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European survey on the content of lead in lip products

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ABSTRACT

223 lip articles (representing 55 brands) were purchased in 15 European Union Member States and analysed for lead content. Various lip products (lipsticks and lip glosses), shades (red, brown, purple and pink) and price ranges (3 categories) were investigated. The analytical method employed a microwave-assisted acid digestion followed by ICP-MS determination. The results revealed that 49 samples (22%) contained lead at a level higher than 1 mg/kg, representing 31% of the tested lipsticks and 4% of the lip glosses. On average, the lead content found in lipsticks (0.75 mg/kg) was nearly double that found in lip glosses (0.38 mg/kg) and this difference was judged statistically significant at 95% probability. Apart from brown, statistically significant higher levels of lead were also found when comparing the average lead contents in lipsticks and lip glosses of the same shade: pink (0.81 and 0.38 mg/kg), purple (0.88 and 0.37 mg/kg) and red (0.58 and 0.25 mg/kg).

The influence of price on lead content was studied on the two lip product types separately. In the case of lip glosses no differences were found. In the case of lipsticks, the more expensive items (price category III) contained a significantly lower quantity of lead in comparison to the cheapest articles (price category I). The lipsticks containing the highest levels of lead belonged to the price category II.

In all cases, however, the actual lead concentration measured in the finished products is far below the recommended limits for Germany (20 mg/kg) or Canada (10 mg/kg). The outcome of this work delivers information about the current situation on the European market and provides information to policy-makers about the quantities of lead in lip articles and technically achievable levels.

1. Introduction

Lead (Pb) is an environmental contaminant that occurs in organic and inorganic forms, both naturally and from diverse anthropogenic activities. The inorganic form is the most common in the environment. Water, air, soil, dust and food, are the sources of human exposure, with the latter being the predominant one. Numerous reports and research papers showed that lead affects almost every system in the body, causing life-long adverse health effects [1–4]. It is also known that skin-absorbed lead might not be detectable in the blood by the usual techniques of electrothermal Atomic Adsorption Spectroscopy (AAS) or Anodic Stripping Voltammetry (ASV) [5]. Thus, in some cases skin absorption of lead may remain undetected. The critical effects for lead risk assessment have been defined as developmental neurotoxicity in young children, cardiovascular effects, nephrotoxicity and reproductive effects in adults [6,7]. Since there is no evidence for a threshold for critical lead-induced effects, measures must be taken to limit human exposure to lead, also by controlling the content of lead in consumer products.

The safety of cosmetic and personal care products has gained attention over the past years, since they are a possible source of daily, population-wide and often long-term exposure to a variety of chemicals. Some studies were conducted on the level of lead and other heavy metals in a number of products such as lipsticks, soaps, creams, hair creams, sunscreen products, eye shadows, henna samples, kohl, eye pencils, eye liners and mascara [8–26]. However, with the exceptions of those carried out by Frontier Global Sciences Inc., on behalf of the Food and Drug Administration (FDA) [9] (400 lipsticks), Ayenimo [11] (85 samples of soaps and creams), Al-Saleh [13] (72 lipsticks and 22 eye shadows), Sainio [24] (49 eye shadows with 88 colours) and Al-Ashban [22] (107 kohl samples), not many items were analysed in each case.

European legislation on the approximation of the laws of Member States related to cosmetic products was introduced in 1976 by the EU Directive 76/768/EEC [27], known as the Cosmetic Products Directive, which has been frequently amended. The new Cosmetic Products Regulation (EC Reg. 1233/2009) [28] was adopted in November 2009 and it is foreseen to replace the Directive 76/768/EEC; however most of its provisions will be only applicable from the 11th July 2013. Lead is listed in Annex II of both Directive 76/768/EEC and EC Reg. 1233/2009, among the substances which must not form part of the composition of cosmetic products. However, as stated in article 17 of EC Reg. 1233/2009, "the

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non-intended presence of a small quantity of a prohibited substance, stemming from impurities of natural or synthetic ingredients, the manufacturing process, storage, migration from packaging, which is technically unavoidable in good manufacturing practice, shall be permitted provided that such presence is in conformity with article 3", which in turn requests the cosmetic products available on the market to be safe for human health when used under normal or reasonably foreseeable conditions of use.

A limit for lead in cosmetics has been defined neither at European nor at international level. The occurrence of lead in cosmetic samples can originate from some ingredients naturally containing lead (such as dyes and pigments) and/or from being introduced during the production process [29].

In 2010, the International Cooperation on Cosmetics Regulation (ICCR) set up a Working Group on Traces in Cosmetics. ICCR is an international group of cosmetic regulatory authorities from the United States (Food and Drug Administration), Japan (Ministry of Health, Labour and Welfare), the European Union (European Commission, Directorate General Enterprise) and Canada (Health Canada). The main goal of this group is to provide the highest level of global consumer protection, while minimising barriers to international trade. In the frame of the activities of the Working Group on Traces, the issue of the possible presence of lead in cosmetics has been taken into consideration with the intention of drafting a recommendation for lead levels in cosmetic products. In this view, the awareness of what is currently available on the market is key data that could provide information on what can be technically achievable and on the levels of lead to which the consumers are exposed. In particular, lip items were considered interesting due to the fact that a significant part of applied product is likely to be ingested so that exposure is not only based on dermal contact.

In this context, the aim of the present work was to take a snapshot of the current situation on the European market with regard to lead content in lip products.

In the search for an efficient lipstick digestion procedure and lead determination technique, an extensive screening of literature was carried out. Several reports and research papers were published on the presence of trace levels of lead in both cosmetic and food products and multiple methods of sample preparation and analysis have been proposed. The amount of Pb extracted depends significantly upon experimental conditions such as sample weight, combination of used acids, temperature and decomposition procedure. In view of sample decomposition various methods are available: the dry ash one [11,21,23,30] and sample digestion by means of a combination of diverse acid mixtures both in an open system and a microwave digestor [10,12–14,17–20,22,24,25,31–37]. The most common analytical techniques applied to quantify lead in cosmetics and food products include: Electrothermal Atomic Absorption Spectrometry (ETAAS) [24], Flame Atomic Absorption Spectrometry (FAAS) [32], Inductively Coupled Plasma-Atomic Emission Spectroscopy (ICP-AES) [12], Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) [14,25,31], and Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) [19]. Other techniques, also applied in the same fields, were: Confocal Raman Microscopy (CRM) [20], Derivative Potentiometric Stripping Analysis (DPSA) [37], Differential Pulse Anodic Stripping Voltammetry (DPASV) [33], Square Wave Anodic Stripping Voltammetry (SWASV) [34], Laser Induced Breakdown Spectroscopy (LIBS) [10], Screen-Printed Silver Electrode (SPSE) [36], X-ray fluorescence (XRF) [22] and electromembrane isolation (EMI) coupled with capillary electrophoresis (CE) and ultraviolet (UV) detection (EMI-CE-UV) [35].

Among all of the above-mentioned sample preparation and lead determination techniques, the US FDA in-house validated method [14] is noteworthy for its robustness, high lead-recovery and efficiency. It involves sample dissolution by a microwave digestion technique followed by ICP-MS analysis. Hepp et al. [14] proved that, in order to achieve a complete mineralisation of lipsticks, the use of the very aggressive hydrofluoric acid is necessary. This is most probably due to the presence of refractory minerals difficult to digest, such as alumina, silica, titanium dioxide and mica.

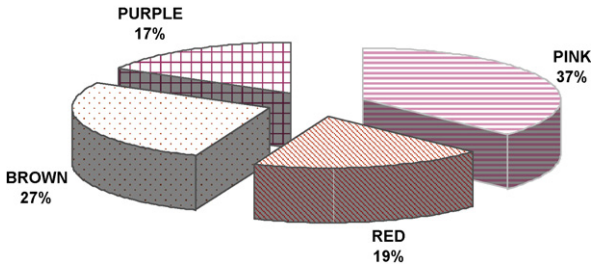

Fig. 1. Colour distribution among the tested samples.

2. Materials, instruments and methods

2.1. Chemicals and instruments

For all the experiments water, 18.2 MΩ quality, was used (Millipore, Vimodrone (MI), Italy). The following acids of the highest purity for metal trace analysis were provided by Sigma–Aldrich (SA Steinheim, Germany): 69% HNO_3, 47–51% HF and 99.97% H_3BO_3. Samples were digested in a microwave oven Multiwave 3000 (Anton Paar, Graz, Austria) and analysed by ICP-MS, SCIEX, ELAN DRC II model (Perkin Elmer, Milan, Italy). Rhenium solution Alfachem (Milan, Italy) was added as an internal standard, before the samples were injected by ICP-MS autosampler. In order to evaluate the extraction efficiency two reference materials, representing different matrix types, were tested: estuarine sediment, SRM 1646a (NIST) (refractory mineral matrix) and lead in base oil 20 standard, ORG-PB8-2Y/Z (SPEX CertiPrep Inc., Metuchen, NJ) (oily matrices containing organically complexed lead).

2.2. Selection of analysed products

In order to have an overview of the current situation of the lead content in cosmetics available on the European market, 223 lip products were purchased for analysis between September 2010 and April 2011 in 15 Member States of the European Union.

It is worth mentioning that there were no strict selection criteria for the sampling (purchase) sites (retail stores, open air markets or shopping malls), brand or product type (lipstick or lip gloss). Various brands, reaching the total number of 55, were tested, covering the majority of those available on the European market. Two different lip product types were considered: lipstick (67%) and lip gloss (33%), in four different shades: pink (37%), purple (17%), brown (27%) and red (19%), reflecting the most common colours of lip products (Fig. 1).

Shades were distinguished mostly by their commercial names, as for majority of them the shade was captured in its name (ex. "red dream" has been categorised as a red shade). In a very few cases, when a commercial name did not state the exact colour, a visual judgement took place.

The country of purchase and production area of the different samples are listed in Table 1. 88% of the samples were produced on the European territory, 6% in the USA and less than 1% in Canada or Japan. For 12 samples (5% of the total), the exact area of production remained unknown.

The market price of the samples varied from "low" (category I – below 5€), to "intermediate" (category II – 5 to 15€), to "high"