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Chromium and cobalt in leather: A Danish market survey

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Abstract

Introduction: Leather has been a significant source of chromium (Cr) allergy in Denmark since the 1990s. More recently, cobalt (Co) allergy has been identified in leather as a source of allergic contact dermatitis.

Objectives: To measure Cr and Co levels in Danish leather goods.

Methods: A total of 87 leather samples were collected, all tanned in Europe. Handheld X-ray fluorescence (XRF) device was used to screen for the presence of Cr and Co. The 20 leather samples with the highest concentrations of Co and Co were tested using International Organization for Standardization (ISO)-standards.

Results: XRF analysis showed Cr in 78/87 (83.9%) samples and Co in 52/87 (59.7%), with average concentrations of 41 mg/kg (range: 0.0–77 mg/kg) and 0.22 mg/kg (range: 0.0–2.9 mg/kg), respectively. ISO 10195 and 17 075–1 testing identified Cr (VI) in 7 out of 20 samples (1.4; 0.3–4.2 mg/kg), while ISO 17072-1 detected Co in 6 of 20 samples, averaging 3.95 mg/kg (range: 0.22–7.9 mg/kg).

Conclusion: Most leather samples contained Cr, which was expected, while Cr (VI) was detectable in seven out of twenty tested samples but only detected in one product above the regulatory limit of 3 mg/kg. A potentially significant concentration was found for Co.

KEYWORDS

allergic cobalt dermatitis, allergic chromium dermatitis, chromium, leather

1 | INTRODUCTION

Nickel (Ni), cobalt (Co) and chromium (Cr) are common metal allergens that cause allergic contact dermatitis (ACD) in individuals exposed to either a high concentration over a short period of time or a low concentration over a prolonged period.[1] Although Co is the second most common metal allergen in ACD patients,[2] determining the clinical relevance of a positive patch test reaction to Co is often challenging due to insufficient knowledge about exposure patterns.[3] Several studies since the 2010s have suggested that contact with bioavailable Co in leather through skin contact might induce contact allergy.[4] Although there is a growing body of evidence confirming the presence of Co in leather products,[5,6] few studies have systematically investigated the occurrence and amount of Co released from such products.

Since the 1990s, leather exposure has become the primary cause of Cr allergy in consecutive dermatitis patients in Denmark.[7–9] Depending on intrinsic antioxidant content, environmental conditions and leather age, Cr-tanned leather might release both trivalent or

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hexavalent forms of Cr [Cr (III) or Cr (VI)] when coming into contact with the skin,[10,11] both capable of causing ACD.[12,13] Cr(VI) is of particular concern due to its potent allergenic properties, lower elicitation thresholds, and higher skin penetration rates.[14] In May 2015, the European Union (EU) implemented a regulation limiting the content of Cr (VI) in leather to a maximum of 3 mg/kg.[15] The question remains whether the proposed reduction in the EU-enforced limit of Cr (VI) from 3 to 1 mg/kg, first suggested in 2021, will be implemented in 2026.[16] There is a lack of systematic post-regulatory market data against these restriction limits.

This study aimed to determine the levels of Cr, Cr (VI), and Co present in leather goods available to consumers in the Danish market.

2 | METHODS

2.1 | Sample collection

In November 2023, Danish leather importers were contacted and asked to provide excess leather samples from production for testing on a voluntary basis. All leather samples had to be imported after 2015 and processed within Europe.

2.2 | XRF screening

To investigate the concentration of metals in the provided leather samples, a handheld XRF scanner (X-MET8000, Uedem, Germany) was used as a screening instrument for the presence of Co and Cr. Previous studies have validated the use of XRF screening for Co in leather, particularly at high concentrations.[4] The device presents the content of each element in weight percentage (wt%) along with a range of statistical measurement errors based on single-point measurements. The manufacturer's recommendation was followed by using the alloy setting for leather. The measurement was conducted with an energy source of 40 kV, an analytical depth of 100 μm, and a measurement time of 8 seconds. The manufacturer did not provide the limit of detection (LOD) for Co and Cr contents in leather.

2.3 | Laboratory analyses

A subsample of the 20 leather items with the highest concentrations of Cr and Co, as determined by XRF, was selected for further testing under International Organization for Standardization (ISO)-standardised protocols. Twenty leather samples were analysed for total Co using an extraction in acid artificial perspiration solution at pH 5.5 (37°C, 4 h) outlined in ISO 17072-1[17] with a detection limit of 0.1 mg/kg. Twenty leather samples were analysed for the release of Cr (VI) following the procedures specified in ISO 17075-2[18] (extraction in a phosphate buffer at pH 8.0 for 3 h) with a detection limit of 3.0 mg/kg and uncertainty of measurement ±10.2%. These analyses were carried out at Eurofins BLC Leather Technology Centre Limited, United Kingdom. For both extraction methods, the leather was preconditioned at 50 ± 5% relative humidity (corresponding to ISO 2419:2012 or ISO 139:2005 + A1:2011),[19,20] according to the Eurofins report.

The same 20 leather samples were also analysed in duplicates for Cr (VI) according to the ISO 17075-1 method,[18] with corresponding pre-conditioning according to ISO 10195[21] (80°C for 24 h of the dry leather). This pre-conditioning was used to remove seasonal and inter-laboratory effects due to differences in relative humidity, and to avoid high relative humidity (>35%), which results in false-negative results.[22] A few measures were taken to reduce the LOD. All samples were screened for the full wavelength range from 300 to 750 nm to account for any interfering peaks. All samples were measured before the addition of the reagents (1,5-diphenylcarbazide and 70% phosphoric acid) to account for any intrinsic colour interference by subtracting this spectrum from the spectrum after the reagent reaction. For solutions with colourants from the leather samples, Reverse Phase C18 Solid Phase Extraction (SPE) (Hypersep©, Thermo Fisher) was used to remove colourants before the analysis. The detection limit was dependent on the sample and ranged from 0.2 to 1 mg/kg. The calibration curve was a seven-point (0, 20, 60, 125, 250, 500 and 1000 μg/L) linear ($R^2 = 0.999$) curve.

3 | RESULTS

Out of the eight leather importers contacted, five responded. We received a total of 89 leather pieces, with 7, 11, 14, 24 and 31 pieces from each importer, respectively. Tables 1 and 2 provide a summary of the main findings from XRF screening, ISO 17072-1 for extractable total Co, and ISO 17075-2 (50% relative humidity pre-conditioning) and ISO 17075-1 (with ISO 10195 pre-conditioning for <10% relative humidity) for extractable Cr (VI).

3.1 | XRF screening

XRF analysis showed Cr in 78 (83.9%) samples and Co in 52 (59.7%), with average concentrations of 41.05 mg/kg (range: 0.0–76.9 mg/kg) and 0.22 mg/kg (range: 0.0–2.9 mg/kg), respectively.

3.2 | ISO standards

ISO 17075-2 testing (50% relative humidity pre-conditioning) identified Cr (VI) in 1 sample (2.2 mg/kg). ISO 17075-1, pre-conditioned with ISO 10195 (<10% relative humidity), detected Cr (VI) in 7 samples, with the highest sample (4.2 mg/kg) being the same sample as detected by the ISO 17075-2 test with the 50% relative humidity pre-conditioning. The other six detected samples had Cr (VI) levels ranging from 0.33 to 1.8 mg/kg, with two samples exceeding 1 mg/kg (1.6 and 1.8 mg/kg). ISO 17072-1 detected Co in 6 of 20 samples, averaging 3.95 mg/kg (range: 0.22–7.9 mg/kg).

Leather colour minimally influenced Cr (VI) detection: detected in 44% of black leathers (avg. 0.51 mg/kg), 40% of brown leathers (avg.

TABLE 1 The 20 leather samples exhibiting the highest Cr content as determined by XRF scan and Cr (IV) release test (EN ISO 17075:2017) with uncertainty of measurement ±10.2%.

		Cr (VI) (mg/kg) (±SD)		
Sample	Description	Lab 1[a]	Lab 2[b]	XRF Cr (%) (±SD)
A	Green leather	ND	ND	67.2 ± 12.5
B	Black leather	ND	0.33 ± 0	67.4 ± 4.1
C	Black leather	ND	ND	67.9 ± 4.1
D	Brown leather	ND	ND	68.4 ± 4.0
E	Blue leather	ND	ND	68.7 ± 0.0
F	Black leather	ND	ND	69.5 ± 8.2
G	Black leather	ND	0.76 ± 0.021	69.7 ± 8.4
H	Grey leather	ND	ND	69.7 ± 0.0
I	Black leather	ND	ND	69.7 ± 2.6
J	Green leather	ND	1.6 ± 0.11[c]	70.4 ± 3.6
K	Brown leather	ND	ND	71.3 ± 15.5
L	Red leather	ND	ND	71.5 ± 0.0
M	Brown leather	ND	ND	71.6 ± 4.8
N	Brown leather	2.2	4.2 ± 0.12	71.6 ± 0.0
O	Black leather	ND	0.35 ± 0.049	72.4 ± 33.8
P	Black leather	ND	ND	72.5 ± 3.4
Q	Black leather	ND	ND	74.9 ± 0.0
R	Black leather	ND	0.61 ± 0.31	74.9 ± 8.7
S	Green leather	ND	ND	76.7 ± 0.0
T	Brown leather	ND	1.8 ± 0.12	76.9 ± 0.0

Note: 1 mg/kg corresponds to 0.0001%.

Abbreviations: Co, Cobalt; ND, not detected; SD, standard deviation; XRF, X-ray fluorescence.

[a] ISO 17075-2 (50% relative humidity pre-conditioning, ISO 2419:2012).

[b] ISO 10195 + ISO 17075-1.

[c] Average and standard deviation (SD) of independent duplicate samples.

3.0 mg/kg) and 33% of green leathers (1.6 mg/kg); absent in grey, red and blue leathers (Table 1). On the contrary, Co detection showed a stronger colour correlation: detected in 75% of green leathers (average 4.5 mg/kg), one yellow leather (7.9 mg/kg Co) and 8% of brown leathers (2.2 mg/kg), but not in white or black leathers (Table 2).

4 | DISCUSSION

The findings of this study provide important insight into the prevalence and concentrations of Cr and Co in leather samples from Danish importers. The detection of Cr with XRF in 83.9% of the samples aligns with previous studies indicating widespread use of Cr in the leather tanning process.[7] However, Cr (VI) was detected in 7 out of 20 samples (35%) at 0.33–4.2 mg/kg, with 1 out of 20 tested samples (5%) exceeding the current regulatory restriction limit of 3 mg/kg. Three out of 20 samples (15%) exceeded the proposed restriction level of 1 mg/kg.

In our study, we observed a significant discrepancy in the metal quantities measured by XRF and ISO methods. For both metals, this is related to the different chemical forms that are measured by the tests. XRF measures the total amount of the metal elements, without distinguishing their exact chemical form, for example, if it is soluble, bioaccessible or in ionic form. XRF further has a limitation for light elements, which means it excludes the organic material, resulting in an overestimation of the heavier elements Co and Cr. In the case of Cr, the difference between the ISO tests and the XRF is caused by several factors. First, the total measured Cr (in weight-%) by XRF is an overestimation because of the exclusion of light elements by XRF. The real total percentage of Cr in Cr-tanned leather is around 2%, and in some cases up to 20%, but never as high as the XRF-measured 60%–70%. The measured percentage values are not wrong, but the percentage values only refer to elements heavier than magnesium. Second, the ISO method only measured the soluble hexavalent Cr form, and it is a fraction far less than 1% of the XRF-measured content, Table 1. This is expected since most of the Cr is in its trivalent form in leather.[11] In the case of Co, the acid artificial perspiration solution extraction (ISO 17072-1:2019) only measures the bioaccessible fraction of released Co. This fraction was less than 1% of the total amount of Co measured by XRF, Table 2. This is because most of the dyes, pigments or impurities of Co contained in the leather will not be chemically soluble at a pH of 5.5 during a 4-h extraction period at 37°C.

TABLE 2 The 20 leather samples exhibiting the highest Co content as determined by XRF scan and extractable (perspiration solution) heavy metals screening (BS EN ISO 17072-1:2019) with detection limit 0.1 mg/kg.

Sample	Description	Co (mg/kg)/Co (%)	XRF Co (%) (SD)
A	Brown leather	ND	0.1 ± 0.1
B	Yellow leather	7.9/0.00079	0.1 ± 0.0
C	Brown leather	ND	0.1 ± 0.1
D	Green leather	5.2 / 0.00052	0.1 ± 0.1
E	Brown leather	ND	0.1 ± 0.0
F	Brown leather	ND	0.2 ± 0.6
G	Brown leather	ND	0.2 ± 0.0
H	Brown leather	ND	0.2 ± 0.0
I	Green leather	ND	0.2 ± 0.0
J	Brown leather	2.2/0.00022	0.2 ± 1.0
K	Brown leather	ND	0.2 ± 0.0
L	Black leather	ND	0.3 ± 0.0
M	Green leather	2.5/0.00025	0.3 ± 0.0
N	Brown leather	ND	0.3 ± 0.1
O	Brown leather	ND	0.6 ± 0.0
P	Brown leather	ND	0.7 ± 0.4
Q	Brown leather	ND	0.9 ± 0.0
R	Black leather	ND	1.6 ± 0.6
S	White leather	ND	2.9 ± 0.0
T	Green leather	5.7 / 0.00057	5.4 ± 2.5

Abbreviations: Co, Cobalt; ND, not detected; SD, standard deviation; XRF, X-ray fluorescence.

Our results highlight the significant role of pre-test conditions on the detectability of Cr (VI) in leather samples. Unfortunately, the ISO 17075 standard test does not require a specific pre-conditioning method but gives a high degree of freedom. There are several possible ISO standard methods to pre-condition leather. The Eurofins lab (lab 1) used a pre-conditioning standard that primarily had been developed for physical and mechanical testing (ISO 2419:2012) while lab 2 used a specially developed pre-condition standard for Cr (VI) testing (ISO 10195). The main difference is 50% (lab 1) versus <10% (lab 2) relative humidity in these pre-conditioning methods.[22] It is well known that the relative humidity prior to ISO 17075 testing influences the results of Cr (VI), often drastically and that relative humidities above 35% in most cases result in non-detectable Cr (VI).[22–24] This is because there is a high abundance of trivalent Cr in leather, which can oxidise to Cr (VI) on the surface of the leather in dry air. Therefore, to ensure consistent test results among labs, pre-conditioning under controlled conditions is necessary. A pre-conditioning at low relative humidity effectively prevents the formation of a moisture film on the leather, thereby facilitating the oxidation of Cr to its hexavalent form, particularly in leathers that do not contain sufficient antioxidants to inhibit this conversion.[10,25] It should be highlighted that relative humidities below 35% are relevant for consumers of leather products, as this relative humidity range is common, for example, in heated indoor environments and under sunlight irradiation.[26,27]

The detection of Co in 59.7% of the samples shows a potential regulatory gap. Although Co is less frequently discussed in legislation, its presence at detectable and variable concentrations could pose a risk similar to Cr (VI). The detection of Co in leather might mean that it should be treated like Cr (VI) in terms of consumer safety.

In 2002, a study by Hansen et al. investigated the prevalence of Cr-induced ACD due to leather exposure from Danish leather samples and, in particular, the risks associated with Cr (VI). Their findings showed that approximately 35% of leather articles on the Danish market had detectable levels of Cr (VI) exceeding 3 mg/kg, with concentrations ranging from 3.6 to 14.7 mg/kg.[28] In comparison, our study presents an optimistic perspective on the effectiveness of the regulatory measures. Our results showed a reduction in the detected concentrations of Cr (VI), with only one leather sample (5%) exceeding the regulatory limit of 3 mg/kg at a concentration of 4.2 mg/kg. This suggests that the legislation may be effective in reducing some of the public health risks associated with leather goods.

Bregnbak et al. investigated the association between Co allergy and leather-induced dermatitis.[29] They found a significant association between Co allergy and dermatitis due to non-occupational leather exposure, highlighting leather as a notable source of Co sensitisation. This is consistent with our observations of detectable levels of Co in leather goods and supports the hypothesis that leather goods may serve as a significant source of exposure to Co, potentially leading to ACD. While the ISO 17072-1 test measures sweat-extractable Co at pH 5.5, earlier research suggests that Co (up to 4.7 mg/kg Co extracted from leather samples at pH 8.0) is also extractable at more neutral pH conditions.[30] Hedberg et al. also suggested that Co release is tannery-dependent and, therefore, relatively easy to eliminate.[30] While there are no studies investigating the sensitization limit of Co, Fischer et al. have reviewed dose–response studies to determine the elicitation threshold for Co chloride.[31] They identified elicitation dose (ED) values, which are doses that cause ACD in a certain percentage of Co-allergic individuals. The ED_{10} values (doses eliciting a response in 10% of sensitised individuals) ranged from 0.07 to 1.95 μg Co/cm^2. In our study, the highest concentration found was 7.9 mg/kg, which for the thickness, dimensions, and mass of our leather is approximately converted to 0.26 μg/cm^2. This value might be sufficient to elicit a reaction.

Future research should focus on longitudinal studies to monitor Cr and Co levels in leather goods over time, especially considering the evolving regulations and tanning practices.

4.1 | Strengths and limitations

The study sourced a diverse range of leather samples from multiple Danish importers, ensuring a broad representation of the market. This diversity helps to assess the presence and concentration of Cr and Co in commercially available leather goods for Danish consumers. Testing metal content and release using current ISO-standardised methods makes the results more reliable and relevant, so they can be compared with other studies and regulatory benchmarks.

The voluntary basis for sample provision might introduce a bias if importers selectively submitted samples they believed were compliant or of higher quality, thus not accurately representing the broader market. This is a cross-sectional study, so we cannot comment on the development of metal concentrations over time after use of the leather. This would require a longitudinal study to assess the release of metals from the leather over time and use. However, it is known from an earlier study that Cr (III) release decreases with usage time while Cr (VI) increases or remains unaltered.[32]

5 | CONCLUSION

Most leather samples contained Cr, which was expected, while Cr (VI) was detected in seven products (35%) but only exceeded the regulatory limit of 3 mg/kg in one product. A potentially significant concentration was found for Co. Currently, there is no EU legislation that establishes a limit value for Co in leather. The authors suggest that prior to the current ISO standard for Cr (VI), leather to be prepared with ISO 10195 to minimise inter-laboratory variability and ensure a more accurate concentration measurement for consumer safety. Further research is needed to ensure consumer safety and inform potential regulatory developments.

AUTHOR CONTRIBUTIONS

Mikkel Bak Jensen: Writing – original draft; project administration; investigation; conceptualization; methodology. **Farzad Alinaghi:** Funding acquisition; writing – original draft; supervision; conceptualization. **Ivan Chen:** Formal analysis; methodology; data curation. **Jonas F. Hedberg:** Investigation; supervision; methodology; formal analysis; data curation. **Yolanda S. Hedberg:** Investigation; validation; methodology; formal analysis; data curation; supervision; writing – original draft. **Claus Zachariae:** Writing – review and editing; resources. **Jeanne Duus Johansen:** Supervision; writing – review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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SUPPORTING INFORMATION

Additional supporting information can be found online in the Supporting Information section at the end of this article.

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Supplementary

Supplementary table 1: Handheld XRF-scan data and respective reference numbers (1-87) for Cr and Co.

Reference	**Chromium (Cr)**			**Cobalt (Co)**		
	%	**Avg.**	**+/-**	**%**	**Avg.**	**+/-**
1	0	0	0	0	0	0
2	1.89	0.63	0.89	0	0	0
3	0.64	0.68	0.032	0	0	0
4	0.82	33.83	28.199	0	0	0
5	72.42	24.68	33.762	0.1	0.05	0.051
6	76.9	76.9	0	0	0	0
7	66.85	22.28	31.513	0	0	0
8	0.46	49.26	34.54	0.13	0.08	0.057
9	53.02	53.02	0	0.85	0.85	0
10	69.45	61.23	8.217	0	0.42	0.423
11	74.85	74.85	0	0	0	0
12	0	37.42	37.432	0	0	0
13	44.17	39.67	30.721	0	0	0
14	67.18	54.7	12.477	1.41	0.7	0.704
15	0.84	36.75	27.357	0.14	0.52	0.633
16	68.71	68.71	0	2.11	2.11	0
17	65.71	67.21	1.5	0.2	1.15	0.956
18	66.41	66.94	1.281	0	0.77	0.95
19	52.07	52.07	0	0	0	0
20	1.34	26.71	25.363	0	0	0
21	0	0	0	0.08	0.08	0
22	0.73	0.37	0.367	0.21	0.15	0.065
23	0	0.24	0.346	0	0.1	0.08
24	63.07	63.07	0	2.9	2.9	0
25	70.38	66.73	3.365	0	1.45	1.449
26	61.33	64.93	3.919	0	0.97	1.366
27	52.89	52.89	0	0	0	0
28	37.77	45.33	7.558	0	0	0
29	0.53	30.4	21.997	0.25	0.08	0.118
30	60.38	60.38	0	0.11	0.11	0
31	62.08	61.23	0.851	1.35	0.73	0.622
32	32.48	32.48	0	0.56	0.56	0
33	71.45	71.45	0	0	0	0
34	0	35.72	35.725	0	0	0
35	60.93	44.13	31.496	5.35	1.78	2.522
36	0.69	0.69	0	0	0	0

37	0.87	0.78	0.091	0	0	0
38	0	0.52	0.375	0	0	0
39	0	0	0	0.72	0.36	0.361
40	0	0	0	0.25	0.25	0
41	0	0	0	0	0.24	0.34
42	0.93	0.93	0	0.19	0.19	0
43	0	0.46	0.464	0	0	0
44	0	0.31	0.438	0	0.06	0.088
45	0	0	0	0	0	0
46	0.49	0.25	0.245	0.11	0.05	0.055
47	0	0.16	0.232	0	0.04	0.052
48	0	0	0	0	0	0
49	0	0	0	0.1	0.05	0.051
50	0	3.33	3.335	0	0	0
51	0	0	0	0	0	0
52	6.67	23.23	28.349	0	0	0
53	57.91	57.91	0	0.13	0.13	0
54	7.47	32.69	25.219	0	0.06	0.063
55	0	21.79	25.719	0	0.04	0.059
56	55.42	55.42	0	0	0	0
57	59.75	59.75	0	0	0	0
58	58.24	59	0.755	0	0	0
59	60.09	60.09	0	0	0	0
60	54.3	57.2	2.897	0	0	0
61	74.85	63.08	8.651	0	0	0
62	8.42	8.42	0	0	0	0
63	60.62	60.62	0	0.11	0.11	0
64	60.62	59.05	1.571	0.11	0.05	0.054
65	67.38	61.83	4.13	0	0.04	0.051
66	64.72	64.72	0	0	0	0
67	64.72	57.37	7.355	0	0.09	0.088
68	69.7	61.48	8.358	0	0.06	0.083
69	40.28	40.28	0	0	0	0
70	71.28	55.75	15.502	0.1	0.05	0.051
71	71.55	66.75	4.8	0.12	0.12	0.004
72	61.95	61.95	0	0.12	0.12	0
73	68.42	67.3	3.99	0	0.08	0.059
74	71.58	71.58	0	0.31	0.31	0
75	63.32	67.45	4.131	0.09	0.2	0.108
76	60.31	65.07	4.766	0	0.13	0.13
77	64.63	64.63	0	0.09	0.09	0
78	63.89	64.26	0.368	0	0.05	0.048
79	69.72	66.08	2.589	0	0.03	0.044

80	76.16	76.16	0	0.09	0.09	0
81	67.87	72.02	4.146	0.11	0.1	0.008
82	72.49	72.18	3.393	0	0.07	0.047
83	62.91	62.91	0	0.21	0.21	0
84	67.18	65.04	2.136	0.14	0.17	0.032
85	62.76	64.28	2.048	0.11	0.15	0.04
86	69.71	69.71	0	0	0	0
87	30.96	50.34	19.374	0	0	0