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FULL LENGTH ARTICLE

Assessment of some heavy metals in vegetables, cereals and fruits in Saudi Arabian markets

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Abstract The concentration of some heavy metals Fe, Mn, Cu, Zn, Pb, Cd and Hg in various vegetables (roots, stems, leafy, fruits, cereals and legumes) grown in four major industrial and urban cities (Tabouk, Riyadh, Damamm and Jazan) in Kingdom of Saudi Arabia was assessed using atomic absorption spectrophotometer. The obtained results declared that concentrations of major studied metals were exceeding than the recommended maximum acceptable levels proposed by the Joint FAO/WHO Expert Committee on Food Additives. Leafy vegetables were found to contain the highest metals values especially parsley (543.2 and 0.048 μg/g for Fe and Hg respectively), Jews mallow (94.12 and 33.22 μg/g for Mn and Zn respectively), spinach (4.13 μg/g for Cd). While peas in legumes group maintained the highest Zn content 71.77 μg/g and finally cucumber had the highest Pb content 6.98 μg/g on dry matter basis. High concentrations of heavy metals in different parts of the vegetables might be related to their concentration in the polluted air with industrial activities especially in middle and eastern districts. The study concludes that atmospheric depositions and marketing systems of vegetables play a significant role in elevating the levels of heavy metals in vegetables having potential health hazards to consumers of locally produced foodstuffs.

Introduction

Heavy metal contamination of vegetables cannot be underestimated as these foodstuffs are important components of human diet. Vegetables are rich sources of vitamins, minerals, and fibers, and also have beneficial antioxidative effects. However, intake of heavy metal-contaminated vegetables may pose a risk to the human health. Heavy metal contamination of the food items is one of the most important aspects of food quality assurance (Marshall, 2004; Radwan and Salama, 2006; Khan et al., 2008). Rapid and unorganized urban and industrial developments have contributed to the elevated levels of heavy

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metals in the urban environment of developing countries such as Egypt (Radwan and Salama, 2006), Iran (Maleki and Zarasvand, 2008), China (Wong et al., 2003) and India (Marshall, 2004; Sharma et al., 2008a,b).

Emissions of heavy metals from the industries and vehicles may be deposited on the vegetable surfaces during their production, transport and marketing. Al Jassir et al. (2005) have reported elevated levels of heavy metals in vegetables sold in the markets at Riyadh city in Saudi Arabia due to atmospheric deposition. Recently, Sharma et al. (2008a,b) have reported that atmospheric deposition can significantly elevate the levels of heavy metals contamination in vegetables commonly sold in the markets of Varanasi, India. The prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases (WHO, 1992; Jarup, 2003). Some heavy metals such as Cu, Zn, Mn, Co and Mo act as micronutrients for the growth of animals and human beings when present in trace quantities, whereas others such as Cd, As, and Cr act as carcinogens (Feig et al., 1994; Trichopoulos, 1997). The contamination of vegetables with heavy metals due to soil and atmospheric contamination poses a threat to its quality and safety. Dietary intake of heavy metals also poses risk to animals and human health. Heavy metals such as Cd and Pb have been shown to have carcinogenic effects (Trichopoulos, 1997). High concentrations of heavy metals (Cu, Cd and Pb) in fruits and vegetables were related to high prevalence of upper gastrointestinal cancer (Turkdogan et al., 2002).

Regulations have been set up in many countries and for different industrial set up to control the emission of heavy metals. The uptake of heavy metals in vegetables are influenced by some factors such as climate, atmospheric depositions, the concentrations of heavy metals in soil, the nature of soil on which the vegetables are grown and the degree of maturity of the plants at the time of harvest (Lake et al., 1984; Scott et al., 1996). Air pollution may pose a threat to post-harvest vegetables during transportation and marketing, causing elevated levels of heavy metals in vegetables (Agrawal, 2003). Al-Rehaili (2009) monitored air quality in Riyadh city. He discussed 10 air pollutants, together with relevant meteorological parameters, his obtained results revealed that most sites had on the average exceeded the recommended standards for SO_2 and NH_3.

The main objectives of the present work are to focus on biomonitoring contamination of heavy metals in different vegetables and to establish some recommendations on human diet foodstuff in order to assure a significant improvement in food safety.

Materials and methods

Study area and sampling locations

The present study was carried out during 2011 in the major four urban cities of Kingdom of Saudi Arabia as follows, capital Riyadh city represents the middle area, Tabouk city represents the northern area, Dammam city represents the eastern area and Jazan city represents the southern area.

Collection of samples

More than 240 samples of harvested fresh vegetables were collected during 2011 grown in different farms. three to five subsamples were collected from different farms for the same vegetable. These samples classified into legumes (haricot, kidney bean, peas, beans), cereals (rice, wheat, barley), leafy vegetables (Jews mallow, spinach, arugula, parsley, cabbage), stems (potatoes, onions), roots (sweet potatoes, carrot, turnips) and fruits (tomatoes, cucumber). All collected samples were stored in clean polythene bags according to their type and brought to the laboratory for analyses.

Preparation and treatment of samples

The collected samples were washed with distilled water to remove the dust particles. Then samples were cut to small pieces using clean knife. Different parts (roots, stems and leaves) of vegetables were dried in an oven at 100 °C. After drying the samples were grinded into a fine powder using a commercial blender and stored in polyethylene bags, until used for acid digestion.

Acid digestion and metals determination of samples

Tri-acid mixture (15 ml, 70% high purity HNO_3, 65% $HClO_4$ and 70% H_2SO_4; 5:1:1) was added to the beaker containing 1 g dry vegetable sample (Allen et al., 1986). The mixture was then digested at 80 °C till the transparent solution was achieved. After cooling, the digested samples were filtered using Whatman No. 42 filter paper and the filtrate was diluted to 50 ml with deionised water. Determination of the heavy metals such as Cu, Zn, Cd and Pb in the filtrate of vegetables and atmospheric deposits was achieved by atomic absorption spectrophotometer (Shimadzu Model 6800 with graphite furnace Model GFA 7000, Hydride unit was used for determination of mercury).

Statistical analysis

The recorded data were subjected to two-way analysis of variance (ANOVA) to assess the influence of different variables on the concentrations of heavy metals in the vegetables tested. ANOVA for each vegetable was performed separately using variables such as sites. All the statistical analyses were computed with STAT5 software version 8.

Results and discussion

Concentrations of Fe, Mn, Cu, Zn, Pb, Cd and Hg in different vegetables collected from main cities of Kingdom of Saudi Arabia are given in Tables 1–4. Concentrations in these samples are varied quietly such as Fe (31.96–543.2 μg/mg), Mn (4.16–94.16 μg/mg), Cu (2.06–33.22 μg/mg), Zn (8.27–71.77 μg/mg), Pb (0.54–6.98 μg/mg), Cd (0.92–4.13 μg/mg), and Hg (0.009–0.048 μg/mg).

The obtained results of the present study showed that the concentration of Fe in the leafy vegetables is much high than other vegetables, however, parsley and Jews mallow maintained highest iron concentration (543.2, 495.9 and 399.1 μg/g

Table 1 Heavy metals concentrations (µg/g dry weight) of studied common vegetables in eastern district during 2011.

	Fe	Mn	Cu	Zn	Pb	Cd	Hg
Roots							
Turnips	110.3 ± 3.63	8.19 ± 0.87	6.21 ± 0.12	15.52 ± 0.64	2.04 ± 0.01	1.34 ± 0.08	0.031 ± 0.001
Carrot	77.9 ± 3.22	7.11 ± 0.45	3.60 ± 0.22	10.28 ± 0.67	1.42 ± 0.06	1.20 ± 0.07	0.023 ± 0.002
Stems							
Onions	50.5 ± 2.83	7.84 ± 0.42	3.29 ± 0.12	20.80 ± 0.92	3.52 ± 0.00	0.93 ± 0.05	0.039 ± 0.001
Potatoes	69.2 ± 2.03	8.80 ± 0.32	6.08 ± 0.23	14.97 ± 0.54	6.19 ± 0.23	0.99 ± 0.08	0.011 ± 0.001
Leafy							
Parsley	495.9 ± 16.91	31.10 ± 1.51	7.82 ± 0.26	26.62 ± 0.87	2.31 ± 0.08	0.98 ± 0.03	0.016 ± 0.001
Jews mallow	204.6 ± 4.20	68.87 ± 2.83	9.07 ± 0.26	22.73 ± 0.71	2.94 ± 0.08	0.94 ± 0.04	0.020 ± 0.001
Spinach	156.4 ± 7.82	26.53 ± 0.95	11.38 ± 0.35	35.54 ± 1.09	2.88 ± 0.17	4.02 ± 0.16	0.015 ± 0.001
Arugula	119.5 ± 5.59	26.67 ± 1.15	6.03 ± 0.14	34.93 ± 1.31	2.19 ± 0.15	2.89 ± 0.26	0.012 ± 0.001
Cabbage	124.2 ± 3.04	21.08 ± 0.35	4.24 ± 0.16	21.54 ± 0.61	5.31 ± 0.07	0.97 ± 0.01	0.018 ± 0.001
Fruits							
Cucumber	112.8 ± 3.30	9.55 ± 0.52	3.21 ± 0.06	29.78 ± 1.28	3.67 ± 3.04	1.13 ± 0.06	0.092 ± 0.093
Tomato	196.1 ± 1.20	10.51 ± 0.52	5.80 ± 0.06	22.44 ± 0.88	3.32 ± 0.02	1.67 ± 0.09	0.025 ± 0.001
Cereals							
Wheat	128.8 ± 3.41	13.88 ± 0.59	4.88 ± 0.17	24.42 ± 0.97	2.81 ± 0.08	1.90 ± 0.11	0.018 ± 0.001
Rice	84.1 ± 2.15	9.76 ± 0.72	7.82 ± 0.26	27.81 ± 1.58	6.16 ± 0.17	0.92 ± 0.08	0.016 ± 0.002
Legume							
Beans	115.4 ± 2.85	15.75 ± 0.66	10.41 ± 0.39	32.62 ± 3.57	4.57 ± 0.12	1.04 ± 0.06	0.018 ± 0.001
Haricot	107.7 ± 2.62	20.30 ± 1.08	7.84 ± 0.12	25.93 ± 0.38	2.23 ± 0.01	1.01 ± 0.01	0.012 ± 0.000
Kidney bean	142.7 ± 2.90	23.07 ± 0.94	7.10 ± 0.13	34.75 ± 0.51	0.54 ± 0.02	0.95 ± 0.02	0.016 ± 0.001

Table 2 Heavy metals concentrations (µg/g dry weight) of studied common vegetables in middle district during 2011.

	Fe	Mn	Cu	Zn	Pb	Cd	Hg
Roots							
Turnips	131.3 ± 6.82	25.19 ± 1.47	14.27 ± 0.2	30.05 ± 1.30	1.27 ± 0.08	1.29 ± 0.07	0.021 ± 0.001
Carrot	85.4 ± 2.23	7.44 ± 0.23	4.49 ± 0.19	26.64 ± 0.98	1.13 ± 0.04	1.43 ± 0.03	0.029 ± 0.001
Sweet Potatoes	79.5 ± 0.72	8.82 ± 0.73	6.29 ± 0.22	23.33 ± 1.39	3.63 ± 0.04	1.22 ± 0.09	0.012 ± 0.001
Stems							
Onions	51.9 ± 1.19	19.05 ± 0.24	10.03 ± 0.31	21.83 ± 0.53	4.28 ± 0.12	0.98 ± 0.02	0.018 ± 0.001
Potatoes	70.2 ± 2.16	7.61 ± 0.57	6.41 ± 0.22	17.65 ± 1.10	1.51 ± 0.06	1.18 ± 0.21	0.009 ± 0.001
Leafy							
Parsley	543.5 ± 14.33	21.58 ± 0.49	7.07 ± 0.26	31.76 ± 0.98	2.71 ± 0.05	0.98 ± 0.02	0.048 ± 0.001
Jews mallow	126.6 ± 0.93	94.16 ± 1.48	33.22 ± 0.22	22.93 ± 0.35	3.51 ± 0.03	1.14 ± 0.01	0.018 ± 0.001
Spinach	94.7 ± 3.34	25.13 ± 0.57	14.07 ± 0.44	30.18 ± 0.78	1.26 ± 0.06	4.13 ± 0.14	0.009 ± 0.001
Arugula	92.6 ± 1.66	86.86 ± 1.05	9.07 ± 0.25	39.46 ± 1.16	2.14 ± 0.05	3.01 ± 0.08	0.011 ± 0.001
Cabbage	76.4 ± 16.88	30.21 ± 1.25	6.85 ± 0.14	24.45 ± 0.87	3.80 ± 0.62	1.34 ± 0.15	0.011 ± 0.001
Fruits							
Cucumber	141.4 ± 5.80	10.92 ± 0.29	7.18 ± 0.25	22.30 ± 0.64	6.98 ± 0.44	1.28 ± 0.08	0.035 ± 0.001
Tomato	364.6 ± 10.45	27.84 ± 0.33	7.46 ± 0.41	22.91 ± 1.02	2.78 ± 0.12	2.45 ± 0.09	0.018 ± 0.001
Cereals							
Wheat	49.4 ± 1.60	25.56 ± 1.09	8.34 ± 0.29	21.47 ± 0.90	1.77 ± 0.08	1.32 ± 0.06	0.010 ± 0.001
Barley	48.5 ± 0.88	19.75 ± 1.02	8.69 ± 0.33	17.65 ± 0.85	2.87 ± 0.05	1.53 ± 0.10	0.009 ± 0.001
Legume							
Beans	99.9 ± 1.92	8.81 ± 0.35	9.04 ± 0.39	29.83 ± 1.15	3.85 ± 0.09	2.31 ± 0.06	0.024 ± 0.001
Haricot	296.6 ± 3.47	34.45 ± 2.58	9.87 ± 0.01	30.41 ± 1.36	3.40 ± 0.05	1.13 ± 0.07	0.017 ± 0.000
Kidney bean	118.6 ± 2.41	52.90 ± 1.69	9.99 ± 0.19	22.71 ± 0.43	0.88 ± 0.01	3.24 ± 0.07	0.026 ± 0.001
Peas	99.3 ± 2.45	4.16 ± 0.05	12.64 ± 0.01	62.47 ± 0.15	3.64 ± 0.05	1.70 ± 0.02	0.015 ± 0.000

dry wet) in middle and northern district respectively (Tables 1–3). Also, tomatoes in middle district have a noticeable high value (364.6 µg/g) (Table 2). Fe contents in cereals record the minimum values amongst the different types of vegetables from different districts especially in wheat and barley (Tables 1–4).

Fe contents in the leafy vegetables of parsley, Jews mallow, spinach, arugula and cabbage were significantly ($p < 0.05$) higher as compared to those in the other vegetables, whereas, in the fruit such as tomatoes and cucumber were found to be significantly higher ($p < 0.05$) although they have lower Fe

Table 3 Heavy metals concentrations (μg/g dry weight) of studied common vegetables in northern district during 2011.

	Fe	Mn	Cu	Zn	Pb	Cd	Hg
Roots							
Turnips	152.9 ± 3.68	6.77 ± 0.31	11.80 ± 0.46	15.79 ± 0.56	4.37 ± 0.03	1.24 ± 0.03	0.022 ± 0.001
Carrot	256.5 ± 2.20	12.04 ± 0.31	4.44 ± 0.26	8.27 ± 0.42	1.64 ± 0.05	1.21 ± 0.05	0.018 ± 0.001
Sweet Potatoes	109.3 ± 2.65	13.58 ± 0.1	14.43 ± 0.59	28.56 ± 0.90	3.17 ± 0.27	1.27 ± 0.09	0.031 ± 0.002
Stems							
Onions	31.9 ± 0.68	8.68 ± 0.51	4.91 ± 0.22	11.36 ± 0.63	4.38 ± 0.02	0.94 ± 0.06	0.021 ± 0.001
Potatoes	49.7 ± 0.94	5.33 ± 0.35	2.06 ± 0.08	14.28 ± 0.82	3.01 ± 0.05	1.15 ± 0.20	0.014 ± 0.001
Leafy							
Parsley	315.0 ± 16.17	39.27 ± 1.84	6.28 ± 0.21	23.30 ± 0.69	1.79 ± 0.15	1.11 ± 0.06	0.030 ± 0.002
Jews mallow	399.1 ± 9.92	60.52 ± 2.32	15.3 ± 0.33	10.47 ± 0.31	3.62 ± 0.04	1.08 ± 0.04	0.020 ± 0.001
Spinach	149.3 ± 1.43	30.60 ± 1.8	7.82 ± 0.26	25.27 ± 1.08	4.14 ± 0.08	3.89 ± 0.17	0.011 ± 0.001
Arugula	516.3 ± 13.96	43.32 ± 2.28	6.81 ± 0.24	29.02 ± 1.41	3.62 ± 0.04	2.67 ± 0.15	0.018 ± 0.001
Cabbage	73.3 ± 1.66	18.66 ± 0.52	2.76 ± 0.02	17.78 ± 0.12	3.49 ± 0.06	1.10 ± 0.01	0.018 ± 0.001
Fruits							
Cucumber	117.1 ± 2.46	7.43 ± 0.26	3.95 ± 0.07	15.71 ± 0.37	4.14 ± 0.06	1.13 ± 0.03	0.014 ± 0.000
Tomato	151.7 ± 3.43	18.05 ± 0.27	3.57 ± 0.25	8.93 ± 0.53	3.49 ± 0.03	1.18 ± 0.02	0.022 ± 0.002
Cereals							
Wheat	41.1 ± 0.93	23.00 ± 1.44	5.15 ± 0.21	25.12 ± 1.40	3.52 ± 0.09	1.52 ± 0.12	0.012 ± 0.001
Barley	60.9 ± 1.48	12.35 ± 1.35	3.61 ± 0.15	16.52 ± 1.44	3.77 ± 0.11	1.05 ± 0.14	0.012 ± 0.001
Legume							
Beans	64.0 ± 2.85	7.40 ± 0.25	12.49 ± 0.54	19.46 ± 0.67	1.51 ± 0.06	1.65 ± 0.05	ND
Haricot	131.5 ± 7.50	23.46 ± 0.7	6.47 ± 0.23	14.71 ± 0.41	2.90 ± 0.09	1.03 ± 0.03	ND
Kidney bean	162.8 ± 5.03	19.64 ± 1.39	5.51 ± 0.18	15.10 ± 0.84	0.88 ± 0.01	1.90 ± 0.14	ND
Peas	51.4 ± 3.20	7.49 ± 0.22	16.58 ± 0.79	71.77 ± 2.79	2.82 ± 0.10	2.12 ± 0.09	0.026 ± 0.002

ND: not detected.

Table 4 Heavy metals concentrations (μg/g dry weight) of studied common vegetables in southern district during 2011.

	Fe	Mn	Cu	Zn	Pb	Cd	Hg
Roots							
Turnips	141.9 ± 2.65	28.74 ± 0.93	7.45 ± 0.27	28.42 ± 0.89	2.94 ± 0.19	1.29 ± 0.08	0.036 ± 0.002
Carrot	168.7 ± 6.86	30.33 ± 0.88	7.82 ± 0.43	12.97 ± 0.55	1.52 ± 0.05	1.25 ± 0.04	0.021 ± 0.002
Sweet Potatoes	62.2 ± 3.30	45.04 ± 1.12	12.99 ± 0.16	21.50 ± 0.41	3.44 ± 0.12	1.54 ± 0.07	0.022 ± 0.001
Stems							
Onions	66.9 ± 1.69	9.77 ± 0.81	7.26 ± 0.01	23.15 ± 0.66	3.15 ± 0.22	1.13 ± 0.10	0.030 ± 0.001
Potatoes	60.0 ± 2.36	8.49 ± 0.31	2.30 ± 0.08	16.41 ± 0.61	5.32 ± 0.68	0.97 ± 0.25	0.017 ± 0.001
Leafy							
Parsley	126.2 ± 6.38	16.74 ± 0.36	9.10 ± 0.20	38.29 ± 0.68	2.20 ± 0.08	1.07 ± 0.03	0.046 ± 0.001
Jews mallow	287.3 ± 7.77	79.67 ± 1.73	24.38 ± 0.86	12.70 ± 0.39	3.49 ± 0.02	1.07 ± 0.02	0.027 ± 0.001
Spinach	145.8 ± 6.51	21.42 ± 1.22	10.00 ± 0.34	59.42 ± 2.32	2.85 ± 0.06	3.95 ± 0.21	0.022 ± 0.001
Arugula	180.9 ± 7.05	66.13 ± 2.42	6.76 ± 0.35	41.83 ± 1.98	4.67 ± 0.14	2.97 ± 0.11	0.019 ± 0.001
Cabbage	79.7 ± 6.52	14.80 ± 0.68	3.88 ± 0.18	22.44 ± 0.81	2.75 ± 0.12	1.09 ± 0.03	0.010 ± 0.001
Fruits							
Cucumber	59.6 ± 4.73	33.98 ± 1.51	5.65 ± 0.15	22.88 ± 0.57	6.70 ± 0.33	1.18 ± 0.04	0.030 ± 0.001
Tomato	164.9 ± 4.41	17.10 ± 1.10	7.30 ± 0.28	27.91 ± 1.52	2.62 ± 0.16	1.85 ± 0.11	0.023 ± 0.001
Cereals							
Wheat	65.3 ± 5.40	24.10 ± 0.5	4.38 ± 0.18	28.78 ± 0.92	3.45 ± 0.05	1.60 ± 0.02	0.022 ± 0.001
Barley	134.3 ± 5.76	22.43 ± 0.90	6.68 ± 0.25	26.05 ± 0.82	3.65 ± 0.14	1.02 ± 0.04	0.012 ± 0.001
Legume							
Beans	137.1 ± 7.50	12.81 ± 0.51	12.26 ± 0.33	42.11 ± 1.24	2.19 ± 0.13	1.47 ± 0.10	0.013 ± 0.001
Haricot	183.3 ± 4.25	43.83 ± 1.72	6.57 ± 0.25	35.67 ± 1.14	3.13 ± 0.03	1.09 ± 0.02	0.013 ± 0.001
Kidney bean	124.5 ± 0.83	26.98 ± 0.48	14.75 ± 0.34	22.95 ± 0.51	2.50 ± 0.07	2.20 ± 0.04	0.018 ± 0.002
Peas	110.5 ± 2.80	8.65 ± 0.61	12.04 ± 0.40	41.48 ± 1.81	3.96 ± 0.33	1.28 ± 0.13	0.019 ± 0.001

content. However, there was no significant variation ($p > 0.05$) in the Fe level in the stem vegetables (range: 31.96–66.9 and 49.7–70.2 µg/g) for onion and potatoes respectively (Tables 1–4). On the other hand, there were significant variation ($p < 0.05$) between Fe content in collected vegetables from middle district and other districts.

The amounts of Fe in the leafy vegetables were higher as compared to those in the other investigated vegetables especially in middle district. The reasonable explanation of this situation is that the Fe uptake can be promoted and accumulated in the leaves as a result of leaves are considered food making factories in plants. Our results were much higher than recorded by Zahir et al. (2009) who analyzed different samples of vegetables and reported a high concentration (7.9–24.8 µg/g) of Fe in Pakistan. In another study, Waheed et al. (2003) investigated that concentration of Fe (17.0–35.60 µg/g) in some raw foodstuffs grown in wastewater industrial areas.

Manganese contents showed a higher value in leafy vegetables as compared to those in the other investigated vegetables. Jews mallow and arugula has the highest Mn concentrations (94.61 and 86.86 µg/g) in middle district followed by Jews mallow in southern district (79.67 µg/g) (Tables 2 and 3). Also, legumes in middle and southern districts have a noticeable high values (52.9 and 43.83 µg/g) for kidney beans and haricot respectively (Tables 2 and 4). Mn contents in peas record the minimum values amongst the different types of vegetables (Tables 2–4).

The analyses of variance for Mn contents showed similar high significance ($p < 0.05$) in leafy vegetables as in iron, whereas in the legumes, especially kidney beans and haricot, there were noticeable significance ($p < 0.05$) although they have lower Mn contents than leafy vegetables. However, there was no significant variation ($p > 0.05$) in the Mn levels in the root and stem vegetables. On the other hand, there were significant variation ($p < 0.05$) between Mn content in collected vegetables from middle and southern districts and other districts in Saudi Kingdom. The obtained results declared that the amounts of Mn in the leafy vegetables were higher as compared to those in the other investigated vegetables especially in middle and southern districts respectively. The reasonable explanation is mainly attributed to Riydah and Damam cities which are heavily traffic and they include the major industries and factories in Saudi Kingdom leading to enforced heavy metals accumulation in the vegetables.

Our results were in agreement with that obtained by Tsoumbaris and Tsoukali-Papadopoulou (1994) who reported high concentration (25.83–70.76 µg/g) of Mn in cereals and fruits respectively in Greece. Manganese contents were in the safe limit permitted for food since WHO recommended 2–9 mg per day for an adult (WHO, 1994).

Concentration of Cu in the leafy vegetables, Jews mallow, spinach, and arugula amounted to 33.22, 24.38, 14.07, 10.0, 9.07 and 6.76 µg/g at middle and southern districts respectively (Tables 2 and 4). There are noticeable high significance level ($p < 0.05$) in Jews mallow and gradually decreased in spinach, arugula and parsley. However, concentration of Cu in the stem and root vegetables did not vary significantly ($p > 0.05$). The lowest Cu value (2.06 µg/g) was recorded in potatoes at northern district (Table 3). On the other hand, cereals and fruits maintained low significant levels ($p < 0.05$) especially between different districts.

It was found that heavy metals accumulated more in leafy vegetables than those in other parts because these leaves considered as entry points of heavy metals from air. Demirezen and Ahmet (2006) reported that levels of Cu (22.19–76.50 mg kg^{-1}) were higher in the leafy species than the non-leafy vegetable species from Turkey. Sharma et al. (2006) reported the concentration of Cu (2.25–5.42 mg kg^{-1}) in vegetables grown in wastewater areas of Varanasi, India to be within the safe limit. Furthermore, Radwan and Salama (2006) carried out a survey of various fruits and vegetables in Egypt and noted that the highest levels of Pb, Cd, Cu and zinc were present in strawberries, cucumber, dates and spinach, respectively. Our study revealed that leafy vegetable accumulated Cu higher than the permissible level (10.00 mg kg^{-1}, WHO, 1994), the concentration was very high in Jews mallow leaves (33.22 µg/g).

The obtained results declared that, zinc values have irregular fluctuation between different vegetables types and different districts (Tables 1–4). The amount of Zn in the samples of legumes was found to be higher than other vegetables especially in peas (71.77, 62.47 and 41.48 µg/g) at northern, middle and southern districts respectively (Tables 2–4). However, there was insignificant difference ($p > 0.05$) in Zn concentration among the cereals, stems, roots and fruits, whereas their values were fluctuated in a narrow range (8.27–30 µg/g).

The results showed a relative increase of zinc contents in most vegetables more than reported by Singh et al. (2004) and Itanna (2002), who reported that the Zn concentration (3.56–4.592 mg kg^{-1}) was within the recommended international standards. On the other hand, the present results were concordant with that obtained by Al Jassir et al. (2005) who reported the levels of Zn between 14.14 and 76.28 µg/g in some vegetables and they found to be higher in the purslane vegetable species for both washed and unwashed samples. Generally, the present study demonstrated the concentration of Zn to be within the set limits of international standards (5.00 mg kg^{-1}, WHO, 1994).

The concentration of Pb in cucumber samples collected from middle and Eastern districts (Riyadh and Dammam cities) have the highest value of 6.98 and 6.7 µg/g respectively (Tables 2 and 4). Pb contents in cucumber and rice were significantly ($p < 0.05$) higher as compared to those in the other vegetables, whereas, the kidney beans samples were found to be less significant ($p < 0.05$) since it has lower Pb contents. However, there was insignificant ($p > 0.05$) variation in the level of Pb in the leafy vegetables of parsley, Jews mallow and cabbage.

Increase of Pb levels in cucumber and rice especially in middle district was attributed to heavily traffic in this area which lead to the accumulation of Pb emitted from cars exhaustions. The recent results were in agreement with that obtained by Sharma et al. (2006) who reported the Pb concentration (17.54–25.00 mg kg^{-1}) in vegetables grown in industrial areas. Muchuweti et al. (2006) reported the level of Pb (6.77 mg kg^{-1}) in vegetables irrigated with mixtures of wastewater and sewage from Zimbabwe to be higher than WHO safe limit (2 mg kg^{-1}). Al Jassir et al. (2005) studied six washed and unwashed green leafy vegetables from Saudi Arabia and noted the highest concentrations of Pb in coriander and purslane.

Cadmium values showed low levels in roots, stems, leaves of parsley and Jews mallow and cereals vegetables as compared with other different vegetables, since the lowest value (0.92 μg/g) was recorded in rice at eastern district. The highest contents of cadmium found in leaves of spinach and arugula at four districts, respectively (Tables 1–4). High significance variation ($p < 0.05$) were be noticed of cadmium values in the samples of spinach, arugula, tomatoes and kidney beans, while a lower significance ($p < 0.05$) in cabbage, wheat and beans. However, there was insignificant ($p > 0.05$) difference between roots, stems and leaves of parsley and Jews mallow vegetables with regard to Cd content.

Fytianos et al. (2001) reported that spinach and lettuce grown in the soil of industrial area of Greece are enriched in Cd. Al Jassir et al. (2005) reported that levels of Cd were higher in the garden rocket vegetable species for both washed and unwashed samples. In our study, concentration of Cd was noted to be above the critical level of 0.1 mg kg^{-1} as reported by WHO (1994) and thus might be a great threat for the human consumers.

Mercury is more toxic than Cd and Pb and causes serious health problems such as loss of vision, hearing and mental retardation and finally death occurs. Mercury was not detected in some legumes species from northern district while it varied in a narrow range and showed their low levels in leafy vegetables except parsley which recorded the maximum mercury value (0.048 μg/g) among the studied vegetables at middle district (Table 2). Cucumber, tomatoes and wheat exhibit slightly high mercury values with relative significance ($p < 0.05$) with other studied vegetables. High significant variation ($p < 0.05$) was noticed in mercury values in the samples of parsley sweet potatoes and turnips, while there was insignificant ($p > 0.05$) difference between cereals and legumes vegetables.

The obtained data ranged between 0.009 and 0.048 μg/g which were in agreement with that obtained by Abbas et al. (2010) who reported that the concentration of mercury in food and foodstuff in Sindh, Pakistan ranged between 0.003 and 0.01 μg/g. The recorded values exceeded the permissible levels (0.03 μg/g) reported by the WHO.

Conclusion

The magnitude of heavy metals detected in different kinds of vegetables was arranged as Fe > Mn > Zn > Cu > Pb > Cd > Hg. The leafy vegetables contained the highest values of most heavy metals especially those collected from middle and eastern districts due to heavy industrial activities and heavy traffic in those districts. Furthermore, the concentrations of most studied metals are above the standard permissible levels thus might be in concern for the human consumers especially in middle and eastern districts.

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