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# Research Article
# Health Risk Assessment of Food Crops Fumigated with Metal Based Pesticides Grown in North-Eastern Nigeria

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## Abstract
**Background and Objective:** Heavy metals contents in food crops and their associated human health through food consumption has been a major health concern. This study was designed to determine the concentrations of heavy metals (Cd, Pb, Cr, Cu, Zn) in food crops fumigated with pesticides as the only source of contamination and their associated human health risk. **Materials and Methods:** Samples of seven selected and commonly consumed food crops and their corresponding soil were randomly collected from farmlands fumigated with pesticides. Heavy metals were measured by atomic absorption spectrometry after acid digestion. The mean difference between the heavy metal in each plant part (root, stem, leave and fruit) was determined using Analysis of variance (ANOVA) with SPSS software. Health risk assessment and pollution indices from the consumption of each plant were computed using the standard formula as described USEPA. **Results:** The concentrations of heavy metals in most of the studied crops varied significantly in different parts of the plants and for different metals. Most of the studied crops showed bioaccumulation factor >1 indicating greater absorption of metals and bioaccumulation factor values were higher for Cd compared to other metals. The Estimated Daily Intake (EDI) of metals were above the oral reference dose (RfD) in all the studied crops for Cd and in *Allium cepa*, *Zea mays* and *Oryza sativa* for Pb, while hazard index showed slightly health risk from the consumption of all metals in *Oryza sativa*. **Conclusion:** The consumption of the studied food crops fumigated with pesticides in the study area may pose a great health risk and hence regular monitoring/screening of heavy metals in pesticides should be enforced.

Key words: Bioaccumulation, heavy metals, hazard quotient, pesticides, *Allium cepa*, *Zea mays* and *Oryza sativa*

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Competing Interest: The authors have declared that no competing interest exists.

Data Availability: All relevant data are within the paper and its supporting information files.

## INTRODUCTION

Heavy metal contamination from pesticides of agricultural soil and crops is one of the serious ecological problems on a global scale, especially in developing countries like Nigeria[1]. However, there are limited studies on the health risk of heavy metals from the consumption of pesticides fumigated food crops. Most studies have attributed the sources of heavy metals in food crops from industrial irrigated wastewater and industrial contaminated sites, while sources of heavy metals from pesticides are mostly overlooked.

Agriculture in Northern Nigeria is the most fundamental form of economic activity and food crops produced in this region are usually distributed to not only the southern parts of the country but including neighboring countries like Niger, Chad, Cameroon etc. The production of chemical pesticides has been on the increase and new pesticides have been infiltrating the Nigerian markets whose chemicals contents for heavy metals and other chemicals unfriendly and toxic to the environment are not known. A survey on pesticide usage in Nigeria indicated that about 15,000 metric tons per annum of pesticides comprising about 135 pesticides chemicals are marketed locally under 200 different product brands and formulation, thus making Nigeria one of the largest pesticide users in sub-Sahara Africa[2]. These records are believed to have substantially increased as more people have gone into farming now. However, despite the beneficial contribution of pesticides in the control of pests and diseases of agricultural importance, studies have shown that they could be detrimental to the soil ecosystem and contamination of food crops through the food chain[3]. More so, studies have revealed the presence of heavy metals in pesticides even at the recommended dilution rate for farmers[4,5]. Heavy metal contents have been banned in pesticides even in developing countries like Nigeria but however, despite the ban, recent studies have revealed heavy metals in pesticides[4,5]. This is thus a case of indiscriminate and illegal infiltration and adulteration of agrochemicals and its massive usage in Nigeria.

The use of large quantities of agrochemicals such as metal based pesticides plays an important role in the contaminating different food crops[6]. Heavy metals have the ability to accumulate in food crops in a contaminated environment and may be toxic to humans, plants even at low concentrations[2]. Heavy metals contamination through the ecological food chain is the main contributing pathway accounting for more than 90% to human exposure[6,7]. Heavy metals are hazardous contaminants in food and the environment and they are non-biodegradable with long biological half-lives[8]. Previous reports showed the persistence of heavy metals in the environment in both non-biodegradable and non-thermo-degradable category which lead to accumulation readily reaching to toxic levels[9]. Excessive accumulations of heavy metals in agricultural soils through the use of agrochemicals particularly pesticides may not only result in soil contamination but also lead to elevated heavy metals uptake by crops and thus affect food quality and safety[10]. Consequently, food crops safety and its associated risk have become an important food quality attribute and a global public health issue within the last decade[11,12]. Crop contamination by heavy metals from pesticides cannot be ignored since crops are crucial to human diet as they comprise important constituents desired by human body, like carbohydrates, protein, vitamins, minerals and trace elements[13]. The aim of this research was to determine the concentrations of heavy metals (Cd, Pb, Cr, Cu, Zn) from pesticides in rain fed agricultural crops and their associated human health risk in some farms in the north eastern part of Nigeria.

## MATERIALS AND METHODS

**Study area:** The study was carried out from February, 2018 to March, 2019. Random samples of leaves, stems, roots and fruits were collected from matured seven crop plants from farms in Gombe. The sampling areas are located at 32P N0749836, 32P E11366869 and N0749941, E1134986. All sampling was replicated five times. At each sampling sites, 20 g each of the seven crops namely (*Spinacia oleracea*, *Capsium cerasiforme*, *Solanum lycopersium*, *Lactuca sativa*, *Allium cepa*, *Zea mays* and *Oryza sativa*) were collected from three different locations in each farm to provide representative samples of each crop. Samples were collected from the roots, stems, leaves and fruits of each of these crops and their corresponding soil. The crops and soil samples were collected in a clean brown envelope, labeled and transported to the laboratory of the Department of Biological Sciences, Abubakar Tafawa Balewa University Bauchi, Nigeria.

**Samples preparation:** Each plant parts were chopped into small pieces and oven-dried at 80°C, pulverized and passed through a 2 mm sieve at Biology laboratory ATBU. The resulting fine powder was kept at room temperature before analysis. Soil samples were air-dried in the laboratory, crushed and passed through a 2 mm mesh size sieve and stored at room temperature before analysis.

**Heavy metal analysis:** Samples (1 g) of both plant and soil were digested using 15 mL each of tri- acid mixture (70% high purity (Nitric acid) $HNO_3$, 65% (Perchloric acid) $HClO_4$ and 70%

(Sulfuric) $H_2SO_4$ in 5:1:1 ratio) The mixture were digested at 80°C till the solution became transparent. The resulting solution was filtered and diluted to 50 mL using deionized water and was analyzed for Cr, Cu, Cd, Zn and Pb using an atomic absorption spectrophotometer[14].

**Health risk assessment**

**Daily intake of metal:** The Daily Intake of Metals (DIM) was calculated to determine the health risk from consuming vegetables contaminated with pesticides with heavy metal contents using the formula given below[14]:

$$DIM = \frac{M \times K \times I}{W}$$

where, M is the concentration of heavy metals in plants (mg $kg^{-1}$), K is the conversion factor, I is the daily intake of vegetables and W is the average body weight

Fresh weight of vegetables was converted to dry weight using the conversion factor 0.085 as described in another study[3]. Where, the daily vegetable Intake (I) for adults in Nigeria was taken as 0.086 kg $day^{-1}$ in accordance with previous study[15], Adult body weight was taken as 60 kg.

**Hazard quotient:** The health risks to the local inhabitants from the consumption of vegetables were evaluated based on the Hazard Quotient, which is the ratio between exposure and oral reference dose (RfD).

Given as:

$$HQ = \frac{Dim}{RfD}$$

The (RfD) is an estimation of human daily exposure that is likely to pose an appreciable risk of adverse health effects during a lifetime. Oral Reference Dose (mg $kg^{-1}$ b.wt./day) for heavy metals was taken as Cr = 1.5, Cd = 0.001, Pb = 0.004, Cu = 0.04, Zn = 0.3[16].

**Hazard index:** Potential risk to human health due to more than one heavy metal known as the Hazard Index (HI) was calculated[16], which is the total sum of all the Hazard Quotients as shown in the Eq:

$$HI = \Sigma HQ = HQ_{Cd} + HQ_{Pb} + HQ_{Cr} + HQ_{Cu} + HQ_{Zn}$$

where, HI is the Hazard Index and HQ is the Hazard Quotients

The hazard index assumes that the magnitude of the adverse effect will be proportional to the sum of the multiple metal exposures.

**Estimated daily intake (EDI):** The degree of toxicity of heavy metals to human upon their daily intake (mg $kg^{-1}$/day) known as the estimated daily intake was computed for each element[16]:

$$EDI = \frac{Cmetal \times Average\ daily\ in\ take}{BA}$$

Cmetal is concentration of heavy metals in plants (mg $kg^{-1}$), Average daily intake of vegetable in Nigeria for adult was taken as 0.086 kg $day^{-1}$ in accordance with Hart *et al.*[15] Adult body weight was taken as 60 kg. To determine the degree of toxicity the computed EDI values were compared with an acceptable level of oral reference dose (Rfd)[16].

**Pollution index (PI):** Pollution index (PI) is the ratio of metal concentration in a biotic or abiotic medium to that of the regulatory Standard of international bodies such as the World Health Organization (WHO), United States Environmental Protection Agency (USEPA) was observed. Values of PI<1 value indicates that the soil or plant material is not yet contaminated whereas PI>1 indicates pollution. On the other hand, PI = 1 reveals a critical state which makes the involved plant useful for environmental monitoring[17].

Mathematically, PI was computed as:

$$PI = \frac{C_{plant}}{C_{USEPA\text{-}STANDARD}}$$

where, PI is the individual pollution index of study metal, $C_{plant}$ is the concentration of the metal in plant and $C_{USEPA}$ is the value of the regulatory limit of the heavy metal by USEPA[16].

Whereas the USEPA, 2006 Oral Reference Dose (mg $kg^{-1}$ b.wt./day) for heavy metals was taken as Cr = 1.5, Cd = 0.001, Pb = 0.004, Cu = 0.04, Zn = 0.3[16].

**Statistical analysis:** The data were analyzed using statistical software "R" 2014 version and SPPS version 8.1. Analysis of variance (ANOVA) was applied for evaluating the significance difference at 0.05 level between heavy metal concentrations in different parts of each vegetable. One way ANOVA was also applied to determine the significance difference between heavy metals concentration in plants tissues and the concentration in corresponding soil for each plant.

## RESULTS AND DISCUSSION

**Heavy metals concentration in crops:** The heavy metal concentrations in different parts of the crops under investigation (roots, stems, leaves and fruits) ranged from 7.67-0.33, 8.17-0.50, 1.17-0.33, 32.83-6.33, 186.08-19.00 mg $kg^{-1}$ for Cd, Pb, Cr, Cu and Zn, respectively (Table 1). There is significant ($p<0.05$) variation in the concentrations of heavy metals in different parts of most of the crops. Variability exist in the uptake and concentration of heavy metals among the different types of crops which could be attributed to individual differences in metal uptake and tolerance coupled with soil types[18]. The finding of this research showed a decreasing order of concentration via Zn>Pb>Cd>Cu>Cr (Table 1). Cadmium concentrations varied from (7.67 mg $kg^{-1}$) in *Spinacia oleracea* root to (0.33 mg $kg^{-1}$) in *Solanum lycopersicum* root. The concentrations of cadmium in all the crops under investigation were above the permissible limits of 0.2 mg $kg^{-1}$ [19], indicating high contamination of the crops. Previous research findings found cadmium as a component of pesticides used by farmers in Northern Nigeria[20]. The cadmium concentrations in this study were higher (0.47 mg $kg^{-1}$) than what was reported[21] in Tunisia and (1.57 mg $kg^{-1}$) in Pakistan[22]. However, the cadmium concentrations reported in the present study were comparatively similar to 4.09 mg $kg^{-1}$ reported in Bangladesh[23] and 2.97 mg $kg^{-1}$ in southern Nigeria by[1]. Conversely, significantly higher concentrations (900 mg $kg^{-1}$) of cadmium was reported in China[14], in different crops compared to the cadmium concentrations reported in this study. Cadmium is a non-essential element, which has no function in the human body but can exhibit toxic tendencies even in low concentrations[22]. Cadmium has been showing to accumulates in the human bones, lungs, liver, kidneys, nerve tissues, leading to their damage and malfunction[24-26].

The concentrations of lead ranged from 8.17 mg $kg^{-1}$ in *Oryza sativa root* to 0.50 mg $kg^{-1}$ in *Capsicum cerasiforme* root (Table 1). The concentration of lead in all the studied crops exceeded the permissible limits of 0.3 mg $kg^{-1}$ [19]. This is an indication of the potential toxicity and health risk to humans. This contamination is due to the presence of lead contents in the pesticides[20] use indiscriminately in the study area. The result revealed high contamination in *Zea mays*, *Oryza sativa* and *Spinacia oleracea* compared to other crops. Interestingly the Lead concentrations recorded in this study were higher compared to 0.034 mg $kg^{-1}$ reported in China[27], 2.2 mg $kg^{-1}$ in Romania[28] and 0.09 mg $kg^{-1}$ reported[2] in Nigeria. However, other findings reported higher values. For example 28.14 mg $kg^{-1}$ was reported in Bangladesh[23] and 374 mg $kg^{-1}$ in Indonesia was reported previously[29]. Lead is non-essential metal that has no function in the human body and exposure to lead has been linked to incidence of neurological disorders, hypertension, cognitive impairment, renal dysfunction, arthritis, hallucination and vertigo[30].

Chromium concentrations varied from 1.17 mg $kg^{-1}$ in *Allium cepa* stem to 0.33 mg $kg^{-1}$ in *Capsicum cerasiforme* stem (Table 1). The concentrations of chromium in all the crops under investigation were above the permissible limits of 2.3 mg $kg^{-1}$ [19]. This chromium contamination could be a potential risk for human consumption. This contamination could be attributed to the use of pesticides with high chromium contents by farmers in the study area. Studies have shown the presence of heavy metals in pesticides at levels above the recommended farmer's dilution rate[20,4,5]. The concentration of chromium in this study was higher compared to 0.077 mg $kg^{-1}$ in China[27], 0.84 mg $kg^{-1}$ in Nigeria[2] and 0.32 mg $kg^{-1}$ reported in India[31]. Conversely, chromium concentrations in this study were significantly lower than concentrations of 17.8 mg $kg^{-1}$ in Romania[28], 10.5 mg $kg^{-1}$ in Pakistan[22] and 33.16 mg $kg^{-1}$ in Bangladesh[23]. The concentration in this study was within the range reported of 2.5 mg $kg^{-1}$ reported in Bangladesh[32]. The chromium concentrations are thus a likely potential health risk to its consumers. Chromium has been shown to cause ulcer, perforation of nasal septum, respiratory cancer and alterations in replication and transcription of DNA and chromosomal aberrations in humans[33,34].

The concentration of copper in the studied crops ranged from (32 mg $kg^{-1}$) in *Spinacia oleracea* root to (6.33 mg $kg^{-1}$) in *Solanum lycopersicum* root (Table 1). The concentrations of copper in different parts of the studied crops are shown in (Table 1). Copper concentrations in this study were below the permissible limits of 40 mg $kg^{-1}$ [19], indicating non Cu contamination. This might infer low copper contents in the pesticides used in the study area. The concentration of copper in this study is higher than (1.68 mg $kg^{-1}$) in Kenya[35], (8.56 mg $kg^{-1}$) in Pakistan[22] and (25.45 mg $kg^{-1}$) in Bangladesh by[23]. Conversely, copper concentrations in this study were substantially lower than (62.58 mg $kg^{-1}$) in Tunisia[21] and (46.53 mg $kg^{-1}$) in Nigeria by[1]. Copper is an essential metal and a component of several enzymes necessary for normal metabolic functions in humans[36,37]. But excessive uptake of copper in humans can cause temporary gastrointestinal distress with symptoms such as nausea, vomiting, abdominal pain, liver toxicity[37].

Table 1: Mean concentration of heavy metals (mg kg$^{-1}$) in crops

| Sampling site | Name of sample | Botanical name | Hausa name | Heavy metals (mg kg$^{-1}$) Cd | Pb | Cr | Cu | Zn |
|---|---|---|---|---|---|---|---|---|
| Gombe | Spinach | *Spinacia oleracea* | Alayyaho | | | | | |
| | Root | | | 7.67$^{a}$ | 5.33$^{a}$ | 0.17$^{a}$ | 32.83$^{b}$ | 19.00$^{a}$ |
| | Stem | | | ND | ND | ND | ND | ND |
| | Leaf | | | 1.33$^{b}$ | 2.33$^{b}$ | 0.58$^{a}$ | 10.92$^{a}$ | 42.67$^{b}$ |
| | Sweet pepper | *C. cerasiforme* | Tattase | | | | | |
| | Root | | | ND | ND | ND | ND | ND |
| | Stem | | | 1.83$^{a}$ | 0.50$^{a}$ | 0.33$^{a}$ | 9.50$^{a}$ | 68.33$^{a}$ |
| | Leaf | | | 1.33$^{a}$ | 3.33$^{a}$ | 0.67$^{a}$ | 17.58$^{b}$ | 66.50$^{a}$ |
| | Fruit | | | ND | ND | ND | ND | ND |
| | Tomato | *S. lycopersicum* | Tomatur | | | | | |
| | Root | | | 0.33$^{a}$ | ND | 0.50$^{a}$ | 6.33$^{a}$ | 26.83$^{a}$ |
| | Stem | | | 1.42$^{b}$ | 4.92$^{a}$ | 0.83$^{a}$ | 16.83$^{a}$ | 61.83$^{a}$ |
| | Leaf | | | 1.42$^{b}$ | 6.33$^{a}$ | 1.00$^{a}$ | 14.25$^{a}$ | 112.92$^{a}$ |
| | Fruit | | | ND | ND | ND | ND | ND |
| | Lettuce | *Lactuca sativa* | Salad | | | | | |
| | Root | | | ND | ND | ND | ND | ND |
| | Stem | | | ND | ND | ND | ND | ND |
| | Leaf | | | 1.17 | 2.50 | 1.00 | 23.67 | 56.17 |
| | Onion | *Allium cepa* | Albasa | | | | | |
| | Root | | | 0.75$^{a}$ | 4.42$^{a}$ | 0.83$^{a}$ | 26.50$^{a}$ | 73.92$^{a}$ |
| | Stem | | | ND | ND | ND | ND | ND |
| | Leaf | | | 1.00$^{a}$ | 3.50$^{a}$ | 1.17$^{b}$ | 22.00$^{a}$ | 99.33$^{a}$ |
| | Maize | *Zea mays* | Masara | | | | | |
| | Root | | | ND | ND | ND | ND | ND |
| | Stem | | | 0.50$^{a}$ | 0.50$^{a}$ | 0.50 | 12.83$^{a}$ | 37.50$^{a}$ |
| | Leaf | | | ND | ND | ND | ND | ND |
| | Fruit | | | 1.42$^{a}$ | 7.17$^{b}$ | ND | 10.92$^{a}$ | 186.08$^{b}$ |
| | Rice | *Oryza sativa* | Shinkafa | | | | | |
| | Root | | | 2.00$^{a}$ | 8.17$^{a}$ | ND | 22.33$^{a}$ | 84.50$^{a}$ |
| | Stem | | | ND | ND | ND | ND | ND |
| | Leaf | | | ND | ND | ND | ND | ND |
| | Fruit | | | 7.33$^{b}$ | 3.50$^{b}$ | 1.17 | 24.83$^{a}$ | 54.00$^{b}$ |
| Safe limit$^{a}$ | | | | 0.2 | 0.3 | 2.3 | 40 | 60 |

Mean followed with the same letter across the column are not significantly different $p>0.05$

The concentration of zinc in the studied crops is shown in (Table 1). Zinc concentrations varied from (186.08 mg kg$^{-1}$) in *Zea mays* fruit to (19 mg kg$^{-1}$) in *Spinacia oleracea* root. The zinc concentrations were above the 60 mg kg$^{-1}$ permissible limits[19], in *Zea mays* fruit (186.08 mg kg$^{-1}$), *Solanum lycopersicum* leave (112.92 mg kg$^{-1}$), *Allium cepa* leave (99.33 mg kg$^{-1}$), *Oryza sativa* root (84.50 mg kg$^{-1}$), *Allium cepa* root (73.92 mg kg$^{-1}$), *Capsicum cerasiforme* root (68.33 mg kg$^{-1}$) and *Capsicum cerasiforme* leave (66.50 mg kg$^{-1}$). Thus the consumption of these crops for zinc could pose potential health risk in humans. This could be attributed to the presence of zinc contents in the pesticides which are used indiscriminately by farmers in the study area (personal communication). The concentration of zinc in the present study was comparatively higher than 32.44 mg kg$^{-1}$ reported in Tunisia[21], 88.4 mg kg$^{-1}$ in Pakistan[22] and 1.68 mg kg$^{-1}$ in Kenya[35]. However, zinc concentration in this study was similar to 171.03 mg kg$^{-1}$ in vegetables from Titagarh, India[38]. Zinc is an essential trace element and has a rather low toxicity[39]. Studies have shown that high dietary doses of zinc can cause clinical symptoms of gastrointestinal distress[38].

The mean concentrations of Cd, Pb, Cr and Co reported in this study were lower compared with some reports from China, Pakistan, Bangladesh, Indonesia and Romania could be due to industrialization, waste water irrigation, mine land sites. However, the higher Zn concentrations observed in this with reports from some countries could be due to indiscriminate use of metal based pesticides[20].

**Heavy metals concentrations in soil:** The mean concentration of heavy metals in the corresponding soil of the crops under investigation is shown in (Table 2). The result showed that the mean heavy metal concentrations in the corresponding soils

Table 2: Mean concentrations of heavy metals (mg kg$^{-1}$) in soils of corresponding crops

| Sampling site | Soil from the land of | Botanical name | Hausa name | Heavy metals (mg kg$^{-1}$) Cd | Pb | Cr | Cu | Zn |
|---|---|---|---|---|---|---|---|---|
| Gombe | Spinach | *Spinacia oleracea* | Alayyaho | 1.92$^{abc}$ | 2.83$^{a}$ | 0.58$^{a}$ | 5.58$^{a}$ | 190.83$^{b}$ |
| | Sweet pepper | *C. cerasiforme* | Tattase | 0.17$^{ab}$ | 4.50$^{a}$ | 1.00$^{a}$ | 7.50$^{b}$ | 48.42$^{b}$ |
| | Tomato | *S. lycopersicum* | Tomatur | 3.67$^{c}$ | 2.33$^{a}$ | 0.58$^{a}$ | 5.17$^{a}$ | 26.67$^{a}$ |
| | Lettuce | *Lactuca sativa* | Salad | 1.50$^{ab}$ | 4.00$^{a}$ | 1.00$^{a}$ | 4.83$^{a}$ | 30.83$^{a}$ |
| | Onion | *Allium cepa* | Albasa | 1.67$^{ab}$ | 2.92$^{a}$ | 0.92$^{a}$ | 10.83$^{b}$ | 37.25$^{a}$ |
| | Maize | *Zea mays* | Masara | 2.50$^{ab}$ | 2.67$^{a}$ | 0.75$^{a}$ | 3.58$^{a}$ | 39.17$^{a}$ |
| | Rice | *Oryza sativa* | Shinkafa | 1.50$^{b}$ | 4.00$^{a}$ | 1.00$^{a}$ | 4.83$^{a}$ | 30.83$^{a}$ |
| Safe limits[a] | | | | 3.00 | 100.00 | 100.00 | 100.00 | 300.00 |

[a]Source: UNEP *et al.* [40]. Mean followed with the same letter across the column are not significantly different p>0.05

Table 3: Bioaccumulation factor (BAF) of heavy metals in the edible parts of crops

| Sampling site | Name of sample | Botanical name | Hausa name | BAF Cd | Pb | Cr | Cu | Zn |
|---|---|---|---|---|---|---|---|---|
| Gombe | Spinach | *Spinacia oleracea* | Alayyaho | 2.35 | 1.35 | 0.64 | 3.92 | 0.16 |
| | Lettuce | *Lactuca sativa* | Salad | 0.78 | 0.63 | 1 | 4.90 | 1.82 |
| | Onion | *Allium cepa* | Albasa | 0.48 | 3.96 | 1.2 | 3.16 | 1.93 |
| | Maize | *Zea mays* | Masara | 0.64 | 0.96 | 0.5 | 2.46 | 3.63 |
| | Rice | *Oryza sativa* | Shinkafa | 14.00 | 1.46 | 1.17 | 4.88 | 2.25 |

BAF value greater than >1 is indicating high uptake of metal

of the crops under investigation ranged from 3.67 mg kg$^{-1}$ in *Solanum lycopersicum* soil to 0.17 mg kg$^{-1}$ in *Capsicum cerasiforme* soil for Cd, 4.50 mg kg$^{-1}$ in *Capsicum cerasiforme* soil to 2.33 mg kg$^{-1}$ in *Solanum lycopersium* soil for Pb, 1.00 mg kg$^{-1}$ in *Lactuca sativa* soil to 0.58 mg kg$^{-1}$ in *Spinacia oleracea* soil for Cr, 10.8 mg kg$^{-1}$ in *Allium cepa* soil to 3.58 mg kg$^{-1}$ in *Zea mays* soil for Cu and 190.83 mg kg$^{-1}$ in *Spinacia oleracea* soil to 26.67 mg kg$^{-1}$ in *Solanum lycopersicum* soil for Zn. The mean concentration of all heavy metals in the corresponding soils of all the studied crops was below the permissible limits set[40] for agricultural soils except for Cd in *Solanum lycopersicum*. The lower mean concentration of metals in soil of the studied crops could be due to the higher uptake of heavy metals by the crops and lower retention in soil. Previous[41] have shown that higher concentration of heavy metals in crops tissues than their corresponding soil reflects relatively poor retention in soil and greater efficiency of crops to absorb metals. The mean soil concentration of cadmium in this study was higher than what was reported in some parts of the world. For example, 0.46 mg kg$^{-1}$ was reported in Tunisia[21], 0.001 mg kg$^{-1}$ in Nigeria by[2]. Lead soil concentrations in the present study were similar to 6.53 mg kg$^{-1}$ reported[2] and 2.31 mg kg$^{-1}$ in India[22] but lower compared to 70.36 mg kg$^{-1}$ in China[11] and 559.5 mg kg$^{-1}$ from near chemical facility in Romania[28]. Chromium concentrations in soils of this study were comparatively lower than 8.9 mg kg$^{-1}$ in Nigeria[2] and 525.8 mg kg$^{-1}$ from chemical facility in Romania[28]. While the soil concentrations of Cu were lower compared to 62.87 mg kg$^{-1}$ reported in Tunisia by[21], however Zn concentrations in soil of the present were higher than 21.42 mg kg$^{-1}$ reported in Tunisia by[21].

**Heavy metals soil plant transfer bioaccumulation factor (BAF):** The values of the estimated bioaccumulation of heavy metals concentration in the corresponding soil of each crop are presented in (Table 3). The bioaccumulation factor of heavy metals is measured by ratio of metal concentrations in plants to metal concentrations in soil[18]. Higher soil to plant transfer of heavy metals means strong accumulation of these metals in plants tissues[14]. The result showed variability in BAF values among the different crops and heavy metals. The result revealed that among the five metals the highest BAF was observed for Cd (14.00) followed by Cu (4.88) in *Oryza sativa*, for Pb (3.96) in *Allium cepa*, for Zn (3.63) in *Zea mays* and (1.17) in *Oryza sativa* for Cr. The trend of metal transfer from the soil to crops was in the order of Cd>Cu>Pb>Zn>Cr and most of the crops showed BAF>1 indicating higher absorption of heavy metals from soil. This is worrisome as bioaccumulation (BAF) has been showing to be a key path way of human exposure to heavy metals through food chain[18]. The variation observed in BAF values among the crops and metals could be due to several factors such as metal concentration in the soil, their chemical forms, the difference in uptake capability and growth rate of different plant species[42]. The BAF values in this study were higher compared

Table 4: Pollution indices of heavy metals in the edible parts of crops

| Sampling site | Name of sample | Botanical name | Hausa name | Pollution index Cd | Pb | Cr | Cu | Zn |
|---|---|---|---|---|---|---|---|---|
| Gombe | Spinach | *Spinacia oleracea* | Alayyaho | 6.67 | 7.78 | 0.25 | 0.27 | 0.71 |
| | Lettuce | *Lactuca sativa* | Salad | 5.83 | 8.33 | 0.43 | 0.59 | 0.94 |
| | Onion | *Allium cepa* | Albasa | 5.00 | 11.67 | 0.51 | 0.55 | 1.66 |
| | Maize | *Zea mays* | Masara | 7.08 | 23.89 | 0.00 | 0.27 | 3.10 |
| | Rice | *Oryza sativa* | Shinkafa | 36.67 | 11.67 | 0.51 | 0.62 | 0.90 |

N.B. PI value greater than >1 is indicating high pollution load

Table 5: Daily intake of metals (EDI) (mg kg$^{-1}$ b.wt./day) through consumption of crops

| Botanical name | Hausa name | Estimated daily intake Cd | Pb | Cr | Cu | Zn |
|---|---|---|---|---|---|---|
| *Spinacia oleracea* | Alayyaho | 0.00191 | 0.00334 | 0.00084 | 0.01565 | 0.06116 |
| *Lactuca sativa* | Salad | 0.00167 | 0.00358 | 0.00143 | 0.03392 | 0.08051 |
| *Allium cepa* | Albasa | 0.00143 | 0.00502 | 0.00167 | 0.03798 | 0.14238 |
| *Zea mays* | Masara | 0.00203 | 0.01027 | 0.00000 | 0.01565 | 0.26672 |
| *Oryza sativa* | Shinkafa | 0.01051 | 0.00502 | 0.00167 | 0.03559 | 0.07740 |
| RfD[a] | | 0.001 | 0.004 | 1.5 | 0.04 | 0.30 |

[a]Source: USEPA[16]

to 0.16, 1.18, 1.19, 1.72 and 0.36 for Cr, Zn, Cd, Cu and Pb reported in Pakistan[22]. Similarly, the Cd BAF values in this study were similar to the 16.79 BAF Cd value reported in Pakistan[21].

**Pollution index determination:** The Pollution Index (PI) is measured by the ratio of metal concentration in plant to that of the regulatory standard (WHO). The PI<1 indicates that the plant is not yet contaminated whereas PI>1 indicates pollution. The result of pollution index in the crops is presented in the (Table 4). The result showed that PI values ranged from 36.67 in *Oryza sativa* to 5.00 in *Allium cepa* for Cd, 23.89 in *Zea mays* to 7.78 in *Spinacia oleracea* for Pb, (0.51) in *Oryza sativa* to, 0.25 in *Spinacia oleracea* for Cr, 0.62 in *Oryza sativa* to, 0.27 in *Zea mays* for Cu and 3.10 in Zea mays to 0.71 in *Spinacia oleracea* for Zn. The result suggest that PI values for all the studied crops for Cd and Pb were >1 indicating high level contamination for these metals and hence unsuitable for human consumption. This is due to the presence of high contents of Cd and Pb in the pesticide use in the study area. However, PI values were <1 in all the studied crops for Cr, Cu, Zn except in *Allium cepa* and *Zea mays.* The trend of PI value was in decreasing risk Cd>Pb>Zn>Cu>Cr.

**Health risk assessment**

**Estimated daily intake (EDI) of heavy metals:** The EDI of the heavy metals was computed based on the heavy metals concentration in the edible part of each food crop and individual consumption rate for adults (Table 5). The result showed that the EDI ranged from 0.01051-0.0014, 0.01027-0.00334, 0.00167-0.00084, 0.03798-0.01565 and 0.26672-0.06116, for Cd, Pb, Cr, Cu and Zn, respectively. The highest daily intake of Cd, Pb and Zn were observed in *Zea mays* and the least was observed in *Spinacia oleracea* for Cr. This could be attributed to higher metals uptake efficiency of *Zea mays* in comparison other studied crops[32]. The result showed all the studied crops exceeded the oral reference dose RfD (the daily exposure of individuals to toxins or pollutants that can pose no appreciable hazard over life time) and are likely to cause human health risk. In the entire crop under investigation Cd exceeded the RfD while *Allium cepa*, *Zea mays* and *Oryza sativa* only Pb exceeded the RfD. The computed daily intake of all the crops for Cr, Cu and Zn were below the RfD dose and are unlikely to cause health risk to humans. The result showed a decreasing order based on daily intake through food consumption as Cd>Pb>Zn>Cu>Cr. This result suggest that Cd, Pb were the major metals contributing significantly to the potential health risk through consumption of the studied food crops in the study area.

**Estimated hazard quotient (HQ) and hazard index (HI):** The potential health to humans through the intake of the contaminated crops was evaluated using the Hazard Quotient (HQ) are presented in (Table 6). The computed hazard quotient from the consumption of the studied crops ranged from 0.8934 in *Oryza sativa* to 0.12183 in *Allium cepa* for Cd, 0.21822 in *Zea mays* to 0.07107 in *Spinacia oleracea* for Pb, (9.47E-05) in *Oryza sativa* to (4.73E-05) in *Spinacia oleracea* for Cr, (0.0807) in *Allium cepa* to (0.03325) in *Spinacia oleracea* for Cu and (0.07557) in *Zea mays* to (0.01733) in *Spinacia oleracea* for Zn. The result showed a decreasing trend of health risk

Table 6: Hazard quotient and hazard index for adult population through the consumption of crops

| Name of sample | Botanical name | Hausa name | Hazard quotient (HQ) Cd | Pb | Cr | Cu | Zn | Hazard index (HI) |
|---|---|---|---|---|---|---|---|---|
| Spinach | *Spinacia oleracea* | Alayyaho | 0.16244 | 0.07107 | 4.73E-05 | 0.03325 | 0.01733 | 0.28414 |
| Lettuce | *Lactuca sativa* | Salad | 0.14214 | 0.07615 | 8.12E-05 | 0.07208 | 0.02281 | 0.31326 |
| Onion | *Allium cepa* | Albasa | 0.12183 | 0.10660 | 9.47E-05 | 0.08071 | 0.04034 | 0.34959 |
| Maize | *Zea mays* | Masara | 0.17260 | 0.21828 | ND | 0.03325 | 0.07557 | 0.49970 |
| Rice | *Oryza sativa* | Shinkafa | 0.89344 | 0.10660 | 9.47E-05 | 0.07564 | 0.02193 | 1.09771 |

from the consumption of the crops viz Cd>Pb>Cu>Zn>Cr. This result revealed <1 HQ values through the consumption of all the investigated crops, indicating that the consumption of these crops will not pose any health risk to human.

The Hazard Index (HI) value expresses the combined non carcinogenic effects of multiple heavy metals (Table 6). The estimated hazard index varied from the highest (1.0977) in *Oryza sativa* to the least (0.28414) in *Spinacia oleracea*. It was observed that current estimates of (HI) from the consumption of all the studied heavy metals in *Oryza sativa* were slightly greater than one (1.0977) and will pose a potential health risk through continues intake. High HI values were reported in Bangladesh 5.23[32] and (1.48) in Nigerian by[2] compared to estimate in this study.

## CONCLUSION

The present study revealed that the concentrations of heavy metals Cd, Pb in all the studied crops fumigated with pesticides as the only source of contamination. The study showed variability in the uptake of heavy metals in different parts of crops and in different plant species. Heavy metals soil plant transfer showed BAF at its highest for Cd in *Oryza sativa* compared to other metals. The pollution indices revealed that all the crops were contaminated with Cd and Pb and *Oryza sativa* and *Allium cepa* were contaminated with Zn hence considered unsafe for human consumption. The estimated daily intake in all the crops for Cd was higher than the RfD while in *Allium cepa*, *Zea mays* and *Oryza sativa* Pb was higher than the RfD values and therefore had the major contribution to potential health risk through food consumption in the study area. Hazard quotient for adult indicated these crops as safe for human consumption. However, the potential risk from the consumption of all the heavy metals combined which is the realistic scenario showed slightly health risk from the consumption of *Oryza sativa*. Thus regular screening of heavy metals in crops and pesticides is suggested.

## SIGNIFICANCE STATEMENTS

This study has uncovered that pesticides application are major contributors of heavy metals accumulation in crops and their link with health risk to humans that consumed them. The study also explores that contaminated irrigated water is not the only source of heavy metal burden in crops. The outcome of this study will help researchers and particularly medical personnel in a proper diagnosis of symptoms associated with heavy metal toxicity in humans. The outcome of this finding will help spark a new frontier of research on the development of new alternative and environmentally safe or less harmful pesticides for now and in the future safe biopesticides. It will also provide clue to government and other environmental and health agencies on the regulation of import and judicious application of these chemical pesticides.

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