i>0.576</i>	0.031	0.023	<i>0.672</i>
A2N	0.055	0.062	0.075	0.033	<i>0.224</i>
A3N	0.076	0.092	0.010	0.019	0.196
B1N	0.042	0.114	0.046	0.020	<i>0.221</i>
B2N	<i>0.298</i>	<i>0.218</i>	<i>2.797</i>	0.027	<i>3.339</i>
B3N	0.059	0.020	<i>1.271</i>	0.025	<i>1.375</i>
C1N	0.018	0.066	0.010	0.017	0.111
C2N	0.007	0.044	0.029	0.015	0.093
C3N	0.077	0.198	0.072	0.021	<i>0.368</i>
Average	0.075	0.154	<i>0.482</i>	0.022	<i>0.733</i>
Maximum tolerable daily intake (MTDI) [67]	<i>0.21</i>	<i>0.21</i>	<i>0.21</i>	<i>0.21</i>	<i>0.21</i>

The italic formatted values have crossed the reference values (MTDI) which are also italicized

that Bangladeshi people's average estimated daily intake is of approximately 66 g of spices [67], but the average daily intake of common spices per person has been estimated as 4.0 g in the USA, 0.5 g in Europe and 1.0 g in New Zealand, and 14.7 g in South Asia [68].

In this situation, the health risk assessment is necessary since we have found a significant total of EDI in spices.

Non-carcinogenic Risk Pattern

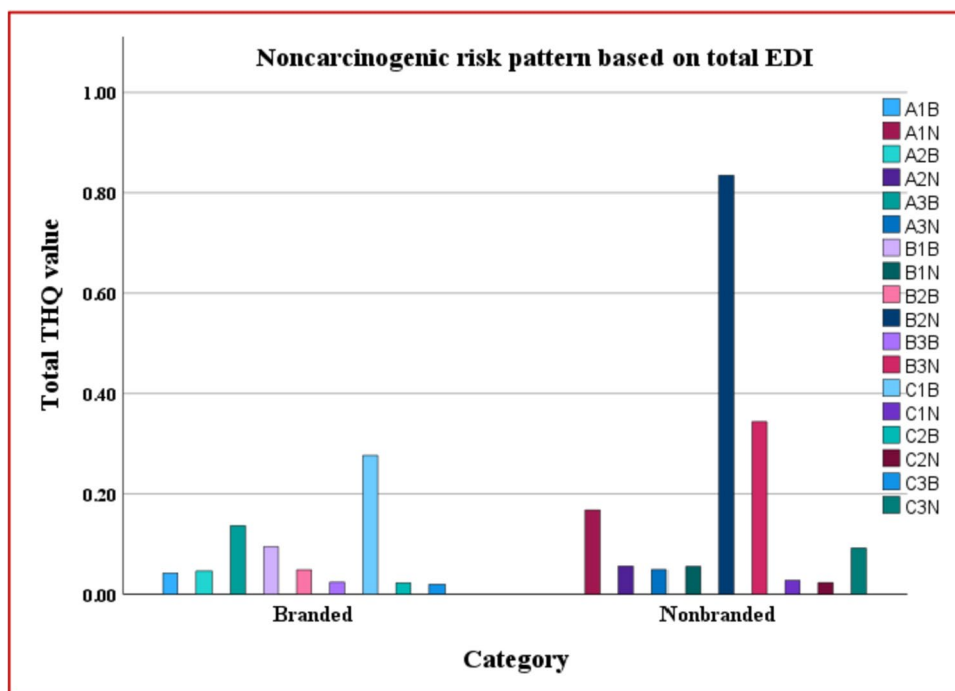
The health risk assessment has been made in two ways like cancerous and non-cancerous health risks. The non-cancerous risk of studied spices was estimated by correlating an exposure level over a definite time period with a reference amount resulting from the corresponding exposure period. The non-cancer risk has been described by a hazard quotient (HQ). The target hazard quotient (THQ) of Pb exposure due to the consumption of spices by Bangladeshi adults is calculated through EDI (Eq. 4). The THQ values of Pb for studied spices with a particular time exposure were less than 1 for all the samples. But the THQ value with respect to the average value of spices indicated the decreasing order of turmeric powder > coriander powder > red chili powder > cumin powder for the branded category, and this order for non-branded items like turmeric powder > red chili powder > cumin powder > coriander powder. The sum of THQ due to the consumption of studied spices is considered as total THQ, the calculated total THQ indicated the non-carcinogenic risk factor did not cross the threshold value (< 1). But the market-based categorical sum of studied branded

spices followed the order C1B > A3B > B1B > B2B > A2B > A1B > C3B > C2B, and for non-branded spices, the order is B2N > B3N > A1N > C3N > A2N > B1N > A3N > C1N > C2N (Fig. 4). From THQ and total THQ values, the inhabitants of study areas will be under satisfactory situation from non-cancer risk in addressed exposure level of Pb through the consumption of the studied spices.

Evaluation of Carcinogenic Risk (CR) Effect

The CR concern owing to Pb exposure in the case of powdered spice consumption for the inhabitant of selected areas of the megacity Dhaka of Bangladesh has been assessed systematically (Eq. 5). The estimated risk pattern for the inhabitant of studied areas was negligible in range (< 10^{-6}) in almost all the samples of spices. However, the average carcinogenic risk factor of spices was found to be turmeric powder (1.54×10^{-6}) > coriander powder (5.76×10^{-7}) > red chili powder (1.72×10^{-7}) > cumin powder (5.74×10^{-8}) for the branded category and turmeric powder (4.1×10^{-6}) > red chili powder (1.31×10^{-6}) > cumin powder (6.36×10^{-7}) > coriander powder (1.87×10^{-7}) for the non-branded category. Also, sum up the carcinogenic risk factor for studied spices to assess the total CR of spices. The summed CR values of the potential risk of Pb also indicated the negligible effect for all market categories of branded and non-branded spices but the two market categories of non-branded spices of market B were shown an acceptable range (10^{-4} to 10^{-6}) of carcinogenic risk factors. The overall CR value

Fig. 4 Non-carcinogenic risk pattern due to the consumption of spices in the selected area



for branded and non-branded spices due to Pb exposure was figured out 2.42×10^{-5} and 5.61×10^{-5} respectively. Generally, CR values lower than 10^{-6} are considered negligible. Above 10^{-4} is considered unacceptable, and values between 10^{-6} and 10^{-4} are considered to be the acceptable range. This implies that the consumers who are taking spices in the studied areas have an acceptable range of carcinogenic threats overall. Therefore, the risk of cancer due to exposure to lead through consumption of the four selected spices will probably not affect the human body in the studied areas.

Conclusions

It can be concluded that the selected spices from the megacity Dhaka of Bangladesh have a considerable amount of toxic Pb in several items of branded and non-branded powder spices. As a developing country like Bangladesh, hundreds of socioeconomic development has been achieved in the last decade. That is related to the selection of branded or non-branded spices in daily food preparation. Different technical reports and articles [40, 43] have tried to describe the accumulation of toxic metal in powder spices but not correlate with the brand and non-brand categories, where we got the non-branded items accumulated more than branded items. Overall, 11% of samples have crossed the allowable limit of Bangladesh Standard. Spices play a significant role in the nutritious and medicinal commitment to consumers. But turmeric powder can consider a prominent adulterate in spice consumption, and loose powdered spices at Kawran Bazar city market have the highest risk of pollution by Pb. So, awareness should be given to non-branded users because of the serious consequence of Pb in the human body, though the studied spices can be considered safe concerning carcinogenic and non-carcinogenic risk factors. This data could be helpful to food safety experts to concentrate more in this area.

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Author Contribution All authors contributed to the study's conception and design. Material preparation was done by D and F, and data collection and analysis were performed by E. The data analysis and figure format with the first draft of the manuscript and revised the submitted version was written by A, B, and C. All authors especially G and H comment on all versions of the manuscript. All authors read and approved the final manuscript.

Data Availability The datasets generated during the analyzed/during the current study are available from the corresponding author upon reasonable request.

Declarations

Ethics Approval This study was performed in line with the continuous monitoring program of the analytical chemistry laboratory and did not require ethical approval.

Consent to Participate Informed consent was obtained from all individual participants included in the study.

Consent to Publish All authors were informed to publish the work.

Competing Interests The authors declare no competing interests.

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