ejpmr, 2020,7(2), 95-101

SJIF Impact Factor 6.222

EUROPEAN JOURNAL OF PHARMACEUTICAL AND MEDICAL RESEARCH

www.ejpmr.com

Review Article

ISSN 2394-3211

EJPMR

KOHL AN EYE COSMETIC PRODUCT: LEAD EXPOSURE AND RISK MANAGEMENT. REVIEW

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Article Received on 02/12/2019 **Article Revised on 22/12/2019** **Article Accepted on 12/01/2020**

ABSTRACT

Kohl (surma) is a traditional cosmetic product for the eyes applied using a wooden or plastic stick. It is a powdery substance, usually of a dark color. It is widely used by women in different region of the world (Asia and North Africa). Kohl is traditionally prepared at home or industrially under different names and a variety of packaging. The chemical composition of kohl is different to from one product to another, but the lead is the most element find in the form of galena (lead sulfide). multiple of analytical methods are used to measure the content of lead and other element in kohl such Atomic Absorption Spectroscopy (AAS), X-ray powder diffraction (XRPD) and Scanning electron microscopy (SEP) and Raman spectroscopy. Many studies correlate lead poisoning with use of Kohl. To reduce the lead content in cosmetics the regulatory agencies in some countries have established prerogatives for the import and use of cosmetics based on lead. The aim of this article is to carry out a literature review on established studies to list the different types of kohl and their composition and analytical techniques used for the control, and also report the declared cases of kohl poisoning.

KEYWORDS: Kohl, lead, poisoning, surma.

I. INTRODUCTION

Kohl is a traditional cosmetic product for the eyes. It is a powdery substance, usually of a dark color.[1]

It is widely used by women, mainly in Asia and North Africa (India, Pakistan, Morocco, Algeria, Tunisia, Middle East ...) as a symbol of beauty and femininity (eye cosmetics). It is also given medicinal virtues.[2]

Its composition varies from one region to another but the main compound is a mineral rock. Its use for aesthetic or cultural purposes is very old.[2]

The objective of this work is to carry out a literature review on established studies and cases of kohl poisoning, to list the different types of kohl and their composition and analytical techniques used for the control in the various published studies.

The makeup of the eyes is very old since it appears from the ancient Egyptian Empire (3000 BC), in the form of green and black makeup, called kohls It is reported that the kohl from Egyptian tombs was composed of antimony and lead sulfide. Analytical investigations on 49 flasks of the Louvre Museum made it possible to identify its components:

- lead derivatives: galena predominantly, but also limonite and phosgenite (for their therapeutic virtues);
- Cerussite (natural carbonate of lead $PbCO_3$) and malachite (mineral species of the group of the mineral carbonate and the subgroup of anhydrous carbonates with foreign anions of formula $Cu_2(CO_3)(OH)_2$), to handle the color.

In cosmetology, malachite has a protective action at several levels of the antioxidant defense process of the cells, and a detoxifying effect. Malachite has been used as a mineral pigment since ancient times to make certain blue-green tones clear and bright, Unsaturated lipids in the form of fatty acids and esters as binders. Today, we are witnessing a falsification of Kohl products through a trade that affects all social categories, the appearance of modern kohl based on dyes, the declaration of more and more clinical cases of chronic intoxication. by lead due to prolonged use of kohl.[1-3]:

II. KOHL IN PRACTICE

1. Denominations

Several names are given to the Kohl or its preparations in different region in the world. The kohl or kuhl for the arabic name used in the arabic region. Kaḥal in the biblical hebrew, Athmad or Al-ithmid to describes stibnite-based kohl, he gave the Latin name 'Athimodium,' 'Atimonium,' 'antimonium. Surma is a name derived from the noun 'antimony' used In Pakistan and Turkey. Kajal is also a name how refers to kohl in some countries (India), Kajal can also refer to the Kohl Liner, and Surma refers to the Kohl powder.[4,5]

2. Presentations

i. Traditionally kohl

Traditionally, kohl is applied using a wooden stick. These are the ancestors of our eyeliners and kohl pencils today. It is an anhydrous product protected by a cap of metal or plastic. True kohls were originally prepared using mineral rock (Stibnite or antimonite) plus plant nuclei or animal black. The major of Kohl containing lead derivatives. However, their use has allowed, in the absence of other antiseptics, to avoid certain infections of the eye. Some countries are still using them. It was not until the end of the 19th century and the beginning of the 20th century that a ban on lead derivatives was introduced in food products and cosmetics.

The Traditionally kohl can contain a micro-pigments of borosilicate type of calcium and aluminum coated with iron oxide providing unprecedented shades: black, brown, blue, violet, green; the vegetable or mineral waxes in large quantities; A liquid fats (vegetable oils: castor oil).[2,5,6]

The list of products added to Kohl is very long and complex depending on the region and the population. The true kohl consists of a mineral powder obtained by grinding a metallized mineral mountainous rock called antimony sulfide (antimonite or Stibnite).[5] Ordinary kohl is simply made up of galena (PbS).[5] Locally prepared or imported from other countries (Algeria, Sudan, India, Saudi Arabia, Morocco ...). Stibnite is a mineral species composed of antimony sulfide of ideal formula Sb_2S_3 with traces of As; Tl; Fe; Pb; Cu; Zn; Co; Ag; Au; Bi.

Its compounds have been used to heal cutaneous and parasitic diseases. In pharmacy, there are ointments with antimony supposed to mitigate the pain. Generally mixed with other substances that has a black hue because usually adds ashes. In cases where the kohl is unmixed, it has hues ranging from gray to blue-green.[5]

ii. Registered Kohl

A registered Kohl marketed under different names prepared industrially and put under a variety of packaging. It is a mainly mixed kohl of various origin (India, Pakistan and Saudi Arabia) based on: galena or antimony, activated carbon or black soot, Zinc oxide, Iron oxide, Camphora officinarum (natural camphor has antibacterial properties and helps prevent inflammation, its astringent properties also help rid the eyes of disgusting red capillaries, leaving light and bright), Almond oil, Laurel water of rose, Clarified butter and castor oil. This kohl is controlled more strictly.

Figure 1: Example of commercial kohl of various origin.

3. The traditional preparation of Kohl

The recipe for this powder varies from India to Morocco, each region and each woman had her own recipe, her own secrets. This blush powder is obtained by grinding stibnite (Antimony) or galena (Lead) a mineral extracted from the mountainous rocks, added with multiple components, below some examples of recipes most used by the Moroccan women:

- Recipe N° 1: Mixed parts of kohl, copper sulfate, calcined alum, copper carbonate and some cloves are mixed. The whole thing will be reduced to a state of fine dust in a mortar, then the powder is collected in an earthen vase, which is exposed to a small flame. Then sifted through a fine handkerchief.
- Recipe N°2: Heating the antimony powder at high temperature so that it is more brittle. Then, using a

mortar, it is stacked until a powder is obtained which is watered with rose water. After the mixture has dried, pile up a second time. Then, to the fine powder thus obtained are added cloves, musk and dates kernels. Add peppercorns eventually. Sift the mixture and arrange in a jar.

- Recipe N°3: Finely chop kohl with charred white broth roots, indigo, black olive stones and charred red sugar. Sift the mixture. Sometimes burnt copper is added.
- Recipe N°4: To finely milled kohl, we add vegetable charcoal from the calcination of gall nuts of the Atlas pistachio, calcinated date cores, dried beef bile, burned copper and peppercorns. In Morocco, we add a few drops of olive oil to make it softer to apply.[7]

III. USES OF KOHL

1. Kohl in traditional medicine

In the traditional pharmacopoeias, it is used as:

- Antiseptic: Its components give it properties of eye drops.
- - 'Preventive and curative medicine for common ophthalmia (conjunctivitis, trachoma, ophthalmia of the newborn).
- - Antiseptic for the navel of the newborn
- On the seventh day of a newborn baby, the midwife put kohl to the baby, to protect her fragile eyes from insect bites and conjunctivitis. Kohl is also used therapeutically (to stop bleeding) or prophylactically (for example, its application is justified as a measure of hygiene after circumcision).[5,8,9]

2. The kohl in cosmetics

Kohl is widely used by women as a symbol of beauty and femininity (eye cosmetics). It gives the eyes more brilliance by framing them in a black or blue border. He beautifies the look. It is considered by some populations as a product that keeps the evil eye away.[5,8]

IV. CHEMICAL ANALYSIS

In order to be able to follow these toxic metals, several analytical methods have been developed:

1. Atomic absorption spectrometry

The most effective way is the acid treatment. The operation of this kind of treatment is simple: the sample is heated in the presence of a strong acid (HCl, H_2SO_4, $HClO_4$, HF…) with or not an oxidant agent like H_2O_2 which allows the destruction of any organic matter. The thermic treatment can be used in a polytetrafluoroethylene mineralization flask with a microwave oven. After the mineralization process sample can be analyzed by an atomic absorption spectrometer equipped with hollow cathode lamps as radiation source, a deuterium lamp or Zeeman correction as background corrector; equipped with a graphite furnace or a flame as an atomizing device.[6,9-11]

2. X-ray powder diffraction (XRPD) and Scanning electron microscopy (SEM)

X-ray powder diffraction (XRD or XRPD) is a rapid analytical technique primarily used for phase identification of a crystalline material. XRPD is widely used for the identification of kohl. The kohl is finely ground, homogenized, and average bulk composition is determined.

A scanning electron microscope (SEM) scans a focused electron beam over a surface to create an image. The beam electrons interact with the kohl, producing various signals that can be used to obtain information about the composition. This method is usually used in parallel with XRPD technique.[12-16]

3. Raman spectroscopy

A confocal Raman microscopy was reported as a new method of analyzing kohl. This non-destructive technique offers an important alternative that provide elemental/atomic composition. Raman spectra of three kohl samples have been measured between 150 cm^{-1} and 3000 cm^{-1} at ambient temperature. The main component of two kohl samples was found to be lead(II) sulfide.[17]

V. THE COMPOSITION OF KOHL

Since the 90s, several articles on the composition of Kohl and its consequences have been published because of several arguments and in particular:

- The increase in clinical cases of poisoning,
- The unknown composition that differs from one region to another
- The presence of very toxic metals (lead and derivatives) in the formulation of Kohl.

A study in 1991 on the hazardous use of eye cosmetics in third world countries and northern countries to analyze samples of kohl of several origins (Kingdom of Saudi Arabia, Pakistan, India and Morocco). The lead concentration is around 70% for some samples (Morocco and India). Others have very minute concentrations.[18]

In 1998 another study on the use of ocular cosmetics in Oman enabled the analysis of 47 samples from several origins (Oman, India, Pakistan, SA, Egypt, Tunisia, US, Germany). The analysis was done by XRPD and SEM. The results show that 32 samples have a lead concentration greater than 70%. 14 are based on PbS (galena as major phase) and 17 based on amorphous carbon as major phase, minium (Pb_3O_4), magnetite (Fe_3O_4), zincite (ZnO), calcite ($CaCO_3$) and sassolite (H_3BO_3) are also found in low content.[15]

In 2000, the Intercommunal Laboratory of Chemistry and Bacteriology of Brussels carried out a survey on the lead content of various over-the-counter Kohls belonging to various origins. The following table shows the results of this study.[19]

Tableau 1: Lead content in over-the-counter kohls in Brussels in 2000.

Provenance	Lead content (%)	Provenance	Lead content (%)
Pakistan	76,8	Pakistan	77,3
Inconnue	< 0,1	Pakistan	68,1
Inconnue	< 0,1	Pakistan	69,6
Pakistan	< 82,0	Pakistan	< 0,1
Pakistan	< 0,1	Pakistan	4,1
Pakistan	2	Pakistan	4,1
Maroc	78,6	Pakistan	< 0,1
Maroc	75,5	Pakistan	< 0,1
Pakistan	78,6	Particulier	82,4
Maroc	48,1		

For samples from Morocco, the lead content varies from 48 to 78%. The first study conducted in Morocco on the study of lead in preparations of the traditional pharmacopoeia involved the analysis of kohl samples from different Moroccan regions (Marrakech, Fez, East and Sahara). The author differentiates the Kohl "Messous" (unmixed) from mixed Kohl or "Harre". The chemical analysis was performed by an atomic absorption spectrophotometer after digestion of the Kohl powder. The results in percentage of lead obtained show very high concentrations in all types of kohl analyzed.[9]

Tableau 2: Results of chemical analysis of different types of Moroccan kohl.

Samples	Origin	Color	% of lead
Kohl messous non-mixed	Miscellaneous	Silver	89
Kohl from Marrakech	Jbilet	Silver	80
Kohl from the Fez	Middle Atlas	Black	76
Kohl from the East	Figig and Oujda	Black	79
Kohl from the Sahara	Tafilalet	Black	82

The results of this study showed that the local kohl is particularly heavy in Lead, especially the unmixed kohl with 89% w / w. The concentration of Lead in mixed kohl is diluted by addition of other vegetable, animal and mineral components: 63% w/w.

Another study on the composition of certain eye cosmetics in United Arab Emirates out of 23 samples (origin: India, Pakistan, SA, France, Yemen) by XRPD and SEM showed the following results: 11 samples based on PbS (galena as major phase). 12 samples based on amorphous carbon, Zincite (ZnO), Sassolite (H_3BO_3), calcite ($CaCO_3$).[14]

A final study in Saudi Arabia out of 107 kohl samples allowed to determination of the presence of lead, antimonium and aluminum by X ray fluorescence spectrometer and atomic absorption spectrometry. The analysis of lead in blood and research of camphor and menthol in the Kohl by GC-MS are also studied. The analyzes showed the following results:

- Very high concentrations of lead (more than 50%),
- Traces of aluminum and antimony,
- A significant increase in blood lead levels in kohl users with a significant decrease in hemoglobin,
- The presence of Camphor and Menthol in some samples.[20]

VI. RISKS OF USE OF KOHL

1 . Toxicity of Lead

Lead is a metal that accumulates in the body and is therefore toxic even at a low level of exposure. Galena is the crystallized natural form of lead sulfide (PbS). It is not always easy to recognize lead poisoning. Symptoms include headache, irritability, abdominal pain, vomiting, general weakness, paleness, weight loss, attention deficit, marked learning disabilities, delayed development of speech and hyperactivity. Prolonged use can lead to excessive impregnation by the lead body which affects the brain and bone marrow causing anemia and convulsions.[21]

Chronic poisoning or lead poisoning induced by prolonged use of kohl is a public health problem. Lead is particularly dangerous for children. Exposure to small amounts of lead before birth, infancy, and childhood can result in low birth weight, behavioral problems, learning disabilities, and cretinism. Lead can increase the risk of miscarriages, stillbirths and preterm births in pregnant women, and may also affect male fertility.[21]

The half-life of lead in adults in the blood is 36 days. Only the 10% free form plasma lead represents the diffusible and potentially toxic fraction, hence the importance of blood lead as a control of the internal dose of the metal. It is considered that intoxication begins in children for a blood lead level greater than or equal to 100µg/l. The rate allowed in subjects not occupationally exposed is ≤110 µg/l(5.31µmol/l). Intestinal absorption

of lead is influenced by the type of ingested derivative, gastrointestinal contents, and age. While an adult absorbs barely 10% of the lead ingested, the child can absorb up to 50%.[21]

2 . Kohl and lead poisoning

The most important routes of entry of lead into the body from kohl are the ocular way, dermal and gastrointestinal. The general intoxication is carried out by the gastrointestinal way (via the rubbing of the eyes, then the fingers brought to the mouth).

The maternal-fetal way is also an important route of entry of lead in the newborn from women prolonged use of kohl.[19, 22]

Numerous observations and studies have correlated the use of ocular cosmetics with abnormally high blood lead levels. A high rate was observed among regular kohl users.[23-25]

Since the last century, several studies have identified several clinical cases of lead poisoning due to kohl. The original description of a case of encephalopathy due to kohl abuse was made in England in 1968. Encephalopathy followed by coma, which occurred in a child of three years of Indian origin, had been caused eyelid and conjunctiva application of a powder that has been tested for high levels of lead.[26]

Twelve cases of pediatric lead poisoning of cosmetic origin. Twelve children from five families and found that ten had blood lead levels of more than 360µg/l (the upper limit of normal for pediatric patients). Three patients (aged 2-3 years) had lead levels ranging from 610 - 690µg/l and required rapid treatment. None of the above had a history of pica, and the homes were checked in some instances for potential sources of lead with no results. Samples of the cosmetic were obtained in three instances and were found to contain from 80 to 85% lead sulphide. The kohl was the origin of this lead poisoning.[27]

At a clinic, two Asian infants aged seven and eight months, who had surma (kohl) on their eyelids, had serum-lead levels of 1.9 and 1.6 µmol/1 (normal range 0 - 1.8 µmol/l). Children of this age would not normally be exposed to many sources of lead, and these concentrations are therefore unexpectedly high. The parents were asked to stop using surma, and within a month the levels had dropped to 1.4 µmol/1 in both infants. The surma was found to contain over 90% lead sulphide.[28]

Blood lead concentrations were measured in sixty-two Asian children, of whom thirty-seven had definitely had surma (kohl) applied to their eyes and twenty-five were thought not to have done. The mean concentration in those who had not used surma was 0.98 µmol/1 (203 µg/l) compared with 1.65 µmol/l (342 µg/l) in those who had. Analysis of 29 different samples of surma showed twenty-three of them to be composed largely of lead sulphide (PbS). The study concludes that the use of surma is associated with high blood lead concentrations.[23]

A US study found a significant difference between the blood lead levels of children in Pakistan and India who were exposed to kohl, and children in these communities who were not exposed to the product (0.62 µmol/l vs. 0.21 µmol/l).[29]

In Israel, blood lead concentrations were significantly higher in infants to whom kohl was applied than in those who did not have kohl applied (112 vs 43 µg/l). Among infants not directly exposed to kohl, blood lead concentrations were significantly higher in infants whose mothers used kohl than in those whose mothers did not (52 vs 28 µg/l). Children who were exposed to surma at least two times a week had median concentrations of lead about 30 µg/l higher than those in other children.[30]

A case of lead poisoning in a young Moroccan woman in Belgium admitted urgently in hospital for headaches, vomiting and abdominal pain like cramps appeared for a week. An initial blood lead level of 4.9 mg/l (226.7 µmol/l) while the rate allowed in subjects not occupationally exposed is ≤ 110µg/l (5.31µmol/l). The patient applied every day on her eyelids as the conjunctiva, kohl bought in Morocco.[19]

In 2011, a chronic impregnation in a woman, aged 39 in France. the hypothesis of lead impregnation by long-term use of kohl was issued. A sample of this cosmetic bought in France was analyzed by atomic absorption spectrometry. This product contains more than 80% of lead, as well as traces of arsenic and mercury, all substances banned in the composition of cosmetics in France.[31]

- A prospective multicenter study on a population of pregnant women giving birth in one of the 3 maternity hospitals in the North of Hauts-de-Seine, France. 1021 pregnant women were included. Eighteen out of 1021 newborns (1.8%) had blood lead > 100 µg/l. The main sources of intoxication were tajine dishes (83.3%), kohl (88.9%) and paints containing lead (22.2%). These secondary sources of exposure to Pb alone account for more than 80% of maternofetal lead intoxications.[32]

VII.REGULATION

Regulatory agencies in some countries have established prerogatives for the import and use of cosmetics based on toxic products. In 2016, FDA based on their exposure assessment, the agency has concluded that a recommended maximum level of 10 ppm (10 µg/L) for lead as an impurity in cosmetic lip products and externally applied cosmetics would not pose a health risk.[33]

In European Union, the directive 76/768/EEC regulates the monitoring of cosmetics. It was first legislated in 1976 and it summarizes that cosmetics should not damage human health when applied in normal or under foreseeable conditions. Later, in 2009 a new directive created the list of prohibited substances including Lead and its compounds in cosmetic products.[34, 35]

In Canada, the Health-Canada regulates for products licensed under the monograph stream of the Natural Health Product Regulations made under the Food and Drugs Act, there is a limit for lead impurities of 10 ppm in products applied to the skin.[36]

In 2006 the World Health Organization (WHO) has estimated that the level of tolerable weekly intake of lead from all sources is 25µg / kg, the acceptable daily intake (ADI) corresponding to a 70 kg adult is 250 µg per day.[21]

VIII. MEANS OF PREVENTION AND RECOMMENDATIONS

1 . Preventive means

The only way to effectively combat lead poisoning by kohl is primary prevention, that is, the removal of risk exposure.

- Inform families of the risk of prolonged use of lead-containing kohl and public awareness.
- Limit the use of kohl for newborns and children by facilitating lead absorption.
- The use of kohl in young children and pregnant women should therefore be avoided unless there is certainty that it is free of lead.
- Avoid using it in case of infection of the free edges of the eyelids, acute or chronic dacryocystitis.
- Avoid the use of the same eyeliner pencil by several people. Eye infections due to the use of the infected eyeliner pencil may be passes the infection from one sick eye to another healthy.

2 . Recommendations

Several recommendations can be applied to limit cases of poisoning by prolonged use of Kohl.

- To make analytical studies to know the levels of lead and other metals in the kohl sold in Market.
- Physicians should be made aware of the problem of chronic lead poisoning in young children, which remains a preventable cause of potentially significant cognitive impairment.
- Regulate or control the import of untagged or unidentified kohl.
- Put information and publicity rules for the general public. The labeling of the vial and/or the accompanying leaflet is often incorrect and misleading.
- Ban the kohl whose origin we do not know.

Conduct studies and local surveys to know exactly the different kohls that circulate, their origins and their compositions.

IX. CONCLUSION

The use of kohl / surma results from a very old cultural practice, deeply rooted among many populations, this source of lead poisoning is now recognized in many countries and is being monitored by health authorities.

The legislative measures proved ineffective, the prohibition of the sale and importation of kohls, the composition of which was unknown, prevented neither of them. A strengthening of control measures, collaboration between the various regulatory and state authorities is recommended.

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