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Heavy metals contamination in vegetable crops irrigated with wastewater: A review

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Abstract

The demand for quality fresh water for agriculture has made waste water application popular for use and heavy metals contamination issues becoming increasingly globally with many documented cases of metal toxicity in mining industries, foundries, smelters, transportation, coal-burning power plants and agriculture. This paper attempted to collate and compile the research literature pertaining to the heavy metal contamination in vegetables irrigation with wastewater. It also reviews deeply about effect, uptake and accumulation of heavy metal in vegetables. Uptake and translocation factor of heavy metal from soil to edible parts of vegetables were quite distinguished for almost all elements examined. However, because of heavy metals bioaccumulation, non-degradability, and its presence in wastewater, contaminates vegetables and cause toxicity to human beings and the functioning of the entire ecology. This is a major issue of concern within the study of environmental science and geochemistry. Although, the issue is a menace, as such requires immediate attention for the developing countries, where the pressure of the teeming population escalates the exigency for human sustainability, food security, and total eradication of hunger. Globally, studies have examined some of these important issues, but most of these studies are fragmented and limited within the purview of mostly individual states and localities within the countries. On a wider geographical scale, the discussion and perspectives of vegetables irrigated with contaminated wastewater offer insufficient insight and expose merely a snapshot of the actual situation in which developing countries are lacking. Thus, the present study synthesizes existing literature and their findings to create a knowledge base on the vulnerability of the vegetables in developing countries. Thus, it is evident from the several research findings that heavy metals contaminate vegetables and has toxic effects humans and animals after certain limits.

Keywords: contamination, metals, irrigation, toxicity, wastewater

1. Introduction

The accumulation of toxic elements in the plants and soil remains a primary environmental concern worldwide due to their ability to accumulate in bio systems (Saglam, 2013) ^[111]. The implications associated with heavy-metal contamination are of great significance, particularly in agricultural production system. Tremendously escalating human population and expanding industry and urbanization has not only used a large area of World productive lands, but is also generating a large volume of wastewater every day (Khurana and Singh, 2012) ^[65].

The rapid increase in human population has also led to the increased demand for food production especially vegetables which are consumed on a daily basis. In most developing countries, rapid industrialization and urbanization in the 21st century have dramatically increased the discharge of large amounts of wastewater accompanied with toxic chemicals.

Estimates revealed an annual production of ~30 million tons of wastewater in the World, of which ~70% is consumed as an agricultural fertilizer and irrigation source (Cheraghi, *et al.*, 2009) ^[28]. With wastewater utilization crop production has gained an acceptance all over the World (Singh *et al.*, 2004) ^[117] and as an economic alternate that could substitute nutrient needs water requirement of plant crops (Rusan, *et al.*, 2007) ^[109].

Artisanal mining which dates back to several decades and have continued till date due to limited government investment in the sector and weak regulatory arising from mining is a major environmental concern on a global scale particularly in developing countries. The waste drains from industries into farmlands or streams, rivers and local channels constitute the primary sources of water for irrigation used in growing vegetables. (Balkhair and Ashraf 2016) ^[16].

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Heavy metal contamination is a major environmental health challenge and is potentially dangerous as metals continue to bioaccumulate through the food chain (Aycicek *et al.*, 2008)^[15], these arises from rapid industrial growth, advances in the use of agricultural chemicals, and the urbanization activities of man. This has now led to the dispersion of heavy metals in the environment, resulting in the impaired health of the population, mainly by the ingestion of vegetables contaminated by these harmful elements (Zukowska and Biziuk, 2008)^[136].

Heavy metals being natural components which cannot be degraded or destroyed biologically are toxic, when in the ecosystem may lead to geoaccumulation and bioaccumulation (Lokeshappa and Anil, 2012)^[74]. The amount of metallic elements in plants is the measure of the level of the metal in the environment in which the plants grows over a period of time (Ahmed *et al.*, 2012; Henry *et al.*, 2018)^[5, 44]. There are also many factors which contribute to heavy metal contamination identified as irrigation water, fertilizers and pesticides applications, emissions of different waste materials from industries, lack of good transportation facilities, harvesting process and storage. The natural contribution of heavy metals (parent material weathering) is very small but major contributing factor is anthropogenic activities which are potentially increasing its concentration in soil, plant and water. These include zinc mining, iron foundries, use of sewage sludge, vehicle exhaust and agronomic practices such as use of city effluent as irrigation in agriculture, fossil fuel combustion, pesticides, application of phosphate fertilizers and several other industrial processes (Iniobong *et al.*, 2012; Bernard, 2008)^[50, 19]. Plants have a natural propensity to take up metals like Cu^{2+} , Co^{2+} , Fe^{2+} , Mo^{2+} , Mn^{2+} , and Zn^{2+} which are essential plant micronutrients, while few others like Hg^{2+} , Ni^{2+} , Cd^{2+} , As^{3+} , Cr^{3+} and Pb^{2+} , are toxic to plants (Achakzai *et al.*, 2011)^[4]. However, such toxic effects are even varying from genotype to genotype of the same crop. Toxic heavy metals are associated with cardiovascular, kidney, nervous and bone diseases. Vomiting, diarrhea, stomach irritation, decreases in reaction time, kidney problems, anemia and blood disorders in humans are some of the diseases associated with heavy metals. They may also cause respiratory tract cancer and mesodermal ulceration (Ahmed *et al.*, 2012)^[5].

This study focuses on the contamination of heavy metals in vegetables irrigated with wastewater, regarding their total concentration levels in the environment. With the early industrialization many developing nations has poor implementation of environmental protection policies and regulations, its environmental pollution and contamination have been a great concern. According to Onakpa *et al.* (2018)^[92], both natural and anthropogenic sources are pathways by which heavy metals and metalloids contaminate many food crops and vegetables.

The findings of these studies are consistent with the general view that heavy metals are non -biodegradable, and metallic elements with relatively high density are toxic or poisonous even at low concentrations. The accumulation of heavy metals in the environment overtime contaminates the food chain, which is a major source of environmental and human health risk (Hawkes, 1997; Ali *et al.*, 2013; 24, Cai *et al.*, 2019; Esmailzadeh *et al.*, 2019)^[43, 7, 23, 33]. Despite the obvious significance of these studies, wide knowledge base of the implication of heavy metals on vegetables is globally lacking mostly in developing countries. This review provides important insight into the toxicity of heavy metals and that particularly vegetables are increasingly vulnerable to heavy metals and hazardous for consumption. The

findings of the current studies will create a knowledge base which somewhat simplifies understanding of the impacts of heavy metals on food chain globally. Given the high population of developing countries, its extreme poverty level, public health challenges (WHO, 2015)^[130], food availability and consumption are key factors to its continuous existence. Therefore, this research informs decision towards mitigating several health issues caused by heavy metals in crops and vegetables for human consumption and capital development.

2. Methods and Data

To achieve the objectives of this review, the authors conducted a structured search process to identify the body of the current literature relevant to the study. In collecting relevant literature, the authors ensured that they maintained quality and academic standard, which are often the basis of most literature review studies, for example, (Manikas and Hansen, 2013; Nkwunonwo *et al.* 2015; Thonemann and Schumann, 2018)^[82, 90, 126]. Therefore, the present literature search considered mainly research articles published in highly reputed journals (indexed by Scopus and Scimago and ranked in Thompson Reuter). The authors consulted EBSCO's environmental database, which hosts top open access environmental research journals: Green file, which archives well-researched debates covering all aspects of human impact on the environment. The study also considered several other libraries including DOAJ (Directory of Open Access Journals) digital Library, Google Scholar, PubMed, E-resources, IEEE Explore, Springer Link, Science Direct, and Thomson Reuters' Web of Science. Key terms such as "heavy metals," "Vegetables," and "Contamination," along with the combination of terms such as "heavy metal in Nigeria," "heavy metal uptake by vegetables," "heavy metals toxicity," and "factors influencing heavy metals in soil" were applied to the search. The criteria adopted by the authors in collection were that literature should address heavy metals as area of core research, must be a research paper, language must be English, current research and the literature must be none of these: an abstract with extended perspective, short communications, letters, and presentations.

2.1 Reviews

Many publications are available in developing and developed nations around the world describing the heavy metal contamination and toxicity; are briefly discussed in the paper. Due to the number of references collected from the variety of sources, some lacks or omissions are possible.

Henry *et al.* 2018^[44] assessed heavy metals (Cu, Cr, Mn, Cd, and Fe) in ex-mining pond water used for irrigation on the soil and two vegetable samples (Cabbage and Tomatoes) and found that all the selected heavy metals detected were within the range of 0.001-216.50 ppm in Cabbage, in tomatoes samples, the heavy metal concentration were in the ranged of 0.131-3.299 ppm. Chromium and cadmium were not detected and Mn, Pb and Fe were above WHO permissible limit. In Cabbage, all metals were detected except chromium and were in the range of 0.006 - 1.900ppm. Mn, Pb, Cd and Fe were above WHO permissible limit except for Cu.

Iyaka, 2007^[52] determined copper and zinc contents in vegetable samples in the Nigeria (middle belt region) and showed that copper concentrations were 5 mg/kg in peppers, 4 mg/kg in onions, and 5.5 mg/kg in tomatoes, while zinc levels were 13.5 mg/kg in peppers, 16.75 mg/kg in onions, and 15.5 mg/kg in tomatoes. The concentrations in plant leaves and crops of cobalt (0.33mg/kg) and iron (0.32mg/kg) in Roselle leaves, copper

(0.71mg/kg) and arsenic (0.37mg/kg) in groundnut, copper (0.48mg/kg) and arsenic (0.28mg/kg) in maize grains, arsenic (0.36mg/kg) and cobalt (0.32mg/kg) in spinach leaves, and copper (0.36mg/kg) and cobalt (0.32mg/kg) in okro in Lafia, Nassarawa State, Nigeria. The concentration of Arsenic in all sample studied were higher than the permissible limit of WHO. Similar by Paluwa *et al.*, 2012^[94] assessed the concentration of Fe and Ni during dry and rainy seasons and noticed that Fe levels reduced by 8.25% in onions and nickel by 45.19% in okro in rainy season samples over those of the dry season, and the mean levels of metals in the okro samples for the dry season were in the order Fe > Cu > Zn > Mn > Ni > Pb > Co > Cr, while those of the rainy season indicated Fe > Cu > Zn > Mn > Ni > Co > Pb > Cr. This trend suggests that okro has a higher retention capacity for essential metals, zinc, manganese, and copper, than for the toxic ones, nickel, lead, cobalt, and chromium (Zhou, *et al.*, 2016)^[135] Mafuyai *et al.* 2020a^[79], determined the accumulation of heavy metals in vegetables (garden egg (*Solanum melongena*), Spinach (*Spinacia oleracea* L.), Tomato (*Lycopersicon esculatum* L.), red pepper (*Capsicum annum*), Carrot (*Daucus carota* subsp. *Sativus*) and cabbage (*Brassica oleracea*) in Jos-South, Plateau State, Nigeria. From his study the concentration the metals in Tomato were; Pb (0.49±0.1), As (0.076 ±0.001, Cr (0.15 ±0.01, Mn (0.45±0.39) mg/kg, while other metals such Cu, Zn, and Fe, has concentrations of 0.071±0.01, 0.376±0.04 and 1.55±0.21 mg/kg, respectively. Garden egg: Pb (0.40 ±0.05), Cr (0.12 ±0.00) and Mn (1.17±0.02) As (0.83 ±0.11) and Cu (0.076±0.01) mg/kg, Pepper: Pb (0.307±0.26), Cu (0.065±0.00), Cd (0.006±0.00), Zn (0.395±0.02) Cr (0.106±0.01), Fe (1.489±0.03), Mn (0.177±0.01) and As (0.033±0.02, Cabbage: Pb (0.164±0.03), Cu (0.065±0.01), Cd (0.01±0.00), Zn (0.18 ±0.02), Cr (0.11±0.01), Fe (1.49±0.07), Mn (0.17±0.01) and As (0.03±0.01) mg/Kg, Carrot: Pb (0.46 mg/Kg) and As (0.47 mg/Kg) were higher compared to other toxic metals in and Spinach: Pb (0.46 ±0.04) Cu (0.06±0.01), Cr (0.014±0.01), Zn (0.45±0.03), Cr (0.15±0.03), Fe (1.55±0.21) The order of heavy metals accumulation in the vegetables was Fe > Mn > Pb > Zn > Cu > Cr > As > Cd. The result revealed that Pb, Cd and Cr were higher than permissible levels in some of vegetables. Similarly, Mafuyai *et al.* 2020b^[81] assessed heavy metals concentration in vegetables garden egg (*Solanum melongena*), Spinach (*Spinacia oleracea* L.), Tomato (*Lycopersicon esculatum* L.), red pepper (*Capsicum annum*), Carrot (*Daucus carota* subsp. *Sativus*) and cabbage (*Brassica oleracea*) irrigated with tin mine pond water in Jos – South, Plateau State and reported that the concentration of the metals was in the range of Pb (0.177 – 0.545), Cu (0.073 – 0.748), Cd (0.005 – 0.019), Zn (0.264 – 0.915), Cr (0.089 – 0.158), Mn (0.162 – 0.253) and As (0.032 – 0.245) mg/kg. The continuous use of tin mined pond water irrigation has changed the soil physicochemical characteristics and has led to heavy metal uptake by food crop, predominantly vegetables. Ibrahim *et al.* 2014^[47] determined metals in Lettuce which revealed that Cu ranged from 0.43 - 0.83; Zn from 2.20 - 3.60; Fe, from 2.60 - 3.03; Pb, from 0.09 - 0.22 mg/kg with a mean value of 0.61, 2.65, 0.56, 1.90 and 1 0.20 mg/kg dried weight of plant, respectively. Lawal and Audu (2011)^[70] similarly analyzed heavy metals (Cr, Cu, Pb and Zn) concentrations in vegetables irrigated with of industrial and domestic wastewaters. Four different vegetables were collected from both fields during dry and rainy seasons including, spinach, okra, onions and tomatoes, grown in effluent irrigated fields and another irrigated field at Thomas Dam in the Kano metropolis, Nigeria. The mean level of metals obtained ranged widely from 0.28 mg/Kg Cr to

18.89 mg/kg Zn. Samples from Jakara garden indicated the mean levels of Cu (7.50 ± 1.08 mg/Kg), Zn (18.89 ± 1.93 mg/Kg) and Cr (0.85 ± 0.10 mg/Kg) while those from Sharada indicated the significant levels of Pb (1.60 ± 0.53 mg/kg). However, the levels were within the National Agency for Food and Drug Administration and Control (NAFDAC) tolerable limits for metals in fresh vegetables.

Determined concentrations of Arsenic (As) and Cadmium (Cd) in three vegetables (*Moringa oleifera*, Spinach and cassava) around river Tudun Wada, Kaduna State, Nigeria. The results of the soil, sampled vegetables revealed that, *Moringa Oleifera* (7.90 ± 0.2 µg/kg As and 14850 ± 100 µg/kg Cd); Spinach (7.70 ± 0.3 µg/kg As and 14800 ± 346 µg/kg Cd) and Cassava (8.10 ± 0.5 µg/kg As and 14950 ± 574 µg/kg Cd) respectively. The concentrations of heavy metals in *Telfairia occidentalis* (fluted pumpkin), *Talinium triangulare* (water leaf) and *Amaranthus hybridus* (spinach) cultivated along the south bank of River Benue in Makurdi determined by Adah *et al.*, 2013^[1] revealed (mg/kg) of Cd, Cr, Cu, Ni and Pb in *T. occidentalis* were 0.0043, 0.0268, 0.2501, 0.2047 and 0.1868; *T. triangulare*: 0.0058, 0.0090, 0.0902, 0.2207 and 0.1641 and *A. hybridus*: 0.0078, 0.0172, 0.0863, 0.0714 and 0.2441, respectively. Generally, the concentrations of the heavy metals in the vegetable were below the permissible limits set by European Union, WHO/FAO, and Nigerian Agency for Food and Drug Administration and Control (NAFDAC).

Edogbo, *et al.*, 2020^[32], reported the heavy metals concentration of (Cd, Cr, Pb and Zn) in vegetables collected from Challawa industrial area in Kano State, as 6.13 ± 2.10, 0.46 ± 0.07, 0.73 ± 0.12 and 23.8 ± 4.30 mg/kg, respectively. The levels of Cd and Cr from the study were above the permissible limits of 0.2 mg/kg for Cd, 2.3 mg/kg for Cr in vegetables, as set by WHO/FAO, (2007)^[35]. The order of concentration of the heavy metals in vegetables were in the order of Fe > Zn > Mn > Cu > Ni > Pb > Co > Cr. Onakpa *et al.* 2018^[92] carried out a similar study on irrigated vegetable farms in Kano Metropolis and found out that the heavy metals in the farm produce followed this sequence Fe > Zn > Mn > Cu > Ni > Pb > Co > Cr.

Dorcas *et al.* 2016^[31] reported Levels of heavy metals: cadmium (Cd), lead (Pb), arsenic (As) and mercury (Hg) investigated in vegetables (carrots and tomatoes) planted on farms near the old mining sites in Jos, Plateau State. The results showed that concentrations of Cadmium (Cd) ranged from 0.01–0.45mg/kg, lead (Pb) ranged from 0.01– 0.43mg/kg, arsenic (As) ranged from 0.02 –1.20mg/kg lastly, mercury (Hg) ranged from 0.01 – 0.39mg/kg above FAO/WHO, 2006. The concentrations of the metals followed the order of Cd < Pb < As < Hg. with the high uptake of metals by tomatoes compared to carrots' uptake.

Determined heavy metals concentration in the edible parts of the vegetables irrigated with the stream water were it noted to be significantly higher with the Cabbage accumulated high levels Cr, Ni and Pb with the concentrations 55.430mg/kg, 26.660 mg/kg, and 26.480 mg/kg, respectively. Similarly, Jimoh and Mahmud, 2011^[55] assessed Cadmium and Lead in Tomatoes Grown in Irrigated Farmland of the Kaduna Metropolis Nigeria. The concentrations of tomatoes samples for cadmium were found to be in the range of 0.10±0.10 - 1.33±0.23 g/g and lead ranged from 0.92±0.08 to 4.67±1.29 g/g. This also shows that concentrations of cadmium and lead in tomatoes samples were above FAO/WHO Allimentarious standard.

Heavy metal analyzed by in different vegetables grown in the vicinity of an industrial area show variable concentrations such as Cd (0.011–0.073 mg kg⁻¹), Pb (1.121–2.652 mg kg⁻¹), Cu (0.161–0.923 mg kg⁻¹), Zn (0.361–1.893 mg kg⁻¹) and Ni

(0.288–0.546 mg kg⁻¹). The magnitude of heavy metals detected in different kinds of vegetables was Cd < Cu < Cr < Zn < Pb. The cabbage (0.073 mg kg⁻¹) exhibited higher levels of Cd than the other vegetables. In contrast, radish (2.652 mg kg⁻¹) contained the highest levels of Pb, while spinach (0.923 mg kg⁻¹) contained the highest Cu concentration. Furthermore, the concentration of Cr was higher in cauliflower, whereas lettuce revealed greater amount of Zn than the other vegetables.

In vegetables study of heavy metals by concentration of Cu, 29.10mg/kg in spinach and Mn has 26.36 mg/kg in round gourd while Cd was found to be 1.61mg/kg in cabbage, and 0.24mg/kg in carrot. Cr concentration was in the range of 0.24 - 3.70 mg/kg in spinach, Cu concentration was in the range of 4.99 - 29.1 mg/kg in spinach, while Mn ranged from 0.88 - 26.09 mg/kg in round gourd, carrot and radish. Ni has 3.58 mg/kg in spinach, 0.13 mg/kg in potato and Pb in vegetables 5.45 mg/kg and 0.05 mg/kg in turnip. In all vegetables general trend found was Cu > Mn > Cr > Ni > Pb > Cd.

Khan *et al.* 2015^[64] determined the Mineral and Heavy Metal Contents of Some Vegetable Available In Local Market of Dhaka City in Bangladesh and the results reveal that the essential mineral Ca, Na, K, Mg and essential trace element Fe, Zn found were ranged from 171.01-932.69, 91.26 - 655.62, 107.50 - 864.28, 56.49 - 920.67 and 3.5 - 74.92, 0.8429.45mg/100g, respectively in three types of vegetables. In case of heavy metals Cu, Cr and Ni were found in all samples ranged from 0.284 - 7.55, 0.034 - 1.10, 0.25 - 1.506 and 0.25 - 1.506 mg/100g respectively. Cd was found in waste water irrigated and fresh water cultivated vegetables ranged from 0.005 - 0.009 and 0.01 - 0.015 mg/100g, respectively. Pb was found in wild vegetables and the range was 0.12-0.96 mg/100g. A significant increase ($p < 0.05$) in Cu, Cr, Ni, Cd and Pb were found when compared to WHO/ FAO permissible value. Hg and Co were absent in all kind of vegetables.

During a population health risk assessment on the consumption of heavy-metal-contaminated food crops and fruits in Owerri, Imo State, reported that the concentration of lead, cadmium, and nickel exceeded the maximum allowable concentrations for agricultural soil as recommended by the European Union. Levels of lead, cadmium, and nickel in food crops were highest in *Oryza sativa*, *Glycine max*, and *Pentabacta microfila* respectively. The highest levels of lead, cadmium, and nickel in fruits were detected in *Canarium schweinfurthii*, *Citrus reticulata*, and *Ananas comosus*, respectively. The concentration of heavy metals in vegetables were in the order Cd < Se < Cr < Cu < Ni < Zn. There were higher heavy metal levels in soil than vegetables samples. The concentration of metals (Pb, Zn, Cu, Cr, Cd, Mn and Ni) in selected fruits and vegetables from Ekan market in Warri Delta state, Nigeria were determined using Atomic Absorption Spectrometry (AAS).

Obi-Iyeke, 2019 determined the concentration heavy metals in Street-Vended Fruits and Vegetables in Warri, Delta State, Nigeria. The concentrations of metals (mg/kg) in all samples ranged from 0.0 - 0.91, 0.0 - 1.12, 0.74 - 1.51, 0.27 - 1.83, 0.02 - 1.74 and 0.15 - 1.93 for Cd, Cu, Ni, Mn, Zn and Pb respectively. Cr was not detected in all the samples. The result also indicated that the concentration of Cd, Cu, Ni, Mn and Zn were within the permissible limit for WHO 2015 while the concentration of Pb in most samples were above the WHO 2015 limit hence pose toxicological risk. Reported the concentration of heavy metals in vegetables obtained from Lagos market Nigeria showing that Cr has the highest concentration in the vegetables studied from among the investigated toxic heavy metals. The range of metals in the vegetables ranged from <0.0 - 17.13, 1.22-14.21, and <0.0

- 14.54 mg/kg in cabbage, potato and khat, respectively. In general, the results revealed that high concentrations of the toxic heavy metals were found in the vegetable samples, which are above the international safe limits.

Deribachew, *et al.*, 2015^[30] also investigated concentrations of the metals cabbage, potato, and khat plants, were found that; Cr [less than method detection limit (0 - 17.13), (11.96-14.21), and (9.04-15.54); Co (5.72-9.72), (5.15-8.72), and (0-8.87); Cd (1.15-2.46), (1.22-1.46), and (0.38-3.22); Pb (5.48-11.95), (5.43-7.78), and (4.49-11) mg/kg, respectively in the vegetables.

Assess the level of toxic metals Pb, Cd, As and Hg in different vegetables grown in Sindh province of Pakistan during 2007-2008. The samples were categorized into four viz., leafy, root and tuberous, cucurbits and fruity. The average concentration ($\mu\text{g g}^{-1}$) of Cd, Pb, As and Hg in leafy vegetables was found 0.083, 0.050, 0.042 and 0.0080, respectively, in roots and tuberous vegetables was 0.057, 0.030, 0.045 and 0.0040, respectively, in cucurbit vegetables was 0.021, 0.051, 0.056 and 0.0089, respectively and in fruity vegetables was 0.035, 0.067, 0.054 and 0.007, respectively. In leafy vegetables, the concentration of cadmium, lead and mercury were found comparatively higher than other three groups of vegetables.

Lente *et al.* (2014)^[71] studied the heavy metal concentrations in irrigation water, soil and edible parts of both exotic and traditional vegetables irrigated with wastewater and groundwater from some parts of Accra and Mampong. The Concentration levels (mg/kg) of heavy metals in vegetables crops analysed from all sites were not elevated except for Pb in cabbage, (10.51), lettuce (10.19), green pepper (9.44), hot pepper (7.61) and ayoyo (9.05) compared to the FAO/WHO (2010)^[34] maximum recommended limit.

Kafeel *et al.* 2014^[59] assessed the heavy metal and metalloid levels in spinach (*Spinacia oleracea* L.) grown in wastewater irrigated agricultural soil of sargodha, Pakistan and revealed that the mean concentrations of metals and metalloids in the vegetable ranged from (Cr ; 16.5 -30.7, Mn; 68.1 – 82.8, Fe; 46.9 – 71.9, Co; 0.64 – 1.11, Zn; 62.6 - 81.9, Cu; 22.8 – 31.8, Ni; 8.97 – 14.6, As; 4.49 – 8.31, Mo; 13.7 – 26.2, Cd; 0.74 – 1.16, Pb; 8.58 – 17.9 and Se; 0.62 – 0.93) mg/kg. Considerably elevated concentrations of metals were observed in the wastewater irrigated vegetable in which, the levels Mo, Cd and Pb in the vegetable exceeded the permissible limit suggested by the World Health Organization.

Mosleh and Almagrabi, 2013^[84] investigated the accumulation of heavy metals in some vegetables irrigated with recycled wastewater in Saudi Arabia. The values of heavy metals in wastewater was 19.98 mg/L for Zn, 0.98 mg/L for Cu, 0.4 mg/L for Cd and 2.1 mg/L for Pb. The high level of Zn and Pb was found in soils (11.24 and 8.32 mg/kg). The concentrations of heavy metal showed a trend of Zn>Cd>Cu >Pb in vegetable fruits, Cd>Zn>Cu>Pb in vegetable leafs. The accumulation of Cu in eggplant fruits was highest 7.67 mg/kg. The low concentrations of Pb (0.21, 0.26 and 0.32 mg/kg) were reported in leaves of lettuce, squash and garden rocket, respectively.

An assessment of some heavy metals concentrations in lettuce, irrigated with wastewater in Tamale Metropolis in Ghana, has been carried out by in the northern region of Ghana. From the analysis it was revealed that the mean concentrations of Fe, Mn, Cu, Zn, Cd and Pb in lettuce were 0.436, 0.345, 0.068, 0.017, 0.04 and 0.038 mg/L and 0.167, 0.163, 0.104, 0.127, 0.142 mg/kg respectively.

Tamene and Seyoum, (2015)^[124] studied the concentrations of As, Cd, Cr, Hg and Pb in soils and garlic, kale, onion, pepper and potato samples from farmlands irrigated with River Mojo

water, at Koka village, Oromia State, East Ethiopia using Flame and Hydride Generation Atomic Absorption Spectrometry. The test results showed that in all soil samples, all the five trace heavy metals exist. The order of the metals mean concentrations (mg/kg) in soil was: As (32.72 ± 22.5) > Pb (22.99 ± 11.09) > Cd (5.23 ± 5.94) > Cr (3.60 ± 2.81) > Hg (2.41 ± 1.24). The mean concentrations of Hg and Cr analyzed were above the FAO/WHO, 2011.

Boamponsem *et al.* 2012^[22] reported that Vegetables such as cabbage (*Brassica juncea*, *Brassica oleracea*) cultivated in wastewater-irrigated soils take up heavy metals in large enough quantities to cause potential health risks to the consumers. The highest concentration (0.221 mg/Kg) of Cu was found in carrot roots and the highest concentration (35.35 mg/Kg) of Zn was found in the roots of *Brassica*. Cd accumulation in *L. sativa* and *B. oleracea* was below detection limit ($< 0.002 \text{ mg/Kg}$). Pb absorbed by the three genotypes was below detection limit ($< 0.005 \text{ mg/Kg}$).

Bambara *et al.* 2015^[17] assessed heavy metals in irrigation vegetables (cabbage, tomato, spinach and lettuce) in selected farms at Loubila and Paspanga, Burkina Faso. The concentrations of the metals Co; 2.1 ± 0.02 , Cr; 0.04 ± 0.002 , Fe; 6.3 ± 0.16 , Mn; 0.4 ± 0.04 , Zn; 0.82 ± 0.6 and As; 0.12 ± 0.01 in cabbage, while in tomato the concentrations were Co; 0.2 ± 0.02 , Cr; 4.5 ± 0.04 , Fe; 28.89 ± 0.09 , Mn; 2.8 ± 0.04 , Zn; 4.52 ± 0.5 and As; 0.06 ± 0.02 , in spinach Co; 1.26 ± 0.1 , Cr; 0.04 ± 0.001 , Fe; 0.65 ± 0.06 , Mn; 0.38 ± 0.064 , Zn; 0.4 ± 0.01 and As; 0.04 ± 0.004 and in lettuce the concentrations was Co; 1.28 ± 0.08 , Cr; 0.04 ± 0.001 , Fe; 3.47 ± 0.04 , Mn; 0.28 ± 0.04 , Zn; 0.40 ± 0.04 and As; $0.04 \pm 0.01 \text{ mg/kg}$. The vegetable data indicate that the average concentration of Cr ($4.5 \pm 0.04 \text{ mg/kg}$) in tomato exceeded the WHO/FAO limit for vegetable.

Singh and Kumar, 2006 assessed heavy metal concentration in spinach and lady's finger. Result show the heavy metal accumulation in mg/kg varied from 7.0 - 50 for Cu, 8.4 - 51 for Zn, 1.4 - 9 for Cd, and 1.7 - 9.1 for Pb in spinach. In Lady's finger heavy metal accumulation varied from 12 - 29 for Cu, 39 - 136 for Zn, 0.4 - 6 for Cd and 0.8 - 7.3 for Pb. It was further observed that the accumulation of all the heavy metal was higher in spinach compared to lady's finger. In the same year Pandey and Pandey, 2009 also worked on accumulation of heavy metals (Cd, Cr, Cu, Ni and Zn) in Radish and Spinach vegetable irrigated with industrial effluent and the results showed the high accumulation of Cr, 302.0; Cu, 81.2; Ni, 155.1 and Zn, 146.8 $\mu\text{g/g}$ dry weight in spinach.

The concentration of heavy metals determined by Benti, (2014) in vegetables (lettuce, spinach and cabbage) in irrigated with Awash River in selected farms around Adama town Ethiopia shows that Pb, Cr and Cd in lettuce 0.53 ± 0.02 , 2.3 ± 0.2 and $0.31 \pm 0.2 \text{ mg/kg}$, spinach 1.86 ± 0.03 , 1.58 ± 0.3 and 0.57 ± 0.3 and cabbage 0.36 ± 0.02 , 0.26 ± 0.2 and $0.22 \pm 0.2 \text{ mg/kg}$, respectively. The result revealed that Pb and Cd concentration in the vegetables exceeds the permissible limit of 0.3 mg/kg and 0.2 mg/kg dry weight FAO/WHO, (2001).

Gupta *et al.* 2012^[41] studied the magnitude of contamination of vegetables (Cauliflower Spinach, Coriander, Parsley, Radish and Pudina) with heavy metals (Pb, Cd, Cr, Cu and Zn) was determined in a long term wastewater-irrigated agricultural land. Among all the vegetables of sewage-irrigated study area, highest contamination of Pb (51.78 mg/kg) in radish followed by Spinach (47.69 mg/kg) and Cauliflower (29.69 mg/kg). Maximum Zinc was found in Spinach (148.04 mg/kg) and the lowest value Cauliflower. The accumulation of cadmium is highest in spinach, (12.46 mg/kg) in cauliflower, highest level of

chromium in spinach (95.79 mg/kg). The level of copper and nickel are highest in spinach. Of all the examined vegetables, Zn showed high and Cd showed low concentration in all the vegetables. In similar year Singh *et al.* worked on heavy metals accumulation and distribution pattern in different vegetable crops. Metal accumulation in edible parts and whole plants, root vegetables namely, radish and carrot registered lower accumulation of almost all heavy metals except Zn in radish root. However, leafy vegetables namely, spinach, amaranthus, mustard and fenugreek recorded higher accumulation of both essential and nonessential heavy metals. Cauliflower and cabbage, however, showed greater accumulation of Pb and Ni, but less accumulation of Cu and Cd.

Taghipour and Mosafari, 2013^[123] analysed heavy metal (Cd, Cr, Cu, Ni, Pb and Zn) in vegetables including kurrat, onion and tomato. The result show that the average mean of Cd, Cu, Cr, Ni and Zn concentration are 0.32 ± 0.58 , 28.86 ± 28.79 , 1.75 ± 2.05 , 6.37 ± 5.61 and 58.01 ± 27.45 . Cr, Cu and Zn were present in all the vegetables. In Nasik city, Maharashtra, Kailas, (2013) detected the concentration of Pb, Cd, As and Cu from randomly collected samples of coriander, spinach, onion, cauliflower, brinjal, cabbage, tomato, cucumber, potato and carrot from four different sites. Pb concentration ranges from 1.60 - 9.70 ppm for Nashik industrial area and 1.90 - 7.10 ppm for Sinnar industrial area Cd concentration in two out of ten samples collected from Nandur has recorded higher than the permissible limits of 1.5 $\mu\text{g/g}$. 30% vegetable sample collected from Nashik and Sinnar industrial area recorded Cd concentration higher than the permissible limit. Concentration ranges from 0.60 - 3.30 ppm for Nashik industrial area and 0.80 - 2.20 ppm for Sinnar industrial area. Cu concentration of most of vegetable sample was within permissible limit. Only two out of 40 samples showed Cu concentration above permissible limit. Concentration ranges from 4.90 - 30.80 ppm for Nashik industrial area, 6.30 - 14.20 ppm for Sinnar industrial area, 3.10 - 15.20 ppm for Nandur village and 2.20 - 7.10 ppm for Dindori. As concentration in all sample collected from Dindori and 90% sample collected from Nandur were within safe limit. Similarly in Raipur city, India, Gupta, 2013^[42] investigated the concentrations of Cu, Cr, Zn, and Pb in the most frequently consumed vegetables including *Pimpinella anisum*, *Spinacia oleracea*, *Amaranthus viridis*, *Coriandrum sativum*, and *Trigonella foenum graecum* in various sites. Heavy metals in vegetables were in the order of $\text{Cr} > \text{Zn} > \text{Cu} > \text{Pb}$.

Swapna *et al.* 2014^[122] was investigated heavy metal contaminations in three leafy vegetables viz., Palak, Thotakura (*Amaranthus*) and Chukkakura, soil and water of Musi River. Results showed that, leafy vegetable Chukkakura had the highest metal accumulation ($801.78 \text{ mg kg}^{-1}$) followed by Palak ($550.97 \text{ mg kg}^{-1}$) and Thotakura ($493.34 \text{ mg kg}^{-1}$). In the similar year, Khan *et al.* 2008^[62] conducted the heavy metals accumulation in vegetables imported from India and compared with same vegetables collected from vegetable market in Pakistan. Green chili, capsicum, tomato and ginger were selected to analyze their heavy metal concentration. Maximum concentration of heavy metals detected by dry ash method in Indian vegetables Cu (0.34 ppm) in capsicum, Cd (0.0 ppm) in capsicum, Cr (0.22 ppm) in Ginger, Pb (0.22 ppm) in ginger and Ni (0.14 ppm) in ginger while in Pakistani vegetables, it were of Cu (0.62 ppm) in Tomato, Cd (0.04 ppm) in Capsicum, Cr (0.17 ppm) in Tomato, Pb (0.36 ppm) in Ginger. Heavy metal contents determined by wet digestion method Cu (0.57 ppm) in Ginger, Cd (0.01 ppm) in capsicum, Cr (0.17 ppm) in Ginger, Pb (0.27 ppm) in capsicum while in Pakistani vegetables these were of Cu (0.19 ppm) in

Ginger, Cd (0.04ppm) in green chili, Cr (0.09ppm) in Tomato, Pb (0.25ppm) in Ginger.

Jaishree and Khan (2015) ^[53] studied the concentration of heavy metals in crops and vegetables (Carrot, Tomato, Wheat, Brinjal, and Barley) irrigated with effluent from textile and tanning industries in Jaipur district. The range of various metals present in the effluent irrigated plants varied from 8.537-14.372 mg/g in wheat, 8.234-15.271 mg/g in carrot, 10.361-13.313 mg/g in Brinjal, 4.024-8.234 mg/g in tomato and 7.234-15.23 mg/g in barley. Nickel and lead found highest and Cadmium and copper found lowest in crops and vegetables. In same year, Verma and bhatiya, 2015 ^[129] determined heavy metal concentration and harmful effects of some edible vegetables around the area of Pariccha Thermal Power Station in Jhansi (Uttar Pradesh India) in Fenu Greek, Cabbage, Cauliflower, Lady's Finger and Brinjal. The maximum concentration of Cu found in Brinjal is 22.412 µg/g. The maximum concentration of Cr was found in Cauliflower as 7.881 µg/g and the value of Cd was found to be < 0.05 µg/g in all samples.

Sulaiman *et al.* 2015 ^[119], determined the levels of heavy metals in tomatoes, and assessed the health risk via consumption of tomatoes grown by the irrigation in Kwadon, Gombe, Nigeria. A total of 45 soil and tomato samples were collected and analyzed for Mn, Cd, Ni, Cr, Pb, Cu and Zn using the concentration of heavy metals ranged from (0.99 - 1.00 mg/kg) Mn, (0.98 - 1.09 mg/kg) Cd, (0.08 - 0.11 mg/kg) Ni, (0.12 - 0.22 mg/kg) Cr, (0.68 - 0.71 mg/kg) Pb, (0.43 - 0.54 mg/kg) Cu and (1.99 - 2.13 mg/kg) Zn.

The levels of Lead (Pb), Cadmium (Cd), Iron (Fe), Zinc (Zn) and Copper (Cu) determined in four different samples of vegetables purchased from Katsina central market were determined by Shuaibu *et al.* 2013 ^[115]. The mean concentrations of metals ranged from (0.07) Pb - (0.632) Cu mg/kg. The relative abundance of metals in vegetables followed the sequence Cu (0.483) > Zn (0.268) > Fe (0.260) > Pb (0.095) mg/kg. The levels of Pb and other metals were below the FAO/WHO recommended limits for metals in vegetables. Low concentrations of Pb and absence of Cd in all the samples are indications that these plants contribute less toxic effects of metals.

Bassey *et al.* 2014 ^[18] determined the concentration of Pb, Cu, Ni, Zn, Cr, Mn and Fe in tomato leaves and fruits from peri-urban environments in Asaba, the result revealed that the metals in the tomato leaves samples were 4.01, 1.91, 1.83, 4.89, 0.16, 4.51 and 7.13 mg/kg in site A; 3.84, 1.56, 2.07, 4.00, 0.41, 4.48 and 8.15 mg/kg in site B, and 4.03, 1.75, 2.01, 4.52, 0.01, 4.42 and 8.11 mg/kg in site C for Pb, Cu, Ni, Zn, Cr, Mn and Fe respectively while in the tomato fruits, the concentrations of metals in mg/kg were 2.96, 0.41, 1.35, 3.33, 0.01, 3.83 and 6.38 mg/kg in site A; 3.01, 1.35, 1.88, 2.98, 0.15, 3.01 and 5.09 mg/kg in site B, and 3.92, 1.44, 1.82, 3.73, 0.01, 3.05 and 6.00 mg/kg in site C for Pb, Cu, Ni, Zn, Cr, Mn and Fe respectively. The values recorded for tomatoes leaves and fruit were below the levels recommended by WHO/FAO and NAFDAC for metals in foods and vegetables but are within the normal range of metals in plants.

Charu Jhamaria *et al.* 2015 ^[25] estimated the accumulation of heavy metals (Pb, Cd, Cr and Ni) in vegetables spinach, tomato, lady finger and brinjal due to wastewater irrigation. In the study Cr was found to be highest (17.26 mg/kg) in spinach followed by tomato (15.26 mg/kg), brinjal (5.1 mg/kg) and lady finger (4.25 mg/kg). Higher levels of Cr in spinach as compared to other studies at wastewater irrigated sites like (13.91 mg/kg) observed by Prabu (2009) ^[104] at Ethiopia. Similarly, in

Hyderabad, India, Karamtothu *et al.* 2015 ^[61] studied effect of levels of some heavy metals like Copper (Cu), cadmium (Cd), chromium (Cr) nickel (Ni), lead (Pb), Iron (Fe), Manganese (Mn) and zinc (Zn) contents of various vegetables Spinach, Ladyfinger, pepper mint, coriander, Tomato. The concentration ranges in mg/kg were (1.45 - 2.55) for Cd, (3.10 to 4.92) Cr, (12.15- 20.50) Cu, (25.00-51.00) for Fe, (7.80 - 15.60) for Mn, (10.16 - 15.42) for Ni, (2.12 - 5.41) Pb and (16.58 - 24.08) for zinc.

In a study by kumari *et al.* 2016 ^[67] in Ludhiana, Punjab the concentration of some heavy metals- Chromium (Cr), Zinc (Zn), Copper (Cu), Cadmium (Cd), Nickel (Ni), and Lead (Pb) in vegetables (Cauliflower, Spinach, and Tomato). Pb and Zn had high transfer factors which are 6.77 and 6.25 respectively. Similarly, the health risk related to vegetables contamination of heavy metals by irrigated with sewage and turbine water in Bahawalpur, Pakistan, the concentration of heavy metals in vegetables irrigated with turbine water was ranged from 0.04 - 0.08, 0.02 - 0.08, 0.03 - 0.42, 0.02 - 0.08 mg/kg for Pb, Cd, Cr, Ni respectively (Iqbal *et al.*, 2016) ^[51].

Study by Jolly *et al.* 2013 to investigate the concentration of different metals in tomato and spinach cultivated agricultural revealed that the concentration of the different metals in tomato is Cr; 0.51±0.03 Mn; 0.06, Fe; 43.86 ±0.84, Co; 0.27, Ni; 0.65, Cu; 3.62 ±0.29, Zn; 31.1 ±0.43, As; 0.05 ±0.00, Se; 0.14 ±0.01, Sr; 0.14, Cd; 0.06 and Pb; 0.12, while the concentration of Fe, Co, Cu, Zn, Se, Sr and Pb in spinach was 0.006, 0.035, 0.106, 1.148, 0.069, 0.167 and 0.065, respectively. The study showed that the concentration of heavy metals in vegetables varied in different samples and hence variations in elemental concentrations among different varieties reflect the difference in uptake capabilities and their further translocation to the edible portion of the plants. Similar study of Pb and Cd in tomato by Gebeyehu and Bayissa, (2020) ^[39] observed higher values; 3.63 and 0.56 mg/kg, respectively, while the corresponding values in cabbage were 7.56 and 1.56 mg/kg for Pb and Cd, respectively. The values of both Pb and Cd metals obtained have shown statistically significant difference in cabbage and tomato vegetable types at 95% probability levels ($P < 0.05$). It can be clearly observed here is that leafy vegetable (cabbage) happened to accumulated more toxic metals than the fruity vegetables (tomato in this particular case).

Sumanta and Srimanta *et al.* 2016 ^[120], reported the order of heavy metal the vegetables Tomato Pb > Mn > Zn > Cr > Cd > Cu Brinjal Zn > Pb > Mn > Cr > Cd > Cu Spinach Mn > Pb > Zn > Cd > Cr > Cu Pea Pb > Mn > Zn > Cr > Cd > Cu Carrot Mn > Pb > Zn > Cr > Cd > Cu Cauliflower Mn > Pb > Zn > Cr > Cd > Cu. The mean concentrations of Cr and Pb in all vegetables, *Amaranthus viridis* (AV) (1.1 µg/g, 2.56 µg/g), *Spinacia oleracea* (0.31 µg/g, 2.78 µg/g), *Trigonella Foeniculum* (1.1 µg/g, 1.55 µg/g) and *Coriandrum Sativum* (1.1 µg/g, 2.49 µg/g) were higher than the values 0.38 µg/g for Cr and 0.47 µg/g for Pb obtained from a suburban area of Zhengzhou city, Henan Province, China and 78.02 µg/g for Cr and 63.1 µg/g for Pb reported in radish collected from treated wastewater irrigated suburban area of Titagarh (Gupta *et al.*, 2012) ^[41]. Derakhshan *et al.* 2016 ^[29] in Iran, reported the mean concentrations of metals Zn, Cu, Pb, and Cd in vegetables as 59, 51, 0.17 and 0.032 mg/kg, respectively. All vegetable samples had less mean contents of Pb, Cd, Zn and Cu than maximum acceptable standard levels. The highest mean levels of Pb, Cd, Zn and Cu were found in basil, Varamin lettuce, purslane and tarragon, respectively. Their lowest mean levels of Pb was detected in Jahrom lettuce.

Mohajer *et al.* 2012^[85] reported that Pb concentration in 75% of the vegetables marketed in Isfahan, Iran exceeded the acceptable level (0.3 mg/kg). Also, reported that Cd content in fresh vegetables of Tehran, Iran, was as high as 0.14 mg/kg which was more than the maximum acceptable level (0.1 mg/kg). According to a survey by Nazemi and Khosravi (2011)^[89], concentration of Pb, Cd, and Zn in vegetable samples of Shahroud, Iran was stated as 23.99, 2.09, and 168.4 mg/kg, respectively which indicated that the concentration of Cd and Pb was higher than the standard level. In the study conducted by Sharma *et al.* (2009)^[68] the mean concentration of heavy metals of Zn, Cu, Cd, and Pb was reported to be 29.6 - 45.5, 9.5 - 25.6, 0.5 - 1.5, 0.3 - 1.4 mg/kg in vegetable.

Ali and Khairia, 2012^[6] examined the concentration of some heavy metals Fe, Mn, Cu, Zn, Pb, Cd and Hg in various vegetables, fruits and cereals in Kingdom of Saudi Arabia using AAS. The obtained result shows that the concentrations of major studied metals were exceeding than the recommended maximum acceptable levels proposed by the Joint FAO/WHO Expert Committee on Food Additives. Leafy vegetables were found to contain the highest metals values especially *parsley* (543.2 and 0.0481 g/g for Fe and Hg, respectively), *Jews mallow* (94.12 and 33.221 g/g for Mn and Zn, respectively), and *spinach* (4.131 g/g for Cd). While peas in legumes group maintained the highest Zn content 71.771 g/g and cucumber had the highest Pb content 6.981 g/g on dry matter basis. High concentrations of heavy metals in different parts of the vegetables might be related to their concentration in the polluted air with industrial activities especially in middle and eastern districts.

Arora *et al.* 2008^[9] carried out a study to assess levels of different heavy metals like Fe, Mn, Cu and Zn, in vegetables irrigated with water from different sources in Rajasthan. The results indicated a substantial build-up of heavy metals in vegetables irrigated with wastewater. The range of various metals in wastewater irrigated plants was 116 – 378, 12 – 69, 5.2 – 16.8 and 22 – 46 mg/kg for Fe, Mn, Cu and Zn, respectively. The highest mean levels of Fe and Mn were detected in mint and spinach, whereas the levels of Cu and Zn were highest in carrot. Pathak *et al.* 2012^[100] reported that concentration of Pb was maximum (57.99±1.54 mg/kg) in roots, Cu (33.91±2.13 mg/kg) in the leaves; Zn (81.70±2.99 mg/kg) in roots, Ni (86.10±3.19 mg/kg) in stem; Cd (20.39±1.99 mg/kg) and Cr (76.78±3.04 mg/kg) in leaves of *A. esculentus* in Dehradu city China.

2.2 Sources of heavy metals

Heavy metals are natural constituents of the earth's crust and are persistent environmental contaminants; they are not degradable and enter the body through food, air, and water and bioaccumulate over a period of time. (Mafuyai *et al.*, 2014)^[77]. Heavy metals enter the environment through natural sources (e.g. volcanic-emissions, erosion of soils, and weathering of parent rocks) and human practices (e.g., agricultural activities, manufacturing, mining etc). Waste water is one the primary source of heavy metals of environmental concern on a global scale particularly in developing countries. Some metals are left tailing during sewage is disposed and scattered in open and partially covered pits, while some are transported through wind creating a range of environmental contamination on crops (Zhou, *et al.*, 2016)^[135]. The results of the metal content in vegetables grown on the farm in the study spinach showed bio-accumulation of the metals Fe, Mn, Zn, Cd, Ni and Pb in the order Fe > Mn > Zn > Cd > Ni > Pb. This could be a reflection of the concentrations of the metals in the soil. Leafy vegetables have been shown to accumulate relatively higher concentrations

of heavy metals than fruit vegetables. Agricultural practices are also considered to elevate the contamination of crops during application of fertilizers, pesticides, composting and manuring. The elements (Pb, Cd, As, Cr and Cd) are contained in phosphatic fertilizer in which Cd is a major cause of concern due to its high mobility and tendency to readily accumulate in vegetables in large quantity without exhibiting any phytotoxic effects (Mafuyai *et al.*, 2020a)^[79].

However, one the major sources of vegetable contamination with heavy metals might be due to the waste water irrigation, solid waste disposal, sludge applications, vehicular exhaust and agrochemicals. Excessive accumulation of heavy metals in agricultural soils through the use of agrochemicals and by other sources may lead to elevated heavy metal up-take by vegetables and thus affect food quality and safety.

2.3 Factors influencing uptake of heavy metals by plants

Heavy metals in the soil are derived from natural components or geological sources as well as from human activities or anthropogenic sources. Normally heavy metals in soil are found in several forms. As noted by Tangahu *et al.* 2011^[125] plants have evolved specific mechanisms through which they translocate as well as store micronutrients. By using the same mechanism, heavy metals and micro nutrients are taken by the plants as they have chemical properties which simulate those of useful elements. The conversion of immobile or non-bioavailable forms of heavy metals to mobile or bioavailable forms is dynamic phenomenon in the soil and occurring continually is regulated by physical, chemical and biological processes and interactions between them. However, it is important to note that the rate of accumulation of metals in plants is dependent on the plant species, type of the soil, growth stage, environmental and weather condition (Singh *et al.*, 2012; Saglam, 2013)^[111]. Vegetables take up and accumulate toxic metals in their edible and non-edible parts not only through the root system from the soil, but also through aerial deposition of contaminated dust from the air (Li *et al.*, 2004). The uptake of metals by plants depends on soil properties and various physiologic-factors of the plant. These factors bring considerable uncertainties to estimating potential doses through oral intake compared to other exposure pathways such as soil ingestion and dust inhalation (Mwegoha and. Kihampa, 2010; Tangahu *et al.*, 2011)^[125].

pH: this is the major factor that influence the solubility of heavy metals in soil for plant uptake. The solubility of metals decreases at high pH and increases at low pH value (Sheoran *et al.*, 2016)^[114]. The pH decrease may be caused, on the one hand, by a release of exchangeable cations during a process of an organic material mineralization brought by wastewater (Kiziloglu *et al.*, 2008)^[66]. This happen due to charge in surface and adsorption of solutes by variable charged soil components like, layer silicates clay, organic matter and oxides of iron and Aluminum. High pH increases the net negative charge (cation exchange capacity), whilst low pH increases net positive charge (anion exchange capacity) (Bhargava *et al.*, 2012)^[21].

Redox Potential determines the tendency of the soil solution to accept or donate the electron, metals are present in their ionic forms in soil solution and their mobility from soil to plants is depended on their oxidation state. Cr which reduced form is Cr³⁺ is quite insoluble in water, while the oxidized form Cr⁶⁺ is highly soluble and readily available for plant uptake (Sheoran *et al.*, 2016)^[114].

Organic matter which is made up of humic acid that has high molecular weight soluble in acid having the tendency to be

removed from solution via precipitation and non-humic substances (fulvic acid) has lower molecular weight soluble at all pH with more active sites. The mechanism involve in heavy metal retention in organic matter are complexation and adsorptive. Other factors that influence the uptake of heavy metals important for noting are temperature, soil texture and cation exchange capacity and selective uptake of ion or diffusion (Mafuyai, 2020b)^[81].

Therefore, the absorption of heavy metals by plants from soil is in response to concentration gradient. Roots play important role in the uptake of ions on the root surface in cationic form with negative cell wall due to the presence of cellulose, pectin and glycoproteins (Patrick, 2006)^[101].

2.4 Contamination of soil and vegetables by heavy metals

There are many sources of trace metals contaminants that can be accumulated in soils and vegetables. The main source of heavy metals in vegetables is the growth media (soil, air, nutrient solutions) from which they are taken up through the roots and foliage. Cultivation areas near highways are also exposed to aerial deposition in the form of metal-containing aerosols which are absorbed by vegetables. This deposition could occur on leaves and fruits and then absorbed in the plant body (Arora *et al.*, 2008; Yusuf and Oluwale, 2009; Iniobong *et al.*, 2012^[9, 132, 50]). The burning of fossil fuels, smelting and other processing techniques release into the atmosphere can be carried for miles and later deposited on the vegetation and soil. Lead, nickel and boron are gasoline additives that are released into the atmosphere and carried to the soil through rain and snow (Holmgren *et al.*, 1993; Igwe *et al.*, 2005)^[45, 48]. Vegetables are an important part of human's diet. In addition to a potential source of important nutrients, vegetables constitute important functional food components by contributing protein, vitamins, iron and calcium which have marked health effects (Arai, 2002)^[3]. Vegetables, especially those of leafy vegetables grown in heavy metals contaminated soils, accumulate higher amounts of metals than those grown in uncontaminated soils because of the fact that they absorb these metals through their leaves (Al Jassir *et al.*, 2005)^[2]. Consumer perception of better quality vegetables is subjective as they consider dark green and big leaves as characteristics of good quality. However, the external morphology of vegetables cannot guarantee wholesomeness because heavy metals rank high amongst the major contaminants of leafy vegetables (Mapanda, *et al.*, 2005)^[83].

The entry of heavy metals from the surface of colloids and clay mineral particles is increased at low pH range by the process of desorption, this increases the available contaminants for plant uptake resulting in concentration of toxic elements (Sheoran *et al.*, 2016)^[114]. Vegetables take up metals by absorbing them from contaminated soil as well as from deposits on different parts of the vegetables exposed to the air from polluted environments (Sobukola, 2003)^[118]. Vegetable plants growing on heavy metal contaminated medium can accumulate high concentrations of trace elements to cause serious health risk to consumers (Long *et al.*, 2010)^[76].

Each anthropogenic activity is associated with some specific heavy metals. Cement industry contributes Pb, Cd, Cu, Cr and Zn in the atmosphere, while Ni, Co, Pb and Cu are used as catalyst, modifiers and dryers (Jan, *et al.*, 2010)^[56]. Gasoline related sources are major contributors in the elevation of Pb, Cd, Cu, Zn and Ni on top soil of roadsides which easily contaminate vegetable plants (Pulles *et al.*, 2012)^[105]. Solid waste which include electronic waste like batteries, painting waste and electroplating waste in dumpsites and leachate produced in

dumpsites are associated with rainfall by slow leaching of heavy metals (Cd, Cr, Cu, Fe, Pb, Mn and Zn) from waste dumpsites that infiltrates and result in the movement into groundwater that contaminate food crops. Industrial effluents, solid waste and smoke released into the soil, air and water, farms near the areas are vulnerable to the heavy metal contamination due to discharge of untreated /poor treated effluents and disposal of wastes.

Despite the physiological characteristics of plants, the heavy metal content in cultures depends also on the physical and chemical nature of the soil, which controls the bioavailability of trace elements. This later depends on many facts like: soil temperature, moisture, organic matter, pH, and nitrogen availability (Sharma *et al.*, 2007; Qishlaqi *et al.*, 2008)^[113, 107]. Many investigations have denoted that a high content of trace elements in the soil does not necessarily reflect their high concentration in cultures. Murtaza *et al.* (2008)^[87] have found that cultivated plants in soils, consisting of a content of Cd inferior to the threshold values, may accumulate this metal with a very important concentrations exceeding the allowed levels. According to Kabata-Pendias (2004)^[58], the soil-plant transfer of trace elements involves the interaction of many parameters: the plants features, features of the soil, the nature of contamination and the environmental facts, etc.

2.5 Toxicity of heavy metals

Heavy metals are considered as one of the most significant environmental concerns because of their toxicity and accumulation in the tissues of living organisms which even at low levels can endanger human health (Arora *et al.*, 2008)^[9]. Heavy metals are those metals which have a specific density of more than 5 g/cm³ and harmfully affect the living organisms as well as environment and living organisms (Jaishankar *et al.*, 2014)^[54]. These metals are essential to maintain the normal body physiology and functioning when present in very low concentrations. However, they become lethal when certain threshold levels exceed (Ugonna *et al.*, 2020)^[128].

Many studies have been conducted throughout the world in relation to plants and soil pollution with heavy metals through irrigation by urban and industrial effluents. Some metal ions like cadmium, lead, As, Hg and chromium have toxic effects on biochemical reactions in our body (Fariha *et al.*, 2018). Heavy metals can be harmful due to their potential accumulation in different tissues of humans. Even at low concentrations, heavy metals have detrimental health effects, because they are non-biodegradable and persistent in nature (Mafuyai, *et al.*, 2019). The health risks of heavy metals through consumption of vegetables from these vegetables are of great concern in the study. The existence of heavy metals in the food chains and their critical concentration can have adverse metabolic and physiological effects on human body (Fariha *et al.*, 2018). The absorption of metals can be affected by several factors such as pH, ionic concentration of the solution, cationic concentration of metal, the presence of competitive metal cations, and organic and inorganic ligands (Gupta *et al.*, 2012)^[41]. Moreover, the shapes and different species of plants can create differences in their ability to absorb and accumulate heavy metals (Nazemi and Khosravi, 2011)^[89].

Lead (Pb) is one of the most known toxic heavy metal that can be great danger to human health because hematopoietic system, nervous system and kidneys are sensitive to it. Toxicological review of Pb has shown inhibition of the activity of *d*-aminolaevulinic dehydratase (prophobilinogen synthase, one of the major enzymes involved in the biosynthesis of heme) and

developmental problems such as impaired cognitive function, behavioral disorder, stunted growth, and impaired hearing at blood lead level as low as $5 \mu\text{g/l}$ (Ihedioha *et al.*, 2017; WHO, 2006)^[49, 131]. Lead also interferes with calcium metabolism, both directly and by interfering with vitamin D metabolism; it concentrated largely in bones in animals and humans and interferes with the normal maturation of erythroid elements in the bone marrow (Plant *et al.*, 2000)^[102]. These effects have been observed in children at blood lead levels ranging from 12 to $120 \mu\text{g/l}$. Lead is toxic to both the central and peripheral nervous systems, inducing subencephalopathic neurological and behavioral effects. The industrial use of Pb is the main cause of environmental pollution by this metal in vegetables (Mafuyai, *et al.*, 2014)^[77]. The adverse effect of Pb is dose and exposure duration dependent, in other words more than the threshold of 0.3 mg/kg for food could be dangerous (Pachathundikandi and Varghese, 2006)^[97].

Cadmium (Cd) is another toxic heavy metal that can lead to kidney damage, high blood pressure, nervous system disorders, anemia, emphysema, anosmia (loss of sense and smell) and carcinogenesis (Mohajer *et al.*, 2012)^[85]. Also, it has been stated that the weekly allowable amount of Cd uptake is $0.4\text{--}0.6 \text{ mg/person}$ (Asadi *et al.*, 1993)^[10]. It is also clear that Pb and Cd are listed as priority hazardous elements and are considered as two of the top 20 contaminants, ranked 2nd and 7th, respectively, by the US-EPA ATSDR, 2012). Pb and Cd are persistent in the environment, and are not removed by normal cropping practices nor easily leached by rainwater because of their strong affinities with the soil solid phase. The toxic heavy metals such as Pb and Cd can have severe impacts on biological processes ranging from microbial activities to primary production of plants (Khan *et al.*, 2016)^[63].

Furthermore, copper (Cu) naturally presented in vegetables can be contaminated after contact with and therefore, it accumulates in the tissues of plants. The most important sources of Cu for contamination of the vegetables are mining activities, agriculture, waste and sludge from wastewater treatment. The small amount of Cu is essential for humans, but if its value increases, it is dangerous to human health. In humans, the maximum allowed daily intake of Cu for adults is 0.9 mg/day (Jaishanka *et al.*, 2014)^[54]. High intake of Cu can cause poisoning, hypotension, jaundice, liver problems, hepatolenticular degeneration with progressive impairment of Cu-laden tissues until death results (APHA, 1999; Asadi *et al.*, 1993)^[8, 10]. Contamination and pollution of vegetable and soils near smelters does occur, and excessive Cu in drinking water has been reported to have caused a toxic syndrome in an infant called pink disease (Plant and Thornton, 1983)^[103].

The toxic effect of Cr is on the respiratory track, these include irritation of the lining of the nose, runny nose, and breathing problems (asthma, cough, shortness of breath, wheezing). Skin rashes and breathing difficulties is a major problem. Cr (VI) causes anemia, ulcer and irritation in stomach. It also destroys male reproductive system and sperm producing ability. Lung and intestinal track cancer, stomach tumors proved to cause by Cr (VI) exposure. (ATSDR). Adverse effects of Mn can result from both deficiency and overexposure, causing neurological effects due to high level in drinking water, which can cause tremor, gait disorders (seen in primate), psychological symptoms such as irritability, and emotional liability^[80]. For instance, amyotrophic lateral sclerosis, according to Hubbs-Tait *et al.* 2005^[46], is a progressive neurological disorder, which appears to be a disease that may reflect a deficiency of Mg or Mn. The decreased intake of Mg or Mn leads to a decreased ability to

store and use thiamin (vitamin B₁). These authors further noted that there is some evidence suggesting that Parkinson's disease may be casually related to Mn, but in this case, to an excess of it (Ugonna *et al.*, 2020)^[128].

Zinc (Zn) is the other metal that is widely distributed in the environment so that it is available in most foods, water, and air. Due to its effect on the activity of enzymes and protein production, Zn is an essential element for human life (Nazemi and Khosravi, 2011)^[89]. However, intake of this element more than needed could be harmful for health. The recommended dietary allowance for Zn intake for men and women is 11 and 8 mg/day, respectively (Grosell *et al.*, 2006)^[40].

Nickel is an essential trace element in animals. Some of its health risk includes fibrosis, chronic bronchitis, impaired pulmonary function, and emphysema. Allergic contact dermatitis is the most prevalent effect of toxicity of nickel in the general population (APHA, 1999; Plant *et al.*, 2000)^[8, 102]. However, it is suspected to be an essential element for some plants and animals. According to Plant and Thornton and Carla, Ni deficiency results in decreased plasma cholesterol, increased liver cholesterol, ultra structural changes in the liver calls, rough hair, impaired reproduction, and poor growth of the offspring (Plant and Thornton, 1983)^[103].

Arsenic is a metalloid whose chronic exposure effects include tingling, numbness, and peripheral neuropathy according to Plant and Thornton, 1983^[103]. These authors also argued that arsenic toxicity in cattle has been found to cause dysentery and respiratory distress. An ecological correlation between the arsenic level of well water and mortality from various malignant neoplasm in China (Province of Taiwan) demonstrated a significant association with the arsenic level in well water ranging from 0.35 to 1.14 mg/l with a median of 0.78 mg/l for cancer of the liver, nasal cavity, lung, skin, bladder, and kidney and hyperpigmentation, hyperkeratosis, Blackfoot disease (a type of gangrene) in both males and females, and prostate cancer in males (Chen and Wang, 1990)^[27].

2.6 The challenges

Food security and safety is of great concern globally due to the toxic effect of heavy metal contamination and their associated health risks (Shaheen *et al.*, 2016; Chen *et al.*, 2016; Yousaf *et al.*, 2016)^[112, 26, 134]. The impact of Pb and Cd on human health has long been and continues to be of great concern, particularly for infants and children (Cao *et al.*, 2016; Pan *et al.*, 2016)^[24, 98]. Thus, long-term consumption of these foods is of public health significance. There is the need for better quality control for food crops to protect consumers from contamination exposure. In addition, close monitoring of the significant possibility of significant harm of these heavy metals on the health of the population in the world is essential. It is also important to use remediation measures in contaminated areas where local foods are grown or produced. In view of the implications of heavy metals in vegetables irrigated with wastewater, some researchers showed insufficient fundamental discussions on the understanding of the true nature of the health risk to inform practical and implementable decision and policy. It therefore, shows that a global knowledge base is lacking. This is a major limitation, which possess potential constraints on insight for future research focus majorly in developing countries. In addition, this review shows that more scientific techniques for mitigating the impacts of these heavy metals' contamination from wastewater irrigation are not intensified globally, although research documents a few primordial and conventional toxicity reduction techniques in wastewater treatment. The spatial

distribution of these toxicities in vegetables is the major issues that suggest the need for further research, which should map and delineate spatially the variations in heavy metal toxicity. Despite the varying dangerous risks on health and environment, direct or indirect use of wastewater in agricultural irrigation in many developing countries is still planned. Also, despite the general low concentration of trace elements in the treated effluents, the impact of this water however, remains a major concern to the environment. Since trace elements generally accumulate in the soil, and because of the biochemical conditions, they pass into the soil solution. They are more or less absorbed by plants and affect the quality of the agricultural products intended for both humans and animals which causes serious health problems.

3. Conclusion

In developing countries, fields and gardens bordering the different conurbations are irrigated with urban wastewater containing relatively weak trace elements content. Heavy metal poisoning of vegetables as a result of wastewater irrigation has stimulated increasing research interests within the field of environmental chemistry and toxicology. Earth materials comprise an abundance of heavy metals that support the ecosystem processes. Several cases of human health impacts have resulted because of bioavailability, bioaccessibility, and excessive concentrations and distribution of contaminants in vegetables. These heavy metals in wastewater contaminate most of the soils, farmlands, and aquatic habitats even though made nutrient available plants.

1. Based on literature thus reviewed, it can be concluded that some industrial effluents hold great promise for the improvement of soil fertility and crop yields, if proper treatment and management practices are adopted.
2. Literature confirms that the Wastewater can effectively increase water resource for irrigation but there is a need for regular monitoring of the heavy-metal and other pollution related parameter concentrations in soil, plants and ground water.
3. Since Wastewater is rich source of micro-nutrients and heavy-metals, the repeated application of Wastes water may accumulate appreciable quantities of heavy-metals and micro-nutrients in the soil. Therefore, such fields need periodic estimates to monitor the build-up of these nutrients.
4. Literature reviewed revealed that safe utilization of Wastewater for irrigation to crops requires several precautionary measures *viz.* treatment, adequate dilution, selection of crop etc. Besides, soil physical properties needs to be reviewed periodically for long-term sustainability of the system.
5. Based on the literature, thus reviewed, it can be concluded that proper management of Wastewater irrigation, periodic monitoring and crop quality are required to ensure successful, safe and long-term use of Wastewater for irrigation.
6. It is desirable that highly contaminated Wastewater from some industries undergo rigorous and suitable treatment in Wastewater treatment plants, before use for irrigation purpose.

Despite the varying dangerous risks on health and environment, direct or indirect use of wastewater for irrigation of vegetables farms in many developing countries is still unplanned. Even at low concentrations, heavy metals have been reported to produce damaging effects on man and animals because there is no good mechanism for their elimination from the body (Arora *et al.*,

2008; Yusuf *et al.*, 2009) [9, 133]. Also, despite the general low contribution of trace elements in the treated effluents, the impact of this water however, remains a major concern to the environment. Since trace elements generally accumulate in the soil and because of the biochemical conditions. They are more or less absorbed by plants and affect the quality of the agricultural products intended for both humans and animals which causes serious health problems. This review was conducted to demonstrate the effect of trace elements resulting from wastewater irrigation and their contamination of vegetable plants which represent not only a significant environmental risk, but also a potential health risk.

4. References

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