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The identification, approaches to cleaning and removal of a lead-rich salt crust from the surface of an 18th century oil painting

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Abstract

A thin, grey surface layer was noticed after removing the varnish of an eighteenth century Dutch oil painting, by an unknown artist titled *Vision of Saint Lutgard of Tongeren*. This layer masked the paint layer almost entirely, casting a greyish veil that greatly influenced the painting's appearance, colour, and saturation. Using scanning electron microscopy coupled with elemental analysis (SEM–EDX), this layer was identified as a lead-rich salt crust. The crust was a distinct layer on top of the paint layer, closely following the topography of the layer underneath but not embedded in it. The article discusses the possible theories behind the formation of this lead-rich salt crust by characterizing the painting's material, past treatments, and environmental history. Furthermore, the article reports the decision-making process behind the removal of the crust, focusing specifically on the ethical and technical challenges of the cleaning treatment. Tests with different cleaning agents targeting separate components of the crust to remove it and different application techniques are described. The chelating agent Ethylenediaminetetraacetic acid (EDTA) was found to be the most effective in the removal of the crust and was chosen for its removal. The pH of the EDTA cleaning solution and its application had to be modified according to the colour of the paint the crust lay upon. Visual observations and analytical assessments using optical microscopy and SEM after cleaning indicate a successful reduction of the crust resulting in a significant improvement in the painting's appearance.

Keywords Lead-rich salt crust, Efflorescence, Conservation Treatment, Ethylenediaminetetraacetic acid (EDTA), Cleaning, Pemulen Gel, NanoRestore gel, SEM–EDX

Introduction

Efflorescence is an all-encompassing term describing the deposition of internal paint components on the surface of the paint layer [1–3]. This migration of mobile components may obscure the surface and affect the aesthetic appreciation of the artwork. Their treatment is usually quite challenging as they are insoluble in most organic solvents deemed safe for aged oil paints or water [4]. Ethically, these disfiguring crusts can be considered part of the original. Cleaning such surfaces requires a delicate balance between material characteristics (solubility of undesired materials) and ethical considerations (removal of original materials). The challenge for a practicing conservator is to find the appropriate cleaning system to remove such crusts.

This paper focuses on the preliminary identification and removal of an efflorescent crust found on the *Vision of Saint Lutgard of Tongeren*, an eighteenth century oil painting by an unknown artist from the collection of the

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Fig. 1 *Vision of St. Lutgard of Tongeren.* 1700–1799. Oil on canvas. 79.0 cm × 63.0 cm. Catharijneconvent Museum, Utrecht. Before treatment. Left—verso; Right—Recto

Catharijneconvent Museum in Utrecht, The Netherlands (Fig. 1).[1] A grey matt surface crust all over the painting became apparent during treatment after varnish removal. The layer was identified using scanning electron microscopy backscattered electron imaging (SEM-BSE) coupled with elemental analysis (SEM–EDX). Theories behind its formation are proposed, established by visual observation of the painting, and cross-referencing these findings with published literature. Following this, an in-depth study of the ethical and technical dimensions relevant to the removal of the crust took place before conducting cleaning tests. Cleaning tests were carried out with aqueous solutions and gelled chelators as well as emulsion gels. Assessment of the cleaning tests and the cleaning processes with the challenges faced during cleaning are described, and further areas for research are proposed. These aspects aim to provide practicing conservators with insight and possibilities when encountering similar problems in other works.

The painting was acquired by the Catharijneconvent Museum in 1979. Not much is known about the origin of the painting, including the artist and its provenance. Stylistically, it has been dated between 1700 and 1799 [5]. The main subject of the painting is Saint Lutgard of Tongeren and her vision of Christ showing her his wounds. There was evidence of past restoration campaigns, which included the application of a reverse priming layer, the repair of a tear, paint consolidation, and the application of two varnish coatings. No report or documentation has been found that indicates when these treatments might have happened or by whom.

A condition assessment employing visual observations, technical photography, and material analysis was undertaken to characterize the materials and techniques of the painting upon its arrival in the conservation studio. A detailed condition report was written, summarizing the findings and establishing a treatment proposal to remove surface dirt, consolidate the tacking margins and paint around the tear, as well as clean the yellowed degraded varnish layers. A matt grey crust especially prominent in the lighter colours was noticed during varnish removal (Fig. 2). It covered the whole surface of the painting, except for the tacking margins and the edges of the painting that had been covered by the frame. The grey material appeared to pool in the valleys of the paint islands and cast a greyish veil on the painting. The visual effect of this layer on the paint surface was clear when comparing the unadulterated borders with the adjacent colours (Fig. 2).

[1] This painting was assigned to the corresponding author as part of the training for the Conservation and Restoration of Cultural Heritage program offered by the University of Amsterdam for technical examination and conservation treatment from 2019 to 2022. The treatment took place in two phases, phase 1 from 2019–2021 at the Ateliergebouw in Amsterdam and then continued at Stichting Restauratie Atelier Limburg (SRAL) in Maastricht from 2021–2022. The painting will subsequently be returned to the Catharijneconvent Museum.

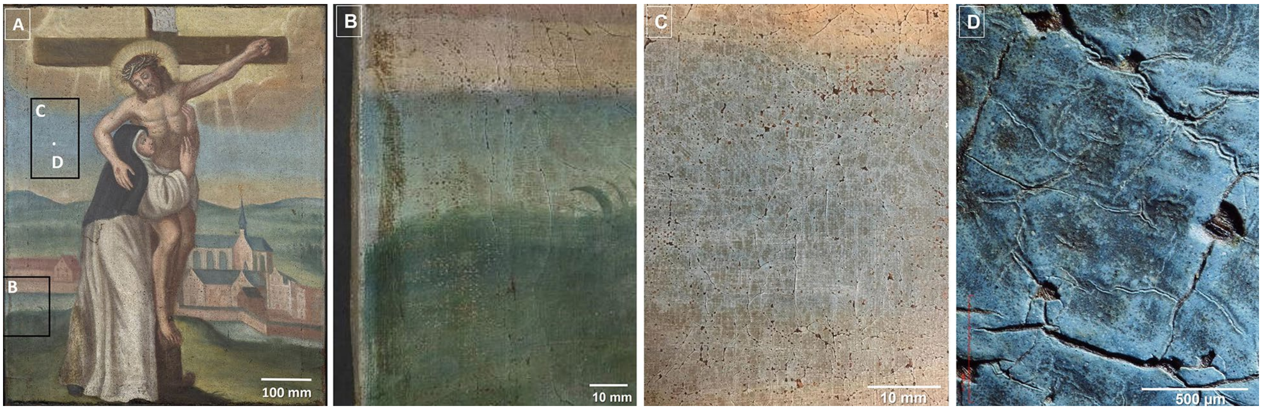

Fig. 2 **A** The painting after varnish removal; **B** Detail photograph of the edge of the painting showing varnish accumulation, the unadulterated border and the visual effect that the crust had on the painting; **C** Detail photograph of the prominence of the crust in lighter colours; **D** Photomicrograph showing the crust pooled in the valley of the paint islands

Methods/experimental

Identification of the crust

Cross-section samples were taken from different areas in the painting at different stages of varnish removal and were investigated with optical microscopy and SEM–EDX.

Samples

Cross-section samples taken at the University of Amsterdam (UvA) were embedded in Technovit 2000 resin and cured under ultraviolet radiation. Samples prepared in Stichting Restauratie Atelier Limburg (SRAL) were embedded in Polypol PS230 resin with MEK hardener. All samples were first wet-grinded with SiC sandpaper (320, 500, 800, 1200) till the sample was almost reached, and then dry polished with the Micro-Mesh polishing cloths (1500, 1800, 2400, 3200, 3600, 4000, 6000, 8000, 12000).

Microscopy

Examination of cross-sections at UvA was done with Leica DM 2500 M research microscope and photomicrographs taken with Zeiss AxioCam. Cross-sections at SRAL were examined with a Lietz Aristomet research microscope with a Leica C3 digital camera.

Scanning electron microscopy

The cross-sections were run on a JEOL JSM-IT700HR scanning electron microscope with a JED-2300-Fully integrated JEOL EDS system (100 mm2 SDD). The SEM was operated in low vacuum mode at a chamber pressure of 30 Pa, with an operating voltage of 20 kV, and a working distance of 10 mm. The cross-section was not coated prior to analysis. Some of the samples were analyzed on a JSM5910LV with a Thermo Scientific SDD EDX detector using NSS7 software.

Cleaning tests

Cleaning tests to remove the crust were carried out in the lighter-coloured areas of the painting where the crust was the most visually disturbing. Ethylenediaminetetraacetic acid (EDTA) and Citric Acid (CA) solutions were tested at pH values starting at 5.5 with a 0.5 increment till pH 8.5 and conductivity between 3000 and 6000 µS (considered safe for aged oil paints).[2] Pemulen® TR-2 emulsion gels were tested at pH 6.5, 7.0, and 7.5. The recipes for the cleaning systems were based on the stock solutions from the Modular Cleaning Program which were first diluted to a working concentration before testing [6]. Assessment of the cleaning was done based on visual and microscopic observations in visible light and ultraviolet radiation. A second series of tests with the EDTA solution at pH 7.5 in a gel form (10% methylcellulose) at durations of 30 and 60 s were carried out under a Hirox digital microscope KH-7700.

Results

Identification of the crust

A distinct thin layer, located directly above the paint layer, was imaged in SEM–EDX and identified to be the crust. Three representative cross-sections of the painting stratigraphy taken before, during, and after varnish

[2] EDTA itself acts as a buffer between pH 5.1–7.1 and CA itself acts as a buffer between pH 2.2–7.4, therefore no extra buffer was added for solutions in this range. For solutions above pH 7.1 for EDTA and above 7.4 for CA, triethanolamine (TEA) was used as a buffer. 10% hydrochloric acid or 10% sodium hydroxide solutions were used to achieve the required acidic and alkaline pH respectively. Pemulen® TR-2 gels were all buffered with TEA.

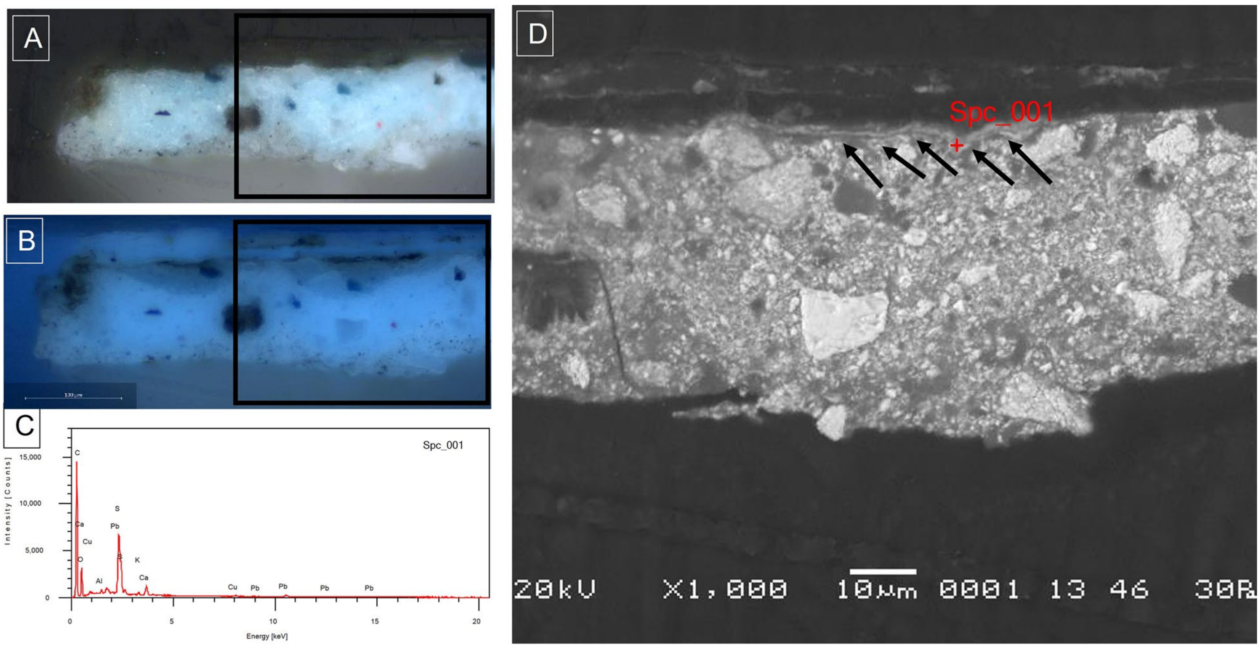

Fig. 3 Cross-section of sample BMRS4 #2 (blue sky) before varnish removal showing the two varnish layers and the blue paint layer; **A** Darkfield illumination; **B** UV radiation; **C** SEM–EDX spectra of the area identified as the crust and shown by the black arrows in **D**; **D** SEM-BSE image of the area shown by the black box in (**A**) and (**B**)

removal are discussed in the following paragraphs. A cross-section sample (BMRS4#2) taken before varnish removal, from the blue-sky area shows the blue paint layer consisting of a copper-based blue pigment, mixed with lead white and chalk, and two varnish layers separated by a layer of dirt (Fig. 3). The ground is missing in the cross-section of the sample. The BSE image indicates where the crust is situated, between the original blue paint and the later applied varnish layers. Under UV radiation the crust appears dark, and in the SEM-BSE image, the contrast pattern is light grey, indicating the presence of a material containing an element with high atomic weight. It is a separate layer, situated on top of the paint layer, closely following the topography of the paint layer but not incorporated into it. The crust has a uniform thickness in the sampled area, measuring 2 µm. The morphology of the crust is different from the other layers, appearing as a single continuous film, rather than having granular particles. EDX spot analysis on the layer detected the presence of lead (Pb) and sulphur (S) as the main components with calcium (Ca) and potassium (K) in lower percentages.

Sample BMRS4#7 is a cross-section from the white robe of Saint Lutgard, taken after the removal of the upper varnish (Fig. 4). A double-coloured ground, white paint layer and the remaining varnish layer are visible. SEM–EDX analysis indicates that there is a high chalk content (Ca) in the red ground layer, in addition to a few particles of lead white (Pb) with ochres or earth pigments (Fe), and some particles of vermillion (Hg). The analysis of the grey ground layer indicates a higher content of lead white (Pb) particles, but also a small quantity of chalk (Ca) and carbon black (C). Under UV radiation the crust appears dark situated between the paint and varnish layers. In the SEM-BSE image, it appears as a light grey layer directly above the paint layer. In this sample, the crust measures 6 µm, which is much thicker than in the sample shown in Fig. 3. EDX spot analysis on the crust gave similar results to sample BMRS4#2.

Sample BMRS4#8 is a cross-section from the blue-sky area, taken after the removal of both varnish layers (Fig. 5). The BSE image shows that the crust remains intact on the paint surface, indicating that it remained unaffected by the use of organic solvents (ethanol, acetone, ethyl acetate, Shellsol® T) used for varnish removal. In this sample the crust measures 4 µm. The EDX spot measurements continue to indicate lead (Pb), sulphur (S), potassium (K), and calcium (Ca).

The EDX spectra shows that the crust is rich in lead (Pb). Cross-referencing this with other published case studies such as the ceiling and chimney painting by G A Pellegrini (1675–1741) and Rembrandt's *Homer* (1663) in the collection of the Mauritshuis, The Hague, The Netherlands, it was inferred that it is a lead-rich salt

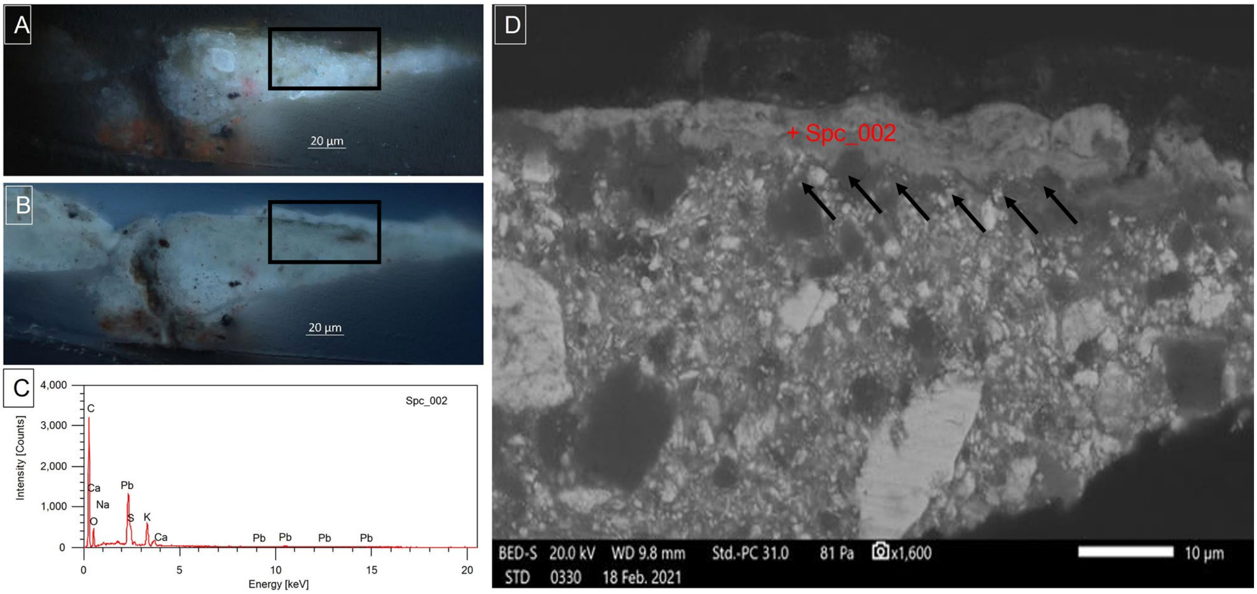

Fig. 4 Cross-section of sample BMRS4 #7 (white robe) after removal of the upper varnish showing the varnish layer, the white paint layer, and the double-coloured ground layers; **A** Darkfield illumination; **B** UV radiation; **C** SEM–EDX spectra of the area identified as the crust and indicated by the black arrows in D; **D** SEM-BSE image of the area shown by the black box in (A) and (B)

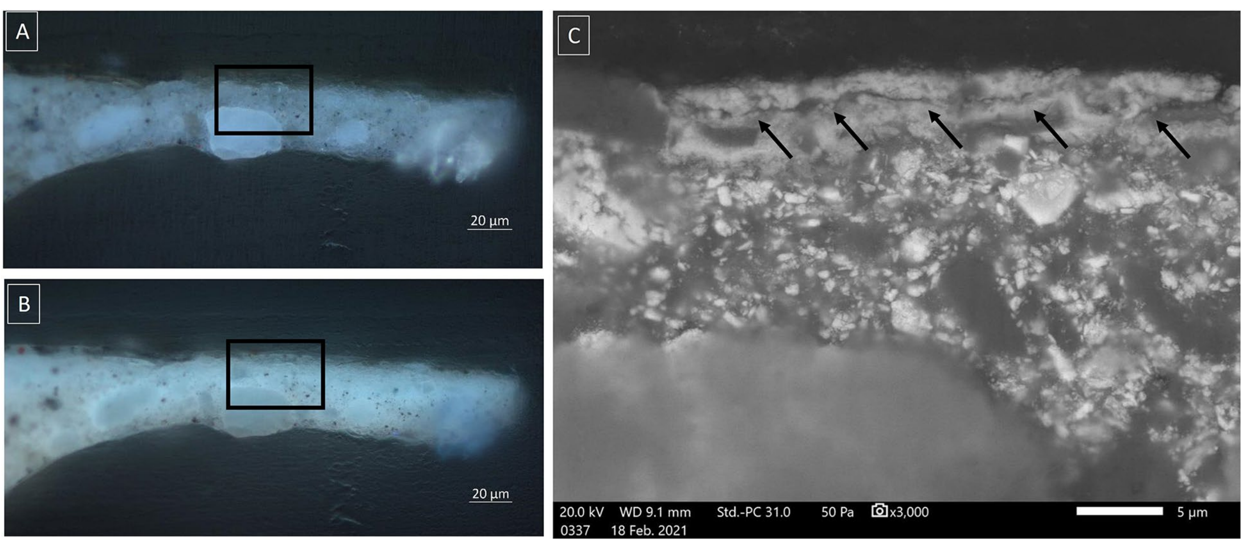

Fig. 5 Cross-section of sample BMRS4 #8 (blue sky) after removal of both varnish layers showing the blue paint layer; **A** Darkfield illumination; **B** UV radiation; **C** SEM-BSE image of the area marked by the black box in (A) and (B) which shows how the crust has a distinct morphology that is not as granular or integrated with the paint layer

crust and most likely a mixture of lead-potassium-sulphate and/or carbonate salt crust [4, 7, 8].[3] Moreover, it is highly possible that the crust also has other particles sourced from dirt and/or soot engrained in it.

Cleaning test results

Three cleaning approaches that would target different components of the crust were tested. The first approach targeted the lead in the crust with the chelating agent

[3] The additional use of infrared spectroscopy could better determine these assumptions. Unfortunately given the limited time frame and resources further analysis could not be undertaken.

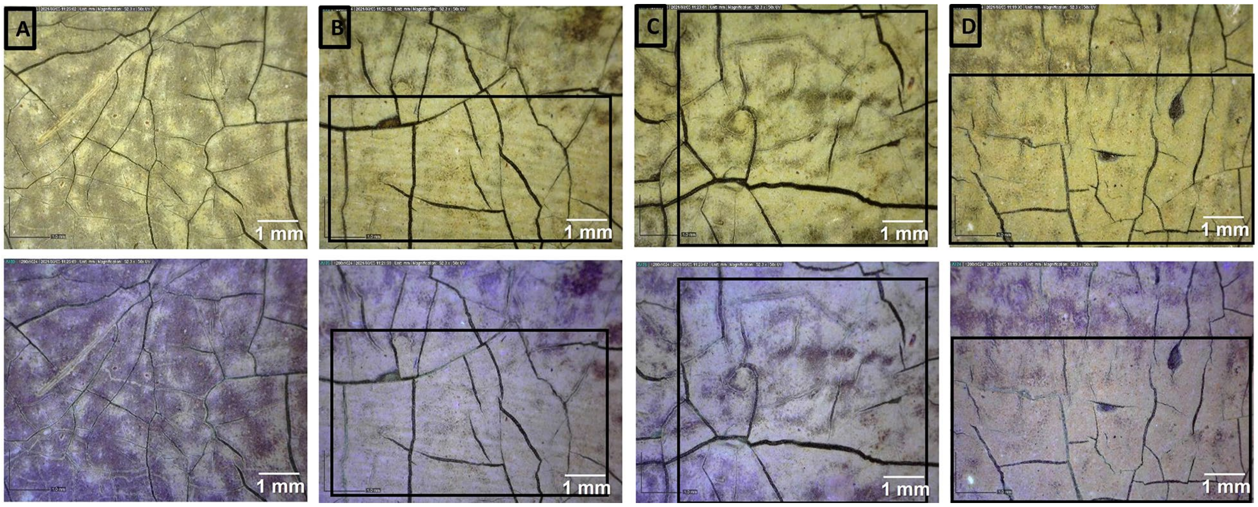

Fig. 6 Dino-Lite microscope images, comparing the different cleaning approaches and their cleaning effect in visible light (top row) and UV radiation (bottom row); **A** Control area showing the crust; **B** The EDTA at pH 7.5 cleans quite uniformly; **C** The CA at pH 7.5 gives an uneven cleaning; **D** The Pemulen® TR-2 gel at pH 7.0 cleans the crust but requires a lot of mechanical action and repeated application to reach the same level of cleaning as the EDTA

Ethylenediaminetetraacetic acid (EDTA) [3]. As the painting has other lead-containing layers, EDTA could have possible side effects on them. Therefore, the second approach focused on the dirt engrained in the crust. The idea was to remove the dirt and make the crust visually less apparent. Citric Acid (CA), a chelating agent with an affinity for iron commonly used for surface dirt removal was tested [9, 10]. The third approach was also directed towards the dirt engrained in the crust, using an aqueous emulsion gel made of Pemulen® TR-2 which would target the hydrophilic and hydrophobic components of the crust simultaneously [11].

The tests showed that EDTA at pH 7.5 and 8.0 immediately dissolved the crust and was able to clean the area. Solutions of CA at pH 6.5 and 7.5 after a lot of mechanical action, seemed to pick up something on the swab (most probably dirt) but resulted in a non-uniform cleaning. Pemulen® TR-2 gels at pH 6.5 and 7.0 had a better cleaning result than the CA. However, the gel required mechanical action to activate its emulsifying properties, which resulted in an increased gloss due to the burnishing effect of the swab. Under UV radiation, the crust appears dark, very different from the paint layer which enabled a straightforward assessment of the cleaning agents (Fig. 6).

Evaluation of the three cleaning approaches showed that the most efficient way to clean the crust would be to sequester the lead using EDTA. However, its application would have to be controlled so that the EDTA would target only the crust, and ideally does not get diffused into the underlying paint and ground layers. Thus, it was decided to gel the cleaning solution which would prevent or at least slow down the diffusion of the EDTA. The need to reduce mechanical action during cleaning, to not abrade the paint layer also favoured the use of gels. The use of gels requires clearance which ultimately results in mechanical action. However, in this case, testing showed that the effect of mechanical action during the rinsing of the gel was lesser than when free liquid was used.

Tests with the gelled EDTA showed that the 60 s gel application gave comparable results to the free liquid cleaning (Fig. 7). Empirical observations during cleaning showed that the gelled EDTA swelled the crust and slight agitation with a sable-haired brush dislodged it and it could then be picked up by a dry cotton swab. Under the Hirox microscope, the paint layer showed no immediate disruptions or craters which indicated that the controlled EDTA application removed the crust without harming the underlying layers. Whether there are any long-term effects or mechanical changes remains to be seen.

Discussion

Influence of treatment history and environmental conditions on the formation of the crust

The painting has undergone restoration treatment in the past. The reverse of the painting has been saturated with a layer that appears black in visible light [12].[4] Gas

[4] It is most probably intended to be a reverse priming application, commonly mentioned in eighteenth and nineteenth century sources, as a preventive measure for flaking, keeping the moisture from the walls from penetrating the canvas or preventing the oil from the paint or ground layers from reaching the canvas support and "burning" the canvas.

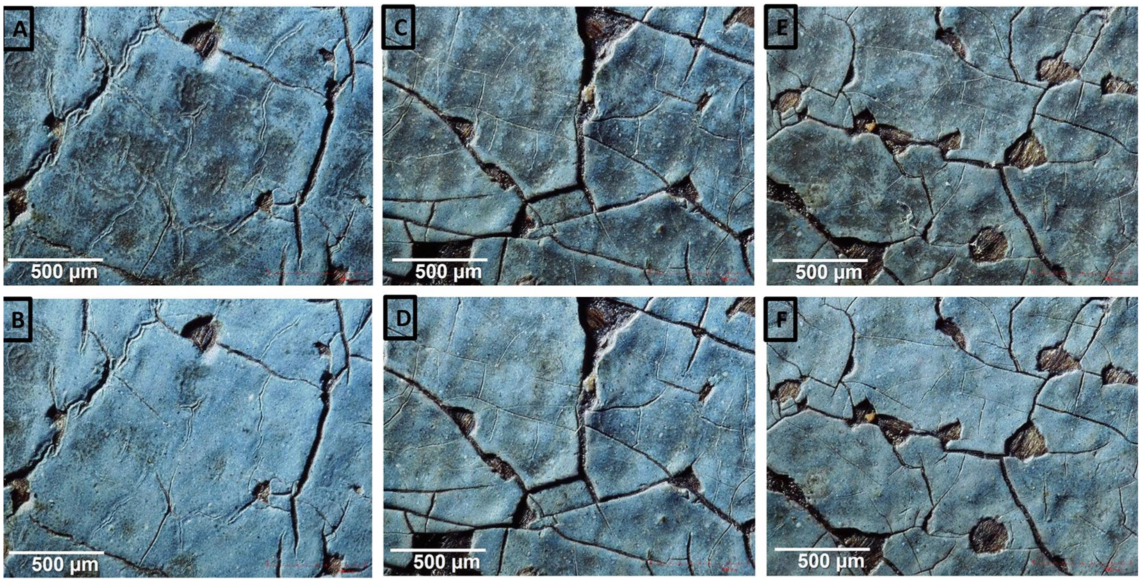

Fig. 7 Photomicrographs before and after cleaning with EDTA solution and gel at pH 7.5; **A**, **C**, and **E** areas before cleaning; **B** After cleaning with EDTA solution; **D** After cleaning with gelled EDTA at 30 s; **F** After cleaning with gelled EDTA at 60 s

Chromatography-Mass Spectroscopy (GC–MS) analysis of a scraping removed from the recto shows that it is composed of a mixture of oxidized Urushi, a drying oil, protein, cellulose compounds, and caffeine. Urushi is the Japanese term for the sap of the tree *Toxicodendron vernicifluum* grown in Asia, containing urushiol which is then processed and used as a lacquer. The lacquer was historically used to preserve, protect, and harden a substrate (usually wood) and as an adhesive [13]. Drying oil, animal glue or egg, and wheat starch would be added to the lacquer probably to modify its working properties, making it less viscous so that it can be easily brushed and leveled. Tea or caffeine would be added to tint it. As the history of this painting is not completely known, the origin of this layer is uncertain.[5] It could have been applied by the artist, the owner, or during a later restoration campaign. However, traces of staining on the sides of the wooden strainer members and the lack thereof under the wooden strainer members or under the tacking margins, suggests that it was applied after the painting was already stretched.

There is extensive craquelure forming a continuous network of paint islands of varying sizes in the painting. The edges of these paint islands can raise upward resulting in cupping and consequent flaking. In this painting, there is physical evidence that the cupped paint was flattened and consolidated with heat and/or pressure. The raised edges of the paint, especially at the intersections with other islands, seem to have chipped off causing numerous losses. This is indicative of what happens when pressure is put on a brittle layer of paint. Another heat related phenomenon observed across the surface are wrinkles and small circular lacunae. These could have been a consequence of softening and blistering of the paint due to overheating during consolidation, and the consequent removal of a facing that could have been applied on the paint surface during treatment.

The lead-rich salt crust was a result of the migration of lead ions to the surface of the paint layer and their consequent interaction with atmospheric gases to form a salt [4]. The lead ions were sourced from the lead white pigment used in the paint and ground layers. The diffusion and migration of lead ions is based on many factors, but it may have been accelerated by the presence of free saturated fatty acids and environmental factors such as high temperatures and humidity [14]. The application of the reverse priming layer, containing a complex mixture including a drying oil, could have provided the additional free fatty acid contributing to the migration and diffusion of the lead ions [15]. Research has shown that high temperatures increase the mobility of lead ions enabling them to migrate to the surface of the paint layer [4, 14, 15]. The consolidation process using heat could have accelerated the reaction. Interestingly, the tacking margins which had not been consolidated previously nor do they have the reverse priming application, show no signs of lead-rich salt crust. It is possible that the edges of the

[5] There was a growing trade for East Asian lacquers in seventeenth-eighteenth century Europe, especially for oriental lacquered furniture.

Arguments to NOT remove the crust	Arguments to remove the crust
- It is original material and one of the first tenets of conservation is to not remove original material from objects.	- Even though it is original material, once the material has migrated to the surface it cannot be put back. Additionally, this phenomenon of migration and crust formation is not something the artist intended.
- It forms a patina giving the painting a certain age and historical value.	- It will improve the readability of the painting.
- Maybe the crust protects the paint layer underneath from more subsequent migration reactions.	- The crust might get embedded into the paint later to a point that it cannot be removed without harming the paint.
- If the crust is removed it might disturb an equilibrium and cause it to form again.	- Since the painting is going back to a stable climate (T 20°C ±2°C and RH 50%±3%) the chances of the crust forming again seem less probable.
- Chelating agents might affect the lead in the ground or paint layers so there is a risk of chelation of original material.	- The crust can be cleaned relatively easily at this point.
- Removing by aqueous methods will require clearance of the cleaning gel with a rinsing solution, so we will be subjecting the painting to liquid water which is not advisable for metal soaps. (This can however be controlled by modifying the application method, rigid gels?)	- Since the crust is probably engrained with dirt, if we don't remove it, the dirt can attract moisture and facilitate other reactions in the future.
- If EDTA is used to clean the lead-rich salt crust, some residues might still be left behind. What are the long-term effects of EDTA residues?	

Fig. 8 Arguments to remove or not remove the crust

painting were protected from the environment and hence show no signs of the crust.

It is hypothesized that the painting was likely unvarnished for some duration of its life. GC–MS analysis of the later applied varnish on the painting indicates that it is dammar. Dammar was first recorded as being used in Europe in 1828 and gained popularity in the mid-nineteenth century [16]. This painting has stylistically been dated to the eighteenth century which means that either there was some other varnish present before the dammar application or no varnish layer. It has been suggested that the painting was hung in a church near a heat source or a window in direct sunlight (M Leeflang, curator at the Catharijneconvent Museum, personal communication, March 16, 2021). In these environmental conditions, the lead ions would have interacted with the surrounding atmosphere to form the crust. Once acquired in 1979, it was stored in the museum depot in more stable conditions.[6]

In addition to the lead-rich salt crust, a few metal soap aggregates were noticed in a few of the cross-sections and some white protrusions were noticed on the surface of the painting. This indicates that there might be additionally reactions happening between the lead ions and the free fatty acids which would need further investigation and were not possible in the scope of this research and conservation treatment.

To clean or not to clean

The cleaning tests showed that the lead-rich salt crust can be removed however, the removal of such crusts should not be undertaken without due ethical consideration. The source of lead are the lead-based pigments used in the painting that have migrated to the surface as a degradation product. The question of whether the crust can be considered an integral part of the artwork remains. The crust forms a patina on the surface of the painting and providing an age/historical value. However, the crust disturbs the tonal values of the painting by casting a greyish veil on it and diminishes the aesthetic value of the painting.

Aspects regarding the future of the crust and the underlying layers, if left untreated were also considered. For example, will the crust become insoluble or deeply embedded into the paint layer over time? Does the crust form a protective layer that prevents further migration of the lead to the surface? And if treated, is it possible that the crust is formed again? Moreover, the crust dissolves in chelating agents but they may affect other lead-containing layers.

To address these questions a table listing the pros and cons of removing the crust was prepared and presented to conservators, scientists, and researchers in the field (Fig. 8). After weighing the ethical, aesthetic and practical

[6] Temperature 19°C ± 2 °C and RH 50% ± 3% in Depot 13 of the Catharijneconvent museum.

aspects carefully, the decision was made in favour of removing the crust as it would greatly improve the visual appearance of the painting. The cleaning tests were also convincing regarding the potential effects of the cleaning agent on the paint layer. The decision was also supported by the current environmental condition of the painting and the future storage conditions of the museum which would minimize the chemical reactivity in the paint film. Additionally, the painting would also serve as a didactic case study allowing for a better understanding and adding to the collective knowledge on the treatment of lead-rich salt crusts. Monitoring the painting to see if there is a reformation of the crust will also continue.

Cleaning of the painting

In practice, there are three possible ways to reduce the visual impact of a lead-rich salt crust [17]. The first method is to saturate the crust to lessen its disturbing appearance with a low molecular weight resin, as it can be considered an original part of the painting. However, in this case, a test patch with Laropal® A81, made the crust even more visually prominent.[7] The second method is to visually reintegrate the crust by retouching. The extent of the crust on this painting made this option undesirable as it would result in considerable overpainting. Therefore, the option of removing or at least reducing it using aqueous cleaning systems was therefore undertaken [18].

The cleaning of the painting commenced with the background and then moved on to the figures. The blue-sky areas of the painting were cleaned first with EDTA at pH 7.5 gelled with 10% methylcellulose. The gel was applied to small areas (5 cm $\times$ 5 cm) and left on for 60 s. It was then moved around with a brush to release the bond of the crust to the paint layer. The gel was wiped off with dry cotton swabs and the area was rinsed with an aqueous solution buffered to pH 7.0. The cotton swabs used for cleaning would be continuously monitored to see if the original paint was removed in the process. Repeat applications of the gel were preferred if felt necessary than a single prolonged exposure allowing for intermediate evaluation of the surface. No visible effects were noted on the paint layer after the first, or second if required, applications of the cleaning system. However, long exposures (180 s or more) or more than two applications, resulted in the blanching of the paint layer. Thus, the cleaning action was stopped after a visual improvement was achieved or after two applications even if some residues of the crust remained.

As the cleaning progressed, it was noticed that the gel with pH 7.5 was not working the same way on other colours in the painting. In a few locations, a lot more mechanical action would be required to achieve the same level of cleaning which sometimes led to pigment pickup and/or abrasion of the paint layer. Tests with a different pH of EDTA showed that the pH of the cleaning solution had a significant effect on the cleaning. It is hypothesized that the crust is differently bound across the whole painting, or it has a slightly different structure depending on the composition of the underlying paint layers [4]. Thus, for each colour in the painting, the EDTA cleaning solution had to be tested at different pHs and the cleaning solution tailored accordingly. Throughout the painting, the range of pH varied from 6.0 to 7.5 (see Fig. 9).

The brown paints in the cross and the figures were sensitive to prolonged exposure to organic solvents and aqueous solutions. Mechanical action was critical as any slight movement of the cotton swab caused pigment pickup. SEM–EDX spot measurements on cross-sections taken from brown-painted areas showed peaks of iron, manganese, and aluminium in addition to lead and calcium, indicating the use of umbers and/or Van Dyke browns. Furthermore, spot tests with EDTA above pH 6.5 led to blanching. Observations under magnification showed that the upper skin of the paint layer was affected. In these more sensitive areas, the cleaning approach was changed to use rigid gels that delivered the cleaning agent as a gel but held their shape for easy clearance. The rigid gel would have to conform to the texture of the paint surface as the crust was pooled in the depressions of the cupped paint islands. It was thus decided to trial the Nanorestore Gel® Peggy 6 to clean the sensitive brown areas [19, 20].[8] These rigid gels consist of a polyvinyl alcohol network into which pH-adjusted aqueous solutions can be distributed.

The Nanorestore Gel® Peggy 6 was cut into small pieces (5 cm $\times$ 3 cm). Half of them were soaked in the EDTA pH 6.0 cleaning solution and the other half were soaked in a rinsing solution buffered to pH 7.0 for at least 12 h before use. The area to be cleaned was masked off with Melinex™. The pre-soaked EDTA gel was first tapped dry on non-woven tissues to remove excess liquid and then placed on the area to be cleaned. After 90 s, the EDTA gel was removed and the rinsing gel (also tapped dry) was placed on the area for 60 s. The gels swelled the crust which was then absorbed with a swab. A final

[7] Saturating with a low molecular weight resin probably works on crusts that are whitish. Since this crust has dirt/soot engrained in it which gives it a grey colour, the saturation increases the tonal contrast with the lighter areas free of crust making the visual impact of the crust even more prominent.

[8] The Nanorestore Gel® Peggy 6 is the most flexible of the gels produced by CSGI (Research Centre for Colloids and Nanoscience, Florence, Italy). It is a hydrogel based on a poly (vinyl alcohol) polymeric network and comes in the form of sheets (10 cm x 15 cm x 2 mm) loaded with water. https://www.csgi.unifi.it/products/peggy.html.

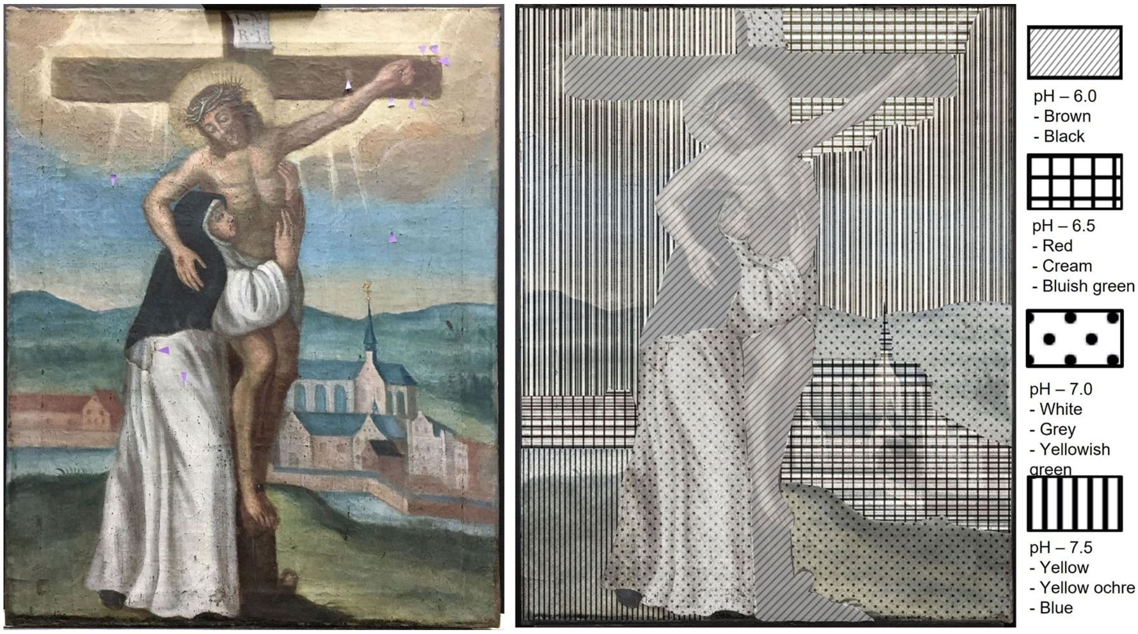

Fig. 9 Mapping of the different pH of the EDTA cleaning solution employed in different colours of the painting

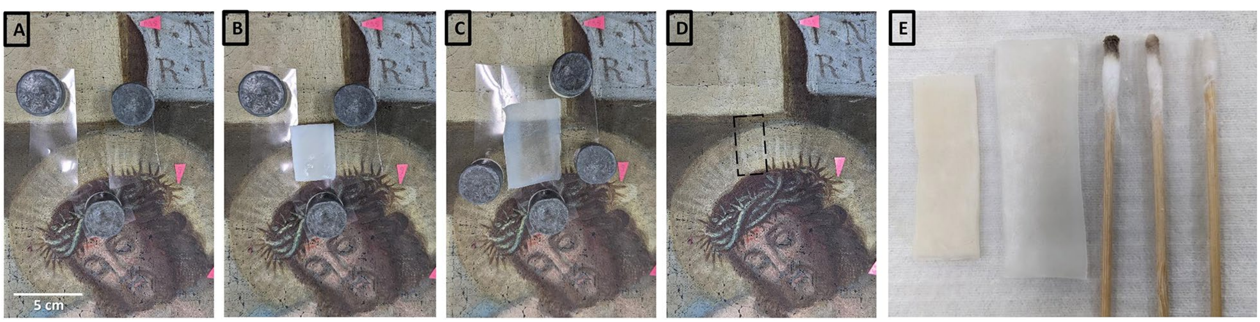

Fig. 10 Using the Nanorestore Gel® Peggy 6 for cleaning the sensitive brown; **A** Isolated the area to be cleaned and masked surroundings with Melinex™; **B** Placed pH 6.0 EDTA loaded Peggy Gel for 90 s and removed; **C** Placed pH 7.0 buffered rinsing solution loaded Peggy Gel for 60 s and removed; **D** Picked up the swollen crust with swabs; **E** The EDTA loaded Peggy Gel, the rinsing Peggy Gel and the swabs that picked up the crust

rinse with a swab was done to make sure no residues of EDTA were left (Fig. 10). Observation of the swabs used to remove the crust under magnification showed no pigment particles.

The crust was not imbibed into the gel pieces, which allowed these to be reused multiple times. However, the Peggy Gel matrix, when left immersed in the EDTA cleaning solution for more than 2–3 days, shrunk, became less flexible and whitened. This was due to the interaction of the polyvinyl alcohol polymeric network with the EDTA which slightly altered the equilibrium solvent content of the system (M Baglioni, personal communication, 30th March, 2022). The shrinking and the consequent whitening were due to the reduction of the size of the nano-domains inside the gel structure. The reduction in flexibility lessened the gel's ability to conform to the textured surface of the paint. Thus, new EDTA gel sections were utilized every two days.

Evaluation of the paint surface after cleaning

Finding the right cleaning agent and modifying its pH, concentration, and conductivity through intensive testing and applying different cleaning methods has resulted in significant visual improvement of the painting (Fig. 11).

Cross-sections and SEM analysis repeated after the removal of the crust validated the cleaning actions. A cross-section of sample BMRS4#12 from the blue-sky area (Fig. 12) shows the two ground layers and the blue paint layer. The SEM-BSE images show no pores or craters in the paint layer that would indicate a loss of

Fig. 11 Detail photograph of crust removal in the background of the painting; Top—Visible light; Bottom—UV radiation; Left—Before crust removal; Right—During crust removal

pigment particles. The surface of the paint layer maintains irregularities, which indicate that it has not been burnished or smoothened by mechanical action.[9]

Sample BMRS4#13 shows a cross-section from the white robe area showing the preparatory layers and the white paint layer (Fig. 13). The SEM-BSE image demonstrates a thin layer of the crust (~310 nm) that still remains, which further corroborates that the cleaning action did not abrade the paint layers. Visually, the cleaned surface looks quite healthy without any noticeable craters even under the microscope. The cleaning appears to be quite uniform, although some residues remain in the lower cups of paint. These residues still affect the visible appearance of the paint below, but to a lesser degree. The risk to the original paint when removing these residues further was deemed too high.

Conclusion

In the last two decades, studies on the identification, analysis, and formation of efflorescent crusts have been published. They have become a recurring concern when treating oil paintings containing lead-rich ground and paint layers. However, publications describing the treatment or removal of these degradation products, especially of lead-rich salt crusts, remain limited. This could partly be because these crusts are not always easy to identify unless the correct analytical technique is available or because they are so embedded in the paint layer that is not possible to remove them without harming the paint layer. The treatment of *Vision of Saint Lutgard of Tongeren* provided a unique didactic opportunity, not only to understand the formation of a lead-rich salt crust and the influence of treatment and

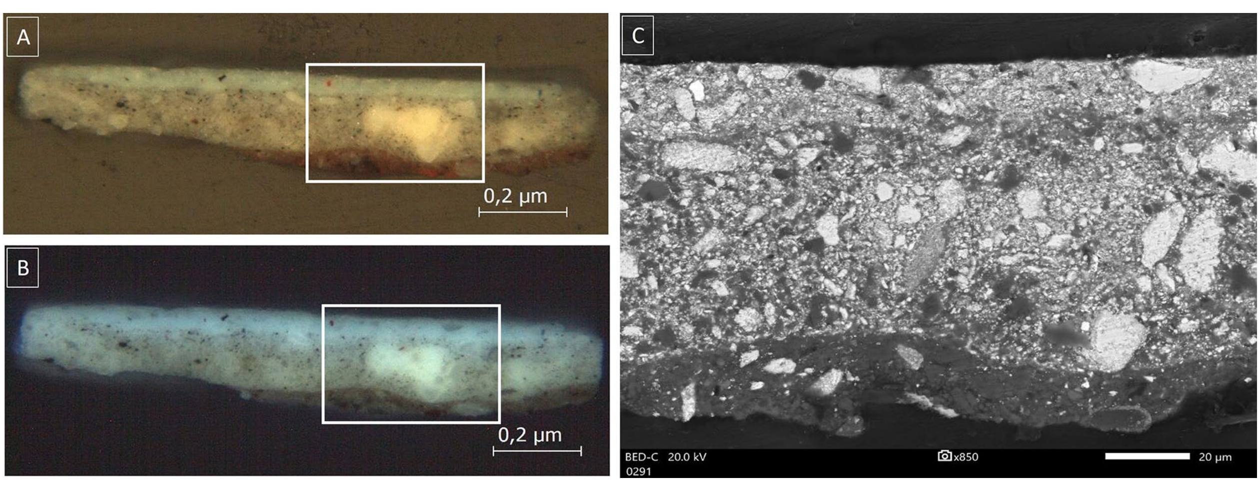

Fig. 12 Cross-section of sample BMRS4 #12 (blue sky) after removal of the crust showing the blue paint layer and the double-coloured ground layers; **A** Darkfield illumination; **B** UV radiation; **C** The SEM-BSE image shows that the paint layer has not been abraded

[9] Personal conversation with Dr. Ineke Joosten (RCE) on 18th March 2022. According to her, if the paint layer had been abraded, it would appear flat as if it was burnished or have a lot more craters if pigment particles were taken away.

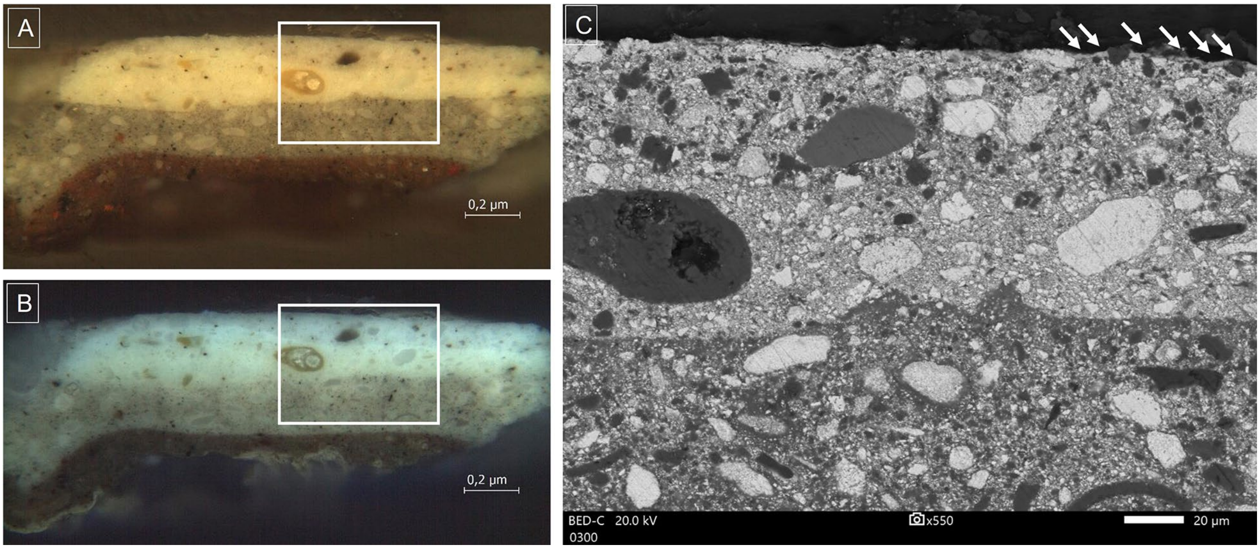

Fig. 13 Cross-section of sample BMRS4 #13 (white robe) after removal of the crust showing the white paint layer and the double-coloured ground; **A** Darkfield illumination and **B** UV radiation; **C** The SEM-BSE images show that a fine layer of the crust is still left behind indicated by the white arrows

environmental history on its development but also how to safely remove it and the practicalities involved in the undertaking of this process.

Abbreviations

CA	Citric acid
EDTA	Ethylenediaminetetraacetic acid
GC–MS	Gas chromatography with mass spectrometry
SEM–EDX	Scanning electron microscopy with energy dispersive x-ray
SEM-BSE	Scanning electron microscopy with back scattered electron
UV	Ultraviolet

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Author contributions
NS did the experimental and practical work on the paintings under the supervision of EF and KS. All authors read and approved the final manuscript.

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Declarations

Competing interests
The authors declare that they have no competing interests.

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