

► INTERNATIONAL PERSPECTIVES



Safety Evaluation of Metal Exposure From Commonly Used Hair Dyes and Tattoo Inks in Nigeria

Abstract In the study described in this article, the concentrations of metals (cadmium, copper, chromium, aluminum, lead, nickel, zinc, cobalt, manganese, and iron) in samples of some commonly used hair dyes and tattoo inks were determined with a view to providing information on the hazards associated with the use of these products. The concentrations of metals were measured after nitric acid/perchloric acid/hydrogen peroxide digestion by atomic absorption spectrometry. Results indicated that the tattoo ink samples contained allergenic metals such as nickel, chromium, and cobalt at concentrations above the suggested limit of 1 µg/g for greater skin protection, and the toxic metals were below their respective prescribed limits, as impurities in ingredients for use for cosmetics, in the majority of the samples.

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Introduction

Tattoos and dying of hair have been practiced for several centuries worldwide. Tattoos have been used as a simple fashionable body ornament as well as to express membership of a particular sociocultural group (Forte, Petrucci, Cristaudo, & Bocca, 2009).

In permanent tattoos, the pigments are deposited into the dermis by means of a needle, which ensures that pigments are permanent. The person is directly exposed to the ingredients in the tattoo color. Tattoo colors are metal based; for instance, dichromate, cobalt, cadmium, and mercury salts are used as the base for green, blue, yellow and red colors, while iron oxide, titanium dioxide, cobalt, and manganese are prevalent in brown, white, black, and violet (Forte et al., 2009; Kaatz, Elsner, & Bauer, 2008). Organics and metals are sometimes combined to create a different hue, brightness, lightness, and shade of colors (Duke, Urioste, Dover, &

Anderson, 1998). The direct contact of the skin with the metal components of these pigments can cause metal-related skin inflammation such as allergic, eczematous, lichenoides, psudolymphomatous, or granulomatous reactions in sensitized persons.

The assessment of metal concentrations in personal care products is a public health concern since the use of these products could represent a possible source of human exposure to a variety of chemicals (Piccinini, Piecha, & Torrent, 2013). Most of these products are applied directly to human skin. While skin provides a good protective barrier, some of the ingredients in cosmetic products can penetrate the skin and reach vital internal organs via the circulatory system (Gondal, Seddigi, Nasr, & Gondal, 2010) where they can exhibit acute or chronic toxicity.

Exposure to low concentrations of lead can cause disorders such as behavioral abnormalities, decreased learning and hearing,

and permanent neurological damage and can have adverse effects on the reproductive, hepatic, and renal systems. The International Agency for Research on Cancer (IARC) has categorized cadmium as a group 2A carcinogen because it is a cell poison, which causes different types of damage including cell death or increased cell proliferation (World Health Organization, 2004).

Other metals such as nickel, chromium, iron, manganese, zinc, and aluminum are essential to humans since they are involved in many biological processes, even though some controversy still exists surrounding chromium. Despite the importance of these metals to humans and other organisms, the presence of these metals in personal products may constitute a serious health risk, e.g., allergic reactions. Nickel, chromium, and cobalt are well-known allergens, while copper, manganese, and zinc are seen as weak allergens. The objective of our study was to

TABLE 1

Hair Dyes and Tattoo Inks Sampled

Brand Name	Color	Country of Origin	Active Ingredients
Hair dyes			
Above Nature	Black	India	Nil
Above Floweriness	Red	India	Nil
Above Violet	Red	India	Nil
Cruset	Deep black	China	Cetearyl alcohol, hydrogen peroxide
Orino	Black	China	Nil
Orino	Wine	China	Nil
Orino	Gold	China	Nil
Orino	Purple	China	Nil
Orino	Coral brown	China	Nil
Orino	Chestnut brown	China	Nil
Orino	Purplish wine	China	Nil
Sabary	Wine red	India	Nil
Sabary	Chestnut brown	India	Nil
Sabary	Grape red	India	Nil
Sabary	Splendid purple	India	Nil
Sabary	Splendid red	India	Nil
Native Dye	Black	Nigeria	Nil
Tattoo inks			
Intenze	Lemon yellow	USA	Nil
Intenze	Bright red	USA	Nil
Intenze	True black	USA	Nil
J.Y.	Red	China	Nil
J.Y.	Black	China	Nil
J.Y.	Blue	China	Nil
J.Y.	Green	China	Nil

Note. Nil = not specified.

evaluate the concentrations of metals (cadmium, copper, chromium, aluminum, lead, nickel, zinc, cobalt, manganese, and iron) in hair dyes and tattoo inks with a view to providing information on the hazards associated with use of these products.

Materials and Methods

Sampling

Samples of commonly used hair dyes and tattoo inks were purchased from markets in Abraka, Warri, Asaba, Benin-City, and Lagos, Nigeria. A total of 24 brands were collected, including some manufactured in Nigeria and some imported from other countries. The choice of brands was carefully made to reflect the brands used by different income classes.

Within each type or brand, a total of three samples with different batch numbers and date of manufacture were collected in order to study the variation in elemental concentrations within a particular brand. All samples collected were within their specified usage periods (shelf life). Information about the brand names, country of origin, and active ingredients of these products is displayed in Table 1.

Reagents

All reagents, nitric acid (69%), hydrogen peroxide (30%), and perchloric acid, were ultra-pure quality. The calibration standards were prepared by diluting 1,000 mg/L commercial standards of cadmium, copper, chromium,

aluminum, lead, nickel, zinc, cobalt, manganese, and iron with 0.25 mol/L nitric acid.

Sample Preparation

A mass of 1.0 g of each sample was placed in a Teflon vessel, to which 20 mL of concentrated nitric acid, 10 mL of perchloric acid, and 5 mL of hydrogen peroxide were added and the mixture (sample + acids) was left to stand overnight. The following day, the sample was heated on a hotplate to 125°C for two hours. The sample solution was allowed to cool, and was then filtered and made up to 25 mL with 0.25 mol/L nitric acid. Four blanks were prepared in an identical manner, but omitting the samples.

Chemical Analysis

All digested samples were analyzed in triplicate for cadmium, copper, chromium, aluminum, lead, nickel, zinc, cobalt, manganese, and iron by using flame atomic absorption spectrometry. Calibration standards and blank solutions were analyzed in the same way as the samples. In each analysis at least three to four blanks were analyzed. The average blank signal was subtracted from the analytical signal of the sample before statistical analysis.

Quality Control and Statistical Analysis

All glassware used in the study was previously soaked in 10% (v/v) nitric acid solution for 24 hours and rinsed with deionized water. The instrument was calibrated after every 10th run. The accuracy of the analytical procedure was verified by using a spike recovery method. A known amount of the test element was introduced into an already analyzed sample and the sample was reanalyzed. The percentage spike recoveries for metals were between 89.0% and 98.9%. The limits of detection for the metals were 0.05, 0.01, 0.03, 0.01, 0.08, 0.01, 0.01, 0.01, 0.02, and 0.03 µg/g while the limits of quantification were 0.15, 0.03, 0.10, 0.25, 0.03, 0.03, 0.03, 0.03, 0.05, and 0.10 µg/g for cadmium, copper, chromium, aluminum, lead, nickel, zinc, cobalt, manganese, and iron, respectively. Analysis of variance (ANOVA) and Tukey's multiple comparison tests were used to determine whether the concentrations of metals varied significantly within the same brand and among the different brands of hair dyes and tattoo inks.

TABLE 2

Metal Concentrations ($\mu\text{g/g}$) in Hair Dyes and Tattoo Inks Purchased in Nigeria

Sample Codes	Cadmium	Copper	Chromium	Aluminum	Lead	Nickel	Zinc	Cobalt	Manganese	Iron
Hair dyes										
HD-1	<0.15	2.00 $\pm$ 0.16	7.50 $\pm$ 0.45	<0.25	3.00 $\pm$ 0.15	0.73 $\pm$ 0.04	32.5 $\pm$ 4.23	<0.03	22.3 $\pm$ 3.34	76.1 $\pm$ 13.7
HD-2	<0.15	7.00 $\pm$ 1.05	8.00 $\pm$ 0.64	<0.25	2.00 $\pm$ 0.24	0.95 $\pm$ 0.08	32.0 $\pm$ 4.80	<0.03	25.8 $\pm$ 2.32	105 $\pm$ 6.28
HD-3	<0.15	8.50 $\pm$ 0.85	7.75 $\pm$ 1.40	<0.25	0.50 $\pm$ 0.07	0.78 $\pm$ 0.05	30.0 $\pm$ 1.80	<0.03	24.0 $\pm$ 1.20	81.6 $\pm$ 4.08
HD-4	<0.15	4.25 $\pm$ 0.72	7.75 $\pm$ 1.24	<0.25	2.25 $\pm$ 0.38	0.78 $\pm$ 0.07	30.0 $\pm$ 2.40	<0.03	41.5 $\pm$ 3.32	188 $\pm$ 20.7
HD-5	<0.15	3.25 $\pm$ 0.39	8.25 $\pm$ 0.83	<0.25	1.25 $\pm$ 0.23	0.78 $\pm$ 0.11	34.8 $\pm$ 3.75	<0.03	26.5 $\pm$ 2.39	86.6 $\pm$ 12.1
HD-6	<0.15	1.50 $\pm$ 0.27	8.25 $\pm$ 0.99	<0.25	1.50 $\pm$ 0.17	0.20 $\pm$ 0.01	20.0 $\pm$ 1.70	<0.03	21.3 $\pm$ 1.49	76.4 $\pm$ 6.11
HD-7	<0.15	3.25 $\pm$ 0.24	8.75 $\pm$ 1.49	<0.25	<0.03	0.43 $\pm$ 0.04	26.8 $\pm$ 2.56	<0.03	23.0 $\pm$ 4.14	77.2 $\pm$ 6.95
HD-8	<0.15	2.00 $\pm$ 0.29	8.50 $\pm$ 1.11	<0.25	4.50 $\pm$ 0.72	0.08 $\pm$ 0.02	34.0 $\pm$ 3.82	<0.03	25.3 $\pm$ 4.04	331 $\pm$ 46.4
HD-9	<0.15	9.50 $\pm$ 0.57	8.25 $\pm$ 1.07	<0.25	1.25 $\pm$ 0.19	0.78 $\pm$ 0.15	32.0 $\pm$ 2.80	<0.03	27.5 $\pm$ 2.48	243 $\pm$ 26.8
HD-10	<0.15	<0.03	<0.10	<0.25	0.75 $\pm$ 0.09	<0.03	<0.03	<0.03	<0.05	<0.10
HD-11	16.8 $\pm$ 2.47	7.75 $\pm$ 0.93	9.0 $\pm$ 1.53	<0.25	2.75 $\pm$ 0.36	1.33 $\pm$ 0.17	155 $\pm$ 24.8	<0.03	<0.05	<0.10
HD-12	<0.15	4.75 $\pm$ 0.33	7.25 $\pm$ 1.09	<0.25	0.75 $\pm$ 0.04	0.75 $\pm$ 0.11	62.5 $\pm$ 4.38	0.50 $\pm$ 0.07	18.8 $\pm$ 2.44	286 $\pm$ 25.7
HD-13	<0.15	2.25 $\pm$ 0.18	8.00 $\pm$ 0.48	<0.25	0.25 $\pm$ 0.26	0.58 $\pm$ 0.06	27.8 $\pm$ 3.89	<0.03	26.0 $\pm$ 3.38	157 $\pm$ 25.1
HD-14	<0.15	2.50 $\pm$ 0.20	7.25 $\pm$ 0.58	<0.25	1.75 $\pm$ 0.21	0.73 $\pm$ 0.04	275 $\pm$ 19.3	<0.03	20.8 $\pm$ 1.87	133 $\pm$ 15.9
HD-15	<0.15	4.00 $\pm$ 0.68	7.25 $\pm$ 0.65	<0.25	3.50 $\pm$ 0.42	0.63 $\pm$ 0.08	298 $\pm$ 17.9	<0.03	26.3 $\pm$ 3.64	77.6 $\pm$ 3.88
HD-16	<0.15	20.5 $\pm$ 2.46	8.25 $\pm$ 0.73	<0.25	2.00 $\pm$ 0.26	1.05 $\pm$ 0.17	133 $\pm$ 10.6	<0.03	29.3 $\pm$ 3.22	99.2 $\pm$ 6.94
HD-17	<0.15	4.25 $\pm$ 0.55	8.50 $\pm$ 0.94	<0.25	1.25 $\pm$ 0.15	0.70 $\pm$ 0.05	37.3 $\pm$ 4.84	1.00 $\pm$ 0.04	18.5 $\pm$ 0.93	128 $\pm$ 14.1
Tattoo inks										
TT-1	<0.15	775 $\pm$ 38.8	16.5 $\pm$ 2.64	878 $\pm$ 105	0.50 $\pm$ 0.08	4.78 $\pm$ 0.76	38.5 $\pm$ 4.24	1.00 $\pm$ 0.17	23.8 $\pm$ 1.19	188 $\pm$ 22.6
TT-2	<0.15	5.50 $\pm$ 0.66	18.0 $\pm$ 3.24	42.0 $\pm$ 2.52	1.25 $\pm$ 0.23	5.28 $\pm$ 0.79	31.5 $\pm$ 4.41	0.75 $\pm$ 0.14	29.8 $\pm$ 2.38	136 $\pm$ 8.15
TT-3	<0.15	2.25 $\pm$ 0.32	14.3 $\pm$ 0.86	78.5 $\pm$ 7.07	31.8 $\pm$ 2.86	3.65 $\pm$ 0.33	138 $\pm$ 19.3	0.75 $\pm$ 0.16	17.8 $\pm$ 1.07	168 $\pm$ 10.1
TT-4	<0.15	3.00 $\pm$ 0.45	8.00 $\pm$ 0.48	<0.25	1.50 $\pm$ 0.08	0.60 $\pm$ 0.03	70.0 $\pm$ 10.5	0.50 $\pm$ 0.08	30.8 $\pm$ 2.77	69.8 $\pm$ 5.58
TT-5	<0.15	57.8 $\pm$ 6.93	9.50 $\pm$ 1.24	<0.25	34.0 $\pm$ 5.10	1.73 $\pm$ 0.42	110 $\pm$ 5.50	0.75 $\pm$ 0.09	28.5 $\pm$ 3.71	74.7 $\pm$ 11.2
TT-6	<0.15	2480 $\pm$ 173	27.0 $\pm$ 0.02	245 $\pm$ 14.7	1.75 $\pm$ 0.16	8.40 $\pm$ 0.40	85.0 $\pm$ 5.95	2.25 $\pm$ 0.12	37.3 $\pm$ 1.86	454 $\pm$ 59.1
TT-7	<0.15	1680 $\pm$ 151	14.8 $\pm$ 1.18	353 $\pm$ 28.2	0.50 $\pm$ 0.03	3.25 $\pm$ 0.46	60.0 $\pm$ 8.40	0.50 $\pm$ 0.02	32.5 $\pm$ 2.28	113 $\pm$ 12.4

Results and Discussion

The concentrations of metals measured in the selected types of hair dyes and tattoo inks are displayed in Table 2. The concentration of the metals varied significantly ($p < 0.05$) among the different types, although the concentrations of chromium in the different types of hair dyes were similar. The metallic concentrations varied from manufacturer to manufacturer and from one color to another, even among like-colored pigments. As a general trend, iron, manganese, and zinc were the dominant metals in the hair dyes, while

iron, manganese, zinc, copper, and aluminum were the dominant metals in the tattoo inks.

The concentrations of cadmium in these samples were below the limit of detection except for HD-11 (16.8 $\mu\text{g/g}$), which is higher than the 3.0 $\mu\text{g/g}$ regulatory limit for cadmium in cosmetic products (Health Canada, 2012). Human exposures to cadmium from the use of these products are very low except in the case of HD-11.

Copper was detected in all samples analyzed except in HD-10. The concentrations of copper in these products ranged from <0.03 to

20.5 and 2.25 to 2,480 $\mu\text{g/g}$ for hair dyes and tattoo inks, respectively. The tattoo inks had higher concentrations of copper than the hair dyes. In our study, HD-16 had a higher concentration of copper than the other hair dyes analyzed, while TT-6 and TT-7 had significantly higher copper levels than the other tattoo inks. The higher concentrations of copper were observed in the blue- and green-colored tattoo inks. The high concentrations of copper in TT-6 and TT-7 may be due to the use of blue pigments from materials such as copper (II) carbonate, calcium copper silicate, and copper

phthalocyanine. Copper phthalocyanine and malachite are used to produce green pigments (Forte et al., 2009). Forte and co-authors (2009) reported similar high concentrations of copper in blue- and green-colored tattoo inks (8,329 and 5,887 µg/g, respectively). The red-colored hair dye had a higher copper concentration than the other colors.

Chromium was found in all samples of the hair dyes and tattoo inks except in HD-10. The concentrations of chromium ranged from <0.10 to 9.00 µg/g and 8.00 to 27.0 µg/g in the hair dyes and tattoo inks, respectively. On average tattoo inks had higher concentrations of chromium than the hair dyes. The highest chromium concentrations were observed in the green-colored tattoo inks, which may be due to the use of dichromate pigments in the manufacture of these products. The concentrations of chromium found in this study were lower than 0.315 to 147.0 µg/g reported for tattoo inks in Italy (Forte et al., 2009).

Nickel concentrations ranging from <0.03 to 1.33 and 0.60 to 8.40 µg/g were found in hair dyes and tattoo inks, respectively. The tattoo inks had elevated levels of nickel when compared with the hair dyes. TT-6 had a higher nickel concentration than the other tattoo inks. The concentrations of nickel found in our samples were higher than those reported in tattoo inks in Italy (Forte et al., 2009), but were lower than nickel concentrations of henna in the Palestinian market (Al-Qutob, Alatrash, & Abol-Ola, 2013).

The concentrations of cobalt in the majority of the hair dye samples were below the limit of detection except in HD-12 and HD-17. The cobalt concentrations in the tattoo inks ranged between 0.50 and 2.25 µg/g. The highest concentration of cobalt was found in a blue tattoo ink, which is due to the presence of cobalt aluminum oxides. Cobalt-based tattoo inks are more toxic than copper-based inks and are linked with urticarial, granulomatous, and pseudolymphomatous reactions (Bagnato et al., 1999; Bjornberg, 1961; Blumental, Okun, & Pontich, 1982; Smith, Odom, & Maibech, 1975). Nickel, chromium, and cobalt are metals of allergological relevance and the levels of nickel, chromium, and cobalt found in our samples can be compared with published maximum levels defined as allergologically safe for consumers exposed to products containing these metals. In this context, Basketter and co-

authors (2003) have shown that the presence of the irritant or following repeated exposures to cobalt, chromium, and nickel, individuals rarely react below 10 µg/g. For this reason, Basketter and co-authors (2003) recommended that consumer products should not contain more than 5 µg/g of cobalt, chromium, or nickel as “good manufacturing practice” while the “target” value to minimize the risk of sensitization in particularly sensitive subjects should be as low as 1 µg/g.

The results of our study indicated that hair dyes and tattoo ink samples contained chromium at concentrations above safe limits while cobalt and nickel were within the safe limit, except for nickel in TT-2 and TT-6. The concentrations of nickel and cobalt found in a henna tattoo mixture ranged from less than 2.5 to 3.96 µg/g and 2.96 to 3.54 µg/g, respectively, while lead and copper were less than their respective detection limits (Kang & Lee, 2006).

Aluminum was found to be less than 0.25 µg/g in all the samples of hair dyes examined, whereas aluminum concentrations of 42 to 878 µg/g were found in five of the tattoo inks examined. The highest aluminum concentration was observed in a yellow-colored tattoo ink. Our samples contained lower concentrations of aluminum than those reported in tattoo ink in Italy (Forte et al., 2009). Al-Qutob and co-authors (2003) reported an aluminum concentration of 142.1 µg/g in henna in the Palestinian market. Aluminum exposure, apart from causing cholinotoxicity, can induce changes in other neurotransmitter levels since neurotransmitter levels are closely related (Al-Ashban, Aslam, & Shan, 2004).

The concentrations of lead in the hair dyes and tattoo inks ranged from <0.03 to 3.50 µg/g and 0.50 to 34.0 µg/g, respectively. Tattoo ink samples TT-3 and TT-5 (black colored) had much higher concentrations of lead than other samples investigated. The United States Food and Drug Administration (FDA) maximum limit for lead in color additives in cosmetics for external use formulated following good manufacturing practice is 20 µg/g, while a 10 µg/g limit for lead in cosmetics was specified by the Canadian Health Authority (Health Canada, 2012). In our study, only TT-3 and TT-5 had lead concentrations above the FDA and Canadian Health specified limits for lead in cosmetics. Apart from these two samples (TT-3 and TT-5), all other samples analyzed had lead con-

centrations in the range reported by Forte and co-authors (2009). Al-Qutob and co-authors (2013) found lead concentrations of 5.35 µg/g in henna from a Palestinian market.

Zinc concentrations of <0.03 to 298 µg/g and 31.5 to 138 µg/g were detected in the hair dyes and tattoo inks, respectively. Higher concentrations of zinc were observed in HD-11, HD-14, HD-15, HD-16, TT-3, and TT-5 than in other samples investigated. Zinc used in anti-dandruff shampoos has been shown to cause allergenic contact dermatitis (Salvador, Pascual-Marti, Arago, Chisvert, & March, 2000) and high levels of exposure to zinc can cause brittle hair and nails, neurological abnormalities, and gastrointestinal disorders and convulsions (Ayenimo, Yusuf, Adekunle, & Makinde, 2010).

The concentrations of manganese in these samples were in the range of <0.05 to 41.5 µg/g and 17.8 to 37.3 µg/g for hair dyes and tattoo inks, respectively. Higher manganese concentrations were found in blue-, green-, and red-colored tattoo inks than in the other colors. The concentrations of manganese observed in our samples were higher than levels reported for tattoo inks in Italy (Forte et al., 2009). Skin diseases associated with manganese seem to be quite rare. Manganese has been reported as a possible cause of swelling and itching, scaling, and cutaneous granuloma in the purple region of a tattoo (Nguyen & Allen, 1979; Schwartz, Mathias, Miller, Rojas-Corona, & Lambert, 1987).

Iron was not detected in two samples of the hair dyes (HD-10 and HD-11), while iron concentrations of 76.1 to 331 µg/g were detected in the other samples of hair dyes. The highest concentration of iron was found in a purple hair dye. Also, the coral brown and wine red hair dyes had significantly higher concentrations of iron than the other colors investigated. Iron concentrations of 69.8 to 454 µg/g were detected in the samples of tattoo inks. High concentrations of iron in a blue tattoo ink may be due to the presence of ferro-ferricyanide (Prussian blue). The lemon yellow, bright red, and black tattoo inks contained high amounts of iron. Iron in a black tattoo ink is associated with the use of magnetite and wüstite (iron oxides) as components of the ink. The concentrations of iron in our samples were higher than concentrations of iron found in tattoo inks in Italy (Forte et al., 2009).

Conclusion

The results of our study revealed that iron, manganese, and zinc are the main components of hair dyes while iron, manganese, zinc, copper, and aluminum are the main components of tattoo inks. Our study indicated that the tattoo ink samples contained allergenic metals such as nickel, chromium, and cobalt at concentrations above the suggested limit of 1 µg/g for greater skin protec-

tion in most samples. The toxic metals (cadmium and lead) were below their respective specified limits in cosmetic products in the majority of these samples. The results of our study demonstrate the need for the establishment of regulatory guidelines for metals in these kinds of cosmetic products. 🐼

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