

ORIGINAL ARTICLE

Chromium and cobalt release from metallic earrings from the Danish market

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Abstract

Background: Chromium and cobalt are important skin sensitizers. It has, however, been difficult to identify causative exposures. Studies on nickel allergy have demonstrated piercing as critical for both sensitization and elicitation. It may be speculated that the same applies for chromium and cobalt.

Objective: To examine the content and release of chromium and cobalt from earrings randomly purchased in Denmark.

Methods: Three hundred four earrings were examined with x-ray fluorescence (XRF) spectrometry. Earrings with measured content of chromium or cobalt were spot tested with diphenylcarbazide spot test (n = 166) or Nitroso-R spot-test (n = 99), respectively. Chromium and cobalt release were quantified in a selected subsample (n = 100) with the artificial sweat test (EN 1811).

Results: Chromium was present in 54.6% (166/304) of earrings and cobalt was present in 72.0% (219/304),— measured by XRF. All chromium spot tests for chromium VI were negative. The cobalt spot test was positive for one component. Chromium release was found from 59/100 (median concentration = $-0.06 \mu\text{g}/\text{cm}^2/\text{week}$) and cobalt release from 29/100 (median concentration = $-0.06 \mu\text{g}/\text{cm}^2/\text{week}$) of earrings in tested subsample.

Conclusion: Earrings for piercing release chromium and cobalt and may on a case basis be a source of chromium and cobalt allergy.

KEYWORDS

chromium, cobalt, contact allergy, diphenylcarbazide, jewelry, Nitroso-R, spot test

1 | INTRODUCTION

Chromium and cobalt are important skin sensitizers. Contact allergy to chromium and cobalt are relatively common affecting, respectively, 0.8%–1.8% and 2.2%–2.7% of the general population.^{1,2} They are even more frequently reported in patients with dermatitis, with 3.7% positive for chromium and 5.3% positive for cobalt of patients who were patch tested with the European baseline series.³

Causative exposures for chromium allergy have been described primarily as exposure to leather objects and previously cement-work.⁴ For

cobalt allergy it has been difficult to identify relevant exposures.^{3,5} However, both chromium and cobalt are present in many common metal alloys and recent studies have shown release of these metal-ions from the metallic parts in various consumer items posing a potential allergy risk.^{6–12} Only a few of these studies have included body piercings.

Two colorimetric spot tests have been developed based on the same principle as the widely used dimethylglyoxime (DMG) spot test for screening of nickel release.¹³ Diphenylcarbazide (DPC) can detect hexavalent chromium (Cr[VI]) release,⁷ and disodium-1-nitroso-2-naphthol-3,6-disulfonate (Nitroso-R salt) cobalt release.^{14,15} Both of

the tests have successfully been used to show release from consumer items.^{6,7,12,16} However, their sensitivity and specificity have been questioned.^{17–19}

The purpose of this study was to evaluate chromium and cobalt content and release from earrings available on the Danish market and to compare the outcome of the DPC and Nitroso-R spot test to the quantitative measurement of metal released in artificial sweat according to EN 1811.²⁰

Additionally, the measured levels of release will be discussed in context of what is currently known about induction and elicitation levels for chromium and cobalt, in an attempt to elucidate the risk.

2 | METHODS

The same earrings from a previously published study on nickel release²¹ were tested for chromium and cobalt content and release. Of the earrings bought as an identical pair, we sought to subject the same earring to the various tests. Tests were done in the following order: x-ray fluorescence (XRF), spot-test, artificial sweat test.

2.1 | Materials

Three hundred four earrings were bought from March to May 2020 from a wide variety of different stores in Denmark. Only unique earrings with a metal perforating post were bought. Approximately 3300 EUR were spent, ranging from 2.5 to 50 EUR (mean = 12.9) per earring. For further details see published article.²¹

2.2 | XRF spectroscopy

The elemental composition of the earrings was measured by a handheld XRF device (X-MET8000 Series, Udem, Germany). The post from all 304 samples was analyzed, and measurements were only done when the lock ($n = 37$), the decorative part with skin contact ($n = 18$), and/or decorative part without skin contact (dangle charm) ($n = 33$) visibly consisted of metal.

Each measurement was taken on the factory default settings for alloy measurements with 10 seconds exposure time as per manufacturer's recommendation. Every sample measurement was done in triplets where the mean is presented as the result in weight percentage (wt%).

The detection limit of the XRF varies depending on the alloy tested and the element recorded. For stainless steel grades, the detection limit for chromium is 25 ppm (0.0025%) and 130 ppm (0.0130%) for cobalt with an average error of, respectively, 0.24% and 0.11%.

2.3 | Spot tests: DPC and NITROSO-R

The elemental composition measured by XRF was used to determine objects for spot testing.

All samples with chromium content in one or more components were tested with the DPC spot test ($n = 166$). Because very low levels of cobalt were detected in the majority of the samples, all samples with cobalt content above 0.1% ($n = 79$) in one or more components were subjected to Nitroso-R spot test. Furthermore, a random selection of samples that contained 0.002%–0.1% cobalt in one component was spot tested ($n = 20$) (Figure 1).

Several days passed between each type of spot-testing to allow re-oxidation of surface metal ions. If a test was positive, the sample was washed with de-ionized water and dried with a paper tissue to avoid cross-discoloration between the tests.²²

Both spot tests were prepared and performed in our laboratory at the Department of Dermatology and Allergy according to previously published methods.^{7,14,15}

For the DPC spot test for Cr(VI), 0.4 g of 1,5-DPC (CAS no. 140-22-7) (Merck KGaA, Darmstadt, Germany) was dissolved in a mixture of 20 mL of acetone (Merck KGaA, Darmstadt, Germany) and 20 mL of 96% ethanol. Then 20 mL 75% H_3PO_4 (Merck KGaA, Darmstadt, Germany) and 20 mL of de-ionized water were added. A cotton swab was soaked in the solution and rubbed on the test area for 30 seconds, and reading was performed after 2 minutes. A positive result was indicated by purple/red coloration of the swab and a negative result was indicated by no change in color.

For the Nitroso-R spot test for cobalt, 0.008 g disodium-1-nitroso-2-naphthol-3,6-disulfonate (Nitroso R salt) (CAS no. 32588-53-7) (BocSciences, Hamburg, Germany), 0.04 g oxalic acid (Merck KGaA, Darmstadt, Germany), and 2 g sodium acetate (Merck KGaA, Darmstadt, Germany) were dissolved in 40 mL de-ionized water. A cotton swab was dipped in the solution and rubbed against the test area for 30 seconds. A positive result was indicated by yellow/red coloration of the swab and a negative result was indicated by no change in color.

All doubtful tests, defined by a discoloration other than a hue of the positive color indication, were retested. If the reaction remained doubtful the result was registered as negative.

2.4 | Metal release in artificial sweat

A subsample of 100 earrings was subjected to metal release testing in artificial sweat according to the current European standard reference test EN1811 for control of compliance with the EU REACH nickel regulation.^{20,23,24} The subsample was chosen with a primary analytical focus on nickel and defined mainly by the nickel content of earrings.²¹ Included in this sample were, 75 of 166 (45%) earrings with chromium content as measured by the XRF and 85 of 219 (39%) earrings with cobalt content; several earrings had content of both metals (Figure 1). The test was performed by Eurofins | BLC Leather Technology Centre Ltd. (Kings Park Road, Moulton Park, Northampton, NN3 6JD, UK) according to BS EN 1811:2011 + A1: 2015,²⁰ with additional measurements of chromium and cobalt release by inductively coupled plasma mass spectrometry (ICP-MS). The resulting release was divided with the measured surface area of the component and reported as $\mu\text{g}/\text{cm}^2/\text{week}$. The lower limit of detection (LOD) was $0.02 \mu\text{g}/\text{cm}^2/\text{week}$.

The chromium analysis did not distinguish between states of chromium (III and VI) and thus represents total chromium.

3 | RESULTS

3.1 | XRF spectroscopy

Chromium was present in 54.6% ($n = 166$) of the 304 earrings in one or more components. Regarding tested components 53.3% posts (162/304, content median = 11.8%), 24.3% locks (9/37, content median = 0.04%), and 51% decorative parts (26/51, content median = 0.4%) had chromium content.

Cobalt was present in 72% ($n = 219$) of the 304 earrings in one or more components.

Regarding tested components 65.5% posts (199/304, content median = 0.07%), 75.7% locks (28/37, content median = 0.06%), and 84.3% decorative parts (43/51, content median = 0.02%) had cobalt content.

3.2 | DPC and NITROSO-R spot tests

All 166 earrings that contained chromium were negative with the DPC spot test.

Of the 99 tested earrings, one lock component of an earring was found positive for cobalt release with the Nitroso-R spot test. Notably, only one lock of an identical earring pair was positive (Figures 1 and 2).

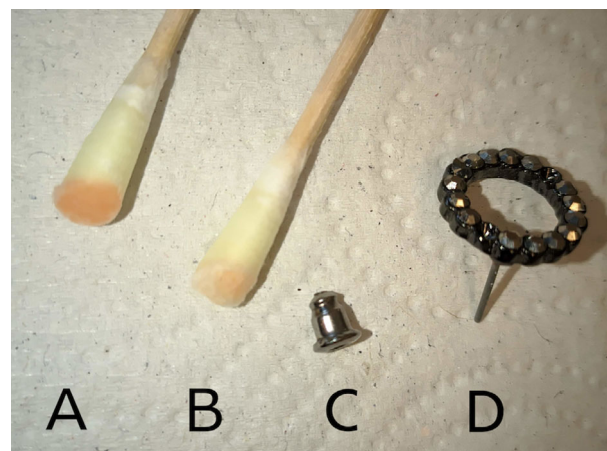
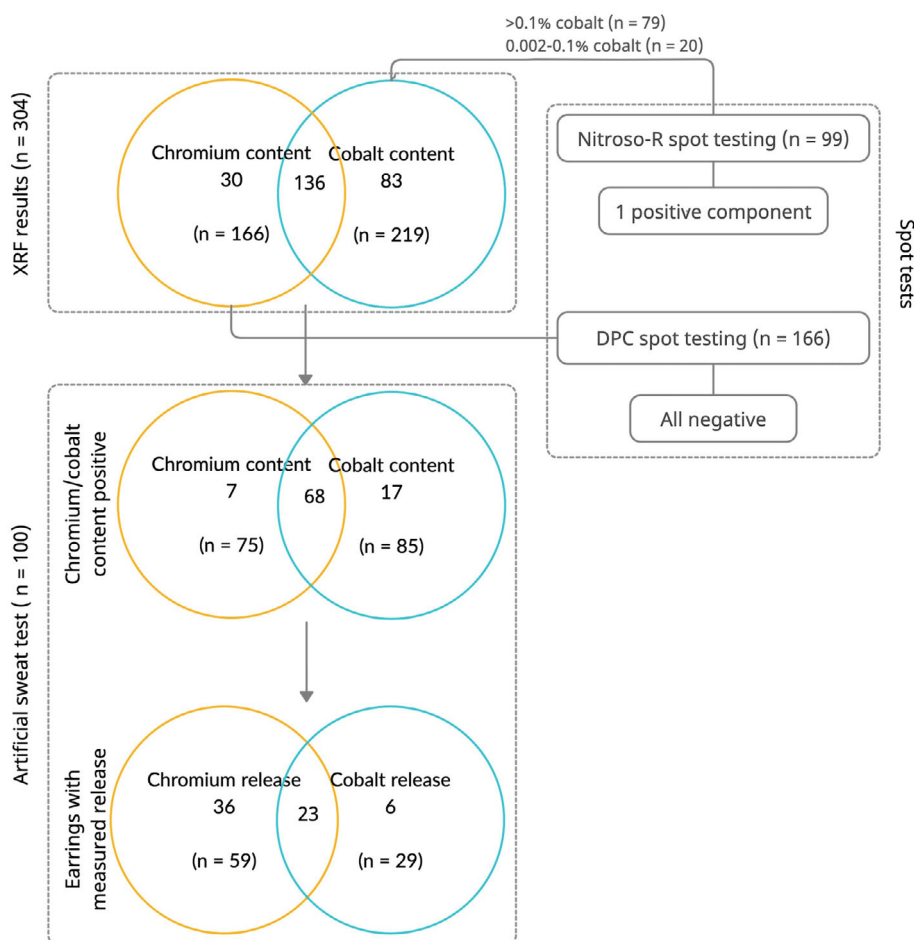


FIGURE 2 Positive Nitroso-R spot test of the lock component of an earring. A, Positive control. B, Positive test. C, Positive lock component. D, The earring post and the decorative part tested negative

FIGURE 1 Summary of study methods and results. Top circles: Earrings positive for chromium and/or cobalt content as measured by the x-ray fluorescence (XRF) analysis of all 304 earrings. All chromium-positive earrings were diphenylcarbazine (DPC) spot tested. A subsample of cobalt positive earrings was Nitroso-R spot tested. Middle circles: Distribution of chromium and/or cobalt content positive earrings in a subsample of 100 earrings chosen for artificial sweat testing (eight earrings did not have any measured chromium or cobalt content). Bottom circles: Earrings positive for chromium and/or cobalt release as measured by the artificial sweat test



	N	Median	Mean	Minimum	Maximum
Post	43	0.06	10.88	0.02	110
95% CI		0.04-0.12	4.33-18.33		
Lock	15	0.06	1.07	0.02	14.00
95% CI		0.03-0.27	0.07-2.96		
Decorative part	10	0.49	6.58	0.02	51.00
95% CI		0.03-4.50	0.58-16.77		
Dangle charm	15	0.07	0.12	0.02	0.88
95% CI		0.04-0.11	0.06-0.24		
Total positive components	83				

TABLE 1 Chromium release from earring components measured by the artificial sweat test

Note: Values are reported in $\mu\text{g}/\text{cm}^2/\text{week}$. A total of 59 earrings were positive in one or more components. Lower limit of detection = $0.02 \mu\text{g}/\text{cm}^2/\text{week}$.

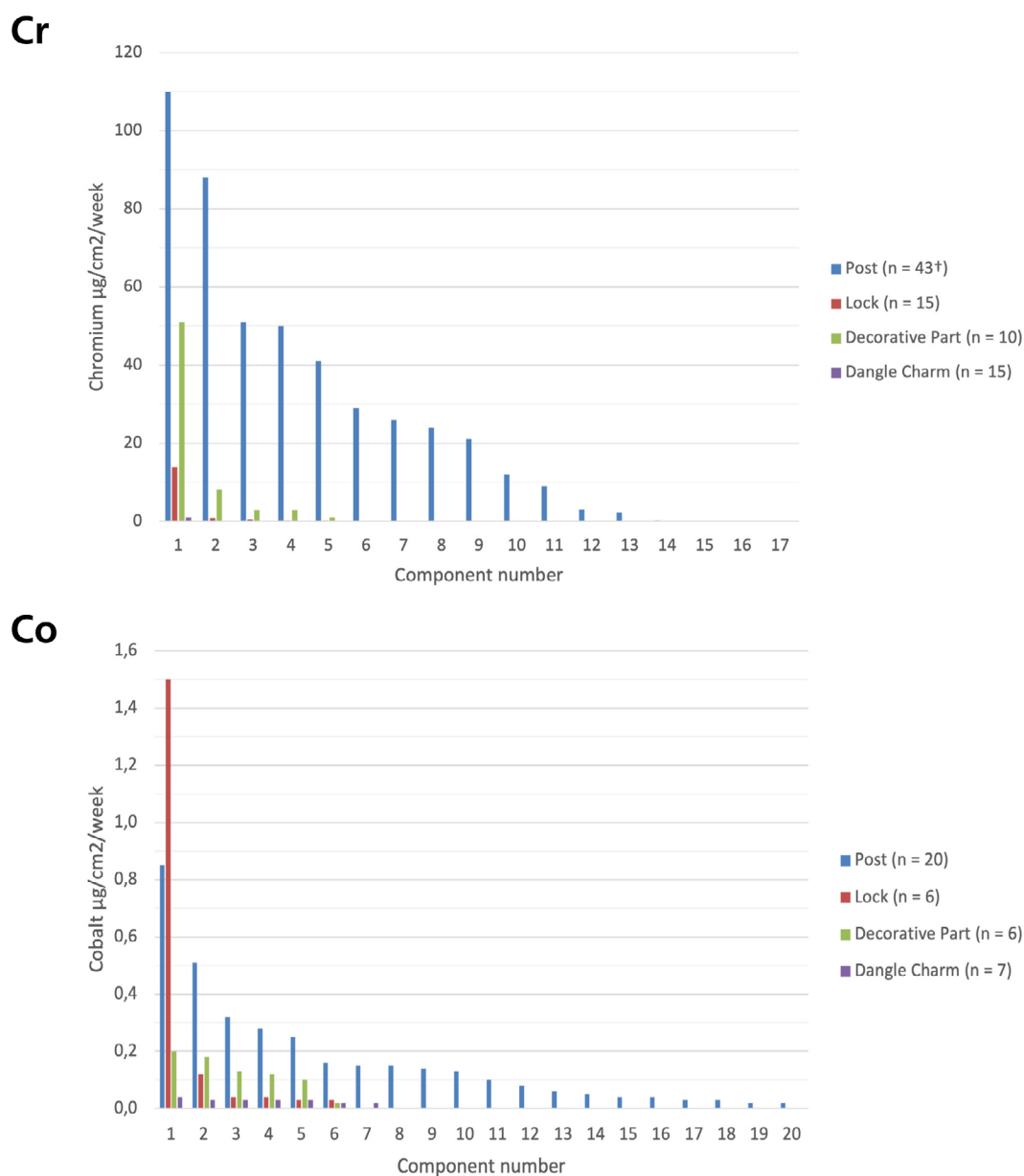


FIGURE 3 Chromium and cobalt release from earring components measured by artificial sweat test. Each bar represents one component. (†Post components with chromium release $<0.1 \mu\text{g}/\text{cm}^2/\text{week}$ are not shown)

TABLE 2 Cobalt release from earring components as measured by the artificial sweat test

	N	Median	Mean	Minimum	Maximum
Post	20	0.12	0.17	0.02	0.85
95% CI		0.05-0.16	0.09-0.27		
Lock	6	0.04	0.29	0.02	1.50
95% CI		0.03-0.81	0.04-0.78		
Decorative part	6	0.13	0.13	0.02	0.20
95% CI		0.06-0.19	0.08-0.17		
Dangle charm	7	0.03	0.03	0.02	0.04
95% CI		0.02-0.03	0.02-0.03		
Total positive components	39				

Note: Values are reported in $\mu\text{g}/\text{cm}^2/\text{week}$. A total of 29 earrings were positive in one or more components. Lower limit of detection = $0.02 \mu\text{g}/\text{cm}^2/\text{week}$.

3.3 | Chromium and cobalt release in artificial sweat

Chromium was released from 59 of the tested earrings (59/100); release from more than two components were found from 20 earrings. A total of 83 components were positive with a median of $0.06 \mu\text{g}/\text{cm}^2/\text{week}$ (0.04-0.11, 95% confidence interval [CI]) with some high outliers. The majority of positive components were posts ($n = 43$), which also had the highest release value ($110 \mu\text{g}/\text{cm}^2/\text{week}$) (Table 1, Figure 3).

Cobalt release was found from 29 (29/100) earrings in one or more components, of which 10 had several positive components. A total of 39 components were positive, with a median of $0.06 \mu\text{g}/\text{cm}^2/\text{week}$ (0.04-0.13, 95% CI). The majority of positive components were posts ($n = 20$); however, a single lock was a high outlier with a value of $1.5 \mu\text{g}/\text{cm}^2/\text{week}$ (Table 2, Figure 3).

A Spearman bivariate correlation analysis showed a moderate correlation between released chromium and cobalt ($r = 0.48$, $P < .01$). There was no significant correlation between either of the two metals and nickel released from the same samples.

4 | DISCUSSION

4.1 | Chromium and cobalt release

In this study, we found that both chromium and cobalt were widely present and often released from earrings on the Danish market.

There have been few published studies assessing the release of chromium or cobalt from earrings and piercing jewelry,^{8,9,11,18,25-27} of which only four (including a follow-up) have performed quantitative measurements.^{8,11,25,27}

With several high outliers, we found a median release of $0.06 \mu\text{g}$ chromium/ cm^2/week ($n = 59/100$) and $0.06 \mu\text{g}$ cobalt/ cm^2/week ($n = 29/100$). In comparison, a Turkish study in 2020 found a mean release of $2.04 \mu\text{g}$ chromium/ cm^2/week (standard deviation [SD] = 4.68) and a mean release of $0.20 \mu\text{g}$ cobalt/ cm^2/week (SD = 2.73) from a sample of 223 cheap earrings.¹¹ The authors did not note how many samples were positive for release.

Our findings of cobalt release are slightly higher than reported in a similar German survey in 2008 (median = $0.013 \mu\text{g}/\text{cm}^2/\text{week}$).⁸ A 2014 follow-up of the survey notes a decline in the levels of cobalt released, however, with no decrease in the number of cobalt releasing earrings.²⁵ In addition, Hamann et al found cobalt release from earrings bought in the United States in levels ranging 0.02 - $0.05 \mu\text{g}/\text{cm}^2/\text{week}$ ($n = 35/96$).²⁷

Of interest, we report a modest correlation in the release of chromium and cobalt but not with the release of nickel, a finding also described in a similar study.¹¹ This is likely due to the alloy composition; however, we are not able to further elaborate with the current data.

Cobalt is frequently seen as a contaminant in various nickel-containing alloys, as cobalt occurs naturally in nickel ores and exists as a trace element in nickel ore extract.²⁸ Nickel was also a common constituent²¹ and might explain some of the very low levels of cobalt content measured by the XRF.

4.2 | Induction and elicitation risk

The sensitization reference dose of chromium for safe human exposure has been calculated to $0.01 \mu\text{g}/\text{cm}^2$, an approximation based on data from animal studies.²⁹ More recent investigations based on patch testing estimated the minimum threshold to elicit dermatitis in 10% (MET₁₀) of sensitized individuals to $0.18 \mu\text{g}/\text{cm}^2$ for Cr(III) and $0.03 \mu\text{g}/\text{cm}^2$ for Cr(VI).³⁰

Our results of chromium release do not distinguish between oxidation states. The conversion between Cr(III) and Cr(VI) is dynamic and depends on a range of environmental factors, such as humidity, pH, and temperature.³¹ From stainless steel, chromium is released primarily as Cr(III) due to the presence of iron as a reductive agent, but other alloys may be more prone to oxidation and the formation of Cr(VI).³² However, once chromium is in contact with or has penetrated the skin barrier, it may be that interactions with immune cells and proteins such as reactive oxygen species may facilitate the conversion to the more potent Cr(VI).^{31,33}

In our study, all chromium positive components (59/100) surpassed $0.02 \mu\text{g}/\text{cm}^2/\text{week}$ (as this was the LOD of the EN 1811);

however, the exact deposition of chromium on the skin under real-life use of earrings is not known.

Cobalt is a strong skin sensitizer, and the dose to elicit dermatitis in 10% (ED₁₀) of sensitized individuals has been estimated to 0.06–1.95 µg/cm².³⁴ In our study, 29% of the subsampled earrings released cobalt with a median of 0.06 µg/cm²/week, making it likely that several earrings may release critical levels of cobalt.

There are no specific studies or approximations on the induction levels for cobalt sensitization, but it is established as a stronger skin sensitizer than chromium or nickel,^{35,36} and safe levels must thus be assumed lower (current European legislative limits nickel in piercing post assemblies to 0.02 µg/cm²/week^{23,24}).

The MET₁₀ and ED₁₀ have been derived from dose-response patch testing on intact skin. From studies of nickel, it is indicated that the elicitation threshold at piercing exposure is lower than at an occluded exposure such as the patch testing.³⁷ Furthermore the piercing exposure has been associated with a higher risk of allergy to nickel.^{38,39} This stresses the potential risk of elicitation from chromium and cobalt released from earrings. Release of chromium and cobalt in our study was seen primarily from the post component, which is in direct contact with the pierced skin canal.

Warshaw et al analyzed the correlation between body piercings and chromium and cobalt allergy and found no association with cobalt allergy alone and a negative correlation with chromium allergy.³⁸ It should be noted, however, that the negative correlation was not significant when adjusting for gender, as few pierced males (4.4%) were included in the study, whereas the male group simultaneously had a higher prevalence of chromium contact allergy.

Nevertheless, along with leather, jewelry has been recorded as the most common source of exposure to cobalt in dermatitis patients.⁵ Our results indicate that on a case basis both chromium and cobalt release from earrings may be of clinical significance.

4.3 | Spot tests

In our spot testing, all samples were negative in the DPC test for chromium release and only one sample (0.3%) was positive in the Nitroso-R test for cobalt.

The DPC spot test only detects Cr(VI) with a detection limit estimated to 0.25 ppm Cr(VI).⁷ We were not able to differentiate between the oxidation states in the artificial sweat test. It is likely that the measured levels of chromium were primarily of Cr(III) and thus not detectable by the DPC spot test; however, the sensitivity and possibility of false negatives of the DPC spot test have not been fully validated to draw this conclusion.⁷

Studies have shown that primarily Cr(III) is released from specific types of biomedical applied steel; however, it is strongly dependent on the environment and alloy production.^{31,33,40,41}

Bregnbak et al¹⁸ did a similar study of DPC spot testing 848 items of jewelry, of which 707 were body piercings. All 848 items tested negative. There was no investigation of chromium release by any other methods.

The detection limit of Nitroso-R is estimated to 8.3 ppm cobalt.¹⁴ The estimated ED₁₀ range of 0.06–1.95 µg/cm² corresponds to 30.8–259 ppm.³⁴ The lock component that was Nitroso-R spot-test positive released 1.5 µg cobalt/cm²/week, which was the component with the highest measured cobalt release. Cobalt release of LOD–0.85 µg/cm²/week were measured for other components. This indicates the usefulness of the Nitroso-R spot test for screening objects for cobalt release of certain clinical relevance.

In 2010, Thyssen et al⁹ performed a Danish cobalt screening survey of 354 jewelry and hair clasps, of which 170 were earrings; 3 earrings (1.8%) released cobalt, found by Nitroso-R spot test. Comparably, we found one (0.3%) Nitroso-R positive earring.

5 | CONCLUSION

In conclusion, earrings for piercing may on a case basis be an overlooked source of chromium or cobalt allergy. The cobalt spot test Nitroso-R seems to be able to detect exposures with some clinical relevance. A spot test for, or including, Cr(III) would be of value.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

AUTHOR CONTRIBUTIONS

Michael Wennervaldt: Conceptualization; data curation; formal analysis; funding acquisition; investigation; methodology; project administration; visualization; writing-original draft; writing-review & editing. **Malin Ahlström:** Conceptualization; formal analysis; funding acquisition; investigation; methodology; supervision; validation; writing-review & editing. **Torkil Menné:** Conceptualization; investigation; methodology; supervision; validation; writing-review & editing. **Morten Haulrig:** Formal analysis; investigation; methodology; writing-review & editing. **Farzad Alinaghi:** Formal analysis; investigation; validation; writing-review & editing. **Jacob Thyssen:** Conceptualization; formal analysis; methodology; project administration; supervision; validation; writing-review & editing. **Jeanne Duus Johansen:** Conceptualization; formal analysis; funding acquisition; methodology; project administration; supervision; validation; writing-review & editing.

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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