

## Heavy Metal Levels in Some Popular Vegetables from Some Selected Markets in Saudi Arabia

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**H**EAVY metals accumulation in vegetables is considered a world problem that may affect food safety and human health. In the present study, fifteen vegetable species consisting of five leafy; Arugula (*Eruca sativa*), Cabbage (*Brassica oleracea*), Corchorus (*Corchorus capsularis*), Dill (*Anethum graveolens*) and Parsley (*Petroselinum sativum*); five fruits; Cucumber (*Cucumis sativus*), Eggplant (*Solanum melongena*), Green pepper (*Capsicum annum*), Tomato (*Solanum lycopersicum*) and Zucchini (*Cucurbita pepo*); in addition to five tubers; Garlic (*Allium sativum*), Onion (*Allium cepa*), Radish (*Raphanus sativus*), Potato (*Solanum tuberosum*) and Yam (*Ipomoea batatas*) were purchased from the local markets in Rafha Governorate at the Northern Border region in Kingdom of Saudi Arabia and tested for levels of cadmium (Cd), lead (Pb), arsenic (As), iron (Fe), copper (Cu) and zinc (Zn) metals using atomic absorption spectrometry. The results obtained showed that levels of Cd, Pb, As, Fe, Cu and Zn ranged from 0.13±0.073 to 1.63±0.96; 0.33±0.18 to 5.38±2.67; 0.020±0.001 to 0.052±0.033; 0.41±0.09 to 10.81±7.29; 0.49±0.13 to 4.37±0.89 and 2.05±0.80 to 94.20±73.28µg/g, respectively, for all vegetable samples. The highest average concentrations of Cd, Pb, As, Fe, Cu and Zn were recorded in leafy species as 0.46±0.19 to 1.63 ± 0.96; 0.00 to 5.38 ± 2.67; 0.00 to 0.049±0.031; 2.95±1.47 to 10.81±1.29; 1.42±0.73 to 4.37±0.89; 4.63±1.37 to 21.99±18.60µg/g, respectively. The highest mean levels 94.20±73.28 and 10.81±7.29µg/g were recorded for Zn and Fe in potato and corchorus, respectively; while the lowest mean levels 0.13±0.073 and 0.33±0.18µg/g were recorded for Cd and Pb in garlic and radish, respectively. The concentration of elements in these vegetables were within safety baseline levels for human consumption.

**Keywords:** Heavy metals, Vegetables, Rafha Governorate, Safe limit, Atomic absorption spectrometry.

### Introduction

Most of the food materials are mainly derived from plants (vegetables, cereals, grains, fruits, etc.) and animals (Izah et al., 2016). Consumption of vegetables on a regular basis is one of the possible health-improving practice thus, vegetables are considered essential component in the human diet. Recently, people all over the world intake the fresh vegetables instead of red meat to reduce the incidence of chronic diseases such as diabetics, cancer, cardiovascular diseases and other age-related diseases (Prakash et al., 2012).

Vegetables act as essential diet because their contributing of vitamins, proteins, carbohydrates, calcium, iron and other short supply nutrients (Amin et al., 2013; Siegel et al., 2014 and Latif et al., 2018). Consumption of vegetables provide the body of the living organisms with trace elements and heavy metals which are essential for the good health (Mohamed et al., 2003).

Vegetables provide the well-known trace elements and heavy metals. Minor or trace elements are essential for good health if they come from an organic or plant source. In contrast, if they come from an inorganic or metallic source,

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they become toxic. The processes of plant growth depend on the cycle of nutrients including trace elements, from soil to plant (Mohamed et al., 2003).

Contamination of food with heavy metals may be considered the most serious problem to our world (Zaidi et al., 2005). This problem can endanger the safety of plant products as feeds and foods in addition to its reduction of the plant productivity (Zheljazkov et al., 2006). Heavy metals are not biodegradable components with long biological half-lives and potential undesirable side effects when accumulated in different body organs (Sathawara et al., 2004 and Radwan & Salama, 2006).

There are several factors for contamination with the heavy metal which including addition of metal-based pesticides and fertilizers, irrigation with contaminated water, industrial emissions and harvesting process (Duran et al., 2007 and Tuzen & Soylak, 2007). Metals of different sources can pollute the soil then mix with soil solution after that they enter into the body of plants and accumulate in the edible parts either in high or in low concentrations (Zhou et al., 2016).

The levels of heavy metals in plants may depend on some factors such as cultivar, plant species, organ of the plant, growth stage, bioavailability of heavy metals in the soil and environmental conditions (Intawongse & Dean, 2006 and Naser et al., 2011). Although vegetables are considered one of the most important dietary source of essential nutrients, it may contain high concentrations of heavy metals (Wiesler, 2012).

Heavy metals may accumulate in plant roots such as Pb, Cr and As or translocate to the shoot system such as Zn, Cd and Fe. Their concentrations are higher in roots, stems, followed by leaves and fruits (Vamerali et al., 2010). For utilization by living organisms, heavy metals can be utilized in small quantities and known as essential metals such as Cr, Cu, Mn, Ni, Zn or toxic and known as non-essential such as As, Pb, Cd, Hg (Vasić-Rački et al., 2010).

The essential micronutrients in higher plants, include B, Cl, Cu, Fe, Mo, Mn, Ni and Zn and non-essential such as Hg, Pb, Cd and As (Kirkby, 2012). The essential heavy metals are involved in many important processes in plant metabolism

that include respiration, photosynthesis, synthesis of proteins, metalloproteins, functions of metalloenzymes, redox processes, plant defense mechanisms and regulation of genes (Broadley et al., 2012).

Nowadays, there is a great demand for food safety hence, the attention of many researchers to estimate the risks associated with consumption of many contaminated foodstuffs; heavy metals, pesticides and/or toxins in vegetables has increased (D'Mello, 2003; Latif et al., 2009 and Ismail & Rashid, 2017). Globally, many people consume street foods every day as a part of their diet. Street food is recognized to pose significant public health risks. Because of persistent of heavy metals in nature, their accumulation in foods especially in vegetables and their potential toxicity, there is a great need to analyze and investigate their levels in the most common foodstuffs. This study therefore, focus on estimation of levels of six metals; cadmium (Cd), lead (Pb), arsenic (As), iron (Fe), copper (Cu) and zinc (Zn) in fifteen vegetable species (five leafy; five fruits and five tubers) which were purchased from some local markets in Rafha Governorate at the Northern Border region in Kingdom of Saudi Arabia.

## **Materials and Methods**

### *Collection of samples*

In the present study, fifteen vegetable species consisting of five leafy (arugula, cabbage, corchorus, dill and parsley); five fruits (cucumber, eggplant, green pepper, tomato and zucchini) and five tubers (garlic, onion, radish, potato and yam) vegetable samples were purchased (3 samples of each species) from local retailers in Rafha Governorate at Northern Border region in Kingdom of Saudi Arabia (during the period of December-2017 to February-2018). Local and botanical names of these vegetable samples are given in Table 1. Parts of the samples that were obviously dust contaminated, damaged by insects or mechanically injured were removed. Then, the selected samples were washed in distilled water. Only edible parts of the samples were blended using a kitchen blender (Clikon) and stored in polyethylene bags at -18°C until analysis.

### *Reagents*

All reagents used were of analytical grade. All the plastic and glassware were cleaned by soaking them in a 10% nitric acid solution and rinsing

them with distilled water prior to use. The element stock solutions (1,000mg/L) and HNO<sub>3</sub> used for digestion were provided by Merck (Darmstadt, Germany).

#### Sample preparation

Wet digestion method was used for sample preparation (Hseu, 2004). Ten grams of samples were weighed into digestion tubes and 10ml of concentrated HNO<sub>3</sub> were added. The samples were digested for 45min at 90°C and for 3h at 130°C, respectively. After digestion, the residue was filtered through blue band filter paper, then, the samples were diluted to 50ml with distilled water. Duplicated analysis was performed on the samples. Blank digestion was also carried out.

#### Standard solutions

The standard solutions were prepared by diluting the stock solutions of 1,000mg/L of cadmium, lead, arsenic, iron, copper and zinc.

#### Heavy metal analysis

An atomic absorption spectrophotometer (AAS, model 220) was used to determine levels

of Cd, Pb, As, Fe, Cu and Zn in the digested solutions. Working standard solutions of each element were prepared from stock standard solutions, this was done to ensure accuracy and that the analytical values were within the range of certified value. The sample preparation and heavy metals analyses were carried out at the Central Unit for Analysis and Scientific Services, National Research Center (NRC), El Bohouth St., Dokki, Giza, Egypt.

#### Daily intake and potential risk assessment

The estimated daily intake (EDI) of heavy metals depends on the corresponding concentrations in food and the daily consumption of food. The highest concentrations of the studied heavy metals were used for the calculation of EDI, assuming that human would expose to those highest levels. The EDI was calculated as follows:

$$EDI = C \times W$$

where: C= Concentration of certain heavy metal in contaminated vegetable and W= Average of the daily consumption of the studied vegetable.

TABLE 1. Vegetable samples, with their local and botanical names.

| Vegetable type   | No. | Local name   | Botanical name                             |
|------------------|-----|--------------|--|
| Leafy vegetables | (1) | Arugula      | <i>Eruca sativa</i>                        |
|                  | (2) | Cabbage      | <i>Brassica oleracea var. capitata</i>     |
|                  | (3) | Corchorus    | <i>Corchorus capsularis</i>                |
|                  | (4) | Dill         | <i>Anethum graveolens</i>                  |
|                  | (5) | Parsley      | <i>Petroselinum sativum</i>                |
| Fruit vegetables | (1) | Cucumbers    | <i>Cucumis sativus</i>                     |
|                  | (2) | Eggplant     | <i>Solanum melongena</i>                   |
|                  | (3) | Green pepper | <i>Capsicum annuum</i>                     |
|                  | (4) | Tomatoes     | <i>Solanum lycopersicum</i>                |
|                  | (5) | Zucchini     | <i>Cucurbita pepo</i>                      |
| Tuber vegetables | (1) | Garlic       | <i>Allium sativum</i>                      |
|                  | (2) | Onion        | <i>Allium cepa</i>                         |
|                  | (3) | Radish       | <i>Raphanus sativus var. Longipinnatus</i> |
|                  | (4) | Potatoes     | <i>Solanum tuberosum</i>                   |
|                  | (5) | Yams         | <i>Ipomoea batatas</i>                     |

Also, the tolerable daily intake (TDI) of the heavy metals for an adult person of 70kg (assumed as an ideal body weight of an Saudi adult person) was calculated by multiply the value of TDI ( $\mu\text{g kg}^{-1}$  b.wt./day) (FAO/WHO, 2001 and Baars et al., 2001).

#### Statistical analysis

Three samples of each vegetable were assayed and analyzed individually. Data was reported as mean  $\pm$  SD. Two-Way ANOVA with replication was used to determine significant difference. Mean levels of metals were correlated with each other for the different vegetables (Neter & Whitmore, 1993).

### Results and Discussion

Presence of heavy metals in vegetables may be coming from deposits of the environmental pollution or from contaminated soils, the metals are then pass through the roots to be deposit on the plant surface or accumulate in their edible parts (Haiyan & Stuanes,

2003 and Nwajei, 2009). Vegetables are known as a good absorber of metals where it contains a wide range of concentrations of both essential and toxic metals (Lokeshwari & Chandrappa, 2006 and Eslami et al., 2007).

Although a hug number of plants are investigated in the phytoremediation researches (Padmavathamma & Li, 2007 and Sarma, 2011), investigation of heavy metal levels in edible plants doesn't have attention with the degree that prevent reaching of the elevated concentrations of heavy metals to the consumer (Steenkamp et al., 2000 and Sharma et al., 2009).

#### Maximum levels for heavy metals in vegetables

Levels of six heavy metals in commonly consumed leafy, fruit and tuber vegetables for Rafha inhabitants are determined based on plants dry weight and recorded as mean $\pm$ SD of three replicates (Tables 2 and 3).

**TABLE 2. Mean concentration, standard deviation and range of Cd, Pb and As metals in different vegetable samples ( $\mu\text{g/g}$  dry wt) (n= 3).**

| Vegetable type   | No. | Name               | Cadmium (Cd) |                  | Lead (Pb) |                 | Arsenic (As) |                   |
|------------------|-----|--------------------|--------------|------------------|-----------|-----------------|--------------|-------------------|
|                  |     |                    | Ranges       | Mean $\pm$ SD    | Ranges    | Mean $\pm$ SD   | Ranges       | Mean $\pm$ SD     |
| Leafy vegetables | (1) | Arugula            | nd           | nd               | nd        | nd              | nd           | nd                |
|                  | (2) | Cabbage            | 0.40-0.77    | 0.46 $\pm$ 0.19  | 2.30-3.65 | 2.34 $\pm$ 0.90 | 0.003-0.090  | 0.031 $\pm$ 0.035 |
|                  | (3) | Corchorus          | 1.10-2.60    | 1.41 $\pm$ 0.75  | 4.80-7.28 | 4.88 $\pm$ 1.66 | 0.001-0.089  | 0.049 $\pm$ 0.031 |
|                  | (4) | Dill               | 1.00-3.20    | 1.63 $\pm$ 0.96  | 4.46-9.54 | 5.38 $\pm$ 2.67 | nd           | nd                |
|                  | (5) | Parsley            | 1.00-2.03    | 1.14 $\pm$ 0.58  | nd        | nd              | nd           | nd                |
|                  |     | <b>Mean values</b> |              | <b>1.16</b>      |           | <b>4.2</b>      |              | <b>0.04</b>       |
| Fruit vegetables | (1) | Cucumber           | nd           | nd               | nd        | nd              | nd-0.021     | 0.020 $\pm$ 0.001 |
|                  | (2) | Eggplant           | 0.30-0.59    | 0.35 $\pm$ 0.14  | 4.10-7.10 | 5.10 $\pm$ 1.54 | nd           | nd                |
|                  | (3) | Green pepper       | 0.25-0.43    | 0.27 $\pm$ 0.10  | 4.30-8.08 | 3.51 $\pm$ 0.83 | nd           | nd                |
|                  | (4) | Tomato             | 0.20-0.44    | 0.25 $\pm$ 0.11  | 2.15-3.93 | 2.40 $\pm$ 0.99 | nd           | nd                |
|                  | (5) | Zucchini           | nd           | nd               | nd        | nd              | nd           | nd                |
|                  |     | <b>Mean values</b> |              | <b>0.29</b>      |           | <b>3.67</b>     |              | <b>0.020</b>      |
| Tuber vegetables | (1) | Garlic             | 0.10-0.20    | 0.13 $\pm$ 0.073 | 3.70-6.15 | 3.93 $\pm$ 1.49 | nd           | nd                |
|                  | (2) | Onion              | nd           | nd               | nd        | nd              | 0.042-0.105  | 0.052 $\pm$ 0.033 |
|                  | (3) | Radish             | 0.10-0.25    | 0.14 $\pm$ 0.063 | 0.30-0.58 | 0.34 $\pm$ 0.15 | nd           | nd                |
|                  | (4) | Potato             | 0.15-0.37    | 0.21 $\pm$ 0.096 | 0.28-0.62 | 0.38 $\pm$ 0.18 | nd           | nd                |
|                  | (5) | Yams               | 0.10-0.29    | 0.19 $\pm$ 0.067 | nd        | nd              | 0.086-0.72   | 0.30 $\pm$ 0.252  |
|                  |     | <b>Mean values</b> |              | <b>0.16</b>      |           | <b>1.55</b>     |              | <b>0.17</b>       |

-nd: Not detected

**TABLE 3. Mean concentration, standard deviation and range of Fe, Cu and Zn metals in different vegetable samples ( $\mu\text{g/g}$  dry wt) (n = 3).**

| Vegetable type   | No. | Name               | Iron (Fe)  |                  | Copper (Cu) |                 | Zinc (Zn)   |                   |
|------------------|-----|--------------------|------------|------------------|-------------|-----------------|-------------|-------------------|
|                  |     |                    | Ranges     | Mean $\pm$ SD    | Ranges      | Mean $\pm$ SD   | Ranges      | Mean $\pm$ SD     |
| Leafy vegetables | (1) | Arugula            | 1.71-6.52  | 2.95 $\pm$ 1.47  | 4.84-5.35   | 4.37 $\pm$ 0.89 | 8.04-12.20  | 9.91 $\pm$ 1.49   |
|                  | (2) | Cabbage            | 3.28-6.45  | 4.31 $\pm$ 1.00  | 3.57-4.67   | 3.47 $\pm$ 0.88 | 2.91-6.74   | 4.63 $\pm$ 1.37   |
|                  | (3) | Corchorus          | 6.21-30.68 | 10.81 $\pm$ 1.29 | 1.00-3.56   | 2.1 $\pm$ 0.90  | 24.75-17.16 | 21.13 $\pm$ 2.69  |
|                  | (4) | Dill               | 4.83-8.40  | 6.53 $\pm$ 1.32  | 1.43-2.69   | 1.65 $\pm$ 0.67 | 2.32-7.86   | 5.09 $\pm$ 2.57   |
|                  | (5) | Parsley            | 3.13-9.14  | 5.40 $\pm$ 1.64  | 1.12-2.56   | 1.42 $\pm$ 0.73 | 2.43-41.55  | 21.99 $\pm$ 18.60 |
|                  |     | <b>Mean values</b> |            | <b>6.00</b>      |             | <b>2.60</b>     |             | <b>12.58</b>      |
| Fruit vegetables | (1) | Cucumbers          | 0.26-0.58  | 0.41 $\pm$ 0.09  | 0.91-0.79   | 0.85 $\pm$ 0.03 | 0.65-3.46   | 2.05 $\pm$ 0.80   |
|                  | (2) | Eggplant           | 1.86-3.63  | 2.41 $\pm$ 0.54  | 0.54-0.53   | 0.49 $\pm$ 0.13 | 2.33-5.48   | 3.90 $\pm$ 0.78   |
|                  | (3) | Green pepper       | 0.81-1.30  | 1.09 $\pm$ 0.17  | 0.92-1.93   | 1.42 $\pm$ 0.20 | 2.52-9.63   | 6.07 $\pm$ 1.89   |
|                  | (4) | Tomatoes           | 1.77-5.70  | 3.65 $\pm$ 1.38  | 0.73-0.59   | 0.66 $\pm$ 0.16 | 14.20-174.2 | 50.48 $\pm$ 19.00 |
|                  | (5) | Zucchini           | 0.97-1.66  | 1.35 $\pm$ 0.27  | 0.75-0.67   | 0.71 $\pm$ 0.07 | 11.40-13.62 | 12.51 $\pm$ 1.46  |
|                  |     | <b>Mean values</b> |            | <b>1.78</b>      |             | <b>0.77</b>     |             | <b>15.0</b>       |
| Tuber vegetables | (1) | Garlic             | 0.80-1.67  | 0.97 $\pm$ 0.44  | 0.73-1.94   | 1.33 $\pm$ 0.49 | 32.47-294.3 | 81.69 $\pm$ 73.68 |
|                  | (2) | Onion              | 1.11-3.78  | 1.78 $\pm$ 1.24  | 0.68-1.75   | 1.21 $\pm$ 0.15 | 6.40-55.34  | 61.74 $\pm$ 29.54 |
|                  | (3) | Radish             | 3.37-5.12  | 3.71 $\pm$ 0.89  | 0.37-1.47   | 0.92 $\pm$ 0.22 | 27.96-131.4 | 79.68 $\pm$ 38.65 |
|                  | (4) | Potatoes           | 4.11-6.56  | 4.30 $\pm$ 1.52  | 0.75-0.93   | 0.51 $\pm$ 0.11 | 21.84-79.13 | 94.20 $\pm$ 73.28 |
|                  | (5) | Yams               | 2.12-8.82  | 5.70 $\pm$ 2.38  | 1.75-5.93   | 2.74 $\pm$ 1.27 | 33.88-131.4 | 82.64 $\pm$ 37.4  |
|                  |     | <b>Mean values</b> |            | <b>3.25</b>      |             | <b>1.34</b>     |             | <b>79.9</b>       |

The results obtained in this study showed that the levels of Cd, Pb, As, Fe, Cu and Zn ranged from 0.13 $\pm$ 0.073 to 1.63 $\pm$ 0.96; 0.33 $\pm$ 0.18 to 5.38 $\pm$ 2.67; 0.020 $\pm$ 0.001 to 0.052 $\pm$ 0.033; 0.41 $\pm$ 0.09 to 10.81 $\pm$ 7.29; 0.49 $\pm$ 0.13 to 4.37 $\pm$ 0.89 and 2.05 $\pm$ 0.80 to 94.20 $\pm$ 73.28 $\mu\text{g/g}$ , respectively for all vegetable samples. These results were compared well with similar samples from other published works in different countries (Table 4).

It was noticed that, the highest average concentrations of Cd, Pb, As, Fe, Cu and Zn were recorded in leafy vegetables as 0.46 $\pm$ 0.19 to 1.63 $\pm$ 0.96; 0.0 to 5.38 $\pm$ 2.67; 0.0 to 0.049 $\pm$ 0.031; 2.95 $\pm$ 1.47 to 10.81 $\pm$ 1.29; 1.42 $\pm$ 0.73 to 4.37 $\pm$ 0.89; 4.63 $\pm$ 1.37 to 21.99 $\pm$ 18.60 $\mu\text{g/g}$ , respectively, compared with the lowest average concentration which detected in fruit vegetables as 0.0 to 0.35 $\pm$ 0.14; 0.0 to 5.10 $\pm$ 1.54; 0.0 to 0.020 $\pm$ 0.001; 0.41 $\pm$ 0.09 to 3.65 $\pm$ 1.38; 0.49 $\pm$ 0.13 to 1.42 $\pm$ 0.20; 2.05 $\pm$ 0.80 to 50.48 $\pm$ 19.00 $\mu\text{g/g}$ , respectively.

At the same time, the tuber vegetables were found to contain moderate average concentrations;

0.13 $\pm$ 0.073 to 0.21 $\pm$ 0.096; 0.0 to 3.93 $\pm$ 1.49; 0.0 to 0.052 $\pm$ 0.033; 0.97 $\pm$ 0.44 to 5.70 $\pm$ 2.38; 0.51 $\pm$ 0.11 to 2.74 $\pm$ 1.27; 61.74 $\pm$ 29.54 to 94.20 $\pm$ 73.28 $\mu\text{g/g}$  of Cd, Pb, As, Fe, Cu and Zn, respectively. Vegetables absorb heavy metals from the soil and polluted air and water and especially leafy vegetables accumulate higher amounts of heavy metals in their leaves (Demirezen & Aksoy, 2006; Sobukola et al., 2008 and Bagdatlioglu et al., 2010).

The results of the analysis showed that the levels of Cd in all samples were between 0.13 $\pm$ 0.073 $\mu\text{g/g}$  in garlic and 1.63 $\pm$ 0.96 $\mu\text{g/g}$  in dill plant with a range of 0.10-0.20 and 1.00-3.20, respectively. The highest levels of Cd in leafy vegetables were observed in cabbage, corchorus, dill and parsley. While the lowest levels of Cd in tuber vegetables were recorded in garlic, onion, radish, yam and potato in that order. Cadmium is a non-essential element in foods and natural waters and it accumulates in the human body, principally in the kidneys and liver. Various sources of environmental contamination have

been implicated for its presence in foods (Divrikli et al., 2006).

Concerning the detected levels of Pb, they were between  $0.34 \pm 0.15 \mu\text{g/g}$  in radish and  $5.38 \pm 2.67 \mu\text{g/g}$  in dill plant with a range of 0.30-0.58 and 4.46-9.54, respectively. The highest levels of Pb were observed in leafy vegetables in corchorus and dill followed by fruit vegetables in eggplant and green pepper then in tuber vegetables in garlic. Pb being a serious cumulative body poison enters into the body system through air, water and food and cannot be removed by washing fruits and vegetables (Divrikli et al., 2003). The high levels of Pb in some of these plants may probably be attributed to pollutants in irrigation water, farm soil or due to pollution from the highway's traffic (Qui et al., 2000). While the lowest levels of Pb in tuber vegetables were recorded in radish and potato, also it was observed that Pb was not detected in some of the studied vegetable species as arugula, parsley, cucumber, zucchini, onion and yam.

Foods considered an important part of the arsenic intake, but because of the low level of arsenic in waters, soils, air, organisms and the other sources of nature, there is a few reports focused on estimation of arsenic level in vegetables especially for those grown in non-polluted soils (Chakraborti et al., 2002; Vela et al., 2004 and Shir Khanloo et al., 2015). In the present study As was not detected in 9 of the tested vegetables, but it was recorded in 6 species at very low concentration ranged between  $0.020 \pm 0.001 \mu\text{g/g}$  in cucumber and  $0.052 \pm 0.033 \mu\text{g/g}$  in onion with a range of 0.0-0.021 and 0.042-0.105  $\mu\text{g/g}$ , respectively. To the best of our knowledge, in Saudi Arabia there are no studies concerned with estimation level of As in vegetables. Only one published work was focused on estimation of As level in some vegetable oils which ranged between 0.005-0.017 and 0.014-0.021  $\mu\text{g/g}$  (Ashraf, 2014).

The highest concentration of Fe was recorded in leafy vegetables in comparing with other vegetable types. Among leafy vegetables both corchorus and dill were the highest species  $10.81 \pm 7.29$  and  $6.53 \pm 1.32 \mu\text{g/g}$ , respectively. The high accumulation of Fe in leafy vegetables than other vegetable types is due to that leaves are considered food making factories in plants. Our results were much lower than recorded by other published works in different regions in Saudi

Arabia (Mohamed et al., 2003; Al Jassir et al., 2005; Ali & Al-Qahtani, 2012 and Ashraf, 2014).

In the present study, the concentration of Cu in all vegetable types ranged between  $0.49 \pm 0.13 \mu\text{g/g}$  in eggplant and  $4.37 \pm 0.89 \mu\text{g/g}$  in arugula with a range of 4.84-5.35 and 0.54-0.53, respectively. The highest levels of Cu were recorded in leafy vegetables; arugula, cabbage, corchorus, parsley and dill in that order, while the lowest levels of Cu were variable in both fruit and tuber vegetables. Because of Cu is necessary for the normal growth of plants, the vegetables contain amount of it in their tissues but the high level of Cu in vegetable may be due to using of copper-based fertilizers (FAO/WHO, 1988) in addition to leaves considered as entry points of heavy metals from the air (Demirezen & Aksoy, 2006). The present study revealed that leafy vegetables accumulated Cu lower than the permissible level;  $10.00 \text{ mg kg}^{-1}$ , (FAO/WHO, 1999), and lower than another study in Saudi Arabia which were 33.22, 24.38, 14.07, 10.0, 9.07 and  $6.76 \mu\text{g/g}$  (Mohamed et al., 2003 and Ali & Al-Qahtani, 2012).

Zinc is one of the most important metals for human growth and development (Divrikli et al., 2006). Its deficiency may be due to different reasons such as dietary inadequate intake or some zinc metabolic disorders as impaired absorption or excessive excretion or inherited defects in its metabolism (Colak et al., 2005 and Narin et al., 2005). In developing countries, the deficiency of Zn which attributed to consumption of plant foods having inhibitory components for Zn absorption is of growing interest (Divrikli et al., 2006). In this study, the recorded levels of Zn were varied between  $2.05 \pm 0.80 \mu\text{g/g}$  in cucumber and  $94.20 \pm 73.28 \mu\text{g/g}$  in potato with range of 0.65-3.46 and 21.84-79.13, respectively. The highest levels of Zn were recorded in tuber vegetables; potato, yam, garlic, radish and onion in that order at the same time the lowest levels were varied between leafy and fruit vegetables.

The maximum permissible levels of toxic metals in human foods are set by different national and international regulations; hence controlling the concentrations of heavy metals in foods is an increasingly important aspect of food quality (Radwan & Salama, 2006 and Sobukola et al., 2010). Comparison of the obtained results with results of similar vegetable species published in different studies are recorded in Table 4.

**TABLE 4. Levels of heavy metals in the selected vegetables from Rafha markets, compared with earlier published results from other studies of the world.**

| No.  | Compared studies          | Levels of heavy metals in leafy vegetables ( $\mu\text{g/g}$ ) |             |              |              |             |             |
|--|---------------------------|--|-------------|--------------|--------------|-------------|-------------|
|  |                           | Cadmium (Cd)   | Lead (Pb)   | Arsenic (As) | Iron (Fe)    | Copper (Cu) | Zinc (Zn)   |
| <b>Leafy vegetables</b>  |                           |  |             |              |              |             |             |
| (1)  | Present study             | 0.46–1.14  | nd–3.34     | 0–0.031      | 2.95–4.31    | 1.42–4.37   | 4.63–21.99  |
| (2)  | Saudi Arabia <sup>a</sup> | 0.94–4.02  | 2.19–5.31   | NS           | 119.5–495.9  | 4.24–11.38  | 21.54–35.54 |
| (3)  | Saudi Arabia <sup>b</sup> | 0–1.22   | 0–14.37     | NS           | 76.9–431     | 0.43–3.34   | 14.9–50.7   |
| (4)  | Saudi Arabia <sup>c</sup> | 0.046–0.384  | 0.059–0.101 | NS           | NS           | 8.63–20.18  | 19.24–41.93 |
| (5)  | Turkey <sup>d</sup>       | nd–0.024   | 0.044–0.249 | NS           | 12.13–149.56 | 0.70–6.60   | 2.41–10.81  |
| (6)  | Bangladesh <sup>e</sup>   | 0.002–0.35   | 1.41–1.68   | 0.37–0.48    | NS           | 4.15–6.28   | 36.40–48.17 |
| <b>Fruit vegetables</b>  |                           |  |             |              |              |             |             |
| (1)  | Present study             | nd–0.25  | nd–2.40     | nd–0.020     | 0.41–1.09    | 0.49–1.42   | 2.05–50.48  |
| (2)  | Saudi Arabia <sup>a</sup> | 1.13–1.67  | 3.32–3.67   | NS           | 112.8–196.1  | 3.21–5.80   | 22.44–29.78 |
| (3)  | Saudi Arabia <sup>b</sup> | 0.59–0.8   | 1.9–4.57    | NS           | 49–431       | 2.48–4.49   | 8.51–50.7   |
| (4)  | Saudi Arabia <sup>c</sup> | NS   | NS          | NS           | NS           | NS          | NS          |
| (5)  | Turkey <sup>d</sup>       | nd   | 0.001–0.005 | NS           | 1.76–5.55    | 0.51–0.85   | 0.40–1.35   |
| (6)  | Bangladesh <sup>e</sup>   | nd–0.47  | 0.090–1.025 | 0.15–0.54    | NS           | 5.78–26.18  | 12.42–56.78 |
| <b>Tuber vegetables</b>  |                           |  |             |              |              |             |             |
| (1)  | Present study             | nd–0.13  | nd–0.33     | nd–0.042     | 0.97–5.70    | 0.51–2.74   | 61.74–94.20 |
| (2)  | Saudi Arabia <sup>a</sup> | 0.93–0.99  | 3.52–6.19   | NS           | 50.5–69.2    | 3.29–6.08   | 14.97–20.08 |
| (3)  | Saudi Arabia <sup>b</sup> | 0.76–0.84  | 2.81–10.29  | NS           | 29.4–93.6    | 0.88–2.81   | 4.5–17.6    |
| (4)  | Saudi Arabia <sup>c</sup> | NS   | NS          | NS           | NS           | NS          | NS          |
| (5)  | Turkey <sup>d</sup>       | nd   | 0.003–0.009 | NS           | 4.99–24.21   | 0.30–3.15   | 2.57–2.95   |
| (6)  | Bangladesh <sup>e</sup>   | 0.001–0.156  | 0.22–0.350  | 0.090–0.058  | NS           | 3.49–13.68  | 21.15–32.19 |
| <b>Permissible limits of the metals (<math>\mu\text{g/g}</math>)</b> |                           |  |             |              |              |             |             |
| (7)  | FAO/WHO <sup>f</sup>      | 0.2  | 0.3         | 0.1          | 425          | 73          | 99          |

-nd: Not detected; ns: Not studied; <sup>a</sup>Ali & Al-Qahtani (2012), <sup>b</sup>Mohamed et al. (2003), <sup>c</sup>Al Jassir et al. (2005), <sup>d</sup>Bagdatlioglu et al. (2010), <sup>e</sup>Alam et al. (2003) and <sup>f</sup>FAO/WHO (1999).

Form the above mentioned results it was clear that, there is no relationship between the highest heavy metal levels and period of storage of the plant part in the soil. It was known that the fruit vegetables have the longest storage period in cultivation comparing with the leafy and tuber vegetables. The obtained results ensured the mean values of the tested metals; Cd, Pb, As, Fe, Cu and Zn were 1.16, 4.20, 0.04, 0.06, 2.60, 12.58 $\mu\text{g/g}$  for leafy species; 0.29, 3.67, 0.20, 1.78, 0.77, 15.0 $\mu\text{g/g}$  for fruits and 0.16, 1.55, 0.17, 3.25, 1.34, 79.9 $\mu\text{g/g}$  for tuber species, respectively. Thus, we can conclude that the longest storage period was arranged as fruits > tubers > leafy vegetables while the highest mean values of each metal in the tested vegetables were arranged as leafy > tubers > fruits vegetables. Assimilation of elements is carried out in the root

system of the plants thus, the direct contact between the soil and plant roots enable different metals to enter the tissues of the plant with nutrients and water (Smical et al., 2008). After the metals enter the plant body through the roots, they either still present and stored in the roots or transported to other plant parts. Thus, plants accumulate the metals in edible or nonedible parts with concentrations enough to cause risks to human and animals that consumes these metals-containing plants (Arora et al., 2008).

In the present study, the obtained heavy metal concentrations are compared with the permissible limits of some international regulations (FAO/WHO, 1999) and it was found that, the maximum levels, 0.042, 5.70, 4.37 & 94.2 $\mu\text{g/g}$  of As, Fe, Cu & Zn are within FAO/WHO safe limits; 0.1, 425, 73 &

99 $\mu\text{g/g}$  for the same metals, respectively. While the obtained maximum levels of both Cd and Pb 1.14 and 3.34 $\mu\text{g/g}$  are higher than the safe limit of FAO/WHO 0.2 & 0.3 $\mu\text{g/g}$ , respectively with about 5 and 10 folds, especially in leafy and fruit vegetable species, but it was lower in the tuber vegetables.

Generally, the levels of heavy metals although they were higher than the permissible levels, they are lower than previously published works especially those in Saudi Arabia County, and those given by various authorities (FAO/WHO, 1988; 1999). At Rafha Governorate, most of the street consumable vegetables are locally cultured in Rafha farmers, where lower industrial activities and lower traffic in this region which result in the lowering of the contamination level.

#### *Correlation coefficient of metals in the tested vegetables*

The correlation between metal concentrations in the edible vegetables and their types was studied. In leafy vegetables, it was found a good positive correlation between the concentration of Cd and Pb ( $r=0.719$ ), Cd and Fe ( $r=0.785$ ), Pb and Fe ( $r=0.725$ ), As and Fe ( $r=0.670$ ), Fe and Zn ( $r=0.521$ ). But there is no significant correlation between Cd and As ( $r=0.146$ ), Cd and Zn ( $r=0.305$ ), Pb and As ( $r=0.468$ ), As and Cu ( $r=0.019$ ), As and Zn ( $r=0.225$ ). While it was noticed a negative correlation between the concentration of Cd and Cu ( $r=-0.929$ ), Fe and Cu ( $r=-0.596$ ), Cu and Zn ( $r=-0.462$ ), Pb and Cu ( $r=-0.424$ ).

In fruit vegetables, it was found positive correlation between Cd and Pb concentration ( $r=0.974$ ), Cd and Fe ( $r=0.604$ ), Fe and Cu ( $r=0.506$ ), Fe and Zn ( $r=0.822$ ). But correlation is not significant between Cd and Cu ( $r=0.005$ ), Cd and Zn ( $r=0.194$ ), Pb and Fe ( $r=0.465$ ), Pb and Cu ( $r=-0.008$ ), Pb and Zn ( $r=-0.015$ ), As and Cu ( $r=0.037$ ), As and Zn ( $r=-0.358$ ), Cu and Zn ( $r=-0.259$ ). While there was a negative correlation between the concentration of Cd and As ( $r=-0.596$ ), Pb and As ( $r=-0.552$ ), As and Fe ( $r=-0.604$ ).

Regarding the tuber vegetables, a positive correlation between Cd and Fe concentration ( $r=0.677$ ), Cd and Zn ( $r=0.956$ ), Pb and Fe ( $r=0.655$ ), As and Fe ( $r=0.644$ ), As and Cu ( $r=0.938$ ). But, correlation was not significant between Cd and Pb ( $r=0.027$ ), Cd and As ( $r=0.229$ ), Cd and Cu ( $r=0.109$ ), Pb and As ( $r=-0.372$ ), Pb and Cu ( $r=0.089$ ), Pb and Zn ( $r=-0.152$ ), As and Zn ( $r=-$

0.025), Fe and Cu ( $r=-0.399$ ), Fe and Zn ( $r=0.494$ ), Cu and Zn ( $r=-0.142$ ). The previously mentioned correlations indicated that the variations in the heavy metals (pollutants) were due to the same factors as soil, water or air pollutions as indicated by their positive correlations. Also, the variations in trace and essential elements depend on their content in the soil and their utilization by the plant organs and/or their partition in the plant. For Cd, Pb, Cu and Zn levels, a significant difference ( $P<0.05$ ) at confidence level 95% was recorded between all vegetable samples. The solubility of heavy metals in the soil and their accumulation in the plants may be the cause for the detected differences in metals concentration for the tested vegetable species (Cordeiro et al., 2013).

#### *Assessment of potential risk (safe dose)*

In each survey study dealing with estimation of food contaminants the safety assessment is essential complementary section. To determine the estimated daily intake (EDI) for the studied 3 vegetables types, the obtained data of the levels of heavy metals were employed. We considered the daily consumption for the adult Saudi person (70kg weight) is 7.8, 80.5 and 61g of leafy, fruits and tuber vegetables, respectively. The data recorded in Table 5 showed that EDI of the metals under investigation were lower than those of tolerable daily intake (TDI) except in case of both Cd and Pb. The results of this study in comparison with TDI values indicate that intakes of metals through leafy vegetables from Saudi Arabia would not mean a health hazards for consumers, but gets significance in light of the reported daily intakes for Cd and Pb through rice in Saudi diet as 67.41 $\mu\text{g}$  Pb and 10.3 $\mu\text{g}$  Cd (Al-Saleh & Shinwari, 2001).

Considering the Saudi nutritional habitats, an adult person (70kg) consumes 7.8, 80.5 and 61g of leafy, fruits and tuber vegetables, respectively in a daily manner. In this regards, calculated data in Table 5 revealed that values of the estimated daily intake (EDI) of the studied heavy metals, for 70kg adult person, were markedly lower than those of tolerable daily intake (TDI) except in case of both Cd and Pb. The results of this study in comparison with TDI values indicate that intakes of metals through leafy vegetables from Saudi Arabia would not mean a health hazards for consumers, but gets significance in light of the reported daily intakes for Cd and Pb through rice in Saudi diet as 67.41 $\mu\text{g}$  Pb and 10.3 $\mu\text{g}$  Cd (Al-Saleh & Shinwari, 2001).

TABLE 5. Estimated daily intake of heavy metals compared to the tolerable daily intake.

| Heavy metals | TDI*<br>( $\mu\text{g kg}^{-1}\text{b.wt/day}$ ) | TDI**<br>(70 $\mu\text{g}$ adult person/day) | EDI*** ( $\mu\text{g}$ ) |                  |                  |
|--------------|--|--|--------------------------|------------------|------------------|
|              |  |  | Leafy vegetables         | Fruit vegetables | Tuber vegetables |
| Cadmium      | 0.2  | 14   | 8.89                     | 91.77            | 69.54            |
| Lead         | 0.3  | 21   | 26.05                    | 268.87           | 203.74           |
| Arsenic      | 0.1  | 7  | 0.327                    | 3.38             | 2.56             |
| Iron         | 425  | 29750  | 44.46                    | 458.8            | 347.7            |
| Copper       | 73   | 5110   | 34.08                    | 351.78           | 266.57           |
| Zinc         | 99   | 6930   | 734.76                   | 6580             | 5746             |

\*FAO/WHO, 1999. \*\*TDI: Tolerable daily intake for assumed ideal Saudi body weight (70kg), \*\*\*EDI: Estimated daily intake (calculated by multiply the highest detected value of certain metal with the daily consumed amount (g) of vegetable according to FAO/WHO (2003).

### Conclusion

In the current study, we can conclude that, the tested heavy metals of the different species of vegetables were arranged as  $\text{Zn} > \text{Fe} > \text{Cu} > \text{Pb} > \text{Cd} > \text{As}$ . The leafy vegetables were found to contain the highest values of most heavy metals but within permissible levels. The heavy metal levels recorded in this study for leafy, fruit and tuber vegetables collected from Rafha Governorate at the Northern Border region in Kingdom of Saudi Arabia are within the standard permissible levels thus these vegetable species are suitable in the food composition for Rafha and Northern Border region inhabitants.

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### مستويات المعادن الثقيلة في بعض أنواع الخضروات الشعبية والمختارة من بعض الأسواق في المملكة العربية السعودية

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تراكم المعادن الثقيلة في الخضروات يُعد مشكلة عالمية خطيرة على سلامة الغذاء وصحة الإنسان. في هذه الدراسة تم إختيار خمسة عشر نوعاً من الخضروات تكونت من خمسة أنواع ورقية هي الجرجير *Eruca sativa*، الملفوف *Brassica oleracea*، الملوخية *Corchorus capsularis*، الشبت *Anethum graveolens* والبقدونس *Petroselinum sativum*؛ خمس أنواع فواكه هي الخيار *Cucumis sativus*، الباذنجان *Solanum melongena*، الفلفل الأخضر *Capsicum annuum*، الطماطم *Solanum lycopersicum* والكوسة *Cucurbita pepo*؛ بالإضافة إلى خمسة أنواع درنية هي الثوم *Allium sativum*، البصل *Allium cepa*، الفجل *Raphanus sativus*، البطاطا *Solanum tuberosum* واليام *Ipomoea batatas* تم شراؤها من بعض الأسواق المحلية في محافظة رفحاء بمنطقة الحدود الشمالية في المملكة العربية السعودية. تم قياس مستويات عناصر الكاديوم والرصاص والزرنيخ والحديد والنحاس والزنك في كل أنواع الخضروات السالفة الذكر باستخدام الامتصاص الطيفي الذري، وأكدت النتائج أن مستويات هذه العناصر تراوحت بين 0.13±0.073 إلى 0.96±1.63 و 0.18±0.33 إلى 2.67±5.38 و 0.001±0.020 إلى 0.033±0.052 و 0.41±0.09 إلى 7.29±10.81 و 0.13±0.49 إلى 0.89±4.37 و 0.80±2.05 إلى 73.28±94.20 ميكروجرام/جرام من الكاديوم والرصاص والزرنيخ والحديد والزنك والنحاس على التوالي. تم تسجيل أعلى تركيزات من مستويات هذه العناصر في أنواع الخضروات الورقية حيث تراوحت النسب من 0.19±0.46 إلى 1.63±0.96 و 0.00 إلى 2.67±5.38 و 0.00 إلى 0.031±0.049 و 1.47±2.95 إلى 1.29±10.81 و 1.42±0.73 إلى 0.89±4.37 و 1.37±4.63 إلى 18.60±21.99 ميكروجرام/جرام على التوالي. تم التوصل من خلال هذه النتائج إلى أن أعلى مستويات العناصر كانت 73.28±94.20 و 7.29±10.81 ميكروجرام/جرام لكل من الزنك والحديد في البطاطا *Solanum tuberosum* والملوخية *Corchorus capsularis* على التوالي؛ بينما كانت أدنى المستويات 0.073±0.13 و 0.18±0.33 ميكروجرام/جرام للكاديوم والرصاص في الثوم *Allium sativum* والفجل *Raphanus sativus* على التوالي. تُظهر نتائج هذه الدراسة أن تركيزات العناصر الثقيلة التي تم قياسها في أنواع الخضروات محل الدراسة ضمن المستويات المسموح بها للاستهلاك البشري.