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CONCENTRATIONS AND HEALTH RISK ASSESSMENTS OF HEAVY METALS IN CRUDE PALM OIL PRODUCED IN SOME AREAS OF THE NIGER DELTA, NIGERIA

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ABSTRACT
Concentrations of Cu, Pb, Cr, Cd, Ni, Fe, Zn, Mg, Ca, K and Na were determined in crude palm oil samples obtained from some selected areas in the Niger Delta region of Nigeria using Atomic Absorption Spectrophotometer and Flame Photometer. The mean concentrations of the metals (mg/100g) in the samples varied significantly ($p < 0.05$) and were: 7.23-24.20 (Fe); 0.55-1.15 (Cu); BDL-0.45 (Cr); 3.73-17.03 (Zn); 0.25-1.03 (Ni); 0.03-0.78 (Pb); 0.15-1.15 (Cd); 12.28-71.53 (Na); 71.03-146.03 (Ca); 14.90-232.00 (Mg); and 21.23-91.53 (K). The estimated daily intakes (EDIs) for Cr, Ni, Pb and Cd (0.0002 – 0.044, 0.024-0.099, 0.003-0.076 and 0.015-0.112, µg/kg bw /day, respectively) were within tolerable levels. The individual and combined metal target hazard quotients had values that were less than one. Consequently, the crude palm oil in the study area were found to be safe for consumption.

Key words: Heavy metals, Estimated daily intake, Palm Oil, Risk assessment, Niger Delta

INTRODUCTION

Oil palm (*Elaeis guineensis* Jacq. (*Arecaceae*) is a versatile monoecious oil bearing plant that thrives in tropical and subtropical regions of the world. Oil palm requires climatic conditions of between 1800 to 5000 mm of rainfall per year, temperature range of 17-28 °C and relative humidity above 75% (Embrandiri *et al.*, 2012, Poku, 2002). The oil palm is propagated by seedling and requires nursery period of 9-12 months before it is planted. Its fruiting begins from between 3-5 years and reaches optimal yield at 10 years after cultivation. It is an economic tree that has a life span of about 200 years, but its economic life is usually between 20 to 30 years (Matthew, 2009)

Nigeria is currently the fifth world's leading producer of palm oil (Ohimain and Izah, 2013). The palm oil industry is a major agro-based enterprise in Nigeria especially in the southern part where palm oil trees are found both in the wild and plantations (Nwaugo *et al*., 2008). The domestic consumption of palm oil in Nigeria in 2016/2017 and 2017/2018 were about 1.34 and 1.40 million metric tons (Statista, 2018). Palm oil processing is carried out using large quantities of water in mills where crude or virgin oil is extracted from the palm fruits and is widely used in African cooking. The refined edible palm oil is utilized in food processing activities (Adepoju-Bello *et al.*, 2012).

Vegetable oils may be contaminated by heavy metals through migration from arable soil (use of fertilizer, location of plantation by highways), technological/ production processes, packaging and storage (Szyczewski, 2016; Izah and Ohimain, 2016, Adepoju-Bello *et al.*, 2012)

The knowledge of metals in foods is essential for calculating the dietary intakes of essential metals and evaluation of human exposure to toxic elements (Iwegbue, 2010). Heavy metals such as Cr, Mn, Cu, Fe and Zn play important biochemical roles in the life processes of many organisms, hence, their presence in foods consumed by humans in trace amounts is essential, however, toxic effects are observed at high concentrations of the metals.

Iron is required for the production of red blood cells, but at high concentrations Fe and Mn can cause pathological effect such as the iron oxides deposition in Parkinson's disease (IOM, 2010). Excess Cu has been associated with liver damage and Zn may produce adverse nutrient interactions with Cu. Also, excess Zn reduces immune function and the levels of high density lipoproteins (Spears, 2000). Nickel is involved in the synthesis of enzymes that are needed in the formation of nucleic acids such as deoxyribonucleic acid (DNA). It is, however, highly toxic at high concentration, and can cause gastrointestinal distress, increase red blood cells and reduce lung functions (Lu *et al.*, 2005). Chromium toxicity is very dependent on the species and oxidation states present. It is normally found in less toxic trivalent state in foods and is poorly absorbed in the gastrointestinal tract. Chromium has been reported to have beneficial effects on type II diabetes (Hague *et al.*, 2008). Other metals such as Pb and Cd are toxic even at low concentration and are not known to have any important biological properties in humans. Lead is known to induce renal tumors, reduce cognitive development, and increase blood pressure in adults. Other symptoms of Pb toxicity include gastrointestinal disorders and some liver impairment. Cadmium may induce kidney dysfunctions, osteomalacia and reproductive deficiencies. It can also cause damage to the central nervous system and produce psychological disorder (Strömgren, 1998).

In order to assess the safety levels of consumers of palm oil from this region, this study evaluated the heavy metals and some essential mineral elements in palm oil from small- scale palm oil processing mills in some areas in Bayelsa state and Elele, Ahoda west Local Government Area of Rivers State within the Niger Delta of Nigeria.

MATERIALS AND METHODS

The study area and Sample collection.

Palm oil samples were collected from a total of nineteen small-scale palm oil production mills situated along high ways and rivers of Ahoda West and East in Rivers State and the local palm oil production mills situated in the rural areas of Ogbia and Yenegoa Local Government Areas in Bayelsa State, Niger Delta Region of Nigeria. Nine samples (A-I) were obtained from oil palm mills in Bayelsa State, and ten samples (J-S) were collected from the mills situated in Rivers State. The samples were collected in aseptically cleaned polyethylene bottles which were tightly covered and kept in the dark, prior to analysis. Appropriate quality assurance procedures and precautions were taken to ensure reliability of the results.

Reagents

All reagents used were of analytical grade. Working standards of Fe, Cu, Cr, Zn, Ni, Pb, and Cd, were prepared by diluting concentrated (1000 mg/L) stock solution of the standard elements (Merck, Darmstadt, Germany) with 0.25 M nitric acid.

Sample preparation

The palm oil samples were digested using concentrated nitric acid (HNO_3, > 90%, 1.48 g/mL at 20 °C) and perchloric acid ($HOCl_4$, 70%, 1.664 g/mL at 25 °C) at ratio 1:1 (v/v) by standard methods (AOAC, 2005). The mineral elements

(Fe, Cu, Cr, Zn, Ni, Pb, Cd and Mg, Ca, K and Na) were determined by using atomic absorption spectrophotometer (AAS) and flame photometer (FP).

Chemical analysis

The digested samples were analyzed in triplicates using AAS (APHA 301A, model: 5100 PC, Perkin-Elmer, Boston, USA) for the mineral elements. Calibration of the AAS was done using working standards prepared from commercially available metals standard solution (Merck, Darmstadt, Germany). The most appropriate wavelength, hollow cathode lamp current, gas mixture flow rate, slit width and other AAS instrument parameters for metals were selected as provided in the instrument guides and background correction was used during the analysis. Procedural reagent blank test was carried out over the entire procedure. Measurements were made within the linear range of working standard used for calibration. The results were expressed as mean ± standard deviation (SD).

Estimated Daily Intake of Metals

The estimated daily intake was evaluated for individual metal based on the consumption of 28 g of palm oil per kg body weight per day using equation 1

$$EDI = \frac{Ef * Ep * FR * Cf * Cm}{WAB * TA} \quad \text{.....1}$$

Where EDI is the Estimated Daily Intake
E_F – the exposure frequency (365 day/year)
E_D – the exposure duration equivalent to life Time (60-64 years and mean value of 62 years was used)
F_R – the fresh food ingestion rate (g/person/day) which was considered to 28g for palm oil;
C_F – the conversion factor (0.208) for fresh weight
C_m – the heavy metal concentration in foodstuff (mg/kg/Dw),

WAB- the average body weight (bw) (average adult body weight was considered to be 60 kg)

T_A – the average exposure time for non-carcinogen (= E_F * E_D).

The Health risk Index (HRI) for a consumer of metal contaminated palm oil was assessed by using equation 2

$$HRI = \frac{EDI}{RfD} \quad \text{.....2}$$

Where EDI- is Estimated Daily Intakes
R_fD- Reference Oral Dose (for Cr = 1.5 x 10^{-3}, Cu = 4.0 x 10^{-2}, Zn =3.0 x10^{-1}, Fe = 7.0 x10^{-1}, Ni = 2.0 x10^{-2}, Mn = 1.4 x 10^{-1} Pb = 1.5, and Cd = 1.0 x 10^{-3}).

Evaluation of the Hazard Target Quotient of Heavy Metals in Palm Oil Samples

The Target Hazard Quotients were evaluated for the metals based on the consumption of 28 g of palm oil per kg body weight per day using the formula in equation 3.

$$THQ = \frac{Efr * EDtot * SFI * MCSinorg * 10^{-3}}{RfD * BWa * ATn} \quad \text{.....3}$$

Where
EF_r – the exposure frequency (day/year) (365days used)
ED_{tot} – Exposure duration (year) (60-64yrs) (62 yrs used)
SFI- the mass of selected dietary ingested (g/day) 28g/day for palm oil
MCS_{inorg} – the concentration of inorganic species in the dietary components (mg/kg wet weight) for each of the elements determined.
RF_D- Oral reference dose (mg/kg)
BWa – average adult body weight = 60kg used
ATn- average time for non-carcinogens (Days) = (EF * ED_{tot})
10^{-3}- unit of conversion factor
RFD (in Equation 2)

Statistical analysis

All the data obtained in the study were subjected to statistical treatment such as analysis of variance (ANOVA) and post-hoc analysis of Tukey's least

significance difference test. (IBM-SPSS, 15.0). Differences at the $P < 0.05$ level were considered significant.

RESULTS AND DISCUSSION

The results of the mineral analysis of the palm oil samples are presented in Tables 1 and 2. The average of mean values for Fe in this study was from 7.23 to 24.20 mg/100g. Samples C and R had the lowest and highest concentrations of Fe respectively. The concentrations of Cu, Cr, Zn, Ni, Pb and Cd were 0.55-1.15, BDL-0.45, 3.73-17.03, 0.25-1.03, 0.03-0.78, and 0.15-1.15 mg/100g respectively. The predominant macro mineral nutrient in the palm oil samples analyzed were Na, Ca, Mg and K with the range of mean values: 12.28-71.53, 71.00 -146.03, 14.90-232.00 and 21.23-91.53 (mg/100g) respectively.

Table 1: Concentrations of Heavy metals (mg/100g) of palm oil samples

SAMPLE	Fe	Cu	Cr	Zn	Ni	Pb	Cd
A	17.03 ± 0.01	1.15 ± 0.01	0.45 ± 0.01	9.15	0.58	0.40	1.15
B	16.60 ± 0.02	0.98 ± 0.08	BDL	7.73	0.50	0.45	0.45
C	7.23 ± 0.01	0.78 ± 0.01	0.23 ± 0.01	4.98	0.58	0.15	0.48
D	12.40 ± 0.03	1.03 ± 0.01	0.23 ± 0.01	9.90	0.35	0.28	0.40
E	19.90 ± 0.01	0.70 ± 0.01	BDL	10.03	0.40	0.45	0.30
F	17.20 ± 0.05	0.98 ± 0.01	0.25 ± 0.01	3.73	0.39	0.15	0.23
G	22.28 ± 0.00	0.73±0.01	BDL	7.40	1.03	0.48	0.35
H	11.03 ± 0.01	0.75 ± 0.01	BDL	10.45	0.40	0.78	0.25
I	21.03 ± 0.01	0.55 ± 0.01	BDL	17.03	0.25	0.48	0.45
J	14.53 ± 0.01	1.15 ± 0.01	0.03 ± 0.01	7.23	0.45	0.48	0.68
K	14.53 ± 0.06	0.90 ± 0.01	BDL	15.28	0.75	0.45	0.15
L	17.10 ± 0.01	1.03 ± 0.01	0.10 ± 0.01	9.20	0.68	0.05	0.40
M	20.60 ± 0.01	0.80 ± 0.08	0.05 ± 0.01	17.03	0.48	0.03	0.40
N	12.40 ± 0.01	0.83 ± 0.01	0.25 ± 0.01	10.40	0.53	0.35	0.30
O	22.78 ± 0.07	0.78 ± 0.01	0.15 ± 0.01	5.70	0.48	0.50	0.30
P	12.40 ± 0.01	1.01 ± 0.04	0.15 ± 0.01	12.40	0.45	0.40	0.45
Q	23.53 ± 0.01	1.15 ± 0.01	0.03 ± 0.01	10.35	0.53	0.28	1.03
R	24.20 ± 0.01	0.73 ± 0.01	0.23 ± 0.01	12.28	0.28	0.45	0.30
S	9.95 ± 0.01	0.75 ± 0.01	BDL	6.05	0.50	0.40	0.28

The above figures are mean values of three replicate determinations. BDL = Below detection limit. (< 0.0001-0.0005) Standard Deviation for Zn, Ni, Pb and Cd in all samples are ± 0.01

Table 2: Mineral content of palm oil samples (mg/100g)

SAMPLES	Na	Ca	Mg	K
A	21.03±0.011	86.05 ±0.011	24.03 ± 0.011	39.73 ± 0.011
B	46.03±0.011	117.03±0.008	46.15 ± 0.016	46.03 ± 0.016
C	71.53±0.011	99.03 ±0.008	50.35 ± 0.011	37.40 ± 0.008
D	12.28 ± 0.01	121.03 ± 0.01	24.53 ± 0.02	46.03 ± 0.01
E	24.10 ± 0.01	71.03 ± 0.01	21.23 ± 0.02	35.40 ± 0.02
F	17.03 ± 0.01	96.00 ± 0.10	16.03 ± 0.02	65.45 ± 0.02
G	21.03 ± 0.01	96.03 ± 0.01	14.90 ± 0.01	37.40 ± 0.01
H	24.60 ± 0.01	119.78 ± 0.01	24.48 ± 0.01	21.23 ± 0.02
I	22.28 ± 0.01	102.15 ± 0.01	22.15 ± 0.02	24.65 ± 0.01
J	24.90 ± 0.01	127.40 ± 0.01	21.23 ± 0.02	51.53 ± 0.02
K	24.65 ± 0.01	74.90 ± 0.06	16.68 ± 0.01	34.53 ± 0.02
L	21.03 ± 0.01	97.10 ± 0.01	232.00 ± 1.58	31.15 ± 0.01
M	15.28 ± 0.01	146.03 ± 0.01	19.90 ± 0.02	91.53 ± 0.02
N	22.40 ± 0.01	72.15 ± 0.01	24.60 ± 0.01	50.40 ± 0.02
O	22.15 ± 0.01	99.15 ±0.02	16.73 ± 0.02	23.23 ± 0.02
P	19.90 ± 0.01	124.90 ± 0.01	21.23 ± 0.02	27.15 ± 0.01
Q	23.20 ± 0.02	82.40 ± 0.02	20.30 ± 0.01	72.03 ± 0.02
R	46.15 ± 0.01	98.03 ± 0.02	14.90 ± 0.01	21.23 ± 0.02
S	17.90 ± 0.01	71.15 ± 0.02	19.90 ± 0.02	37.40 ± 0.01

The above figures are mean values of three replicate determinations. BDL = Below detection limit. (< 0.0001-0.0005)

Estimated dietary intake of metals

The estimated daily intakes of the studied metals in μg/kg bw/day are presented in Table 3. The recommended dietary allowance values for Fe ranged from 10 to 18 mg/day/ person (Table 4). The estimated intake of Fe from consumption of palm oil samples from the Niger Delta, Nigeria, ranged from 0.70 to 2.35 μg/kg bw/day. The percentage intake of Fe in this study, ranged from 2.3-7.6% which are below the recommended dietary allowance value for Fe.

The recommended dietary allowance (RDA) for Cu ranged from 0.9 to 30 mg/day/person, these values are similar to World Health Organization's recommendation (15–500 μg/kg bw/day) (WHO, 1993). The intake values for Cu in this study, ranged from 0.05 to 0.11 μg/kg bw/day. The highest intake value of Cu was obtained for the palm oil sample M collected from the oil palm mill situated in the rural area of Bayelsa State, Nigeria. The estimated intake values for Cu constitutes about 3.17–6.70% of the upper limit of the recommended dietary allowance value.

Table 3: Estimated Daily Intakes (EDI) of heavy metals in palm oil samples in μg/kg bw/day

Samples	Fe	Cu	Cr	Zn	Ni	Pb	Cd
A	1.653	0.112	0.044	0.888	0.056	0.039	0.112
B	1.611	0.095	0.0003	0.750	0.049	0.044	0.044
C	0.701	0.076	0.022	0.483	0.056	0.015	0.047
D	1.204	0.099	0.022	0.961	0.034	0.027	0.039
E	1.932	0.068	0.0002	0.974	0.039	0.044	0.029
F	1.670	0.095	0.024	0.362	0.038	0.015	0.022
G	2.163	0.071	0.0003	0.718	0.099	0.047	0.033
H	1.071	0.073	0.0002	1.014	0.038	0.076	0.024
I	2.041	0.053	0.0005	1.653	0.024	0.047	0.044
J	1.410	0.112	0.003	0.702	0.044	0.047	0.066
K	1.410	0.087	0.0002	1.483	0.073	0.044	0.015
L	1.660	0.099	0.009	0.893	0.066	0.005	0.039
M	1.999	0.121	0.005	1.653	0.047	0.003	0.039
N	1.204	0.081	0.024	1.009	0.051	0.034	0.029
O	2.211	0.076	0.015	0.553	0.047	0.049	0.029
P	1.204	0.098	0.015	1.204	0.044	0.039	0.044
Q	2.284	0.112	0.003	1.004	0.051	0.027	0.099
R	2.350	0.071	0.022	1.192	0.027	0.044	0.029
S	0.966	0.073	0.0003	0.587	0.049	0.039	0.027

Table 4: Comparison of Recommended Dietary Allowances (RDA) of the elements in mg/kg/day and mg/l/day.

Parameters	This study	Iwegbue *et al.*, (2014) in alcoholic drinks	Foods and wine
Fe	7.23-24.20	0.30-10.30	10-18 (WHO, 1993)
Cu	0.70-1.15	0.09-0.60	30 (WHO,1993)
Cr	0.10-0.45	0.005-0.15	1.0 (WHO,1993),
Zn	3.73-17.03	0.12-3.86	4.2 (EVM, 2003),5.0 (OIV, 2008)
Ni	0.25-1.03	0.005-0.11	0.02 (SON, 2007)
Pb	0.03-0.45	0.001-0.047	3.6 (WHO,1993)
Cd	0.15-1.15	0.02-0.05	1.0 (WHO, 1993)
Mg	14.90-232.00	2.08-301.33	400–420 and 310–320 (Male and Female respectively, IOM, 2010)

Ca	71.00-146.03	2.21-49.23	28-37 (Ethiopian wine , IOM, 2010) 1000 mg (IOM, 2010)
Na	12.28-46.15	4.00-34.40	60 (WHO, 1993)
K	21.23-91.56	21.65-926.10	467-1147 (USA wine, IOM, 2010) 694-767 (Ethiopian wine, IOM 2010)

WHO- World Health Organization, 1993; OIV -Organization *Internationale de la Vigue et du* VIN, 2008; EVM-Expert Group on Vitamins and Minerals, 2003; IOM - Institute of Medicine 2010.

In this study, the highest dietary intake of Cr (0.044 μg/kg bw/day) was obtained for the palm oil sample A (mill located along the highways of Rivers State), which, however, was below the recommended dietary allowance for Cr (130 μg/ day/person and 2.2 μg/kg bw/day).

The estimated intakes of Zn from the consumption of palm oil samples analysed, ranged from 0.36 to 1.65 μg/kg bw/day (Table 3). The Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a provisional maximal daily intake of 1000 μg/kg bw/day for Zn (tolerable WHO, 2003). The Expert Group on Vitamins and Minerals of the Food Safety Authority (EVM) recommended a safe upper limit (SUL) of 4.2 mg/day for Zn (equivalent to 700 μg/kg bw/day in a 60 kg adult) for total dietary intake (EVM, 2003). The estimated intake values of Zn in this study ranged from 13.9 to 41.5 (%) of the provisional maximal tolerable daily intake of Zn.

The tolerable daily intake of Ni as established by the European Food Safety Authority (EFSA) is 2.8 μg Ni/kg bw/day (EFSA, 2015). The estimated daily intake of Ni in the analysed palm oil ranged from 0.02 to 0.10 μg/kg bw/day (Table 3), these values are within 0.71 – 3.57 (%) of tolerable daily intake value for the element, nickel. The estimated daily intake value of Pb from this study ranged from 0.003 to 0.076 μg/kg bw/day (Table 3), which is approximately 0.44–11.09 (%) of the tolerable intake value (3.6 μg/kg bw/day) The highest intake value of Pb was found in the palm oil sample H, obtained in Bayelsa State, Nigeria.

The daily intake of Cd from the analysed palm oil ranged between 0.02 and 0.11 μg/kg bw/day (Table 3). However, the tolerable intake value of Cd is 1 μg/kg bw/day (WHO, 1993). The estimated intake value of Cd from the palm oil is within approximately 1.85–13.83 (%) of the tolerable intake value.

The recommended dietary allowance (RDA) for Mg for male and female healthy adults are 400–420 and 310–320 mg of magnesium per day respectively (IOM, 2010) while the recommended dietary allowance value for Ca is set at 1000 mg Ca per day (IOM, 2010). The estimated intake values of Mg and Ca varied considerably among the analysed palm oil samples. The highest dietary intakes of Mg and Ca were found in palm oil samples L and M (Table 3), both were obtained from oil mills located in Balyesa State, Nigeria

Comparison of Recommended Dietary Allowance

A comparison of Recommended Dietary allowance (RDA) values of the elements analysed in this study was made with some values obtained by Iwegbue *et al*. (2014) for some drinks from Nigeria and for food and wine from some International and National Standards organizations (Table 4)

Target hazard quotients

The estimated target hazard quotients (THQ) values (Table 5) for individual metals were less than 1 and they indicate safe levels for the metals. Any THQ values above unity indicate level of concern to health. The combined THQ values for all metals examined were ≥1 <2 for palm oil, while the combined THQ values for palm oil samples analyzed were <1. There is no lifelong health concern from the metals associated with the palm oil samples analyzed.

Table 5: Estimation of target hazard quotients (THQ) of heavy metals in the palm oil samples

Samples	Fe	Cu	Cr	Zn	Ni	Pb	Cd
A	0.0114	0.0134	0.1400	0.0142	0.14	0.0001	0.5400
B	0.0011	0.0114	9.3E-08	0.0120	0.12	0.0001	0.2100
C	0.0048	0.0091	0.0720	0.0077	0.14	0.0001	0.2200
D	0.0083	0.0120	0.0720	0.0154	0.08	0.0009	0.1900
E	0.0133	0.0082	6.2E-0.8	0.0156	0.09	0.00001	0.1400
F	0.0115	0.0011	0.0780	0.0058	0.09	0.00001	0.1100
G	0.0148	0.0085	9.3E-08	0.0115	0.24	0.0002	0.1600
H	0.0073	0.0088	6.2E-0.8	0.0162	0.09	0.0002	0.1200
I	0.0140	0.0064	1.6E-07	0.0264	0.06	0.0002	0.2100
J	0.0096	0.0134	0.0093	0.0112	0.11	0.0002	0.3200
K	0.0096	0.0105	6.2E-0.8	0.0236	0.18	0.0004	0.0700
L	0.0114	0.0120	0.0311	0.0143	0.16	0.00002	0.1900
M	0.0137	0.0093	0.0156	0.0265	0.11	9E-06	0.1900
N	0.0083	0.0097	0.0778	0.0162	0.12	0.0001	0.1400
O	0.0152	0.0091	0.0467	0.0887	0.11	0.0002	0.1400
P	0.0083	0.0118	0.0467	0.0193	0.11	0.0001	0.2100
Q	0.0157	0.0134	0.0093	0.0161	0.12	0.00009	0.4800
R	0.0161	0.0085	0.0720	0.0191	0.07	0.0001	0.1400
S	0.0066	0.0088	9.3E-08	0.0094	0.12	0.0001	0.1300

CONCLUSION

In conclusion, the mean mineral concentrations for the palm oil samples in the Niger Delta region were within the allowable limits. Therefore, the palm oil samples do not pose any risk to human health.

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