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Determination of different heavy metals concentrations in cosmetics purchased from the Palestinian markets by ICP/MS

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Abstract. Cosmetic products are a possible source of heavy metal exposure to human beings. This research investigated the content of heavy metals in various types of cosmetics: lipsticks, Kohl (eyeliner), henna (hair dye or temporary tattoo), eye shadows, cream; freckles, moisturizing and foundation, and face powders. Eighteen products of different brands or misbrands cosmetics (expensive and inexpensive) that are found in the Palestinian markets were analyzed to determine the concentrations of thirteen metals (Ba, Pb, Bi, Al, Cr, Mn, Co, Ni, Cu, Zn, Mo, Ag and Cd) in these products using Inductive Couple Plasma Mass Spectrometry (ICP-MS). Results showed that two brands of cosmetics have lead content higher than 10 ppm. 50% of the samples contained nickel by more than 5 ppm while one compact powder contained 214.54 ppm. Furthermore, results showed that the concentrations of Cr, Cd and Co are found to be higher than the allowed regulation limits in 69%, 81% and 12.5% of the analyzed samples, respectively. In general, eleven of examined metals were found in 100% of samples. Two heavy metals (Ba and Cd) were found in 94, and 81% of analyzed samples, respectively. Among the different cosmetics products studied, the highest heavy metal contaminations was found in a very cheap brand of lipstick and in a cheap brand of kohl which is used inside the eyes and also in a brand of compact face powder. Additionally, results revealed that 75% of the cosmetics contained more than five parts per million of at least one of lead, cobalt, nickel, and chromium, and that 100% of the products contained more than one part per million of at least one of those metals. Our findings call for an instant mandatory regular testing program to check lead, cadmium, cobalt and other toxic heavy metals in cosmetic products imported to Palestine in order to limit their overabundance and protect consumer health.
Key Words: heavy metals, cosmetics, Palestinian markets, ICP-MS.

Resumen. Los productos cosméticos son una posible fuente de exposición a metales pesados para seres humanos. Este estudio investigó el contenido de metales pesados en diferentes tipos de cosméticos, lápices labiales, Kohl (delineador), la henna (tintura para el cabello o tatuaje temporal), sombras de ojos, crema, pecas, hidratante y fundación, y polvos para la cara. Dieciocho productos de distintas marcas o misbrands cosméticos (cara y barata) que se encuentran en el los mercados palestinos fueron analizadas para determinar las concentraciones de trece metales (Ba, Pb, Bi, Al, Cr, Mn, Co, Ni, Cu, Zn, Mo, Ag y Cd) en estos productos mediante ICP-MS. Los resultados mostraron que dos marcas de cosméticos tienen contenido de plomo superiores a 10 ppm. Un 50% de las muestras contenían níquel por más de 5 ppm mientras que un polvo compacto contiene 214.54 ppm. Por otra parte, los resultados mostraron que las concentraciones de Cr, Cd, Co se encuentran algo por encima de los límites de regulación en un 69%, 81% y 12,5% de las muestras analizadas, respectivamente. En general, once de los metales se encuentran en el 100% de las muestras. Dos metales pesados (Ba y Cd), se encontraron en el 94 y el 81% de las muestras analizadas, respectivamente. Entre los diferentes productos cosméticos estudiados, la contaminaciones de metal más alto se encuentra en una marca de lápiz labial barata y en una marca de kohl que se utiliza dentro de los ojos y también en una marca de polvo facial compacto. Además, los resultados revelan que el 75% de los cosméticos contenían más de cinco partes por millón de por lo menos plomo, cobalto, níquel y cromo, y que el 100% de los productos contenían más de una parte por millón de uno de esos metales. Nuestros resultados sugieren que un instante programa obligatorio de pruebas regulares de plomo, cadmio, cobalto y otros metales pesados tóxicos en los productos cosméticos importados a Palestina a fin de limitar su sobreabundancia y proteger la salud de los consumidores.
Palabras Clave: metales pesados, cosméticos, los mercados palestinos, ICP-MS.

Introduction. There is a growing concern about the physiological and behavioral effects of trace metals in human population. Contamination by these metals is an important environmental and health issue as they are toxic even at low concentrations (Chukwuma 1997; Vodela et al 1997; Marcovecchio et al 2007). There are 35 metals that concern us because of occupational or residential exposure; 23 of these are heavy metals: antimony, arsenic, bismuth, cadmium, cerium, chromium, cobalt, copper, gallium, gold, iron, lead, manganese, mercury, nickel, platinum, silver, tellurium, thallium, tin, uranium, vanadium, and zinc (Ferner 2001). Small amounts of these elements are common in our environment and diet and some are necessary for good health, but large amounts of any of them may cause acute or chronic toxicity. Health risks of heavy metals include reduced growth and development, cancer, organ damage, nervous system damage, and in extreme cases, death. Exposure to some metals, such as mercury and lead, may also cause development of autoimmunity, in which a person's immune system attacks its own cells. Heavy metals become toxic when they are not metabolized by the body and accumulate in the soft tissues. Heavy metals may enter the human body via food, water, air, or absorption through the skin in agriculture, industrial, or residential settings (Dupler 2001).

Many studies have been made to determine the amount of heavy metals in cosmetics all over the world. Hepp et al (2009) have examined the lead content of 20 shades of lipstick (different brands) sold in the USA. The study showed that all the lipsticks had contained detectable amounts of Pb, with values ranging from 0.09 to 3.06 μg g^{-1} and an average amount of 1.07 μg g^{-1}. The Campaign for Safe Cosmetics (CSC's) in the United States (CSC 2007) raised concern about the presence of lead in lipsticks. They found that more than half of the tested 33 brand-name red lipsticks contained detectable lead in the range of 0.03–0.65 ppm. Since the US Food and Drug Administration has not set a limit for lead in lipsticks, the CSC's results were evaluated based on the acceptable FDA limit of lead in candy assuming that lipstick can be swallowed like candy. They found that one third of the tested lipsticks had lead levels exceeded 0.1 ppm FDA's lead limit in candy (US FDA 2006). A research was done at the Canadian Environmental Defence (2011) to examine the make up used by women in Canada for arsenic, cadmium, lead, mercury, beryllium, nickel, selenium, and thallium. All of the 49 items tested contained nickel, 96% contained lead, and 90% contained beryllium. The four metals of most concern for this testing were arsenic, cadmium, lead, and mercury. Only one product was found to not contain a single metal of most concern. Some of these cosmetics contained lead 10 times and arsenic 20 times higher than the limit set out in the Health Canada Draft Guidance on Heavy Metal Impurities in Cosmetics. Adebajo (2002) has investigated heavy metals concentration in Nigerian cosmetics. They had assessed the levels of metals (arsenic, cadmium, lead, mercury and nickel) in different cosmetic products (30 creams and 20 lipsticks and lip glosses) sold at local shops in Lagos, Nigeria. All of the tested samples contained a detectable amount of all the metals of interest which ranged from 0.006 to 0.207 ppm. Omolaoye et al (2010) have analysed Chinese-made eye shadows sold in Nigeria to determine the levels of heavy metals (Pb, Cd, Ni, Cu, Zn, Cr, Co, and Mn) in them. Ni, Cu, Zn, Co and Mn were detected in all of the eye shadows, except for one, all of the colors contained Cr, 35% of the brands contained Cd at low concentrations, and 2 brands (3 colours) have lead content higher than 20 μg g^{-1}. Al-Dayel et al (2011) examined 9 brands of the most expensive brands names of Mascara and Eye Shades from the Saudi market for 28 elements of the metals using Inductive Couple Plasma Mass Spectrometry (ICP-MS) and Flow Injection Mercury System (FIMS). Lead, arsenic, cadmium, mercury and antimony level in the samples under investigation were within the normal levels. The Ni concentration reached a high level of 46.8 ppm in one of the mascara samples. While Al concentration reached 50000 ppm in two of the eye shadows. Cr concentration was high in one eye shadow sample (7000 ppm). Hardy et al (2004) examined 18 samples of eye cosmetics (kohl) obtained in Egypt (Cairo). Out of the 18 samples studied, 33% of them were found to contain lead compounds. A study from Al-Saleh et al (2009) has revealed that lead from kohl can enter the body through skin absorption in children's as well as in their parents. They found that few brands had lead content above 20 ppm that might put

consumers at the risk of lead poisoning. Children are particularly at risk for the subtle adverse effects of chronic low-dose lead exposure, as are pregnant women/fetuses (Al-Saleh et al 2009). Studies of cosmetic talcum powder have shown that these products were rarely the pure mineral talc, but rather were mixtures of various minerals (Rohl et al 1976). Kohl is composed of galena (PbO), amorphous carbon, zincite (ZnO), sassolite (H_3BO_3), minium (Pb_3O_4), magnetite (Fe_3O_4), goethite (FeO(OH)), cuprite (Cu_2O), and talc ($Mg_3Si_4O_{10}(OH)_2$) (Ali et al 1978; Hardy et al 1998, 2002; Worthing et al 1995; Healy et al 1982).

Many other studies have proven that cosmetic products are a possible source of heavy metal exposure to human beings (Al-Ashban et al 2004; Al-Hazzaa & Krahn 1995; Bruyneel et al 2002; Funtua & Oyewale 1997; Hardy et al 2002 and others). However and according to our best knowledge there are no studies conducted on the analysis of heavy metals in cosmetics products of west bank. The objectives of the current study are therefore to study the concentration of different trace metals, and to determine the concentration of different heavy metals (Li, Na, Mg, Ca, Sr, Ba,Tl, Pb, Bi, Al, K, Cr, Mn, Co, Ni, Cu, Zn, Mo, Ag and Cd) using ICP-MS in the west bank region of Palestine.

Material and Method. Eighteen cosmetics samples (lipsticks, Kohl (eyeliner), henna (hair dye or temporary tattoo), eye shadows, cream; freckles, moisturizing and foundation, and face powders) were purchased from open markets in the West Bank /Palestine. One gram of each samples were digested with HNO_3 using MARS 6 microwave digestor with the addition of HF to digest any siliceous materials. The samples were then analyzed for heavy metals content (Ba, Pb, Bi, Al, Cr, Mn, Co, Ni, Cu, Zn, Mo, Ag and Cd) by ICP/MS (Agilent technologies 7500 series).

For accurate quantitative determination of heavy metals in these samples, an internal standard method was used using (In) as internal standard and a multi-standard cali-bration method: 22 metals standard (Ag 10 ppm, Al 50 ppm, B 50 ppm, Ba 10 ppm, Bi 100 ppm, Ca 10 ppm, Cd 10 ppm, Co 10 ppm, Cr 50 ppm, Cu 10 ppm, Fe 10 ppm, K 100 ppm, Li 50 ppm, Mg 10 ppm, Mn 10 ppm, Mo 50 ppm, Na 50 ppm, Ni 50 ppm, Pb 100 ppm, Sr 10 ppm, Tl 50 ppm, Zn 10 ppm, matrix 5% HNO_3). Samples were prepared by dilution of 1.0 mL of the water samples to 10.0 mL with 0.3% ultrapure nitric acid and analyzed by ICP/MS. Each sample was analyzed three times and the results are expressed as mean ± SD (SD: standard deviation). Relative standard deviation (RSD) of the three results are calculated and found to be less than 5% for all samples for all metals analyzed in this study, reflecting the precision of the method for the analysis of these heavy metals. Calibration curves for all metals analyzed were constructed by plotting the ratio of the intensity of the analyte metal to that of the internal standard (In) vs. concentration of the trace metal (in ppb), and results showed that the calibration curves are linear with correlation coefficient (r2) greater than 0.999 for the trace metals analyzed.

The Agilent Technologies 7500 Series ICP- MS (Agilent 7500) can measure trace elements as low as one part per trillion (ppt) and quickly scan more than 70 elements to determine the composition of an unknown sample with a MassHunter Workstation software automates the analysis and accurately interprets the resulting data. The ICP/MS instrument consists of an on-board peristaltic pump that controls the flow of sample solution into and waste (drain) out of the instrument, a nebulizer (Micro Mist nebulizer) that uses a stream of argon to disperse the sample, an ICP Argon plasma torch using Argon as plasma gas, auxillary gas and nebulizer (carrier) gas, two pumps for evacuation, quadrupole mass analyzer with 0.8 amu resolution at 10% height, an octapole reaction system (ORS), and electron multiplier detector. The operating conditions are as follows: nebulizer gas (argon) flow rate: 0.9 L/min, auxiliary gas (argon) flow 0.3 L/min, plasma (Argon) gas flow: 15 L/min, reaction gas flow (helium) 4mL/min, lens voltage 7.25 V, ICP RF power: 1100 W, CeO/Ce = 1%, and Ce+2/Ce+1 = 1%.

Results and Discussion. In this study, we have tested eighteen cosmetic products for the presence of thirteen heavy metals (Ba, Pb, Bi, Al, Cr, Mn, Co, Ni, Cu, Zn, Mo, Ag and Cd) in

these products using ICP-MS. The distribution of heavy metals in samples studied is shown in Table 1. Metal elements were detected in most samples in varying concentrations.

Heavy metals were measured in four brands of lipstick samples. Results (Table 1) showed that almost 9 metals (Ba, Pb, Bi, Al, Cr, Mn, Ni, Cu, Zn) were detected. Cadmium (Cd) was detected in one sample of lipsticks with a concentration value of 0.14 ppm. Very high lead concentration was measured also in one lipstick sample which reaches 15.92 ppm. This high lead value knocks the alarm for all users of this product. Ladies and young palestinian girls buy this kind of lipstick because it is very cheap without worrying about its harmful components which unfortunately, are not registered on the wrapping. Licking lips, eat and drink while wearing lipstick can cause swallowing the lipstick's ingredients. Glamour magazine's had reported in June 2002 the "Beauty Quickie Tip" repeats a commonly quoted statistic, "Women inadvertently (but harmlessly) eat about 4 lbs. of lipstick" in a lifetime. Unfortunately, the latest science shows that no level of lead is "harmless" (Cynthia 2002). The use of lead contaminated lipstick or eye shadows by pregnant or/and lactating women could expose the fetus and infants to the risk of lead poisoning. The Centre for Disease Control and Prevention (CDC) has even gone so far as to recommend that parents should avoid using cosmetics on their children as that could be contaminated with lead (ATSDR 2007).

For the majority of the cosmetic products that were tested in this study, the lead contents was lower than 10 ppm. Only in two brands the lead contents were higher than 10 ppm and that might put consumers at the risk of lead poisoning (Al-Saleh et al 2009). The highest lead concentration was detected in lipstick (15.92 ppm) followed by kohl (10.51 ppm), beauty code compact face powder (9.38 ppm). While eye shade 7.27 and 5.35 ppm for both Henna and compact powder puff. Other products showed lowest lead content. The regulations relating to cosmetics products gave no limit values for toxic elements such as metals occurring as impurities in cosmetics products. However, it has been proposed that the levels of lead should not exceed 20 ppm, while the regulations in Canada include statements about heavy metals, and low concentrations that may be difficult to avoid and put a limit to the lead concentration that not exceed 10 ppm (CSC 2007, Canadian Environmental Defense 2011). Worth mentioning that The World Health Organization (WHO) has established 25 micrograms of lead per kilogram of body weight per week as a provisional tolerable weekly intake (PTWI) for children (WHO 2003).

Many metals were detected in different concentrations in the sample of hair dye (Henna) and also in the sample of eye shade. Remarkable concentrations for three toxic metals (Pb, Cr, Ni) were detected for Pb: 5.35 & 7.27, Cr: 13.36 & 36.73, Ni: 29.65 & 75.54 ppm for Henna and eye shade, respectively. High level of nickel in the eye shade is considered a cautionary warning of the allergic eye infections. It may cause the risk of allergic contact dermatitis for Henna (Kang & Lee 2006).

Two brands of Kohl were tested in this study, high concentration of heavy metals was remarked in one of these two samples. Cadmium concentration of 0.96 ppm in this Kohl sample is far exceeds permissible limits, where it is estimated that an exposure to more than 0.005 ppm of cadmium can be risky to human health (ATSDR 2012). However, level of exposure that is considered as a high limit is varied because the cadmium effect may increase or decrease depending on other factors of the form, type and duration of exposure (ATSDR 2012).

Also for facial products (two samples of compact powder, four samples of foundations and two samples of freckles and moisturizing creams) showed that the concentrations of chromium and cobalt exceed 5 ppm for 6 samples (75%), 2 samples (25%) respectively. Noteworthy that one of the compact powders had high concentrations of Pb, Cr, Co, Ni, and Cd with: 5.35, 10.53, 13.02, 214.54 and 0.08 ppm respectively. Also, high concentrations of cadmium (0.66 and 0.51 ppm) were found in two foundations.

For nickel, significant concentrations of nickel have been found in a number of cosmetics ranging from 127.15 ppm in lipstick to 214.54 ppm in compact powder. The presence of high concentrations of nickel in some types of facial beauty products especially powders (eyeshade and face powder) have been detected. Fifty (50) percent of the samples that were tested contained nickel by more than 5 ppm. Commonly, nickel salts are used as dye mordant and pigments. Nickel allergy caused by eye shadow has been reported by Sainio et al (2000). They had reported that even 1 ppm may trigger a pre-existing allergy.

Table 1

Concentration of trace metals detected in cosmatic samples analyzed in this study. Results expressed as average ± SD for three samples (SD: standard deviation)

Cosmetics number (type)	*Concentration of metals (ppm)*												
	Ba	*Pb*	*Bi*	*Al*	*Cr*	*Mn*	*Co*	*Ni*	*Cu*	*Zn*	*Mo*	*Ag*	*Cd*
Lipstick 1	10.37 ±4.42	15.92 ±1.61	21.30 ±4.02	397.5 ±3.99	81.60 ±4.97	48.89 ±3.99	---	---	118.6 ±2.25	345.8 ±3.27	---	---	---
Lipstick 2	1895± 0.11	1.32± 0.88	---	10.98± 1.30	7.20± 2.87	1.19± 0.49	---	4.94± 0.18	---	---	---	---	0.14± 0.07
Lipstick 3	36.04± 4.05	---	698.1± 4.05	478.9± 4.39	---	3.02± 5.95	---	---	---	---	---	---	---
Lipstick 4	---	---	32.07± 3.51	694.5± 0.71	1.30± 1.11	0.89± 0.13	---	---	---	10.62± 0.28	---	---	---
Henna	17.46± 2.21	5.35± 1.72	38.58± 3.26	142.1± 1.52	---	---	---	29.65± 3.53	95.92± 4.65	89.93± 3.43	---	---	---
Eye shade	---	---	---	62.17± 14.18	---	---	---	18.45± 12.30	18.95± 14.29	54.91± 14.16	---	---	---
Kohl 1	10.20± 0.21	----	66.15± 3.06	1009.3± 0.52	2.16 ±0.32	---	---	---	---	5.39± 3.15	---	---	---
Kohl 2	2.50± 0.151	10.51 ±0.06	---	56.75± 0.03	8.57± 0.362	1.31± 0.20	---	6.014± 0.405	2.464± 0.484	284634± 0.304	1.040± 0.026	1.780± 0.448	0.958± 0.24
Compact powder 1	103.68± 0.60	5.346 ±0.73	12.22 ±0.36	18661.5± 0.18	10.53 ±0.16	33.88 ±0.26	13.02 ±0.33	214.54± 0.25	9.690± 0.26	1.676± 0.22	---	---	---
Compact powder 2	93.75 ±1.01	9.38 ±0.10	0.37 ±0.15	8530.25± 0.18	4.45 ±0.123	18.12 ±0.19	---	3.232± 0.312	0.886± 0.295	270.122± 0.126	0.227± 0.108	---	---
Foundation cream 1	---	5.38± 3.63	---	69.85± 0.05	9.51± 0.11	5.76± 0.07	5.33± 0.10	4.92± 0.13	4.84± 0.16	25398± 0.64	1.69± 0.05	---	---
Foundation cream 2	---	---	---	33.26± 0.92	6.22± 0.84	3.00± 0.41	---	4.24± 0.79	1.05± 0.91	9.64± 0.28	0.71± 0.28	0.82± 0.79	---
Foundation cream 3		5.78± 0.83	---	902.2± 0.36	15.75± 0.28	13.54± 0.19	0.47± 0.21	10.53± 0.15	2.95± 0.11	36.99± 0.19	2.18± 0.26	---	---
Foundation cream 4	79.57± 0.66	2.12± 0.30	---	1103± 0.13	8.72± 0.04	11.04± 0.10	1.06± 0.09	7.12± 0.30	1.65± 0.23	16.69± 0.34	1.02± 0.36	---	---
Freckles cream	---	---	---	62.17± 4.18	---	---	---	18.45± 2.30	18.95± 4.29	54.91± 4.16	---	---	---
Moisturizing cream	---	---	---	15.31± 0.13	4.30± 0.12	0.70± 0.01	---	2.89± 0.04	0.84± 0.02	996.3± 0.10	0.51± 0.06	0.07± 0.05	---

For Aluminum, 50% of analyzed samples contained more than 300 ppm of Al. Significant concentrations of aluminum (18661.51 ppm) have been found in one compact powder sample and 8530.25 ppm in the second compact powder. Al concentration in fouandation cream was found to be 1103 ppm. Noteworthy, Al-Saleh et al (2009) had highlighted the adverse developmental effects of aluminum on children and infants.

Barium was detected in all samples except the freckles creams, while its high concentration (1895.85 ppm) was found in a lipstick sample. Cadmium concentrations were detected in one lipstick and one Kohl samples.

Ninety percent of the tested samples contained chromium (ranging from 1.24 to 81.6 ppm) where 70% of the samples contained chromium with a concentration more than 5 ppm.

The metals analyzed in this study are not listed as ingredients on any of the products. Due to a lack of manufacturer testing and regulatory oversight, it is possible that the companies are not even aware that the products are contaminated. These contaminants likely get into the products when poor-quality ingredients are used. Most likely the metals are contaminants from one or more of the inorganic base ingredients. Since all the metals are found in various environments, manufacturers would have to test the raw ingredients before they are gathered into the final products in order to track the origin of these contaminants.

Conclusions. This study has revealed that continuous use of these cosmetics could result in an increase in the trace metal levels in human body beyond acceptable limits. Our findings call for an instant mandatory regular testing program to check heavy metals in cosmetic products that are imported to Palestine in order to limit their overabundance and protect consumer health. Also efforts should be made at enlightening the users and the general public on the dangers involved especially for unknown misbranded products that are pumped in large quantities to the Palestinian markets.

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