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A survey of copper, lead, cadmium and zinc residues in cocoa beans obtained from selected plantations in Nigeria

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ABSTACT

Black pod disease is the most prevalent disease of Theobroma cacao L. The most popular means of controlling the disease among cocoa farmers in Nigeria is the use of copper-based fungicides. Due to the possibility of fraction of applied fungicide being retained in the beans as residue, the European Union set maximum residue limits for all pesticides used on cocoa during production. Since a considerable proportion of Nigerian cocoa beans is marketed in Europe, it became necessary to assess the levels of some heavy metals in cocoa beans obtained from selected plantations within three cocoa producing States in Nigeria. Ripe cocoa pods were collected from the farmers' field and processed according to standard procedures. Portion of the processed samples were digested and analyzed for Cu, Pb, Cd and Zn using Buck Scientific Atomic Absorption Spectrophotometer. Result show that, copper residues in all the samples were below maximum residue limits while cadmium was not detected in most of the samples except in few samples from Ondo State. The few samples that have detectable cadmium were below the maximum residue limit for cadmium. However, the level of Pb in most of the samples obtained from Cross River and Ondo States were moderately high. This may be due to inherent Pb content of the soils on which the crop is grown.

Keywords: Cocoa, heavy metals, black pod disease,

INTRODUCTION

Cocoa is a commodity of commerce in many West African nations of the world. West Africa produces 73% of the world cocoa production while Nigeria, currently produces 230 metric tonnes making her the fourth largest producer of cocoa in the world [1]. Cocoa consumption continues to increase globally basically due to its health benefits. Recent research activities have revealed additional more important health benefits which have further enhanced the attractiveness of cocoa products. In the last decade, studies have shown that cocoa consumption can play important role in the reduction of risks or delaying the development of cardiovascular diseases, cancer and other age-related diseases. Cocoa consumption has been positively connected to anticarcinogenic activity in human cells, hypertension, diabetes and sexual weakness as well as an aphrodisiac which stimulates the hypothalamus, which induces pleasure sensation and affects the level of serotonin in the brain [2].

However, the cultivation of cocoa in Nigeria and other cocoa producing countries encounter several challenges in which infestation by black pod disease is major [3]. The most predominant means of controlling the menace of black pod disease among Nigerian cocoa farmers is the use of copper-based fungicides of various brands. When crops are treated with pesticides, only 15% of the applied pesticide is taken by the target while the remaining is distributed within the air and the soil [4]. Due to the ability of copper to permeate the cuticle of cocoa pods after application, it was reported that, 11% of the total copper residue in cocoa beans was absorbed from the applied copper fungicide via the pod [5].

Heavy metals such as Pb, Cd, and Cu are generally regarded as environmental contaminants and their presence in foods can have some toxic effects in human. High levels of Pb in food may result to food poisoning in humans either in acute of chlorine exposure [6]. Accumulation of lead produces damaging effects in the hematopoetical, hematic, renal, gastrointestinal systems [7]. Conditions associated with increased copper in human body are arthritis, fatigue, insomnia, scoliosis, osteoporosis, heart disease, cancer, migraine, heart seizures, gum diseases, memory loss [8].

The possibility of detecting some heavy metals as residues in cocoa cannot be avoided due to the fact that, heavy metal like copper is a major component of most fungicides used on cocoa globally. The presence of Pb in soils on which cocoa is grown is another means through which Pb can get into cocoa beans. This however depends on the parent material from which the soils are formed. Due to health importance of heavy metals in human body, the European Union has set up maximum residue limits for specific heavy metals in cocoa beans meant for sales within European Union zone [9]. Stringent measures are equally attached to the maximum residue policy. One of the major measures to enforce total compliance is the outright rejection of cocoa beans that exceeds the maximum limit.

Due to the fact that, the largest proportion of cocoa beans produced in Nigeria is sold to the European Union zone, it became necessary to assess the levels of copper, lead, cadmium and zinc in cocoa beans obtained from selected cocoa producing states in Nigeria in order to determine the levels of these heavy metals and compare with the standard maximum residue limits for the various metals.

EXPERIMENTAL SECTION

Cocoa samples were collected from selected cocoa plantations in Iloro-Idanre (7^o N, 5^{o}E), Bamikemo (7^o 18'N, 4^o 54'E) and Owena (7^o 10'N, 4^o 59'E) in Ondo State; Boki-Biakwan (6^o 03'N, 8^o 53'E), Efraya (5^o 53'N, 8^o 45'E), Ajassor (5^o 52'N, 8^o 48'E), Okundi. (5^{o}57'N 8^{o}46'E) and Bendege (6^o 0'N, 8^o 49'E) in Cross River State; and Bodo (7^o 13'N, 3^o 42'E), Bangboye (7^o 12'N, 3^{o}43'E), Sotayo (7^o 12'N, 3^o 44'E), Ogunmakin (7^o 08'N, 3^o 47'E and Sora-Bale (7^{o}07'N, 3^{o}43'E) in Ogun State.

In each of the three States considered in the study, ten cocoa farms were selected at different locations. In each of the selected farms, ten cocoa trees were selected randomly and three ripe cocoa pods were collected from each of the trees. The pods were transported to the fermentation unit of Cocoa Research Institute of Nigeria, Ibadan, Nigeria where the pods were broken with wooden stick and later fermented for six days.

The fermented beans were sun-dried for six days. The beans were later oven-dried at the temperature of 60^{o}C for 48 hours until constant bean weight was attained. The beans were pulverized with ceramic mortar and pestle. 0.25g of each of the sample was digested with 10 ml of perchloric: nitric acids in a ratio of 1: 3 in a digester for 2 hours until a clear solution of the digest was attained. The sample was made up to 50ml with double distilled water and a portion of the digest was analyzed for Cu. Pb, Cd and Zn with Buck Scientific Atomic Absorption Spectrophotometer.

RESULTS AND DISCUSSION

Copper residues in cocoa beans:
Result of heavy metal analysis in cocoa beans obtained from the various locations within the three States is presented in Table 1. In samples from Ogun State, copper residue ranged from 22 – 27 mg kg-1 with an average value of 25 mg kg-1. Sample from Bangboye 1 had the lowest Cu residue while sample from Bodo 2 and Sora-Bale3 had the highest value. There was however, no significant difference in copper residue among the sample analyzed within Ogun State.

Cu residue in the beans obtained from Ondo state ranged from 16 to 31 mg kg^{-1} with a mean of 26.10 mg kg^{-1}. Samples obtained from cocoa plantation in Idanre 4 and OD 1 were significantly lower in Cu residue than the remaining samples. The highest value of Cu residue was recorded for the sample from Bankemo 1. In cocoa beans obtained from Cross River State, Cu residue ranged from 10 – 24 mg kg^{-1} with an average value of 18 mg kg^{-1}. On the other hand, cocoa beans from Ajassor 3, Adiginpo 1 and Boki-Biakwan 2 had significantly lower Cu residue than the rest of the analyzed samples.

Determination of some heavy metals in cocoa beans collected from the selected cocoa farms across Ondo, Cross River and Ogun States shows that, the average concentration of Cu in cocoa samples from Ondo State were higher than those from Cross River and Ogun States. However, none of the analyzed cocoa beans from the various States exceeded the maximum residue limit of Cu (50 mg kg^{-1}) set by the European Union. The higher average copper residue in cocoa beans from Ondo State was indicative of the correlation between Cu in soil and Cu residue in beans. From the baseline study, selected cocoa plantations in Ondo State were older than the plantations in Cross River and Ogun States. Since application of Cu-based fungicide is a regular activity among active cocoa farmers, the build-up of copper in soil and cocoa tissues might have been responsible for the higher mean value of Cu in selected plantations in Ondo State compared to the selected cocoa plantations in Cross River and Ogun States. The ranges of copper residue in cocoa beans from Ondo (16 - 31 mg kg^{-1}), Cross River (10 – 24 mg kg^{-1}) and Ogun (22 – 27 mg kg^{-1}) are much higher than the range of Cu (0.08 - 0.22 mg kg^{-1}) in cocoa beans reported by [10] and (15.22 - 24.50 mg kg^{-1}) reported by Lee and Low [11] . Chronic low-level intakes of heavy metals have damaging effects on human beings and other animals since there is no good mechanism for their elimination. Metals such as copper, lead, mercury and cadmium are cumulative poisons. They cause environmental hazards and are reported to be exceptionally toxic [12].

Lead residues in cocoa beans:

Pb residue in cocoa beans obtained from Ondo State ranged from 0.85 to 3.00 mg kg^{-1} with an average value of 1.97 mg kg^{-1}. Cocoa beans obtained from cocoa plantation in Idanre 4 had Pb residue below average and significantly lower than any other sample within the group while samples obtained from Idanre 1, Bankemo 1 and OD 1 were higher in Pb residue than the rest of the beans. Cocoa samples from selected plantations in Cross River had Pb residue ranging from 0.40 – 2.70 mg kg^{-1} with a mean of 1.66 mg kg^{-1}. Sample obtained from Okundi 2 and 3 had significantly ($P < 0.05$) higher Pb residue than the rest cocoa beans samples while cocoa samples obtained from Adiginpo 2 and Boki-Biakwan 2 had significantly ($P < 0.05$) lower Pb residue compared to other samples.

Pb concentration in cocoa samples ranged from 0.40 to 3.45 mg kg^{-1} with a mean value of 1.25 mg kg^{-1}. Cocoa beans from Bangboye 1, Sora-Bale 3 and Bodo 1 had significant ($P < 0.05$) lower quantity of Pb in them compared with the rest of the samples while sample from Sora-Bale 1 had a significantly ($P < 0.05$) higher value of Pb than the rest samples.

All the cocoa beans from Ondo State with the exception of sample from Idanre 4 had Pb residue above the maximum Residue Limit (1.0 mg kg^{-1}) set by the European Union. This implies that, 90 % of the cocoa beans from Ondo State exceeded the MRL. In cocoa beans obtained from Ogun State, 50 % of the samples had Pb residue within the maximum Residue limit while 50 % had Pb residue above the limit. On the other hand, 82% of cocoa samples obtained from Cross River State had Pb residue above the limit. The high concentration of lead (Pb) in the various beans suggests the possibility of health threat on the consumers of cocoa products made from these contaminated beans. Lead can be very harmful even at low concentration when taken over a long time [13, 14, 15]. After ingestion, the typical absorption rate of lead ranges from 3 to 80 %, whereas the typical absorption rates of dietary lead in adults and infants are 10 and 50% respectively. After absorption, lead is initially distributed to soft tissues throughout the body via blood and then deposited in bone. Lead is a chronic toxic chemical. It may cause damages to kidneys, the cardiovascular, immune, hematopoietic, central nervous and reproductive systems.

Several reports suggest that, contamination of food by Pb was due to leaded gasoline. A potentially important source of lead contamination in cocoa beans is the tetra-ethyl lead (TEL) additive in gasoline, which is still common in many African countries [16]. For example, Nigerian gasoline contains 0.4–0.8 g/L lead, which is among the highest in the world [17]; approximately 90% of the lead pollution in Nigeria is derived from the combustion of leaded gasoline, with total estimated annual lead aerosol emissions of 2,800 metric tons [18]. Those emissions are reflected in the contamination of Nigerian dusts, plants, and foods [19, 20] and the elevated blood lead concentrations noted in several studies of Nigerian people [21, 22, 23]. This ongoing contamination from leaded gasoline emissions is consistent with the report by [24] that lead concentrations of some foods grown on the African continent still exceed both WHO and the United Nation’s Food and Agricultural Organization permissible levels of 5 ng/g. Analysis of a variety of chocolate products from various global locations carried out by a Swiss group in 2002 found that the lead content of food items ranged from 0.0011 to 0.769 mg kg^{-1}, below the International Standards [25]. The low levels of lead in the products were attributed to successful phasing out of leaded gasoline in European countries when leaded fuel is burned. However, heavy metals assessment of various brands of cocoa beverages in South Western Nigeria by Shittu and Badmus, reported higher residue of Pb than the COPAL's recommendation. The issue of high Pb content of Nigerian cocoa has been raised by Cocoa Producer Alliance [26].

On the contrary, the high concentration of Pb in the investigated cocoa beans may not be a consequence of leaded gasoline due to the fact that, all the cocoa plantations where the cocoa beans were collected were far from urban areas where vehicular movement is high. The pod breaking, fermentation and sun-drying were done in an environment free of smoke from automobiles. It then suggests that, the accumulation of lead in the beans was mainly from the soil since the beans did not undergo any industrial process where metallic parts of industrial machine could be sources of Pb contamination.

In a study carried out by [27], which evaluated the contribution of different natural and industrial Pb in cocoa beans, cocoa and chocolate products, they reported that, the presence of contaminant Pb in cocoa beans shell was substantiated by the concentrations observed in the various soil profile composites through the use of isotopic Pb as tracer. They further stated that the average Pb concentration in top soil was 14.20 mg kg^{-1} which was consistent with the survey of [28], who reported the lowest lead value measured in Nigerian soils as 10 mg kg^{-1}. They concluded that, the similarities in the isotopic compositions of Pb in cocoa beans shell were indicative of a single predominant source of Pb contamination at the cocoa farms.

There are three possible sources of Pb contamination in cocoa plantations. One is the level of Pb in native soil which is determined by the parent rock materials and weathering processes; the second is from the application of old lead-acid batteries on cocoa farms by farmers whenever there was outbreak of termite attack on cocoa plantations (personal communication with farmers) and the third source is Pb impurity in applied agrochemicals and fertilizers. On overall, it might be difficult to say precisely the source of Pb in cocoa plantations but the fact remains that, the level of Pb in cocoa beans is traceable to Pb content of the soil. It has been earlier reported by Charley et al., and Chukwuma that, the contribution from the native soils may be the major source of Pb in Nigerian cocoa.

Cadmium residues in cocoa beans:
In all the samples collected from Ondo State, 50% of the sample population had cadmium residue ranging from 0.04 – 0.08 mg kg^{-1} with an average of 1.97 mg kg^{-1}(Table 1). Among the samples obtained from Ogun State, cadmium residue was only detected in cocoa beans obtained from Sotiya at a concentration of 0.14 mg kg^{-1}. There was however, no detectable cadmium residue among cocoa beans collected from Cross River State.

Analysis of cadmium residue in the cocoa beans shows that, 50% of cocoa beans obtained from Ondo State had detectable cadmium ranging from nd – 0.08 mg kg^{-1}, 10 % of samples from Ogun State (nd - 0.14 mg kg^{-1}) while none of the samples from Cross River State had Cd residue (Table 1). Data on Cd residue in cocoa beans is scarce probably due to the fact that, Cd contamination in agricultural soils is rare. However, due to health challenges experienced globally through food contamination, the issue of heavy metal contamination in cocoa beans is taken with all seriousness especially by the European Union who is the main importer of cocoa beans from West Africa.

In recent time, European food Safety Authority in a scientific report has shown that, sugar and confectionery was seen to contribute 4.3% of adult's dietary exposure to cadmium. The figure increased to 7.3% in children and cocoa products has been implicated as been responsible for Cd in confectionery [29].

According to Brussel, cadmium is a substance which poses a risk to human health because it can lead to kidney failure, bone problems and reproductive difficulties [30]. Consequently, the European Union has set a maximum residue limit of 0.5 mg $kg^{-1}$for Cd in cocoa powder. The values obtained for Cd in all the analyzed cocoa beans show that, cadmium residue in Nigerian cocoa beans is well below the maximum residue limit. However, few data (though old) available for Cd residues in cocoa beans show that, Nigerian cocoa have very low Cd level (nd – 0.14 mg kg^{-1}) compared with cocoa beans in other countries. [31] reported 0.48 – 1.83 mg kg^{-1} Cd in Malaysian cocoa; Lee and Low reported 0.89 – 1.10 mg kg^{-1} Cd in Malaysian cocoa and [32] reported average of 0.183 mg kg^{-1} Cd in cocoa beans analyzed in Europe.

Rock phosphate fertilizer has been implicated as the main agricultural input source of cadmium in the soil [33]. Although cadmium occurs naturally as a contaminant in all phosphate rock, the concentrations however, vary considerably depending on the origin of the material. Igneous rock or apatite (found in Russia - 0.3mg kg^{-1} and South Africa - 0.1 mg kg^{-1}) has low concentrations of cadmium (often less than 1 mg per kg P_2O_5). Sedimentary rock, which accounts for some 85-90 % of world production, contains cadmium in concentrations ranging from less than 20 to more than 200 mg per kg P_2O_5 (Togo-162 mg kg^{-1}; Algeria- 60 mg kg^{-1}; Morocco-100 mg kg^{-1}; North Carolina, USA-166 mg $kg^{-1}$and Senegal- 203 mg kg^{-1}) [34]. Cadmium analysis of Sokoto rock phosphate revealed a mean value of 0.08 mg kg^{-1} P_2O_5. The mean value of Cd in Sokoto rock phosphate is lower than cadmium content of many phosphate rocks in the world. This implies that, the application of Sokoto rock phosphate as fertilizer or soil conditioner may not lead to Pb contamination in cocoa plantations.

Zinc residues in cocoa beans:
Zinc concentration ranged from 78 - 175 mg kg^{-1} with a mean of 109 mg kg^{-1}. Cocoa beans obtained from OD 1 and 2 were significantly higher in Zn content than the remaining samples. Zinc concentration in the cocoa beans was lowest in sample from Bendege 1 (77.00 mg kg^{-1}) and highest in sample obtained from Okundi 1 (111.00 mg kg^{-1}). There was however, no significant difference in Zn content among the various sample analyzed. Zinc content of cocoa beans obtained from Ogun state ranged from 79 - 180 mg kg^{-1} with a mean value of 108 mg kg^{-1}. Cocoa obtained from Sora Bale 4 had the least concentration of zinc while sample from Sora-Bale 1 had the highest concentration.

The average zinc residue in cocoa beans from the three States considered in the study is higher than zinc content reported in groundnut cake (45.5mg kg^{-1}) and Melon cake (19.3 mg kg^{-1}) collected from selected motor parks in Ogun State, Nigeria. The difference may be due to high inherent ability of *Theobroma cacao* as a crop to absorb zinc from soil. Different species of crops have different capacity to absorb micronutrients from soil. This is due to the fact that, no record at the moment has shown that, cocoa plantations in Nigeria are contaminated with zinc. Available data on heavy metal contaminations in cocoa plantations in Nigeria showed that, the source of zinc in most cocoa plantations has been reported to be from parent materials from which the soils were made [35, 36]. Knowledge of zinc toxicity in humans is minimal [37]. The most important information reported is its interference with copper metabolism [38]. The symptoms that an acute oral Zn dose may provoke include: tachycardia, vascular shock, dyspeptic nausea, vomiting, diarrhea, pancreatitis and damage of hepatic parenchyma [39]

Linear correlation and regression among the various metals considered in the study:
Linear correlation of the various heavy metals considered in the study is shown in Table 2. Result shows positive correlation among Pb, Cu and Zn residues in cocoa beans obtained from Cross River State. There was however, no significant correlation among heavy metal contents of the beans. In samples obtained from Ondo State, Zn residue had negative correlation with Cu while there was positive correlation among the rest metal content of the beans. The residue of Pb in cocoa beans obtained from Ogun State had positive correlation with Zn at significant ($P < 0.05$) level while Cu had negative correlation with Zn. Except for samples obtained from Cross River State, copper residue in cocoa beans obtained from the various study area had negative correlation with zinc. This is an indication that, zinc distribution in cocoa beans reduces with copper increase in beans. A similar observation was made in the study of [40] where Cu negatively affected the distribution of Zn in cocoa beans. The antagonistic effects of copper on Zn concentration in cocoa beans may be due to the interference of copper with the transport mechanism within cocoa plant. The findings of Pearson et al., showed that, excess Cu reduced phloem transport of zinc into *T. aestivum* grains. The authors concluded that, the reduction was because copper and zinc compete for the same phloem loading sites in plant. In a recent study carried out by Aikpokpodion et al., where various concentrations of copper fungicides was used on cocoa, it was observed that, zinc distribution decreased in cocoa beans with increase in copper concentration applied on cocoa pods.

The linear regression of the various heavy metal residues in cocoa beans obtained from the studied areas is presented in figures 1-12. Among the heavy metal residues, only Pb and Zn in cocoa samples obtained from Ogun State showed significant linear relationship (R^2 = 0.679). This suggests that, no direct link or relationship between applied copper-based fungicides and Pb, Cd and Zn exist in cocoa plantations.

Table 1: Concentration of various heavy metal residues in cocoa beans obtained from Ogun, Ondo and Cross River States.

Ogun State					Ondo State mg kg^{-1}					Cross River State				
Location	Zn	Pb	Cu	Cd	Location	Zn	Pb	Cu	Cd	Location	Zn	Pb	Cu	Cd
SoraBale 1	180^{a}	3.45^{a}	23^{a}	nd	Idanre 1	96^{b}	2.95^{a}	36^{a}	0.08^{b}	Bendege 1	77^{a}	1.40^{c}	20^{a}	nd
SoraBale 2	132^{ab}	2.30^{b}	25^{a}	nd	Bamkemo1	110^{b}	3.10^{a}	41^{a}	0.04^{b}	Adiginpo 2	87^{a}	0.45^{e}	21^{a}	nd
SoraBale 3	97^{b}	0.45^{e}	27^{a}	nd	OD1	175^{a}	3.00^{a}	19^{c}	nd	Ajassor1	110^{a}	1.60^{c}	23^{a}	nd
Bangboye1	82^{b}	0.40^{e}	22^{a}	nd	OD 2	171^{a}	2.50^{ab}	25^{b}	nd	Okundi 1	111^{a}	2.70^{ab}	22^{a}	nd
Bodo 1	85^{b}	0.50^{e}	23^{a}	nd	Idanre 2	95^{b}	1.70^{b}	26^{a}	0.06^{a}	Efraya	103^{a}	1.30^{cd}	24^{a}	nd
Bangboye2	87^{b}	0.70^{d}	24^{a}	nd	Bamkemo2	94^{b}	1.50^{b}	27^{b}	nd	Okundi 2	88^{a}	2.10^{a}	22^{a}	nd
SoraBale 4	79^{b}	0.90^{d}	26^{a}	nd	Idanre 3	86^{b}	1.30^{b}	26^{b}	0.06^{b}	Boki-Biakwan1	97^{a}	2.45^{b}	15^{b}	nd
SoraBale 5	83^{b}	2.00^{b}	26^{a}	nd	Idanre 4	89^{b}	0.85^{c}	16^{c}	0.07^{b}	Ajassor 2	94^{a}	1.73^{c}	11^{c}	nd
Bodo2	126^{ab}	1.75^{bc}	27^{a}	nd	Owena 1	98^{b}	1.10^{b}	28^{b}	nd	Adiginpo 1	93^{a}	1.00^{d}	10^{c}	nd
Sotiya	137^{ab}	1.45^{c}	25^{a}	0.14	Owena 2	78^{b}	1.70^{b}	31^{b}	nd	Boki-Biakwan2	81^{a}	0.40^{e}	11^{c}	nd
Mean	108	1.25	25	0.14	Mean	109	1.97	26	0.062	Mean	93	1.66	18	nd
Std	34	1.03	3	-	Std	34	1.10	3	0.02	Std	11	1.64	4	nd

Key: nd- not detected; std – standard deviation
Same alphabets on the same column are not significantly different.

Table 2: Linear correlation among heavy metal residues in cocoa beans

	Cross River			Ondo				Ogun		
	Pb	Cu	Zn	Pb	Cu	Zn	Cd	Pb	Cu	Zn
Zn	0.544	0.373	1.0	0.592	-0.278	1.0	-0.440	0.82**	-0.71	1.0
Pb	1.0	0.270	0.544	1.0	0.479	0.592	-0.043	1.0	0.018	0.82**
Cu	0.270	1.0	0.373	0.479	1.0	-0.278	0.104	0.018	1.0	-0.71
Cd	-	-	-	-0.043	0.104	-0.440	1.0	-	-	-

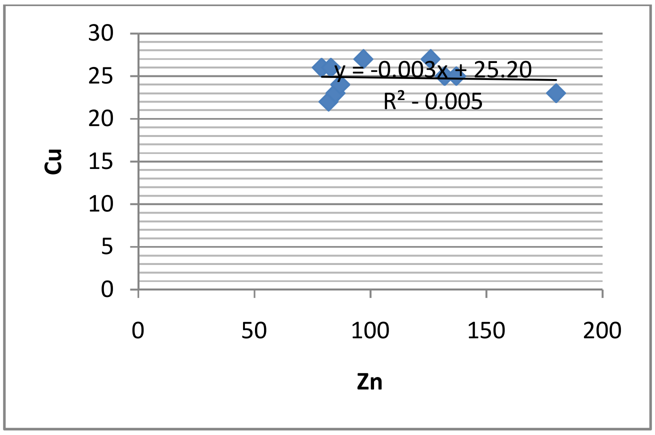

Figure 1: Relationship between Cu and Zn in beans from Ogun State

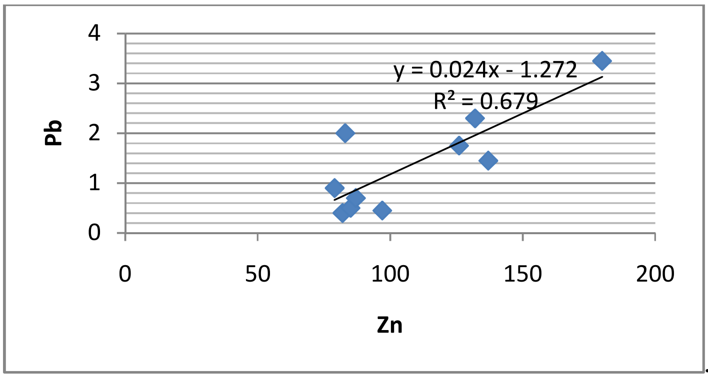

Figure 2: Relationship between Pb and Zn in beans from Ogun State

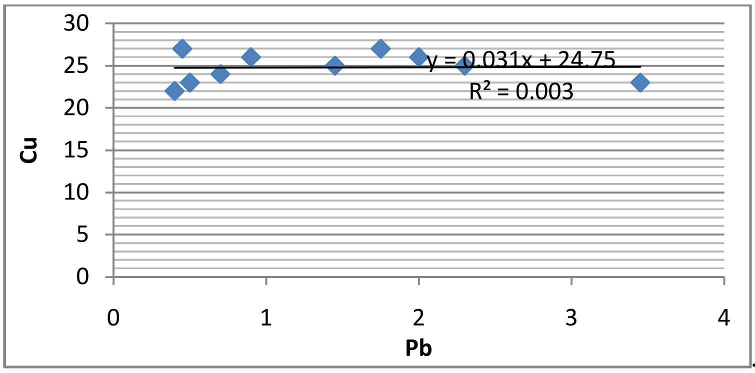

Figure 3: Relationship between Cu and Pb in beans from Ogun State

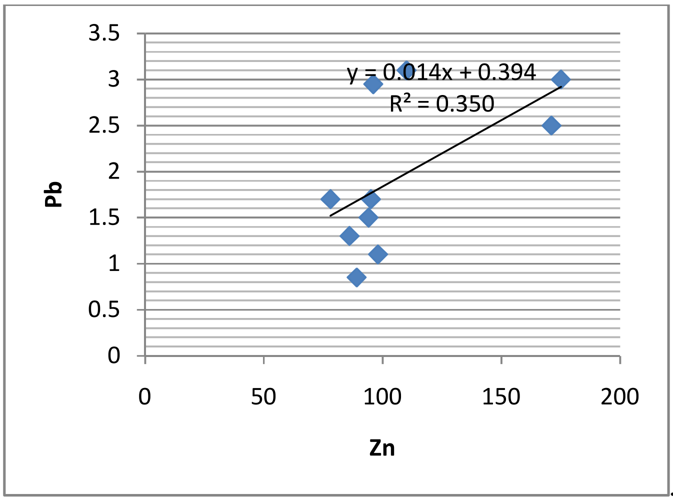

Figure 4: Relationship between Pb and Zn residues in cocoa from Ondo State

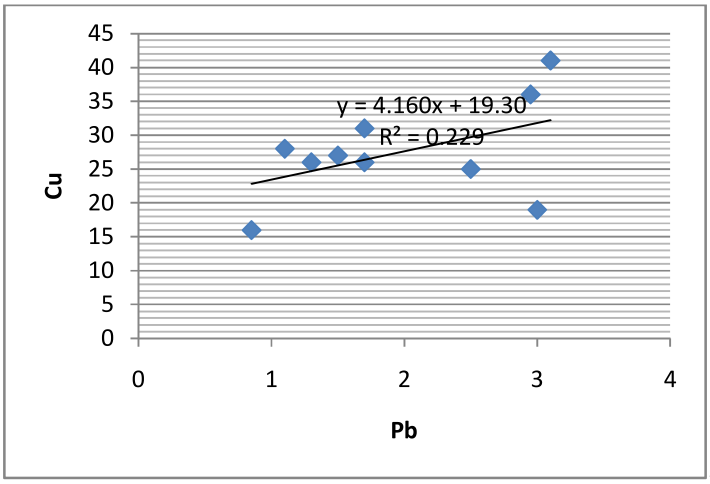

Figure 5: Relationship between Cu and Pb residues in cocoa from Ondo State

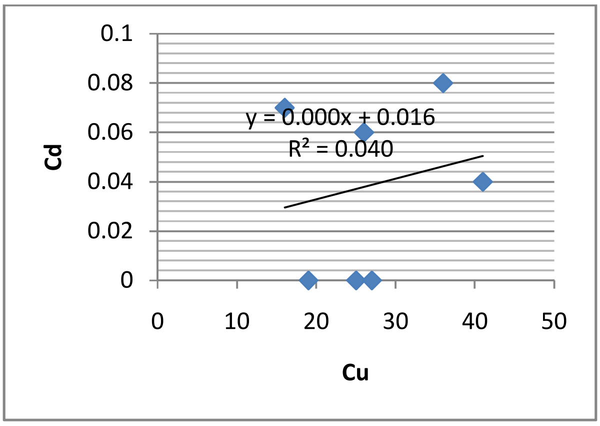

Figure 6: Relationship between Cu and Cd residues in cocoa from Ondo State

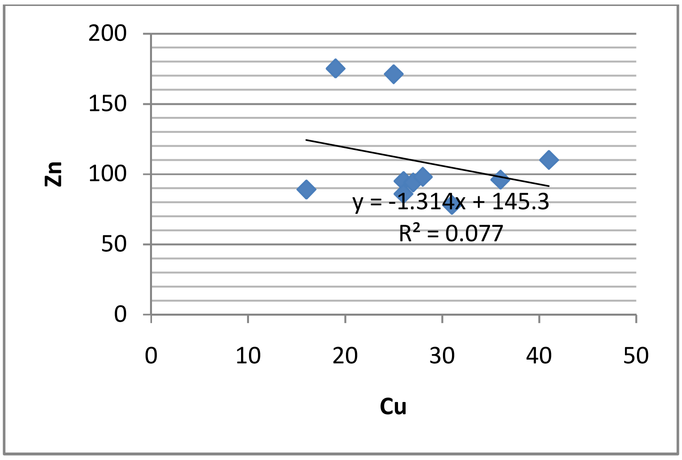

Figure 7: Relationship between Cu and Zn residues in cocoa from Ondo State

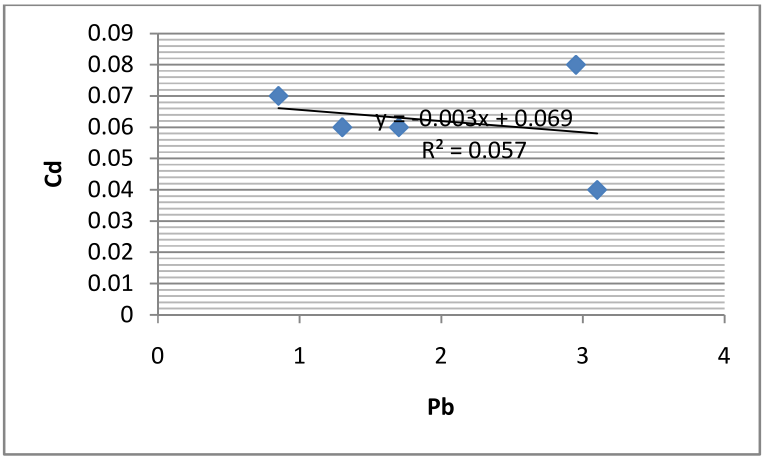

Figure 8: Relationship between Cd and Pb residues in cocoa from Ondo State

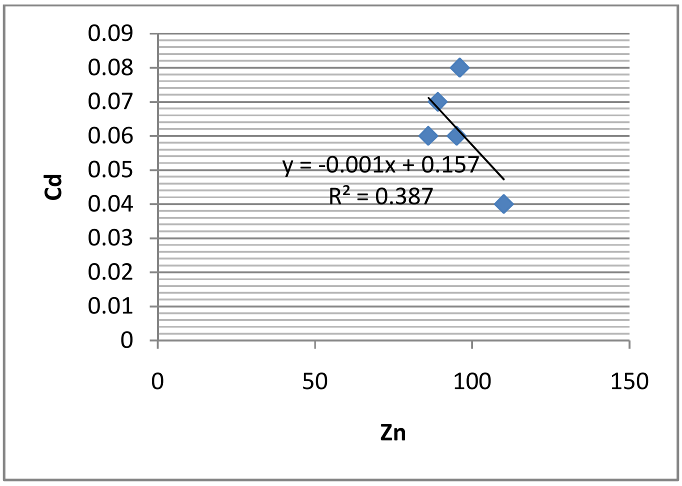

Figure 9: Relationship between Cd and Zn residues in cocoa from Ondo State

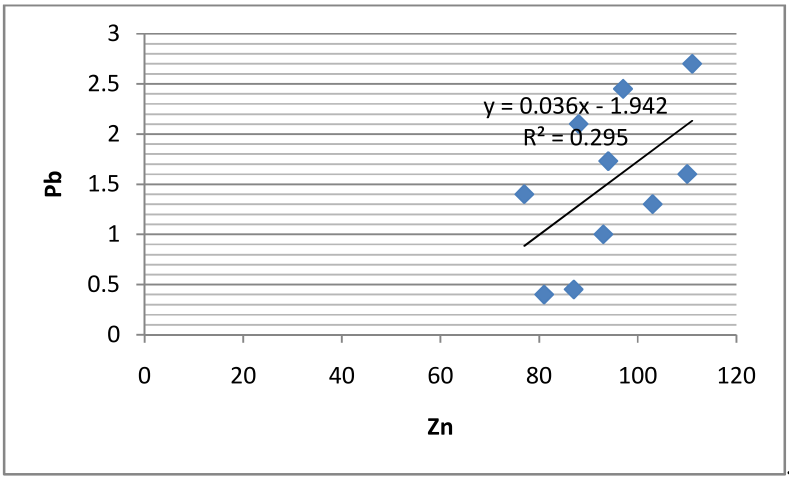

Figure 10: Relationship between Pb and Zn residues in cocoa from Cross River State

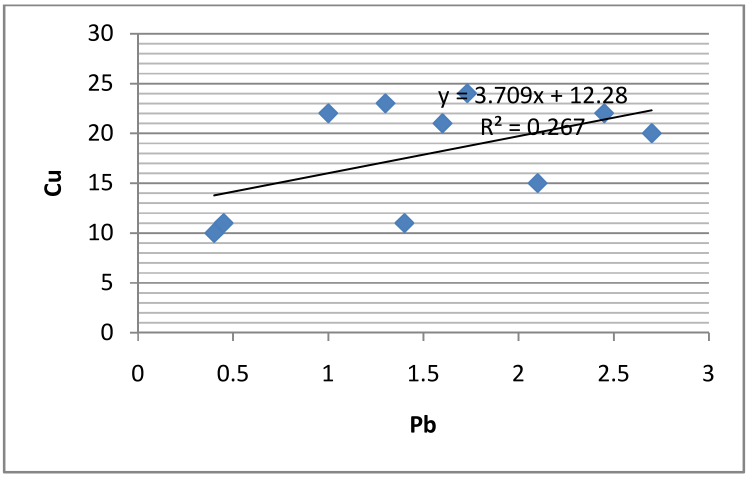

Figure 11: Relationship between Cu and Pb residues in cocoa from Cross River State

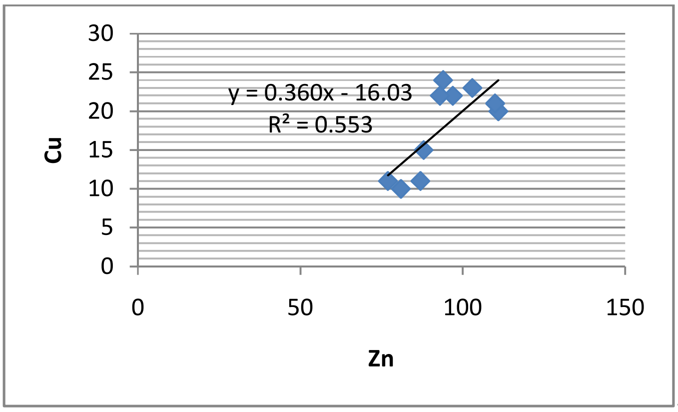

Figure 12: Relationship between Cu and Zn residues in cocoa from Cross River State

CONCLUSION

The study has shown that, cocoa beans obtained from the selected plantations in Ondo, Cross River and Ogun State, had copper residue below the maximum residue limit. Cadmium was not detected in most of the cocoa beans and those samples with detectable cadmium were much lower than the maximum residue limit set for cadmium in cocoa. However, lead residue in samples from Cross River and Ondo States were moderately high.

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REFERENCES

[1] www.worldcocoafoundation.org/about-cocoa/cocoa-market-statistics
[2] EO Afoakwa, *J. clin. Nutri.* **2008**, 21(3):107-113
[3] SO Agbeniyi; AR Adedeji, *In Proceedings of 14th International Cocoa Research Conference* Malaysia, **2003**, 14:1377-1380
[4] MV Leonila (2002) *University of the Philippines at Los banos Bulletin*., **2002**, 5:1-5
[5] PE Aikpokpodion; L Lajide; AF Aiyesanmi, *World Journal of Agric Science*, **2013**, 9(1):10-16
[6] TA Shittu; BA Badmus, *J. food composition and Anal.*, **2009**, 22:212-217
[7] RPM Correia; E Oliveira; PV Oliveira *Analytical Chimica Acta* **2000**, 405:205-211
[8] TY Lanre-Iyanda; IM Adekunle I.M, *Afr. J. Food, Agric. Nutr. and Dev*., **2012**, 12(3):6156-6169
[9] EU (European Union) Commission Regulation (EC) No 396/2005
[10] JK Koka; DK Dodoo; PK Kwakye; J Kambo-Dorsa *J. Chem. and Pharm. Research* **2011**, 3(4):467-471
[11] S Rehman; SM Husnain, *Trace Element Analysis*, **2012**, 1:1-11
[12] G Ellen; JW Loon; K Tolsma, *Z. Lebensm Unters Forsch*, **1990**, 190(1):34-39
[13] U Celik; J Oehlenschlager, *Food Chemistry*., **2004**, 87:343-347
[14] M Tuzen, *Food Chemistry*, **2003**, 80:119-123
[15] RB Voegborlo; AM El-Methnani; MZ Abedin, *Food Chemistry*, **1999**, 67: 341-345
[16] JO Nriagu; CC Jinabhai; R Naidoo, *Science of Total Environment*,. **1996**, 191:69–76
[17] OJ Ogunsola; AF Oluwole; OI Osubiojo; MA Durosinmi; AO Fatusi; W Ruck, *Science of Total Environment*., **1994**, 146/147:111–116.
[18] IB Obioh; AF Oluwole; FA Akeredolu, *CEC Consultants*, **1993**, 2: 271–274.
[19] PC Onianwa; A Egunyomi, *Environmental Pollution* B., **1983**, 5:71–81.
[20] JO Nriagu, *Science of Total Environment*. **1992**, 121:1–37.
[21] O Ademuyiwa;T Arowolo; DA Ojo; OO Odukoya; AA Yusuf; TF Akinhanmi, *Trace Element Electrol*., **2002**, 19:63–69.
[22] J Nriagu; NT Oleru; C Cudjoe; A Chine, *Science of Total Environment*, **1997**, 197:13–19.
[23] FO Omokhodion, *Science of Total Environment* , **1994,** 151:187–190
[24] TE Bahemuka; EB Mubofu, *Food Chemistry*. **1999**, 66: 63–66.

[25] S Mounicou; J Szpunar; D Andre, *Food Additives and Contaminants.* **2003**, 20: 343-352
[26] COPAL (Cocoa Producer Alliance). **2004** ,http://www.copal-cpa.org/lead.html
[27] WR Charley; JO Nriagu; JK Aggarwal; TA Arowolo; K Adebayo, *Environmental Health Perspectives* **2005**, 113(10):1344-1348
[28] C Chukwuma, *Ambio.* **1997**, 26:399–402
[29] www.confectionerynews.com)
[30] http/agritrade.cta.int/en/layout
[31] G Knezevic, G, *Disch. Lebensm-Rundsch* **1982**, 75(10):305-306
[32] EFSA (European Food Safety Authority) *EFSA Journal* **2012**, 10(2):2551
[33] S Satarug; JR Baker; S Urbenjapol; M Haswell-Elkins; PEB Reilly; DJ Williams; MR Moore, *Toxicology Letters* **2003**, 137:65-83
[34] FH Oosterhuis; FM Brouwer; HJ Wijnants, *Final Report to the European Commission* **2000**, 85p
[35] PE Aikpokpodion *J. Appl. Biosciences* **2010**, 33:2037-2046
[36] PE Aikpokpodion; L Lajide; AF Aiyesanmi *American-Eurasian J. Agric. & Environment Sci*. **2010**, 8(3)268-274
[37] E Islam; X Yang; Z He; Q Mahmood *J. Zhejiang University Science B* **2007**, 8(1): 1-13
[38] A Barone; O Ebesh; RG Harper; RA Wapnir *J. Nutr* **1998**, 128(6):1037-1041
[39] MJ Salgueno; M Zubillaga; A Lysionek; MI Sarabia; R Caro; TD Paoli; A Hager; R Weill;J Boccio *Nutr. Res* **2000**, 20(5): 737-755
[40] PE Aikpokpodion; CI Iloyanomon; SM Omotoso, *Proc. 44th Annl. Conf. Agric. Soc. Nigeria* **2010**, 44:1203-1208