

HEALTH RISKS EVALUATION OF HEAVY METALS IN DRUMSTICK LEAF SAMPLES CULTIVATED IN KATSINA STATE, NIGERIA

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ABSTRACT

This work contributes to the monitoring of heavy metals in agricultural produce in Katsina State, Northwest Nigeria, and the possible health risk to the consumer population. Drumstick leaf samples from the three senatorial zones that constitute to make up Katsina state in the North West of Nigeria were collected and the concentrations of seven heavy metals (Pb, Cd, Cr, Fe, Zn, Mn and Ni) in all the samples were evaluated by atomic absorption spectrometry. The health risk assessment methods developed by the United States Environmental Protection Agency (US EPA) were employed to explore the potential health hazards of heavy metals in the samples on the children and adult population. The highest concentration (mg/kg) was observed for Zn (range: 0.826-1.428), followed by Fe (range: 0.596-1.369), Pb (range: 0.521-1.251) and Cr (range: 0.138-0.310). While Cd has the lowest concentration (range: 0.039-0.073) with the heavy metals Mn and Ni being below detection level (BDL). The mean Pb concentrations in all the samples were above the permissible regulatory limit. The target hazard quotient (THQ) and the hazard index (Hi) for the heavy metals evaluated were within the safety limit. The overall cancer risk to the adults based on pseudo-total metal concentrations exceeded the target value, mainly contributed by the heavy metal Pb. Cd is the primary heavy metal posing non cancer risks while Pb caused the greatest cancer risk. It was concluded that consumption of the Drumstick leaf samples from Katsina State may contribute to the population cancer burden.

KEY WORDS: Vegetables, Pollution, Environmental pollutants, Population, Health hazard

1. INTRODUCTION

The leading causes of morbidity and mortality in the developed countries, and now increasingly becoming prevalent in developing countries are chronic disease conditions that include Cardiovascular and cerebrovascular diseases, cancer, diabetes, metabolic syndrome, obesity, neurocognitive disorders, and immune dysfunction such as autoimmune diseases (WHO, 2011). The manifestation of heavy metal toxicity commonly involves the brain and kidney and can also occur in other parts of the body, for example the metalloids arsenic, is clearly capable of causing cancer, and hypertension can result in individuals exposed to lead, and renal toxicity can result in individual exposed to cadmium (Gottipolu *et al.*, 2012).

It has been shown over the years that various anthropogenic and natural activities contribute to heavy metal build up in the environment (Rayhan and Hosna, 2021).

Moringa oleifera (Zogale Gandi in Hausa language) is cultivated for its food, medicinal and culinary value and its leaves, fruits and roots are the parts used. It is commonly known as the 'horseradish' tree arising from the taste of a condiment prepared from the roots or 'drumstick' tree due to the shape of the pods. *M. oleifera* has a host of other country-specific vernacular names, an indication of the significance of the tree around the world (Nautiyal and Venkataram, 1987).

The present study investigated the presence of heavy metals in Drumstick leaf which is a component of the diet among the population in Katsina state, Nigeria. "Kwadon Zogale" a salad prepared from boiled drumstick leaves mixed with groundnut cake and spices is a delicacy of inhabitants of Katsina state.

2. METHODOLOGY

2.1 Sampling Area

The study was conducted during 2017-2018 in Katsina State, Nigeria located between latitude 12°15'N and longitude 7°30'E in the North West Zone of Nigeria, with an area of 24,192km² (9,341 sq. meters) and a population of 7.6 million with the growth rate of 3.0% per annum. With more than 50% of the population between the ages of 15-64 years and 80% of the population engaged in subsistence farming and livestock rearing (Katsina State, 2016). The State has a rainy season that begins in April and ends in October, while the dry season starts in November and last till March. The average annual rainfall, temperature, and relative humidity of Katsina State are 1,312 mm, 27.3°C and 50.2%, respectively.

2.2 Sampling and Sample Preparation

The leaves of the vegetable sample were collected with the consent of the farmers from the sampling site using a cleaned and decontaminated polyethylene bag. The edible portion of the vegetable sample was cut into small pieces, washed with tap water and then rinsed with distilled deionized water. These were placed on cardboard papers and dried in open-air in the laboratory for three weeks. The dried samples were then grinded into fine powder using a ceramic pestle and mortar and stored in a stoppered plastic bottle.

2.3 Sample Digestion

The plant sample was digested according to the procedure adopted by Awofolu (2005); whereby 0.5 g of the powdered sample was weighed into a 100 mL beaker and 5 mL of concentrated HNO_3 and 2 mL HClO_4 were added. The mixture was then heated on a hot plate at 95°C until the solution became clear. It was then filtered into a 100 mL volumetric flask and made up to the mark with distilled water.

2.4 Heavy Metal Determination

The concentration of heavy metals in the sample was determined using Atomic Absorption Spectrophotometer (Buck 210 VGP Model) equipped with a digital read-out system. Working standards were used, after serial dilution of 1000 ppm metal stock solution in each case. Calibration curves were generated by plotting absorbance values versus concentrations. By interpolation, the concentration of the metals in sample digest was determined as described by Audu and Lawal (2006).

2.5 Heavy Metal Health Risk Assessment

2.5.1 Daily Intake of Metals (DIM)

The ingestion of heavy metals in the samples depicted as the daily intake of metals (DIM) was calculated using the following equation.

$$\text{DIM} = (\text{C}_{\text{metal}} \times \text{C}_{\text{factor}} \times \text{D}_{\text{intake}}) / \text{B}_{\text{weight}} \dots\dots\dots \text{eqn. (1)}$$

With C_{metal} standing for heavy metal concentration in the sample, C_{factor} representing the conversion factor (Cf) which was taken as 0.085 (Jan *et al.*, 2010) used in converting the sample to its dry weight, D_{intake} representing the daily intake of the sample taken from literature as 0.527 kg person⁻¹ d⁻¹ (Balkhaira *et al.*, 2015), and B_{weight} representing the average body weight which is also taken from the literature as 60 kg (Orisakwe *et al.*, 2015) for adults and 24 kg (Ekhatior *et al.*, 2017) for children. The same values were used to evaluate the HRI.

2.5.2 Non-Cancer Risks

Non-carcinogenic risks for individual heavy metal of the samples will be evaluated by computing the target hazard quotient (THQ) using the following equation (Micheal *et al.*, 2015).

$$\text{THQ} = \text{CDI} / \text{RfD}$$

CDI was the chronic daily heavy metal intake (mg/kg/day) obtained from the previous section and RfD is the oral reference dose (mg/kg/day) which represents an estimation of the maximum permissible risk on human population through daily exposure, taking into consideration a sensitive group during a lifetime (Li and Zhang, 2010). The following reference doses were used ($\text{Pb} = 0.6$, $\text{Cd} = 0.5$, $\text{Zn} = 0.3$, $\text{Fe} = 0.7$, $\text{Ni} = 0.4$, $\text{Mn} = 0.014$, $\text{Cr} = 0.3$) (Li *et al.*, 2013; US-EPA, 2002). To evaluate the potential risk to human health through more than one heavy metal, chronic hazard index (HI) will be obtained as the sum of all hazard quotients (THQ) calculated for individual heavy metals for a particular exposure pathway (NFPCSP Nutrition Fact Sheet, 2011). It was calculated as follows:

$$\text{HI} = \text{THQ}_1 + \text{THQ}_2 + \dots + \text{THQ}_n$$

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

It is based on the assumption that the magnitude of the effect is proportional to the sum of the multiple metal exposures and that similar working mechanism linearly affects the target organ (RAIS, 2007). The population is assumed to be safe when $\text{HI} < 1$ and in a level of concern when $1 < \text{HI} < 5$ (Guerra *et al.*, 2012).

2.5.3 Cancer Risks

The possibility of cancer risks in the studied samples through intake of carcinogenic heavy metals will be estimated using the Incremental Lifetime Cancer Risk (ILCR) (Liu *et al.*, 2013).

$$\text{ILCR} = \text{CDI} \times \text{CSF}$$

Where, CDI is chronic daily intake of chemical carcinogen, mg/kg BW/day which represents the lifetime average daily dose of exposure to the chemical carcinogen.

The US EPA ILCR is obtained using the cancer slope factor (CSF), which is the risk produced by a lifetime average dose of 1 mg/kg BW/day and is contaminant specific (Micheal *et al.*, 2015). The following cancer slope factor for

specific heavy metals will be used; Pb = 0.0085 mg/kg/day (Kamunda *et al.*, 2016), Cd = 0.38 mg/kg/day (Yang *et al.*, 2018), Ni = 1.7 mg/kg/day (Javed *et al.*, 2016). ILCR value in sample represents the probability of an individual's lifetime health risks from carcinogenic heavy metals' exposure (Pepper *et al.*, 2012). The level of acceptable cancer risk (ILCR) for regulatory purposes was considered within the range of 10^{-6} to 10^{-4} (Li and Zhang, 2010). The CDI values will be calculated on the basis of the following equation and CSF values for carcinogenic heavy metals will be used according to the literature (Liu *et al.*, 2013).

$$CDI = (EDI \times EFr \times ED_{tot})/AT$$

where EDI is the estimated daily intake of metal via consumption of the samples; EFr is representing the exposure frequency (365 days/year); ED_{tot} is the exposure duration of 60 years, average lifetime for Nigerians; AT is the period of exposure for non-carcinogenic effects ($EFr \times ED_{tot}$), and 60 years' life time for carcinogenic effect (Micheal *et al.*, 2015). The cumulative cancer risks in the samples as a result of exposure to multiple carcinogenic heavy metals due to consumption of a particular type of food were assumed to be the sum of the individual heavy metal increment risks and calculated by the following equation (Liu *et al.*, 2013).

$$\sum I_n = ILCR_1 + ILCR_2 + \dots + ILCR_n$$

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

3. RESULTS AND DISCUSSION

A total of 11 Drumstick leaf samples were analyzed for the presence of heavy metals in this study. As shown in Figure 1, among the heavy metals evaluated, the highest concentration (mg/kg) was observed for Zn (range: 0.826-1.428), followed by Fe (range: 0.596-1.369), Pb (range: 0.521-1.251) and Cr (range: 0.138-0.310). While Cd has the lowest concentration (range: 0.039-0.073) with the heavy metals Mn and Ni being below detection level (BDL). In contrast a study conducted in Jibia local Government area of Katsina State Nigeria on the health risks evaluation of heavy metals in drumstick leaf samples have reported the detection of the heavy metal Ni (Yaradua *et al.*, 2023), an observation that was attributed to E-waste and illegal mining in the area the study was conducted. The mean concentrations of the heavy metal Pb (Figure 1), were observed to be higher than the 0.01 mg/kg which is the maximum permissible limit set by WHO/FAO and also the maximum allowable concentration of 0.02 mg/kg by EU and 0.05 mg/kg limit set by USEPA (Landrigan *et al.*, 2017). The high percentage of samples which were in violation of the maximum permissible limits of Pb set by the WHO, EU, and US EPA is a cause for public health concern. Within permissible mean concentrations were observed for the heavy metals Zn, Fe, Cr and Cd in the present study. The possible explanation for the observed values of within permissible limit of the heavy metals may be due to their low values and lower pollution indices in the agricultural sites where the vegetables were cultivated (Yaradua *et al.*, 2020; Yaradua *et al.*, 2022).

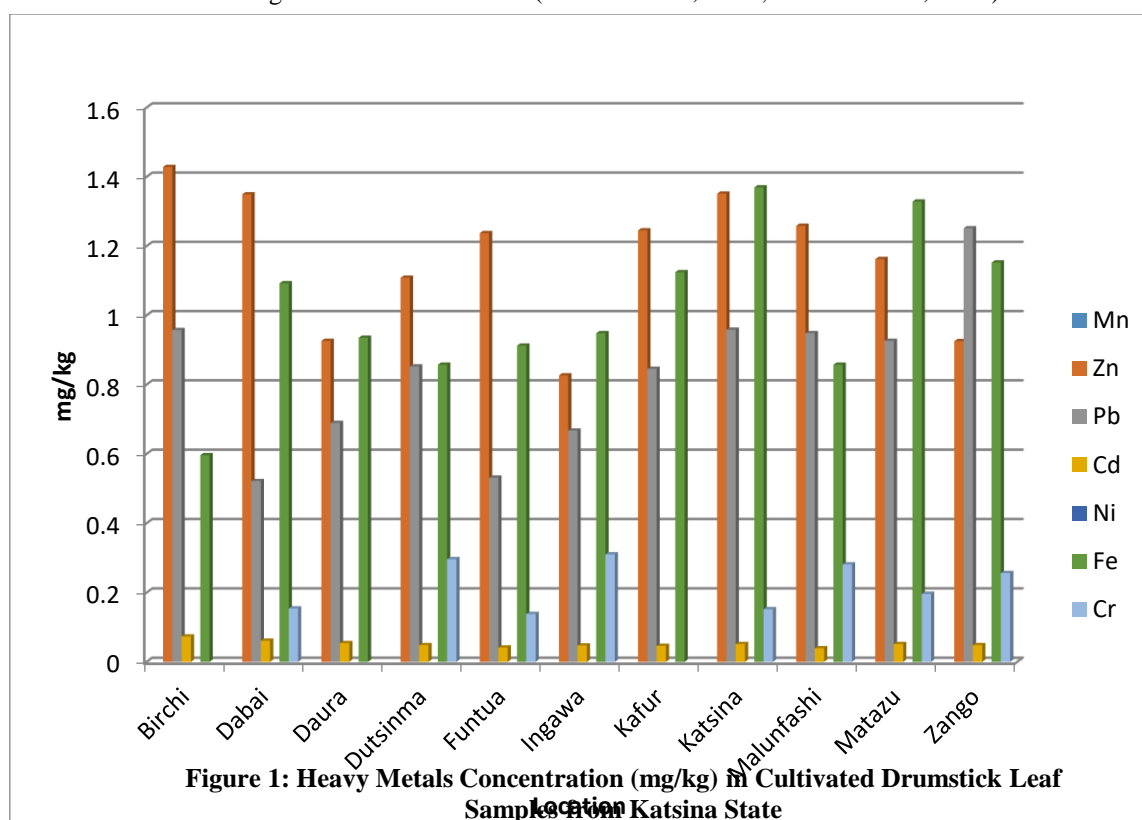


Figure 1: Heavy Metals Concentration (mg/kg) in Cultivated Drumstick Leaf Samples from Katsina State

The results for the estimated daily intake (EDI) of the heavy metals on consumption of the samples were given in Tables 1 and 2. From the Tables the estimated daily intake of the heavy metals (Pb, Zn, Cd, Cr and Fe) were lower than the tolerable daily intake limit set by the USEPA (2013) in all samples. The order of sequence of daily metal intake in both adult and children from consumption of the Drumstick leaf samples in the various sampling sites were as follows: Birchi (Zn>Pb>Fe>Cd); Dabai (Zn>Fe>Pb>Cr>Cd); Daura (Fe>Zn>Pb>Cd); Dutsinma (Zn>Fe>Pb>Cr>Cd); Funtua (Zn>Fe>Pb>Cr>Cd); Ingawa (Fe>Zn>Pb>Cr>Cd); Kafur (Zn>Fe>Pb>Cd); Katsina (Fe>Zn>Pb>Cr>Cd); Malunfashi (Zn>Pb>Fe>Cr>Cd); Matazu (Fe>Zn>Pb>Cr>Cd); Zango (Pb>Fe>Zn>Cr>Cd).

Table 1 Daily Intake of Heavy Metal in Adults from Consumption of Cultivated Drum Stick Leaf Samples from Katsina State

Location	Zn	Pb	Cd	Fe	Cr
Birchi	0.001066	0.000715	0.000055	0.000445	BDL
Dabai	0.001007	0.000389	0.000046	0.000815	0.000115
Daura	0.000691	0.000514	0.000040	0.000698	BDL
Dutsinma	0.000827	0.000636	0.000035	0.000639	0.000221
Funtua	0.000924	0.000396	0.000031	0.000681	0.000100
Ingawa	0.000617	0.000498	0.000035	0.000708	0.000231
Kafur	0.000930	0.000631	0.000034	0.000839	BDL
Katsina	0.001001	0.000715	0.000038	0.001022	0.000
Malunfashi	0.000939	0.000705	0.000029	0.000640	0.000210
Matazu	0.000868	0.000691	0.000038	0.000991	0.000146
Zango	0.000690	0.000934	0.000036	0.000860	0.000191

Table 2 Daily Intake of Heavy Metal in Children from Consumption of Cultivated Drum Stick Leaf Samples from Katsina State

Location	Zn	Pb	Cd	Fe	Cr
Birchi	0.002665	0.001786	0.000136	0.001112	0.000623
Dabai	0.002518	0.000972	0.000114	0.002038	0.000287
Daura	0.001728	0.001286	0.000101	0.001600	BDL
Dutsinma	0.002068	0.001590	0.000090	0.001600	0.000553
Funtua	0.002309	0.000991	0.000026	0.001702	0.000258
Ingawa	0.001542	0.001245	0.000088	0.001863	0.000579
Kafur	0.002324	0.001577	0.000086	0.003218	BDL
Katsina	0.002522	0.001788	0.000095	0.002551	0.000284
Malunfashi	0.002348	0.001769	0.000073	0.001600	0.000525
Matazu	0.002169	0.001728	0.000095	0.002408	0.000366
Zango	0.002335	0.002335	0.000090	0.002150	0.000478

The non-cancer risks (THQ) of the investigated heavy metals through the consumption of the Drumstick leaf samples for both adults and children inhabitants of the study area were determined and presented in Tables 3 and 4. Risk level of Target Hazard Quotient (THQ < 1) was observed for all the evaluated heavy metals for both adults and children. It indicates that intake of these heavy metals through consumption of the samples does not pose a considerable non-cancer risk. The THQ for the samples was in the decreasing order Mn>Zn>Fe>Pb>Cd, for all the samples respectively. The sequence of risk was the same for both adults and children although the children had higher THQ values in all cases. Yaradua *et al.* (2019), Mahfuza *et al.* (2017), Micheal *et al.* (2015) and Liu *et al.* (2013) have reported similar observations previously.

Table 3 Heavy Metal Target Hazard Quotient and Health Risk Index in Adults from Consumption of Cultivated Drum Stick from Katsina State

			Target Hazard Quotient			Health Risk Index (HRIs)
Location		Heavy Metal				
	Zn	Pb	Cd	Fe	Cr	
Birchi	0.003554	0.001191	0.000109	0.000636	BDL	0.002280
Dabai	0.003357	0.000648	0.000091	0.001165	0.000383	0.002372
Daura	0.002302	0.000857	0.000081	0.000997	BDL	0.001944
Dutsinma	0.002757	0.001060	0.000072	0.000914	0.000737	0.002581
Funtua	0.003078	0.000660	0.000061	0.000673	0.000343	0.001835
Ingawa	0.002256	0.000830	0.000070	0.001611	0.000772	0.002889
Kafur	0.003098	0.001051	0.000069	0.001199	BDL	0.002434
Katsina	0.003362	0.001592	0.000076	0.001460	0.000378	0.001990
Malunfashi	0.003131	0.001172	0.000058	0.000914	0.000699	0.002521
Matazu	0.002892	0.001152	0.000076	0.001416	0.000488	0.002735
Zango	0.002302	0.001557	0.000072	0.001229	0.000637	0.002712

Table 4 Heavy Metal Target Hazard Quotient and Health Risk Index in Children from Consumption of Cultivated Drum Stick from Katsina State

			Target Hazard Quotient			Health Risk Index (HRIs)
Location		Heavy Metal				
	Zn	Pb	Cd	Fe	Cr	
Birchi	0.008884	0.002977	0.000273	0.001589	0.002078	0.015760
Dabai	0.008343	0.001621	0.000227	0.002912	0.000958	0.014116
Daura	0.009095	0.002143	0.000202	0.002285	BDL	0.013724
Dutsinma	0.006894	0.002650	0.000179	0.002285	0.001842	0.013850
Funtua	0.007696	0.001652	0.000523	0.002432	0.000859	0.012690
Ingawa	0.005139	0.002075	0.000176	0.002661	0.001929	0.019790
Kafur	0.007746	0.002629	0.000172	0.004597	BDL	0.015143
Katsina	0.008405	0.002980	0.000190	0.003650	0.000946	0.016172
Malunfashi	0.007827	0.002949	0.000146	0.002285	0.001748	0.014955
Matazu	0.007229	0.002881	0.000190	0.003541	0.001219	0.015060
Zango	0.005755	0.003892	0.000179	0.003072	0.001593	0.016103

The computed ILCR and cumulative incremental lifetime cancer risk (Σ ILCR) for Cd, and Pb through the vegetable samples in both adult and children are presented in Tables 5 and 6. From the results, the ILCR for Pb in the studied Drumstick leaf has reached the moderate risk limit ($>10^{-3}$) in adults while in children it was above the moderate risk limit ($>10^{-2}$). The ILCR for Cd in children has reached the moderate risk limit ($>10^{-3}$) while in adults it was below the threshold limit ($>10^{-5}$). The trend of risk for developing cancer as a result of consuming the studied samples showed: Zango > Birchi > Katsina > Matazu > Malunfashi > Dutsinma > Kafur > Daura > Ingawa > Dabai > Funtua. The calculated cumulative cancer risk (Σ ILCR) of all the studied samples was at the moderate risk limit ($>10^{-3}$) in adults while in children it was above the limit ($>10^{-2}$).

The range of ILCR and Σ ILCR from consumption of all the evaluated samples as highlighted above, which raises the level of health concern for the consumer population as they may contribute to the population cancer burden was similar to what was reported by Gebeyehu and Bayissa (2020), in vegetables from Mojo Ethiopia. It was also similar to the ILCR and Σ ILCR reported from consumption of food samples from China, Jamaica and Bangladesh (Chen *et al.*, 2011; Chang *et al.*, 2014; Antoine *et al.*, 2017; Mahfuza *et al.*, 2017; Li *et al.*, 2018; Islam *et al.*, 2018). However, the results differ from the results for vegetables from some selected communities from ONELGA Rivers State Nigeria. A study that reported non-carcinogenic cancer risks from the vegetable samples in the study (Ogbo and Patrick-Iwuanyanwu, 2019).

Table 5 Incremental Life Time Cancer Risk in Adults from Consumption of Cultivated Drumstick Leaves from Katsina State

Location	ILCR		Σ ILCR
	Pb	Cd	
Birchi	4.5010E-03	9.3000E-05	4.5940E-03
Dabai	2.4510E-03	7.8000E-05	2.4280E-03
Daura	3.2410E-03	6.9000E-05	3.3090E-03
Dutsinma	4.0070E-03	6.1000E-05	4.0680E-03
Funtua	2.3580E-03	5.2000E-05	2.4100E-03
Ingawa	3.1370E-03	6.0000E-05	3.1970E-03
Kafur	3.9740E-03	5.8000E-05	4.0330E-03
Katsina	4.5060E-03	6.5000E-05	4.5710E-03
Malunfashi	4.4310E-03	5.0000E-05	4.4800E-03
Matazu	4.3550E-03	6.5000E-05	4.4200E-03
Zango	5.8840E-03	6.1000E-05	5.9450E-03

Table 6 Incremental Life Time Cancer Risk in Children from Consumption of Cultivated Drumstick Leaves from Katsina State

Location	ILCR		Σ ILCR
	Pb	Cd	
Birchi	1.8755E-02	4.0875E-03	2.2843E-02
Dabai	1.0211E-02	3.4059E-03	1.3616E-02
Daura	1.3724E-02	3.0236E-03	1.6747E-02
Dutsinma	1.6697E-02	2.6877E-03	1.9385E-02
Funtua	1.0406E-02	7.8390E-04	1.1190E-02
Ingawa	1.3072E-02	2.6319E-03	1.5703E-02
Kafur	1.6885E-02	2.5757E-03	1.7142E-02
Katsina	1.8775E-02	2.8556E-03	2.1630E-02
Malunfashi	1.8579E-02	2.1837E-03	2.0762E-02
Matazu	1.8148E-02	2.8557E-03	2.1003E-02
Zango	2.4517E-02	2.6877E-03	2.7204E-02

4. CONCLUSION

This study explores the possible health risks to the population from consumption of Drumstick leaf samples. The results have revealed that the mean concentration for the heavy metal Pb in all the samples evaluated were above the permissible regulatory level. The heavy metals Mn and Ni were below the detection level. While the heavy metals Zn, Fe, Cr and Cd have mean concentration values that were within the permissible regulatory values. The target hazard quotient and

health risks indices to the population from the evaluated metals indicated a non-cancer risk. While the cancer risks evaluation indicated a possible cancer risk to the population from consumption of the samples.

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