# Health Assessment of Heavy Metal Pollution (Cadmium, Lead, Arsenic) in Citrus Marketed in Tehran, Iran, 2015

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## A-R-T-I-C-L-EI-N-F-O

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### A-B-S-T-R-A-C-T

Background & Aims of the Study: Today, the environment pollution with heavy metals has increased. It is important to study various types of pollutions specially those regarding fruits. The effect of pollutions on food safety for human consumption is a global concern. This study was conducted for health assessment of heavy metals pollution (cadmium, lead, and arsenic) in citrus marketed in Tehran, Iran in 2015. Materials & Methods: After collecting and preparing 2 samples from each citrus species (tangerine, grapefruit, sweet lime, sour orange, orange) with acid digestion method, the citrus pulp and peel were surveyed. Inductively coupled plasma optical emission spectrometry (ICP-OES) was used to determine the concentrations of heavy metals with three replications. Moreover, SPSS version 19 was employed to perform statistical analysis. Results: The results showed that the concentration average of Cadmium, Lead and Arsenic in citrus samples of the pulp parts were 19.73, 42.95 and 2.30 mg/kg and in peel parts were 20.09, 42.71 and 2.12 mg/kg, respectively. The average concentrations of heavy metals were higher than WHO maximum permissible limits. **Conclusions:** Based on these results, consumption of citrus species has no adverse effect on the consumers' health (except Sweet lime, Orange, Tangerine and Grapefruit in lead is risky for adults and Sweet lime and Orange that Health Index in Lead and Arsenic and Sour Orange, Tangerine and Grapefruit that Health Index in Lead is more than 1 and is risky for children). Thus, individuals living in Tehran should be cautious about using these citrus fruits and researchers should try to obtain national standards in the field of entering these metals to food in environmental conditions that are in Iran.

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# Background

Fresh fruits and vegetables are considered as food because they have vitamins, mineral salts, water, calcium, iron, sulphur and potash (1). Citrus are the most important tropical and subtropical fruits in the world named scientifically "Citrusspp.", as family "Rotaceae" and subfamily "Auranitideae". They are rich in vitamins A, B, C, fiber, carbohydrate, calcium, potassium, niacin and folic acid. They also reduce blood cholesterol, prevent viral infections and reduce the risk of colon and stomach cancers (2). The vitamin C is in citrus increases iron absorption (3,4). They usually compose 85- 90 percent of water (5). They grow virtually in 50 countries around the world which are known for their good taste and quality. Their genus is about 60 species. Species such as orange, lemon, grapefruit and sour orange are important from an economic standpoint (6). Fruits are useful for human health in order to prevent and treat diseases.

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Food safety is an important issue in the world. Over the past few decades, increasing demand for food safety has raised research regarding the risk which is associated with consumption of food contaminated by pesticides, heavy metals and toxins (7). Heavy metals are main pollutants of food supply and the most important problem for our environment (8). Pollutants are disrupting agents in ecosystems and heavy metals even have been important in low levels because of problems with toxicity, persistence and bioaccumulation in the environment (9). Eight cases of the most common contaminated metals were presented by American Environmental Protection Agency (AEPA) which are Arsenic, Cadmium, Zinc, Lead, Nickle, Mercury, Copper and Chromium (10). Today, human activities in mining, smelting, refining of metals, industrial effluents and the use of chemical fertilizers have led to release large amounts of heavy metals into the environment and have created serious problems for environment and human health (11-13).

In recent years, heavy metal elements in fertilizers that cause environmental pollution are used in agriculture widely. Some of these harmful heavy metal elements such as lead, cadmium, mercury, chromium and arsenic are transferred to fruits and other farm crops (14). Heavy metals are considered as one of the main obstacles in the production of horticultural crops in many parts of the world, especially in arid and semi-arid areas like Iran (15). They are produced by agricultural technologies in fruits and vegetables. Some of these technologies are irrigation with wastewater, use of organic and mineral fertilizers containing heavy metals and use of pesticides that have chemical elements in their structures (16,17). One of the major pollutants is heavy metals in fruits and vegetables (18). Monitoring of heavy metals in fruits is an important issue with regards to human health (19). Nutritional adverse effects of heavy metals, as well as some of their beneficial role in human life have been reported (20).

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The major contaminants of food supply was cadmium, lead and mercury, also these metals may be considered as the most important environmental contaminants, some metal such as iron, zinc and copper are necessary for human body metabolism (8). The accumulation most of heavy metal in different body organs cause the side effects because they are not biodegradable and have long biological half-lives (20,21).

Cadmium has caused acute inflammation of the gastrointestinal tract (22). It is an effective factor in heart diseases and blood pressure (23). The most important effects of Lead can be named disorder in nerve system (central CNS, peripheral PNS), reduction in neural connection and behavioral difficulties (22). Arsenic leads to dwindle bone marrow, abnormal black in skin, red blood cells rupture, an enlarged liver and peripheral neural disorders in humans (24). Apau et al (2014) studied health risk assessment of Zn, Fe, Cu and Cd in tubers from Markets in Ghana; they posed consumers are not in danger as far as these metals are concerned (25). Chen et al (2014) studied health risk assessment of Cr, Cd, Pb contents of vegetables which are grown around the battery production area and indicated that vegetable consumption does not create a problem for the health of the local population in Changxing County, China (26). Mausi et al (2014) reported that mango and orange fruits sold in Eldort town of Kenya had no health risks to consumers based on their Daily Intake of Metal (D.I.M) levels, as the values were within provisional daily tolerable intake standards of World Health Organization (WHO) (27).

# Aims of the study:

Since these heavy metals (Arsenic, Lead and Cadmium) have adverse effects on human health and also because of the importance of these metals in terms of health and hygiene in individuals, this study aims to investigate the health assessment of heavy metals pollution (Cadmium, Lead, and Arsenic) in citrus marketed in Tehran city, Iran, 2015.

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# **Materials & Methods**

## Sampling and sample analysis

In this study, 10 samples of citrus species (tangerine, grapefruit, sweet lime, sour orange, orange) were bought from different markets that are located in Mirdamad Boulevard in Tehran in 2015 in order to analyze their Arsenic, Lead, and Cadmium contents in pulp and peel of these samples.

All specimens were washed with water and cleaned by distilled water to remove airborne pollutants. Then, their pulp and peel were airdried separately to reduce water content of fruit samples. Afterwards, all samples were dried by oven at 70-80 C for 24 h in order to eliminate all moistures. Finally, dried samples were powdered by the grinder (28). After samples were powdered and transported to the laboratory, 0.2 g of each powdered sample was weighted and each sample was poured into special lidded cans and 4 mL of concentrated nitric acid was poured on each samples. Afterwards, they were put in water bath model WNB-14 Memmert at 65 °c for 60 minutes. Afterthat, temperature was increased to 100 °C and samples were placed in water bath for 90 minutes. Next, cans containing samples were removed from water bath and they reached to laboratory temperature, 0.2 cc hydrogen peroxide 0.037 was added to them and they were released half an hour for completing the process. Then, we used 20 flasks (25 mL) for 20 samples that were laid funnel and paper filter on them. Samples were poured on each of these filters. Then, they were released until they became smooth and were extracted completely. Afterwards, extracts were reached to a volume of 25 cc by deionized water (29). Finally, ICP-OES (Varian 710-Es) was used for analyzing the content of Arsenic, Lead and Cadmium in samples with three replications.

### Human health risk assessment

Estimated Average Daily Intake (EADI) was used to determine the long-term health risks

creating for consumers who use metals in food that is as follows Equation (25):

$$EADI = \frac{C \times F}{W \times D}$$

Where C represents the concentration of metal in each commodity (mg/kg); F indicates the mean annual intake of food per person; D indicates the number of days in a year (365 days) and W represents the mean body weight (70 kg for adult and 15 kg for children)

The WHO has set values for toxicity, called acceptable daily intakes (ADIs) for a large number of chemicals, including some essential trace elements (30). The health index was computed by dividing the EADI by the ADIs (mg/kg/day) established by FAO/WHO Codex Committee (31,32). The following equation is Health Index:

 $HI = \frac{EADI}{ADI}$ 

When the HI>1, the food is a risk for consumers. When the HI<1, the food is acceptable for consumers (25).

# **Statistical analyses**

SPSS version 19 was used in order to analyze the obtainable data. Data were analyzed, using One-sample t-test in order to compare the average concentration of these heavy metals in peel and pulp of citrus with international standards.

# **Results**

The contents of cadmium, lead and arsenic in the analyzed citrus samples are shown in Table 1. The contents of Lead, Arsenic and Cadmium in peel of samples (P-value was less than .05) were higher than WHO maximum permissible limits (0.2, 0.5, 0.1 mg/kg, respectively). Peel had the highest content of Lead in comparison with Arsenic and Cadmium (42.71 mg/kg) while the lowest content of heavy metal was Arsenic in peel (2.12 mg/kg). Moreover, the contents of Lead. Arsenic and Cadmium in the pulp of citrus were higher than WHO standards (0.2, 0.5, 0.1 mg/kg). Pulp had the highest

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content of Lead (42.95 mg/kg) while the lowest content of heavy metals was Arsenic (2.30 mg/kg).

The health risk assessment due to heavy metals encountered in citrus species is summarized in Table 2 and 3. The results showed that EADIs ranged between 0.0001 to 0.046 mg/kg/day and HI ranged from 0.03 to 15.3 for analyzed heavy metals.

	ulp of citrus (mg/	g/kg).				
Citrus	Heavy Metal	Standard level	Т	Degree of	Mean	Standard deviation
				Freedom		
	Arsenic	0.5	4.81	9	$2.12 \pm 0.001$	1.06
Peel	Cadmium	0.1	15.13	9	$20.09 \pm 0.00$	4.17
	Lead	0.2	2.91	9	42.71±0.017	46.16
	Arsenic	0.5	4.08	9	$2.30 \pm 0.003$	1.39
Pulp	Cadmium	0.1	25.73	9	19.73±0.00	2.41
	Lead	0.2	3.60	9	$42.95 \pm 0.006$	37.46

Table 2) Acceptable and estimated daily intakes and health index for heavy metals found in citrus samples in adults

		Heavy Metal			
Citrus	health index	Arsenic	Cadmium	Lead	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Sweet lime	EADI (mg/kg/day)	0.0004	0.0028	0.008	
	HI	0.2	0.04	2.6	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Orange	EADI (mg/kg/day)	0.0004	0.0023	0.01	
-	HI	0.2	0.03	3.3	
<b>C</b>	ADI (mg/kg/day)	0.002	0.06	0.003	
Sour orange	EADI (mg/kg/day)	0.0002	0.0024	0.001	
	HI	0.1	0.04	0.3	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Tangerine	EADI (mg/kg/day)	0.0001	0.0026	0.0036	
	HI	0.05	0.04 1.2		
<b>a a b</b>	ADI (mg/kg/day)	0.002	0.06	0.003	
Graperruit	EADI (mg/kg/day)	0.0001	0.04 0.06 0.0023 0.03 0.06 0.0024 0.04 0.06 0.0026 0.04 0.06 0.002 0.04 0.06 0.002 0.03	0.0036	
	HI	0.05	0.03	1.2	

 Table 3) Acceptable and estimated daily intakes and health index for heavy metals found in citrus samples in children

		Heavy Metal			
Citrus	health index	Arsenic	Cadmium	Lead	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Sweet lime	EADI (mg/kg/day)	0.0022	0.013	0.03	
	HI	1.1	0.21	10	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Orange	EADI (mg/kg/day)	0.002	0.011	0.046	
	HI	1.02	0.18	15.3	
G	ADI (mg/kg/day)	0.002	0.06	0.003	
Sour orange	EADI (mg/kg/day)	0.001	0.01	0.009	
	HI	0.5	0.16	3	
	ADI (mg/kg/day)	0.002	0.06	0.003	
Tangerine	EADI (mg/kg/day)	0.0005	0.01	0.017	
	HI	0.25	0.16	5.6	
Cara a farait	ADI (mg/kg/day)	0.002	0.06	0.003	
Graperfult	EADI (mg/kg/day)	0.0007	0.012	0.017	
	HI	0.35	0.2	5.6	

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# Discussion

Contamination of food in developing countries is 13% more than developed countries. Sources of pollution are different completely, ranging from industrial factors and traffic problems to the purification of sludge, agricultural activities such as the use of chemical fertilizers, animal fertilizers, compost and pesticides (33,34). Heavy metals and all of their destructive effects are main air pollutants of large cities (35).

Fruits and vegetables are essential parts of human diet; so, the impact of heavy metals pollution in them is important. Consumption of fruits contaminated with heavy metals are hazardous to human health; hence, heavy metal contamination in food is one of the main aspects of food quality assurance (36-38).

The results of this study indicate that the average concentrations of Arsenic, Cadmium and Lead in the peel of citrus were  $2.12\pm0.001$ ,  $20.09\pm0.00$ ,  $42.71\pm0.017$  mg/kg and in the pulp of these samples were  $2.30\pm0.003$ ,  $19.73\pm0.00$ ,  $42.95\pm0.006$  mg/kg that were higher than standard levels of WHO and FAO.

In this regard, studies have shown that the high concentration of these heavy metals can be due to the use of pesticides, fertilizers, sewage for cultivating fruits (27) and other pollutants that are in irrigation water and farm soil which can be attributed to pollutions that resulted from the highways traffic (39). Another possible explanation to high levels of these heavy metals in citrus of this study could be atmospheric emissions from vehicles or machines which placed the metals on fruits during transportation and the places where fruits are for sale in open markets along roadside (40).

Results of this study are compatible with those of Kalagbor et al that their studies conducted on sweet orange in Khanaand concentration of Chromium, Copper, Nickle and Lead in peel and pulp of orange are more than standard level (41); also, with the findings of Mausi et al in study of the concentration of heavy metals

(Lead and Chromium) in fresh orange in Eldoret of Kenya that is more than standard level (27). Also, the results of Kalagbor and Diriin, a study on a farm in Kaani, Bori and Rivers states in Nigeria that concentration of Lead in orange is more than standard level (42). As shown in table 2 and 3, citrus consumption (except Sweet lime, Orange, Tangerine and Grapefruit that Health Index in Lead is more than 1 and is risky for adults) is not risky for adults because health index is less than 1. Moreover, for children (except Sweet lime and Orange that health index in Lead and Arsenic and Sour Orange, Tangerine and Grapefruit that Health Index is in Lead more than 1 and is risky for children), it is not risky either because health index is less than 1.

Ferre- Huguet et al. studied the risk assessment of metals from consuming vegetables, fruits and rice grown on soils irrigated with waters of the Ebro river in Catalonia, Spain. They found that when the minimum concentration of elements in food is considered, the possible risk of heavy metals is low in all of the studied age groups; however, when a high level of heavy metals concentration in food is considered, the possible risk of being polluted with arsenic is high among children for both boys and girls as well as elderly men (43).

Bo et al study about the risk assessment which has been done in Beijing, China, through consumption of vegetables; their results revealed that among seven studied elements (cadmium, lead, nickel, copper, chromium, arsenic, and zinc), arsenic had the highest amount in HI level (44).

# Conclusion

This study surveyed the health assessment of Arsenic, Lead and Cadmium in five consumed citrus species (orange, grapefruit, sweet lime, tangerine and sour orange) in Tehran city, Iran, comparing the average concentration of Arsenic, Lead, and Cadmium in pulp and peel of citrus with international standard levels

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shows that there is a significant difference between the concentration of these metals and standard level of WHO/FAO at a significant level of .05. As a result, the average concentration of Lead, Cadmium and Arsenic in peel and pulp of citrus is more than specified permissible level that is set by WHO/FAO and consumable citrus are polluted with these metals in Tehran. Therefore, researchers should try to obtain national standards in the field of entering these metals to food in environmental conditions that are in Iran.

Concentration of metallic pollutants in citrus is determined annually and the use of international experience to investigate the adverse effects of heavy metals on human and the ways of reducing pollution with heavy metals in citrus are considered.

# Footnotes

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### **Conflict of Interest:**

The authors declared no conflict of interest.

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