Journal of Pharmaceutical and Health Sciences 2016;4(3), 227-235.

Original Article

JPHS

Study on Chemical Contamination Problem in Macaroni and Pasta Production Technology

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Abstract

This study was done to determine the concentration of heavy metals including Lead (Pb) and Cadmium (Cd) and nutrients element Copper (Cu) and Zinc (Zn) in a simple and whole wheat Macaroni in the valid different supermarkets in Tehran. 254 samples from seven Iranian simple Macaroni, four foreign simple Macaroni, and two Iranian whole wheat macaroni were purchased consecutively in 3 seasons of 2015. Results were determined as mean ± SD of dry weight from three replicates in each test. The samples were analyzed by wet digestion method and standardized international protocols were followed for the preparation of material and analysis of mineral and heavy metals contents and analyzed by Atomic Absorption Spectrophotometer. The mean concentration in milligrams per kilogram of dry matter for Lead, Cadmium, Copper and Zinc were determined. According to the results, the mean contents of Lead in the simple Iranian, imported and Iranian whole wheat Macaroni were 2.897, 3.070 and 0.636, amount of Cadmium were 0.469, 0.620 and 0.920, Copper mean contents were 1.563, 11.866 and 7.085 and finally mean level of Zinc were 16.296, 28.425 and 36.318 mg/kg respectively. The mean concentration of Lead, Cadmium, Copper and Zinc between in whole wheat and simple macaroni samples has a significant difference (P<0/05). The results revealed that the Lead and Cadmium contents in all studied analyzed pasta samples exceeded limits set for them according to the national standard Iran, International Codex and World Health Organization, while Zinc content was lower and Copper content in imported studied brands was slightly higher.

Keywords: Macaroni, Atomic Absorption Spectroscopy, Lead, Cadmium, Copper, Zinc.

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Introduction

Nowadays due to industrial development, pollution in the environment and consequently in agricultural raw materials are appearing as safety major concerns across the world and especially in developing countries. In the case of environmental pollution, heavy metals' contamination is one the major issues which impending danger safety threats. Considering soil, atmosphere, surface water and underground pollution, foods and beverages are getting contaminated with heavy metals. Reported effect of heavy metals in the environmental pollution on consequent contamination of foods and on their safety for human consumption is a serious global public health issue and widely consigned (Alegeria et al., 1990; Mirmohammad-Makki and Ziarati, 2014; Heidari and Ziarati, 2015). Several reports have focused on residues of numerous heavy metals in foodstuffs (Zhang et al., 1997; Zhang et al., 1998). Other reports have outlined on the heavy metal contamination of cereal and cereal products (www.internationalpasta.org; Watt & Merrill, 1967). Hubbard and Lindsay in 1979 reported that ingestion was the major route of man's exposure to the heavy metal. Some heavy metals such as Nickel, lead and cadmium are extensively used in various industries. Their level with an anthropogenic origin is much higher than from natural sources and actually are non-essential for almost all living organisms. The largest amount of trace elements found in the human body has been concentrating via food. Pb and Cd can enter wheat flour and its processed food stuff such as Pasta and Macaroni. Pasta is the hard wheat product formed from the dough, but not leavened. Pasta occupies a substantial position in human nutrition due to its vitamin, mineral and carbohydrate contents.

Italy is the first as the ranking of pasta consumers, with 26 kilos per capita annually. Second-ranked Venezuela consumes slightly less than half that much, 13 kilos per head, while Tunisia is the third-ranking consumer worldwide with 11.9 kilos per head. On other continents, the USA is in ninth place with 8.8 kg; while Iran (7 kg) and Turkey (6.1 kg) precede Japan (1.7 kg) (Pérez et al.,2000).

The generic name pasta includes products obtained from flour mixtures, dried or not, in a no fermented dough with or without the addition of eggs. The commercial names (macaroni, spaghetti, vermicelli, cannelloni, etc.) allude to products of different shapes and sizes. In the United States, pasta that contains yoke is called "noodles" and those that do not, macaroni (Balden, 1838). The spread of pasta manufacturing outside Italy started in the period between World War 1 and World War 2. At present, Iran society is a large pasta-consuming market due to the high expenses of protein and other food stuff. For economic reasons and a new market for low-calorie products, there is a trend for using lower priced raw matter with a smaller starch and higher fiber content whenever the final product is of fairly good quality and acceptable (Mortazavi, 1996).

The main production step of pasta and macaroni involve mixing, kneading, extrusion, drying and packing. In preparing pasta dough, the semolina, flour and water and in some cases egg emulsion and other ingredients are measured in a pre-determined ratio and put into a mixer where they are mixed into a consistency of wet sand i.e. a conglomeration of millions of tiny moist granules. The mixing is normally accomplished for 12-15 minutes and the mixture is usually made to have about 30% moisture. The quantity of water depends on the drying temperature employed in the manufacturing process. In continuous press, mixing is effected under the application of vacuum. The presence of air bubbles in the pasta dough gives the product a chalky appearance and reduces its mechanical strength. At the end of the mixer, the dough is received into a specially designed auger, which is mounted in tightly sealed cast housing. Here, the kneading of the dough, the feeding of the pasta/macaroni

- forming die with the dough and the creation of pressure required for forcing the dough through the extrusion die opening is affected. The kneading operation is necessary to give uniform texture and color to the finished pasta/ macaroni product(Ziarati, 20112; Ziarati et al., 2013). One of the major issue that should be noted is that flour bleaching materials can be carcinogenic. Oxidation of stale flour during long storage, lack of complete separation of weed seeds from wheat, use of animals and poultry feed concentrate powder, uses of damaged poor – quality, germinated wheat or infected by worms, pests, and field – storage fungi including, Eurygaster integricep, ergot, bunt (smut) and rust reduce the quality of flour and its products (Ziarati and Tosifi, 2014; Ziarati and Azizi 2014).

In this research, levels of lead and cadmium as heavy metals and Zinc and Copper as essential elements in samples from different imported and Iranian popular production brands in the forms of simple and whole wheat pasta and macaroni were investigated. In addition, determination of variations in the levels of these elements in Macaroni and pasta samples in the two formulation forms of simple and whole wheat are evaluated and assessing the health risk for Iranian people was another objective of this study.

Materials and methods

Sample collection

206 Iranian and 48 imported pasta samples in two forms of simple and whole wheat were purchased from recognized markets during 3 consequent seasons in 2015.

The determination of humidity of each sample was done in fresh pasta before analyzing essential mineral elements (zinc and copper) and heavy metals (lead and Cadmium) in order to refer all the mixtures to a dry sample and make them comparable.

Lead, Cadmium, Zinc, and Copper Determination

For Lead, Cadmium, Zinc and Copper concentrations in pasta samples, powered samples were dried in an oven for 24 hours at a temperature of 85°C. The samples then were ground and sieved through 0.5 mm sieve. The powdered samples then subjected to the acid digestion using Sulfuric acid (96.5% Merck), concentrated nitric acid (65% Merck), and perchloric acid (70% sigma). Analar grade hydrogen peroxide (about 30%) also was used at the end of the digestion process. Application of concentrated HNO3 along with thirty percent hydrogen peroxide H2O2 (Merck) for mineralization of samples to the complete digestion of samples (Masamba and Kazombo-Mwale,2010; ORA LABORATORY MANUAL FDA, 2014; Hernandez et al., 2004) following Environmental Protection Agency (EPA) Method 3052 was done (ATSDR,1996; Iranian National Standardization Organization, 2013; Jafari-Moghadam et al., 2015).

Two gram of air-dried of each homogeneous pasta samples accurately weighed and 30.0 mL of the digestion mixture (4 parts by weight of nitric acid: 2 parts of Sulfuric acid & 3 parts by weight perchloric acid) and heated slowly by an oven and then rise the temperature. The remaining dry inorganic residues were dissolved in 30.0 mL of concentrated nitric acid and the solution used for the determination of trace and essential mineral elements. Blanks and samples were also processed and analyzed simultaneously. All the chemicals used were of analytical grade (AR). Standardized international protocols were followed for the preparation of material and analysis of heavy metals contents (Ziarati and Tosifi, 2014; Ziarati and Azizi 2014). The samples were analyzed by Flame Emission Spectrophotometer Model AA-6200 (Shimadzu, Japan) using an air-acetylene, flame temperature: 2800°C, acetylene pressure: 0.9–1.0 bar, air pressure: 4.5–5 bar, reading time: 1–10 sec (max 60

sec), flow time: 3-4 sec (max 10 sec), using at least six standard solutions for each metal .The accuracy was checked using quality control test for fungi and their substrate samples to show the degree of agreement between the standard values and measured values; the difference was less than 5%.

Statistical Method

State differences on the basis of the formulation type (simple or whole wheat) and country (Iranian or imported) of samples were determined by student t-test. The changes were calculated by one-way ANOVA and for analysis of the role of multiple factors univariate analysis was used by SPSS 17.Probability values of <0.05 were considered significant. Concentrations were expressed in terms of mg/Kg on a dry weight basis.

Risk Assessment

To evaluate the potential risk of pasta consumption containing the heavy metals, Provisional Tolerable Daily Intake (PTDI) for a 60kg adult person was calculated by the following equation in which C is the heavy metal concentration in pasta, Cons is the average consumption of pasta in country (20 g per capita per day) (Pérez et al., 2000) and BW is body weight of an Iranian adult person (60kg). The output was compared with the WHO/FAO and Iranian standard level.

PTDI = C × Cons / Bw

The Iran standard PTDI limits have been recommended for, Cd and Pb 0.001, 0.0036 and mg/day/kg Bw, respectively (Iranian National Standardization Organization, 2013; Debeca et al., 2003).

Results and Discussion

Results were determined as mean ± SD of dry weight from three replicates in each test. The samples were analyzed by wet digestion method and standardized international protocols were followed for the preparation of material and analysis of mineral and heavy metals contents which analyzed by Atomic Absorption Spectrophotometer in Research Laboratory in Pharmaceutical Sciences Branch, Islamic Azad University.

All of 7 simple Iranian macaroni brands showed the high content of Cd and Pb (Table II), while Iranian and imported whole wheat had a low content of lead and significant higher cadmium levels ($p < 0.01$) as it is demonstrated in Figure 1.

Table 1: Lead content in different simple Iranian Brand samples

Brand	STDEV	Mean content of Lead n=186	Mean of Lead Content		
			Winter 2015	Spring 2015	Summer 2015
A	1.275	[b]2.548	4.002	2.022	1.620
B	0.540	[a]3.332	3.168	3.936	2.892
C	0.812	[a]4.222	5.130	3.972	3.564
D	0.644	[a]3.516	3.360	2.694	4.224
E	0.382	[b]2.928	2.790	2.634	3.360
F	0.514	[c]0.568	0.096	1.116	0.492
G	0.529	[b]2.470	2.226	3.078	2.106
Mean	0.671	2.797	2.967	2.817	2.608

The dietary intake of cadmium was evaluated by the Committee at its fifty-fifth and sixty-first meetings (FAO/ WHO, 2005). In each of these assessments, intakes of cadmium were calculat-

Table 2: Cadmium content in different simple Iranian Brand samples

Brand	STDEV	Mean content of Cadmium n=186	Mean of Lead Content (mg/kg DW) in different studied seasons		
			Winter 2015	Spring 2015	Summer 2015
A	0.042	[d]0.132	0.174	0.132	0.090
B	0.057	[c]0.268	0.254	0.330	0.216
C	0.078	[b]0.376	0.450	0.384	0.294
D	0.034	[b]0.446	0.408	0.474	0.454
E	0.142	[a]0.584	0.738	0.558	0.458
F	0.075	[a]0.788	0.822	0.840	0.702
G	0.067	[a]0.692	0.708	0.618	0.750
Mean	0.068	0.469	0.508	0.476	0.423

ed from available data on concentrations and food consumption taken from the GEMS/ Food regional diets. Total intakes of cadmium estimated by the Committee at its sixty-first meeting ranged from 2.8 to 4.2mg/kg of body weight per week, which equate to 40–60% of the current PTWI of 7mg/kg of body weight per week. The seven commodity groups that contributed significantly to total intake of cadmium included rice, wheat, root vegetables, tuber vegetables, leafy vegetables, other vegetables. These commodities accounted for 40–85% of the total intake of cadmium in the five GEMS/ Food regions. To conduct intake and impact assessments for the seven commodity groups, taking into account three different MLs (the draft Codex ML proposed by CCFAC and one level lower and one level higher than the proposed ML). The draft Codex MLs were as follows: rice, 0.4mg/kg; wheat, 0.2 mg/kg; potatoes, 0.1 mg/kg; stem/ root vegetables, 0.1mg/kg; leafy vegetables, 0.2 mg/kg; other vegetables, 0.05mg/kg (FAO/ WHO, 2005).

Total intakes ranged from 2.8 to 4.2mg/kg of body weight per week, which corresponds to 40–60% of the PTWI of 7mg/kg of body weight per week (FAO/ WHO, 2005).

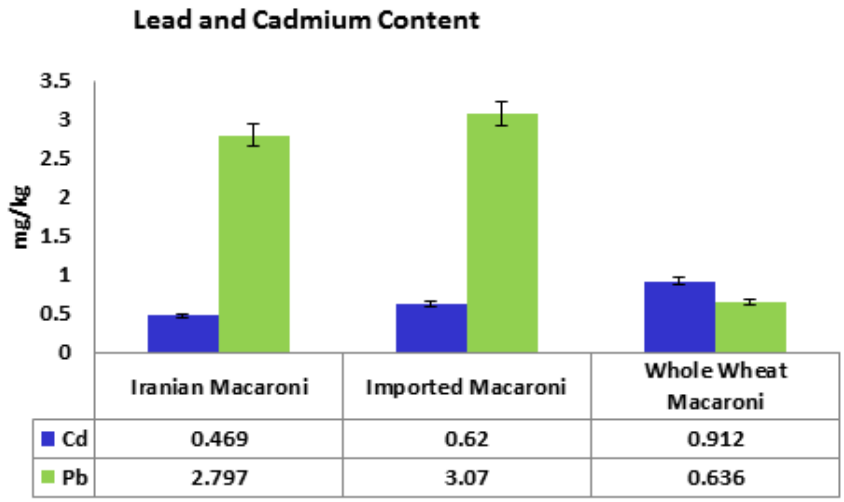

Figure 1- Mean level of Lead and Cadmium Content (mg/kg DW ±SD) in different studied Macaroni samples

Cd and Pb contents were lower in Iranian sample than imported macaroni samples. The highest content of Cd was observed in whole wheat imported brands. Pb levels were lower in Iranian whole wheat samples than other samples. Among all Iranian simple studied macaroni , simple imported winter production date had the highest level of lead by 3.054 738 mg kg-1 dry weight (P<0.05) in comparison to others and among Iranian simple samples Brand D (summer production) showed the highest level od Pb by 4.224 mg kg-1 dry weight (P<0.05) which is demonstrated in Table 1. The highest content of Cd in Iranian was showed in E brands which were produced in winter with 0.738mg kg-1 dry weight (P<0.05).

Among the most collected macaroni samples imported and expensive whole wheat, brands contained more Cd about 0.0.932 mg kg-1 dry weight, whereas the summer production of Iranian brand A contained lower Cd about 0.090mg kg-1 dry weight, all P<0.05 (Table 2).

Cu levels of simple and whole wheat macaroni samples in Iranian and foreign brands were showed wide range varieties (Figure 2). As in imported simple samples copper contents observed much higher than Iranian ones, the significant differences were observed (P<0.01). The season of production in Iranian samples had a significant role in Cu contents and the results revealed that spring date of production had the highest level of copper (Table 3).

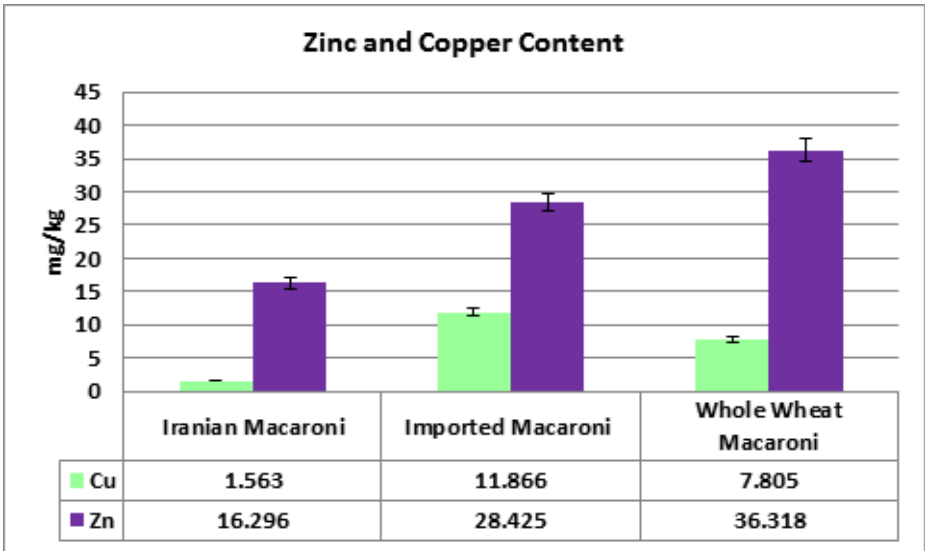

Figure 2: Mean level of Zinc and Copper Content (mg/kg DW ±SD)

The highest levels of Cu content was in imported samples while the highest level of Zinc content was observed in Iranian and imported whole wheat samples (Figure 2). A wide range of zinc content in Iranian simple samples can be seen in Table 4 and Zn content significantly was changed by different brands and time of production (Table 4).

In a study conducted in Canada, variable levels of lead ranging between 30.5 and 36.7 ng/g, and 14.10 and 26.10 ng/g in wheat and cooked pasta collected from different resources were obtained, respectively (Debeca, 1993). Another study in Spain carried out on different kinds of pasta products including spaghetti and vermicelli, indicates a range 26.5 and 37.1 ng/g (Fetter et al., 2011), whereas in Greece the level of 156.3 ng/g was observed in pasta (Tsoumbaris and Tsoukali-Papadopoulou, 1994). When these levels are compared with the results obtained in this study, lead levels in wheat samples were not very different from

Table 3: Copper content in different simple Iranian Brand samples

Brand	STDEV Spring 2015	Mean content of Copper n=186	Mean of Cu Content (mg/kg DW) in simple Iranian Macaroni studied samples in different seasons		
			Winter 2015	Spring 2015	Summer 2015
A	1.672	[a]3.292	1.698	5.034	3.144
B	1.109	[c]0.504	0.600	0.384	0.528
C	1.159	[c]0.682	0.570	0.612	0.864
D	0.028	[c]0.548	0.570	0.516	0.558
E	1.186	[a]3.128	2.940	2.048	4.398
F	1.289	[b]2.186	2.448	3.324	0.786
G	1.153	[c]0.602	0.440	0.624	0.744
Mean	1.085	1.563	1.323	1.791	1.574

Table 4: Zinc content in different simple Iranian Brand samples

Brand	STDEV	Mean content of Zinc n=186	Mean of Zn Content (mg/kg DW) in simple Iranian Macaroni studied samples in different seasons		
			Winter 2015	Spring 2015	Summer 2015
A	4.157	[a]25.234	21.234	24.936	29.532
B	2.968	[a]21.182	24.426	18.600	20.520
C	2.154	[c]10.392	12.846	8.814	9.516
D	2.754	[b]16.268	16.914	18.642	13.248
E	1.702	[b]15.023	14.208	13.883	16.980
F	2.361	[b]16.262	13.692	16.754	18.336
G	1.920	[c]9.716	11.310	10.254	7.548
Mean	2.573	16.296	16.375	15.983	16.530

those obtained in Canada. However, the lead levels in pasta samples (107.08 - 147.58 ng/g) were close to those obtained in Greece.

Conclusion

According to our results, the average amount of Lead in the simple Iranian, imported and Iranian whole wheat Macaroni were 2.897, 3.070 and 0.636, amount of Cadmium were 0.469, 0.620 and 0.920, amount of Copper were 1.563, 11.866 and 7.085 and finally amount of Zinc were 16.296, 28.425 and 36.318 milligrams per kilogram respectively. The average concentration of Lead, Cadmium, Copper and Zinc between in whole wheat and simple macaroni has a significant difference (P<0/05). The results revealed that the Lead and Cadmium contents in all studied analyzed pasta samples exceeded limits set for them according to the national standard Iran, International Codex and World Health Organization, while Zinc content was lower and Copper content in imported studied brands was slightly higher.

In this study, the results related to the pasta production line showed that a decrease in the lead but increasing the cadmium and zinc levels took place during the processing of whole wheat production. On the other hand

during pasta production, the possible sources of contamination are metal surfaces in contact with the material and those present in the air. In conclusion, the present study provides a useful guide for pasta and macaroni choices and pasta preparation taking into consideration the heavy metal toxicity effects. In general, during pasta production, the possible sources of contamination are metal surfaces in contact with the material and those present in air and environment. Kind of processing fuel is also an influence on residues of heavy metals.

Acknowledgment

Pharmaceutical Sciences Branch, Islamic Azad University (IAUPS) is gratefully acknowledged.

Conflicts of Interest

None of the authors have any conflict of in terest associated with this study.

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