Evaluation of Carcinogenic and Non-Carcinogenic Health Risk of Heavy Metal Exposures from Some Local Fruits in Katsina State, North West Nigeria

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Authors’ contributions
This work was carried out in collaboration among all authors. Author AIY designed the study, wrote the protocol and wrote the first draft of the manuscript. Authors JIB, LS, AN, AU and UB performed the statistical analysis and manage the analysis of the study. Authors ZAS, IAY and AIY managed the literature searches. All authors read and approved the final manuscript.

Article Information
DOI: 10.9734/AJACR/2022/v11i430264

Open Peer Review History:
This journal follows the Advanced Open Peer Review policy. Identity of the Reviewers, Editor(s) and additional Reviewers, peer review comments, different versions of the manuscript, comments of the editors, etc are available here: https://www.sdiarticle5.com/review-history/90015

Received 05 June 2022
Accepted 10 August 2022
Published 16 September 2022

ABSTRACT
This work was carried out to evaluate the heavy metals (Cr, Cd, Fe, Ni, Mn, Pb and Zn) pollution load and health risks to the population of some local fruit samples in Katsina state Nigeria. Results from this study have shown that except for the mean concentration of Pb (1.152-1.623) and Cd (0.053-0.092), the mean concentration (mg/kg) range values of Fe (1.728-1.954), Mn (0.321-0.502) and Zn (0.263-0.967 ppm) in the samples were generally below the maximum allowable concentration for these metals. The risk level of Target Hazard Quotient below 1(< 1) was recorded for all the evaluated heavy metals for the population, indicating a non-carcinogenic adverse health effect on the population. The Incremental Lifetime Cancer Risk for the heavy metal Cd was at the threshold risk limit (>10−4) in all the sampled fruits in adults, while Pb for adults and Pb and Cd

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for children were within the moderate risk limit (>10−3). The cumulative cancer risk (∑ILCR) of the studied fruits was within the moderate risk level (>10−3) in adults and above the level (>10−2) in children. The results of the study have suggested that the intake of the local fruits in Katsina State is of public health concern as it may lead to an increase in the population cancer cases.

**Keywords:** Heavy metals; health risk; cancer; pollution; fruits; Katsina State; Nigeria.

### 1. INTRODUCTION

The beneficial effect of fruit consumption on human health is well documented. Fruits have become a part of people’s daily diets more and more with the improvement of living conditions in many parts of the world [1]. The quality and safety of fruits are compromised by their contamination with heavy metals [2,3,4,5].

Heavy metals are known to be non-biodegradable, with long biological half-lives, and possess the ability to bioaccumulate in different body organs, resulting in unwanted side effects [6,7].

The paucity of information on the heavy metals content of locally consumed fruits in Katsina State and the health implication of consuming food with heavy metals at levels detrimental to the health of the consumer warrant this study. Studies on heavy metals in various food samples have been carried out in Katsina State Nigeria [8-17], but no study has been carried out on the levels of heavy metals in local fruits in Katsina State. Hence, the necessity to carry out the study.

The information on heavy metals in the fruits generated will provide an insight into the extent of metal contamination and by extension the impact on food safety standards and risk to consumers.

### 2. MATERIALS AND METHODS

#### 2.1 Study Area

The study was conducted in Katsina State, with a location of between latitude 12015’N and longitude of 7030’E in the North West Zone of Nigeria, possessing a land area of 24,192 km² (9,341 sq. meters). Rainy and dry seasons are the two observable seasons in Katsina State. The rainy season starts in April and ends in October, while the dry season starts in November and ends in March [18]. This work was carried out during the dry season. The average annual rainfall, temperature, and relative humidity of Katsina State are 1,312 mm, 27.3°C and 50.2%, respectively.

#### 2.2 Sample Collection

The fruits samples comprising of Aduwa (Balanites aegyptica), Dinya (Vitex doniana), Faru (Lannea acida), Goruba (Hyphaene thebaica), Kadanya (Vitellaria paradoxa) and Kanya (Diospyros mespiliformis) were collected based on the method of Pehluvan et al. [1]. Briefly, in each sampling point, three fruit trees were selected in terms of the age and growing status. From fruit samples that were at the full maturity stage, about 1 to 4 kg of fruit samples were taken depending on fruit species. The fresh fruit samples were placed into clean plastic bags and conveyed to the laboratory. The location of sampling is as follows: Aduwa = Kaita local government area; Dinya = Funtua local government area; Faru = Kurfi local government area; Goruba = Jibia local government area; Kadanya = Kafur local government area; Kanya = Katsina local government area.

#### 2.3 Sample Preparation

The collected samples were cleaned, fragmented with a clean plastic spoon and knife and dried at ambient temperature. After drying, the seeds were removed from the dried fruits.

#### 2.4 Heavy Metals Determination

About 0.5 g of each dried sample was weighed and ashed at 550 °C for 24 hours in an electric muffle furnace (Thermolyne FB131DM Fisher Scientific). The ash was diluted with 4.5 mL concentrated hydrochloric acid (HCl) and concentrated nitric acid (HN0₃) mixed at a ratio of 3:1. The diluent is left for some minutes for proper digestion in a beaker. 50 mL of distilled water was added to the diluents to make up to 100 mL in a volumetric flask. The levels of heavy metals (Pb, Zn, Ni, Cd, Cr, Mn and Fe) were determined using AA210RAP BUCK Atomic Absorption Spectrometer flame emission spectrometer filter GLA-4B Graphite furnace (East Norwalk USA), according to standard methods [19] and the results were given in (mg/kg).
Chart 1. Map of Katsina State showing the location of the local government areas used for fruits sampling

2.5 Heavy Metal Health Risk Assessment

2.5.1 Daily Intake of metals (DIM)

The daily intake of metals from the consumption of the local fruit sample was calculated using the following equation:

$$\text{DIM} = \frac{C_{\text{metal}} \times C_{\text{factor}} \times \text{Dintake}}{\text{Bweight}}$$  \hspace{1cm} (1)$$

Where, $C_{\text{metal}}$ represents the heavy metal concentrations in the sample, $C_{\text{factor}}$ represents the conversion factor, $\text{Dintake}$ represent daily intake of the sample and $\text{Bweight}$ represents the body weight, respectively. The conversion factor (CF) of 0.085 [20] was used for the conversion of the samples to dry weights. The average daily intake of the fruits was 300g kg$^{-1} \text{d}^{-1}$ [21] and the average body weight for the adult and children population was taken from literature as 60 kg [22] and 24 kg [23] respectively.

2.5.2 Non-cancer risks

The non-cancer risks that may result from ingestion of the fruit samples containing the evaluated heavy metals by the population were computed as the target hazard quotient (THQ) using the following equation [24].

$$\text{THQ} = \frac{\text{CDI}}{\text{RfD}}$$  \hspace{1cm} (2)$$

Where, CDI is the representation of the chronic daily heavy metal intake (mg/kg/day) calculated in the previous section and RfD is the
representation of the oral reference dose (mg/kg/day) which is a quantification of the maximum permissible risk on the human population through daily exposure, bearing in mind a sensitive group in the course of a lifetime [25]. The following reference doses from the literature were used (Pb = 0.6, Cd = 0.5, Zn = 0.3, Fe = 0.7, Ni = 0.4, Mn = 0.014, Cr = 0.3) [26,27]. To evaluate the potential risk to human health through more than one heavy metal from consumption of the fruit samples, the chronic hazard index (HI) was obtained as the sum of all hazard quotients (THQ) computed for individual heavy metals for a particular exposure pathway [28]. This is calculated as follows:

\[ HI = THQ_1 + THQ_2 + \cdots + THQ_n \]  

Where, 1, 2..., n are the individual heavy metals for the fruit samples.

It is normally understood that the enormity of the effect is equal to the sum of the multiple metal exposures and that a similar working mechanism linearly affects the target organ [29]. The calculated HI was then compared to standard levels: the population is presumed to be safe when HI < 1 and at a level of concern when 1 < HI < 5 [30].

### 2.6 Cancer Risks

The likelihood of cancer risks to the adult and children consumer population in the studied fruit samples through the ingestion of carcinogenic heavy metals was calculated by the use of the Incremental Lifetime Cancer Risk (ILCR) [31].

\[ ILCR = CDI \times CSF \]  

Where, CDI represents the chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure in the adult and children population to the chemical carcinogen.

The US EPA ILCR is obtained by the use of the cancer slope factor (CSF), which is the representation of the risk produced by a lifetime average dose of 1 mg/kg BW/day and is specific to the individual contaminant [24]. The ILCR value in fruits signifies the probability of an individual's lifetime health risks from carcinogenic heavy metals' exposure [31]. For regulatory purposes the level of acceptable cancer risks (ILCR) is considered within the range of $10^{-6}$ to $10^{-4}$ [25]. The CDI value was calculated based on the following equation and CSF values for carcinogenic heavy metals were used according to the literature [32].

\[ CDI = \frac{(EDI \times EFr \times ED_{tot})}{AT} \]  

where EDI represents the estimated daily intake of metal via consumption of the samples; EFr is the exposure frequency (365 days/year); EDtot is the exposure duration of 60 years, which is the average lifetime for Nigerians; AT is the period of exposure for non-carcinogenic effects (EFr × EDtot), and 60 years lifetime for carcinogenic effect [24]. The cumulative cancer risk as a result of exposure to multiple carcinogenic heavy metals from intake of a particular type of food was taken to be the sum of the individual heavy metal increment risks and calculated by the following equation [32].

\[ \sum_{n=1}^{n} ILCR_1 + ILCR_2 + \cdots + ILCR_n \]  

Where, n = 1, 2 ..., n is the individual carcinogenic heavy metal.

### 3. RESULTS AND DISCUSSION

The present study investigated the presence of heavy metals in some local fruit samples in Katsina State, Nigeria. A total of 6 fruit samples were investigated for the presence of heavy metals in this research. As depicted in Table 1, among the heavy metals investigated, the highest concentration (mg/kg) was recorded for Fe (range: 1.728-1.954) followed by Pb (range: 1.152-1.623), Zn (range: 0.263-0.967) and Mn (range: 0.321-0.502). While Cd recorded the lowest concentration (range: 0.053-0.092) with the concentrations of Ni and Cr being below detection level (BDL).

Results from the study have pointed out that the mean Fe concentration in the sampled fruits was lower than that reported in previous studies that evaluated heavy metals in fruits from Turkey, Bangladesh, Pakistan and Poland [1,33-35]. All the samples Fe concentrations were lower than the maximum regulatory limit of 425 mg/kg for Fe in fruits [36].

In this study the Pb content of all samples exceeded the permissible limit of regulatory bodies [37] that recommended Pb concentration limits for berries and other small fruits as 0.2 mg/kg and foods as 0.5 mg/kg. These results for the mean Pb in the samples were lower than the
results recorded by Rahman and Islam [38] in a study on heavy metals in fruits from Bangladesh, Pehluvan et al. [1] for a sweet cherry, apple, plum and apricot in a study carried out in Turkey and the study of Ali et al. [39] that reported between 2.1 and 7.0 mg/kg of Pb in apricot.

The Pb result of the present study was also lower than the values reported by Hamurcu et al. [40] in apple (2.21 mg/kg), plum (2.82 mg/kg) and rosehip (1.54-2.86 mg/kg). Likewise the values reported for Pb in the present study are higher than the values reported by Nawab et al. [41] in fruits from Pakistan and Prabagar et al. [42] in fruits from Jaffna, Sri Lanka and the results of Uroko et al. [43] that reported non detection of Pb in fruit samples from Umuahia market, Nigeria.

The differences in the reported mean Pb concentration in the current study as compared to the above cited literatures may likely arise from the nature of the studied fruits, as plants have different absorptive capacities for heavy metals; another likely assumption is the nature of the heavy metal contamination source.

All the fruit samples have Cd concentrations that were above the permissible limit (0.05 mg/kg) proposed by the FAO/WHO [37]. As the sampling locations were located near the high way the above permissible mean values can be attributed to vehicular exhaust and the use of Cd containing fertilizers. But the concentration of Cd (mg/kg) in this study was much higher than those reported by Krejpcio et al. [35] in sweet cherry (0.021 mg/kg) in Poland, Pehluvan et al. [1] and Duran et al. [44] in white mulberry (0.015 and 0.63 mg/kg, respectively) in Turkey, Radwan and Salama [45] in apple (0.05 mg/kg) in Egypt and Hamurcu et al. [40] in plum (0.14 mg/kg) in Turkey.

But the Cd results are similar to those reported by Pehluvan et al. [1] for sweet cherry and plum and the Cd mean concentration range of various fruits from Anambra State, Nigeria [5]. However, the results obtained in the present study for Cd concentration were lower than those detected in some fruit and vegetable with a 25 mg/kg average value [46] and the reported Cd in fruits from Bangladesh [38].

The mean Zn values in the studied fruits fall below the WHO permissible limit of 5.00 mg/kg for Zn in fruits [47]. But the values are similar to those reported in fruit samples from some markets in Anambra State Nigeria [5] and the results from a study conducted by Dhar et al. that evaluated heavy metals in fruits from Khulna, Bangladesh [48], but are far below the range reported by Pehluvan et al. [1] (10.24-30.24 mg/kg) in fruits and the Zn concentrations in fruits reported in a study conducted by Mahfuza et al. [33].

Manufacturing concerns such as galvanization, paint, batteries, smelting, fertilizer and pesticides, fossil fuel combustion, pigment, polymer stabilizer have been reported to emit the heavy metal Zn to the environment [49], which may explain the observed values in the present study. As Katsina State is mostly an agrarian state with a handful of industries that are mainly located in the state capital.

In the present work, the Mn content of fruit samples is lower than that reported in fruits from Turkey [1] and Poland [35].

The values of the estimated daily intake (EDI) of the heavy metals from ingestion of the fruits were given in Figs. 1 and 2. From the Figures, the EDI of the heavy metals (Pb, Zn, Cd, Fe and Mn) was lower than the tolerable daily intake limit proposed by the US EPA [50] in all the samples for the children and adult population.

The calculated THQ through the consumption of the present study fruit samples provides a measure of the evaluated metal contaminants although it does not measure the risk of exposure but rather give an estimate of the level of concern [43]. The consumption of the study local fruits pointed to an indication of no adverse health effect for both the children and adults population of the study area, because the calculated fruit sample THQs are <1; hence, it may be assumed that the study fruit samples are safe for consumption (Tables 2 and 3). The THQ for the samples sequentially decreases in the order; Mn>Zn>Fe>Pb>Cd, for all the samples respectively. Similar observations of THQs <1 have been reported previously in studies that have investigated metals concentrations in foods from Katsina State, Nigeria [10-17]. The cumulative HI to the population which is the representation of the non-cancer risk for all studied fruits, revealed the risk level (HI < 1) with the highest value in Dinya fruit and the lowest in Goruba fruit (Tables 4 and 5).
Table 1. Heavy metal concentration (mg/kg) in local fruit samples

<table>
<thead>
<tr>
<th>SL</th>
<th>Sample</th>
<th>Mn</th>
<th>Zn</th>
<th>Pb</th>
<th>Cd</th>
<th>Ni</th>
<th>Fe</th>
<th>Cr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Katsina</td>
<td>Kanya</td>
<td>0.351±0.0004</td>
<td>0.952±0.0012</td>
<td>1.362±0.0001</td>
<td>0.078±0.0003</td>
<td>BDL</td>
<td>1.728±0.0004</td>
<td>BDL</td>
</tr>
<tr>
<td>Kafur</td>
<td>Kadanya</td>
<td>0.423±0.0012</td>
<td>0.263±0.0006</td>
<td>1.520±0.0002</td>
<td>0.065±0.0008</td>
<td>BDL</td>
<td>1.825±0.0012</td>
<td>BDL</td>
</tr>
<tr>
<td>Kurfi</td>
<td>Faru</td>
<td>0.368±0.0003</td>
<td>0.967±0.0002</td>
<td>1.623±0.0003</td>
<td>0.053±0.0002</td>
<td>BDL</td>
<td>1.936±0.0008</td>
<td>BDL</td>
</tr>
<tr>
<td>Funtua</td>
<td>Dinya</td>
<td>0.502±0.0001</td>
<td>0.925±0.0016</td>
<td>1.152±0.0002</td>
<td>0.092±0.0001</td>
<td>BDL</td>
<td>1.932±0.0025</td>
<td>BDL</td>
</tr>
<tr>
<td>Jibia</td>
<td>Goruba</td>
<td>0.321±0.0003</td>
<td>0.958±0.0006</td>
<td>1.274±0.0002</td>
<td>0.074±0.0006</td>
<td>BDL</td>
<td>1.951±0.0001</td>
<td>BDL</td>
</tr>
<tr>
<td>Kaita</td>
<td>Aduwa</td>
<td>0.357±0.0003</td>
<td>0.941±0.0006</td>
<td>1.331±0.0002</td>
<td>0.085±0.0003</td>
<td>BDL</td>
<td>1.748±0.0006</td>
<td>BDL</td>
</tr>
</tbody>
</table>

Values are expressed as Mean ± Standard Deviation
Key= SL= Sampling location
Fig. 1. Daily intake of heavy metal (DIM) in adults from consuming local fruit samples from Katsina State

Fig. 2. Daily intake of heavy metal (DIM) in children from consuming local fruit samples from Katsina State
Table 2. Heavy metal target hazard quotient and health risk index in adults from consuming local fruit samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Target Hazard Quotient</th>
<th>Heavy Metal</th>
<th>Health Risk Index (HRIs)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mn</td>
<td>Zn</td>
<td>Pb</td>
</tr>
<tr>
<td>Kanya</td>
<td>0.023367</td>
<td>0.002475</td>
<td>0.001695</td>
</tr>
<tr>
<td>Kadanya</td>
<td>0.032664</td>
<td>0.002352</td>
<td>0.001891</td>
</tr>
<tr>
<td>Faru</td>
<td>0.025729</td>
<td>0.002552</td>
<td>0.002020</td>
</tr>
<tr>
<td>Dinya</td>
<td>0.037836</td>
<td>0.002302</td>
<td>0.001433</td>
</tr>
<tr>
<td>Goruba</td>
<td>0.021652</td>
<td>0.002454</td>
<td>0.001585</td>
</tr>
<tr>
<td>Aduwa</td>
<td>0.023897</td>
<td>0.002342</td>
<td>0.001656</td>
</tr>
</tbody>
</table>

Table 3. Heavy metal target hazard quotient and health risk index in children from consuming local fruit samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Target Hazard Quotient</th>
<th>Heavy Metal</th>
<th>Health Risk Index (HRIs)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mn</td>
<td>Zn</td>
<td>Pb</td>
</tr>
<tr>
<td>Kanya</td>
<td>0.046795</td>
<td>0.005923</td>
<td>0.004202</td>
</tr>
<tr>
<td>Kadanya</td>
<td>0.056394</td>
<td>0.005879</td>
<td>0.004728</td>
</tr>
<tr>
<td>Faru</td>
<td>0.049061</td>
<td>0.006016</td>
<td>0.005049</td>
</tr>
<tr>
<td>Dinya</td>
<td>0.066926</td>
<td>0.005755</td>
<td>0.003584</td>
</tr>
<tr>
<td>Goruba</td>
<td>0.042795</td>
<td>0.005960</td>
<td>0.003963</td>
</tr>
<tr>
<td>Aduwa</td>
<td>0.047375</td>
<td>0.005855</td>
<td>0.004140</td>
</tr>
</tbody>
</table>

Table 4. Incremental life time cancer risk in adults consuming local fruit samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>ILCR</th>
<th>∑ILCR</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pb</td>
<td>Cd</td>
</tr>
<tr>
<td>Kanya</td>
<td>6.406000E-03</td>
<td>8.730000E-04</td>
</tr>
<tr>
<td>Kadanya</td>
<td>7.149000E-03</td>
<td>7.280000E-04</td>
</tr>
<tr>
<td>Faru</td>
<td>7.634000E-03</td>
<td>5.930000E-04</td>
</tr>
<tr>
<td>Dinya</td>
<td>5.418000E-03</td>
<td>1.030000E-03</td>
</tr>
<tr>
<td>Goruba</td>
<td>5.992000E-03</td>
<td>8.850000E-04</td>
</tr>
<tr>
<td>Aduwa</td>
<td>6.260000E-03</td>
<td>9.520000E-04</td>
</tr>
</tbody>
</table>

Table 5. Incremental life time cancer risk in children from consuming local fruit samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>ILCR</th>
<th>∑ILCR</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Pb</td>
<td>Cd</td>
</tr>
<tr>
<td>Kanya</td>
<td>1.601533E-02</td>
<td>2.183745E-03</td>
</tr>
<tr>
<td>Kadanya</td>
<td>1.787320E-02</td>
<td>1.819785E-03</td>
</tr>
<tr>
<td>Faru</td>
<td>1.908434E-02</td>
<td>1.483830E-03</td>
</tr>
<tr>
<td>Dinya</td>
<td>1.354601E-02</td>
<td>2.576100E-03</td>
</tr>
<tr>
<td>Goruba</td>
<td>1.498056E-02</td>
<td>2.211750E-03</td>
</tr>
<tr>
<td>Aduwa</td>
<td>1.365081E-02</td>
<td>2.379720E-03</td>
</tr>
</tbody>
</table>
The International Agency for Research on Cancer (IARC) has listed the heavy metals Cd and Pb as metal carcinogens [51,52]. Prolonged exposure to low doses of Cd, and Pb could therefore manifest into various types of cancers [6]. Tables 4 and 5 are the representations of the total cancer risk of heavy metals to the children and adults consumer population in the study fruit samples. From the tables a deduction can be made that the cancer risks for the heavy metal Cd in the adults population were at the threshold of the risk limit (ILCR > 10$^{-3}$) for cancer, while the metal Pb in adults and the metals Pb and Cd in children were above the risk threshold (ILCR ≤ 10$^{-5}$) for cancer. The trend of risk for developing cancer due to the ingestion of the fruit samples revealed; Faru> Kadanya > Kanya > Goruba > Dinya> Aduwa.

4. CONCLUSION

The study evaluates heavy metal concentrations and their possible health risks indices in local fruit samples from Katsina State, Nigeria. Findings from this work have revealed that concentration values of Pb in the samples were generally above the permissible limit of Pb in fruits set by the regulatory bodies. While the rest of the evaluated heavy metals have concentrations that fall within acceptable safe limits. The results have revealed that the computed EDIs of the heavy metals were lower than the tolerable daily intake limit set by the regulatory bodies in all the fruit samples. Target Hazard Quotient of less than 1 (THQ < 1) was recorded for all the evaluated heavy metals. The calculated ILCR for Cd was at the threshold risk limit for cancer (>10$^{-4}$) in all the studied fruits in adults. While Pb for adults and Cd for children lies within the moderate cancer risk limit (>10$^{-5}$) and the ILCR for Pb in children was above the moderate cancer risk limit (>10$^{-5}$) in children. Moreover, cumulative cancer risk (ΣILCR) of all the studied fruits were within the moderate cancer risk level (>10$^{-5}$) in adults while in children it was above the level (>10$^{-2}$). The study has revealed that the consumption of the studied local fruits in Katsina State is of a public health concern as they may add to the population's cancer risk.

With the ability of heavy metals to biomagnify and bioaccumulate in the feeding chain it is recommended that continuous routine monitoring of fruits consumed in the state be carried out, to have a wider picture and history of these pollutants for possible intervention if such a need is to arise.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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