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Bioaccessible trace metals in lip cosmetics and their health risks to female consumers☆

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ABSTRACT

Females can be exposed to toxic elements in lip cosmetics following ingestion. The bioaccessibility of Cr, Mn, Co, Ni, Cu, Cd, Sb and Pb in lip cosmetics (n = 32) were assessed via the dilute HCl extraction method, *In Vitro* Gastrointestinal protocol (IVG) and the United States Pharmacopeia Methodology (USPM), and then health risks were characterized. The total concentrations of trace metals (TMs) in lip cosmetics were in the range of 15.55–111.97 mg/kg (Mean: 60.99 mg/kg). Cu, Pb and Cr were the three major TMs and accounting for >75% of the total concentrations. Except Sb and Pb in 4/32 and 4/32 samples were higher than the US FDA (Food and Drug Administration of the United States) limits, the other TMs were lower than that limits. Only bioaccessible Pb in all samples significantly exceeded the FDA limit 0.1 mg/kg in candy. Using IVG or USPM might be preferable for evaluating the TMs exposure over HCl since they better represent gastrointestinal physiology. The estimated average daily intake (ADI) of bioaccessible ∑TMs through lip cosmetics ingestion of career women and female college students were under safety level. The long-term exposure of bioaccessible TMs by lip cosmetics using would inevitably cause non-carcinogenic health risk. This is the first report on the *in vitro* tests used for evaluating bioaccessible TMs in lip cosmetics.

1. Introduction

To moisturize lips or appear more beautiful and charming, lip cosmetics has become one of the most commonly used cosmetics among modern women around the world. However, to increase the brightness of lip cosmetics to achieve better quality and enhanced effects, some additives such as natural mineral mica or other pigments, dye, and shine are added into these products, resulting in the introduction of trace metals (TMs) into lip cosmetics, including chromium (Cr), manganese (Mn), cobalt (Co), nickel (Ni), copper (Cu), cadmium (Cd), antimony (Sb), and lead (Pb), etc. (Al-Saleh and Al-Enazi, 2011; Lemaire et al., 2013; Liu et al., 2013; Nourmoradi et al., 2013).

TMs can get released from the lip cosmetics to the digestive tract following ingestion, and significant amounts of TMs may become bioaccessible and harm various organs once they reach systemic circulation. According to the classification of the International Agency for Research on Cancer (IRAC), Cr, Cd and Ni are Group 1 human carcinogens (IARC, 2011). Chronic exposure to Pb, Cu, Ni, Cr and Cd has been associated with an increased risk of numerous diseases, including cardiovascular diseases and neurologic defects (Al-Saleh et al., 2009; Al-Saleh and Al-Enazi, 2011; Bocca et al., 2014; Hepp et al., 2014).

Bioaccessible TMs are the fraction of TMs that become soluble in the oral cavity-gastrointestinal tract, thus becoming available for absorption (Ruby et al., 1996). The bioaccessibility of TMs are usually analyzed through *in vivo* tests, i.e., animal experiments, which are expensive, time consuming, and hurt the tested animals (Basta

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and Gradwohl, 2000; Cabanero et al., 2004; Finch et al., 1978; Intawongse and Dean, 2008). Thus, for the evaluation of the bioaccessibility of TMs, *in vitro* tests might be more appropriate than *in vivo* tests owing to their cost advantage and ethical considerations. With the continuous development of *in vitro* tests, many studies have gradually started using these methods for the risk assessment of ingestion of various substances (Bruce et al., 2007; Furman et al., 2006; Guney and Zagury, 2013; Oomen et al., 2000; Rotard et al., 1992; Schroder et al., 2004; Tao et al., 2009; Versantvoort et al., 2005). Although many investigations on TM concentrations and human ingestion of lip cosmetics have been conducted in the USA and other countries (Al-Saleh et al., 2009; Al-Saleh and Al-Enazi, 2011; Brandao et al., 2012; Gardner et al., 2013; Gondal et al., 2010; Gunduz and Akman, 2013; Hepp et al., 2009; Liu et al., 2013), very few studies have examined the bioaccessibility of TMs in lip cosmetics (Gao et al., 2015; Zhao et al., 2016).

Unlike the unintentional exposure to TMs in dusts and metal jewelries, the release of TMs with the use of lip cosmetics is an active exposure among female consumers. Hence, after the bioaccumulation of TMs following long-term use of lip cosmetics, high amounts of TMs can become bioaccessible by reaching the systemic circulation and potentially damage various target organs depending on the chemical nature of the TMs, physiological and behavioral exposure parameters, and bioaccessibility of the TMs (Ruby et al., 1996).

In view of their potential for occurrence and toxicity, determination of TMs in lip cosmetics and evaluation of exposure from the ingestion of lip cosmetics are imperative to the assessment of risks and development of strategies to mitigate exposures. In the present study, eight TMs, including Cr, Mn, Co, Ni, Cu, Cd, Sb and Pb were evaluated in 32 lip cosmetic samples collected from various cosmetic stores. The bioaccessible concentrations of TMs were assessed by three *in vitro* digestion methods, namely, dilute HCl extraction method (HCl), *in vitro* gastrointestinal protocol (IVG), and the United States Pharmacopeia Methodology (USPM). The TM exposures via lip cosmetics ingestion were estimated in three female groups (American women, Chinese career women, and Chinese female college students) based on the measured concentrations established in our previous research (Gao et al., 2015; Liu et al., 2013). The present study is the first to report on the occurrence of bioaccessible TMs in lip cosmetics by *in vitro* tests and evaluate the TMs exposure among American and Chinese women users who showed different characteristics when using lip cosmetics.

2. Materials and methods

2.1. Sampling and preparation

A total of 32 lip cosmetic samples were selected and purchased through the internet (5 lip balms, 8 lip glosses and 9 lipsticks) and from shopping malls (3 lip balms, 4 lip glosses and 3 lipsticks) in Harbin, northeast China, in 2015. The purchased lip cosmetics were grouped into three categories, namely, the most commonly used brands of lip balms ($n = 8$, 1 colors in eight brands), lip glosses ($n = 12$, 3 colors in nine brands), and lipsticks ($n = 12$, 3 colors in nine brands). The colors studied for each brand of lip balm were colorless; each brand of lip gloss were elegant pink, coral pink, and elegant red; and each brand of lipstick were orange red, true red, and burning red. Detailed information including types, prices (USD) and colors of these lip products are provided in Table S1.

2.2. Exposure scenarios

The study estimated the health risks following six exposure scenarios according to the users (Chinese career women, Chinese female college students, and American women) and the frequencies of lip cosmetics usage (moderate use and high use). Among the Chinese female college students, only a small proportion of them used lip glosses and lipsticks (5%), and hence, only their use of lip balms was evaluated. The exposure scenarios examined in this study were as follows (Gao et al., 2015; Liu et al., 2013):

- Scenarios 1 and 2, Chinese career women exposed to moderate (33.42 mg/day) and high use (66.84 mg/day) of lip cosmetics.
- Scenarios 3 and 4, Chinese female college students exposed to moderate (10.32 mg/day) and high use (51.60 mg/day) of lip balms.
- Scenarios 5 and 6, American women exposed to moderate (24 mg/day) and high use (87 mg/day) of lip cosmetics.

2.3. Determination of total TM concentration

The analysis of TMs in lip cosmetics was performed according to the National Institute for Occupational Safety and Health (NIOSH) standard method (NIOSH, 2003), with slight modifications. Approximately 0.5 g of each sample was transferred into a clean, 100-mL Teflon digestion tube, and digested with 2.0 mL of concentrated nitric acid (HNO_3) in a block digester at 130 °C for 15 h. The tubes were covered with glass funnels to allow HNO_3 reflux during digestion. The residual material was diluted with deionized water to a final volume of 50 mL and the solution was filtered to remove material that did not completely dissolve.

2.4. Determination of oral bioaccessibility

The bioaccessibility of the lip cosmetic samples were tested by HCl, IVG, and USPM, and the test conditions and comparisons are shown in Table S2.

2.4.1. HCl

Approximately 100 mg of the lip cosmetic samples were leached with 5 mL of 0.07 M HCl in a water bath at 37 °C for 2 h (acid volume to sample mass ratio was 50:1). Subsequently, after 1 min of agitation, the pH was checked, and adjusted to 1.8 with concentrated HCl dropwise. (EC, 2009a).

2.4.2. IVG

Rodriguez's IVG protocol was used with slightly modified. The gastric phase solution was 0.15 M NaCl and 1% porcine pepsin (Sigma Chemical), and the pH of the gastric solution was adjusted to 1.8 following the addition of lip cosmetics. The intestinal phase solution comprised porcine bile extract (0.35%; Sigma Chemical) and porcine pancreatin (0.035%; Sigma Chemical). The bioaccessible fractions of TMs at the end of gastric and intestinal digestion were labeled as IVG-G and IVG-I, respectively. (Welfringer and Zagury, 2009).

2.4.3. USPM

The digestive fluids were simulated on the basis of human digestive fluids compositions such as α-amylase, pepsin, and bile salts. Saliva, gastric, and intestinal digestive fluids were prepared according to Table S3 and preheated to 37 °C. To prevent microbial contamination, sodium azide (200 mg/L) was added to all solutions (Wang et al., 2011). The digestion was started by incubating the lip cosmetic samples with 6 mL of saliva ($pH = 6.8 \pm 0.1$) at 37 °C and 55 rpm. Furthermore, to simulate gastric digestion, 12 mL of gastric juice ($pH = 1.3$) were added to the mixture and incubated on a rotator at 37 °C for 2 h. Lastly, 12 mL of duodenal juice ($pH = 8.1$),