Research Paper: Health Risk Assessment of Heavy Metals Via the Consumption of Spaghetti

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ABSTRACT

Background: It has long been recognized that the intake of foods contaminated with chemicals, especially heavy metals can give rise to acute intoxications. The toxic properties of heavy metals are due to their long biological half-lives, as well as non-biodegradable and persistent composition. Considering the importance of the cereal products in the human diet, the present investigation was carried out to assess the human health risk of residue levels of Al, Cr, Pb, and Zn in the domestic and imported spaghetti.

Methods: In this descriptive study, 18 samples of Iranian and imported spaghetti were collected from the market in Kermanshah City, Iran, in 2015. After the preparation and processing of the samples in the laboratory, the concentration of metals was determined using the inductively coupled plasma optical emission spectroscopy. Then, statistical analyses were performed in SPSS (version 19) by running the Shapiro-Wilk test, one-way ANOVA (Duncan multiple range test), Independent t-test , and Pearson correlation coefficient.

Results: The results showed that the mean concentrations of Al, Cr, Pb, and Zn in Iranian spaghetti samples were 655.56±400.35, 211.11±92.80, 2666.67±948.68, and 588.89±116.67 µg/kg, respectively and in the imported spaghetti samples, these values were 2022.22±1940.86, 677.78±393.0, 3300.0±728.0, and 866.67±685.57 µg/kg, respectively. Moreover, Health Risk Index (HRI) values in adults and children via consumption of Iranian and imported spaghetti were within the safe limits (HRI <1).

Conclusion: Considering the serious contamination of some samples of Iranian and imported spaghetti by Al and Pb, the control of heavy metals content during the whole production processing of spaghetti is suggested.

1. Introduction

Because of some properties of heavy metals such as long biological half-lives, non-biodegradable potential, and bioaccumulation potential, they can threaten human health through the soil-plant-food chain contamination. Heavy metals are potential environmental contaminants with the ability to cause adverse health effects such as causing diseases, disorders, malfunctions, and malformations in humans if they appear in excess amounts in the foods consumed by the human being [1-4].

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Some of the trace elements such as Zinc (Zn), iron (Fe), and Copper (Cu) are found in living organisms and considered essential components (i.e. indispensable for health and growth); whereas, other metals are not [5-7]. However, it should be noted that some of these non-essential trace elements can be beneficial to human health via their pharmacological properties [6, 8, 9].

Aluminum (Al) is not an essential element for humans; nevertheless, it is prevalent in water, air, plants, and consequently in all foods [3, 10]. Chromium (VI) or Cr (VI) is toxic and known human carcinogens; whereas, Cr (III) is an essential element. Chromium is mainly used in stainless steel production. Therefore, stainless steel vessels seem to be the main source of Cr contamination [11, 12]. Lead (Pb) is present in the environment because of air, water, and soil pollution. Major sources of this element include fuel combustion, industrial gases, liquid effluents, and agricultural inputs such as some pesticides and phosphate fertilizers [11, 13]. Lead poisoning in adults can affect renal blood flow as well as peripheral and central nervous systems [14]. Furthermore, fetuses, infants, and children are at particularly high risk from neurotoxic and development disorders of this element [15, 16]. However, Zn is known as an essential structural and functional element for numerous cellular processes [17].

Risk assessment includes scientific analyses whose results can be presented quantitatively or qualitatively the likelihood of harm associated with exposure to chemical compounds. In this regard, the human health risk assessment requires identification, collection, and integration of information on health hazards of the chemicals, exposure of humans to the chemicals, and relationships between exposure, dose, and adverse effects [18, 19].

The Acceptable Daily Intake (ADI) of heavy metals was compared with the oral Reference Dose (RfD) or Provisional Tolerable Weekly Intake (PTWI) to examine the potential health risks. Following the standard methods (US EPA), the Health Risk Index (HRI) or Target Hazard Quotients (THQ) are explained as the ratio of the dose resulting from exposure to site media to the dose that is believed to be safe, even in sensitive individuals such as children and elderly. If the HRI<1, no significant risk of chronic-toxic effects exists. If the HRI>1, chronic-toxic effects may occur [20, 21].

The literature review mentions that the estimated dietary intakes of Cr, Pb, and Zn via the consumption of potato/corn chips were 0.37, 0.23, and 33.75 µg/kg/d for children, respectively and were 0.06, 0.04, and 5.19 µg/kg/d for adults, respectively [22]. Filon et al. (2013) reported that the mean content of Pb in wheat flour consumed in Poland was 0.076±0.0448 mg/kg [23]. Iwegbue (2012) reported that the contents of Cr, Pb, and Zn in some brands of biscuits marketed in Nigeria were in the range of 0.39-0.72, <0.001-1.07, and 29.06-49.27 µg/g, respectively [24].

Owing to the possibility of heavy metals accumulation in agricultural products such as cereals and cereal products through irrigation with contaminated water or wastewater, overuse of metal-containing pesticides or chemical and organic fertilizers, atmospheric deposition of heavy metals, production process and packaging [2, 3, 25], the present study was conducted to analyze and assess the non-carcinogenic health risk of the Al, Cr, Pb, and Zn through consumption of some Iranian and imported spaghetti marketed in the city of Kermanshah in 2015.

2. Methods

In this study, the standard stock solutions of different metal ions at the concentration of 1000 µg/mL were used to prepare working solutions after appropriate dilution. Standard solutions were of analytical grade (Merck, Darmstadt, Germany). Distilled deionized water was used in all dilution procedures.

The sample collection was performed according to the Cochran formula of SS=Z2×(p)×(1 - p) /c2 and also considering the constraints of time and budget. For this purpose, 9 samples of Iranian raw spaghetti and 9 samples of imported raw spaghetti were chosen randomly from the market basket of people inhabited in the city of Kermanshah, Iran, in autumn 2015. In the laboratory, 5 g of each sample was placed in crucibles and few drops of concentrated HNO3 were added as an ashing aid. The dry-ashing process was carried out in a muffle furnace by a stepwise increase of the temperature up to 550° C and then left to ash at this temperature for 4 h. Next, the ash was left to cool down and rinsed with 1M HNO3. The ash suspension was filtered by Whatman filter paper grade 42 (Sigma-Aldrich, Germany) and the filtrate made up to the volume of 25 mL with 1M HNO3 [2]. The analysis of Al, Cr, Pb, and Zn was performed by inductively coupled plasma mass spectrometry (Varian, 710-ES, Australia) with three replications (wavelength for Al, Cr, Pb, and Zn were 308.2, 267.7, 220.3, and 206.2 nm, respectively). To check the accuracy of the analytical method, a multi-element standard solution (Merck, Germany) with different concentrations of Al, Cr, Pb, and Zn (100, 500, 1000, 2500, 5000 ppb) were used for the calibration.

In the current study, the potential health risks due to exposure to elements were assessed. For doing so, the average Daily Intake of Metal (DIM) was computed with Equation 1 [4, 26-28]:

$$1.\ DIM = \frac{\text{Cmetal} \times \text{Cfactor} \times \text{Dfood intake}}{\text{Baverage weight}}$$

where Cmetal, Cfactor, Dfood intake, and Baverage weight are respectively the heavy metal concentrations in spaghetti (mg/kg), conversion factor, daily intake of spaghetti (kg/d), and average body weight (kg). The conversion factor (0.085) is used to convert fresh weight into dry weight [29]. The average daily intake of spaghetti for adults and children is considered 0.02 kg per person per day [29]. The average adult and children's body weight are considered as 70.0 and 15.0 kg, respectively [30-32].

The HRI for the local population through the consumption of spaghetti was assessed using Equation 2 [26-28]:

$$2.\ HRI = \frac{\text{DIM}}{\text{RfD}}$$

Here, DIM and RfD represent daily intake of metal and reference dose of metal, respectively. The oral reference doses for Al, Cr, Pb, and Zn were 0.02, 0.003, 0.0035, and 0.30 mg/kg/d, respectively. An HRI of <1 means the exposed population is assumed to be safe [21, 33, 34].

The total HRI (THRI) of heavy metals for the spaghetti was calculated as the mathematical sum of each metal HRI value according to Equation 3 [32]:

3.

$$\textbf{THRI} = \text{HRI (toxicant 1)} + \text{HRI (toxicant 3)} + \ldots + \text{HRI (toxicant } n)$$

The statistical analysis of the obtained results was performed first by the Shapiro-Wilk test for checking data normality, followed by the One-way Analysis of Variance (ANOVA) for statistical grouping of the mean concentrations of Al, Cr, Pb, and Zn between samples. Moreover, the concentrations of the analyzed elements between Iranian and imported samples were compared by the independent t-test. Finally, to study a correlation between the metals in the different samples, the Pearson correlation coefficient was performed. A probability level of P=0.05 was considered statistically significant.

3. Results

Table 1 presents the concentrations of Al, Cr, Pb, and Zn in the analyzed spaghetti samples. Data in Table 1 show that among the analyzed Iranian spaghetti samples, Al, Cr, Pb, and Zn were detected in amounts ranging from 300 to 1500, 100 to 400, 2000 to 5000, and 400 to 800 µg/kg, respectively. Furthermore, among the analyzed imported spaghetti samples, Al, Cr, Pb, and Zn were detected in amounts ranging from 300 to 5000, 300 to 1400, 2200 to 4300, and 200 to 2500 µg/kg, respectively.

However, HRI values in adults and children via consumption of Iranian and imported spaghetti were within the safe limits (HRI<1) (Table 2). The THRI values for adults via consumption of Iranian spaghetti varied from 1.51×10-2 to 3.98×10-2 and for children from 7.04×10-2 to 1.86×10-1 and so below the safe limit (THRI<1). Similarly, the THRI values for adults via consumption of imported spaghetti varied from 1.81×10-2 to 4.74×10-2 and for children from 8.43×10-2 to 2.21×10-1, so they were below the safe limit (THRI<1). Therefore, the consumption of Iranian and imported spaghetti has no potential health risk for adults and children.

Based on the results of the Pearson correlation coefficient, no significant correlations were found between the concentration of the metals in the Iranian spaghetti samples. However, a significant positive correlation was found between Al and Cr concentrations (r=0.850) and between Al and Zn concentrations (r=0.597), and also between the Cr and Zn concentrations (r=0.878) in the imported spaghetti samples.

The results of the independent t-test and Duncan's multiple range test demonstrated that a significant difference was found between the Iranian and imported spaghetti samples and also between some brands of spaghetti in terms of the contents of Al, Cr, Pb, and Zn (P<0.05), respectively.

4. Discussion

The current study was conducted to analyze the content and also health risk assessment of heavy metals (i.e. Al, Cr, Pb, and Zn) through the consumption of some Iranian and imported spaghetti marketed in the city of Kermanshah, Iran, in 2015.

Exposure to high amounts of Al can lead to several serious adverse effects such as osteomalacia, colic, fatigue,

Table 1. Heavy metal concentrations (Mean±SD) of the spaghetti samples (µg/kg, DW)

	Sample	Mean±SD			
		Metal Concentration			
		Aluminum	Chromium	Lead	Zinc
Domestic Spaghetti	1	400.0±82.0^{ab}*	200.0±85.0^{ab}	2200.0±1000.0^{ab}	600.0±64.0^{abc}
	2	600.0±128.0^{b}	400.0±123.0^{c}	2300.0±970.0^{bc}	500.0±44.0^{ab}
	3	400.0±90.0^{ab}	300.0±96.0^{bc}	2100.0±800.0^{ab}	600.0±49.0^{abc}
	4	1000.0±289.0^{c}	100.0±29.0^{a}	2000.0±640.0^{a}	600.0±55.0^{abc}
	5	1500.0±314.0^{d}	200.0±81.0^{ab}	2500.0±810.0^{c}	600.0±61.0^{abc}
	6	900.0±270.0^{c}	200.0±70.0^{ab}	2000.0±500.0^{a}	500.0±57.0ab
	7	400.0±70.0^{ab}	200.0±62.0^{ab}	2900.0±1100.0^{d}	800.0±245.0^{c}
	8	300.0±49.0^{a}	100.0±37.0^{a}	3000.0±820.0^{d}	400.0±173.0^{a}
	9	400.0±68.0^{ab}	200.0±92.0^{ab}	5000.0±1210.0^{e}	700.0±112.0^{bc}
	Concentration	655.56±400.35	211.11±92.80	2666.67±948.68	588.89±116.67
Imported Spaghetti	1	300.0±75.0^{a}	500.0±105.0^{bc}	2500.0±740.0^{b}	600.0±53.0^{b}
	2	500.0±118.0^{a}	300.0±57.0^{a}	4300.0±900.0^{f}	700.0±60.0b
	3	500.0±66.0^{a}	400.0±92.0^{ab}	3500.0±710.0^{d}	600.0±41.0^{c}
	4	5000.0±0.00^{f}	1000.0±149.0^{d}	2200.0±620.0^{a}	1000.20±615.0^{c}
	5	3500.0±0.00^{e}	1400.0±111.0^{e}	4100.0±664.0^{ef}	2500.20±917.0^{d}
	6	5000.0±0.10^{g}	1100.0±101.0^{d}	3100.0±542.0^{c}	1200.00±588.0^{c}
	7	1500.0±410.0^{d}	600.0±160.0^{c}	3000.0±497.0^{c}	700.00±302.0^{b}
	8	800.0±190.0^{b}	300.0±98.0^{a}	4000.0±491.0^{e}	200.00±99.0^{a}
	9	1100.0±270.0^{c}	500.0±123.0^{bc}	3000.0±386.0^{c}	300.00±103.0^{a}
	Concentration	2022.22±1940.86	677.78±393.0	3300.0±728.0	866.67±685.57

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* The letters (a, b, c, d, …) represent the significant difference between the mean content of elements in spaghetti specimens that computed by 1-way ANOVA and Duncan multiple range test (P=0.05).

dementia dialectica, anemia, and other blood disorders, neurodegenerative disorders, reduction of renal function, dental caries, and kidney and liver dysfunctions along with Parkinson and Alzheimer diseases [3]. The results showed that the mean concentrations of Al in Iranian and imported spaghetti samples were 655.56±400.35 and 2022.22±1940.86 µg/kg, respectively, and those were higher than maximum permissible limits (MPL). It has been proved that the root apex (root cap, meristem, and elongation zone) is highly sensitive to Al and accumulates Al very easily [35].

It has been proven that skin irritation, running nose, irritation to the lining of the nose, nose ulcers, liver damage, kidney failure, breathing problems, and circulatory and nerve disorders are the main effects arising from breathing and or long-term exposure to Cr (VI) [12]. Major sources of Cr-contamination in the soil include the disposal of Cr containing wastes and electroplating processes [36]. High contents of Cr causes severe chlorosis, necrosis, and a host of other growth abnormalities and anatomical disorders in plants [37].

Table 2. Daily Intakes of Metals (DIM) in mg and Health Risk Index (HRI) for individual heavy metal caused by the spaghetti

Target Population		Domestic Spaghetti			
		Aluminum	Chromium	Lead	Zinc
Adults	DIM, mg	1.59×10-5	5.13×10-6	6.48×10-5	1.43×10-5
	STD	9.72×10-6	2.25×10-6	2.30×10-5	2.83×10-6
	Min	7.29×10-6	2.43×10-6	4.86×10-5	9.71×10-6
	Max	3.64×10-5	9.71×10-6	1.21×10-4	1.94×10-5
	HRI	7.96×10-4	1.71×10-3	1.85×10-2	4.77×10-5
	STD	4.86×10-4	7.51×10-4	6.58×10-3	9.44×10-6
	Min	3.64×10-4	8.10×10-4	1.39×10-2	3.24×10-5
	Max	1.82×10-3	3.24×10-3	3.47×10-2	6.48×10-5
Children	DIM	7.43×10-5	2.39×10-5	3.02×10-4	6.67×10-5
	STD	4.54×10-5	1.05×10-5	1.08×10-4	1.32×10-5
	Min	3.40×10-5	1.13×10-5	2.27×10-4	4.53×10-5
	Max	1.70×10-4	4.53×10-5	5.67×10-4	9.07×10-5
	HRI	3.71×10-3	7.97×10-3	8.63×10-2	2.22×10-4
	STD	2.27×10-3	3.51×10-3	3.07×10-2	4.41×10-5
	Min	1.70×10-3	3.78×10-3	6.48×10-2	1.51×10-4
	Max	8.50×10-3	1.51×10-2	1.62×10-1	3.02×10-4

Target Population		Imported Spaghetti			
		Aluminum	Chromium	Lead	Zinc
Adults	DIM	4.91×10-5	1.65×10-5	8.01×10-5	2.10×10-5
	STD	4.71×10-5	9.54×10-6	1.77×10-5	1.66×10-5
	Min	7.29×10-6	7.29×10-6	5.34×10-5	4.86×10-6
	Max	1.21×10-4	3.40×10-5	1.04×10-4	6.07×10-5
	HRI	2.46×10-3	5.49×10-3	2.29×10-2	7.02×10-5
	STD	2.36×10-3	3.18×10-3	5.05×10-3	5.55×10-5
	Min	3.64×10-4	2.43×10-3	1.53×10-2	1.62×10-5
	Max	6.07×10-3	1.13×10-2	2.98×10-2	2.02×10-4
Children	DIM	2.29×10-4	7.68×10-5	3.74×10-4	9.82×10-5
	STD	2.20×10-4	4.45×10-5	8.25×10-5	7.77×10-5
	Min	3.40×10-5	3.40×10-5	2.49×10-4	2.27×10-5
	Max	5.67×10-4	1.59×10-4	4.87×10-4	2.83×10-4
	HRI	1.14×10-2	2.56×10-2	1.07×10-1	3.27×10-4
	STD	1.10×10-2	1.48×10-2	2.36×10-2	2.59×10-4
	Min	1.70×10-3	1.13×10-2	7.12×10-2	7.56×10-5
	Max	2.83×10-2	5.29×10-2	1.39×10-1	9.44×10-4

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Huang et al. (2008) reported that Cr is the least mobile element in wheat grains [38]. The results showed that the mean concentrations of Cr in Iranian and imported spaghetti samples were 211.11±92.80 and 677.78±393.0 μg/kg, respectively and those were lower than MPL. Chromium contents in the literature have been reported in the mean concentration of 5490 μg/kg in raw spaghetti samples consumed by students in Italy [11].

Lead is well known for its toxic and adverse health effects. In this regard, the absorption of ingested lead may constitute a serious risk to public health. Some chronic effects of Pb poisoning are anemia, constipation, and colic [6, 39]. The results showed that the mean concentrations of Pb in domestic and imported spaghetti samples were 2666.67±948.68 and 3300.0±728.0 μg/kg, respectively and those figures were higher than MPL. The literature review mentioned that the plants do not absorb or accumulate Pb. However, in soils contaminated with a high content of Pb, some Pb can be taken up [36].

Lead contents in the literature have been reported in the mean concentration of 89±46 and 245±140 µg/kg in pasta marketed in Poland and Egypt, respectively [2, 23]. Besides, the obtained results are much higher than the findings of Alberti-Fidanza_et al. (2002) who reported that the concentrations of Pb in raw spaghetti samples consumed by students in Italy were 2120 µg/kg [11] and also Cuadrado et al., who reported that the concentrations of Pb in pasta samples collected from Spain were 18.70 µg/kg [40].

Zinc is naturally found in soil, but Zn concentrations are rising unnaturally due to man-made activities, especially mining, coal, and waste combustion, industrial activities, and steel processing. Many foodstuffs contain certain concentrations of Zn. Plants often have a Zn uptake that their systems cannot handle due to the accumulation of this element in soils [36]. The results showed that the mean concentrations of Zn in Iranian and imported spaghetti samples were 588.89±116.67 and 866.67±685.57 µg/kg, respectively and those amounts were lower than MPL. Zinc content in the literature has been reported in the mean concentration of 3395±640 µg/kg in spaghetti marketed in Egypt [2]. Moreover, the obtained result is much lower than the findings of Cuadrado et al. (2000) who reported that the concentration of Zn in pasta samples collected from Spain was 14100 µg/kg [40].

Considering the heavy metal concentrations in studied spaghetti samples with the maximum permissible limits (MPL) established by WHO (200 µg/kg for Al and Pb, 1300 µg/kg for Cr, and 50000 µg/kg for Zn) [2, 22, 33], our findings indicated that the mean concentrations of Al and Pb in both Iranian and imported spaghetti samples are higher than MPL.

As shown in Table 2, the average HRI values through the consumption of Iranian spaghetti were 2.11×10-2 and 9.82×10-2 for adults and children, respectively. While, the average HRI values through consumption of imported spaghetti were 3.10×10-2 and 1.44×10-1 for adults and children, respectively. Therefore, the results showed that the mean HRI values of analyzed metals for adults and children through the consumption of Iranian and imported spaghetti are lower than 1. In this regard, the HRI of all analyzed metals were minimal.

5. Conclusion

According to the results, although HRI values in adults and children via the consumption of Iranian and imported spaghetti were within the safe limits (HRI<1), the mean concentrations of Al and Pb in both Iranian and imported spaghetti samples were higher than MPL. Therefore, the control of heavy metals content during wheat cultivation, harvesting process, transportation and storage, and the production process of spaghetti are recommended.

Ethical Considerations

Compliance with ethical guidelines

Ethical approval for this article was registered under license No. 98/154 historian January 14, 2020.

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Conflict of interest

The author declared no conflict of interest.

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