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HEALTH RISK ASSESSMENT OF TOXIC METALS IN FOREIGN AND LOCAL WHEAT-BASED LIQUORS

Francis Olumide Oladapo# and Abigael Aderonke Falola
Department of Chemical Sciences, Tai Solarin University of Education,
Ijagun, Ijebu-Ode, Ogun State, Nigeria.
#Correspondence address: email: oladapofo@tasued.edu.ng; phone: +248032426294

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Abstract This study investigated targeted toxic heavy metal contents and their health risks in local and foreign wheat-based liquors obtained in local markets in Ijebu-Ode, Nigeria. Liquor samples were treated with the concentrated mixture of nitric acid-hydrogen peroxide solution and the resulting solution was analyzed using Microwave Plasma-Atomic Emission Spectrometry (MP-AES). The mean contents of toxic heavy metals in the wheat liquors ranked in the order of Pb (1.96149 mg.L^{-1}) > Cd (1.39746 mg.L^{-1}) > Ni (1.06424 mg.L^{-1}) > As (0.0356 mg.L^{-1}) for the local wheat liquors and Pb (1.84044 mg.L^{-1}) > (1.05842 mg.L^{-1}) > Cd (0.91676 mg.L^{-1}) > As (0.03344 mg.L^{-1}) for the foreign wheat liquors. The non-carcinogenic risk analysis of the wheat liquors produced a total HI of 0.1454, indicating a low-risk hazard index while the carcinogenic risk indicators suggested a high carcinogenic risk tendency due to individual contributions of Cd, Ni, and Pb ions. The total carcinogenic risk potential due to additive toxic metal effect was considered highly significant.

Keywords: Carcinogenic risk, Hazard Index, Heavy metals, MP-AES, Wheat liquors.

Introduction

Domesticated cereals are common plants grown and consumed worldwide and are a member of a Poaceae family. They are comprised of wheat, maize, oat, rice, rye, millet and sorghum, and are common sources of carbohydrates, protein, energy, vitamins, and some minerals (Blandino *et al*., 2003; Goesaert *et al*., 2005). Wheat grains derived from wheat-grass are widely processed not only as food constituents for daily dietary consumption but also as essential raw materials in the production of wheat-based beverages. Wheat beverages commonly produced include whiskey, tequila, gin, rum, brandy, and bourbon. Alcoholic beverages are usually used for entertainment, celebration of milestones and rare achievements, and for some ritual rites across global cultures and social communities of the world. For instance, red wine has been reported to contain phytochemical-inhibiting aromatase which catalyzes critical reaction that will activate estrogen synthesis (Eng *et al*., 2002; Kijima *et al*., 2006). Polyphenols found in red wine can protect erythrocytes in human from oxidative stress (Tedesco *et al*., 2016). In medicine, alcohols are used as a disinfectant of hands by hospital workers and body skin of patients before the administration of injection (Pederson *et al*., 2005; Lawrence *et al*., 1994).

Heavy metals are ubiquitous group of elements found in the environment. Whilst metals such as lead and mercury have no beneficial functions, certain others are essential in living organisms as sources of vitamins and minerals for the cell functions (Lane and Morel, 2009). Generally, metals can induce toxicity in living organisms at elevated concentrations; however, some metals, such as mercury, arsenic and lead, can modulate toxicity at low concentrations after bioaccumulation in key organs such as liver, kidney and brain (Meharg, 2011). Consequently, the metal exposure may result in carcinogenic and mutagenic health problems (Jarup, 2003; Patlola & Tchounwou, 2005a, 2005b).

Occurrence of metals in agro-allied foods and beverages has evoked huge concern in recent decades considering their health implications at sub-lethal concentrations. Maduabuchi *et al* (2007) reported arsenic and chromium in the range of 0.002-0.261 mg/L in canned and uncanned beverages obtained in Nigeria. Ibanez *et al*. (2008) documented the sources of heavy metals at various production stages of alcoholic beverages in industries. Similar studies have reported heavy metals in some beverages which are components of daily drink menu (Musche, 1976; Onianwa *et al*., 1999; Redan *et al*., 2019; Izah *et al*., 2016). Consequently, agro-food industries, the food researchers, and various regulatory agencies of the governments are deeply concerned about the possible outbreak of food poisoning globally.

This study focused on potentially toxic metals in foreign and local wheat-based alcoholic liquors available in local markets and stores in Nigeria. Onianwa and co-workers (1999) reported substantial amounts of non-essential heavy metals in various categories of beverage foods and drinks in Nigeria. Although, their study targeted a wide range of different classes and types of beverages, it did not capture the wheat-based alcoholic beverages which have been widely consumed by the youth and adults across class and income-earning stratification. This investigation determined the presence of arsenic, cadmium, nickel and lead ions in local and foreign wheat-based liquors and comprehensively assessed the non-carcinogenic and carcinogenic risks associated with consumption of wheat liquors over a lifetime.

Materials and Methods

Materials

Analytical grades of trioxonitrate (V) acid, hydrogen peroxide, distilled water, sodium borohydride ($NABH_4$), and sodium hydroxide (NaOH) were purchased from Sigma Aldrich, United Kingdom. The laboratory hardware was purchased from Pyrex, United Kingdom. The MP-AES was purchased from Agilent Technologies Incorporated (USA).

Methods

Sample Collection and Digestion

Ten local and five foreign wheat liquors were collected from Oke-Aje Market and two other Shopping Malls all in Ijebu-Ode, Nigeria and preserved at -20^{0}C temperature in the laboratory. The foreign liquor collections were from Indian, France, Scotland and United Kingdom. All the samples collected were properly labelled. 1 mL of each wheat liquor collected was digested separately with 40 mL of peroxide-nitric acid (3:1; v/v) mixture in an oven at 70^{0}C temperature for 40 mins. The solution was cool and transferred into a clean plastic bottle and then stored at -20^{0}C temperature for MP-AES analysis.

Microwave Plasma-Atomic Emission Spectrometry Analysis

The Agilent 4200 MP-AES instrument was used to analyze cadmium, nickel and lead with the viewing position, nebulizer pressures and sample introduction mode operated automatically. The other instrument parameters used for the analysis are presented in Table 1.0.

Table 1.0: Viewing position, Read time and Nebulizer flow for total metal concentrations using Agilent 4200 MP-AES.

Analyte	Wavelength (nm)	Viewing Position (mm)	Read Time (s)	Nebulizer Flow (L/min)	Background Correction
Arsenic	228.812	Auto	3	0.75	Auto
Cadmium	361.051	Auto	3	0.5	Auto
Nickel	352.454	Auto	3	0.7	Auto
Lead	405.781	Auto	3	0.75	Auto

The MP-AES instrument was pre-cleaned with nitrogen for 2 hours before analysis. Quantification of each metal was done individually under nitrogen atmosphere prior to sample analysis. Five levels of calibration ranging from 0.2, 0.5, 1.0, 2.0 and 4.0 mg.L^{-1} were prepared to match the expected metal concentrations using a multi-element calibration instrument. The analytical cycle of 85 seconds was operated at interval of sample injection, but the MP-AES was rinsed with 5% HNO_3 solution during the last 40 seconds of the analytical cycle. The HNO_3 conditioned the system operating atmosphere to that of the sample. The multi-purpose sample introduction system connected to nebulizer, the double pass glass cyclonic spray chamber and pump operating at a speed of 15 rpm ensured a well-controlled matrix loading into the plasma. For arsenic quantification, hydride generation method modulated with Multimode Sample Introduction System (MSIS) accessory was used. This consisted 1.2% Borohydride ($NaBH_4$), and 1.0% sodium hydroxide (NaOH) solution.

Health Risk Assessment Analysis

Non-Carcinogenic Risk Effect

Risk assessment of heavy metals can be expressed as the various means of determining the likelihood of toxic heavy metals occurring in concentrations that would compromise human health over a lifetime (Bempah and Ewusi, 2016). The common category of health risks peculiar with heavy metals are carcinogenic and non-carcinogenic risks and they represent the total sum of the health hazards posed by individual toxic heavy metals.

The non-cancerous risk posed by toxic heavy metals in humans was calculated using equation (1) where CDI indicates the estimated chronic daily intake ($mg.kg^{-1}.day^{-1}$), MC stands for the median concentration of heavy metals in the beverages ($mg.L^{-1}$), CR indicates the daily average rate of beverage consumption ($L.person^{-1}.day^{-1}$), FE represents the frequency of exposure to the beverages ($365\ days.year^{-1}$) and ED is the annual and/or lifetime exposure durations ($365\ days.year^{-1}$ and 52 years estimated based on 70 years lifetime average and 18 years as permissible age). BW stands for the average body weight of adults (average: 65 kg) and AT represents the exposure time (in days which is 18980 days). According to Richie *et al.* (2018), the annual global average alcohol consumption is 6.4 L per person which corresponds to 17.53 cm^3 per day (0.175 L per day).

$$\mathrm{CDI} = \frac{\mathrm{MC\ x CR\ x\ FE\ x\ ED}}{\mathrm{BW\ x\ AT}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Also, the non-carcinogenic exposure risks exhibited by each toxic metals were calculated with equation (2) based on the hazard quotient (HQ) based on Li *et al.*, (2016) and Zhang *et al.*, (2018) as follows:

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

The RfD represents the reference dose of each toxic metal ($mg.kg^{-1}.day^{-1}$). The respective RfD values for As, Ni, Cd and Pb are 0.0003, 0.02, 0.001 and 0.004 $mg.kg^{-1}$ body weight per day. (Wu *et al.*2009; Jan *et al.*, 2010).

The hazard index (HI) of these beverage liquors were calculated as the sum of HQ of each toxic metals detected in the beverage liquors expressed in equation (3) as follows:

$$\mathrm{HI} = \sum_{i=1}^{i=n} \mathrm{HQ}_i \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (3)$$

When the values of HI and HQ are less than unity, the non-carcinogenic risk posed by the toxic metals in the beverage liquors is negligible (Table 2.0). However, when the values are greater than 1, the non-carcinogenic risks presented by these metals in the beverage liquors are palpable as they have exceeded an acceptable safety limit.

Table 2.0: Ranking of Non-carcinogenic Risk (USEPA, 1989)

Risk Ranking	Hazard Risk	Chronic Risk Index
1	< 0.1	Negligible
2	≥ 0.1 < 1.0	Low
3	≥ 1.0 < 4.0	Moderate
4	≥ 4.0	High

Carcinogenic Risk Effect

The carcinogenic risks of the toxic metals in the beverage liquors were calculated using equation (4) as suggested by Heshmati *et al*, (2018), Kim *et al.*, (2013) and USEPA, (2000).

$$\mathrm{CR} = \mathrm{CDI\ x\ CSF\ x\ AF} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (4)$$

The CSF, which expresses the probable potential of a substance to raise the cancer risk through oral exposure, is measured in $kg.day.mg^{-1}$. The AF is the age-dependent adjustment factor.

Statistical Analysis

The results obtained from MP-AES were analyzed statistically using the Statistical Package for Social Science (SPSS, version 20). The significance of differences between heavy metal contents in local and foreign wheat liquors was determined using paired t-test and Pearson correlation.

Results and Discussion

Heavy Metal Analysis

This study assessed the contents of arsenic, cadmium, nickel and lead ions present in assorted local and foreign wheat-based liquors and the statistical data from this work revealed some interesting findings presented in Tables 3.0 and 4.0.

Table 3.0: Levels of Heavy Metals Present in Assorted Local Wheat-based Liquors Analyzed (n=3)

Samples	Arsenic (mg/L)		Cadmium (mg/L)		Nickel (mg/L)		Lead (mg/L)	
	Mean	RSD	Mean	RSD	Mean	RSD	Mean	RSD
WWL1	0.0435	27.09	3.9885	9.98	3.3504	2.51	6.4916	0.71
WWL2	0.0538	21.42	3.1343	6.84	2.4512	1.49	4.3336	0.86
WWL3	0.0535	24.58	4.9025	9.21	3.4020	1.47	5.9391	1.26
WWL4	0.0237	24.34	0.1677	3.15	0.0282	7.03	0.1383	5.89
WWL5	0.0171	19.28	0.1354	5.65	0.0161	3.38	0.1185	2.55
WWL6	0.0215	60.45	0.5150	6.24	0.5286	3.42	0.8830	0.48
WWL7	0.0258	29.98	0.3572	0.83	0.3033	1.61	0.3990	1.11
WWL8	0.0174	54.57	0.1102	6.24	0.0209	2.58	0.1819	1.82
WWL9	0.0261	42.05	0.4451	4.29	0.4397	5.02	0.7696	1.54
WWL10	0.0228	20.06	0.2187	8.95	0.1020	1.71	0.3603	0.31
Average	0.0356		1.39746		1.06424		1.96149	

Table 4.0: Levels of Heavy Metals Present in Assorted Foreign Wheat-based Liquors analyzed (n=3)

Samples	Arsenic (mg/L)		Cadmium (mg/L)		Nickel (mg/L)		Lead (mg/L)	
	Mean	RSD	Mean	RSD	Mean	RSD	Mean	RSD
WWF1	0.0521	8.82	2.9857	7.41	2.5137	2.23	4.9978	0.31
WWF2	0.0280	21.72	0.1128	7.41	0.0313	2.95	0.1153	8.68
WWF3	0.0328	35.68	1.5483	8.44	1.5828	1.80	3.0469	0.85
WWF4	0.0298	50.42	0.3454	6.69	0.2888	1.41	0.5304	0.74
WWF5	0.0245	15.67	0.2999	6.69	0.1672	1.36	0.5118	1.30
Average	0.03344		1.05842		0.91676		1.84044	

The local wheat liquors recorded mean concentrations ranging from 0.0171 to 0.0538 $mg.L^{-1}$ for As, 0.1102 to 4.9025 $mg.L^{-1}$ for Cd, 0.0161 to 3.4020 $mg.L^{-1}$ for Ni and 0.1185 to 6.4916 $mg.L^{-1}$ for Pb while the foreign wheat liquors range from 0.0245 to 0.0521 $mg.L^{-1}$ for As, 0.0245 to 0.0521 $mg.L^{-1}$ for Cd, 0.0313 to 2.5137 $mg.L^{-1}$ for Ni and 0.1153 to 4.9978 $mg.L^{-1}$ for Pb. The mean average of the assorted local Arsenic contents exceeded the foreign liquors by 2.16 $\mu g.L^{-1}$ and both values were below the US regulatory agency permissible limit of 0.05 $mg.L^{-1}$ (USEPA, 2002) but higher than 0.01 $mg.L^{-1}$ set by WHO (2011). The mean cadmium concentration of 1.05842 $mg.L^{-1}$ recorded in assorted foreign wheat liquor was lower than 1.39746 $mg.L^{-1}$ detected in local wheat liquors. Both cadmium values exceed the permissible limit recommended by WHO (2011). Nickel mean content of 1.06424 $mg.L^{-1}$ found in assorted local wheat liquors was higher than 0.91676 $mg.L^{-1}$ obtained in the assorted foreign wheat liquors while both concentrations exceed the maximum acceptable limit (0.07 $mg.L^{-1}$) set by WHO (2011). For lead, a mean concentration value of 1.96149 $mg.L^{-1}$ was detected in assorted local wheat liquors compared to 1.84044 $mg.L^{-1}$ found in assorted foreign wheat liquors. This value exceeds 0.01 $mg.L^{-1}$ recommended by World Health Organization as the maximum limit for drinking water (WHO, 2011).

The profiles of metal contents in the wheat liquors presented in Figures 1.0, 2.0 (a) and (b) suggest that samples WWL1, WWL2 and WWL3 of the local brand and WWF1 and WWF3 of the foreign brand were highly contaminated with the four toxic metals studied. The levels of contamination reported in these identified rice samples may be associated with the agricultural practices or the fields from where they were harvested.

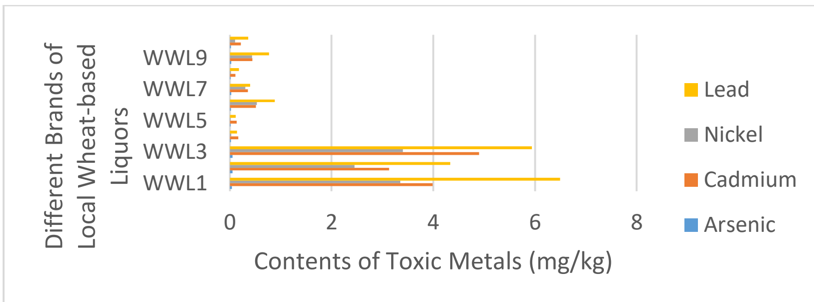

Figure 1.0 (a): Contents of Assorted Local Wheat-based Liquors

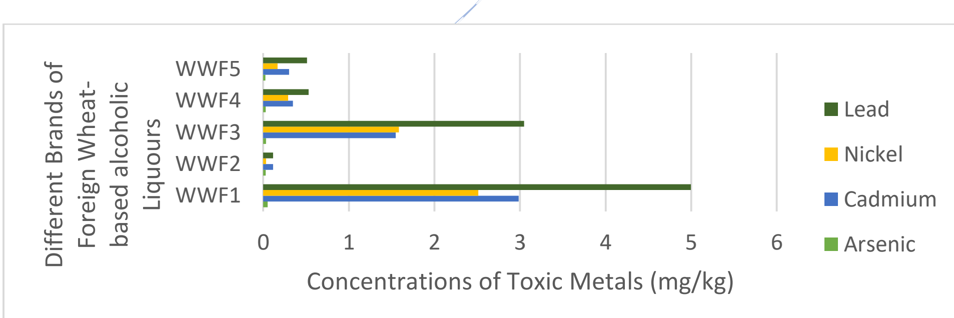

Figure 1.0 (b): Contents of Assorted Foreign Wheat-based Liquors

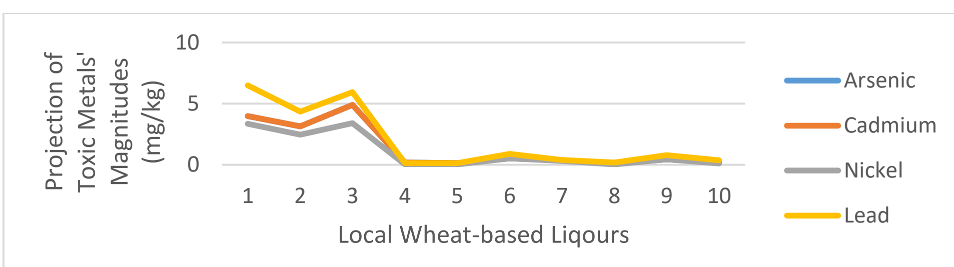

Figure 2.0 (a): Projection of Toxic Metal Levels in Assorted Local Wheat-based Liquors

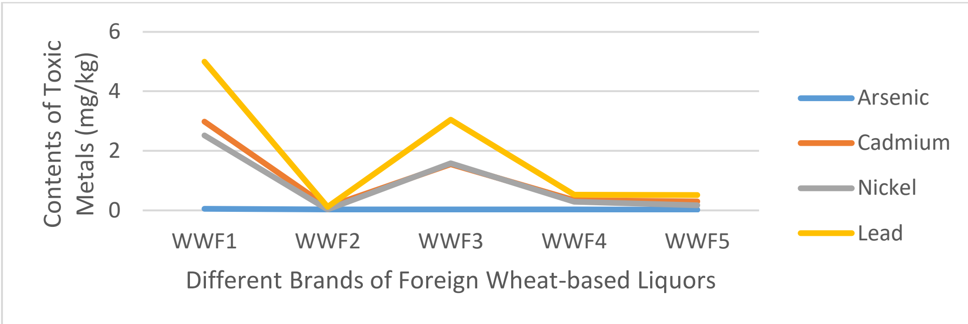

Figure 2.0(b): Projection of Toxic Metal Levels in Assorted Local Wheat-based Liquors

Human Health Assessment Analysis

Comparative analysis of the metal contents in both local and foreign wheat liquors revealed marginal differences in their statistical values. The carcinogenic and non-carcinogenic risk analyses of the wheat liquors revealed the magnitude of toxic heavy metals contamination of the wheat liquors. The metal contents in local and foreign liquors showed no significant difference when statistically analyzed. The paired t-test results [t(3) = 2.185, CV =2.353, p = 0.117] showed a t-value less than the critical value and p-value greater than 0.05 at two-tailed test analysis. This imply that the two data were not significantly and statistically different. Similarly, the Pearson analysis showed significant correlation between both liquors [r = 0.988; p = 0.012]. Since the two data are not statistically and significantly different, the average contents of metals in both local and foreign liquors were used for health risk evaluation.

Non-carcinogenic Risk Assessment

The rank order of heavy metals in wheat liquors according to HQ was Cd > Pb > As > Ni (Table 5.0), confirming Cd to have the highest pollution factor in wheat liquors in Nigeria. The ∑HI of this work was 0.1454, regarded as low risk hazard Index {where 0.1 < ∑HI < 1.0; low risk} (Table 2.0). This indicates that it is less likely for the metals to cause non-cancerous health problems for wheat beverage consumers. It is noteworthy that factors such as average exposure time, inaccurate oral reference dose and metal bioavailability can affect the ∑HI estimated in this study.

Carcinogenic Risk Assessment

The CR values for all the heavy metals exceeded the minimum acceptable risk limit of 1 x 10^{-6} set by US regulatory authority (USEPA, 2015). However, the CR values for Cd and Pb exceeded the maximum acceptable risk limit of 1.0 x 10^{-4} (USEPA, 2015). These results indicated high possibility for an individual exposed to Cd, Ni and Pb to suffer cancerous risk via wheat liquor consumption in addition to the health problems caused by alcoholic liquors. The carcinogenic risk posed by the four toxic heavy metals in wheat liquors was quantified as 1.274 x 10^{-3}. It is known that As, Cd, Ni and Pb are toxic carcinogenic and mutagenic metals which are harmful to human biosystem even at low concentrations (ATSDR, 2007). While toxic metals may have been recognized as causing harmful environmental and nutritional problems in humans, their carcinogenic risks constituted in food and beverages are significantly worrisome.

Conclusion

The presence of toxic heavy metals in wheat-based liquors poses serious health problems for consumers. The findings of this study have shown that toxic heavy metals contained in wheat beverages may not pose non-carcinogenic risks to consumers. However, the threats of carcinogenic risks due to human exposure are very significant and worrisome. The food and beverage regulatory bodies need to step up action against the concerned drinks for the safety of the consuming public.

Conflict of Interest

The authors declare no conflict of interest.

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