Determination of trace and toxic metals in pulses, spices and sauces

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Abstract: Some trace metals such as Fe, Cu, Mg, Co, Zn and toxic metals like Pb, Cd, As, Cr, Ni were determined in pulses, spices and sauces of Chittagong city of Bangladesh by using Atomic Absorption Spectrophotometer(AAS).Wet digestion method was applied for the digestion of the samples. Spike recoveries (90 %-103 %) and analysis of NIST standard reference metal solutions demonstrated the reliability and accuracy of the analytical methodologies employed in this study. The amount of toxic metals in pulses were determined that ranged from 0.71 to 3.34 mg kg^{-1} of Pb, 0.01 to 0.5mg kg^{-1} of Cd, 0.004 to 0.03 mg kg^{-1} of As, 0.22 to1.06 mg kg^{-1} of Cr and 0.4 to 9.7 mg kg^{-1} of Ni. In the case of spices and sauces they were in the ranges: Pb: 0.08-2.79 mg kg^{-1}, Cd : 0.02-0.67 mg kg^{-1}, As: 0.012-0.293 mg kg^{-1}, Cr : 0.24-68 mg kg^{-1}, Ni: 0.3-9.32 mg kg^{-1} and Cd: 0.013-0.026 mg kg^{-1}, As : 0.007-0.033 mg kg^{-1}, Cr: 5.17-6.37 mg kg^{-1} and Ni: 0.07-3.03 mgkg^{-1}, respectively. The concentration of Pb, Cd and As in all the samples under the study were lower than those of recommended daily allowance that indicates the foodstuffs are almost free from contamination of the metals. But the concentrations of Cr and Ni in some of them were higher than those of the tolerance limits in human body. The intake of such foods can cause the accumulation of these hazardous metals in various parts of the body. The essential trace elements in the said food items are in the range of human necessity.

Key words: Pulses, spices, sauces, essential metals, toxic metals, AAS.

Introduction

Toxic metals are considered to be severe pollutants as they enter into foodstuffs easily through bioaccumulation. They can cause a great concern to the public health if the threshold limit is exceeded. Amidst growing concern of human health, these are being paid special attention throughout the world due to their toxic effects even at very low concentrations in human body [1]. Several cases of human disease, disorder, malfunction and deformation of organs due to metal toxicity have been reported [2].Pulses and spices are essential foodstuffs in a country like ours. Many studies pointed out that pulses and spices contaminated the principal food with different degrees of toxic metals in East Asia [3], Egypt [4], China [5], Nigeria [6] and Turkey [7].

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Jarup *et al*. pointed out that cadmium is present in most foodstuffs with varied concentrations[8]. Cadmium exposure may cause kidney damage and/or skeletal damage[9]. Airborne lead can be deposited on soil, water and plants thus reaching human body via the food chain. Lead is preferred to be accumulated in the skeleton and cause renal tubular damage and kidney damage[10]. International Agency for Research on Cancer (IARC) classified cadmium and lead as human carcinogen[11].Food consumption is the main source of daily chromium exposure for most people. Chromium is usually accumulated in liver, kidney, skin and lung. Small amount of Cr(III) is essential in controlling insulin, cholesterol and lipid biosynthesis[12]. Hexavalent chromium, Cr(VI) is known to be responsible for causing cancer in human body which is yet to be explored fully. However, it has been reported that Cr(VI) binds to double stranded deoxyribonucleic acid (DNA), therefore altering gene replication, repair and duplication. Severe and often deadly pathological changes (asthma, bronchitis, dermatitis, renal failure etc.) are occurred in cases of excessive intake of Cr, especially Cr(VI) [13].Approximately 90% of total intake of nickel comes from food. Excess amount of nickel can be stored in skin, lung, throat and kidney. Small amount of Ni(II) in human body play an important role in regulating prolactin and stabilization of RNA and DNA structures. Excess nickel in human body may lead to lung cancer, nose cancer, larynx cancer, prostate cancer, asthma and chronic bronchitis, heart disorders etc[14]. Nickel in zero oxidation state is said to be more toxic than its other state[15].Arsenic enters into the human body mainly through drinking water, smoking and accumulate in liver, muscle, hair, nail and skin. The International Agency for Research on Cancer (IARC) has listed arsenic as a human carcinogen since 1980. Elemental arsenic is not toxic but trivalent form of arsenic [As(III)] is more toxic than pentavalent form of arsenic[As(V)]. Chronic arsenic exposure is responsible for loss of appetite, nausea and vomiting, dry throat, pain, diarrhoea, tingling of the hands and feet, skin cancer, jaundice etc[16]. Contrary to the toxic metals, Iron, copper, magnesium, cobalt and zinc are known to be essential which are so significant for nutrition and other metabolic functions in human body.

The information gained from this study is expected to be very useful to the general people of this region either to select such kind of food on the basis of the abundances of the essential trace metals or to avoid/limit the food that exceeding the permissible concentration of the toxic metals.

Materials and method

Sampling

Pulses and spices were collected randomly from six different markets of Chittagong city area with adequate number of replicates. Selected sauces were collected randomly from the market of the chosen reputed companies. The samples were stored in refrigerator until required for the preparation of solution.

Sample Preparation

The pulses and spices were washed with water repeatedly to remove the unwanted particles. The washed and clean samples were dried at room temperature for 10 hours and ready for digestion. Sauces were taken for digestion immediately after collecting the samples. Pre weighed (20-25 g) of the samples were taken in a three neck round bottom flask and 50 mL of water was added with the sample. These were boiled for about four hours and then the solvent was evaporated. Then at room temperature, 100 mL HNO_3 and $HClO_4$ (1:5, v/v) was added. This was refluxed at 120-125^{0}C (20 hours) until a clear solution appeared. The volume of the solution was reduced to about 10-15 mL by condensation. Few mL of water was added with the solution and filtered through Whatman-40 filter paper into a 100 mL volumetric flask and made up to the mark with deionized distilled water. All the sample solutions under the present investigation were prepared in similar manner and stored at room temperature for spectroscopic measurements. A blank solution was also prepared for each group of sample by using all reagents except the sample.

Spectroscopic analysis

The sample solutions were subsequently analyzed for toxic metal and essential metal contents, as dry weight basis, using an Atomic Absorption Flame Emission Spectrophotometer (Model AAS-240FS, Varian Australia). But the concentration of As was determined by hydride vapour generation. The analysis was carried out using respective hollow cathode lamps under standard instrumental conditions (Table 1). The flame type used for all elements was air-acetylene. Working solutions were prepared by dilution just before the use of 1000 ppm (BDH, England) standard solutions for atomic absorption spectroscopy. For the determination, two solutions were prepared for each sample and three separate readings were made for each solution. The means of these figures were used to calculate the concentrations.

Table 1: *Instrumental set up of AAS for the elements studied*

Condition	Fe	Cu	Mg	Co	Zn	Pb	Cd	As	Cr	Ni
Wavelength (nm)	248.3	324.8	285.2	240.7	213.9	217	228.8	193.3	357.9	232
Slit (nm)	0.5	0.5	0.5	0.2	1.0	1.0	0.5	0.5	0.2	0.2
Lamp Current (m A)	4	4	4	7	5	10	4	10	7	4
Air flow (L/m)	13.5	13.5	13.5	13.5	13.5	13.5	13.5	13.5	13.5	13.5
Acetylene flow (L/m)	2.0	2.0	2.0	2.0	2.0	2.0	2.0	2.1	2.0	2.0

Quality assurance

Appropriate quality assurance procedures and precautions were carried out to ensure reliability of the results. Samples were generally carefully handled to avoid contamination. Glassware was properly cleaned, and the reagents were of analytical grade. Double distilled deionized water was used throughout the study. Reagents blank determinations were used to correct the instrument readings. A recovery study of the

analytical procedure was carried out by spiking and homogenizing several previously analyzed samples with varied amounts of standard solutions of the metals. Average recoveries obtained were 90-97, 95-99, 97-103, 98-102, 94-97, 95-100, 96-98, 95.2-101, 92-97 and 96-103 % for Fe, Cu, Mg, Co, Zn, Pb, Cd, As, Cr and Ni, respectively. The detection limit of these elements for the instrument under the experimental conditions was determined and the values were 0.01, 0.01, 0.01, 0.01, 0.003, 0.003, 0.03, 0.002, 0.003, 0.05 and 0.02 ppm, respectively. Analysis of NIST standard reference metal solutions also demonstrated the reliability and accuracy of the analytical methodologies employed in this study (Table 2).

Table 2: *Accuracy of the analytical data with reference to the NIST standard metal solution*

Element	Measured Value (ppm)	Certified Value (ppm)	Deviation	Element	Measured Value (ppm)	Certified Value (ppm)	Deviation
Fe	0.974±0.01	1	2.60%	Pb	0.534 ± 0.01	0.5	6.80%
Cu	0.196±0.01	0.2	2%	Cd	0.197 ± 0.02	0.2	1.50%
Mg	0.104±0.01	0.1	4%	Cr	0.504±0.02	0.5	0.80%
Co	0.496 ± 0.01	0.5	0.80%	Ni	0.491±0.01	0.5	1.80%
Zn	0.196 ± 0.01	0.2	2%	As	0.533±0.01	0.5	6.80%

n=7 (seven replicate measurements), ± = Standard deviation of the measurement

Results and Discussion

The amount of essential and toxic metals determined in different pulses, spices and sauces are presented in Tables 3, 4, 5, 6, 7 and 8 respectively. The essential metals were determined as follows:pulses (Fe : 4.67-55.34 mg kg^{-1}, Cu: 0.75-8.36 mg kg^{-1}, Mg :15.31-30.26mg kg^{-1}, Co : 0.08-8.15 mg $kg^{-1}$and Zn : 21.97-58.01 mg kg^{-1}) ; spices(Fe: 8.46-79.43 mg kg^{-1}, Cu: 0.11-13.54 mg kg^{-1}, Mg : 12.18-56.31 mg kg^{-1}, Co: 0.08-8.05mg kg^{-1} and Zn : 2.16-51.97 mg kg^{-1}) and sauces(Fe : 2.23-23.01 mg kg^{-1}, Cu : 1.74-4.98 mg kg^{-1}, Mg: 20.25-35.72 mg kg^{-1}, Co: 0.04-5.56 mg kg^{-1} and Zn:1.25-2.49 mg kg^{-1}). The variations are due to the genetic nature of the species of pulses, spices and their abundances in soils from which they enter to the species[18].The amounts of the essential elements in our findings, in some of the cases are either higher or lower than those reported in literature. The concentration level of Fe, Cu and Mg of the species are fairly good. So these may be considered as a good source of these metals as human necessity. Similarly pulses, spices and sauces contain higher amount of Co and Zn than those reported in the literature. The study revealed that most of the studied samples contain the trace metals within the recommended levels. The recommended daily allowance (RDA) of Fe, Cu, Mg, Co and Zn are 10-15, 1.5-3.2, 350-500, 1-1.5 and 12-15 mg per day, respectively for a man having weight 70 kg[17].Pulses and spices are consumed by the general people in daily diet. So these food items contribute an adequate amount of these

essential metals in human body. Moreover, the higher percentage of organic materials of proteins and digestive enzymes in pulses and spices made them significant foodstuffs[19].The study on sauces is very limited. However our finding shows that these foodstuffs are also fairly good sources of Mg, Co, Zn and Cu but the concentration of Fe in sauces is comparatively lower than those in pulses and spices. Our finding shows no harmful effect on human health for these sorts of pulses, spices and sauces.

Table 3: *Amounts (mg kg^{-1}) of essential metals of different pulses*

Sample	Fe	Cu	Mg	Co	Zn
Gram	55.34±2.33	2.33±0.04	BDL	5.67±0.06	58.01±2.05
Lentil	33.03±1.25	4.22±0.50	28.33±1.04	5.07±0.23	57.27±1.04
Grass pea	38.14±0.53	5.02±1.02	21.31±0.67	8.15±0.07	32.74±1.03
Green gram	25.45±1.20	8.36±0.31	28.04±2.04	6.09±0.12	37.7±0.45
Pea	50.16±0.43	5.01±0.04	15.31±0.87	2.64±0.14	43.89±1.04
Garden pea	4.67±1.01	0.75±0.04	30.26±1.63	0.08±0.03	21.97±1.11
Lablab	28.52±0.51	3.74±0.03	19±2.03	5.71±0.5	23.06±1.02
Cow pea	6.17±0.05	7.19±0.02	19.38±1.06	5.07±0.45	50.33±0.78
Black gram	14.88±1.07	3.44±0.23	25.23±0.75	0.09±0.02	42.24±1.04
Pea	26.39±0.07	5.10±0.13	21.61±1.08	0.39±0.01	37.24±0.04

BDL = Below Detection Limit

Table 4: *Amounts (mg kg^{-1}) of essential metals of different spices*

Sample	Fe	Cu	Mg	Co	Zn
Green chilli	24.38±1.12	0.66±0.04	47.13±2.32	0.14±0.04	2.62±0.78
Onion	17.47±0.34	0.31±0.56	26.1±1.08	0.26±0.33	2.16±0.32
Garlic	17.89±0.25	1.34±0.76	35.1±2.05	0.22±0.04	7.97±0.89
Zinger	8.46±0.24	0.11±0.04	13.88±2.12	0.08±0.01	BDL
Cumin	30.52±1.03	10.38±0.76	41.27±3.06	2±0.5	41.24±2.66
Coriander	24.54±0.54	12.77±1.04	28.28±1.07	5.98±0.78	45.63±1.65
Poppy seed	18.93±0.45	13.54±1.23	56.31±1.02	1.69±0.35	51.97±1.87
Cinnamon	9.05±1.07	3.75±0.45	17.25±1.10	4.47±1.04	4.97±0.85
Turmeric	45±2.02	6.59±0.76	22.8±1.32	1.7±0.67	18.46±0.45
Red chilli	79.83±1.75	10.87±1.67	12.18±0.54	8.05±0.34	20.51±2.07

Table 5: *Amounts (mg kg^{-1}) of essential metals in different sauces*

Sample	Fe	Cu	Mg	Co	Zn
Chilli (*BD foods*)	4.09±0.12	1.74±0.43	29.15±2.32	0.04±0.01	1.65±0.23
Tamarind (*Pran*)	5.97±0.32	2.13±0.45	32.98±1.43	5.56±0.45	1.93±0.43
Tomato (*Meridian*)	8.18±1.02	2.53±0.16	34.21±2.54	0.39±0.02	1.51±0.5
Tomato (*Kwality*)	2.23±0.34	2.62±0.15	21.29±2.22	0.21±0.03	1.25±0.15
Tomato (*Nur foods*)	22.95±2.23	3.77±0.72	20.25±2.54	0.13±0.04	2.49±0.32
Tomato (*Ahmed foods*)	22.66±1.57	3.43±0.73	25.94±1.43	5.48±1.11	2.05±0.37
Tomato (*BD foods*)	23.01±1.87	4.98±0.75	35.72±3.32	0.26±0.23	1.89±0.33

The concentration of Pb was found below the detection limit in most of the pulses samples (Table 6). Lablab contains the highest amount of Pb (3.34 mg kg^{-1}), which is significantly higher because the maximum permissible amount of Pb is 0.3 mg per day from all sources of diet for human [17]. From the same consideration, gram (1.68 mg kg^{-1}) and garden pea (1.56 mg kg^{-1}) also contain elevated amount of Pb. Nevertheless most of the pulses are safe to human health from Pb toxicity. Though cinnamon and red chilli contains 2.47 and 2.79 mg kg^{-1} of Pb, respectively (Table 7), these are not harmful as their intake is not more than 10-15 gm per day per person usually [20].

Table 6: *Amounts (mg kg^{-1}) of toxic metals of different pulses*

Sample	Pb	Cd	As	Cr	Ni
Gram	1.68±0.01	0.5±0.02	0.02±0.01	1.06±0.21	5.9±0.31
Lentil	0.71±0.02	0.22±0.01	0.03±0.01	0.7±0.03	2.44±0.15
Grass pea	BDL	0.03±0.01	0.01±0.01	0.22±0.04	0.4±0.03
Green gram	BDL	0.03±0.01	0.02±0.01	BDL	BDL
Pea	BDL	0.04±0.03	0.02±0.01	0.7±0.04	2.29±0.16
Garden pea	1.56±0.01	0.01±0.01	0.01±0.01	0.56±0.03	BDL
Lablab	3.34±0.02	0.25±0.01	0.01±0.01	0.23±0.12	9.7±0.18
Cow pea	BDL	0.02±0.01	0.03±0.01	0.26±0.03	4.51±0.17
Black gram	BDL	0.01±0.01	0.02±0.01	0.5±0.10	6.63±0.17
Pea	BDL	0.02±0.01	0.02±0.01	0.31±0.04	BDL

As a result, the amount of Pb that is present in the spices is not harmful to health because the contribution of Pb from the food would be far below the maximum permissible value. Hossain *et al*. reported that the Pb concentration in pulse of Bangladesh was in the range 0.5-3.5 mg kg^{-1} and Cd in pulses of Bangladesh was in the range 0.5-1.0 mg kg^{-1} [21]. Krejpeio *et al*. reported the Pb concentration in spices of Poland was in the range 0.17-5.43 mg kg^{-1} [22].

Ozkutlu *et al*. reported that spices of Turkey had 0.018-0.206 mg kg^{-1} of Cd[24]. But our study shows only poppy seed (0.67 mg kg^{-1}) and cinnamon (0.42 mg kg^{-1}) contain higher amount of Cd and rest of the samples contain lower amount of Cd compared to the reported data. The amount of Cd found in sauces is very negligible compared to the MPL value (0.2 mg per day) [17]. Since the consumption of spices and sauces is limited, these foods supply a little amount of Cd into human body. Therefore, they are safe for human health.

The permissible daily allowance of As is 0.7 mg kg^{-1} and the maximum permissible limit of As in foodstuffs is 1 mg kg^{-1} [17].

Table 7: *Amounts (mg kg^{-1}) of toxic metals of different spices*

Sample	Pb	Cd	As	Cr	Ni
Green chilli	ND	0.02±0.01	0.1±0.01	0.24±0.14	0.76±0.06
Onion	ND	0.02±0.01	0.01±0.01	0.26±0.03	7.9±0.45
Garlic	ND	0.02±0.01	0.02±0.01	0.35±0.06	0.37±0.06
Zinger	0.08±0.02	0.03±0.01	0.01±0.01	ND	0.51±0.52
Cumin	0.38±0.05	0.12±0.01	0.1±0.01	8.77±0.76	0.3±0.16
Coriander	1.06±0.23	0.1±0.01	0.29±0.12	36.77±0.16	4.41±0.14
Poppy seed	ND	0.67±0.05	0.23±0.13	15.9±1.03	0.69±0.87
Cinnamon	2.47±0.44	0.42±0.03	0.1±0.01	68.19±2.87	2.25±1.04
Turmeric	0.19±0.05	0.06±0.01	0.16±0.01	17.92±1.04	1.66±0.32
Red chilli	2.79±0.03	0.4±0.04	0.12±0.01	11.81±1.06	9.32±0.44

Table 8: *Amounts (mg kg^{-1}) of toxic metals in different sauces*

Sample	Pb	Cd	As	Cr	Ni
Chilli (*BD foods*)	ND	0.03±0.01	0.01±0.01	5.84±0.55	0.07±0.01
Tamarind (*Pran*)	ND	0.02±0.01	0.02±0.01	5.74±0.22	2.84±0.24
Tomato (*Meridian*)	ND	0.02±0.01	0.02±0.01	5.17±0.67	0.35±0.10
Tomato (*Kwality*)	ND	0.01±0.01	0.01±0.01	5.17±0.52	0.21±0.04
Tomato (*Nur foods*)	ND	0.02±0.01	0.01±0.01	6.37±0.44	0.37±0.12
Tomato (*Ahmed foods*)	ND	0.02±0.01	0.03±0.01	5.3±0.33	3.03±1.03
Tomato (*BD foods*)	ND	0.01±0.01	0.01±0.01	5.43±1.21	ND

ND=Not Determined

The present investigation shows that the amount of As in all the pulses, spices and sauces are very negligible. Thus these foodstuffs are safe in terms of As contamination.

Ahmad and Goni in a study of bioaccumulation of arsenic in crops: Case study, Singair Upzila in Manikgonj District reported the higher concentration of 0.35mg kg^{-1} in corn ear, 0.32mg kg^{-1}in lentil 0.28mg kg^{-1}in field pea and 0.24mg kg^{-1} in kheshari pulse[23]. The value found in our study is much lower.

The concentrations of Cr in pulses, spices and sauces are in the ranges 0.22-1.06 mg kg^{-1}, 0.24-68.19 mg kg^{-1} and 5.17-6.37 mg kg^{-1}, respectively (Table 6, 7 and 8). The concentration of Cr in pulses of Bangladesh is in the range 1.0-3.0 mg kg^{-1} and that of Pakistan is in the range 115-368 mg kg^{-1} [20-21]. So the concentration of Cr in the pulses found in the present study is lower than the reported value and that in spices is far below than those reported previously. According to the permissible value of Cr (0.06 mg per day)[17]all the pulses contain slightly higher level of Cr. But a number of spices, e.g., cinnamon (68.19mgkg^{-1}), coriander (36.77 mg kg^{-1}), turmeric (17.92 mg kg^{-1}), poppy seed (15.9 mg kg^{-1}), red chilli (11.81 mg kg^{-1}) and cumin (8.77 mg kg^{-1}) contain significant amount of Cr. On the basis of the values determined it can be said that all the pulses are

safe in terms of chromium contamination but few spices (Cumin, coriander, cinnamon, turmeric and red chilli) having Cr concentration more than the recommended value are the sources of Cr in body and are thus harmful to health. Though the amount of Cr in sauces (5.17-6.37 mg kg^{-1}) are in consistent and comparatively higher than those found in pulses and spices, but the consumption of sauces is very limited, So the people need not worry on the toxicity of Cr for this foodstuff i.e. sauces.

The highest amount of Ni was found in lablab pulse (9.7mg kg^{-1}) followed by black gram (6.63 mg kg^{-1}), gram (5.90 mg kg^{-1}) and cow pea (4.51 mg kg^{-1}) (Table 6). Red chili, onion, coriander and cinnamon contain 9.32, 7.9, 4.41 and 2.25 mg $kg^{-1}$of Ni, respectively (Table7). Tomato sauces of Ahmed brands and tamarind sauces of Pran brand contain 3.03 mg kg^{-1} and 2.82 mg kg^{-1} of Ni, respectively (Table 8). The presence of Ni in other samples is negligible compared to the MPL value of Ni (0.45 mg per day, Toxicology 2nd ed., 1980)and safe from Ni toxicity[17].Hossain et al. reported that the amount of Ni in pulses of Bangladesh was 9-37mgkg^{-1}that is higher than our study[21].

Conclusion

All kinds of pulses, spices and sauces contain appreciable amount of nutrients and toxic elements within consumable limits. Most of them are found to be safe from cadmium and arsenic toxicities. But few of them contain noticeable amount of toxic metals such as Pb, Cd, Cr and Ni. The present study suggests avoiding these food items as much as possible. The accumulation of heavy metals might have mid term and long term health risks. So strict and periodic surveillance is advisable. Moreover, concerned authority should take necessary steps for finding out the ways in reducing the contamination of toxic metals into the food chain. This study also provides baseline data of toxic metals in pulses, spices and sauces in this region of Bangladesh.

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