Monitoring of essential and toxic metals in imported herbal teas marketed in selected cities in Southern Nigeria: A health risk assessment study

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ABSTRACT: Teas are the most consumed beverage worldwide after water, and its consumption among Nigerians has increased over the past decades due to its documented health potentials. The objective of this study was to investigate the levels of heavy metals (Pb, Ni, Cr, Zn, Fe and Cu) in herbal teas and also to estimate human health risk associated with their consumption. Twenty eight samples of commonly consumed herbal tea bags were purchased from major supermarkets in Port-Harcourt, Yenagoa and Owerri in Southern Nigeria. They were digested, ashed and analysed using atomic absorption spectroscopy (AAS). Heavy metal concentration varied among the different brands of herbal teas in the study whereas the concentrations of Cd, Cr and Ni in some herbal teas were higher than the permissible limit in China. The estimated daily intake (EDI) of heavy metals via consumption of herbal teas showed that Fe contributed the most to the daily intake of metals while Pb contributed the least. The mean average of EDI of heavy metals were 0.657, 0.118, 0.014, 0.014, 0.008 and 0.0009 mg/kg/day Bw in the order of Fe > Zn > Cu > Cr > Ni > Pb respectively. The THQ and HI values were < 1. In conclusion, the study suggests that the risk level of heavy metal exposure via consumption of herbal teas is low with no significant health implications to consumers and may not pose a threat to food safety. © JASEM

Tea is a popular beverage consisting of processed and dried tea leaves (Moghaddam et al., 2008). It is widely consumed around the globe because of its aroma, taste, smell, variety of types and most especially for its multiple promoting health effects (Salahinejad and Aflaki, 2010; Ipeayieda and Dawodu, 2011). Presently in Nigeria there is a growing acceptance of these imported herbal teas among the population as it is believed to have numerous beneficial health effects, such as cancer prevention (Ipeayieda and Dawodu, 2011), diabetes (Vinson and Zhang, 2005; Sajilata et al., 2008) as well as reducing the risk of cardiovascular diseases (Davies et al., 2003). Among the different types of popular teas (Green, Black, Oolong and Puerh teas), green tea is documented to be the most beneficial since it has been proven to contain high amounts of antioxidants which protects against oxidative damages in humans (Kaushik et al., 2011). Herbal teas are highly reputed to restore the body's proper balance and to maintain optimum health by many Nigerians (Orisakwe et al., 2015). Also it has the ability to detoxify the body of alcohol and toxins (Dufresne and Farnworth, 2000).

Contamination of food by heavy metals is detrimental to health and there are currently strict regulations by developed countries to reduce the concentration of toxic metals in food and beverages with regards to the permissible limits outlined by WHO/JECFA, US EPA, and EFSA (Seenivasan et al., 2008). In Nigeria more than 95% of teas consumed are imported from China which is the highest exporter of tea globally with 1,726,023 tons produced in 2011 (FAOSTAT 2014).

The quality of tea depends on several factors which includes the cultivated soil, the degree of contamination and atmospheric conditions (Brezicha-cirocka et al., 2016). It has been reported that the different procedure of processing tea leaves which includes biological and industrial methods does not remove heavy metals, rather they become more concentrated and in turn enter the food chain and ultimately pose serious human health risk to consumers (Ansari et al., 2007; Weldegebriel et al., 2012; Rezaee et al., 2014). However, the evaluation of essential and toxic metals in teas is of great importance considering the potential health risk to habitual tea drinkers if these metals are not within the safe limits (Szymczycha-madeja et al., 2012). Since food ingestion is a major route of exposure to toxic substances (Zhu et al., 2011; Liu et al., 2013), prolonged consumption of toxic metals (Pb, Cd, Cr and Hg) which have been reported in teas (Wang et al., 2008; Mania et al., 2014; Li et al., 2015;; Orisakwe et al., 2015) might lead to toxic outcomes as a result of bio-accumulation over a life time (Zhou et al., 2016). Also, the presence of essential metals in teas can also exceed the limit associated with their toxicities due to regular consumption.
 Herbal teas sold in Nigeria are consumed indiscriminately for therapeutic purposes among the population, therefore there is the need to monitor the

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levels of heavy metals in imported herbal teas with respect to their permissible limits. This study is aimed at investigating the concentration of toxic (Pb, Ni and Cr) and essential (Zn, Fe and Cu) metals present in various brands of herbal teas, calculate the Estimated Daily Intake (EDI) of metals, Target hazard quotient (THQ), Hazard Index (HI) and also the cancer risk associated with consumption of herbal teas available in the market in southern Nigeria.

MATERIALS AND METHODS

Collection of samples: A total of 28 samples of imported herbal tea bags sold in major supermarkets were bought from three different cities in Southern Nigeria namely Port-Harcourt, Yenagoa and Owerri in September 2015.

*Sample Preparation***:** The glass beakers and containers used for analysis were washed with detergents and rinsed with double distilled water. 0.5g of each herbal tea sample was weighed accurately, ground and transferred into a glass beaker. HNO_3 and $HClO_4$ were added in a ratio of 5:1. The samples were kept standing overnight at room temperature for pre-digestion. The mixture was heated on a hot plate (130^0C, 30 mins-1hr) until the solution became transparent and semi-dried. It was allowed to cool, afterwards and then transferred to a 50ml volumetric flask by adding deionized distilled water (Du et al., 2003; Hseu, 2004). Blanks were also prepared using the same procedure.

Quality Assurance; Analysis of Sample; The concentrations of copper ($\lambda = 324.7nm$), Lead ($\lambda = 283.3nm$), Iron ($\lambda = 248.3nm$), Zinc ($\lambda = 213.9nm$), Chromium ($\lambda = 357.9nm$) and Nickel ($\lambda = 232.0nm$) were determined using solar thermo flame atomic absorption spectrometer model S4 71096 with percentage recovery between 92%-100%. The digestion of all tea samples was triplicated and only the mean was reported afterwards.

*Health Risk Assessment of Heavy Metals***:** The following equations were used to calculate the exposure assessment to heavy metals from consumption of herbal teas.
Estimate Daily Intake (EDI): To calculate the estimated daily intake of heavy metals (mg/kg/day Bw) via consumption of herbal teas we used the equation below;

EDI = $\underline{Ci\ X\ Di\ X\ ED\ X\ Ef}$ (Chary et al., 2008) (1)
Bw X ATn
Where:
Ci = Concentration of metals

Di = Amount of daily intake (8g/person/day Cao et al., 2010; Shen Chen et al., 2008)
ED = Human lifetime exposure (50 years)
Ef = Exposure Frequency (350 days/year)
Bw = Mean body weight (70 kg was adopted for Nigerian Adults)
ATn = Average Time for non-carcinogen (17,500)
Sf = Slope factor of the metal (mg/kg/day)
Reference dose = Fe (0.7), Zn (0.3), Cu (0.04), Pb (0.004), Ni (0.02), Cr (1.5). (US EPA 2015)

Target Hazard Quotient **(THQ):** To calculate for Non-carcinogenic risk of heavy metals exposure. The hazard quotient of a single contaminant is determined by the equation below:

Target Hazard Quotient = $\underline{Exposure\ dose}$ (Wang et al., 1998)   (2)
Reference dose

Hazard Index: To calculate the total risk for a combination of heavy metals, the hazard index of the heavy metals is calculated from the equation below;
Hazard Index = ΣTHQ   (US EPA 1986)   (3)

RESULTS AND DISCUSSION

Levels of Heavy Metals in Herbal Teas: The concentration of heavy metals (Cu, Zn, Fe, Pb, Cr, and Ni) in imported herbal tea samples sold and commonly consumed in southern Nigeria are presented in Table 1. The maximum levels of copper observed in herbal tea samples were seen in liver cleanser, MWT, LGT, TFR and BOD samples with average concentrations of 6.59 mg/kg while the least levels were seen in SLT and Rheumatic and Arthritis tea samples with a mean concentration of 2.66 mg/kg. Essential metals required by humans have also been documented to accumulate in tea despite the many reports about toxic heavy metals (Rezaee et al., 2011). In recent years researchers have demonstrated that regular consumption of tea can significantly contribute to the daily dietary intake of essential elements (Karak and Bhagat, 2010; Malik et al., 2013; Li et al., 2016). Although tea is described as the most consumed beverage after water, as well as a rich source of some essential dietary metals (Tea Council, 2004).The highest concentration of Fe was observed to be in Liver cleanser, ETM and LGT with 453, 318 and 317 mg/kg respectively, the least levels were 141.3 and 141 mg/kg as detected in TTG and ADT respectively. Heavy metal concentration varied among the different brands of herbal teas in the present study. This may be attributed to diverse sources of these herbal teas which are marketed in Nigeria under different labels laying claim to

multiple therapeutic potentials. Fernandez-Carcecres et al., in 2001 confirmed in their study that concentration of metals in tea varieties can be an adequate parameter to differentiate teas and their geographical origins. This statement is in agreement with the large variation of heavy metal concentrations in our study. Trade liberalization in Nigeria has led to the flooding of the Nigerian market with sundry items of which herbal teas is not an exception (Orisakwe et al., 2015). Therefore there is the need to constantly monitor the level of contaminants in imported products especially food products.

On the other hand, MWT, BOD and Anti-Cancer had the highest concentration of Zinc (Zn) with levels of 126.1, 54.7 and 57.2 mg/kg respectively while levels of 22.5, 23.3 and 24.9 mg/kg detected in STE, SLT and TTG respectively were the least. Furthermore, the concentration of Chromium (Cr) was highest in Liver cleanser and UOG teas which recorded concentration levels of 9.16 and 9.64 mg/kg while the least levels were observed in MNHT and CCT teas with concentrations of 0.39 and 1.38 mg/kg. Lead levels in herbal teas were found to be highest in neck and spinal cord, BFR, TFR and MNHT teas with concentrations ranging from 0.52 – 0.8 mg/kg while the minimum concentrations ranged from 0.04 - 0.07 mg/kg in after stroke recovery, Rheumatic and Arthritis and STE teas respectively. Nickel (Ni) concentrations were highest in SBT, LYL and MNHT with levels ranging between 5.14 and 5.85 mg/kg with least levels found in ADR, ETM, CCT and UOG teas with a concentration of 0.01 mg/kg.

Over the years, researchers have documented in literature that tea consumption is a potential source of toxic metal exposure (Salahinejad and Aflaki, 2010; Schwalfenberg et al., 2013 and Li et al., 2015). Also, tea plants show strong evidence in its ability to take up non-essential metals (Karak and Bhagat, 2010) from the soil and inevitably move to the tea leaves (Jin et al., 2005) which is the primary raw materials used in the production of tea. The individual concentrations of Pb in herbal tea samples ranged between 0.005-0.8 mg/kg in the present study, these values are in accordance with the levels of Pb found in teas marketed in India (Seenivasan et al., 2008) but lower than the values reported in China (Li et al., 2015). Also high levels of Pb were found in tea samples from Japan ranging between 0.51-1.5 mg/kg

(Brzezicha-Cirocka et al., 2015). The high levels of Pb in Tea samples could be attributed to dust particles during tea processing and the solder used for packaging (Ashraf and Mian, 2008).

The analyzed herbal tea samples were seen to contain high levels of Nickel, though this toxic metal was detected in 100% of the analyzed tea samples. The present study showed that the level of Ni in herbal teas ranged from 0.01- 6.72 mg/kg, this values are in contrast and found to be lower than those reported by Marcos et al., 1998 with range between 2.99-22.6 mg/kg also herbal teas from Thailand with concentration ranging from 2.281- 9.19 mg/kg (Nookabkaew et al., 2006). In another study, tea samples from Turkey contained Ni concentrations ranging between 10.8-38.8 mg/kg (Narin et al., 2004).

Various studies have reported the health implications of Cr as a major food contaminant (Cao et al., 2010; Guerra et al., 2012) because of its well-known toxic potential. Cr is usually available in two forms, Cr^{3+} and Cr^{6+}. In our present study the determination of Cr was not specie specific so we assume Cr^{3+} since Cr^{6+} could be reduced to Cr^{3+} in the stomach under acidic pH (De flora et al., 1997). The concentration of Cr in our study ranged from 0.39-7.61 mg/kg which is relatively higher than the values reported for teas from China, India and Japan (Brzezicha-cirocka, 2016). A study by Ferrara et al., 2001 reported Cr concentrations in Chinese teas to be between 17.9-115.4 mg/kg. These values are higher than the result of our study. Though few studies have assessed the content of Cr in teas, the varying concentrations from different studies which were neither consistent nor corresponded with our study could probably be as a result of geographical origin or the different processing procedures involved in tea production (Seenivasan et al., 2008).

Therefore the incessant consumption of these herbal teas by Nigerians in search of health benefits could reach limits that are linked to its toxicity. The content of Cu in the study ranged from 2.49-6.72 mg/kg with an average content of 4.54 mg/kg which is low when compared with the result of Qin and Chen 2007 (17.13 mg/kg), Jin et al., 2008 (13.26 mg/kg) and Li et al., 2015 (18.42 mg/kg).

Table 1: Concentrations (mg/kg) of heavy metals from Herbal tea samples marketed in Southern Nigeria

TEA SAMPLES	Cu	Fe	Zn	Cr	Pb	Ni
Black tea						
Ttg	5.12	141.30	24.90	4.08	0.17	4.08
Sbt	3.68	183.20	36.20	2.69	0.34	5.14
Lyl	4.27	171.60	32.10	5.07	0.2	5.14
Anti-diabetic tea						
Adr	4.15	134.60	29.20	3.99	0.27	0.01
Adt	3.97	141.00	27.80	2.77	0.42	2.16
Sexual enhancement tea						
Mnht	3.69	245.40	33.30	0.39	0.52	5.85
Etm	5.16	318.90	34.10	6.68	0.21	0.01
Mwt	6.68	204.10	126.10	2.35	0.31	0.85
Anti-hypertensive & cholesterol						
Cct	3.84	261.20	32.90	1.38	0.22	0.01
Ctt	4.11	175.10	36.10	3.12	0.2	2.2
Hcs	3.89	163.30	29.60	4.13	0.27	4.19
Fat reducing						
Bfr	3.88	161.30	29.90	3.82	0.54	1.84
Tfr	6.56	211.10	71.90	5.98	0.8	3.49
Green tea						
Qgt	4.13	152.70	32.40	5.49	0.26	1.49
Lgt	6.64	317.10	31.80	7.61	0.33	1.69
Organic green tea						
Uog	5.96	161.80	32.80	9.64	0.39	0.01
Bod	6.34	202.20	54.70	5.12	0.31	4.98
Sleeping tea						
Slt	2.94	154.10	23.30	2.6	0.41	4.33
Ste	3.16	192.70	22.50	5.1	0.05	0.34
Others	3.13	293.30	57.20	6.61	0.14	6.72
Anti-cancer tea						
Ginseng tea	4.08	187.90	31.20	2.58	0.33	4.54
Stop smoking tea	5.16	280.80	31.40	5.07	0.41	0.86
Neck & spinal cord tea	4.24	187.30	43.10	3.98	0.64	0.75
After stroke recovery tea	3.26	279.70	35.10	6.64	0.04	1.53
Rheumatic & arthritis	2.97	190.90	37.70	4.17	0.07	3.6
Digestive system	5.54	236.10	24.50	4.18	0.22	0.86
Liver cleanser	6.72	453.10	28.80	9.16	0.47	3.58
Anti-malaria	3.89	169.10	39.50	2.66	0.29	1.69

With respect to Fe, Mihaljev et al., 2014 reported a range of 61.87-673 mg/kg among herbal teas in Serbia. Similarly, Fe content ranging between 141-453 mg/kg observed in our study can be compared with the result of Salahineijad and Aflaki, 2009 who reported a mean concentration of 169.19 mg/kg in teas consumed in Iran. According to the report of Ghoochani et al., 2015,
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Zn levels among black teas cultivated in Iran had a concentration ranging from 17.43-113 mg/kg. On the other hand, Mmoghaddam et al., 2007 reported an average Zn content of 50.7 Mg/kg in Iranian consumed tea which can be compared with the result of our study with an average concentration of 38.2 mg/kg in herbal tea samples

Table 2: Estimated Daily Intake (EDI) of heavy metals (mg/kg day $^{-1}$ Bw^{-1}) from consumption of Herbal Teas marketed in Southern Nigeria

TEA SAMPLES	Cu X 10^{-4}	Fe X10^{-2}	Zn X 10^{-3}	Cr X 10^{-4}	Pb X 10^{-5}	Ni X 10^{-4}
BLACK TEA						
TTG	5.63	1.55	2.74	4.49	1.87	4.49
SBT	4.05	2.02	3.98	2.96	3.74	5.65
LYL	4.69	1.89	3.53	5.58	2.20	5.65
ANTI DIABETIC TEA						
ADR	4.57	1.52	3.21	4.39	2.97	0.01
ADT	4.37	1.55	3.06	3.05	4.62	2.38
SEXUAL ENHANCEMENT TEA						
MNHT	4.06	2.69	3.66	0.43	5.72	6.44
ETM	5.68	3.51	3.75	7.35	2.31	0.011
MWT	7.35	2.25	13.87	2.59	3.41	0.94
ANTI HYPERTENSIVE & CHOLESTEROL						
CCT	4.22	2.87	3.62	1.52	2.42	0.011
CTT	4.52	1.93	3.97	3.43	2.2	2.42
HCS	4.28	1.79	3.26	4.54	2.97	4.61
FAT REDUCING						
BFR	4.27	1.77	3.29	4.2	5.94	2.02
TFR	7.22	2.32	7.91	6.58	8.80	3.84
GREEN TEA						
QGT	4.54	1.68	3.56	6.04	2.86	1.64
LGT	7.30	3.48	3.49	8.37	3.63	1.86
ORGANIC GREEN TEA						
UOG	6.56	1.78	3.61	10.6	4.29	0.011
BOD	6.97	2.22	6.02	5.63	3.41	5.48
SLEEPING TEA						
SLT	3.23	1.69	2.56	2.86	4.51	4.76
STE	3.48	2.12	2.48	5.61	5.50	0.37
OTHERS	3.44	3.23	6.29	7.27	1.54	7.39
ANTI CANCER TEA						
GINSENG TEA	4.48	2.07	3.43	2.84	3.63	4.99
STOP SMOKING TEA	5.68	3.09	3.45	5.58	4.51	0.95
NECK & SPINAL CORD TEA	4.66	2.06	4.74	4.38	7.04	0.83
AFTER STROKE RECOVERY TEA	3.59	3.08	3.86	7.30	4.40	1.68
RHEUMATIC & ARTHRITIS	3.27	2.09	4.15	4.59	7.70	3.96
DIGESTIVE SYSTEM	6.09	2.59	2.69	4.6	2.42	0.95
LIVER CLEANSER	7.39	4.98	3.17	10.08	5.17	3.94
ANTI MALARIA	4.28	1.86	4.35	2.93	3.19	1.86

Health Risk Assessment: The calculated estimated daily intake (EDI) of essential and toxic metals for individual herbal teas is presented in Table 2. The EDI of heavy metals via consumption of herbal teas showed that Fe contributed the most to the daily intake of metals while Pb contributed the least. The mean average of EDI of heavy metals were 0.657, 0.118, 0.014, 0.014, 0.008 and 0.0009 mg/kg/day Bw in the order of Fe > Zn > Cu > Cr > Ni > Pb respectively. The calculated EDI when compared with the Rfd of the studied heavy metals were all within the Provisional Tolerable Daily Intake (PTDI).

The THQ values of individual metals in herbal teas were all below 1 as outlined in Table 3 while on the other hand the effect via consumption of multiple metals in herbal teas expressed by the total hazard index (HI) value of all investigated metals were all below 1 as shown in Table 3.

The science of risk assessment provides researchers with severe tools in order to perform computational risk of hazards associated with foods. Although many recent studies only reported the levels/concentrations in teas, this study however, assessed the potential risk of heavy metal exposure via consumption of herbal teas by calculating the EDI of both toxic and essential metals, the THQ and HI proposed by the US EPA to assess the possible non-carcinogenic effects of heavy metals on human (Fu et al., 2015). The calculated EDI via consumption of herbal teas as shown in Table 2 was compared with the concentration of oral exposure to heavy metals that is likely to be without an appreciable risk of deleterious effects over a

lifetime (IRIS 2011). There are relatively no background data and standards currently available in Nigeria for the assessment of human health risk associated with exposure to heavy metals, so we compared the EDI with the reference doses. The dietary intake of heavy metals arising from consumption of herbal teas tends to be lower than the reference dose in all the samples. The THQ values of individual metals through consumption of herbal teas were all < 1 as shown in Table 3. Lastly, the combined effects of all the studied heavy metals expressed as HI were all < 1, suggesting no significant health implications to the exposed population.

Table 3: Target hazard quotient (THQ) and Hazard index (HI) for Population exposed to Herbal teas in Southern Nigeria.

TEA SAMPLES	Cu X 10^{-2}	Fe X 10^{-2}	Zn X 10^{-2}	Cr X 10^{-4}	Pb X 10^{-3}	Ni X 10^{-2}	Hazard Index
Black tea							
Ttg	1.41	2.22	0.91	3.00	4.70	2.24	0.05
Sbt	1.01	2.88	1.33	2.00	9.40	2.83	0.06
Lyl	1.17	2.69	1.18	4.00	5.50	2.83	0.06
Anti-diabetic tea							
Adr	1.14	2.12	1.07	3.00	7.40	0.01	0.05
Adt	1.09	2.22	1.02	2.00	11.60	1.19	0.06
Sexual enhancement tea							
Mnht	1.02	3.86	1.22	0.30	14.30	3.22	0.08
Etm	1.42	5.01	1.25	5.00	5.80	0.01	0.08
Mwt	1.84	3.21	4.62	2.00	8.50	0.47	0.11
Anti-hypertensive & cholesterol							
Cct	1.06	4.11	1.21	1.00	6.10	0.01	0.07
Ctt	1.13	2.75	1.32	2.00	5.50	1.21	0.06
Hcs	1.07	2.57	1.09	3.00	7.40	2.31	0.05
Fat reducing							
Bfr	1.07	2.54	1.09	3.00	14.90	1.01	0.06
Tfr	1.80	3.32	2.64	5.00	22.00	1.92	0.10
Green tea							
Qgt	1.14	2.39	1.19	4.00	7.20	0.82	0.05
Lgt	1.83	4.98	1.17	6.00	9.10	0.93	0.09
Organic green tea							
Uog	1.64	2.54	1.20	7.00	1.07	0.01	0.07
Bod	1.74	3.18	2.01	4.00	8.50	2.74	0.08
Sleeping tea							
Slt	0.81	2.42	0.85	2.00	11.30	2.38	0.05
Ste	0.87	3.03	0.83	4.00	1.40	0.19	0.05
Others							
Anti-cancer tea	0.86	4.61	2.09	5.00	3.90	3.69	0.08
Ginseng tea	1.12	2.95	1.14	2.00	9.10	2.49	0.06
Stop smoking tea	1.42	4.41	1.15	4.00	11.30	0.47	0.08
Neck & spinal cord tea	1.17	2.94	1.58	3.00	17.60	0.41	0.07
After stroke recovery tea	0.89	4.39	1.29	5.00	1.10	0.84	0.07
Rheumatic & arthritis	0.82	2.99	1.38	3.00	1.90	1.98	0.05
Digestive system	1.52	3.71	0.89	3.00	6.10	0.47	0.07
Liver cleanser	1.85	7.12	1.06	7.00	12.90	1.97	0.11
Anti-malaria	1.07	2.66	1.45	2.00	7.90	0.93	0.06

Conclusion: Continuous intake of these imported herbal teas is likely going to increase the body burden of toxic metals in habitual tea drinkers. The present study reveals that imported herbal teas are contaminated with toxic metals (Pb, Ni and Cr) at concentration higher than the maximum permissible limit. Although the health risk assessment result showed no possible health risk, the fact that the exposed population are exposed to other sources of heavy metal pollution is of public health concern as combined exposure through oral, dermal and inhalation routes could lead to toxic outcomes. Consequently there is the need to enforce strict regulations to monitor the level of heavy metals in imported food products.

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