HEAVY METALS IN SOIL AND VEGETABLES AND THEIR EFFECT ON HEALTH

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ABSTRACT

The present study was carried out to assess heavy metals Cadmium (Cd), Lead (Pb), copper (Cu), zinc (Zn), Chromium (Cr) and Nickel (Ni) levels in vegetables like Cauliflower (Brassica oleracea var. botrytis), Cabbage (Brassica oleracea), Carrot (Daucus carota), Brinjal (Solanum melongena), Spinach (Spinacia oleracea) and Radish (Raphanus sativus) irrigated with domestic wastewater. The vegetable samples were randomly collected from the farmlands irrigated with domestic wastewater around the Hisar district. Spinach, cabbage, brinjal and carrot accumulated higher Cd (1.30±0.31 mg kg\(^{-1}\)), Pb (4.23±0.32 mg kg\(^{-1}\)), Cu (1.42±0.25 mg kg\(^{-1}\)), Zn (3.4±0.28 mg kg\(^{-1}\)), Cr (1.16±0.11 mg kg\(^{-1}\)) and Ni (2.45±0.86 mg kg\(^{-1}\)) respectively. Transfer Factor (TF) of Cd, Pb, Cu, Zn, Cr and Ni are more in spinach (0.0306), cabbage (0.4448), spinach (0.2642), cauliflower (0.2494), carrot (0.0764) and spinach (0.7469) respectively. The health risk assessment has been calculated followed by Estimated Daily Intake Metal (EDIM) and Estimated Health Risk Index (EHRI). The present study highlights that both adults and children consuming vegetables grown in wastewater irrigated soils accumulate significant amount of these metals. However, the values of these metals were lesser than recommended maximum tolerable levels proposed by the FAO/WHO (1999).

Keywords: Daily Intake; Heavy Metals; Plant Uptake; Risk Assessment; Reference Dose; EDIM; EHRI.


1. INTRODUCTION

Indian economy is based on agriculture and having second largest population in the world. Most of its states are depends on the monsoon. There are two main sources for irrigation. First one is canal and second is ground water but the quality of ground water is so poor for long sustainability of agriculture system. This water is not adequate to fulfill the crop water requirement and needs additional extra water for agricultural purposes. To cater the need of the present demand for irrigation, use of municipal domestic wastewater, is becoming a common practice in urban areas of Haryana, India. This causes serious problems of salinity and ultimately reduction in agriculture production [1]. The large amounts of untreated industrial and domestic wastewater are used for year round irrigation of vegetables. Such waste water usually contains...
Heavy Metals which accumulate in the soil. The use of such untreated wastewater has been reported to cause contaminations of the food chain [2-3].

Some trace amount of heavy metals such as Zn and Cu are essential for the growth of organisms while others such as Cd and Pb are toxic [4-5]. Dietary uptake pathway could be through crops irrigated with contaminated wastewater and have been reported to contain large amount of toxic heavy metals which may lead to health disorders in humans depending on the uptake of these metals into plant and consumed by animals or humans [6-9]. Consumption of metal contaminated vegetables may lead to a weakened immune system, intra-uterine growth retardation, impaired psycho-social behavior, high prevalence of upper gastrointestinal cancer and other disorders typically associated with malnutrition [10].

Potential health risk to human being from consumption of vegetables may be due to heavy metal uptake from contaminated soils via plant roots as well as direct deposition of contaminants from the atmosphere onto plant surfaces [11]. A number of previous studies from developing countries have reported heavy metal contamination in wastewater and wastewater irrigated soil [5, 12-15]. Dietary intake is the main route of exposure of heavy metals for most people [16]. The information about heavy metal concentration in different types of vegetables and their dietary intake is very important for assessing their risk to human health. Heavy metals in the nutrient cycle have seriously threatened health and environmental integrity, therefore, problem of heavy metal contamination in vegetables requires detailed study so as to develop central strategies. The objective of present study was bioaccumulation of heavy metals in vegetables irrigated by domestic wastewater and assessment of health risk due to these heavy metals.

2. MATERIALS AND METHODS

2.1. Study Area and Sampling

All the samples of vegetables such as Cauliflower (Brassica oleracea var. botrytis), Cabbage (Brassica oleracea), Carrot (Daucus carota), Brinjal (Solanum melongena), Spinach (Spinacia oleracea) and Radish (Raphanus sativus); and soils were randomly collected from the farmlands irrigated with domestic wastewater around the three different locations such as Rishi Nager (L1), New Police Line (L2) and Ludass village (L3) of Hisar district of Haryana, India. Soil samples were collected at the surface depth of 10 cm using stainless steel spade sampling tools and plastic buckets to avoid any contamination of samples with traces of elements from the tools. At each sampling site, scrape away surface debris and remove a core sample to the appropriate depth. Soil samples were air dried, ground, passed through a 2 mm sieve and stored in plastic bags for further analysis. Five ground water and seven wastewater samples were also collected from each location. All samples were collected, stored and kept at 4°C for further analysis in polythene bags/canes according to their type and brought to the laboratory for metal quantification.

2.2. Sample Preparation

All the collected Vegetables were washed with double distilled water to remove airborne particles. The edible parts of the samples were weighed and soil samples were air-dried at room temperature, to reduce water content. All the samples were then oven-dried in a hot air oven at
70±5 °C for 24 h. Dried samples were powdered using a pestle and mortar and sieved through muslin cloth.

2.3. Digestion of the Vegetable and Soil Samples

For each vegetable, three powdered samples from source irrigated by domestic wastewater (1.0 g each) were accurately weighed and placed in crucibles, three replicates for each sample. The soil and vegetable ash samples were digested with per chloric acid and nitric acid (1:4) solution. The samples were cooled and contents were filtered through Whatman filter paper No. 40. Each sample solution was made up to a final volume of 50 ml with distilled water. Concentration of each heavy metal was analyzed by atomic absorption spectrophotometer (ASS: model AA6300, Shimadzu). Five samples of each vegetable were assayed and analysed individually in triplicate. Data were reported as mean ± SD. One way analysis of variance (ANOVA) by Scheffe test was used to determine significant difference between groups by using SPSS INS Version 16.0 statistical software.

2.4. Risk Assessment

2.4.1. Transfer Factor (TF)

The transfer factor (TF) of Cadmium (Cd), Lead (Pb), copper (Cu), zinc (Zn), Chromium (Cr) and Nickel (Ni) from the soil to vegetables was calculated using equation 1 [17-18]:

\[
\text{Transfer Factor (TF)} = \frac{\text{Metal concentration in vegetable plant at sampling field}}{\text{Metal concentration in soil at that field}}
\]  

2.4.2. Estimated Daily Intake of Metal (EDIM)

The Estimated daily oral intake of metals from soil through vegetables in mg was calculated by equation 2:

\[
\text{EDIM} = \frac{C_m \cdot C_F \cdot D_I}{B_{Aw}}
\]  

Where, \(C_m\) is heavy metal concentration in vegetable plants (mg/kg), \(C_F\) is conversion factor, \(D_I\) is daily intake of vegetables (kg/ person/day) and \(B_{Aw}\) is average body weight (kg). The conversion factor used to convert fresh green vegetable weight to dry weight was 0.085 [10]. The average daily vegetable intake for adults and children were considered to be 0.250 and 0.150 kg/ person/day, respectively, while average body weights were taken as 55 and 25 kg of the age of 35 and 16 years, respectively, for adult and child.

2.4.3. Estimated Health Risk Index (EHRI)

Estimated health risk index (EHRI) is the ratio of estimated daily intake of metal (EDIM) to the reference dose (RD) is defined as the maximum tolerable daily intake of a specific metal that does not result in any harmful health effects. If the value of EHRI is less than one, the exposed population is said to be safe and if greater than one indicating that there is a potential risk associated with that metal was calculated by equation 3 [19]:

3. RESULTS AND DISCUSSIONS

3.1. Metal Concentration Level in Water and Soil

The quality of domestic wastewater and ground water (tube well) was assessed for irrigation with respect to their pH, EC, OC and some of the heavy metals. The pH of the sewage water in the range of 6.8-7.3 was lower than the ground water as collected from the nearby area in the range of 7.2– 7.9) while its salt content (EC) was in the range of 175.8-195.3 mS/m considerably higher than those of ground water (148.9-158.6 mS/m). The concentration for heavy metal contents in domestic wastewater and in ground water samples shows that Cd, Pb, Cu, Zn, Cr and Ni are well within the permissible limit shown in figure 1. In general, concentration of heavy metals were on higher side in domestic wastewater than in the ground water (tube well) which could be toxic to some crops and human health. In the studied area, the concentration of all the heavy metals in water and domestic wastewater were found to be higher except Zn from the permissible limits of Indian standards and WHO/FAO [20-22]. The total concentrations of heavy metals (Cd, Pb, Cu, Zn, Cr and Ni) in soil sampled at three different sites are shown in figure 2. The average pH at the location-I (7.04±0.11), location-II (7.02±0.19) and location-III (7.0±0.03) are almost nearly neutral. The electrical conductivity (EC) was 190.2-273.6 mS/m for location-I, 202.8-247.2 mS/m for location –II and 170-271.3 mS/m for location-III. The percent organic carbon content in soil was higher due to constant domestic wastewater irrigation and range from 3.48-5.2% for location-I, 3.85-5.54% for location-II and 3-69-5.6% for location-III. As the sub soils were clayey, the organic carbon was found to be in high percent. Soil organic carbon is the most important indicator of soil quality and in addition to acting as a store-house of the plant nutrients plays a major role in nutrient cycling [23].

Figure 1: Heavy metal concentration in water and wastewater

Figure 2: Heavy metal concentrations in soil at different locations
Across the study area, wide ranges of heavy metal concentration in soil were observed and are shown in figure 3. The observed concentration of heavy metal range between 37.76-73.5 mg kg\(^{-1}\) for Cd, 2.74-22.44 mg kg\(^{-1}\) for Pb, 0.54-24.16 mg kg\(^{-1}\) for Cu, 0.96-12.44 mg kg\(^{-1}\) for Zn, 3.21-72.85 mg kg\(^{-1}\) for Cr, 1.37-6.47 mg kg\(^{-1}\) for Ni, and 249.3-773.4 mg kg\(^{-1}\) for Fe for location-I. For location-II, heavy metals range between 23.7-103.7 mg kg\(^{-1}\) for Cd, 5.67-30.99 mg kg\(^{-1}\) for Pb, 3.04-16.47 mg kg\(^{-1}\) for Cu, 3.16-16.88 mg kg\(^{-1}\) for Zn, 3.19-41.35 mg kg\(^{-1}\) for Cr and 4.68-13.77 mg kg\(^{-1}\) for Ni; 6.31-66.61 mg kg\(^{-1}\) for Cd, 3.20-19.60 mg kg\(^{-1}\) for Pb, 1.35-11.81 mg kg\(^{-1}\) for Cu, 4.47-15.55 mg kg\(^{-1}\) for Zn, 1.38-3.54-46.64 mg kg\(^{-1}\) for Cr and 1.38-9.99 mg kg\(^{-1}\) for Ni for location-III. The heavy metal (Cd, Pb, Cu and Cr) concentration was not significantly different in all the three locations and concentration of Zn in location-I is significantly different from location-III at \((P<0.05)\) and location-III is significantly different from location-II at \((P<0.05)\) level. For Ni, the concentration of Ni in location-I is significantly different from location-II at \((P<0.05)\) and location-III is is significantly different from location-I at \((P<0.05)\) level. The results clearly indicate higher concentration of metals in soils which had prolonged irrigation by sewage wastewater. If the same trend continues the concentration of metals will accumulate in the soil. Relatively higher amounts of these heavy metal which attributes to the reduction in soil pH to moderately acidic conditions as well increase in organic carbon due to the continuous use of sewage effluents. This may be a cause of prime concern in near future. Long-term application of domestic wastewater will result in increase in organic carbon and decrease in pH.

![Figure 3: Concentration of heavy metal in different vegetables](http://www.ijoest.com)

### 3.2. Heavy Metal Accumulation in Vegetables

The concentration of heavy metals in edible part of vegetables was investigated in vegetables which are commonly grown. The bioaccumulation of heavy metals concentration in all the vegetables are different, so no similar trend have been observed for heavy metal concentration. The heavy metal bioaccumulation in cauliflower was Zn > Pb > Cu > Ni > Cd > Cr, for cabbage was Pb > Zn > Cd > Cr > Cu > Ni, for carrot was Zn > Pb > Ni > Cu > Cr > Cd, for brinjal Zn > Pb > Cd > Ni > Cr > Cu, for spinach was Zn > Ni > Pb > Cd > Cu > Cr, a similar trend have also been observed for radish Zn > Ni > Pb > Cr > Cu > Cd [24].

The concentrations of heavy metals (Cd, Pb, Cu, Zn, Cr and Ni) in vegetable samples are quite variable and shown in figure 3. The spinach exhibited higher concentration levels of Cd (1.30 mg kg\(^{-1}\)) and Ni (2.45 mg kg\(^{-1}\)) than other vegetables. In contrast, cabbage (4.23 mg kg\(^{-1}\)) contained...
the highest levels of Pb, while carrot (1.42 mg kg\(^{-1}\)) contained the highest Cu concentration. Furthermore, the concentration of Cr was higher in radish (1.16 mg kg\(^{-1}\)), whereas cauliflower (4.26 mg kg\(^{-1}\)) revealed greater amount of Zn than other vegetables. The concentration of Cd (1.30 mg kg\(^{-1}\)) in spinach is significantly different from the cauliflower, radish at \((p<0.05)\) and \((p<0.001)\) level, respectively and for carrot \((p<0.05)\) level. The results of the present analysis showed that the concentration of Pb (4.23 mg kg\(^{-1}\)) in cabbage is significantly different from cauliflower, brinjal and radish, from the vicinity of three locations at \((p<0.05)\) and \((p<0.001)\) level. The amounts of Cu (1.42 mg kg\(^{-1}\)) in carrot is significantly different from cabbage and brinjal at \((p<0.05)\) level. The amount of Zn in all vegetables i.e., cauliflower, carrot, brinjal, cabbage, spinach, and radish were found to be not significantly different \((p>0.05)\) level. Concentration of Cr (1.16 mg kg\(^{-1}\)) in carrot is more significantly different from the cauliflower at \((p<0.05)\) and \((p<0.001)\) level and concentration of Cr in radish is significantly different from cauliflower at \((p<0.05)\) level. Concentration of Ni (1.42 mg kg\(^{-1}\)) in spinach is significantly different from the cauliflower and cabbage at \((p<0.05)\) level. The range of Cd concentration in brinjal (0.50 - 1.10 mg kg\(^{-1}\)) recorded in this study was lower than the range (1.10 - 9.20 mg kg\(^{-1}\)) [25]. Vegetables (spinach and cabbage), range of Ni concentration was highest in spinach (0.01 - 3.80 mg kg\(^{-1}\)). These values were lower than the range (5.55 - 15.00 mg kg\(^{-1}\)) in spinach from Dinapur area as well as the range (0.2 - 3.0 mg kg\(^{-1}\)) in spinach from waste water irrigated areas of Hyderabad [25-26]. In cabbage concentration of Pb, Cu, Zn, Cr, Ni were lower during the present study. The present concentration (mg kg\(^{-1}\)) of 0.30±0.03 for Cd, 0.94±0.10 for Pb, 0.83±0.18 for Cu and 0.94±0.09 for Cr in radish were lower than the values obtained for radish [27]. The higher concentration of Cd, Pb, Cu, Cr and Ni and in radish plants grown at waste water irrigated areas than the clean water irrigated ones [28]. Among all the heavy metals, Zn concentration (4.26±0.06) was higher and Cr (0.09±0.07) was lower in cauliflower. Radwan and Salama have also found highest concentration of Zn in vegetables collected from Egyptian markets [29]. The bioaccumulation of Cd in spinach is higher than all the tested vegetable samples and similar result have also observed by Zhuang and his coworker [30]. The bioaccumulation of Pb, Cu, Zn, Cr and Ni were found higher in cabbage, carrot, cauliflower, carrot and radish respectively.

### 3.3. Heavy Metal Transfer Factor

Due to industrialization and urbanization, the heavy metal concentration of soil has increased worldwide. Soil-to-plant transfer of heavy metal is major pathway of human exposure to soil contamination [17]. The high transfer factor from soil to plants indicates a strong accumulation of the particular metal by vegetable crops [28]. The results indicated that TF values were lower for Cd, Pb, Cu and Cr, and higher TF for Zn and Ni at all locations. The transfer factor values in soil to plant of studied metals such as Cd, Pb, Cu, Zn, Cr and Ni for various vegetables varied between vegetable plants and locations and are shown in table 2. The results revealed that there is a large variation in transfer factor of Cd, than other metals in all three locations. The spinach (0.0306) had very high transfer factor for Cd and followed by brinjal (0.0207), cabbage (0.0165), carrot (0.0108), radish (0.0071) and cauliflower (0.0049). Similarly, The cabbage (0.4448), had very high transfer factor for Pb and followed by carrot (0.2376), spinach (0.2187), brinjal (0.0207), cauliflower (0.1672) and radish (0.0988); The carrot (0.2977), had very high transfer factor for Cu and followed by spinach (0.2642), cauliflower (0.2537), radish (0.1740), cabbage (0.0881) and brinjal (0.0818); The carrot (0.0764) had very high transfer factor for Cr and
followed by radish (0.0619), spinach (0.0527), cabbage (0.0454), brinjal (0.0408) and cauliflower (0.0054) for location-III; The cauliflower (0.2494) had very high transfer factor for Zn and followed by carrot (0.1991), brinjal (0.1909), cabbage (0.1862), spinach (0.1739) and radish (0.0984) for location-II; The spinach (0.7469) had very high transfer factor for Ni and followed by carrot (0.4451), radish (0.4542), brinjal (0.2348), cauliflower (0.0732) and cabbage (0.0335) for location-I. The transfer factor of Cd, Pb, Cu and Cr is more in Location III, similarly, Zn in location II and Ni in location-I. Therefore, vegetable crops growing on polluted site can accumulate high concentration of trace elements to cause serious health risk to consumers.

3.4. Risk Assessment

In order to check the health risk of any toxicity from wastewater, it is necessary to estimate the level of exposure of heavy metals through food chain. In this study, six vegetables were selected and calculate the health risk assessment in terms of estimated daily intake of metal (EDIM) and estimated health risk index (EHRI) by considering the intake of metal through vegetables by the human being.

3.4.1. Estimated Daily Intake of Metal (EDIM)

The daily intake of heavy metals was estimated on basis of the average consumption and concentration of metal in particular vegetable spices. The average concentration of Cd, Pb, Cu, Zn, Cr, Ni and Fe are shown in figure 3. Based on the above, the estimated daily intake of metal (EDIM) for adults and children through food chain were calculated and is shown in Table 1. The highest daily intake of metal such as Pb, Zn, Ni, Cu, Cd and Cr were from spinach, cabbage, cauliflower, spinach, carrot, spinach and Carrot respectively grown in domestic wastewater for both adults and children. The results of the study revealed that EDIM suggest that the consumption of vegetables grown in domestic wastewater polluted location is high but is free from any risk as the dietary intake of Cd, Pb, Cu, Zn, Cr and Ni in adults is below than the permissible limits [31].

3.4.2. Estimated Health Risk Index (EHRI)

In order to investigate the estimated health risk index (EHRI) associated with domestic wastewater irrigated soil, it is essential to estimate the level of exposure by quantifying the route of exposure of a heavy metal to the target person. The results indicate that the EHRI (table 2) values were less than one for Cd, Pb, Cu, Zn, Cr and Ni in all tested vegetables consumption. Therefore, the health risk of heavy metals such Cd, Pb, Cu, Zn, Cr and Ni exposure through vegetables was of no consequence and generally assumed to be safe. All the estimated dietary intake of Cd, Pb, Cu, Zn, Cr and Ni were far below the tolerable limits. The oral reference dose (Rd) for Cd, Pb, Cu, Zn, Cr and Ni are 1.0E-03, 3.5E-03, 4.0E-02, 3.0E-01, 1.5E-00 and 2.0E-02 mg kg\(^{-1}\) day\(^{-1}\), respectively given by US-EPA [32]. The results of the study showed that EDIM and EHRI suggest that consumption of vegetables grown in polluted soil irrigated with domestic wastewater is nearly free of risk. But there are other sources of metal exposure such as dust inhalation, vehicular exhaust which were not included in this study.
Heavy Metals in Soil and Vegetables and their Effect on Health

Table 1: Transfer factor (TF) of heavy metals from contaminated soil to vegetable plant

<table>
<thead>
<tr>
<th>Vegetable</th>
<th>Location</th>
<th>Cd</th>
<th>Pb</th>
<th>Cu</th>
<th>Zn</th>
<th>Cr</th>
<th>Ni</th>
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<tbody>
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<td>Cauliflower</td>
<td>I</td>
<td>0.0039</td>
<td>0.1518</td>
<td>0.1929</td>
<td>0.2391</td>
<td>0.0025</td>
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<td></td>
<td>II</td>
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Table 2: Estimated Daily Intake (EDIM) and Estimated Health Risk Index (EHRI) of heavy metals in vegetables

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<td>1.0E-1</td>
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<td>EDIM</td>
<td>2.7E-4</td>
<td>3.6E-4</td>
<td>1.6E-3</td>
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4. CONCLUSIONS & RECOMMENDATIONS

The prolonged irrigation by domestic wastewater increases heavy metal accumulation in the soil which leads to contamination of food crops. This study reveals that edible vegetables show...
significant bioaccumulation of heavy metals in vegetables grown by sewage wastewater irrigation. These vegetables are supplied to local markets and there is possibility of associated health hazard with consumption of these contaminated vegetables over a long period of time. The concentration of all heavy metals in domestic wastewater was found to be higher except Zn from the permissible limits of national and international standards. The results indicated higher concentration of metals in soils contributed by prolonged irrigation with sewage wastewater. If it is continued, the concentration of metals will accumulate in the soil. The higher amount of these heavy metals which attributes to reduction in soil pH to moderately acidic conditions and as well as increase in organic carbon due to the continuous use of domestic wastewater. The transfer factor of Cd, Pb, Cu and Cr is more in Location III, of Zn in location II and Ni in location-I. Therefore, vegetable crops growing on polluted site can bio-accumulate high concentration of trace elements to cause serious health risk to consumers. The highest daily intake of metal such as Pb, Zn, Ni, Cu, Cd and Cr were from consumption of the spinach, cabbage, cauliflower, spinach, carrot, spinach and Carrot, respectively, grown in domestic wastewater for both adults and children. The results of study also revealed that EDIM and EHRI suggest that the consumption of vegetables grown with domestic wastewater in polluted location is high but is free from any risk. Therefore, the health risks from the heavy metals such Cd, Pb, Cu, Zn, Cr and Ni exposure through vegetables was of no consequence and it is generally assumed to be safe. Therefore, in order to reduce risk, plants with lower accumulative nature should be grown. In this scenario, the present study indicates that there is need of proper treatment and disposal of domestic wastewater with less cost and worldwide acceptable technology.

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Heavy Metals in Soil and Vegetables and their Effect on Health


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