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Health risk assessment of exposure to natural radionuclides and heavy metals in indigenous eyeliners used in Benue State, Nigeria

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Abstract

This study investigates natural radionuclides (NRs) and heavy metals (HMs) in locally produced eyeliners. Untreated (market-bought) and treated (laboratory-prepared) eyeliners were analysed for NRs and HMs using a NaI(Tl) detector and an atomic absorption spectrophotometer. Potential health risks from exposure were assessed using the margin of safety (MoS), systemic exposure dose (SED), carcinogenic risk, and radiological health indices. Results revealed activity concentrations of ^{40}K, ^{238}U, and ^{232}Th as 44.87 ± 2.40, 0.98 ± 0.07, and 6.07 ± 0.34 Bq kg^{-1} in the untreated eyeliner and 43.58 ± 2.38, 1.93 ± 0.20, and 7.81 ± 0.44 Bq kg^{-1} in the treated eyeliner, respectively. The observed cancer risk from exposure to NRs was within the safe range (1×10^{-6}–1×10^{-4}) for cosmetic products; hence, no radiological health risk from exposure to the eyeliners was indicated. Furthermore, significant reductions were observed in the concentrations of Cd, Co, Cr, and Pb in the treated eyeliner, except for Ni. This was ascertained from the statistical analysis. This indicates improvement, as the impact of Ni may be less significant than that of Pb, especially in children. The observed SED values imply that the effects of HMs in the sampled eyeliners on children were more severe than on adults. To mitigate the impact of HMs, prolonged exposure should be discouraged to avoid bioaccumulation. This study provides scientific insight into the concentrations of NRs and HMs in locally produced eyeliners.

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1. Introduction

Over the years, cosmetic products have been used for decorative, therapeutic, facial ornamentation, and aesthetic purposes [1]. In recent decades, the demand for cosmetic products has significantly increased globally. In the formulation of cosmetic products, natural and synthetic ingredients are used as emollients, surfactants, preservatives, colourants, fragrances, thickeners, sunscreen agents, and antioxidants. Radioactive materials are also employed in cosmetics and healthcare products [2].

The importance of radiation to man cannot be overemphasised; however, the type of radiation, its sources, uses, and their interactions with matter in our day-to-day activities are of great concern to scientists [3]. Radionuclides, also known as radioactive isotopes,

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which include Potassium (^{40}K), Uranium (^{238}U), and Thorium (^{232}Th), are atoms with unstable nuclei that undergo radioactive decay [4]. It is an unstable nuclide and therefore degenerates by emitting ionising radiation in the form of alpha particles, beta particles, gamma rays, and other subatomic particles.

Besides the increased use of radioactive materials in cosmetics and healthcare products, which probably pose health risks, natural cosmetics extracted from plants and mineral resources (e.g. locally produced eyeliner) are, in most cases, applied directly without properly assessing the NRs and HMs in them. This suggests the need for continuous assessment of cosmetic products (e.g. eyeliner).

Eyeliner is a traditional eye-lining cosmetic used for facial ornamentation. It is mostly used by nursing mothers for their infants and children. Over the years, eyeliner has been applied to the eyes because of its beautifying and attractive effect. It is a greyish-black antimony ground into fine powder. Eyeliner is applied to the dermal surfaces of the eyelids of mostly infants and children. The name varies based on culture. It is called "Otanjele", "Toji", "Atovi", "tiro", "Uhie", and "Kwali" by the Idoma, Tiv, Ebira, Yoruba, Igbo, and Hausa, respectively. Generally, many believed that eyeliner kills germs and cleanse dirt in the eyes. It is also used to treat ophthalmologic infections and to relieve eyestrain, pain, or soreness. Thus, it helps in the cleansing of the eyes, thereby promoting visual development in children [5, 6].

It has, however, been reported that eyeliner contains a high percentage of lead (Pb) and traces of other elements like Aluminum, Carbon, Iron, and Zinc; thereby exposing infants, children and other users of this product to HMs [5]. Traditional eye cosmetic (kohl) has been reported to contain toxic metals, especially Pb. High levels of Pb were reported in the blood of the kohl user due to the high contents of Pb in kohl [7].

HMs are metallic elements that have high atomic weights and are toxic to living organisms when they accumulate in the body. They can be derived from both natural sources (rocks, soil, minerals, etc.) and human activities (industrial, agricultural, mining activities, etc.). HMs such as Mercury (Hg), Nickel (Ni), Cobalt (Co), Cadmium (Cd), Chromium (Cr), Arsenic (As), and Lead (Pb) are among the most toxic HMs.

Symptoms of exposure to HMs include headaches, nausea, fatigue, muscular weakness, and organ failure. Health problems associated with the accumulation of HMs in the body are skin irritation, cancer, organ damage, reproductive toxicity, etc.; thus, the concentration of HMs in cosmetics is of great concern to scientists worldwide [8]; hence, the need for this study.

This study hopes to update earlier reports as well as improve the quality of locally produced eyeliner commonly used in Benue State over the years. Furthermore, the activity concentration of radionuclides in eyeliner has not been considered by earlier studies. This study seeks to assess the concentrations of NRs and HMs in the eyeliner, as it could be a source of exposure to infants and children who mostly use them. This study is significant as it will create awareness of the presence, concentrations, as well as the potential health risk of NRs and HMs in the surveyed eyeliners. This will safeguard the health and safety of consumers of these products.

1.1. Theoretical framework

Health risk due to exposure to NRs and HMs are assessed employing health hazard indices outlined as follows.

1.1.1. Estimation of radiological health hazard indices

Activity concentration C, in Bq kg^{-1} measured in the sample was estimated using the expression [9]:

$$C = \frac{C_a}{\varepsilon M_s I_{\text{eff}}}. \tag{1}$$

where C_a, ε, I_{eff} and M_s are the net gamma counting rate for the peak at energy, detected efficiency of the gamma ray, intensity of the gamma line in the radionuclides and mass of the sample respectively.

Radium equivalent activity (Ra_{eq}) in Bq kg^{-1} is used to assess the health hazard effects produced from the activity concentrations of ^{238}U, ^{232}Th, and ^{40}K. It is given as [10, 11]:

$$Ra_{eq} = A_U + 1.43A_{Th} + 0.077A_K, \tag{2}$$

where A_U is activity concentrations of ^{238}U, A_{Th} is activity concentrations of ^{232}Th, and A_K is activity concentrations of ^{40}K.

Absorbed dose rate (D) expressed in $nGyh^{-1}$due to the effect of gamma radiation originating from radioactive sources in the environment is expressed in terms of the total gamma radiation absorbed dose rate in air given as [12, 13]:

$$D = 0.427A_U + 0.662A_{Th} + 0.043A_K. \tag{3}$$

Annual effective dose equivalent (AEDE) (outdoor) in $mSvy^{-1}$ is concerned with radiation exposure due to radionuclides (^{238}U, ^{232}Th, and ^{40}K) in our environment, expressed as [14]:

$$\text{AEDE}_{\text{outdoor}} = D(\text{nGy h}^{-1}) \times 24\,\text{h} \times 365.25\,\text{d} \times 0.2 \times 0.7\,\text{Sv Gy}^{-1} \times 10^{-3}. \tag{4}$$

Annual gonadal equivalent dose expressed in mSv y^{-1} is given by the expression [9]:

$$\text{AGED} = 0.31A_K + 3.09A_U + 4.18A_{Th}. \tag{5}$$

External and internal radiation hazard indices (H_{ex}) and Internal Radiation Hazard Index (H_{in}) of natural radionuclides (^{238}U, ^{232}Th and ^{40}K) are given as [10]:

$$H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}, \tag{6}$$

$$H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810}. \tag{7}$$

Activity concentration index (I_γ) expressed in Eq. (8) is concerned with radiological health hazard level due to exposure to radionuclides [10].

$$I_\gamma = \frac{A_U}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500}. \tag{8}$$

Excess lifetime cancer risk is the probability of developing cancer due to long time exposure to radionuclides. ELCR is given by Eq. (9) [9].

$$\text{ELCR} = \text{AEDE}_{\text{outdoor}}(\text{mSv y}^{-1}) \times DL \times RF(\text{Sv}^{-1}), \tag{9}$$

where DL is duration of life span (70 years) and RF is the risk factor (0.05 Sv^{-1}).

1.1.2. Health risk assessment due to exposure to heavy metals

Health risks due to exposure to metallic impurities in samples are assessed as follows:

Margin of safety (MoS) is concerned with health risk from exposure to HMs in cosmetic products. It is expressed as [15]:

$$\text{MoS} = \frac{\text{NOAEL}}{\text{SED}}, \tag{10}$$

where NOAEL is No Observed Adverse Effect Level and SED is Systemic Exposure Dosage.

No observed adverse effect level (NOAEL) The NOAEL is given by the expression [16]:

$$\text{NOAEL} = RFD \times UF \times MF, \tag{11}$$

where *MF* and *UF* are the modifying and uncertainty factors with default value of 1 and 100 respectively. *RFD* is Reference Doses (mg/kg/day) for the HMs. The *RFD* for HMs considered are: Cd (1 x 10^{-3}), Co (3 x 10^{-3}), Cr (3 x 10^{-3}), Ni (2 x 10^{-2}), and Pb (4 x 10^{-3}) [16].

Systemic exposure dose (SED) The SED ($mg/kg/d$) is concerned with the quantity of chemicals that enter into human body due to exposure. It is expressed as [16]:

$$\text{SED} = \frac{C_s \times AA \times SSA \times F \times RF \times BF}{BW} \times 10^{-3}, \tag{12}$$

where *Cs* is metal concentration in the eyeliner (ppm), AA is the daily quantity of eyeliner applied (0.3 g), SSA is the skin surface area to which the product was applied (60 cm^2), computed from the density of the eyeliner assume to be 0.005 g cm^{-2} and quantity used, F is application frequency (1.0), RF is the retention factor (1.0), BF is the bio-accessibility factor, 10^{-3} is the unit conversion factor, and BW is the average body weight: 16 kg for children and 60 kg for adult as default values [16, 17].

Hazard quotient (HQ)

The hazard quotient (HQ) relates the SED to the RfD of each metal. It is given by the expression [2]:

$$\text{HQ} = \frac{\text{SED}}{RfD}. \tag{13}$$

Carcinogenic risk

Carcinogenic risk (CR) is usually investigated for carcinogenic metals (Cd, Cr, Ni and Pb). It is computed from the *SED* and slope factor, *SF* in mg/kg/d [2].

$$\text{CR} = \text{SED} \times SF. \tag{14}$$

The SF for Cd, Cr, Ni and Pb are 6.7, 0.5, 0.91 and 0.0085 respectively [2].

2. Materials and methods

The following procedures were employed in sample preparation, collection and analysis.

2.1. Washing of all glassware and accessories

To avoid contamination of the collected samples and ensure the accuracy of results, all apparatus were thoroughly cleaned before use. Glassware was washed, rinsed and soaked in 5 % nitric acid, HNO_3 solution for 24 hours and then rinsed with deionised water.

2.2. Sample collection and preparation

Untreated eyeliners (market-bought) these are eyeliners that were purchased from the market. Eyeliners sold in the market are usually produced by boiling Antimony in water for about 1 hour to soften it. The samples were then cooled and later ground into fine powder. In this study, about 500 g of this eyeliner (sample) was purchased from North Bank Market, Makurdi, Benue State.

Treated eyeliner (laboratory-prepared) These are eyeliners prepared in the laboratory. In the laboratory preparation of the eyeliners, about 1000 g of Antimony were collected at North Bank Market, Makurdi, Benue State. The sample (Antimony) was properly ground and then sieved using a mesh of 200 mm to obtain a homogeneous fine powder. This was then used for further analysis.

2.3. Extraction and purification

The extraction and purification of the sample (laboratory-prepared) were carried out using a hydrometallurgical process modified from Ref. [18]. Exactly 50 g of the eyeliner was dissolved in 200 mL of 6 M H_2SO_4. The suspension was agitated for 2 hours; thereafter, it was filtered using Whatman No. 1 (110 mm) filter paper to obtain the filtrate containing Antimony solution. The pH of the filtrate was adjusted with 6 M NaOH to obtain Antimony precipitate. Again, the precipitate was washed several times with cold distilled water and filtered to remove dissolved impurities. The precipitate was dried at 105°C for 24 hours and further dried at 500°C for 2 hours to convert Antimony hydroxide to Antimony oxide (Eq. (15)):

$$Sb(OH)_{3(aq)} \rightarrow Sb_2O_{3(s)}. \tag{15}$$

The untreated eyeliners (market-bought) and the treated eyeliners (laboratory-prepared) were then used for heavy metal analysis using an atomic absorption spectrophotometer (AAS), radiological analysis using a NaI(Tl) detector and elemental composition of HMs in the eyeliners using XRF.

2.4. Elemental composition of untreated and treated eyeliners using XRF

About 10 g of untreated and treated eyeliners were used to assess the elemental composition of the eyeliners using XRF analysis at Ahmadu Bello University, Zaria, Nigeria.

2.5. Radiological analysis

About 320 g of both untreated and treated samples were sealed for at least 28 days in clean, uncontaminated, airtight, random impermeable plastic containers for gamma detection analysis using a NaI(Tl) detector at the National Institute of Radiation Protection and Research (NIRPR), Ibadan. The activity concentration of the radionuclide (C) measured in each sample was then determined (Eq. (1)).

2.6. Heavy metal analysis

For HM analysis, the samples were first digested using a block-digester following the standard procedures [18]. Each sample (about 10.0 g) in triplicate was put in a calibrated digestion tube of 200 mL. Concentrated HNO_3 (5 mL) was added, and the digestion tube was then carefully swirled. With a glass funnel on the neck of the tube in the rack, the tube racks were then placed in the block-digester. The temperature was gradually increased to about 150 °C for 1 hour, and concentrated perchloric acid, $HClO_4$ (7 mL), was added. This was then heated further for 1 hour to about 235 °C. The block-digester was opened, and the tube racks were removed and allowed to cool for about 1 hour to room temperature. Whatman No. 42 filter paper was used to filter it and then diluted with deionised water up to 100 mL for heavy metal analysis. Each sample solution was analysed for Cd, Co, Cr, Ni, and Pb. Buck Scientific atomic absorption spectrophotometer (MODEL 210 VGP) was used for HMs analysis at Federal University, Wukari. For each of these HMs, calibration curves were prepared with different concentrations. The working conditions used for the determination of each HM are shown in Table 4.

Table 1: Activity concentration of ^{40}K, ^{238}U, and ^{232}Th (Bq kg^{-1}) in the sampled eyeliners.

Sample	^{40}K	^{238}U	^{232}Th
Untreated	44.87 ± 2.40	0.980 ± 0.070	6.070 ± 0.34
Treated	43.58 ± 2.38	1.930 ± 0.20	7.810 ± 0.44

Table 2: Radiological health indices in the sampled eyeliners.

Sample	D nGy h^{-1}	AEDE mSv y^{-1}	Ra_{eq} Bq kg^{-1}	H_{ex} mSv y^{-1}	H_{in} mSv y^{-1}	I_γ mSv y^{-1}	AGED mSv y^{-1}	ELCR $\times 10^{-3}$
Untreated	5.990	0.007	13.115	0.035	0.038	0.097	42.488	0.022
Treated	7.426	0.009	16.453	0.044	0.049	0.120	52.293	0.027

3. Results and discussion

3.1. Activity concentration of radionuclides and radiological indices in the eyeliner

The activity concentrations of ^{40}K, ^{238}U and ^{232}Th are 44.87±2.40, 0.98±0.07 and 6.07±0.34 Bq kg^{-1} in the untreated eyeliner and 43.58±2.38, 1.93±0.20 and 7.81±0.44 Bq kg^{-1} in the treated eyeliner, respectively (Table 1). This shows that the activity concentrations of the NRs in the sampled eyeliners are relatively low. Similarly, the radiological health indices are relatively low (Table 2). Notable are the H_{ex} and H_{in}, which are below the safe limit of unity [10]. The observed effective dose is well below the reference limit of 50 mSv y^{-1} recommended for the general public [1].

Working on activity concentration and dose estimation of natural radium isotopes in 18 natural cosmetic products, [1] reported the activity concentrations for ^{226}Ra and ^{228}Ra in the range of 7.9±5.0 – 37.6±12.5 Bq kg^{-1} and 2.5±1.7 – 35.4±2.6 Bq kg^{-1}, respectively, with mean effective dose in the skin as 13.1 ± 4.9 μSvy^{-1}. These values are higher than the activity concentration of ^{238}U and the effective dose observed in this study. The ELCRs observed for the untreated and treated eyeliners are 0.022 x 10^{-3} and 0.027 x 10^{-3} respectively, which are within the safe limit of 1.0 x 10^{-6} - 1.0 x 10^{-4} for ELCR in cosmetic products [2, 19]. This depicts that exposure to these eyeliners poses no cancer risk. Generally, there is no significant difference in the activity concentration of ^{40}K, ^{238}U and ^{232}Th and the radiological health indices in the untreated and treated eyeliners. Our observation depicts little or no potential radiological health risk from exposure to the surveyed eyeliners.

3.2. Concentration of elements in the untreated and treated eyeliners using XRF

It could be observed from the XRF analysis of both untreated and treated eyeliners that the concentrations of the elements vary in the untreated and treated eyeliners (Table 3). A notable observation is that P and K were not detected in both eyeliners. Similarly, Co, Ag and Sn were not detected in the untreated eyeliner, while in the treated eyeliner, V and Ba were not detected.

The concentration of O was high in both eyeliners (19.547 % and 26.244 %). Also, concentrations of S and Zn are higher in the untreated (6.457 % and 2.096 %) as compared to the treated eyeliner (0.381 % and 0.008 %). Similarly, a major noticeable observation is the high concentration of Pb in the untreated eyeliner (64.497 %) as compared to the treated eyeliner (0.385 %). This clearly shows the high content of Pb in eyeliner as reported by earlier studies [5]. It is interesting to note that the observed concentration of Pb is lower than the 80.00 % reported earlier for local eyeliner in Nigeria by the Centers for Disease Control [5]. This could be attributed to the mineral (Antimony), ingredients, formulation and methods of preparation of the eyeliners. This suggests that the concentration of Pb in locally produced eyeliner may vary from one location or region to another.

Furthermore, twenty-seven elements were observed in the treated eyeliner against twenty-six in the untreated eyeliner. Notable is Sb, which was absent in the untreated eyeliner, but present with the highest concentration (67.779 %) in the treated eyeliner. This could be attributed to the leaching and chemical reaction of Pb in the untreated eyeliner, thereby resulting in the formation of Sb and other elements. The presence and high concentration of Sb pose little or no danger in comparison to the health risks associated with Pb, especially in children. Hence, our finding generally depicts improvement of the treated eyeliner.

3.3. Concentrations of HMs and potential health risk in the eyeliners

The working conditions used for the determination of each HM are shown in Table 4. The mean concentrations of Cd, Co, Cr, Ni, and Pb are 0.1337±0.002, 0.182±0.001, 0.003±0.002, 1.500±0.001, 4.259±0.002 ppm in the untreated eyeliner and 0.0659±0.002, 0.098±0.002, 0.002±0.001, 3.900±0.002, and 4.012±0.001 ppm in the treated eyeliner, respectively (Table 5). It is pertinent to note that the concentrations of Cd, Co, Cr and Pb were reduced by 6.78 %, 8.40 %, 0.10 % and 24.70 % respectively, in the treated eyeliner; however, that of Ni increased by 240 %. Since the impact of Ni is not as significant as Pb, especially in children, this indicates improvement of the locally produced eyeliner as compared to the high percentage of Pb in locally produced eyeliner reported by earlier studies [5, 7, 16].

Table 3: Concentration of elements in the untreated and treated eyeliners using XRF.

S/No.	Element	Untreated eyeliner (%)	Treated eyeliner (%)
1	O	19.547	26.247
2	Mg	1.157	0.639
3	Al	1.169	1.537
4	Si	0.897	0.812
5	P	0.000	0.000
6	S	6.457	0.381
7	Cl	0.772	0.281
8	K	0.000	0.000
9	Ca	0.780	0.990
10	Ti	0.029	0.000
11	V	0.063	0.000
12	Cr	0.123	0.032
13	Mn	0.066	0.031
14	Fe	1.090	0.097
15	Co	0.000	0.030
16	Ni	0.075	0.025
17	Cu	0.147	0.058
18	Zn	2.096	0.008
19	Zr	0.149	0.001
20	Nb	0.021	0.002
21	Ag	0.000	0.032
22	Sn	0.000	0.601
23	Sb	–	67.779
24	Ba	0.185	0.000
25	Ta	0.133	0.017
26	W	0.022	0.013
27	Pb	64.497	0.385
Total		99.4756	99.998

Table 4: Working conditions for HM determination using AAS.

Parameter	Cr	Ni	Cd	Pb	Co
Lamp current (mA)	20	20	10	15	20
Wavelength (nm)	357.9	232	228.9	217	240.7
Detection limit (mg L^{-1})	0.04	0.05	0.01	0.04	0.05
Slit width (nm)	0.7	0.2	0.7	0.7	0.2
Linear range (mg L^{-1})	5.00	4.00	2.00	10.00	5.00
Flame type (colour)	A–A lean/blue	A–A rich/yellow	A–A lean/blue	A–A lean/blue	A–A lean/blue

Table 5: Measured averages of concentrations of HMs (ppm) in eyeliner.

S/No.	Sample	Cd	Co	Cr	Ni	Pb
1	Untreated	0.1337 ± 0.002	0.182 ± 0.001	0.003 ± 0.002	1.500 ± 0.001	4.259 ± 0.002
2	Treated	0.0659 ± 0.002	0.098 ± 0.002	0.002 ± 0.001	3.900 ± 0.002	4.012 ± 0.001

Among the HMs considered, Pb has the highest concentration, while Cr has the lowest concentration. This is consistent with earlier studies on eyeliner [20]. The decreasing order of HMs in the treated and untreated eyeliner is Pb > Ni > Co > Cd > Cr. This is in contrast to the decreasing order of HMs (Co > Cu > Ni > Cr > Pb > Cd) in local eyeliners reported by [21].

The concentrations of HMs were compared with international permissible limits for cosmetic products, since there are no available guidelines for HMs in cosmetic products in Nigeria. New Zealand's Environmental Protection Authority (EPA) and World Health Organisation (WHO) guidelines were used in this study [21, 22]. It is seen that the concentration of Cd in the treated eyeliner was below the permissible limit of 0.2 ppm recommended by the EPA and 0.3 µg/g by WHO [23], while in the untreated eyeliner, it was above the limit (Table 3). Similarly, Co and Cr in both samples were below the 25 ppm and 0.2 ppm safe limits, respectively.

Table 6: Systemic exposure dose (SED) in the sampled eyeliners for adults and children.

Group	Sample	50% bio-accessibility factor					100% bio-accessibility factor				
		Cd	Co	Cr	Ni	Pb	Cd	Co	Cr	Ni	Pb
Adults	Untreated	2.01E-05	2.73E-05	4.50E-07	2.25E-04	6.39E-04	4.01E-05	5.46E-05	9.00E-07	4.50E-04	1.28E-03
Adults	Treated	9.89E-06	1.47E-05	3.00E-07	5.85E-04	6.02E-04	1.98E-05	2.94E-05	6.00E-07	1.17E-03	1.20E-03
Children	Untreated	7.52E-05	1.02E-04	1.69E-06	8.44E-04	2.40E-03	1.50E-04	2.05E-04	3.38E-06	1.69E-03	4.79E-03
Children	Treated	3.71E-05	5.51E-05	1.13E-06	2.19E-03	2.26E-03	7.41E-05	1.10E-04	2.25E-06	4.39E-03	4.51E-03

Table 7: Margin of safety (MoS) in the sampled eyeliners for adults and children.

Group	Sample	50% bio-accessibility factor					100% bio-accessibility factor				
		Cd	Co	Cr	Ni	Pb	Cd	Co	Cr	Ni	Pb
Adults	Untreated	4.99E+03	1.10E+03	6.67E+05	8.89E+03	6.26E+02	2.49E+03	5.49E+02	3.33E+05	4.44E+03	3.13E+02
Adults	Treated	1.01E+04	2.04E+03	1.00E+06	3.42E+03	6.65E+02	5.06E+03	1.02E+03	5.00E+05	1.71E+03	3.32E+02
Children	Untreated	1.33E+03	2.93E+02	1.78E+05	2.37E+03	1.67E+02	6.65E+02	1.47E+02	8.89E+04	1.19E+03	8.35E+01
Children	Treated	2.70E+03	5.44E+02	2.67E+05	9.12E+02	1.77E+02	1.35E+03	2.72E+02	1.33E+05	4.56E+02	8.86E+01

Table 8: Hazard quotient (HQ) in the sampled eyeliners for adults and children.

Group	Sample	50% bio-accessibility factor					100% bio-accessibility factor				
		Cd	Co	Cr	Ni	Pb	Cd	Co	Cr	Ni	Pb
Adults	Untreated	2.01E-08	9.10E-10	1.50E-11	7.50E-09	2.13E-08	1.34E-09	1.82E-09	3.00E-11	1.50E-08	3.19E-07
Adults	Treated	9.89E-09	4.90E-10	1.00E-11	1.95E-08	2.01E-08	6.59E-10	9.80E-10	2.00E-11	3.90E-08	3.01E-07
Children	Untreated	7.52E-08	1.02E-07	1.69E-09	4.22E-06	1.20E-05	7.52E-07	1.02E-06	1.69E-08	8.44E-06	2.40E-05
Children	Treated	3.71E-08	5.51E-08	1.13E-09	1.10E-05	1.13E-05	3.71E-07	5.51E-07	1.13E-08	2.19E-05	2.26E-05

Table 9: Carcinogenic risk in the sampled eyeliners for adults and children.

Group	Sample	50% bio-accessibility factor				100% bio-accessibility factor			
		Cd	Cr	Ni	Pb	Cd	Cr	Ni	Pb
Adults	Untreated	1.34E-04	2.25E-07	2.05E-04	5.43E-06	2.69E-04	4.50E-07	4.10E-04	1.09E-05
Adults	Treated	6.62E-05	1.50E-07	5.32E-04	5.12E-06	1.32E-04	3.00E-07	1.06E-03	1.02E-05
Children	Untreated	5.04E-04	8.44E-07	7.68E-04	2.04E-05	1.01E-03	1.69E-06	1.54E-03	4.07E-05
Children	Treated	2.48E-04	5.63E-07	2.00E-03	1.92E-05	4.97E-04	1.13E-06	3.99E-03	3.84E-05

Table 10: One-way ANOVA results for HM concentrations in untreated and treated eyeliner samples.

Metal	F-value	*p*-value	Significance	Interpretation
Cd	2688.00	8.28×10^{-7}	Significant	Treatment significantly reduced Cd concentration in the eyeliner samples.
Co	4234.00	3.34×10^{-7}	Significant	Treatment significantly reduced Co concentration.
Cr	0.60	0.482	Not significant	No significant difference was observed between untreated and treated samples for Cr.
Ni	3,456,000.00	5.02×10^{-13}	Significant	Treatment significantly increased Ni concentration.
Pb	36,605.00	4.48×10^{-9}	Significant	Treatment significantly reduced Pb concentration.

However, Ni and Pb in both samples were above the 0.2 ppm and 2 ppm safe limits by the EPA's guidelines. Among the HMs considered, Pb has the highest concentrations, which is consistent with earlier works on eyeliners [7, 24].

The concentrations of HMs obtained in this study were compared with those in earlier studies. The concentrations of Co (0.5 – 1.7 ppm), Ni (0.65 – 0.74 ppm), Cr (0.16 – 0.42 ppm), Pb (0.20 – 0.30 ppm), and Cd (0.02 – 0.03 ppm) were reported by [21] in the eyeliners commonly used in Adamawa. According to Ref. [25] the concentrations of Cd, Pb, Cr, Ni, Cu, Co, Fe, Mn, and Zn reported from their assessment of HMs in domestic eyeliner were 5.10 ± 3.00, 49.9 ± 102, 9.40 ± 4.10, 26.8 ± 15.9, 2.50 ± 2.9, 9.9 ± 4.2, 8750 ± 26600, 230 ± 626, and 26.4 ± 25.4 μgg^{-1} respectively. Similarly, the concentration of Cd, Cr, Ni, and Pb observed by [26] in

Table 11: Summary of treatment effects on heavy metal concentrations.

Metal	Untreated Mean (ppm)	Treated Mean (ppm)	Effect of Treatment
Cd	0.1337	0.0659	Strong reduction
Co	0.1820	0.0980	Strong reduction
Cr	0.0030	0.0020	Slight reduction
Ni	1.5000	3.9000	Strong increase
Pb	4.2590	4.0120	Moderate reduction

eyeliner used in Rivers State was in the range of 0.73±0.03 - 0.95±0.03, 0.026±0.03 - 1.78±0.08, 0.10±0.01 - 0.15±0.08, 0.22±0.08 - 0.28±0.02 mgkg^{-1} respectively. According to Ref. [16], the concentrations of Cd, Cr, Ni, and Pb in eyeliner used in Nigeria range from 0.009±0.00002 - 1.116±0.004, 0.001±0.00005 - 33.814±0.287, 0.027±0.001 - 0.352±0.015 and 2.004±0.003 - 56.815±0.038 ppm, respectively. Notable variation was observed, which generally suggests that the concentrations of HMs in eyeliners vary based on factors such as the mineral constituent and ingredients used in the formulation.

Potential health risks of exposure to HMs in the eyeliners were assessed. The SED of metals in the sampled eyeliners is presented in Table 6. The SED values are higher at 100 % (i.e. worst scenario) than at 50 % (i.e. midpoint) scenario. The SED values imply that the impact of the eyeliner on children is severe compared to adults, and this also depends on the age group of the children. This clearly shows the vulnerability of children to environmental contaminants, consistent with previous findings [23]. This suggests that the effect of the eyeliner varied based on exposure time (bio-accessibility factor) and age (adults and children).

The MoS in sampled eyeliners for adults and children at both 50% and 100% systemic levels shows varying values (Table 7). Notably, Pb has MoS values of 83.5 (untreated eyeliners) and 88.6 (treated eyeliners) at the 100 % level in children. These values are below the minimum acceptable safe limit (MoS > 100) for human skin products [27]. This depicts significant health risks and impacts on children. This may explain the high Pb levels found in the blood of children exposed to local eyeliner in Nigeria, as reported by the Centres for Disease Control [5]. According to Ref. [26], the MoS for Cd, Cr, Ni, and Pb in eyeliner used in Rivers State, Nigeria, were 5.26 x 10^{-5} ppm, 9.70 x 10^{-5} ppm, 7.69 x 10^{-5} ppm, and 7.81 x 10^{-4} ppm, respectively. These are consistent with those observed in this study. Similarly, [16] observed MoS for HMs in eyeliner used in Nigeria: Cd, Cr, Ni, and Pb. These values vary slightly from those in this study.

The HQ observed in the eyeliner is relatively low, less than the recommended value of unity (Table 8). This implies that short-term exposure to this eyeliner is safe; however, long-term exposure poses health risk. This is in agreement with the result of [26] on eyeliner used in Rivers State. Carcinogenic risk due to exposure to carcinogenic HMs (i.e. Cd, Cr, Ni and Pb) was assessed. Varying values of carcinogenic risk were observed among the HMs (Table 9). Notably is Cd in untreated (1.01×10^{-3}) for children, Ni in treated (1.06×10^{-3}) for adults, Ni in untreated (1.54×10^{-3}) and treated (3.99×10^{-3}) for children at 100 % accessibility level are above the acceptable range of $1.0 \times 10^{-6} – 1.0 \times 10^{-4}$ for carcinogenic risk in cosmetic products [2]. This indicates potential health risk of developing cancer from long-term exposure due to bioaccumulation of HMs.

3.4. Statistical analysis (one-way ANOVA) for heavy metal concentrations in the untreated and treated eyeliner samples

One-Way ANOVA analysis for heavy metals (HMs) concentrations in untreated and treated eyeliner samples and the treatment effects on the concentrations of HM are presented in Tables 10 and 11 respectively. The statistical analysis revealed that the treatment exerted varying effects on the concentrations of HMs present in the eyeliner samples. Significant differences ($p < 0.05$) were observed for Cd, Co, Ni, and Pb, indicating that the treatment process substantially altered the concentrations of these metals. However, no significant difference was observed for Cr ($F = 0.60$, $p = 0.482$), suggesting that the treatment had little influence on chromium levels.

Cadmium (Cd) exhibited a highly significant reduction following the treatment ($F = 2688.00$, $p < 0.001$), with the mean concentration decreasing from 0.1337 ppm in the untreated sample to 0.0659 ppm in the treated sample. This shows the effectiveness of the treatment in removing Cd contamination. Similar behaviour was observed for cobalt (Co), where treatment reduced the mean concentration, resulting in a highly significant difference between treated and untreated samples ($F = 4234.00$, $p < 0.001$).

Chromium (Cr) showed a decrease in mean concentration from 0.0030 ppm to 0.0020 ppm due to the treatment effects. However, the ANOVA result indicated that this reduction was not statistically significant ($p = 0.482$). This suggests that the observed difference may be attributable to random variation rather than a true treatment effect. Consequently, the treatment method appears ineffective for Cr removal under the conditions employed in this study.

Nickel (Ni) exhibited a contrasting trend. The mean concentration increased substantially from 1.500 ppm in the untreated sample to 3.900 ppm in the treated sample. The ANOVA result confirmed that this increase was highly significant ($F = 3{,}456{,}000.00$, $p < 0.001$). This finding suggests that the treatment process may have introduced additional Ni contamination or enhanced Ni mobilization within the sample matrix. Further investigation is therefore necessary to identify the source of the elevated Ni concentration.

Lead (Pb) concentrations decreased significantly following the treatment, with the mean concentration declining from 4.259 ppm to 4.012 ppm. Although the reduction was smaller than that observed for Cd and Co, the difference was statistically significant

(F = 36,605.00, $p < 0.001$), indicating that the treatment consistently reduced lead levels across the replicate samples.

Overall, the results demonstrate that the treatment was effective in reducing Cd, Co, and Pb concentrations in eyeliner samples, while having no significant effect on Cr. The significant increase in Ni concentration suggests a potential unintended consequence of the treatment process and highlights the need for optimization of the treatment procedure before its application for cosmetic safety enhancement.

3.5. Implication of the findings

The findings of this study suggest that long-term exposure to the surveyed eyeliner has bio-accumulation implications, especially on children, which can be fatal. Our findings generally depict improvement of HMs in the sampled eyeliner (treated), especially Pb. We, however, recommend further studies for more improvement, as the eyeliner still poses some health risks based on the MoS values if exposed for a long time, since it is the commonly used eyeliner in the state. To mitigate the impact of the HMs, prolonged exposure should be discouraged to avoid the accumulation of the HMs in the body.

4. Conclusion

This study investigated the presence of natural radionuclides (NRs) and heavy metals (HMs) in locally produced eyeliner used in Benue State, Nigeria. Both untreated (market-bought) and treated (laboratory-prepared) eyeliner samples were analysed. The study assessed potential health risks using indices such as margin of safety (MoS), systemic exposure dose (SED), carcinogenic risk, and radiological health indices. The results indicate low activity concentrations of NRs in both eyeliner types, posing no significant radiological health risk. The treated eyeliner showed reduced concentrations of several HMs (Cd, Co, Cr, and Pb), except Ni, suggesting improvement. Furthermore, the MoS value of Pb observed for children at the 100% systemic availability level was below the safe limit of 100. This implies a significant health risk to children, a possible explanation for the high Pb levels reported in the blood of children exposed to local eyeliner in Nigeria by the Centers for Disease Control and Prevention [5].

Data availability

The data used for this study are available on request.

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

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