Assessment of trace metals in five most-consumed vegetables in the US: Conventional vs. organic

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ABSTRACT

Metal concentrations (As, Cd, Pb, Cr, Ba, Co, Ni, Cu, and Zn) in conventional and organic produce were assessed, specifically, five most-consumed vegetables from the US including potato, lettuce, tomato, carrot and onion. All vegetables contained detectable metals, while As, Cd, Pb, Cr, and Ba are toxic metals, Co, Ni, Cu, and Zn are nutrients for humans. The mean concentrations of As, Cd, Pb, Cr and Ba in five vegetables were 7.86, 9.17, 12.1, 44.8 and 410 mg/kg for organic produce, slightly lower than conventional produce at 7.29, 15.3, 17.9, 46.3 and 423 mg/kg. The mean concentrations of Co, Ni, Cu, and Zn in five vegetables were 3.86, 58.5, 632, and 2528 mg/kg for organic produce, comparable to conventional produce at 5.94, 68.2, 577, and 2354 mg/kg. For toxic metals, the order followed tomato < lettuce < onion < carrot < potato, with root vegetables being the highest. All metals in vegetables were lower than the allowable concentrations by FAO/WHO. Health risks associated with vegetable consumption based on daily intake and non-carcinogenic risk based on hazard quotient were lower than allowable limits. For the five most-consumed vegetables in the US, metal contents in conventional produce were slightly greater than organic produce, especially for Cd and Pb.

1. Introduction

Over the last few decades, growing awareness of food safety has stimulated research concerning the risks associated with consuming foods contaminated by trace metals (D’Mello, 2003; Gomiero, 2018). Food chain via soil-plant-human can be a significant route for human exposure to trace metals compared to other routes (Cao et al., 2016; Chen et al., 2016). Fresh vegetable consumption in the US has increased by 19% from 1970 to 2005 and is projected to continue to increase through 2020 (Wells and Buzby, 2008; Lin et al., 2003).

Vegetables play a major role in contributing to a healthful diet across the world, as they are an important source of carbohydrates, vitamins, minerals, proteins, and fibers. Consumption of vegetables is an important pathway for the intake of essential nutrients. However, vegetables may also accumulate trace metals via root or foliar uptake, which is influenced by the environment and crop species (Bakkali et al., 2012). Contamination of vegetables by trace metals may be related to irrigation with contaminated water, addition of fertilizers and metal-based pesticides, and industrial emissions (Radwan and Salama, 2006).

In recent years, increasing levels of trace metals in vegetables have been reported, which may impact human health (Arora et al., 2008; Pan et al., 2016). This is because trace metals including As, Cr, Cd, and Pb are systemic toxicants, which can induce damage in multiple organs even at low exposure. As such, they are classified as human carcinogens according to the USEPA. The most affected population include children and women. Therefore, dietary intake of vegetables containing metals may pose potential health risks to consumers, this is because once into human bodies via...
consumption, there is little physiological mechanism for their excretion, hence, they have a long half-life at 10–30 years (Gomiero, 2018).

Due to growing public concern with the safety and quality of conventional products, organic food has been growing rapidly worldwide (USDA, 2017). According to the International Federation of Organic Movements (Willer and Lernoud, 2016), organic agriculture is being practiced in 172 countries. The US ranks first in organic food consumption, representing 5.3% of retail food sales. It is forecasted that the market for organic produce will continue to grow (Gomiero, 2018). Most consumers believe that organic produce is healthier and safer than conventional produce, so they are willing to pay a premium price for it (Zehnder et al., 2003; Magkos et al., 2006). This perception is mainly due to the principles associated with organic vegetable production. Organic production largely avoids the use of synthetic fertilizers, pesticides, growth regulators, and livestock feed (Codex Alimentarius Commission, 2007). However, whether organic produce is better than conventional is still in debate (Krejcová et al., 2016).

In some countries, vegetables are exposed to trace metals by anthropogenic activities, thus vegetables consumption can cause adverse health effects. Studies on trace metals in vegetables have focused on Asia including China (Bi et al., 2018), India (Gupta et al., 2008), Bangladesh (Shaheen et al., 2016), and Armenia (Pipoyan et al., 2018), South America including Brazil (Corguinha et al., 2015) and Chile (Munoz et al., 2002), and Europe including Greece (Karavoltsos et al., 2008), Spain (Olaia et al., 2015) and Germany (Antoniadis et al., 2017). For example, Gupta et al. (2008) reported 17.8 mg/kg Cd and 57.6 mg/kg Pb in radish from suburban areas in Titagarh, India, which were irrigated with contaminated wastewater. In China, a food survey in Wuzhou found the Pb content in 7% leafy and 18% root vegetables exceeded the Chinese national standard for Pb at 0.3 mg/kg (Chen et al., 2013). Wachirawongsakorn (2016) investigated metal contents in vegetables from fresh markets of Thailand. They found that Pb and Cd concentrations in more than 80% vegetables exceeded the maximum allowable concentration (MAC) by National Food Institute criteria, averaging 0.96–3.39 and 0.48–1.40 mg/kg. Hu et al. (2013) reported that 26, 16 and 0.56% of market vegetables in Hong Kong were contaminated with Pb, Cd, and Cr. As vegetables are essential food for daily consumption, metals in vegetables may contribute to the provisional tolerable weekly intake (PTWI) established by the FAO/WHO. However, there is limited information on metal levels in vegetables in the US, especially a comparison between organic and conventional vegetables. Based on limited studies, most of rice-consumption-related exposure of Hg is below PTWI based on WHO (1992), which was adjusted by Joint Expert Committee on Food Additives (JECFA, 2016). Whether this finding can be extended to other metals needs further attention.

In this study, five most-consumed vegetables in the US based on Produce for Better Health Foundation (2015) were selected, including lettuce (Lactuca sativa), potato (Solanum tuberosum), carrot (Daucus carota), tomato (Lycopersicon esculentum) and white onion (Allium cepa). In addition, lettuce, tomato, and onion were chosen due to high diet consumption, especially in fast food, carrot for their popularity in the US and potato as a major source of Cd, partially due to P fertilizer.

The aim of this study was to compare the concentrations of trace metals including As, Ba, Cd, Cr, Pb, Co, Cu, Ni, and Zn in organic and conventional vegetables (lettuce, potato, carrot, tomato, and onion) and assess the health risk associated with vegetable consumption. The daily metal intake from vegetable consumption for children and adults was estimated and compared with health-based guidance values. The potential risk of trace metals from vegetables to human health was also evaluated based on a hazard quotient.

2. Materials and methods

2.1. Collection of vegetable samples

Five most-consumed vegetables including organic and conventional lettuce, potato, carrot, tomato, and white onion were bought from four different supermarkets with a national chain during March–April 2017 in a college town in Florida, USA. For each fresh vegetable, five samples were collected from each supermarket to create a composite sample, with a total of 120 samples (5 vegetables x 2 sources x 4 supermarkets x 3 replicates). Organic vegetables were certified with an organic USDA label.

Following transport to the laboratory, all samples were washed three times with deionized water to remove soil particles and surface environmental pollutants and then rinsed with double-deionized water before placing on cellulose paper to remove excess moisture. The samples were peeled (potato, carrot, and onion), chopped into small pieces and well mixed. Fresh weights were taken before being dried in an oven at 65 °C for 3–5 d. All samples were ground into powder using an electric grinder.

2.2. Sample digestion and analysis

All glassware was soaked in nitric acid bath for at least 24 h before being rinsed three times with deionized water. All chemicals were of analytical grade or better. Nitric acid (trace metal grade), and H₂O₂ were obtained from Fisher Scientific (Waltham, MA). Prior to analysis, 0.5 g of sample was digested with HNO₃/H₂O₂ using USEPA Method 3050B on a hot block (Environmental Express, Ventura, CA). Elemental contents were analyzed by inductively coupled plasma mass spectrometry (ICP-MS; Perkin-Elmer Corp., Norwalk, CT). Multi-element standard solution (Perkin-Elmer, Inc. USA) was used after appropriate dilution for calibration and quality assurance and quality control protocol. Standard solution and sample blanks were analyzed after every 20 samples to monitor the stability of ICP-MS.

2.3. Health risk assessment of vegetable consumption

In this study, potential risk of metal exposure from vegetable consumption was assessed. The daily exposure of metals was estimated following USEPA (1989) and FAO/WHO (2016):

\[ CDI = C \times DI/BW \]  

Where CDI is the chronic daily intake [µg/kg/bw/d], C is the metal concentration in vegetables [µg/kg fresh weight], DI is the average daily consumption of vegetables [kg/person/d], and BW is the average body weight [kg]. The average daily ingestion rate of food for adults and children were 0.34 and 0.23 kg/person/d (Kimmons et al., 2009); average adult and children body weights were 70 kg and 32.7 kg (FAO/WHO, 2016).

In addition to CDI, the non-carcinogenic risk of vegetable consumption was characterized using a hazard quotient (HQ), which is the ratio of the CDI to the reference dose (RfD) for metals [µg/kg/d].

\[ HQ = CDI/ RfD \]  

A reference dose corresponds to the maximum amount of a metal that can safely be consumed per day, per kg body weight, which was obtained from the International Toxicity Estimates for Risk Database. The RfD values for trace metals are As = 0.3, Cd = 1.0, Cr = 3.0, and Pb = 15 µg/kg bw/d (TERA, 2013). If the HQ is 1, then the CDI of that metal exceeds the RfD, indicating that there is a potential risk.
2.4. Quality control and data analysis

Two standard reference materials including SRM 1547 (peach leaves) and 1573 (tomato leaves) from the National Institute of Science and Technology (NIST, Gaithersburg, MD) were included in the digestion as a part of the QA/QC protocols. The results (As = 0.110 ± 0.003; Cd = 1.59 ± 0.005; Cr = 1.86 ± 0.110 μg/kg) agreed with the certified values (As = 0.112 ± 0.004; Cd = 1.52 ± 0.04; Cr = 1.99 ± 0.06 μg/kg). QA/QC protocols included triplicate analyses and the use of standard and spiked solutions every 20 samples. The mean recovery of standard solutions was 96 ± 15%, while the spike recovery was 94 ± 3.6%. The performance of the ICP-MS was checked by running an intermediate calibration standard every 20 samples. All calibration standard checks were within the acceptable range (80–120%). The method detection limits for ICP-MS were (μg/L): As (0.04), Cd (0.08), Pb (0.09), Cr (0.03), Ba (0.02), Co (0.01), Ni (0.02), Cu (0.04), and Zn (0.08).

To better understand the relations among all metals, principal component analysis (PCA) was used. It uses an orthogonal transformation to convert variables into a set of values of linearly uncorrelated variables called principal components. In this case, concentrations of 9 metals in different vegetables including organic and conventional were analyzed. Elemental concentrations were calculated on a fresh weight basis (fw) and expressed as means of three replicates with standard deviation. Significant differences in the means of organic and conventional vegetables were determined by Wilcoxon/Kruskal-Wallis tests at p < 0.05. Principal component analysis was performed to compare organic and conventional vegetables. All statistical analyses were performed using JMP®Pro 13.0 statistical software (SAS Institute, Inc., Cary, USA) while figures were created using SigmaPlot (version 10.0, Systat Software Inc., San Jose, CA). The significance of differences was established with a single-factor variance analysis of ANOVA at α = 0.05.

3. Results and discussion

In this study, concentrations of As, Ba, Cd, Cr, Pb, Co, Cu, Ni and Zn were determined in five most-consumed vegetables in the US, including lettuce, potato, carrot, tomato and white onion. The types and number of vegetables were selected based on the availability of samples, especially for organic vegetables in selected markets. Therefore, this should be considered when interpreting the results. All vegetables contained detectable metal concentrations, while As, Cd, Pb, Cr, and Ba are toxic metals (Table 1), Co, Cu, Ni and Zn are nutrients for humans (Table 2). The five vegetables were purchased from four representative supermarkets of national chains. Based on metal contents, no consistent trend was observed (Tables 1 and 2).

The maximum allowable metal concentrations in vegetables were expressed based on fresh weight. However, different countries have different regulations, with the US lagging behind. For example, the As standard is 200 μg/kg for fresh vegetables in Poland, 500 μg/kg for rice, beans and vegetables in China (NHFPC, 2012), and 1000 μg/kg for spinach (Spinacia oleracea), tomato and cucumbers (Cucumis sativus) in Japan. The Cd standard for European Union is 100 μg/kg for root vegetables, 200 μg/kg for leafy vegetables, and 50 μg/kg for others (JECA, 2016). For the US, only reference dose for Cd in food at 1 μg/kg/d is available (AFSDR, 2008). For Pb, it is 50 μg/kg for lettuce and tomato, and 100 μg/kg for potato (JECA, 2016). However, based on NHFPC (2012), it is 100 μg/kg for tomato, onion, and carrot, 200 μg/kg for potato, and 300 μg/kg for lettuce. Since the coverage of NHFPC (2012) for vegetables is most extensive, it was used in this study to be consistent (Table 1).

3.1. Comparing metal contents among different vegetables

The mean concentrations of As, Cd, Pb, Cr and Ba in all vegetables were 7.86, 9.17, 12.1, 44.8, and 410 μg/kg for organic produce, and 7.29, 15.3, 17.9, 46.3 and 423 μg/kg for conventional produce (Table 1). Based on their characteristics, the five vegetables can be grouped into leafy (lettuce), fruit (tomato), and root (onion, carrot, and potato) vegetables. When considering all five metals, root vegetables had the highest metal contents, followed by leafy and fruit vegetables. Generally, the order followed tomato < lettuce < onion < carrot < potato (Table 1).

For example, the Cd content in tomato (0.93–4.30 μg/kg), lettuce (4.87–14.5 μg/kg), onion (8.00–19.8 μg/kg), carrot (5.20–13.8 μg/kg), and potato (14.0–33.5 μg/kg) generally followed this trend (Table 1). This is because more metals tend to accumulate in leafy and root vegetables than storage organs or fruits (Douay et al., 2013). As such, leafy and root vegetables accumulate higher levels of metals, showing higher health risk than fruit vegetables (Hu et al., 2013). However, our data were different from Shaheen et al. (2016) who determined As, Cd, Pb, and Cr concentrations in different vegetables, including bean (Phaseolus vulgaris), green chili pepper (Capsicum annuum), carrot, onion, potato and tomato, from 30 sites in Bangladesh. They found little differences in metal concentrations among vegetables except higher Cd was observed in tomato, i.e., 56 vs. 0.62–6.80 μg/kg, which was much greater than those in this study (Table 1). In addition, the As concentrations in potato were 8.00–8.66 μg/kg in this study, which were much lower than that reported by Munoz et al. (2002) who found 85 μg/kg As in potato from Chile. However, high As levels were found in potato from northern Chile at 241–860 μg/kg, which were attributed to both volcanic activity and irrigation water (Queirolo et al., 2000), Radwan and Salama (2006) found low concentrations of Cd and Pb in vegetables from markets in Egypt (μg/kg), including lettuce (3.43 and 28.4), tomato (0.69 and 16.9), carrot (1.20 and 21.6), onion (2.41 and 16.8), and potato (4.38 and 2.19), all below MAC proposed by FAO. However, Ali and Al-Qahtani (2012) found high concentrations of Cd and Pb (μg/kg) in vegetables including tomato (76.7 and 226), carrot (145 and 196), onion (521 and 111), and potato (251 and 659) from Saudi Arabian markets, with all vegetables exceeding the MAC for Cd (50–100 μg/kg) and Pb (100–200 μg/kg). Different metal levels in vegetables from different countries were probably due to contamination from irrigation water and being close to the highways. Furthermore, agronomic practices such as the application of fertilizer, pesticides and water management can also affect metal accumulation.

Based on mean values, metal contents varied greatly with vegetables. While tomato had the lowest As and Ba (1.60 μg/kg As and 46.6 μg/kg Ba), carrot had the highest As and Ba (13.3 μg/kg As and 1.18 mg/kg Ba; Table 1). While potato had the highest Cd (33.5 μg/kg), Pb (32.6 μg/kg) and Cr (80.0 μg/kg), tomato (0.93 μg/kg Cd), onion (3.00 μg/kg Pb), and lettuce (12.7 μg/kg Cr) had the lowest Cd, Pb, and Cr. So, the highest metal content in the vegetables was 6–36 fold greater than the lowest, with the lowest variation being Cr and the largest variation being Cd. Among five toxic metals, Ba had the highest concentrations, consistent with literature. McBride et al. (2014) conducted a survey on vegetables in three supermarkets in New York. They found Ba concentrations of 2.7–33 mg/kg in leafy vegetables and 9.4–32 mg/kg in root vegetables. Compared to 0.047–1.18 mg/kg in vegetables in our study, their levels were much greater.

3.2. Comparing metal contents for a given vegetable

Since the MACs in vegetables vary with metals (NHFPC, 2012), it is difficult to compare the values of different metals for a given
Table 1
As, Cd, Pb, Cr, and Ba content (μg/kg fw) in organic and conventional vegetables from four supermarkets in Florida.

<table>
<thead>
<tr>
<th>Vegetable</th>
<th>As (μg/kg fw)</th>
<th>Cd (μg/kg fw)</th>
<th>Pb (μg/kg fw)</th>
<th>Cr (μg/kg fw)</th>
<th>Ba (μg/kg fw)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean ± SD</td>
<td>Mean ± SD</td>
<td>Mean ± SD</td>
<td>Mean ± SD</td>
<td>Mean ± SD</td>
</tr>
<tr>
<td>Lettuce</td>
<td>0.10 ± 0.05</td>
<td>0.20 ± 0.05</td>
<td>0.30 ± 0.05</td>
<td>0.40 ± 0.05</td>
<td>0.50 ± 0.05</td>
</tr>
<tr>
<td>Potato</td>
<td>1.00 ± 0.10</td>
<td>2.00 ± 0.20</td>
<td>3.00 ± 0.30</td>
<td>4.00 ± 0.40</td>
<td>5.00 ± 0.50</td>
</tr>
<tr>
<td>Onion</td>
<td>2.00 ± 0.20</td>
<td>3.00 ± 0.30</td>
<td>4.00 ± 0.40</td>
<td>5.00 ± 0.50</td>
<td>6.00 ± 0.60</td>
</tr>
<tr>
<td>Carrot</td>
<td>3.00 ± 0.40</td>
<td>4.00 ± 0.50</td>
<td>5.00 ± 0.60</td>
<td>6.00 ± 0.70</td>
<td>7.00 ± 0.80</td>
</tr>
</tbody>
</table>

The results represent mean of three values ± standard deviation; MAC – maximum allowable concentration.
Table 2
Co, Cu, Ni, and Zn content (μg/kg fw) in organic and conventional vegetables from four supermarkets in Florida.

<table>
<thead>
<tr>
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<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Store 1</td>
<td>6.50 ± 0.49</td>
<td>0.50 ± 0.50</td>
<td>76 ± 2.00</td>
<td>12.5 ± 0.50</td>
<td>379 ± 2.00</td>
<td>127 ± 1.49</td>
<td>1840 ± 51.5</td>
<td>1810 ± 3.50</td>
</tr>
<tr>
<td>Store 2</td>
<td>3.00 ± 0.49</td>
<td>5.50 ± 0.49</td>
<td>56.5 ± 13.5</td>
<td>55.5 ± 2.50</td>
<td>444 ± 7.65</td>
<td>259 ± 7.51</td>
<td>1129 ± 52.0</td>
<td>1390 ± 27.0</td>
</tr>
<tr>
<td>Store 3</td>
<td>2.50 ± 0.50</td>
<td>1.00 ± 0.50</td>
<td>30.0 ± 0.40</td>
<td>12.5 ± 3.00</td>
<td>216 ± 2.10</td>
<td>219 ± 6.00</td>
<td>1290 ± 162</td>
<td>1925 ± 37.0</td>
</tr>
<tr>
<td>Store 4</td>
<td>3.00 ± 0.49</td>
<td>3.00 ± 3.33</td>
<td>32.5 ± 2.50</td>
<td>505 ± 30.0</td>
<td>330 ± 33.5</td>
<td>176 ± 7.9</td>
<td>1845 ± 161</td>
<td>1775 ± 329</td>
</tr>
<tr>
<td>Mean</td>
<td>3.75 ± 1.06</td>
<td>2.50 ± 1.44</td>
<td>46.0 ± 12.1</td>
<td>32.7 ± 13.5</td>
<td>342 ± 55.5</td>
<td>195 ± 32.7</td>
<td>1525 ± 215</td>
<td>1725 ± 134</td>
</tr>
</tbody>
</table>

Tomato

| Store 1 | 1.20 ± 0.60 | 1.20 ± 0.60 | 192 ± 1.80  | 47.3 ± 43.1  | 405 ± 13.1 | 462 ± 5.98    | 2611 ± 15.5 | 1672 ± 90.4   |
| Store 2 | 1.20 ± 0.18 | 1.80 ± 0.18 | 988 ± 67.0  | 246 ± 11.4   | 1204 ± 14.0 | 1952 ± 99.9   | 1311 ± 478 |               |
| Store 3 | 2.99 ± 0.99  | 49.1 ± 22.1 | 418 ± 50.2  | 344 ± 10.9   | 2470 ± 345 | 2130 ± 130    | 2654 ± 452 |               |
| Mean    | 2.10 ± 1.20  | 1.94 ± 0.43 | 14.7 ± 3.56 | 54.8 ± 18.1  | 325 ± 112  | 642 ± 309     | 2281 ± 465 | 1633 ± 194    |

Onion

| Store 1 | 3.00 ± 0.99 | 3.00 ± 0.99 | 40.0 ± 24.0 | 40.0 ± 27.0  | 344 ± 10.9 | 2470 ± 345    | 2130 ± 130 | 2654 ± 452    |
| Store 2 | 2.00 ± 1.00 | 2.00 ± 1.00 | 29.0 ± 10.0 | 27.0 ± 7.0   | 379 ± 49.6 | 187 ± 15.0    | 3310 ± 299 | 2642 ± 352    |
| Store 3 | 1.80 ± 1.19 | 1.80 ± 1.19 | 56.0 ± 48.0 | 40.0 ± 20.0  | 548 ± 59.7 | 498 ± 13.9    | 2140 ± 148 | 2400 ± 136    |
| Store 4 | 6.67 ± 0.82 | 11.0 ± 1.06 | 70.0 ± 15.9 | 45.0 ± 44.7  | 1320 ± 816 | 542 ± 66.6    | 2777 ± 528 | 2596 ± 325    |

Potato

| Store 1 | 4.80 ± 1.59 | 1.60 ± 1.60 | 94.4 ± 6.40 | 94.8 ± 24.0  | 475 ± 11.1 | 594 ± 28.5    | 2659 ± 83.2 | 3724 ± 216    |
| Store 2 | 3.20 ± 1.60 | 3.84 ± 4.80 | 275 ± 13.7  | 427 ± 3.19   | 721 ± 62.4 |               |             |               |
| Store 3 | 4.80 ± 1.59 | 4.80 ± 1.59 | 38.4 ± 6.40 | 137 ± 11.1   | 432 ± 9.50 | 762 ± 96.0    | 3880 ± 489 | 3950 ± 457    |
| Store 4 | 1.60 ± 1.60 | 4.80 ± 1.59 | 217 ± 31.9  | 104 ± 1.60   | 1557 ± 14.4 | 587 ± 12.7    | 3040 ± 662 | 3234 ± 244    |
| Mean    | 3.60 ± 0.88 | 3.73 ± 1.06 | 97.0 ± 48.6 | 108 ± 18.6   | 722 ± 321  | 647 ± 70.0    | 3072 ± 347 | 3639 ± 255    |

Total Mean 3.86 5.94 58.5 68.2 632 577 2528 2354

The results represent mean of three values ± standard deviation; MAC = maximum allowable concentration.

Kohrman and Chamberlain (2014) did a survey in three grocery stores in San Francisco. They recorded low levels of Cd (3 μg/kg) and Pb (8 μg/kg) in squash, consistent with our data (Table 1). In the present study, vegetables samples were peeled, which may be the reason for the low metal accumulation. The data suggested that peeling and thorough washing are important to reduce metal contamination in vegetables.

3.3. Essential nutrient contents in vegetables

Trace metals including Co, Cu, Ni, and Zn are nutrients required for biologic functions, however, in excess, they cause cellular and tissue damage. Their contents in all vegetables were low (Table 2). Among the four nutrients, Cu and Zn were more abundant in vegetables.

Vegetable species differ widely in their ability to accumulate nutrients, even varying among cultivars in the same species (Zhou et al., 2016). Among 5 vegetables, potato had the highest Co and Cu concentrations, with carrot having the highest Ni and Zn, and Co levels being the lowest among all nutrients. Bakkar et al. (2012) reported higher Ca concentrations in onion, carrot and spinach from Spain (4.5–6.8, 10.1–16.9, and 5.5–13.8 μg/kg) and Morocco (14.5–17, 15.1–25 and 11–14.7 μg/kg) than those reported in this study (18.0–110 μg/kg; Table 2). The highest Cu content was in potato and onion (10.0 and 11.0 μg/kg), lower than Osali et al. (2016) who found high Co concentrations in carrot and potato at 294 and 401 μg/kg in Jordan. The amounts of Cu in vegetables were 195–1320 μg/kg (Table 2). Bigdelli and Seilsepour (2008) reported much lower Cu levels in spinach, green pepper, and tomato at

vegetable. For better comparison, we used the ratios of mean metal concentration to its MAC, which is termed MM value. For example, the mean As concentration in tomato was 1.60 μg/kg and its MAC is 500 μg/kg, so its MM value was 0.32% (Table 1). The larger the MM value, the closer it is to its MAC for a given metal.

Among all metals, the highest MM value at 34% was observed in potato for Cd and the lowest at 0.32% in tomato for As (Table 1). For Cd and Pb, similar to potato (34 and 16%) and lettuce (7.3 and 8.4%), high MM values were also observed for Cd and Pb. Though metals in all vegetables were below MACs, based on the MM values, Cd and Pb concentrations were closer to the MAC than As, Cr and Ba in the vegetables. Though relatively speaking, Cd and Pb were of more concern, their levels were lower than those reported in the literature. For example, the Cd concentrations in vegetables at 0.93–33.5 μg/kg (Table 1) were lower than those reported by Nawab et al. (2017) who found Cd concentrations exceeded its safe limit of 100 μg/kg in 46% of vegetables in Pakistan. Martorell et al. (2011) found Cd concentrations in potato at 53 μg/kg in Spain. The Pb contents in vegetables at 3.00–32.6 μg/kg were lower than those from Australia (80.0–200 μg/kg; Angelova et al., 2010) and Turkey (102–154 μg/kg; Navare et al., 2009). Elbagermi et al. (2012) collected vegetables from markets of Libya, found higher Pb concentrations in tomato, onion, carrot and potato at 30.6, 19.1, 4.96, and 30.2 μg/kg compared to 7.7, 19.0 and 16.5 μg/kg in this study (Table 1). Mehari et al. (2015) conducted a survey in North Carolina and found that Cd, Ni, and Cr concentrations were undetectable in potato, squash (Cucurbita maxima), and eggplant (Solanum melongena), with Pb being detected in some vegetables.
Fig. 1. Comparison of trace metals content (µg/kg fw) in organic and conventional vegetables collected from four supermarkets in Florida. Data represent the mean of all samples from four supermarkets, with three samples from each supermarket. Metal concentrations marked with * are significantly different at α = 0.05 for a given vegetable.

Fig. 2. Comparison of nutrient content (µg/kg fw) in organic and conventional vegetables from four supermarkets in Florida. Data represent the mean of all samples from four supermarkets, with three samples from each supermarket. Metal concentrations marked with * are significantly different at α = 0.05 for a given vegetable.
vegetables (Table 1). These results are compatible with literature (Malmauaret et al., 2002; Ghidini et al., 2005; Karavolotsos et al., 2008), suggesting that organic agriculture may help to reduce metal content (Pussemier et al., 2006).

However, studies have also reported similar metal contents between organic and conventional vegetables (Hoeckens et al., 2009; Olaia et al., 2015). For example, Krejcová et al. (2016) reported no difference in As, Cd, Pb and Cr contents between organic and conventional carrot in Czech Republic. Arslanbaş and Baydan (2013) also did not find difference in Cd and Pb contents between the two in Turkey.

In this study, organic lettuce (342 and 195 μg/kg) and potato (1320 and 542 μg/kg) had higher Cu contents than conventional samples (Fig. 2c). However, the opposite was true for tomato and onion. The Cu content was higher in conventional tomato (642 and 325 μg/kg) and onion (877 and 416 μg/kg) than organic (Fig. 2c). When considering all four mineral nutrients, their contents in organic vegetables were similar to conventional ones, which was different from Kelly and Bateman (2010) who reported lower mineral contents in conventional vegetables. Similarly, higher mineral contents in organic vegetables were also observed by Olaia et al. (2015) and Siderer et al. (2005).

To better compare metal contents in organic and conventional produce, we also conducted principal component analysis (PCA). According to PCA, the first two components explained 57% of total variance. The vector diagram showed the direction of separation of two types of vegetables based on the contents of different metals (Fig. 3a). The vegetables having higher metal contents were on the right side while those having lower contents were on the left side. In general, there seemed an even distribution between conventional and organic vegetables across metals (Fig. 3a), indicating limited difference in their composition. While component 1 explained 32% of total variance and was positively correlated with Ni, Cd, and Co, component 2 explained 25% of total variance and was positively correlated with Cu, Cr, Pb, and Zn (Fig. 3b). The data indicated that Cu, Cr, Pb, and Zn may come from similar sources whereas Ni, Cd, and Co may come from different sources.

3.5. Chronic daily intake and hazard quotient

The chronic daily intakes (CDI) from vegetables were calculated for children and adults based on Eq. (1). The CDI values were compared with the oral reference dose (RfD) to assess their potential health risks. Slightly higher contribution to metal intake came from conventional vegetables. However, the daily intake was lower than the tolerable daily intake for all metals (JECFA, 2016). The chronic daily intakes of As, Cd, Pb, and Cr through vegetable consumption were <0.48 μg/kg bw/d (Table 3), much lower than the recommended oral reference doses (USEPA, 1989). The chronic daily intake of Cd in this study was lower than that in literature at 1.8–52 μg/kg bw/d (Santos et al., 2004; Tripathi et al., 1997).

The hazard quotient (HQ) was developed by USEPA (1989) as a quantitative way to evaluate potential health risks associated with long-term exposure to pollutants in foodstuffs, which is the ratio of measured dose to RfD. When HQ < 1.0, the risk of metals from vegetables is lower than RfD, presenting little health risk (Ji et al., 2016). In this study, the HQ for all metals at 0.01–0.3 was <1, showing little health risk from vegetable consumption (Table 3).

Though metal exposure through vegetable consumption was limited, it is important to consider exposures through other sources. However, high HQs of Pb were reported for vegetables in China (Pan et al., 2016) and Uganda (Nabulo et al., 2010).

Metal concentrations in all vegetables were low and within the maximum allowable limits established by FAO/WHO (JECFA, 2016). Metal concentrations in conventional produce were slightly higher...
than those in organic produce, especially for Cd and Pb. Metal concentrations also varied with vegetables, with root vegetables having greater metal contents than leafy and fruit vegetables. On a relative basis, among five toxic metals, Cd and Pb were of more concern as their levels were closer to MAC based on MM values. According to this study, based on both chronic daily intake and hazard quotient, the risk of metal exposure through consumption of the 5 most-consumed vegetables was probably of limited concern.

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