ASSESSMENT OF HEAVY METAL CONCENTRATIONS IN PUMPKIN GROWN ALONG RIVER BENUE, MAKURDI LOCAL GOVERNMENT AREA OF BENUE STATE, NIGERIA.

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ABSTRACT

This study aimed at detecting and assessing the concentrations of heavy metals in Pumpkin irrigated using surface and groundwater sources grown along the banks of River Benue, Makurdi, Nigeria. Eight heavy metals were detected to include cadmium, chromium, cobalt, copper, iron, manganese, lead and zinc. The mean concentration of heavy metals in pumpkin irrigated with surface water obtained from Fiidi, Wurukum, New garage and Wadata areas of makurdi was determined using triplicate values obtained from heavy metal analysis. The results show the mean concentrations of heavy metals to include Cd (2.07 ± 0.1) , Cr (2.07 ± 0.1) , Co (0.803 ± 0.01) , Cu (3.14 ± 0.1) , Mn (2.51 ± 0.02) , Pb (1.05 ± 0.05) and Zn (9.03 \pm 0.03). Fiidi had Cd (1.02 \pm 0.03), Cr (2.24 \pm 0.04), Co (0.417 \pm 0.02), Cu (2.61 \pm 0.01), Fe (1.03 \pm 0.02), Pb (1.22 \pm 0.02) and Zn (6.81 \pm 0.001). Wurukum had mean concentration of Cd Cr, Co, Cu, Fe, Mn, Pb, and Zn to be 0.920 ± 0.03 , 1.61 ± 0.02 , 0.410 ± 0.02 , 2.42 ± 0.02 , 1.02 ± 0.02 , 1.02 ± 0.02 , 1.02 ± 0.02 , $0.02 \pm$ 0.03, 1.22 ± 0.02 , 1.41 ± 0.01 and 8.54 ± 0.04 for the heavy metals respectively. On the other hand, pumpkin irrigated with surface water around New garage had a mean concentration of 0.710 ± 0.03 , $2.13 \pm 0.03, 0.410 \pm 0.01, 2.90 \pm 0.01, 1.11 \pm 0.02, 2.03 \pm 0.06, 0.527 \pm 0.03, 3.11 \pm 0.01, 1.22 \pm 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01, 0.01,$ $0.07, 1.81 \pm 0.02, 1.22 \pm 0.03$ and 6.83 ± 0.06 for Cd, Cr, Co, Cu, Fe, Mn, Pb and Zinc respectively. The mean concentrations of heavy metals in pumpkin irrigated with groundwater were Manganese was highest in Zua (4.64 \pm 0.03) and cobalt (0.410 \pm 0.02) was lowest, Fiidi had Zinc (9.01 \pm 0.01) as the highest whereas cobalt had 0.517 ± 0.02 as the lowest mean concentration. The new garage had 7.85 ± 0.06 and 0.517 ± 0.02 whereas Wadata had 8.24 ± 0.2 and 0.4 ± 0.02 for zinc and cobalt as the highest and lowest mean concentration respectively. The presence of the heavy metals in pumpkin planted in the locations studied did not vary significantly since all the heavy metals detected were present in all the samples studied at P > 0.05.

Keywords: Heavy metals, pumpkin, irrigation.

INTRODUCTION.

Irrigation is the controlled application of water for agricultural purposes through man-made systems to supply water requirements not satisfied by rainfall. Crop irrigation is vital throughout the world in order to provide the world's ever-growing populations with enough food. An irrigation activity that is gradually gaining recognition is the one being practiced under the urban and peri-urban agriculture (UPA). The system involves the use of stream water to irrigate lands at the banks with the objective of producing fruits and vegetables for the consumption of city dwellers (Binns *et al.*, 2003). The heavy metals pollution of Rivers effects on the irrigation water quality along the river, as well as crops irrigated with it. The irrigated crops may take-up these heavy metals and consequently, introduce them into food chain, resulting to gradual accumulation in the humans, and thereafter, present health hazard.

Studies of Baes *et al.* (1984) and Reilly (1991) have shown that intake of trace metals from dietary sources may represent a significant exposure pathway for human populations. However, dietary exposure to trace metals is highly variable. For example, Lopez-Artiguez *et al.* (1993) has observed that for Cd, the principal exposure route for the general

population is through uptake by food plants. Where metal concentrations in crops exceed the limits, it may be possible to use this produce in animal feeds in order to minimize the effect upon the human diet. However, animals fed on a metal-enriched diet may have elevated concentrations of these metals in their tissues and milk. Reilly (1993) noted that regular consumption of metal-enriched animal products may lead to adverse health effects in humans. Furthermore, Beresford *et al.*(1999) have observed that the greatest degree of metal accumulation occurs in offal, such as livers and kidneys.

Heavy metals such as Cd, Zn, Pb, Cr, Fe and Cu are commonly found on contaminated sites (Oathman, 2001). Sources of heavy metal contamination to the environment have been identified by various researchers to include; exhaust pipes, wastewater effluents and oil exploration activities. The implication of gas explosive on human health is all related to the exposure of those hazardous air pollutant emitted during incomplete combustion, acid rain effect and subsequent acidification of the soil (Akhionbare,2011). Bioaccumulation of these heavy metals has been identified mainly in surface water, soil and in plants (Akoto et al.2008).

The supply of clean and uncontaminated water is a great challenge facing developing nations. In Nigeria, pollution is a major threat to both surface and underground water bodies (Oathman, 2001; Osabolien et al. 2013). Green vegetables such as pumpkin leaf form a substantial portion of the daily human diet of the southern part of Nigeria. These are grown on all types of available land such as banks of polluted rivers, alongside road and areas enclosing wastewater ponds. These plants absorb some of these chemicals which are toxic to human health (Baranowska et al. 2002) Industrial emissions of contaminant to the atmosphere which is finally deposited on soil may cause some pollution problem. The contaminant concentration in soil mainly depends on the adsorption properties of soil matter (Deka and Sarma 2012). The high toxic and persistent nature of heavy metals in the environment has made them priority pollutants. The present study quantified the metals; cadmium, heavy chromium. cobalt,copper, iron, lead manganese and zinc (Cd, Cr, Co, Cu, Fe, Pb, Mn, Zn) in pumpkin irrigated with ground and surface water from different locations in Makurdi the Benue State capital of Nigeria.

METHODOLOGY

Sample

Pretreatment of Samples for analysis of metals in Vegetable Samples.

The plant Samples will be thoroughly washed rinsed with tap water and then with double distilled water to remove any attached soil particles. It will then be cut into small portions and placed in a large crucible where they will be oven dried at 60 ^oC overnight. The dried plant will then be grounded into fine particles using a clean mortar and pestle.

The triacid method of digestion will be employed. The acids to be used will be Nitric acid (HNO₃), Perchloric acid (HCLO₄) and concentrated Sulphuric acid (H₂SO₄) in the ratio of 68:8:2 respectively.

0.2 g of the powdered crop sample will be weighed into a 100 ml glass beaker. 30 ml of the acid mix will be added and the content swirled and placed on a hot plate. The beaker will be heated until the brown fumes of the Nitric acid go off, and the heating continued until the content of the beaker is reduced to about 5 ml. The content will be allowed to cool and little amount of distilled water will be added and the beaker swirled again. The content will then be poured into a 50 ml volumetric flask and made up to the required mark. The digest obtained will be used for the determination of the metals (Salawu *et al.*, 2015).

Determination of Heavy Metals.

The measurement of heavy metal concentration was carried out with an Atomic Absorption Spectrophotometer (AAS). All concentrations were determined using the 3

absorbance made air-acetylene flame. Eight working solutions were prepared from the stock solution for each of the metals (Cr, Cu, Cd, Zn, Co, Fe, Pb, and Mn) by successive serial dilution and each of the standard solutions were aspirated into the flame of AAS and the absorbance recorded in each case. A plot of the concentration against the corresponding absorbance gave the calibration curve of each of the metals. The samples were aspirated into the flame and the absorbance obtained (Chiroma and Ebawele, 2003).

RESULTS

Figure 1shows the occurrence of heavy metals in pumpkin plants for irrigated with surface water obtained from Fiidi, Wurukum, New garage and Wadata areas of makurdi. The result show the mean concentrations of heavy metals to include (in mg/kg) Cd (2.07 \pm 0.1), Cr (2.07 \pm 0.1), Co (0.803 \pm 0.01), Cu (3.14 \pm 0.1), Mn (2.51 \pm 0.02), Pb (1.05 \pm 0.05) and Zn (9.03 \pm 0.03). Fiidi had Cd (1.02 \pm 0.03), Cr (2.24 \pm 0.04), Co (0.417 \pm 0.02), Cu (2.61 \pm 0.01), Fe (1.03 \pm 0.02), Pb (1.22 \pm 0.02) and Zn (6.81 \pm 0.001). wurukum had mean concentrations of Cd Cr, Co, Cu, Fe, Mn, Pb, and Zn to be 0.920 ± 0.03 , 1.61 ± 0.02 , 0.410 $\pm 0.02, 2.42 \pm 0.02, 1.02 \pm 0.03, 1.22 \pm 0.02,$ 1.41 ± 0.01 and 8.54 ± 0.04 for the heavy metals respectively. On the other hand,

pumpkin irrigated with surface water around New garage had a mean concentration of 0.710 \pm 0.03, 2.13 \pm 0.03, 0.410 \pm 0.01, 2.90 \pm 0.01, 1.11 \pm 0.02, 2.03 \pm 0.06, 0.527 \pm 0.03, 3.11 \pm 0.01, 1.22 \pm 0.07, 1.81 \pm 0.02, 1.22 \pm 0.03 and 6.83 \pm 0.06 for Cd, Cr, Co, Cu, Fe, Mn, Pb and Zinc respectively.

Figure 2 shows the mean concentration of heavy metals in pumpkin irrigated using ground water in the locations studied. The mean concentration of manganese was highest in Zua (4.64 ± 0.03) and cobalt (0.410 ± 0.02), Fiidi had Zinc (9.01 ± 0.01) and cobalt 0.517 ± 0.02 as the heavy metals with highest and convert mean concentration respectively. New garage had 7.85 ± 0.06 and 0.517 ± 0.02 whereas Wadata had 8.24 ± 0.2 and 0.4 ± 0.02 for zinc and cobalt respectively.

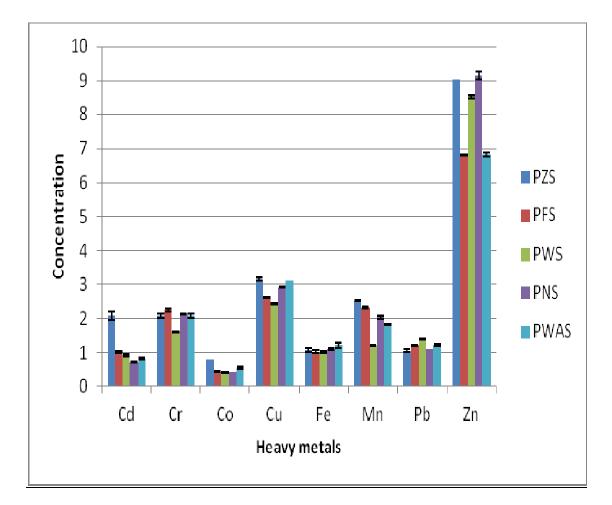


Figure 1: Heavy metal concentration in pumpkin irrigated with surface water

Key:

- PZS= Pumpkin crop-Zua-Surface water irrigated
- PFS= Pumpkin crop-Fiidi-Surface water irrigated
- PWS= Pumpkin crop-Wurukum-surface water irrigated
- PNS=Pumpkin crop-New Garage-Surface water irrigated
- PWAS= Pumpkin crop-Wadata-surface water irrigated.

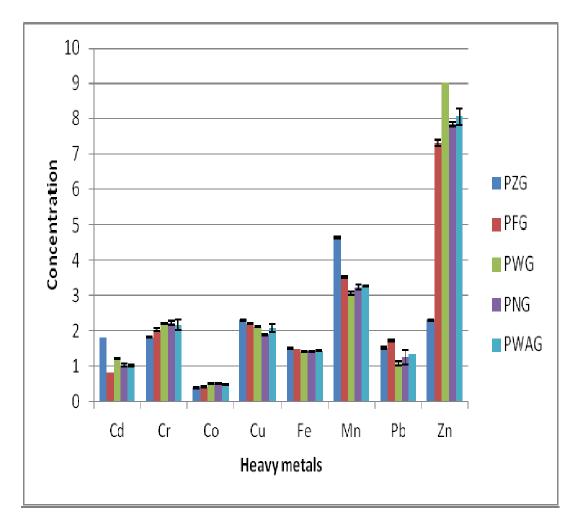


Figure 2: Heavy metal concentration in pumpkin irrigated with ground water

Table 1: Concentration of Heavy metals in Pumpkin irrigated with Grounnd water in Wadata (in mg/kg).

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|-------|-------|------|-------|------|------|------|-------|------|
| PWAG1 | 0.800 | 2.00 | 0.400 | 2.00 | 1.20 | 2.80 | 0.900 | 8.30 |
| PWAG2 | 0.800 | 2.20 | 0.400 | 2.00 | 1.23 | 2.82 | 0.910 | 8.43 |
| PWAG3 | 0.830 | 2.30 | 0.430 | 2.20 | 1.20 | 2.83 | 0.920 | 8.00 |
| Mean | 0.810 | 2.17 | 0.41 | 2.07 | 1.21 | 2.82 | 0.91 | 8.24 |
| SD | 0.02 | 0.2 | 0.02 | 0.1 | 0.02 | 0.02 | 0.01 | 0.2 |
| RSD | 2.1 | 7.1 | 4.2 | 5.6 | 1.4 | 0.5 | 1.1 | 2.7 |

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PNG1 | 1.00 | 2.20 | 0.500 | 1.90 | 1.40 | 3.20 | 1.30 | 7.80 |
| PNG2 | 1.00 | 2.20 | 0.520 | 1.90 | 1.43 | 3.20 | 1.04 | 7.91 |
| PNG3 | 1.08 | 2.30 | 0.530 | 1.86 | 1.45 | 3.32 | 1.46 | 7.83 |
| Mean | 1.03 | 2.23 | 0.517 | 1.89 | 1.43 | 3.24 | 1.27 | 7.85 |
| SD | 0.05 | 0.06 | 0.02 | 0.02 | 0.03 | 0.07 | 0.21 | 0.06 |
| RSD | 4.5 | 2.6 | 3.0 | 1.2 | 1.8 | 2.1 | 16.7 | 0.7 |

Table 2: Concentration of Heavy metals in Pumpkin irrigated with Grounnd water in New Garrage (mg / kg).

Table 3: Concentration of Heavy metals in Pumpkin irrigated with Grounnd water in Fiidi (mg/kg).

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|-------|------|-------|------|------|------|------|------|
| PFG1 | 0.800 | 2.00 | 0.400 | 2.20 | 1.50 | 3.50 | 1.70 | 7.20 |
| PFG2 | 0.820 | 2.00 | 0.430 | 2.20 | 1.50 | 3.56 | 1.72 | 7.40 |
| PFG3 | 0.810 | 2.08 | 0.440 | 2.23 | 1.50 | 3.54 | 1.73 | 7.32 |
| Mean | 0.810 | 2.03 | 0.423 | 2.21 | 1.50 | 3.53 | 1.72 | 7.31 |
| SD | 0.01 | 0.05 | 0.02 | 0.02 | 0.0 | 0.03 | 0.02 | 0.1 |
| RSD | 1.2 | 2.3 | 4.9 | 0.8 | 0.0 | 0.9 | 0.9 | 1.4 |

Table 4: Concentration of Heavy metals in Pumpkin irrigated with Grounnd water in Zua (in mg/kg).

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PZG1 | 1.80 | 1.80 | 0.400 | 2.30 | 1.50 | 4.60 | 1.50 | 2.30 |
| PZG2 | 1.82 | 1.80 | 0.400 | 2.32 | 1.53 | 4.65 | 1.53 | 2.31 |
| PZG3 | 1.80 | 1.83 | 0.430 | 2.30 | 1.54 | 4.66 | 1.55 | 2.33 |
| Mean | 1.81 | 1.81 | 0.410 | 2.31 | 1.52 | 4.64 | 1.53 | 2.31 |
| SD | 0.01 | 0.02 | 0.02 | 0.01 | 0.02 | 0.03 | 0.03 | 0.02 |
| RSD | 0.6 | 1.0 | 4.2 | 0.5 | 1.4 | 0.7 | 1.6 | 0.7 |

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PWG1 | 1.20 | 2.20 | 0.500 | 2.10 | 1.40 | 3.00 | 1.10 | 9.00 |
| PWG2 | 1.23 | 2.24 | 0.520 | 2.13 | 1.41 | 3.09 | 1.10 | 9.00 |
| PWG3 | 1.20 | 2.21 | 0.530 | 2.10 | 1.44 | 3.07 | 1.00 | 9.02 |
| Mean | 1.21 | 2.22 | 0.517 | 2.11 | 1.42 | 3.05 | 1.07 | 9.01 |
| SD | 0.02 | 0.02 | 0.02 | 0.02 | 0.02 | 0.05 | 0.06 | 0.01 |
| RSD | 1.4 | 0.9 | 3.0 | 0.8 | 1.5 | 1.5 | 5.4 | 0.1 |

Table 5: Concentration of Heavy metals in Pumpkin irrigated with Grounnd water in Wurukum (mg/kg)

Key:

PZG= Pumpkin crop-Zua- Grround water irrigated

PFG= Pumpkin crop-Fiidi- Ground water irrigated

PWG= Pumpkin crop-Wurukum- Ground water irrigated

PNG= Pumpkin crop-New Garage-Ground water irrigated

PWAG= Pumpkin crop- Wadata- Ground water irrigated.

Discussion

Anthropogenic activities, land use, lithogenic contribution by surface runoff erosion, deposition of eroded materials, temperature etc can artificially change organic matter content as well as a heavy metal concentration in the soil. The heavy metals studied in pumpkin with the exception of chromium had values higher than FAO/WHO recommended permissible levels (Pb 0.3, Cd 0.2, Fe 425, Zn 99.40, Cu 73.0, Cr 0.05, Mn 1.0 (in mg/kg). The bioaccumulation of these metals into the pumpkin leaf may be due to non - formation of complexes with the organic matters in the soil thereby making it possible for these metals to leach from the soil into the plant. (Deko and

Sarma, 2012;Kabatta and Henryk 1984).

Akhionbare and Akhionbare, (2004) reported that increase in total zinc content of the soil could lower fluted pumpkin lead accumulation. Zhangreen et al. 2002, also reported that the soil, root and stem play a role in the process of heavy metal transport. These The level of Pb,Cr and Zn reported in the pumpkin leaf from these locations were in agreement with those reported in the study of Oshman, (2001) for pumpkin and Amaranths leaf. Comparing the average heavy metal contents obtained in vegetable to the maximum levels permitted by FAO/WHO, we observed that people eating vegetable grown at these locations, were consuming it at concentration levels that are potentially hazardous to their health.

The results of the present study showed high concentration levels of the heavy metals in pumpkin samples irrigated using ground water appreciably above FAO threshold limit though with the exclusion of lead and iron which was below the threshold. The high concentration of the heavy metals in the crops reflected the concentration in ground water source used for irrigation. Kumar et al. (2007) and Conor (2002) opined that heavy metals can be transferred from soil to other components of the ecosystem such as underground water or crops. Studies of Al-Jaboobi et al. (2014) observed normal levels of Fe, Cu, Zn, Mn, Ni and Cd in edible portions of vegetable crops irrigated groundwater sources with the exception of Cr and Pb which exceeded the FAO/ WHO limits of 0.5 mg/kg. High accumulation of Pb in vegetables irrigated with heavy metal contaminated groundwater sources was previously reported in Nebal (Sharma et al., 2007).

The results of analysis of heavy metals in pumpkin, spinach and tomato samples irrigated using surface water appreciably above FAO threshold limit though with the exclusion of lead and iron which were below the threshold. Previous research findings of Nazir et al. (2015) in four wild plants namely Xanthium strumarium. Acacia modesta. Dodmea viscosa and Tamarix aphyda showed that cadmium, zinc, iron, copper, nickel, chromium and lead were present. Plants samples were analyzed separately for their root, stem and leaves. Flame absorption spectrometer was used for analyzing the samples and concentrations of cadmium, chromium, iron and lead in water were recorded above the permissible limits set by WHO while zinc and copper were recorded below the permissible limits and no concentration of nickel was recorded in water samples.

Chiroma *et al.* (2003) in their study on heavy metal contamination of vegetables and soils irrigated with sewage water in Yola, Nigeria reported high concentration of heavy metals (especially Fe, Zn, Cu, Mg, Mn and Pb) contamination in the soils irrigated with sewage water. Their study also found out that there is accumulation of these metals in different parts of plant cultivated on the soil. The study also shows that the heavy metal concentration vary in different parts of the plant with Fe shown to accumulate in roots and leaves while Zn accumulates in roots and translocates gradually to the leaves and Mn and Mg show greater accumulations in unwashed leaves. Arora et al. (2008) carried out a study in Rajasthan India to assess the level of different heavy metals such as iron, manganese, copper and zinc, in vegetables irrigated with water from different sources. The results indicated a substantial build-up of heavy metals in vegetables irrigated with wastewater.

The heavy metals detected in both surface and ground water sources were cadmium (Cd), Chromium (Cr), cobalt (Co), Copper (Cu), iron (Fe), Manganese (Mn), lead (Pb) and Zinc (Zn). The results revealed high level concentration of the detected heavy metals in both sources above FAO limits for irrigation water except for Lead and Iron. Previous studies of Taghipour et al. (2012) revealed the presence of cadmium (Cd), chromium (Cr), Copper (Cu), lead (Pb) Nickel (Ni) and zinc (zn) in ground water used for irrigation in Tabriz city in Iran. The average and even maximum concentrations of heavy metals in ground water was reportedly less than the toxicity threshold limits of agricultural water, thus signifying the effectiveness of the efforts of authorities in reducing the rate and level of heavy metal contamination, hence posed no health risk as compared to the findings of our study.

As an interim measure, the source of pumpkin in the affected areas studied should be regularly checked so as to avoid food and vegetables irrigated with polluted water since the linkage between crops, heavy metals content and heavy metals in irrigation water has been established in previous studies. REFERENCES.

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APPENDICES

| | Cd | Cr | Co | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PZS1 | 2.00 | 2.00 | 0.800 | 3.10 | 1.00 | 2.50 | 1.00 | 9.00 |
| PZS2 | 2.02 | 2.12 | 0.810 | 3.20 | 1.11 | 2.50 | 1.09 | 9.05 |
| PZS3 | 2.20 | 2.10 | 0.800 | 3.13 | 1.10 | 2.54 | 1.07 | 9.03 |
| Mean | 2.07 | 2.07 | 0.803 | 3.14 | 1.07 | 2.51 | 1.05 | 9.03 |
| SD | 0.1 | 0.1 | 0.01 | 0.1 | 0.1 | 0.02 | 0.05 | 0.03 |
| RSD | 5.3 | 3.1 | 0.7 | 1.6 | 5.7 | 0.9 | 4.5 | 0.3 |
| | | | | | | | | |

| | Cd | Cr | Co | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PFS1 | 1.00 | 2.20 | 0.400 | 2.60 | 1.00 | 2.30 | 1.20 | 6.80 |
| PFS2 | 1.06 | 2.27 | 0.430 | 2.62 | 1.00 | 2.34 | 1.23 | 6.82 |
| PFS3 | 1.00 | 2.26 | 0.420 | 2.61 | 1.08 | 2.30 | 1.22 | 6.80 |
| Mean | 1.02 | 2.24 | 0.417 | 2.61 | 1.03 | 2.31 | 1.22 | 6.81 |
| SD | 0.03 | 0.04 | 0.02 | 0.01 | 0.05 | 0.02 | 0.02 | 0.01 |
| RSD | 3.4 | 1.7 | 3.7 | 0.4 | 4.5 | 1.0 | 1.3 | 0.2 |
| | | | | | | | | |

| | Cd | Cr | Co | Cu | Fe | Mn | Pb | Zn |
|------|-------|------|-------|------|------|------|------|------|
| PWS1 | 0.900 | 1.60 | 0.400 | 2.40 | 1.00 | 1.20 | 1.40 | 8.50 |
| PWS2 | 0.900 | 1.63 | 0.400 | 2.42 | 1.00 | 1.21 | 1.42 | 8.57 |
| PWS3 | 0.960 | 1.61 | 0.430 | 2.44 | 1.05 | 1.24 | 1.40 | 8.56 |
| Mean | 0.920 | 1.61 | 0.410 | 2.42 | 1.02 | 1.22 | 1.41 | 8.54 |
| SD | 0.03 | 0.02 | 0.02 | 0.02 | 0.03 | 0.02 | 0.01 | 0.04 |
| RSD | 3.8 | 0.9 | 4.2 | 0.8 | 2.8 | 1.7 | 0.8 | 0.4 |

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|-------|------|-------|------|------|------|------|------|
| PNS1 | 0.700 | 2.10 | 0.400 | 2.90 | 1.10 | 2.00 | 1.10 | 9.10 |
| PNS2 | 0.690 | 2.13 | 0.420 | 2.91 | 1.10 | 2.00 | 1.10 | 9.08 |
| PNS3 | 0.740 | 2.15 | 0.410 | 2.90 | 1.13 | 2.10 | 1.12 | 9.30 |
| Mean | 0.710 | 2.13 | 0.410 | 2.90 | 1.11 | 2.03 | 1.11 | 9.16 |
| SD | 0.03 | 0.03 | 0.01 | 0.01 | 0.02 | 0.06 | 0.01 | 0.1 |
| RSD | 3.7 | 1.2 | 2.4 | 0.2 | 1.6 | 2.8 | 1.0 | 1.3 |

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|-------|-------|------|-------|------|------|------|------|------|
| PWAS1 | 0.800 | 2.00 | 0.500 | 3.10 | 1.20 | 1.80 | 1.20 | 6.90 |
| PWAS2 | 0.860 | 2.10 | 0.550 | 3.12 | 1.30 | 1.84 | 1.20 | 6.79 |
| PWAS3 | 0.810 | 2.12 | 0.530 | 3.10 | 1.16 | 1.80 | 1.26 | 6.80 |
| Mean | 0.823 | 2.07 | 0.527 | 3.11 | 1.22 | 1.81 | 1.22 | 6.83 |
| SD | 0.03 | 0.06 | 0.03 | 0.01 | 0.07 | 0.02 | 0.03 | 0.06 |
| RSD | 3.9 | 3.1 | 4.8 | 0.4 | 5.9 | 1.3 | 2.8 | 0.9 |

| | Cd | Cr | Co | Cu | Fe | Mn | Pb | Zn |
|------|-------|------|-------|------|------|------|------|------|
| PZG1 | 1.80 | 1.80 | 0.400 | 2.30 | 1.50 | 4.60 | 1.50 | 2.30 |
| PZG2 | 1.82 | 1.80 | 0.400 | 2.32 | 1.53 | 4.65 | 1.53 | 2.31 |
| PZG3 | 1.80 | 1.83 | 0.430 | 2.30 | 1.54 | 4.66 | 1.55 | 2.33 |
| Mean | 1.81 | 1.81 | 0.410 | 2.31 | 1.52 | 4.64 | 1.53 | 2.31 |
| SD | 0.01 | 0.02 | 0.02 | 0.01 | 0.02 | 0.03 | 0.03 | 0.02 |
| RSD | 0.6 | 1.0 | 4.2 | 0.5 | 1.4 | 0.7 | 1.6 | 0.7 |
| | | | | | | | | |
| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
| PFG1 | 0.800 | 2.00 | 0.400 | 2.20 | 1.50 | 3.50 | 1.70 | 7.20 |
| PFG2 | 0.820 | 2.00 | 0.430 | 2.20 | 1.50 | 3.56 | 1.72 | 7.40 |
| PFG3 | 0.810 | 2.08 | 0.440 | 2.23 | 1.50 | 3.54 | 1.73 | 7.32 |
| Mean | 0.810 | 2.03 | 0.423 | 2.21 | 1.50 | 3.53 | 1.72 | 7.31 |
| SD | 0.01 | 0.05 | 0.02 | 0.02 | 0.0 | 0.03 | 0.02 | 0.1 |
| RSD | 1.2 | 2.3 | 4.9 | 0.8 | 0.0 | 0.9 | 0.9 | 1.4 |
| | | | | | | | | |
| | | | | | | | | |
| | Cd | Cr | Co | Cu | Fe | Mn | Pb | Zn |
| PWG1 | 1.20 | 2.20 | 0.500 | 2.10 | 1.40 | 3.00 | 1.10 | 9.00 |
| PWG2 | 1.23 | 2.24 | 0.520 | 2.13 | 1.41 | 3.09 | 1.10 | 9.00 |
| PWG3 | 1.20 | 2.21 | 0.530 | 2.10 | 1.44 | 3.07 | 1.00 | 9.02 |

2.11

0.02

0.8

1.42

0.02

1.5

3.05

0.05

1.5

Mean

SD

RSD

1.21

0.02

1.4

2.22

0.02

0.9

0.517

0.02

3.0

1.07

0.06

5.4

9.01

0.01

0.1

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|------|------|------|-------|------|------|------|------|------|
| PNG1 | 1.00 | 2.20 | 0.500 | 1.90 | 1.40 | 3.20 | 1.30 | 7.80 |
| PNG2 | 1.00 | 2.20 | 0.520 | 1.90 | 1.43 | 3.20 | 1.04 | 7.91 |
| PNG3 | 1.08 | 2.30 | 0.530 | 1.86 | 1.45 | 3.32 | 1.46 | 7.83 |
| Mean | 1.03 | 2.23 | 0.517 | 1.89 | 1.43 | 3.24 | 1.27 | 7.85 |
| SD | 0.05 | 0.06 | 0.02 | 0.02 | 0.03 | 0.07 | 0.21 | 0.06 |
| RSD | 4.5 | 2.6 | 3.0 | 1.2 | 1.8 | 2.1 | 16.7 | 0.7 |

| | Cd | Cr | Со | Cu | Fe | Mn | Pb | Zn |
|-------|-------|------|-------|------|------|------|-------|------|
| PWAG1 | 0.800 | 2.00 | 0.400 | 2.00 | 1.20 | 2.80 | 0.900 | 8.30 |
| PWAG2 | 0.800 | 2.20 | 0.400 | 2.00 | 1.23 | 2.82 | 0.910 | 8.43 |
| PWAG3 | 0.830 | 2.30 | 0.430 | 2.20 | 1.20 | 2.83 | 0.920 | 8.00 |
| Mean | 0.810 | 2.17 | 0.41 | 2.07 | 1.21 | 2.82 | 0.91 | 8.24 |
| SD | 0.02 | 0.2 | 0.02 | 0.1 | 0.02 | 0.02 | 0.01 | 0.2 |
| RSD | 2.1 | 7.1 | 4.2 | 5.6 | 1.4 | 0.5 | 1.1 | 2.7 |