

Chromate and amine contact allergies in workers manufacturing precast concrete elements

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Summary

Background. Five workers from a plant manufacturing concrete wall panels and beams were referred to our department because of suspected occupational dermatitis. When patch tested, 3 workers reacted to potassium dichromate. Four workers reacted to ethylenediamine dihydrochloride, without any obvious exposure. Owing to the high proportion of workers with recent-onset skin disease, an investigation of all workers at the plant was initiated.

Objectives. To investigate the prevalence of occupational dermatitis and contact allergy in the workers at the plant.

Methods. All 24 workers at the plant underwent a clinical investigation and were patch tested.

Results. Four cases of allergic occupational contact dermatitis and 3 cases of irritant occupational contact dermatitis were diagnosed. Contact allergy to potassium dichromate was found in 4 workers. All 4 also reacted to ethylenediamine dihydrochloride and/or amines that were present as additives in the cement.

Conclusions. Chromate contact allergy can still be found in concrete workers, despite the legislation regulating the amount of hexavalent chromium (chromate) in cement. Occupational contact allergy to amines can be found in workers exposed to cement and concrete, so amines should be tested in these workers.

Key words: allergic contact dermatitis; amines; cement; chromium; concrete; ethylenediamine; irritant contact dermatitis; occupational.

Within a 3-year period, 5 workers from a plant producing concrete wall panels and beams were referred to our department with suspected work-related skin diseases. When patch tested with our baseline series, 3 of the 5 workers reacted to potassium dichromate. Legislation stating that the water-soluble hexavalent chromium (chromate) content in dry cement must not exceed 2 mg/kg was passed in Sweden in 1989, so it could be

assumed that cement with a reduced chromate content had been used at the plant for several years. Surprisingly, 4 of 5 workers reacted to ethylenediamine dihydrochloride in our baseline series, without any obvious previous exposure. Owing to the high proportion of employees with work-related skin disease of recent onset at this plant, a study on occupational skin disease was performed in collaboration with the management at the plant.

Materials and Methods

The plant manufactures concrete wall panels and beams. Concrete is produced by mixing cement with water, ballast material and additives in an automated process. When the concrete products are casted, the casting moulds are first treated with mould release oil. After

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the concrete has been applied to the casting mould, the surface is smoothed manually. This work is performed while the concrete is still wet, and the workers have no protection other than textile clothes, leather/textile gloves, and shoes.

A worksite visit was performed when 1 of the 5 original patients was investigated, in order to determine how the workers were exposed to wet concrete and other products at the plant. Another worksite visit was performed prior to the study, in order to collect material to be used for patch testing. At the time of the investigation, the plant had a total of 24 workers (all male). The mean age of the workers was 42 years (range 21–64 years), and the mean duration of employment was 7.5 years (range 1–31 years).

After an oral presentation of the study at the plant, a questionnaire was handed out to all workers. The questionnaire included questions regarding duration of employment, work tasks, previous employment, and past and present skin diseases.

All employees at the plant were offered a consultation, including an interview, clinical investigation, and patch testing. The consultation and patch testing were performed at our department. The study was approved by the Regional Ethical Review Board in Lund, Sweden. Written informed consent was obtained from each participant.

Patch testing was performed with our baseline series supplemented with preservatives, metals, textile dyes, and plastics chemicals, and a series consisting of preparations of work material and workplace-related allergens. All test preparations were purchased from Chemotechnique Diagnostics (Vellinge, Sweden), unless otherwise stated. Test preparations made from chemicals available at our laboratory and from products collected at the plant were prepared at our laboratory in water, acidic buffer solution (1) or pet. (Snow White Quality E; Apoteket Produktion & Laboratorier, Göteborg, Sweden).

Because we had found unexplained contact allergies to ethylenediamine, we contacted the manufacturer of the cement concerning possible additives. We were informed that a mixture of amines called Berolamine 20 (Akzo Nobel Functional Chemicals, Stenungsund, Sweden) was added to the cement in small amounts [0.03–0.05% (wt/wt) of the dry cement] as a grinding aid during the production process. Prior to the study, 19 controls were tested with a 1% preparation of Berolamine 20 in pet. Among these, 18 had negative results and 1 had a doubtful reaction. The work environment series included Berolamine 20 and four amines present in Berolamine 20: aminoethyl ethanolamine, diethanolamine (both from Sigma-Aldrich, Steinheim, Germany), tetraethylenepentamine (Akzo Nobel Functional Chemicals), and triethylenetetramine. Furthermore, the work

Table 1. The work environment series and number of employees reacting to the test preparations

Substance/product	Concentration	No. positive
Potassium dichromate	0.5% aq.	3
Berolamine 20	1% pet.	3
Berolamine 20	0.1% pet.	–
Aminoethyl ethanolamine*	1% acidic buffer	–
Diethanolamine*	1% acidic buffer	–
Tetraethylenepentamine*	0.5% pet.	–
Triethylenetetramine*	0.5% pet.	1
Abrasive soap Ekofekt Grovtvål	5% aq.	–
Abrasive soap Ekofekt Grovtvål	0.5% aq.	–
Soap Ekofekt Kombitvål	10% aq.	3
Soap Ekofekt Kombitvål	1% aq.	–
Cocamide DEA†	0.5% pet.	–
Cocamidopropyl betaine†	1% aq.	–
Propylene glycol†	5% pet.	–
Grease from bolt‡	10% pet.	–
Grease from bolt‡	1% pet.	–
Mould release oil Differol T08	50% pet.	–
Mould release oil Differol T08	5% pet.	–
Hot-melt adhesive‡	'as is'	–
Styrofoam insulation‡	'as is'	–
Superplasticizer Sikament 20HE	10% aq.	–
Superplasticizer Sikament 20HE	1% aq.	–
Leather from glove Active Wear	'as is'	–
Fabric from glove Active Wear	'as is'	–

*Amines present in Berolamine 20.

†Ingredients in the soaps.

‡Unknown brand.

environment series included an aqueous solution of potassium dichromate (Janssen Chimica, Geel, Belgium), the mould release oil, grease from the bolts used in building of the moulds, a hot-melt adhesive, a superplasticizer additive, protective gloves (both the leather and textile parts tested separately), two liquid soaps, and selected components of the soaps that were available at our laboratory (Table 1). Finn Chambers® (Smart Practice, Phoenix, AZ, USA) were used for the patch tests. According to recommendations, 20 mg of each pet. preparation was applied in the test chambers (2). For patch testing with solutions, 15 µl was micropipetted onto the filter papers in the test chambers (3). The patch tests were removed by the participants after 48 h, and readings were performed on day (D) 3 and D7, according to ICDRG criteria (4).

Subjects with positive or doubtful reactions to potassium dichromate 0.50% pet. or aq. at D3 were also tested with a dilution series of potassium dichromate (0.50%, 0.16%, 0.050%, 0.016% and 0.0050% (wt/vol) aq.). Subjects with positive reactions to any of the mixes in the baseline series were additionally tested with the individual components of the mix. Subjects with doubtful reactions to patch test preparations in the work

environment series were retested with the preparation at the originally used concentration and also at a lower concentration.

Skin diseases were regarded as work-related if they had begun or worsened after employment at the plant, and improved when the worker was off work. When contact allergies to substances/products that the workers were exposed to in their work environment were found, the dermatitis was regarded as allergic occupational contact dermatitis. When no work-related contact allergies could be diagnosed, the dermatitis was regarded as irritant occupational contact dermatitis.

Samples of cement used in the manufacturing of wall panels and beams, and the Berolamine 20 sample, were analysed for hexavalent chromium by use of a spot test method with *sym*-diphenylcarbazine (Acros Organics, Geel, Belgium) as a reagent (5).

Results

The questionnaire was answered by all 24 workers, and all workers also agreed to participate in the clinical investigation and to be patch tested. Eighteen of the workers had worked with cement only in Sweden. Eleven workers reported dermatitis during their present employment. The most common sites of dermatitis were the legs (7 workers), arms (6 workers), and hands (6 workers). In 7 of these, the dermatitis was regarded as work-related, according to the criteria specified above. The dermatitis was regarded as allergic occupational contact dermatitis in 4 workers and as irritant occupational contact dermatitis in 3 workers. On clinical examination, 11 of 24 workers showed current skin disease. The dermatitis was localized on the arms in 6 workers, the hands in 5 workers, the legs in 6 workers, and on the feet in 1 worker (Table 2).

The results of patch testing are shown Tables 1–4. Eleven workers reacted to at least one preparation in the patch test series. Four workers had positive reactions to potassium dichromate. Three of 4 had simultaneous positive reactions to Berolamine 20, and 1 had a doubtful reaction. One worker reacted to ethylenediamine dihydrochloride. Three workers reacted to a soap used at the plant, and 2 of these had doubtful reactions to cocamidopropyl betaine, which was one of the ingredients in the soap.

The water-soluble chromate content in the cement used for the wall panels was estimated to be 1 mg Cr/kg with the *sym*-diphenylcarbazine spot test. The same amount was found in the cement used for the beams. A higher level of chromate (~2 mg Cr/kg) was found in cement collected from an open sack of cement that was occasionally used for repair work. No chromate was

found in the Berolamine 20 sample analysed with the same method.

Discussion

Chromate in cement has long been a well-known cause of chromium contact allergy, often resulting in persistent eczema in workers handling cement. Legislation stating that the water-soluble hexavalent chromium (chromate) content in dry cement must not exceed 2 mg Cr/kg came into force in Sweden in 1989 and in the EU in 2005 (6). In order to obtain cements with such low chromate levels, iron sulfate is added to the cement to reduce the chromate to trivalent chromium compounds, which are almost completely insoluble in water under alkaline conditions (7).

The prevalence of chromate contact allergy is decreasing in Europe, probably as a result of the legislative measures regarding hexavalent chromium in cement (8), but cases of chromate allergy among concrete workers are still being observed (9). Whereas cement has been a historically important source of sensitization in Europe, it is currently thought that exposure to chromium-tanned leather is of greater importance (10, 11). Since 2015, the content of hexavalent chromium in leather articles that come into contact with the skin has been regulated in the EU. According to this regulation, the leather may not contain >3 mg/kg hexavalent chromium (12).

The reduction of chromate is a reversible process, and the trivalent chromium formed can, upon air exposure, be oxidized back to chromate. This has been observed, for example, when opened sacks of cement have been stored for a longer period of time (13). We found chromate levels below the threshold limit value stated in the legislation when cement used in the production of wall panels and beams was analysed. Higher chromate levels (~2 mg Cr/kg) were found in a sample of cement taken from an open sack used for repair work. However, the exposure to this cement was limited.

Chromate allergy was diagnosed in 4 of 24 workers. As a comparison, the frequency of positive reactions to potassium dichromate among consecutive patch tested male dermatitis patients at our unit between 2005 and 2014 was 3.1% (14). The duration of employment of the 4 chromate-positive workers ranged from 2 to 21 years. Three of these had only handled cement in Sweden, and 1 had handled cement in Bosnia, but not professionally. All 4 reported onset of their present skin problems during their employment at the plant. We regard the detected chromate contact allergy as most probably being caused by work with chromate-reduced cement. Many of the workers were heavily exposed to wet concrete, especially when manually smoothing the surfaces of the freshly

Table 2. Summary of questionnaire and clinical investigation

Case no.	Age (years)	Skin status according to questionnaire				Work tasks			Outcome of clinical investigation			
		Duration of employment (years)	Skin disease prior to present employment	Skin disease during present employment	Improved skin status during vacation	Mixing cement	Casting beams	Casting wall panels	Ongoing dermatitis/ localization	Contact allergies	Work-related skin disease	
1	57	8	–	X	X	–	X	X	Hands	Potassium dichromate Ethylenediamine dihydrochloride* Soap Ekofekt Kombitvål Thiuram mix	AOCD	
2	38	17	X	X	X	X	–	X	Hands	Potassium dichromate Berolamine 20	AOCD	
3	40	21	–	X	X	X	–	X	Forehead, wrists, arms, legs	Potassium dichromate Berolamine 20 Triethylenetetramine Ethylenediamine dihydrochloride	AOCD	
4	22	2	–	X	X	–	–	X	Legs, feet	Potassium dichromate Ethylenediamine dihydrochloride* Berolamine 20 Cobalt chloride	AOCD	
5	59	3	–	–	–	–	–	–	Hands, elbows	Nickel sulfate Palladium chloride 4,4'-Diaminodiphenyl-methane	–	
6	44	25	–	–	–	X	–	X	–	Neomycin sulfate Nickel sulfate	–	
7	64	9	–	–	–	X	–	–	–	Soap Ekofekt Kombitvål	–	
8	49	7	–	–	–	–	–	–	Arms, legs	Soap Ekofekt Kombitvål <i>Evernia prunastri</i>	–	
9	48	2	–	X	X	–	X	–	Arms, legs	–	IOCD	
10	27	3	–	X	X	–	X	–	Arms, legs	–	IOCD	
11	21	1	–	X	–	X	–	X	Hands	–	–	
12	62	31	–	–	–	X	X	–	–	–	–	
13	46	2	X	X	–	–	–	–	Elbows, legs	Fragrance mix II Quaternium-15	–	
14	36	1	–	X	–	–	–	X	Hands	–	–	
15	37	10	–	–	–	–	X	X	–	Fragrance mix II	–	
16	46	10	–	–	–	X	X	X	–	Aluminium chloride	–	
17	28	3	–	–	–	–	–	X	–	–	–	
18	34	2	–	–	–	–	–	X	–	–	–	
19	43	1	X	X	–	–	–	–	–	–	–	
20	41	2	–	X	X	X	X	X	–	–	IOCD	
21	25	1	–	–	–	X	–	–	–	–	–	
22	36	8	–	–	–	X	X	X	–	–	–	
23	47	8	–	–	–	–	–	–	–	–	–	
24	49	2	–	–	–	–	–	–	–	–	–	

AOCD, allergic occupational contact dermatitis; IOCD, irritant occupational contact dermatitis; X, yes. *Two workers did not react to ethylenediamine dihydrochloride when tested within the study, but had positive reactions to ethylenediamine dihydrochloride in the baseline series when investigated earlier at our department.

Table 3. Positive reactions to substances tested in the baseline series in 24 workers

Substance	Concentration (% in pet.)	No. positive
Potassium dichromate	0.5	4
Ethylenediamine dihydrochloride	1	3*
Nickel sulfate hexahydrate	5	2
Fragrance mix II	14	2
Thiuram mix	1	1
Neomycin sulfate	20	1
Cobalt chloride	0.5	1
<i>Evernia prunastri</i>	2	1
Palladium chloride	2	1
Quaternium-15	1	1
4,4'-Diaminodiphenylmethane	0.25	1
Aluminium chloride hexahydrate [†]	20	1

*Two workers did not react to ethylenediamine dihydrochloride when tested within the study, but had positive reactions to ethylenediamine dihydrochloride in the baseline series when investigated earlier at our department.

[†]From MP Biomedicals, Santa Ana, CA, USA.

casted wall panels. When performing this task, they were crawling on the wall panels, and both their gloves and trousers became wet. In addition, the workers used gloves partly made of leather, as well as leather shoes. The study was performed before the EU regulation on chromate in leather articles came into force. Thus, chromium-tanned leather may have been an additional exposure source, especially as the release of chromate is increased in an alkaline environment (15, 16). However, the dermatitis of the chromate-sensitized workers was not exclusively located on the hands or feet.

Three of the 4 chromate-allergic workers showed simultaneous reactions to Berolamine 20, and 1 had a doubtful reaction. Two of 3 workers who reacted to Berolamine 20 and 1 worker with a doubtful reaction to Berolamine 20 also reacted to ethylenediamine dihydrochloride, either when tested within the study or when tested earlier at our department. In addition, 1 worker who had been investigated at our department earlier, but had stopped working at the plant when the study was performed, had a ++ reaction to ethylenediamine dihydrochloride. Ethylene diamine has been used as a stabilizer in topical steroid creams (17, 18), but is no longer used for this purpose in Sweden. Occupational cases of ethylenediamine contact allergy have been reported among workers handling epoxy systems, in which ethylenediamine and related amines are used as curing agents (17, 19), and in workers exposed to ethylenediamine-containing coolant oils and synthetic coolants (20, 21). Furthermore, allergic reactions to ethylenediamine have been reported in healthcare workers handling theophylline and aminophylline

(22, 23). No history of exposure to ethylenediamine could be found in the ethylenediamine-allergic workers. According to the safety data sheet of Berolamine 20, it does not contain ethylenediamine. It is therefore likely that the reactions to ethylenediamine dihydrochloride are attributable to cross-reactivity to the amines in Berolamine 20, which have similar structures. Cross-reactivity between ethylenediamine and other aliphatic amines such as diethylenetriamine and triethylenetetramine has previously been reported (18, 24, 25). The unexpected reactions to ethylenediamine dihydrochloride in 4 of 5 patients from the plant investigated prior to the study had made us inquire about amine-related additives in the cement. If these reactions had not been observed, we would not have learned about the addition of Berolamine 20 to the cement. It is remarkable that the exposure to the low concentrations of Berolamine 20 in cement seems to be the source of sensitization. However, the workers reacting to Berolamine 20 all had a damaged skin barrier, which may facilitate the penetration of the components of Berolamine 20. As all workers who reacted to Berolamine 20 also reacted to potassium dichromate, the Berolamine 20 sample was analysed for hexavalent chromium, but none was found. It may be speculated that the amines present as additives in the cement are able to form complexes with metal ions, such as chromate ions, which could facilitate the skin penetration of chromate ions through the skin.

One worker who reacted positively to potassium dichromate and ethylenediamine dihydrochloride when patch tested before the study now also reacted to thiuram mix. According to his record from his first patch test, he had used underarm protection made of rubber, and had experienced itching where it was in contact with the skin; thus, the thiuram allergy may be occupationally acquired. One worker who did not react to ethylenediamine dihydrochloride or to Berolamine 20 reacted to 4,4'-diaminodiphenylmethane (MDA), which was included in our extended baseline series. He denied any present exposure to MDA, but he may have been exposed to products containing MDA or its corresponding isocyanate, 4,4'-diphenylmethane diisocyanate, in his previous employment as a construction worker.

Three workers had + reactions to a 10% aq. solution of a soap used at the plant. The soap was also tested at 1%; 2 of the workers showed doubtful reactions to this preparation, and the third had a negative result. Two of the workers reacting to the soap also had doubtful reactions to cocamidopropyl betatine, which was one of the ingredients in the soap. After the study had been performed, 19 controls were patch tested with a 10% solution of the soap. One of the controls had a

Table 4. Work-related contact allergies in 6 workers

Test preparation	Concentration	Case no.					
		1	2	3	4	7	8
Potassium dichromate	0.50% pet.	++	++	++	++	(+)	–
	0.50% aq.	++	++	–	++	–	–
	0.16% aq.	++	(+)	–	++	–	–
	0.050% aq.	–	–	–	(+)	–	–
	≤ 0.016% aq.	–	–	–	–	–	–
Ethylenediamine dihydrochloride	1.0% pet.	+	–	++	++*	–	–
Berolamine 20	1.0% pet.	(+)	++	++	++	–	–
	0.10% pet.	–	–	(+)	–	–	–
Triethylenetetramine	0.50% pet.	–	–	++	(+)	–	–
Tetraethylenepentamine	0.50% pet.	–	–	(+)	–	–	–
Soap Ekofekt Kombitvål	10% aq.	+	(+)	–	–	+	+
	1.0% aq.	(+)	–	–	–	(+)	–
Cocamidopropyl betaine	1.0% aq.	(+)	–	–	–	–	(+)
Thiuram mix	1.0% pet.	++	–	–	–	–	–

*Two workers did not react to ethylenediamine dihydrochloride when tested within the study, but had positive reactions to ethylenediamine dihydrochloride in the baseline series when investigated earlier at our department.

morphologically positive reaction, and two had doubtful reactions. Thus, it cannot be ruled out that the observed weak positive reactions to the soap were false-positive irritant reactions. Nevertheless, we recommended that the soap, which also contained fragrance, should be replaced with a fragrance-free alternative.

In general, the workers at the plant were poorly protected, and thus exposed to both irritants and allergens. For example, they used gloves made of fabric and leather, which, in some cases, became soaked with wet concrete. At one worksite visit performed prior to the study, a pH value of ~11 was measured on the inside of a glove with a pH indicator paper, showing that the hands were exposed to an alkaline environment under occlusion of

the gloves. We advised the staff at the plant to use better protective clothes and gloves, for example to use rubber gloves instead of textile/leather gloves. If better protective equipment had been used earlier, we believe that the work-related skin diseases could have been avoided, at least in some of the workers.

This study shows that chromate contact allergy still can be found in concrete workers, despite the legislation regulating the amount of water-soluble hexavalent chromium in cement. Furthermore, it shows that occupational contact allergy to amines can be found in workers exposed to cement and concrete. When occupational contact allergy is suspected in these workers, amines should be tested.

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