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Heavy metal contamination in selected cruciferous vegetables grown in Jos, Nigeria

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Abstract

The levels of different heavy metals (As, Cd, Hg, Pb, Sn, Cr and Cu) were determined in six cruciferous vegetables: white cabbage (*Brassica oleracea* var. *capitata*); broccoli (*Brassica oleracea* var. *italica*); cauliflower (*Brassica oleracea* var. *botrytis*); red cabbage (*Brassica oleracea* var. *rubra*); curly kale (*Brassica oleracea* var. *sabellica*); and radish (*Raphanus sativus*) commonly grown and locally consumed in Jos, Plateau State of Nigeria. These vegetables are consumed raw, steamed or cooked. Several cultivars of each vegetable were collected during harvest, processed, digested and analysed for their content of heavy metals using atomic absorption spectrometry (AAS). The results showed that the mean-concentrations of the heavy metals in the vegetables were in the following ranges: As 0.552-3.010 mg/kg; Cd 0.009-0.064 mg/kg; Hg 0.001-0.057 mg/kg; Pb 0.331-0.451 mg/kg; Cr 0.251-0.529 mg/kg; and Cu 1.892-3.510 mg/kg. The concentrations of the metals in the samples were compared with WHO/FAO permissible limits. The vegetables were most contaminated by Cr followed by Pb, As and Cd, but showed no contamination by Hg, Cu and Sn. The results further showed that the public is at risk of Cr, Pb, As and Cd toxicities on consumption of cruciferous vegetables grown in the study area.

Keywords: Cruciferous vegetables, heavy metals, concentration, contamination.

I. Introduction

Heavy metals, according to chemical properties, are elements with metallic properties and are described based on their density, atomic number or atomic weight, chemical property and/or toxicity (Taghipour and Mosaferi, 2013). They can also be seen as a heterogeneous group of elements which vary widely in their chemical property and biological importance. The term heavy metals can be defined as a metal which has a specific gravity (or density) more than 5g/cm^3 and an atomic number greater than 20. The term (heavy metals) has been regarded as misinterpretation in a technical report of the International Union of Pure and Applied Chemistry (IUPAC) because of its contradictory description and lack of coherent scientific basis. Hence, the term "toxic metals" is more preferable even though there is no consensus of exact definition for it (Hezbullah *et. al.*, 2016).

Heavy metals do not degrade and cannot be broken down into less toxic constituents. They accumulate where they are deposited or released. In other words, they are non-biodegradable and non-thermo degradable (Li *et. al.*, 2016; Hezbullah *et. al.*, 2016). Thus, they persist and accumulate to toxic levels (Shamar *et. al.*, 2005) in environmental and biological matrices. Some are known to play biologically important roles while others do not have any known biological value (role) (Taghipour and Mosaferi, 2013;

Dabak *et. al*., 2013).Heavy metals can be classified into four groups on the basis of their biological importance: essential heavy metals (e.g. Cu, Zn, Fe, Co) which are toxic only at significantly high concentrations; non-essential heavy metals (e.g. Ba, Al, Li); less toxic heavy metals (e.g. Sn, Al); and highly toxic heavy metals (e.g. Pb, As, Cd, Hg) which are toxic even at low concentrations (Hezbullah *et. al.*, 2016).

Living organisms require optimum concentrations of trace metallic elements of biological importance such as Na, Ca, Mg, Zn, Cu, Cr, Co, Se, etc. for normal physiologic functioning. High concentrations of these elements can be toxic to the organism−humans and other animals (Brandl, 2012; Dabak *et. al.*, 2013). Other elements such as Pb, Cd, Hg, Au, As, Sn, etc. have no biological significance and prolonged exposure to them leads to bio-accumulation that causes serious health problems over time (Sharma *et. al.*, 2009; Guerra *et. al.*, 2012). Toxicity of these toxic, non-essential heavy metallic elements is explicable to their ability to displace essential metals (of biological importance) from their binding sites or forming complexes with them (formation of ligands). This leads to damaging of cell membranes, altering enzyme activities, disrupting cellular functioning and damaging of DNA structure (Brandl, 2012; Dabak *et. al.*, 2013). Heavy metal contamination of food has become an important aspect of food quality assurance in recent decades (Sharma *et. al.*, 2009; Shuaibu *et. al.*, 2013). This is because of inefficiency of effective detoxification and excretion mechanisms for such heavy metals (Chen *et. al.*, 2013; Hezbullah *et. al.*, 2016) and ingestion of food stuff, including vegetables, has become one of the major ways of entry of these metals into the human and animal systems (Guerra *et. al.*, 2012).

The source of heavy metals is not only the bedrock itself, but also anthropogenic sources including solid and liquid waste deposits, agricultural inputs (metallo-organic herbicides, pesticides, insecticides and fertilisers) and fallouts of industrial and urban emissions (Morias *et. al.*, 2012; Dabak *et. al.*, 2013; Ifenkwe *et. al.*, 2015; Hezbullah *et. al.*, 2016) and another salient source is atmospheric deposition (Chary *et. al.*, 2008; Guerra *et. al.*, 2012). Human activities such as mining, industrial processes such as battery and car manufacturing, agricultural processes (such as waste-water irrigation), breaking (weathering) of rocks and construction have increased the chances and risk of exposure to these heavy metals (Morias *et. al.*, 2012; Czech *et. al.*, 2012). Disposing of industrial wastes into streams and rivers along which farmlands are located and whose water is used for irrigation has added to the menace (Singh *et. al.*, 2010; Nazemi, 2012). Heavy metals contaminate vegetables when they occur in concentrations that are above permissible limits. Vegetables take up heavy metals and bio-accumulate them in their edible and non-edible parts in concentrations high enough to cause clinical problems in humans as well as animals (Brandl, 2015; Hezbullah *et. al.*, 2016).

Cruciferous vegetables have been found to be chemo-preventive against cancer (Soni and Kohli, 2015; Kim, 2016), anti-microbial (Dias *et. al.*, 2014) and contain ample amounts of anti-oxidants. These properties of this class of vegetable are attributed to a distinct class of phytochemicals, glucosinolates (Soni and Kohli, 2015) that are unique to them. These glucosinolates are hydrolysed to, among other hydrolysis products, isothiocyanates and indoles (El-Sayed *et. al.*, 2015; Mizani *et. al.*, 2016) which are chemo-preventive against cancer and some other degenerative disease conditions. Cruciferous vegetables are also good sources of vitamins, minerals, dietary fibres, and other phytochemicals that function to benefit both the vegetable plants and their consumers − humans and animals (Achikanu *et. al.*, 2013; Taljera and Moon, 2014; Ramteke *et. al.*, 2016). Considering these health and nutritional benefits of cruciferous vegetables and the fact that they are a major class of vegetables consumed in large quantities worldwide and particularly in Jos, Nigeria, there is need to assess the concentrations of heavy metals in locally grown crucifers (since they are usually cultivated by irrigation using water from abandoned mining ponds) in order to ensure their safety for consumption and to advise the public accordingly in order to minimise the intake of these toxic metals and the possible health risks so associated. Also, since heavy metals are almost indispensable in nature and cruciferous vegetables are an indispensable part of human diet, there is need to regulate the quantity of daily consumption of these vegetables for their health and nutritional benefits as well as reducing the daily intake of heavy metals.

II. Materials and Methods

2.1 Reagents and materials

All reagents (Sigma-Aldrich, Britain) used were of analytical grade and were obtained at the Soil Laboratory of the Federal Ministry of Agriculture, Kaduna, Nigeria. Metal analyses were carried out using Atomic Absorption Spectrometer (AAS-PG 990). Sample preparation was done at the Department of Biochemistry, University of Jos.

2.2 Study area

Jos is a small industrialised rocky city and administrative capital of Plateau State in Central Nigeria. The city is located on coordinates $9^0$56'N, $8^0$53'E, with a population of about 1,000,000 inhabitants and enjoys a more temperate climate than much of rest of Nigeria being 4200ft (1280m) above sea level. It has an average monthly temperature ranging between 21-25^{0}C and annual rainfall of about 1400mm (55 inches) (NIMET, 2017).

Jos is known for large-scale tin mining which has left mining tailings (ponds) well scattered within Jos and its environs which serve dry-season farmers with water for irrigation. Crops mostly irrigated include Irish potatoes and vegetables (Dabak, *et. al.*, 2013).

The city also records series industrial activities. Some of these industries include the NASCO Group, Vita Foam, Nigeria Bottling Company, Grand Cereals and Oil mills, Diamond Paints and Sharon Paints among many others. Industrial wastewater from most of these factories is disposed into streams along which many irrigation farms are located. The effects of mining and industrial activities and indiscriminate disposal of industrial and urban wastewater into irrigation channels (streams) formed the basis for the choice of this study area.

2.3 Sample collection

All cruciferous vegetables used for this research were obtained during harvest in January, 2017 from several irrigation fields within Jos Metropolis. These are Farin Gada, Vom, Anglo-Jos and Foron areas which are major production sites for vegetables consumed within and outside Jos. Vegetables collected include white cabbage (*Brassica oleracea* var. *capitata*); broccoli (*Brassica oleracea* var. *italica*); cauliflower (*Brassica oleracea* var. *botrytis*); red cabbage (*Brassica oleracea* var. *rubra*); curly kale (*Brassica oleracea* var. *sabellica*); and radish (*Raphanus sativus*).

Cultivars of each vegetable were randomly collected, wrapped in polythene bags, clearly labelled and immediately taken to the laboratory for processing, digestion and subsequent analyses.

2.4 Sample preparation

Sample preparation was done according to Dabak *et. al.* (2013) with some modifications: The sample were removed from polythene bags in the laboratory, thoroughly washed under running water to completely remove particles of sand and debris. Edible portions of each vegetable were collected as composite sample, rinsed again with clean tap water and then air-dried under shade for two weeks until completely dry.

Each dried sample was ground into fine powder using Teflon mortar and pestle, sieved using muslin cloth and then kept in well-labelled, air-tight containers at room temperature until digestion and subsequent analyses.

2.5 Sample digestion

Sample digestion was done according to Dabak *et. al.* (2013) with some modifications: 0.2g of sieved sample was accurately weighed into a beaker and 30ml of digestion mixture, ($HClO_4$:H_2SO_4:HNO_3) in the ratio 1:4:33, was added. The beaker, containing the sample was placed on a hot plate in a fume cupboard and the mixture was gently heated at 150^{o}C until mixture boiled. The heating continued till the brown fumes disappeared leaving white dense fumes and the content reduced to about 5ml. This was then cooled, transferred into a 50ml volumetric flask and made up to mark with distilled water. The digest was stored at room temperature until analyses of metals.

2.6 Metal analyses

The concentrations of the metals: As, Cd, Hg, Pb, Cr, Cu and Sn were analysed in the sample digests using Atomic Absorption Spectrometer (AAS-PG 990) at the Soil Laboratory of the Federal Ministry of Agriculture, Kaduna-Nigeria.

2.7 Statistical analysis

Mean values of the concentrations of each metal in individual samples were computed from three replicates each. One-way ANOVA was used to analyse the difference in the concentration of each heavy metal across the various cruciferous vegetables and p-value less than or equal to 0.05 was considered to be significant. Chi-square was used to determine the significant levels ($P \leq 0.05$) of each metal in the vegetables as compared to WHO/FAO maximum safety limit (maximum permitted level) for each metal. Calculations were done using the statistical software SPSS V17.5.

III. Results

Table 1 below shows the concentrations of As, Cd, Hg, Pb, Cr, Cu and Sn in the various selected cruciferous vegetables obtained within the study area. Sn was below detectable levels in all vegetables. The concentration of As ranged between 0.552-3.010 mg/kg; Cd 0.009-0.064 mg/kg; Hg 0.001-0.057 mg/kg; Pb 0.331-0.451 mg/kg; Cr 0.251-0.529 mg/kg; and Cu 1.892-3.510 mg/kg.

Table 1: Concentration of As in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	As
1.	White cabbage	3.010±0.01
2.	Broccoli	1.010±0.01
3.	Cauliflower	0.781±0.03
4.	Red cabbage	0.591±0.05
5.	Curly kale	0.611±0.01
6.	White radish	0.552±0.02
	WHO/FAO	0.500*****

All values are expressed as mean±SD (n=3); *****Codex Alimentarius Commission's CODEX STAN 193-1995 as amended 2011 and 2015

There is significant difference (P≤0.05) in the concentration of As across the vegetables and all have concentrations significantly higher than WHO/FAO maximum limit.

Table 2: Concentration of Cd in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	Cd
1.	White cabbage	0.016±0.15
2.	Broccoli	0.009±0.01
3.	Cauliflower	0.064±0.02
4.	Red cabbage	0.054±0.01
5.	Curly kale	0.061±0.03
6.	White radish	0.031±0.01
	WHO/FAO	0.050*****

All values are expressed as mean±SD (n=3); *****Codex Alimentarius Commission's CODEX STAN 193-1995 as amended 2011 and 2015

There is significant difference (P≤0.05) in the concentration of Cd across the vegetables. Cauliflower, red cabbage and curly kale have concentrations higher than WHO/FAO safe limit.

Table 3: Concentrations of Hg in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	Hg
1.	White cabbage	0.016±0.01
2.	Broccoli	0.011±0.01
3.	Cauliflower	0.052±0.02
4.	Red cabbage	0.001±0.00
5.	Curly kale	0.031±0.01
6.	White radish	0.057±0.02
	WHO/FAO	0.100

All values are expressed as mean±SD (n=3); *****Codex Alimentarius Commission's CODEX STAN 193-1995 as amended 2011 and 2015

There is significant difference (P≤0.05) in the concentration of Hg across the vegetables. All have concentrations below WHO/FAO maximum limit

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Table 4: Concentrations of Pb in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	Pb
1.	White cabbage	0.451±0.04
2.	Broccoli	0.415±0.01
3.	Cauliflower	0.394±0.02
4.	Red cabbage	0.344±0.04
5.	Curly kale	0.331±0.03
6.	White radish	0.451±0.02
	WHO/FAO	0.100*

All values are expressed as mean±SD (n=3); *Codex Alimentarius Commission's CODEX STAN 193-1995 as amended 2011 and 2015

There is significant difference (P≤0.05) in the concentration of Pb across the vegetables and all have concentrations higher than WHO/FAO maximum permissible limit.

Table 5: Concentration of Cr in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	Cr
1.	White cabbage	0.323±0.01
2.	Broccoli	0.384±0.03
3.	Cauliflower	0.529±0.02
4.	Red cabbage	0.491±0.02
5.	Curly kale	0.511±0.01
6.	White radish	0.251±0.05
	WHO/FAO	0.050*

All values are expressed as mean±SD (n=3); *Nazemi, 2012; Dabak *et. al.*, 2013

There is significant difference (P≤0.05) in the concentration of Cr across the vegetables and all have concentrations higher than WHO/FAO permissible limit

Table 6: Concentrations of Cu in the Cruciferous Vegetables in mg/kg

S/N	Vegetable	Cu
1.	White cabbage	2.250±0.05
2.	Broccoli	3.510±0.01
3.	Cauliflower	3.200±0.01
4.	Red cabbage	2.952±0.03
5.	Curly kale	1.892±0.01
6.	White radish	2.510±0.01
	WHO/FAO	40.000*

All values are expressed as mean±SD (n=3); *Bvenura and Afolayan, 2012

There is significant difference (P≤0.05) in the concentration of Cu across the vegetables and all have concentrations significantly lower than the WHO/FAO limit.

Tin was below the detectable level in all vegetable samples. Hence was not detected.

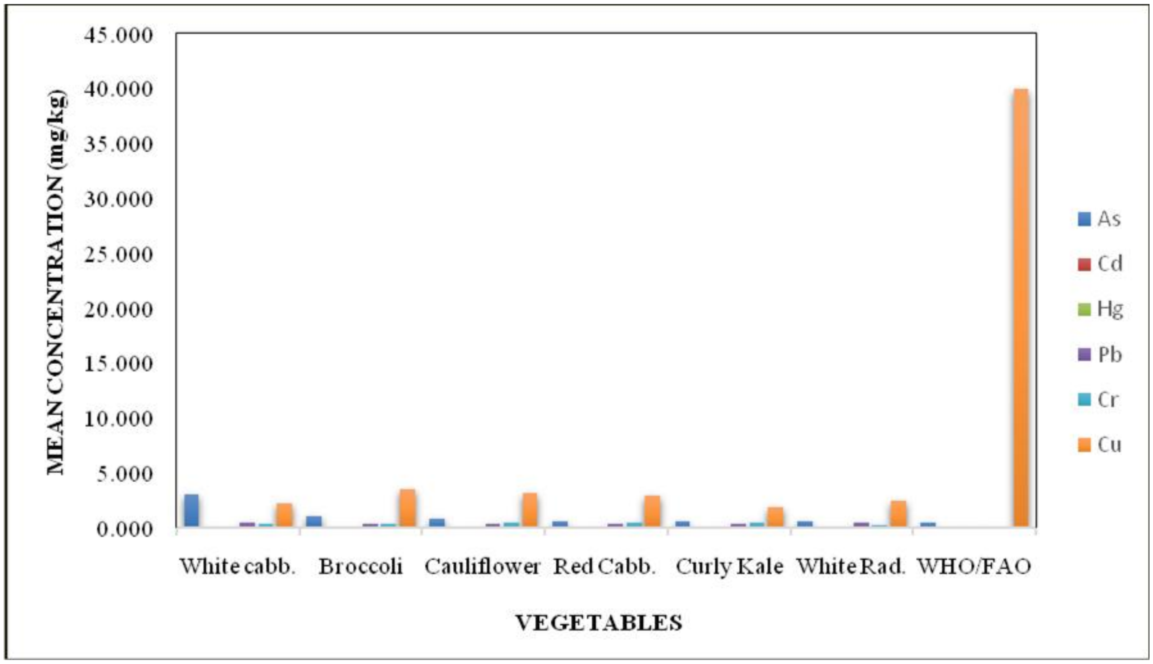

Figure 1: Bar chart showing the mean-concentrations (in mg/kg) of As, Cd, Hg, Pb, Cr and Cu in the selected cruciferous vegetable samples. At P≤0.05, there is significant difference in the concentrations of heavy metals detected in all the vegetables.

IV. Discussion

The concentration of heavy metals in vegetables is influenced by fertilisers, agrochemicals, bio-solids and manures, wastewater, metal mining processes, industrial and urban activities (Wuana and Okieimen, 2011; Taghipour and Mosaferi, 2013) which as well affect the soil concentration of heavy metals (Dabak *et. al.*, 2013; Ifenkwe *et. al.*, 2015). Also, the uptake of heavy metals by vegetables is affected by the plant species, chemical properties of the heavy metals and the concentration of the metals in the soil among other factors (Tangahu *et. al.*, 2011). Hence, low concentration of tin in the soil, its chemical property and/or state and the species of the vegetable samples possibly formed the basis for the absence of Sn in all vegetables analysed. Sn has long been reported as occurring in trace amounts in vegetables and other natural, unprocessed and unpackaged foods (De Groot *et. al.*, 1973), but is found in significant amounts mostly in foods stored or packaged in unlacquered tin cans which may account for up to 98% of all ingested Sn (Blunden and Wallace, 2003). Shin *et. al.* (2013) reported tin in significant amounts in commercial yam powder products sampled across South Korea. It can, therefore be deduced that cruciferous vegetables are generally partially or completely irresponsive to Sn and its compounds in the soil.

Suruchi and Khanna (2011) and Ogunkunle *et. al.* (2014) reported Cd concentrations of 0.064mg/kg and 0.061mg/kg respectively in cabbage which are similar to those obtained for curly kale, red cabbage, and cauliflower in this study. Also Ogunkunle *et. al.* (2014) recorded high contamination of cabbage by As (1.840mg/kg) which was far above the maximum level permitted by WHO/FAO. This was also obtained in this study. Dabak *et. al.* (2013) conducted a comparative study on heavy metal levels in vegetables obtained from four different Local Government Areas of Plateau State, Nigeria and results obtained indicated that cabbage from Jos North LGA, the present study area, had high concentrations of Cd and Cr (6.168 mg/kg and 16.280 mg/kg respectively) as obtained in this research as all samples were contaminated by Cr but only cauliflower, red cabbage and curly kale were contaminated by Cd. Oluwole *et. al.* (2013) reported that vegetables grown by roadsides can be contaminated by heavy metals which deposit from atmosphere as dust, vapour and/or particulates. Such deposits can also come from fumes of industrial and urban combustion as well as vehicular activities (Chary *et. al.*, 2008). This also adds to soil concentration of heavy metals.

All the vegetables were contaminated by As, Pb and Cr. The occurrence of arsenic in the samples was due to its presence in the soil and water which probably resulted from the interaction of water with or soil weathered from arsenic-rich rock (Ratnaike, 2003)and could also result from the use of arsenic-based herbicides and pesticides (Hezbullah *et. al.*, 2016). Also, Pb, being a functional component of paints, batteries, lead alloys and other lead- containing wares, could contaminate the soil and hence, the vegetables by indiscriminate disposal of these lead-containing substances. Lead is associated with devastating health conditions including miscarriages, low birth weights in infants, inborn defects, decreased erythrocyte life span, hypertension and cardiovascular malfunctioning (Hezbullah *et. al.*, 2016). Chromium could arise in the soil via waste containing tanned leather, chrome plates, paints, wood treatment agents, textile products, cement, antifreeze, glass and ceramics (ATSDR, 2008), plastic stabilisers, electroplated wares, and phosphate fertilisers (Ali *et. al.*, 2013). Therefore, the presence of As and Cu in the vegetables could be attributed to the use of copper and arsenic-containing pesticides and herbicides used on the farms. Pb and Cr could have come from industrial waste from the paint factories scattered within the Jos Metropolis. Pb could also have possibly come from indiscriminate disposal of lead batteries and other lead-containing materials. Possible sources of Hg and Cd contamination in the study area are phosphate fertilisers, sewage sludge, cement-containing wastes and coal and petroleum wastes.

Both Cr and Cd are associated with increased cancer risks, impaired organ functioning and death among other health challenges (Tchounwou *et. al.*, 2012; Jaishankar *et. al.*, 2014; Yang and Shu, 2015; Adal, 2016).Heavy metals are well-known to be non-biodegradable and non-thermo degradable (Li *et. al*., 2016). Therefore the menace of heavy metals can best be tackled by phytoremediation technologies: the use of selected plant species to clean up contaminants in the environment (Tangahu *et. al.*, 2011). Placek *et. al*. (2016) determined the phytoremediation potentials of three tree plants: Scots pine (*Pinus silvestris* L.), Norway spruce (*Picea abies* L.) and oak (*Quercus robur* L.) on Cd, Zn and Pb using sewage sludge as additive for supporting the process. Results from this study showed that Scots pine and Norway spruce are excellent remediators of the soil from Cd, Zn and Pb. Amuda *et. al.* (2007) used chitosan-coated, acid treated coconut shell carbon (a composite adsorbent) to adsorb heavy metals from industrial wastewater. This was effective on Zn which was readily absorbed. Khan *et. al.* (2000) reported *Mycorrhizae* fungi (*Ectomycorrhizae* and *Arbuscularmycorrhizae*) and the unique superfamily of thiol-containing metal-binding proteins called metallothioneins as important phytoremediation and phytochelation agents. Metallothioneins are also known as phytochelators and are known to modulate internal levels of metal concentrations between deficient and toxic levels by binding toxic metals. These phytochelators are found in good amounts in roots of *Brassica spp.* L., *Acer pseudoplatamus* and *S. cucubalus* which are effective on Cd, Zn and Cu. This is reflected the presence of Cu in the samples used in this study. When soil was dosed sufficiently with EDTA prior to planting, *Zea mays* showed enhanced uptake of Fe, Mn and Cu after a six-week growth (Khan *et. al.*, 2000).

V. Conclusion

When compared with the WHO/FAO permissible limits for heavy metals, the results show that the selected cruciferous vegetables were most contaminated by Cr followed by Pb, As, and then Cd. The vegetables were least contaminated by Hg and Cu, and Sn was not detected in any of the samples. It can be seen that irrigation with water contaminated by industrial and urban wastes as well as mining activities have increased the levels of heavy metals in cruciferous vegetables within the study area.

In spite of the nutritional and health benefit of these cruciferous vegetables, especially their anticancer activity which has informed their increased consumption, it is imperative to also pay attention to the safety profile of these vegetables. There is need to frequently monitor the heavy-metal profile of farm soils and irrigation water as well as proper regulation of waste disposal and treatment in order to reduce the danger posed by heavy metals. We therefore recommend that the public within the study area should reduce the intake of large quantities of cruciferous vegetables grown within the area in order to avoid the bioaccumulation of these heavy metals in the body and to prevent any associated risks.

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