

Contamination and health risk assessment of heavy metals in juices and nectars of five fruit species from Egypt's market

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Abstract:

Background: The evaluation of commercial fruit juices is a vital issue for consumer safety, as they are commonly consumed worldwide. Heavy metals end up in food because they are part of the make-up of air, water and soil. Also, manufacturing and packaging processes can also introduce heavy metals to food.

Materials and Methods: The objective of this study was to estimate heavy metal contamination and potential health risks of exposure to heavy metals along in both juice and nectar from five commonly consumed fruit species (citrus, peach, guava, apple, and mango) in Egypt. We compared the heavy metals content found in our analyzed fruit juices and nectars with different international organizations (USEPA, WHO, EU).

Results In general, the analyzed heavy metal concentrations were the highest in juice samples, particularly of guava and apple, whereas nectar samples were the lowest. The obtained values of iron and copper in juice samples of all five fruit species, besides lead in citrus and peach nectar, were above the maximum permissible limits set by USEPA and WHO. Heavy metal concentrations varied among the fruit samples. The values of calculated target hazard quotients (THQ) demonstrated that consumption of the fruit juices or nectars posed very little potential health risk, where THQ values were less than 1.0. These indicated that no direct hazard could be happened to human health in spite of presence of the investigated metals in analyzed juice or nectar at those limits of concentrations. For the non-carcinogenic health risk, according to the relative risk (RR) value, the data showed high value of RR, which was more than 1.0, for copper of juice samples from guava, apple and mango. For the carcinogenic health risk (CR), our data revealed that values for lead of guava and apple juice, were more than the acceptable or tolerable risk of 1×10^{-6} , indicating that there was a potential carcinogenic risk when both adults and children were exposed to lead. For carcinogenic health risk for adults and children, our data revealed that children were more vulnerable and sensitive than adults when exposed to the same metal.

Conclusion: These findings may be valuable for taking protective measures in reducing contamination levels of heavy metals in fruit juice.

Key Word: heavy metals; human health risk; fruit juices and nectars; non-carcinogenic risk; carcinogenic risk.

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I. Introduction

Growing and an increasing demand for food safety has drawn the attention of many researchers to assess the potential risks associated with consumption of contaminated foodstuffs that is pesticides, heavy metals, and/or toxins in vegetables¹. In recent years, an increase of interest in fruit and vegetable juices is observed in the world. This is due to increased consumer awareness in the aspect of search for the products rich in health-promoting components². The fruit juices and nectars dominate an important part of the beverage market in Egypt and other countries. They are sources of vitamins, especially vitamin C, folic acid, and vitamins from group B, as well as minerals mainly manganese and potassium but also phosphorus, magnesium, calcium, zinc, and selenium. They contain fiber, which regulates the metabolic processes, as well as antioxidant and bioactive substances. In addition, they enhance the attractiveness of meals due to their pleasant taste, flavor, consistency, and color. In human diet, juices are an important source of water, which is an essential component for life. Juices contain minerals, which are naturally present in the fruit components and are associated to the uptake of elements by plants from the surrounded environment (soil, water, air)³.

The products labeled as "juice" must contain 100% fresh fruit, that has been processed after extracting water from it, without preservatives or sweeteners and no artificial colors and may contain pulp of the fruit itself;

whereas nectar is diluted fruit juice which has been mixed in with water, additives, sweeteners, and preservatives. Therefore, nectars are generally cheaper than the soluble solids of the fruit itself, making this category more affordable for consumers in the intermediate per-capita income range⁴.

Fruits contamination by heavy metals is one of the main problems that happen due to an economic development by the industrial and novel agricultural practices⁵, and to fulfill the advanced demands of food production for human consumption⁶. The heavy metals can reach inside the human body system via different routes, food, air, and water; they have long biological half-time and are not biodegradable⁷. The elements are divided into two groups, first, minerals as the main and beneficial group for human health such as Na, Ca, Fe, Mg, K and P; second, heavy metals such as Hg, Sn, Pb, As and Cd that cause serious damage for human health even at low concentrations, 10–50 mg/L^{8,9,10}. Bioaccumulation of heavy metals may occur over a period of time because of their long half-time and hence the undesirable side effects can be occurred as a result of accumulation in the different body organs¹¹. For example, serious risks to human health could be happened as a result of chromium consumption at high concentrations leading to disruption of numerous biochemical processes and diseases such as stomach upset, skin rashes, and lung cancer¹². Likewise, contaminated food with high concentrations of lead and cadmium are found to be associated with severe diseases, teratogenesis and carcinogenesis¹³. Copper toxicity can induce iron deficiency, lipid peroxidation and destruction of membranes¹⁴.

Organic pollutants in food increasing health risks in many worldwide countries including Egypt did not provide safety hazardous chemical standard values in foods¹⁵. Unfortunately, there is no documented database in Egypt so far about the average daily consumption of fruit juices or nectars for adults and children, as well as the estimated rate of contamination with common heavy metals, and whether it falls among the permissible limits approved by specialized international organizations such as USEPA and WHO or exceeds them. However, there are some research studies that have been carried out in Egypt to monitor the rates of contamination of juices with common heavy metals and their associated with the potential human health risk. Some efforts have been done to monitor residues of heavy metals and persistent organic pollutants in various environmental matrices¹⁶. The heavy metals contents in samples from fruit juice and nectar which collected from some supermarkets in Egypt showed that most investigated samples had high limits of iron than the maximum permissible limits (MPL), whereas lead, zinc and cadmium were less than MPL¹⁷. Likewise, juice samples of four fruit species collected from the Egyptian markets had high level of lead exceeding the MPL set by European Union¹⁸.

Therefore, the main objective of this study was to estimate the content of selected trace elements (Cd, Pb, Cr, Fe, Cu, and Zn) in commercial fruit juices and nectars, with determination of the health risk assessment associated with the consumption of that assorted products.

II. Material And Methods

Sample collection:

A total of one hundred juice and nectar samples were collected from different local markets. Fruit juices and nectar samples of five different types (apple, guava, mango, orange, and peach) were bought from different supermarkets. The most frequently consumed brands were selected from local companies during 2020. A total of 50 fruit juice and 50 nectar samples were studied.

Chemicals and Heavy metal analysis:

All chemicals used were Analytical Grade Reagents purchased from Sigma-Aldrich Corp, Chem Service (West Chester, PA, USA). The element standard solutions used for creating the calibration curves were prepared from a 1000 mg/L Merck[®] stock solution of the relevant element. Thermo Elemental model, Solar M Atomic Absorption Spectrophotometer (SOLARR M5, Ser No GE600531, made in England) was used for all the measurements. The current, wavelength and slit band width of each element were adjusted automatically by the instrument software (Thermo elemental version, 2003). Determination of investigated heavy metals in fruit juice samples were performed according to the official method of analysis¹⁹. About 1 gram of juice sample, in triplicates, was accurately weighed and transferred into Kjeldahel flask and digested with 10 mL of HNO₃ acid and 1 mL of H₂O₂, then made up the volume to 25 mL by deionized water. Percentage recovery tests were carried out for the six metals by spiking 0.002, 0.05, 0.05, 1.0, 0.1 and 0.1 µg mL⁻¹ of Cd, Pb, Cr, Fe, Cu and Zn; respectively; to the similarly prepared samples as those unknown samples. The range of these percentage recoveries were between 93% -98% Table No 1.

Table No. 1: The Limits of Detection, Limit of Quantification, Percentage Recoveries and Maximum Permissible Limits (mgL⁻¹ or mgKg⁻¹) of investigated heavy metals by different international organizations.*

| | Cadmium | Lead | Chromium | Iron | Copper | Zinc |
|-------------------|---------|--------|----------|--------|--------|--------|
| LOD | 0.003 | 0.014 | 0.01 | 0.0045 | 0.0043 | 0.0033 |
| LOQ | 0.0093 | 0.0433 | 0.03 | 0.015 | 0.0143 | 0.011 |
| % Recovery | 95 | 97 | 98 | 96 | 95 | 93 |
| MPL ²⁰ | 0.003 | 0.010 | 0.05 | 0.3 | 2.00 | 3.0 |
| MPL ²¹ | 0.005 | 0.015 | 0.10 | 0.3 | 1.30 | 5.0 |
| MPL ²² | 0.030 | 0.300 | 0.050 | 0.2 | 3.50 | 5.0 |
| RfD | 0.001 | 0.004 | 0.003 | 0.30 | 0.040 | 0.30 |

*LOD, Limit of Detection; LOQ, Limit of Quantification; MPL, Maximum Permissible Limits; RfD, Oral Reference Dose.

Health risk assessment:

Non-carcinogenic health effect:

Human health risk assessment is a process used to estimate the health effects that might result from exposure to carcinogenic and non-carcinogenic chemicals. The risk assessment process is made up of four basic steps: hazard identification, exposure assessment, toxicity (dose-response) assessment, and risk characterization²³.

1. Estimated Daily Intake (EDI):

EDI is the calculated amount of intake of heavy metals per kilogram of body weight and RfD is the reference dose of heavy metal oral intake (mg kg⁻¹ day⁻¹). The RfDs for cadmium (Cd), Lead (Pb), chromium (Cr), Iron (Fe), copper (Cu) and zinc (Zn) are 0.001, 0.004, 0.003, 0.3, 0.04 and 0.3 mg kg⁻¹ day⁻¹, respectively Table No 1. The value of EDI was calculated using equation NO.1^{24,25}:

$$EDI = (C \times IR \times EF \times ED) / (BW \times AT) \dots (1)$$

Where: C is the trace element concentration in Juice and nectar (mg/kg), IR is average daily consumption of juice in Egypt (0.32 kg person⁻¹day⁻¹)²⁶, EF is exposure frequency (365 days year⁻¹), ED is exposure duration (70 years, equivalent to the average lifetime), BW is the average body weight (kg) (70 kg for adults, 20 kg for children) and AT is the average exposure time for non-carcinogens (365 days year⁻¹ × ED = 25,550).

2. Target hazard quotient (THQ):

THQ is the ratio of the exposure dose to the reference dose (RfD). The THQ used for calculation of non-carcinogenic hazard of ingestion of heavy metals. When THQ > 1, there is a probability of potentially harmful effects, and when THQ ≤ 1, there is no probability of unfavorable effects. The dose calculations were performed using standard assumption from the integrated USEPA risk analysis equation No.2^{27,25}:

$$THQ = EDI / RfD \dots (2)$$

3. Total Target Hazard Quotient (TTHQ)

TTHQ is used to evaluate the potential risk to human health when more than one heavy metal is involved²⁸. TTHQ was calculated as the sum of target hazard quotients (THQ) for trace metals. Since different pollutants can cause similar adverse health effects, its often appropriate to combine THQS associated with different substances, TTHQ was calculated using equation NO.3^{29,25}:

$$TTHQ = THQ (\text{toxicant 1}) + THQ (\text{toxicant 2}) + \dots THQ (\text{toxicant n}) \dots (3)$$

Carcinogenic health effect (Cancer risk, CR):

For carcinogen, which USEPA identifies by a weight-of-evidence classification of the chemical, the estimated daily dose, and the cancer slope factor (CSF) are multiplied together to find the LCR posed by the chemical. CSFs are estimates of carcinogenic potency and are used to relate estimate daily dose of a substance over a lifetime probability of excess tumors. CR was calculated using equation NO.4²⁷:

$$CR = EDI \times CSF \dots (4)$$

Where, CR is carcinogenic risk, EDI is the estimated daily intake of each heavy metal (mg kg⁻¹ day⁻¹), and CSF (mg kg⁻¹ day⁻¹) is the cancer aslope factor and is defined as the risk generated by a lifetime average amount of one mg kg⁻¹ day⁻¹ of carcinogen chemical and is contaminant specific. For a single heavy metal, a RISK of less than 1 × 10⁻⁶ is regarded as inconsequential, and the cancer risk can be ignored; while a RISK of more than 1 × 10⁻⁴ is regarded as unacceptable, and the cancer risk is concerning²⁸.

Relative risk (RR):

The RR of analyzed heavy metals for both carcinogenic and non-carcinogenic effects, which can be helpful to recognize the most harmful contaminants. RR was calculated using equation No.5³⁰, where all parameters have been previously defined:

$$RR = C / RfD \dots (5)$$

III. Result and Discussion

Element quantification:

The mean levels of the analyzed metals (Cd, Pb, Cr, Cu and Zn) in the juice and nectar samples reported in this study are shown in Table No. 2. The concentrations of these metals are compared with the Maximum Permissible Limits (MPL) set by different international organizations, viz.^{20,21,22} Table No. 1. The results showed that iron was the most abundant element with a range of 1.8-14.75 mg kg⁻¹ in all juice and nectar samples, while Cd was the least concentration with mean value 0.0033 mg kg⁻¹ that was detected in only one sample among all juice and nectar samples.

Cadmium: Among 100 juice and nectar samples, cadmium not detected except in only one sample (Guava juice), and its mean concentration was 0.003 mg kg⁻¹ Table No. 2. Cadmium is a toxic metal and there is no evidence of its essentiality to humans. Oral ingestion of cadmium lead to increases in hematological, liver, kidney, gastrointestinal, neurological, and testicular effects³¹. In the present study, cadmium concentration in guava juice is 0.003 mg kg⁻¹, which is within or lower than the MPL set by^{20,21} or²² in drinking water.

Lead: Lead, Pb, is another highly toxic element³². The mean concentration of lead in the present study ranges from 0.023-1.00 mg kg⁻¹ (Table 2). The higher concentration of Pb was detected in peach nectar sample, and the lowest concentration was detected in Guava juice. The mean concentration of lead in peach nectar samples is higher than the maximum permissible limits (MPL) set by different international organizations^{20,21,22} Table No. 1. Also, Pb concentrations in juice samples of guava and apple were higher than Pb limit set by both^{20,21}. The main symptoms of lead poisoning are of three types: gastrointestinal, neuromuscular, and neurological³³. Gastrointestinal absorption of water-soluble lead appears to be higher in children than in adults³⁴. Similar study conducted by¹⁴ showed that Pb concentrations in most samples of juice or nectar of five fruit species (apple, guava, mango, orange and peach) collected from local markets in Egypt were lower than the allowed limits except four orange samples, one apple juice sample and one peach nectar sample which exceeded the MPL set by²².

Chromium: Chromium element was detected only in apple juice and mango nectar samples with mean concentrations of 0.002 and 0.0002 mg kg⁻¹, respectively, at which are lower than Cr limit set by different international organizations^{20,21,22} Table No. 2. Chromium has oxidation states ranging from chromium (II) to chromium (VI). In humans, chromium (III) is an essential nutrient. Cr plays a role in glucose, fat, and protein metabolism. However, exposure to chromium (VI) via ingestion may cause adverse health effects including respiratory and gastrointestinal problems³⁵.

Iron: Iron element was detected in all analyzed juice and nectar samples with mean concentration level ranges from 1.8-14.75 mg kg⁻¹ Table No. 2. The data showed that all samples of fruit juices and nectars had iron levels that exceeded the MPL of 0.30 mg L⁻¹ (mg Kg⁻¹) set by^{20,21}. The highest levels of iron were detected in juice samples from guava (12.07 mg kg⁻¹), apple (13.13 mg kg⁻¹) and mango (14.75 mg kg⁻¹). Similar findings were reported by¹⁷ who found that most samples from fruit juices or nectars of five fruit species (apple, guava, mango, orange and peach) collected from local markets in Egypt had higher iron levels than the allowed limits. Iron is an essential element in human nutrition and is required in the production of red blood corpuscles, oxygen transportation and the functioning of many enzymes in the organism. It also plays a significant role on vitamin A and iodine metabolism³⁶.

Copper: The mean concentrations of copper in this study ranges from 0.5-11.01 mg kg⁻¹ Table No. 2. The data showed that juice samples from peach, guava, apple, and mango had iron levels exceeding the MPL (1.3 ~ 3.5 mg kg⁻¹) set by different international organizations, i.e.^{20,21,22} Table No. 1. Copper is an essential for good health, it is required in normal carbohydrate and lipid metabolism and blood formation³⁷. However, exposure to higher doses of copper can be harmful causing gastrointestinal problems, nausea, vomiting, diarrhea as well as liver and kidney damage.

Zinc: In the present study, the mean concentration of zinc ranges from 0.4-1.65 mg kg⁻¹ Table No. 2. The data showed that Zn levels found in all samples of fruit juices or nectars were less than these set by different international organizations^{20,21,22} (3.0 - 5.0 mg Kg⁻¹) Table No. 1. Zinc is an essential nutrient for humans, at which involved in glucose and lipid metabolism, hormone function, wound healing and it also helps in proper hair growth³⁷, however, high levels of Zn can cause gastrointestinal distress³⁸.

High concentration of the heavy metals found in fruit juices or nectars is due to the concentration of these metals in raw materials and also influenced by the manufacturing process⁸. The high metal concentration in raw materials depends on several factors including, the nature of the fruit, the mineral composition of the soil from which it originated, the composition of the irrigation water, the weather conditions, the agricultural practices such as the types and amounts of fertilizers and pesticides used, and soil composition. However, consuming fruit juices that

exceed maximal permissible values does not necessarily imply an increased risk for human health, since the amount of fruit juice consumed per day is expected to be lower than the amount of water³⁹.

Table No. 2: Concentrations of heavy metals in juice and nectar samples of five fruit species (mg kg⁻¹)

| Metal | Citrus species samples | | | | | | | | | |
|-------|-----------------------------------------------|----------------|----------------|----------------|------------------|----------------|-------------------|----------------|-----------------|--------------------|
| | Mean concentration (mg kg ⁻¹ ± SD) | | | | | | | | | |
| | Citrus | | Peach | | Guava | | Apple | | Mango | |
| | Juice | Nectar | Juice | Nectar | Juice | Nectar | Juice | Nectar | Juice | Nectar |
| Cd | ND | ND | ND | ND | 0.003 ± 0.001 | ND | ND | ND | ND | ND |
| Pb | ND | 0.11 ± 0.01 | ND | 1.10 ± 0.30 | 0.023 ± 0.002 | ND | 0.14 ± 0.038 | ND | ND | ND |
| Cr | ND | ND | ND | ND | ND | ND | 0.002 ± 0.0006 | ND | ND | 0.0002 ± 0.0001 |
| Fe | 5.10 ± 1.80 | 1.80 ± 0.80 | 8.10 ± 2.30 | 3.10 ± 0.80 | 12.07 ± 3.05 | 4.48 ± 1.64 | 13.13 ± 2.38 | 2.24 ± 1.01 | 14.75 ± 2.75 | 4.63 ± 1.11 |
| Cu | 2.50 ± 0.80 | 0.55 ± 0.01 | 3.66 ± 0.80 | 0.50 ± 0.10 | 11.01 ± 3.4 | 1.34 ± 0.56 | 6.70 ± 1.31 | 1.25 ± 0.53 | 9.55 ± 2.93 | 1.05 ± 0.16 |
| Zn | 0.70 ± 0.20 | 0.44 ± 0.01 | 0.92 ± 0.20 | 0.70 ± 0.20 | 1.50 ± 0.75 | 0.59 ± 0.09 | 1.45 ± 0.34 | 0.61 ± 0.26 | 1.65 ± 0.75 | 0.69 ± 0.17 |

Human health risk assessment:

1. Estimated daily intake (EDI) and target hazard quotient (THQ)

The estimated daily intake (EDI) values for the analyzed metals from citrus juice and nectar for adult and children are presented in Table No. 3. The EDI values for all the investigated metals in either juice or nectar samples from the selected fruit species are less than the oral reference doses (RfD) of these metals based on the recommendations of United States Environmental Protection Agency (USEPA). The EDI values for the analyzed metals ranged from 0.00032 (Zn) to 0.00816 (Fe) in juice samples; and ranged from 4.57E-05 (Pb) to 0.00288 (Fe) in nectar samples. The data showed also that the Target Hazard Quotient (THQ) of each metal from the ingesting of citrus juice and nectar was generally less than 1.0. Total Target Hazard Quotient (TTHQ) values were also less than 1.0. This suggests that consuming of either juice or nectar from citrus contaminated with these limits of metal concentrations might not have adverse effects on the human health Table No. 3.

The EDI, THQ and TTHQ values of analyzed metals from peach juices and nectars for adults and children are summarized in Table 3. The calculated EDI values for juice and nectar samples were lower than the RfD values. The EDI values for the analyzed metals in juice ranged between 0.000411 (Zn) to 0.01296 (Fe) mg/kg and ranged between 0.000046 (Pb) to 0.00496 (Fe) in nectar samples. In both cases for the adults and children, the THQ values for the analyzed metals were lower than 1.0 and ranged from 0.001371 (Zn) to 0.144 (Cu) 0.1440 in juice samples, and from 0.001067 (Zn) to 0.40 (Pb) in nectar samples. TTHQ values were also below 1.0. These results suggested that no direct hazard to human health could be happened, in spite of presence of the investigated metals in pear juice or nectar at those limits of concentrations.

Likewise, the EDI of analyzed metals from guava juices and nectars for adults and children were lower than the RfD values Table No. 3. The EDI values reported for metals in juice ranged between 0.151E-06 (Cd) to 0.019312 (Fe) mg/kg and ranged from 0.00027 (Zn) to 0.007168 (Fe) in nectar samples. In both cases for the adults and children, the THQ values for the analyzed metals were less than 1.0 and ranged from 0.001509 (Cd) to 0.4404 (Cu) in juice samples, and from 0.000899 (Zn) to 0.0536 (Cu) in nectar samples. TTHQ values for total detected metals were less than 1.0. These results suggested that the populace consuming juices or nectars of the selected fruits would not experience significant health risks from the intake of individual or total analyzed metals.

Similar data for the calculated EDI of analyzed metals from apple juices and nectars either for adults or children are reported in Table No.3. All EDI values for the analyzed metals were lower than the RfD, where ranged from 9.14E-07 (Cr) to 0.021008 (Fe) in juice samples and ranged from 0.000279 (Zn) to 0.0020 (Cu) in nectar samples. In both cases for the adults and children, the THQ values for the analyzed metals were less than 1.0 and ranged from 0.000305 (Cr) to 0.268 (Cu) in juice samples, and from 0.00093 (Zn) to 0.050 (Cu) in nectar samples. Also, TTHQ values were below 1.0. These results suggested that the populace consuming juices or nectars of the selected fruits would not experience significant health risks from the intake of individual metals.

The EDI values for the investigated metals in all mango juice or nectar samples were lower than the RfD Table No. 3 indicating no direct hazard could be happened to human health in spite of their presence. The EDI values reported for metals in juice ranged between 0.000755 (Zn) to 0.0236 (Fe) mg/kg; and between 0.9.14E-08 (Cr) to 0.0074 (Fe) mg/kg in nectar samples. The THQ values in both adults and children for the analyzed metals were less than 1.0, where ranged from 0.002517 (Zn) to 0.38192 (Cu) in mango juice; and between 3.05E-05 (Cr) to 0.04192 (Cu) in nectar samples. TTHQ values were also less than 1.0. These indicated that no direct hazard could be happened to human health in spite of presence of the investigated metals in mango juice or nectar at those limits of concentrations.

Table No. 3: Estimated Daily Intake (EDI), Target Hazard Quotient (THQ) and Total Target Hazard Quotient (TTHQ) for analyzed heavy metals from fruit juice and nectar samples consumed by adults and children.

| Metal | EDI (mg Kg ⁻¹ day ⁻¹ (Adults)) | | THQ (Adults) | | EDI (mg Kg ⁻¹ day ⁻¹ (Children)) | | THQ (Children) | |
|--------|------------------------------------------------------|----------|--------------|---------|--------------------------------------------------------|---------|----------------|---------|
| | Juice | Nectar | Juice | Nectar | Juice | Nectar | Juice | Nectar |
| Citrus | | | | | | | | |
| Cd | ND | ND | ND | ND | ND | ND | ND | ND |
| Pb | ND | 4.57E-05 | ND | 0.01143 | ND | 0.00016 | ND | 0.04 |
| Cr | ND | ND | ND | ND | ND | ND | ND | ND |
| Fe | 0.002331 | 0.000823 | 0.003331 | 0.00118 | 0.00816 | 0.00288 | 0.011657 | 0.00411 |
| Cu | 0.001143 | 0.000229 | 0.028571 | 0.00571 | 0.004 | 0.0008 | 0.100 | 0.020 |
| Zn | 0.00032 | 0.000183 | 0.001067 | 0.00061 | 0.00112 | 0.00064 | 0.003733 | 0.00213 |
| TTHQ | | | 0.032969 | 0.01893 | | | 0.11539 | 0.06625 |
| Peach | | | | | | | | |
| Cd | ND | ND | ND | ND | ND | ND | ND | ND |
| Pb | ND | 0.000046 | ND | 0.01143 | ND | 0.0016 | ND | 0.400 |
| Cr | ND | ND | ND | ND | ND | ND | ND | ND |
| Fe | 0.003703 | 0.001417 | 0.00529 | 0.00202 | 0.01296 | 0.00496 | 0.018514 | 0.00709 |
| Cu | 0.001646 | 0.000229 | 0.04114 | 0.00571 | 0.00576 | 0.0008 | 0.144 | 0.020 |
| Zn | 0.000411 | 0.00032 | 0.001371 | 0.00107 | 0.00144 | 0.00112 | 0.0048 | 0.00373 |
| TTHQ | | | 0.047791 | 0.02023 | | | 0.167314 | 0.04308 |

Table No. 3: (continued)

| Metal | EDI (mg Kg ⁻¹ day ⁻¹) (Adults) | | THQ (Adults) | | EDI (mg Kg ⁻¹ day ⁻¹) (Children) | | THQ (Children) | |
|-------|----------------------------------------------------------|----------|-----------------|---------|------------------------------------------------------------|---------|-------------------|---------|
| | Juice | Nectar | Juice | Nectar | Juice | Nectar | Juice | Nectar |
| Guava | | | | | | | | |
| Cd | 1.51E-06 | ND | 0.001509 | ND | 5.28E-06 | ND | 0.00528 | ND |
| Pb | 1.05E-05 | ND | 0.002629 | ND | 3.68E-05 | ND | 0.0092 | ND |
| Cr | ND | ND | ND | ND | ND | ND | ND | ND |
| Fe | 0.005518 | 0.002048 | 0.007822 | 0.00293 | 0.019312 | 0.00717 | 0.027589 | 0.01024 |
| Cu | 0.005033 | 0.000613 | 0.125829 | 0.01531 | 0.017616 | 0.00214 | 0.4404 | 0.0536 |
| Zn | 0.000686 | 0.00027 | 0.002286 | 0.00089 | 0.0024 | 0.00094 | 0.008 | 0.00314 |
| TTHQ | | | 0.140075 | 0.01919 | | | 0.490469 | 0.06699 |
| Apple | | | | | | | | |
| Cd | ND | ND | ND | ND | ND | ND | ND | ND |
| Pb | 0.00046 | ND | 0.016 | ND | 0.000224 | ND | 0.056 | ND |
| Cr | 9.14E-07 | ND | 0.000305 | ND | 3.2E-06 | ND | 0.001067 | ND |
| Fe | 0.006002 | 0.001024 | 0.000858 | 0.00146 | 0.021008 | 0.00355 | 0.030011 | 0.00512 |
| Cu | 0.003063 | 0.000571 | 0.076571 | 0.01429 | 0.01072 | 0.0020 | 0.268 | 0.0500 |
| Zn | 0.000663 | 0.000279 | 0.00221 | 0.00093 | 0.00232 | 0.00098 | 0.007733 | 0.00325 |
| TTHQ | | | 0.1064 | 0.01668 | | | 0.362811 | 0.05837 |
| Mango | | | | | | | | |
| Cd | ND | ND | ND | ND | ND | ND | ND | ND |
| Pb | ND | ND | ND | ND | ND | ND | ND | ND |
| Cr | ND | 9.14E-08 | ND | 3.0E-05 | ND | 3.2E-07 | ND | 0.00011 |
| Fe | 0.006743 | 0.002114 | 0.00963 | 0.00302 | 0.0236 | 0.0074 | 0.033714 | 0.01057 |
| Cu | 0.004365 | 0.000479 | 0.10912 | 0.01198 | 0.015277 | 0.00168 | 0.38192 | 0.04192 |
| Zn | 0.000755 | 0.000315 | 0.002517 | 0.00105 | 0.002643 | 0.00110 | 0.008811 | 0.00367 |
| TTHQ | | | 0.12127 | 0.01605 | | | 0.424445 | 0.05627 |

ND, not detected.

2. Estimated cancer risk (CR) and relative risk (RR)

RISK is assessed by calculating the incremental probability of an individual developing cancer over a lifetime due to exposure to a potential carcinogen. According to the U.S. EPA, the value of cancer risk in the range of 10^{-6} to 10^{-4} is an acceptable or tolerable risk, a risk of less than 10^{-6} can be ignored, and a risk exceeding 10^{-4} is considered to unacceptable⁴⁰.

For carcinogenic risk, due to the lack of carcinogenic slope factors for Fe, Cu, and Zn, only the carcinogenic risks for other three metals (Pb, Cd, and Cr) were estimated. Among them, the carcinogenic risk of Pb were considered for nectar samples from both citrus and peach. In both adults and children, the calculated cancer risk (CR) values of Pb were 0.0005378 and 0.018824, respectively, for citrus nectar, where were 0.05378 and 0.188253, respectively, for peach nectar, which were higher than the acceptable range of 10^{-6} to 10^{-4} , implying a

potential carcinogenic risk for human health. However, the CR values of Cr for mango nectar, for both adults (1.83E-07) and children (6.4E-07), were lower than the set tolerable limit, indicating that the consumption of mango nectar might not pose carcinogenic risk for Cr Table No. 4.

For juice samples from guava and apple, the data also revealed that the carcinogenic risk of Pb were considered Table No. 4. In both adults and children, the calculated cancer risk (CR) values of Pb were 0.001237 and 0.004329, respectively, for guava juice, where were 0.007529 and 0.026353, respectively, for apple juice, which were higher than the acceptable range of 10^{-6} to 10^{-4} , indicating that the consumption of guava and apple juices might pose carcinogenic risk for human health for Pb. Moreover, the calculated CR value of Cr for apple juice, in both adults (1.83E-06) and children (6.4E-06), showed a slight increase over the acceptable or tolerable risk of 1×10^{-6} , revealing that there was a potential carcinogenic risk when both adults and children were exposed to chromium. Likewise, the carcinogenic risk of cadmium for guava juice was higher than the acceptable range of 10^{-6} to 10^{-4} , especially for the children, indicating that consumption of guava juice might pose carcinogenic risk for cadmium.

Accordingly, by comparing the RISK values, it seems that the Pb was the main pollutant for calculated carcinogenic risk, for samples of citrus and peach nectar, and also for samples of guava and apple juices. Generally, it was obvious that children suffered from more carcinogenic risk than adults, implying that children were more sensitive than adults and vulnerable to heavy metals in the analyzed samples. Similar findings were reported by several researchers^{41,42,43}. This finding may be because the body weight of children is less than that of adults, and compared with adults, children participate in outdoor activities more often^{42,41}.

3. Estimated non-carcinogenic relative risk (RR)

The non-carcinogenic relative risk assessment, or the Relative Risk (RR) is evaluated by comparing an exposure level over a specified time period (e.g., lifetime), with a reference dose derived for a similar exposure period. The Relative carcinogenic risk (RR) values can be also characterized as a Hazard Quotient (HQ)^{43,28}. If the value of HQ or RR is less than one, it is unlikely to create adverse health effects for exposed populations. If the value of RR exceeds one, it is not in the acceptable range, and the greater the value, the greater the probability of the occurrence of adverse health effects⁴⁰.

The results of no-carcinogenic risks (RR) for the analyzed heavy metals are summarized in Table No. 4. Due to the high concentration of Cu and the low value of RfD, this metal showed a higher RR of the juice from guava, apple and mango than other metals, with values more than 1.0, as follows: 2.75, 1.67 and 2.38, respectively. This suggest that consuming such samples of fruit juices may present a high risk to the health, therefore should be highly stressed. For the nectar samples, the data show that the RR for only Pb was > 1.0 , in only peach nectar, indicating a value of 2.50, whereas RR values for the other investigated metals were < 1.0 . It was also obvious that the contribution of the investigated metals to the non-carcinogenic health risk for the selected fruit juices and nectars were found in the order of $Cu > Fe > Zn$.

As such, consuming fruit juices that exceed the maximal permissible values does not necessarily imply an increased risk for adverse human health effects^{44,39}. In general, these results indicated that except such samples, the health risk estimation of other heavy metals, Fe, Zn, Cr and Cd, revealed RR values less than 1.0 suggesting an acceptable level of non-carcinogenic harmful health risk in all samples taken. Similar findings were reported by several researchers^{41,42,43}. This finding may be because the body weight of children is less than that of adults, and compared with adults, children participate in outdoor activities more often^{42,41}.

In order to avoid environmental contamination with heavy metals, a recognized public health hazard worldwide, results reported by another researcher suggested that it should struggle for minimization of metal releases and discharges from anthropogenic activities, thus decreasing metal levels in drinking-water, since this is likely the major source of metal contamination in fruit juices³⁹. This type of study is important as it quantifies the dietary intakes of metals present in fruit juices and broaden the understanding on how fruit juices should be evaluated and considered for balanced diets due to its and nutritional importance³⁹.

Table No. 4: Cancer slope factor (CSF), Estimated cancer risk (CR) and relative risks (RR) of analyzed metals for different types of juice and nectar consumption ♣

| Metal | CSF | CR | | RR | |
|---------------|--------|------------------------------|------------------------------|--------|---------|
| | | Juice | Nectar | Juice | Nectar |
| Citrus | | | | | |
| Cd | 0.380 | -* | - | - | - |
| Pb | 0.0085 | - | AD: 0.005378 CH: 0.018824 | - | 0.2500 |
| Cr | 0.500 | - | - | - | - |
| Fe | - | - | - | 0.0728 | 0.0257 |
| Cu | - | - | - | 0.6250 | 0.1250 |
| Zn | - | - | - | 0.0233 | 0.0133 |
| Peach | | | | | |
| Cd | 0.380 | - | - | - | - |
| Pb | 0.0085 | - | AD: 0.053780 CH: 0.188253 | - | 2.500 |
| Cr | 0.500 | - | - | - | - |
| Fe | - | - | - | 0.1157 | 0.0443 |
| Cu | - | - | - | 0.9000 | 0.1250 |
| Zn | - | - | - | 0.0300 | 0.0233 |
| Guava | | | | | |
| Cd | 0.380 | AD: 3.97E-05 CH: 1.39E-05 | - | 0.0330 | - |
| Pb | 0.0085 | AD: 0.001237 CH: 0.004329 | - | 0.0575 | - |
| Cr | 0.500 | - | - | - | - |
| Fe | - | - | - | 0.1724 | 0.0640 |
| Cu | - | - | - | 2.7530 | 0.3350 |
| Zn | - | - | - | 0.0500 | 0.0197 |
| Apple | | | | | |
| Cd | 0.380 | - | - | - | - |
| Pb | 0.0085 | AD: 0.007529 CH: 0.026353 | - | 0.3500 | - |
| Cr | 0.500 | AD: 1.83E-06 CH: 6.4E-06 | - | 0.0067 | - |
| Fe | - | - | - | 0.1876 | 0.0320 |
| Cu | - | - | - | 1.6750 | 0.3125 |
| Zn | - | - | - | 0.0483 | 0.0203 |
| Mango | | | | | |
| Cd | 0.3800 | - | - | - | - |
| Pb | 0.0085 | - | - | - | - |
| Cr | 0.500 | - | AD: 1.83E-07 CH: 6.4E-07 | - | 0.00067 |
| Fe | - | - | - | 0.2107 | 0.0661 |
| Cu | - | - | - | 2.3870 | 0.2620 |
| Zn | - | - | - | 0.0551 | 0.0229 |

*AD, Adult; CH, Children.

IV. Conclusion

The aforementioned findings revealed that among the analyzed heavy metals, concentrations of iron and copper in juice samples of all five fruit species, besides lead in citrus and peach nectar, were above the maximum permissible limits set by USEPA and WHO. Heavy metal concentrations varied among the fruit samples; they were the highest in juice samples, particularly of guava and apple, whereas nectar samples were the lowest. By calculating the estimated daily intake (EDI) levels and the target hazard quotients (THQ), we concluded that consumption of the fruits studied posed very little potential health risk, where THQ values were less than 1.0. For the potential carcinogenic health risk (CR), our data revealed that values for Pb of guava and apple juice, were more than the acceptable or tolerable risk of 1×10^{-6} , revealing that there was a potential carcinogenic risk when both adults and children were exposed to Pb. Moreover, the results of no-carcinogenic health risk (RR) showed high value of RR, which was more than 1.0, for Cu of juice samples from guava, apple and mango. This suggest that consuming such samples of fruit juices may present a high risk to the health, therefore should be highly stressed. By comparing the health risk to adults and children of analyzed heavy metals, it was obvious that children were more susceptible to the potential health risk than adults. thus, more attention should be given to children to avoid the harmful effects of pollutants. Regular monitoring of heavy metals in fruit juices and nectars supplied commercially to the market is necessary to reduce the potential health risk of the people.

References

- [1]. D'Mello JPF. Food Safety: Contaminants and Toxins. CABI Publishing, Wallingford, Oxon, UK, Cambridge, MA. 2003.
- [2]. Kowalska G, Pankiewicz U, Kowalski R, Mazurek A. Determination of the content of selected trace elements in Polish commercial fruit juices and health risk assessment. De Gruyter Public License. Open Chemistry. 2020;18: 443–452. <https://doi.org/10.1515/chem-2020-0043>
- [3]. Fathabad AE, Shariatifar N, Moazzen M, Nazmara S, Fakhri Y, Alimohammadi M, et al. Determination of heavy metal content of processed fruit products from Tehran's market using ICP-OES: a risk assessment study. Food Chem Toxicol. 2018;115:436–46. <https://doi.org/10.1016/j.fct.2018.03.044>
- [4]. Neves MF, Trombin VG, Lopes FF, Kalaki R, Milan P. The orange juice business. A Brazilian Perspective. In: Neves MV, Trombin VG, Lopes F, Kalaki R, Milan P. (Eds.), Definition of juice, nectar and still drink. Wageningen Academic Publishers, Wageningen. 2011;pp:117-118. doi.org/10.3920/978-90-8686-739-4_30
- [5]. Otitolaju AA. Today's Apple: Perspective of an Environmental Toxicologist. 12th Inaugural Lecture, University of Lagos Press. 2016;pp.1–77. <https://doi.org/10.12895/jaeid.20171.560>.
- [6]. Kooner R, Mahajan BV, Dhillon WS. Heavy metal contamination in vegetables, fruits, soil, and water—a Critical Review, Int. J. Agric. Environ. Biotechnol. 2014;(7):603–612, <https://doi.org/10.5958/2230-732X.2014.01365.5>.
- [7]. Jarup L, Hazards of heavy metals contamination, Br. Med. Bull. 2003;(68):67–182, <https://doi.org/10.1093/bmb/ldg032>.
- [8]. Dehelean A, Magdas DA. Analysis of mineral and heavy metal content of some commercial fruit juices by inductively coupled plasma mass spectrometry. Sci. World J. 2013. <https://doi.org/10.1155/2013/215423>
- [9]. Savić SR, Petrović SM, Stamenković JJ, Petronijević ŽB. The presence of minerals in clear orange juices. Adv. Technol. 2015;(4):71–78. DOI: 10.5937/savteh1502071S
- [10]. Salma I, Sajib M, Motalab M, Mumtaz B, Jahan S, Hoque M, Saha B. Comparative evaluation of macro and micro-nutrient element and heavy metal contents of commercial fruit juices available in Bangladesh. Am. J. Food Nutr. 2015;(3):56–63. DOI: 10.12691/ajfn-3-2-4
- [11]. Taghizadeh S, Davarynejad G, Asili J, Nemati S, Rezaee R, Goumenou M, Tsatsakis A, Karimi G. Health risk assessment of heavy metals via dietary intake of five pistachio (*Pistacia vera* L.) cultivars collected from different geographical sites of Iran, Food Chem. Toxicol. 2017;(107):99–107. <https://doi.org/10.1016/j.fct>.
- [12]. Sathawara NG, Parikish DJ, Agrwal YK. Essentials heavy metals in environmental samples from western Indian. Bull. Environ. Cont. Toxicol. 2004;(73):756–761. DOI: 10.1007/s00128-004-0490-1
- [13]. IARC. Cadmium and cadmium compounds, Beryllium, Cadmium, Mercury, and Exposure in Glass Manufacturing Industry. IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, International Agency for Research on Cancer, Lyon. 1993;(58):119–237.
- [14]. Zaidi MI, Asrar A, Mansoor A, Farooqui MA. The heavy metal concentrations along roadsides trees of Quetta and its effects on public health, J. Appl. Sci. Faisalabad 2005;(5):708–711. doi.org/10.3923/jas.2005.708.
- [15]. Li Z, Jennings A. Worldwide regulations of standard values of pesticides for human health risk control: A review. Int. J. Environ. Res. Public Health. 2017;14:826. DOI: 10.3390/ijerph14070826
- [16]. Tawfic MT. Persistent organic pollutant in Egypt- An overview. Soil and Water Pollution Monitoring. 2006;(pp):3-38. DOI: 10.1007/978-1-4020-4728-2_2
- [17]. Hassan A, Abd El-Rahman T, Marzouk A. Estimation of Some Trace Metals in Commercial Fruit Juices in Egypt. International Journal of Food Science and Nutrition Engineering. 2014;(4):66-72. 10.5923/j.food.20140403.02
- [18]. Osman MA, EL Badry N, Shreif RM, Youssef M. Safety of commercial fruit juices available on the Egyptian markets with regards their content from determined by heavy metal and aflatoxins residues. Current Science International. 2014;(3):159-171. <https://www.researchgate.net/publication/264586085>
- [19]. A.O.A.C. Official methods of analysis. Pesticide and Industrial Chemical Residues, 16th ed. A.O.A.C. Int., Arlington, Virginia, USA. 1995.
- [20]. WHO. Guidelines for Drinking Water Quality, World Health Organization, 3rd ed. Incorporating, The First and Second Addenda. Volume 1, Recommendations, Geneva, Switzerland. 2008.
- [21]. USEPA. Supplementary Guidance for Conducting Health Risk Assessment of Chemical Mixtures. Agency. 2000.

- [22]. European Union (EU). (Official Journal of the European Union) Commission Regulation (EC) No 1881/2006 Setting Maximum levels for Certain Contaminants in Foodstuffs. 2006.
- [23]. USEPA. United state environmental protection agency. Quantitative Risk Assessment Calculations. 2015;7–9. <https://www.epa.gov/sites/production/files/2015-05/documents/13.pdf>.
- [24]. USEPA. Integrated risk information system of US environmental protection agency. 2012.
- [25]. Adebisi FM, Ore OT, Ogunjimi IO. Evaluation of human health risk assessment of potential toxic metals in commonly consumed crayfish (*Palaemon hastatus*) in Nigeria. *Heliyon*. 2020;(6),e03092. doi.org/10.1016/j.heliyon.2019.e03092
- [26]. Turner T, Burri BJ. Potential Nutritional Benefits of Current Citrus Consumption. *Agriculture*. 2013;(3);70-187. doi:10.3390/agriculture3010170
- [27]. U.S. Environmental Protection agency. Supplementary Guidance for Conducting Health Risk Assessment of Chemical Mixtures. 2000.
- [28]. USEPA. Risk Assessment Guidance for Superfund: Human Health Evaluation Manual [Part A]: Interim Final. U.S. Environmental Protection Agency, Washington, DC, USA. 1989. [EPA/540/1-89/002].
- [29]. Chien L, Hung T, Choang K, Yeh C, Meng P, Shieh M, Ha B. Daily intake of TBT, Cu, Zn, Cd and As for fishermen in Taiwan. *Science of The Total Environment*. 2002;285(1-3),177-85. [http://dx.doi.org/10.1016/S0048-9697\(01\)00916-0](http://dx.doi.org/10.1016/S0048-9697(01)00916-0)
- [30]. Yu Y, Wang X, Yang D, Lei B, Zhang X, Zhang X. Evaluation of human health risks posed by carcinogenic and non-carcinogenic multiple contaminants associated with consumption of fish from Taihu Lake, China. *Food Chem. Toxicol*. 2014;(69):86–93. DOI: 10.1016/j.fct.2014.04.001
- [31]. Faroon A, Wright S, Tucker P, Jenkins K, Ingerman L, Rudisill C. Toxicological Profile for Cadmium Agency for Toxic Substances and Disease Registry. ATSDR, Atlanta, GA. 2012.
- [32]. Abadin H, Ashizawa A, Stevens YW, Lladós F, Diamond G, Sage G, Citra M, Quinones A, Bosch SJ, Swarts SG. Toxicological Profile for Lead. Agency for Toxic Substances and Disease Registry (ATSDR), Atlanta, GA. 2019.
- [33]. Pearce JMS. Burton's line in lead poisoning. *European Neurology*. 2007;(57):118-119. DOI: 10.1159/000098100
- [34]. Mushak P. Gastro-intestinal absorption of lead in children and adults: overview of biological and biophysico-chemical aspects. *Chem. Speciat. Bioavailab*. 1991;(3):87–104. doi.org/10.1080/09542299.1991.11083160
- [35]. Wilbur S, Abadin H, Fay M, Yu D, Tencza B, Ingerman L, Klotzbach J, James S. Toxicological Profile for Chromium Agency for Toxic Substances and Disease Registry. ATSDR, Atlanta, GA. 2012.
- [36]. Allen LH. Iron supplements: scientific issues concerning efficacy and implementations for research and programs. *J. Nutr*. 2002;132 (4 Suppl). 813S–9S. DOI: 10.1093/jn/132.4.813S
- [37]. Hambidge KM, Cassey CE, Krebs NF. in: Mertz, W. (Ed.), Zinc in Trace Element in Human and Animal Nutrition. Academic Press, Orlando. 1987;(pp):1–138.
- [38]. Roney N, Smith CV, Williams M, Osier M, Paikoff SJ. Toxicological Profile for Zinc Agency for Toxic Substances and Disease Registry. Atlanta, GA. 2005.
- [39]. Anastácio M, dos Santos APM, Aschner M, Mateus L. Determination of trace metals in fruit juices in the Portuguese market. *Toxicology Reports*. 2018;(5):434-439. <https://doi.org/10.1016/j.toxrep.2018.03.010>
- [40]. Liang Y, Yi X, Dang Z, Wang Q, Luo H, Tang J. Heavy Metal Contamination and Health Risk Assessment in the Vicinity of a Tailing Pond in Guangdong, China. *Int. J. Environ. Res. Public Health*. 2017;(12):1557. doi: 10.3390/ijerph14121557.
- [41]. Yang G, Li Y, Wu L, Xie L, Wu J. Concentration and health risk of heavy metals in topsoil of paddy field of Chengdu Plain. *Environ. Chem*. 2014;(33):269-275.
- [42]. Che F, Yu J, Hu XN, Duan XL, Li Q, Lin HP. Preliminary health risk assessment of heavy metals in soil in Shen-fu irrigation area. *J. Agro-environ. Sci*. 2009;(28):439-1443.
- [43]. Duan B, Zhang W, Zheng H, Wu C, Zhang Q, Bu Y. Comparison of health risk assessment of heavy metals and As in sewage sludge from wastewater treatment plants (WWTPs) for adult and children in urban district of Taiyuan, China. *Int. J. Environ. Res. Public health*. 2017;(14):1194. DOI.10.3390/ijerph14101194
- [44]. Tvermoes BE, Banducci AM, Devlin KD, Kerger BD, Abramson MM, Bebenek IG, Monnot AD. Screening level health risk assessment of selected metals in apple juice sold in the United States, *Food Chem. Toxicol*. 2014;(71):42–50. DOI: 10.1016/j.fct.2014.05.015