Heavy Metals Accumulation in the Edible Parts of some Sewage Wastewater Irrigated Vegetable Crops

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Abstract
Nowadays, using sewage wastewater in the irrigated agriculture causes hazardous environment impacts. Assessment of these negative effects is vital issue to prevent heavy metals to be introduced in the food chain. Field and laboratory studies were conducted at Ellwan, Mangabad and El-Madabegh villages, Assiut Governorate, in order to evaluate heavy metals concentrations in the edible parts of carrot, turnip and onion plants which were irrigated with sewage water (SW). The obtained results indicated that, the soils of the studied villages were contaminated by heavy metals. The concentrations of Zn, Cu, Pb, Cd and Ni concentrations in the edible portion of these vegetables plants ranged between 45-70, 13-19, 3.8-6.2, 2.85-3.85 and 2-6 mg kg⁻¹, respectively. The obtained results showed that the concentrations of Pb, Cd and Ni in the edible plants were higher than the permissible limit values but those of Zn and Cu were within the safe limit levels. It is worthy to mention that the irrigated edible vegetable crops especially onion, turnip and carrot with SW should be avoided. This study highlights the potential hazard for human health due to uptake of high concentrations of heavy metals especially Ni, Cd and Pb by the studied vegetable crops.

Keywords: Contaminated soils, Safe limits, Human consumption, Heavy metals, Carrot, Turnip, Onion.

Introduction
Water is important for all human activities. Water accounted for about 50-97% of plant and animal bodies; moreover, it is a vital for all biological processes in plant and animal cells (Buchholz, 1993). Fresh water recourses in the world are very limited and only 0.6% of the total world water resources is fresh water (FAO, 2015). Fresh water resources have been decreased in an alarming rate and they may not able to meet the requirements of the different human in the future (Qadir et al., 2007). The agriculture sectored uses about 80% of the water resources in irrigation purposes. Some of agriculture lands located near urban areas irrigated by wastewater because of the low availability of fresh water for crop production (Qadir et al., 2007).

The increasing of population and human activities enlarged the volume of sewage wastewaters (SW) (Qadir et al., 2007; Ismail et al., 2014). In many developing countries, these water resources may, in most cases, use in the form of diluted raw sewage, even if it is considered illegally (Qadir et al., 2007). Wastewaters quality differs both between and within countries. In many poor countries in Africa, Asia and Latin America, the untreated wastewaters are used widely in agriculture production, while in middle-income countries treated wastewater is used (Qadir et al., 2007; Ismail et al., 2014).
The use of sewage wastewater in irrigation provides the soil with nutrients and organic matter; moreover, it is an inexpensive system for wastewater disposal (Ullah et al., 2011; Gosh et al., 2012). In many situations, Egyptian farmers use wastewater in irrigation even when the fresh one is available, due to the high profits earned by using waste water. Sewage wastewaters (SW) are usually rich in nitrogen (N), phosphorus (P) and potassium (K) and farmers use these waters at a low price fertilizers (Chhabra, 1989). Nutrient concentrations in sewage waste waters are varied widely and Chhabra (1989) in India found that the SW contained 48.3, 7.6, 72.4 and 34.6 mg L\(^{-1}\) of N, P, K and S, respectively besides micro-nutrients contents of 0.34, 10.8, 0.2 and 0.36 mgL\(^{-1}\) for Zn, Fe, Cu and Mn, respectively. Therefore, ten SW irrigations of 7.5 cm each could add about 362, 58, 540 and 260 kg ha\(^{-1}\) of N, P, K and S, respectively, to the soil which are more than the nutrient requirements of most crops (Eissa, 2016). These findings recapitulate that wastewater has great potential as a manure when it is used to irrigate crops (Khurana and Singh, 2012).

Wastewaters contain high levels of Cd, Pb, Cr and As which are not essential for plant and animal nutrition (Kanwar and Sandha, 2000). The use of sewage wastewater in the irrigation processes may cause remarkable increases in soil heavy metal concentrations (Khan et al., 2008; Ullah et al., 2011; Gosh et al., 2012). The raising of soil heavy metal content will lead to introduce the metals to the vegetables and cereals crops causing a potential health risk to human and animal (Sharma et al., 2006; Singh et al., 2010; Gupta et al., 2011). The concentrations of heavy metals in plants cultivated on wastewater-irrigated soils are significantly higher than in those grown on fresh water-irrigated ones (Khan et al., 2008; Singh et al., 2010; Gupta et al., 2011).

The use of sewage wastewater to irrigate plants is an old action in many areas in Egypt due to the complexity of its treatment and disposal as well as the scarcity of fresh irrigation water. Its use is obligatory in order to provide foods to the ever-increasing population. It may cause soils and plants to be contaminated with heavy metals. The present study was undertaken to assess heavy metal contents of carrot, turnip and onion plants irrigated with sewage wastewater.

Materials and Methods

Site Description and Sampling

Composite plant samples of carrot, turnip and onion crops irrigated with sewage waste water were collected from Ellwan, Mankabad and Arab Elmadabegh villages, Assiut governorate, Egypt which are located at 27° 12’ 16.67” N latitude and 31° 09’ 36.86” E longitude to evaluate levels of some heavy metals (Zn, Cu, Pb, Ni and Cd) in these vegetables plants. The soils in these villages have been irrigated by raw sewage water for more than 50 years. Table 1 shows the main soil properties of the studied site. Each composite sample included the edible portion of ten plants of each crop. The plant samples were washed twice by tap water, rinsed by distilled water, air-dried,
oven-dried at 70°C to a constant weight, grounded and then were kept for chemical analysis. Soil (0-20 cm) and sewage wastewater samples were also taken from each study site.

Table 1. Some physical and chemical characteristics of the soils in the studied sites

<table>
<thead>
<tr>
<th>Soil property</th>
<th>Ellwan</th>
<th>Mankhabad</th>
<th>Arab-Elmadabegh</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay (g/kg)</td>
<td>100</td>
<td>120</td>
<td>110</td>
</tr>
<tr>
<td>Silt (g/kg)</td>
<td>200</td>
<td>180</td>
<td>190</td>
</tr>
<tr>
<td>Sand (g/kg)</td>
<td>700</td>
<td>700</td>
<td>700</td>
</tr>
<tr>
<td>Texture</td>
<td>Sandy loam</td>
<td>Sandy loam</td>
<td>Sandy loam</td>
</tr>
<tr>
<td>CaCO₃ (g/kg)</td>
<td>62</td>
<td>75</td>
<td>58</td>
</tr>
<tr>
<td>pH (1:2)</td>
<td>7.43</td>
<td>7.44</td>
<td>7.45</td>
</tr>
<tr>
<td>CEC (cmol/kg)</td>
<td>18</td>
<td>22</td>
<td>17</td>
</tr>
<tr>
<td>Total organic C (g/kg)</td>
<td>5.2</td>
<td>5.1</td>
<td>5.8</td>
</tr>
<tr>
<td>EC (1:2) (dS/m)</td>
<td>1.5</td>
<td>1.6</td>
<td>1.5</td>
</tr>
</tbody>
</table>

Soil, water and plant analysis
The physical and chemical properties of the studied soil samples were determined according to Burt (2004) as they are shown in Table 1. The available heavy metals (Zn, Cu, Pb, Cd and Ni) were extracted from the soil samples using a 0.005 M DTPA (diethylen triamine penta acetic acid) solution buffered at pH 7.3 as described by Lindsay and Norvell (1978). To determine the total heavy metals, the soil samples were digested according to the procedure given by the US EPA (1996). A known volume of each sewage wastewater sample was oven-dried and then was digested using concentrated HNO₃ at 80 °C (Table 2). The soil samples were air-dried and sieved with a 2-mm diameter sieve and kept for analysis. The metals in the soil, water and plant digest as well as DTPA soil extract were measured using the Inductivity Coupled Plasma Emission Optical Emission Spectrometry (ICP–OES thermo iCAP 6000 series). The ground plant samples were digested using concentrated acids of HNO₃ and HClO₄.

2.5. Statistical analysis
One-way ANOVA was used to test the significance of different between the studied plants and Duncan test was used to compare between means. The collected data were statically analyzed using SPSS statistical software.

Table 2. Chemical analysis of the irrigation sewage wastewater in the studied sites

<table>
<thead>
<tr>
<th>Site</th>
<th>pH</th>
<th>EC (dS m⁻¹)</th>
<th>Zn (mgkg⁻¹)</th>
<th>Cu (mgkg⁻¹)</th>
<th>Pb (mgkg⁻¹)</th>
<th>Cd (mgkg⁻¹)</th>
<th>Ni (mgkg⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ellwan</td>
<td>7.25</td>
<td>3.2</td>
<td>0.42</td>
<td>0.25</td>
<td>1.1</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Manghabad</td>
<td>7.56</td>
<td>3.5</td>
<td>0.34</td>
<td>0.35</td>
<td>1.2</td>
<td>0.06</td>
<td>0.01</td>
</tr>
<tr>
<td>Arab-Elmadabegh</td>
<td>7.11</td>
<td>3.7</td>
<td>0.52</td>
<td>0.38</td>
<td>1.0</td>
<td>0.08</td>
<td>0.03</td>
</tr>
<tr>
<td>PL*</td>
<td></td>
<td></td>
<td>0.20</td>
<td>0.20</td>
<td>5.0</td>
<td>0.01</td>
<td>0.20</td>
</tr>
</tbody>
</table>

*Permissible limits according to FAO (1985). Each value represents the mean of three replicates.
Results and Discussion
Soil and Sewage Wastewater Heavy Metals

Concentrations of Zn, Cu, Pb, Cd and Ni in the investigated wastewater samples differed from 0.34 to 0.52, 0.25 to 0.38, 1.0 to 1.2, 0.05 to 0.8 and 0.01 to 0.05 mg L, respectively (Table 2). The levels of Zn, Cu and Cd were higher than the permissible limits of the irrigation water according to the FAO (1985). On the other hand, Pb and Ni concentrations in these wastewater samples were lower than the FAO (1985) allowable limits. The highest Zn, Cu and Cd concentrations were recorded for Arab-Elmadabegh, where the sewage wastewater collection and treatment station of Assiut city is located.

The total concentrations of Zn, Cu, Pb, Cd and Ni in the studied soil samples varied from 620 to 640, 302 to 310, 300 to 305, 4.8 to 6.5 and 120 to 150 mgkg⁻¹, respectively (Table 3). The total concentrations of the investigated metals (Zn, Cu, Pb, Ni and Cd) were above the maximum permissible limits recorded by EU (2002) and USEPA (1997). These obtained values confirmed that the soils under study are contaminated with these heavy metals. Similar results were found by Eissa (2016). The long-term use of treated and untreated wastewater in irrigation was reported to cause significant buildup of the heavy metals in the soils (Khan et al., 2008; Ullah et al., 2011; Gosh et al., 2012; Uzma et al., 2016; Zia et al., 2017).

Table 3. Available and total heavy metals concentrations (mgkg⁻¹) in the soils of the studied sites

<table>
<thead>
<tr>
<th>Study site</th>
<th>Zn</th>
<th>Cu</th>
<th>Pb</th>
<th>Cd</th>
<th>Ni</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ellwan</td>
<td>5.0</td>
<td>5.2</td>
<td>5.1</td>
<td>0.30</td>
<td>1.0</td>
</tr>
<tr>
<td>Mankhabad</td>
<td>6.0</td>
<td>5.6</td>
<td>5.3</td>
<td>0.50</td>
<td>1.5</td>
</tr>
<tr>
<td>Arab-Elmadabegh</td>
<td>6.9</td>
<td>6.5</td>
<td>6.8</td>
<td>0.40</td>
<td>1.9</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Total metal</th>
<th>DTPA-extractable metal</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Zn</td>
</tr>
<tr>
<td>Ellwan</td>
<td>640</td>
</tr>
<tr>
<td>Mankhabad</td>
<td>620</td>
</tr>
<tr>
<td>Arab-Elmadabegh</td>
<td>620</td>
</tr>
<tr>
<td>PL</td>
<td>200-300</td>
</tr>
</tbody>
</table>

*Permissible limits according to European Union Standards (EU, 2002) and U.S. Environmental Protection Agency (USEPA, 1997).

Heavy Metals in the Edible Parts of Carrot, Turnip and Onion Plants
Zinc (Zn)

The concentrations of Zn in the edible parts of the studied plants ranged between 45 and 70 mgkg⁻¹ (Figure 1). The highest Zn concentration was recorded in the edible part of carrot collected from Arab-Elmadabegh. The soil of Arab-Elmadabegh contained the highest available level of soil Zn (6.9 mgkg⁻¹). This may explain the highest concentration of Zn in the plant grown in this soil. Concentrations of Zn in the edible portions of the studied plants were found to decrease in the order: carrot > turnip > onion. According to
EU (2006) and WHO/FAO (2007), the maximum permissible Zn limit for human consumption is between 60-80 mg kg\(^{-1}\) dry weights. Thus, the concentrations of Zn in the current study were less than the allowable level and these plants are safe for human consumption.

**Figure 1.** Zinc (Zn) concentrations (mg kg\(^{-1}\)) in the edible parts of carrot, turnip and onion plants

**Copper (Cu)**

The concentrations of Cu in the edible parts of carrot, turnip and onion plants varied from 13 and 19 mg kg\(^{-1}\). The highest value of Cu concentration was recorded for carrot plants grown on the soil of Arab-Elmadabegh which contained the highest value of the available soil Cu (6.5 mg kg\(^{-1}\)).

The edible portions of the studied plants exhibited Cu concentrations were decreased in the order of: carrot > turnip > onion.

The maximum permissible concentration of Cu in the edible parts for human consumption is 40 mg kg\(^{-1}\) dry weight (EU, 2006; WHO/FAO, 2007). Therefore, the edible parts of these plants are safe to be used by human being.

**Figure 2.** Copper (Cu) concentrations (mg kg\(^{-1}\)) in the edible parts of carrot, turnip and onion plants
Lead (Pb)

The Pb content of the edible parts of carrot, turnip and onion plants differed between 3.8 and 6.2 mg kg\(^{-1}\) (Figure 3). According to EU (2006) and WHO/FAO (2007) the maximum Pb permissible level for human consumption is 0.3 mg kg\(^{-1}\) dry weight. Thus, the concentrations of Pb in the edible plant parts were higher than this permissible level indicating that they are unsafe to be used. The results indicated that sewage wastewater use in irrigating these vegetables caused a significant increase in Pb content in their edible portions. These results are in an agreement with those of Rattan et al. (2005) and Ismail et al. (2014). Also, Eissa (2015) found that the Pb stored in plant roots was from 93 to 98% from the total Pb absorbed by these plant roots. Moreover, Fahr et al. (2013) reported that plant roots can absorb Pb 3-50 times more than the leaves. This may explain the high levels of Pb in the roots of turnip, onion and carrot.

Cadmium (Cd)

The edible parts of carrot, turnip and onion plants contained Cd levels varied between 2.85 and 3.85 mg kg\(^{-1}\) (Figure 4). EU (2006) and WHO/FAO (2007) indicated that the maximum permissible level of Cd for human consumption is 0.2 mg kg\(^{-1}\) dry weight. Thus, the concentrations of Cd in these edible parts were higher than that permissible limit which mean that are not safe to be consumed. These results coincide with those of Rattan et al. (2005) and Ismail et al. (2014). High concentrations of Cd in roots of carrot, turnip and onion plants may be related to its low translocation within plants (Voutsa et al., 1996). Similar results were reported by Uzma et al. (2016) and Zia et al. (2017).
Figure 4. Cadmium (Cd) concentrations (mg kg⁻¹) in the edible parts of carrot, turnip and onion plants

Nickel (Ni)

Nickel (Ni) levels in the edible parts of carrot, turnip and onion plants varied from 2 to 6 mg kg⁻¹. The highest level of Ni concentration was recorded in the edible part of carrot plants collected from Arab-Elmadabegh site. As it shown in Table 2, the soil of Arab-Elmadabegh contained the highest available soil Ni (1.9 mg kg⁻¹) it may explain the highest levels of Ni in the investigated plant grown on such soils, which is the point source of the sewage water.

The maximum permissible Ni level for human consumption is 1.5 mg kg⁻¹ dry weight (EU, 2006 and WHO/FAO, 2007). Thus, levels of Ni in the edible parts of these plants were higher than its permissible limit level. These results are in the same line with those of Rattan et al. (2005) and Ismail et al. (2014). The prolonged application of treated and untreated wastewaters results in significant buildup of heavy metals in the soils (Khan et al., 2008; Ullah et al., 2011; Gosh et al., 2012) and grown vegetables and cereals which are subsequently transfer to the food chain causing potential health risk to consumers (Sharma et al., 2006; Singh et al., 2010; Gupta et al., 2011). Heavy metals concentrations in plants grown on wastewater-irrigated soils were reported to be significantly higher than those grown on fresh water-irrigated soils (Khan et al., 2008; Singh et al., 2010; Gupta et al., 2011; Zia et al., 2017).
Figure 5. Nickel (Ni) concentrations (mg kg\(^{-1}\)) in the edible parts of carrot, turnip and onion plants

**Conclusion**

It might be concluded that heavy metals could accumulated in the edible vegetables that irrigated with untreated sewage wastewater. In this study, the obtained results showed that the concentrations of Pb, Cd and Ni in the edible parts of carrot, turnip and onion plants were higher than their permissible limit levels. Therefore, the edible parts of carrot, turnip and onion plants are not safe for human consumption. It is worthy to mention that irrigated edible vegetable crops irrigated with sewage wastewater should be avoided and Egyptian guidelines should be developed for the reuse of these waters in agriculture. Therefore it is recommended to never use sewage wastewater to irrigate vegetables unless it is obligated. Sewage wastewater might be used to irrigate other plants such as woody trees that can be used as a wind break as well as energy producer plants.

**References**


تراكم العناصر الثقيلة في الجزء المأكل لبعض محاصيل الخضر المروية بعوامل الصرف الصحي

ناديه محمد كمال رشدي
قسم الأراضي والمياه – كلية الزراعة – جامعة أسيوط

الملخص

اجريت بعض الدراسات الحقلية والعملية على بعض القرى بمحافظة اسيوط وهي: علوان، منقباد، والخانقانة، وذلك لتقييم تركيز العناصر الثقيلة في الجزء المأكل من بعض محاصيل الخضر وهي: الجزر، اللفنت، البصل حيث تم رؤى هذه المحاصيل بواسطة مياه الصرف الصحي.

أشهرت النتائج المحصلة عليها أن الأراضي تحت الدراسة كانت ملوثة بالعناصر الثقيلة، كما وجد أن تركيز كل من الزنك، النحاس، الرصاص، الكادميوم، النيكل في الجزء المأكل من محاصيل الخضر تتراوح بين 0.04-0.07، 0.16-0.19، 0.24-0.38، 0.24-0.38، 0.42-0.62 ملجم / كجم على الترتيب.

كما أوضحت النتائج المحصلة عليها أن تركيز كل من الرصاص والكادميوم والنيكل في الجزء المأكل من النباتات كان أعلى من الحدود المسموح بها أما تركيز الزنك والنحاس كان أقل من الحدود المسموح بها في النبات.

وبالتالي فإن هذه الدراسة تبين مدى خطورة استخدام مياه الصرف الصحي في رى محاصيل الخضر لما تحتويه على تركيزات عالية من العناصر الثقيلة التي تضر بصحة الإنسان وتصيبه بالمراجعات الخطيرة، إذا يجب تجنب الري بعوامل الصرف الصحي خصوصا إذا كانت غير معالجة للمحاصيل التي تدخل في السلسلة الغذائية للإنسان أو الحيوان.