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# Cobalt and lead concentrations in cosmetic products sold at local markets in Saudi Arabia

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#### ABSTRACT

Cosmetics are products that can be used on the human body for cleaning, beautification or enhancing perceived attractiveness. Cosmetics may contain a variety of heavy metals such as cobalt (Co) and lead (Pb), which at high concentrations may pose adverse effects on human health.

This study focuses to measure the concentration of heavy metals (Co and Pb) in some cosmetic samples of four types (foundation, skin lighteners, kajal (kohl) and lawsone (henna)) available at local markets in Saudi Arabia.

The total number of all cosmetic samples understudy was 41. The samples were analysed using atomic absorption spectroscopy to measure the content of Co and Pb. Quality control of the data was performed using Standard Reference Materials [IAEA-V-10] hay powder.

For all cosmetic types and qualities combined, the Co concentration range was determined to be 21.14  $\pm$  3.70–144.91  $\pm$  2.27 µg/g and the Pb range 0.75  $\pm$  0.00–10.60  $\pm$  1.24 µg/g.

The Co concentration in all cosmetic types understudy was higher than the recommended level. Pb concentration was within the range recommended by the United States Food and Drug Administration in all types of cosmetics except for kohl, for which 22 % of the samples contained concentrations higher than the permissible limit.

The findings of this study call for immediate and ongoing testing to monitor the concentrations of toxic metals in cosmetic products used in Saudi Arabia to ensure that established limits are respected and thereby protect consumer health.

## 1. Introduction

Cosmetics can be defined as any product that can be used on any part of the human body for cleaning, beautification, or enhancing perceived attractiveness, and includes all products applied for use as cosmetics [1, 2]. Depending on their ingredients, cosmetics can be either herbal or synthetic; either can serve the intended purpose. Generally speaking, cosmetics are applied to improve the appearance of skin, hair, nails, teeth or to modify one's appearance [3].

"Cosmetic products" are applied to external body parts with the intention of moisturising, perfuming, improving or changing one's appearance, protecting the body or maintaining it in good condition [4]. Such products are often incorporated as part of routine body care. Lipsticks, lip glosses, eyeliners, eye pencils, eye shadows, mascara, blushers, foundations, powders and facial cleansers are all examples of cosmetic products [5]. Cosmetic products represent a part of daily body care for beautification for many people [6,7]. As the value placed on physical appearance has increased, so too has the demand for cosmetics. Their pervasive use has led researchers to focus on the potential health impacts of such products, including side effects that might be linked to heavy metal content [34].

A variety of heavy metals with known significant toxicological properties — such as Co and Pb at high concentrations — may pose adverse effects for human health [8]. As many cosmetics contain chemical compounds which include heavy metals, this is a serious potential health issue, especially in light of the recent rapid increase of

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cosmetic use in Saudi Arabia [9,10].

Previous studies have shown variable concentrations of heavy metals in different types of cosmetics.

In Korea, Kang et al. [11] evaluated heavy metal concentrations in henna using inductively coupled plasma atomic emission spectroscopy (ICP-AES) and mercury analysis to quantify the presence of chromium (Cr), mercury (Hg) and Pb. In Pakistan, Ullah et al. [7] determined the heavy metal content of Pb, Co, Cr, cadmium (Cd), copper (Cu), iron (Fe), nickel (Ni) and zinc (Zn) in different cosmetic types using AAS. A 2011 study on the level of Pb concentration in commonly used cosmetics in South Africa also examined samples using AAS [12]. Sani et al. [4] also used AAS to assess toxic metals [manganese (Mn), Ni, Cu, Cd, Cr and Pb] in various Nigerian cosmetic products.

Kilic et al. [13], investigated the levels of some toxic metals in different homemade cosmetic products using ICP-MS. They found that the highest mean level was of Pb, also concluded that the concentration of Pb was higher than the maximum allowable level set by WHO.

Selected heavy metals were evaluated in different cosmetic brands collected from local markets in Pakistan, the study concluded that the increase in the concentrations of the heavy metals in the cosmetic products was mainly due to the type and source of raw materials used, processing techniques, storage, and mode of transportation [14].

Ashraf et al. [15] determined selected heavy metals in different types of skin-whitening creams from local markets, Pakistan. They found that all the metals detected were found in the study samples. They concluded that all samples (except one brand) were contaminated by the heavy metals under study compared with the permissible limit of metals reported by the US FDA and WHO.

Alqadami et al. [16] determined some heavy metals in skin whitening cosmetics from Saudi Arabia, using solid-phase extraction as apreconcentration method for heavy metals, before using ICP-AES for the analysis of the heavy metals.

There is currently a lack of information regarding the concentrations of heavy metals in commonly available Saudi Arabian cosmetics. The current study was thus conducted to assess the levels of Co and Pb in cosmetics such as foundation, skin lighteners, kajal (kohl) and lawsone (henna).

# 2. Materials and methods

Four different types of commonly used cosmetics, specifically foundation, skin lighteners, kohl and lawsone (henna), available in local markets in Al-Ahsa, Northern Province, Saudi Arabia were collected [nine samples of henna, nine samples of foundation cream, six samples of kajal (kohl) and six samples of skin-whitening cream]. The samples were collected from nine shops. Another four samples from high-quality brand cosmetic products were collected for each type of cosmetic (three samples of each type). The total number of all samples collected was 42 samples. The samples were classified as shown in Table 1 with abbreviations used to denote samples, brand names and number of samples. Note that the abbreviation for the brand samples is "R" for each type of cosmetic.

# 2.1. Sample preparation

# 2.1.1. Cleaning of apparatus

The glassware and plastic containers used were first washed with liquid soap, then rinsed with water, soaked in 10 % volume/volume nitric acid for 24 h, cleaned thoroughly with distilled water and then dried in an oven at 50  $^\circ$ C.

# 2.1.2. Digestion of samples and blanks

Around 1.00 g of each sample was accurately weighed using a sensitive balance (Shimadzu ATY224) and placed into a 50 mL conical flask. The standardised international protocols as pertain to the wet digestion method were applied for sample preparation. A mixture of acids was

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TOXICOLOGY REPORTS 8	(2021)	1093-1098

# Table 1

Al	obre	viations	used	to c	lenote	the	sampl	les.
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Abbreviation	Sample name	Brand name	Country of manufacture	No. of samples
А	Henna "Lawsonia inermis"	Rani Kone	SA	3
В	Henna "Lawsonia inermis"	Henna Arose Alhilal	SA	3
С	Henna "Lawsonia inermis"	Black Gold	India	3
R1	Henna "Lawsonia inermis"	Natural Henna	SA	3
D	Foundation cream	Silver Moon	PRC	3
Е	Foundation cream	Vivadana	PRC	3
F	Foundation cream	Kiss Beauty	PRC	3
R2	Foundation cream	Elf	US	3
M	Kajal (kohl)	Rani	SA	3
Ν	Kajal (kohl)	Alathmad	SA	3
R3	Kajal (kohl)	Elf	US	3
J	Skin-whitening cream	Agader	Morocco	3
K	Skin-whitening cream	Pohli	SA	3
R4	Skin-whitening cream	Fair and Lovely	India	3

added to each sample [using HNO<sub>3</sub> and HClO<sub>4</sub> (3:1)] [17] and placed on a 200–300 °C hot plate until it completely digested (the process takes about 4–5 h for henna samples and approximately 10–28 h for all other samples). Samples then remained on the hot plate to evaporate acids. Once cooled to room temperature the samples were filtered with Whatman filter paper No. 42 into a 50 mL volumetric flask. Distilled water was added to achieve the required volume. The prepared samples were transferred to polyethene containers until analysed by AAS. Blank samples were prepared as above for each set of samples. Standard Reference Material (SRM), namely [IAEA-V-10] hay powder produced by the IAEA [18] was also digested with blank and cosmetic samples.

# 2.2. Sample analysis

The sample solution was analysed for Co and Pb using a flame atomic absorption spectrophotometer (FAAS, iCE 3000 series, Thermo Scientific) with the results expressed in parts per million (ppm). Table 2 displays the FAAS working conditions and determination parameters. The readings were rounded off suitably according to the value of standard deviation from measurements in triplicate.

# 2.3. Statistical analysis

A two-sided t-test [Eqs. (1) and (2)] was applied to compare the brand and non-brand cosmetics.

# Table 2

Operating conditions for atomic absorption spectrophotometer (AAS).

Elements	Flame type	Fuel flow (L/ min)	Burner height (mm)	Wavelength (nm)	Band pass (nm)	Lamp current (mA)
Со	Air- CaHa	1.1	7.0	240.7	0.2	75%
РЪ	Air- C <sub>2</sub> H <sub>2</sub>	1.1	7.0	217	0.5	75%

The two-sided *t*-test is applied to verify if the items under comparison provide the same results by comparing their means and standard deviations. For the comparison of the means, equal variances were not assumed [19].

$$t_{cal} = \frac{\bar{x}_1 - \bar{x}_2}{\sqrt{\left(\frac{s_1^2}{n_1} + \frac{s_2^2}{n_2}\right)}}$$
(1)

where  $\overline{x}_1$  is the mean of the first method,  $\overline{x}_2$  is the mean of the second method,  $s_1^2$  and  $s_2^2$  are the respective variances and  $n_1n_2$  are the respective number of measurements.

The test is performed by calculating the tvalue from Eq. (1) and comparing it with  $t_{\alpha/2}$  the tabulated t-distribution at **the**  $\alpha$  level of significance and with degrees of freedom *df* [20], as in Eq. (2).

$$df = \frac{\left(\frac{s_1^2}{n_1} + \frac{s_2^2}{n_2}\right)^2}{\sqrt{\left(\frac{\left(\frac{s_1^2}{n_1}\right)^2}{n_1 - 1} + \frac{\left(\frac{s_2^2}{n_2}\right)^2}{n_2 - 1}\right)}}$$
(2)

# 3. Results

## 3.1. Sample results

Standard Reference Material [IAEA-V-10] hay powder produce by the IAEA [33] was used for quality control purposes.

Table 3 shows the values found, the certified values, the error and the recovery for the studied metals based on CRMs. Note that the recovery and the error are acceptable for both Co and Pb.

Table 3 also displays the results of the Co and Pb concentrations detected in the cosmetic brands under study [(Lawsonia) henna,

#### Table 3

Illustrate the concentrations of Co and Pb ( $\mu$ g/g) in different cosmetics, the standard reference materials values for the SRM hay powder and the permissible limits set by USFDA for Co and Pb.

Sample code	Co (µg/g)	Pb (µg/g)
Lawsonia (henna)		
Α	$143.05\pm6.64$	$2.21\pm0.38$
В	$138.22\pm2.18$	$\textbf{2.70} \pm \textbf{0.49}$
С	$144.91\pm2.27$	$\textbf{3.04} \pm \textbf{0.06}$
Mean	$141.71\pm4.86$	$\textbf{2.60} \pm \textbf{0.48}$
R <sub>1</sub>	$66.10\pm3.70$	$\textbf{2.28} \pm \textbf{0.28}$
Foundation cream		
D	$62.05\pm0.46$	$3.82\pm0.31$
E	$\textbf{57.19} \pm \textbf{4.45}$	$\textbf{2.27} \pm \textbf{0.63}$
F	$\textbf{58.75} \pm \textbf{7.44}$	$\textbf{2.27} \pm \textbf{0.14}$
Mean	$59.33 \pm 4.84$	$\textbf{2.79} \pm \textbf{0.86}$
R <sub>2</sub>	$66.35\pm5.41$	$\textbf{1.79} \pm \textbf{0.19}$
Kajal (kohl)		
M	$41.33\pm2.50$	$\textbf{3.83} \pm \textbf{0.66}$
N	$36.71 \pm 4.35$	$10.60 \pm 1.24$
Mean	$39.02\pm4.06$	$\textbf{7.22} \pm \textbf{3.81}$
R <sub>3</sub>	$21.14\pm3.70$	$1.769 \pm 0.52$
Skin-whitening cream		
J	$\textbf{42.97} \pm \textbf{4.05}$	$1.32\pm0.26$
K	$68.26 \pm 5.05$	$\textbf{2.18} \pm \textbf{0.33}$
Mean	$55.62 \pm 15.07$	$1.75 \pm 0.56$
Standard Reference Materials [SRM] Hay Powder		
Value found (µg/g)	0.15	1.90
Certified value (µg/g)	0.13	1.60
Recovery %	115.40	118.75
Permissible limit		
USFDA range		
Henna	10-20	5.00
Foundation	10-20	5.00
Skin-whitening cream	10-20	5.00
Kajal (kohl)	10-20	5.00

foundation cream, kajal (kohl) and skin whitening cream], which were collected from local markets in the Kingdom of Saudi Arabia. The analysed samples showed the existence of Co and Pb in all samples with varying concentrations.

Co was detected in all four types of cosmetic products with average concentrations of 141.71  $\pm$  4.86, 59.33  $\pm$  4.84, 39.02  $\pm$  4.06 and 55.62  $\pm$  15.07  $\mu$ g/g for henna, foundation cream, kohl and skin-whitening cream samples, respectively. For brand samples the Co results were found to be 66.1, 66.35, 21.14 and 114.33  $\mu$ g/g for henna, foundation cream, kohl and skin-whitening cream samples, respectively.

The Pb concentration range in the cosmetic samples was found to be 2.60  $\pm$  0.48, 2.79  $\pm$  0.86, 7.22  $\pm$  3.81 and 1.75  $\pm$  0.56 µg/g for henna, foundation cream, kohl and skin-whitening creams, respectively. For brand samples, Pb concentration was 2.28, 1.79, 1.77 and 0.75 µg/g for henna, foundation, kohl and skin-whitening creams, respectively.

# 3.2. Comparison between brand and non-brand cosmetics

A two-sided *t*-test [Eqs. (1) and (2)] was applied to compare the brand and non-brand cosmetics.

When  $t_{cal} > t_{a/2}$  it can be concluded that the difference between the means obtained by the two methods is statistically significant. If  $t_{cal} \le t_{a/2}$ , it can be concluded that there is no significant difference between the two methods. The results are tabulated in Table 4.

# 3.3. Relationship between the Co and Pb concentration ranges in the cosmetic products under study and proposed limits

Table 6 shows the relation between Co concentration in the four cosmetic types and a proposed limit. The results in Table 6 as determined for 41 cosmetic samples from four types of cosmetics show that all samples have a Co concentration of more than 5  $\mu$ g/g and that 0% of samples have a Co concentration of less than 1  $\mu$ g/g.

From Table 7 100 % of the henna and foundation samples have a Pb concentration between 1–10  $\mu$ g/g, while seven samples of kajal (kohl) (78 %) and four samples of whitening cream (80 %) have a concentration between 1–10  $\mu$ g/g. No sample from all the cosmetics understudy has a Pb concentration of more than 10  $\mu$ g/g, except for kajal, for which two samples (22 %) have a concentration of more than 10  $\mu$ g/g. Just one sample of whitening cream (20 %) has a concentration that low.

# 4. Discussion

The difference between the Co and Pb concentrations among the samples in this study is due to variants in the crude materials and assembling methods. Differences found between samples from the same producer can be attributed to fluctuations in batch-to-batch fabricating, differing work practices and contamination from exogenous sources, as highlighted by Nnorom [21] and Iwegbue et al. [22].

The level of Co in all cosmetic types was found to be higher than the acceptable limit, which as proposed by Iwegbue et al. [23] is 5  $\mu$ g/g. A comparison of Co concentrations in cosmetics from literature and the Co results of this study is found below (Table 5).

In a study of heavy metals in Iraqi henna, Al-Dahhan [24] found no Co in the samples tested, while Kang et al. [11] found a Co concentration range of 2.96–3.54  $\mu$ g/g in Korean henna dye. These results were lower than those found for henna in this study.

In Pakistan, Ullah et al. [7] investigated heavy metals in different cosmetics, examining both imports and locally manufactured products; they concluded that the concentration of Co in foundation cream was 0.5  $\mu$ g/g. In his study of heavy metals in different types of cosmetics in Nigeria, Iwegbue et al. [23] found a Co concentration of 11.1  $\pm$  3.4  $\mu$ g/g in foundation cream. These Co concentrations in the foundation were lower than that measured in this study.

#### Table 4

Comparison of the metal concentration between brand and non-brand samples in the cosmetic products under study.

	Flomonto	Non-brand sample	es	Brand samples	Brand samples		đf	t tobulated
	Elements	Mean (µg/g)	SD	Mean (µg/g)	SD	t-calculated	u	t-tabulated
Honno	Со	141.71	4.86	66.10	3.7	27.58	1.32	12.76
Heima	Pb	2.60	0.48	2.28	0.28	1.37	2.03	4.303
Providenting	Со	59.33	4.84	66.35	5.41	-1.99	0.80	12.76
Foundation	Pb	2.79	0.86	1.79	0.19	3.26	7.83	2.36
	Со	55.62	15.07	114.33	31.34	-3.07	0.62	12.76
Skin-whitening cream	Pb	1.75	0.56	0.75	0.00	4.37	5.00	2.57
Kaial (hahl)	Со	39.02	4.06	21.14	3.70	6.613	1.24	12.76
кајаг (кош)	Pb	7.22	3.81	1.769	0.52	3.44	5.31	2.57

#### Table 5

Comparison of concentrations of Co and Pb with the previous studies.

Samples	Co (µg/g)	Reference	Pb (µg/g)	Reference
			11.51	[35]
	2.96-3.54	[11]	$\begin{array}{c} 1.2 \pm 0.01 {-} 0.8 \pm \\ 0.01 \end{array}$	[13]
Henna	0.00	[24]	$\begin{array}{c} 0.16 \pm 0.01  2.63 \pm \\ 0.24 \end{array}$	[25]
			0.60-0.93	[26]
	$141.71\pm4.86$	This study	$\textbf{2.60} \pm \textbf{0.48}$	This study
	0.50	[7]	7.8-32.9 1.4	[12]
	$11.1 \pm 3.4$	[23]	26.84	[2]
	11.1 ± 0.1	[20]	2.8 - 6.4	[14]
Foundation	> 0.50	[13]	$2.79 \pm 0.86$	This
		and .		study
	$59.33 \pm 4.84$	This study		
	0.20	[7]	0.5-4.5	[22]
	< 0.02 - 2.5	[23]	0.2997-4.7287	[15]
			0.05-0.14	[4]
Skin-	0 1002 1 0031	[15]	$5.52 \pm$	
whitening	0.1992-1.9931	[13]	0.046–632.60 $\pm$	[16]
			2.250	
	$55.62 \pm 15.07$	This	$1.75 \pm 0.56$	This
		study		study
	5.3–103	[23]	12.92–76.14	[27]
			51.1-4839.5	[28]
Kohol			$2.4\pm0.01$	[13]
	$39.02\pm4.06$	This	$\textbf{7.22} \pm \textbf{3.81}$	This
		study		study

Regarding Co concentration in kajal (kohl), Iwegbue et al. [23] found Co concentrations between  $5.3-103 \mu g/g$  in eyeliner samples, which was higher than the Co concentration range in kajal samples in this study. In eye pencil samples, however, Iwegbue et al. [23] found Co

concentrations between 1.4–43.6  $\mu$ g/g, which is similar to those found in kajal in this study.

For skin-whitening cream, Ullah et al. [7] found a Co concentration of 0.2  $\mu$ g/g, while Iwegbue et al. [22] found that Co concentrations in commonly used skin-whitening creams and moisturisers in Nigeria was  $< 0.02-2.5 \mu$ g/g. Both of these results are quite low as compared to the Co concentrations found in whitening creams in this study.

Ashraf et al. [15] determined selected heavy metals in different types of skin-whitening creams from local markets, Pakistan. They found that the range of Co in their study samples was 0.1992–1.9931 ppm, which is lower than the value of Co in this result.

Kilic et al. [13] investigated the levels of some toxic metals in different homemade cosmetic products using ICP-MS. Their results showed that Co concentration in all types of cosmetics in their study is lower than the results of Co in the four types of cosmetics in this study.

The results for Pb were within the acceptable limit of 10–20 µg/g [12]. The results of Pb in different cosmetic types from previous studies are compared to those from this study as follows (Table 5). Bobaker et al. (2019) assessed the concentration of toxic heavy metals in Libyan henna, establishing a Pb concentration of 11.51 µg/g, which was higher than Pb concentration in henna in this study. Results from studies of heavy metals in henna samples with lower Pb concentrations than those found in this study include Tsanuo [25], who found a Pb concentration range of  $0.16 \pm 0.01$ – $2.63 \pm 0.24$ . Similarly (Ozbek et al (2016) [26] evaluated heavy metals in henna and other hair dyes in Turkey. His results showed a Pb concentration in henna of 0.60–0.93 µg/g. Also, Kilic et al. [13] found the range of Pb in henna was  $1.2 \pm 0.01$ – $0.8 \pm 0.01 µg/g$ .

In 2012, Brandao et al. [12] investigated the Pb content in different cosmetic products in South Africa, they found that the range of Pb in foundation was7.8–32.9  $\mu$ g/g. In 2018, Ababneh and Al-Momani [2] assessed the heavy metal content of cosmetics from Jordan, with results

# Table 6

Relationship between the concentration range of	Co in cosmetic produ	ict and the acceptable limit	: (10 µg/g) acc	ording to Iwegbue et al	. [23].
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Henna			Foundation Cream			Kajal (Kohl)			Skin-Whitening Cream		
Concentration Range ppm	No. of Samples	Percentage									
Less than 1	0	0%									
1-5	0	0%	1-5	0	0%	0.1 - 5	0	0%	0.1 - 5	0	0%
More than 5	9	100 %	More than 5	12	100 %	More than 5	9	100 %	More than 5	5	100 %

Table 7

Relationship between the concentration range of Pb in cosmetic products and the FDA limits (10  $\mu$ g/g).

Henna			Foundation Cream			Kajal (Kohl)			Skin-Whitening Cream		
Concentration Range ppm	No. of Samples	Percentage	Concentration Range ppm	No. of Samples	Percentage	Concentration Range ppm	No.of Samples	Percentage	Concentration Range ppm	No. of Samples	Percentage
Less than 1 1–10	0 9	0% 100 %	Less than 1 1–10	0 12	0% 100 %	Less than 1 1–10	0 7	0% 78 %	Less than 1 1–10	1 4	20 % 80 %
More than 10	0	0%	More than 10	0	0%	More than 10	2	22 %	More than 10	0	0%

showing a Pb concentration in foundation samples of 26.84  $\mu$ g/g. Pb concentrations in foundation samples in the current study were lower than both of these results.

Iwegbue et al. [22] found Pb concentrations in skin-whitening cream of 0.5–4.5  $\mu$ g/g, a result comparable to that found in this study. Another comparable result for Pb in skin whitening creams was found in a study done by Ashraf et al. [15], in which the range was found to be 0.2997–4.7287  $\mu$ g/g. Also in Nigeria, Sani et al. [4] found a Pb concentration range in skin-whitening creams of 0.05–0.14  $\mu$ g/g, which was less than that for Pb in whitening creams in this study.

Alqadami et al. [16]in their study found that the range of Pb in skin whitening cosmetics was  $5.52 \pm 0.046$ – $632.60 \pm 2.250 \mu g/g$ , which is quite high than that for Pb in whitening creams in this study.

For kajal (kohl), Belaid et al. [27] investigated Pb in kohl procured from Libyan markets, finding a Pb concentration range of 12.92–76.14 %. While conducting a risk assessment of heavy metals in Tunisian cosmetics, Nouioui et al. [28] reported a Pb concentration range in kohl samples of 51.1–4839.5 µg/g. Both sets of results found greater Pb concentrations in kohl than those found in the samples investigated in this study. On other hand, Kilic et al. [13] in their study of heavy metals in different cosmetic products, reported that the concentration of Pb in eyeliner was 2.4 ± 0.01 µg/g, which is lower than the result of Pb in kajal in this study.

Arshad et al. [14] found that the range of Pb concentration in different types of cosmetics was 2.8-6.4 ppm which is approximately within the range of Pb concentration in this study.

Another comparison that can be made is that between the Pb concentrations found in this study and the permissible limits in Canadian cosmetics [29]. This comparison is favourable in that it highlights the fact that the Pb concentrations in the four samples types in this study are lower than the allowable limit of element concentrations in Canadian cosmetics.

As concerns the results of the comparison between brand and nonbrand cosmetics (see Table 5), the following observations can be made. For Co, there is no significant difference between brand and nonbrand samples for all cosmetic types understudy except henna, where a significant difference is found between non-brand samples and natural henna.

For Pb, there is a significant difference between the brand and nonbrand samples, again except for henna, for which there is no significant difference between non-brand samples and natural henna samples.

There are currently no international limits of Co in cosmetic products. However, many studies have found that extended exposure to Co may cause irritations. Symptoms appear when the level of Co is  $\leq 10 \,\mu\text{g/g}$  [30,31]. In light of this information, Basketter et al. [31] proposed that the amount of Co in cosmetic products should not exceed 5  $\mu$ g/g, and an abundantly cautious limit would be 1  $\mu$ g/g [23]. This latter limit was suggested in light of the findings of Uter et al. [32], who reported that Co can cause allergenic contact dermatitis in individuals diagnosed with pre-senility at concentrations of more than 1.0  $\mu$ g/g. Table 6 shows the relation between Co concentration in the four cosmetic types and the proposed limits.

Regarding allowable concentrations of Pb in cosmetic products, in 2006 the USFDA placed the limit for Pb colour additives at  $10-20 \ \mu g/g$  [12]. Table 7 shows the relation between Pb concentration in the four cosmetic types and the USFDA limit.

# 5. Conclusions

In the present study, Co and Pb concentrations were examined in various brands of henna, foundation creams, kohl and skin-whitening creams, using inductively coupled plasma mass spectrometer (ICP-MS).

In all cosmetic types, Cobalt (Co) was found with highest concentration than the accepted permissible limits, being ranged from 21.14  $\pm$  3.70 to 144.91  $\pm$  2.27 µg/g.

The content of Co overall cosmetic products as follows: henna >

foundations > skin whitening creams > kajal.

Lead (Pb) was detected within the permissible limits for all cosmetic products except for kajal, of which two samples (22 %) exceeded the permissible limit.

The concentration of Pb was found as follows: Kajal > foundations > henna > skin whitening creams.

Continuous use of these cosmetic kinds could result in an increase of heavy metals in human body which make it exposed to several health risks.

The careful selection of raw materials free from these toxic metals that used in cosmetics manufacturing could reduce the health risk of extensive use of such products.

## CRediT authorship contribution statement

**Amel Y. Ahmed:** Conceptualization, Methodology, Supervision, Writing- Original draft preparation. **Ageela Asada**: Visualization, Investigation. **Izzedin A A Hamza**: Statistical Analysis, Writing-Reviewing and Editing.

# **Declaration of Competing Interest**

The authors declare no conflict of interest.

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# References

- R. Sahu, P. Saxena, S. Johnson, H. Mathur, H. Agarwal, Heavy Metals in Cosmetics, Centre for Science and Environment, 2014, pp. 1–28.
- [2] F.A. Ababneh, I.F. Al-Momani, Assessments of toxic heavy metals contamination in cosmetic products, Environ. Forensics 19 (2) (2018) 134–142.
- [3] N.M. Alsaffar, Determination of heavy metals in some cosmetics available in locally markets, IOSR J. Environ. Sci. Toxicol. Food Technol. 8 (2014) 09–12.
- [4] A. Sani, M.B. Gaya, F.A. Abubakar, Determination of some heavy metals in selected cosmetic products sold in kano metropolis, Nigeria, Toxicol. Rep. 3 (2016) 866–869.
- [5] O. Al-Dayel, J. Hefne, T. Al-Ajyan, Human exposure to heavy metals from cosmetics, Orient. J. Chem. 27 (1) (2011) 1–11.
- [6] V.J. Brown, Metals in Lip Products-A Cause for Concern? National Institute of Environmental Health Sciences, 2013.
- [7] H. Ullah, S. Noreen, A. Rehman, A. Waseem, S. Zubair, M. Adnan, I. Ahmad, Comparative study of heavy metals content in cosmetic products of different countries marketed in Khyber Pakhtunkhwa, Pakistan, Arab. J. Chem. 10 (1) (2017) 10–18.
- [8] S. Samia, R.M.E. Barghasha, Hatem Abdel Moniem Ahmedd, Ruba Al-Otaiqe, Banan Aba Alkhail, detection of heavy metals in cosmetic creams and antiperspirants marketed in Saudi Arabia, World J. Pharm. Med. Res. 3 (2017) 30–39.
- [9] E. Tambo, H. Almeer, Y. Alshamrani, Health and beauty cosmeceuticals industry and market expansion in Saudi Arabia, Pharmaceut. Reg. Affairs 5 (172) (2016) 2.
- [10] W. Alqulaity, A. Sheitt, D. Bakheet, M. Tallab, S. Zimmo, Skin care routine among female medical students at King Abdulaziz University Hospital (KAUH), Jeddah, Saudi Arabia, Int. J. Adv. Res. 5 (2017) 400–405.
- [11] E.K. Kang, S. Lee, J.H. Park, K.M. Joo, H.J. Jeong, I.S. Chang, Determination of hexavalent chromium in cosmetic products by ion chromatography and postcolumn derivatization, Contact Derm. 54 (5) (2006) 244–248.
- [12] J. Brandao, O. Okonkwo, M. Sehkula, R. Raseleka, Concentrations of lead in cosmetics commonly used in South Africa, Toxicol. Environ. Chem. 94 (1) (2012) 70–77.
- [13] S. Kilic, M. Kilic, M. Soylak, The determination of toxic metals in some traditional cosmetic products and health risk assessment, Biol. Trace Elem. Res. 199 (6) (2021) 2272–2277.
- [14] H. Arshad, M.Z. Mehmood, M.H. Shah, A.M. Abbasi, Evaluation of heavy metals in cosmetic products and their health risk assessment, Saudi Pharm. J. 28 (7) (2020) 779–790.
- [15] T. Ashraf, M. Taneez, S. Kalsoom, T. Irfan, M.A. Shafique, Experimental calculations of metals content in skin-whitening creams and theoretical investigation for their biological effect against tyrosinase enzyme, Biol. Trace Elem. Res. 199 (2021) 3562–3569, https://doi.org/10.1007/s12011-020-02441-z.
- [16] A.A. Alqadami, M.A. Abdalla, Z.A. AlOthman, K. Omer, Application of solid phase extraction on multiwalled carbon nanotubes of some heavy metal ions to analysis

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of skin whitening cosmetics using ICP-AES, Int. J. Environ. Res. Public Health 10 (2013) 361–374.

- [17] A.B.M.H. Uddin, R.S. Khalid, M. Alaama, A.M. Abdualkader, A. Kasmuri, S. A. Abbas, Comparative study of three digestion methods for elemental analysis in traditional medicine products using atomic absorption spectrometry, J. Anal. Sci. Technol. 7 (1) (2016) 6.
- [18] L. Pszonicki, A. Hanna, Report on Intercomparison IAEA/V-10 of the Determination of Trace Elements in Hay Powder, International Atomic Energy Agency, 1985.
- [19] Z.B. Alfassi, Z. Boger, Y. Ronen, Statistical Treatment of Analytical Data, Wiley Online Library, 2005.
- [20] P.C. Meier, R.E. Zünd, Statistical Methods in Analytical Chemistry, John Wiley & Sons, 2005.
- [21] I. Nnorom, Trace metals in cosmetic facial talcum powders marketed in Nigeria, Toxicol. Environ. Chem. 93 (6) (2011) 1135–1148.
- [22] C.M. Iwegbue, F.I. Bassey, G.O. Tesi, S.O. Onyeloni, G. Obi, B.S. Martincigh, Safety evaluation of metal exposure from commonly used moisturizing and skinlightening creams in Nigeria, Regul. Toxicol. Pharmacol. 71 (3) (2015) 484–490.
- [23] C.M. Iwegbue, F.I. Bassey, G. Obi, G.O. Tesi, B.S. Martincigh, Concentrations and exposure risks of some metals in facial cosmetics in Nigeria, Toxicol. Rep. 3 (2016) 464–472.
- [24] W.H. Al-Dahhan, Determination of heavy metals in natural hair dye selected from Iraqi plants, Noble Int. J. Sci. Res. 4 (2) (2020) 11–16.
- [25] K.M. Tsanuo, Determination of Lead, Cadmium and Nickel in Henna Leaves and Cosmetic Henna Products within Lamu County, Kenya, Kenyatta University, 2018.
- [26] N. Ozbek, S. Akman, Determination of lead, cadmium and nickel in hennas and other hair dyes sold in Turkey, Regul. Toxicol. Pharmacol. 79 (2016) 49–53.

- [27] A. Belaid, A. Eswayah, I. El-Sharaa, S. Shmaghi, Determination of lead and cadmium contents and microbial contamination assessment in Kohl used in Libyan markets, IOSR J. Pharm. Biol. Sci. 12 (2017) 34–38.
- [28] M.A. Nouioui, S. Mahjoubi, A. Ghorbel, M. Ben Haj Yahia, D. Amira, H. Ghorbel, A. Hedhili, Health risk assessment of heavy metals in traditional cosmetics sold in Tunisian local markets, Int. Sch. Res. Notices 2016 (2016).
- [29] H. Canada, Guidance on Heavy Metal Impurities in Cosmetics, Government of Canada, 2012.
- [30] D.J. Gawkrodger, Nickel dermatitis: how much nickel is safe? Contact Derm. 35 (5) (1996) 267–271.
- [31] D.A. Basketter, G. Angelini, A. Ingber, P.S. Kern, T. Menné, Nickel, chromium and cobalt in consumer products: revisiting safe levels in the new millennium, Contact Derm. 49 (1) (2003) 1–7.
- [32] W. Uter, W. Aberer, J.C. Armario-Hita, J.M. Fernandez-Vozmediano, F. Ayala, A. Balato, A. Bauer, B. Ballmer-Weber, A. Beliauskiene, A.B. Fortina, Current patch test results with the European baseline series and extensions to it from the 'European Surveillance System on Contact Allergy'network, 2007–2008, Contact Derm. 67 (1) (2012) 9–19.
- [33] L. Pszonicki, A.N. Hanna, Reference Material IAEA-V-10 Trace Elements in Hay (Powder), International Atomic Energy Agency, 2000.
- [34] R. Mayildurai, A. Ramasubbu, N. Velmani, ICP–OES investigations of heavy metal contents in cosmetic products, J. Pharm. Res. 9 (2015) 27–30.
- [35] Amin M Bobaker, Intisar Alakili, Sukiman B Sarmani, Nadhir Al-Ansari, Zaher Mundher Yaseen, Determination and Assessment of the Toxic Heavy Metal Elements Abstracted from the Traditional Plant Cosmetics and Medical Remedies: Case Study of Libya, International journal of environmental research and public health 16 (11) (2019) 1957, https://doi.org/10.3390/ijerph16111957.