

1 Kohl containing lead (and other toxic elements) is widely available in Europe

2 Montserrat Filella<sup>a</sup>, Agathe Martignier<sup>b</sup>, Andrew Turner<sup>c</sup>

3 <sup>a</sup>Department F.-A. Forel, University of Geneva, Boulevard Carl-Vogt 66, CH-1205 Geneva,

4 Switzerland

5 <sup>b</sup>Department of Earth Sciences, University of Geneva, CH-1205 Geneva, Switzerland

6 <sup>c</sup>School of Geography, Earth and Environmental Sciences, University of Plymouth, Drake

7 Circus, Plymouth PL4 8AA, UK

8

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13 **Abstract**

14 Kohl is an eye cosmetic that was traditionally used in many Asian and African countries but  
15 that is now more widely available. Ingredients of kohl reported in previous studies seem to be  
16 rather variable but mention is frequently made of minerals based on Pb whose use in cosmetic  
17 products is prohibited in Europe. We purchased 23 products of kohl from retail outlets in five  
18 different European countries and over the internet and analysed their chemical composition by  
19 XRF and SEM-EDXS. The majority of the products ( $n = 17$ ) did not conform with European  
20 legislation based on the presence of Pb (often as galena), whose concentrations ranged from a  
21 few  $\text{mg kg}^{-1}$  to over  $400000 \text{ mg kg}^{-1}$ . Lead appeared to be present as galena in many cases.  
22 Cadmium, another element prohibited in cosmetic products in Europe, was also present as a  
23 contaminant in 13 products at concentrations up to a few hundred  $\text{mg kg}^{-1}$ . In addition to heavy  
24 metals, minerals of other metals (e.g. Fe and Zn) appeared to be present in the nano-size range  
25 and might represent an additional health hazard. Clearly, the lack of quality control in the  
26 manufacture of kohl results in the widespread occurrence of toxic and unwanted elements and  
27 the trade of illegal products in Europe. In principle, shop sales would be relatively  
28 straightforward to prevent, but products traded through internet are more difficult to regulate.

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30 Keywords: kohl; XRF; SEM-EDXS; lead; cadmium; toxicity

31

## 32 1. Introduction

33 Kohl is an eye cosmetic that has been used since antiquity to darken the eyelids and as mascara  
34 for the eyelashes. It is documented to have been used by ancient Egyptians as a cosmetic and  
35 as a medicinal collyrium (Ebers Papyrus, c. 1550 BCE; Hirschberg, 1982). Kohl is still in use in  
36 the Middle East, the north and Horn of Africa, the Indian subcontinent (India, Iran, Pakistan,  
37 Bangladesh, Nepal), and southern regions of the former Soviet Union. Depending on  
38 geographical area, different terms are used to name similar products (e.g., kajol, al-kahal,  
39 surma, tiro, kwalli) but we will only use the generic term kohl in this study. It should be noted,  
40 however, that the term kohl has been adopted by some commercial cosmetic firms for some  
41 mascara products that bear no relationship with the 'ethnic' kohl products discussed here.

42 In the literature regarding the metalloid, antimony (Aldersey-Williams, 2011; Schwarz-  
43 Schampera, 2014), and in much of the literature about Arab cultural traditions ancient kohl is  
44 identified with stibnite ( $\text{Sb}_2\text{S}_3$ ). More recently, however, the composition of kohl appears to  
45 vary widely and lead sulphide seems to be commonly present (Table SII). It has been suggested  
46 that the originally used Sb sulphide was gradually substituted by Pb sulphide because Sb  
47 became scarce and too expensive (Al-Hazzaa and Krahn, 1995; Hardy et al., 1998; Vaishnav,  
48 2001; Al-Ashban et al., 2004; Jallad and Hedderich, 2005) but no author appears to provide a  
49 precise timeline. Interestingly, evidence gathered towards the end of the nineteenth century by  
50 Fischer (1892) and corroborated by more recent analyses points to Pb as the main constituent  
51 of Egyptian eye preparations rather than Sb (Walter et al., 1999). Some authors assert that the  
52 confusion might arise from the closeness of the Egyptian name for lead sulphide, galena (*stim*)  
53 and the Latin name for antimony (*stibium*) (Mahmood et al., 2019).

54 Traditionally, the kohl powder is applied to the conjunctive surface of the eyelids in the same  
55 way as mascara is applied to the outer surface for a variety of reasons: to make the eyes  
56 beautiful; as a daily tradition stemming from both cultural and religious backgrounds; and for  
57 its perceived therapeutic properties. It is also applied to the umbilical stump of newborn babies,  
58 to the eyes of babies and infants to 'improve' eye health and to protect them against 'bad eye',  
59 and to stop bleeding after newborn circumcision. The use of Pb-containing kohl, particularly  
60 in children, has been repeatedly associated with high Pb concentrations in blood, with excellent  
61 reviews provided by Tiffany-Castiglioni et al. (2012) and, from a more historical perspective,  
62 by De Caluwé (2009).

63 Kohl is now also used by immigrant Asian populations in western countries. The first warning  
64 of the implication of kohl as a cause of Pb poisoning among the Asian community in the UK  
65 was issued by Warley and co-workers more than 50 years ago (Warley et al., 1968). The case  
66 described an Asian child in London with Pb encephalopathy and led to a warning from the  
67 Home Office asking for a voluntary sale restriction in 1968. The subject continued to gain  
68 attention in the UK in the 1970s (Betts et al., 1973; Snodgrass et al., 1973; Josephs, 1977; Ali  
69 et al., 1978; Aslam et al., 1979; Green et al., 1979) but interest subsequently waned. The  
70 presence of Pb was also reported in cosmetics bought in the US nearly 30 years ago (Parry and  
71 Eaton, 1991) and resulted in a request for a control of lead “originating from a variety of  
72 unorthodox sources”. In continental Europe, the presence of Pb in kohl products has been more  
73 sparsely documented, with isolated studies conducted in Denmark (Bernth et al., 2005), France  
74 (Sainte et al., 2010; Kervegant et al. 2012) and Belgium (Bruynel et al., 2002).

75 In the US, kohl is defined as a colour additive and is not permitted in cosmetics or in any other  
76 FDA-regulated product (FDA, 1996, 2001). Kohl is not specifically regulated as such in Europe  
77 but Pb and its compounds have been banned in cosmetics since 1976 (Council of the European  
78 Communities, 1976) and are subjected to current European legislation for cosmetics (European  
79 Parliament and Council of the European Union, 2009). According to the EC legislation, the  
80 main regulatory framework for finished cosmetic products when placed on the EU market,  
81 elements that cannot be present are Sb, Cd, Pb and Se (with the exception of SeS<sub>2</sub> in  
82 antidandruff shampoos), with the presence of Zn also controlled.

83 In the frame of our research on the presence of toxic elements such as Pb and Sb in consumer  
84 products (Turner and Filella, 2017; Turner, 2018), we have investigated the availability and  
85 characteristics of kohl in Europe that does not comply with EC legislation on cosmetics. We  
86 have combined measurements of two complementary techniques: portable x-ray fluorescence  
87 (XRF) spectrometry and scanning electron microscopy-energy-dispersive x-ray spectroscopy  
88 (SEM-EDXS). While XRF provides a rapid semi-quantitative chemical composition of the  
89 whole sample, SEM-EDXS yields an insight into the aspect and visual heterogeneity of the  
90 analysed objects at the microscale range accompanied by the possibility of performing semi-  
91 quantitative analysis of individual components of the samples.

## 92 2. Materials and Methods

93 Products were purchased in Europe through retail shops in Belgium, France, Spain, Switzerland  
94 and the UK or via the internet (Amazon.de, Amazon.uk, eBay) and delivered to Luxembourg  
95 or the UK. All samples were coded, photographed in their original packaging and any details  
96 of composition and origin noted.

### 97 2.1 XRF analysis

98 A battery-operated Niton XL3t 950 He GOLDD+ portable XRF spectrometer housed in a  
99 laboratory accessory stand and remotely operated via a laptop was used for XRF analysis.  
100 Between 2 and 5 g of powdered samples, pastes or kohl stones (used to manually prepare kohl)  
101 purchased in jars or bottles were transferred to polyethylene XRF cups (Chemplex series 1400;  
102 21-mm internal diameter) and collar-sealed with 3.6  $\mu\text{m}$  SpectraCertified Mylar polyester film;  
103 softer material in kohl pencils or sticks (50 to 100 mg) was scraped directly onto Mylar film.  
104 Measurements of elements of atomic mass  $\geq 40$  were conducted in a mining mode and with a  
105 beam width of 8 mm for a total time of 60 s, comprising successive counting periods of 30 s at  
106 50 kV/40  $\mu\text{A}$  (main filter), 15 s at 20 kV/100  $\mu\text{A}$  (low filter) and 15 s at 50 kV/40  $\mu\text{A}$  (high  
107 filter). Elemental concentrations (in  $\text{mg kg}^{-1}$ ) were derived from fluorescent x-ray spectra using  
108 fundamental parameters encoded in the Niton Data Transfer software. The performance of the  
109 instrument was checked by measuring two certified reference sediments (NIST 2709 and  
110 GBW07318) contained in XRF cups with each batch of samples. Detection limits for the kohl  
111 samples, based on the lowest counting errors multiplied by three, ranged  $< 20 \text{ mg kg}^{-1}$  for As,  
112 Pb and Rb to about  $150 \text{ mg kg}^{-1}$  for Ba, Mn, Sn, Ti.

### 113 2.2 SEM-EDXS analysis

114 While XRF provides the elemental composition of whole samples (or areas of 8 mm in  
115 diameter), EDXS measurements are performed on a volume of 2 to 3  $\mu\text{m}$  in diameter that  
116 targets specific grains or groups of grains.

117 For EDXS, samples of kohl were mounted on aluminium stubs using double-sided conductive  
118 carbon tape and coated with Au (ca. 10 nm) using low vacuum sputter coating. A JEOL JSM-  
119 7001F scanning electron microscope, equipped with an EDXS detector (model JEOL EX-  
120 94300S4L1Q), was used to perform analyses and to obtain secondary electron images of the  
121 samples. Imaging was also accomplished with backscattered electrons where elements with

122 high atomic number appear brighter in the image. EDXS measurements were acquired with an  
123 accelerating voltage of 15 kV, a beam current of 7 nA and an acquisition times of 30 s.  
124 Semiquantitative EDXS analyses of elemental concentrations were made without taking C, N  
125 and O into account (elemental quantification is not as good for light elements). The presence  
126 of C can nevertheless clearly be identified in the EDXS spectrum by the presence of a peak at  
127 0.27 keV representing the characteristic carbon  $K_{\alpha}$  line. EDXS results are all presented as mol  
128 %, with detection limits dependent on several parameters but estimated to be around 1%.

### 129 **3 Results**

130 Information about the 23 samples studied is shown in Table 1, with content data available on  
131 the container or packaging given where available. Table SI2 shows pictures of the samples as  
132 purchased and illustrates that some products are sold under different names depending on the  
133 provider and country of purchase (for example, compare samples #5 and #6, and samples #2,  
134 #9 and #10). All products are black or grey, with the exception of a blue pencil (# 12), and  
135 exhibited a variety of textures and lustres. Some products are powders, some are pastes, a few  
136 are sticks or pencils with a soft interior, and two are ‘kohl stones’ used to prepare final products  
137 either at home or by local retailers.

138 The elemental compositions of the kohl samples (atomic mass  $\geq 40$ ) as obtained by XRF  
139 analysis are shown in Table 2 and SI3 and reveal marked variations in chemical makeup. Lead  
140 is detectable in 17 out of the 23 products tested and three distinct groups can be observed. Thus,  
141 some samples contain very high concentrations of Pb in the 400000 mg  $\text{kg}^{-1}$  range (# 1, 3, 13,  
142 18, 19, 23) and some samples contain high concentrations of Pb in the 25000-40000 mg  $\text{kg}^{-1}$   
143 range (# 20, 21, 22), while remaining samples exhibit low concentrations of Pb in the  
144 approximate range of 40 to 1400 mg  $\text{kg}^{-1}$  (# 2, 6, 7, 8, 9, 10, 15, 17). The first two groups point  
145 to a Pb mineral being the main constituent, either alone or mixed with other compounds, while  
146 the third group suggests that Pb is present as a contaminant. Cadmium is detectable in 12 kohl  
147 samples over a concentration range of 14.5–369 mg  $\text{kg}^{-1}$ , with concentrations above 160 mg  
148  $\text{kg}^{-1}$  always associated with Pb-rich samples. Iron and zinc are detected in a total of 18 and 17  
149 samples, respectively, with concentrations ranging two or three orders of magnitude and from  
150 about 100 to 77000 mg  $\text{kg}^{-1}$  for Fe and about 100 to 40000 mg  $\text{kg}^{-1}$  for Zn. It would appear,  
151 therefore, that some samples contain either a Fe or Zn mineral as the main, or as an important,  
152 component. While some samples were relatively ‘clean’ with respect to the aforementioned

153 elements (# 8, 14, 16), a general characteristic of the products is that they all contain traces of  
154 an array of unwanted chemical elements like Ba, Co, Cr, Cu, Ni, Sb, Sn and W.

155 Although SEM-EDXS does not provide quantitative measurements on whole kohl samples, it  
156 provides information on the structure and particle size of the materials and a semi-quantitative  
157 analysis of selected objects and as such is complementary to XRF. Images derived from this  
158 approach are shown in the SI file and a summary of the observations is given in Table 3. SEM-  
159 EDXS confirmed the variety of products sold under the kohl umbrella as well as the  
160 heterogeneous composition of most products. As explained above, EDXS cannot provide semi-  
161 quantitative information on C but a distinctive peak at 0.27 eV in a few samples (#s 11, 14, 16)  
162 suggests material of an organic matrix.

163 Some samples contained a Pb-S mineral that was often mixed with other components (#s 1, 3,  
164 13, 18, 19, 20, 21). This compound revealed stoichiometries of Pb:S of 60:40, 66:34 or 69:31  
165 and appeared as well-defined cubes (Figure 1a, 1b and 1f), embedded in other matrices (Figures  
166 1c and 1d), or as very small particles covering a larger one (Figure 1e and 1f). Galena, a PbS  
167 mineral, is typically cubic, octahedral or a combinations of the two, and it would be reasonable  
168 to assume that the compound we observe is this. Note that the inconsistencies in the  
169 stoichiometries observed above and the stoichiometry of pure galena may be attributed to  
170 instrument response (e.g., an underestimation of the quantity of S, a relatively light element)  
171 and/or surface oxidation and the formation of secondary minerals like Pb sulphate and Pb  
172 carbonate

173 Aluminosilicates and Si-Mg and Si-Na compounds were present in a number of kohl samples.  
174 The Si-Mg compound could be talc, whose presence has already been mentioned in previous  
175 studies (Table SII). Also of interest is the presence of metal oxides in the nano-size or micro-  
176 size range (Zn, #s 4, 6; Cu, # 12; Fe, #s 5, 11) that were located using backscattered images,  
177 and as exemplified in Figure 2. In other cases, the presence of Zn and Fe were registered  
178 without nano-particles being spotted (#s 2, 9, 17).

179 Overall, there is good agreement between the results of the kohl sample analyses arising from  
180 XRF and SEM-EDXS.

#### 181 4. Discussion

182 From the compositional perspective, our results confirm those of previously published studies  
183 that have investigated the makeup of kohl (Table S11). Our findings are, however, both  
184 surprising and cause for concern in that so many products that are readily available for purchase  
185 in Europe at retail outlets and through the internet contain very high concentrations of Pb.  
186 While some of the products containing the highest levels of Pb do not provide quantitative or  
187 qualitative information on kohl composition, others give information that is not accurate or that  
188 is misleading. For instance, two products containing Pb at above 40% by weight declare or  
189 imply the contents to be 'lead-free', with one of the products referring to an Sb-based material  
190 when the metalloid was not detected by our XRF analysis (Sb was only found at trace levels in  
191 four products). The labelling of another product states that no Pb is present according to  
192 laboratory tests but contains well over 1000 mg kg<sup>-1</sup> of the heavy metal according to XRF.  
193 SEM-EDXS results suggest that high levels of Pb across a range of products and texture types  
194 that are manufactured in different countries throughout Asia and Africa arise from the common  
195 use of the sulphidic mineral, galena.

196 An additional concern unveiled by our study is that most kohl products analysed not only  
197 contain lead but also a heterogeneous array of other chemical elements, including the heavy  
198 metal Cd, that is also highly toxic. This suggests that these products lack any quality control  
199 during material sourcing and manufacture and are produced from low-grade and impure  
200 materials. This problem may be more acute than our results indicate because portable XRF is  
201 not a particularly sensitive technique and many of the elements we analysed may be distributed  
202 in a larger number of samples at lower concentrations. Based on the detection of Pb and/or Cd  
203 by our XRF analyses, 21 out of 23 products are non-compliant with respect to current European  
204 legislation.

205 SEM observations also reveal an extremely heterogeneous particle size distributions, often with  
206 very small particles present. Of potential significance from a toxicological viewpoint,  
207 therefore, is the presence of some metal compounds, such as Zn oxide, at the nano- or micro-  
208 sized particle range. Apart from the problems directly associated with the higher solubility of  
209 very small particles in the eyes and mouth, exposure to potentially harmful elements can also  
210 take place via the respiratory system, if particles below PM10 or PM2.5 present in many of the  
211 products investigated were to become airborne. Concerns about the presence of minerals in the  
212 nanosize range in cosmetics usually refer to ZnO, a widely used ingredient in sunscreens (Kim



213 et al., 2017), but in kohl products nano-sized particles composed of a variety of additional  
214 elements (including Pb and Cd) may be present.

215 The presence of seemingly non-toxic compounds such as talc might also be problematic from  
216 a health perspective. Pure talc itself has been implicated in various respiratory illnesses (Wild  
217 et al., 2008) but is not currently classified as carcinogenic according to the IARC (American  
218 Cancer Society, 2020). However, given the apparent lack of quality control in the manufacture  
219 of the kohls referred to above, the presence of talc could be associated with the occurrence of  
220 more harmful asbestos (Rohl et al., 1976).

## 221 **Summary**

222 The results obtained in this study indicate that kohl products available in Europe, but  
223 manufactured elsewhere, have a very physical and chemical heterogeneous composition and  
224 that many of them contain significant amounts of toxic elements like, Pb and Cd, whose  
225 presence is not permitted according to current European legislation. Such products used to be  
226 obtained by European-based users through friends and relatives (Ali, 1978) but now they are  
227 either imported and sold in retail shops or purchased via the internet. While, in principle, shop  
228 sales would be relatively straightforward to prevent, that products can be so readily obtained  
229 through internet sources is particularly worrisome. More generally, the study confirms that the  
230 internet is an essentially unregulated market for consumer goods with health and safety  
231 regulations that are either virtually absent or unenforced.

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235

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- 326

327 Table 1. Description of the products analysed. Photographs of the products are shown in Figure S1.

ID	Product	Origin	Date of purchase	Information as provided in the products
1	Kohl Ithmid by SunnaHealing UK	Bought from Amazon UK, delivered in UK; delivery offered in other countries	22 June 2019	Lead-free antimony according to Amazon webpage; no composition given
2	Kohl Al-Sherifan	Bought in a shop in Madrid, Spain Origin unknown	28 May 2019	Free from lead Active ingredients: Sankh (42.5 %), Sufoof-E-Syah (42.5 %), Bhimseni Kafoor (15 %)
3	Hashmi Kohl Aswad bottle	Bought in a shop in Madrid, Spain Made in Pakistan	28 May 2019	Ingredients: Kohl powder, zinc oxide, amorphous black, blends of herbs Dose (both eyes) 1.4 mg corresponds to Pb 0.00%
4	Hashmi Kajal tube	Bought in a shop in Madrid, Spain Made in Pakistan	28 May 2019	Ingredients: Zinc oxide, waxes, processed carbon black, herbs, clarified butter, <i>Cinnamomum camphora</i> Dose (both eyes) 0.5 mg, Pb 0.00%
5	Khojati Mumtaz Herbal Kajal with almond oil	Bought in a shop in Manchester, UK Made in India	11 Sep 2019	Contents: Cow's glee (30.5 % w/w), Black iron oxide (8.0 % w/w), Waxy base (ad 100 % w/w) Tested in laboratory, free from lead
6	Khojati Mumtaz Delux Kajal (special quality)	Bought in a shop in Manchester, UK Made in India	11 Sep 2019	Contents: Coconut oil (31.3 w/w), Cow's ghee (18.0 % w/w), Sufoof-e-Syah (6.0 w/w), Bhimseni Kafoor (2.0 w/w), Roghan-e-Baid Anjeer (1.8 w/w), Sange Basari (0.4 w/w), Waxy base (ad 100 % w/w) Tested in laboratory, free from lead

7	Khojati Mumtaz Delux Kohl	Bought in a shop in Manchester, UK  Made in India	11 Sep 2019	Contents: Gile Surkh (50.0 w/w), Sufoof-e-Syah (33.0 w/w), Sange-e- Basari (17.0 w/w)  Soaked in 5% almond oil  Tested in laboratory, free from lead
8	Natural kohl powder Black natural eyeliner  No brand	Bought from eBay; delivered in Luxembourg, sent from Germany  Made in Morocco	13 Oct 2019	No composition
9	Kohl Al-Sherifain <sup>a</sup> (2)	Bought from Amazon.de; delivered in Luxembourg, sent from Spain  Origin India	12 Oct 2019	Free from lead  Active ingredients: Sankh (42.5 %), Sufoof-E-Syah (42.5 %), Bhimseni Kafoor (15 %)
10	Kohl Al-Sherifain <sup>a</sup> (1)	Bought from Amazon.de; delivered in Luxembourg, sent from Spain  Origin India	12 Oct 2019	Free from lead  Active ingredients: Sankh (42.5 %), Sufoof-E-Syah (42.5 %), Bhimseni Kafoor (15 %)
11	Sheida. Kohl powder	Bought from Amazon.de; delivered in Luxembourg, sent from Germany  Made in Turkey	12 Oct 2019	Ingredients: Talc, mica, nylon-12, ethyleneacrylic acid copolymer, isoestearyl neopentanoate, phenoxyethanol, caprylic glycol, sorbic acid
12	Intense kohl, crayon yeux  No brand	Bought in a shop in Brussels, Belgium  Origin unknown	30 Oct 2019	Ingredients: triethylhexanoin, cera carnauba, candeilla cera, caprylic/capric triglyceride, polybutene, bis-diglyceryl polyacyladipate-2, talc, cetyl palmitate, tocopheryl acetate, parfum, benzyol salicylate, buthylphenyl methylpropional, linalool

13	No label, no brand	Bought in a shop in Brussels, Belgium Origin unknown	30 Oct 2019	No composition
14	Al-Ameerah eye make-up	Bought in a shop in Brussels, Belgium Made in Pakistan	30 Oct 2019	No composition in English
15	Hatimi Eye-liner	Bought in a shop in Brussels, Belgium Made in India	30 Oct 2019	Ingredients: Carbon black, comphor, alum, menthol
16	Seeme Kajal	Bought in a shop in Brussels, Belgium Made in Pakistan	30 Oct 2019	No composition
17	Mumtaz Delux Surma (export quality)	Bought in a street market in Gaillard, France Made in India	14 Dec 2019	Contents: Ochre rouge 50.00%, carbone noire naturelle 33.00%, oxyde de zinc 17.00%. Imbibé d'huile d'amande à 5% (in French in the original)
18	Al-Asmad Alharmain	Bought in a shop in Geneva Made in IRP; address in Dubai	16 Dec 2019	Zam Zam & Rose water
19	Pure kohl Ithmid eyeliner powder Antimony Health Means Wealth	Bought from eBay; delivered in UK, sent from UK Origin: Yemen	31 December 2019	No composition



20	Kohl. No brand	Bought from eBay; delivered in UK, sent from UK Origin: Morocco?	31 December 2019	No text in English Text in eBay: Fair trade Moroccan kohl. Black eyeliner-hand carved wooden bottle and kajal
21	Kohl. No brand	Bought in eBay, delivered in Luxembourg, sent from Algeria Origin: Morocco?	31 December 2019	No composition Text in eBay: kohl Moroccan wooden bottle hand-made mascara surma eyes attractive eyeliner new
22	Surma stone	Bought from eBay; delivered in UK, sent from UK	31 December 2019	No composition Text in eBay: 100g surma Ithmid kohl pure stone
23	Surma stone	Bought from eBay; delivered in Luxembourg, sent from India Origin: India	31 December 2019	No composition (the stone arrived wrapped in a newspaper) Text in eBay: Surma stone antimony kohl stibine kajal 100 g natural eyeliner Hindu Muslim cool

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Table 2. Elemental concentrations (mg kg<sup>-1</sup>) in the kohl samples determined by XRF. Concentrations of other elements present are in table S2.

Sample	Ba	Ca	Cd	Co	Cr	Cu	Fe	K	Mn	Ni	Pb	Sb	Sn	Ti	Zn
1	987	11501	369	241		471	1838		766	576	444 507		344		2522
2		32 625	19.8		59	207	18 416	1213			112			943	37 994
3		1358	311	229		498	547		735	455	440 957	217	286		4273
4	294	435			64										380 827
5			17.2		79		121 861								
6		18 708			224	140	77 867	867			1431			7394	144 241
7			17.6				367				8				7730
8		5549				107					323			169	
9		44 426	14.5	134		190	12 934	560			39			668	34 892
10		66 367	17.3	236	60	255	15 072	872			36			1046	41 150
11		2017			303		371 485	7261	1098				370	20 796	2159
12		3653	18.4				34 201	11 606					222	44 918	
13	2648	2814	270	356		143	403	1191	777	448	441 466	217	236		122
14		165	17.9												
15	156 380	5245		268		178	6375		6905	1061	62				7502
16		276	17.4												
17		8298			241	213	133 247	1184	193		203			17 889	149 673
18	621	993	160	177		428	338		547	371	311 820	174	208		492
19	650	2348	275	311		710	995		762	637	467 632	149	315		7580
20		1448		0	983	184	354 929	536	3836	269	25 988			590	94
21		26 268				216	619	158 184			41795		155		180
22		10 267						1839			40 321				
23	1033	34 685			169	164	21 594	20987	1384	826	438 351			3650	6903

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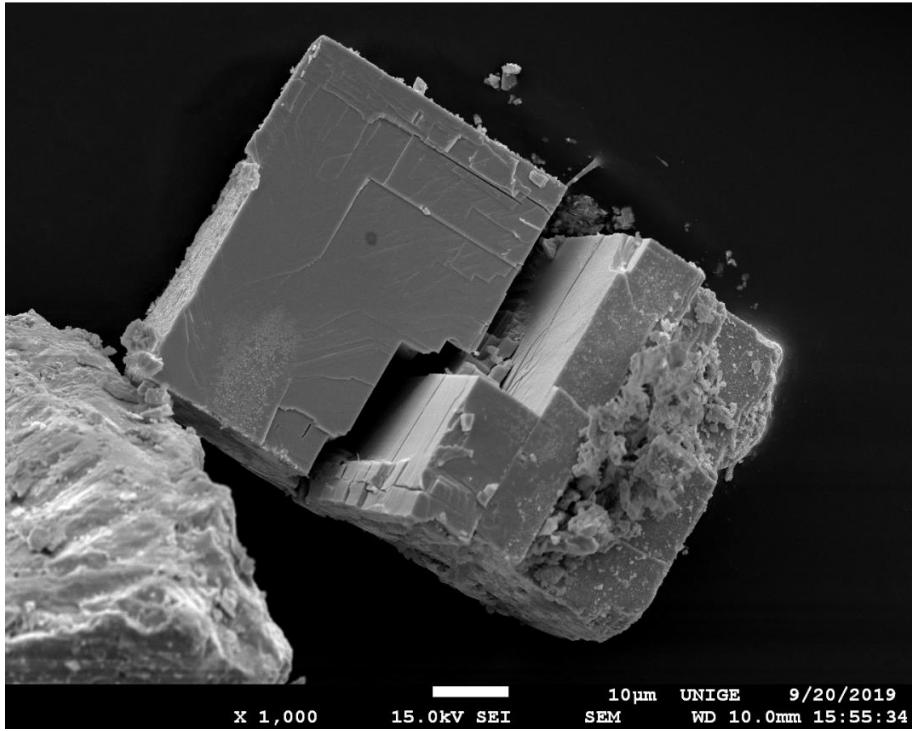
332 Table 3. Key morphological and chemical characteristics of the samples obtained by SEM-EDXS analysis. Complete information, including  
 333 SEM pictures of all samples, is found in Table S3. All values are in molar percentages.

ID	Observations
1	Many size homogeneous cubes observed, with composition Pb:S = 60:40 Higher variability in the composition of particles in the agglomerates, ranging from Pb-S only particles to particles that, while containing a high % of Pb, also had Ca, Ti, Fe, Cu
2	Rather heterogeneous sample with probable presence of quartz and talc. Presence of Zn
3	Heterogeneous sample with, at least, three types of particles present: particles with Pb and S only (60:40 ratio), particles with C only and particles mostly containing Si and Mg (possibly talc)
4	C matrix that contains many small particles (size range: 150 to 800 nm with many at ~300 nm), detected in backscattered images. These particles contain Zn only by EDXS, possibly made of ZnO
5	C matrix with weak Fe signal. Analysis of backscattered images confirmed the presence of Fe nanoparticles. The presence of Fe was indicated by the producer (size range: 200 nm to 1 $\mu$ m)
6	Rather heterogeneous sample with C only particles, particles rich in Si-Mg, Si-Al or Ca. Some contained Zn. Analysis of backscattered images confirmed the presence of nanoparticles containing high amounts of Zn, possibly ZnO (size range: 150 to 800 nm)
7	EDXS analysis showed either an entirely C matrix or C with a bit of Fe but Fe nanoparticles could not be found in backscattered images
8	All organic C
9	Two types of particles: Si-Mg and Zn-S. The Si-Mg particle has a similar composition as particles found in sample 10. Backscattered electron-imaging did not help to identify metal-rich particles
10	Organic matter with, at least, two types of minerals compounds, Si-Mg and Si-Na; the Si-Mg could be talc

11	On a substrate of aluminosilicates, abundant nanoparticles with Fe only (size range: 100-300 nm)
12	Organic matrix with aluminosilicates particles. Small particles containing Cu only were also spotted in backscattered images. Their sizes ranged from 1.5 to 5 $\mu\text{m}$
13	Many particles containing Pb and S only sitting on a C matrix. Pb:S ratio: 60:40
14	All organic C
15	Highly heterogeneous sample with different types of particles present (Si-Mg and pure organic C) including a lot of possible $\text{BaSO}_4$
16	All organic C
17	Heterogeneous sample. Results suggest the presence of particles are rich in Zn or Fe among others that contain Si, Al, Ti
18	Two different types of material. One consists of particles with Pb and S only (Pb:S ratio: 66:34) and the second type contains O and C only
19	All particles analysed contained Pb. Most had Pb and S only ((Pb:S ratio: 69:31) but one was 100% Pb, probably and Pb oxide None contained Sb, as said by the seller
20	Three types of particles detected by EDXS: one formed by organic C only, one 100% Fe and one with Pb and S, ratio 66:34
21	Two types of particles detected by EDXS: one formed by organic C and one with Pb and S, with a mean ratio 66:34
22	It is a compound of Pb and S, ratio 69:31
23	Not studied by SEM-EDXS

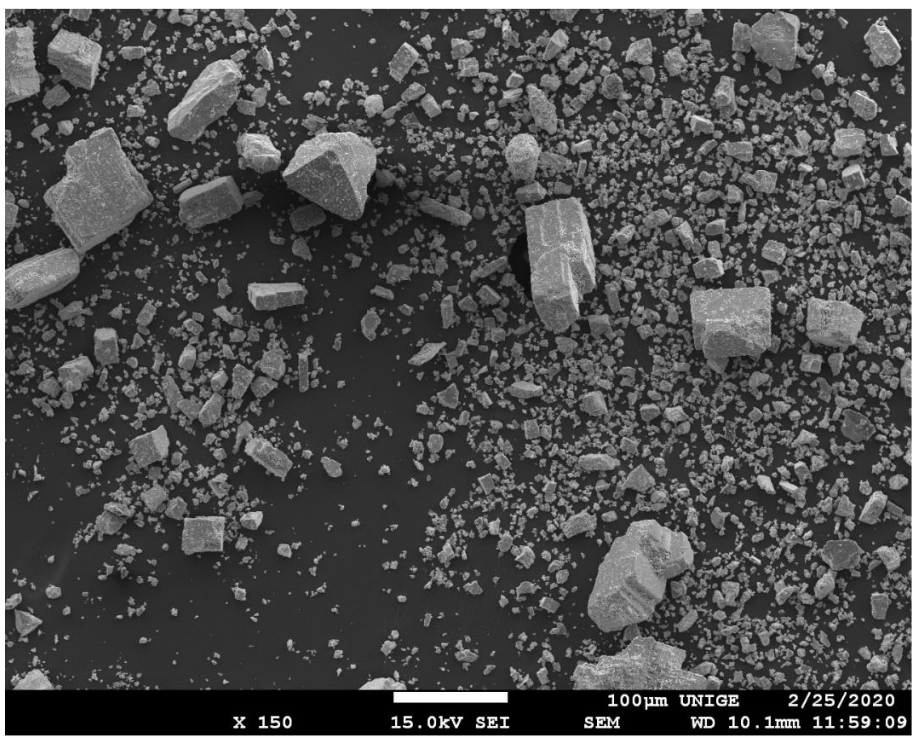
335 Figure 1. Pb-S compounds found in different samples: (a) #1, (b) #19, (c) #13, (d) #3, (e) #18.  
336 Compounds are revealed as isolated minerals of well-defined shape, as in (a), (b) and (e), or  
337 embedded in organic matrices, as in (c) and (d). Note that (c) is a backscattered electron image.  
338 Size is indicated at the bottom of the pictures.

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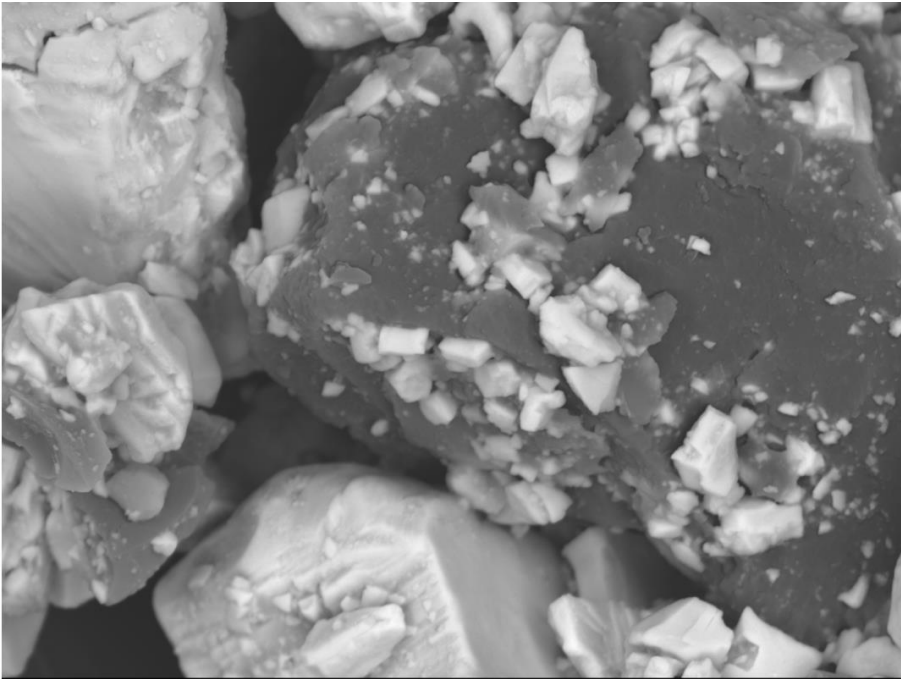
341 (a)

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343 (b)

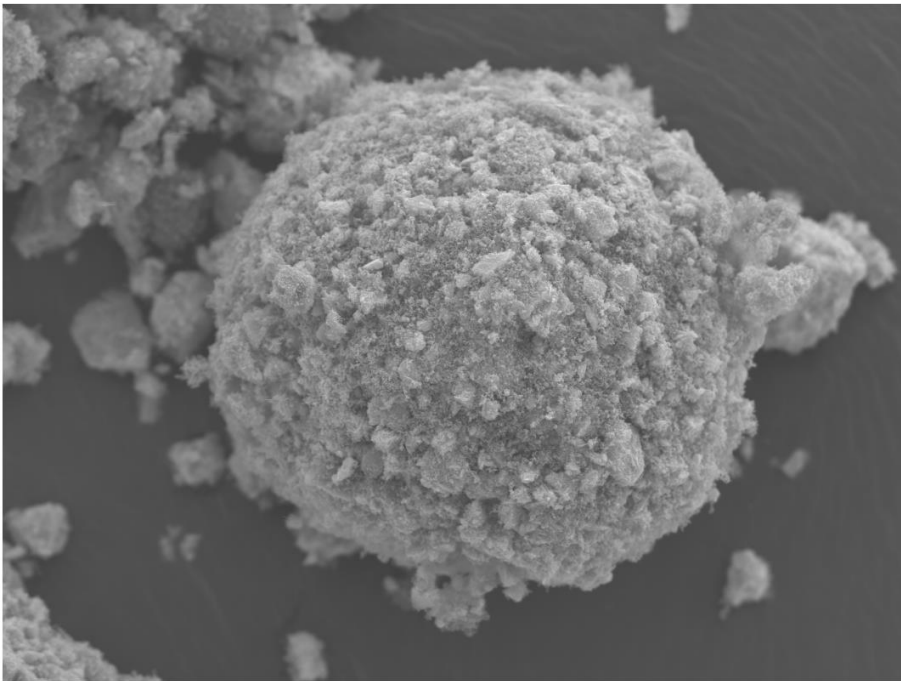
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1µm UNIGE 11/22/2019  
X 5,000 15.0kV BEI\_COMP SEM WD 10.0mm 11:25:35

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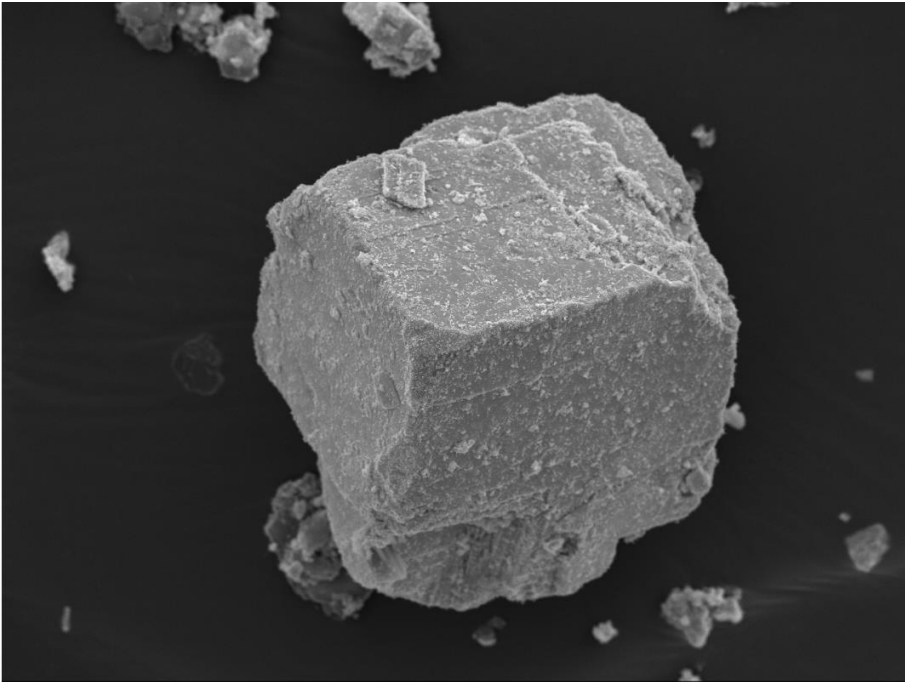
(c)



10µm UNIGE 10/4/2019  
X 2,300 15.0kV SEI SEM WD 10.0mm 14:33:31

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(d)



X 1,100 15.0kV SEI SEM 10µm UNIGE 2/25/2020 WD 9.9mm 11:48:04(e)

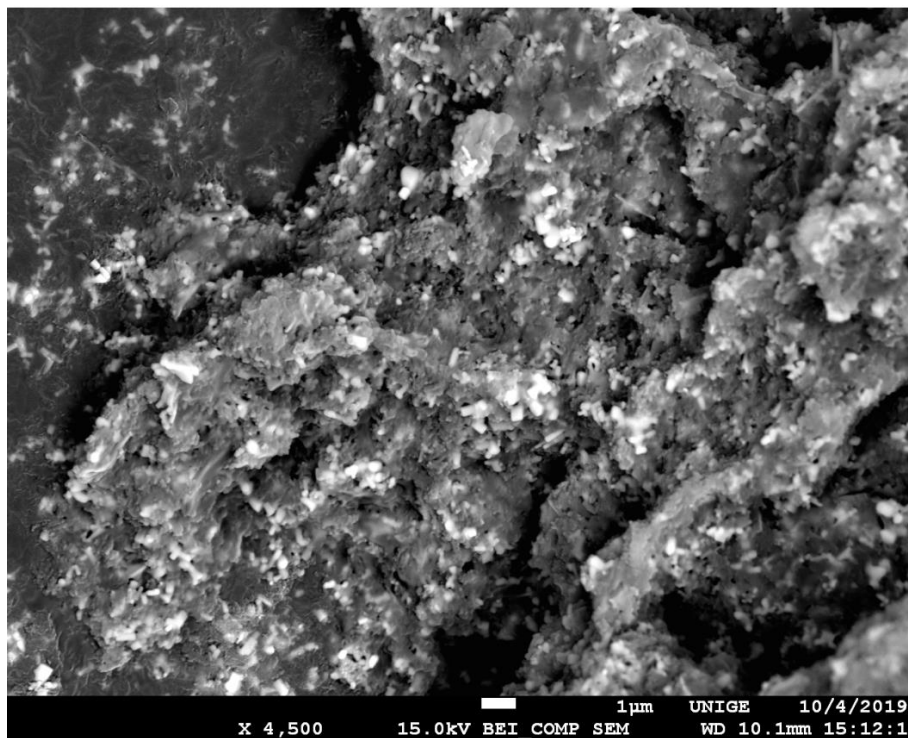
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351 Figure 2. Backscattered electron images showing, in white against the darker surrounding  
352 material, (a) Zn nanoparticles in sample # 4, (b) Zn nanoparticles in sample # 6 and (c) Fe  
353 nanoparticles in sample # 5.

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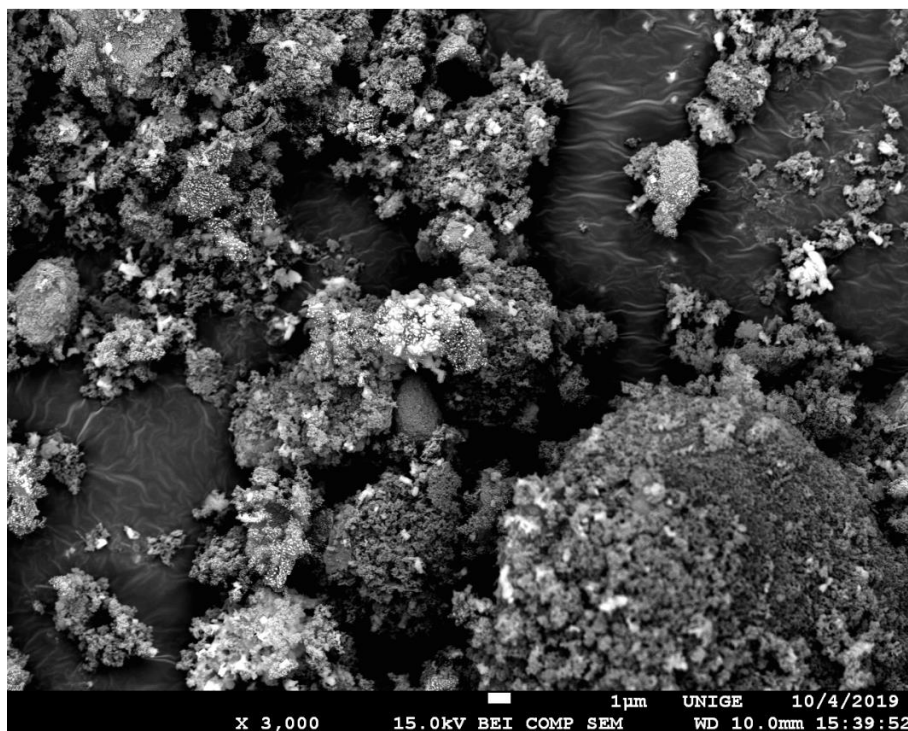
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(a)

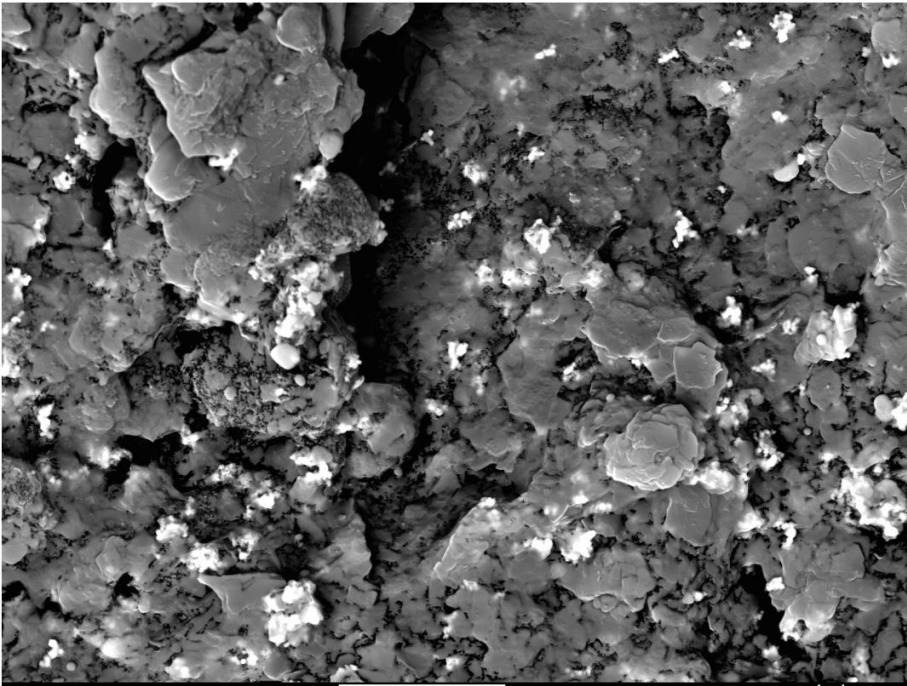


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(b)





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X 2,200 15.0kV BEI COMP SEM 10µm UNIGE 10/4/2019  
WD 9.8mm 16:42:14

(c)