A Large Unvarnished Oil Painting on Canvas by Edvard Munch

No original varnish has been reported or observed on "Chemistry", one of eleven large Aula paintings by Edvard Munch. The problems specific to the cleaning of unvarnished oil paintings on canvas, especially in the case of Munch and other painters working in the early twentieth century, have received relatively little study in comparison to varnished oil paintings. "Chemistry" provides a compelling example of the problems faced by such works. It has been severely soiled by airborne pollutants and its surface is more difficult to clean and more fragile than comparable varnished oil paintings. During two periods in 2008, a total of 88 cleaning tests were performed on "Chemistry", 80 prior to and eight during one week of EU-ARTECH MOLAB Access Service in July. The aim was to propose a method to assess the cleaning system. The non-invasive mid-FTIR spectroscopy measurements allowed the identification and mapping of various types of surface contaminants like sulphates, silicates, metal soaps and zinc oxalates. Visual examinations and infrared measurements were applied to evaluate the effectiveness and harmfulness of the cleaning tests.

Introduction
Soiling and cleaning of Edvard Munch’s unvarnished oil paintings on canvas (1903–1916) in the Aula assembly hall of Oslo University (UO) was cyclical and the consequences were severe.1 The eleven large scale murals, all titled "Chemistry" had been cleaned up to six times in the seven decades between their installation in 1916 and the last treatment in 1986.2 In the course of the same period, the average interval between cleanings was eleven years and never exceeded sixteen.3 Comparable cycles for similar paintings and treatments at the neighbouring National Museum of Fine Arts, Nasjonalgalleriet are forty-six years.

In 2008, clean colours were found under random splashes of flour paste from the mounting of the Aula canvases on museum boards in 1946 (Fig. 1) (the boards were held together by a wooden framework on the reverse side).4 It seems the aim of the first three surface cleanings in 1925, 1937 and 1946 was to remove the airborne dirt. The same may also have been the case of the two later cleaning projects in 1957 and 1973 but the sixth and last cleaning only consisted of limited dirt removal (three paintings: "The Sun", "History" and "Awakening Men in the Flood of Light").5 This means that the dirt on the surfaces of the other eight had accumulated since 1973 and without intrusion of any known conservation treatment. The previous cleanings involved the use of various mechanical methods and solvents.6 Low-suction vacuum cleaners, common school sponges and hand-drawn sponges of bread dough rolled, pressed and rubbed over the surfaces were employed to remove the dirt.7 The first reported cleaning also involved the use of compressed air dipped in distilled water and in a fast evaporating petrolether (boiling point 45–60°C). During the last cleaning of the three paint-
Research questions
Clearing tests and MOLAB analyses addressed the following questions: Which of the tested cleaning agents are suitable for removing dirt from funerary unvarnished oil paintings in the Aule of UoT? How do they affect a selection of Munch’s colours when examined using visual methods commonly applied by conservators? How can conservation scientists expand on these results by employing mobile non-invasive examination techniques? What are the surface contaminants and what may be their causes? Finally, can the tests and the results of the examinations be used to propose a future cleaning regimen for Munch’s Aule paintings?

Materials and methods
A preliminary, non-invasive study of Munch’s technical procedures and materials in "Chemistry" was published in 2009. The investigation established that the painting consists of two twill-weave canvases. Results from a hand-held XRF suggest that the white, lean and porous ground contains zinc, lead and chalk. The greyish black underdrawings are an organic material, presumably a carbon-based black. Various other colour mixes can be found, with a mixture of pigments. Another major finding was that Munch used the same support, ground and painting technique without variation for these four paintings. A recent examination of the other seven Aule paintings indicates that fewer examples have been used in various combinations throughout the work. This implies that the results from the cleaning tests on "Chemistry" may also apply to the other Aule paintings as well as more contemporary paintings by Munch. However, Munch’s Aule paintings are quite young and some of the paintings may still be quite soluble. Thus organic solvents were considered unsuitable. The research therefore concentrated on a selection of aqueous systems such as natural water, non-ion-surfactant aqueous solutions and gels, a chelating agent as well as dry mechanical cleaning.

Cleaning agents, application and removal
Selection of cleaning agents for "Chemistry" was guided by a rather defined review of publications on dirt removal from unvarnished painting. However, restrictions were imposed by the young paint film and the large size of the painting. Another important factor was the availability of the cleaning materials. Cleaning agents had to be prepared, applied and removed by painting conservators working without the support of a specialist laboratory for their individual design (as most conservators did). The following cleaning tests (see Table 2), their application and removal 29 were carried out as follows:

- **Test 1**: Natural salve 29 was applied with a cotton swab for 16 seconds.
- **Test 2**: Marilgap 161/25 (2 w/v%) in distilled water with pH 6.5 was applied with a polystyrene sponge 29 for 16 seconds. The sponge was slightly moistened with the cleaning agent and softly pressed onto the test area. The sponge was never rubbed, wiped or rolled back and forth. Application of the cleaning agent was followed by two subsequent removals with a cotton swab dipped in distilled water.
- **Test 3**: Bri 700-gel 29 (pH c. 9) was applied by brush. The gel was immediately removed with a dry cotton swab. This was followed by two additional removals with a cotton swab dipped in distilled water.
- **Test 4**: Triton X-100-gel 29 (pH 9) was applied by brush. The gel was immediately removed with a dry cotton swab. This was followed by two additional removals with a cotton swab dipped in distilled water.
- **Test 5**: Tris-ammonium citrate (TAC) 36: 1w/v% TAC in distilled water was applied with a cotton swab for 16 seconds. Application was immediately followed by two removals with a cotton swab dipped in distilled water.
- **Test 6**: TAC: 1w/v% TAC in distilled water was applied with a polystyrene sponge. The sponge was slightly moistened with the cleaning agent and softly pressed onto the test area. The sponge was never rubbed, wiped or rolled. Application of the cleaning agent was followed by two individual removals with a cotton swab dipped in distilled water.
- **Test 7**: A sponge of vulcanized natural rubber gum was rolled using slight pressure for c. 30-60 seconds.
- **Test 8**: A small lump of fresh white bread with washing soda on the crust 29 was rolled back and forth until the lump picked up dirt and turned greyish (not tinted). All the tests except the sixteen preliminary tests (see below) were performed upon 2 x 2 cm squares. Their dimensions were controlled by using same Malmex frames. Each application was then followed by a stoppage. When cotton swabs were employed, they were rolled in two opposite directions, except on the dark blue and dark green colours, where the cotton swabs were rolled only in one direction to reduce the risk of removing original pigment.

Location of the cleaning tests
According to empirical experiences gained at the Munch-Museum, some of the paints used by Munch have proven to be more vulnerable to dirt removal than others: lean blues and greens, yellows, inorganic reds and white, weakly bound grounds all bound in oil. 27 "Chemistry" contains all these colours, but its yellows were omitted from testing 29 with the selected set of cleaning agents. The recent XRF examinations suggested that the majority of colours in "Chemistry" is composed of more than one individual pigment. 29 Including rather large areas of bare white ground (both lean and water sensitive), the painting contains more than thirty different applications of oil colours. Half of these are painted as rather slim lines. When the number of cleaning agents to be tested was decided, it became clear that many lines and colour applications in "Chemistry" were too small to accommodate all the eight cleaning tests, including adequate uncleaned areas of the same colours.

The cleaning tests were performed in two periods in 2008, prior to and in the course of MOLAB's one week visit in July. First, two sets of eight tests were applied as preliminary tests and not intended to make a definitive evaluation of cleaning agents. These preliminary tests are used here as a comparison between various cleaning agents. The second stage of testing with eight additional sets of tests was intended to compare and evaluate the cleaning agents.

Table 2 shows the first set of 64 individual cleaning tests on Chemistry and their visual appearance judged with the naked eye, a microscope (magnification 43x) and a UV lamp.

| Application area | Colour | Pigments | No pigment loss | Pigment loss | Pigment loss | Good cleaning but pigment loss | Good cleaning but pigment loss | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning | Good cleaning 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Mould fungi analysis

In March 2009, after dismantling the other Aula paintings, some small amounts of visible mould were observed on some of their reverse sides only, which might pose a risk to the painted surfaces [Fig. 4]. By then, the surface of "Chemistry" had already been cleaned mechanically with polyurethane sponges.\(^{25}\) "Harvesting Women\(^{26}\), which was also mounted on an external wall and has similar materials, techniques and cleaning history, was examined for possible biological contamination. Samples of surface dirt were taken from its surface, edges and reverse side.\(^{27}\) This sampling was performed with tape; brush onto tape and with polystyrene sponges. In addition, on the reverse side of "Women Reaching Towards the Light\(^{2}\), two pieces of paper with visible dark spots as well as scratchings from the same surfaces were examined. All samples were studied under a microscope (JSM-840; magnification 400×). Cotton blue dye was added to highlight fungi.

Visual examination

The visual appearance of the sixteenth preliminary cleaning tests and the first sixty-four tests, their corresponding colours and the used cotton swabs were evaluated with the naked eye, under a microscope (magnification 63×) and with a UV lamp. The eight additional tests were judged only with the naked eye.

Non-invasive reflective FTIR

The MOLAB instrument consists of a portable JASCO\(^{25}\) FTIR 9500 spectrophotometer (50 x 50 x 50 cm\(^3\)), 35 kg equipped with a HENSCHEL mid-Infrared fibre-optic sampling probe. The bench components include a MOLAC irradiation lamp source, a Michelson interferometer and a liquid nitrogen cooled MCT (Mercury Cadmium Telluride) detector. The fibre-optic probe, a bifurcated cable containing 18 chloroformide glass fibres, is used for the collection of spectra from 6500 to 900 cm\(^{-1}\). An excellent signal to noise ratio (S/N) is obtained throughout the range with the exception of the 2000-200 cm\(^{-1}\) interval, where the S/N ratio is by approximately one order of magnitude as a result of the fibre 56-H stretching absorption. The investigated sample area, as determined by the probe diameter is approximately 1.2 cm\(^2\). Correct reflectance measurement is achieved by obtaining the reflection spectrum of an Alflatop mirror. The spectrum intensity was defined as the pseudo absorbance A with A = log(1/R). The fibre-optic probe was positioned perpendicularly (\(90^\circ\) geometry) and at a distance of approximately 2 mm from the sample surface. In "Chemistry\(^{2}\), spectra were collected with 40 scans at a spectral resolution of 4 cm\(^{-1}\). Each spectrum was averaged over 10 spectra carried out for each individual cleaning area resulting in about 130 spectra of the surface. In addition, for comparison, spectra were also collected from painted areas that had been covered by the picture frame and from the picture frame itself.

Acceptable

Minimal but even improvements after cleaning imply that colour and gloss were still obscured by dirt, although there might be circumstances where this is the only option for dirt removal. There were four-

teen tests that were classified as acceptable, that is, no pigment loss, and the results were even but were minimally effective (see value 2 in Tables 1 and 2). The best cleaning results were found among the remaining twenty cleaning tests characterized as average and good. Three tests were given the value of 4 and seventeen tests 6 (see Table 2).

Colours in relation to the cleaning agents

To sum up the sixty-four tests, only colour (dark green) can be cleaned with all of the cleaning agents, one (blue mixed with white) with seven agents, one (red mixed with white) with five, four (dark blue, green mixed with white, red mixed with white and bare ground) with three and finally, one (dark red) with only two. Simultaneously, the cleaning agents can be arranged according to the evaluated results: natural ash best (1) seemed useful for three colours, Mural 1819/5 test 2 for four, Brij 70-gel test 3 for two, Triton-X-gel (test 4) for five, 1%w/v TAC (applied with a cotton swab, test 6) for four, 1%w/v TAC (applied with a sponge, test 8) for two, natural rubber gum (test 7) for eight and finally, bread dough (test 8) for six (see Table 2).

Analysis of the surface contaminants

Fibre-optic FTIR spectrometry allowed analysis of the composition of the paint layers and of the superficial deposits. The equipment works in \(0^\circ\) geometry collecting both specular and diffuse reflected light. For this reason, fibre-optic FTIR measurements are very sensitive to surface materials active in infrared.\(^{37}, 38\) The FTIR measurements on the uncleaned areas provided a basis for the identification of variable amounts of the following compounds: metal (Pb/Zn) carbonate, sulphates, silicates and zinc oxides (Fig. 7). Metal carbonate is also known as metal soaps, are identified by the absorption at 1560, 1540, 1510 cm\(^{-1}\) assigned to asymmetric stretching \(\nu_a\) and symmetric stretching \(\nu_s\) of CO\(_2\), appearing as derivative bands due to the effect of specular reflection. Sulphates and silicates are identified by the infrared bands at about 1140 and 1100 cm\(^{-1}\), respectively, attributable to the asymmetric stretching of Si-O and Si-O\(^{3}\) O\(_2\) species. Identification by observation of signals in the region of 1300-1370 cm\(^{-1}\), in particular by the presence of 4 absorption bands located at 1320 and 1360 cm\(^{-1}\) assigned to asymmetric stretching of \(\nu(C-O) + \alpha (D-C-O)\) and 1344 cm\(^{-1}\) \(\nu(C-O) + \alpha (\Pi-C-O)\) that are specific for Zn-oxalates. Moreover, the small signal at 3386 cm\(^{-1}\) (OH stretching) characterises them as hydrated zinc oxalates.

The results were compared with spectra collected on the areas that had been covered by the pic-
In the present case, no residuals of chelating and surfactant agents were observed in “Chemical” because they had been removed.

Biological contamination? None of the twenty samples of surface dirt taken from the surface, edges and reverse side of “Her-Vestering Woman” showed evidence of mold or any other biological activity.

On the reverse side of “Woman Reaching Towards the Light”, however, seven species of fungi were found in a few areas on the paper and in the glue attaching it to the wood.

A general framework (Uromyces sp., Aspergillus glaucus, Cladosporium cladosporioides, Eurotium herbarium and Ulocladium chartarum). Fungi on the reverse side of the rigid support may contribute to some biological activity on the surface of this single painting and must remain open to discussion.

Discussion

Implications for prospective dirt removal

The Aula paintings were contaminated with large accumulations of loose and firmly attached dirt. While this is unfortunate for their condition and visual characteristics, the present cleaning tests and their examination proved that every prospective cleaning project involves limitations regarding both the available range of cleaning agents and their manner of application and removal.

Given the visual impact, none of the present cleaning materials and techniques was adequately good (value 6) for all the eight colours tested. The non-ionic surfactant, like the Triton-X-gel (test 4), which had the highest number of good results, did nevertheless remove original pigments on dirt, diluted blue, dilute red and on the semi-solid paint on the underradiations. With minor exceptions, these colours were difficult to clean by all the tests and Methods of illustration. The treatment on “Chemistry” is much wider than these eight and other tints may be more or less resistant than the tested ones.

Only the two dry mechanical cleaning methods (tests 7 and 8) released original pigments or metallic soaps. Although, the latter resulted in two uneven areas (value 3). In just a few cases, the appearances of these tests were appreciated as average or good (values 4 and 6).

Neither the two mechanical tests nor saliva (test 1) decreased the invisible zinc oxalates on the surface of “Chemistry”, but the non-ionic surfactant agents (tests 2, 3, 5 and 6) rendered two results. Only the two tests with the chelating agent tri-ammonium citrate (tests 5 and 6) efficiently removed all of the oxalates (see Fig. 8). At the same time, original pigments were released by the mechanical cleaning at least in two campaigns (1987 and 1992). Visible zinc soaps are accordingly widespread. They were for instance analysed in “Falling Leaves” from 1888 by Vincent van Gogh (1853–1890). The painting contained white or transparent zinc soaps which occurred following a rather recent application of varnish. This surface defect was described as transparent material slightly elevating through the paint and standing upright through cracks in the paint layer. It was suggested that several applications of varnish including the repeated use of solvents had triggered the formation of the aggregates.

From early on, the paintings from 1927–1932 by Stanley Spencer (1891–1959) at the Sandham Memorial Chapel have endured recurring surface deterioration of isolated material.

By 1975, they had almost been treated twice in order to reduce the thickness and were probably varnished for the first time. By 1993, white bloom had been removed at least three more times and further costs of varnish had been applied. The white deposit could easily be removed with a soak. Analysis suggested that the blanched material contained degradation products such as fragments of varnish, fatty acids, inorganic pigments and possibly soaps. The influence of air pollution and moisture on the blanched problem could not be ruled out, and frequent varnishing did not prevent the continual whitening. On the contrary, it may be possible that all these applications were crucial for the whitening as in the case of “Falling Leaves” by van Gogh. In experiments, applications of varnishes containing polar, oxygenated resins dissolved in polar solvents were reported to extract soluble components from oil based films.

In another, although unvarnished oil painting from the twentieth century, such as “Composition Without Title” by George Pollock (1920–1960) from the late 1950s, visible surface blooming caused by the formation of metal soaps was reported in 1999, about thirty years after application. In this case, zinc soaps migrated and/or precipitated on the paint surface, with an increase of the blooming with rising humidity was observed. In the late 1980s, this bloom was removed with a dry scrap and the surface was subsequently covered with a 5% solution of tween in white spirit.

To our knowledge, no occurrence of whitening on this painting has been reported.

Those few cases suggest that the growth of white surface crusts (as metal soaps and other materials) can be activated by solvents used in conservation treatments, pollution, high temperatures and relative humidity. So far, visible white crusts of metal soaps have not been analyzed in any painting by the RISM team. It seems that the Aula paintings may be candidates for similar, prospective and undisclosed changes in texture, colour and transparency. The treatment history of the Aula paintings involved solvents for at least two campaigns (1987 and 1992), cleaning at least in two campaigns (1987 and 1992), and more than 90 years of accumulation (six known cleanings) and relative humidity (RH) up to 80%.
In addition, the paintings also have grounds containing white pigments of zinc and lead, which easily react with fatty acids in the oil. Still, neither "Chemistry" nor the other Aula paintings have any perceptible surface whitening or increased transparency; however, this may only be a question of time (see below). Since invisible metal soaps of zinc and lead were found on "Chemistry" and both contain exposed and unpainted surfaces (covered by the frame), they must be related to the artist's painting materials (including possible paint driers) and their aging. This also implies that the same process is going on in every Aula painting, as well as in other similar paintings by Munch.

The sources of oxalate Zinc oxalates were discovered on exposed grounds on "Chemistry." They are, at present invisible and their presence was surprising because this phenomenon had never been reported. The formation of other oxalate patinas as a degradation phenomenon has been widely observed and studied in the case of outdoor marble artefacts such as sculptures and monuments. Generally, these patinas are made of calcium oxalate (CaCO₃·H₂O), gypsum (CaSO₄·2H₂O), silicates and phosphates together with iron compounds which are responsible for their characteristic yellow colour. In particular, calcium oxalate may be present in two different forms, hemihydrate (CaCO₃·H₂O) and dehydrate (CaCO₃·2H₂O) according to the number of crystallization water molecules. The fact that oxalate patinas are degradation phenomena is clear considering the reaction that is behind their formation: CaCO₃·H₂O → CaCO₃ + H₂O + CO₂.

Indeed, there is a consumption of original calcium carbonate due to the acid-basis interaction with oxalic acid. On the other hand, it must be noted that once formed the oxalate patina may act as protective layers on marble due to the lower solubility of calcium oxide with respect to calcium carbonates including their great variety of textures and light reflections. Finally, and for the same reasons as for the formation of metal soaps, zinc oxalates may be found on the other Aula paintings and also on other works by Munch.

Cleaning proposal For the Aula paintings, dry and mechanical cleaning may represent the only cleaning option since these techniques remove neither original pigments, metal soaps nor zinc oxalates. If the zinc salt is interpreted as disposable both before it turns visible and after, it will have to be dealt with in both cases. On the other hand, if the invisible oxalates are considered as part of the aging products closely connected to original material before they become visible, it will be disputable to try to remove them before we have sufficient knowledge about their future formation and characteristics.

On other paintings, visible crusts of metal soaps have been removed mechanically or by increasing their saturation by impregnation. However, it is still an open question whether filling micro fractures in surface soap deposits, for instance with a low viscosity resin solution may worsen the problem. Munch's Aula paintings are neither suitable for impregnation nor application of varnish. Furthermore, organic solutions could intensify the soap formation, and any type of surface saturation will change both the artist's intent and the appearance of these paintings. Such treatments will alter the large areas of matte, opaque and groundy grounds by making them transparent and by revealing the darkened canvas below. Moreover, they will diminish the tremendous variation in surface glosses.

Conclusions In general, "Chemistry" shares the same materials and techniques with the other ten paintings in the Aula. The present study has revealed that "Chemistry" has started to form reaction products of metal soaps and zinc oxalates on its surface which are so far, detectable only with the aid of a mobile and non-invasive FTIR.

The preliminary cleaning tests on "Chemistry" demonstrated that any prospective dirt removal must be performed without the use of our selection of aqueous systems because they can remove original pigments. In the case of the other Aula paintings and prior to a future development of more suitable cleaning agents for monumental and unvarnished oil paintings, mechanical cleanings seem to be the only preferable approach. Since vulcanized natural rubber gum and bread dough used on "Chemistry" only slightly removed parts of the dirt, but not pigments, metal soaps or zinc oxalates, we had to look for another mechanical approach to improve the dirt pick-up. Following access to the MOLAB-diphosphonate sponges were used for this purpose on the Aula paintings. Although not tested to the same extent as the areas cleaned with rubber gum or dough on "Chemistry", the efficiency of these sponges when used dry (no water or organic solvent) is apparent (Fig. 9).

Before the present renovation of the building the indoor climate of the Aula contained quite extensive amounts of dirt, and the cleaning history of its paintings clearly indicated short intervals between the previous treatments. It is hoped that the renovation will generate less airborne pollution when the paintings are reinstated in spring 2011, but the prospective and extensive use of the room also implies continuous dust production. To extend future intervals between major cleaning of the paintings, we think it would be important to introduce minor cleaning each year to brush off the inevitable accumulation of loose dust (into a tip of a suitable vacuum cleaner) before it becomes firmly attached to their surfaces.

Suggestions for further research Sooner or later the soaps of lead and zinc and the zinc-oxalates may occur as visible impurities on the painted surfaces of the Aula paintings. To be able to delay this process and to prevent possible expansion of these products with proper means for a good preventive control is of utmost importance. As an updating of their prospective aging processes, since visible metal soaps have been reported to be removable whereas visible oxalates of copper and zinc have been characterized as insoluble, priorities should be given to gain more insight about zinc oxalate formation.

Following renovation of the building and subsequent reintroduction of the paintings, the quality of the air inside should be evaluated and a corresponding aging experiment conducted in situ on the reaction between zinc oxide and oxalic acid in the presence of water. The latter can be performed by a suitable arrangement close to one of the paintings with a battery of test pieces of canvas (2 x 2 cm²) covered with a layer of fresh and unvarnished zinc oxide. Each summer and prior to the suggested minor removal of dust, one of the test pieces can be taken down, sealed and sent to the MOLAB laboratory for analysis.

With this arrangement we trust that the proposed monitoring test materials and further analyses will provide reliable data to act upon if and when necessary.

Acknowledgements Many thanks to Harri Miettunen and Pekka Kemppi for discussions and to Noliville Streeton for editing (both IAIX, UO). Access to the MOLAB was supported by the EU within the 8th Framework Programme (Contract EU-ART-05, RIS-CT-2004-00177). The 2009 Aula cleaning project was funded by the Technical Department, UO.