HEAVY METALS CONTAMINATIONASSESSMENT FOR SOME IMPORTED AND LOCAL VEGETABLES

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ABSTRACT

The objective of this study, to was measure concentrations of some heavy metals in various imported and locally produced vegetable crops, including root crops (Turnips *''Brassica rapa*'' and Carrots *''Daucus carrota*''), stem crops (Potatoes *''Solanum tuberosum''* and Onion ''*Allium sativa*''), leaves (Lettuce *''Lactuca sativa*''), and fruits (Tomatoes *''Lycopersicon esculentum'')*.These crops were collected from Baghdad central whole sales. X-ray fluorescence spectrometry technique was applied to measure heavy metal concentrations. In the imported vegetables, heavy metal mean concentrations were arranged in the following increasing order: Fe>Zn> Cu>Ni>Co>Cd>Cr>Pb, whereas higher levels of Cr, Fe, Cu, Zn, Cd and Pb (1.2075, 165.995, 37.2275, 43.775, 6.0375, 1.48)mgkg^{-1}, respectively, were found within the locally produced vegetables. High level of Co 3.09625 mgkg^{-1} was also detected in onion, while, increased concentrations of Ni 7.8675mgkg^{-1} were found in lettuce collected from local market. Overall significant differences in the heavy metals concentration between imported and locally produced vegetables were observed. The daily intake of four main heavy metals (Cd, Cu, Zn and Pb) had been estimated which revealed highly consumption of Cd (310 and 372 µg per day) for imported and locally vegetables respectively. This study suggests raising the concern of the society and Iraqi government to this problem and to take in consideration its impact on general health and environment.

Keywords: X-ray spectrophotometer, Vegetable Crops, Baghdad local markets.

مجلة العلوم الزراعية العراقية -2018 :49(5):794-802 حسون

تحديد التلوث بالمعادن الثقيلة لبعض الخضروات المستوردة والمحلية

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المستخلص

تهدف هذه الدراسه الى تقدير بعض تراكيز العناصر المعدنيه الثقيله في محاصيل خضر مختلفه مستورده و محليه و التي تضمنت الجذور (الشلغم "*Brassica rapa*" و الجزر "*Daucus carrota*") و السيقان (البطاطا "*Solanum tuberosum*" و البصل "*Allium sativa*") و الاوراق (الخس "*Lactuca sativa*") و الثمار (الطماطا "*Lycopersicon esculentum*"). جمعت هذه المحاصيل من اربعة أسواق رئيسيه في بغداد.اجري الكشف عن المعادن الثقيله بتطبيق التقنيه الضوئية لمطياف اشعة X.كان متوسط المحتوى المعدني في الانواع المستورده بالتسلسل الاتي:Fe>Zn>Cu>Ni>Co>Cd>Cr>Pb. في حين أن اعلى مستوى للمعادن Cr وFe و Cu و Zn و Cd و Pb(1.2075،165.995،37.2275،43.775،6.0375،1.48) ملغم كغم$^{-1}$بالتتابع. كما لوحظ اعلى مستوى لعنصر Co (3.09625)ملغم كغم$^{-1}$في عينات البصل. بينما سجل عنصرNi(7.8675) ملغم كغم$^{-1}$زياده في التركيز في عينات الخس.ظهرت فروقا معنويه بين الخضروات المستورده و المنتجه محليا. اضافه الى ذلك فقد تم تقدير المعدل اليومي لتناول اربعة معادن رئيسيه و هي Cd و Cu و Zn و Pb والتي اظهرت استهلاكا عاليا لعنصر Cd(310 و372 مايكروغرام في اليوم الواحد) للمستوردة والمحلية بالتتابع. ان هذه الدراسه تقترح زيادة اهتمام الحكومه و المجتمع بهذه المشكله و الاخذ بنظر الاعتبار تأثيراتها على الصحه العامه و البيئه.

الكلمات المفتاحية: مطياف الاشعه السينيه، محاصيل الخضروات، أسواق بغداد المحلية.

***Received:25/2/2018, Accepted:3/5/2018**

INTRODUCTION

Vegetables provide a fast, low cost and an adequate source of vitamins, minerals, and fibers (2).Vegetables contain fundamental nutrients such as proteins, calcium, which is part of the essential life- supporting materials for humans and animals (34). During the digestion process, vegetables play an important role in neutralizing agents for acidic substances that arise during this process (35). Moreover, the most important benefit of vegetables is their anti-oxidative effects against different toxicants (11). Various plant species have their nutritive requirements, including minor and macro elements which support the budding and growth rate (14).

Several traces of heavy metals are regarded as micronutrients which plants and higher animals demand in fewer quantities, so health problems might be caused when these metals concentrations are elevated more than acceptable limits (22). A wide range of minerals are utilized by the plants through their roots from the contaminated soil and are transported to the shoot apex and accumulate in different edible plant parts (25). Another pathway of entrance is already identified via the atmospheric deposition on the leaves surfaces, then absorbed by the plant issues (16). This explains why these contaminants are reaching human food chain, leading to many health issues within populations exposed to them. Heavy metals are classified depending on their chemical properties which include atomic number, atomic weight, and toxicity (27). Their harmful effects are due to their non-biodegradability, long biological half-life and potential accumulation in the body organs (29).Heavy metals emission sources in the environment are both; natural, such as soil, rock erosion, and dust or anthropogenic, such as industrial activities, mining, rapid organization, transportation and pesticides (38). Extended and exacerbated application of organic manures and both organic, inorganic fertilizers, can lead to high levels of heavy metals as well as other ions (2).In addition this random application of fertilizer not dissolved soil fertility problems and not increased its efficiency to provide crops with nutrients unless its insure a good nutrient balance in order to produce an ideal yield (3). Recent studies revealed that wastewater effluents are loaded with different types of heavy metals, including cadmium (Cd), capper (Cu), zinc (Zn) and nickel (Ni), which their effect is going beyond the physiological necessity of plant but instead tending to be magnified in next steps of food chain (30). This is already proved by their binding ability to different protein molecules, preventing the replication of DNA and affecting cells division (8). It has become obviously, that prolong consumption of heavy metals can lead to numerous defects in biochemical processes of nervous, vascular systems as well as bone diseases (15). Some metals such as Cu and Zn, when exceeding allowable limits, are acting as oxidative stressing factors through oxidative -reduction reaction (redox), thus altering the production -utilization of energy in living systems (31).Lead (Pb) has the same oxidative effect and can cause mental retardation in children (7). In contrast, chromium (Cr), especially (CrVI) form, is carcinogenic. Whereas glomerular damage and metabolic alteration are caused by cadmium (Cd) long term exposure (24). Simultaneously, cobalt (Co), nickel (Ni), iron (Fe), have received more attention because of their adverse effect on health (18). Hence, these facts insure their association with many chronic diseases in humans (10). Heavy metals harmful influence and persistence in vegetables have been well studied and documented extensively in many countries around the world (5). This study aimed to measure the concentration of heavy metals in imported and local vegetables at four markets of Baghdad city.

MATERIALS AND METHODS

Four local sites were chosen for samples collection during 2016 (Al-Taji, Al-Shuala, Al-Zaafaraniya and Al-Rahased markets), which represent the northern, western, eastern and southern sites of Baghdad city, respectively. All those markets serve as the whole suppliers and sales sites for vegetables to other internal or minor markets in the city. For local produced crops, selected vegetable were collected in two different times. These samples were classified to root crops (Turnips "*Brassica rapa*" and Carrots "*Daucus carrota*"), stem crops (Potatoes "*Solanum tuberosum"* and Onion "*Allium sativa*"), leaves

(Lettuce *"Lactuca sativa*") and fruits (Tomatoes *"Lycopersicon esculentum"*). Samples were designed in such way to cover common edible parts of vegetables which repeatedly consumed by people at Baghdad city. Regarding imported vegetables, two batches were chosen within two time intervals during the year in which the study was carried on. This was done to insure more accuracy of the collected data. Samples were put in plastic bags and closed tightly to prevent any cross contamination, then brought to the laboratory for further analysis. The collected samples were washed thoroughly with tap water first and then with deionized water to remove dust particles. These samples were cut into small pieces using a clean knife. Different parts of roots, stems, leaves and fruits were air dried for two days in shade and then kept in hot oven not more than 70ºCto dry and left to cool at room temperature (25).Each sample was grinded in to a fine powder, using mortar at first and then a commercial blender. The powders were passed through a 212 µm mesh sieve and finally stored in air tight sealed screw cups appropriately labeled until for analysis (19). Three grams of the prepared vegetables powder were weighed and taken to analysis (4).Determination of heavy metals, such as (Cr, Fe, Co, Ni, Cu, Zn, Cd and Pb) levels was carried out using x-ray fluorescence spectrometry according to methods described previously(6) (9). The following equation was applied to calculate the daily intake of heavy metals within the vegetables tested in this study by Elbagermi *et al.* (11):

$$\boldsymbol{Daily\ intake\ of\ heavy\ metals}\left(\frac{\mu g}{day}\right) \boldsymbol{or} \left(\frac{mg}{day}\right) = \boldsymbol{daily\ vegtables\ consumption} \times \boldsymbol{concentration\ in\ eaten\ parts}$$

This equation was mainly used to calculate the concentration of Pb, Cd, Cu and Zn as recommended by Joint FAO/WHO (17). Data was analyzed and displayed using GraphPad virgin 6 (v6) and significance was determined using multiple T-test analysis, depending on data normalcy.

RESULTS AND DISCUSSION

The mean concentrations of heavy metals for imported batches of collected vegetable samples are given in Table 1. The results demonstrated that the detected heavy metals are present in all samples and their concentrations in these samples are varying. Lead (Pb) ranged from 0.325 to 1.5575mgkg^{-1} in tomato and lettuce. It was observed that Fe had the maximal values in lettuce samples (411.625mgkg^{-1}). While the highest concentration of Cu (13.65 mgkg^{-1}) was observed in tomatoes. In the case of Zn, this element value was between 33.525 mgkg^{-1} in onion and reached to 24.88mgkg^{-1} in carrots. In the terms of its concentration, Ni was found to be variable as following:

Lettuce>tomatoes>turnips>onion>carrot> potatoes.

In table1, the average of Cd was ranging from (1.625-5.575) mgkg^{-1}. Interestingly, Co had similar concentration in all samples which was about (3.1) mgkg^{-1}. Cr was present in close values in almost all collected samples except with turnips which had the highest one (1.7475mgkg^{-1}). From what is previously mentioned, results showed that the concentrations of heavy metals in vegetable samples were found according to their abundance in the following order: Fe>Zn> Cu>Ni>Co>Cd>Cr>Pb.

Mean concentrations of heavy metals in different local vegetables are given in Table2.

Approximately, the same variation in heavy metal concentrations for imported groups was observed in local types.Results showed that value of Fe in lettuce (leafy vegetables) is higher than other vegetables. Pb concentration in tomatoes 0.465 mgkg^{-1}. Whereas the highest value of Pb was observed in turnips (1.48mgkg^{-1}). Simultaneously, Table 2 illustrated an interesting value of Cu (37.2275mgkg^{-1}) and Zn (43.775mgkg^{-1}) in turnip samples, when compared with other vegetables. The highest concentrations of Ni were noticed in lettuce (7.8675mgkg^{-1}) followed by a 7.7775 mgkg^{-1}in turnips. However, Cd maintained its range 1.525mgkg^{-1} in turnips and 6.0375mgkg^{-1} in onion. Interestingly, Cr and Co concentrations in local vegetables were similar to those recorded in imported types (1.03 and 3.1 mgkg^{-1} respectively).In spite of new values such as Cr (1.2075) mgkg^{-1} in lettuce, and for Co (2.8375) mgkg^{-1} in potatoes. Statistical analysis revealed that significant differences were noticed between local and imported vegetables collected from various sites. The obtained results were compared with recommended limits established by FAO/WHO and other international guide lines of heavy metals in food (edible parts of different vegetables) to assess levels suitable for human consumption (17). The most important factor is the intake by human. In contrast, there is no national legislation dealing with standard levels of heavy metals in vegetables, so that most of published studies depend on these guidelines to insure improvement of food safety (21).As mentioned before, the concentration of Fe in lettuce was high. Fe concentrations were significantly ($p<0.01$) higher as compared to other vegetables.

Table 1. Heavy metal concentrations ($mgkg^{-1}$) dry matter represented as (means $\pm$ SD)for 1st and 2nd batches of imported vegetables

Metals	Root		Stem		Leaf	Fruit	
	Turnips	Carrot	Onion	Potatoes	Lettuce	Tomato	
Cr	1.7475	1.1725	1.03	1.03	1.03	1.025	mean
	1.435	0.285	0	0	0	0.005773503	standard deviation
Fe	15.875	8.64	59.625	29.475	411.625	34.325	mean
	3.73396572	11.28396207	29.49727389	3.685444342	30.7432784	8.054553578	standard deviation
Co	3.0925	3.0825	3.1	3.09	3.09	3.1	mean
	0.015	0.020615528	0	0.02	0.02	0	standard deviation
Ni	8.53	7.15	8.3825	6.4125	10.35	9.6	mean
	0.349666508	0.602771377	1.043020454	0.698349244	0.86986589	0.565685425	standard deviation
Cu	6.35	10.875	10.15	11.23	8.0325	13.65	mean
	0.802080628	0.579511288	1.011599394	0.801415415	2.124984314	0.946924847	standard deviation
Zn	22.2	24.88	33.525	19.525	20.35	17.325	mean
	0.716472842	1.763027699	1.873276986	0.699404509	1.024695077	0.298607881	standard deviation
Cd	2.6	1.625	3.875	5.575	1.675	1.95	mean
	1.366260102	1.452297031	3.314991202	3.298863441	1.337597348	1.201388086	standard deviation
Pb	0.74	0.53	0.63	1.09	1.5575	0.325	mean
	0.127279221	0.282842712	0.579827561	0.579827561	0.141421356	0.141421356	standard deviation

Table 2.Heavy metal concentrations ($mgkg^{-1}$) dry matter represented as (means $\pm$ SD) for local vegetables

Metals	Root		Stem		Leaf	Fruit	
	Turnips	Carrot	Onion	Potatoes	Lettuce	Tomato	
Cr	1.0375	1.02875	1.0275	1.02875	1.2075	1.11375	mean
	0.017525492	0.003535534	0.0046291	0.003535534	0.244992711	0.171041975	standard deviation
Fe	21.58222222	22.33333333	33.55888889	17.32555556	165.9955556	22.28888889	mean
	11.88904418	11.8759962	34.19678721	23.28998033	116.1660165	23.12992805	standard deviation
Co	3.0825	3.095	3.09625	2.8375	3.08625	3.0875	mean
	0.028660575	0.014142136	0.010606602	0.730415733	0.019226098	0.017525492	standard deviation
Ni	7.7775	7.2675	7.15375	7.44125	7.8675	6.31375	mean
	0.955013089	0.386032937	1.223098378	3.587933896	1.231871399	0.764253838	standard deviation
Cu	37.2275	9.5375	10.515	18.8625	11.10125	7.25875	mean
	37.50777891	2.525123759	1.556213353	5.993791431	1.812814997	1.349777098	standard deviation
Zn	43.775	33.775	31.4875	26.1	29.1125	25.30125	mean
	28.37653709	16.84329031	6.521270144	3.318347265	4.428297803	2.127375563	standard deviation
Cd	1.525	4.0875	6.0375	5.975	1.925	3.4375	mean
	0.936177639	2.46254079	2.773825363	4.10182886	1.393607856	2.985171687	standard deviation
Pb	1.48	0.6825	0.6825	0.7875	0.835	0.465	mean
	1.260249408	0.301128829	0.361494912	0.333969631	0.209965984	0.320847093	standard deviation

Similarly, this was found by Santamaria *et al.* (28) that different parts of plant had varied concentrations of heavy metals. Moreover, they reported that leaves had higher levels of these contaminants. In present study, Fe recorded concentrations reached to (411.625) $mgkg^{-1}$ (table, 1) which is considered much higher than those reported by Zahir and Mohi (36) when they observed that the Fe concentration in different analyzed vegetables ranged from 7.9-24.8 $mgkg^{-1}$.The present results were in agreement with those found by Ali and Al-Qahtani (5) who reported Fe concentrations 31.9$mgkg^{-1}$and 543.5$mgkg^{-1}$ for onion and parsley, respectively. The reason behind the Fe high concentrations in leaves is that leaves are considered as food making factories in plant; therefore Fe can be promoted and accumulated in them. This explains why carrot (root) contains concentration (8.64$mgkg^{-1}$ Table, 1).

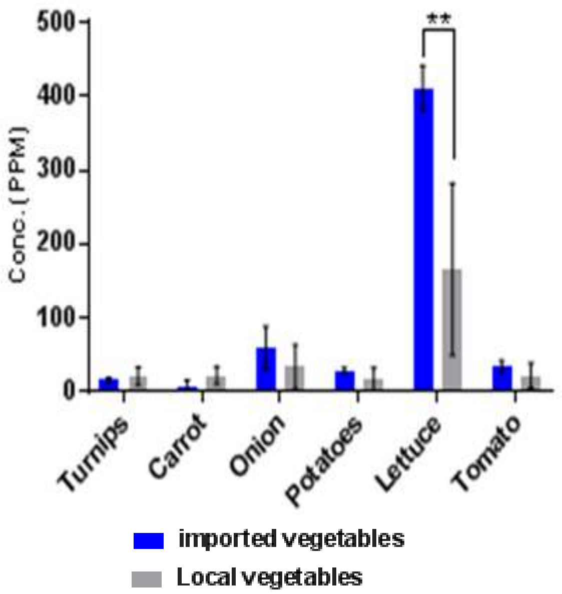

Figure 1. Mean concentrations of Fe levels ($mgkg^{-1}$) in imported and local vegetables ** (p <0.01)

Results showed that some Cu values are within acceptable limits (10$mgkg^{-1}$) as recommended by FAO/WHO (21).There was a significant variation ($p<0.001$) between imported and locally samples of tomato.

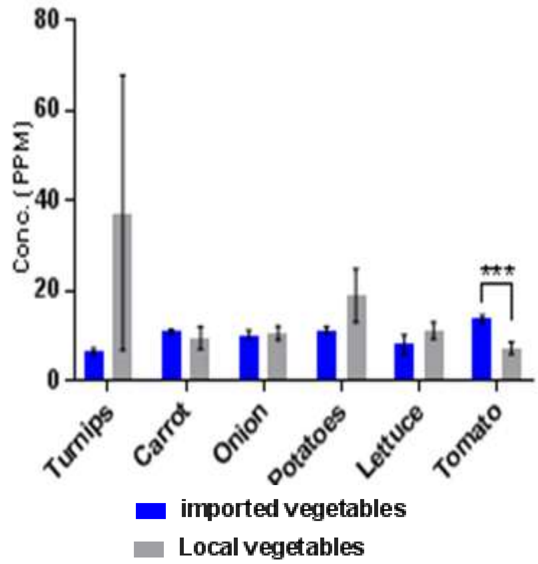

Figure 2. Mean concentrations of Cu levels ($mgkg^{-1}$) in imported and local vegetables * (p <0.001)**

In some lettuce samples, Cu was in safe limits and this interpreted by Parvin *et al.* (25) who pointed that cupper concentrations are corresponding to chlorophyll richness in leafy vegetables. However, imported turnips indicated a low cupper value (6.35 $mgkg^{-1}$Table 1) which was similarly obtained by Mills and Jones (23) when they suggested two forces are controlling Cu absorption in vegetables. These forces are including: pH imbalance and an excess of nutrients such as phosphorus. Regarding the Zn concentration results, an increase above the permitted standards (5-20 $mgkg^{-1}$) was observed (32).

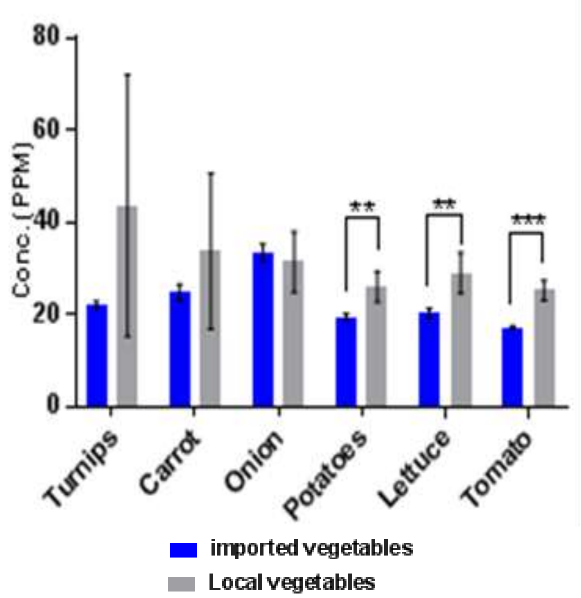

Figure 3. Mean concentrations of Zn levels ($mgkg^{-1}$) in imported and local vegetables ** (p <0.01), * (p<0.001)**

There were a significant differences ($p<0.01$) for potatoes and lettuce. Moreover, tomatoes showed the same high significance ($p<0.001$). In contrast, Pb contents occurred within steady limits (0.465-1.5575) $mgkg^{-1}$ for local tomatoes and imported lettuce.

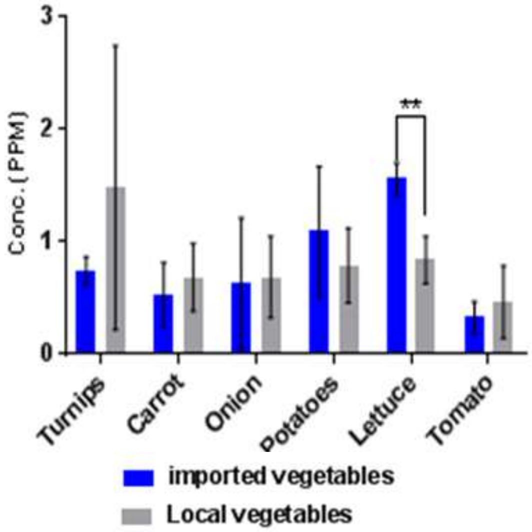

Figure 4. Mean concentrations of Pb levels ($mgkg^{-1}$) in imported and local vegetables
**** ($p<0.01$)**

Figure 4 indicated significance of variation ($p<0.01$) with in lettuce. The variations in Pb concentration were because of the traffic intensity effect, since this heavy metal is emitted from cars exhaustion (5). In Iraq, leaded fuel is still used for vehicles and diesel generators, resulting in emitting large amounts of Pb. Lokeshwari and Chandrappa (20)mentioned that some soil factors might have an impact on stimulating plants to increase their Pb uptake, including: pH, particle size, cation exchange capacity of soil, root exudation and other physico-chemical parameters. Taghipour and Mosaferi, (32) described more factors such as moisture, temperature, soil properties and degree of plant maturity that could influence Pb uptake by plants. Fergusson (13) found that the ability of heavy metals to transmit and form stable coordinated compounds with organic and inorganic matter makes them potentially toxic. The same approach was obtained by Rapheal and Adebayo (26) when they suggested that heavy metals are able to interact with other soil organic compounds and absorbed by growing plants and that these elements become more dangerous in the form cation or when bound to short chain of carbon atoms rather than free state. In general, Iraqi soil is lacking Pb element therefore, it is obviously a substantial amount to be added along with aqueous phosphate fertilizers (6). The present results indicated various concentrations of Cd most of them are higher than normal levels (0.05-1 $mgkg^{-1}$) as reported by (33).Enrichment of the soil with cadmium leads to its higher presence due to two combined factors; the first is its relative mobility whereas the second is its affinity to associated with organic matters (26).

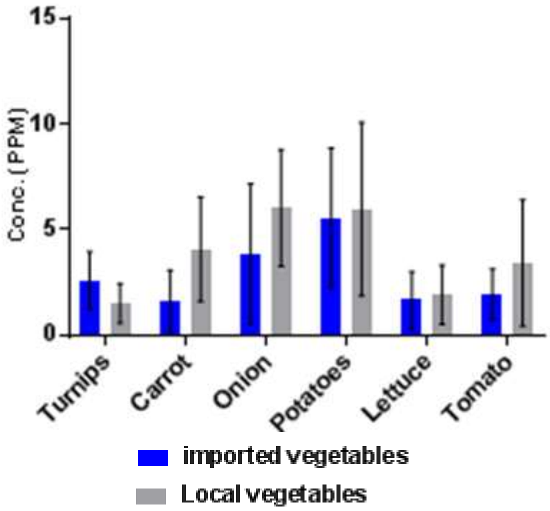

Figure 5. Mean concentrations of Cd levels ($mgkg^{-1}$) in imported and local vegetables

No significant differences were found among collected vegetables. In this study, Cr levels ranged between (1.025-1.7475) $mgkg^{-1}$ which was close to what was noted by Mutune, *et al.* (24) in an industrial area when they set limits between 1.19and1.24 $mgkg^{-1}$ for spinach and spider plants respectively. Chrome plates corrosion of vehicular motors may increase the emissions of this element (12).

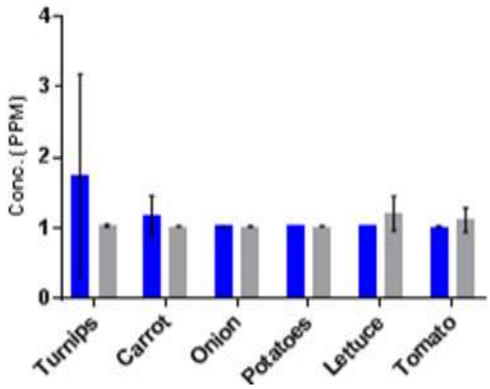

Figure 6. Mean concentrations of Cr levels ($mgkg^{-1}$) in imported and local vegetables
*** ($p<0.05$**

Significant differences ($p<0.05$) are scored among imported and local samples. Co levels demonstrated a significant difference ($p<0.05$) as shown in figure 7.

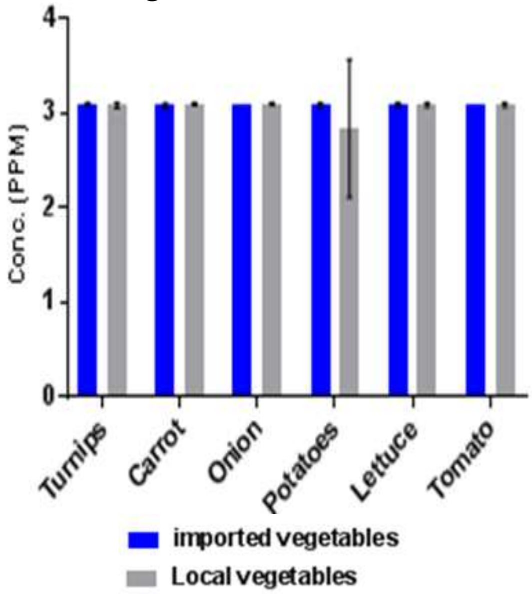

Figure 7. Mean concentrations of Co levels (mgkg^{-1}) in imported and local vegetables * ($p <0.05$)

Generally, cobalt concentration in this study was higher than those obtained by Lawal and Audu (19). They found 0.12 to 1.14 mgkg^{-1} in onion and the lowest was recorded in tomatoes. Nickel, determined in this work, was above the critical limits (2.7 mgkg^{-1}) which were recorded by the national agency for food and drug administration and controls (19).

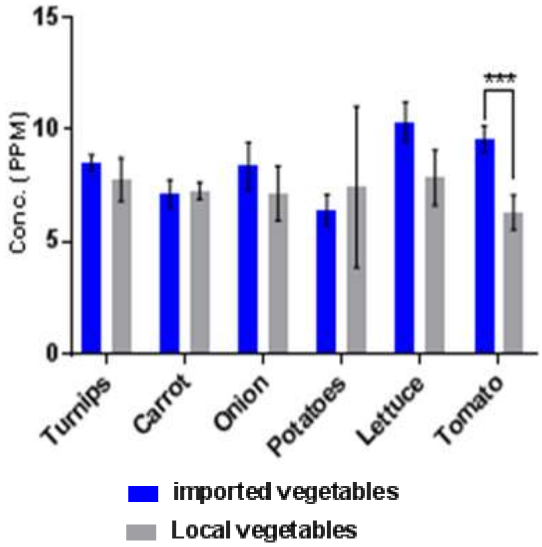

Figure 8. Mean concentrations of Ni levels (mgkg^{-1}) in imported and local vegetables * ($p<0.001$)**

Statistical analysis for Ni concentrations indicated a significant variation ($p<0.001$) in tomato. Lettuce as (GLV) reported high value (table 1) as compared with the study of Abdulazeeza and Azizb (1) which fall in the range between 0.037-0.503 mgkg^{-1}.Another source of Ni is the corrosion of vehicular engines, especially the old ones (12). In a relevant research, Mahakalkar, *et al.* (22) described what is known as transfer factor (TF) of different heavy metals from soil to vegetables. They show that TF varied from metal to another depending on efficiency of plant species to accumulate a given metal. This might explain the variance of metals values observed among different vegetables in this study. Furthermore, Zeng and Mei (37) expressed that transportation and marking system is involving in elevating metals level than production sites. Recently, in Iraq groceries are scattered near the main road and high ways that make them in contact with automobile emissions. Furthermore, washing vegetables with river water at farming sites may increase these contaminants. FAO/WHO set certain limits for the daily intake of some heavy metals underlying terms of Provisional Tolerable Daily Intake (PTDI) (17). These standard limits depending on an average weight of adult person (60 kg) consuming at least 98 g of vegetables per day. According to equation mentioned before by Elbagermi, *et al.* (11), the obtained results for imported and local vegetables were shown in Table 3. Table 3 exhibited the values of heavy metals daily intake for both imported and local vegetables.

The results revealed that estimated daily intake of heavy metals in this study is above those documented in Misurata markets (11). But still within acceptable limits, as recommended by FAO/WHO (214µg, 3mg, and 60mg) for Pb, Cu and Zn respectively except Cd reported more than its permissible limits (60µg per day)(17). It is recommended that healthy diet should include the consumption of large amount of vegetables to provide the body with essential and necessary compounds which supporting the survival underlying what is known as protective food. Unfortunately, vegetables as plants have the ability to accumulate higher concentrations of heavy metals from different pathways. They are the main supplier of these contaminants to food chain due to their location in life system. This study represented an attempt to evaluate the values of these heavy metals in various vegetables edible parts widely consumed by peoples in Baghdad city and to ensure their adverse effect on human being. Most of detected metals, in this study are above the recommended levels proposed by FAO/WHO and other International Agencies. Consequently, it would be helpful to investigate the importance of long term exposure to food stuffs with such contaminants and giving awareness to their influences on public health. Moreover, the daily up take of some heavy metals was calculated in order to estimate the limits of those contaminants in daily meal

Table 3.The daily heavy metals intake through consumption of some imported and local vegetables

Food stuff (98) g g/person/day	Pb	intake µg/day	Cd	intake µg/day	Cu	intake mg/day	Zn	intake mg/day
Imported vegetables, Mean level ($mgkg^{-1}$)								
	0.812	79.576	3.163	310	10.05	0.985	23	2.25
Local vegetables, Mean level ($mgkg^{-1}$)								
	0.822	80.6	3.8	372	15.8	1.55	31.6	3.1

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