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Heavy metals health risk assessment through consumption of some foodstuffs marketed in city of Hamedan, Iran

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ABSTRACT

Since global studies on the health risk assessment of heavy metals in foodstuffs, including biscuits, potato chips, chocolates, and traditional pastries are limited, hence, this study was conducted to determine the contents and the human health risk assessment of lead (Pb), cadmium (Cd) and chromium (Cr) via the consumption of different brands of these foodstuffs.In this descriptive study, a total of 84 samples of 28 brands/kinds of food under consideration were collected from a market basket in city of Hamedan, Iran in 2016. After preparation and processing the samples in the laboratory, the metal concentrations were measured using the inductively coupled plasma-optical emission spectrometer (ICP-OES). In addition, all statistical analyses including One-Way ANOVA and one-sample t test were performed using the SPSS statistical package (Version 19). The results showed that the mean concentrations (mg kg^{-1}) of Pb in all the foodstuff samples (2.25 ± 0.76 for biscuits, 1.60 ± 0.35 for potato chips, 2.61 ± 0.68 for chocolates, and 3.66 ± 0.64 for traditional pastries) were higher than the Maximum Permissible Limit (MPL).Furthermore, the computed health risk index showed no potential risk for adults and children via consumption of the studied foodstuffs under the current consumption rate.Based on the results, as the mean contents of Pb observed in all the foodstuff samples exceeded the MPL, serious attention toward the discharge of pollutants into the environment and chemical residue monitoring, especially of heavy metals in foodstuffs, is recommended.

Key words: Carcinogens, Environmental monitoring, Food safety, Heavy metals, Health risk.

INTRODUCTION

Toxic heavy metals (e.g. Hg, Cd, Cr, and Pb), unlike essential elements (e.g. Ca, Co, Fe, Mg, Mn, and Zn), have no known biological function; yet, as they are not biodegradable and can enter the human body via food, water, and air. They can accumulate in human vital organs and thereafter exhibit toxicological effects, even at trace levels (Sobhanardakani & Jamshidi 2015).

Toxic metals such as Cd, Cr, Hg, and Pb are known to disrupt kidney function, cause liver damage, induce reproductive capacity problems in men and women and also induce hepatic dysfunction, hypertension and tumors. Lead, at elevated levels, can cause renal failure, mental retardation, impaired hearing, shortened gestation period in women, and retard physical development in young children (Wagner 1995; Abou-Arab *et al.* 1996; Iwegbue 2011; Sobhanardakani & Kianpour 2016; Samiee *et al*. 2019; Vahidinia *et al*. 2019). Ingestion of food can cause human exposure to metals, not only because many metals are natural components of this product but also due to contamination during food processing (Ashraf 2006). Therefore, as heavy metals are a potential threat to human health, there is a growing interest in assessing the levels of heavy metals in foodstuff as well as the health risk assessment of their traces (Sobhanardakani 2016a,2017a,2017b; Sobhanardakani & Taghavi 2017).

Risk assessment is a part of risk analysis, resulting in qualitative or quantitative explanations of the likelihood of harm associated with the exposure to a chemical. So that, human health risk assessment requires identification, collection, and integration of information on a chemical health hazards, exposure of humans to the chemical, and relationships between exposure, dose, and adverse effects (Sobhanardakani 2016b).

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At present, sweets (chocolate, candy, biscuit, and pastry) and potato chips are the favorite common food items of people at all ages, especially children who consume them in large amounts. These items are often given to them by their families and friends as signs of affection (Dahiya *et al.* 2005).
Since these foodstuffs may be contaminated with heavy metals and the metal levels of food are directly taken into the body (Gopalani *et al.* 2007), children are the most vulnerable age group to heavy metal contamination by these food items.
Therefore, this study was conducted to assay Pb, Cd, and Cr through the consumption of different brands of commonly consumed biscuits, potato chips, chocolates and traditional pastries as well asto determine human health risk in Iran.

MATERIALS AND METHODS

Sample collection

In this study, a total of 84 samples of different brands were purchased from markets and confectioneries including10 biscuit brands (N =30), six potato chips brands (N = 18), sixcocoa-based chocolate brands (N = 18), and two kinds of traditional pastries named Komaj and Sheermal (N = 18) in city of Hamedan, Iran during 2016. Food samples were stored in polyethylene bags at room temperature until considered for analysis of the metal contents.

Chemical analyses

According to the method suggested by Narin *et al.* (2005), for the wet digestion of potato chips, biscuits, and traditional pastries, 1.0 g of each specimen was taken using a mixture of 12 mL $HN0_3$:HCl (8:4 v/v). Furthermore, the mixtures were heated for 3 h up to 130°C. After cooling, 5 mL of double distilled water (DDW) was added to the samples and mixed. The residue was filtered through Whatmann filter paper No. 42.
The sample was then diluted to 25 mL with DDW (Narin *et al.* 2005). For the digestion of chocolate samples, after removing them from their wrappers, 1 g of each sample was placed into an Erlenmeyer, then 6 mL concentrated nitric acid and 2 mL concentrated hydrogen peroxide were added. This mixture was heated at 100°C for 1 h until the solubilization of the sample was completed and then diluted with 10 mL DDW (Duran *et al.* 2009). Blank digestions were also performed in a similar manner to the samples.
The standard solutions of each element used for calibration were prepared by diluting a stock solution of 1000 mg L^{-1} supplied by Sigma-Aldrich. The contents of the metals in the final solutions were determined using ICP-OES (710-ES, Varian, Australia) with three replications. All the instrumental conditions applied for Pb, Cd and Cr determinations were set in accordance with general recommendations by manufacturer (the wavelengths for Pb, Cd, and Cr were 220.35, 226.50 and 267.72 nm, respectively).
For the quality control of metals, the accuracy of the analytical procedure was controlled by introducing known metal standards into already analyzed samples and reanalyzing after subjecting them to the aforementioned digestion methods. Spiked recoveries were 98% for Pb, 101% for Cd, and 93% for Cr.

Statistical analyses

The statistical analyses of the obtained results consisted of a first Shapiro–Wilk normality test, followed by the study of variance homogeneity using an ANOVA parametric test with a DMS post hoc and a Duncan multiple-range test.
The mean levels of heavy metals were compared with international standards using a one-sample t test. The statistical calculations were performed using the SPSS Version 19.0 (SPSS Inc., Chicago, Il, USA) statistical package.

Potential health risk assessment

In the present study, the potential human health risks due to chronic exposure to heavy metals were assessed. For computing potential risk assessment, the average daily intake of metal (DIM) was calculated using equation 1 (Guo *et al.* 2016):

$$\mathrm{DIM} = \frac{\mathrm{Cmetal} \times \mathrm{Cfactor} \times \mathrm{Dfood\ intake}}{\mathrm{Baverage\ weight}} \quad \mathbf{(1)}$$

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where C_{metal}, C_{factor}, and $D_{food\ intake}$ represent the heavy metal concentrations of examined foodstuffs (mg kg^{-1}), the conversion factor (0.085), and daily intake of foodstuffs (1.4E0–3 kg per person per day for biscuits, 6.0E0–3 kg for potato chips, and also 5.5E0–3 kg for chocolates and traditional pastries, respectively). In addition, $B_{average\ weight}$ indicates the average body weight, which equals to 70.0 kg for adults and 15.0 kg for children (Falco *et al.* 2006; Omar *et al.* 2013; Tang *et al.* 2015).
The health risk index (HRI) for the local population through the consumption of foodstuffs was assessed using equation 2 (Guo *et al.* 2016):

$$HRI = \frac{DIM}{RfD} \quad \textbf{(2)}$$

where DIM and RfD indicate the daily intake of metal (mg) and the reference dose of metal, respectively.
The oral reference doses were 0.0035 mg/kg/day for Pb, 0.001 mg/kg/day for Cd, and 1.50 mg/kg/day for Cr. An HRI < 1 means that the exposed population is assumed to be safe (Xue *et al.* 2012; Liang *et al.* 2015; Zeng *et al.* 2015).
The total HRI (THRI) of heavy metals for the foodstuffs were calculated as the mathematical sum of each individual metal HRI value in accordance with equation 3 (Sobhanardakani 2017).

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

RESULTS

The contents of Pb, Cd, and Cr in the analyzed foodstuff samples are presented in Table 1. As shown in this table, the contamination rate (%) of foodstuff samples by heavy metals reached 100%. Among the samples, Pb was detected in amounts ranging from 1.0 to 4.13, Cd from 0.002 to 0.090, and Cr from 0.01 to 2.20 mg kg^{-1}. Comparing the heavy metal levels in the examined foodstuffs with the maximum permissible limits (mg kg^{-1})established by WHO and the EU (Iwegbue 2011; Hariri *et al.* 2015)—0.30 for Pb, 0.30 for Cd, and 1.30 for Cr in biscuits, potato chips, and traditional pastries; and 1.0 for Pb, 0.05 for Cd, and 1.30 for Cr in chocolates—show that the mean Pb levels in all examined foodstuffs exceeded the MPL, whereas the mean levels observed in biscuits, potato chips, chocolates, and traditional pastry samples never exceeded the MPL for Cd and Cr. Therefore, the contribution of the examined foodstuffs to the total body burden of Cd and Cr can be considered negligibly small.

Table 1. Residual levels of examined heavy metals in foodstuff samples (mg kg^{-1}, wet weight)**.**

Metal	Positive samples (> LOD*)		Min.	Max.	Mean	S.D
	No.	%				
				Biscuit		
Pb	28	93.33	1.000	4.000	2.250	0.760
Cd	29	96.67	0.010	0.090	0.080	0.070
Cr	29	96.67	0.010	2.200	0.370	0.440
				Potato Chips		
Pb	16	88.88	1.300	2.100	1.600	0.350
Cd	16	88.88	0.003	0.020	0.012	0.010
Cr	17	94.44	0.050	1.600	0.500	0.740
				Chocolate		
Pb	17	94.44	1.800	3.521	2.610	0.680
Cd	17	94.44	0.002	0.031	0.012	0.010
Cr	15	83.33	0.205	0.850	0.490	0.230
				Traditional Pastry		
Pb	17	94.44	2.500	4.130	3.660	0.640
Cd	17	94.44	0.010	0.040	0.020	0.010
Cr	17	94.44	0.030	0.710	0.250	0.240

* Limit of Detection (LOD).

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Table 2. Daily intakes of metals (DIM, mg) and health risk index (HRI) for individual heavy metal caused by the examined foodstuffs.

		Pb	Cd	Cr
			Biscuit	
Adults				
DIM		3.82E–06	1.36E–07	6.29E–07
	STD	1.29E–06	1.19E–07	3.49E–06
	Min	1.70E–06	1.70E–08	7.93E–08
	Max	6.80E–06	1.53E–07	1.74E–05
HRI		1.09E–03	1.36E–04	4.19E–07
	STD	3.69E–04	1.19E–04	2.33E–06
	Min	4.86E–04	1.70E–05	5.29E–08
	Max	1.94E–03	1.53E–04	1.16E–05
Children				
DIM		1.78E–05	6.35E–07	2.93E–06
	STD	6.03E–06	5.55E–07	3.49E–06
	Min	7.93E–06	7.93E–08	7.93E–08
	Max	3.17E–05	7.14E–07	1.74E–05
HRI		5.10E–03	6.35E–04	1.96E–06
	STD	1.72E–03	5.55E–04	2.33E–06
	Min	2.27E–03	7.93E–05	5.29E–08
	Max	9.07E–03	7.14E–04	1.16E–05
			Potato Chips	
Adults				
DIM		1.17E–05	8.74E–08	3.64E–06
	STD	2.55E–06	7.28E–08	5.39E–06
	Min	9.47E–06	2.18E–08	3.64E–07
	Max	1.53E–05	1.46E–07	1.17E–05
HRI		3.33E–03	8.74E–05	2.43E–06
	STD	7.28E–04	7.28E–05	3.59E–06
	Min	2.71E–03	2.18E–05	2.43E–07
	Max	4.37E–03	1.46E–04	7.77E–06
Children				
DIM		5.44E–05	4.08e–07	1.70E–05
	STD	1.19E–05	3.40e–07	2.52E–05
	Min	4.42E–05	1.02e–07	1.70E–06
	Max	7.14E–05	6.80e–07	5.44E–05
HRI		1.55E–02	4.08e–04	1.13E–05
	STD	3.40E–03	3.40e–04	1.68E–05
	Min	1.26E–02	1.02e–04	1.13E–06
	Max	2.04E–02	6.80e–04	3.63E–05
			Chocolate	
Adults				
DIM		1.74E–05	8.01E–08	1.53E–05
	STD	4.54E–06	6.68E–08	1.54E–06
	Min	1.20E–05	1.33E–08	1.37E–06
	Max	2.35E–05	2.07E–07	5.68E–06
HRI		4.98E–03	8.01E–05	1.02E–05
	STD	1.30E–03	6.68E–05	1.02E–06
	Min	3.43E–03	1.33E–05	9.13E–07
	Max	6.72E–03	2.07E–04	3.78E–06
Children				
DIM		8.13E–05	3.74E–07	1.53E–05
	STD	2.12E–05	3.12E–07	7.17E–06
	Min	5.61E–05	6.23E–08	6.39E–06
	Max	1.10E–04	9.66E–07	2.65E–05
HRI		2.32E–02	3.74E–04	1.02E–05
	STD	6.06E–03	3.12E–04	4.78E–06
	Min	1.60E–02	6.23E–05	4.26E–06
	Max	3.13E–02	9.66E–04	1.77E–05
			Traditional Pastry	
Adults				
DIM		2.44E–05	1.33E–07	1.67E–06
	STD	4.27E–06	6.68E–08	1.60E–06

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Min	1.67E–05	6.68E–08	2.00E–07
Max	2.76E–05	2.67E–07	4.74E–06
HRI	6.98E–03	1.33E–04	1.11E–06
STD	1.22E–03	6.68E–05	1.07E–06
Min	4.77E–03	6.68E–05	1.33E–07
Max	7.88E–03	2.67E–04	3.16E–06
Children			
DIM	1.14E–04	6.23E–07	7.79E–06
STD	1.99E–05	3.12E–07	7.48E–06
Min	7.79E–05	3.12E–07	9.35E–07
Max	1.29E–04	1.25E–06	2.21E–05
HRI	3.26E–02	6.23E–04	5.19E–06
STD	5.70E–03	3.12E–04	4.99E–06
Min	2.22E–02	3.12E–04	6.23E–07
Max	3.68E–02	1.25E–03	1.47E–05

In addition, in the present study, all the calculated HRI values of heavy metals were within the safe limits (HRI < 1) (Table 2). Furthermore, the variations in the range of THRI values—5.03E–04 to 2.01E–03 and 2.35E–03 to 9.80E–03 in the biscuit samples; 2.73E–03 to 4.52E–03 and 1.27E–02 to 2.11E–02 in the potato chips; 3.44E–03 to 6.93E–03 and 1.61E–02 to 3.23E–02 in the chocolates; as well as 4.84E–03 to 8.15E–03 and 2.25E–02 to 3.81E–02in traditional pastries for adults and children respectively—were also within the safe limit (THRI < 1). Therefore, it can be concluded that people might have no potential significant health risk through only consuming the examined foodstuffs.

DISCUSSION

One of the major sources of human exposure to toxic metals is through food ingestion. It is obvious that for normal physiological functions, humans require an adequate intake of some essential elements. However, humans may be exposed to toxic elements mainly through the consumption of different kinds of foodstuffs including fresh and processed foods, drinking-water, and even through occupational exposures (Hosseini *et al.* 2013). Lead is well known for its toxicity and its serious adverse effects on human health, especially on the development of abnormalities in children. It has been proved that about 85% of daily doses of this element are absorbed into the human body through food consumption. Therefore, food is the main source of non-occupational human exposure to Pb (Krejpcio *et al.* 2005; Liu *et al.* 2010; Hariri *et al.* 2015). In the present study, the mean Pb levels (mgkg^{-1}) in biscuits, potato chips, chocolates, and traditional pastries were 2.25 ± 0.76, 1.60 ± 0.35, 2.61 ± 0.68 and 3.66 ± 0.64, respectively; all these values were higher than the MPL. So that, even though the computed health risk index of Pb shows no potential risk for adults and children via the consumption of the examined foodstuffs, since Pb, like other toxic metals, is removed very slowly from the human body, consumption of food containing this element during a short period can cause some digestive problems including weight loss, constipation, vomiting, abdominal pain, colic, and other chronic problems such as language development delay, behavior change, lethargy, and anemia (Needleman 2004; Uluozlu *et al.* 2007; Duran *et al.* 2009). The same studies observed that the mean Pb level (mgkg^{-1}) in potato chips consumed among the Lebanese population was 0.42 ± 0.41 (Hariri *et al.* 2015), in some brands of biscuits consumed in Nigeria was 0.51 ± 0.36 (Iwegbue 2012), and, similarly, in the present study, it was higher than the MPL. In another study, Iwegbue (2011) reported that the mean Pb levels in some chocolate brands consumed in Nigeria were 0.80 ± 0.80, hence,in contrast tothe present study, it was lower than the MPL.In addition, Gopalani *et al.* (2007) reportedthat the mean Pblevels in different brands of biscuits available in Nagpur, India were lower than the MPL. Salama & Radwan (2005) analyzed different brands of biscuits marketed in Alexandria, Egypt, reporting that the mean Pb levels were 0.127 mg kg^{-1}.

Cadmium is a toxic element and due to its ability to accumulate in the human body, long-term exposure to Cd can cause adverse health effects such as kidney disease, lung damage, fragile bones, anemia, hypertension, arthritis, cardiovascular disease, diabetes, hypoglycemia, osteoporosis, kidney disease, and cancer. So that, another important complication of Cd is its effect on the reproductive system of female (Duran *et al.* 2009; Ju *et al.* 2012; Chen *et al.* 2014; Liao *et al.* 2015; Hosseini *et al.* 2018). The results of the present study showed that the mean Cd levels (mg kg^{-1}) in biscuits, potato chips, chocolates, and traditional pastries were 0.08 ± 0.07, 0.012 ± 0.01, 0.012 ± 0.01, and 0.02 ± 0.01, respectively, hence, lower than the MPL in all the samples.

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The mean Cd levels (mg kg^{-1}) reported in the literature were 0.64 ± 0.79 in potato chips consumed in Lebanon (Hariri *et al.* 2015), 0.07 ± 0.06 in chocolates consumed in southern Nigeria (Iwegbue 2011), 0.07 ± 0.08 in different brands of biscuits marketed in Egypt (Salama & Radwan 2005), and 0.04 ± 0.008 in different biscuit types consumed in Nigeria (Iwegbue 2012). Furthermore, for other similar food products, Iwegbue (2011) reported that the mean Cd levels in candies consumed in Nigeria were 0.03 ± 0.00 mg kg^{-1}. Duran *et al*. (2009) also reported that Cd levels in chewing gums and candies marketed in Turkey were in the range of 0.027–0.825 mg kg^{-1}.

Chromium, in its trivalentform, is an essential trace element. It is biologically usable to potentiate insulin action, and therefore, plays a significant role in the metabolism of lipid, glucose, and protein. However, hexavalent Cr is a carcinogen (Ikem & Egiebor 2005; Janbakhsh *et al.* 2018). Sources of contamination by this element are mainly from the use of raw materials, manufacturing processes, and leaching Cr from vessels in which chromium is stored (Dahiya *et al.* 2005). Breathing high levels of Cr and chronic exposure to this element can cause nose ulcers, irritation to the lining of the nose, running nose, breathing problems, damage to liver, skin irritation, kidney failure, as well as circulatory and nerve disorders (Kabata-Pendias 2010). The results of the present study showed that the mean Cr levels (mg kg^{-1}) in biscuits, potato chips, chocolates, and traditional pastries were 0.37 ± 0.44, 0.50 ± 0.74, 0.49 ± 0.23, and 0.25 ± 0.24, respectively, thus lower than the MPL in all of these foodstuffs. The mean Cr levels (mg kg^{-1}) reported in the literature were 0.68 ± 0.86 in potato chips consumed in Lebanon (Hariri *et al.* 2015), 1.90 ± 0.60 in chocolates in southern Nigeria (Iwegbue 2011), and 0.56 ± 0.13 in the different biscuit types in Nigeria (Iwegbue 2012). In addition, for other similar food products, Iwegbue (2011) reported that the mean Cr levels in candies consumed in Nigeria were 1.80 ± 0.90 mg kg^{-1}, higher than the MPL. Duran *et al*. (2009) reported the Cr levels in the chewing gums and candies marketed in Turkey to be in the range of 0.740–6.265 $mgkg^{-1}$. Gopalani *et al*. (2007) also reported that the mean Cr levels in some biscuit brands consumed in India were higher than the MPL.

As shown in Table 2, the HRI values of Pb, Cd, and Cr for children and adults are less than 1. Hence, the average HRI values in biscuit samples were 4.09e–04and 1.91E–03; in potato chips were 1.14E–03 and 5.31E-03; in chocolates were 1.69E–03and 7.86E–03; and also in traditional pastrieswere 2.37E–03and 1.11E–02for adults and children respectively. Therefore, it can be concluded that a target population might have no potential significant health risk by only consuming the analyzed foodstuffs from the study area. However, the non-carcinogenic risks are greater for children than for adults.

CONCLUSIONS

The element contents increase according to the following descending order for all foodstuff: Cd > Cr > Pb. Based on the results the mean Pb concentrations in all the foodstuff samples were higher than the MPL. The results of HRI showed that there are no potential risks for adults and children through consumption of the biscuits, potato chips, chocolates, and traditional pastries under the current consumption rate. In addition, notably since the mean Pb levels in all foodstuff samples were found to be higher than the MPL, the special attention must be paid for hazardous substances due to human exposure to their harmful effects through food consumption.

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ارزیابی مخاطره سلامت فلزات سنگین ناشی از مصرف برخی مواد غذایی عرضه شده در شهر همدان، ایران

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چکیده

از آنجا که تاکنون در سطح بین‌المللی در مورد ارزیابی مخاطره سلامت فلزات سنگین در مواد غذایی، از جمله انواع بیسکویت، چیپس سیب‌زمینی، شکلات و شیرینی‌جات سنتی مطالعات اندکی انجام شده است، بنابراین این مطالعه با هدف تعیین محتوی و ارزیابی مخاطره سلامت عناصر سرب، کادمیوم و کروم ناشی از مصرف علائم تجاری مختلف از این مواد غذایی انجام یافت. بدین منظور در این مطالعه توصیفی، ۸۴ نمونه از ۲۸ علامت تجاری مواد غذایی مدنظر از بازار مصرف شهر همدان در سال ۲۰۱۶ تهیه شد. پس از آماده‌سازی و فرآوری نمونه‌ها در آزمایشگاه، محتوی عناصر در آن‌ها توسط دستگاه طیف‌سنج نشری پلاسمای جفت القایی خوانده شد. همچنین پردازش آماری داده‌ها نیز با استفاده از نسخه ۱۹ نرم‌افزار SPSS و کاربرد آزمون‌های تحلیل واریانس یک‌طرفه و تی تک‌نمونه ای انجام شد. نتایج نشان داد که میانگین غلظت عنصر سرب بر حسب میلی‌گرم در کیلوگرم در نمونه‌های بیسکوئیت، چیپس، شکلات و شیرینی سنتی به‌ترتیب با ۰/۷۶ ± ۲/۲۵، ۰/۳۵ ± ۱/۶۰، ۰/۶۸ ± ۲/۶۱ و ۰/۶۴ ± ۳/۶۶، از بیشینه رواداری این عنصر بیش‌تر بود. از طرفی، مقادیر شاخص مخاطره سلامت بیان‌گر آن بود که با توجه به نرخ مصرف فعلی، مصرف مواد غذایی مورد مطالعه بزرگسالان و کودکان را با خطر بالقوه مواجه نمی‌کند. به هر حال، با توجه به تجاوز میانگین غلظت سرب در همه نمونه‌های مواد غذایی از بیشینه رواداری، در خصوص توجه جدی به تخلیه آلاینده‌ها در محیط‌زیست و پایش باقی‌مانده مواد شیمیایی، به‌ویژه فلزات سنگین در مواد غذایی توصیه می‌شود.

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