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HEAVY METAL CONTAMINATION OF SOME VEGETABLES FROM PESTICIDES AND THE POTENTIAL HEALTH RISK IN BAUCHI, NORTHERN NIGERIA

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ABSTRACT

Vegetable farming in developing countries is characterized by the indiscriminate application of pesticides and the resultant pollution of agricultural soil with heavy metals that form constituents of these pesticides. These heavy metals have long term toxicity to human and other biota in the ecosystem. This problem is exacerbated by lack of monitoring to regulate the excessive use of pesticides. The objective of this study was to determine the presence and concentration of some heavy metals in pepper, tomato and onion grown in pesticide contaminated farm and the human health risk associated with their consumption. Pepper (*Capsicum annuum*), Tomato (*Solanum lycopersicum*) and Onion (*Allium cepa*) and their corresponding soils were collected from three vegetable farms. The concentrations of Pb, As, Cd, Cr and Zn in the plants and soil were determined. The potential health risk from the consumption of these vegetables was assessed using the methods developed by World Health Organization (WHO) and Food and Agricultural Organization (FAO). Results obtained showed presence of heavy metals (Pb, As, Cd, Cr and Zn) Sin different parts of the plants and at different concentrations, with some above the WHO/FAO permissible limits. The edible part of these vegetables (fruits) had the least mean contents of all heavy metals while roots recorded the highest mean concentration. All vegetables were found to be hyper accumulators of Cd and Cr. The Hazard Index (HI) of heavy metal contamination in these vegetables suggested a potential human health risk. The need by government for monitoring and regulation of pesticides application needs urgent attention.

Key Words: Vegetables, pesticides, heavy metals, health risk, toxicity

INTRODUCTION

Agricultural lands in northern part of Nigeria which produces food crops that the nation almost entirely rely on suffers a great deal from environmental pollution caused by pesticides and fertilizer applications. The present agricultural production has not been proportionating with the ever-increasing population in developing countries. This is partly due to the menace of plant and animal pests of agricultural importance and due to the increase in human population that has not match the pace of increased food production. The need to strive to feed this ever-increasing population through increased crop production and the prevention of pre-and post-harvest crop diseases has led to the desperate and indiscriminate use of agrochemicals. Agrochemicals especially herbicides, insecticides and fungicide have resulted in the increase in crop production to a large extent (Yuguda *et al.*, 2015, Kranthi *et al*., 2002), but their residual concentrations in the soil and potential environmental hazards in the ecosystem is of great concern. The presence of heavy metals in pesticides has been established from previous studies (Yuguda *et al.*, 2015, FAMIC, 2014, Alloway, 1990; Kabata-pendias and Pendias, 1992, Wang *et al*., 2005). They have been implicated in causing human disorders such as nervous, renal, their carcinogenic, mutagenic disorders among others (WHO, 1992, 1995, Jarup, 2003, Radwan and Salama, 2006).

It is not debatable to assert that heavy metal pollution from pesticides and the cumulative effects on the soil biota and human through consumption of crops treated with these agrochemicals poses a great health risk (Opaluwa *et al*., 2012, Wilson and Pyatt, 2007; Guerra *et al*., 2012). Thus heavy metals from agrochemicals particularly pesticides not only contaminate soil and affect food quality and subsequently transferred to human through food chain (Premarathna *et al*., 2011; Olowoyo *et al*., 2011, Antonious and Konchhar, 2009; Mutune *et al*., 2012) but responsible for series of health ailments in humans.

Vegetables that provide daily and steady supply of vitamins, minerals and fiber to people in the developing countries are contaminated with high doses of pesticides. This can pose serious health risk to millions of people that consume them. These pesticides have heavy metals such Zn, Cd, Pb, Cu, Cr etc. as their constituents (Yuguda *et al.,* 2015). Thus, the importance of heavy metals in vegetables as a vital consideration and recognition in food quality assurance cannot be emphasized. There are differences though, in what appears to be considered maximum limits or allowable concentrations of heavy metals in the edible parts of plants such as vegetables. However, international and national regulations on food quality have being in favor of lowering the maximum allowable levels of heavy metals in food materials due to increasing risk of human contamination (Sharma *et al*., 2009).

Not much attention has been focused on the international and even national regulations of food quality when it comes to vegetables consumed in developing countries. There is in fact, indiscriminate and uncontrollable application of high doses of pesticides in northern Nigeria. A lot of new pesticides are continually infiltrating the markets and from all indication, no screening of these agrochemicals was conducted if at all these chemicals have been duly permitted for entry into the country. This could not be much different from other developing countries. The result of this is the high accumulation of these agrochemicals in the environment (Wang *et al*., 2013) with their consequent risk hazards (Liu *et al.,* 2013).

Soil is the main reservoir of heavy metal and is the main source of pollution to the ecosystem. There are pathways by which humans could be exposed to heavy metals contaminations. These could be through direct ingestion of the vegetables (food chain), direct ingestion of soil particles, dermal contact with soil particles, inhalation of soil dust and other particles from the air, oral and or dermal intake from groundwater (Liu, *et al.,*2013). A lot of pesticides whose actual chemistry are not known are infiltrating the markets in the study area and the potential hazards should be of great concern.

Recent studies have shown that heavy metals contamination of food crops and its resultant health consequences is becoming a global issue (Sharma *et al*., 2008 *et al*., Guerra *et al*., 2012; Shah *et al*., 2013; Nazir *et al*., 2015; Yuguda *et al.,* 2015). There is paucity of information on the heavy metals contents of vegetables from agrochemicals and their health risks in Northern Nigeria that produce them. Hence, this study was designed with the aim of determining heavy metals (Cd, Pb, As, Zn and Cr) contents in soil and some vegetables produced in Bauchi and distributed throughout Nigeria for consumption and the health risk associated with them.

MATERIALS AND METHODS

Samples of roots, stem, leaves and fruits of Pepper (*Capsicum annum*), Tomato (*Solanum lycopersicum*) and Onion (*Allium cepa*) and their corresponding soils were randomly collected from three farms in Bauchi located in North Eastern Nigeria. Soil collected outside the farm that has no pesticides application was considered control samples. All samples were packaged in well labeled polythene bags. Samples were transported to the laboratory of Abubakar Tafawa Balewa University (ATBU) Bauchi, Nigeria for processing and metal analysis.

1. Preparation of Samples

In the laboratory, the vegetable samples were washed with tap water and thoroughly rinsed with deionized water to remove any soil particles or debris that must have been attached to the plants. They were then separated into non-edible parts (roots, stem, and leaves) and edible parts (fruits). Soil and plant samples were air dried for 24 hours to reduce moisture content and then dried in an oven at 70^{o}C for another 24 hours until they are completely dried. The dried parts were pulverized and sieved through a mesh size of 2mm. The powdered samples were then kept in well labeled plastic containers before analysis.

2. Heavy Metal Analysis

Samples of both soils and vegetables (1.00±0.001g each) were placed into 100ml beakers separately, to which 15ml of tri-acid mixture (70% high purity $HN0_3$, 65%, $HCl0_4$ and 70%

H_2SO_4 in 5:1:1 ratio) were added. The mixture was digested at 80^0C till the solution became transparent. The resulting solution were filtered and diluted to 50ml using deionized water and analyzed for Pb, Cr, Cd, and Zn, by atomic absorption spectrophotometry.

RISK TO HUMAN HEALTH

Daily intake evaluation was determined by multiplying the average metal content of each vegetable by the estimated consumption rate of vegetable in Nigeria. The consumption rate of children was taken as one third of adults (IBGE, 2004). Risk to health due to intake of metal contaminated vegetables was calculated using the hazard quotient (HQ) as described by United State Environmental Protection Agency (USEPA, 1989), where HQ is taken as the ratio between exposure and oral reference dose (R_fD) Given as;

$$HQ = \frac{Div \ X \ C_{metal}}{RfD \ X \ Bo}$$

Div = daily intake of vegetables (kgd^{-1})

C_{metal} = concentration of metal in vegetables ($mgkg^{-1}$)

R_fD = oral reference dose for the metal

Bo = human body mass (kg).

Where the Daily Vegetable intake (DiV) for adults was taken as $400gd^{-1} = 0.4kgd^{-1}$, Daily Vegetable Intake (DiV) for children was taken as 1/3 of adults = $0.13kgd^{-1}$, Adult body weight (Bo) was taken as 70kg, and Children body weight 0 – 6yrs (Bo) was taken as 19.25kg,
The Oral Reference Dose (RfD) for HMs was taken as: As = $0.0030mgd^{-1}$, Cd = $0.0010 \ mgd^{-1}$, Cr = $1.50 \ mgd^{-1}$, Pb = $0.0035 \ mgd^{-1}$, Zn = $0.30 \ mgd^{-1}$

When the ratio is < 1 there is no obvious risk (USEPA (1989),

1. Hazard Index (HI)

Potential risk to human health due to more than one heavy metal known as the Hazard Index (HI) was calculated as described by USEPA (1989), which is the total sum of all the Hazard Quotients as shown in the equation below;

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

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

2. Bio-accumulation Factor

Bio-accumulation factor is an index of the ability of the vegetable to accumulate a metal to its concentration in the soil substrate.

$$\mathbf{BAF = C_{Plant}/C_{Soil}}$$

Where; C_{Plant} = heavy metal concentration in edible part of vegetables

C_{Soil} = heavy metal concentration in edible part of vegetables.

STATISTICAL ANALYSIS

Analysis of variance (ANOVA) using MINITAB 17 statistical software was used to calculate the differences between the heavy metals concentrations across the vegetables at 0.05 level of significance.

RESULTS AND DISCUSSION

1. *Soil Heavy Metals Contents*

The mean concentrations (mg kg^{-1}) of heavy metals in the soils of the vegetables farm are presented in Table 1. The mean Zn content recorded the highest values compared to all other metals. It ranged between 21.38 to 28.58 mg kg^{-1} in tomato and pepper farms respectively. Chromium had the least concentration with mean values of 0.01 (tomato and onion) to 0.02 mg kg^{-1} (pepper). Generally, the soil heavy metal concentrations decreased in the order Zn >Pb> As > Cd > Cr.

Table 1: Mean Soil Heavy Metal Concentrations ($mgkg^{-1}$) of vegetable farm.

Farm Soil	Pb	As	Cd	Cr	Zn
Pepper	2.77 ± 0.435^{b}	1.65 ± 0.264^{a}	0.22 ± 0.023^{a}	0.02 ± 0.003^{a}	28.58 ± 2.660^{a}
Tomato	3.27 ± 2.347^{ab}	0.69 ± 0.565^{b}	0.14 ± 0.019^{a}	0.01 ± 0.002^{a}	21.38 ± 2.726^{b}
Onion	4.31 ± 0.473^{a}	2.12 ± 0.244^{a}	0.03 ± 0.003^{b}	0.01 ± 0.001^{a}	23.06 ± 7.778^{b}
Control	0.31 ± 0.135^{c}	0.18 ± 0.029^{c}	0.01 ± 0.001^{c}	0.01 ± 0.001^{a}	2.93 ± 1.568^{c}
WHO/FAO PL	**100.00**	**20.00**	**3.00**	**100.00**	**300.00**

Means with the same letter across the column are not significantly different p > 0.05.

PL = Permissible Limit

The control soils had significantly lower heavy metals than all the farm soils except Cr. The lower Cr in the pesticides contaminated farm soil could be attributed to very low Cr levels in the Pesticides. The soil heavy metals in the vegetable farm soils were below the WHO/FAO permissible limits.

2. *Heavy Metal Concentration in the Vegetables*

One of the main concern in this study is the heavy metal concentrations in the edible parts of the vegetables which are the fruits in the tomato and pepper and in onion its bulbs and to a little extent its leaves. This, however, was the major focus and concern and the basis for quantifying the risk to humans in this study. The mean concentrations of heavy metals in the vegetables are shown in Table 2. In the different parts of the vegetables analyzed for heavy metals and regardless of whether or not they exceed the WHO/FAO permissible limit, the roots which is the main organ of uptake of these metals in the pesticide fumigated soil generally had higher heavy metals concentrations than the other parts of the vegetables. These differences were significant (P<0.05) for Pb, Cd, Cr and Zn in pepper, Pb and Zn in tomato and Pb and Cd in onion. Heavy metals and nutrients absorbed by the roots are usually translocated and allocated to different parts of the plants which could limit the concentrations in the roots. However, availability of metals in the soil and continuous absorption by the roots could lead to higher concentration in the roots.

The mean concentrations of the heavy metals in pepper (*Capsicum annuum*) decreased in the order root > leaf > stem > fruit. The only exception, is As which had the highest concentration (0.39 $mgkg-^{1}$) in the leaves, though this difference was not significant (P>0.05). The root had significantly (P>0.05) higher Pb, Cd, Cr and Zn than that of stem and fruit. The values for the root were 4.64, 0.94, 0.33 and 17.19$mgkg^{-1}$ respectively and did not exceed the WHO/FAO permissible limits. The As and Cd in all the parts of pepper exceeded the WHO/FAO permissible limits. The range was from 0.13 to 0.39 $mgkg^{-1}$ and 0.32 to 0.94 $mgkg^{-1}$ respectively. While Cr and Zn were lower than the WHO/FAO limits and ranged from 0.15 to 0.33 $mgkg^{-1}$ and 3.95 to 17.19 $mgkg^{-1}$ respectively. The fruit which is the edible part of this vegetable recorded heavy metal concentrations that did not exceed the WHO/FAO permissible limits. As was the only heavy metal that slightly (0.13$mgkg^{-1}$) exceeded the permissible limit (0.1 $mgkg^{-1}$).

Table 2: Mean Concentration ($mgkg^{-1}$) of Heavy Metals in Different Parts of Vegetables.

Vegetable	Parts	Pb	As	Cd	Cr	Zn
**Pepper (*Capsicum*	Fruit	1.36 ± 0.119^{c}	0.13 ± 0.003^{a}	0.32 ± 0.024^{c}	0.15 ± 0.012^{c}	3.95 ± 0.915^{d}
***annuum*)**	Leaf	3.26 ± 0.316^{ab}	0.39 ± 0.346^{a}	0.69 ± 0.042^{b}	0.27 ± 0.043^{ab}	11.23 ± 1.291^{b}
	Stem	2.08 ± 0.297^{bc}	0.15 ± 0.008^{a}	0.48 ± 0.029^{c}	0.19 ± 0.012^{bc}	7.98 ± 1.982^{b}
	Root	4.64 ± 0.944^{a}	0.27 ± 0.024^{a}	0.94 ± 0.130^{a}	0.33 ± 0.049^{a}	17.19 ± 2.022^{a}
	Fruit	0.79 ± 0.129^{bc}	0.08 ± 0.120^{a}	0.27 ± 0.044^{a}	0.02 ± 0.012^{a}	4.19 ± 0.324^{bc}
Tomato	Leaf	2.28 ± 0.462^{ab}	0.18 ± 0.038^{a}	0.52 ± 0.099^{a}	0.09 ± 0.059^{a}	6.49 ± 1.152^{b}
(*Solanum*	Stem	0.36 ± 0.044^{c}	0.25 ± 0.295^{a}	0.21 ± 0.021^{a}	0.05 ± 0.058^{a}	2.99 ± 0.187^{c}
***lycopersicum*)**	Root	3.77 ± 1.037^{a}	0.26 ± 0.039^{a}	0.66 ± 0.089^{a}	0.13 ± 0.039^{a}	10.48 ± 1.218^{a}
	Bulb	0.79 ± 0.061^{b}	0.02 ± 0.005^{a}	0.11 ± 0.023^{b}	0.02 ± 0.004^{a}	4.70 ± 1.119^{a}
Onion	Leaf	1.21 ± 0.594^{ab}	0.07 ± 0.022^{a}	0.13 ± 0.036^{ab}	0.02 ± 0.012^{a}	4.25 ± 1.398^{a}
(*Allium cepa*)	Root	$2.46 + 0.593^{a}$	$0.13 + 0.084^{a}$	$0.19 + 0.021^{a}$	$0.07 + 0.040^{a}$	$6.19 + 2.940^{a}$
WHO/FAO PL		**5.00**	**0.10**	**0.20**	**5.00**	**60.000**

Means with the same letter in a column across a vegetable row are not significantly different $p > 0.05$.

PL = Permissible Limit

In tomato (*Solanum lycopersicum*), there was variations in the mean concentrations of heavy metals in the different parts with no clear pattern. Comparing the different part of tomato, there is no significant difference (P>0.05) in As, Cd and Cr. The fruit heavy metal contents were generally not different from other parts of the vegetable. Only Cd (0.27 $mgkg^{-1}$) in the fruits was slightly higher than WHO/FAO (0.20 $mgkg^{-1}$) permissible limit.

The mean heavy metal concentration in onion (*Allium cepa*) was highest in the root for all heavy metals. The bulb that is the most edible part had the least value except for Zn which was higher in the leaf. All heavy metals in the different parts of onion were generally not significantly different (P>0.05). The only metal that was slightly higher than the permissible limit was As.

The presence of heavy metals in varying concentrations in different parts of the vegetables under investigation could be attributed to the presence of these trace metals in the pesticides indiscriminately used in the study area. Previous studies have reported the presence of these metals in pesticides (Yuguda *et al*., 2015, Nazir *et al*., 2015, Shah *et al*., 2013, Singh *et al*., 2012, Guerra *et al*., 2012). The differences in the accumulation of these metals in the different parts of the vegetables under study could be attributed and not limited to their varying physiological phenomenon such as absorption rate of different metals viz a viz soil physicochemical properties, choice of plants in selecting which mineral is allocated and stored in its parts among other factors (Alloway, 1990). It is understandable however, that the roots which absorb these accumulated metals from the soil had generally higher concentrations than the stem, leaves and fruits.

3. *Bio-accumulation Potentials of Vegetables under Study*

Bioaccumulation factor (BF) of heavy metals from soil to vegetables was calculated as the ratio between the concentrations of heavy metals in the edible part of the vegetables and their respective concentrations in soil (Table 4). The BF is one of the key components of human exposure to metals through the food chain. The results obtained when the bio-accumulation potentials of the different vegetables was computed, showed that in pepper the order of heavy metals uptake followed Cr (10.20) > Cd (1.46) >Pb (0.49) > Zn (0.14) > As (0.08). That of tomato follows the same order with values as Cr (2.00) > Cd (1.96) >Pb (0.24) > Zn (0.19) >As (0.12) while in the case of onion it is of the order Cd (3.31) > Cr (1.75) > Zn (0.20) >Pb (0.18) > As (0.01). Any value of BF >1 indicates the plants as hyper accumulator and could be used in bioremediation of heavy polluted soil. The results of the BF suggest that all the vegetables under study viz, pepper, tomato and onion were hyper accumulators of Cr and Cd with the highest accumulation of Cr in pepper (10.20). Factors responsible for the differences in the accumulation of heavy metals in plant include but not limited to soil physical and chemical properties, physiological processes in individual species etc.

Table 4: Bio-accumulation Factors (BF) of Heavy Metals in the edible part of the vegetables

Vegetable	Pb	As	Cd	Cr	Zn
Pepper	0.49	0.08	1.46	10.20	0.14
Tomato	0.24	0.12	1.96	2.00	0.19
Onion	0.18	0.01	3.31	1.75	0.20

The different concentrations of these heavy metals in the soil and the resultant BF is attributed to the varying concentrations of the heavy metals in the pesticides widely used in this area and mobility of metals from soil to plant (Zurera *et al.,* 1987). Most studies though, attributed heavy metal pollution of vegetables to irrigation water from untreated sewage, industrial waste etc. (Khan *et al*., 2008), the heavy metal inputs from agrochemicals used in vegetable farms is usually excluded. This assumption could be misleading as the concentration of heavy metals from pesticides might exceed or equals the input from such polluted irrigation water. In this study, the sources of the irrigation are rain fed and bore holes sunk within the vicinity of the farms with no source of any contaminants except the agrochemicals used by farmers.

4. *Human Risk of Heavy Metal Contamination from Vegetables*

Risk to human health due to intake of metal contaminated vegetables was calculated using the hazard quotient (HQ) shown in Table 5. The USEPA (2006) maximum limit (≥1) was considered in this study. The results of HQ obtained in this study suggest high health risk from Pb, As and Cd contamination in both adults and children and only a slight health risk of Zn (1.058) contamination in children consuming onion. The HQ for Pb ranged between 1.29 to 2.22 in adults and 2.63 to 15.30 in children. In the adult's category, the HQ for As was 1.29 to 2.44 while 0.52 to 2.89 was recorded for children. The HQ of Cd for adults ranged from 0.61 to 1.95 and 2.30 to 17.89 in children. The result, thus indict these vegetables as a serious potential health risk to the communities in the study area and many other areas that consume these vegetables through market chain. The values for Zn and Cr in the vegetable under study showed no health risk in adults. The HQ showed children appeared to have higher risks than adults. When the different vegetable types in the adult category are compared, pepper had the highest HQ for Pb, As and Cd than tomato and onion with a decreasing order pepper>tomato> onion. This trend is like that of the children's category except that HQ for Pb was higher in tomatoes and onion than pepper. The order is tomato = onion > pepper.

The hazard index (HI) of consuming pepper was 6.69 for adults and 8.49 for children. For tomato HI was 4.41 in adults and 35.94 in children, whereas HI of consuming onion was 3.29 for adults and 24.01 for children. Any health risk associated with their consumption could be regarded as a national or regional health risk. This is because vegetables cultivated in the study area are transported to other parts of Nigeria in the south and to neighboring countries in the sub-Sahara.

Table 5: Hazard Quotient and Hazard index of Heavy Metals Consumed in Vegetables

Vegetable	Human Category	Hazard Quotient (HQ) of Heavy Metal					
		Pb	**As**	**Cd**	**Cr**	**Zn**	**HI**
Pepper	Adults	2.22	2.44	1.95	BDL	0.08	6.69
	Children	2.63	2.88	2.30	0.01	0.89	8.71
Tomato	Adults	1.29	1.52	1.51	BDL	0.08	4.41
	Children	15.30	1.80	17.89	0.001	0.95	35.94
Onion	Adults	1.29	1.29	0.61	BDL	0.09	3.29
	Children	15.30	0.52	7.16	BDL	1.06	24.04

NB: Hazard Quotient Values ≥1 are very Hazardous (USEPA, 2006); BDL = Below Detectable Limits

Lower HQ values <1 have been reported. This could be attributed to the region's robust regulations on heavy metal pollution (Guerra *et al.,* 2012). There were however previous reports that showed higher values than what is reported in this study. For example, Zhuang *et al*. (2008) reported higher HQ values for Pb and Cd through food consumption of rice and suggested adverse health effect as such.

The Hazard Index HI is the sum of all the HQ which determines the cumulative health risk. The results for adults showed that pepper had the highest HI (6.69) than tomato (4.41) and onion (3.29). In children, however, tomato had the highest HI (35.94). This was followed by 24.04 and 8.71 for onion and pepper respectively. Children had the greatest health risk than adults from heavy metal contaminated vegetables fumigated by pesticides.

CONCLUSION

The presence of heavy metals (Pb, Cd, Cr, As and Zn) from pesticides application in some vegetable farms was detected in pepper, tomato and onion and their corresponding farm soil. The bioaccumulation factor for the vegetables showed they are hyper accumulators of Cd and Cr. The concentrations of these heavy metals varied with the metal species and different parts of the vegetable exceeding in some instances the WHO/FAO permissible limits. In the edible parts of these vegetables, As and Cd in pepper exceeded WHO/FAO permissible limits while in tomato only As exceeded this limit.

The hazard index (HI) of all the vegetables considered in this study exceeded the high-risk limit (>1). This showed that consumption of these vegetables in the study area pose health risk from heavy metal contamination because of uncontrolled pesticides applications. This risk could be considered a national risk since all vegetables produced in the study are transported and consumed by the populace in the southern parts and neighboring countries as well. There were previous work, though using different vegetables that reported similar higher heavy metal accumulation beyond WHO/FAO maximum limits (Arora *et al*. 2008) The need for concerted effort in monitoring and regulating the use of agrochemicals in Northern parts of Nigeria being the sole producer of pepper, tomato and onion cannot be over emphasized.

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