EUROPEAN COMMISSION

Health & Consumer Protection
Directorate-General

Scientific Committee on Health and Environmental Risks

SCHER

Scientific opinion on

The Danish EPA Survey and Health Risk Assessment of Lead in Jewellery

(Danish EPA report on the environmental and health risks posed by heavy metals in jewellery)

Scientific Committees

on consumer safety

on emerging and newly identified health risks

on health and environmental risks

The SCHER adopted this opinion by written procedure on 22 February 2010

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SCHER

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It may also address questions relating to examination of the toxicity and eco-toxicity of chemical, biochemical and biological compounds whose use may have harmful consequences for human health and the environment. In addition, the Committee will address questions relating to methodological aspect of the assessment of health and environmental risks of chemicals, including mixtures of chemicals, as necessary for providing sound and consistent advice in its own areas of competence as well as in order to contribute to the relevant issues in close cooperation with other European agencies.

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http://ec.europa.eu/health/scientific_committees/environmental_risks/members_wg/index_en.htm

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1. BACKGROUND

The Danish Environmental Protection Agency (EPA) analysed a number of items of jewellery present on the Danish market for their release of arsenic, barium, cadmium (Cd), chromium, copper (Cu), mercury, nickel (Ni), lead (Pb), antimony and selenium[1]. Four metals migrated into artificial sweat in concentrations above the detection limit: Cd, Cu, Ni and Pb. They were selected for exposure and risk assessments for consumers. Exposures were calculated for wearing the jewellery for 16 or 24 hours/day, or for sucking on it for 2 hours/day. Exposure levels were then compared with Margin to Tolerable Daily Intake (Margin TDI) values, which took Danish background exposures to Cd, Cu, Ni and Pb into account.

Exposure through the skin was shown to be a risk for Cd, and in certain cases also for Ni. No health risk was related to the exposure of the skin to jewellery containing Cu or Pb. When considering oral exposure through sucking of the jewellery, potential health risks were identified for Cd, Ni and Pb. Cu was not found to cause health risks by oral exposure.

Based on these results, the authors concluded that it cannot be excluded that potential health risks could arise from wearing or sucking some of the metal jewellery examined in this study. Thus, the Danish Ministry of the Environment invited the Commission to consider whether the results of this study, and the possible health risks to consumers (e.g. kidney toxicity for Cd; reduced IQ in children for Pb) should justify regulatory action.

Currently, migration limits exist at European level for Ni in jewellery (Entry 27, Annex XVII of REACH)[2]. Cd is also regulated under REACH (Entry 23, Annex XVII) but specific limits do not yet exist for Cd in jewellery. A review is currently ongoing and the Commission intends to propose amendments to Annex XVII of REACH relating to further restrictions on the uses of Cd, notably in jewellery. At present, no specific limits apply for Pb in jewellery at European level.

2. TERMS OF REFERENCE

Against the above background, the Scientific Committee is requested to:

(1) Critically review the Danish EPA Survey and health assessment of chemical substances in jewellery and comment in particular on its completeness, reliability and on the validity of its conclusions. The Committee is also asked to comment on the chosen methodological approach and assumptions made for the risk assessments (e.g. 2 hours/day sucking time for oral exposure assessments, calculation methods, reference Margin to TDI values, etc).

(2) In light of its response to question 1, pronounce itself as to whether there may be reasons for concern arising from the exposure of consumers from jewellery containing Pb, as concluded in the report. In elaborating its point of view, the Committee is asked to take into account all known sources of exposure of consumers to Pb. If the Scientific Committee disagrees with the conclusions of the report, it is invited to elaborate on the reasons and provide comments.

(3) In light of its response to question 2, assess whether the exposure of consumers to Pb from jewellery should be mitigated. If so, the Committee is asked to assess whether a limit of Pb in jewellery can be established that will lead to exposure of consumers not giving reasons for health concerns (e.g. the Danish limit of 100

[1] http://www2.mst.dk/udgiv/publications/2008/978-87-7052-853-5/pdf/978-87-7052-854-2.pdf

[2] http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=OJ:L:2007:136:0003:0280:EN:PDF

mg/kg Pb in products, including jewellery[3]). In answering this question, consideration should also be given to other sources of Pb than jewellery.

3. OPINION

3.1. Review the Danish EPA Survey

"Survey and health assessment of chemical substances in jewelleries" (Danish-EPA, 2008) investigates potential risks that may be associated with a release of heavy metals from these products commonly used by the consumer. The project was initiated on the basis of reports that cheap items of jewellery may contain problematic substances, for example large amounts of lead (Pb), and the death of a boy attributed to Pb intoxication after swallowing a piece of jewellery containing >99% Pb. One of the purposes of the project was to provide an overview of Pb (and several other heavy metals) content in items of jewellery on the Danish market and to evaluate whether Pb (and other heavy metals) released from jewellery during use may cause health problems due to skin contact or after sucking.

Wood-based items of jewellery, which may be painted with paints containing Pb, were not represented. In total, 318 parts of 170 of metal items of jewellery (both of low and relatively high price categories: 34 finger rings, 51 necklaces, 43 bracelets, 34 ear rings, 3 pieces of piercing jewellery, 6 ankle chains and 6 other products, for example toe ring) purchased from the Danish retail market were screened for the content of Pb and several other metals including gold (Au) and silver (Ag) by X-ray fluorescence spectroscopy (XRF). The Au and Ag jewellery parts were divided into "coated", "alloyed" and "-like" respectively, because the Pb content of the jewellery parts might be released differently, depending on whether the product is coated with precious metals, alloyed with precious metals, or does not contain precious metals at all. On the basis of Ag and Au content, the products were assigned "Gold coated" (Au 0-33%), "Gold like" (Au <LOD (limit of detection)), "Silver coated" (Ag 0-20%), "Silver alloy" (Ag 20-80%), and Silver-like (Ag <LOD).

Some of the conclusions from the screening of the metal jewellery were:

- 58% of all examined items of jewellery contained Pb above 0.01% (w/w). The maximum content of Pb in a jewellery part was 69.6 % (w/w).

- The data do not indicate that jewellery imported from a certain country have a larger probability of containing Pb. However, 30% of the 37 items of jewellery originating from China contained Pb above 0.01% w/w.

- There appeared to be a greater chance of a high content of Pb in the cheaper metal jewellery (0 – approx. 10 DKK. per gram)

- The results do not indicate a correlation between the Pb content and the product category (i.e. gold coated, silver-like, non-precious metal etc.).

However, the conclusions based on the XRF-measurements have limitations. The XRF technique can only semi-quantitatively determine the metal composition on the surface of a material (thin samples) as X-rays are not able to penetrate deeper into thicker metal samples such as jewellery. In the present study, results of XRF screening (based on signal intensities) are considered to be appropriate for selecting the jewellery parts for migration testing. This rationale for selecting the samples for migration analysis, based on signals in XRF analysis, is supported by SCHER because migration mainly takes place from the surface.

Based on the screening results, 25 jewellery parts (from 21 products) were selected for the migration test to determine release of heavy metals including Pb. The jewellery parts

[3] Statutory order on ban on import and sale of products containing lead - Stat. Ord. no. 1082 of 13.09.2007

selected covered the different product types (rings, necklaces, bracelets etc.) and product categories (silver-coated, golden-like, non-precious metal etc.); however, the primary criterion was that they represented a part of the jewellery coming in contact with skin. It was attempted to select jewellery items representing the entire concentration span related to the content of Pb. However, focus was primarily on contents between 100 and 2 000 mg Pb/kg because products containing >100 mg/kg Pb should not be placed on the Danish market (Danish-Legislation, 2007). Four of the tested jewellery items did not contain lead, but they were probably included because of additional focus on other heavy metals.

The migration analysis method used was "Migration to artificial sweat" according to DS/EN 1811:2000. The migration test was performed on 2 x 2 g jewellery parts, without giving any rationale. The report indicated that the areas of the tested samples were estimated, and the results of Pb migration (μg/g) were then converted to migration based on area (μg/cm^2). For exposure assessments both by oral and dermal route, jewellery area in contact with the skin or in the mouth should be determined more precisely. SCHER considers potential contact area of a jewellery item as more relevant to exposure assessment and recommends a more detailed assessment of the area of the jewellery item that my come into contact with skin.

The results of the migration analysis in artificial sweat resulted in measurable Pb release from 14 of the 21 Pb containing jewellery parts. Surprisingly, the results of the migration analysis did not show a correlation between migration and Pb content of the tested jewellery parts. In addition, significant Pb migration was measured even when the tested material was coated with gold/silver.

Considering that finger rings, ear rings, bracelets, ankle chain, piercing and other parts are not likely to be mouthed, only necklace (chain and pendant from 8 products, total of 10 parts) should be considered for oral exposure by mouthing.

In the survey, only necklaces (chain and pendant from 8 products, total of 10 parts) have been considered for oral exposure by mouthing, since finger rings, ear rings, bracelets, ankle chain, piercing and other parts were not considered likely to be mouthed. However, SCHER recommends including finger rings, bracelet pendants and special tongue/lip piercing among the possibly mouthed items.

Regarding dermal exposure assessment, the low uptake of the metals under study through the skin is considered by the Danish EPA and the uptake factors are supported by the references given.

The results from the migration testing were then introduced into health risk assessment. Health risk assessment is based on review of the toxicology of the metals under consideration and applies TDI-values derived by WHO or other organizations. Regarding Pb, the authors decided to use only 50 % of the TDI derived in 1995 (based on effects of Pb at blood concentrations of 10 μg Pb/dL, which translates to a TDI of 3.6 μg Pb/kg body weight /day) to account for results of newer studies showing on IQ development in children at blood Pb concentrations lower then 10 μg/dL. This approach is acceptable.

Due to a considerable exposure to Pb from a variety of sources with food as the most important exposure pathway in both children and adults, the authors use a "margin to TDI" approach to define additional tolerable exposures. In the end, this "margin to TDI" subtracts the known exposures to a metal (or other chemical) from specific sources (e.g. food) from the TDI and thus gives an indication of how much of the TDI is already covered by these sources and a maximal possible contribution of the source under study. The approach therefore does not make recommendations on risk management options and gives an indication of the magnitude of a potential problem.

The review of SCHER identified several issues with analytical determinations and the transformation of the results into the exposure assessment:

1. The Pb-release from jewellery items is higher when contrasted with long-term Pb release rates found in other studies. This may be due to an inappropriate sample preparation for the migration analysis. The migration data reported (2-280 µg Pb/cm^2 in 4 hours) are up to two orders of magnitude higher than long-term runoff rates under natural rain from pure lead sheets subjected to atmospheric corrosion. The composition of rain obviously differs from saliva. Rain does not contain Pb^{2+} chelating compounds (that facilitate dissolution) such as lactic acid present in saliva. However, rain is more acidic than saliva and the pH effects generally dominate over the effects of chelating compounds (see below). Long-term runoff rates of pure lead sheets range from 90 to 500 µg Pb/cm^2/year equivalent to 0.04-0.22 µg/cm^2 in 4 hours (Matthes *et al.*, 2002; Wilson, 2003). The high migration noted could be due to a number of causes including the "first flush" effect (see point 2 below) and/or to exposure of freshly cut surfaces of the jewellery items. Samples for migration testing were cut from the jewellery items and the sites thus exposed were covered with wax to avoid migration from the recently cut surface. However, under the experimental conditions used (4 h at 40 °C), the wax may have been perforated or loosened from the jewellery part, and thus allow migration of Pb from the sites exposed after cutting.

2. An assumption in the report with regard to Pb-migration is that a 4 hour static extraction test represents a model for long term exposure because the migration data are contrasted with human health thresholds for chronic exposure. SCHER questions the assumption that the initial migration of Pb to the extraction fluid is representative for repeated events of sucking which is a dynamic process. Repeated discontinuous extractions separated by a 'dry spell' of the metal may better mimic this exposure situation. Corrosion kinetics of metals or alloys often show that metal release rates in biological fluids or water slow down after an initial fast release, commonly denoted as the "first flush" (Skeaff et al., 2000; Herting et al., 2007). This decrease is a consequence of the time-related formation of a more corrosion-resistant surface (Herting et al., 2007). Moreover, the effects of sucking and cleaning the jewellery should also be integrated in the experimental approach.

The extents of migration and "first-flush" effects have a major impact on exposure assessment. The document assumed constant rates since the risk ratio (TDI/Exposure) is inversely proportional to the corrosion rate. For risk assessment, it needs to be verified if repeated (consecutive) extractions give similar release rates as the first flush.

3. The results from the artificial sweat test (performed according to a European standard, see Annex 1) are used for the estimation of dermal and oral exposure. The dermal exposure is not critical for Pb according to this study. The oral exposure suggests a possible risk. However, migration of Pb to saliva should have been performed under dynamic conditions to mimic sucking. An extrapolation from artificial sweat to saliva cannot be made due to differences in chemical composition such as pH and presence of chelating agents.

4. The detection limit (DL) of the method to quantify Pb is not sufficient to permit conclusions on potential risks. If the DL (about 2.5 µg Pb/g/4h, see appendix H of the report) is introduced in the calculation of the potential exposure, the daily dose at the 'detection limit' ($I_{oral,pot}$) would be 1.25 µg/kg bw/day[4]. This value already exceeds the tolerable 'margin to TDI' in children at the background exposure of 1.02 µg/kg/day. However, because of the limitation of the analytical method applied (inappropriate sensitivity and DL), assessment at migration rates below the detection limit is not possible. Based on all available literature, risks at levels below the used DL, especially for children, cannot be excluded. Therefore, a limit for Pb in jewellery items translating to consumer exposures without health concerns cannot be defined based on the Danish EPA study.

[4] Calculation made according to the equation on p.81 of the document for a jewellery of 40 g

3.2. May there be reasons for concern arising from the exposure of consumers from jewellery containing Pb?

Based on the study data, the initial flush of Pb from jewellery items into sweat may be in the order of several 100 µg Pb/g jewellery in 4h (Matthes *et al.*, 2002). Assuming that the data obtained with artificial sweat are sufficiently representative for migration of lead into saliva and translating this into an exposure assessment, even when using a sucking time of only 1h/day and the 4 g weight used in the study, Pb ingestion by sucking could be in the order of 100 µg Pb/day, well exceeding the TDI for children. However, the actual data in the study are questioned since average runoff rates of pure lead sheet in the environment is app. two orders of magnitude lower then the release rates of Pb from the jewellery items. Therefore, no conclusion regarding a potential concern should be made based on the study. SCHER recommends performing of an optimized migration study with repeated extractions as described above to simulate the sucking behaviour especially by children and apply a sensitive analytical method for Pb-determination at lower levels. SCHER also recommends to use surface area instead of weight for the risk assessment.

A potentially significant release pathway of Pb may also come from piercings that are in direct contact with mucous membranes and tissues. This issue is not addressed in the Danish EPA report and cannot be commented in the opinion due to lack of information.

It also needs to be considered that the large amount of lead in jewellery items, which may be ending up in waste collection, could result in an environmental issue. A total sale of 312 tons of jewellery items made of non-precious metals in Denmark (in 2005) suggests that many thousand tons of such items may be sold annually in Europe. A considerable part of the sold cheap jewellery may be discarded as waste.

3.3. Can a limit be defined with these data, e.g. the Danish 100 mg/kg limit?

Based on the study results, a scientifically justified limit for Pb in jewellery cannot be derived. The document concludes 'it is seen that none of the jewellery containing a maximum of 100 mg/kg of lead caused health problems'. That conclusion is incorrect since it is based on observations below the detection limit. Therefore, risks cannot be excluded for the 'undetectable Pb' in the migration test when using the study data.

For an appropriate limit for Pb in jewellery to be set there is a need for a detailed risk assessment considering EU wide exposure to Pb from different routes and sources within the EU for general population and vulnerable groups, in particular children. It is also important to apply an appropriate migration study protocol and sensitive analytical method.

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Table 1: Content of Pb in jewellery parts investigated for Pb migration in artificial sweat. Both results were reported, when duplicate analysis revealed significant differences in the results.

Jewellery				Pb content (% w/w)	Migration of Pb in artificial sweat	
Product category	Appearance* (Coating/alloy metal content)	Part Analysed			microg/g	microg/ccm
		No.	Description			
Necklace	Gold coated (Au 1.51%)	95.3	Catch	66.59	250	190
Necklace	Gold coated (Au 0.98%)	70.3	Catch	40.95	140	100
Other**	Silver coated (Ag 10.35%)	136.2	Disc	33.49	100	100
Necklace	Silver-like (Ag <LOD)	56.1	Pendant, back side	26.14	23/64	6/18
Necklace	Gold coated (Au 0.30%)	70.1	Pendant	21.42	-	-
Necklace	Silver-like (Ag <LOD)	62.1	Pendant	15.62	69	53
Ring	Silver coated (Ag 7.70%)	68.1	Ring	14.35	140	110
Necklace	Gold coated (Au 0.69%)	38.1	Piece of jewellery	9.21	220	210
Necklace	Gold coated (Au 0.17%)	6.1	Angle	7.64	6/160	5/130
Bracelet	Gold coated (Au 1.49%)	107.1	Chain with balls	3.86	-	-
Necklace	Gold coated (Au 0.51%)	101.3	Cornet/cone	3.65	-	-
Necklace	Gold coated (Au 0.44%)	101.1	Back side of heart	2.58	73	140
Necklace	Silver alloy (Ag 21.8%)	138.1	Pendant from behind	1.77	150/210	190/280
Ear ring	Silver-like (Ag <LOD)	130.1	Needles	1.21	-	-
Finger ring	Silver coated (Ag 2.83%)	125.1	Ring	1.20	93/540	60/260
Ankle chain	Silver coated (Ag 2.7%)	169.1	Heart	0.36	17	15
Ring	Non-precious metal	152.2	Matt ring	0.16	-	-
Piercing	Silver coated (Ag 4.73%)	164.1	Heart back side	0.11	2/4	3/7
Bracelet	Golden-like (Au <LOD)	60.1	Bracelet	0.03	-	-
Necklace	Non-precious metal	91.3	Chain	0.03	-	-
Necklace	Non-precious metal	91.1	Big ball	0.02	-	-
Ear ring	Gold coated (Au 18.95%)	99.1	Twisted ring	0.00	-	-
Ear ring	Gold coated (Au 18.92%)	99.2	Non-twisted ring	0.00	-	-
Ring	Silver-like (Ag <LOD)	88.1	Ring without stone	0.00	2/5	2/5
Other	Non-precious metal	26.2	Catch	0.00	-	-
Necklace	Silver-like (Ag <LOD)	56.1	Pendant, back side	26.14	23/64	6/18
Necklace	Gold coated (Au 0.30%)	70.1	Pendant	21.42	-	-
Necklace	Silver-like (Ag <LOD)	62.1	Pendant	15.62	69	53
Necklace	Gold coated (Au 0.69%)	38.1	Piece of jewellery	9.21	220	210
Necklace	Gold coated (Au 0.17%)	6.1	Angle	7.64	6/160	5/130
Necklace	Gold coated (Au 0.51%)	101.3	Cornet/cone	3.65	-	-
Necklace	Gold coated (Au 0.44%)	101.1	Back side of heart	2.58	73	140
Necklace	Silver alloy (Ag 21.8%)	138.1	Pendant from behind	1.77	150/210	190/280
Necklace	Non-precious metal	91.3	Chain	0.03	-	-
Necklace	Non-precious metal	91.1	Big ball	0.02	-	-

LOD: Limit of detection
*Appearance, Gold coated: Au 0-33%, Golden like: Au <LOD, Silver coated: Ag 0-20%, Silver alloy: Ag 20-80%, Silver-like: Ag <LOD
**Other: For instance toe ring etc.