https://dx.doi.org/10.4314/dujopas.v10i1b.2
ISSN (Print): 2476-8316
ISSN (Online): 2635-3490
Dutse Journal of Pure and Applied Sciences (DUJOPAS), *Vol. 10 No. 1b March 2024*

Detection of Mercury in Skin Lightening Cosmetics Marketed in Katsina Metropolis, Katsina State, Nigeria

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Abstract

Deliberate use of metals as active cosmetic ingredients has been banned in many countries due to their persistent nature, toxicity and transdermal exposure over a certain period of time. Fifteen (15) samples were purposively sampled to cover the most frequently used brands as the samples for the study. The samples cover five (5) of each of the skin lightening creams, lotions and soaps from different cosmetics shops in Katsina central market and other outlets within the metropolis of Katsina. Qualitative inorganic analysis was conducted for the detection of the presence of mercury in the samples. Also quantitative analysis of the mercury was carried out using Atomic Absorption Spectrometry (AAS). The results for the qualitative inorganic analysis showed that all the samples were mercury (II) positive with 0.5N KI reagent through the formation of reddish orange precipitate, and no positive reaction of mercury was detected using 2.0N NaOH reagent. Quantitative analysis of the mercury revealed the presence of mercury in all the analyzed samples. The concentration of mercury ranged from 27.7 ± 0.0006 to 61.5 ±0. 0007 ppm. From the results obtained, it is thus evident that the analyzed samples were mercury (II) positive and above the recommended WHO permissible limit. Therefore, there may be a serious health hazard to the users of such products in Katsina metropolis. It is recommended that periodic analysis of such skin lightening products should be conducted by the regulatory bodies to ascertain their safety to the users.

Keywords: Mercury, Atomic Absorption Spectrometry, Lotions, Soaps, Creams

INTRODUCTION

Heavy metals are metals that have density range of 3.5 to 7.0 g/cm^3. They are inorganic chemicals, toxic and poisonous even in trace amount. They are not biologically degradable but easily assimilate and bio-accumulated in the protoplasm of living organism. Some common examples of these heavy metals are Mercury (Hg), Cadmium (Cd), Arsenic (As), Chromium (Cr), Copper, (Cu), Lead (Pb), Manganese (Mn), Molybdenum (Mo), Nickel (Ni) (Appenroth, 2010)

Hg as a heavy metal is well known for its toxicity. It is broadly distributed in the environment by natural and anthropogenic activities including mining, agriculture, and industrial activities (Peregrino *et al*., 2011). However, its presence in cosmetic products is a less explored route for

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Hg exposure (Peregrino *et al.,* 2011). It is categorized in a group of three heavy metals beside Pb and Cd with high toxicity which might be absorbed into the human body via cosmetics, food, or air (Islam, 2018). Hg exists in three forms namely; elemental, organic and inorganic. In cosmetic products such as skin lightening products, inorganic Hg is most widely used which can be present as ammoniated Hg, Hg iodide, mercurous chloride, mercurous oxide, or mercuric chloride (WHO, 2019). Organic Hg compounds like thiomersal which contains ethyl mercury an diphenyl mercuric salts are present as cosmetic preservatives in eye makeup including mascara and eye makeup cleansing products (Samia *et al.,* 2017; WHO, 2019). These types of Hg are dissimilar in their levels of toxicity and effects on the nervous, digestive and immune systems, as well as the lungs, kidneys, skin and eyes (WHO, 2019).

Interestingly, Hg has been used as preservative and skin lightening agent in cosmetic and pharmaceutical products for several years (Mallkuci, 2014), thus these skin lightening products are manufactured in many countries in the world, like Bangladesh, China, Hong Kong, Jamaica, Lebanon, Malaysia, Mexico, Pakistan, Philippines, USA, Republic of Korea, etc. (WHO, 2019). It is used as an antifungal and antiseptic agent in many drug products such as vaccines, ointment, and contact- lens solution and in ear, eye and nasal drop solutions (Mallkuci, 2014). Currently, Hg can be present in pharmaceutical and cosmetic products even when it is not listed on the label or information leaflet (Mallkuci, 2014). The routes of Hg exposure may involve skin absorption into the blood stream from cosmetic, inhalation of Hg vapor by the user and even those in the household of the user (Peregrino *et al.,* 2011).

Hg salts inhibit the formation of melanin by competing with copper in the tyrosinase active site and also inhibit the production of L-Dopaquinone, thereby resulting to a lighter skin tone (Burger *et al.,* 2016). The mixture of Hg (II) salt is usually used which is more reactive due to its lightening effect within 5-7 days (Islam, 2018). Adverse effect of the inorganic Hg contained in skin whitening products including creams and soaps include kidney damage, skin rashes, skin discoloration and scarring, reduction in the skin's resistance to bacterial and fungal infections, anxiety, depression, psychosis and peripheral neuropathy (WHO, 2019). Other complications associated mercurial toxicity include; allergic reaction, skin irritation, neurotoxic manifestation (Samia *et al.,* 2017), exogenous ochronosis, dermatitis, facial acnes, facial hypertrichosis, hyper and hypo- pigmentation, impaired wound healing, the fish odor syndrome, steroid addition syndrome and predisposition to diseases (Akiibinu *et al.,* 2019).

Short- term exposure to Hg in high dosage might lead to vomiting, diarrhea, permanent damage to brain and kidney, impaired fetal growth, cancer, tremors, and weakness, memory loss (Islam, 2018; Akiibinu et al., 2019). According to Yetunde (2017), some mothers have started to bleach the skin of their babies. This author reiterated further that one of the mothers interrogated in her study affirmed that "black babies are not cute". Mercurial toxicity may also be transferred from a bleaching practicing mother to her nursing infant during feeding (Mallkuci, 2014). High concentration of Hg in pregnant women is known to increase the risk of permanent brain damage, pink baby syndrome (Akiibinu *et al.,* 2019). Long- term use of Hg based skin lighteners often produce a characteristic slate gray skin color, accumulated levels in hair, blood and urine (Akiibinu *et al.,* 2019). Adulterated skin lightening cosmetics containing Hg are widely available in beauty stores in Nigeria despite it is hazard, mostly above recommended permissible limit of 1 ppm (WHO, 2019). More so there was no work on ground to ascertain the safety of such products used by the people of Katsina metropolis as such products are not properly labeled by the manufactures. The aim of this research therefore was to determine the presence of Hg, its type, as well as its concentration levels in skin

lightening creams, soaps and lotions as the samples marketed in the metropolis of Katsina, Katsina State.

MATERIALS AND METHODS

Chemicals and Reagents

The chemicals used were of analytical grades. Nitric acid (69%), Mercury (ii) oxide (HgO), Sodium Hydroxide (NaOH), Potassium Iodide (KI), Hydrochloric acid (HCl) were obtained from Umaru Musa Yar'adua University laboratory. Distilled water and Whatman® paper no. 1 were equally employed in this study.

Sample Collection

For the purpose of this study, a total of Fifteen (15) skin lightening cosmetic samples: five (5) of the skin lightening creams, soaps and lotions were procured from different cosmetic shops in Katsina central market and other outlets within Katsina metropolis. The choice of the brands was purposively made to reflect the brands mostly used by the people of different income groups. The samples were anonymously coded as C1, C2, C3, C4 and C5 for skin lightening creams, S1, S2, S3, S4 and S5 and L1, L2, L3, L4, and L5 for skin lightening soaps and lotions respectively. The samples' information as written by the manufacturers is presented in Table 1.

Table 1: Information contained on the packages of sampled skin lightening cosmetics

S/N	Sample Code	Country of Origin	NADFDAC No.	Expiry Date
1	C1	Cote D' Ivoire	02-6454	11/23
2	C2	Cote D' Ivoire	Nil	06/24
3	C3	USA	Nil	07/24
4	C4	Cote D' Ivoire	Nil	03/24
5	C5	Togo	Nil	02/25
6	L1	Cote D' Ivoire	Nil	06/24
7	L2	Cote D' Ivoire	Nil	01/24
8	L3	USA	Nil	03/24
9	L4	Cote D' Ivoire	Nil	07/24
10	L5	Togo	Nil	11/24
11	S1	Cote D' Ivoire	A2-0423	-
12	S2	Cote D' Ivoire	Nil	-
13	S3	USA	Nil	-
14	S4	Cote D' Ivoire	Nil	-
15	S5	Togo	Nil	10/25

Key: c: creams, l: lotions, s: soaps

Instrumentation

Atomic Absorption Spectrophotometer (AAS) model Agilent 55A was used for the quantitative determination of Hg. It is specification include lamp current (4mA), fuel (Acetylene), Support (Air), Flame Stoichiometry (Oxidizing), Wavelength (253.7 nm), Slit width (0.5 nm), Optimum working range (2-400 ppm), Analytical balance (AR 2140) with a readability of 0.0001 g was used for sample measurements and Stuart® SB 160 Hot plate capable of attaining a surface temperature of 300 °C was used for sample digestion.

Preparation of 2.0 N NaOH

About 8.0 g of NaOH pellets were measured, dissolved in 30 mL distilled water, and then diluted in a 100 mL volumetric flask marked with the same solvent.

Preparation of 0.5 N KI
About 8.3 g of KI was measured, dissolved in 30 mL distilled water, and then diluted in a 100 mL volumetric flask marked with the same solvent.

Sample Test with 2.0 N NaOH
About 4 mL of the sample solution was measured into the test tube and added with 3 drops of 2.0 N NaOH. Black precipitate; ppt, was observed if it was mercury (I) positive and yellow ppt for mercury (II) positive.

Sample Test with 0.5 N KI
About 4 mL of the sample solution was measured into the test tube and added with 3 drops of 0.5 N KI. Green ppt was observed if it was mercury (I) positive and red ppt for mercury (II) positive.

Preparation of Mercury Stock Solution (1000 ppm)
About 1.08 g of mercury (II) Oxide; HgO was measured, dissolved in 20 mL 5 Molar hydrochloric acid, and then diluted to 1 L volumetric flask with distilled water (Perkin, 1996).

Preparation of Mercury Working Standards
Working standards of Hg were prepared from the prepared 1000 ppm stock solution of mercury. By serial dilution, 200, 400, and 600 ppm were prepared by measuring 20, 40, and 60 mL respectively of the solution into the 100 mL volumetric flask and set to the mark with distilled water. The prepared working standards were used for the calibration curve.

Sample Preparation and Determination of Mercury Level
The sample solutions were prepared in wet destruction according to the proposed Islam (2018) procedures. 2.0 g of each of the samples was measured 10 mL concentrated HNO_3 was added, the mixture was heated on a hot plate at 80 °C until it got soluble, and 20 mL of distillate was added into the evaporation residue and reheated for 15 minutes. The solution was allowed to cool and filtered using Whatman® filter paper no. 1. The blank sample was also prepared using the same condition. Hg was determined directly from the final solution in the sample bottles using an Agilent 55A AAS whereby the instrument gave out the absorbence of the metal. The concentration of the metal was then determined from the calibration curve.

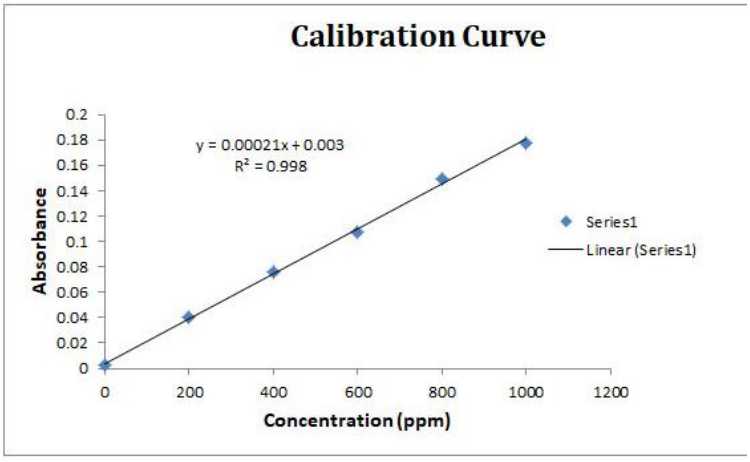

Figure 1: Mercury standard calibration curve

RESULTS

Tables 2 to 7 showed the results for the qualitative inorganic analysis of Hg in skin lightening creams, lotion, and soaps as samples obtained within the Katsina metropolis, using test reactor 2.0 N NaOH and 0.5 N KI to detect the presence of Hg and its type in the samples. Table 2 shows the qualitative inorganic analysis of Hg in skin lightening creams using 0.5 N KI. All the samples analyzed formed reddish orange precipitate which indicated the presence of the Hg (II). Table 3 shows results for qualitative inorganic analysis of Hg in skin lightening lotions. The analyzed samples all formed reddish orange precipitate which is an indication of the presence of Hg (II). Table 4 on the other hand shows the qualitative Hg inorganic analysis in skin lightening soaps. The analyzed samples revealed positive reaction of Hg (II) through the formation of reddish orange precipitate. Tables 5, 6, and 7 present the results for Hg analysis using 2.0 N NaOH reagent, the results show no colored precipitate observed in all the samples.

The concentration levels of Hg in different skin-lightening cosmetic samples analyzed are presented in Tables 8, 9, and 10 and Figure 2. The results reveal that all samples contained Hg with concentration range from 27.7± 0.0006 ppm to 61.5 ± 0.0007 ppm, even though no manufacturer claims its presence. Table 8 depicts the concentration levels of Hg in skin lightening creams, the highest level was detected in sample C4 with a concentration level of 37.6± 0.0004 ppm, and the lowest level was detected in sample C2 with a concentration level of 27.7± 0.0006 ppm. Concentration levels of Hg in skin lightening lotions was presented in Table 9, the highest value was detected in sample L5 with a concentration level of 48.5± 0.0003 ppm, and the lowest concentration level was detected in L1 with value of 39.8± 0.0003 ppm. The Hg concentration level in skin lightening soaps presented in Table 10, the lowest value in the samples was detected in sample S2 with a concentration level of 53.9± 0.0006 ppm and the highest level was detected in sample S5 with a concentration level of 61.5± 0.0007 ppm. From Figure 2, S5 recorded the overall high Hg concentration level (61.5± 0.0007 ppm) and C2 recorded the overall low concentration Hg level (27.7± 0.0006 ppm).

Table 2: Qualitative inorganic analysis of mercury in skin lightening creams using 0.5N KI

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	C1	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
2	C2	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
3	C3	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
4	C4	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
5	C5	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++

Key: C1= carotone, C2= bronz tone, C3= gluta white, C4= rich & pure, C5= clinic clear
++ = colored precipitate formed ppt= precipitate

Table 3: Qualitative inorganic analysis of mercury in skin lightening lotions using 0.5N KI

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	L1	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
2	L2	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
3	L3	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
4	L4	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
5	L5	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++

Key: L1= carotone, L2= bronz tone, L3= gluta white, L4= rich & pure, L5= clinic clear
++ = colored precipitate formed ppt= precipitate

Table 4: Qualitative inorganic analysis of mercury in skin lightening soaps using 0.5N KI

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	S1	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
2	S2	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
3	S3	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
4	S4	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++
5	S5	sample solution + 0.5 N KI	reddish orange ppt	reddish orange ppt	++

Key: S1= carotone, S2= bronz tone, S3= gluta white, S4= rich & pure, S5= clinic clear
++ = colored precipitate formed ppt= precipitate

Table 5: Qualitative inorganic analysis of mercury in skin lightening creams using 2.0 N NaOH

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	C1	sample solution + 2.0 N NaOH	+	+	+
2	C2	sample solution + 2.0 N NaOH	+	+	+
3	C3	sample solution + 2.0 N NaOH	+	+	+
4	C4	sample solution + 2.0 N NaOH	+	+	+
5	C5	sample solution + 2.0 N NaOH	+	+	+

Key: C1**=** carotone, C2= bronz tone, C3= gluta white, C4= rich & pure, C5= clinic clear
+ = no colored precipitate formed

Table 6: Qualitative inorganic analysis of mercury in skin lightening lotions using 2.0 N NaOH

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	L1	sample solution + 2.0 N NaOH	+	+	+
2	L2	sample solution + 2.0 N NaOH	+	+	+
3	L3	sample solution + 2.0 N NaOH	+	+	+
4	L4	sample solution + 2.0 N NaOH	+	+	+
5	L5	sample solution + 2.0 N NaOH	+	+	+

Key: L1= carotone, L2= bronz tone, L3= gluta white, L4= rich & pure, L5= clinic clear
+ = no colored precipitate formed

Table 7: Qualitative inorganic analysis of mercury in skin lightening soaps using 2.0 N NaOH

S/N	Sample Code	Treatment	Replicate Observation		Justification
			1st	2nd	
1	S1	sample solution + 2.0 N NaOH	+	+	+
2	S2	sample solution + 2.0 N NaOH	+	+	+
3	S3	sample solution + 2.0 N NaOH	+	+	+
4	S4	sample solution + 2.0 N NaOH	+	+	+
5	S5	sample solution + 2.0 N NaOH	+	+	+

Key: S1= carotone, S2= bronz tone, S3= gluta white, S4= rich & pure, S5= clinic clear
+ = no colored precipitate formed

Table 8: Mercury concentration level in skin lightening creams

S/N	**Sample Code**	**Concentration (ppm) n=3**
1	C1	28.0± 0.0009
2	C2	27.7± 0.0006
3	C3	34.0± 0.00010
4	C4	**37.6± 0.0004**
5	C5	33.0± 0.0004

Key: C1= carotone, C2= bronz tone, C3= gluta white, C4= rich & pure, C5= clinic clear

Table 9: Mercury concentration level in skin lightening lotions

S/N	**Sample Code**	**Concentration (ppm) n=3**
1	L1	39.8± 0.0003
2	L2	46.3± 0.0006
3	L3	44.5± 0.0004
4	L4	43.8± 0.0003
5	L5	**48.5± 0.0004**

Key: L1= carotone, L2= bronz tone, L3= gluta white, L4= rich & pure, L5= clinic clear

Table 10: Mercury concentration level in skin lightening soaps

S/N	Sample Code	Concentration (ppm) n=3
1	S1	54.9± 0.0002
2	S2	53.9± 0.0006
3	S3	60.2± 0.0005
4	S4	58.9± 0.0007
5	S5	**61.5± 0.0007**

Key: S1= carotone, S2= bronz tone, S3= gluta white, S4= rich & pure, S5= clinic clear

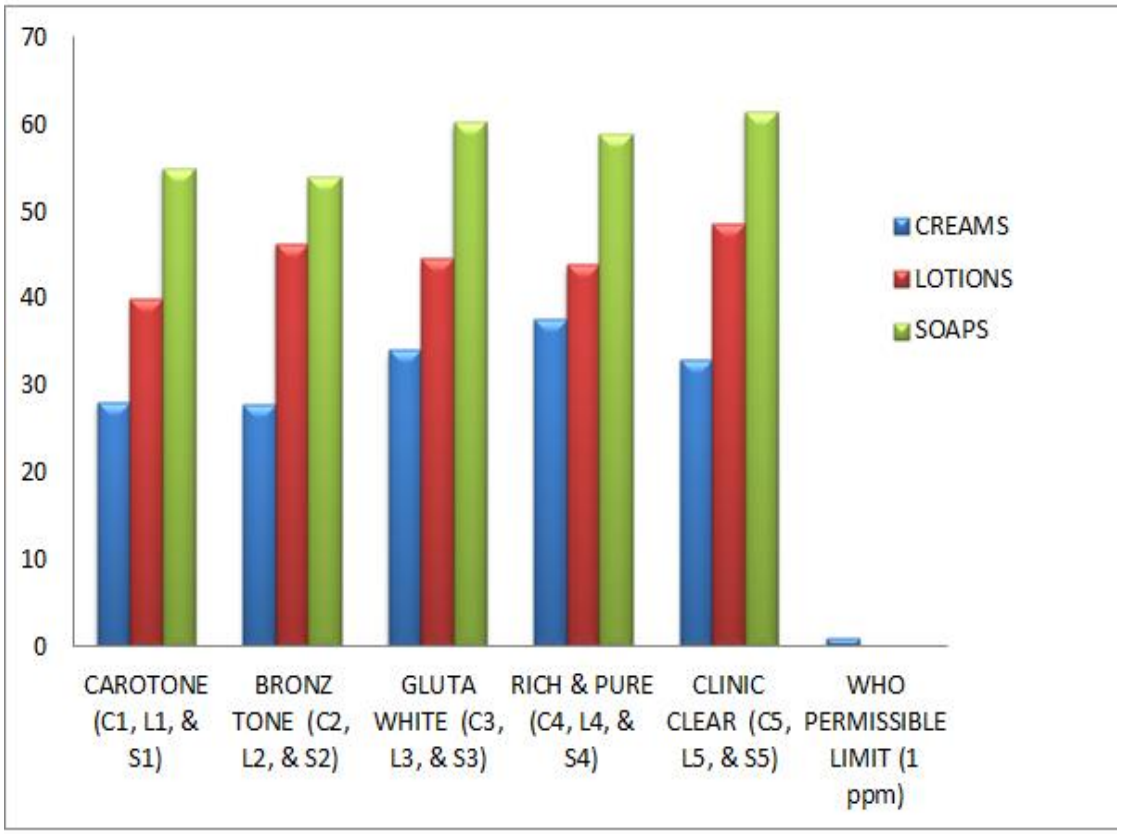

Figure 2: Mercury concentration level in skin lightening creams, lotions, and soaps

DISCUSSION

Short-term exposure to Hg in higher dosages might lead to vomiting, diarrhea, permanent damage to the brain and kidney, cancer, body weakness, and memory loss (Islam, 2018; Akiibinu et al., 2019). Many studies have reported the presence of Hg as a skin lightening agent in cosmetics products (Mallkuci, 2014; Agorku *et al.*, 2016; Islam 2018), due to it is lightening effect within short period of five (5) to seven (7) days ((Islam, 2018). The findings of this study supports the chemical reactivity of Hg as reported by Svehla (1979). The positive reaction of Hg detected in this study is comparable to that of Rohaya & Ibrahim (2017) in which all the analyzed samples show the presence of mercury using 20 % KI.

The detected Hg concentration level in this study is above that of Samia (2017) and Agorku *et al.* (2016) with Hg concentration range of 5.16 to 8.438 mg/L, and 0.098± 0.082 µg/g and 0.152±0.126 µg/g respectively in skin lightening creams and soaps. Similarly, the Hg level of this study is much lower than that of Peregrino *et al.* (2011) which reported the Hg level of 878 to 36,000 ppm in the analyzed facial creams samples. Phylicia (2019) also reported higher Hg concentration level of up to 17,547 ppm which is higher than that detected in this study. Hg

or Hg containing compounds were not clearly stated on the packaging by all the manufacturers. However, the Hg level detected in this study was higher than that of (WHO, 2019) recommended 1 ppm permissible limit.

CONCLUSION

The results of this study revealed that all the analyzed samples are qualitatively Hg positive and above the recommended permissible limit of 1 ppm by WHO (2019). The analyzed skin lightening soaps reveled to have the highest Hg concentration level, followed by skin lightening lotions then skin lightening creams. The concentration ranged from 27.7± 0.0006 to 61.5 ±0.0007 ppm. Therefore, continuous and excessive use of such products may lead to serious health hazard to the users in Katsina metropolis.

It is therefore recommended that concentration label of Hg in the skin lightening creams, lotions and soaps should be precisely monitored and controlled by the concerned authorities so as to minimize or eliminate it is presence in the products.

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