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Lead exposure from the use of *Lawsonia inermis* (Henna) in temporary paint-on-tattooing and hair dying

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ABSTRACT

This study reports the evaluation of a number of spectroscopic techniques used in identifying and quantifying the presence of lead in twelve commercial and traditional henna samples. The lead levels found in henna were low with concentrations ranging from 2.29 ppm to 65.98 ppm. Henna is used as a traditional cosmetic and remedy in the Middle East, Far East, and North Africa. The very low concentrations of lead measured in these henna samples were reassuring; however, the cumulative effects of prolonged lead exposure may be of concern. Thus, the use of henna especially among children may constitute a public health risk.

1. Introduction

Henna, a traditional product with religious associations, has been widely used over the centuries for medical and cosmetic purposes in Africa, Asia, the Middle East and many other parts of the world (Al-Tufail et al., 1999). Henna is believed to give healthy and beautiful hair and to reduce body temperature in cases of high fever. Henna is also applied as temporary paint-on tattoos on the hands and feet for cosmetic purposes thus providing an alternative to permanent tattoos. Henna is a finely ground brown or green powder originating from dried leaves of the plant *Lawsonia inermis* which is grown in dry tropical and subtropical zones, including North Africa, India, Sri Lanka, and the Middle East (Chung et al., 2002). The traditional plant derived henna contains the active dye (red-orange pigment) lawsone [2-hydroxy-1,4-naphthoquinone] (Lekouch et al., 2001). Henna is usually mixed with water and made into a paste which is then applied on the hair, hands, and feet. More recently, henna preparations have been fortified with various herbs or materials in order to give it a stronger color. The added material is suspected to include various mineral products which are very rich in heavy metals such as Mercury (II) Sulfide, Lead (II) Sulfate, Lead (II) Carbonate, and Lead (II) Acetate (Lekouch et al., 2001).

The wide use of henna on the hair and hands of both children and adults (Al-Saleh and Coate, 1995) encouraged us to investigate whether henna is a possible source of lead exposure. In this study, the presence of lead in henna samples will be tested using different spectroscopic techniques. The techniques included both destructive instrumentation as scanning electron microscopy (SEM) with energy dispersive X-ray analysis (EDAX), X-ray powder diffraction (XRPD), inductively coupled plasma optical emission spectroscopy (ICP-OES) and non-destructive instrumentation as confocal Raman microscopy. SEM with EDAX microanalysis, XRPD, and

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ICP-OES require both the introduction of vacuum and energy (high excitation source) and sample preparation, thus modifying the sample to be analyzed. In contrast, Raman spectroscopy is non-destructive, may be performed at ambient conditions, and requires no sample preparation. Atomic composition of henna samples can be either quantitatively determined using ICP-OES or qualitatively/semi-quantitatively determined using SEM with EDAX microanalysis. The crystalline phases of the lead containing material believed to be added to commercial henna products can be qualitatively identified either by using XRPD or by using Raman spectroscopy where the molecular vibrational signatures obtained for a Raman spectrum are very sensitive to chemical structure and bonding.

2. Materials and methods

2.1. Samples

Twelve samples of henna were obtained from local consumer products' superstores in Sharjah and Dubai in the United Arab Emirates. Four of those were black color henna, four were green, and two were red. The remaining two henna samples (paste) were for hand use only since they were fortified with chemical products like phenylene diamine and had a pungent odor.

2.2. Analytical procedure

Henna samples (1 g) were oven-dried overnight at 400 °C. Weighed samples of 0.5 g (oven-dried henna) were reacted with 5 ml concentrated nitric acid on an electric plate for 1 h at 350 °C. After digestion, henna samples (usually 1 ml) were mixed with four volumes of distilled water (Lekouch et al., 2001). Precision for lead analysis in henna samples was calculated on a daily basis and expressed as percent relative standard deviation (RSD). Precision varied between 1 and 6%, depending on the concentration of lead found in the samples.

2.3. Instrumentation

All digested henna lead analyses were performed on a Varian Vista MPX CCD simultaneous ICP-OES spectrophotometer set at 220.353 nm. Calibration of the ICP-OES was performed using standards of known concentrations of aqueous lead solutions prepared each day. The concentrations covered a range from 0 to 100 ppm. Lead contents were expressed as ppm (micrograms of lead per gram of sample; dry weight). The henna specimens were coated with a thin film (1–2 nm) of palladium–gold for SEM (Jeol JSM-5900LV—Conventional high vacuum/high energy mode) observation and energy dispersive X-ray analysis. The henna samples were finely crushed and prepared for XRPD (Philips PW 1050, 3710 Diffractometer—Conventional high vacuum/high energy mode) analysis. In the case of confocal Raman microscopy (ambient conditions), the henna samples (used as is) were globally illuminated by a 20 mW Helium–Neon (He–Ne) laser operating at 632.5 nm with only 5 mW reaching the sample field of view. The resulting Raman spectra covered a range spanning from 150 to 3000 cm^{-1}.

3. Results and discussion

The analysis of lead in the digested henna samples using ICP-OES (lead detection limit 1.5 ppb at 220.353 nm) spectroscopy yielded concentrations ranging from 2.29 ppm to 65.98 ppm as shown in Table 1. The highest lead concentration was in the henna paste with added materials. However, the concentrations of lead in all kinds of henna samples were generally low. In 1994, the United States Food and Drug Administration set an action level (enforceable) of 0.5 μg/ml or 0.5 ppm for lead in products for use by infants and children. Using SEM with EDAX microanalysis for elemental identification, lead was only detected in henna paste samples with total lead concentrations greater than 40 ppm. The SEM image (penetration depth ≤ 2.0 μm) along with the EDAX spot analyses spectrum and the percent by weight elemental composition of the imaged area (275 μm × 275 μm) for the red henna sample (from Emirates—lead content 9.94 ppm), shown in Fig. 1 and Table 2, respectively, clearly indicated the failure of SEM (detection limit 0.1%) to identify the presence of lead. However, in the case of the black henna paste sample (from Emirates—lead content 44.01 ppm), SEM was able to confirm the presence of lead as shown in the EDAX spot analyses spectrum (Fig. 2) and the percent by weight elemental composition of the imaged area (275 μm × 275 μm) illustrated in Table 3. Since XRD can give a clear indication of the crystalline phases, it was used to test the henna samples for lead presence. XRD failed to detect any lead containing crystals (detection limit 10%); instead it was able to identify silicon dioxide (SiO_2) crystals as shown in the XRD spectra of both the green henna (from Emirates—lead content 2.32 ppm) sample (Fig. 3a) and of the red henna (from India—lead content 8.00 ppm) sample (Fig. 3b). SiO_2 (silica) is known to be a main constituent of the Horsetail extract powder derived from the plant *Equisetum hiemale*. Horsetail extract powder is usually mixed with henna powder to stimulate, heal, and soften both hair and skin. In addition, it is thought to increase the skin's defense mechanism, to regulate the skin due to the plant's rich mineral (mainly silica) content, and to strengthen connective tissue due to the presence of silicic acid (a chemical compound containing silicon, hydrogen and

Table 1 – Results for chemical analysis of henna samples

Henna sample identifier	Lead (ppm) (mean ± S.D.)	Color
Emirates 1 (paste)	65.98±0.91	Black
Emirates 2 (paste)	44.01±0.88	Black
Emirates 3	9.94±0.33	Red
India 1	8.00±0.21	Red
Pakistan	6.53±0.13	Black
India 2	4.90±0.28	Green
India 3	4.45±0.22	Black
Sudan	3.98±0.17	Green
India 4	3.73±0.08	Black
India 5	3.20±0.19	Black
Emirates 4	2.32±0.05	Green
Egypt	2.29±0.11	Green