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Understanding and optimizing Evolon® CR for varnish removal from oil paintings

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Abstract
Evolon® CR is increasingly used in paintings conservation for varnish removal from oil paintings. Its key benefits over traditional cotton swabs are limiting solvent exposure and reducing mechanical action on the paint surface. However, this non-woven microfilament textile was not originally engineered for conservation use and little is known about its chemical stability towards organic solvents. Moreover, the physical processes of solvent loading and release by Evolon® CR, as well as solvent retention inside paint after cleaning, have not been studied. These three topics were investigated using a multi-analytical approach, aiming for an improved understanding and optimized use of Evolon® CR for varnish removal. Our results show that the tissue is generally chemically and physically stable to organic solvents when exposed on timescales that are typical in conservation practice. However, a pre-treatment step of Evolon® CR is necessary to avoid the release of unwanted saturated fatty acids into the paint during varnish removal. We show that the primary mechanism of solvent uptake by the fibers is *ad*sorption rather than *ab*sorption and that the dominant factor dictating the maximum solvent load is the volume of the voids between the fibers. Finally, solvent induced dynamics after application of solvent-loaded Evolon® CR within the paint film was monitored using portable laser speckle imaging on model paints. A method to quantify solvent-retention in real-time was developed and revealed that the presence of varnish on paintings results in lower dynamics of solvents within the paint in comparison to unvarnished paint. Comparing various solvents, it was found that cleaning with acetone resulted in a roughly six-fold increase in dynamics compared to ethanol and isopropanol.

Keywords: Oil paint cleaning, Evolon® CR, Varnish removal, Portable laser speckle imaging

Introduction

The growing corpus of scientific literature dealing with the physicochemical aspects of oil paint alterations has led to concerns by conservators about unwanted adverse effects of solvents on paint films [1]. Due to the limited knowledge regarding the short- and long-term chemical and physical stability of oil paintings, exposure of paint films to solvents should be kept to a minimum. To address these concerns, cleaning methods employing gels, gel composites or emulsions (with or without the aid of tissues) have been developed as alternatives to traditional swab cleaning [2–13]. Gel cleaning can reduce the amount of solvent or water released into a paint film [1]. However, the application and clearance of spreadable gels using a brush or swab requires mechanical action and poses the risk of leaving behind gel residues with unknown long-term effects on the stability of the paint [14]. The use of rigid gels with either physical or chemical cross-links can reduce the risks of residues but such gels are not always compatible with organic solvents used for varnish removal on oil paint.

In the last 5 years, a new method of minimising solvent exposure has been developed using Evolon® CR, a speciality-grade, non-woven microfiber cloth. This tissue is loaded/humidified with the conservator's choice of

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solvent, and subsequently placed in direct contact with the paint surface, upon which the solvent is released and the varnish is solubilised and poulticed into the cloth. This procedure aims to reduce the amount of solvent necessary for varnish removal, and results in significantly reduced mechanical action and solvent exposure.

Evolon® fabrics have been developed for a range of applications, including dust-mite-proof mattress covers, anti-allergen pillows, cosmetic wipes, filtration, technical packaging, kitchen towels, and more [15]. Evolon® CR is a specific type within the Evolon® family that has been designated for use in conservation (C = Conservation, R = Restoration). Manufactured by Freudenberg and marketed by Deffner & Johann, it is a blend of 70% polyester and 30% polyamide fibers spun into segmented filaments, and features a specific density of 77.4 g/m^2 [16, 17].

Previous studies

In a 2018 publication describing the application of Evolon® CR in a conservation treatment, Tauber et al. presented a procedure called *controlled loading*, where the amount of solvent loaded into the tissue is regulated and kept below its maximum saturation. Controlled loading is achieved by weighing known amounts of Evolon® CR and solvent, and sealing them in a closed container (glass tube) overnight or 12 h. This procedure aims to achieve a homogeneous distribution of the solvent or solvent vapour throughout the tissue [17]. It is assumed that a homogeneous distribution of the solvent inside the tissue would directly translate to a homogeneous controlled release of the solvent into the varnish layer, which appears confirmed by the homogeneity of the resulting varnish removal as observed by Tauber et al. [17].

The use of Evolon® CR has numerous advantages in addition to reduced solvent exposure of the painting. Firstly, it enables a systematic approach to varnish removal, which is especially beneficial when working on large-scale paintings with different conservators. Secondly, it permits a more reproducible method of applying solvent compared to swab cleaning, potentially permitting a means of standardising varnish removal. Thirdly, the application process of lightly pressing the fabric to increase contact with the painted surface involves little mechanical action. As a fabric, Evolon® CR can be used in sheets of any size, and can be custom cut to match the shape of elements in painting. Additionally, discoloured varnish and retouches poulticed into the fabric can provide invaluable information about the varnish and retouching used in previous restorations. Finally, the used sheets of Evolon® CR can be kept as documentation of the cleaning process and analysed [17]. For example, the elemental composition of overpaint visible on used sheets of Evolon® CR can be analyzed using XRF and the composition of the removed varnish using GC/MS.

Several researchers have already made seminal contributions to the improved understanding of Evolon® CR's use in conservation practice. In 2019, Vergeer et al. used ethanol mixed with a fluorescein isothiocyanate protein indicator dye to visualise solvent distribution in a paint film and found considerable variation depending on the method of application [16].

Baij et al. systematically compared the use of Evolon® CR with traditional swab and rigid gel cleaning [17, 18]. By monitoring the saturated fatty acid (SFA) extraction from the paint film, it was found that the use of Evolon® CR with controlled loading and rigid Nanorestore® gel cleaning led to significantly reduced SFA extraction compared to the use of free solvents and cotton swabs [18].

In a continued study, Baij and Buijs et al. employed portable Fourier transform laser speckle imaging (FT-LSI) to visualise and monitor solvent retention in model oil paint films. The experiments demonstrated that Evolon® CR with controlled loading resulted in a decreased solvent retention inside the paint compared to both swab and rigid gel cleaning [19].

Though it has many promising benefits, Evolon® CR remains a new material in conservation practice. Despite the aforementioned research, Evolon® CR's chemical stability and the physical mechanism of solvent uptake and release remain unknown. Moreover, practical and procedural aspects for use by conservators are not yet optimized or fully researched.

This study

This work aims to build upon the existing body of research and focuses on exploring three main questions related to Evolon® CR's material stability, physical mechanism of action and practical considerations for conservators.

1. *Is Evolon® CR chemically inert to the organic solvents ethanol and acetone*, which are commonly used for varnish removal? Is pre-treatment necessary? From a material standpoint, Evolon® CR was not specifically engineered for conservation [20]. Considering the common practice of overnight solvent loading, it is important to understand the material tolerance for solvents and whether chemicals or additives used in the manufacturing process are released from the tissue into the painting during its use.
2. *What is the physical process of solvent uptake and release during controlled loading of Evolon® CR?* Understanding the mechanism of solvent uptake in Evolon® CR is important in developing an accurate protocol for controlled loading. Adsorbed vs.

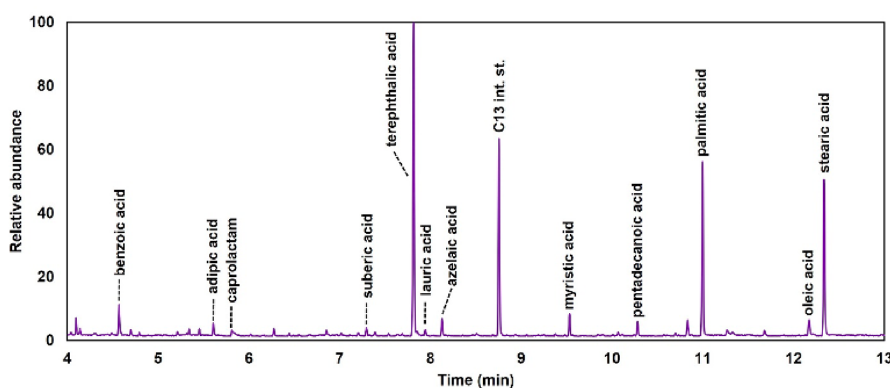

Fig. 1 THM-py-GC/MS chromatogram showing the extracted components from unwashed Evolon® CR

absorbed solvent may not equally be available for subsequent release to the varnish layer on the painting. Understanding the physical process of solvent uptake and to what extent these processes are able to release solvent is critical for obtaining an accurate understanding of the true quantity of solvent the painting will be exposed to.

3. *What is happening in the paint film during and after cleaning with Evolon® CR?* Here we focus on the cleaning process from the painting's perspective, and attempt to understand what is occurring chemically and physically in the dammar varnish and in our zinc white-based oil paint models during and after varnish removal using Evolon® CR.

This work evaluates the first two questions focusing on the chemical stability and physical mechanism of action of Evolon® CR using thermally-assisted hydrolysis and methylation pyrolysis gas chromatography mass spectrometry (THM-Py-GC/MS) to study chemical degradation and optimal pre-treatment procedure, scanning electron microscopy (SEM) to assess physical degradation post solvent exposure, HIROX microscopy to investigate in situ change upon solvent exposure, gravimetric analysis to determine the maximum solvent loading, and nitrogen adsorption and mercury intrusion experiments to determine absorption vs. adsorption processes in solvent uptake. We aim to improve our understanding of the release of solvents by Evolon® CR into mock up paint films and use attenuated total reflection Fourier transform infrared (ATR-FTIR) spectroscopy to investigate the possibility of triggering a reaction between components released by unwashed Evolon® CR and components present in the paint. Portable FT-LSI is used to study solvent retention in paint films, and UV photography to quantify the effectiveness of varnish removal. Finally, an overview of the most important implications for the conservation field will be presented in the conclusion.

Results

THM Py-GCMS

Characterisation of Evolon® CR The characterisation of the composition of Evolon® CR revealed two characteristic monomers: caprolactam and terephthalic acid (TPA) (Fig. 1). Both compounds are commonly used during the production of nylon and polyester fibers [21]. Benzoic acid (BZA), styrene and benzyl derivatives were also detected. Although BZA is used in the manufacture of caprolactam and TPA [22], these fragments mainly originate from TPA fragmentation during pyrolysis [23]. Surprisingly, SFAs, such as adipic acid (2C6), suberic acid (2C8), lauric acid (C12:0), azelaic acid (2C9), myristic acid (C14:0), pentadecanoic acid (C15:0) palmitic acid (C16:0), oleic acid (C18:1), stearic acid (C18:0), were also detected in the chromatogram (Fig. 1). This might suggest the addition of SFAs during the polymerisation process, since SFAs and their aluminium salts are commonly added to tailor the physical properties of many polymers [24].

Influence of washing protocol In order to propose the most optimal pre-treatment prior to the solvent loading step, solvent extracts containing small amounts of monomers extracted from the Evolon® CR tissue were analysed using Py-GC/MS. Measurements were performed on solvent extracts from Evolon® CR after being immersed in acetone or ethanol as part of the washing protocols examined in this study: untreated or machine washed Evolon® CR followed by (1) a single wash with a 24 h solvent exposure time, or (2) five iterative washing steps with a 1 min solvent exposure time (see Materials and methods).

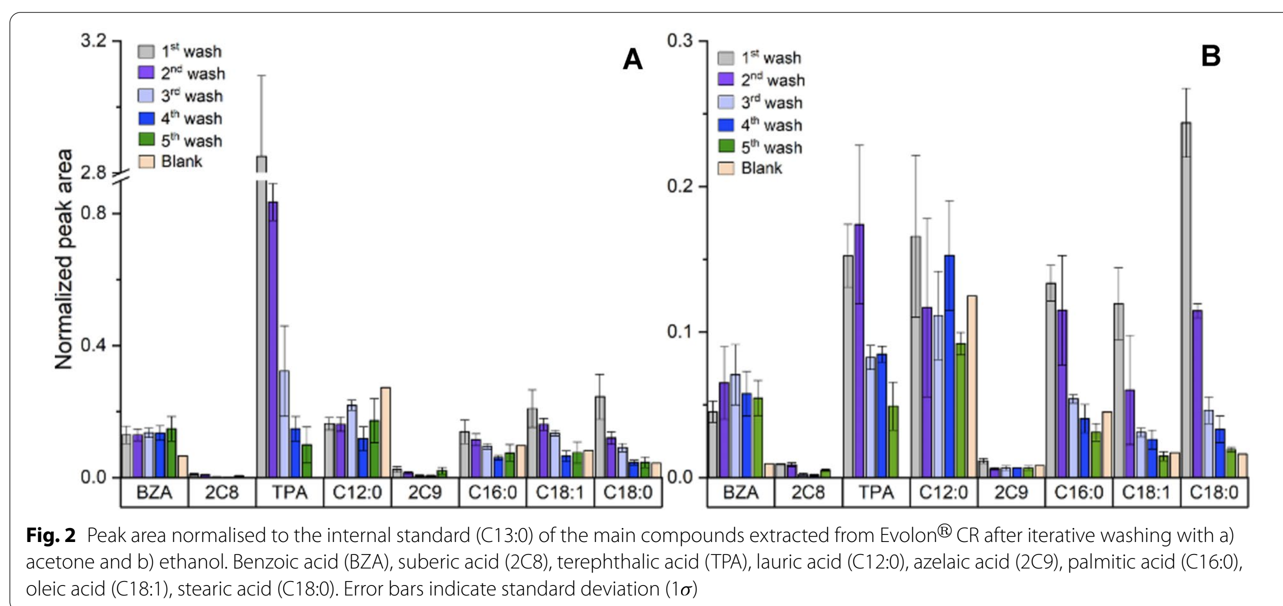

Fig. 2 Peak area normalised to the internal standard (C13:0) of the main compounds extracted from Evolon® CR after iterative washing with a) acetone and b) ethanol. Benzoic acid (BZA), suberic acid (2C8), terephthalic acid (TPA), lauric acid (C12:0), azelaic acid (2C9), palmitic acid (C16:0), oleic acid (C18:1), stearic acid (C18:0). Error bars indicate standard deviation (1σ)

Figure 2a and b show the main compounds extracted from the untreated Evolon® CR after the sequential rinsing with acetone or ethanol normalised to a tridecanoic acid (C13:0) internal standard. Although the pure solvents used in this study also contained SFAs and BZA in small quantities, a clear increase of these compounds was observed after the first washing of the Evolon® CR when compared to the blank. After each solvent exposure, a clear decrease in the extracted amount of TPA, 2C8, C12:0, 2C9, C16:0, C18:1 and C18:0 was observed. However, the amount of BZA remained constant after each extraction with no significant variation observed for the remaining SFAs monitored. After the 5th extraction, the extracted amounts of SFAs were within the same range as the signals for pure solvents, indicating that the tissue no longer contains solvent-releasable components. Thus, five washes appear to be optimal. Although the amounts of TPA found in the extracts are greatly reduced after washing with acetone and ethanol, small amounts of TPA are detected even after three or five washing iterations.

The second washing procedure tested was the full immersion of untreated Evolon® CR in the designated solvent (either acetone or ethanol) for 24 h (Fig. 3). The most significant difference with respect to the iterative washing protocol was the high release of TPA into both acetone and ethanol (Additional file 1: Figs. S1 and S2). It is important to note that after 24 h continuous immersion, the extracted amounts of SFAs are in the same range as those after the 2nd or 3rd iterative wash, indicating that a washing procedure that involves sequential rinsing steps with fresh solvent each time is more effective than one, single long immersion (Additional file 1: Figs. S1 and S2).

By comparing the extraction efficiency of both acetone and ethanol, the data shows that the amount of TPA detected in the solvent extracts was 18 times higher with acetone than with ethanol for 1 min iterative washes, and 5 times higher for the 24 h wash. Moreover, acetone shows a slight increase in SFA extraction efficiency for both washing protocols.

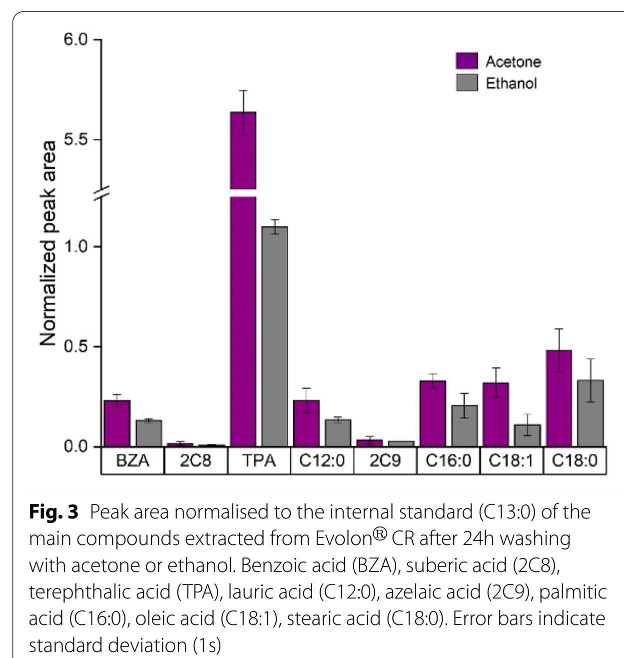

Fig. 3 Peak area normalised to the internal standard (C13:0) of the main compounds extracted from Evolon® CR after 24h washing with acetone or ethanol. Benzoic acid (BZA), suberic acid (2C8), terephthalic acid (TPA), lauric acid (C12:0), azelaic acid (2C9), palmitic acid (C16:0), oleic acid (C18:1), stearic acid (C18:0). Error bars indicate standard deviation (1s)

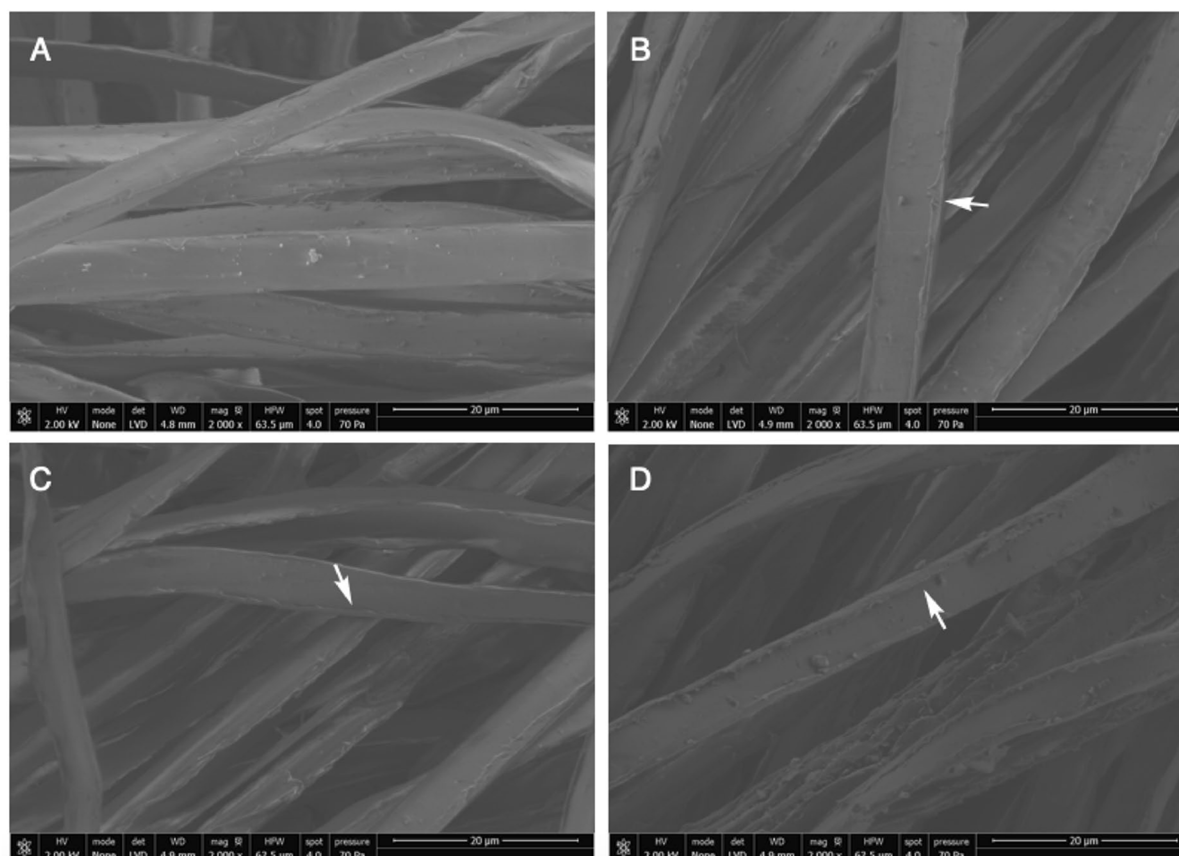

Fig. 4 SEM images at 2000× magnification of **A** no solvent exposure, **B** 1 min exposure, **C** 24 h exposure, and **D** 7 day consecutive exposure in acetone. White arrows indicate fraying along squared edges of individual fibers

The extraction of SFA followed the same behaviour when Evolon® CR was washed in a washing machine at 60 °C (data not shown). The only significant difference was that caprolactam, not detected in any of the solvent extracts of the untreated Evolon® CR, was detected when washed at 60 °C in the washing machine. This finding may be explained by the mechanical action of the washing machine in combination with the elevated temperature of the wash cycle, causing greater extraction of residual monomers from the manufacturing process or hydrolysis of the fibers during washing.

SEM

Scanning electron microscopy (SEM) was used to evaluate potential physical degradation or changes to fiber structure upon solvent exposure to acetone and ethanol. Evolon® CR tissues fully submerged in solvent for 1 min, 24 h, and 7 days were studied.

In general, no fiber rearrangement or reorganisation on a global level was observed after solvent exposure of any duration. The main differences observed were changes to the appearance of individual fibers, with greater solvent exposure times resulting in fraying of the fibers. At 2000× total magnification, it is clear that individual Evolon® CR fibers are square shaped. Fraying manifests itself primarily along the longitudinal edges of the square shaped fibers (Fig. 4B–D).

We observed a distinct difference in fiber-edge fraying in the tissue that had been exposed for 7 consecutive days (Fig. 4D) compared to the tissues exposed for 24 h and 1 min (Fig. 4B and C), but only a subtle difference between tissues exposed for 24 h and 1 min. The tissues exposed for 24 h and 1 min appear similar to the non-solvent exposed control, with the exception of having fewer particulate material adhered on individual fibers in the solvent exposed tissues. This observation could be the result of rinsing of the fibers upon solvent exposure.

The same general effect was observed for tissues exposed to ethanol, except fraying occurs with shorter exposure times (1 min exposed tissues already have frayed edges, see Supplementary Information). The frayed edges of ethanol exposed tissues also generally

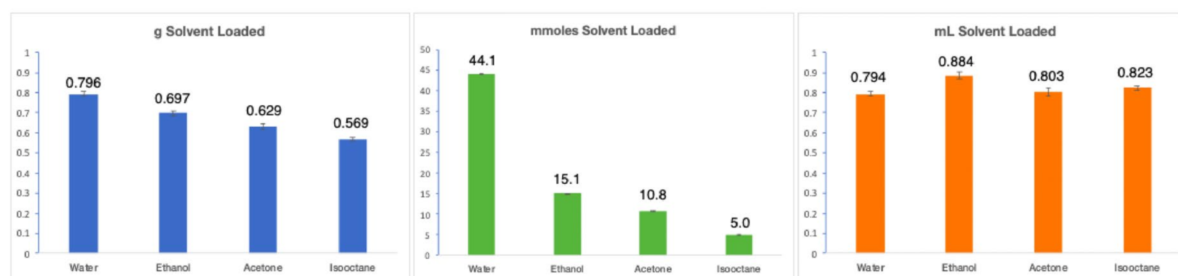

Fig. 5 Graphical representation of total mass, molarity, and volume of solvent taken up by the tissue for four chemically different solvents, demonstrating that maximum absorption appears to be dictated by filling volumetric space

have a smoother look compared to acetone exposed fibers. Finally, we observed little difference between the non-solvent exposed control and the tissue that had been machined washed at 60 °C (Additional file 1: Figs. S3 and S4)).

HIROX microscopy

To compliment the SEM analysis of dried tissues post-solvent exposure, HIROX microscopy was used to visualise any potential in situ change upon solvent wetting with water, ethanol, or acetone.

Average fiber widths for dry fibers were 5.60 μm (± 0.69 μm) and solvent-wetted fibers were 6.03 μm (± 0.83 μm) for water, 6.13 μm (± 1.38 μm) for ethanol, and 6.06 μm (± 0.79 μm) for acetone. All solvent-wetted fibers appear to swell to a similar value (ca. 6.06 μm). While this value represents a slight increase over the dry fiber width, it should be noted that once wetted, fiber edges appear fuzzier. Diameter measurements were taken by measuring the outermost edge to the outermost edge of the fiber, thus this optical effect contributes to an apparent widening of fiber diameters when measured under the microscope. We attribute the measured increase to these optical effects than a true swelling of solvent-wetted fibers. Stereomicrograph images and diameter measurements are shown in Additional file 1: Fig. S5.

Gravimetric analysis

To empirically determine the maximum solvent load of a fully saturated tissue, Evolon® CR tissue was investigated using gravimetric analysis. For analysis, 5×5 cm squares of Evolon® CR were weighed dry, submerged in solvent and reweighed while fully saturated. The solvents evaluated: –water, ethanol, acetone, and isooctane– were chosen for their chemical- and polarity differences.

Weight gain results were averaged over at least five measurements and converted into molarity and volume. Assessing these three methods of representing solvent load - mass, molarity, and volume (Fig. 5)—proved useful. We observed moderate variation when represented in weight (range of 0.535 g to 0.716 g) and large variation when represented as mmoles (range of 4.70 mmol to 39.7 mmol). When represented by volume, the maximum solvent loads appears to be in the range of 0.716–0.798 mL. Figure 5 clearly shows that the volume gain remains fairly constant while weight and molarity strongly vary. The same trend was reproduced when using a slightly modified method of assessing the weight-gain of a fully saturated tissue (Materials and methods).

Nitrogen adsorption and mercury intrusion

To determine the porosity of Evolon® CR, nitrogen adsorption experiments (capable of detecting pore sizes in the sub-nanometer to nanometer range) were performed on the tissue. In these experiments, materials are measured to determine the volume of an adsorbent gas that can be retained in its bulk, which is a quantitative measure of that material's porosity. The adsorption isotherm Fig. 6 obtained for Evolon® CR shows an essentially constant P/P_0 until nitrogen condensation occurs as evidenced by the infinite spike at $P/P_0 = 1$. The very low volume of nitrogen uptake of 0.4 cm^3/g (associated with a surface area of 1.6 m^2/g) and the constant adsorption isotherm suggest that Evolon® CR is effectively non-porous.

To compliment the nitrogen adsorption experiment, mercury (Hg) intrusion was used to analyse pores and voids by filling the sample with Hg at increasing pressure. Full raw data plots (Section 5, Supporting Information) show that the majority of the Hg intrusion is complete at low pressures which is indicative for a large inter-particulate space, and that the intrusion process occurs in a calculated pore size range of 100–1000 nm. The latter finding corroborates the nitrogen adsorption results that the tissue is in fact non-porous and suggests that the

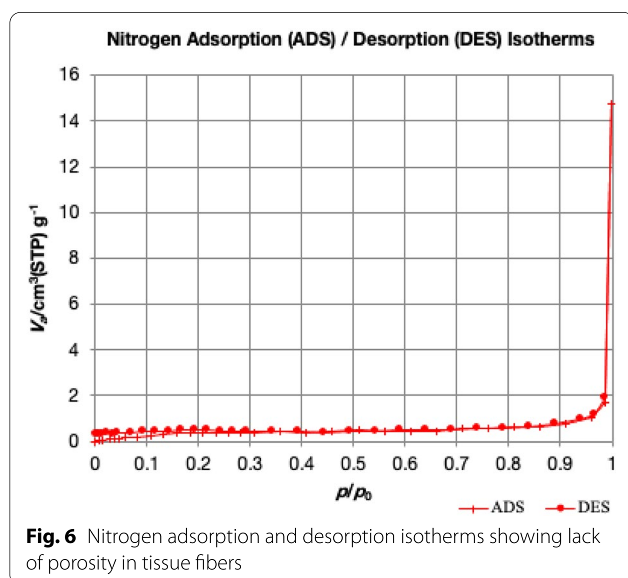

Fig. 6 Nitrogen adsorption and desorption isotherms showing lack of porosity in tissue fibers

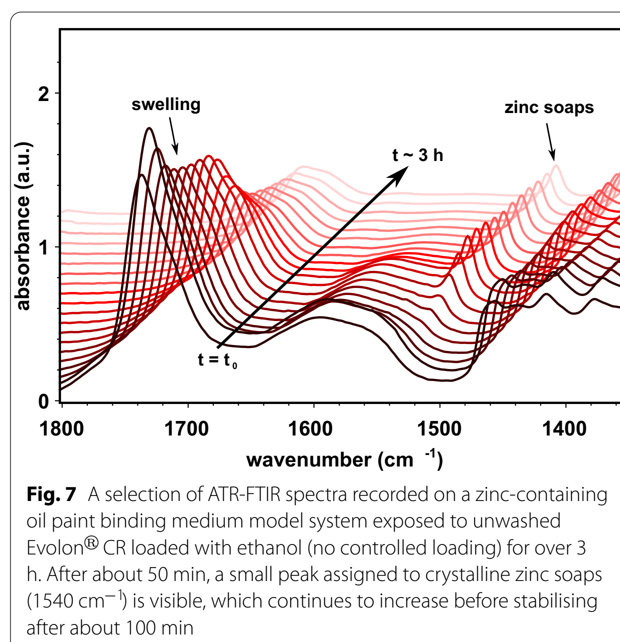

Fig. 7 A selection of ATR-FTIR spectra recorded on a zinc-containing oil paint binding medium model system exposed to unwashed Evolon® CR loaded with ethanol (no controlled loading) for over 3 h. After about 50 min, a small peak assigned to crystalline zinc soaps (1540 cm^{-1}) is visible, which continues to increase before stabilising after about 100 min

intruded Hg is purely filling interstitial space between fibers.

Time-dependent ATR-FTIR

To probe potential chemical reactions of SFAs and other solvent extractable components if released into a paint film during cleaning, time-dependent ATR-FTIR spectroscopy was used to track the development of new functional groups (indicative of chemical reactivity) in paint films during cleaning. A zinc-containing oil paint binding medium model system was exposed to unwashed, ethanol-saturated Evolon® CR.

From Fig. 7, it is clear that crystalline zinc soaps are formed in the zinc ionomer model system when exposed to ethanol-saturated unwashed Evolon® CR. The formation of crystalline zinc soaps was not observed when pure ethanol was delivered to the ionomer films in absence of Evolon [25], demonstrating that untreated Evolon® CR releases enough SFAs to promote zinc soap formation after 50 min of solvent exposure (Fig. 7, sixth spectrum counting from t_0). Attempts to conduct this measurement procedure [25] on Evolon® CR with controlled solvent loading were unsuccessful since the amount of solvent was too small to be detected before evaporation.

Figure 7 shows that the swelling of the zinc ionomer model system (as indicated by the decrease of the ester band at 1738 cm^{-1}) is distinctly different from the swelling profiles measured in previous studies on the uptake of solvents released by conservation cleaning gels [26] and pure solvents [25]. Since the unusual swelling-behaviour only becomes apparent at timescales that are longer than typically used in conservation practice (> 30 min), the in-depth investigation of this phenomenon is beyond the scope of this article.

Portable FT-LSI & UV photography

We here employ portable laser speckle imaging (LSI) to measure and quantify solvent action after application of Evolon® CR on varnished and unvarnished oil paint model systems. Various solvents are tested for the removal of artificially aged dammar varnish (see Materials and methods) and the uptake of solvent by the oil paint models.

LSI can be used to track the motion of light-scattering particles in real-time and spatially-resolved. Using this technique, solvent action inside a painting can be observed in situ [19, 27]. Since translucent solvent molecules do not scatter light, the motion of opaque pigment particles mobilised through the solvation process are measured and used as a proxy for solvent induced dynamics. These dynamics comprise solvent sorption, desorption and evaporation from the paint film, which together define the 'retention time' (the time that solvents are retained inside the paint layer). Knowing the solvent retention time and the rate of signal decay (the rate at which solvents disappear from the paint layer) can help to decrease the risks associated with solvent-based cleaning because these parameters determine how long the paint is plasticised by solvent.

Varnished versus unvarnished paint To investigate the possible 'protective' capacity of varnishes against solvent-uptake by the paint, a series with increasing exposure

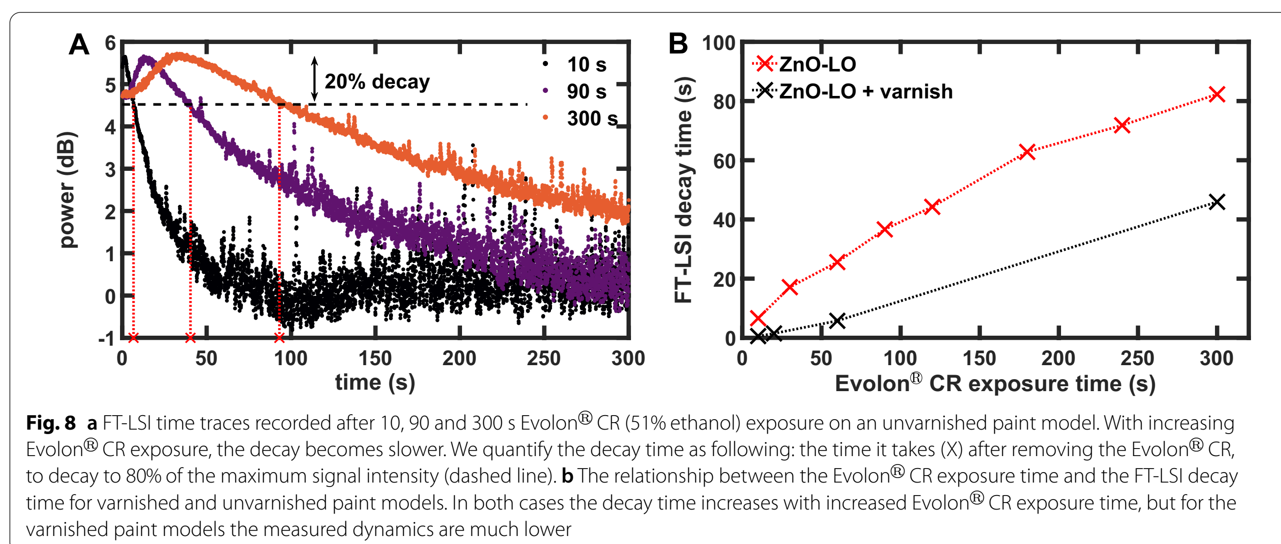

Fig. 8 **a** FT-LSI time traces recorded after 10, 90 and 300 s Evolon® CR (51% ethanol) exposure on an unvarnished paint model. With increasing Evolon® CR exposure, the decay becomes slower. We quantify the decay time as following: the time it takes (X) after removing the Evolon® CR, to decay to 80% of the maximum signal intensity (dashed line). **b** The relationship between the Evolon® CR exposure time and the FT-LSI decay time for varnished and unvarnished paint models. In both cases the decay time increases with increased Evolon® CR exposure time, but for the varnished paint models the measured dynamics are much lower

times of Evolon® CR on varnished and unvarnished paint models was measured and the decay times compared. For these experiments, the Evolon tissue was loaded with ethanol at 51% loading capacity according to the method by Tauber et al. [17].

Investigating the rate of LSI signal decay (referred to here as time trace) for Evolon® CR exposure times of 10, 90 and 300 s (Fig. 8a), it is clear that the intensity of the dynamics within the paint increases with increasing exposure time. The resulting decay times (see Materials and methods) are plotted for different Evolon® CR exposure times in Fig. 8b. The transformation of the time traces in Fig. 8a to the decay times in Fig. 8b is visualised by the red dashed lines in Fig. 8a. Figure 8b shows the decay times for a series of Evolon® CR exposure times on model paints that contain a thin layer of varnish. The measured decay times for varnished paint are consistently lower, indicating that solvent uptake by the paint is slower due to the presence of varnish.

Comparison of solvents An advantage of Evolon® CR is that it can be loaded with different solvents or combinations of solvents to tailor the method to dissolve a specific varnish. We here investigate the intensity and decay of dynamics caused by different solvents using real-time analysis of LSI data for ethanol, acetone and isopropanol. Both varnished and unvarnished paint models are studied and the LSI measurements are combined with UV-photography to assess the effectiveness of varnish removal.

Differences are most easily visualised by plotting the decay time versus the exposure time, as done in Fig. 9a, b. Comparing the decay times obtained for 51%-loaded acetone, ethanol and isopropanol tissues on unvarnished paint (Fig. 9a), large differences are immediately apparent. For example, the decay time for 10 s acetone exposure is roughly equal to the decay time for 90 s ethanol exposure, and slightly higher than 120 s isopropanol. This agrees with our previous result that the diffusion rate is leading in how long the solvent stays in the paint layer [19], and we now confirm that this is also true for controlled application with Evolon® CR.

Figure 9b displays the decay times after application of Evolon® CR loaded with 51% acetone, ethanol and isopropanol on varnished paint models. As expected, the decay time increases with increasing exposure time for all solvents. Comparing the results obtained for model paints without varnish (Fig. 9a) and with varnish (Fig. 9b), the protective capacity of the varnish layers can be observed: the decay time of the solvent signal in the paint is roughly two-times smaller in the presence of varnish.

To asses the effectiveness of the varnish removal procedures we also performed UV-fluorescence imaging on each treated area. The mean intensity value of each treatment, a measure for the amount of varnish present, is shown in Fig. 9c and the collected image in figure 9d. In general, longer exposures result in better varnish removal with acetone being the fastest (40 s), isopropanol the slowest (90 s) and ethanol in the middle (60 s). We did not observe significant differences in the spatial homogeneity of varnish removal.

Both FT-LSI and UV-fluorescence data from cleaning tests contain valuable information that can support conservators in making an informed decision on how to proceed with cleaning a painting. Taking as an example the data obtained from the model system in Fig. 9, we envision that this data could be applied to aid conservation decision making through the following logic: 90 s

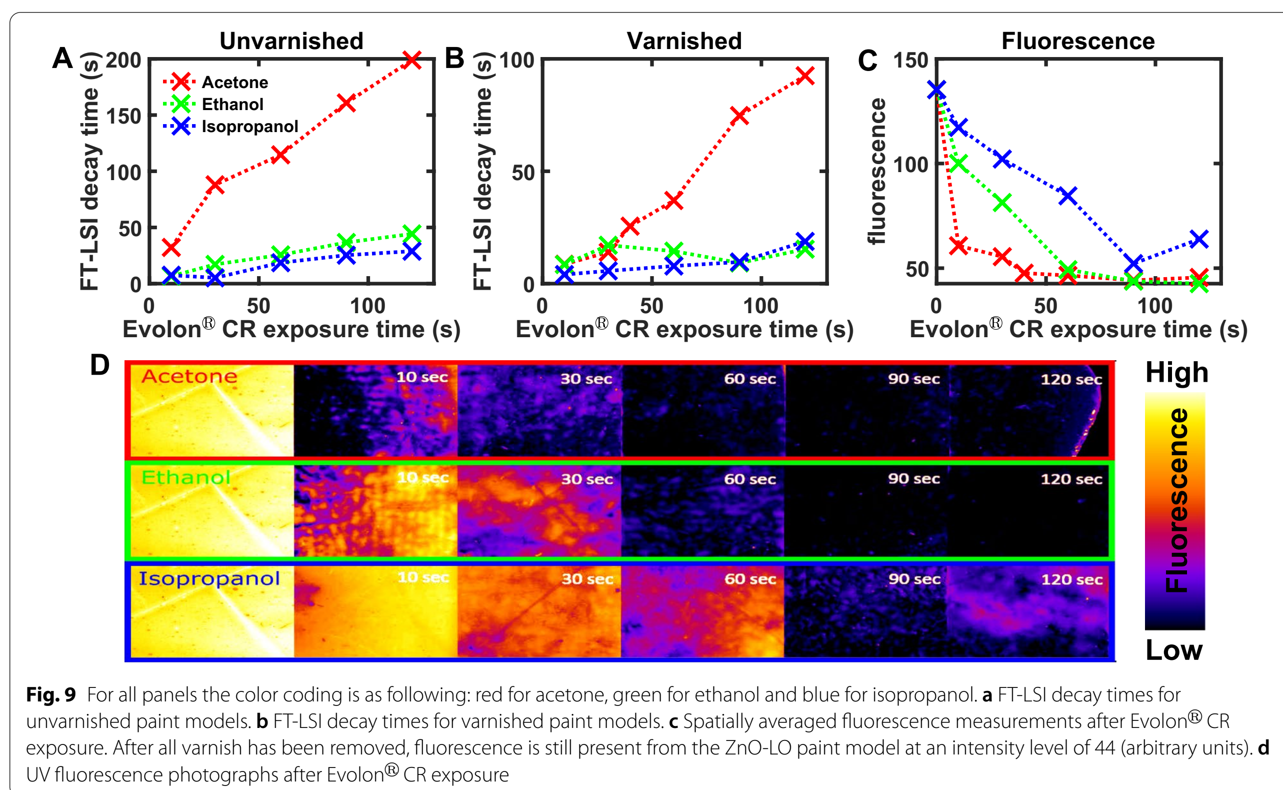

Fig. 9 For all panels the color coding is as following: red for acetone, green for ethanol and blue for isopropanol. **a** FT-LSI decay times for unvarnished paint models. **b** FT-LSI decay times for varnished paint models. **c** Spatially averaged fluorescence measurements after Evolon® CR exposure. After all varnish has been removed, fluorescence is still present from the ZnO-LO paint model at an intensity level of 44 (arbitrary units). **d** UV fluorescence photographs after Evolon® CR exposure

isopropanol is not the cleanest based on UV-fluorescence imaging so it is not further considered; 60 s acetone and 90 s ethanol have comparable UV-fluorescence data and thus good cleaning performance, however acetone has a much higher FT-LSI decay time, thus ethanol is the better choice. Having selected ethanol as the best solvent, routine decisions can be made (i.e. can 60 s instead of 90 s ethanol be used to minimize solvent exposure while maintaining acceptable varnish removal.

Solvent loading A series of measurements using Evolon® CR with different solvent loading was tested on paint models with and without varnish. The loading percentages and procedures used by conservators were taken from the aforementioned publication by Tauber et al. [17]. Figure 10a shows a collection of decay times for Evolon® CR loaded with 24–51% ethanol and one saturated (gently squeezed) Evolon® CR tissue for different application times (10–300 s). Surprisingly, our experiments do not show a significant influence of the solvent loading on the decay time for unvarnished model systems (Fig. 10a). The fact that a higher loading does not result in longer decay times may be explained by either the solvent uptake by the paint being rate-limiting, or by the rate of solvent release from Evolon® CR being independent of the loading. However, the experiments on varnished paint models (Fig. 10b), clearly show that 24% and 34% loadings result in shorter decay times over the whole range of exposure times, compared to 44%, 51% and saturated loading. The inset in Fig. 10c shows an expansion of the UV fluorescence intensity for the most relevant exposure times: 10–60 s. In the first 60 s, the different loadings perform similar in terms of residual fluorescence, which is below 50 (arbitrary intensity) units in all cases. However, the higher decay times in Fig. 10b for 44%, 51% and saturated loading, indicate that more solvent is taken up by the paint. These results lead to the conclusion that for these models 24% and 34% loadings minimize the decay time while retaining optimal varnish removal. We note again that such a procedure is varnish-dependent and will in practice be required to be carried out for every unique varnish layer (assuming different solubilities) and each individual painting.

Discussion

The analyses detailed above provide an improved understanding of key questions regarding the use of Evolon® CR for varnish removal: its chemical stability, the mechanics of solvent uptake and release in the tissue, and solvent action on the paint film.

1. Is Evolon® CR chemically inert to solvents used for varnish removal? Is pre-treatment necessary?

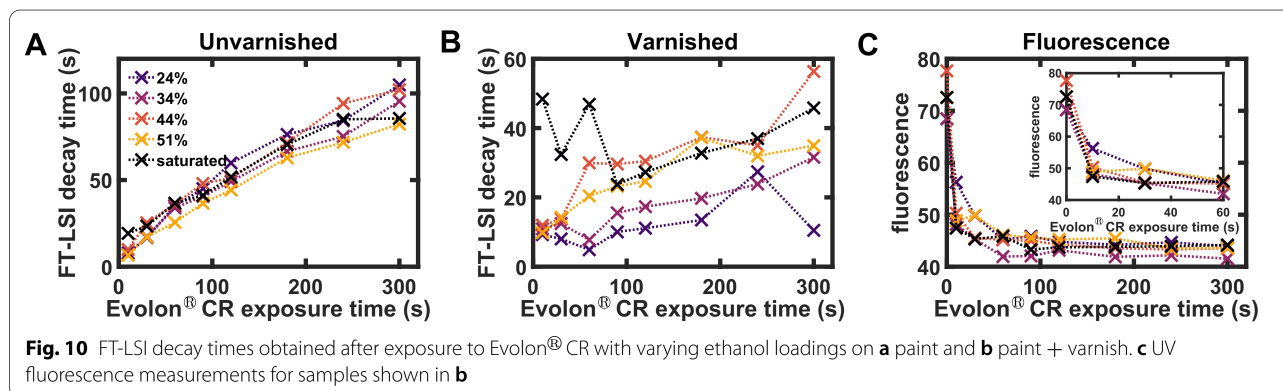

Fig. 10 FT-LSI decay times obtained after exposure to Evolon® CR with varying ethanol loadings on **a** paint and **b** paint + varnish. **c** UV fluorescence measurements for samples shown in **b**

Py-GC/MS and SEM analysis demonstrated that Evolon® CR is chemically and physically stable on typical time-scales used by conservators for varnish removal (less than 24 h), and that acetone extracts greater amounts of monomers compared to ethanol. The effect of long solvent exposure on the physical degradation of Evolon® CR was also confirmed by SEM analyses after immersing the tissue in solvent for 7 days. These results support that solvent exposure of Evolon® CR should be kept to a minimum, and that Evolon® CR should not be consistently reloaded with solvent due to solvent-induced physical degradation of the tissue. To minimize waste, it is currently common practice to re-use Evolon® CR squares previously loaded with solvent. Our results suggest this practice should be avoided considering the physical degradation to the tissue with prolonged solvent exposure. Combined results from Py-GC/MS and ATR-FTIR analyses show that pre-treatment is required to extract SFAs from the Evolon® CR, limiting the risk of unwanted reactions [18] (e.g. metal soap formation) in the painting. Different washing methods/steps were evaluated by Py-GC/MS, showing that the amount of SFAs released from the Evolon® CR depends on the solvent and immersion time. Untreated Evolon® CR followed by iterative washing (at least 3 times) with short time exposures of solvent showed the best compromise between SFA-extraction efficiency and retained physical properties of the tissue.

2. *What is the physical process of solvent intake and release during controlled loading?*

Results obtained from the nitrogen adsorption and Hg intrusion experiments showed that solvent uptake by Evolon® CR occurs predominantly through adsorption in the interstices between fibers, with negligible amounts of absorption into the fibers themselves. Gravimetric analysis also demonstrated that the total volume of solvent loaded was roughly consistent with widely differing solvents, corroborating that the tissue's maximum loading capacity is dictated by the filling of voids in its interstices. This interpretation is supported by HIROX analysis, which showed subtle to negligible amounts of fiber swelling upon wetting.

Our findings suggest that calculations for controlled loading should be based on available volumetric space in fiber interstices. Table 1 highlights the empirically determined maximum solvent load for acetone and ethanol based on volume (taken from the gravimetric analysis experiments) and compares it with those obtained using the currently practised method that a solvent's maximum uptake capacity is 4 times the weight of the utilised Evolon® CR tissue (based on information provided from the manufacturer that Evolon® CR is capable of absorbing up to 4 times its own weight in water). As shown, there is a discrepancy in the calculated maximum load, explained by the fact that water is more dense than both acetone and ethanol. Thus, using water as the benchmark to calculate maximum loading for organic solvents results in an overestimation of the maximum loading capacity.

3. *What happens in the paint film during and after cleaning with Evolon® CR?*

ATR-FTIR measurements demonstrated that zinc soaps can potentially form in the treated paint film after using Evolon® CR that has *not* been pre-washed. It should be noted that although the ionomer model system used in this ATR-FTIR study readily reacts with SFAs, the peaks associated with crystalline zinc soaps were detected only after approximately 50 min of solvent exposure (Figure 7, sixth spectrum counting from t_0). The relatively slow rate of crystalline zinc soap formation may be explained by a slow release of SFAs by the untreated (no pre-washing with ethanol) tissue, a slow rate of diffusion of SFAs in the ionomer film or a combination of these factors. Previous experiments indicated that fast-diffusing solvents, such as acetone, can accelerate SFA diffusion and zinc soap formation [28]. Based on this

Table 1 Comparison of two methods of calculating maximum solvent loading in Evolon® CR; all values reported are an average of measurements on 6 independent squares of tissue

	Maximum loading by interstitial volume		**Maximum loading by weight of Evolon® CR**	
	5 cm x 5 cm tissue	per cm^2 of Evolon® CR	5 cm x 5 cm tissue	per cm^2 of Evolon® CR
Acetone	0.803 mL	0.032 mL	1.059 mL	0.042 mL
Ethanol	0.884 mL	0.035 mL	1.105 mL	0.044 mL

data, we believe SFA diffusion is unlikely to be the rate-determining factor for zinc soap formation.

Finally, we note that although typical contact times between the loaded Evolon® CR tissue and paint surface during a treatment are significantly shorter than 50 min (usually on the order of 1 min), it is still possible that small amounts of SFAs could be released into the paint film potentially inducing metal soap formation over the long-term in the dry state (after the Evolon® CR is removed) [18].

The potential formation of zinc soaps using Evolon® CR with controlled solvent loading was not investigated with ATR-FTIR due to current limitations in experimental set up. Since the extraction of soluble components from the painting is lower for Evolon® CR with controlled solvent loading, [18] and the pre-treatment procedure (washing with ethanol) effectively removes SFAs from Evolon® CR, we assume that the combination of controlled loading and pre-treating the Evolon® CR will lead to a low risk of solvent-mediated metal soap formation.

Specific studies dealing with the possible harmful effects of TPA on paint layers were not found. However, the breakdown of TPA into aliphatic carboxylic acids under strong UV conditions is known to be accelerated by the presence of photoactive pigments such as ZnO or TiO_2 [29]. TPA can also react with zinc ions when heated in dimethylformamide at 100 °C for 4 days [30]. These studies suggest that the possible effects of TPA-deposits on paint surfaces need attention.

The varnished versus unvarnished FT-LSI studies showed consistently lower decay times for varnished model systems, illustrating that the presence of varnish hinders the uptake of solvent in the painting. This important research outcome reflects empirical knowledge of conservators that varnish delays paint swelling and is consistent with previous LSI studies [19]. To give a quantitative representation of this phenomenon, our data shows that the dynamics caused by 120 s Evolon® CR on unvarnished paint is equal to the dynamics caused by 300 s Evolon® CR on top of a thin varnish layer.

The evaluation of various solvents with FT-LSI shows that the signal decay times vary considerably for different solvents. These results are in good agreement with previous results obtained for pure diffusion coefficients of solvents in oil paint models [25]. However, it remains difficult to relate decay times or time traces measured with LSI to pure diffusion coefficients of solvents because the FT-LSI signal is composed of both the swelling of the paint matrix due to solvent sorption and the variation in thermal motions inside the paint. Both these parameters depend on the rate of evaporation of solvents from the paint surface. Since the time traces are in agreement with the order of diffusion and swelling coefficients for solvents, we hypothesise that evaporation is likely not the rate-determining factor for the retention of solvents in paint. Such a situation is expected when the uptake of solvent is much faster than solvent evaporation. It would be interesting to investigate if the (relative) decay times for solvents changes for increasingly porous paints or models on canvas supports, because that would suggest that evaporation becomes dominant for such systems.

Finally, FT-LSI studies on solvent loading demonstrated that when varnish is not yet fully removed, acetone and ethanol cause comparable dynamics, but when the varnish is removed, acetone causes roughly six times longer-lasting dynamics than ethanol. Isopropanol causes the shortest-lasting dynamics, but is also slowest in the removal of aged dammar varnish. It should be noted that when varnish is still present, the LSI signal is composed of both dynamics inside the varnish and dynamics inside the paint and it has thus far not been possible to separate these contributions. Therefore, the LSI signal that is detected before the varnish is fully removed cannot be taken as a pure measure of the dynamics inside paint (Figure 9d). In contrast, the dynamics measured after the varnish is fully removed are fully representative of solvents that plasticise the paint.

We thus arrive at an ideal solvent choice for the removal of varnish from our model systems: ethanol, which combines the generation of relatively low-intensity dynamics and a short retention of solvent in the paint with effective varnish removal within 60 s. It should be noted that this conclusion is valid specifically for the combination of aged dammar varnish on ZnO-based oil paint that we used for our experiments.

Conclusions

As the paintings conservation field has rapidly discovered the potential of Evolon® CR for varnish removal from oil paintings, the need to advance our understanding of this material and the steps necessary for safe and optimal

usage increases. This analytical study contributes to the growing corpus of scientific cleaning studies by researching Evolon® CR's chemical stability, physical mechanisms of solvent uptake and release, and solvent action on a painted surface. The findings presented here have important practical implications that should be considered when proceeding with an Evolon® CR-mediated varnish removal.

Primarily, we suggest a solvent-based pre-treatment of Evolon® CR (as received from the manufacturer) to remove residual SFAs before proceeding with solvent loading for varnish removal. This washing procedure is expected to significantly reduce the probability of Evolon® CR-induced metal soap formation, and is therefore an important step in safeguarding the chemical and structural integrity of the pictorial layers for the future. Studies dealing with the possible harmful effects of residual TPA on paint layers were not found and this topic may need attention.

The current practice of *controlled loading* is still encouraged with the additional caveat that solvent loading should ideally occur the day before intended use, as this work shows that Evolon® CR is chemically and physically stable on timescales of less than 24 h. For the same reasons, our results suggest that during the testing phase pre-solvent loaded, but unused tissue squares should not be dried and re-subjected to controlled loading in an effort to conserve the Evolon® CR tissue. Finally, it was found that solvent-uptake occurs predominantly through adsorption in the interstitial voids. Therefore, to provide greater consistency in solvent loading irrespective of the choice of solvent, a solvent loading method based on solvent volume is preferred over calculating solvent weight or mmoles.

FT-LSI analyses revealed that a higher solvent loading does not result in significantly longer solvent decay times (i.e. longer solvent retention), but rather increasing solvent exposure times *do*. This is particularly observed with prolonged (over)exposure to Evolon® CR, as the initial hindrance of solvent uptake by the paint due to the presence of varnish is eventually overcome. Since acetone induces roughly six times longer-lasting dynamics (i.e. solvent induced motions) of pigment particles in a paint layer as compared to ethanol or isopropanol when used for varnish removal, conservators should be especially reserved with its use if other effective and less invasive solvents are available.

Finally, with regard to the adoption of FT-LSI analyses in conservation practice, FT-LSI in combination with UV photography is a relatively quick method to assess the effectiveness and the retention of solvent inside the paint when evaluating a particular varnish removal method. Since there are no other methods that can quantitatively and non-invasively assess the retention of solvents with high temporal and spatial resolution, our methodology provides a significant improvement in terms of reducing the risks associated with solvent-based varnish removals. However, a few challenges also remain. On a practical level, minimisation of canvas sway and vibration is critical for success in measuring larger paintings during testing. These movements tend to dominate the exceptionally minute displacements detectable by FT-LSI. We have obtained FT-LSI timetraces with comparable quality to those from paint models on small canvas and panel paintings, but for larger canvasses ($> 0.5 \times 0.5$ m), further research is still needed to find optimal adjustments to the setup (e.g. via use of a larger vibration damping table or conservation methods such as the fitting of padded backing boards to minimize canvas sway). The second challenge is accessibility and adoption. We hope that our development of a self-constructed, portable FT-LSI setup, constructed from a commercially available green laser and camera will facilitate easy accessibility of this technique in the future. However, until these limitations have been resolved and FT-LSI secures a definitive place in the conservator's analytical toolkit, tailoring of solvent testing with Evolon® CR will still need to be done empirically through gradual testing of different solvents, solvent loading, and contact times aided by careful visual observation.

This study improved our understanding regarding the use of Evolon® CR for varnish removal and showed that it can be safely employed when adhering to the recommended guidelines. Apart from continuing the development and implementation of FT-LSI for the paintings conservation field, future research avenues could explore less wasteful pre-treatments, and how support (e.g. lined and unlined canvas supports), paint type and structure (e.g. medium rich, porous and wax-resin impregnated paint) influence solvent decay times measured with FT-LSI when using Evolon® CR for varnish removal. Outstanding questions that concern the more practical aspects of using Evolon® CR for varnish removal, such as line formation, will be addressed in future publications.

Materials and methods

THM-Py-GC/MS

Sample preparation First, untreated Evolon® CR tissue was characterised by THM-Py-GC/MS (from now on Py-GC/MS) to identify its monomer composition. 100 µg of untreated Evolon® CR was cut, inserted in an Eco Cup SF (Frontier Laboratories, Japan) and placed with the sampler in the pyrolyzer.

To identify the substances released from the Evolon® CR during the washing process, the solvent extracts were

analysed by Py-GC/MS. Solvent extracts were obtained by exposing 1 × 1 cm of Evolon® CR to 1 mL of the designated solvent for a set period of time. Two washing methods were evaluated: (1) a single 24 h exposure to solvent, and (2) iterative 1 min exposures totalling five iterations per series, wherein the solvent extract is analysed after each iteration. The iterative washing protocol consisted of immersing the Evolon® CR for 1 min in clean, fresh solvent (either acetone or ethanol), letting the tissue dry to completion, and repeating for a total of five washes (see Supplementary Information for detailed procedure). Both methods of washing were set up in triplicate. Solvent extracts were analysed by Py-GC/MS as follows: 100 μL of the solvent extract was taken and evaporated under a stream of nitrogen at room temperature. This dry residue was redissolved in 10 μL of 25 % TMAH in methanol. 3 μL of the final solution were transferred to an Eco Cup SF and placed in the pyrolyzer for analysis.

Another pre-treatment option was explored in parallel; washing the Evolon® CR in a washing machine (at 60 °C, without detergent) as a more environmental and cost-friendly alternative to solvent washing. After this pre-treatment, Evolon® CR was subsequently exposed to solvents according to the methods explained above.

Instrument and analysis parameters Untreated Evolon® CR tissue and solvent extracts were analyzed using a multi-shot pyrolyzer PY-3030D (Frontier Laboratories, Japan) coupled with a Thermo Trace 1310 GC system interfaced to a ISQ 7000 single quadrupole mass spectrometer (Thermo Fisher Scientific, USA). The pyrolyzer was used in the programmable temperature mode; after sample introduction the pyrolyzer was heated from 350 °C to 660 °C at 500 °C/min, total time of 1 min. Pyrolyzer interface was set at 290 °C. The temperature of the SSL injector was 200 °C and the split ratio was 1:32. The septum purge flow was set at 0.5 mL/min. The GC separation was achieved using a Supelco SLB5 MS (20 m × 0.18 mm × 0.18 m) capillary column. Chromatographic conditions: 35 °C (1.5 min), 60 °C/min to 100 °C, 14 °C/min to 250 °C, 20 °C/min to 300 °C (2 min), helium flow rate (range of 0.6–1.3 mL/min). MS parameters: electron impact ionization (EI, 70 eV) in positive mode; ion source temperature was kept at 240 °C; transfer line temperature was 270 °C; scan range 29–600 amu with a dwell time of 0.2 min. Py-GC/MS data was processed with Xcalibur software (Thermo Fisher Scientific, USA) and the NIST 14 mass spectral library.

SEM

Sample preparation Solvent exposed Evolon® CR tissue samples were obtained by exposing 1×1 cm cut squares of Evolon® CR to 1 mL of solvent for a set period of time, and subsequently analysing the solvent exposed tissue. Set time periods were 7 days continuous exposure, 24 h, and 1 min. Additionally, a non-solvent exposed tissue was analysed as a control and a tissue that had been washed in a commercial washing machine at 60 °C (no detergent). All solvent exposure lengths were timed such that all tissues were removed from solvent, prepped for analysis and analysed on the same day. All samples were analysed dry and within 3 h after having been exposed to solvent.

Upon complete drying of each solvent exposed tissue, the tissues were cut into squares of roughly 5 ×5 mm and mounted onto an aluminium slot head stub by double sided copper tape. These discs were then positioned on the analysis stage and inserted into the SEM chamber for analysis.

Instrument and analysis parameters Analyses were performed on an XL30 SFEG electron microscope (FEI, Eindhoven, The Netherlands) on low vacuum with GAD. Images were captured at 2 kV accelerating voltage, 70 Pa, and spot size of 4. For each sample, at least two locations were documented at 350× and 2000× total magnification. On average, one image was captured at 350× as a general overview image of the chosen location, and 2–3 different areas imaged at 2000× to give representative detail images.

HIROX microscopy

Instrument and analysis parameters Optical imaging was performed on a RH-2000 HIROX digital microscope under bright field illumination, and imaged at 140×, 400×, and 700×. fiber measurements were carried out using the HIROX RH-2000 software, and measured at 2000× magnification for the dry and water-wetted tissues, and 1000× magnification for the ethanol- and acetone-wetted tissues. The use of a lower magnification reduced time needed for focusing on individual fibers, which was necessary for the more volatile solvents.

The tissues were laid over a sheet of Mylar (to protect the HIROX analysis stage). The dry tissue was imaged as such and the solvent-wetted tissues were imaged immediately after applying drops of solvent directly onto the tissue. 30 independent measurements were taken and averaged for the dry tissue and water-wetted tissue. 40 independent measurements were taken and averaged for the ethanol- and acetone-wetted tissues. The higher number of measurements was done to compensate for potential inaccuracies in measurement due to fiber movement during the solvent evaporation process. The tissue was regularly re-wetted during the measurement process for both ethanol and acetone.

During experimentation, it was difficult to obtain a good focus at high magnification on individual fibers after wetting, rendering it impossible to track changes to a single fibers as it was being wetted. In the case of acetone, fibers were observed to move subtly after wetting (likely due to acetone's high rate of evaporation). Hence, it was decided to image multiple locations and use average fiber widths.

Gravimetric analysis

Sample preparation Evolon® CR tissue was accurately cut into 5×5 cm squares using a grid and scalpel roller along guide lines. Subsequently, the solvent was poured into a shallow, circular tin plate, with a diameter large enough that the cut squares could float/lie without folding.

Experimental procedure Each cut square was weighed dry on a Sartorius BP211D balance accurate to 0.01 mg, and its weight recorded. The tissue was subsequently laid to float on the surface of the solvent and left to stand until the tissue was fully saturated. With water this process took a few seconds, but with the organic solvents, saturation was immediate. Upon saturation, the tissue was removed by tweezers, and rapidly laid on a clean paper towel to blot dry excess solvent. The blotting process was rapid - solvent soaked Evolon® CR tissue was simply left to make full contact with the paper towel, after which it was immediately lifted off by tweezer and weighed on the balance. No pressure or squeezing was applied to the tissue during the blotting process. To compensate for inherent variation due to rapid evaporation of the organic solvents and the blotting process, the weighing experiment outlined above was done in quintuplicate for each solvent.

To further ensure accuracy in how 'full' saturation was assessed and to ensure our finding were reproducible, a second method was tested in sextuplicate. For this method, the tissue was shaken until drops of excess solvent ceased to drip. At this point only the very bottom corner of the tissue was briefly allowed to touch a dry paper towel to remove the building droplet and the tissue immediately weighed.

Nitrogen adsorption

Sample preparation Analysed tissues were pre-treated under vacuum at 200 °C for 6 h to remove residual moisture from tissue interstices.

Instrument and Analysis Parameters The pre-treated tissues were analysed using a BELSORP-maxII porosimeter using the standard conditions of nitrogen as the adsorptive gas at 77 K. Results were analysed on a BELSORP version 1.1.1.4 software.

Mercury intrusion

Instrument and analysis parameters Evolon® CR tissues directly cut off the roll were analysed on a PASCAL 400 porosimeter using step-wise increases in pressure. The tissue was subjected to incremental increases in pressure up to a maximum of 200 MPa pressure, and the interparticle space calculated using the SOLID version 1.6.5 software.

Time-dependent ATR-FTIR spectroscopy

Time-Dependent ATR-FTIR Spectroscopy was carried out by placing a piece of untreated Evolon® CR immersed in ethanol on top of the zinc ionomer model paint film. With the piece of Evolon® CR on top, the bottom of the zinc ionomer film was then measured continuously for over 3 with ATR-FTIR Spectroscopy. This method and the synthesis of the zinc ionomer binding medium model system (Znpol) is described in more detail in the 'Portable FT-LSI & UV photography section.

ATR-FTIR spectra were measured on a Perkin-Elmer Frontier FT-IR spectrometer fitted with a Pike GladiATR module and a diamond ATR-crystal. Spectra were averaged over 4 scans. For analysis, 5×5 mm squares of the films were cut and lifted off the glass. During time-dependent measurements, the ATR module was flushed with nitrogen to ensure a constant background signal. In order to measure spectra of polymer samples while they were exposed to solvents or solutions, a custom built stainless steel cylinder was used as described in [28]. The cell volume was sealed with two solvent resistant O-rings between the top plate and the pressure clamp of the ATR module. The polymer sample was covered by a $\varnothing = 10$ mm porous sintered metal disk, and a small but constant pressure was applied to the polymer sample by a spring placed between the pressure clamp of the ATR module and the porous disk. The inlet was kept sealed with parafilm during measurements to avoid solvent evaporation.

Portable FT-LSI & UV photography

Sample preparation Model paint samples containing ZnO (Sigma Aldrich, ≥99%) were made by grinding the pigments with cold-pressed untreated linseed oil (Kremer Pigmente) in a 1:1 (w/w) ratio to a smooth paste with mortar and pestle. The Pigment Volume Concentration (PVC) for wet paint was 14% in all samples. The mixture was applied to 50×75 mm glass slides and spread with a draw-down bar to achieve a wet thickness of 190 μm. The samples were cured in the dark in air at 60 °C for 7 days at 97% RH. Humidity was controlled using a saturated K_2SO_4 solution (for 97% RH) in a closed container and was determined using a Rotronic HL-1D temperature and humidity data logger. After drying, the paint

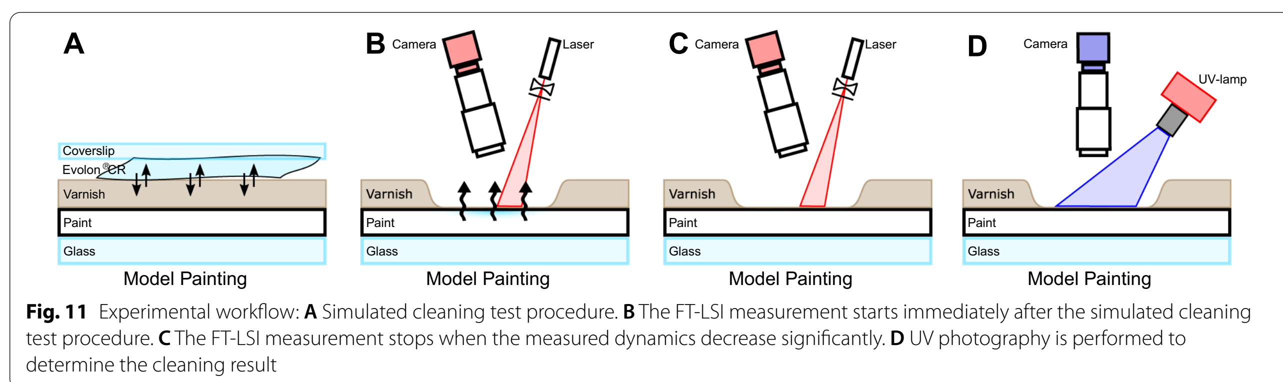

Fig. 11 Experimental workflow: **A** Simulated cleaning test procedure. **B** The FT-LSI measurement starts immediately after the simulated cleaning test procedure. **C** The FT-LSI measurement stops when the measured dynamics decrease significantly. **D** UV photography is performed to determine the cleaning result

reaches a dry thickness of about 150 μm and a dry PVC around 18% for all samples. Samples were varnished using a brush with a dammar solution in 35 wt% Shellsol A and Shellsol T and subsequently aged for 7 days under UV-A and UV-B radiation. The total radiation dosage was 1.4×10^7 J/cm^2 (UV-A) and 5.2×10^7 J/cm^2 (UV-B).

Simulated cleaning test procedure Model oil films were prepared in the same manner as those for ATR-FTIR studies. Evolon® CR tissue was cut into 1×1 cm squares, washed with a large excess of acetone and ethanol using a Buchner funnel, dried and subsequently loaded with solvent.[1] For fully saturated tissues (no controlled loading), the Evolon® CR was dipped into the solvent, squeezed using nitrile gloves and directly used for cleaning. For controlled loaded tissues, loadings of 24%, 34%, 44% or 51% loading were assessed, and prepared as described in Tauber et al. [17]. During solvent application, Evolon was covered with a glass cover slip to avoid solvent evaporation from the top. LSI data acquisition is started immediately upon removal of the Evolon® CR.

FT-LSI Instrument and analysis parameters LSI measurements were performed on a custom-built portable FT-LSI set-up as schematically demonstrated in Figure 11. Previously published FT-LSI analyses as described in Baij and Buijs et al. [19] mostly employed a non-portable, lab-based instrument. We here employ a portable set up which can be easily dismantled and transported for analysis of objects in situ. To our knowledge, this is the first report of portable LSI coupled with real-time analysis and UV photography.

In all measurements, paint surfaces were illuminated by a 685 nm CNI laser at 70 mW, amplified using a single bi-concave lens and ground glass diffuser. The effective light intensity at the sample was 45 mW and covered an area with a diameter of 8 cm, resulting in a light intensity of 0.90 mW/cm^2 on the sample. Back-scattered light was captured using a Navitar variable zoom objective camera, and speckle patterns captured with a Thorlabs DCC3240N camera at medium frame-rates (50 fps). The solvent induced dynamics were quantified in real-time by Fourier transform analysis on speckle fluctuations as previously described in Buijs et al. [27]. The dynamics of the pigment particles (*i.e.* thermal or solvent induced motions) were measured in terms of power. The measured power was expressed in decibel (dB) using dry paint as the reference such that a power above zero dB corresponds directly to solvent-induced dynamics. Slow dynamics were observed by analysing batches of 128 speckle patterns from a 15 fps stream while faster dynamics employed batches of 32 speckle patterns from a 50 fps stream.

The 'decay time' is defined as the time it takes for the measured signal intensity to decay to 80% of its original intensity. A smoothed version of the time trace was used for decay time determination to reduce the effect of noise. For all other purposes the original time-trace was used.

UV-photography After each FT-LSI measurement, varnish removal was determined with UV-photography using a 365 nm Thorlabs UV LED lamp (1150 mW, M365LP1) and Genie Nano M640 Mono camera (640× 480 resolution, 40 ms exposure time). A 400 nm long-pass UV filter (Thorlabs, FEL0400) was used to remove UV light from the lamp and ensure measurement of fluorescence exclusively from the sample. The entire set-up was assembled inside a dark box to avoid interference of other light sources.

Abbreviations

ATR-FTIR: Attenuated total reflection Fourier transform infrared spectroscopy; BZA: Benzoic acid; COOR: Ester; COOM: Metal carboxylate; 2C6: Adipic acid; 2C8: Suberic acid; 2C9: Azelaic acid; C12:0: Lauric acid; C13:0: Tridecanoic acid; C14:0: Myrisitc acid; C15:0: Pentadecanoic acid; C16:0: Palmitic acid; C18:0:

[1] This washing method is not representative of conservation practice, but is expected to yield efficient extraction of SFAs.

Stearic acid; C18:1: Oleic acid; FT-LSI: Fourier transform laser speckle imaging; Hg: Mercury; LO: Linseed oil; LSI: Laser speckle imaging; PVC: Pigment volume ratio; SEM: Scanning electron microscopy; SFA: Saturated fatty acid; THM-Py-GC/MS: Thermally-assisted hydrolysis and methylation pyrolysis gas chromatography mass spectrometry; TMAH: Tetramethylammonium hydroxide; TPA: Terephthalic acid; UV: Ultraviolet; ZnO: Zinc oxide.

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40494-021-00627-9.

Additional file 1. Supplementary information for Understanding and optimizing Evolon® CR for varnish removal from oil paintings.

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Authors' contributions
LB, CL, JB, AAM, LR, JS, and KK designed the experiments. AAM carried out and interpreted the py-GC/MS experiments. KK performed the SEM analysis, and CL, AAM, and KK interpreted the SEM data. CL performed and interpreted the data from the HIROX microscopy and gravimetric analysis experiments. NG performed and interpreted the data from the Nitrogen Adsorption and Mercury Intrusion experiments. LB performed and interpreted the data for the ATR-FTIR experiments. LB, JB, DW, and JS. performed and interpreted the data for the portable FT-LSI and UV Photography experiments. LB, CL, JB, AAM, and LR wrote the manuscript. LB, CL, JB, AAM, LR, PN, and KK edited the manuscript. KK supervised the project. All authors read and approved the final manuscript.

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Availability of data and materials
Supplementary Information contains additional graphical representations and sample preparation information for py-GC/MS, and raw data for SEM, HIROX Microscopy, Nitrogen Adsorption, and Mercury Intrusion experiments.

Declarations

Consent for publication
Not applicable.

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

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