The Wall Paintings of Bamiyan, Afghanistan

Technology and Materials

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Front cover: Seated Buddha figure discovered during conservation intervention at Cave N(a). Photo: H. Otake, courtesy of NRICPT, 2006.

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Contents

Foreword

Located in Eurasia's centre, Afghanistan is an isolated mountainous region with a culture that has persisted through the vicissitudes of its rich and varied history. The Bamiyan Buddhist archaeological site is the product of a people who built an 800-year-old kingdom in the Hindu Kush and Koh-i Baba mountain ranges that, in the words of Herodotus, was a lasting 'gift of peace'.

In 1964, the Silk Road was peaceful and vibrant, and Bamiyan was bustling with travellers coming and going from the east, the west and the south. Travellers looked up at the Eastern and Western Giant Buddhas towering over the steep southern cliffs of the Hindu Kush and marvelled at the wonder of the iconography on the surrounding walls. In July of that year, on a crisp summer night, I was dreaming by the light of the lamp in my lodgings at Bamiyan when I felt a mysterious energy emanating from one of the hundreds of caves. It was an invitation to visit Cave N.

Cave N, which had survived the surrounding destruction, welcomed me again in 2005 when I brought along Dr Yoko Taniguchi, a young colleague and friend. The Bodhisattva of Cave N welcomed us with a faint smile as we gazed upon it with bright eyes. Yoko's keen discernment recognised that the Bodhisattva's expression contained something not yet found in prior research here: Cave N was filled with wall paintings that carried the flourish of the Middle Ages with their exuberant motifs, uninhibited style, variations in grounds and culturême (crossing over of the elements). Each of these stirred her gaze. As Aby Warburg, the German art historian and cultural theorist commented: 'God is in the details'.

Research in 1923 on the Bamiyan Buddhist site, which began with the archaeological survey by the French archaeological delegation in Afghanistan, DAFA (Délégation archéologique française en Afghanistan), has already spanned a century, albeit intermittently. Research on the wall paintings has deepened from its initial examination of motifs and styles, to later analysis and interpretation of their meaning, and now to today's reconstruction of 'visual écriture' based on precise scientific analyses.

The Wall Paintings of Bamiyan, Afghanistan: Technology and Materials offers a marvellous opportunity for us to refresh our understanding of Afghanistan as it stands: a model of a splendid polyphonic culture, even amidst its current ongoing struggle against hardship.

Kosaku Maeda
May 2022

Professor Maeda points out (to the author) the site of an ancient Buddhist monastery in the Bamiyan Valley. (Photo taken in Cave D, 29 June 2007)

Preface

I visited the Bamiyan site as a team member of the National Research Institute for Cultural Properties, Tokyo (NRICPT), conducting research and conservation work on wall paintings in the summer/winter of 2005, spring/autumn of 2006 and the summers of 2007, 2010 and 2013 with Professors Kosaku Maeda and Kazuya Yamauchi, as well as other colleagues, as part of the UNESCO Japanese Funds-in-Trusts project for safeguarding the Bamiyan site. Security issues in the summers of 2009 and 2012 prevented us from fulfilling our plans to travel to the site, and from 2013 on, security had deteriorated to such an extent that it was not possible to even plan any further surveys.

The demolition of the Giant Buddhas in 2001 marked a rise in the use of cultural heritage in political propaganda. Not only were the Buddhas repeatedly hit with explosives, but wall paintings in surrounding caves were also destroyed and later stolen, piece by piece, to be sold abroad. One project to safeguard the Bamiyan site was launched under UNESCO's leadership in 2003. Institutions from Germany, Italy and Japan undertook core roles in the project, and I worked as a research fellow at Japan's lead institution: the Research Center for International Cooperation in Conservation at NRICPT. I was asked to analyse the composition of wall paintings at the Bamiyan site to better determine appropriate conservation materials and methodologies.

At the time that I started in 2004, 80% of Bamiyan's wall paintings had been lost relative to a count from the 1970s. I was informed by my superiors that it would be dangerous to leave the artworks in place and that the remaining wall paintings should be detached and stored at a safe site for the duration of conservation activities. However, we insisted on preserving the wall paintings *in situ* owing to our belief that it was imperative they remain in place within their landscape, carved into the conglomerate cliffs, and to our hope that local people would have access to their cultural heritage for hundreds of years to come. For these reasons, we left the few remaining wall paintings in place together with the damage they had incurred from bullets and blades.

The Bamiyan site has suffered numerous disasters throughout its history, and we do not yet know how much longer it will remain intact. Afghanistan has been ruled by various political powers throughout its history, possibly more than most places in the world due to its central location between Eastern and Western Eurasia. The Buddhist era is only one brief part of Afghanistan's long and complicated history. I believe that Afghanistan's mosaic history is imbued with rich cultural depth and allure.

Since 2005, I have worked in numerous laboratories with many collaborators using myriad analytical techniques in research on painting materials. In the winter of 2006, together with Marine Cotte and Emilie Checroun, I was researching painted materials at the European Synchrotron Radiation Facility (ESRF) where we obtained astonishing results from minute samples taken from a damaged site. The discovery of oil paintings in Bamiyan, which captured the oldest example of oil-based paints in the world, evidences the remarkable academic value of Afghanistan's cultural heritage. Moreover, many more of our material and technical discoveries offer a fascinating window into Bamiyan's historical connections to regions to its west, south and east on the Eurasian continent.

Stemming from our involvement in the conservation of cultural heritage in West and Central Asia, we feel that local education is of paramount importance. For people, learning about the history and culture of their region can bolster respect for their identity and improve historical awareness. While we were conducting our research, numerous young people affiliated with Bamiyan University repeatedly asked us to teach them about Bamiyan's history, as they were being taught very little. We held a small workshop that inspired a lively discussion among the many local youth in attendance. I have embraced writing this book in the hope that it provides them with further inroads to the joys and wonder of their own cultural heritage. Our only regret is that the book was not published sooner, as nearly 10 years have passed since our analyses and results were initially compiled.

What is certain is that the Buddhist wall paintings of Bamiyan are remarkably valuable for the abundant information they carry on the history of painting and its technologies. Oil paintings were once believed to have emerged in their most perfected form in the first half of the 15th century in the Netherlands (although it was known that this was undoubtedly a much older technique). However, Bamiyan hosts the world's earliest known examples, dating back to the mid-7th century. Many hope that a broader geographic scope of analysis pertaining to mural painting techniques and materials will offer greater insight into the Buddhist world under the umbrella of the Hephtalites, spanning the Gupta dynasty in the south, the Roman and Sasanian Empires in the west, and various nomadic states in the east. I personally wish to see further analysis of techniques and materials to help us understand how Bamiyan interacted with its neighbouring regions. We also hope that it will give Afghans greater opportunity to know and express pride in their cultural heritage. Any lack of understanding found in this book is the responsibility of the author. I would like to express our sincere gratitude to the many people who have helped to produce it.

We sincerely hope that our friends in Afghanistan will not suffer too much and that we can work together to help rebuild Afghanistan in the near future.

Yoko Taniguchi
November 2022

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We would like to express sincere gratitude to all the people who joined us on this journey, and especially to colleagues and friends in Afghanistan and across the world who have participated in this work which was supported by JSPS Kakenhi Grant numbers JP18700680, JP21700845 and JP17K01192, and Grant-in-Aid for Publication of Scientific Research Results in 2022. The Bamiyan wall painting project has been supported by the National Research Institute for Cultural Properties, Tokyo (NRICPT) as a part of safeguarding projects by Japanese Funds-in-Trust UNESCO project, and Culture of Agency, Ministry of Foreign Affairs, Japan.

I would especially like to thank Professor Kazuya Yamauchi, as a leader of the Bamiyan project. Sadly, Professor Kosaku Maeda, who was a guardian of our study and project as a cornerstone and source of knowledge, passed away during production of this book. He will be greatly missed.

Apart from work at the European Synchrotron Radiation Facility (ESRF) in France, the Getty Conservation Institute (GCI) in the USA kindly analysed select paint samples from the wall paintings to help us gain additional insight into binding media found at Bamiyan. I owe a special debt of gratitude to the GCI, especially to Joy Mazurek, the director Tim Whalen and to the former head of GCI Science Giacomo Chiari. Without my study period at the Getty in 2007, this binding media study would not have been possible. In addition, Prof. Takashi Nakazawa of Nara Women's University, Prof. Kazuki Kawahara of Osaka University, Yuki Kumazawa and Yuki Taga of Nippi Research Institute of Biomatrix, and Miho Takashima of National Museum of Western Art have contributed extensively to the challenging analyses of organic substances from wall paintings.

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Yoko Tanuguchi

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Marine Cotte

Introduction

Sadly, repeated vandalism has caused significant damage to the Giant Buddhas of Bamiyan in Afghanistan, as well as to the wall paintings in their vicinity. The devastation began with the Mongol invasions of the 13th and 14th centuries, and continued more recently in 2001 with the use of explosives and rockets against the Eastern and Western Giant Buddhas. At that time, all the wall surfaces in the niches surrounding the Giant Buddhas were destroyed while countless wall surfaces in nearby caves were also devastated and looted. Today, very little remains of the Eastern and Western Giant Buddhas and over 80% of the wall paintings in their surrounding grottoes have been lost.

This book presents important data collected through chemical analyses aimed at determining the materials and techniques used in the Buddhist wall paintings at Bamiyan. It discusses the results of these studies from a broad perspective, addressing the cultural interactions between the geographical east and west of the Eurasian continent and includes:

- an overview of research previously conducted on Bamiyan sites over the last 1500 years;
- an overview of the techniques and materials used in wall paintings across Central Asia;
- a discussion of physical and chemical analyses of Bamiyan's wall paintings;
- an insight into connections between cave styles and radiocarbon dates, and how these reflect changes in Buddhism and oil painting techniques introduced via the Silk Road.

This study is based on research carried out by the National Research Institute for Cultural Properties, Tokyo (NRICPT) from 2003 to 2007 within the framework of the Bamiyan Safeguarding Project,[1] based on an interest in wall painting conservation and preservation of the two Giant Buddhas and the site (Petzet 2009; Toubekis *et al.* 2011; Nagaoka 2020). First, a comprehensive survey was conducted targeting Buddhist wall paintings across four cave groups in the Bamiyan Valley using scientific methods to identify painting materials and manufacturing techniques. These results are discussed from a broad perspective, honouring the significance of centuries of transcontinental intercultural exchanges along the Silk Road.

Bamiyan's Buddhist wall paintings were produced over the course of about five centuries, from the time that Buddhist monasteries began to emerge in Bamiyan from the early 5th century until the end of the 9th, when Buddhism's influence waned. As cave temples appeared sporadically in this region over hundreds of years, techniques and materials underlying the production of wall paintings shifted alongside patterns of trade and dominant schools of Buddhism.

This research was originally launched as part of an effort to preserve what remained of the wall paintings. Early on, we sought to identify the original materials and techniques to best determine how to clean and protect the wall paintings without causing further damage. From this research, it became clear that Bamiyan's cave walls had been decorated in a variety of colours using painting techniques more complex than previously thought. Today, as these archaeological sites are threatened by human destruction, very minute samples of wall fragments have become the source of a great deal of valuable new information – so vast, in fact, that it would take years to analyse it all. Therefore, this study deals only with the data compiled so far.

This study was the first to detect drying oil in Buddhist wall paintings in Central Asia, confirming a tempera painting technique. This book offers a series of exhaustive analyses to illustrate comprehensively the structure of the wall paintings. It seems that the oil painting techniques involved first appeared in Bamiyan in the mid-7th century. Bamiyan's earliest oil paintings currently stand as the oldest examples of oil painting in the world, not just among those of Buddhist origin in Central Asia.

The paintings in Bamiyan not only used drying oils as a binding medium, but also involved the sophisticated use of organic materials such as gum and proteins in multiple layers that included glazing and sizing. The use of lead white – an artificially processed white substance made from lead, together with drying oil and possibly associated with lead oxide – succeeded in achieving a distinctive white colour and facilitated polymerisation of the drying oil due to lead oxide's function as a drying agent. One technique of applying natural resin over tin foil in imitation of gold leaf was also very similar to the medieval European *mecca* technique. These materials and techniques are neither accidental nor primitive but sophisticated and well planned. They are also historically important evidence of active networks of exchange linking Central Asia and medieval Northern Europe, from where it was once believed that oil paintings originated. In particular, the technique's presence in Central Asian Buddhist repertoires raises groundbreaking questions.

Objectives and significance of the research on painting techniques

Background and related literature on Central Asian wall painting techniques

Wall paintings are a kind of immovable property adapted to local climates and produced from materials available in nearby regions, as seen with lime in frescoes (*affresco*) and clay in walls. The colours used in the frescoes combine locally available materials, such as pigments and binding media, with other materials, techniques and skills available through trade routes. Mobile, professional artisans such as the *ebusshi* (Buddhist painters) incorporated artistic ideas, materials and skills from throughout the region into their wall paintings. This reflects a technical background that differs slightly from that of worshippers and practices between religious groups and tribes.

Various historical art and archaeological studies of the Buddhist wall paintings at the Bamiyan site, as well as those at the Kizil and Bezeklik grottoes in the Tarim Basin (today's Taklamakan Desert in China's Xinjiang Uygur Autonomous Region), have shown that these wall painting techniques developed under Indian, Persian, Hellenistic Roman and Chinese influences, and then spread along the Silk Road through economic and cultural exchange. Afghanistan, in particular, may be regarded as 'the missing link on the route between the eastward spread of Greek culture, the westward spread of Indian Buddhist culture, and the spread of mixed Buddhist art to China' (Miyaji 2002: 42). For this reason, it presents a culturally complex picture. In recent years, the possibility that the motifs of the early Bamiyan cave paintings are related to western China has also been discussed (Iwai 2008).

Afghanistan is a region with a very cosmopolitan history. The painting materials, tools and methods employed to produce the Buddhist wall paintings reflected the best techniques of painters and artisans recruited by wealthy donors who funded the construction of Buddhist monasteries and complexes far and wide. The materials used were of the highest quality, and transported along the Silk Road together with knowledge of new techniques from various regions across the East and the West. In addition to detailed analysis of individual sites, this book offers extensive consideration of the geographies of Central Asia, the origins of raw materials and the routes of material exchange in ancient times.

There are no records of the painting techniques employed at Bamiyan and the Tarim Basin, like ancient technical treatises written by contemporaneous painters. Therefore, we employ direct analysis of what remains of the wall paintings today to understand the techniques and materials that were used historically. For additional insight, we also refer to sources such as *De lapidibus* by Theophrastus (4th century BC), *De architectura libri decem* by Vitruvius (1st century BC), *Materia medica* by Dioscorides (1st century AD), *Naturalis historia* by Pliny the Elder (1st century AD), and the *Citrasutra* of the Gupta dynasty in India. The Roman texts include information on architecture, medicine and alchemist craftsmanship, as well as on minerals and plants, all of which closely relate to materials used in painting. Written hundreds of years before Bamiyan's cave paintings emerged, these records show that many of the materials and techniques used were already known in Central Asia and the wider Eastern world.

Several South Asian texts have also described painting techniques. One representative treatise on art is the 7th-century compilation *Vishnudharmottara*,[2] a supplement to the *Vishnupurāna*, which contains dialogues by sages on pictorial styles (Kramrisch 1928; Ueno 1973: 398; Sadakane 1988). In one sutra, the *Citrasutra* (*Sutra of Painting*) (Sivaramamurti 1978; Nardi 2006: 120–142), we find an important reference to painting techniques. Moreover, it should be noted that the sutra developed from artistic views of the Gupta period (AD 320–600), when Hinduism flourished.

In contrast to the Greek and Roman accounts, which contain a great deal of geographical and technical information, the treatises present in India today

Table 1 List of major painting materials used in this book: common names of pigments, mineral names and chemical compositions.

Colours	*Pigment*	*Mineral*	*Chemical formula*
Whites	Gypsum	Gypsum	$CaSO_4.2H_2O$
	Anhydrite	Anhydrite	$CaSO_4$
	Chalk	Calcite	$CaCO_3$
	Lead white	Hydrocerussite/cerussite	$2PbCO_3.Pb(OH)_2$ / $PbCO_3$
	Kaolinite	Kaolinite	$Al_2Si_2O_5(OH)_4$
	Talc	Talc	$Mg_3Si_4O_{10}(OH)_2$
Yellows	Yellow ochre	Goethite	α-$FeO(OH)$
	Orpiment	Orpiment	As_2S_3
	Massicot	Massicot	PbO
	Litharge	Litharge	PbO
	Gamboge		natural resin from Guttiferae trees
Reds	Lead red	Minium	Pb_3O_4
	Red ochre	Haematite	α-Fe_2O_3
	Cinnabar	Cinnabar	HgS
	Vermilion	Cinnabar	HgS
	Realgar	Realgar	α-As_4S_4
	Madder lake		$C_{14}H_8O_4$
Blues	Lapis lazuli (ultramarine)	Lazurite (calcite, pyrite as impurities)	$Na_7Al_6Si_6O_{24}S_3$
	Azurite	Azurite	$2CuCO_3.Cu(OH)_2$
	Indigo		$C_{16}H_{10}N_2O_2$
Greens	Malachite	Malachite	$CuCO_3.Cu(OH)_2$
	Atacamite	Atacamite, paratacamite	$Cu_2Cl(OH)_3$ $Cu_2(OH)_3Cl$
	Verdigris	Copper acetate	$Cu(C_2H_3O_2)_2.2Cu(OH)_2$
	Verdigris	Copper resinate	$Cu(C_{19}H_{29}COO)_2$
	Chrysocolla	Chrysocolla	$(Cu,Al)_2H_2Si_2O_5(OH)_4\ n(H_2O)$
	Green earth	Celadonite	$K(Mg,Fe)(Fe,Al)Si_4O_{10}(OH)_2$
	Green earth	Glauconite	$(K,Na,Ca)_{1.2-2.0}(Fe^{+3},Al,Fe^{+2},Mg)_4\ (Si_{7-7.6}Al_{1-0.4}O_{20})(OH)_4\cdot 7nH_20$
Blacks	Bone black		$C + Ca_3(PO_4)_2$
	Charcoal black (carbon black)		C
	Lamp black		C
	Magnetite	Magnetite	Fe_3O_4
Metal leaves	Gold leaf	Gold	Au
	Tin leaf	Tin	Sn + Pb (impurity)

are canons of divinity on Buddhist and Hindu deities and lack specific material and technical detail. However, only the *Citrasutra* describes the technique of painting on earthen renders, as opposed to the Greek and Latin texts that specify painting only on lime or marble.

Significance of the study of painting techniques and view of the present study

Painting is the process of adding colour to an object or surface, with paint being one of the most common materials used for this purpose. Paints are particles of colour or pigments that have been blended with a binding medium, defined as an organic substance with an adhesive effect. In order to paint a surface (such as a wall, canvas or board) with colours, it is essential to have knowledge of, and experience with, paints. For example, the most commonly used pigments are powders, each with a specific colour, that are made from various substances such as natural minerals, earths, artificially synthesised metal salts and organic dyes. There are two main types: inorganic pigments made from sand or stone such as ochre and cinnabar (mercury sulfide), and organic pigments produced from plants and animals, for example madder roots, cochineal, indigo and gamboge.

Natural inorganic pigments (e.g. malachite and cinnabar), which are obtained by crushing minerals into a coloured powder, are well known. However, minerals with bright, deep colours (e.g. blue, green and yellow) are particularly precious and must have been very difficult to obtain. In some cases, the value of the pigment was enhanced by its rarity, as it could only be found in certain areas, as was the case with the green earth of Cyprus or the lapis lazuli of Afghanistan's Badakhshan region and the Kokcha River basin. The wisdom and alchemy of knowing which colours were available from where, or what mixtures could produce which colours, was of great interest to Egyptian, Greek and Roman painters, as documented as early as Roman times. For this reason, inorganic and organic synthetic pigments were used as substitutes for natural minerals that remained out of reach (Table 1).

Table 2 The relationship between the type of glue (organic material) and the painting technique.

	Binding Media	*Painting Techniques*
Sap or other resinous material associated with plants	a Gums (polysaccharide): Gum arabic, guar, tragacanth gum, etc. b Gums (resinous): Cherry, plum gums, etc. c Resins	Watercolour Tempera
Proteins	Animal glue Egg Milk Casein	Tempera
Drying oils	Linseed, poppy, walnut, tung, perilla, safflower, sunflower oils	Oil paintings
Polymeric carbohydrate consisting of glucose units, produced by plants	Starch	

For example, Vitruvius and Plinius the Elder demonstrated the chemical reaction of lead at Rhodes by pouring vinegar over a lump of lead and keeping the vat closed to produce a white powder, *psimithium*[3] (*Naturalis historia* XXXIV: ch. 54), which could be further burnt to produce a red lead. Alternatively, the same process produces green verdigris (copper acetate) when copper is substituted for lead[4] (*Naturalis historia* XXXIV: ch. 26; Rowland and Howe 1999: 94). Other examples include the creation of artificial barium copper silicate and calcium copper silicate in imitation of hard-to-find natural minerals that are ground into a blue powder known as Han blue or Egyptian blue. Instead of using the precious natural cinnabar (HgS), which gives a bright red colour, artificial vermilion was produced by heating sulfur and mercury. Furthermore, when some pigments are mixed with certain others, they may produce discoloration because of chemical incompatibilities or optical problems relating to refractive indices, issues that will ultimately prevent the desired colours from being obtained. Therefore, experience and knowledge of these pigments were crucial for ancient painters.

While synthetic inorganic pigments may be prepared from natural minerals, or produced by transforming metals or glass into coloured substances, organic pigments of animal or plant origin were also widely used. Organic pigments are insoluble organometallic complexes obtained by depositing a coloured dye of animal or plant origin – for example, cochineal, saffron or madder – on an extender pigment (i.e. an inorganic substrate such as calcite or alumina). Depending on the method of extraction, the refining process and the substance on which they are deposited, a variety of colours can be produced and used in paintings, crafts and textiles.

The use of pigments as paint therefore requires an adhesive, binder or medium to adhere them to the surface. Five techniques are of importance: *buon fresco* involves the reaction between a lime mortar and carbon dioxide in the air to produce a calcium carbonate used as a binder; *watercolour* uses a water-soluble gum arabic as a binder; *tempera* employs proteins such as egg, animal glue and casein; *Enkaustik* (encaustic) utilises beeswax as a binder (Table 2); and lastly *oil painting* uses a type of oil (known as a drying oil) that dries by oxidative polymerisation when exposed to oxygen. In a broader sense, tempera techniques involve the application of various minerals, synthetic metal salts and earthen ochres, while animal glue is used as a binder.

When making paint, artists take into account the differences in colour and hue caused by the relationship between the pigment's refractive index and an adhesive, or the uniformity in thickness of the coloured film, among other things. Various additives and extender pigments containing white particles (in an inorganic matrix) may be mixed with the paint. For example, a variety of red colours can be produced by layering or mixing different hues of red. The painter's palette is only complete when the most suitable paints for the desired hue and painting technique have been found. In other words, colour is the result of a complex process that involves blending pigments, binding media and various additives, which are then applied to a surface as paint.

Most prior research on materials used in wall paintings, especially in Asia, has concerned pigments. While there is abundant elemental analysis of inorganic pigments available – primarily mineral pigments – there are few examples of research on organic materials in pigments, binding media, organometallic salts, glazes, buffer layers and sizing (i.e. a preparatory layer that prevents the paint from penetrating the wall and improves brushstrokes) due to the technical difficulties inherent to analysis.

To reveal the techniques and technologies embedded in painting – including for the purpose of preservation – we must identify not only inorganic pigments, but also analyse organic substances, examine the relationship between the paint and its supports, recognise how pigment particles relate to binding media, and study layering, tools and the order of painting, while also determining states of deterioration and the origin of any degradation. Only after such extensive work may we confidently clarify the painting techniques used.

Significant progress has been made involving research on painting techniques and materials, owing largely to the rapid development of analytical methods over the past 20 years. This progress is rooted in the increased use of instruments capable of analysing small samples of organic and other materials in investigations of cultural heritage, as well as in new methods for analysing small samples of inorganic materials with high precision.

The application of such analytical methods to the study of cultural heritage seems to have started in the European Union, where multidisciplinary research bodies have engaged in cross-border and academic research. Such bodies also carry out analytical chemical research on cultural heritage in close collaboration with a wide range of more specialised (e.g. in physics, chemistry and biology) research organisations. In addition, non-invasive methods have become the norm thanks to the development of analytical instruments and optical methods such as multispectral imaging, thereby allowing for a quick and easy survey of multiple sites without physically disturbing the materials.

The methodological limitations of non-invasive analysis, despite its increased accuracy, mean that in some cases it is necessary to use minute samples in conjunction with an analysis. The amount of information that can be obtained from such small samples, even if only a few micrograms, is considerable. In particular, a non-invasive elemental analysis of a surface may not provide enough information to identify techniques such as the layering of colours or the adjustment of pigment particles to produce rich colour and colour depth. The use of both non-invasive and microanalytical methods is therefore essential for the study of cultural heritage. For example, when we look at the walls of the Kizil and Bezeklik grottoes today, we may see that the paintings are made from bright lapis lazuli blue, atacamite green, strong 'shading' dark brown and white. However, studies and analyses have shown that, in fact, most of the yellow has faded and the red has darkened, so that only the less discoloured areas (i.e. the blue, green, brown and white colour combinations) are now visible. In other words, chemically stable inorganic pigments remain relatively well preserved, but unstable inorganic pigments and many organic substances have faded over time, making it difficult to infer the original colours just by looking at the surface. Recent advances in science and technology have allowed us to identify the presence of organic substances and certain inorganic pigments that have faded, revealing the original colours such as bright yellow or red.

Therefore the colours we see today in the Kizil caves were not those originally intended. Similarly, at Bamiyan's Cave N(a), the wall painting with the Thousand Buddhas appears to be dominated by red and contains yellow that has faded and black that originated from darkened green. A red colour that was less susceptible to fading was used at the Bamiyan site while another red colour more susceptible to darkening was used only at the Kizil caves. A similar situation is evident at the Mogao grottoes in Dunhuang, where the colours appear to be well preserved. Today, a variety of analytical techniques can be used to reveal an artwork's original colours and tones, and even to create a virtual 3D rendering of its original form.

Outside of Europe and the United States, a greater focus has been placed on the analysis of inorganic pigments as until recently it was difficult to identify organic pigments and binding media. However, research has increasingly shown that pigments derive from more than just ground and powdered natural minerals, and that Asian paintings have used techniques that require more than just dissolving pigments in glue solutions. In fact, Asian paintings, like Mediterranean artworks, incorporate complex techniques and the use of varied and diverse hues alongside a variety of colour-extending and binding materials.

European preconceptions of so-called Western painting have also strongly influenced impressions of the painting techniques used in Central Asian and Asian countries, including interpretation of paintings within art history. In fact, most early medieval prescriptions for painting techniques were written in Latin and Greek by ancient Romans and Greek artisans, while many of the materials and techniques they used originated from throughout South, Southeast, Central and East Asia, brought by traders along the Silk Road. Our collective historical understanding of painting has long been based on European perceptions of history. However, with the greater knowledge we have now accumulated, we know that painting techniques were not developed in a one-way flow from the West to the East in antiquity and the early Middle Ages. Therefore, this book examines the Buddhist wall paintings at the Bamiyan site with a focus not only the study of pigments, but also of the painting techniques used by ancient painters, as well as centuries of transcontinental cultural exchange.

Fresco techniques in the West and secco techniques in the East

When describing wall painting techniques, terms such as 'fresco' and 'tempera' are often used but they are also confusing as they fail to accurately convey the specific materials and techniques involved. The Buddhist wall paintings at Bamiyan were painted using the so-called *a secco* (dry) technique, which is similar to wall painting techniques prevalent in early medieval Eurasia and regions to the east of Central Asia. This technique differs from the *affresco* (fresh) technique by its use of a dry wall surface for painting, comprised of a mixture of pigments and organic binding media such as drying oil, egg whites, plant gums and resin. Such paintings are also referred to as tempera.

The so-called tempera technique originally referred to paintings made with various organic binding media such as animal glues, plant gums and eggs. However, today, the term is only applied to paintings made using eggs. While, in a broader sense, it can be said that Central Asian wall paintings were rendered using tempera techniques, this book refers to them as secco wall paintings in order to avoid confusion with the more narrowly defined tempera derived from eggs. Examples of secco painting on earthen renders can be seen throughout Asia, including the Ajanta caves, India; the Kizil, Kumtura and Bezeklik caves; the Tarim Basin; the Mogao-ku in Dunhuang, Tibet; and the wall paintings in the Golden Hall of Japan's Horyuji Temple. In fact, the Ajanta wall paintings are considered to be 'fresco-secco' as the earthen render was topped with lime mortar before the paint was applied with a mixture of binding media (Mitra 1981).

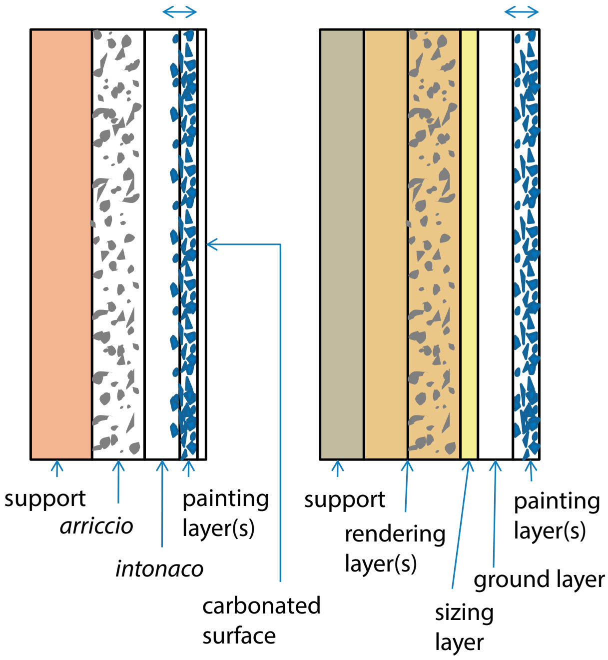


Figure 1 Stratigraphy of wall paintings that are: *affresco* (left) and *a secco* on earthen plaster (right). For a *buon fresco*, a layer of mortar made of lime and sand (*arriccio*) is applied to a rock or brick support, over which an ochre underdrawing (*sinopia*) is painted, followed by layers of lime mortar (*intonaco*) and paint. For an *a secco* wall painting, a render with sand and/or straw is applied to a rock or brick support, followed by an organic sizing layer to prevent the paint from penetrating. A white ground layer is then applied to create a blank surface on which to apply underdrawings and paint.

The *affresco* technique, on the other hand, uses pigments loosened in water to paint on a fresh wall while it is still wet, just after the lime mortar has been added. The wetted lime in the lime mortar reacts with carbon dioxide in the air by forming carbonate, which allows the pigments to set on the surface. The fresco technique is widespread along the Black Sea coast in Western Asia and the Mediterranean. One typical schematic is shown in Figure 1. In *a secco* wall painting, the paint is applied to the dry wall and the paint layers exist independently, while in a fresco painting, the pigment is applied when the lime mortar is only partially dry so that pigment particles penetrate the *intonaco* (or final) very thin layer of plaster.

From a practical point of view, lime used as a support for the wall would preferably be procured in the vicinity of where the mortar is placed, as the process requires calcination of heavy limestone using a large amount of fuel, followed by hydration with water. At the same time, expensive pigments can be transported from remote places through trade and the mobility of artisans. Thus, it seems natural that lime mortar was used as a support in areas where limestone was geologically available, such as in West

Figure 2 Winged angels painted inside a Buddhist niche from Hadda, collection of the Musée national des arts asiatiques-Guimet. Photo: Y. Taniguchi, 2006.

Asia and the Mediterranean, whereas the use of earthen plaster as a render and the secco technique are more commonly found in areas where limestone was scarce.

Painting practices travelled to distant places through the Silk Road. Most Greek and Roman wall paintings were executed on lime mortar: many in fresco, but some in secco. Many of these paintings – especially those on panels, vases and marble objects – were painted using the secco technique and various organic materials as binders. To date, beeswax, gum arabic and eggs have been identified consistently as binding media in analyses. For instance, 'mummy portraits' (1st–4th century) in Faiyum, Egypt used the beeswax painting technique (encaustic) (Newman and Serpico 2000: 489). Similarly, the surface of Greek red-figure vases contained plant gums used as binders (Scott *et al.* 2002).

The easternmost known frescoes in Eurasia are the wall paintings of the Brihadisvara Temple (Thanjāvūr, India),[5] while the westernmost examples of secco painting on earthen renders are found in the Sasanid domain.[6] Moreover, a fusion of Greek and Roman motifs and composition styles, albeit with the use of gypsum and clay in a stucco technique, can be seen at the sites of Begram in the southern foothills of the Hindu Kush Mountains, Hadda, in the middle reaches of the Kābul River, and Mīrān on the southern rim of the Taklamakan Desert in Xinjiang, northwest China. One example of a wall painting from Hadda can be found in the collection of the Musée national des arts asiatiques-Guimet in Paris: it shows a statue of Buddha in a Buddhist niche adorned by a pair of winged angels holding flowered vines (Fig. 2) in a Greco-Buddhist style.

According to one analysis carried out in Italy, some wall fragments excavated from Hadda – such as those depicting a standing Buddha (MG17451) – were painted using the lime fresco technique (Cambon 2004: 97). The mixture of Greco-Buddhist style wall paintings with lime frescoes is an important example of the fusion of East and West. Sir Arthur Herbert Church, who analysed the Buddhist walls at Mīrān (also representative of the Greco-Buddhist style with figures holding flowered vines and winged angels), concluded that the wall paintings differed from

European frescoes: the paintings were depicted on a white gypsum ground layer over a chaff-tempered earthen render (Church 1921).

The wall paintings from the Tarim Basin, which have not been analysed and lack any records of the materials used for painting, include those from:

- Kara-tepe (late 1st–2nd century) (Mkrtichev 2002)
- Hadda (2nd–5th century, Kushano-Sasanian)
- Mes Aynak (3rd century)
- Fayaz-tepe (2nd–4th century, Kushano-Sasanian) (Kageyama *et al.* 2021, 2022; Kageyama 2022)
- Karadon (late 4th–early 5th century) (Rhie 2007a)
- Balalyk-tepe (5th–6th century) (Al'baum 1960: 219)
- Ajina-tepe (7th–8th century) (Litvinsky *et al.* 1971)
- Kafir-Kara (2nd–3rd century, mid-6th century–740s) (Litvinsky *et al.* 1990)
- Niya (late 4th–early 5th century) (Hansen 2004)
- Dandan Oilik (7th century) (Gu Libiya 2007)
- Tavka (6th–7th century) (Rahmonov 2001: 139)
- Kala-i Kafirnigan (Tokkuz-tepe) (early 7th–mid-8th century AD) (Litvinskij 1981)
- Penjikent (5th century–AD 722) (Marshak 2016)

According to available information in the reports, they are all considered secco wall paintings executed on earthen render (Marshak *et al.* 1990). In other words, the wall paintings along the Silk Road can be divided roughly into two groups: the lime and secco paintings located to the west of Central Asia, and the secco paintings on earthen render located to the east. Materials used in the construction of the walls, which necessarily required large quantities, undoubtedly reflect local geological properties.

Notes

1. This project was carried out as part of the Cooperation Project for the Conservation of Cultural Heritage in West Asian Countries and as part of the Bamiyan Safeguarding Project funded by the UNESCO Japanese Funds-in-Trust for the Conservation of Cultural Heritage in collaboration with the Ministry of Information and Culture of the Islamic Republic of Afghanistan. Due to organisational changes, the name of the implementing institution was changed from the National Research Institute for Cultural Properties (2003–March 2007) to the National Research Institute for Cultural Properties, Tokyo and the Nara National Research Institute for Cultural Properties (April 2007 onwards).
2. Analysis of wall paintings from the Deokheung-ri tumulus (AD 408), one of the Goguryeo tumuli, indicates that the wall paintings were depicted over wet lime mortar. Mazzeo *et al.* (2006), who carried out the analysis, have suggested that the wall paintings are frescoes. The lack of historical documentation on the use of such techniques makes it impossible to conclude whether this was a deliberate decision by the painter or if it was frescoed by chance. As there are no examples from the surrounding areas connecting the Goguryeo tumuli with the frescoes, we would like to keep this for reference.
3. Bostock n.d.
4. *Ibid*.
5. The wall paintings of the Brihadisvara Temple (Thanjāvūr, India), in which the use of the *buon fresco* technique has been reported, belong to the 11th or 12th century, which is somewhat later than the period discussed in this book (Paramasivan 1937). See also note 2 above.
6. A wall painting from the manor house of a Sasanid lord at Hājiābād in southern Iran, 280 km east of Shiraz, probably dating from the 4th century. There are few descriptions of the painting technique, so details remain unclear, but it is believed that the painting was executed on an earthen wall render mixed with chaff fibres, followed by an underdrawing in red ochre and final outlines and details in black (Azarnoush 1994).

Chapter 1: Past studies on materials and techniques used in Bamiyan's wall paintings

1.1 Bamiyan and the wall paintings in its surrounding caves

1.1.1 About the wall paintings

Bamiyan is one of several historical Buddhist towns located on the periphery of intersecting routes connecting China in the east, Iran and the Mediterranean world in the west, India in the south and nomadic tribes and Central Asian oasis states in the north (Fig. 1.1). Of the many structures once built here, all that remains today are two large Buddhas carved into the cliffs, ruins of one stupa, and hundreds of caves and dozens of wall paintings.

Bamiyan has three valleys – Bamiyan, Foladi and Kakrak – each named for the rivers that flow through them (Fig. 1.2). The area is sandwiched between the Hindu Kush and the Koh-i Baba Mountains, and takes the shape of a long and narrow basin running from east to west at an altitude of about 2500 m. The Hindu Kush Mountains track diagonally across the centre of Afghanistan, forming a boundary between South and Central Asia. They are also a major transportation artery: leading eastwards across the Pamirs to the western edge of Chinese territory around the Tarim Basin and westwards along the Bamiyan Valley through Herat and into Iran.

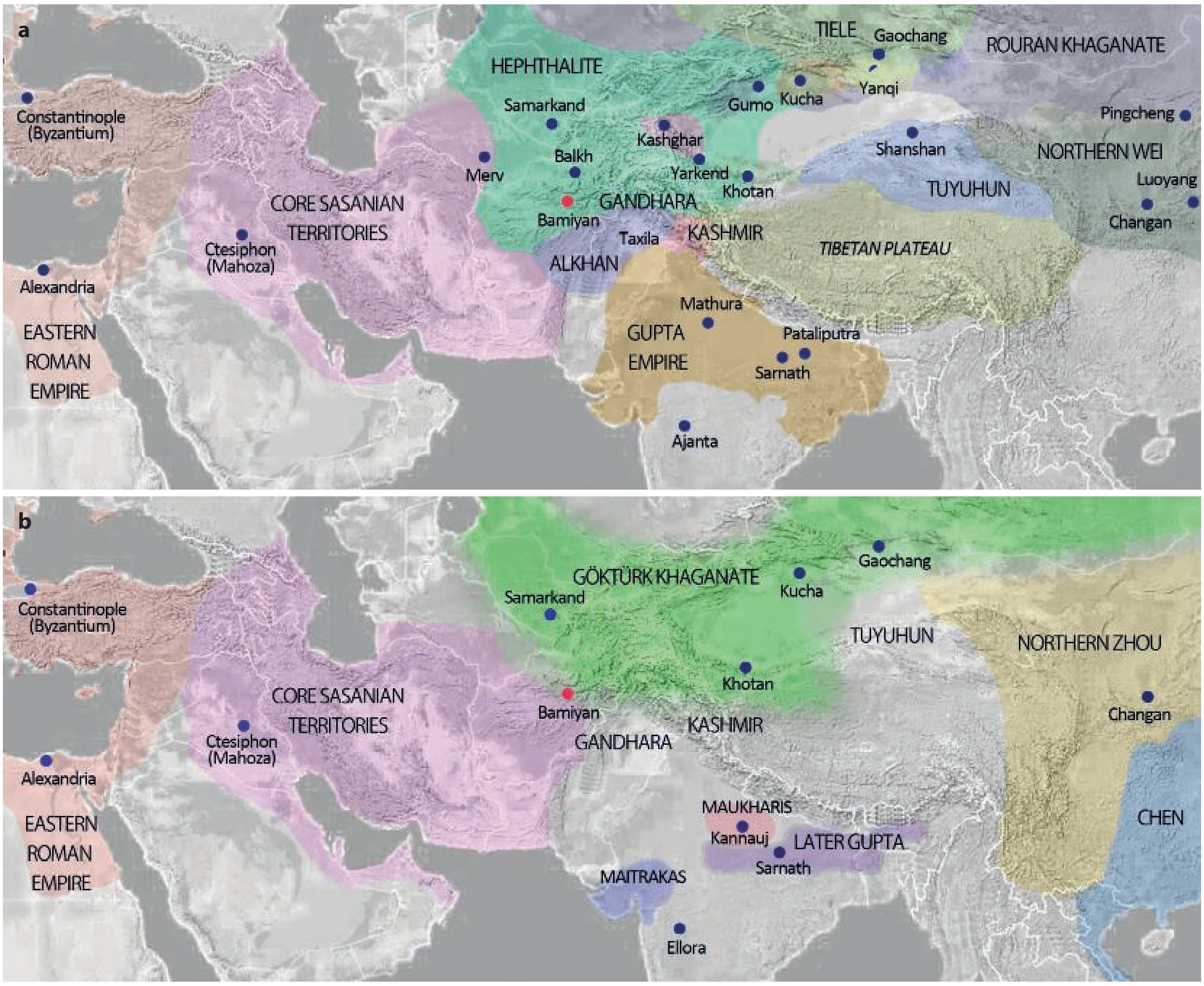


Figure 1.1 Location of Bamiyan sites in Eurasia: political entities in (a) the late 5th to early 6th century AD and (b) in the late 6th century AD.

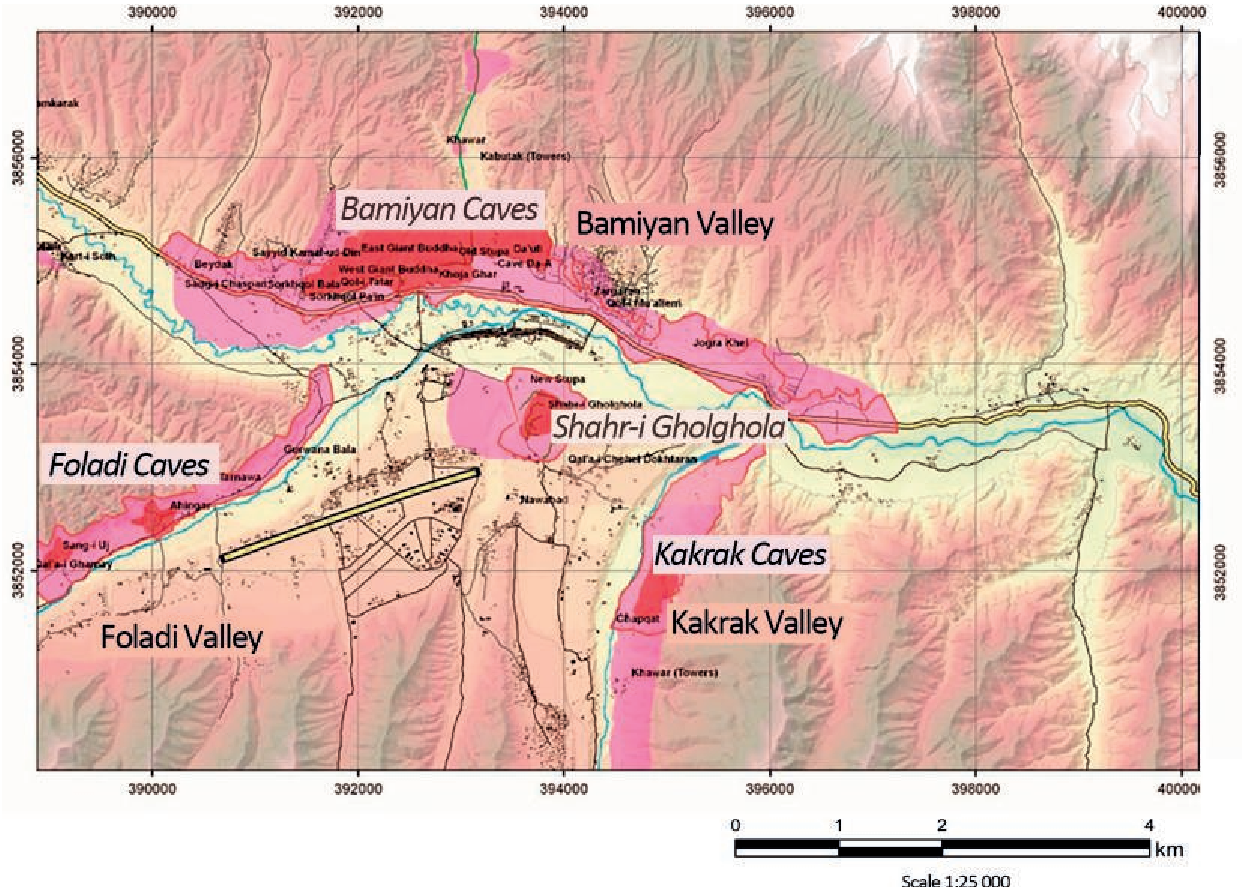


Figure 1.2 Location of the Bamiyan, Foladi and Kakrak caves in the Bamiyan Valley.

The northern and southern sides of the Hindu Kush Mountains are zones of mixed peoples and cultures of different lineages: Persian and Mediterranean in the west, South Asian in the southeast and Central Asian nomads in the north. This diversity makes it difficult to compile a unified historical interpretation. For this reason, debate has proliferated in the fields of Buddhist art history, world history, linguistics, architectural history and archaeology. Many previous studies have attempted to date the wall paintings using iconography and Buddhist history to better clarify the complex ethnic and religious trends pervading Central Asia.

Topographical maps of the Bamiyan Valley show that on the northern side of the valley is a series of mountains with sparse greenery, while the basin on the southern side holds an oasis of lush green land, fed by water from the Bamiyan River. The Batakhshan region, which is home to the natural blue mineral lapis lazuli, lies some 500 km northeast of Bamiyan. The region also produces iron, lead, sulfur, alum, ochre and sal ammoniac, and once had gold and silver mines in its vicinity (Bowersox *et al.* 1995: 40).

There are numerous grottoes in the Bamiyan Valley, mainly in the caves on the Bamiyan main cliff, the Foladi caves in the west and the Kakrak caves in the east (Figs 1.3–1.8). More than 750 caves have been found in total. The Qol-e Jalal caves, discovered during a survey in 2005, are located about 4 km to the west of the Bamiyan main cliff. Nearby, the remains of a Buddhist temple in the Chil-Borji Citadel also hold wall paintings (Irisawa 2006: 7).

Two surveys – one conducted by Kyoto University in 1974, 1976 and 1978 (Higuchi 1983–1984) and another by the National Research Institute for Cultural Properties, Tokyo (NRICPT) in 2003–2013 and 2017 – compiled the number of caves with wall paintings in Bamiyan, with the latter finding approximately 50 (NRICP 2004). Most wall paintings were found in smaller caves (square, octagonal or circular in shape) and were most likely used as shrines for meditation. Bamiyan's wall paintings depict various motifs such as Buddhas, Bodhisattvas, royalty, monks, Mithra, the Sun God, the Moon God, the Wind God, griffins, animals such as boars, dogs and hamsa birds, and plants such as lotuses (Maeda 2006). Styles vary from cave to cave with influences from Greece and Rome, Sasanid Persia, India and the Tarim Basin. The fields of Buddhist art history and world history each include extensive research into their subject, iconography and stylistic characteristics.

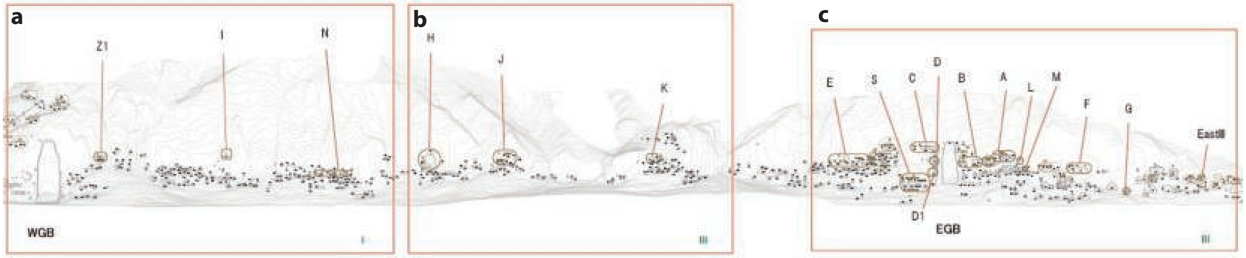


Figure 1.3 Layout of the Eastern and Western Giant Buddhas and the caves at Bamiyan's central cliffs (after figs 4 and 5 in Yamauchi 2011).

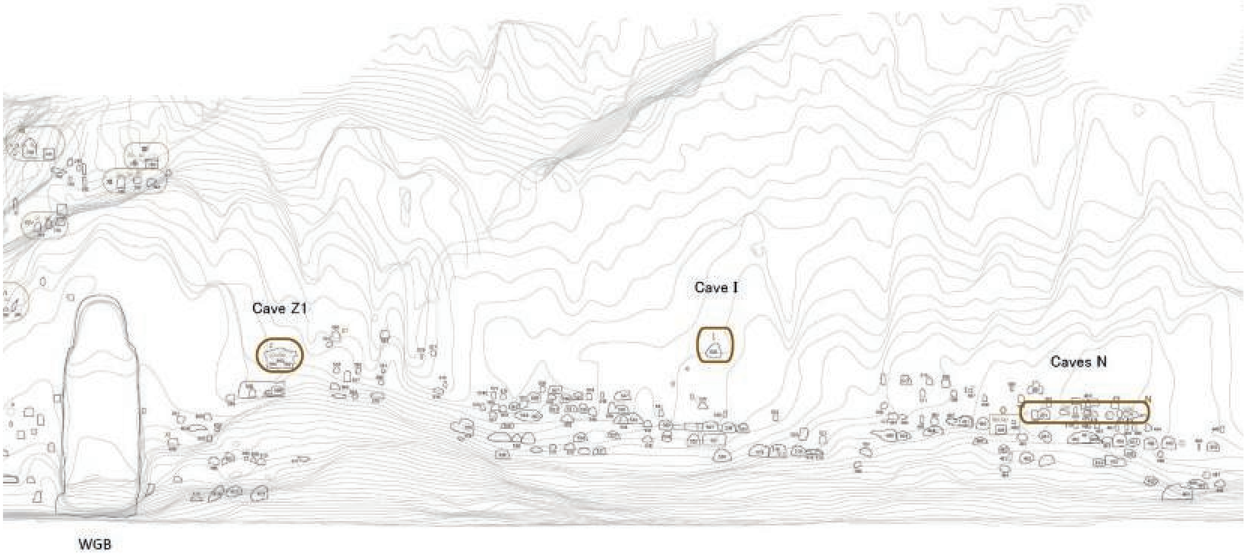


Figure 1.4 Layout of the Western Giant Buddha and caves at Bamiyan's western cliffs (enlarged view of Fig. 1.3a).

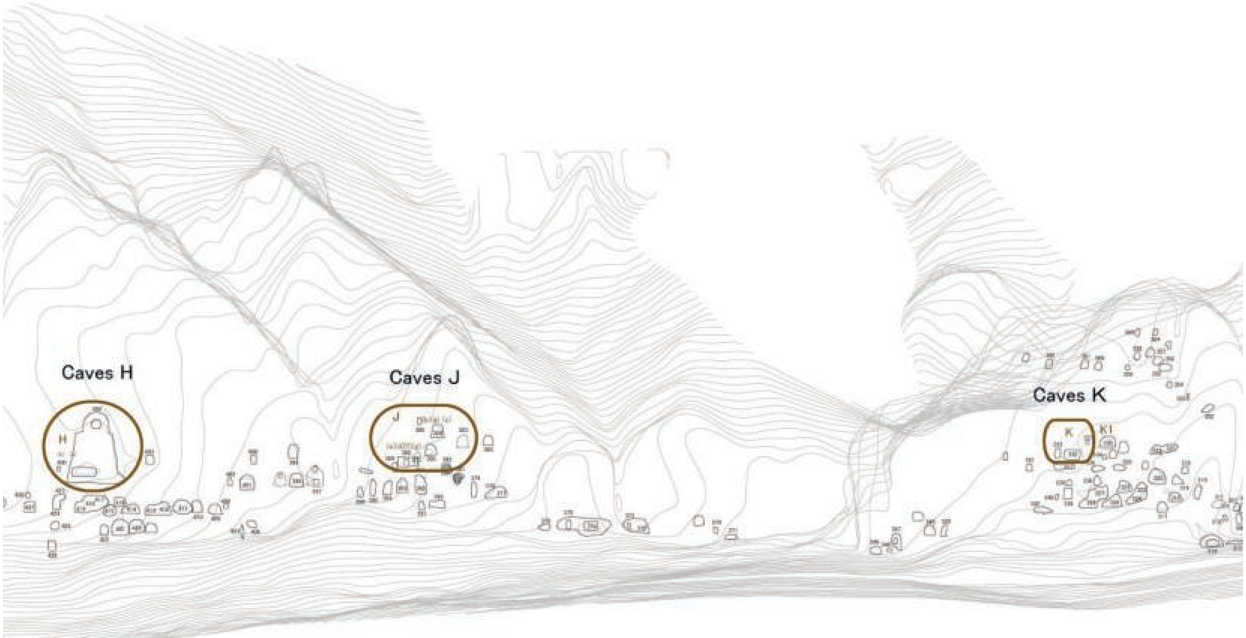


Figure 1.5 Layout of the caves in the central cliffs of Bamiyan (enlarged view of Fig. 13.b).

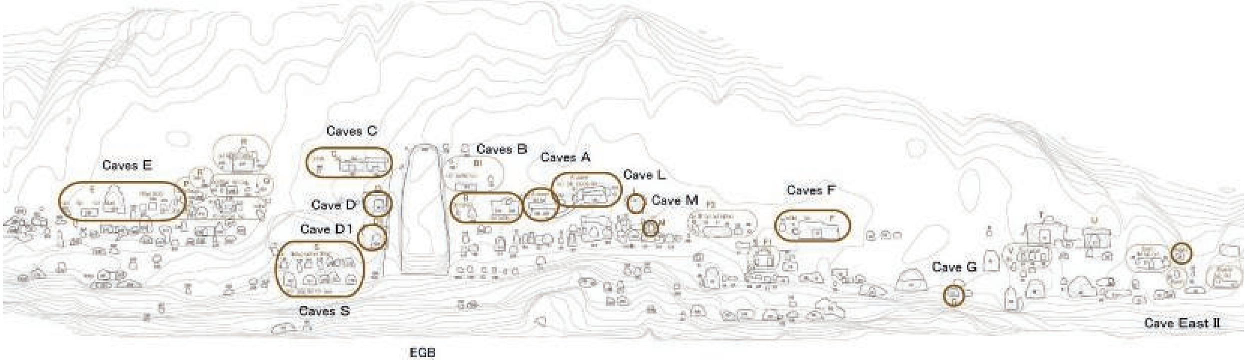


Figure 1.6 Layout of the Eastern Giant Buddha and caves at Bamiyan's eastern cliffs (enlarged view of Fig. 1.3c).

Figure 1.7 Map of the distribution of caves in the Foladi Valley (after Higuchi 1983–4: vol. IV, plan 55, modified).

Figure 1.8 Distribution of caves in the Kakrak Valley (after Iwade *et al.* 2013: 212, fig. 602).

First, in the following section, we consider the results of radiocarbon dating of wall paintings using chaff tempers (straw fibre) procured from renderings. An overview is then provided of the dates and stylistic characteristics associated with each site. By clarifying when Buddhism was introduced to the Bamiyan Valley and when the Eastern and Western Giant Buddhas were built, we may be able to distinguish the times and regions of influence. This examination will illustrate not only the northward movement of Buddhism after the fall of Gandhara, but also the possibility that Buddhism, once spread to the Tarim Basin and its environs, moved westwards to Bamiyan. Ultimately, we shall see how centuries of wall paintings at Bamiyan were influenced by these different regions, and how painting materials and techniques changed over time.

1.1.2 The chronology of wall paintings determined by radiocarbon dating

The historical origins of Bamiyan have been debated vigorously, but are thought to stem from the fall of Gandhara and the spread of Buddhism through minority and indigenous nomadic peoples such as the Hephthalites. Kuwayama (2002: 155) elaborates:

> [F]rom that time [mid-6th century] onward, the earlier [Karakorum] route was completely discarded [and] a new route was opened as a highway running beyond Nagarahara towards the west and crossing the Hindu Kush at Bamiyan. Supported by more and more frequent exchanges of wealth from both sides of the Hindu Kush, Bamiyan, and Kapisi appeared from an earlier, rather isolated position in both geographical and hence economical contexts … The Bamiyan colossi emerged when Bamiyan became quite prosperous on the highway, that is, from the middle of the sixth century on.

This section reconsiders the chronology of Bamiyan's caves and wall paintings based on findings from newly collected samples. Radiocarbon dating using chaff fibres and ropes as well as wood fragments was carried out in collaboration with the Nagoya University Center for Chronological Research and the NRICPT. The data include results from samples collected in October 2003 (including straw fibres, ropes and wood pegs), September–October 2006 (including straw fibres), and June and July 2006 (including straw fibres from the Giant Buddhas, and ropes and large wooden pegs from the Western Giant Buddha). Samples from wall paintings were collected by the Japanese mission between 2003 and 2004, and samples from surface fragments of both Buddhas were

Table 1.1 Radiocarbon dating results and calibrated dates.

No.	*Sample ID*	*Cave*	*Material*	*^{14}C age (Uncal BP)*	*error （±1σ）*	*Cal AD （±1σprobability 95.4%）*	*Lab ID （NUTA2-）*
1	A lower salle-1	Bamiyan A lower salle	Chaff fibre	1447	27	567-651	7441
2	A lower salle-2	Bamiyan A lower salle	Chaff fibre	1462	27	556-646	7442
3	B(a)	Bamiyan B(a)	Chaff fibre	1348	27	641-766	7443
4	B(d)	Bamiyan B(d)	Chaff fibre	1296	28	663-772	7444
5	C(a)	Bamiyan C(a)	Chaff fibre	1506	27	442-637	7642
6	C(a) forecourt	Bamiyan C(a) forecourt	Chaff fibre	1501	27	438-631	7641
7	C(b)	Bamiyan C(b)	Chaff fibre	1496	27	467-640	7640
8	D	Bamiyan D	Chaff fibre	1386	27	605-669	7644
9	D forecourt	Bamiyan D forecourt	Chaff fibre	1394	27	609-672	7643
10	D1-1	Bamiyan D1	Chaff fibre	1484	27	541-640	7645
11	D1-2	Bamiyan D1	Chaff fibre	1471	27	549-643	7787
12	E(e)-1	Bamiyan E(e)	Chaff fibre	1289	27	666-774	7782
13	E(e)-2	Bamiyan E(e)	Chaff fibre	1280	28	664-779	7783
14	E(e)-3	Bamiyan E(e)	Chaff fibre	1308	28	657-772	7784
15	E(e)-4	Bamiyan E(e)	Chaff fibre	1278	27	665-779	7785
16	East III	Bamiyan EIII	Chaff fibre	1347	28	641-767	7780
17	F(a)	Bamiyan F(a)	Chaff fibre	1307	27	658-772	7648
18	F(c)-1	Bamiyan F(c)	Chaff fibre	1245	27	682-870	7650
19	F(c)-2	Bamiyan F(c)	Chaff fibre	1208	27	713-891	7651
20	H(a)	Bamiyan H(a)	Chaff fibre	1416	30	584-663	13842
21	I niche	Bamiyan I niche	Chaff fibre	1270	27	665-808	7795
22	J(b)-1	Bamiyan J(b)	Chaff fibre	1550	27	428-570	7788
23	J(b)-2	Bamiyan J(b)	Wood	1557	27	426-564	7801
24	J(d)	Bamiyan J(d)	Chaff fibre	1560	27	426-562	7792
25	J(e)	Bamiyan J(e)	Chaff fibre	1543	26	430-577	7793
26	J(g)-1	Bamiyan J(g)	Chaff fibre	1538	26	431-590	7790
27	J(g)-2	Bamiyan J(g)	Chaff fibre	1559	27	426-563	7791
28	K-1	Bamiyan K3	Chaff fibre	1283	25	669-774	8614
29	K-2	Bamiyan K3	Chaff fibre	1283	25	669-774	8615
30	L	Bamiyan L	Chaff fibre	1291	29	664-774	13841
31	M	Bamiyan M	Chaff fibre	1559	28	425-565	7786
32	N(a)	Bamiyan N(a)	Chaff fibre	1375	27	613-680	7794
33	S(a)-1	Bamiyan S(a)	Chaff fibre	1397	25	606-666	8618
34	S(a)-2	Bamiyan S(a)	Chaff fibre	1455	29	556-650	13825
35	S(b)	Bamiyan S(b)	Chaff fibre	1528	31	432-602	13826
36	S(c)	Bamiyan S(c)	Chaff fibre	1325	30	650-771	13827
37	Z1	Bamiyan Z1	Chaff fibre	1216	27	695-889	7796
38	Foladi 2	Foladi 2	Chaff fibre	1347	27	641-766	7436
39	Foladi 4-1	Foladi 4	Chaff fibre	1258	28	671-862	7433
40	Foladi 4-2	Foladi 4	Chaff fibre	1268	28	666-854	7434
41	Foladi 4-3	Foladi 4	Chaff fibre	1274	27	666-802	7435
42	Foladi 5 front room	Foladi 5 front room	Chaff fibre	1228	27	690-883	7437
43	Foladi 6	Foladi 6	Chaff fibre	1254	27	674-863	7440
44	Kakrak 43	Kakrak 43	Chaff fibre	1303	27	660-772	7799
45	Kakrak 44	Kakrak 44	Chaff fibre	1112	26	885-991	7800
46	EGB-A12P	Eastern Giant Buddha	Chaff fibre	1543	32	428-591	12512
47	EGB-A123P	Eastern Giant Buddha	Chaff fibre	1571	31	420-556	12513
48	WGB-A07	Western Giant Buddha	Rope	1442	31	564-655	12443
49	WGB-A08	Western Giant Buddha	Rope	1436	30	570-655	12444
50	WGB-A09	Western Giant Buddha	Rope	1413	31	582-665	12445
51	WGB-A10	Western Giant Buddha	Wooden pile	1439	31	566-655	12446
52	WGB-A11P	Western Giant Buddha	Chaff fibre	1457	30	554-649	12511

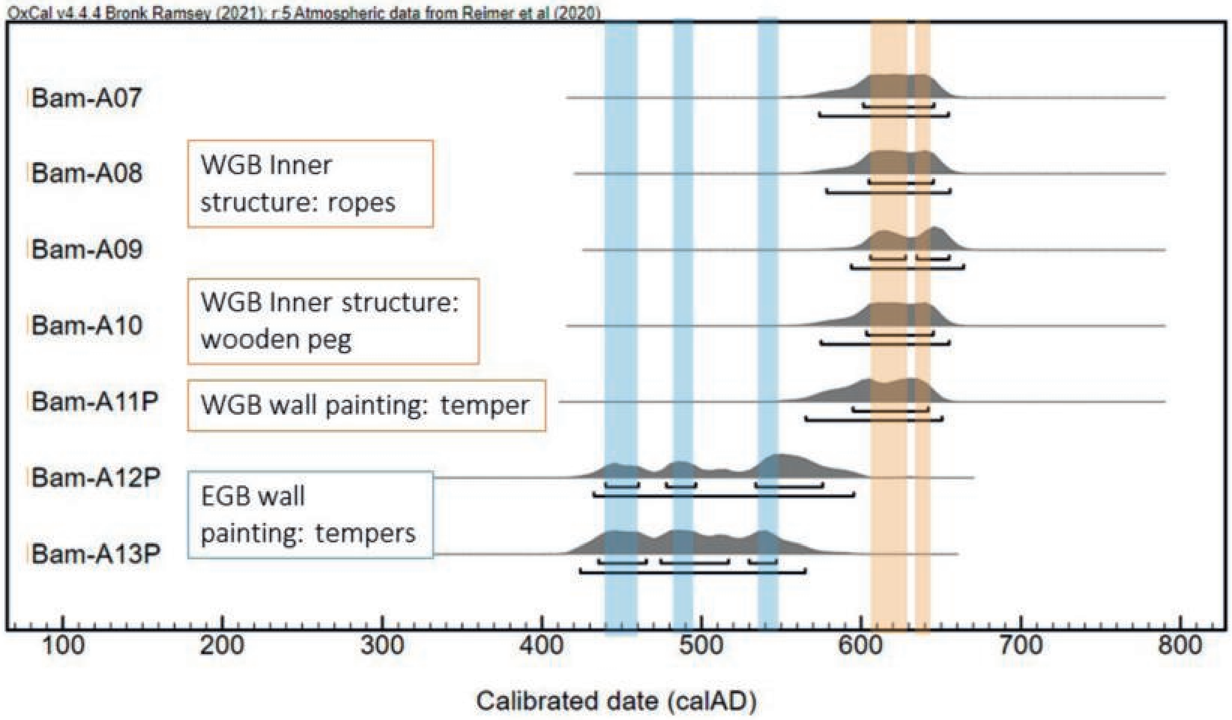


Figure 1.9 Plot of calendar-year probability distributions based on the calibrated dates for the Giant Buddhas (from red chaff fibre samples from earthen renders) (data from NRICPT/Nagoya University).

provided by the German Committee of International Council on Monuments and Sites (ICOMOS). The location of the samples collected in 2003 and the methodology and results of their analysis have been reported in detail by Iwai and others (Yamauchi 2006a; Iwai 2006c). The ICOMOS Germany findings have also been previously reported in detail (Blänsdorf *et al*. 2009a).

Some radiocarbon dates determined by the Japanese and ICOMOS Germany teams were recalibrated in 2022 – specifically, 52 dates for wall paintings and the Eastern and Western Giant Buddhas. These were then calibrated and plotted using OxCal v. 4. 1. 1 with IntCal 04 (Bronk Ramsey 2009) and OxCal v. 4.4 with IntCal20 (Reimer *et al.* 2020). Figure 1.9 shows the probability distributions of the calibrated dates for both Giant Buddhas in calendar years. Figures 1.10 and 1.11 illustrate the probability distributions for the wall paintings in calendar years but rearranged in chronological order of Bamiyan, Foladi and Kakrak. Table 1.1 shows the samples and dates obtained for each.

The ropes buried between the rock body and the earthen render of the Western Giant Buddha were dated to roughly the same period as the straw fibres associated with the wall paintings. The consistency between the dates suggests that the wall paintings and the decoration covering the surface of the Western Giant Buddha are contemporaneous.

These findings are generally consistent with the results of ICOMOS Germany's radiocarbon dating of chaff fibres from earthen renders on the surface of each Giant Buddha (Melzl and Petzet 2007). The aforementioned results placed the Eastern Giant Buddha in the late 6th century (1σ: AD 549–579, 2σ: AD 544–595) and the Western Giant Buddha in the early 7th century (1σ [68%]: AD 605–633, 2σ [95%]: AD 591–644) (Blänsdorf *et al*. 2009a: 235). The recalibrated Japanese results (Nakamura 2006) fall into dates spanning AD 440–460, AD 478–496 and AD 534–546 (1σ) for the Eastern Giant Buddha, and AD 606–642 (1σ) for the Western Giant Buddha.

There has been extensive discussion regarding the construction period of both Buddhas, especially regarding the earlier Eastern Giant Buddha and references by the Chinese scholar Xuanzang (Rowland 1974; Tarzi 1977; Carter 1985; Miyaji 2008, 2022; Klimburg-Salter 1988, 2003, 2019; Kuwayama 1985, 1987, 1990, Tanabe 2001/2). Xuanzang's text mentions the presence of two colossi in Bamiyan, therefore indicating that the Western Giant Buddha could not have been constructed later than 629. In addition, determination of the dates is complicated by possible contamination of chaff fibres from subsequent ancient repairs, including the possible reuse of old wooden pegs. There are evident differences in the plastering techniques used in each Buddha: in the Eastern Giant Buddha, surface holes were coated with rock pebbles

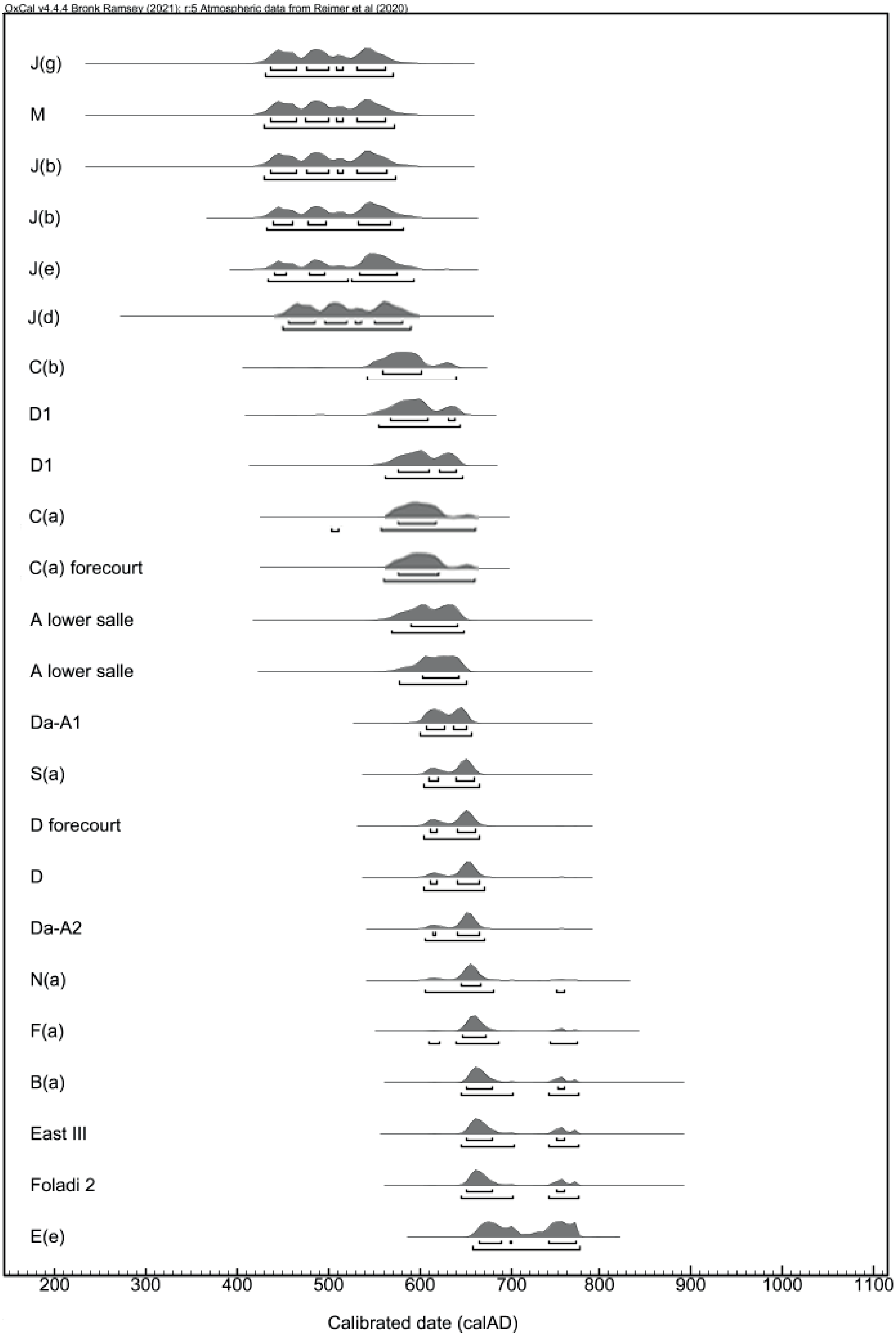


Figure 1.10 Plot of calendar-year probability distributions based on the calibrated ages of the wall paintings (1).

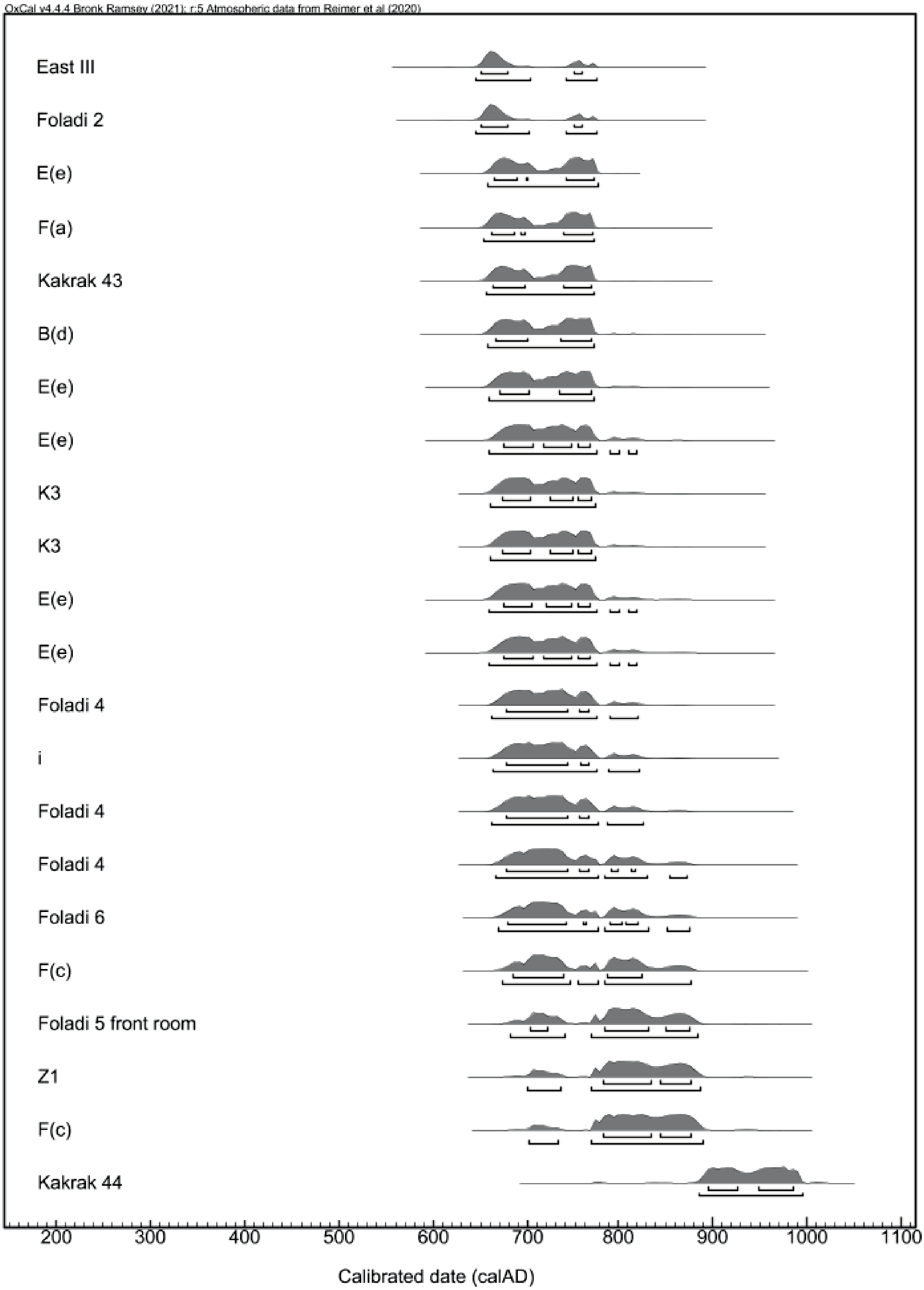


Figure 1.11 Plot of calendar-year probability distributions based on the calibrated ages of the wall paintings (2).

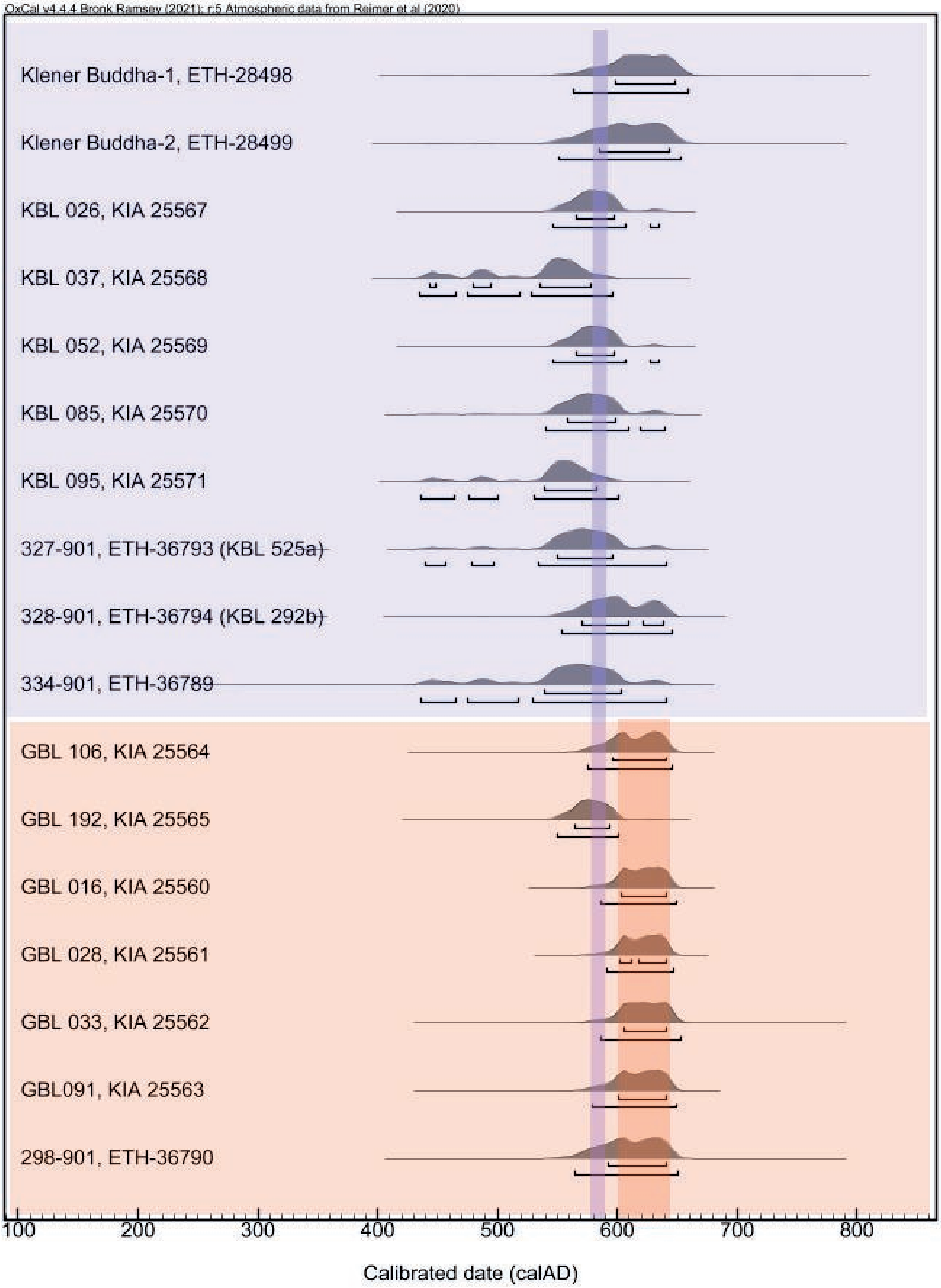


Figure 1.12 Plot of calendar-year probability distributions based on the calibrated ages of the EGB (blue) and WGB (orange) (data from ICOMOS Germany) (Blänsdorf *et al.* 2009a).

adhered to earthen plaster (Praxenthaler 2014: 267–268) while in the Western Giant Buddha the holes were filled using rock pebbles alongside wooden pegs with ropes (Blänsdorf *et al.* 2009b), therefore taking into account differences in plant samples is of critical importance. Although the date of origin of the Eastern Giant Buddha remains unclear, it is possible to locate the origin of the Western Giant Buddha in the first half of the 7th century AD (Fig. 1.12) (Blänsdorf *et al.* 2009a: 235). Because the scope of our research does

Table 1.2 Chronological sequence of caves based on radiocarbon dating by NRICPT/Nagoya University (2006).

Phase	*Date*	*Caves*
Phase I	5th–late 6th century	EGB, Caves J(b), J(d), J(g), M
Phase II	early 6th–mid-7th century	Caves C(a), C(b), D1, S(a), A lower salle, H(a), WGB Caves E, K_3, I
Phase III	mid-7th–late 8th century	Caves B(d), F(c), N(a), S(a), L, Foladi Cave 4, 6, Kakrak Cave 43

* Dating indicated here is based on the ^{14}C result of Nagoya University (Nakamura 2006), however, there are various ongoing debates in history and art history over the date of the earliest phase of Bamiyan

not include entering into the debates on chronology, this text only discusses work based solely on the radiocarbon dating results obtained by both ICOMOS Germany and the Japanese teams. The wall paintings of the Eastern Giant Buddha possibly date back to the late 6th century AD.

Bamiyan's caves are unique in that they are interconnected via staircases and passages (Iwade and Kubodera 2013: 237). For this reason, the Nagoya University and French teams identified the caves in groups as Caves A, Caves C, and so on, with each group represented by capital letters. The radiocarbon dating results for each group showed dates that were close in range, which suggests that the estimated construction dates are reliable. Identifying the exact dates for groups would require collecting and dating organic samples from all the locations including caves surrounding the Eastern Giant Buddha. Miyaji (2005b) summarised the chronological order of cave construction as follows:

Phase I
(1) mid-5th–mid-6th century: Caves M, J(b), J(d), J(e), J(g)
(2) mid-6th–late 6th century: Caves C(a), C(a) anterior chamber, C(b)
(3) mid-6th–early 7th century: Caves DI, A lower
Phase II
(1) early 7th– late 7th century: Caves D anterior chamber, D, N(a), S(a)
(2) mid-7th– late 7th century: Caves F(a), B(a), East III
Phase III
(1) late 7th– late 8th century: Caves B(d), E(e), K3, I
(2) late 8th– late 9th century: Caves F(c) Z1

Caves J, which show the earliest dates, include sculptures and wall paintings with a stupa in the centre. Caves J(g) and J(d) both had nirvana Buddha figures. Caves J have often been described as belonging to a later period of style in Bamiyan, but the radiocarbon dating results here triggered an important debate on the chronological interpretation of the whole. Cave M also featured one painting of a donor wearing a common Toharistan tunic, as well as Sun and Moon gods showing Gupta influence. According to Miyaji (2005b), the art historical context of such motifs must be reconsidered if in fact they were introduced to Bamiyan between the mid-5th and mid-6th century. In this book, the chronological phases indicated are based on the Nagoya University results (Nakamura 2006). However, there are many ongoing debates in both history and art history as to the earliest dates (Iwai 2007, 2014; Klimberg-Salter 2019, 2020; Miyaji 2005b), therefore further discussion of the chronologies will be addressed using both scientific and historical facts.

The earliest Bamiyan wall paintings date back to the late 5th century while the most recent belong to the end of the 9th century. The Eastern Giant Buddha and some of the caves may have appeared in the Bamiyan Valley earlier than the mid-6th-century emergence of the trans-Hindu Kush route described by Kuwayama and Buddhism's eventual flourishment in Bamiyan. The caves at Foladi and Kakrak, which date from the mid-7th century to the end of the 9th century, are somewhat more recent than Bamiyan's wall paintings. The anterior chamber of Foladi Cave 5 is slightly older than the other Foladi caves, dating between the late 7th and the late 9th century. Only one cave, Cave 44 of the Kakrak caves, has been dated to the 10th century, which is considerably more recent than the other wall paintings.

The above probability distributions have been organised in chronological order, and the following trends can be identified roughly. For convenience, time periods are given from the first to the third phase (Table 1.2).

Since there are about 750 caves without wall paintings, the findings here do not provide a comprehensive account of the rise and fall of Buddhist cave temples in the Bamiyan Valley. The samples were taken only from the 50 caves with wall paintings, which span a period from around AD 450 to 850 based on radiocarbon dating results. We may also deduce the time at which some cave temples were created in the Bamiyan Valley using the dates attributed to their wall paintings.

Based on the above timeframe, in the first phase, it appears that the earliest Buddhist structures in Bamiyan were the Eastern Giant Buddha, Cave M, which is located to the east of the Giant Buddha, and Caves J located about 1 km west of the Giant Buddha. The surrounding caves (Caves C, D, D1, A lower) were created after the Buddha's construction. These caves are interconnected by a passage that circles around the Giant Buddha, therefore, the Giant Buddha was constructed first, followed by the surrounding caves, and not vice versa (Miyaji 2005b).

From the early 6th century to the mid-7th century, in the second phase, the array of cave temples gradually expanded to the west and east of the Bamiyan cliffs as seen in the Seated Buddha of the Cave I niche, Cave K_3, Cave E and Cave B(d). Cave D, which dates to the mid-7th century, includes the Sasanid plant motif on the ceiling. From the mid-7th century to the late 8th century, in the third phase, cave temples were also being built in the Foladi and Kakrak valleys, a short distance from Bamiyan. Hence, it is likely that over time the cave temples spread from the area around the Eastern Giant Buddha towards the east and other nearby valleys. This chronology is discussed further in the next section on wall paintings and in Chapter 4 (section 4.1) on cave styles.

1.2 Wall paintings around the Bamiyan Valley

1.2.1 Wall paintings by the niches surrounding the two Giant Buddhas in Bamiyan

Alexander Burnes, who visited Bamiyan in the mid-19th century, noted that the local population referred to the 'male and female' colossi as *Salsal* and *Shahmama* (Burnes 1833). He also reported that the niches surrounding the Giant Buddhas were decorated with colours as vivid and painting styles as distinct as Egyptian tombs. When the Giant Buddhas were destroyed, the wall paintings along both Buddha niches were also completely lost. Because they can no longer be seen today, it is difficult to discuss the wall paintings. However, surveys and photographic documentation carried out by many countries through the 1970s ignited much debate about iconography. Some of the earlier studies are reviewed here.

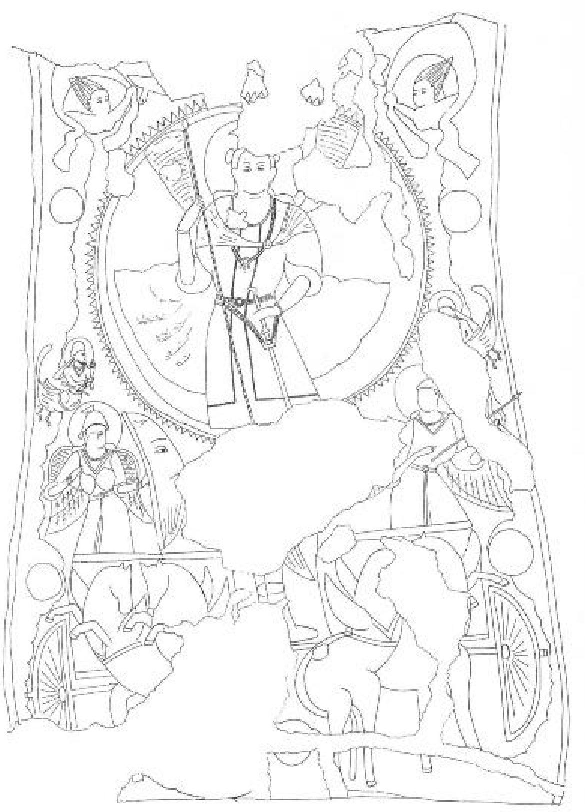


Figure 1.13 Reconstruction of the large composition on the ceiling of the niche by the Eastern Giant Buddha (after Miyaji 2002: 75).

According to Akira Miyaji, the ceiling of the Buddha's shrine at the Eastern Giant Buddha was once covered with a blue lapis lazuli background and numerous illustrations including a four-headed two-wheeled chariot, a winged deity, a winged goddess, a Buddha, a prince and prince offering, a bird, a wind god, and other figures not commonly found in Buddhist wall paintings (Miyaji 2002: 74–77) (Fig. 1.13). The figure of the Sun God riding on a two-wheeled chariot may have originated with Helios in the Greek world, Surya in India, or Mithra in Iran, among other possibilities. Rowland's theory is that the image derives from a combination of beliefs about the Sun God with ideas of the Buddha, as the former was worshipped before the introduction of Buddhism to Bamiyan (Rowland 1938a). According to Avesta's text, *Mihr Yasht*, the Sun God is regarded positively as Mithra, a god who guarantees the safety of nomadic space and the god of contracts (Grenet 1993 [1995]; Maeda 1999; 2007). Miyaji's theory is that the Buddha's Sun God character and his heavenly salvation are both expressed in this single figure (Miyaji 2002: 87).

Grenet suggests that the figure of the winged goddess on the ceiling was the Iranian goddess Arštāt,

Figure 1.14 Bodhisattva image on the west wall of the niche by the Western Giant Buddha (before its destruction). Photo: K. Maeda, 1970s.

who was identified with the Greek goddess Athena, and that the image opposite her is the goddess Cištā. He hypothesises that Arštāt was depicted with wings because the artist was faithful to the sacred text of the Avesta. Miyaji (2002) describes images of royal princes wearing Sasanian tunics on the east–west strip of the niche surrounding the Eastern Giant Buddha. He suggests that they represent Bamiyan's royal families who took refuge in Buddhism and donated to monks from economic prosperity achieved through trade, as recorded by Xuanzang.

On the other hand, however, the wall paintings adorning the niche of the Western Giant Buddha are thought to represent the heavenly world with a seated Maha Bodhisattva at the centre accompanied by groups of Bodhisattvas, musicians, flying celestial beings and statues of offerings dressed in Central Asian clothing, all surrounded by depictions of lotuses, flowered vines and hanging curtains (Fig. 1.14). Miyaji suggests that, based on the motifs and painting styles, the wall paintings of the Western Giant Buddha Temple reveal three influences: those of the Indian Gupta dynasty, the Gandharan region and Central Asia's Sasanian Empire; these can be attributed to the itinerant artisans who carried influences from different regions with them (Miyaji 2002: 103).

The influence of the Gupta dynasty is visible in the fingers of the Mudras, the Tribhaṅga[1] of the Bodhisattva figures, and the form of the half-naked celestial maidens. The Gandharan influence is identifiable by the floral rope patterns, the hanging curtain patterns, and the pose of the maidens flying through the sky, while the Sasanian Central Asian influence can be seen in the 'bejewelled Buddha' figures and the dress of the royal princes and attendants.

Based on these prior studies, we may surmise that the style of the wall paintings in the niche surrounding the Eastern Giant Buddha of the first phase was influenced by the Hellenistic world and the Sasanian dynasty. The style of the wall paintings in the niche surrounding the Western Giant Buddha of the second phase, however, was influenced by the Gupta dynasty, the Gandhara region and the Sasanian Empire.

1.2.2 Wall paintings in the Bamiyan caves

As mentioned above, Caves J and M fall in the first phase. The wall paintings in Caves J are particularly striking in that they are not smooth and the lines are rough, and those in Cave M, to the east of the Eastern Giant Buddha, included an image of a large standing Buddha on the north wall, flanked by images of a Bodhisattva and a seated Buddha. Two circles rested in a niche on that same wall, one signifying the Sun God and the other the Moon God. Nearby, an image of a winged god appeared to represent the Sun God in a manner reminiscent of depictions at the Eastern Giant Buddha's niche. A figure of a local donor behind the Buddha's mandorla wears a caftan with an open collar (Fig. 1.15). This caftan's fabric (with manifold grid lines) appears structurally similar to a depiction from Kizil Caves 84 and 207, and are possibly representations of Kucha-made silks recorded in two Turfan manuscripts (Hiyama 2018). This depiction is widely seen in Buddhist and non-Buddhist sites in Central Asia and Northwest China (early 5th–mid-6th century) that are as old as the Eastern Giant Buddha.

In Cave J, located at the eastern end of Bamiyan's western cliffs, Caves J(d) and J(g) contain the best-preserved wall paintings. Both caves are square in shape with domed ceilings, and Cave J(d) features a seated Bodhisattva in the dome's centre surrounded by a seated Buddha in a Thousand-Buddha composition. The wall paintings in Cave J(d) have a pink ground layer with a rough surface (Fig. 1.16) similar to the base of the Eastern Giant Buddha (Blänsdorf *et al.* 2009c).

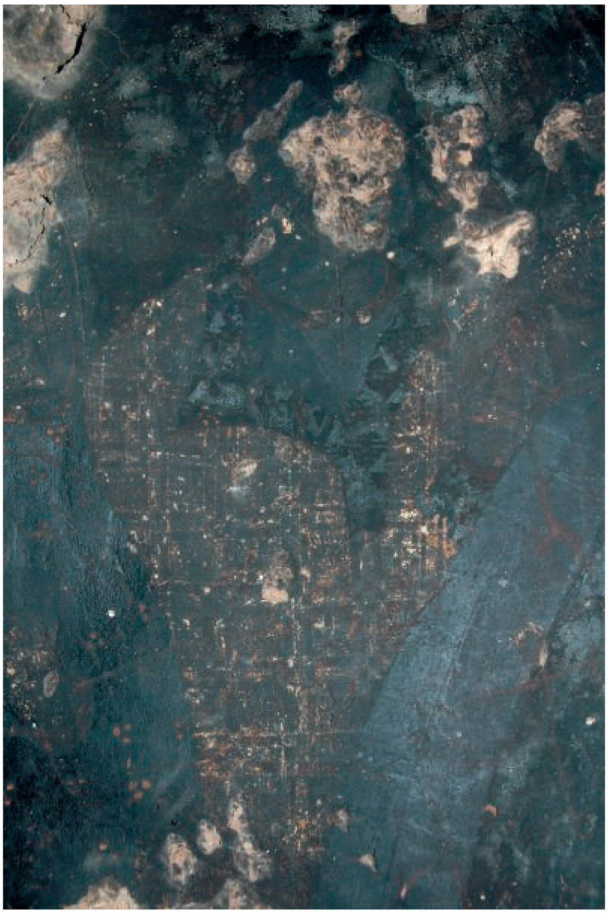


Figure 1.15 The figure of a donor, wearing a caftan with one collar folded over, painted on the south wall of Cave M. The wall painting is covered with a black deposit. Photo: H. Otake, courtesy of NRICPT, 2005.

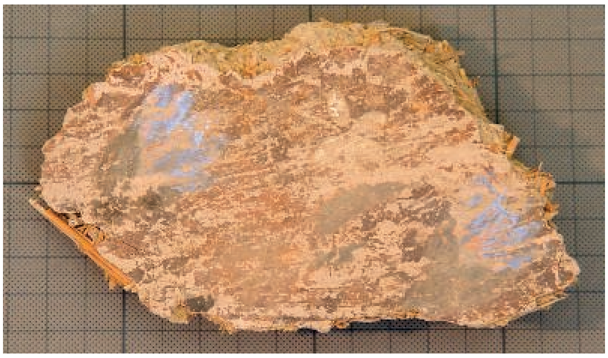


Figure 1.16 Wall painting fragment recovered from Cave J(d) (inv. no. 1077). Photo: H. Otake, courtesy of NRICPT, 2005.

Figure 1.17 Wall painting fragment recovered from Cave C (inv. no. 1349). Photo: Y. Taniguchi, courtesy of NRICPT, 2005.

Figure 1.18 Wall painting fragment recovered from Cave S(a) (inv. no. 1382). The margins between the figures are covered with tin foil. Photo: H. Otake, courtesy of NRICPT, 2005.

The wall paintings from the second phase are a mixture of different types, so it is not possible to summarise their regional and cultural context in a single sentence. They can be divided into three groups: those of Caves C, with their rough wall paintings, simple lines and quick strokes (Fig. 1.17); those of Cave H(a), with its Gupta dynasty influence; and those of Caves S(a) and N(a) with their smooth wall painting surfaces, fine lines and blurred colours (*ungen*), often decorated with metallic leaf (Figs 1.18–1.20).[2] There are also circular medallions on the ceiling of Cave D, a finely designed boar's head, horses and birds, within a circular border of pearls, typical of Sasanian patterns.

Caves S(a) and N(a) also share other similarities such as the Laternendecke ceilings comprised of beams organised in a square lantern-like shape that are finely detailed with seated Buddhas in a mandala-like pattern. Additionally, foliated scroll patterns featuring animals are painted on the middle beams of Cave N(a). In both caves, the inner sides of the central quadrangle display four bands of six birds holding ribbons

Figure 1.19 A tree deity and Bodhisattvas on the east wall of Cave N(a) before its destruction. Photo: K. Maeda, 1970s.

Figure 1.20 Seated Buddha figure discovered during conservation intervention at Cave N(a). Photo: H. Otake, courtesy of NRICPT, 2006.

Figure 1.21 Relief of a row of hamsa birds in Bamiyan's Cave 53-V in the Musée national des arts asiatiques-Guimet. Photo: Y. Taniguchi, 2006.

Figure 1.22 A grey-haired man's head with a beard, Cave N(a). Photo: M. Momii, courtesy of NRICPT, 2016.

between their beaks. They also share foliated scrolls (arabesques) that are attached to projections and resemble so-called octopus foliated scrolls. This style of octopus foliated scrolls perched on a red ground and decorated with lotus flowers in gradient colouring are reminiscent of the Gupta influence, as seen in wall paintings at the Jain temple of Sittannavāśál in Tamil Nadu (7th century) and in Cave 2 of the Ajanta caves.

The foliated scroll design and rows of hamsa birds described above were also once found in other caves in Bamiyan, although they are now lost. Cave 53-V, the foremost cave on the east wall by the Western Giant Buddha's foot, is a cave shaped like a square with a Laternendecke ceiling, a tambour and a cornice. The septum of the tambour's trilobate niche was once covered with a three-dimensional spiral foliated scroll pattern made of earthen plaster. The sides of the ceiling beams were also originally lined with rows of hamsa birds in relief (Higuchi 1980: 79), but these are now lost. A fragment stripped from this cave with hamsa birds holding ribbons between their beaks and foliated scrolls can now be seen in the Musée national des arts asiatiques-Guimet (Fig. 1.21). Cave F(a) has a domed ceiling with a niche located just over paintings on its side wall. The arch in the niche was decorated with a similar foliated scroll pattern and a row of birds feeding on flowers (Higuchi 1980: 40).

During conservation work, several figures were found on the ceiling of Cave N(a), details of which have not been reported until now. These included the right half of a grey-haired man's head with a beard (Fig. 1.22); a balding man with grey hair seen with his right arm bent and thrust upwards at a right angle (Fig. 1.23); a young man with a pronounced nose and large eyes wearing scale armour (Fig. 1.24); a young woman wearing a crown-shaped adornment with long hair hanging past both shoulders (Fig. 1.25); and two women's foreheads (Fig. 1.26). It is not known who requested these representations, each of which is only about 2 cm. These tiny figures are painted in the gaps between the regularly arranged seated Buddhas. They appear to be part of a story, but that story's substance is not yet known.[3]

The wall paintings of the third phase seem to blend different aspects, as seen in the second phase,

Figure 1.23 A balding man with grey hair seen with his right arm bent and thrust upwards at a right angle, Cave N(a). Photo: M. Momii, courtesy of NRICPT, 2006.

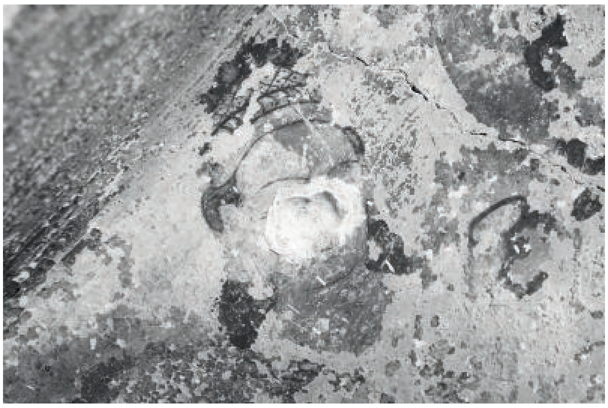


Figure 1.25 A young woman wearing a crown-shaped adornment with long hair hanging past both shoulders, Cave N(a). IR image: Y. Taniguchi, courtesy of NRICPT, 2013.

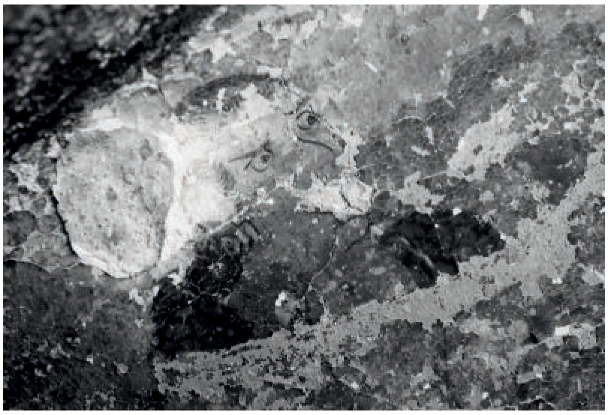


Figure 1.24 A young man with a pronounced nose and large eyes wearing scale armour. IR image: Y. Taniguchi, courtesy of NRICPT, 2013.

Figure 1.26 Two human figures with bunned hair, Cave N(a). Photo: M. Momii, courtesy of NRICPT, 2006.

Figure 1.27 A row of seated Buddhas on the vaulted ceiling of Cave K_3. Photo: F. Colombo, courtesy of NRICPT, 2006.

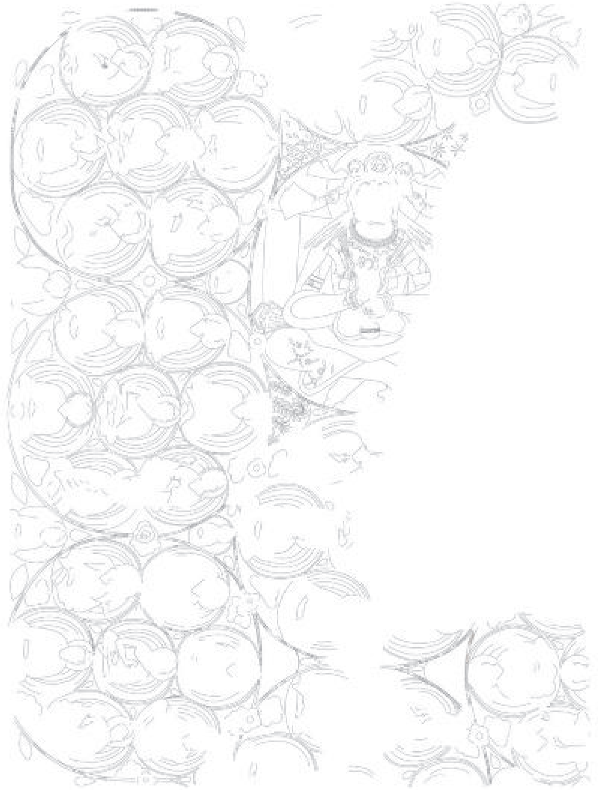


Figure 1.28 The vaulted ceiling of Cave K_3, based on field observation records and sketches by Y. Taniguchi from 2006. Line drawings: M. Aso, 2021.

Figure 1.29 Colour scheme of the vaulted ceiling of Cave K_3, based on field observation records and sketches by Y. Taniguchi from 2006. Line drawings: M. Aso, 2021.

but contain no crude lines or rough wall surfaces. Generally, they are located in smaller, square-shaped caves with Laternendecke ceilings such as Caves S(a), N(a), L and F(c). These wall paintings depict a Thousand Buddha motif in elaborate colours and have mandala-like features sometimes referred to as 'proto-mandalas' (Klimburg-Salter 2003). The entrance to Cave F(c) also has a painting of the Parinirvana (reclining) Buddha. Cave K_3, with its vaulted ceiling, has a large circular composition featuring a circle of six seated Buddhas around a Bodhisattva with alternating green and blue circles in the background (Fig. 1.27). Like Cave E(e), which is square with a domed ceiling, the composition has a Thousand Buddhas circling around the Bodhisattva image.

Figures 1.28 and 1.29 are line drawings based on field observation records and sketches from 2006. The initial records were corrected by Miki Aso who accounted for photographic distortions of the Thousand Buddhas on the vaulted ceiling by referring to drawings flattened through the works provided by the PASCO Corporation.[4] She also reconfigured the line drawings digitally (using Adobe Photoshop and Illustrator) while developing a colour scheme by taking into account discoloration identified in scientific analysis. The seated Buddhas did not follow a regular pattern of intermittent blue and green, and the arrangement of the head and body of these Buddhas changed according to whether the interior was green or blue. Although the flora in the background were not analysed, they appear to have a similar colour scheme to one example from Cave 285 in the Mogao grottoes, which changed from dull blue to green by mixing orpiment and indigo (Takabayashi *et al.* 2008).

The wall paintings of the fourth phase are essentially similar to those of the third phase. From the third phase onwards, there is a mixture of styles, but the influence of the Hellenistic world seems to diminish and a greater proportion of wall paintings contain foliated scroll and early mandala-like compositions.

1.2.3 Wall paintings in the Foladi caves

There are approximately 50 grottoes in the Foladi Valley, which lies about 3 km southwest of the Bamiyan main cliff (see Fig. 1.7). Wall paintings can be found in Foladi Caves 2, 4, 5 and 6, each of which are small with Laternendecke ceilings and domes. Because cracks in the cliffs in the Foladi Valley have led to the collapse of wall paintings and damage to

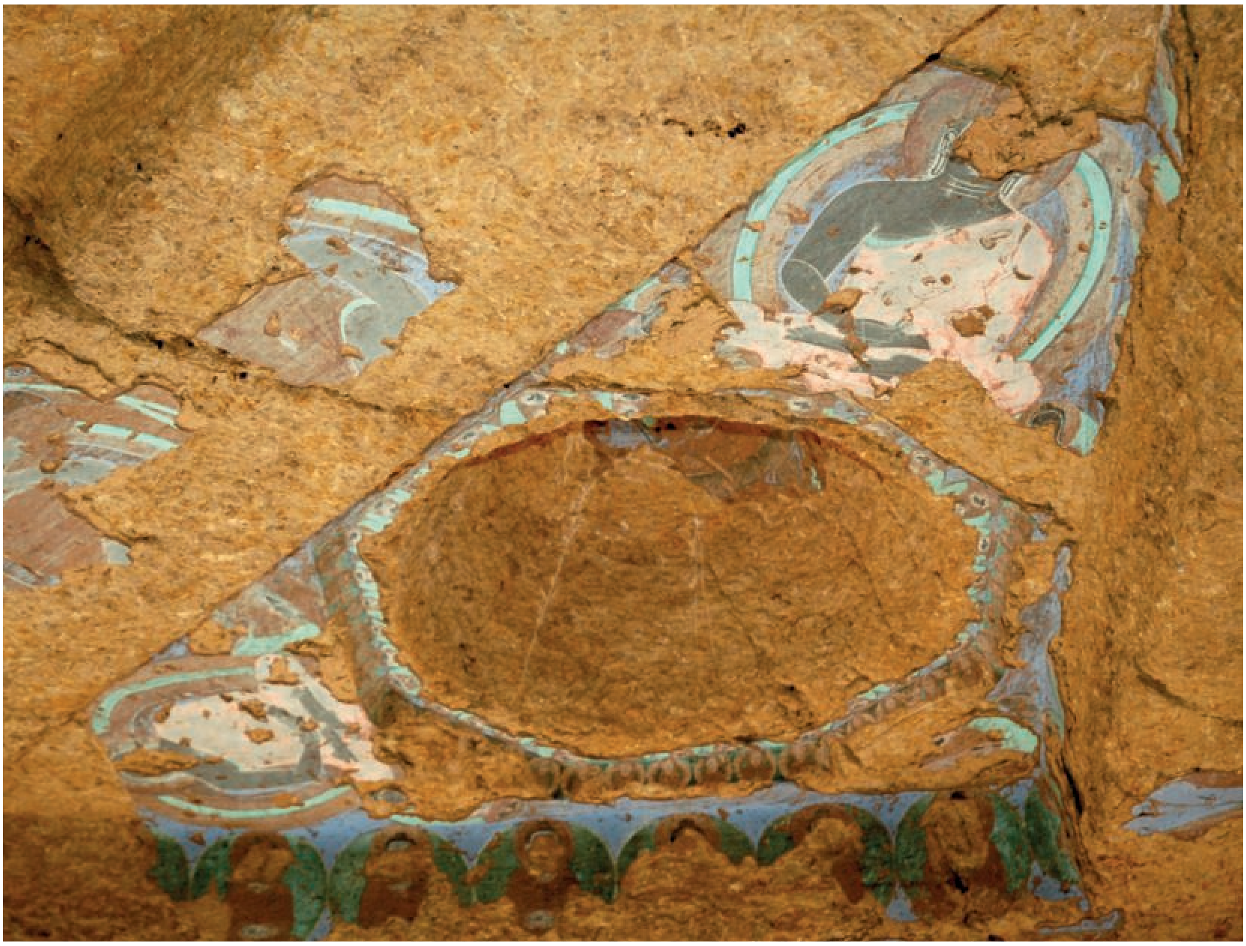


Figure 1.30 A row of seated Buddhas depicted on the square vaulted ceiling of Foladi Cave 4. Photo: H. Otake, courtesy of NRICPT, 2006.

the cave floors, it is difficult to assess the original state of the wall paintings.

Compositions in the Foladi caves typically consist of a seated Buddha in the centre of the dome, with several seated Buddhas and stupas in an alternating pattern around it (Fig. 1.30). At the centre of a group of seated Buddhas on Foladi Cave 4's dome ceiling is an image of the crowned, or bejewelled, Buddha. Radiocarbon dating indicates that the caves with wall paintings emerged relatively late: two date to the second phase in the mid-7th century, Foladi Caves 4 and 6 date to the third phase, and the front chamber of Foladi Cave 5 dates to the fourth phase.

There are few stylistic differences between paintings from the second phase as seen in Foladi Cave 2 and the rest, which are believed to be later. Additionally, each cave has its own distinctive colour palette. In Foladi Cave 4, the ground is white-green and light blue (only green and brown paints are conspicuous) and the skin of the seated Buddhas appears to have turned dark brown (Fig. 1.31). In Foladi Cave 5, the ceiling is covered with a sooty black substance obscuring a clear view of the wall paintings, but

Figure 1.31 The Thousand Buddha motif on the ceiling of Foladi Cave 6. Note: the space between the Buddhas has turned black. Photo: F. Colombo, courtesy of NRICPT, 2007.

Figure 1.32 Enlargement of the row of seated Buddhas of Foladi Cave 6. Photo: H. Otake, courtesy of NRICPT, 2006.

colours including white-green, light blue and pink are visible. In Foladi Cave 6, seated Buddhas are placed in a mandala-like arrangement on each level of the ceiling, and the overall colour scheme appears to be black and white (Fig. 1.32). However, observations suggest that this is merely a blackening of the green colour. Just below the ceilings of Foladi Cave 4, there is a pattern similar to the foliated scroll pattern seen in the Bamiyan Caves F(c) and N(a), particularly with regard to the way lines are drawn and how the seated Buddhas have been composed.

1.2.4 Wall paintings of the Kakrak caves

The Kakrak caves are located about 3 km southeast of the Bamiyan main cliff. They consist of a large Buddhist niche and a group of about 100 caves (see Fig. 1.8). However, only a few caves are clearly defined as Buddhist temples owing to the varying ways they were used over subsequent centuries. In addition, more recent events – including the stripping of works by a foreign team and destruction during the civil war– have left the caves with few wall paintings remaining today.

The dome ceiling in Kakrak Cave 43,[5] stripped by a French expedition, shows a seated Bodhisattva surrounded by a double circle at its centre. The dome's outer zone is a mandala consisting of seven circular rings with 11 seated Buddhas surrounding a seated Buddha. There is also a hunting king dressed in Sasanian-style clothing. According to radiocarbon dating of fragments from the cave, this wall, like the Foladi wall paintings, belongs to the third phase.

Figure 1.33 shows one fragment of a Kakrak wall painting from an unknown cave that is held in the collection of the Musée national des arts asiatiques-Guimet. The fragment displays a group of seated Buddhas arranged in a circle against a green background. They are surrounded by body nimbi, lotus flowers and trees with red branches that shift in hue from yellow to red. The red lines of the robes, the gradient colouring in the faces, the precision of the brushwork and the iron-wire line strokes[6] suggest that the painting techniques used were very similar to those of the wall paintings in Cave N(a), and may even have originated from the same workshop. The line strokes resemble an iron wire due to their hard, evenly drawn lines with no inflection. The wall paintings at Kakrak were made in the third phase.

1.2.5 Wall paintings in the Qol-e Jalal caves

In 2005, a new cave with Buddhist wall paintings was identified at Qol-e Jalal, located at the western end of the Bamiyan Valley about 6 km west from the Western Giant Buddha (Iwai 2006a). Approximately 50–60 caves, now used as dwellings, stand on the cliffs of Qol-e Jalal. The cave with paintings at Qol-e Jalal is believed to have originally consisted of a front chamber and a main chamber, but the front chamber is now collapsed. The chamber in which the wall paintings are located is small, square and topped by

Figure 1.33 Kakrak wall painting fragment (cave unknown) in the Musée national des arts asiatiques-Guimet. Photo: courtesy of Musée national des arts asiatiques-Guimet (2008, cat. no. 284, p. 362).

a Laternendecke ceiling with a dome at its centre, much like caves in the Foladi Valley. Most of the inner surface of the cave is covered with soot-like black material. In some parts, the earthen plasters have fallen, leaving the wall paintings in a poor state.

In a few places, such as the west wall, wall paintings with vivid blue and crimson colours have been found. The image on the west wall depicts a seated Thousand Buddha motif surrounded by smaller seated Buddhas with fine brushwork in iron-wire lines, some of which are decorated with fine strips of gold leaf. The body of the Buddha is painted in a gradient of hues that shift from red to blue. In the space between the Thousand Buddhas are designs of lotus flowers, also in gradients of red to white (Fig. 1.34). The lower part of the wall is punctuated with strips of the Sasanian medallion motifs (Fig. 1.35). One example of this curling foliage-like design can be seen on the ceiling of Cave D.

Because the layer of rendering for the wall painting at Qol-e Jalal was very thin and contained very little straw fibre, we were unable to acquire the necessary samples for radiocarbon dating; therefore the radiocarbon date for the Qol-e Jalal paintings is not known. However, the painting is believed to belong to the third or fourth phase based on its Thousand Buddhas composition, the use of gradient colouring and its brushwork.

Figure 1.34 A section of a wall painting with a floral pattern using colour gradation in the Qol-e Jalal cave. Photo: S. Iwai, courtesy of NRICPT, 2005.

Figure 1.35 Wall paintings at the Qol-e Jalal cave, with their Sasanid pearl-roundel motif. Photo: S. Iwai, courtesy of NRICPT, 2005.

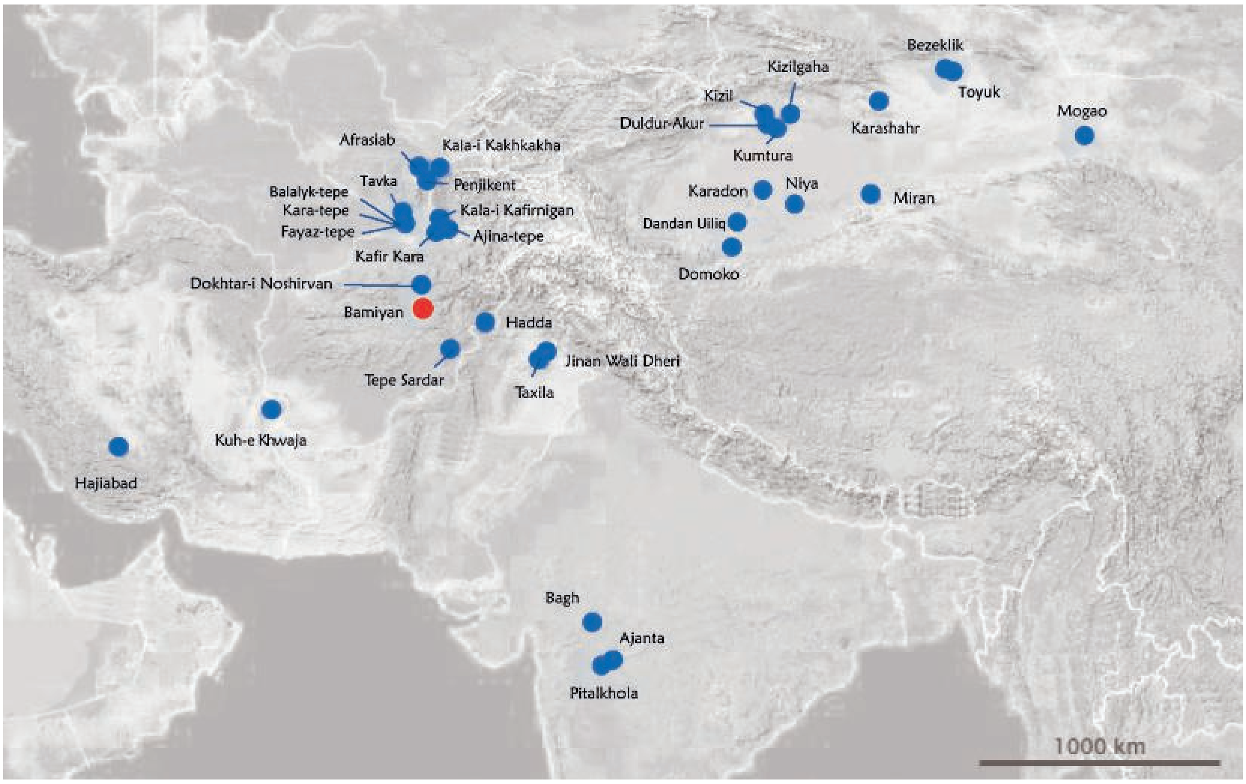


Figure 1.36 Map of the distribution of major archaeological and Buddhist sites with secco wall paintings on earthen walls (2nd–8th century).

1.3 The chronology of Bamiyan's wall paintings and their relationships to neighbouring works

When Buddhist grottoes were emerging in Bamiyan, artisans influenced by the Greco-Roman world and the Sasanian dynasty began creating wall paintings such as those in the niche surrounding the Eastern Giant Buddha in the first phase. The wall paintings of the second phase, including those in the niche surrounding the Western Giant Buddha, were painted by a variety of artisans influenced by materials, methods and techniques from the Gupta dynasty, Gandhara, Sasanian dynasty, Central Asia and the Tarim Basin (Fig. 1.36).

In the 7th and 8th centuries, Greco-Roman motifs depicting Sun Gods and princes in Central Asian costumes disappeared, and seated Buddhas painted with fine brushstrokes and vivid colours, much like a mandala, began appearing throughout Bamiyan and the surrounding valleys. Wall paintings at Foladi and Kakrak share some similarities with Bamiyan Caves B(d) and F(c) from the third phase, particularly regarding the composition of lines and the seated Buddhas.

It is difficult to determine from where the artisans who created these mandala-like wall paintings with exquisite brushwork came, as no clear links exist. Viewed more broadly, however, this is a reminder of the significance of esoteric Buddhism introduced through Kashmir and Tibet from the 7th century onwards.

Stone carvings and stucco work are prevalent in the Gandhara region in today's Pakistan, whereas wall paintings are less common: the few found appear to have been executed in the *a secco* technique, closer to the techniques used in the Ajanta caves in India. Similarly, there are very few examples of wall paintings in Iran. Sasanian painting is characterised by strong black outlines and detailed lines, but these have little in common with the Bamiyan wall paintings, which feature iron-wire lines. Iranian wall paintings appear to have used the *a secco* technique and a limited range of earth colours such as ochres.

The Sogdiana wall paintings were produced in a different cultural context from that of Buddhism, so any direct comparison with the Bamiyan wall paintings should be made with caution. However, there are several aspects that are reminiscent of the first and second phases of the Bamiyan wall paintings: the use of Sasanid motifs from Persia, the iron-wire strokes, the use of gypsum as a white ground, and the application of a black layer underneath a blue layer of lapis lazuli (see Chapter 2).

Figure 1.37 (Left) A wall painting fragment (02CDF1:001) from the Buddhist temple at Dandan Oilik; (top) a composite drawing of the wall painting fragment and (bottom) a painting to the western side of the south wall showing iron-wire brushstrokes (after Kojima *et al.* 2007: 295).

Techniques similar to those employed in Bamiyan's second to fourth phases (from the 7th century onwards) can be found in the Tarim Basin, as evidenced in the common use of iron-wire brushstrokes. In particular, these techniques stand out in Buddhist monuments on South Tianshan Road, as well as in wall paintings around Kucha including those at Kizil, Kizilgaha and Simsim.

Wall paintings at the Dandan Oilik and Domoko sites along the South Western Highway share some commonalities with Bamiyan's second to fourth phases, such as the use of lead-containing pigments, gradient colouring, iron-wire strokes, foliated scroll patterns, and other techniques and motifs. Furthermore, the work of 7th-century painter Yü-eh'ih I-sêng, particularly his outstanding iron-wire strokes, is also known here. It is possible that the wall paintings at the Dandan Oilik site and elsewhere reflect colouring techniques characteristic of the Tarim Basin area, such as those of the Yü-eh'ih I-sêng school (Fig. 1.37).

The wall paintings around Khotan by the Southwest Passage are considered to hold a key to identifying the origins of the iron-wire stroke style visible in Bamiyan's wall paintings as well as links between Sasanid Iranian, Kashmiri Buddhist, Tibetan Buddhist and post-Gupta techniques. In particular, Kashmiri Buddhism and Tibetan Buddhism appear to share esoteric elements that may have influenced the Bamiyan wall paintings from the second phase onwards, especially with regard to wall paintings of the third phase.

1.4 Past research on technical materials and problems

1.4.1 Wall paintings removed by the Central Asian expeditions of the Great Powers

1.4.1.1 Wall paintings removed by the Central Asian Expeditions

The history of research on Central Asian wall painting techniques starts with expeditions to Central Asia sent by the so-called Great Powers, or imperialist states of the mid-to-late 19th century. Tables 1.3 and

1-4 list the results to date of representative analyses of pigments from Central Asian wall paintings as carried out by researchers from different countries.

From the late 19th to the early 20th century, imperialist rivalry between Britain and Russia led to state-sponsored geographical and military surveys across Central Asia and around the Tarim Basin. Over the course of these surveys, several important archaeological and historical discoveries were made, which led to the dispatch of organised scientific expeditions backed by state governments and their museums. Among the leading explorers were Sven Anders Hedin (Sweden), Albert Grünwedel (Germany), Albert von Le Coq (Germany), Sir Marc Aurel Stein (UK), Paul Eugène Pelliot (France) and Sergey Fyodorovich Oldenburg (Russia) as well as the Otani Expedition by Otani Kozui (Japan) and others of the Otani sect of Jōdo Shinshū (also known as Shin-Buddhism/True Pure Land Sect). Many of the major sites along the Silk Road – such as Bamiyan, Dunhuang, Khotan, Turfan, the Kizil Thousand Buddha caves, the Bezeklik Thousand Buddha caves and the Mogao caves – were explored during this period. Competition for the procurement of cultural heritage was fierce, with the first to arrive often claiming wall paintings and clay sculptures.

These Central Asian expeditions resulted in the plunder of a wide variety of materials, including scriptures, textiles and artefacts such as wall paintings, many of which have since been kept at the National Museum New Delhi, the State Hermitage Museum of St Petersburg, the Musée national des arts asiatiques-Guimet in Paris, the Tokyo National Museum, the National Museum of Korea, the Asian Art Museum, Berlin State Museums and the Ryukoku Museum, Kyoto, among others.

The majority of the wall painting fragments from Bamiyan are housed in the Musée national des arts asiatiques-Guimet and the National Museum of Afghanistan in Kabul. The 102 fragments and sculptures, purchased by Professor Ikuo Hirayama and protected by the Japan Committee for the Protection of Displaced Cultural Properties, were returned to the National Museum in 2015. These objects had been stolen from the museum and archaeological sites such as Bamiyan. The fragments were cut from wall paintings at the Bamiyan and Foladi sites and sold to antique dealers overseas in the 2000s, largely due to Afghanistan's deteriorating security situation at the time. Fragments of wall paintings were also collected during Benjamin Rowland's Bamiyan survey in 1936.

Prior studies on materials used in wall paintings have generally concentrated on inorganic pigments. The history of pigment research dates back to its first scientific surveys in the 1930s, studying materials brought back by Rowland from Bamiyan and the Kizil caves (Gettens 1938a,b). Other than these, however, scant scientific analysis of the materials exists, probably due to the fact that at the time, wall painting fragments were brought back to museums and art galleries, but then often ignored and left unattended. As a result, apart from lacking any form of organisation, there was a general absence of records of the origin of the fragments and other data.

1.4.1.2 Harvard University's material survey of wall painting fragments from Bamiyan and the Kizil caves

In 1936, a Harvard University team led by Benjamin Rowland and Johnson Townshend conducted a photographic survey of the Bamiyan Valley and interpreted the iconography found in ceiling paintings by the Eastern Giant Buddha. In total, 13 samples were taken from caves surrounding the Eastern Giant Buddha: 4 from Caves B, 1 from Cave B-1, 6 from Caves C and 2 from Cave G. These samples were then taken back to the Fogg Art Museum at Harvard for analysis of pigmentation and binding media carried out by Rutherford J. Gettens, a pioneer in the study of pigments: one of the earliest scientific analyses of materials used in Bamiyan's wall paintings (Gettens 1938a).

According to Gettens, wall paintings consisted of an earthen render with organic fibres, a white ground layer made of gypsum and a layer of paint. After soaking the specimen in water and viewing it in its swollen state under the microscope, he found that animal glue served as its binding media, as traditionally seen in Asia. For their pigments, Gettens identified minium (lead oxide), red ochre, yellow ochre, natural ultramarine (lapis lazuli), charcoal black and chrysocolla (hydrated copper phyllosilicate, a copper-based green pigment). He also recorded a layer of charcoal black mixed with gypsum of similar thickness beneath layers of natural ultramarine.

In addition, the Fogg Art Museum purchased some wall painting fragments from the Kizil caves brought back by Le Coq in 1926. Rowland found a correlation in the colour and style of wall paintings from around the Tarim Basin (Kizil) with those from Afghanistan (Bamiyan) (Rowland 1938b). Using the same samples, Gettens (1938b) conducted a comparative study of painting materials and techniques, which found that the wall paintings at the Kizil caves also consisted of

Table 1.3 Results from analysis of pigments in Central Asian wall paintings (1).

Location	*Site*	*Analysed by*	*Whites*						*Yellows*				*Reds*			
Gansu	Mogao	Warner		Lime	Lead white			Kaolinite							Cinnabar/vermilion	Madder
Gansu	Mogao	Gettens		Lime	Lead white			Kaolinite				Gamboge	Red lead	Haematite	Cinnabar/vermilion	
Gansu	Mogao (Cave 285)	Takabayashi								Orpiment			Red lead	Iron oxide	Vermilion	Organic red
Gansu	Mogao (Cave 220, 223, 257)	Duang *et al.*			Lead white			Ca, K, si, Al, Mg					Red lead or litharge		Cinnabar/vermilion	
Gansu	Mogao	Li	Gypsum/Anhydrite	Calcite	Cerussite, cotunnite, anglesite	Mica	Talc	Kaolinite					Red lead	Iron oxide	Cinnabar/vermilion	
Turfan	Sänggim-aghiz	Riederer	Gypsum/Anhydrite												Cinnabar/vermilion	
Turfan	Turfan	Riederer								Orpiment					Cinnabar/vermilion	
Turfan	Bezeklik	Gairola	Gypsum										Red lead	Siena		
Turfan	Bezeklik	Riederer	Gypsum/Anhydrite		Lead white				Yellow ochre	Orpiment	Massicot		Red lead	Red ochre	Cinnabar/vermilion	
Turfan	Bezeklik	Li	Gypsum/Anhydrite										Red lead	Red ochre	Cinnabar/vermilion	
Turfan	Chotscho	Riederer	Gypsum/Anhydrite		Lead white				Yellow ochre		Massicot		Red lead	Red ochre	Cinnabar/vermilion	
Turfan	Murtuk	Riederer	Gypsum/Anhydrite												Cinnabar/vermilion	
Karashar	Shorchuk	Riederer	Gypsum/Anhydrite		Lead white				Yellow ochre				Red lead	Red ochre	Cinnabar/vermilion	
Kucha	Kizil	Li	Gypsum/Anhydrite	Calcite		Quartz							Red lead	Haematite	Cinnabar/vermilion	
Kucha	Kizil	Hermitage Museum	Gypsum (ground)	Lime + qz									Red lead			
Kucha	Kizil	Gettens	Gypsum										Red lead	Red ochre		
Kucha	Kizil	Riederer	Gypsum/Anhydrite						Yellow ochre	Orpiment	Massicot		Red lead	Red ochre		
Kucha	Kumutra	Riederer	Gypsum/Anhydrite		Lead white						Massicot		Red lead	Red ochre		
Kucha	Simsim	Riederer	Gypsum/Anhydrite								Massicot			Red ochre		
Kucha	Duldur-Akur	Hermitage Museum	Gypsum		Lead white (Hydrocerussite)									Red ochre		Realgar
Khotan	Miran	Church	Gypsum											Haematite		
Kashgar	Tumxuk	Riederer	Gypsum/Anhydrite						Yellow ochre				Red lead	Red ochre	Cinnabar/vermilion	
Tajikistan	Ajina Tepa	Hermitage Museum							Yellow ochre				Red lead		Cinnabar/vermilion	
Tajikistan	Shafristan	Hermitage Museum	Gypsum								Litharge			Red ochre		
Tajikistan	Penjikent	Hermitage Museum	Gypsum (ground)	Lime + qz						Orpiment	Litharge			Red ochre		
Afghanistan	Kakrak	Hermitage Museum											Red lead			
Afghanistan	Bamiyan	Hermitage Museum	Gypsum						Yellow ochre				Red lead			
Afghanistan	Bamiyan	Gettens	Gypsum						Yellow ochre				Red lead	Red ochre		
Uzbekistan	Afrasiab	Hermitage Museum	Gypsum		Lead white (Hydrocerussite)					Orpiment					Cinnabar/vermilion	
Uzbekistan	Er Kurgan	Hermitage Museum		Lime										Haematite		
India	Ajanta	Ghosh	Gypsum	Lime				Kaolinite	Yellow ochre					Red ochre		
India	Ajanta (Cave 17)	ICR		Lime?	Lead white			Kaolinite	Yellow ochre	Orpiment	Litharge		Red lead	Red ochre	Cinnabar	
India	Sittanavasal	Paramasivan		Lime					Yellow ochre					Red ochre		

Table 1.4 Results from analysis of pigments in Central Asian wall paintings (2).

Location	*Site*	*Analysed by*	*Blues*			*Greens*				*Blacks*				
Gansu	Mogao	Warner	Lapis lazuli			Malachite				Carbon black				
Gansu	Mogao	Gettens		Azurite	Indigo	Malachite				Carbon black				
Gansu	Mogao (Cave 285)	Takabayashi	Lapis lazuli		As-pigment + indigo	Copper-containing pigment								
Gansu	Mogao (Cave 220, 223, 257)	Duang *et al.*												
Gansu	Mogao	Li	Lapis lazuli	Azurite, azurite + malachite, azurite		Malachite	Atacamite			Carbon black			Magnetite	
Turfan	Sänggim-aghiz	Riederer		Azurite/ indigo	Indigo									
Turfan	Turfan	Riederer			Indigo					Carbon black	Lamp black			
Turfan	Bezeklik	Gairola				Malachite				Carbon black				
Turfan	Bezeklik	Riederer	Lapis lazuli	Azurite	Indigo		Atacamite			Carbon black				
Turfan	Bezeklik	Li		Azurite			Atacamite							
Turfan	Chotscho	Riederer	Lapis lazuli				Atacamite			Carbon black	Lamp black			
Turfan	Murtuk	Riederer	Lapis lazuli		Indigo					Carbon black				
Karashar	Shorchuk	Riederer					Atacamite			Carbon black				
Kucha	Kizil	Li	Lapis lazuli				Atacamite							
Kucha	Kizil	Hermitage Museum	Lapis lazuli	Azurite			Atacamite	Chrysocolla?				Bone black		
Kucha	Kizil	Gettens	Lapis lazuli					Chrysocolla						
Kucha	Kizil	Riederer	Lapis lazuli		Indigo		Atacamite	Chrysocolla? Copper silicate		Charcoal black				
Kucha	Kumutra	Riederer	Lapis lazuli	Azurite		Malachite	Atacamite	Chrysocolla		Charcoal black				
Kucha	Simsim	Riederer	Lapis lazuli				Atacamite							
Kucha	Duldur-Akur	Hermitage Museum						Water soluble-copper product		Carbon black (plant ash)				
Khotan	Miran	Church				Malachite					Lamp black			
Kashgar	Tumxuk	Riederer	Lapis lazuli				Atacamite			Carbon black				
Tajikistan	Ajina Tepa	Hermitage Museum	Lapis lazuli						Green earth (Celadonite)					
Tajikistan	Shafristan	Hermitage Museum	Lapis lazuli			Malachite								Tenorite
Tajikistan	Penjikent	Hermitage Museum											Magnetite	
Afghanistan	Kakrak	Hermitage Museum	Lapis lazuli				Paratacamite			Carbon black				
Afghanistan	Bamiyan	Hermitage Museum	Lapis lazuli				Atacamite/ paratacamite							
Afghanistan	Bamiyan	Gettens	Lapis lazuli					Chrysocolla		Carbon black (plant ash)				
Uzbekistan	Afrasiab	Hermitage Museum	Lapis lazuli											
Uzbekistan	Er Kurgan	Hermitage Museum												
India	Ajanta	Ghosh	Lapis lazuli						Green earth (Glauconite)					
India	Ajanta (Cave 17)	ICR	Lapis lazuli						Green earth					
India	Sittanavasal	Paramasivan	Lapis lazuli						Green earth					

Each analytical result is shown as uniforming the style of presentation (mineral name, pigment name). Each result is based on the reports from the State Hermitage Museum data (Kossolapov *et al.* 1999), Riederer 1977, Warner from (Gray 1959), Gettens 1938a; b, Takabayashi *et al.* 2007, Duang, *et al*. 1987, Li 2005, Church 1921, Gairola 1960, Ghosh 1967, and Paramansivan 1938.

a rendering layer made of clay with organic fibres, a white ground layer made of gypsum, and layers of paint with animal glue serving as the binding media. Gettens also observed the mineralogical properties using polarised light microscopy (PLM) and wet analysis, which indicated that the green colour was copper silicate, possibly chrysocolla; the blue colour was artificially refined lapis lazuli; and the red colour was red ochre and blackened red lead (minium). In the 1990s, the Conservation Institute of Dunhuang Academy in China reported that all the green colour in the Kizil caves was atacamite ($Cu_2Cl(OH)_3$) and not chrysocolla (Su *et al*. 2003).

Gettens made a special note not only of the absence of vermilion, malachite and azurite in the Bamiyan pigments but also of the extensive use of

Table 1.5 Result of pigment analysis by Riederer (1977), modified after Vignato and Hiyama (2022: 287, table 2).

Style	*Cave number*	*German name*	*Inventory number*	*Whites*	*Reds*	*Blues*	*Greens*	*Yellows*
A	77 (statue)	Statuenhöhle	III 8184	gypsum/ anhydrite	red ochre		atacamite	
	77 (wall paintings)	Statuenhöhle	III 8838, 8839, 8840, 8841 a, 8842	gypsum/ anhydrite	red ochre, minium		atacamite	massicot
	84	Schatzhöhle C	III 8444, 8444 b, 8444 c	gypsum/ anhydrite	red ochre, minium	lapis lazuli	atacamite	yellow ochre, massicot
	118	Hippokampenhöhle	III 8412	gypsum/ anhydrite	red ochre, minium		atacamite	massicot
	129	Kleine Kuppelhöhle	III 9277	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	massicot
	207	Malerhöhle	III 8690a, 9148, 9148 b	gypsum/ anhydrite	red ochre		atacamite, chrysocolla	yellow ochre
	212	Seefahrerhöhle	III 8398, 8399, 8401	gypsum/ anhydrite	red ochre, minium		atacamite	yellow ochre, massicot
	Kumtura South Monastery Cave 22	2. Mittlere Schlucht 2. Kuppelhöhle	III 9053	gypsum/ anhydrite	red ochre		atacamite, chrysocolla	
Mixed style A + B	67	Rotkuppelhöhle	III 8403	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	massicot
B	8	Höhle mit den 16 Schwertträgern	III 8691	gypsum/ anhydrite		lapis lazuli	atacamite	massicot
	13	5. Höhle neben Sechzehnschwertträger	III 8859, 8373a, 8373b*	gypsum/ anhydrite	red ochre, minium	lapis lazuli	atacamite	massicot, orpiment
	114	Höhle mit der Gebermühle	III 9084, 9103, 9140 b	gypsum/ anhydrite	red ochre, minium	lapis lazuli	atacamite	massicot
	123	Höhle mie den ringtragenden Tauben	III 9066	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	massicot
	171	Höhle über den Kassettenhöhlen	III 8420, 8891	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	massicot, orpiment
	184	Drittletzte Höhle	III 8372 b	gypsum/ anhydrite	minium	lapis lazuli	atacamite	massicot
	188	Dritte Höhle	III 9030	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	
	198	Teufelshöhle C	III 8428 a	gypsum/ anhydrite		lapis lazuli, indigo	atacamite	massicot
	199	Teufelshöhle A	III 8431, 8432	gypsum/ anhydrite	red ochre, minium	lapis lazuli, indigo	atacamite	massicot
	219	Ajataśatenhöhle	III 8885	gypsum/ anhydrite	red ochre	lapis lazuli	atacamite	massicot
	224	Mayahöhle	III 8836, 8861, 8864, 8879, 9075	gypsum/ anhydrite	red ochre, minium	lapis lazuli	atacamite, chrysocolla	massicot

* For the identification of the mural fragments III 8373a, 8373b* as those detached from Kizil Cave 13 see the latest study by Konczak-Nagel (2020).

Original table was modified. Cave numbers have been corrected according to the latest research state, and some data which are mentioned in Riederer's text but not in the table are supplemented as well. Selected cave data are shown in this table, the exact provenance of which has been identified and is related to the Styles A and B with certainty. Identification and correction of Riederer's table were done by Satomi Hiyama.

lapis lazuli. However, as the samples brought back by the Harvard team only came from around the Eastern Giant Buddha, it would be presumptuous to infer that they represent wall paintings from Bamiyan as a whole.

In 1960, Kazuo Yamasaki of Nagoya University published a paper that summarised Gettens' findings and his own analysis of wall painting fragments from Kizil and Bezeklik, alongside wall paintings from the Main Hall (Kondo) of Japan's Horyuji Temple (Yamasaki 1960). Yamasaki's analysis used wall fragments from the Otani expedition and Le Coq, as well as those brought back by Namio Egami from Ch'ing-Ling (11th-century imperial mausoleums in eastern Mongolia). This analysis showed that pigments in the Kizil wall paintings resembled those presented by Gettens. Yamasaki identified two types of Bezeklik wall paintings: those with gypsum as a ground layer and those with lime. The results of Gettens' analysis of the Kizil and Bamiyan wall paintings were long presumed to be representative of wall materials in Central Asia.

1.4.1.3 Riederer's analysis of pigments in wall paintings from the Asian Art Museum, Berlin State Museum collection originating in and around the Tarim Basin

The Indian Art Museum (now the Asian Art Museum, Berlin State Museums) holds a collection of archaeological artefacts and wall fragments from sites around the Tarim Basin, including Kizil, Kumtura, Bezeklik, Khocho, Simsim and Tomshuk. These were obtained by four German expeditions to Central Asia between 1902 and 1914, led by Grünwedel, Le Coq and Waldschmidt. In particular, wall fragments recovered from the Kizil caves were large in scale and quantity. Some 80 wall painting fragments and painted clay sculptures from Tarim Basin sites were chemically analysed by Josef Riederer in the 1970s (Riederer 1977) (Table 1.5).

Riederer analysed mainly inorganic and organic pigments using PLM on pigment particles, microscopy on cross-sections, elemental analysis by emission spectroscopy, X-ray diffraction (XRD) and Fourier transform infrared spectroscopy (FTIR). Because the wall painting fragments had undergone consolidation and reinforcement treatments using synthetic resins for conservation, no binding media analysis was carried out. Riederer's series of studies is the only clear example of a comparative study of Central Asian wall painting technologies from a more geographically expansive perspective. That is, he considered not only data on pigments but also painting techniques and historical documentation such as the Indian text *Citrasutra* and others in classical Greek and Roman languages.

Riederer compared the results of his analysis of painting materials, and broadly categorised them into two groups: those belonging to the western part of the Tarim Basin periphery near Kucha (Kizil, Simsim, Kumtura, etc.) and those from the eastern part around Turfan (Bezeklik, Khocho, etc.). The following sections discuss some key painting materials that gave rise to regional differences based mainly on previous analyses.

In the wall paintings at Kucha, Karashahr and Turfan, a mixture of gypsum (dihydrate calcium sulfate, $CaSO_4 \cdot 2H_2O$) and anhydrite ($CaSO_4$) is often found as a white ground. However, anhydrite has not been detected in the Sogdiana wall paintings or in Bamiyan. As discussed in detail in Chapter 2 (section 2.2.5.3), the difference in mineral form due to varying hydration of gypsum may reveal whether the gypsum is of natural mineral origin or artificially calcined.

While most of the white is plaster, it is worth noting that lead white has been found in five wall paintings and painted clay sculptures from Bezeklik, Kumtura, Khocho, Sholchuk and Qum-Ariq. As all five pieces originated from sites in the eastern part of the Tarim Basin area, Riederer believed that lead white was synthesised in the east and thus discussed the relationship between lead white and Chinese artisans.

From among the pigments, three types of blue have been identified: lapis lazuli, azurite ($Cu_3(CO_3)_2(OH)_2$) and indigo. Most of the Tarim Basin's blue pigments were made from lapis lazuli, although some examples of azurite have been found at Kumtura and Bezeklik. Therefore, Riederer concluded that the western part of the Tarim Basin was dominated by lapis lazuli, while the eastern part, influenced more by China, showed the use of azurite. Indigo has also been found at Kizil and Sengum-Agis, but mostly mixed with lapis lazuli; it also tends to be detected more frequently at sites in the east.

Looking at the greens, atacamite has been found in most of the sites, whereas malachite has only been identified in the Kumtura caves. It is also interesting to note that, as suggested by Gettens' earlier work, chrysocolla has been detected in some of the wall paintings at the Kizil and Kumtura caves. Chrysocolla gives a blue-green colour with a wax-like gloss and is so rare that it was probably only used in a limited number of areas.

1.4.2 Survey of wall paintings undertaken by the USSR and Russia

1.4.2.1 The State Hermitage Museum's analysis of Central Asian wall painting pigments

When the Central Asian countries were a part of the USSR, many of their cultural heritage sites such as Tajikistan's Ajina-tepe were the focus of large-scale archaeological research, often by Russian archaeologists. The research teams collected and carried materials back to Moscow and St Petersburg. In the 1970s, Russian scientists analysed these materials (Abdurazakov *et al.* 1975), notably using wet chemical analysis and analysis of inorganic pigments and agglomerates with XRD and gas chromatography with mass spectrometry (GC-MS). For many years, this research was not accessible to much of the outside world, partly because most publications were in Russian. Moreover, scientists outside of Russia have not carried out analysis of materials used in wall paintings in Tajikistan and Uzbekistan.

Since the early 1990s, the State Hermitage Museum in St Petersburg, with support from the National Gallery of Art in Washington (CASVA) and in collaboration with Russia, France, the USA and Uzbekistan, has carried out research on wall paintings at Bamiyan and nine other sites, including Penjikent, Afrasiyab and the Kizil Caves. The findings have been compiled and reported in both Russian and English (Kossolapov and Marshak 1999).

Appendix IV.6 includes a summary of findings by Russian researchers who analysed wall paintings in the northern foothills of Hindu Kush and Sogdiana. Specifically, Russian analysis of Central Asian wall paintings has been extremely valuable as few analyses of binding media exist. In the 1970s, the Russian chemist Vadim Birstein carried out a detailed analysis of wall paintings and concluded that fruit gums such as cherry and apricot would have been used as binding media (Birstein 1975, 1977).

Following Birstein's work, Kamila Kalinina at the State Hermitage Museum has conducted further analyses of binding media using GC-MS since the 1990s. For example, polysaccharides containing large amounts of galactose and mannose, and small amounts of arabinose and xylose, have been detected in wall paintings from Penjikent. Since galactose is often obtained from leguminous species such as guar or jackfruit, binding media were likely produced by mixing gums from one of these with gums from fruit trees such as peach or apricot (Kossolapov and Kalinina 2007).

Although plant polysaccharides such as gum were often used as binding media in the Greco-Roman world, this is the first chemical identification of the use of gum to the east of the Silk Road. In particular, the verifiable use of fruit gums such as guar and apricot, each of which seem to have been adapted to the vegetation of Central Asian lands, has brought about a significant change in our understanding of painting materials in Central Asia and the relationship of trade along the Silk Road to their selection.

1.4.3 Conservation surveys of Bamiyan's wall paintings

1.4.3.1 Conservation problems pertaining to Bamiyan's Buddhist wall paintings

If the first phase of analysis was the scientific research carried out by imperial states on the detached fragments of wall paintings, the second phase would be research carried out in the field and in museums during conservation. This section gives an overview of the second phase of research on Bamiyan's wall paintings. The conservation of Bamiyan's wall paintings was a measure taken to address the effects of Central Asia's harsh natural environment and various acts of human destruction (Taniguchi and Aoki 2005).

The Bamiyan cliffs are made of fragile conglomerate and contain silt layers that are prone to structural collapse when flooded (Margottini 2004). It has also been observed that many of the wall paintings collapsed due to a loss of adhesion between the rock and the rendering layer. In addition, many of the wall paintings have been darkened by the accumulation of dust, smoke and tar-like substances from fires set by people who inhabited the caves during the winter and by refugees who lost their homes during the civil war. The pigments in many areas have undergone discoloration (e.g. whitening or blackening) due to the use of red lead, copper-based green pigments and orpiment.

One of the more serious conservation problems is vandalism. Historically, the invasion of non-Buddhists from the 9th and 10th centuries onwards is believed to have led to the destruction of faces and eyes. By the 1920s, when expeditions were visiting, many of Bamiyan's wall paintings were thought to have been severely damaged by natural collapse and the effects of sunlight, wind and rain. Later, during the civil war, many wall paintings were damaged by bullets, arrows and lacerations, and the Giant Buddha statues were destroyed by large quantities of gunpowder. The resulting vibrations and blasts had profound effects, triggering the collapse of many wall paintings. Wall paintings were also cut or removed with sharp knives and sold to collectors abroad so today, only a small number remain in the caves. Since the 1920s, French, Indian and Japanese conservators have been working intermittently on the conservation of the wall paintings during periods of stability.

1.4.3.2 Conservation and restoration by the French Corps

The first conservation project at Bamiyan started in 1923 during an archaeological survey by a French team that had exclusive survey rights (Anon. 1924a,b). The French government obtained exclusive rights to conduct archaeological research in Afghanistan for 30 years under its so-called 30-year plan[7] (Olivier-Utard 1997: 37; Fenet 2011: 121). Thereafter, it established the French archaeological delegation in Afghanistan, DAFA (Délégation archéologique française en Afghanistan) under the leadership of Sorbonne University professor Alfred Fouché, and conducted archaeological research at various sites in Afghanistan including Hadda, Ay Khanum and Bagrām. The Bamiyan site was also the subject of pioneering archaeological research. Fouché, along with the architect André Godard and Musée Guimet curator Joseph Hackin, catalogued the caves and wall paintings (Godard *et al.* 1928; Hackin and Carl 1933; Hackin *et al.* 1959).

While conducting archaeological and art historical fieldwork, DAFA also provided 'first aid' to the missing parts of wall paintings and their surrounding areas using gypsum mortar. In addition, a massive brick buttress was constructed on the west side of the Eastern Giant Buddha to provide structural reinforcement for the crumbling cliff façade where the caves had been opened (Taniguchi and Aoki 2005). It appears that pigment analysis of the wall paintings was not carried out at the time of this survey.

1.4.3.3 Conservation and material surveys by Indian and Afghan teams

Following the end of France's exclusive survey rights in Afghanistan in 1952, new Bamiyan surveys were launched by various countries. The most extensive conservation and restoration work was carried out by the Indo-Afghan Corps, led by conservation scientist R. Sengupta from the Archaeological Survey of India (ASI), between 1969 and 1976 (Lal 1970; Sengupta 1975, 1977, 1984, 2002; Goryacheva 2002; Warikoo 2002). They cleaned, strengthened and reinforced the wall paintings in the caves around the Eastern and Western Giant Buddha statues (namely, Caves A, B, C, D) as well as the Giant Buddhas. The Corps also set drains to prevent snowmelt water from entering the niches, engaged in buttressing (such as removing large antechambers and cementing cracks) and restored the stairways to the caves. The main restoration work was limited to the two Giant Buddhas and the surrounding caves and wall paintings, while the many other difficult-to-access caves were left untouched. At the time of this conservation project, the ASI carried out a material survey of the wall paintings in Caves A, B, C and D (Lal 1970). This was the second such study after Gettens' analysis in the 1930s.

The analytical report by the ASI did not clarify details about the samples or from where they were taken but it is thought that they originated from the caves around the Eastern Giant Buddha. The results of the analysis correlate with those of Gettens: the white ground layer is gypsum, the yellow is yellow ochre, the red is red ochre, the blue is natural ultramarine (lapis lazuli), and the black is carbon black or lampblack. The ASI did not identify any green pigments, such as malachite or atacamite, and no red lead or lead white was detected. No analyses of binding media were carried out.

B.B. Lal, an ASI archaeologist, found that the rendering of Bamiyan's wall paintings, in common with traditional wall structures in India and other parts of Central Asia, consists essentially of mortar mixed with sand and gypsum. Furthermore, his analysis of the rather simple variation of pigments in Bamiyan's wall paintings pointed to a similarity with Ajanta, rather than the Mogao caves and the Mīrān site, where the use of organic dyes and lead white has been reported (Lal 1970). As with Gettens' analysis, because the wall paintings were concentrated in caves around the Eastern Giant Buddha, Lal's findings were limited to the early wall paintings and did not cover all four phases.

1.4.3.4 The Japanese survey of the 'displaced cultural property' of wall painting fragments

Since the two Giant Buddhas were hit with explosives in 2001, numerous archaeological sites and museums in Afghanistan have been looted and their collections sold overseas. Therefore, in 2001, the Japan Committee for the Protection of Displaced Cultural Properties was established to protect 99 cultural heritage items that were displaced from Afghanistan (102 items at the time of return in 2015), including 41 wall painting fragments that had been cut from the Bamiyan site (Bamiyan, Foladi) (42 fragments in 2007).

From 2004 to 2005, the Tokyo National University of Fine Arts and Music (now Tokyo University of the Arts) and the NRICPT jointly carried out a research project on the displaced wall paintings of Bamiyan (Yamauchi 2006b). The aim of this research was to create a photographic record of wall fragments using various light sources and methods, and to investigate painting materials by adopting non-invasive analytical methods such as visible light reflection spectroscopy, portable X-ray fluorescence (pXRF) and XRD analysis. Observations and ultra-high resolution digital photographs were obtained in natural and raking light, as well as by infrared (IR) and ultraviolet (UV) fluorescence.

Since 2006, conservation and mounting work has been carried out to ensure the return of these wall fragments to Afghanistan (Kijima *et al.* 2008, 2009) (Fig. 1.38). The non-invasive analytical survey was the first step in researching the techniques and materials associated with the wall paintings. The wall fragments protected as displaced artefacts are known to have been removed from Caves K_3, E(e) and I at Bamiyan as well as Foladi Cave 4.

1.4.3.5 Painting techniques and materials pertaining to Bamiyan's wall paintings (Caves K_3, E(e), and I)

The raking light photographic records of wall painting fragments from Bamiyan Caves K_3, E(e) and I show that the surface of the rendering wall has been smoothened. Infrared photographs clearly show outlines and underdrawings. In particular, strong and weak strokes are visible on a fragment from Cave K_3, revealing the lines of the Buddha's fingers and the folds of his robe while he is gesturing. UV fluorescence photography showed different fluorescent colours such as blue, purple, red and yellowish-white, but it was not possible to identify the specific pigments or organic materials responsible for the fluorescence.

Similarities and differences in pigments and painting techniques exist across Caves K_3, E(e) and I. All the fragments were covered with a thin, white, calcium-based ground layer over two layers of earthen rendering with chaff straw fibres, but whether this white ground was gypsum or lime could not be confirmed as light elements such as sulfhur were undetectable. The white ground layer was applied roughly with a brush and atop this layer, the Buddha, Bodhisattvas and haloes were roughly outlined in reddish-brown and then coloured. The garments were generally red in colour, often produced from a mixture of cinnabar/vermilion and red lead.

The wall painting fragments from Cave K_3 show a band from part of a nimbus with a blue-grey colour containing lapis lazuli and what appears to be a blackened copper-based green pigment. Portable XRD analysis revealed gypsum in the white ground. Other materials detected in the coloured areas included cinnabar/vermilion, minium, red ochre, yellow ochre, lapis lazuli and some kind of blackened pigment containing copper, probably from a copper-based green pigment. The red areas were often the result of the application of a layer of cinnabar/vermilion on top of minium.

The wall painting fragments in Cave E(e) are almost entirely covered with black deposits. It appears that originally they were painted with various pigments, although only black, red and yellow are now visible. Analysis confirmed minium, cinnabar/vermilion, orpiment (As_2S_3), and some kind of pigment containing copper, which had turned black.

Wall painting fragments from Cave I included shading in yellow ochre and skin colouring on the hands and face. Vivid bands of green, blue, yellowish-brown and red were preserved, and the head of the Buddha was painted with coloured spiral-shaped curls. Portable XRD analysis revealed lapis lazuli in the blue areas and gypsum as the white ground layer. Yellow pigments (possibly arsenic-containing yellow: orpiment) were detected along with minium, red and yellow ochre.

In wall fragments from the three caves, at least three types of red pigments were identified – apparently red, crimson and reddish-brown; these were thought to be minium, cinnabar/vermilion (on minium) and red ochre, mainly composed of lead, mercury and iron, respectively. Different shades of red were used in the representation of red garments and spheres, as were layering techniques, as seen in the wall paintings of Caves K_3 and E(e).

The discoloration of minium, as seen in wall painting fragments in Caves K_3 and E(e) had already been pointed out by Gettens (1938a). The blackening of minium has been found in many places – for example, the discoloration of minium in Central Asia has recently been attributed to the presence of soluble salts and the formation of laurionite (lead hydroxychloride, PbClOH) and plattnerite (lead oxide, PbO_2) (Kossolapov and Marshak 1999).

Figure 1.38 After conservation and mounting work at Tokyo University of the Art for the displaced wall painting fragment from Cave K_3. Photo: T. Kijima, courtesy of Tokyo University of the Art, 2008.

Photographic documentation of raking light on wall painting fragments at Foladi Cave 4 also revealed the technique used to smooth the wall surface. The rest of the fragments differed significantly from the Bamiyan fragments from Caves K_3, E(e) and I. First of all, unlike wall paintings at Bamiyan, the rendering of wall paintings from Foladi Cave 4 apparently consists of two layers: a thick, light brown earthen render with chopped straw fibres, and a reddish-brown, hard upper layer of about 3 mm, slightly coarse but with homogeneous sand grains. The reddish-brown upper layer, which is composed of heavy sand grains tightly packed, may have been kneaded with some kind of resinous material as used in Indian earthen wall construction techniques.

A thick yellow transparent resinous layer is always found between the paint, ground and rendering layers. This resinous layer, which has scaly cracks that also appear in the upper painted layer, often exudes from the cracks in the painted layer, and in some places is also responsible for the staining of the wet colour. It is assumed that this layer was applied to the wall paintings as a sizing.

The major difference between Foladi and Bamiyan (from Caves K_3, E(e) and I) wall paintings is the white ground layer. Lead was detected in the white layer, and analysis by pXRD revealed hydrocerussite

($Pb_3(CO_3)_2(OH)_2$), the main component of lead white, and realgar (As_4S_4). Its pale yellowish-white colour is due to the presence of small amounts of orange-coloured realgar particles in the white substrate.

Some of the Buddha's faces are dark reddish-brown with white lines for detail, while some details of the face and hands have faded and are no longer recognisable. Most of the body nimbus is white-green with copper, and the visible light reflectance spectrum is similar to that of malachite (basic copper carbonate) or atacamite (basic copper chloride). Portable XRD also detected peaks that may contain both phases, but identification was difficult. The garment is coloured crimson with cinnabar/vermilion or salmon pink with lead white, and the space outside the body nimbus is filled with two layers of lapis lazuli blue or black, possibly discoloured by some copper pigment. The blue colour of the wall paintings in Foladi Cave 4 is characterised by fine and coarse grains of lapis lazuli particles. Cinnabar/vermilion was used for the fine outlines and details of clothing.

Copper was detected in areas that now exhibit black coloration. It is thought that some pigment containing copper has turned black, and that this was probably where a copper-based green pigment and an organic material were used, rather than a green pigment from a simple copper compound such as atacamite or malachite. The darkening of these green copper salts is discussed further in Chapter 2 (section 2.3.1).

1.4.3.6 Current status of conservation and analysis of Bamiyan's Buddhist wall paintings by Japan[8]

Following the destruction of the two Giant Buddhas, international momentum for the protection of archaeological sites and cultural heritage has grown, and research on protection has made significant progress. In June 2003, a meeting of the International Coordination Committee for the Rescue of Afghanistan's Cultural Heritage was held at the UNESCO headquarters in Paris, and the Bamiyan Site Safeguarding Project was launched by UNESCO, Japan, Germany and Italy (Yamauchi 2005). Japan has been involved in the conservation of the remaining wall paintings at the site since 2004, including the protection and management of fragments from wall paintings destroyed and scattered during the civil war, as well as the conservation of the wall paintings in Caves I and N(a) (Taniguchi 2007).

Many of the wall paintings at the Bamiyan site are in poor condition due to the risk of collapse, rainwater penetration, discoloration and black sooty material and require urgent measures to be taken. In order to conserve the wall paintings using appropriate materials, it was necessary to know from what materials they were made and the causes of discoloration. Therefore, samples were taken and analysed from as many of the caves as possible, representing a variety of painting materials, in order to cover the whole area (Taniguchi 2006; Taniguchi and Cotte 2008; Taniguchi and Mazurek 2008; Taniguchi *et al.* 2006, 2007). This series of analyses is discussed in Chapter 2.

Notes

1. A posture in which the hips are twisted to one side and the centre of gravity is placed on the twisted leg, with the other leg slightly bent at the knee.
2. A painting method in which a colour's hue graduates from dark to light and/or light to dark.
3. Some of these images were recorded using infrared photography.
4. https://www.pasco.co.jp/eng/.
5. Of the wall paintings stripped from the dome, half are on display at the Musée national des arts asiatiques-Guimet and half at the National Museum of Afghanistan. The Japanese team visited both collections in 2005 and 2006.
6. One of the techniques evident in *Eighteen Outlinings (shi-ba-miao-fa)* (Yamamoto 1883), in which Zhou Lüjing of the Ming dynasty (1368–1644) divided types of clothing used in successive generations of figure painting into 18 categories. According to Zhang Yanyuan's *Lidai Minghua Ji (Famous Paintings through History),* Yü-ch'ih I-sêng was the first painter in the Tang dynasty (618–690, 705–907) to bring Western painting techniques to the Tang court (Zhang 1977). Yü-ch'ih I-sêng's father, Yü-ch'ih Pa-chih-a, a native of the Khotan, was also a painter in the Sui dynasty (581–618). He has been credited with having helped advance the Western technique of using lines of unvarying thickness to outline figures, as seen in the 'iron-wire' line in Buddhist temples of the great cities of China (Schafer 1963: 32).
7. Texte de la Convention concernant la concession du privilège des fouilles archéologiques en Afghanistan, Kaboul, 9 septembre 1922.
8. As of 2021.

Chapter 2: The painting structure and composition of the Bamiyan wall paintings

2.1 Objectives and methodologies for analysing wall painting materials and techniques

Of the more than 700 caves remaining in the Bamiyan Valley, approximately 50 contain wall paintings.[1] Identifying painting techniques used in wall paintings requires comprehensively analysing as many as possible and understanding the centuries of influence exerted by Bamiyan's Buddhist monasteries. This chapter discusses the scientific analysis of Bamiyan's Buddhist wall paintings and its findings.

In most of Bamiyan's grottoes, wall paintings incorporate multiple colours such as the basic colours of white, yellow, red, blue, green and black, as well as metal leaf, therefore several colours from each cave were selected for investigation. Moreover, because the surfaces of the Eastern and Western Giant Buddhas were also painted, they were included in the survey. The principle of this study was fundamentally: (i) to observe the wall paintings *in situ* and research available knowledge regarding their colours and painting techniques, and (ii) to analyse wall paintings in the field as well as in the laboratory by sampling and microanalysis of minute fragments. However, many of Bamiyan's caves and artefacts have been destroyed, and only a few paintings remain on the walls. As a result, fragments have been scattered on cave floors or stolen and sold overseas. We collected samples for analysis from cave walls and fallen fragments. Security issues complicated our ability to carry out long-term research in Bamiyan and restricted observation and recording in the field.

Analyses of the wall paintings required not only examining pigments, as in most prior studies, but also consideration of how the wall itself was constructed and painted. Thus, we collected data to aid in the determination of the structures and steps involved in producing each painting. Results from

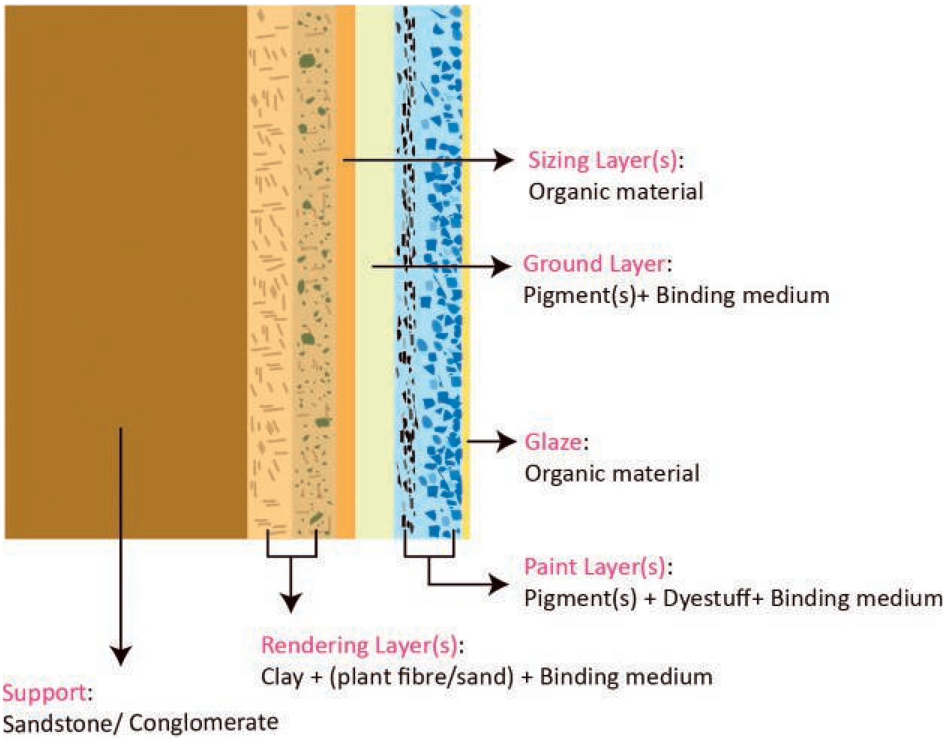


Figure 2.1 Schematic drawing of the layered structure of Bamiyan's wall paintings.

previous analyses (Yamauchi 2006b) show that the wall paintings have a multilayered structure with an open-cut support of cliff-forming conglomerate and sandstone, an earthen layer of rendering containing straw fibres and animal fibres, an organic sizing layer, a buffer layer, a white ground layer and multiple layers of paint (Taniguchi 2006). Figure 2.1 is a schematic diagram of one of the structures found among Bamiyan's Buddhist wall paintings.

Although there is no written record of the painting techniques employed in Central Asia, it is possible that organic materials such as resins, plant gums, animal glues and oils were used in different layers on the walls such as in the earthen renders, the ground and painting layers, as seen in the *Citrasutra* of India (Appendix I). Therefore, we decided to analyse the layered structure by studying cross-sections using optical, electron and synchrotron microscopes. Wall paintings were expected to contain a wide range of substances, such as pigments (including organic, inorganic, mineral and artificial, as well as dyes), binding media (organic) and sizing (organic). We analysed both organic and inorganic substances contained in each layer to capture their chemical elements and mineral or crystalline states.

The description of the paint stratigraphy was first performed on raw fragments and resin-embedded cross-sections using optical microscopy (in visible and UV light), complemented by field observation of the paintings. A scanning electron microscope with an energy-dispersive X-ray spectrometer (SEM-EDS) was then used to obtain elemental information on inorganic materials, particularly for the measurement of micro areas in each layer. Approximately 50 samples were analysed using synchrotron radiation (SR)-based techniques at the European Synchrotron Radiation Facility (ESRF). Most of the work was carried out in 2006 at the ID21 and ID18F beamlines but some samples were re-analysed in 2021 at the ID13 and ID21 beamlines. Synchrotron radiation-based micro-X-ray fluorescence (SR-μXRF) mapping was employed to identify and locate elements, particularly metals. The crystalline components were further identified and mapped using synchrotron radiation-based micro-X-ray diffraction (SR-μXRD) mapping. Synchrotron radiation-based micro-Fourier transform infrared spectroscopy (SR-μFTIR) was used to identify and map molecular groups, giving access to both organic and some inorganic components.

FTIR is very useful for determining the material structure of organic compounds as it gives information on the molecular structure. However, this information is qualitative and does not facilitate distinctions between sub-types of components (such as types of oils or proteins). More precise information is needed to quantitatively measure and identify organic substances. Therefore, for this purpose, gas chromatography with mass spectrometry (GC-MS) was used to break down and detect the organic substances. Since GC-MS provides both chromatograms and mass spectra simultaneously, the identification of each peak component is relatively easy. The analysis was carried out using settings for amino acids (animal glues, eggs, casein, etc.), fatty acids (oils, beeswax, resins) and polysaccharides (plant gums).

However, GC-MS alone cannot reveal the origin of proteins composed of amino acids (animal glue, egg white, egg yolk, casein, etc.) therefore enzyme-linked immunosorbent assay (ELISA) was used to identify the origin of proteins from cows, poultry and fish. Although still in its trial stage, this method allows researchers to detect and quantify the concentration of antibodies or antigens in a sample. Animal species were also identified using liquid chromatography with mass spectrometry (LC-MS).

By identifying the variety of organic and inorganic materials found in wall paintings, we may cultivate a comprehensive understanding of how these paintings were produced, but only to the extent that analytical techniques are executed in a complementary manner. For instance, identifying particular layering strategies may lead to a better understanding of visual aesthetics and painting techniques. Similarly, comparisons across eras could provide insight into historical contexts. In addition, findings could also be useful when deciding upon the most appropriate materials and methods for conservation.

The methodology is described in detail in Appendix II. The next section gives an overview of the results obtained from each analysis with a summary provided in Tables 2.1 and 2.2. Information on individual pigments and colorants identified using SR-μXRF, SR-μXRD and SR-μFTIR and the results of the GC-MS analysis of fatty acids, beeswax and resins, and the ELISA analysis of proteins and polysaccharides are summarised in Appendix III. Section 2.2 summarises the overall structure of the wall paintings and includes a discussion of materials used in each layer of the composition. Chapter 3 then discusses the results with a focus on oil painting techniques commonly found in Bamiyan.

Table 2.1 Summary of identified and possible grounds, pigments, binding media and other decorations in Bamiyan wall paintings.

Location	Ground layer	Pigments	Binding medium	Sizing	Glaze	Mordant	Alteration products	Organic substances in any layer
WGB	None							
J(b)	Partially no ground	Lapis lazuli, copper-containing blue (azurite?), atacamite, gypsum						
C(a)		Copper-containing green, arsenic containing pigment, lime	No amino acid					
E(e)	Gypsum	Copper/zinc-containing mineral						
G		Yellow ochre	Amino acid (protein/ polysaccharide)					
H(a)			Amino acid (protein/ polysaccharide)					
H(b)								
I niche		Cinnabar/vermilion, minium, gypsum, copper containing pigment	No amino acid				Laurionite (lead halide mineral), anglesite (lead sulfate mineral), cotunite (lead chloride)	
J(c)		Copper-containing green						Egg yolk
K		Cinnabar/vermilion, minium						
EGB			Amino acid (protein/ polysaccharide)					
M		Cinnabar/vermilion, minium	Amino acid (egg yolk)					
B(d)	Lead white		Drying oil (walnut or poppy seed)					Animal glue, polysaccharide (honey?)
East III		Lead white	Amino acid (protein/ polysaccharide)				Gypsum	
F(c)		Lapis lazuli, cinnabar/vermilion, lead white	Drying oil (walnut or poppy seed)				Copper alteration product	Amino acids (protein/ polysaccharide)
L			Drying oil (walnut or poppy seed)					Amino acids (protein/ polysaccharide)
N(a)		Ultramarine blue, sodalite, copper-containing greens (chrysocolla, atacamite), minium, cinnabar/vermilion, red ochre, yellow ochre, lead white, gypsum, carbon black, tin leaf	Drying oil (walnut or poppy seed)	Protein	Resin (yellow)	Drying oil	Moolooite (copper oxalate), calcium oxlate, lead soaps, plumbonacrite ($Pb_5O(OH)_2(CO_3)_3$)	Amino acids (protein/ polysaccharide), egg white
S(a)			Drying oil (walnut or poppy seed)					
Foladi 2		Copper-containing pigment, cinnabar/vermilion, minium, gypsum (in render)	Drying oil (walnut or poppy seed)				Copper alteration product	
Foladi 3			Drying oil (walnut or poppy seed)					Amino acids (protein/ polysaccharide)
Foladi 4		Copper-containing greens (atacamite, paratacamite), minium, ochre (goethite), orpiment, lead white	Drying oil (walnut or poppy seed)	Protein/polysaccharide/resin			Moolooite, lead palmitate, copper palmitate	Amino acids (protein/ polysaccharide)
Foladi 6		Cinnabar/vermilion„ minium, ferric red pigment (red ochre), lead white	Drying oil (walnut or poppy seed)				Lead alteration product	Amino acid (egg yolk)
Kakrak 43		Lead white	Drying oil (walnut or poppy seed)					Amino acid (animal glue)
Kakrak 44			Drying oil (walnut or poppy seed)					Amino acid (egg yolk)
Qol-e-Jalal		Copper-containing green, minium, ferric red pigment (red ochre as bole), gold leaf	Drying oil (walnut or poppy seed)	Protein		Drying oil	Calcium oxalate, lead soaps	Amino acids (protein/ polysaccharide)

2.2 Structure of the wall paintings

2.2.1 Overview of the structure and paintings

2.2.1.1 Basic structure of the wall paintings in Bamiyan

The structures of wall paintings across Bamiyan, Foladi and Qol-e Jalal are relatively similar: most use the inner walls of caves as a support, since these wall surfaces are made of relatively soft substances such as sandstone or conglomerate. First, a product made from cow dung is applied to the wall surface followed by several smooth coats of a conglomerate of soil and admixtures such as binding media, animal and plant fibres, and sand to form a rendering layer. Most renders consist of two layers – the lower and upper layer – the composition of which varies from one cave to another (see section 2.2.3). The surface of the smooth, polished clay render is then covered with an organic, glue-like material that serves as a sizing layer to prevent the absorption of water and improve the mobility of brushes and bristles with paint. Field observations and microscopic examination of samples confirm that wall paintings were produced using a white ground layer followed by several coloured layers, for example black, white and blue paints. Table 2.1 summarises the distinguishing characteristics of each wall painting with regard to its ground and sizing layers. The majority of ground layers in wall paintings are made of white materials, although the wall painting in Cave J(d) has a light pink substrate containing red particles in a white pigment, similar to wall paintings at Mīrān.

Sizing layers generally conform to one of two types: either a thick yellow transparent resinous layer with strong white UV fluorescence, or a thin transparent layer with blue, white or yellow UV fluorescence, each of which may be composed of different organic substances. Almost all the wall paintings have multiple layers of paint or painting layers. For example, a dark blue colour was obtained by applying a blue layer of lapis lazuli over a black layer, bright garments were made from two different layers of red, and a clear deep green colour was achieved by layering green, white and orange. Paintings in Caves N(a) and F(c) included a transparent yellow or blue layer on the surface of the painting layer; this layer was probably glazing applied to add depth to the colours.

The head and body nimbus and the petals of the lotus flowers are all decorated with gradient colouring of darker and lighter hues, or so-called '*ungen*' colours. This is distinct from Greek and Roman colours as well as the monochromatic tones of Iranian and Sogdian wall paintings.

Figure 2.2 Unfinished wall painting at the entrance to the front chamber of Cave C(a). Photo: Y. Taniguchi, courtesy of NRICPT, 2005.

Figure 2.3 Unfinished wall painting on the north wall of the front chamber of Cave C(a). Photo: Y. Maekawa, courtesy of NRICPT, 2007.

2.2.1.2 Layered painting structures in unfinished wall paintings

Along the western side of the Eastern Giant Buddha, a wall painting on the northern wall of Cave C(a)'s front chamber is painted with red and white (Fig. 2.2). On the wall's surface is a rough white ground

Table 2.2 Possible binding media based on GC-MS and ELISA analyses.

			Results				*Interpretation*					*Ground layer*	
Sample number	*Cave*	*Details*	*Fatty acids*	*Amino acids*	*Polysaccharides*	*ELISA (GCI)*	*Drying oils*	*Resins*	*Plant gums*	*Proteins*		*Lead white*	*Gypsum*
BMM 128	J(b)	blue/black, white ground	×	●					○	○			●
BMM 112	J(c)	green on white ground	×			● Egg yolk			○	◎			●
BMM 113	J(c)	green on white ground	×		● Tragacanth				○	○			●
BMM 120	J(d)	blue, pink ground	×	●					○	○			
BMM 169	J(f)	red, render	×	● Casein					○	○			
※BMM 053	M	darkened surface, later soot?	×	●					○	○			●
BMM 054	M	red, white ground	●	● Egg yolk	● Plant gum		○	○	○	◎			●
BMM 091	C(a)	unusual light blue	×	×					○				●
BMM 209	C(b)	green, white ground	×						○	○			
BMM 099	D	red, white ground	×	●					○	○			
※BMM 108	E(c)	ground and soot deposit	×						○	○			
※BMM 101	E(e)	darkened surface, later soot?	×						○	○			●
BMM 045	G	yellow, white ground	×	● Casein					○	○			●
BMM 212	H(a)	yellow, white ground	×	●					○	○			●
BMM 211-2	H(b)	red and white ground	×	●					○	○			●
BMM 009	I	red, white ground	×	×					○				●
BMM 073	I	blue, black, white ground	×						○	○			●
BMM 080	K$_3$	flesh colour, white ground	×						○	○			●
BMM 134	K$_3$	brown deposit, red, white ground	×	●					○	○			●
※BMM 135	K$_3$	brown deposit on chaff	●	●				○	○	○			●
BMM 157-2	East Displaced	blue and render	×						○	○			
BMM 063	B(d)	red and ground	●	● Glue	● Tragacanth		◎	○	◎	◎		●	
BMM 067	E III	yellow, white ground	×	●					○	○		●	
※BMM 083-1	F(c)	translucent yellow glaze and render	×						○	○		●	
BMM 083-2	F(c)	black darkened layer, yellow glaze, white ground	●	●			◎	○	○	○		●	
BMM 203	L	green, white ground, yellow size	●	●			◎	○	○	○		●	
※BMM 204-2	L	black deposit	×						○	○		●	
BMM 035	N(a)	resin, green, black oils, white ground, yellow size	●	●			◎	◎	○	◎	µFTIR	●	
BMM 040	N(a)	red/oily red, white ground	●			● Egg white	◎	○	○	◎		●	
BMM 183	N(a)	deep red glaze on orange	●	×	● Tragacanth		○	○	○			●	
※BMM 184-1	N(a)	yellow resin on tin leaf					○	○	○	○		●	
BMM 184-2	N(a)	tin leaf and white ground					○	○	○	○		●	
BMM 163	S(a)	grey/green, white ground, size	●		● Fruit gum		◎	○	○	○		●	
BMM 189	EGB	blue in white, white ground	×						○	○			●
BMM 190	EGB	red, white ground, render	×						○	○			●
BMM 191	EGB	blue in white, white ground	×	●					◎	○			●
BMM 199	WGB	red	×						○	○			
BMM 201	WGB	deep red	×						○	○			
FDM 059	Foladi 2	red and white ground	●	×			◎	○	○			●	
※FDM 043-1	Foladi 3	yellow translucent size	●	●			○	○	○	○		●	
FDM 043-2	Foladi 3	white, white ground, size	●	●			◎	○	○	○		●	
FDM 014	Foladi 4	salmon pink, yellow, white ground, size	●	●			◎	○	○	○		●	
FDM 055-1	Foladi 4	green, white ground, size, sandy render	●				◎	◎	◎	◎	µFTIR	●	
※FDM 055-2	Foladi 4	reddish sandy upper render	×						○	○		●	
FDM 023	Foladi 4B	white, orange, white ground, size	●				◎	○	○	○			
FDM 003	Foladi 5	red, white ground, size	×	● Egg yolk					○	◎			
FDM 026	Foladi 6	black (alteration of green), white ground, white size	●				◎	○	○	○		●	
FDM 060	Foladi 6	brown green, red, white ground, size	●	● Glue			◎	○	○	○		●	
KAK 03	Kakrak 43	red, black organic, white ground	●	● Glue			◎	○	○	◎		●	
KAK 10	Kakrak 44	white and ground	●	● Egg yolk			◎	○	○	◎		●	
※QJM 06	Qol-e Jalal	gold leaf and mordant	●				◎	○	○	○		●	
QJM 07	Qol-e Jalal	brownish colour, whole paint layer	●	●			◎	○	○	○		●	

●Detected ×ND ◎Confirmed ○Possible µFTIR : confirmed by layer to layer

layer with only underdrawings and incomplete work, revealing that the wall painting was unfinished: this is most unusual and, as such, serves as a valuable example of an artist's sequence and process.

At the entrance to Cave C(a)'s front chamber (Fig. 2.3) is an outline of a seated Buddha drawn on a white ground in reddish-brown lines applied roughly with visible brushstrokes. The lines of the reddish-brown underdrawing are not very thick and seem to have been drawn with a hard rather than soft brush. Details of the face and limbs are not clear, as they were later destroyed, but the eyes, nose and fingers appear to have been sketched in at a preliminary stage. The Buddha's red robe is composed of the same reddish-brown colour as the underdrawing, and lacks any folds or other detail. The same reddish-brown colour also fills the standing Buddha's head and body nimbus and the stupa. The artist may have intended to cover this layer with another shade of red, such as cinnabar, vermilion or minium. To the naked eye, the background may look bluish-grey but on closer observation it is not made of blue but of a thin layer of black particles. As discussed in section 2.2.7, this may have been an area that the artist originally intended to colour in by adding a layer of blue lapis lazuli on top of the black.

The painting's current unfinished state suggests that when the composition was initially drawn on the ground, the artist was following a plan for applying colours. He applied this first layer with the expectation that others would follow later. It is not known what prevented completion of the work in Cave C(a) but one reason may have been a donor's financial difficulties. However, it is likely that the wall painting was abandoned before the final layers of paint were ever applied.

2.2.2 The supports

Supports undergirding the Bamiyan, Foladi and Qol-e Jalal wall paintings are similar. In most cases, the grottoes are found in relatively soft rock, such as sandstone or conglomerate, and the rock surface inside the cave is used as a support. The cliffs are composed of alternating layers of sandstone, silt and conglomerate, so the caves are concentrated in conglomerate and sandstone layers and developed horizontally. Artisans appear to have avoided the siltstone when carving grottoes, probably due to its brittleness.

The caves were carved and sculpted with chiselling tools. The chisel marks are relatively coarse and appear to have been applied deliberately to create unevenness: when caves are built into conglomerate rock, a coarse surface seems distinctly conducive to the rendering layer's adhesion. Larger caves are often built in conglomerate layers.

Preparations for the application of the kneaded clay renders vary depending on the nature of the rock. When grottoes comprise coarse conglomerate rock, a substance containing a large amount of very fine plant fibre, probably cow dung, is applied to the rock surface in order to increase the adhesive strength between the rock surface and the kneaded soil, thereby preventing them from separating (Bamiyan Caves C, E(c), I, K_3, Kakrak Cave 44, etc.) (Fig. 2.4). As cattle have four stomachs and extensive processes for masticating and digesting plant fodder, their excrement consists mainly of fine, short plant fibres and mucus. This is used as adhesive material for connecting interfaces and is widely visible in Central and South Asian earthen walls beyond Bamiyan. Once the fine fibrous material enhances the surface of the conglomerate, owing to the irregularity produced by the chisel marks, the thicker, heavier kneaded clay of the rendering will adhere to the wall.

In sandstone grottoes, on the other hand, no traces of any cow dung-like adhesive have been observed. Sandstone, due to its relatively smooth and easily detachable surface, seems to be suitable for the use of light, thin rendering layers. In order to improve sandstone's properties, holes were drilled into its surface and wooden rods inserted as dowels to increase adhesion between the earthen renders and rock wall (Foladi Cave 6) (Fig. 2.5). In addition, in some grottoes wall paintings have noticeably detached and fallen because rock surfaces were not treated with cow dung or wooden dowels (e.g. East Displaced Cave, Cave M).

Figure 2.4 A layer comprised of fine fibres, possibly from cow dung, between the rock surface and the earthen render (Cave I, niche). Photo: Y. Taniguchi, courtesy of NRICPT, 2006.

Figure 2.5 Holes in sandstone walls with small wooden pegs (Foladi Cave 6). Photo: Y. Taniguchi, courtesy of NRICPT, 2006.

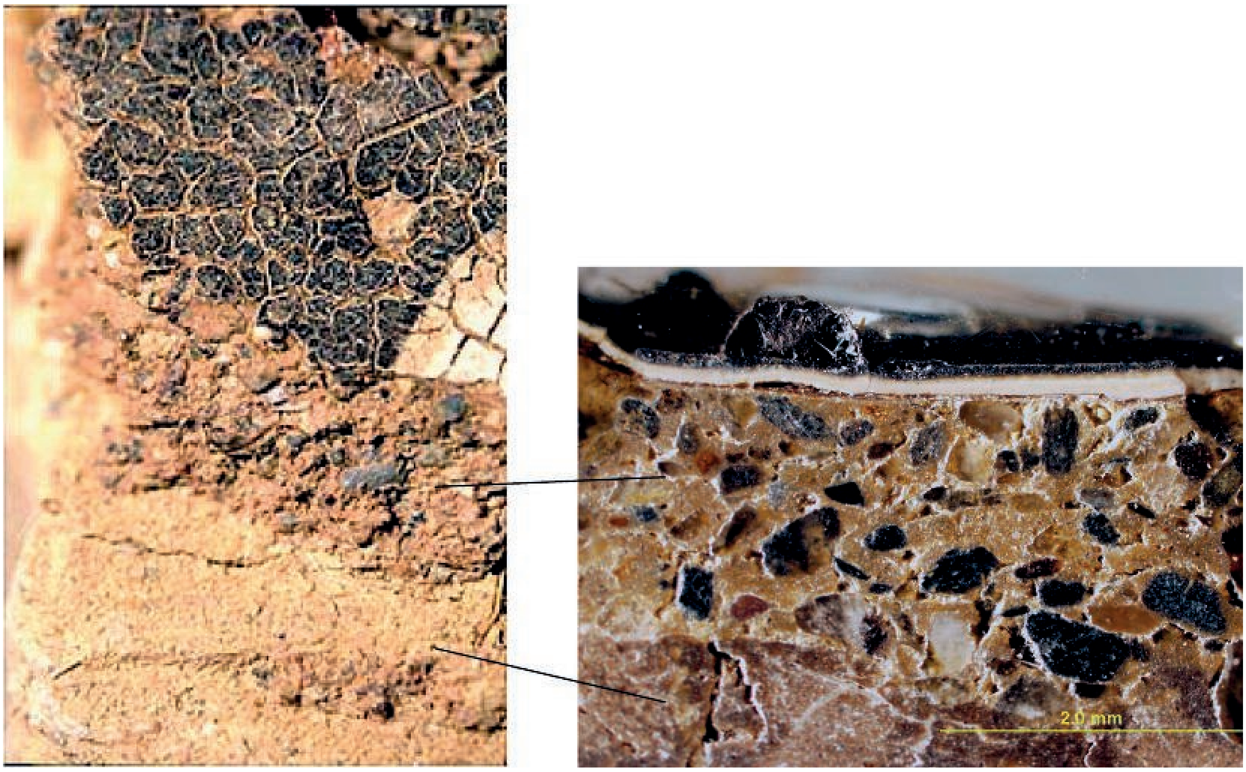


Figure 2.6 Cross-section of a wall painting fragment from Foladi Cave 4 (left: 'Displaced Cultural Property') and a cross-section of a wall painting from Cave F(c) (BMM082) (right). The rendering layers consist of a sand-rich layer (upper) and a straw fibre-rich layer (lower).

2.2.3 The rendering layer

After application of the supports to the rock wall, its surface is smoothed by adding several coats of kneaded clay with a mixture of soil, animal and plant fibres and sand on top to form the rendering layer. As already mentioned, most renderings consist of two layers (Fig. 2.6): the lower layer contains a large proportion of straw fibres 3–5 cm in length while the upper layer tends to contain fine-grained soil with a high proportion of sand. The amount of straw fibre used will vary but the thicker renders on conglomerate tend to contain more straw fibre of substantial length, while the thinner renders on sandstone contain less straw fibre of shorter length.

The rendering may also contain other types of animal fibres (e.g. Cave D1, East Displaced Cave) (Fig. 2.7) and plant fibres such as small pieces of dyed textile (Kakrak Cave 44) (Fig. 2.8), woven yarn (Cave J(g)) (Fig. 2.9) and clumps of cotton (Caves F(c), S(b)). It is believed that dyed and woven materials were reused and added to prevent the render from cracking. In a study by the Technical University of Munich, kneaded clay on the surface of the Western Giant Buddha, unlike that on wall paintings, was found to contain wheat, barley fibres, berries and husks (Pfeffer and Blänsdorf 2009a). They also included goat, sheep and

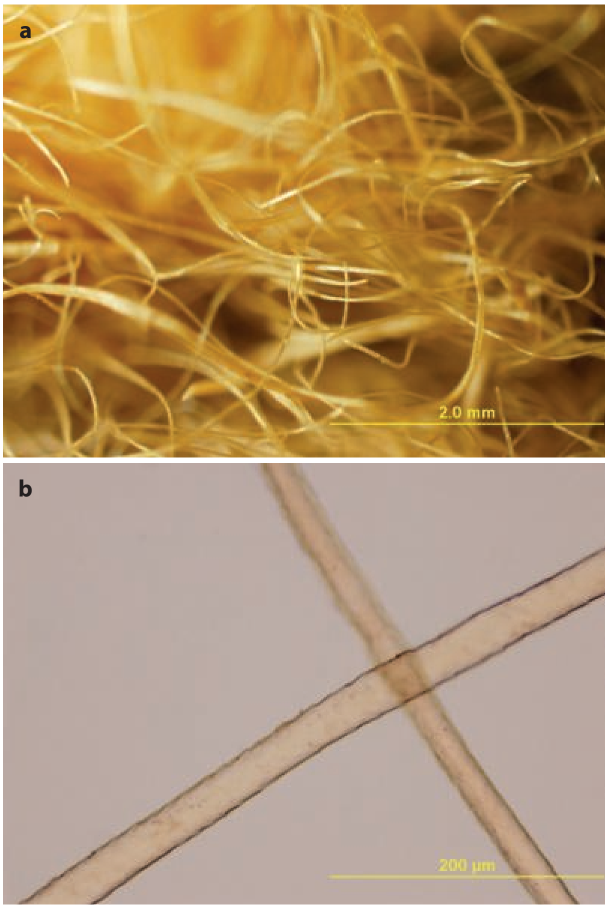


Figure 2.7 Stereomicrograph (a) and PLM photomicrograph (b) of fibres in a rendering layer at Cave D1. The fibres appear to be from an animal such as a camel (BMM161).

Figure 2.8 Fragments of a textile, containing fibres of two different thicknesses, in a rendering layer at Kakrak Cave 44 (KAK012).

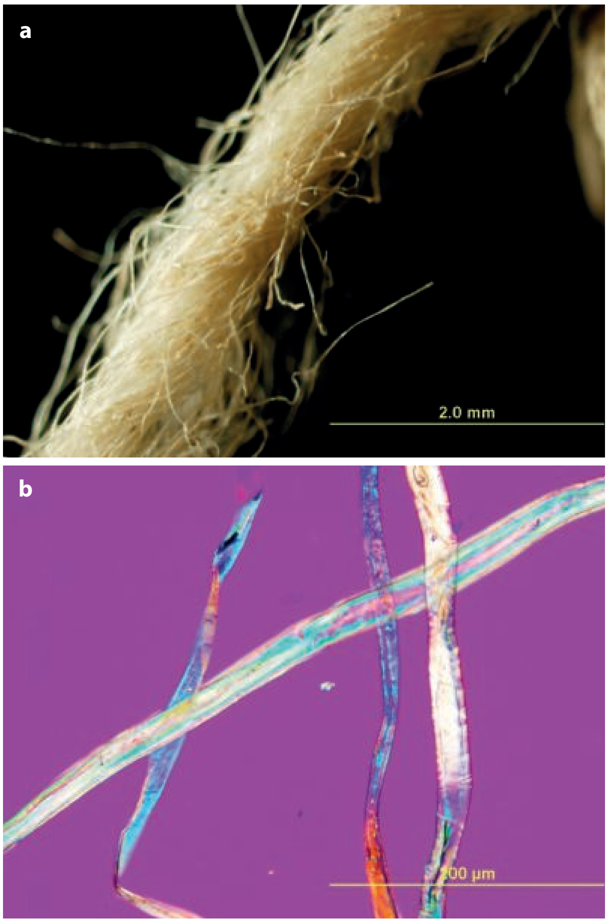


Figure 2.9 Stereomicrograph (a) and PLM photomicrograph (b) of cotton fibres in a rendering at Cave J(g) (BMM159).

other animal hairs, as well as ropes and textile fragments identified as *Dom-i-shutur* ('herd of camels') (*Astragalus cuneifolius* Bunge) from the mountainous area around Lake Band-e Amir (Pfeffer and Blänsdorf 2009b). Goat hairs are thin and cut short in the upper layers of the render and remain thick and long in the lower layers (Grunwald 2009).

Some rendering layers, as in Caves F(c) and N(a) and the Foladi wall paintings, consist of a thick layer of light brown soil with straw fibre added, and a reddish-brown, slightly coarse but homogeneous upper layer composed of many sand particles of relatively large size (about 3 mm thick), but the particles are bonded together and the earth is compacted.

In India, the traditional construction of earthen walls is known to involve kneading clay with a vegetable mucilage or resin (Sengupta 1973). Methods for building walls in the Gupta period, as described in Chapter 40 of the *Citrasutra* (Appendix I), involve the use of substances such as fragrant gum resin,[2] beeswax, honey, kundara grass (liquorice), molasses, safflower soaked in oil, powder of lime, bel fruit (*Feronia elephantum*) pulp,[3] lampblack and sand (Sivaramamurti 1978: 180–182).

Rendering in the Ajanta caves tends to be thin and dense, as if some organic substance has been added as an adhesive. The renders at Bamiyan are slightly thicker and less dense. However, rendering layers in Caves F(c) and N(a), as well as in the Foladi wall paintings, are more robust than the other renders, therefore it is natural to assume that they contained some organic material. In order to verify whether rendering layers contained some kind of adhesive, we analysed polysaccharides using GC-MS (Foladi Cave 4: reddish-brown rendering, upper layer FDM055-2). No fatty acids or terpenoids, indicative of the use of oil, resin or beeswax, were detected, but it is possible that polysaccharides such as plant mucilage were used. However, we could not separate and identify any organic matter in the analysis. Some rendering layers were completed without any finishing adjustment, as in the case of wall paintings in Caves C, M and J from the first and second phases of the cave chronology (Fig. 2.10), while others were completed with a smoothed surface, for example, the wall paintings in Caves I and K_3 and the Foladi caves. However, the Buddha niche in Cave I has trowel marks and fingerprints that may be those of an artisan (Fig. 2.11), suggesting that the surface was polished when the walls were only partly dry. This particular method of smoothing and polishing the surface of the rendering is considered a relatively new technique within the Bamiyan chronology.

2.2.4 The sizing layer

The surface of the rendering layer is coated with an organic glue-like substance. Painting with water-soluble paints on porous and water-absorbent earthen walls requires the surface of the wall to be sized first in order to prevent moisture in brushes and bristles from being absorbed too quickly, which

Figure 2.10 Fragment of a wall painting from Cave J(d). The surface of the priming layer is unadjusted and straw soot is visible on the surface. Photo: H. Otaki, courtesy of NRICPT, 2005.

Figure 2.11 West wall of the niche in Cave I: trowel marks and fingerprints on the rendering layer. A thin layer of white ground has been applied with a coarse brush. Photo: Y. Taniguchi, courtesy of NRICPT, 2006.

would prevent smooth lines or result in blurring. This kind of sizing can also be seen in other paintings. In modern canvas painting, an insulating layer comprised of natural resins, shellac, glue or synthetic resins is used (Wehlte 1975: 71): these organic layers play an important role in preventing paint delamination and cracking. The sophistication that sizing adds to the layer structure in Bamiyan creates a healthy foundation for the paint.

Chapter 40 of the *Citrasutra* (Appendix I) describes the process of smoothing walls by applying several coats of earth mixed with Śāla resin[4] and sesame oil when the walls are fresh and dry. Milk is then repeatedly applied and wiped off. In this process, resin, oil and casein in the milk act as sizing. Cennino Cennini, in his 14th-century *Il libro dell'arte*, also refers to the use of a sealant made from whole eggs and water that 'goes evenly over the whole work you paint in *secco* and also embellish with gold'.[5] The wall in this case is essentially a lime mortar support. Cennini instructs:

> take a well-diluted size. Making a better *tempera* starts with beating a whole egg in a porringer with

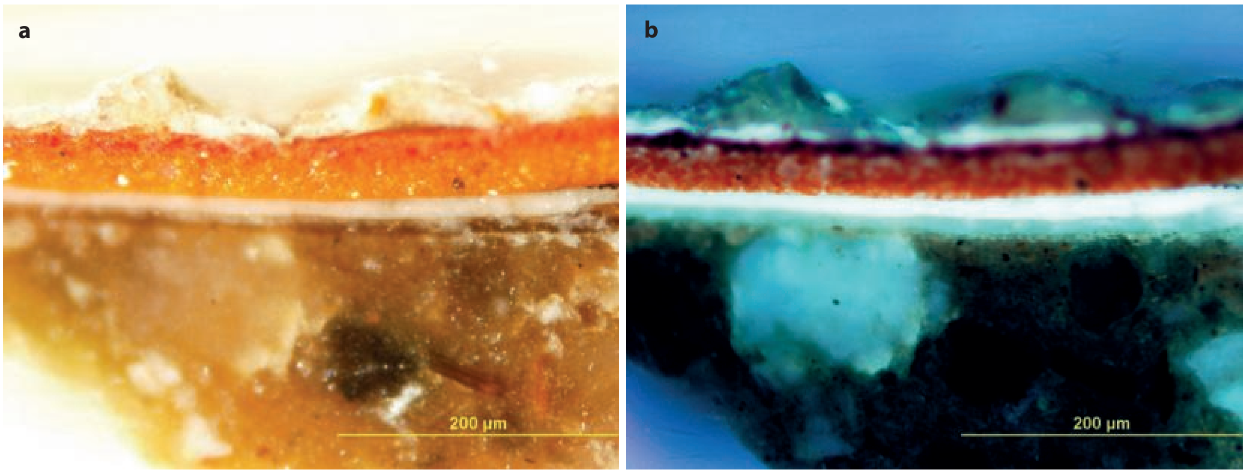


Figure 2.12 Cross-section of the red area on the north wall of Cave N(a) (BMM036). Photomicrographs under normal diffused light (a) and UV light (b). The white fluorescent layer is a white ground layer composed of lead white and drying oil.

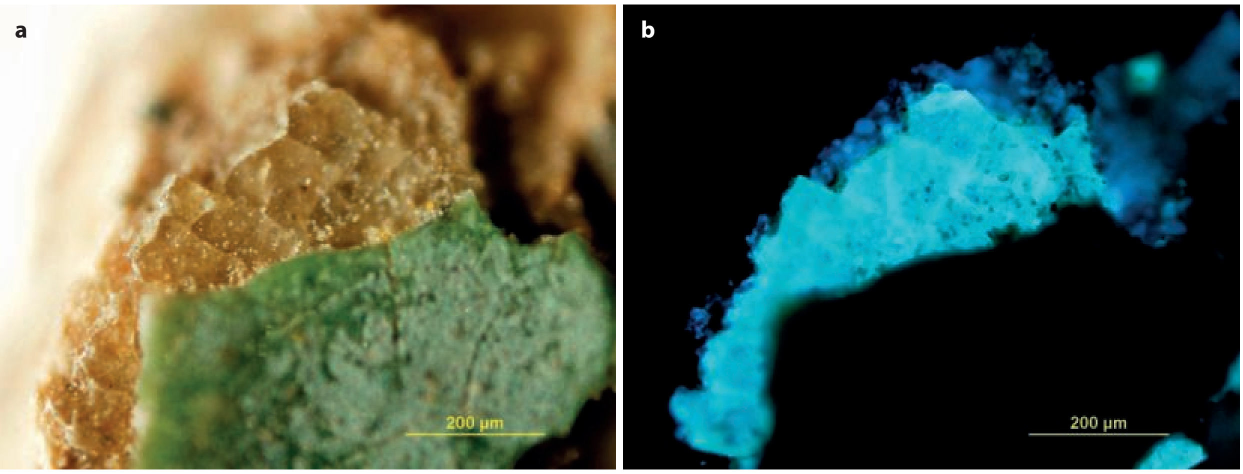


Figure 2.13 Stereomicrograph of the green part of Foladi Cave 3 (FDM038) under normal light (a) and UV light (b). The yellow transparent layer is sizing which fluoresces white-yellow. The layer below this fluoresces pale blue.

> fig-tree latex, and pouring a goblet of water over the egg. Then, either with a sponge or with the soft, rather blunt brush, apply one coat of this mixture all over the ground which you have to prepare; and let it dry for at least one day.[6]

In this preparation, the proteins in the eggs and the hide glue work together with polysaccharides in the figs as a sizing. Applying organic materials to wall surfaces in this way was a critical step in painting, whether the wall was an earthen or lime mortar. Now, both sizing layers in Bamiyan's wall paintings are assumed to derive from different organic materials: one appears as a thick, transparent yellow resinous layer that emits white UV fluorescence, while the other, a thin transparent layer, emits blue-white, white or yellow UV fluorescence (Taniguchi *et al.* 2006).

As shown in Appendix III (Fig. A.III.18), the µFTIR analysis of sample BMM035 from the green part of the east wall of Cave N(a) detected localised absorption specific to amide I (1650 cm^{-1}) in part of the sizing layer. This clearly indicates that the thick yellow transparent layer contains some proteins assumed to derive from animal sources such as collagen from cows, rabbit bones and skin, fish[7] or albumin from egg whites. The species of animal was later identified by LC-MS as equine (see section 2.2.8), therefore, there is a possibility that animal glue from horse collagen was used here.

Egg white albumin was identified by enzyme-linked immunosorbent assay (ELISA) from the red part (BMM040) of the same sample in Cave N(a) (Appendix III, Table A.III.35a). However, the sizing layer of BMM035 was not found to have absorbance of fatty acid (Appendix III, Fig. A.III.18), suggesting that the protein used in this sizing layer was a purer protein such as glue or egg white. Analysis of amino acids using GC-MS of sample BMM035 (Appendix III, Table A.III.33) showed that alanine, leucine,

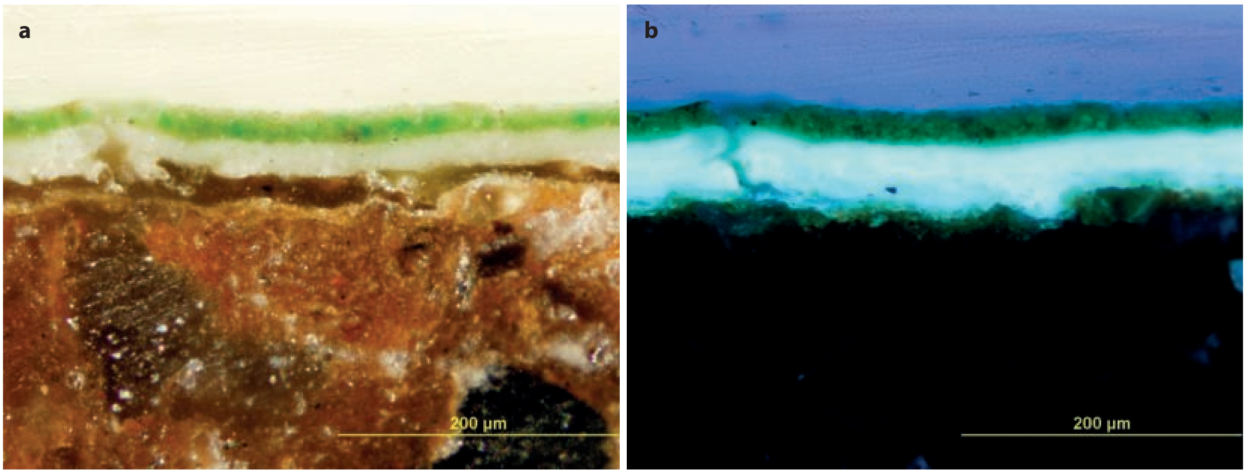


Figure 2.14 Cross-section of Foladi Cave 3 green area (FDM038). Photomicrograph under normal light (a) and UV light (b). The yellow transparent layer below the white ground is the sizing layer which fluoresces pale blue.

isoleucine and glycine were present, but the correlation coefficient of the egg was insufficient to identify the protein type (Gautier and Colombini 2007). This kind of structure is commonly observed in Bamiyan (i.e. BMM036, Cave N(a)) (Fig. 2.12).

The sample from the green part of Foladi Cave 4 (FDM055) has a transparent layer and another transparent yellow layer applied on top of the rendering layer. As shown in Appendix III (Fig. A.III.96, spectrum 5), the µFTIR analysis confirms that this transparent yellow layer (5) has absorption attributed to polysaccharides (around 1050 cm^{-1}), to amide I (1650 cm^{-1}) and to amide II (1550 cm^{-1}). This layer is believed to consist of proteins (e.g. animal glues and egg white) and polysaccharides (e.g. peach gums and gum arabic). The lower transparent sizing layer (6) shows FTIR absorption similar to that of resin acids therefore it is likely that this layer included some kind of natural resin. There are a number of candidates, such as mastic and dammar, but it is difficult to identify the exact type of resin based on information from FTIR spectroscopy. The abovementioned sizing structure is also seen in the wall paintings of Foladi Cave 3 (FDM038) (Figs 2.13 and 2.14). It is clear, however, that a variety of organic materials, including proteins, resins and plant gums, was applied to earthen walls to act as sizing layers.

2.2.5 The ground layer

2.2.5.1 Different types of ground layers in Bamiyan's wall paintings

Before painting on clay renders, a white or coloured ground layer is first applied. The ground layer acts as a reflector to redirect incoming light, which is essential for painting subsequent layers. Prior studies have already identified numerous kinds of ground layers in wall paintings around Central Asia (Chapter 1, Tables 1.3 and 1.4). In many of the analyses mentioned here, the age of the wall paintings in question is not known, so the chronology of the substrate over time is unclear. Five different white substrates have been reported: gypsum, lime, lead white, white clay (kaolinite) and talc; gypsum, in particular, is commonly selected and used extensively. The use of talc is limited to the Mogao grottoes in Dunhuang (Gansu Province) and white clay to the area around the Ajanta caves (India). White clay and talc have not been identified in Central Asia's wall paintings; the use of lime is found mainly in India. Interestingly, in some cases more than one white material, such as gypsum and lead white, might be used at a single site.

Gypsum, lead white and, in some cases, lime, are produced through artificial processes. For example, two calcium sulfate minerals may be used to make gypsum: one in the dihydrate form ($CaSO_4$. $2H_2O$) and the other in anhydrite form ($CaSO_4$). Both occur naturally and can be calcined and used as either semi-hydrated gypsum (bassanite: burnt gypsum) or fully dehydrated (anhydrite: hard gypsum). Both dihydrate and anhydrate gypsum have been used as ground materials for wall paintings, with the latter most commonly identified in the wall paintings of Kucha, Turfan and Kharashahr (see Chapter, Table 1.3).

Lead white, another synthetic inorganic pigment produced when lead is corroded by vinegar vapours, is also found in Central Asia, India and Gansu. The term 'lead white' usually refers to a mixture of lead carbonate (cerussite, $PbCO_3$) and hydroxycarbonate (hydrocerussite, $Pb_3(CO_3)_2(OH)_2$) (Gettens *et al.* 1993: 67). However, as detailed below, lead whites

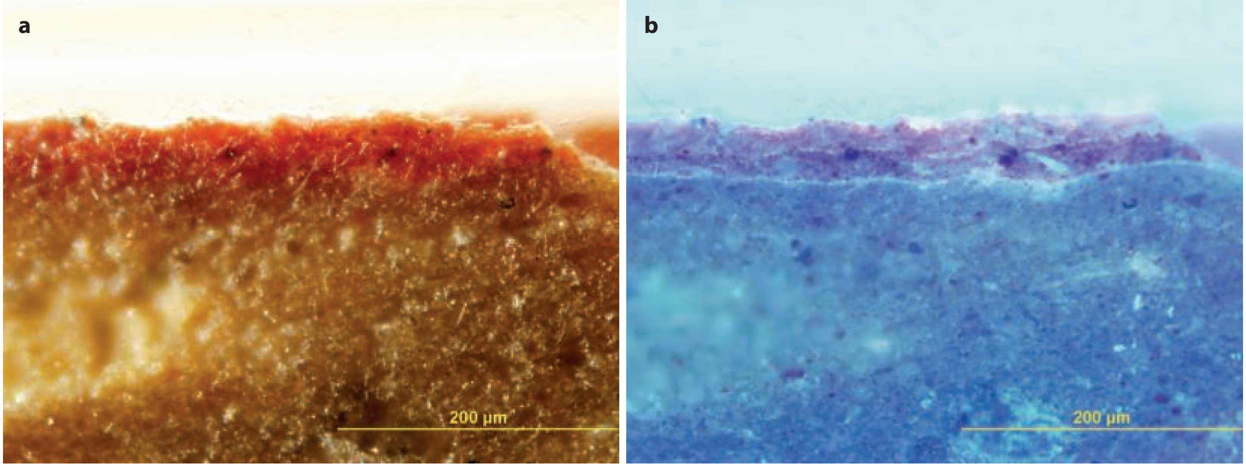


Figure 2.15 Photomicrograph of a cross-section of a red portion of a wall painting from Cave J(b) (BMM121). (a) PLM photomicrograph under a normal diffused light source. Two layers of different shades of red have been applied on top of a rendering layer. (b) In the UV fluorescence image, two layers of red particles can be seen on top of a fluorescing sizing layer on earthen render. The lower layer contains large red particles.

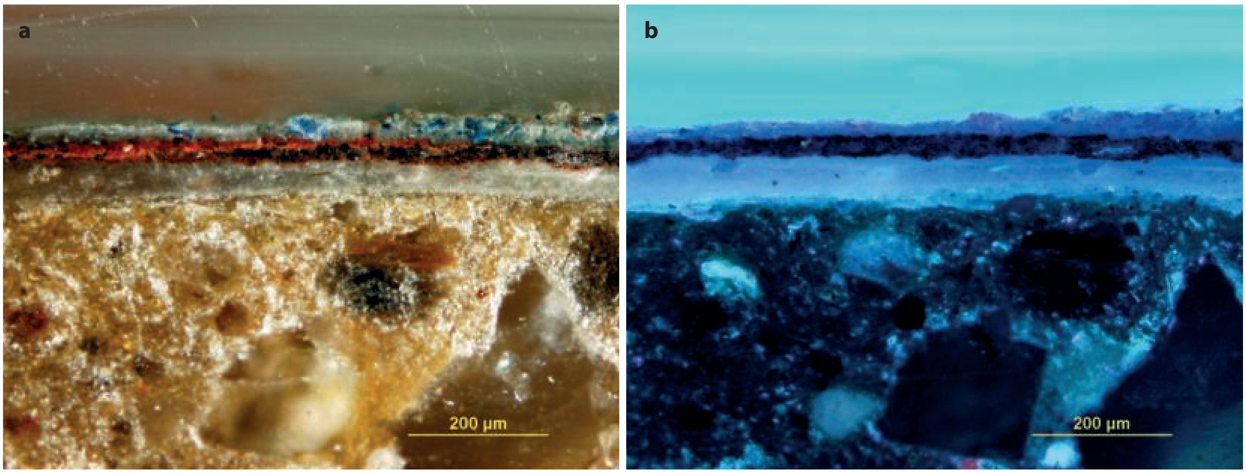


Figure 2.16 (a) PLM photomicrograph of a cross-section of a wall painting from Cave J(b) (BMM126) with blue overlaid on red (left). A clear white ground layer can be seen. The black layer is partly overlaid with red, and a layer containing blue particles is applied on top. (b) In the UV fluorescence image, the white base round layer and the white part of the top layer fluoresce bluish-white.

with a very unusual composition have been detected in some of Bamiyan's wall paintings. In the present analysis, two main types of substrate were identified from Bamiyan's wall paintings, as summarised in Table 2.1. Most of Bamiyan's wall paintings have a white ground consisting of gypsum or lead white, but some do not have a ground layer:

- Partially absent ground layer: Caves J(b), C(a)
- Gypsum ground: Caves C(a), E(e), G, H(a), H(b), I, J(b), J(c), K, M
- Lead white ground: Caves B(d), East III, F(c), L, N(a), S(a); Foladi Caves 2, 3, 4, 6; Kakrak Caves 43, 44; Qol-e Jalal

Wall paintings have been organised into the following periods, classified according to radiocarbon dating of materials in their ground layers. While gypsum grounds were used continually throughout Bamiyan's history, it was not until the mid-7th century that lead-white grounds were used in some caves.

2.2.5.2 Paintings with a partially absent ground layer

Samples in Caves J(b) and C(a) had a white ground consisting essentially of gypsum, however, in some places a coloured layer was applied directly on top of the render without any white ground layer observed. Above its rendering, one sample from Cave J(b) (BMM121) had a thin sizing layer that was difficult to decipher in natural light but visible under UV light as well as two layers of red paint (Fig. 2.15). The upper layer of paint is a brighter red, while the lower layer is a relatively transparent red and contains larger red particles. However, in the same cave, a clear white ground (gypsum) can be observed in an area where blue is overlaid on red (BMM126). The

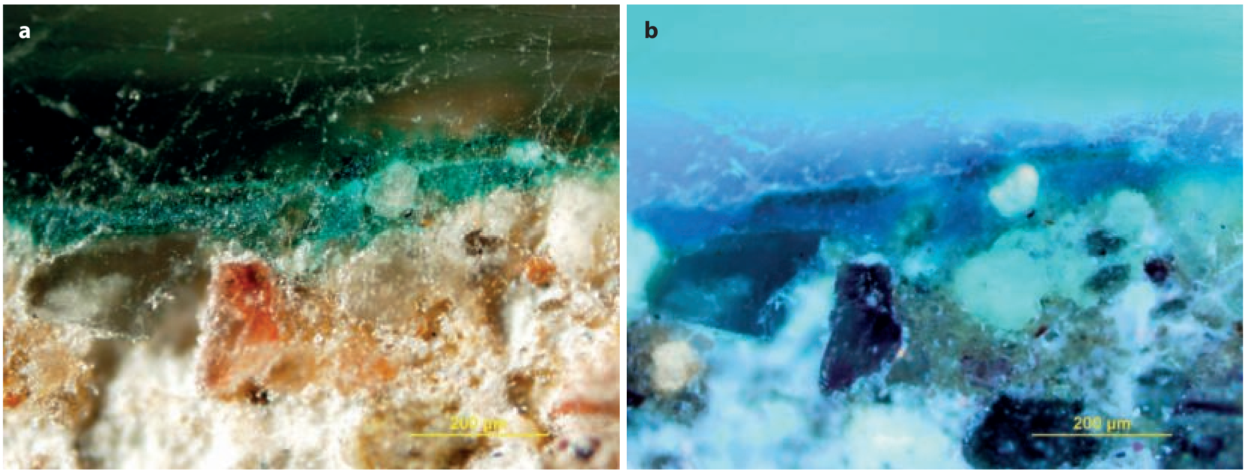


Figure 2.17 (a) PLM photomicrograph of a cross-section of a blue portion of a wall painting from Cave C(a) (BMM091). The blue-green and green layers are applied on top of an earthen render. (b) UV fluorescence image.

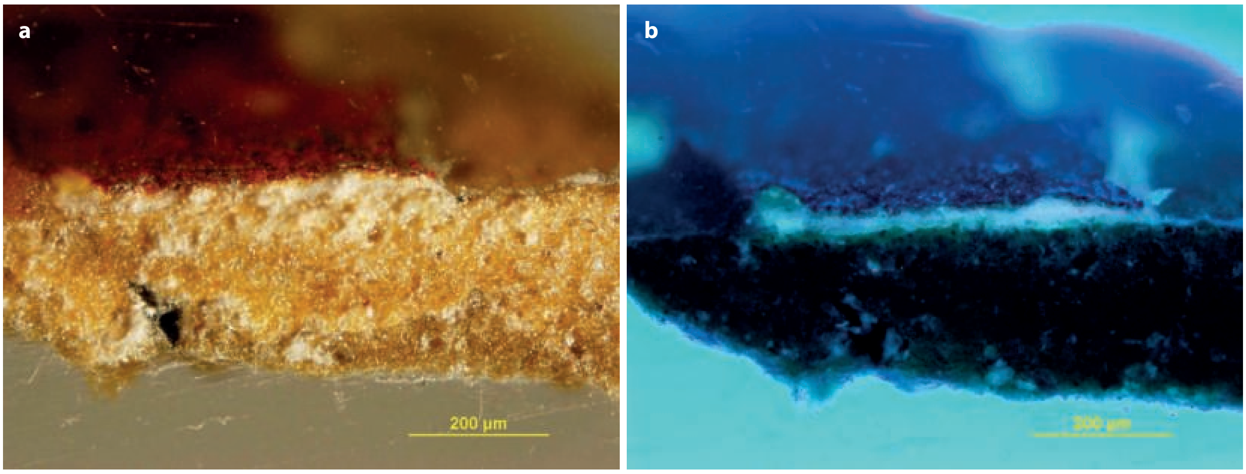


Figure 2.18 (a) PLM photomicrograph of a cross-section of a red portion of a wall painting from Cave C(a) (BMM095). (b) In the UV fluorescence image a thin red coloured layer is visible on a white base.

red colour is partly overlaid on a black layer, and a layer containing blue particles is applied to the top layer (Fig. 2.16). It is likely that the black layer was applied beforehand in order to obtain an optical effect when applying the blue (see section 2.2.7). In both cases, the rendering layer is relatively smooth and paint is applied on top of sizing.

A similar case was observed in a sample from Cave C(a) (BMM091) (Fig. 2.17) in which a blue-green layer on top of a render is covered by a green layer. There is no UV fluorescence indicating the presence of organic materials. However, in another sample (BMM095) (Fig. 2.18), a thin white layer is covered by a thin dark red layer. In both cases, the surface of the render is not smooth and there is no uniformity in the thickness of either the painting layer or the white ground. There are numerous similarities between the surface treatment of the render, the sizing and other layers, namely, a white ground is present in some areas but absent in others, suggesting that the ground was not applied in certain areas accidentally but deliberately in others. For example, in Cave 69 of the Kizil grottoes, only the upper parts of the wall paintings seem to have been depicted directly on the sandstone substrate (Fig. 2.19) (Vignato and Hiyama 2022: 365). An analysis of the sizing has not been conducted, but the surface looks to be well sized.

2.2.5.3 Gypsum grounds

As mentioned earlier, 'gypsum' may refer to two calcium sulfate compounds: dihydrate (gypsum) and anhydrite. In some cases, gypsum was used in its natural form, however, there are also accounts suggesting that it may have been synthesised by adding water to semi-hydrated gypsum (bassanite) ($CaSO_4 \cdot 1/2H_2O$) obtained by calcination, which would cause a reaction leading the materials to hydrate

Figure 2.19 Wall paintings on the upper part of the vaulted ceiling of Cave 69, Kizil grottoes. Earthen render on the sandstone substrate is not visible. Photo: Y. Taniguchi, courtesy of Tokyo University of the Arts/Kizil Research Institute, 2014.

and solidify as gypsum (Thompson 1960). Semi-hydrated gypsum is the same as the plaster used for moulding. A typical example of gypsum is gesso, a white powder mixed with an adhesive and used as a ground for *gesso Bologna* paintings in medieval Europe. This white powder, which occurs naturally in Bologna, was not entirely dihydrate, but partially anhydrite (Frederspiel 1995). Anhydrite is also a naturally occurring mineral, but it can be produced artificially by calcining gypsum at 300–650 °C to remove all the hydrates. The resulting white mass is then powdered and mixed with an adhesive to make white paint.

Gypsum has been identified in many of the lower layers of Central Asian wall paintings at Shahristan, Penjikent and Ajina-tepe, suggesting that it was probably the main material used for white ground layers in Central Asia. Plaster containing a large amount of anhydrite was commonly used in wall paintings in Kucha, Turfan and Kharashahr (see Chapter 1, Table 1.3 and 1.4). However, gypsum found in wall paintings in Domoko (including in Shahristan and Penjikent) and those in the Tarim Basin (including in Duldul-akur) contained no anhydrite when analysed by the State Hermitage Museum in St Petersburg. Because of this peculiarity, Kossolapov suggests that the latter 'rather pure' mineral gypsum ores that could be used without calcination must have been readily available where they were found (Kossolapov and Marshak 1999: 77). Similarly, gypsum substrates at Bamiyan have been found to contain mainly dihydrate calcium sulfate, and to date, no anhydrate form. However, in Bamiyan, the gypsum layer has a few impurities and is of a uniform texture, so it is unlikely that it originates simply from the use of a natural gypsum ore of high purity, as suggested by Kossolapov – it is more likely that the gypsum was manufactured in some way.

Distinctions of the usage between dihydrate and anhydrate are currently ambiguous, but both were probably produced deliberately and used regularly until at least the Middle Ages. For example, analysis of plaster grounds in 14th-century paintings from northern Italy shows that the coarse plaster (*gesso grosso*) consists of anhydrite (100%), anhydrite:dihydrate (75:25) or anhydrate:dihydrate (50:50), while the fine plaster (*gesso sottile*) used as a finish in the top layer is dihydrate (100%) or anhydrate:dihydrate (25:75) (Martin *et al.* 1992). It is not known whether gypsum or anhydrite were chosen based on the texture of their particles, origins, temperatures during the firing process or their other properties such as insolubility in water.[8] Where gypsum was used as a ground, it appears to have been applied with a large brush in a relatively short period of time; coarse brushstroke lines can be observed in the wall paintings of Caves I, K_3 and H.

2.2.5.4 Gypsum grounds containing red pigments

Most of Bamiyan's wall paintings have a white ground, with the exception of wall paintings in Cave J(d), which have a pale pink ground made from red pigments in a white ground (BMM120) (Fig. 2.20). The wall paintings at Foladi Cave 4 have two or three layers of ground, including a pale yellow ground of lead white with orange realgar (As_4S_4) (Yamauchi 2006b). In Cave J(d), one of the oldest caves at Bamiyan, the wall paintings rest on top of white gypsum mixed with red ochre.

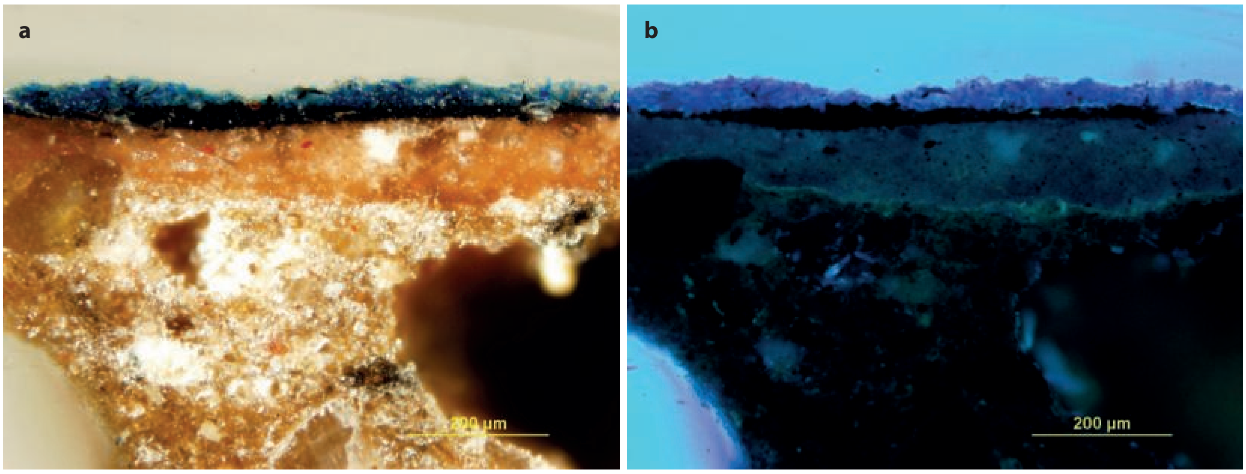


Figure 2.20 (a) PLM photomicrograph of a cross-section of a blue portion of a wall painting from Cave J(d) (BMM120). Red iron oxide particles can be seen underneath. Under the blue layer of lapis lazuli, a black layer of carbon black has been applied. (b) In the UV fluorescence image, a thin, fluorescent sizing layer is visible on top of the base coat.

A similar ground is found in wall paintings at Mīrān (3rd–4th century) (Stein 1980: appendix D). Wall paintings at the Mīrān Buddhist site are in the Greco-Buddhist style, one of the most famous of which is that of a winged angel (see Fig. 2 in the Introduction). Rowland (1947: 86) suggests that this work was stylistically and conceptually influenced by Western art:

> since colossi do not appear in Buddhism before the Gandhāra school, it may be that among the contributions of this hybrid art was the plastic realization of the superhuman nature of the Buddha contained in the texts, aided and abetted by the Greco-Roman artists' knowledge of over life-sized figures of gods and kosmokrators in the West.

According to Sir Arthur Church, a British chemist who analysed the wall paintings at Mīrān, a pinkish gypsum ground layer containing iron oxide (Fe_2O_3) was applied over the earthen renders. Paint was then applied to the ground using an *a secco* technique (Church 1921). Although there is a gap in time between the two, these techniques are common to the early wall paintings found in Cave J(d), and may be an important key to the emergence of wall paintings in Bamiyan. It has also been reported that the ground of the painted surface of Bamiyan's Eastern Giant Buddha is also a ground of pink gypsum mixed with red ochre (see Chapter 3, section 3.5) (Blänsdorf *et al.* 2009b). The date of this Buddha's construction is similar to that of Cave J, which also dates back to Bamiyan's early days. More recently, coloured ground layers were very fashionable in 17th- and 18th-century European paintings because they provided a warmer tone than white grounds. Ochre and red oxides were typically used to give this reddish hue (Duval 1992).

2.2.5.5 Lead white grounds

As mentioned above, 'lead white' is usually a synthetic pigment made of two crystalline phases: cerussite and hydrocerussite. The synthesis of lead white remained almost unchanged from antiquity through to the 19th century (Gonzalez *et al.* 2017). The historical and geographical production and use of lead white is detailed in the next section. In brief, it was traditionally produced by corroding metallic lead plaques by suspending them above vinegar in jars, stacked under horse manure (hence the name 'stack process'). Carbon dioxide and heat produced by the decomposition of the manure together with acetic acid vapours induced corrosion and the formation of cerussite and hydrocerussite on the lead's surface. The pigment was collected, treated by different post-synthesis processes (grinding, washing etc.) before being sold.

SR-XRD has shown that different grades of lead white can be classified based on the ratio of cerussite to hydrocerussite and on the crystallite size of pigments. These chemical signatures can be connected to different synthesis conditions or post-synthesis treatments employed by paint manufacturers and artists (e.g. when white powder produced by lead corrosion was washed in acidic solution or heated in water) (Gonzalez *et al.* 2017). Recently, SR-XRD revealed that Leonardo da Vinci employed two different lead whites in *The Virgin and Child with St Anne* (Musée du Louvre, Paris): one in the white ground layer and the other in the blue sky layer. Each offers different scattering properties and consequently differing white hues (Gonzalez *et al.* 2020). The best technique for distinguishing hydrocerussite and cerussite is XRD but some information on lead white composition can also be obtained with FTIR. Specifically, absorption of around 3530 cm^{-1} is characteristic of an O-H stretching bond in lead-hydroxide compounds (Siidra *et al.* 2018). Cerussite can be distinguished from hydrocerussite by its peak at 838 cm^{-1} ($n_2(CO_3)$), whereas hydrocerussite's spectrum is weaker (Brooker *et al.* 1983).

In the present corpus, a group of hydrocerussite/cerussite lead white grounds (12 caves: Bamiyan Caves B(d), F(c), L, N(a), S(a), Foladi Caves 2, 3, 4, 6, Kakrak Caves 43, 44 and Qol-e Jalal) appear at Bamiyan in the mid-7th to late 8th century, according to radiocarbon dating. This is common to the Bamiyan, Foladi and Kakrak caves. Although no dating has been carried out, the Qol-e Jalal cave is also considered to be newer than the Bamiyan site. Thus there is little doubt that wall paintings with lead white grounds tend to appear from the mid-7th century onwards. Caves with lead white in their wall paintings share a common square layout with a domed or Laternendecke ceiling. The style of the paintings is also similar, as is the regular arrangement of the Thousand Buddhas on walls like a mandala. Some examples of the use of lead white as a ground layer in the South and Central Asian region can be found at Ajanta, Kumtura, Bezeklik, Khocho, Duldul-akur and other sites, but the timing of their use is not well understood. In Sogdiana, a gypsum ground was used for wall paintings in Penjikent and Shahristan, and even in neighbouring regions. In the late medieval period (11th and 12th centuries), lead white was used as a ground for wall paintings excavated from the site of the Islamic capital city of Khulbuk (Shimadzu 2009).

Interestingly, different lead white compositions were detected in several layers within the stratigraphy of a single sample, notably in the ground and painting layers. In a wall painting sample (BMM083) from Cave F(c) and a wall painting sample (BMM035) from Cave N(a), the lead white in the ground layers mainly contained hydrocerussite. At the same time, the brown copper layer of the former and the green copper layer of the latter contained a significant amount of cerussite together with hydrocerussite. Conversely, in sample FDM055 from Foladi Cave 4, there are two white layers with high amounts of cerussite, while the lead white mixed with copper pigment in the green layer contains mainly hydrocerussite (Appendix III, Fig. A.III.104 and Tables A.III.5, A.III.20 and A.III.27).

A specific composition of lead white ground, hydrocerussite and susannite ($Pb_4SO_4(CO_3)_2(CH_2)$), was found in four samples: BMM035, BMM039, BMM040 and BMM177. The presence of this combination is highly unusual and has rarely been reported previously (Eastaugh *et al.* 2004: 230). Usually, when lead white is found with sulfates, this is due to degradation – in Bamiyan's paintings, anglesite ($PbSO_4$) and palmierite ($K_2Pb(SO_4)_2$) were detected in whitish veils covering the surface of some paintings (see section 2.3.1). In such a case, their specific distribution at the surface indicated that they formed during degradation processes due to reactions with volatile sulfur compounds from the environment, as observed in fragments from Rembrandt's *Homer* (Price *et al.* 2019). Here, however, susannite is present as micrometric particles, as is hydrocerussite, and both types of particles are homogeneously distributed in the lead white layer.

This confirms that susannite is part of the original lead pigment. Interestingly, leadhillite, another lead carbonate sulfate hydroxide of the same composition but different crystalline structure (leadhillite crystallises as a monoclinic form and susannite as a trigonal form) has been reported in Tang dynasty samples from the Tiantishan grottoes in Wuwei, China (Zhou *et al.* 1993). In this work, leadhillite is considered to be a natural mineral. In total, five types of lead white were identified (lead white, lead sulfate, lead hydroxychloride, lead chloride and leadhillite) from the wall paintings in the Tiantishan grottoes. More research would be necessary to identify the origin of the Bamiyan susannite (natural or synthetic) and to establish whether susannite is associated with a specific quality of lead white. In summary, these results show a large variation in the composition of lead white, which may be related to different grades and varying uses of this type of pigment.

Compared to lime and gypsum, lead white is a pigment with a prodigious capacity for obscuring underlying layers – even when applied in thin layers, it is capable of producing a bright white colour independent of any underlying colour. Artificially synthesised lead white crystals are generally uniform and fine in size and texture. The crystals are plate-like and arranged in layers within the coating, which boosts its adherence and resistance to peeling. Historically numerous attempts in alchemy have been made to produce lead white, especially in the Mediterranean world. The earliest record is probably in Theophrastus' treatise, *On Stones* (56) in which he describes a process involving a lump of metallic lead that is placed in a vessel and exposed to acetic acid vapours. The lead reacts with the vapours by forming a white lead acetate on its surface, which is scraped off. This then further reacts with carbon dioxide in the air and produces lead carbonate (Caley *et al.* 1956: 57). In another known process recorded by Vitruvius (Morgan 1960: 219, Ch. XII) the first step is to insert lead shavings in a jar of vinegar and cover it with a lid which produces a white substance that is collected, heated and then washed in water. Both of these processes are essentially similar to those used to synthesise copper acetate (verdigris) from copper (Rowland and Howe 1999: 94–95). Pliny (*Naturalis historia* XXXIV, ch. 54) cites two processes mentioned by Theophrastus and Vitruvius to create lead white (*psimithium* or *ceruse*); the former from Rhodes was more highly esteemed (Bostock n.d.).

Lead white was used as a painting material, primarily in secco wall paintings. In frescoes, it is thought that the lime mortar, which contains a lot of water and is a strong base, causes the lead white to darken (Giovannoni *et al.* 1990). However, the exact mechanisms underlying this discoloration still remain unclear. It is also known that when mixed with sulfur compounds such as cinnabar/vermilion (HgS) or when exposed to a sulfide-rich environment, lead white is converted into lead sulfide (PbS) and blackens. Fresco wall paintings are considered more susceptible to discoloration because there is no coating to shield the pigment particles from the surrounding environment. In any case, known problems of lead white cæruleum's discoloration meant that its use in frescoes was avoided (*Naturalis historia* XXXV, ch. 31) (Bostock n.d.).

Lead white has been found on painted sculptures on the island of Delos, on Macedonian tombs and on wall paintings in Alexandria. These were painted with the *a secco* technique using plant gums and other binding media. It is often used alone as a ground for sculptures, for example, but sometimes it can be mixed with Egyptian blue to produce a light blue tone, as seen in the painted marble throne of Eurydice found inside the Tomb of Aigai (Vergina) in Macedonia (340 BC). On the same throne, lead white also served as the base material for a red lake (Kakoulli 2009: 57).

Synthesised lead white was used not only as a painting material but also as a cosmetic and medicinal product. According to Dioscorides, the best lead white (*psiuthios*) for use in cosmetics is that from Corinth, Rhodes or Lacedaemonia (Laconia) in Greece (*De materia medica* 5: 103) (Osbaldeston 2000: 796). In another case, al-Khwārizmī (a mid-10th-century scribe) wrote in his *Mafāfīḥ al-'ulūm* (*Key to the Sciences*) that lead white was mixed with ointment to treat tumours in Arabic medicinal practices (Sato 2008: 101). Another book written approximately two centuries later by al-Shayzarī (d. 1193), *Kitāb nihāyat al-rutba fī talab al-hisba* (*The Book of the Islamic Market Inspector*) (12th century), states that in order to improve a drink's colour, taste and flavour, one must 'make a drink of sugar cane juice with milk, vinegar and lead-white (*isfidaj* [today's *sbidag*])', although the lead in the lead white meant that 'such a drink would be poisonous to the body' (Sato 2008: 100–101).

Thus, lead white was evidently used widely in the Mediterranean region, mainly for medicinal and cosmetic purposes as well as for painting. However, the history of its use in Central Asia is not yet clear. In Bamiyan, lead white is a non-native material that suddenly appeared in the mid-7th century, but where it was synthesised and how it came to Bamiyan still remains unknown. Lead white was imported to Japan in the Tang dynasty as *gofun* and in the Nara period as Tang *gofun*. In the Tang dynasty, the word '*hu*' referred not to the Persian or Iranian world, but also to other countries west of Persia. If the origin of *gofun* from the Nara period can be determined, it may provide a major clue to the origins of the lead white brought to Bamiyan.

Chapter 4 (section 4.2.2) describes an attempt to estimate provenance by analysing the lead isotopes in lead white. A larger number of cases using lead white would be necessary to accumulate data through isotope ratio analysis and other methods and, ultimately, obtain more reliable results. In addition to the identification and mapping of hydrocerussite and cerussite, among other compounds, μFTIR revealed the presence of lead soaps in lead white grounds (Appendix III, Fig. A.III.103). These probably emerged after a lead white pigment or lead drier (e.g. PbO) reacted with long-chain fatty acids and therefore evidence the presence of an oil-based binding media, as discussed below in section 2.2.9.

When lead white or lead oxide is mixed with an oil-based binding media, saponification can take place to form lead soaps. Saponification of lead oxide is generally faster and preferred over lead white (Cotte *et al.* 2006b). Mixing fats or oil with lead compounds has been a regular practice in the production of pharmaceutical lead plasters and ointments, as well as lead-oil

Figure 2.21 Lower half of the seated Buddha underneath the lower strata of Foladi Cave 4. Photo: H. Otake, courtesy of NRICPT, 2006.

paint media (Cotte *et al.* 2019). Many European treatises (e.g. *Recueil des essaies des merveilles de la peinture* by Pierre Lebrun in 1635 and the Brussels Manuscript) describe the preparation of 'fat oil' (*huile grasse*) (Merrifield 1967b: ccxxxviii), '*olio cotto*' or '*olio coto*' (Filippo Baldinucci and the Volpato Manuscript) (Merrifield 1967b: ccxxxix; Cotte *et al.* 2017), 'boiled oil' (Paduan Manuscript) (Merrifield 1967b: ccxxxviii, 692), 'fat Oyl' (Smith 1687; Cotte *et al.* 2017), and '*huile de litharge*' (Sloane MS 2052: De Mayerne 1620–1646), which involve cooking oil with litharge (lead oxide) and sometimes adding water (Cotte *et al.* 2017). Reproducing such recipes leads to the formation of a partially saponified oil (Cotte *et al.* 2006b). This kind of cooked oil possesses excellent properties for the preparation of ground or *imprimatura* layers, recommended by Sir Theodore Turquet de Mayerne 'to print canvases that won't crack or peel.' The noteworthy amount of lead soaps in Bamiyan supports the hypothesis that saponified oil was prepared following a similar recipe. Lead acts as a catalyst and accelerates the oxidative polymerisation of the drying oil, which then acts as the binding media, thus improving the drying properties of the paint film.

2.2.5.6 Overview of the ground materials used in Bamiyan's wall paintings

Wall paintings in Cave J(d) have a gypsum ground containing red pigment, reminiscent of the wall paintings at Mīrān. Most of Bamiyan's later wall paintings have a gypsum ground as used predominantly in Central Asia. However, some also have lead white grounds. Artificially produced lead white known as '*gofun*'[9] may have been one of many materials exchanged along East–West trade corridors by Sogdian merchants, who had close relations with the Iranian world but its specific origin is not clear. It may have been chosen as a material for painting due to lead white's excellent hiding power and its compatibility with oil-based binding media.

2.2.6 The underdrawings

In many of Bamiyan's wall paintings, the lines of underdrawings appear to be executed in reddish-brown red ochre (see Figs 2.2 and 2.3). The final details of an image, such as the eyes and nose, are then drawn in fine black or crimson lines. This technique can be seen widely in wall paintings in Ajanta, the Mogao caves (Dunhuang), the Kizil grottoes, the Bezeklik caves and at Kala-i Kafirnigan. In *buon fresco*, also, the underdrawings, now called *sinopia*, were painted with a reddish-brown ochre.

One of the main differences in underdrawings in secco wall paintings, as seen in Bamiyan, is the *buon fresco* process of applying one layer of lime mortar on top of the reddish-brown underdrawings, and then recreating the underdrawings with dots and scratched lines while the lime mortar is still fresh; the underdrawings may then be painted over. Traces of compass needles, or instruments used to draw

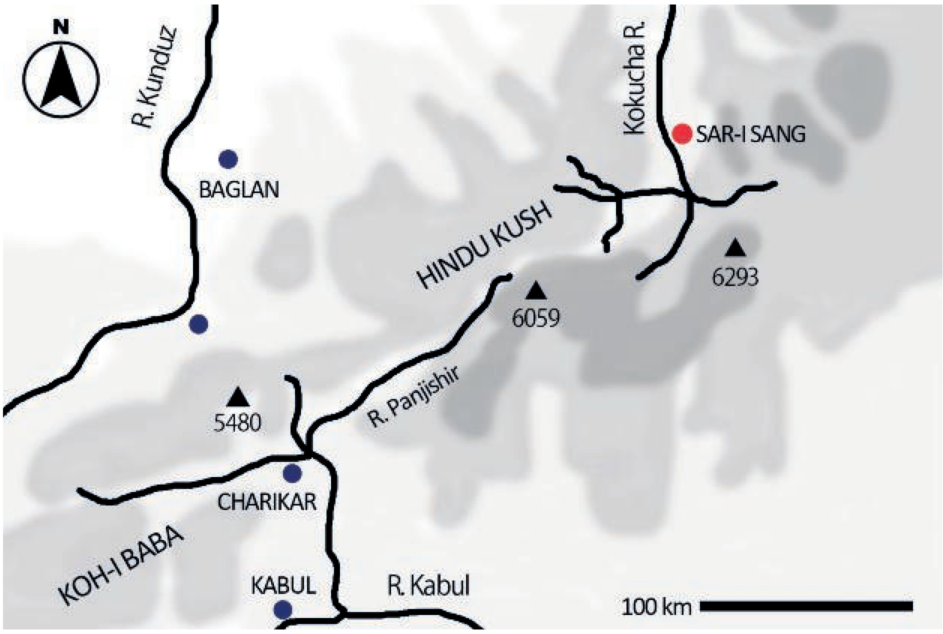


Figure 2.22 Map of a lapis lazuli mine in Badakhshan, Afghanistan (adapted from Herrmann 1968: fig. 1).

circles accurately, are often found in *buon fresco*. However, in Bamiyan there are no traces of nails, holes or anything else indicative of the use of a compass-like instrument to create circular compositions or a nimbus.

In exceptional cases, black lines of an underdrawing can be seen beneath a white ground layer (Fig. 2.21). On the walls of Foladi Cave 4, one thick black underdrawing remains. It is possible that this area was repaired sometime later as it has a yellowish-brown rendering that differs from the reddish-brown rendering on the ceiling. The white ground and the painted layer on top have disintegrated and fallen off, leaving only a few traces of red, green and blue colours. It is possible that a rough composition, like a *sinopia* in *buon fresco*, was painted under the white ground, but there are no examples of this type of painting outside Foladi Cave 4.

2.2.7 The coloured layers: pigments

2.2.7.1 Blue pigments: lapis lazuli and ultramarine

With the exception of one copper-based blue pigment, most blue pigments used in Bamiyan were made from lazurite $((Na,Ca)_8(AlSiO_4)_6(SO_4,S,Cl,OH)_2)$, a pigment extracted from natural lapis lazuli. Indigo may also have been used in some areas (e.g. the dark blue of the Śāla tree at Cave K$_3$), but this has not been confirmed scientifically as no analysis has been carried out.

Lapis lazuli has long been known to originate from the Sar-e Sang of the Kokcha River basin in the Badakhshan region of Afghanistan (Bowersox and Chamberlin 1995: 47) and has been used extensively as a blue gem and a pigment throughout the ages (Fig. 2.22). The earliest lapis lazuli was brought to Mesopotamia from Badakhshan as beads in the Hassuna period (6000 BC) of Yalim Tepe (Merpert *et al.* 1976), in the Halaf period (5500–5200 BC) of Tall Arpachiyah (Mallowan and Cruikshank Rose 1935), and in the Halaf period (400–5200 BC) of Nineveh (Thompson and Mallowan 1933). It arrived in Tepe Gawra in the late Ubaid period (c.4000 BC) and was used as a seal (Herrmann 1968). It is also known that lapis lazuli was used extensively in beads in the Indus civilisation (Early Harappan/Ravi phase: c.3700–2800 BC) and in the Caucasus of the Neolithic period (Dikshit 2012–13). Lapis lazuli was particularly popular and frequently used in medieval Europe from the 14th to the mid-15th century (Plesters 1993). The relative proximity of Bamiyan to the lapis lazuli mining area (about 500 km) suggests that it was abundant. Lapis lazuli may also have been widely traded as Buddhist culture and industries were thriving.

As a natural mineral, lapis lazuli contains not only blue lazurite, which is composed of light elements such as sodium, aluminium and sulfur, but also white calcite ($CaCO_3$), pyrite (FeS_2), diopside ($MgCaSi_2O_6$) or wollastonite ($CaSiO_3$). The quality of blue, therefore, varied with the amount of impurities as well as the temperature of the lapis lazuli

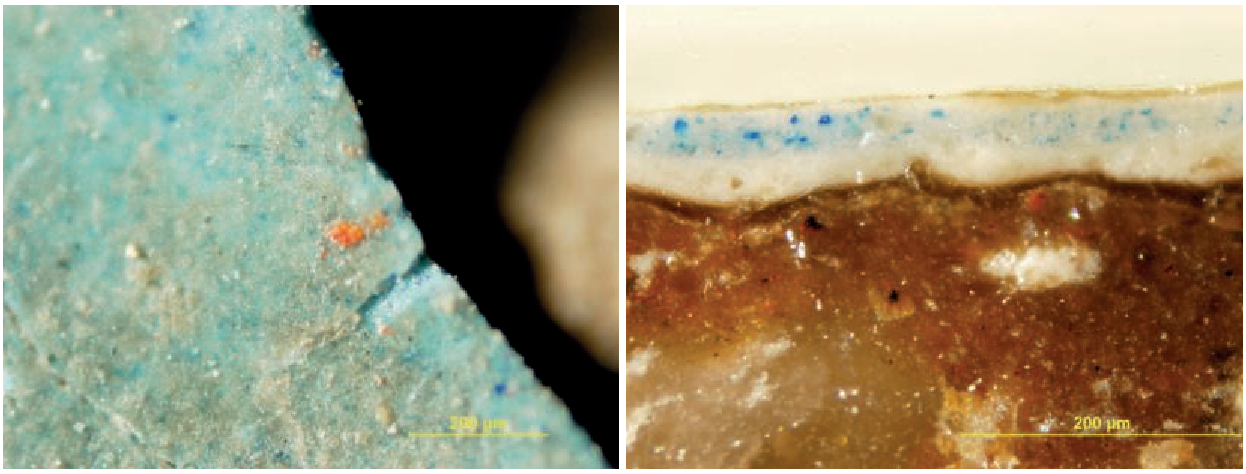


Figure 2.23 Stereomicrographs of a light blue portion (BMM001) in Cave N(a).

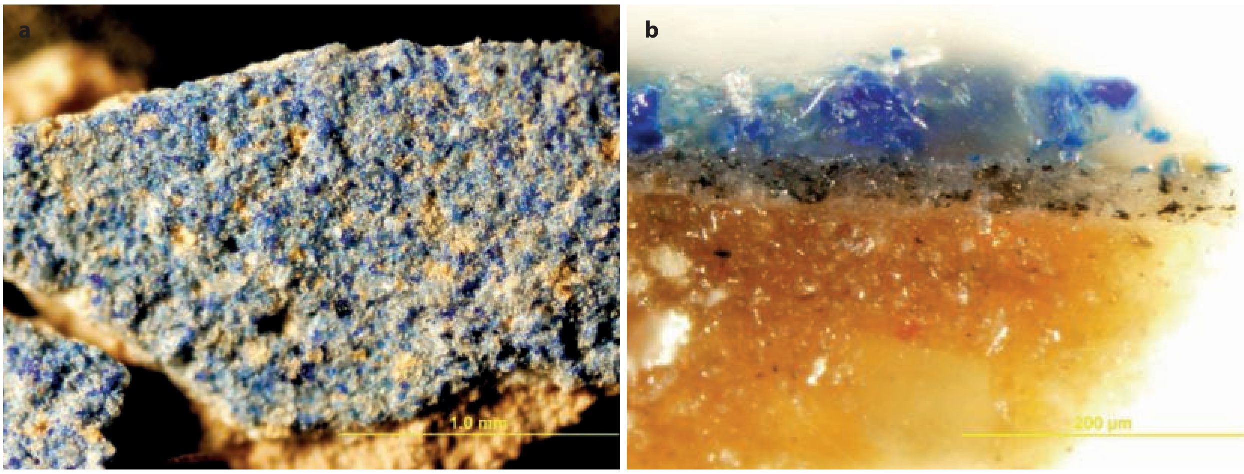


Figure 2.24 (a) A stereomicrograph and (b) a PLM photomicrograph of a blue portion of a wall painting from the East Displaced Cave (BMM155).

when the lazurite was extracted (Gambardella *et al.* 2020). In order to obtain a bright blue, it is necessary to separate the lazurite from any impurities although it is difficult to separate the blue particles. Because the specific gravities of lazurite and calcite are similar, if the original lapis lazuli stone is ground and sedimented, the minerals will not be successfully separated and only a dull blue-grey powder will be obtained. Around the 13th century, a method finally emerged for refining natural ultramarine blue by removing its impurities and isolating only the clean, blue lazurite. The most famous book describing this technique is *Il libro dell'arte* by Cennino Cennini (Thompson 1960) which includes a method for mixing and kneading ground lapis lazuli powder with pine resin, mastic resin[10] and beeswax, and then adding linseed oil and rubbing it in warm lye to obtain a blue pigment (Thompson 1956). This seems to be based on the principle of using a heavy liquid similar in density to that of the substance to be separated.

There are two different uses for lapis lazuli. The first is the so-called natural ultramarine blue, in which only the finely textured, fine blue lazurite is separated from the lapis lazuli and placed over a white ground (lead white) to produce a bright light blue colour (Cave N(a)) (Fig. 2.23). The second involves taking coarsely crushed lapis lazuli particles and laying them on top of a black layer to create a deep blue colour (Cave East Displaced Cave) (Fig. 2.24). The first technique suggests that, as a natural ultramarine pigment, lapis lazuli may already have been artificially refined and used in different colours as far back as Cennini's time. This technique can be seen in use with lead white in an Italian fresco painting of the Monastery of St Anne in Camprena, Tuscany (15th–16th century) (Fig. 2.25).

The black layer used in the second technique is a mixture of white pigments such as gypsum and carbon-based substances, for example, lampblack or charcoal black. Such paintings, with a black layer underneath the crushed lapis lazuli, can be seen

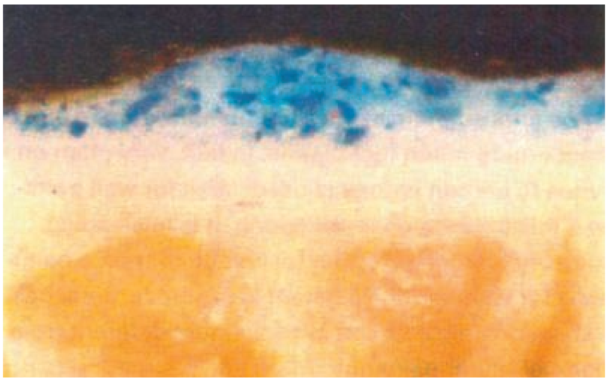


Figure 2.25 Fresco painting of the Monastery of St Anne in Camprena, Tuscany (Sienese Sodoma, 15th–16th century). Blue layer with purified ultramarine blue and lime white (after Malaguzzi-Valerj 1973).

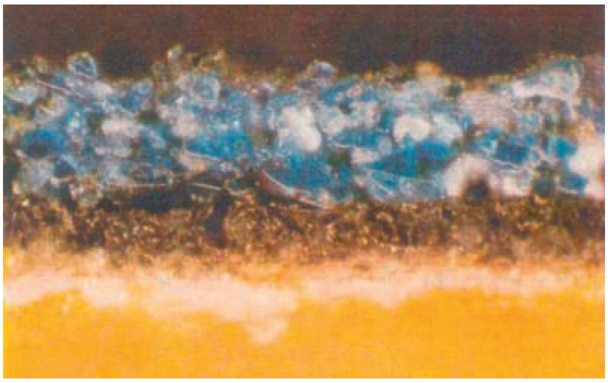


Figure 2.26 Fresco by Simone Martini (14th century): a black layer under a coarse blue (after Malaguzzi-Valerj 1973).

in the wall paintings of Bamiyan Caves C(a), D, G, I, J(b), J(c), J(d), K_3 and the East Displaced Cave. Basically, the black layer under coarse lapis lazuli tends to be found in gypsum ground wall paintings. A similar layering mechanism can also be seen in the fresco painting by Simone Martini (14th century) (Fig. 2.26). Both the layering and the refining techniques show that, despite Bamiyan's geographical advantage of being relatively close to the lapis lazuli mines, raw lapis lazuli was not used in unnecessarily large quantities, but instead incorporated skilfully and efficiently, a common practice in medieval Europe in a later period.

Lapis lazuli was not only engineered to produce different blue hues, such as by modifying particle sizes and degrees of refinement – white grounds were also adjusted with lead white used alongside refined lapis lazuli, and black layers with gypsum together with coarse lapis lazuli. Therefore, the decision as to what ground to use and how to refine the lapis lazuli was considered jointly. As it would have been difficult to refine natural ultramarine blue, it is possible that the ultramarine pigments traded on the Silk Road at that time were ready-made products, having already been separated and refined by specialists.

Further back in time, there is one example from the Mogao grottoes, Dunhuang (Chapter 4, section 4.2.1) of a paint mixture of gypsum and lapis lazuli particles such as the one seen in Bamiyan. It involves a blue pigment lump excavated from a site in front of Caves 53–55, which was processed into a hardened mixture of crushed lapis lazuli powder, gypsum and glue (Mizuno *et al.* 1996). This product could be prepared for use as a watercolour paint simply by adding water.

2.2.7.2 Red pigments: cinnabar/vermilion

Red mercury sulfide (HgS) is a pigment produced from a crushed natural mineral called cinnabar and a fine-grained synthetic (produced by sulfurising mercury) called vermilion. According to Pliny, cinnabar was imported from the Almaden mines in the Baetica region of the Iberian Peninsula and smelted in Rome (*Naturalis historia* XXXIII, ch. 40) (Bostock n.d.). According to Theophrastus, there is an earlier reference to a manmade vermilion (synthesised mercury sulfide) from near Ephesus in Turkey, but this is not well known (Eastaugh *et al.* 2004: 386).

Large quantities of cinnabar were mined from Liaoning Province in China, and may have been brought to Central Asia as Chinese vermilion (Kossolapov and Marshak 1999: 71, 79), although the exact source of the cinnabar is not known. In Bamiyan, cinnabar/vermilion is found in the wall paintings of various caves, but generally finely and uniformly grained, with no impurities such as quartz, suggesting that synthetic vermilion was used. Historically, compared to minium, cinnabar/vermilion would have been expensive and difficult to obtain due to its scarcity, but it would have also been an effective way to obtain a highly saturated red colour in small amounts.

2.2.7.3 Green pigments: copper-based green compounds

Analysis has confirmed the use of copper-based compounds for green pigments in Bamiyan. Among the blue-greens, atacamite ($Cu_2Cl(OH)_3$) has been detected. Much of the green used in Bamiyan and Foladi has turned blackish-brown, notably in Cave K_3 but underneath these dark brown areas, some layers still retain a bright green colour. Since X-ray fluorescence analysis has detected copper in these degraded areas (Yamauchi 2006b), it is likely that they were originally coloured with a copper salt.

Therefore we can assume that the blackened area is a discoloration of the green copper salt. This green colour is discussed further in section 2.3.1 and is considered to be the result of copper-based green pigments transforming into black alteration due to adverse environmental conditions.

Observation of the cross-section shows a green transparent substance in green paint from part of the painting that did not blacken (FDM011) (Appendix III, Figs A.III. 82 and A.III.83), which suggests that it contains a large amount of organic material. Absorption tests indicated the presence of a natural resin, which may have been a verdigris pigment such as copper resinate. Atacamite was also detected in Caves J(b), N(a) and Foladi Cave 4 (Table 2.1). Paratacamite ($Cu_2(OH)_3Cl$) was also identified in Foladi Cave 4, but as it is a naturally occurring substance associated with atacamite, its presence is not surprising.

Examples of the actual use of chrysocolla ($Cu_{2-x}Al_x(H_{2-x}Si_2O_5)(OH)_4 \cdot nH_2O$, $x < 1$) as a green pigment was found not only by Gettens (1938a,b) in Bamiyan's wall paintings and in the Kizil caves but also by the State Hermitage Museum and Riederer in the Kizil and Kumtura caves (Kossolapov and Marshak 1999; Riederer 1977). Riederer (1977: 377) observed that the bluish-green outlines of green areas were typically chrysocolla. However, despite our extensive analyses, we were unable to identify any chrysocolla from green areas in Bamiyan and the unique glossy greenish-blue areas in Kizil (Li 2005; Taniguchi 2016). A sample of greenish-blue pigment taken from Kizil Cave 69 was identified as a high arsenic-containing copper (Murofushi and Kijama 2014), a material not mentioned in prior studies. According to Theophrastus and Vitruvius, the term 'chrysocolla' was used in the same context as malachite (Eastaugh *et al.* 2004: 103). Vitruvius, for example, describes chrysocolla as being obtained from a copper mine in Macedonia (Osbaldeston 1999: 93), but it is not clear whether he was confusing it with malachite.

Atacamite is a green crystalline substance found in copper deposits and also obtained from reactions of metallic copper to brine. It has been suggested that a substance known in China as 'stone green' (Laufer 1967: 510) may indeed be atacamite. In China's Sui dynasty (581–618), stone green was attributed to Persia of the Sasanian dynasty, and in China's Northern Zhou dynasty (557–581) to Kucha. Su Gong of the Tang dynasty (7th century) is said to have pointed out that stone green was a product from Kalashar found in water collected using a concave stone (Laufer 1967: 510). Pliny describes a blue substance, *chalcanthum*, obtained by putting water into (copper mining) wells or pits in Spain (*Naturalis historia* XXXIV, ch. 32) (Bostock n.d.). Riederer (1977) considers that this may indicate the manufacture of manmade atacamite.

Although malachite was believed to have been the most common green copper mineral historically and geographically in Central and East Asia, it is now clear that atacamite was actually very common in these areas. It was frequently used not only in Bamiyan, but also in wall paintings in the caves of Turfan, in Kucha and other areas west of Gansu Province, and in Kashgar (Chapter 1, Tables 1.3 and 1.4). There are also examples of its use in treasures in the Shōsōin Repository (Nara, Japan) with strong connections to the West, such as in the underpainted inlays on the back of the octagonal mirror with Turbo marmoratus (green turban) inlay in the Shōsōin's northern storehouse (North 42) (Naruse 2004).

According to researchers at the Dunhuang Research Institute, on the other hand, there is little evidence of the use of atacamite in wall paintings in Tang dynasty tombs around Xi'an. They believe that in ancient times in China, atacamite was derived as a secondary product of copper deposits, and later artificially synthesised from metallic copper using vinegar (Wang *et al.* 2002). Another example from Yuan dynasty wall paintings is the production of green pigment from bronzeware, which later deteriorated to form atacamite (Twilley and Garland 2005). The production of green pigments, especially atacamite, is a complex issue.

Atacamite was identified in the green painted area of a polychromed sculpture in Cave 6 (AD 465–494) at the Yungang grottoes in Northern Wei. This atacamite is assumed to be manmade atacamite due to the pigment's characteristic 'globular rosettes with undular extinction, the occasional presence of central dark core, and high relief' viewed under a transmitted light microscope (Piqué 1997: 353). Regardless, there is no doubt that this 'salt green' was brought to China via Central Asia over the course of trade along East–West routes as evidenced by the accounts of Li Xun and Li Shizhen.

In *Oversea Materia Medica* (*Hai Yao Ben Cao*)[11] of c.750, Li Xun stated: 'salt green is produced in Persia and adheres to stones; that imported by ship is called stone green. Chinese imitations, made of copper and vinegar, should not be used for medicine, and their colour does not last long'. 'Green salt', possibly the same as salt green, was described by Schafer as similar to azurite, used for curing eye disease and brought to Tang by ship from the Karashahr region and Iran. He also identified it as crystallised copper sulfate, sometimes called 'blue vitriol' (Schafer 1963: 194). Salt green is known as *zingār* in Persian and, because it is made by corroding copper or brass using vinegar vapours, it is believed to be copper acetate (Laufer 1967: 510–511).

At the same time as the natural salt green, a synthetic salt green appeared. Copper is a highly reactive metal and has therefore been used since antiquity to cultivate pigments by reacting it with a variety of substances. It is known that 'verdigris' can generate a wide range of colours and crystal structures, depending on the conditions and processes of synthesis (Scott *et al.* 2001). This salt green is made by creating a reaction between vinegar and copper and, judging by how it is described, tarnishes easily. This artificial salt green, 'verdigris', is a copper-based pigment that does not have a specific chemical composition. Generally considered to be a hydrated copper(II) hydroxy-acetate ($Cu_x(CH_3COO)_y(OH)_z.nH_2O$), it can also exist as a copper(II) acetate ($Cu(CH_3COO).nH_2O$), or a copper(II) hydroxychloride ($Cu_xCl_y.(OH)_x$) (Roja *et al.* 2007). A basic method for synthesising verdigris, as described by Theophrastus (*On Stones,* 57) (Caley *et al.* 1956) is the same as that for synthesising lead white, except that lead is replaced by copper. A piece of metallic copper is exposed to vinegar vapours to form copper acetate rust crystals on the surface of the copper plate, which are then scraped and collected to form a blue-green pigment.

Another facet of verdigris discoloration is its tendency to react with organic substances. For example, a wide variety of forms of copper resinate and protein salts can be produced by mixing verdigris with natural resins. Such organically produced salts have a deep and lustrous glaze, unlike the inorganic salts of copper, such as malachite and atacamite, used on their own. Many of the transparent greens observed in Bamiyan's wall paintings probably contained such organic salts. While there are historical accounts of the synthetic production of verdigris, it is a pigment that remains little known today since no specific artefacts have been found to utilise it.

Few examples of unaltered verdigris exist in historical sources – even if more examples were located, verdigris would be difficult to identify through simple analytical methods such as XRF because of its nature as an organometallic compound, making it difficult to verify the regional and historical extent of its use. The only case where a connection can be made is the analysis of the green colour of the Duldul-akur site (around the Tarim Basin) which showed copper salts absorbed on gypsum (Kossolapov and Marshak 1999). In this case, copper was detected as an element, but no inorganic compounds such as malachite or atacamite in crystalline form were found, so it is likely to have been an organic copper salt of some kind. This and other examples of green pigments in wall paintings along the Silk Road could be re-examined in the future to identify more cases.

2.2.7.4 Yellow pigment

The most common colours seen at the Bamiyan and Foladi sites are blue, red, reddish-brown and white; yellow is also present, albeit to a lesser extent. In some cases, such as at Foladi Cave 4, the greenish-white colours are more pronounced, but in the majority, green and yellow are less common. It should be noted that the materials analysed are not the same as they were in the original compositions. Many of Bamiyan's wall paintings have not retained their original colours – they have faded or changed. In their original state, Bamiyan's wall paintings would have been extremely colourful with many yellow and green hues. In most cases, orpiment was assumed with the presence of arsenic in the altered areas.

2.2.8 The binding media

In order to be used in painting, binding media must possess a certain degree of stickiness, dry or harden with time and should not dramatically modify a pigment's colour.[12] As described in the Introduction to this book, historically a variety of materials has been used as binding media, including animal glues, plant gums (polysaccharide), resins, beeswax, eggs, casein and drying oil. Binding media may include not only items available locally but also imported through trade. Because pigments and binding media have physical properties that are compatible with each other, it is necessary to consider the palette and painting techniques of the painter, rather than just the influence of materials in isolation.

Analysis of binding media in Central Asian wall paintings was carried out in Penjikent and Ajinatepe, where gums from fruit trees (sap from plum and apricot trees) were detected alongside tragacanth gum and guar gum traceable to plants in South Asia and Asia Minor (Birstein 1977; Kossolapov and Kalinina 2007). In a previous study of binding media in Bamiyan's wall paintings, Gettens found animal glue in fragments of wall paintings from the caves around the Eastern Giant Buddha (Gettens 1938a), and a survey team from the Archaeological Survey of India (ASI) identified both plant gum and animal glue (Lal 1970). The exact location of the wall paintings studied is unknown, but it is likely that restoration work was carried out in the caves surrounding both Giant Buddhas, and therefore the analysis was limited to the same areas.

In previous studies, it was not clear whether the results obtained were representative of the region or the period because no comprehensive analyses of binding materials were conducted across various periods and styles at a single site. For this reason, we attempted to carry out a diachronic analysis of as many cave wall paintings as possible. Microsamples taken from wall paintings included all the painting layers, the ground layer, the sizing layer and the glaze. In addition to binding media, each sample was expected to contain various types of organic material. The minute samples were then analysed directly by GC-MS. As the thickness of each layer ranged from a micrometre to tens of micrometres, it was not possible to mechanically separate out the layers. The same samples were also analysed in turn in settings for the identification of fatty acids, amino acids and polysaccharides (plant gums). In the same way, the samples were tested to identify proteins and gums using the ELISA method and examined to investigate whether any reinforcing information could be obtained (the quantitative values of fatty acids, amino acids and sugars for each sample are given in Appendix III, Tables A.III.32 and A.III.33). GC-MS fatty acid analysis was carried out on 52 samples (from 30 caves and both Giant Buddhas), amino acid analysis on 29 samples (from 24 caves and both Giant Buddhas) and plant gum analysis on 52 samples (from 28 caves and both Giant Buddhas). The sample preparation was not sufficient for analysis of polysaccharides, so data were only obtained from two samples: Cave B(d) (BMM063), fructose, glucose, arabinose; and the Eastern Giant Buddha (BMM191), xylose (Appendix III, Table A.III.3).

Using ELISA at the Getty Conservation Institute (GCI), Los Angeles, two of 25 samples (22 caves) showed positive results: egg white from BMM040 (Cave N(a)) and casein and animal glue from BMM082 (Cave F(c)) (Appendix III, Tables A.III.35a and A.III.35b). Positive results were achieved in 26 tests (16 samples) of 26 samples at the National Museum of Western Art (NMWA) in Tokyo (Appendix III, Table A.III.36). From these analyses at NMWA, the following materials were identified:

- plant gum: BMM096 (Cave C(a)); BMM082 (Cave F(c)); BMM115 (Cave J(c)); BMM203 (Cave L)
- casein: BMM191 (EGB); BMM094 (C(a)); BMM095 (C(a)); BMM210 (C(a)), BMM209 (C(b)); BMM129 (J(b)); BMM113 (J(c)); BMM115 (J(c)); BMM120 (J(d)); BMM169 (J(f)); BMM137 (Cave K_3); BMM203 (Cave L); BMM082 (Cave F(c)); KAK05 (Kakrak Cave 44)
- egg: BMM210 (Cave C(a)); FDM003 (Foladi Cave 5); FDM019 (Foladi Cave 4)
- animal glue: BMM191 (EGB); BMM210 (Cave C(a)).

However, there is a fundamental methodological difficulty using the ELISA technique: it is problematic when it comes to aged materials, especially those from an external environment (Cartechini *et al.* 2010). For example, from BMM082 (Cave F(c)), animal glue and casein were identified by the GCI, and from the same sample, plant gum and casein were detected by the NMWA. The reason for the discrepancy may be because reaction degrees also differ owing to the different antibodies employed at the two laboratories. Different antibodies recognise different parts of the collagen molecules, therefore the reaction differs depending on whether or not that part of sequence is broken. Due to the manufacturing process of animal glues, reaction with metal ions in coexisting pigments, the raw animal species and aging causes disruptions of the sequence of collagen or loss of epitope. Therefore, antibodies often fail to react with collagen of old animal glues in paint samples. ELISA can also sometimes give both false/positive and false/negative results, especially when analysing aged samples, so differing results are not unusual. In this case, therefore, two different tests were attempted. Additional organic analyses using different methods such as GC-MS and LC-MS are indispensable to obtain results.

In addition, SR-μFTIR maps were acquired on 23 multilayered fragments (prepared as thin sections or by gently pressing raw paint fragments with a diamond compression cell) to identify the different classes of organic material and to locate them (as well as some inorganic components) in the different layers (Appendix III).

A comprehensive list of GC-MS and ELISA results obtained from the analysis of organic matter

Table 2.3 Summary of organic analysis by GC-MS and ELISA.

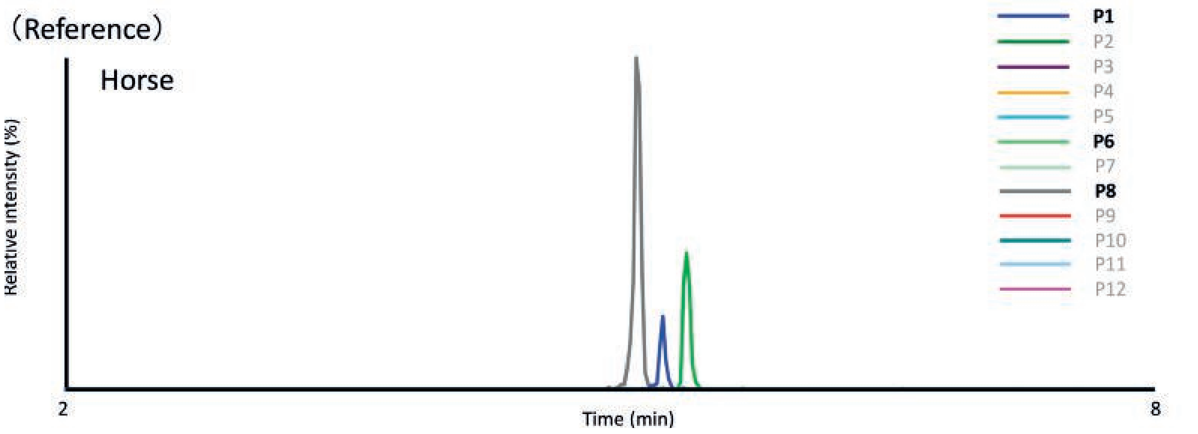
Site	*Cave*	*Ground layer*		*Proteins*		*ELISA (US)*	*ELISA (Japan)*	*Polysaccharides*		*Fatty acids*	
		Gypsum	*Lead white*					*plant gums*	*Other*	*Drying oils*	*Other*
Bamiyan	A lower salle	◎									
	C(a)	◎					Egg white, animal glue, casein, plant gums		●		
	East III	◎		●							
	E(e)	◎		●							●
	G	◎		●	Casein						
	H(a)	◎		●							
	H(b)	◎		●	Egg						
	I niche	◎									
	J(b)	◎		●							
	J(c)	◎				Egg yolk		●			
	J(d)	◎					Casein, plant gums				
	J(f)	◎		●	Casein		Casein				
	K_3	◎		●			Casein				●
	M	◎		●	Egg			●			●
	EGB	◎		●	Cow skin glue		Animal glue, casein, tragacanth				
	WGB	◎		●	Cow skin glue						
	B(d)		◎	●	Animal glue			●		●	
	C(b)						Casein, tragacanth				
	D			●							
	E(c)								●		
	F(c)		◎	●			Plant gums, casein	●		●	●
	L		◎	●			Casein, plant gums			●	
	N(a)		◎	●	Horse glue	Egg white		●		●	
	S(a)		◎					●		●	
	East Displaced								●		
Foladi	2		◎							●	
	3		◎	●						●	
	4		◎	●	Animal glue		Egg white	●		●	
	5			●	Egg		Egg white				
	6		◎	●	Animal glue					●	
Kakrak	43		◎	●	Animal glue			●		●	
	44		◎	●	Egg		Casein			●	
Qol-e Jalal			◎	●						●	

is given in Table 2.3. The following indices were used as a basis for preparation of Table 2.3. In the fatty acid analysis, quantitative detection of fatty acids included pimelic, suberic, lauric, azelaic, sebacic, myristic, palmitic, stearic, arachidic and oleic acids. The analysis was conducted with the aim of identifying oils, beeswax and resins. When concentrations of azelaic acid, a saturated dicarboxylic acid, were found to be high (0.6 or above), the material was considered to be a possible oil. When the azelaic acid to palmitic acid radio (A/P value) was above 1, it was considered to contain highly oxidised drying oil. Artificially oxidised oils such as stand oils and sun-thickened oils are used today in oil paintings. Historically, however, they were also used in oil paintings, as described by Cennini in passages on how to make a boiled (Thompson 1960: 58, ch. LXXXXI) or sun-thickened (Thompson 1960: 50, ch. LXXXXII) linseed oil for mordant and other uses. Intentionally oxidised drying oil might also be used.

If amino acids are detected but no fatty acids, the proteins may be attributed to glues, eggs, casein, or proteins contained in plant gums. In cases where neither fatty acids nor amino acids are identified, it is possible that a completely different substance was used as the binding media, such as a polysaccharide. Identifying binding media in wall paintings is quite challenging due to exposure to the external environment. In addition, the original binder is likely to have been modified due to aging and environmental degradation (Schilling *et al.* 2010; Sotiropoulou *et al.* 2018).

Results from ELISA and GC-MS analysis of the two cases of plant gums are also listed as well as the results of a layer-by-layer analysis by µFTIR of the same cases. Cross-checking has been carried out with certainty. In addition, materials used in the ground, such as lead white or gypsum, are also given for reference, as it is thought that the ground material is key to the overall coherence of painting techniques. The samples can be divided into five

Sample	*P1*	*P2*	*P3*	*P4*	*P5*	*P6*	*P7*	*P8*	*P9*	*P10*	*P11*	*P12*
BMM 194	○	-	-	-	-	○	-	○	-	-	-	-



Chain	*Location*	*Marker peptide*	*Sequence*	*Bovine (cow)*	*Horse*	*Sus scrofa domesticus (pig)*	*Sheep*	*Goat*	*Deer*	*Rabbit*	*Sturgeon*
α1 (I)	316-327	P1	GFOGADGVAGPK	○	○	○	-	-	○	○	-
	316-327	P2	GFOGSDGVAGPK	-	-	-	○	○	-	-	-
	688-704	P3	GAAGPOGATGFOGAAGR	-	-	-	-	-	-	-	○
	733-756	P4	GETGPAGROGEVGPOGPOGPAGEK	○	-	-	-	○	○	○	-
	741-756	P5	AGEVGPOGPOGPAGEK	-	-	-	○	-	-	-	-
	889-906	P6	GEAGPAGPAGPIGPVGAR	-	○	-	-	-	-	-	-
	889-906	P7	GETGPAGPAGPAGPAGAR	-	-	-	-	-	-	-	○
α2 (I)	238-252	P8	GIOGPAGAAGATGAR	-	○	○	-	-	-	-	-
	253-264	P9	GLVGEOGPAGTK	-	-	-	-	-	-	○	-
	361-374	P10	GFOGSOGNVGPAGK	-	-	○	-	-	-	-	-
	978-990	P11	IGQOGAVGPAGIR	○	-	-	-	-	-	-	-
	978-990	P12	TGQOGAVGPAGIR	-	-	○	○	○	○	-	-

O indicates 4-Hyp.
The numbering of residues begins with the triple-helical region of the chains.

Figure 2.27 List of marker peptides for animal species identification of glue and result of BMM194.

groups: (A) those based on drying oils; (B) those using both protein and plant gums; (C), those using plant gums/sugar; (D) those containing proteins; and (E) those that would have used something else.

Group (A) includes 12 caves: Bamiyan Caves B(d), F(c), L, N(a) and S(a); Foladi Caves 2, 3, 4 (4B) and 6, Kakrak Caves 43 and 44 and Qol-e Jalal. In addition to the detection of fatty acids, group (A) was judged to have a highly oxidised drying oil because of its high azelaic acid concentration and high A/P value. The P/S value (palmitic acid/stearic acid ratio), which is constant regardless of the degree to which an oil dries, is useful in order to identify types of drying oil (Mills and White 1994). The P/S values in these samples generally show a value of 3 (Appendix III, Table A.III.32), which is close to that of walnut oil (3.1) when compared with typical reference data. It should be noted that the identification of drying oils using P/S values can be impacted by the degradation and preferential evaporation of palmitic acid (Schilling *et al.* 1999; Bonaduce *et al.* 2012). Sesame oil and tung oil, which may have been used in East Asia, have P/S values of about 1, as does linseed oil (perilla oil: 1.2, tung oil: 0.9, linseed oil: 1.2) (Taniguchi *et al.* 2022). Accordingly, the type of oil here is closer to walnut or perhaps poppy seed (both walnut trees and poppies grow near Central Asia). From the data obtained, it is difficult to distinguish whether the fatty acids are derived from walnut or poppy seed oil using current methods. Interestingly, all 12 of these caves have one thing in common: a lead white and oil ground layer. This kind of lead white-oil *imprimatura* is commonly found in early European oil paintings.

Group (B) contains both proteins and polysaccharides but no fatty acids. Protein analysis has since evolved from the time of this study, but the methodologies utilised here were modified because the amino acid technique was heavily impacted when silica, a common mineral detected in wall painting samples, was present; either way, the paint samples are water-soluble. The wall paintings included here are those of Bamiyan Caves C(b), E(c), J(c), J(d) and

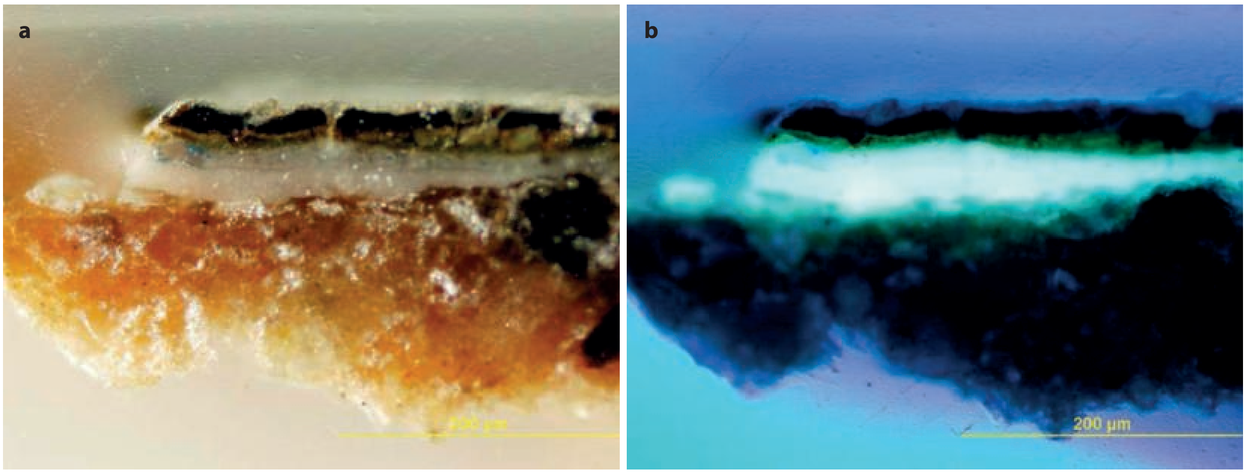


Figure 2.28 (a) PLM photomicrograph of a cross-section from a black portion of a wall painting (FDM026) on the west wall of Foladi Cave 6. (b) In the UV fluorescence image, the green transparent layer is still visible under the darkened surface layer.

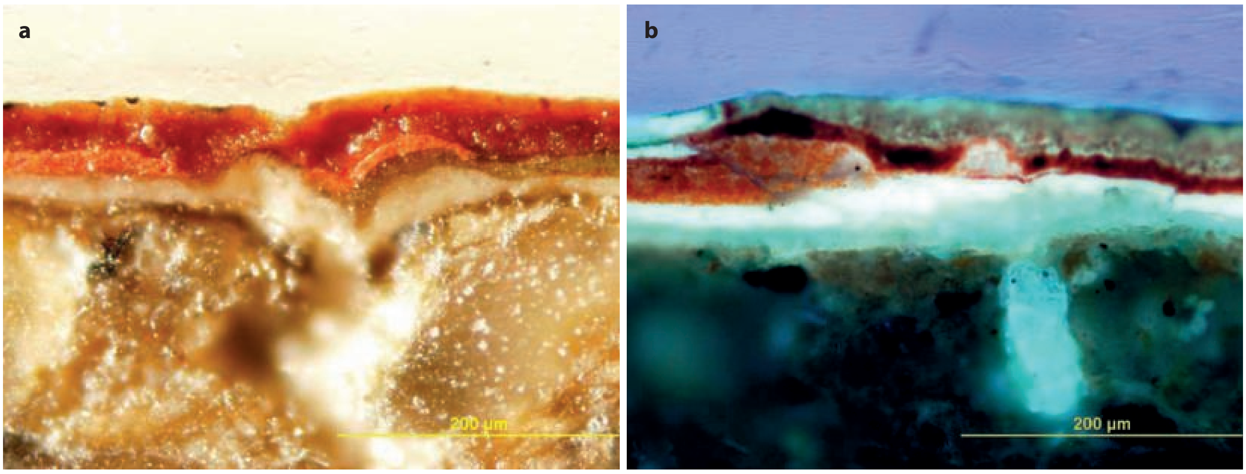


Figure 2.29 (a) PLM photomicrograph of a cross-section of wall paintings from Cave N(a), the red area (BMM002). The transparent red colour is superimposed on the opaque red colour. (b) In the UV fluorescence image only the area of the transparent red line shows strong UV fluorescence.

M and the surface painting of the Eastern Giant Buddha. It is not known if this proteinaceous material was all from binding media or whether it was partially applied in a certain layer. Group (C) contains polysaccharides not found from any caves. Polysaccharides are always found with proteins.

Group (D) contains proteins: wall paintings of the Bamiyan Caves East III, E(e), D, G, H(b), J(b), J(f) and K_3, Foladi Cave 5, and the surface painting of the Western Giant Buddha. Casein was identified from Caves C(a), C(b), E(c), F(c), G, J(d), J(f), K_3 and L, and Kakrak 44, EGB, egg from Caves J(c), M and N(a), Foladi Cave 5 and Kakrak 44, and animal glue from Caves B(d), H(b), K_3 and N(a), the Eastern Giant Buddha and Foladi Cave 4.

The wall paintings in the last group (E), in which neither fatty acids nor amino acids were detected or not fully tested, include those of Caves A lower salle, East Displaced and I. By elimination, but without any proof, these paintings could be made of plant gum/honey or another substance altogether. All the wall paintings were applied on top of gypsum.

The wall paintings in the 12 caves where drying oil served as a binding media are characterised by the use of oil-lead white as a ground material. The rest of the wall paintings are made of gypsum white ground. Egg has been detected in some of the wall paintings containing protein; other possible sources of protein include animal glue and casein. In contrast to the aforementioned oil paintings, some wall paintings can be regarded as watercolour paintings. However, in one sample (BMM194) from Cave N(a) – that was reanalysed at the Nippi Research Institute of Biomatrix (NRIB) in Tokyo using collagen analysis with LC-MS – collagen from a horse was found (Taniguchi 2020b) (Fig. 2.27). In this cave, horse collagen may have been used as sizing on the clay render, although the work was painted using oil painting techniques and the

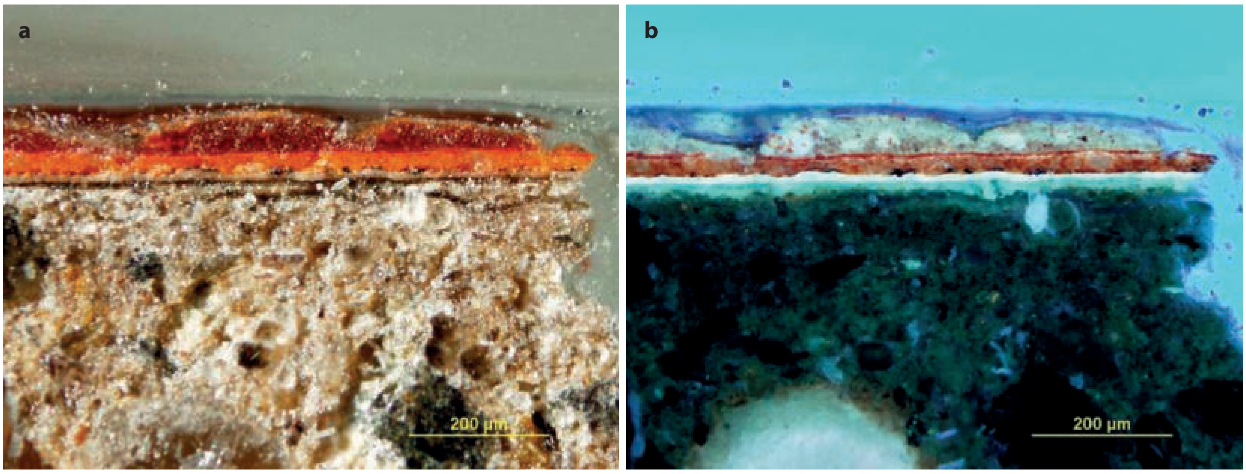


Figure 2.30 (a) PLM photomicrograph and (b) UV fluorescence image of a cross-section of a red portion of a wall painting (BMM183) from Cave N(a) showing the application of two layers of different shades of red.

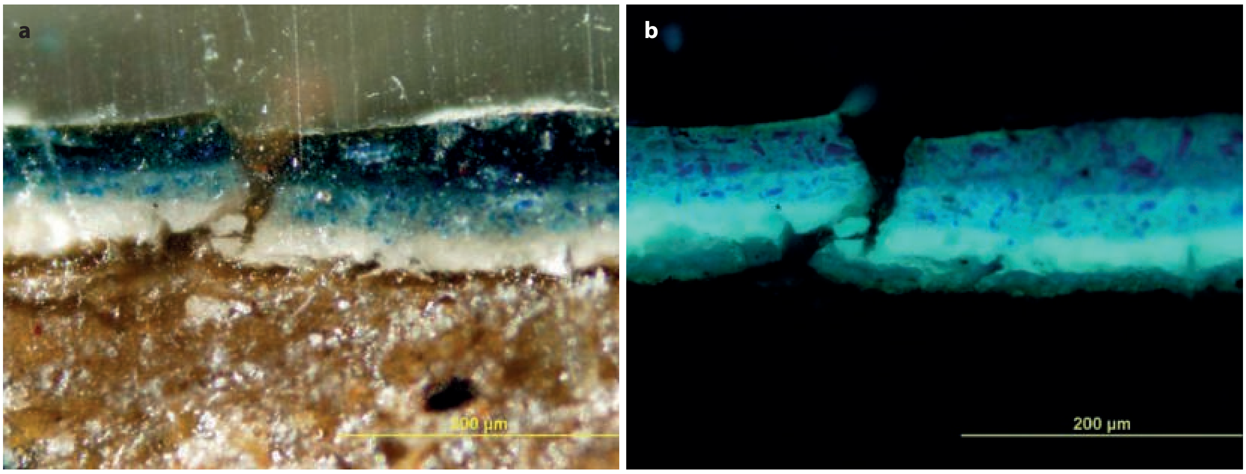


Figure 2.31 (a) PLM photomicrograph and (b) UV fluorescence image of a cross-section of a blue area of a wall painting (BMM003) from Cave N(a). Two layers of different shades of blue were applied (a), however, the amount of blue lazurite particles in both layers is comparable (b).

binding media in the painting layer is drying oil. This is the first time that horse collagen has been found in Bamiyan. From BMM063 (Cave B(d)), animal glue was also identified from a red paint layer and white ground by GC-MS. In previous studies, cow skin glue was detected from surface polychromies of the Eastern and Western Giant Buddhas (Kawahara *et al.* 2013). From BMM082 (Cave F(c)), animal glue and casein were both identified by ELISA. As drying oils were detected in both paint layers, it is reasonable to assume that proteins were also used in other layers for such cases.

For the plant gums, 9 samples were identified: Bamiyan Caves B(d), F(c), J(c), N(a) and S(a), Foladi Cave 4 and Kakrak 43 (Appendix III, Table A.III.34). Galactose was identified from the sample of Cave M. Some other sugars were also detected from C(a), E(c), the East Displaced Cave, M and the surface deposit of walls in Cave L, but these are not plant gums. Caves with plant gums somewhat overlap with caves with drying oils.

2.2.9 The glazes

One group of Bamiyan's wall paintings evidence a special painting technique. According to naked-eye observations, a transparent, deep colour can be seen in wall paintings in Caves N(a), B(d) and F(c). The cross-section reveals a thick layer of transparent organic material on the surface. This thin layer of a coloured, transparent finish consists of a small amount of pigment and a large quantity of resinous organic material. For example, in the case of one wall painting in Foladi Cave 6, where the surface has become rough or discoloured likely due to exposure to the external environment, the naked eye cannot see this transparent layer. However, the cross-section

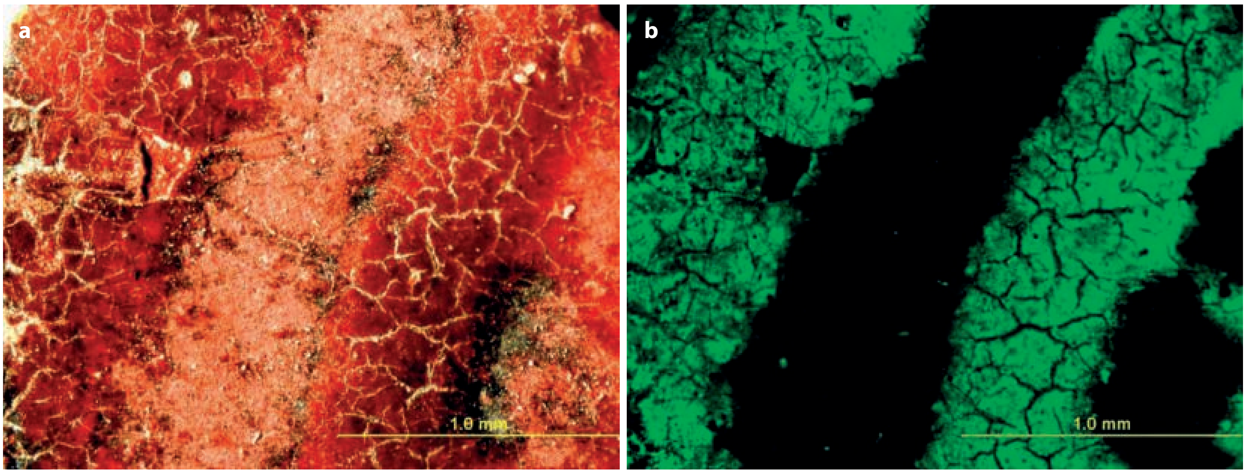


Figure 2.32 (a) Stereomicrograph of a red portion (BMM002) in Cave N(a). The robe is represented by transparent red lines. (b) Ultraviolet fluorescence image: only the transparent red lines show strong UV fluorescence.

reveals a pigmented layer with a transparent green colour on the inner surface (Fig. 2.28). Painting techniques in post-medieval Europe included applying transparent glazes over a painting to create an opaque layer that gives other colours the effect of 'depth' (Nicolaus 1979 [1985]: 68). These glazes were made by diluting paint with larger amounts of lakes, organic substances or binding media.

The transparent organic layers found in Bamiyan and analysed in this study are green, red, blue and yellow (Figs 2.29–2.32). Basically, the transparent organic layer and the opaque layer underneath do not differ in the type or amount of pigment, but in the higher amount of drying oil and the presence of resin in the upper organic transparent layer (BMM035 green part) (Appendix III, Fig. A.III.18). There are also examples of the use of transparent glazes as decoration in localised areas. For example, the folds of the Buddha's garments in Cave N(a) have been coloured to produce a three-dimensional effect with transparent red lines (Fig. 2.32). This particular area has been coated with a substance that fluoresces under UV light. This, like the glaze described above, is thought to be the result of the use of a paint with a high amount of drying oil and resin.

The wall paintings with glazes observed here comprise a unique set of wall paintings with an oil-lead white ground, in which drying oil was used as a binding media. To date, there have been no reports of such a glaze on Central Asian wall paintings. The Greek and Roman frescoes and the Sogdiana wall paintings have rather simple colour palettes, which seem to be only distantly related to the use of glazes to create graduated levels of colour depth. This painting technique is strongly reminiscent of the glossy finishes and glazes found on medieval European panel paintings.

Figure 2.33 Seated Buddha on the ceiling of Cave F(c): the robe has a darkened grid pattern and losses. Photo: Y. Taniguchi, courtesy of NRICPT, 2006.

2.2.10 Metal leaf decoration and the 'pseudo' gold leaf technique

2.2.10.1 Gold and tin foils

Other than glazing, some of the wall paintings at Bamiyan are decorated with metal leaf. A discussion of the recesses caused by subsequent scraping of the gold leaf is given in section 2.2.10.2. During restoration work in the field, metallic lustre was identified in several cave paintings and in some of the recovered fragments of wall paintings, two types were observed by the naked eye. The first is a golden substance found on the Buddha's votive offerings and on the white halo of his forehead in Caves N(a) and F(c). The gold leaf seems to have been cut in straight lines and applied using a technique known in Japanese as *kirikane*. For example, in wall paintings at Qol-e Jalal and Caves F(c), N(a) and S(a), gold leaf stretched

Figure 2.34 Standing Buddha on the east wall of Foladi Cave 5: the lattice-like patterns in the robe is visible. Photo: Y. Taniguchi, courtesy of NRICPT, 2006.

very thin (about 1 μm) is cut into lines and triangles and pasted to the image to create decorative patterns (Iwai 2006a). Gold leaf was applied to the wall paintings at Qol-e Jalal using drying oil as a mordant (QJM06) (Appendix III, Figs A.III.117 and A.III.119).

At present, there are very few examples of gold leaf in Bamiyan. On the ceiling of Cave F(c), the gilded portion of lattice patterning on the main deity of the three Buddhas, which represents gold embroidery on his robe, is evenly depressed possibly as a result of gold leaf being scraped off the earthen walls (Figs 2.33 and 2.34) (Miyaji 2002: 166). Similar shallow depressions and remnants of gold leaf can be seen very clearly in the Kizil grottoes of 'Tradition B' (Hiyama 2021, 2022; Taniguchi 2022: 437), such as those in Caves 38, 171 and 224, which flourished between the mid-6th century and the 7th century. Silvery metal leaf has also been observed in a number of places, such as in body nimbi along the north wall of Cave N(a), on the ceiling of Cave S(a) at the base of foliated scroll patterns, in lattice patterning on robes, and in spaces between the seated Buddhas (Fig. 2.35).

The use of tin foil has been confirmed by non-invasive analysis using XRF and SEM-EDS (Taniguchi *et al.* 2007). The tin foil is approximately 20 μm thick and contains some lead (e.g. BMM186) (Appendix III, Fig. A.III.74), probably due to the fact that lead in the tin could not be removed using the tin refining technology of the time. The techniques involved in applying the foil – from preparation of the ground to the many finishing touches – required a great deal of ingenuity to achieve an effective gold or silver appearance (Otake 2003, 2004). For example, the gold foil is rendered so thin that the colour of the ground underneath shows through it therefore a realistic gold colour can be obtained by applying a red colour beneath the foil. A coloured varnish can also be applied to the surface of the foil, both as a protective layer and to enhance its golden 'expensive' appearance. An adhesive is required to attach the foil and in Kizil Cave 171, a reddish-purple lac resin was used as an adhesive (mordant) for gold leaf, and drying oil for tin leaf. Red lac resin is an alkali-extracted reddish substrate which also has a transparent glazing effect. It has been identified in wall paintings in Kizil Caves 171 (Zhou *et al.* 2020a,b, 2021), 224 (Taniguchi 2022: 292) and Ajanta Cave 2 (Shimadzu 2021) as a red colorant. In the Translator's Temple and the Upper Temple of Nako (12th century) in the Western Himalayas, laccaic acids A and D, and erythrolaccin were identified by high-performance liquid chromatography (HPLC) (Bayerová 2018). Since erythrolaccin is a non-water-soluble molecule, the colorant could be extracted using alkali as lac resin (Kirby 2008).

Glossy finishes were also applied to metal leaf in Europe in the Middle Ages. At the time, a yellow or red varnish called *auripetrum*,[13] coloured with saffron, aloes, the inner bark of black plum or dragon's blood, was added to tin and silver foils to imitate gold leaf. The substances were dissolved in melted pine balsam, which could then be diluted with boiled oil and turps (Laurie 1910: 203–204). The '*mecca*' technique, for example, found in medieval tempera paintings, is a similar technique (Kii 2006: 94, 107–108). This coloured transparent varnish is fundamentally identical to the glaze mentioned above. Different materials and techniques were used for various parts of the wall paintings in the same cave; for instance, real gold leaf was used in the Buddha's garments, while tin foil with golden varnish was used elsewhere to emulate the appearance of gold.

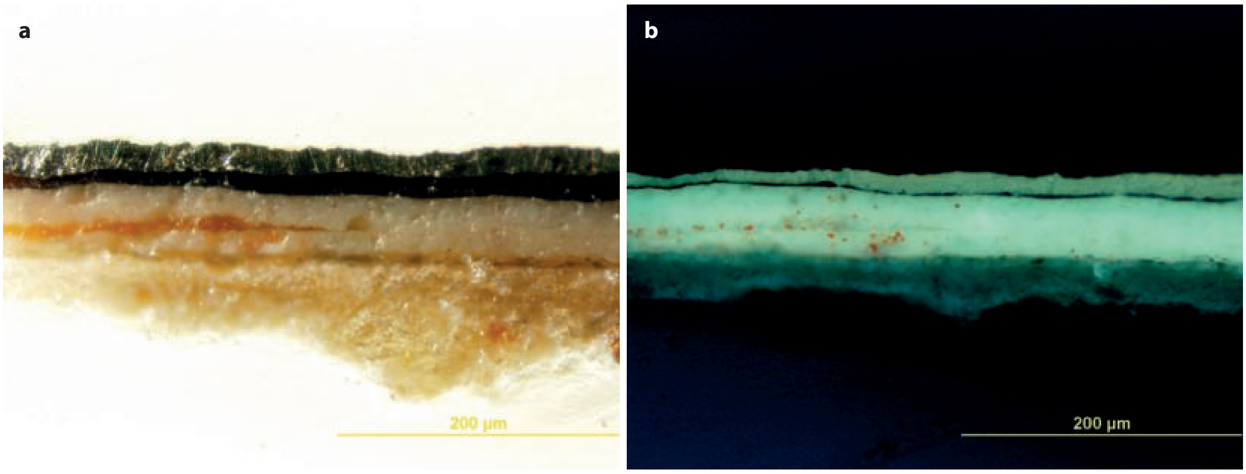


Figure 2.35 (a) PLM photomicrograph of a cross-section of a piece of tin foil in a Thousand Buddhas wall painting (BMM165) in Cave S(a) and (b) UV fluorescence image. A dark reddish-brown layer of mordant can be seen beneath the thick, silvery tin foil.

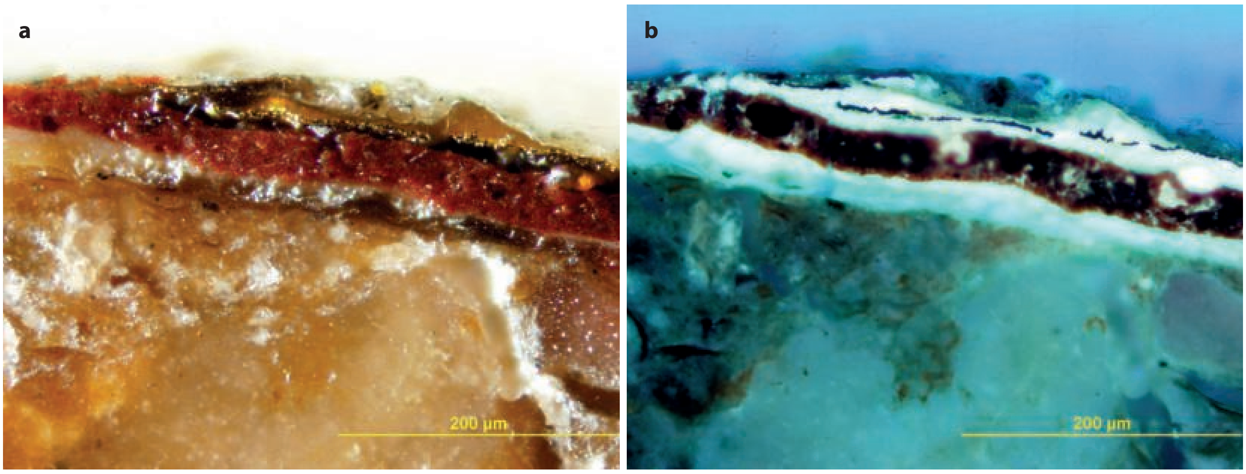


Figure 2.36 (a) PLM photomicrograph of a cross-section of a gold leaf decoration on a wall painting from Qol-e Jalal (QJM007). A dark reddish-brown layer of mordant can be seen beneath the thin gold leaf, and a yellow transparent layer is seen above the gold leaf. (b) In the UV fluorescence image both the dark reddish-brown and yellow transparent layers are fluorescing yellowish-white.

Glazes were used not only on tin and silver leaf, but also on gold leaf to enhance the gold colouring. These techniques are also found in medieval Europe, such as in panel paintings from the 12th century onwards (Nicolaus 1979 [1985]: 123). Examples of this technique can also be found in Central Asia dating as far back as the 12th century, as seen in the cross-section of the cut-gold work (QJM007) in a wall painting at Qol-e Jalal (Fig. 2.36). The technique of applying a gold varnish over tin foil to create a 'pseudo' gold leaf can be seen in animal figures on the ceiling beam of Cave N(a). A cross-section of this area revealed that it was in fact a silvery metal foil coated with some kind of yellow transparent glaze which fluoresces (BMM186) (Appendix III, Fig. A.III.73). Underneath the metal foil there was also a fluorescent layer thought to be the glue or mordant used to attach the foil. Analysis revealed that the wall painting was executed using black lines on a tin-based metal leaf, with animals and foliated scroll patterns, and a background painted in deep red (see Chapter 3, section 3.1). Some transparent yellow resin was found on the silver-coloured metal foils that formed the animal bodies and foliated scroll patterns, which may have given the patterns and figures a golden glow (Taniguchi *et al.* 2007).

Historical references to the formulation of such a golden varnish can be found in the *De coloribus et artibus romanorum* by Eraclius (late 13th century). In a section on the production of golden varnish, called *auripetrum*, he describes, 'the process of heating a mixture of "linseed oil, (*vesprum*) bark, myrrh,[14] aloes, sandalac or amber"' (Merrifield 1967a: 220). In Petrus de S. Audemaro's *Method of Making Pigments for Painters and Manuscript Decorators* (13th–14th century), 'saffron and glue or varnish' is mentioned as a material for making golden varnish (Merrifield 1967a: 158). Oleoresinous varnishes on metal leaf were described as a solution of sandarac, amber or mastic in heated oil, which had already been mentioned by the monk Theophilus Presbyter in c.1100 (Nicolaus 1979 [1985]: 314). It is further explained as a coloured varnish using linseed oil, copal resin, pine resin, frankincense, cherry gum and painting agents such as camphor, sandalwood and madder-root lake (Nicolaus 1979 [1985]: 124). Myriam Serck-Dewaide (1991) pointed out that large areas of gold or silver leaf were only found on sculptures for a few decades in the 12th century. Extensive use of red and green glazes over silver emerged in the mid-13th century (Plahter 2004: 195–198). Only a few of Bamiyan's wall paintings exhibit metal leaf decoration, such as gold leaf and tin leaf, or the 'pseudo' gold gilding technique with tin leaf, as described here: a group of wall paintings in which lead white, drying oil and glaze have been identified but the relationship between them is unclear.

In summary, there are two main trends pertaining to the techniques used in Bamiyan's wall paintings: the first is the use of gypsum as a white ground and a water-based adhesive, and the second is the use of lead white as a white ground and an oil-based binding medium. The latter group of wall paintings shows a pronounced layering with the occasional use of glazes and metal leaf. This group of wall paintings, which suddenly appeared in Bamiyan from the mid-7th century onwards, is very different in terms of technique and materials from wall paintings produced after Bamiyan became established as a Buddhist capital.

2.2.10.2 Loss of metal foils

When observing wall paintings, loss was apparent in certain areas. Figure 2.34 shows a standing Buddha painted on the east wall of Foladi Cave 5. All the Buddha's garments have a lattice-like pattern, but the lattices appear to have been engraved into the rendering. A similar example can be seen in Cave F(c): the garments of the seated Buddha on the ceiling seem to be decorated with a pattern of red and black alternating lattices (Fig. 2.33). Closer examination reveals that the black lattice comes from a metal foil, like tin foil, that has blackened where once it had been cut and pasted onto the surface. In addition, the recessed areas were probably covered originally with square-cut gold leaf, as a few pieces of gold-coloured metal foil are still visible nearby. A small amount of gold gilding was also observed around the lattice-like recesses of Foladi Cave 5. It is more than likely that a number of the wall paintings in Bamiyan were once decorated with gold leaf but many were looted in later times. The use of gold leaf in decoration is also common in wall paintings in the Kizil grottoes – again there are many traces of gold leaf that was deliberately removed.

In Kizil, gold leaf was used in Style B wall paintings (also known traditionally as the Second Indo-Iranian style) of the central pillar caves, such as Caves 38, 171 and 224, classified as 'Tradition B' (Vignato and Hiyama 2022; Hiyama 2021, 2022; Taniguchi 2022: 347). The gold leaf decoration is not seen in Style A wall paintings (or the First Indo-Iranian style) of 'Tradition A' caves that flourished in Kucha before being replaced by 'Tradition B'. At least some parts of the Style B paintings contain several motifs that can be associated with the historical occupation of Kucha by the West Turkic Khaganate in the mid-6th–mid-7th century (Hiyama 2015, 2021).

Green and yellow colours, both of which were used in Bamiyan, easily discolour or undergo change and there is also evidence that gold leaf was applied, as seen in the lattice patterning of the Buddha's garments, although little remains today. This confirms that the current state of these compositions differs greatly from their original colours and brilliance.

2.2.11 Optical effects of a multilayered structure

In order to produce different shades of colour, several pigments can be mixed (i.e. blended together) or painted on top of one another. Observation of cross-sections has revealed that most of Bamiyan's wall paintings comprise a multilayered structure in which numerous layers of pigment were applied to create vivid colours (Taniguchi 2006). In particular, many wall paintings with lead white grounds tend to have a variety of colour combinations and a well-recognised multilayered structure. As seen in section 2.2.1.2, the unfinished wall paintings in the front room of Cave C(a), for example, evidence a systematic process of painting with ordered layers of colour.

Multiple layers were involved in the composition, preparation and application of a variety of blue hues. For example, Cave N(a) (BMM001) (Appendix III, Table A.III.1) has a bright light blue, while the East Displaced Cave (BMM155) has a deep blue produced from a highly technical, traditional technique (Taniguchi *et al.* 2006). The blue used in each consists of a pigment made from the lapis lazuli mineral, while that found in Cave N(a) was produced by crushing lapis lazuli into small pieces and separating

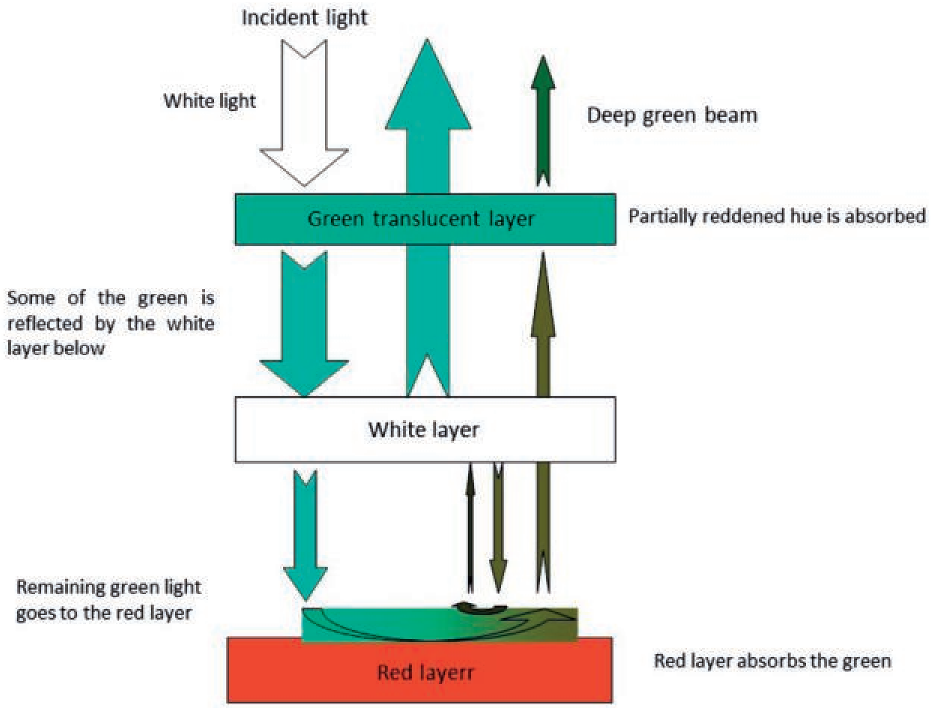


Figure 2.37 Optical mechanisms in the multilayered structure of the green portion, coloured with red, white and green.

Figure 2.38 Photograph and photomicrograph of the *urushi* lacquer coating on a red earplug from the late Jomon period (2540–1270 BC) (after Nagashima 2006). Multiple layers of cinnabar with different grain sizes.

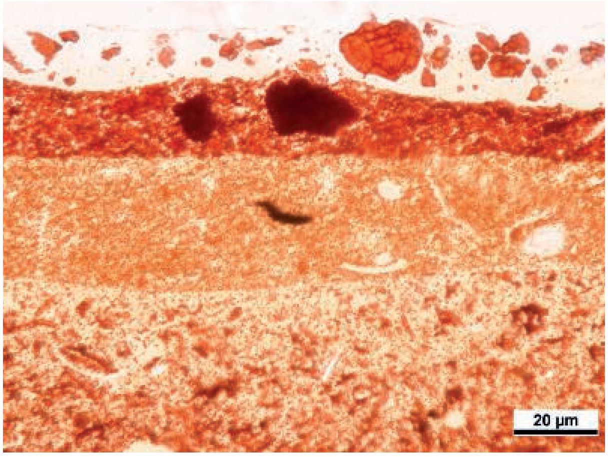


Figure 2.39 Photomicrograph of a thin section of a wooden comb from the Juno site in Saitama, Japan, mid–late Jomon period (1950–1470 BC). The multilayered structure is comprised of a transparent *urushi* lacquer, red *urushi* (micro pipe-shaped iron oxide), red *urushi* (fine cinnabar), red *urushi* (fine and coarse cinnabar) and transparent *urushi* (coarse cinnabar) layers from the bottom to the surface (Naruse *et al.* 1994).

only the blue lazurite. Lazurite is an important component of the natural ultramarine pigment: it was found mixed with lead white and applied on top of a lead white ground (Fig. 2.23). In the East Displaced Cave, a black colour containing lampblack was applied first followed by a layer of a blue pigment made from crushed and unrefined lapis lazuli still containing calcite and other impurities (Fig. 2.24). A deep blue colour is then produced by placing a black layer under the blue. This technique of mixing purified lapis lazuli with white pigment to produce a bright blue colour, or applying black underneath blue hues, can also be found in medieval European frescoes (Figs 2.25 and 2.26).

Regarding red pigments, the bright red garments (BMM081) in the wall painting of Cave K$_3$ are made of cinnabar/vermilion over orange minium (Appendix III, Table A.III.10). Many wall paintings use overpainting with minium and cinnabar/vermilion, as seen in Caves E(e), N(a) and Foladi Cave 4. Historically, compared to red lead, cinnabar/vermilion was probably expensive and difficult to obtain due to its rarity (Eastaugh *et al.* 2004: 387), and it is likely that this two-layer system was a way of obtaining a small quantity of an effectively saturated red

Figure 2.40 The Thousand Buddhas image in Cave K_3. Note: one part of the green body nimbus has turned black. Photo: H. Otake, courtesy of NRICPT, 2005.

colour. A colorimetric examination confirmed that a more yellowed, saturated colour could be obtained by applying an overlay of the two colours of red lead and cinnabar/vermilion relative to the red colour of cinnabar/vermilion alone (Seki *et al.* 2006).

A similar multilayer structure was observed in the green parts of Cave East III (BMM068) (Appendix III, Fig. A.III.34) and Foladi Cave 4 (FDM055) (Appendix III, Fig. A.III.93), where the combination of successive red, white and green layers was used to create a clear, deep green. The principle of this method is that green and red are complementary colours, therefore as light in a certain wavelength range is reflected by the white layer in the middle of the spectrum, giving a green colour (1), some reaches the red layer and is reflected back as a deeper green (2 and 3) (Fig. 2.37). The combination of these two types of light produces a deep, vivid green, therefore it is unlikely that this three-layered structure and its order was accidental but rather a well-calculated and highly technical painting technique (Taniguchi *et al.* 2006). In fact, examples of these kinds of multilayered painting techniques date back to prehistoric times.

In Japan, for instance, cinnabar was used for red paint in the middle of the Jomon period (3500–2400 BC). At least one wooden earplug[15] has been found with layers of natural cinnabar, each mixed with different particle sizes, on top of a layer of lacquer mixed with iron oxide (Fig. 2.38) (Nagashima 2006). Another wooden piece found was coated with lacquer, followed by layers of red lacquer (pipe-shaped iron oxide + *urushi* lacquer), red lacquer (coarse-grained cinnabar + iron oxide + *urushi* lacquer) and finally red lacquer (coarse-grained cinnabar + *urushi* lacquer) (Fig. 2.39). In both cases, the red lacquers of different shades were applied in layers to produce vivid reds.

In the case of Bamiyan's wall paintings and the red lacquer of the late Jomon period, the techniques used incorporated a thorough understanding of both optical effects and economic considerations for the use of precious pigments. In particular, when several pigments with different specific gravities are mixed, the resulting paint tends to separate easily, resulting in uneven colours. This layering technique utilises the optical behaviour of pigments, as ordinarily light reflected from a surface is gloss and light reflected from an interior is usually responsible for the colour (Stearns 1953). The use of layers seems to indicate that the artisans of the time were well aware of both the optical and economic qualities of materials used.

2.3 Composition of surface degradation

2.3.1 Degradation/alteration of copper- and lead-based pigments

Bamiyan's wall paintings include numerous products of degradation or alteration, differentiating what is now seen from the original work. Some elements used in paintings are highly reactive chemically, such as copper and lead, and tend to undergo change in their chemical states (Coccato *et al.* 2017). Copper-based pigments frequently transform into copper oxalate (moolooite: $CuC_2O_4{\cdot}nH_2O$) owing to the biodegradation of an organic binder. In Bamiyan's wall paintings, there were cases of atacamite (a copper chloride used as a green pigment) turning into copper oxalate. As a pigment, atacamite is not stable in acidic conditions, i.e. when oxalic acid is produced by biodegradation resulting in the formation of mooloite (Castro *et al.* 2008). The blackening of copper pigments is often observed in Bamiyan. Copper resinate was employed as a greenish glaze and is highly chemically reactive in certain environments. It is prepared by mixing copper acetates (verdigris) with natural resins. When the copper resinate comes into contact with a proteinaceous binder, metalloproteins may be formed if an egg tempera was used (Cartechini *et al.* 2010). The copper resinate is known to darken with light exposure (Franceschi *et al.* 2011) as seen in most cases in Bamiyan.

Very few intact green pigments were observed at Bamiyan. For example, only black, white and pale red can be seen in the Laternendecke ceiling of Foladi Cave 6 (Chapter 1, Figs 1.31 and 1.32). The body nimbus of the seated Buddhas and the spaces between the Thousand Buddhas are black. A similar black colour can be seen in the wall painting on the vaulted ceiling of Cave K_3 (Fig. 2.40). In this case, only the right side of the seated Buddha's body nimbus is black, while the rest remains green. This green colour is thought to result from a change of the copper-based green pigment to a black product, possibly triggered by environmental factors.

Lead pigments can also be highly reactive: products resulting from such changes include oxide, sulfates and chlorides. Frequently used lead pigments include: lead(II) oxide, litharge, massicot (PbO), lead(IV) oxide (PbO_2), plattnerite (PbO_2), lead(II) and lead(IV) oxide and minium ($2PbO.PbO_2$; historically Pb_3O_4) (Eastaugh *et al.* 2004: 228). Lead white as a pigment refers to a wide variety of white lead-based materials, as well as basic lead carbonate such as hydrocerussite and cerussite (Eastaugh *et al.* 2004: 233–235; Gettens and Stout 1967).

The blackening of lead white and minium is attributed to the formation of either plattnerite, following exposure to oxidising agents or microbiological activity, or galena (PbS) (Saunders 2000; Gettens *et al.* 1993: 72; Petushkova and Lyalikova 1986; Rosado *et al.* 2015) from atmospheric conditions such as sulfuric environments, acidic solutions (rainwater, carbon dioxide, microbial activity), and light. Anglesite ($PbSO_4$), cerussite ($PbCO_3$), hydrocerussite and galena then form as a result (Pérez-Rodríguez *et al.* 1998). Anglesite identified on wall paintings (Barone *et al.* 2016) reportedly blackened in glue tempera paintings exposed to light and humidity (Gettens and Stout 1966: 153; Saunders *et al.* 2004). The parameters responsible for the blackening of minium are identified as light (Gettens and Stout 1967), the pigment's composition and climate. Plattnerite might form when biological colonisation is present (Aze *et al.* 2008). Laurionite ($PbCl(OH)$), anglesite and hydrocerussite were identified in darkened areas in minium-painted shrines (Kuchitsu 1997). Laurionite is water-soluble, while anglesite and hydrocerussite are insoluble in water. The darkening occurs as laurionite and anglesite's white crystals form in the presence of saline water, and then collect dust particles. The presence of soluble salts in the wall paintings may provoke the alteration of lead pigments into laurionite.

Figure 2.41 Standing Buddha image on the west wall of the niche at Cave I. One part of the painting (arrow) is missing, exposing the earthen render. Photo: Y. Taniguchi, courtesy of NRICPT, 2005.

Palmierite has already been reported as a degradation product of lead pigments, notably in old master paintings (Van Loon *et al.* 2017). It is often associated with degraded smalt, lakes or ultramarine in combination with lead white (Van Loon *et al.* 2011). Cotunnite ($PbCl_2$), a colourless crystal, has also been identified from degraded lead pigments on painted statues in the Sacred Mount of Varallo (Vercelli, northern Italy, 16th–17th century) (Possenti *et al.* 2021). However, it is important to note that various artificial lead-based colorants were used in the past – namely, laurionite, phosgenite ($Pb_2Cl_2CO_3$) (Walter *et al.* 1999), lead sulfate ($PbSO_4$), lead chloride ($PbCl_2$) and blixite ($Pb_8O_5(OH)_2Cl_4$) (Naruse 2004) – which complicates our ability to interpret whether they have resulted from chemical reactions or were part of the original manmade lead-based colorants.

Regarding red colours, in some cases, including in the red parts of the Buddha's niche in Cave I (BMM009) (Appendix III, Figs A.III.4 and A.III.5), wall paintings that have been exposed to the open air and lack oil-based binding media exhibit red colours that have turned slightly purplish. HgS discoloration has been studied in other paintings, such as in Pompeii (Cotte *et al.* 2006a), the Pedralbes Monastery in Barcelona (Cotte *et al.* 2008) and in works by Rubens (Radepont *et al.* 2011). The mechanisms of discoloration are complex and manifold

(Radepont *et al.* 2015). In the presence of chlorine ions, and under the effects of light, the sulfide from HgS is progressively substituted by chlorides, forming different white/grey/purple compounds such as $Hg_3S_2Cl_2$ and Hg_2Cl_2 (calomel). The decomposition of HgS also leads to the formation of black metallic mercury (Anaf *et al.* 2013) and of a sulfate that, in the presence of calcium, can lead to the formation of gypsum.

In addition, the green pigment, verdigris – whether copper acetate or organic copper salts such as copper resinate – has a chemically unstable structure and, as in the case of Li Xun, has been known to easily change in colour. Organic salts of copper are incompatible with numerous pigments (typically gypsum, lapis lazuli, realgar and orpiment), including those containing sulfur, for example, and may turn black when mixed. Light and UV radiation can also cause blackening (Kühn 1993). Recently, a peroxo-Cu^{II} dimer was identified by electron paramagnetic resonance (EPR) and optical absorption spectroscopy (OAS) when model films of paints prepared by mixing copper acetate and copper resinate pigments in linseed oil were photo-aged (Alter *et al.* 2019).

2.3.1.1 Yellow degradation/alteration

Because Bamiyan's Buddhist wall paintings are religious, it is likely that yellow was used to represent light, as seen in the head and body nimbus of seated and standing Buddhas. However, as Figure 2.41 shows, some of the nimbi, which would have been yellow, have been lost along with the white ground (indicated by the green arrow). In addition to the blue, red and green bands in the body nimbus, yellow was also used and may have been coloured with orpiment, which produces a golden colour that has now become black. As a yellow pigment, orpiment is known to be easily degraded by light into arsenic oxide species (Dubois *et al.* 2001; Coccato *et al.* 2017; Keune *et al.* 2015).

Notes

1. Caves were selected from the archaeological report by Kyoto University (Higuchi 1983–1984), based on descriptions such as 'wall painting' and 'painting'. Their locations are illustrated elsewhere (NRICP 2004).
2. Extracted from *Amyris agallochum*.
3. *Aegle marmelos*, modern name: bel.
4. Śāla, a species of tree from the Dipterocarpaceae family. The resin from its trunk is called *sarjarasa* in Sanskrit, and is said to have been translated into Chinese as *bái jiāo xiāng* (Feng Xiang). It appears in the Chinese *Materia medica* as *Liquidambar.*
5. Thompson 1960: 50, ch. LXXII: 'the way to paint on a wall in *secco*; and the *tempera*s for it'.
6. Thompson 1960: 57–58, ch. LXXXX: 'how you should start working in oil on a wall'.
7. Swim bladder.
8. Solubility in water (20 °C): $CaSO_4.2H_2O$ (0.205 wt%), $CaSO_4.1/2H_2O$ (0.298 wt%), $CaSO_4$ (0.72 wt%) (Chemical Society of Japan 1975: 779).
9. In Japan today, a white substance (calcium carbonate) made from the crushed shells of oysters and clams is often referred to as *gofun,* but it originally meant lead white (lead carbonate).
10. In the late Tang dynasty, in Sichuan's Azusa state, Venusense sold fragrant medicines or drugs imported from abroad. Because of his Persian grandfather, Li Xun's family was well acquainted with Western medicines of the time and introduced knowledge of them to China, such as by compiling the *Oversea Materia Medica* with explanations (Zhang 2013).
11. See note 10 above.
12. Binders, binding media and vehicles are a combination of agglutinants with colour spreaders.
13. Also known as Peter's gold.
14. This refers to the resin secreted by trees of the genus *Conmiphora* (Myrtaceae) (Laufer 1967: 460–464) and collected from the Arabian Gulf and Indian Ocean coasts. Pliny and Dioscorides describe seven and two species, respectively (Otsuki 2009a).
15. Excavated at the Kita-Ekoda site in Nakano-ku, Tokyo dating back to the late Jomon period (2500–1300 BC).

Chapter 3: Oil painting techniques in Bamiyan's wall paintings

3.1 Case study 1: The foliated scroll design with animals on the ceiling of Cave N(a)

Cave N(a) has a square-vaulted Laternendecke ceiling whose design appears to hold two smaller squares, each rotated 90 degrees from the other, within a broader square enclosure (Fig. 3.1). The inner side of the middle square is decorated with a band of foliated scroll designs, approximately 6 cm wide and 40–50 cm long. The inner side of the central square is decorated with a row of four bands of six birds (24 birds in total), each with a ribbon in its mouth. Ceiling beams were initially covered with black soot that obscured the foliated scroll design. However, cleaning during conservation work revealed the animal and foliated scroll designs against a red background and a small amount of gold-coloured material on the animals (Otake *et al.* 2007).

The centre of each foliated scroll features a pair of animal figures that naturally merge into it. Originally, there may have been 12 pairs (24 animals), but about half of the figures have been damaged or lost. The animals and foliated scroll-creeper include a bull and a griffin on the northwest underside of the second square, a boar and a lion on the northeast underside, an unidentified animal and a dog on the southwest underside, and a monkey and a half-man on the northeast underside (Figs 3.2–3.4). If the figure depicted here is the upper half of a man or a half-human hybrid such as a triton or centaur is unclear due to damage (Yamauchi 2006).

A similar design fusing arabesques with animal figures on a red background found at Ajanta Cave 17 in India (Fig. 3.5) is thought to have originated in art of the Gupta dynasty (Koezuka and Miyaji 1999). Observation with the naked eye shows that the entire strip is covered with a silver-coloured metal leaf on which foliated scrolls and animals are drawn in thin black lines. The background behind the arabesques and animals is coloured with red paint, while outlines of the animals and arabesques are left unpainted. The surfaces of the corners of the beams are also uneven, as if the foil (metal leaf) had been creased. We conducted a chemical analysis using SEM-EDS, SR-μXRF, SR-μXRD and SR-μFTIR to identify the materials.

Figure 3.1 The ceiling of Cave N(a). Photo: M. Momii, courtesy of NRICPT, 2007.

Figure 3.2 A boar and a lion. Photo: M. Momii, courtesy of NRICPT, 2007.

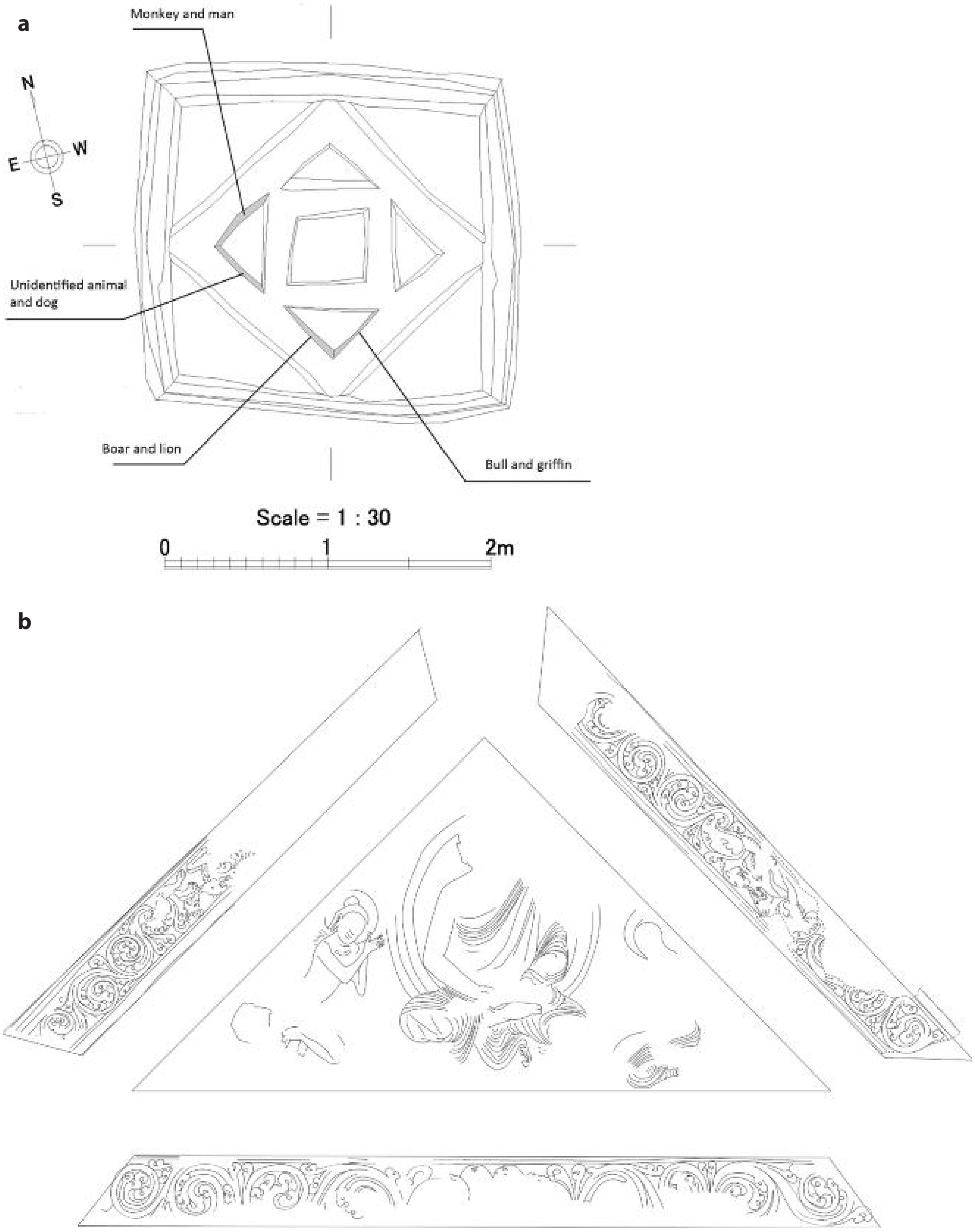


Figure 3.3 (a) The ceiling of Cave N(a) and locations of each pattern. (b) Line drawing of the patterns in a triangular section in the south end of the ceiling, Cave N(a).

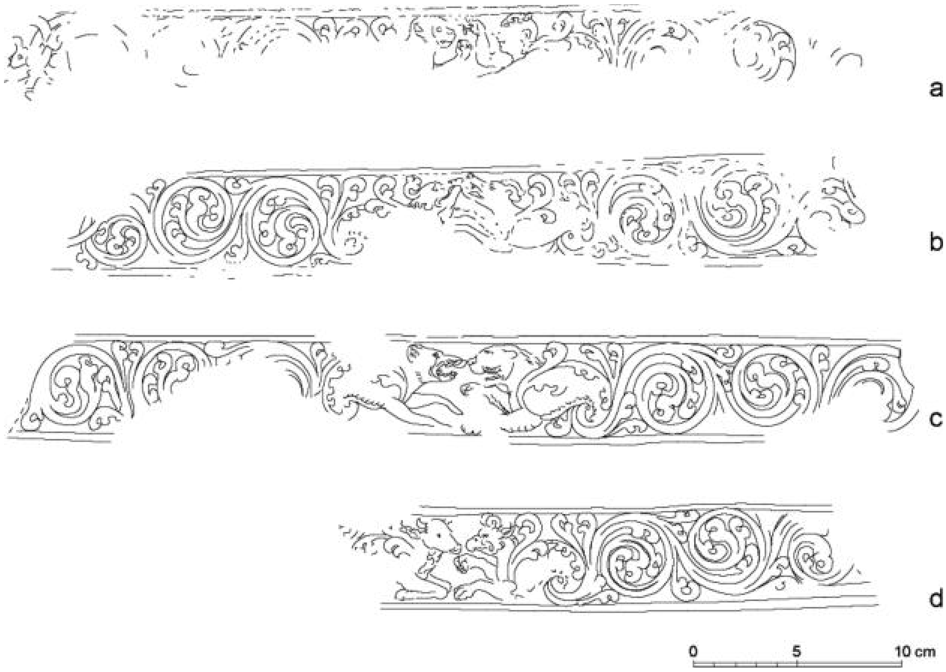


Figure 3.4 Line drawings of animal and foliated scroll patterns identified at Cave N(a). a: monkeys and figures; b: unidentified animals and dogs; c: boars and lions; d: bulls and griffins.

Figure 3.5 Animal and foliated scroll patterns on the ceiling of Ajanta Cave 17. Photo: H. Otake, courtesy of NRICPT, 2006.

3.1.1 Observation of animal and foliated scroll patterns in Cave N(a)

Samples taken from one patterned area exhibiting a golden lustre (BMM184) as well as from the boar (BMM186) were examined under a stereomicroscope. Normal light and UV fluorescence observations of cross-sections revealed that in fact the gold is not gold leaf but a transparent yellow varnish-like organic material applied over a silvery foil about 20 μm thick, which gives the patterns and figures their golden glow (BMM177: Appendix III, Fig. A.III. 69; BMM186: Appendix III, Fig. A.III.73). It was also observed that the silver-coloured foil had been applied over a white ground layer with a white fluorescent organic adhesive (mordant). A white fluorescent organic material and a thin organic material with a bluish-white fluorescence were also found between the white ground layer and the rendering layer. The ground layer itself emits white fluorescence.

3.1.2 Elemental and structural analysis of the lead white ground and metal foils using SEM-EDS, SR-μXRF and SR-μXRD

SEM-EDS was used to analyse a cross-section of BMM186 (Appendix III, Figs A.III.72–A.III.74) and a thin section of BMM177 was analysed using SR-μXRF/SR-μXRD. SEM-EDS detected lead in the underlying white ground layer while SR-μXRD revealed that the white layer contains hydrocerussite. In addition, susannite, a lead hydroxycarbonate sulfate, was also detected in the white layer. Tin and lead were identified by SEM-EDS in the silver-coloured

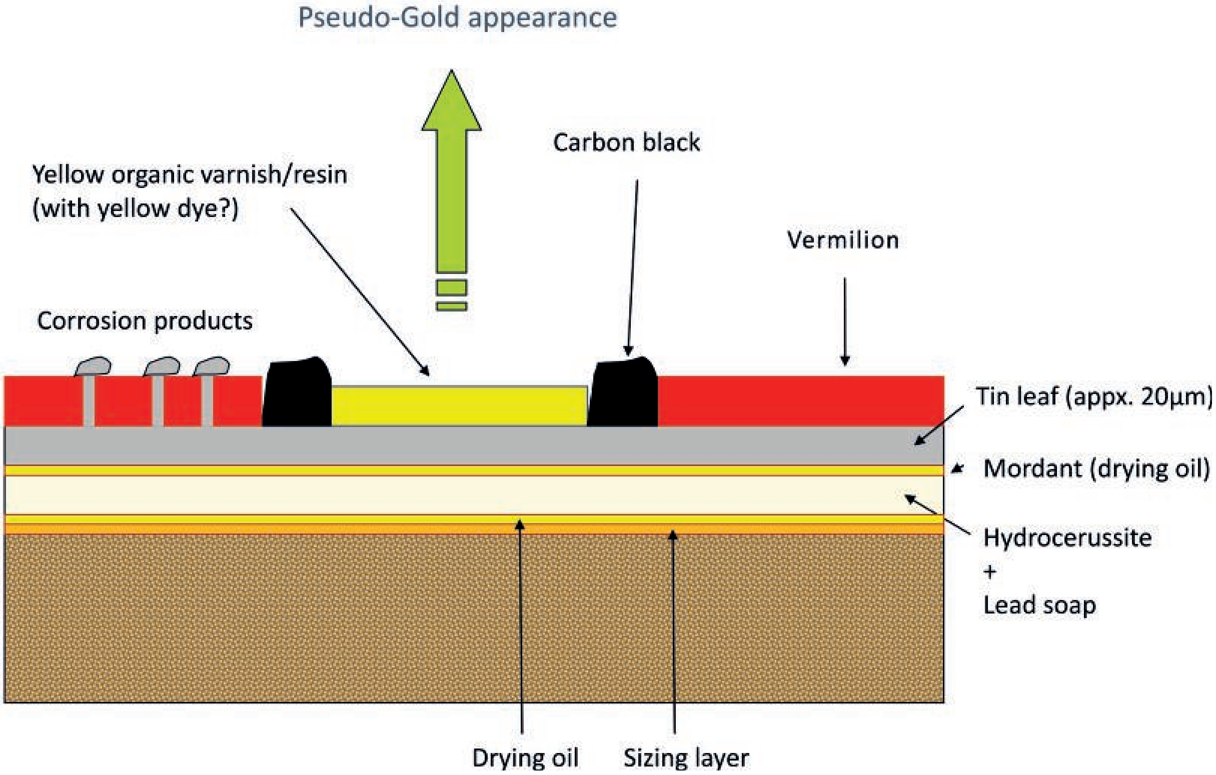


Figure 3.6 Schematic diagram of the technique for decorating with tin foil, as seen in wall paintings at Cave N(a).

metal foil, suggesting that it contained lead in parts. SR-μXRD on sample BMM177 confirmed the presence of metallic lead together with metallic tin. Altogether, these findings confirm that the tin-refining technology at the time was not yet capable of removing lead. A thin layer of calcium was detected on the surface of the yellow varnish-like layer of organic material, probably a deposit that formed after the transparent yellow layer was added to the foil.

3.1.3 Analysis of organic matter by SR-μFTIR and GC-MS

One portion of the boar sample (BMM178) was sliced with a microtome to obtain thicknesses ranging from 5 to 50 μm. A sample of a small amount of compacted powder from all the layers was also taken to preserve the layered structure. Both the microtome and compacted samples were subjected to SR-μFTIR point analysis and mapping by transmission. Analytical conditions were in accordance with Appendix III. The FTIR spectra data are shown in Appendix III, Figure A.III.71. The sample is composed of three main layers: a white ground layer, a translucent mordant and the metal foil. FTIR absorption intensity was integrated over six regions of interest, characteristic of the mordant (strong and sharp absorption ν(C-H) (a) and ν(C=O) (b), δ(C-H), ν(C-O) (c)) and of the white ground ((d) carbonate ν_3(C-O), (e) lead hydroxide ν(O-H)) and (f) lead carboxylate ν_{AS} (CO)). These maps show good agreement with the layers as seen in the optical microscopy image. Average FTIR spectra were calculated over each layer. The signal in the mordant layer suggests that the material is composed mainly of oil. However, the possibility of a mixture of oil and resin cannot be ruled out. As this mordant forms a thick layer, it is reasonable to assume that the oil mainly comprises a drying oil. Analysis of fatty acids by GC-MS (BMM184-1: Appendix III, Table A.III.1) also detected a small amount of palmitic acid and azelaic acid, giving results favourable to the possible presence of a drying oil.

Analysis of the white ground layer (B) revealed that, in addition to the oil peaks, there is absorption (d) characteristic of lead white (d) and (e). A strong absorption (f) at 1520 cm^{-1} is also distributed in the same area, indicating the presence of lead carboxylates vas(C=O) groups or lead soap. Therefore, this white base layer is comprised of lead white and partially saponified oil.

In an additional sample (BMM184) the transparent yellow organic material on the surface of the tin foil is believed to be a yellow dye in natural resin.

However, this has not been clearly identified. The surface of the yellow varnish-like layer shows absorption characteristic of calcium oxalate at 1320 cm^{-1}, which is consistent with the detection of calcium by SEM-EDS. This calcium oxalate may be a common degradation product in paintings (Rampazzi 2019).

3.1.4 Layered structure of the animal and foliated scroll designs

In summary, the animal and foliated scroll designs on Cave N(a)'s ceiling beams are thought to have been produced in the following manner (Fig. 3.6). First, a sizing layer containing proteins was applied to the rendering wall, followed by a layer comprised mainly of drying oil, and then a white ground layer consisting of lead white (with an unusual composition: hydrocerussite, susannite and partially saponified drying oil). On top of these, a metallic tin foil was applied to the side of the beam using a substance consisting mostly of drying oil as a mordant. The tin foil has a thickness of 20 μm and contains metallic lead as an impurity.

A pattern was drawn in black lines on the pasted tin foil and the background filled in with a red pigment (vermilion). The remainder of the tin foil was covered with a yellowish (or yellow-coloured) natural resin, most likely to give the impression of gilding. This technique for creating a 'pseudo' gold leaf resembles the technique used in medieval European panel paintings.

Figure 3.7 The tree deity of Cave N(a) before destruction. Photo: K. Maeda, 1970s.

Figure 3.8 General view of the tree deity on the east wall of Cave N(a) (after destruction), with the area from which sample BMM035 was taken (yellow dot). Photo: H. Otake, courtesy of NRICPT, 2005.

3.2 Case study 2: Green colouring in the tree deity on the east wall of Cave N(a)

3.2.1 The tree deity image on the east wall of Cave N(a)

In the centre of the north side of the east wall in Cave N(a) is an image of what is probably a tree deity: it has a red ivy trunk and branches spreading out from its body, along with a row of heart-shaped leaves similar to those of a lime tree. At the time of a survey carried out by Nagoya University in the 1970s, the figure of the tree deity was clearly preserved (Fig. 3.7). However, most of it was damaged and lost during the civil war (1978–1992), leaving only a small portion remaining today (Fig. 3.8). The red trunk and branches spread out against a black background, and the leaves are a deep, shiny green colour. Between the leaves are small white petal-like structures. The entire image is finely painted with a thick coat of paint in which small strokes of the paintbrush are visible. Sample BMM035 was taken from a green leaf for analysis.

3.2.2 Description of the stratigraphy using optical microscopy

Observation of the sample's cross-section reveals a multilayered structure. The green area of the leaf has a transparent yellow layer, a white ground layer and a black organic layer on top of a clay render. In UV light, the transparent yellow layers emit blue-white fluorescence and the white ground layer a strong white fluorescence. A layer containing black organic matter in sample BMM035 also emits UV fluorescence and contains a large number of black particles (Appendix III, Fig. A.III.13).

3.2.3 SR-μXRF analysis of the sample

SR-μXRF mapping was performed under vacuum at the ID21 beamline and in air at the ID18F and the ID13 beamlines (Appendix III, Fig. A.III.14 and Fig. A.III.15). The μXRF map at ID21 detected calcium, potassium and chlorine in the yellow-glazed surface, while the green layer was found to contain mainly a matrix of lead and copper, together with particles of iron, silicon, aluminium, potassium and calcium; lead was detected in the black and white coloured layers. A higher concentration of lead is found in the white ground layer. The particles in the black layer could have been carbon black or a black substance made from burnt calcium phosphate such as ivory black. However, since phosphorous has not been found in the black layer, it is unlikely to be ivory black or bone black, but rather a carbon black or lampblack.

3.2.4 SR-μXRD analysis of the sample

SR-μXRD diffraction patterns were collected at ID18F and ID13 simultaneous to the collection of μXRF spectra, over a 2D region, on a thin microtome section. Figure A.III.16 in Appendix III shows an example of the analysis of the green layer, with an average of about 300 diffraction patterns obtained per sample. Gypsum (calcium sulfate) and weddellite (calcium oxalate) are clearly identified in the calcium yellow varnish-like layer, both probably the products of degradation.

In the green layer, both hydrocerussite and cerussite, characteristic of lead white, were identified, as well as quartz. The identification of the copper compound was not easy. Chrysocolla, a green copper silicate, is thought to have been used as a green pigment: Gettens (1938a) detected it in the green pigment of the wall paintings in caves around the Eastern Giant Buddha in the 1920s. However, XRD analysis could not confirm its presence and Si and Cu do not show co-localisation. Atacamite, a basic copper chloride, malachite and azurite were not detected either, therefore it seems that Cu is mainly in an amorphous form. In summary, the green layer appears to be a lead white (hydrocerussite/cerussite) paint coloured by an unidentified green copper pigment. The white layer contains mainly hydrocerussite mixed with susannite. Interestingly, two different qualities of lead whites were used for the white and the green layers.

3.2.5 SR-μFTIR analysis of the green part of the tree deity image

Results of SR-μFTIR analysis of ID21 are detailed in Appendix III, Figure A.III.17. The analysis was conducted using the same methodology as outlined in section 3.1.3. In addition, statistical analyses (non-negative matrix approximation, or NNMA, of 7 components) were carried out using the PyMCA ROI imaging tool (Cotte *et al.* 2016) (Appendix III, Fig. A.III.18). The render layer (NNMA2) contains mostly inorganic compounds, such as kaolinite. The next brown transparent layer (NNMA0) is composed primarily of proteins (possibly animal glue or egg white with characteristic amide I, II and III bands) with some silicates and oxalates (peak at 1320cm^{-1}). Calcium oxalates were also detected in the brown layer at the surface of the green layer. As already mentioned, these are common degradation compounds in paintings.

The white ground layer is composed of lead white (small and sharp ν(OH) at 3537cm^{-1}, broad and intense $\nu_3(CO_3)$ at 1420 cm^{-1} and small and sharp ν_1 and $\nu_2(CO_3)$ at 1047 and 839 cm^{-1}) (Brooker *et al.* 1983), mixed with a partially saponified oil (ν(CH) series at 2927 and 2855 cm^{-1}, ν(CO ester) at 1740cm^{-1}, and ν(CO lead carboxylate) at 1544cm^{-1}). This component (NNMA1) is also present in the green layer. The signal of the black layer (NNMA4) was not clearly identified but it seems to indicate the presence of an oil, but with a higher content of acid (ν(CO) at 1734cm^{-1}), The green layer is a mixture of NNMA4 (lead white and saponified oil as in the white layer) and NNMA3 (oil partially saponified with copper soaps) (sharp and intense ν(CH) peaks at 2925 and 2854 cm^{-1} and ν(CO) (copper carboxylate) at 1585cm^{-1}).

3.2.6 Layered structure of the sample

Considering the above results from SR-μXRF, SR-μXRD and SR-μFTIR analyses, the green leaves on the north wall of Cave N(a) were likely produced

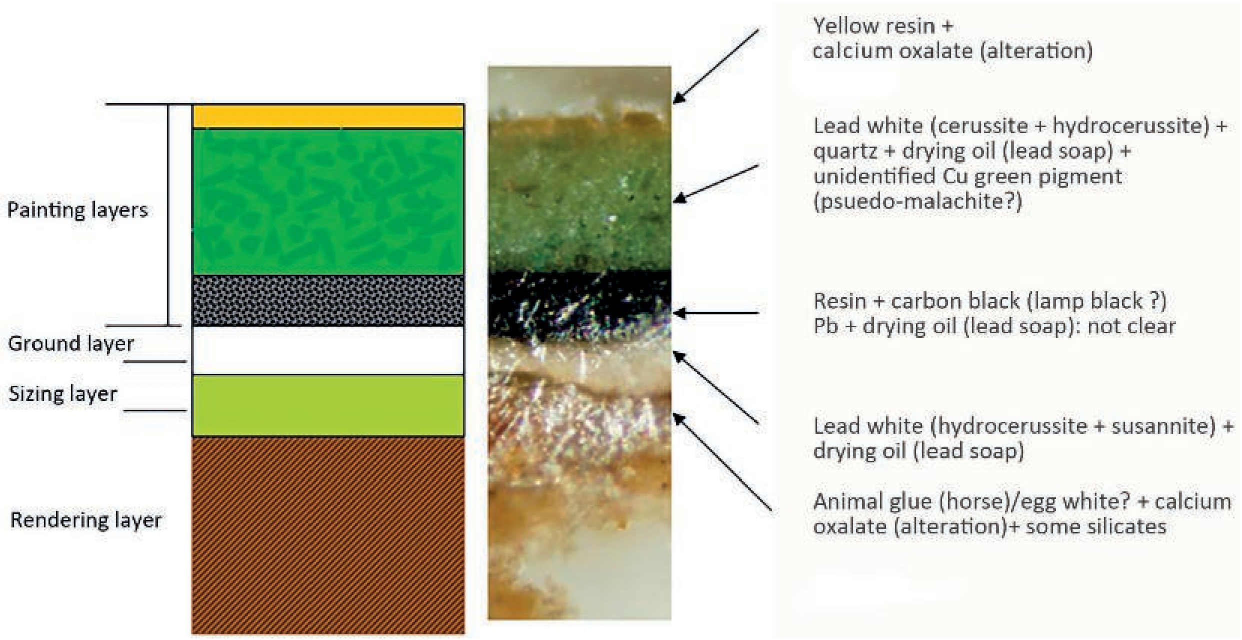


Figure 3.9 Schematic diagram of the layered structure of a green portion (BMM035) of the tree deity on the east wall of Cave N(a).

as follows (Fig. 3.9). Initially, the earthen render was coated with a protein-based material such as an animal glue milk or egg white, probably as a sizing to facilitate painting. This is supported by the amino acid detected in the GC-MS amino acid analysis (BMM035: Appendix III, Table A.III.33), although we could not identify any specific type of proteins. A white ground layer consisting of lead white (hydrocerussite and susannite) and partially saponified (lead soaps) drying oil was applied on top of the sizing layer. The black layer contains an unidentified organic component with a carbon-based pigment, probably lampblack. Lightweight pigments such as lampblack do not mix well with oil and are known to float. We suspect, therefore, that the resin was used to improve dispersion.

The green colour was produced through a complex formulation: it contains lead white (hydrocerussite and cerussite) and partially saponified drying oil. In addition to the lead soaps, SR-μFTIR analysis also detected copper carboxylates, which may indicate that copper mineral pigments were transformed into copper resinate or copper oleate by contact with organic substances such as resin or drying oil. The paint layer contains a small amount of copper mineral particles which have no clear phase structure, probably pseudo-malachite, which is visible as a green colour. A yellow ochre is also likely to be present in the green layer as analysis identified particles containing iron with a yellow colour.

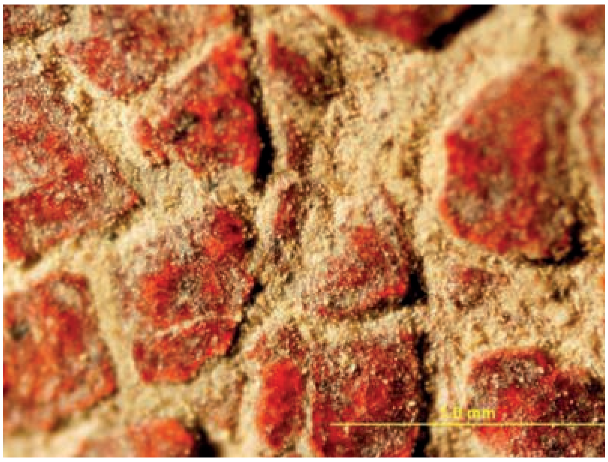


Figure 3.10 Stereomicrograph of a red portion (BMM063) of a wall painting in Cave B(d).

3.3 Case study 3: Binding media for wall paintings in Cave B(d)

3.3.1 Description of the sample BMM063

Sample BMM063 was taken from Cave B(d), one of the caves around the Eastern Giant Buddha. Radiocarbon dating indicates that the wall paintings in this cave were produced sometime around the late 7th or 8th century (AD 681–768) (Nakamura 2006: 121). The cave has a cross-vaulted ceiling with a distinctive ladder-shaped section. The central cross-vault is formed by a two-tiered Laternendecke ceiling with a dome in its centre, which is one of the most unique structures in the Bamiyan caves (Kubodera and Iwade 2006).

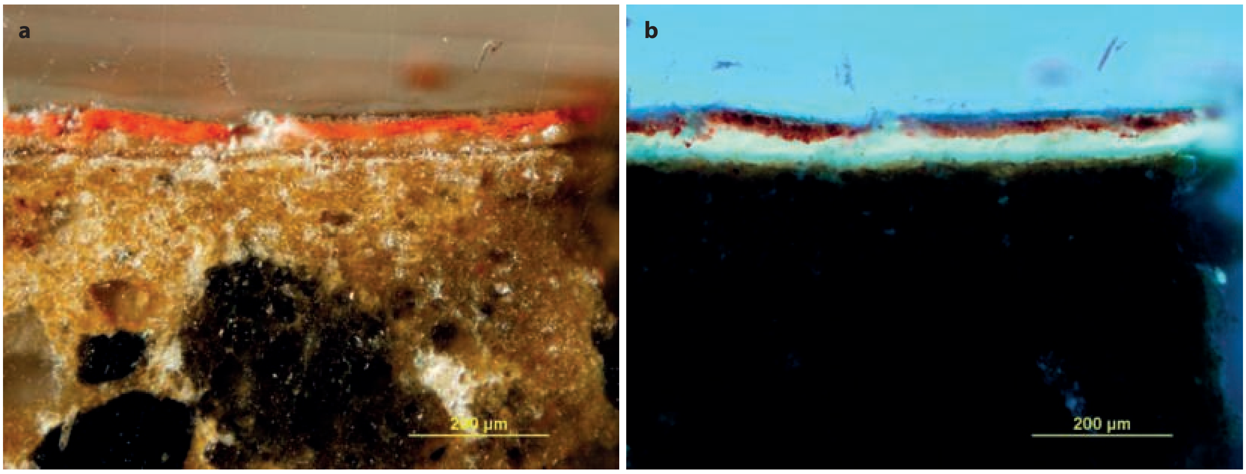


Figure 3.11 (a) PLM photomicrograph and (b) UV fluorescence image of a cross-section of a wall painting (BMM063) from Cave B(d).

The sample was taken from a wall painting fragment (inv. no. 0617) recovered from Cave B(d). It is cracked, weathered and coloured by a red pigment (Fig. 3.10). Observation of cross-sections indicate that the painting has a multilayered structure consisting of a rendering layer made of kneaded clay, a transparent yellow sizing layer (emitting bluish UV fluorescence), a white ground layer (emitting yellow UV fluorescence), a red pigment layer and a red glaze layer (emitting white UV fluorescence) (Fig. 3.11). Under a stereobinocular microscope, the transparent yellow sizing layer, the white ground layer, the red pigment layer and the red glaze layer were mechanically and manually separated from the rendering layer, which consisted of clay. All parts other than the rendering layer were used collectively in the analysis sample. Based on the analysis of similar samples by SR-µFTIR, it was assumed that the sizing layer would contain amino acids and that the white ground and red pigment layers would contain drying oil. Further analysis was then carried out to identify the specific organic substances such as proteins and oils in these paints. The samples were analysed by GC-MS (details, including the conditions of analysis, are given in Appendix II.5).

3.3.2 GC-MS analysis of oils, beeswax and resins

The analysis of fatty acids (Fig. 3.12) revealed high concentrations of azelaic acid and palmitic acid. The concentration of azelaic acid, a saturated dicarboxylic acid, was 123.2 ppm (Table 3.1). In addition, the A/P value (azelaic acid/palmitic acid ratio) was 2.4, suggesting that this sample contains highly oxidised drying oil. This probably indicates that the drying oils used in Cave B(d) have been oxidised for over a millennium due to exposure to the cave's semi-external environment. The sample has a P/S value of approximately 3. It is difficult to identify the plant species from which the oil originated with any certainty, as the reference used to compare the quantitative values obtained does not seem to cover all the oil species that would have been available in the ancient Central Asian region. However, it is noteworthy that the drying oils analysed here are closer to walnut or poppy seed oil, both of which have P/S values of around 3, much higher than other drying oils of vegetable origin used in oils and paints (i.e. linseed, sesame, almond and tung oil).

The interpretation of fatty acid profiles can be problematic when there are mixtures of drying oils or when egg and non-drying oils are suspected. An A/P ratio of between 0.6 and 1 or higher indicates the presence of a drying oil. A/P values between 0.2 and 0.5 are indicative of egg yolk or semi-drying oils that only partially oxidise when exposed to oxygen (sesame and grapeseed oil) and are identified here as 'oil'. Environmental contamination due to microbial growth, lipids, fats, soot and grease have extremely low A/P values (less than 0.2) and are reported here as 'fatty acids'.

3.3.3 GC-MS analysis of proteinaceous substances

Sample BMM063 was then subjected to amino acid analysis in order to clarify the origin of the proteins. The chromatogram obtained is shown in Figure 3.13 and the quantitative values of the amino acids in Table 3.2. Data obtained from this sample were compared with references including egg, collagen

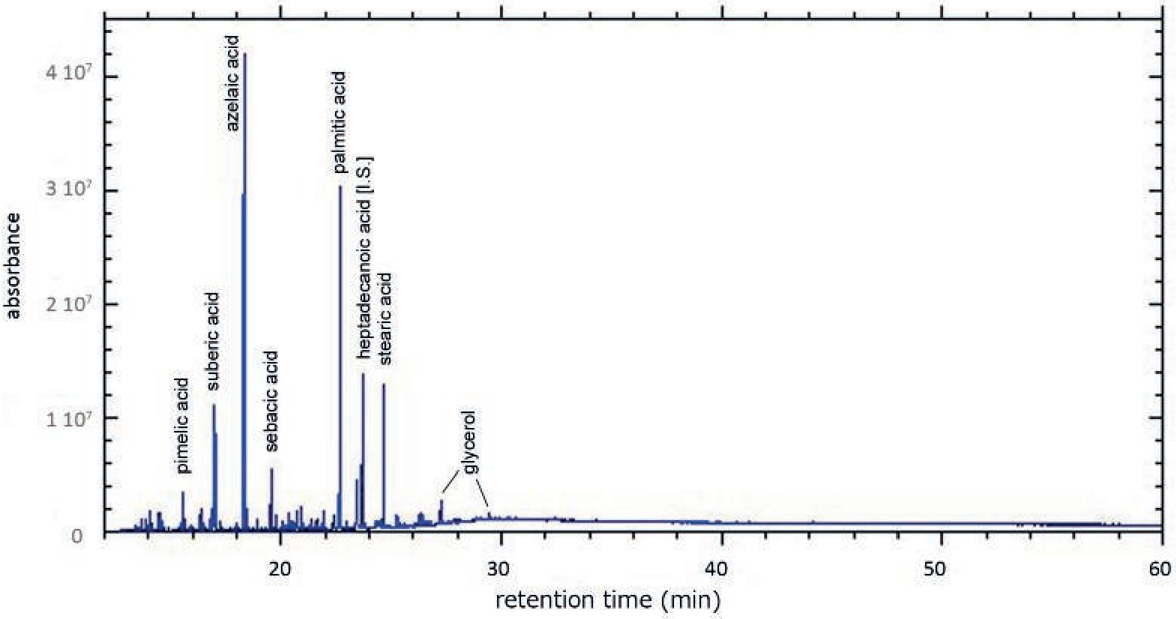


Figure 3.12 Chromatogram of fatty acids (methyl esters) by GC-MS for oil, beeswax and resin analysis of a sample (BMM063) from a red portion of a wall painting from Cave B(d).

Table 3.1 Quantitative values (ppm) of fatty acids in sample BMM063 and reference data: P/S, %FA percentage of total fatty acids in the sample.

Sample	*Cave*	*Description*	*Sample Weight ug*	*Final Volume ul*	*pimelic acid*	*suberic acid*	*lauric acid*	*azelaic acid*	*sebacic acid*	*myristic acid*	*palmitic acid*	*stearic acid*	*eicosanoic acid*	*oleic acid*	*P/S*	*A/P*	*% FA*
BMM063	B(d)	red and ground	76	15	6.97	25.7	0.0	123.2	10.7	1.2	52.4	17.0	0.0	0.0	3.1	2.4	4.7
Reference Oils																	
Plane tree gum		modern ref, Bamiyan 2006			0.38	0.9	0.0	2.0	0.2	0.4	3.8	1.5	1.7	1.3	2.5	0.5	
Almond oil Fowler (film)		M. Schilling 1989			0.53	3.5	0.0	15.9	0.7	0.0	4.6	4.2	0.4	1.5	1.1	3.4	
Linseed oil refined Winsor & Newton		M. Schilling 1989			4.93	35.5	0.0	365.2	14.5	0.6	165.0	134.7	3.4	25.1	1.2	2.2	
Poppy seed oil Grumbacher		M. Schilling 1989			7.02	70.2	0.0	655.7	20.9	0.6	365.7	116.6	4.4	18.7	3.1	1.8	
Poppy seed oil cold press Grumbacher		M. Schilling 1989			6.77	72.2	0.0	591.3	19.6	0.8	414.0	117.8	4.1	107.0	3.5	1.4	
Poppy oil sunbleached		M. Schilling 1989			5.05	46.2	0.0	414.7	12.4	0.4	328.6	74.1	3.1	5.2	4.4	1.3	
Sesame oil Arrowhead Mills		M. Schilling 1989			1.11	7.5	0.0	56.5	1.9	0.3	105.4	89.5	6.6	227.9	1.2	0.5	
Soy oil Spectrum Naturals		M. Schilling 1989			3.44	26.0	0.0	201.2	9.7	0.6	252.8	116.3	7.1	21.7	2.2	0.8	
Sunflower cold pressed Schminke Tung oil		M. Schilling 1989			0.4	2.9	0.0	21.2	1.1	0.0	6.3	12.2	0.7	2.4	0.5	3.4	
China		M. Schilling 1989			0.22	2.7	0.0	31.8	0.4	0.8	102.0	109.9	6.1	325.0	0.9	0.3	
Walnut Spectrum Naturals		M. Schilling 1989			5.37	40.2	0.0	363.5	11.6	0.4	117.5	37.3	1.6	2.5	3.1	3.1	
Walnut oil Rougie		M. Schilling 1989			4.54	35.4	0.0	339.9	12.4	0.5	324.5	142.3	4.1	12.5	2.3	1.0	
Safflower		M. Schilling 1989			4.81	38.0	0.0	286.2	12.0	0.8	147.8	112.6	12.5	16.3	1.3	1.9	

and casein. The correlation coefficients for sample and reference data on the seven stable amino acids (alanine, valine, isoleucine, leucine, glycine, proline and hydroxyproline) are shown, all of which are less likely to be oxidised or degraded over time. Methods for identifying protein and procedures for calculating correlation coefficients are based on those described in detail by Schilling and Khanjian (1996).

We concluded that the amino acids in the sample are highly compatible with some animal-derived binding media, such as isinglass and collagen, and that the amino acids originated from the same. However, it is not possible to determine from GC-MS results alone whether the binding media were derived from fish, deer or rabbit. This was tested using the ELISA technique, which facilitates identification of animal species using antibody reactions. However, the tests provided no reaction capable of positively identifying any species. Different organic substances were detected in the red portion of the sample using GC-MS analysis. A high content of fatty acids in the binding media was confirmed, as well as a similarity between fatty acid values for the drying oil and walnut and poppy seed oils. In other words, this confirms that the wall paintings in Cave B(d) were made using a technique similar to that used in oil paintings.

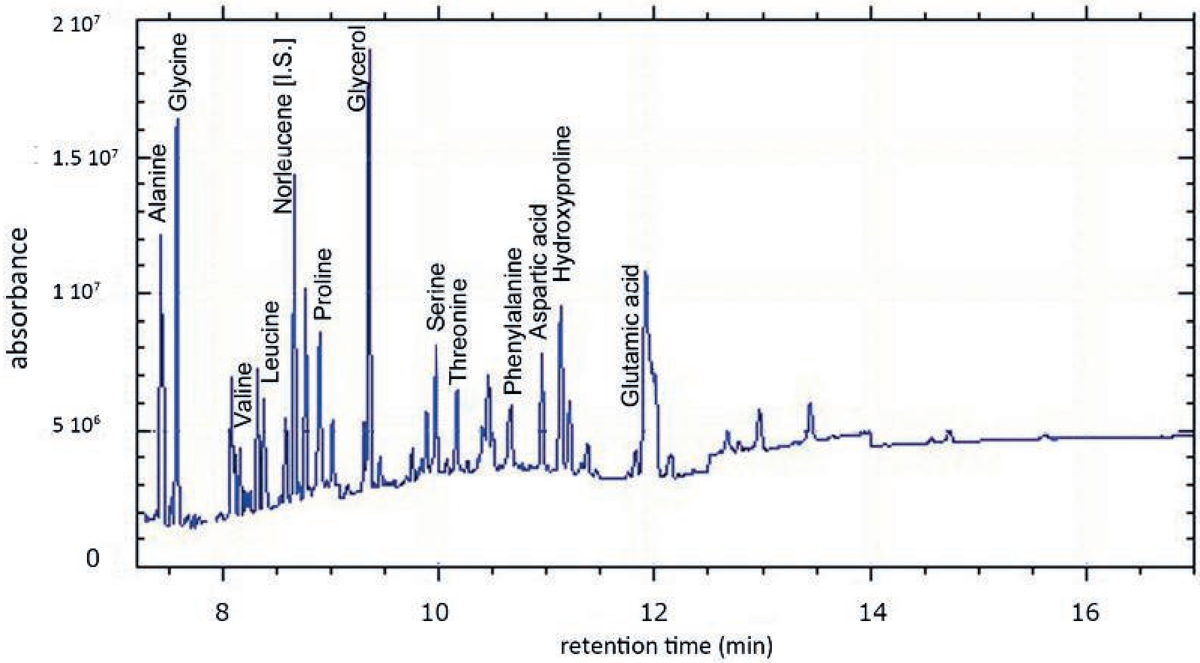


Figure 3.13 Chromatogram of amino acids by GC-MS of a sample (BMM063) from a red portion of a wall painting from Cave B(d).

Table 3.2 Quantitative value (ppm) of amino acids in sample BMM063 and correlation coefficients with reference data.

Sample	*Cave*	*Description*	*Sample Weight ug*	*Final Volume ul*	*oil %*	*protein %*	*alanine*	*valine*	*isoleucine*	*leucine*	*glycine*	*proline*	*hydoroxyproline*	*candidates/ correlation coefficient*	*results*	*GC/MS results of plant gums*
BMM 063	B(d)	red and ground	76	30	13.2	5.3	20.7	3.8	2.8	7.4	36.1	15.0	14.3	isinglass 0.976, collagen 0.971	glue	fructose, glucose, arabinose (honey?)
Reference Proteins																
Isinglass (Zecchi)							18.1	3.4	2.2	4.1	46.9	15.0	10.3			
Collagen & gelatine (mean)							15.7	3.0	1.8	3.7	46.7	16.7	12.4			
Egg white (mean)							22.0	18.3	12.8	21.6	14.8	10.5	0.0			
Whole egg (mean)							22.0	17.4	13.2	22.0	14.5	10.9	0.0			
Egg yolk (mean)							20.9	16.6	13.3	23.2	14.1	11.9	0.0			
Casein (mean)							10.9	16.9	12.8	22.0	8.6	28.8	0.0			

3.4 Case study 4: Green colour in Foladi Cave 4 and its discoloration

3.4.1 Description of the sample FDM055

Many of the wall paintings in Foladi Cave 4 have been lost to vandalism (Figs 3.14 and 3.15): large fragments have been removed and sold to buyers overseas while smaller fragments remain scattered on the floor. The fragment (inv. no. 0891) analysed here was recovered from the floor of Foladi Cave 4. The sample (FDM055) was taken from a green area. Observation of the cross-section (Appendix III, Fig. A.III.93) shows that this green area consists of an earthen render, a clear sizing layer, a transparent yellow sizing layer, a white ground covered with a multilayered structure of red, white and green on top of a white ground layer. This layered structure makes the green colour appear deeper and more vivid, as discussed in section 3.2.

3.4.2 SR-μFTIR analysis of the sample

The sample was analysed using μFTIR according to procedures described in Appendix II.4.6. A 2D map acquired from the entire stratigraphy and average FTIR spectra were calculated for each layer (Appendix III, Fig. A.III.95 and Fig. A.III.96). Their analysis and comparison revealed interesting results. Moreover, the integrated intensity of specific absorption bands provided useful information about the distribution of different components (Appendix III, Fig. A.III.103). First, the uppermost light green layer (1) of the coloured surface shows absorption characteristic of atacamite or paratacamite (basic copper chloride) as well as absorption characteristic of moolooite (copper oxalate), which may be responsible for the whitish degradation veil (Appendix III, Fig. A.III.97). This probably formed from the reaction between a copper-based green pigment and

Figure 3.14 The entire ceiling of Foladi Cave 4. Photo: Y. Maekawa, courtesy of NRICPT, 2007.

carboxylic acid produced from the degradation of organic binding media.

The next green layer (2) shows absorption characteristic of copper carboxylate, probably the result of a reaction between copper pigments and oil. Lead carboxylates are also detected that may derive from the reaction of lead white pigment or a lead drier and drying oil (Appendix III, Fig. A.III.98). The FTIR spectrum of the red layer (3) is very similar to that of the green layer (2): both primarily contain a partially saponified drying oil. We calculated the difference between the two spectra to identify the components in each layer (Appendix III, Fig. A.III.99). The calculations show the specific presence of goethite (brown iron oxide) in layer (3), and a higher degree of oil saponification in layer (2). The white ground layer (4) showed absorption associated with sharp ν(OH) at 3535 cm^{-1}, indicating the presence of hydrocerussite, as well as a peak at 838 cm^{-1}, which is characteristic of cerussite. The maps of these two peaks indicate that hydrocerussite is concentrated in layer (4) and is more diluted in layers (1) and (2), while some high spots of hydrocerussite correspond to visible white grains. At the same time, cerussite is mainly concentrated in white layer (4) (Appendix III, Fig. A.III.100).

Figure 3.15 Fragment of a wall painting from the ceiling of Foladi Cave 4. The fragment was once held by the Japan Committee for the Protection of Displaced Cultural Properties and returned to the Afghanistan National Museum in 2016 with 101 other pieces. Photo: Japan Committee for the Protection of Displaced Cultural Properties/Tokyo University of the Arts.

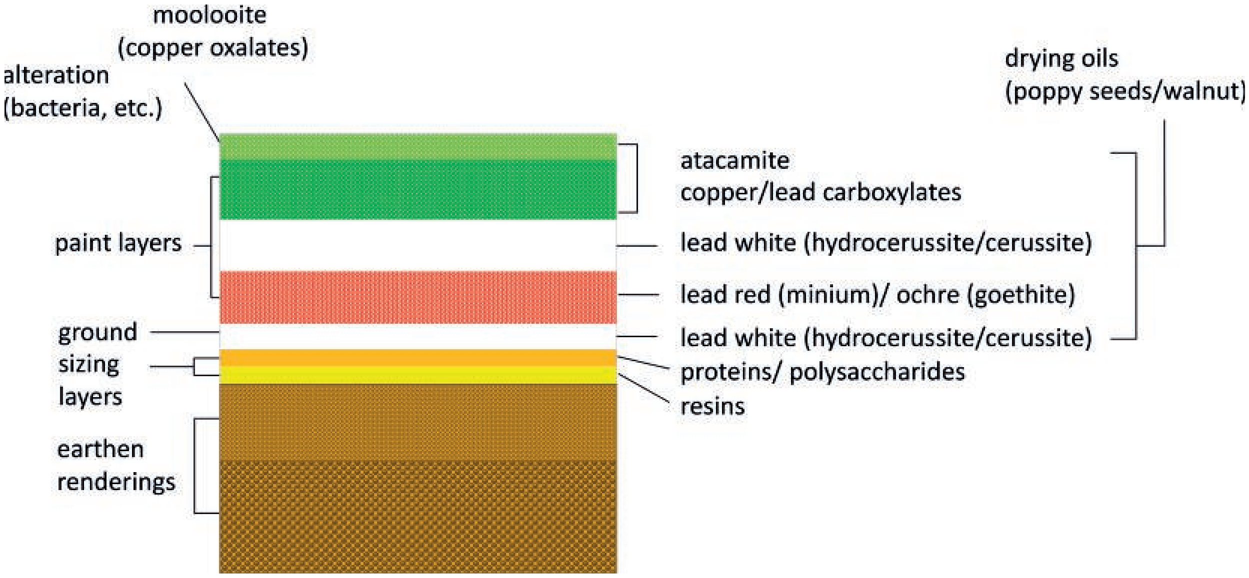


Figure 3.16 Schematic diagram of the multilayered structure of a green portion of a wall painting from Foladi Cave 4.

The transparent yellow sizing layer (5) shows absorption characteristic of amide as well as polysaccharide absorption of around 1050 cm^{-1} (Appendix III, Fig. A.III.101). It is believed that this layer contains both proteins, such as animal glues and egg whites, as well as gums (plant polysaccharides), possibly peach gum and gum arabic. The bottom transparent layer shows absorption derived from resinate (Appendix III, Fig. A.III.102), therefore it is probably a layer composed of some kind of natural resin. This is an excellent example of partially saponified drying oils, proteins, resins and plant gums all being present in one sample (Appendix III, Figs A.III.103 and A.III.104).

3.4.3 µXRF and µXRD analyses of the sample

Simultaneous µXRF and µXRD analyses were carried out on thin slices of the same sample. SR-µXRF elemental mapping is shown in Appendix III, Fig. A.III.94 and SR-µXRD mapping in Appendix III, Fig. A.III.104. The µXRF analysis detected lead in the ground layer and the coloured painting layers, iron in the red layer and copper in the green layer (as well as in the uppermost layer). SR-µXRD analysis confirmed the main FTIR results with the presence of moolooite and atacamite in the green surface areas, cerussite and hydrocerussite in the white ground layer and the white intermediate paint layer with higher cerussite to hydrocerussite ratios in the ground layer, and goethite in the red areas. In addition, SR-µXRD analysis revealed the presence of minium in the red layer. This component is transparent in the mid-IR region and could not be detected with SR-µFTIR. Finally, µXRD revealed the presence of two additional lead sulfate-based degradation products: palmierite and anglesite.

3.4.4 GC-MS analysis of fatty acids

SR-µFTIR analysis revealed that the green area of Foladi Cave 4 (FDM055-1) contains drying oils, proteins, resins and plant gums. In order to study the drying oil, the sample was subjected to fatty acid analysis using GC-MS: this revealed high concentrations of azelaic acid (399.4 ppm) and palmitic acid (217.6 ppm), as well as stearic acid and suberic acid (Appendix III, Table A.III.32), confirming the presence of drying oil. The P/S value of this fatty acid was 2.8, similar to the P/S values of walnut, poppy seed or perilla oil. Based on the findings from the SR-µFTIR analysis, drying oils such as these were probably used in the green, white and red coloured layers and in the white ground layer.

3.4.5 Layered structure of the sample

In summary, the wall paintings in Foladi Cave 4 were probably produced as follows (Fig. 3.16). First, two layers of an organic paste-like substance were applied on the surface of an earthen render. Because water-soluble paints would be applied on the porous, water-absorbent earthen walls, the surface was first sealed to ensure that brushes could retain their moisture and draw smooth lines. Thus, sizing was applied to the surface of the wall, either to seal or buffer it. The lower transparent yellow layer is made of natural resin while the upper transparent yellow layer is a mixture of proteins, for example, animal glue or egg white, and vegetable polysaccharides such as gum.

The white ground layer is made of paint consisting of lead white (hydrocerussite and cerussite) and partially saponified drying oil. The painting layers, from top to bottom, are green, white and red, and are pigmented with atacamite, lead white and a mixture of minium and ochre (goethite) respectively. All the layers have oil as a binding medium. It was observed that the oil partially reacted with metallic pigments to form carboxylates such as copper and lead. The copper carboxylates contribute to the green colours. As mentioned in Chapter 2, section 2.2.7.3, there are a few cases of atacamite being detected in the green areas of Asian paintings (Naruse 2004). However, it is still unclear whether atacamite was intentionally collected as a natural green mineral and used as a pigment, if it was artificially synthesised using seawater, or whether it was the result of a reaction that transformed a copper-based pigment such as malachite into atacamite. In addition, moolooite was detected on the surface of the green layer as a degradation product from copper pigment.

The above discussion has focused on oil wall paintings in Caves B(d), N(a) and Foladi Cave 4. It was found that samples taken from each of these caves had complex multilayered structures, with each layer containing a variety of organic materials such as oil and resin, which also reacted with the inorganic pigments to form a variety of secondary products. The Bamiyan oil paintings include not only the use of drying oil, but also involve complex processes surrounding preparation of the wall, the application of colours and surface finishing. The choice of materials and oil painting techniques required skill and planning, techniques that were probably already well established when introduced to Bamiyan.

3.5 Painting techniques at the Eastern and Western Giant Buddhas

A description of the two Giant Buddhas can be found in the *Great Tang Records on the Western Regions of the Great Tang Dynasty* by Xuanzang, who visited Bamiyan in the early winter of AD 630. It is likely that Bamiyan's stone cave temples were being actively constructed at that time, probably shortly after the completion of the Western Giant Buddha. ICOMOS Germany estimated the date of the Western Giant Buddha's construction as between 591 and 644 (2σ) (Blänsdorf *et al.* 2009a). On a hill to the northeast of the royal city stands a giant 150 ft high figure of the Buddha made of stone. To the east of this figure is a Buddhist monastery, to the east of which is another standing figure of Buddha Shakyamuni, measuring 100 ft high and made of brass. In the monastery is a lying figure of the Buddha in nirvana, 1000 ft long (Beal 1884: 50 ff.).

Figure 3.17 East Giant Buddha (EGB: small 38 m Buddha) in Bamiyan, as the first photograph from J.A. Gray, *At the Court of the Amir* (1895) (after Morgan 2012: 2, fig. I).

The 'standing figure of the Buddha' (Western Giant Buddha) and the 'standing figure of the Buddha Shakyamuni made of brass' (Eastern Giant Buddha) are carved from the conglomerate rocky cliffs (Fig. 3.17). Artisans carved their outer shape from the cliffs and then shaped the figures with thick kneading clay. These figures were not cast as Xuanzang describes: observations show that the rock in the Giant Buddhas was prepared to facilitate the adhesion of the clay renders by drilling holes in its surface and placing round stones and wooden pegs in the holes. The wooden pegs were then connected to each other with ropes made from plants, *Dom-i shutur* (*Astragalus cuneifolius* Bunge), harvested from the mountainous area. These piles were then covered with a mixture of straw thatch and animal hair, ranging in thickness from a few centimetres to over 20 cm (Grunwald 2009; Pfeffer and Blänsdorf 2009a). The folds of the Buddhas' robes were then created in three dimensions by applying a band of clay paste. A few millimetres of fine clay paste mixed with sand was then applied to the surface.

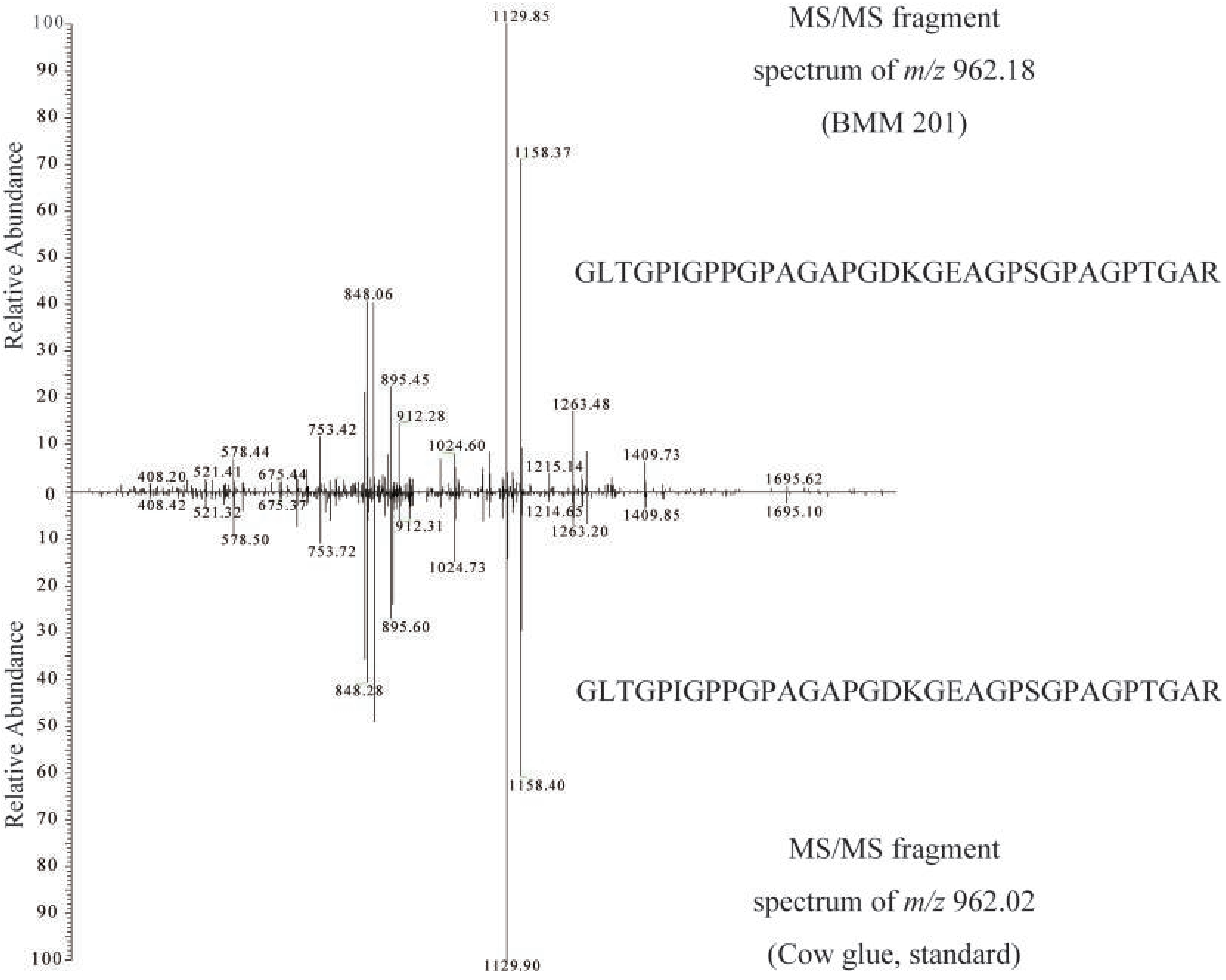


Figure 3.18 Comparison of MS-MS spectra (from Kawahara *et al.* 2013).

Analysis by ICOMOS Germany and the Technical University of Munich detected red, blue and yellow colours on the surface of the Eastern Giant Buddha, which dates from the 5th and 6th centuries (Blänsdorf *et al*. 2009b). In blue areas, a blue layer of lapis lazuli was applied on top of a black layer, which seems to be particularly unrefined due to its high content of calcite and other impurities relative to the lapis lazuli used in the wall paintings of Cave N(a). The application of coarse lapis lazuli over a black layer is similar to the techniques used in the gypsum-based wall paintings of Caves C(a), D, G, I, J(b), J(c), J(d), K_3 and the East Displaced Cave.

The original positions of the painted fragments related to both Giant Buddhas are not known, as the fragments were damaged and repainted many times, and layered structures vary from place to place. Findings from analyses were difficult to interpret but judging from the cross-section, it is believed that both Giant Buddhas were repainted two or three times. The presence of a first layer of pink gypsum/clay ground and a second subsequent painted layer of gypsum or lead white (Blänsdorf *et al.* 2009b) indicates that the lead white substrate is of a later date than the pink substrate.

The Eastern Giant Buddha may have had blue in its garment while the lining remained white (Blänsdorf 2021). Analysis of samples provided by ICOMOS Germany showed that the blue colour included high levels of impurities in lapis lazuli, calcite and pyrite. It is possible that lapis lazuli, a relatively affordable pigment, was chosen in consideration of the Giant Buddha's large surface area and the distance from which the work would be viewed. In some places, the black and blue layers appear twice, suggesting that they were repainted at some point. In this case, it is interesting to note that not only was the blue surface repainted but the black layer also appears below it.

On the other hand, the surface of the Western Giant Buddha, which was constructed in the early 7th century, shows red (red ochre) for the most part, with only a few exceptions such as yellow and white, and no blue (Melzl and Petzet 2007). It is possible that the red colour was used as a ground layer for more expensive pigments such as red lead or cinnabar/vermilion, as is the case with the wall paintings in the front chamber of Cave C(a). In other words, the painting of this Buddha may have been left unfinished, although it is unclear whether this was for financial reasons. At present, there is no evidence of gold leaf on the surface of what remains, therefore Xuanzang's description of the Western Giant Buddha as being gold in colour may be an error or misunderstanding. In addition, there are places where no white ground layer can be found (Blänsdorf *et al.* 2009b). This differs greatly from the wall paintings, all of which have some kind of ground layer, such as gypsum, lime or lead white.

Another example of the Giant Buddha of Central Asia is the 13 m long Reclining Buddha made of kneaded clay at Ajina-tepe (Tajikistan), built in the 7th–8th century. It was excavated by the Russians in the 1960s and 70s and moved to the National Museum of Antiquities in Dushanbe. In 2007, the surface of the Buddha was observed to have a white ground layer with some red (red ochre) in its robe and blue (lapis lazuli) in its hair. The surface of the Buddha is white. The way in which the Reclining Buddha at Ajina-tepe has been painted is reminiscent of the polychrome style of Bamiyan's Eastern Giant Buddha.

GC-MS analyses for detecting fatty acid, resin and beeswax were carried out on three painted fragments taken from the Eastern Giant Buddha and two taken from the Western Giant Buddha (Appendix III, Table A.III.32). GC-MS analyses for detecting amino acids were also conducted on one painted fragment from the Eastern Giant Buddha. The results for plant gums are shown in Appendix III, Table A.III.34. All the samples were provided by ICOMOS Germany. GC-MS analysis of the painted fragments did not reveal any fatty acids in either of the Giant Buddhas. Because fatty acids would have indicated the use of a drying oil, it is therefore likely that a water-soluble binding media was employed in both cases.

The samples were also studied in Japan and the United States using PLM, SEM-EDS, GC-MS and nanoscale liquid chromatography coupled with tandem mass spectrometry (nano-LC-MS-MS) techniques. Results showed that the Eastern Giant Buddha had been painted numerous times in red, blue and yellow on a white gypsum ground with a water-soluble binder containing xylose and amino-acids (Taniguchi 2016: 185). Figure 3.18 shows a comparison of two GC-MS/MS spectra (above: WGB sample BMM201; below: cow glue, standard) (Kawahara *et al.* 2013; Taniguchi 2016: 186; Taniguchi *et al.* 2022). It was previously believed that a cowhide glue could not have been used in Buddhist contexts such as Bamiyan; therefore, these results gave new insight into the palette of artisans employed by the kings and monks of ancient Bamiyan. LC-MS analysis carried out by the Nippi Research Institute of Biomatrix, Japan, on the sample from the Eastern Giant Buddha (BMM194) identified cow glue (Taniguchi 2022). Analyses by the University of Pisa in Italy found egg protein in samples provided by ICOMOS Germany from the surface of both Giant Buddhas as well as cow, goat and sheep milk casein, egg and animal glue in repainted areas. Tragacanth gum was also identified (Blänsdorf 2021).

When the Giant Buddhas were being constructed, painting their surface using only unstable footholds would have required remarkable speed and efficiency, a task very different from creating wall paintings in a rock-cut cave. It is likely that water-soluble binding media and relatively inexpensive pigments were used to paint the expansive surface with large brushes. As the painting techniques used were similar to the gypsum-based wall painting techniques employed in the caves around the Eastern Giant Buddha, we may assume that both the artisans and techniques involved in their production differed from those of wall paintings produced using oil painting techniques.

Chapter 4: Study of variations in painting techniques at the Bamiyan site

4.1 Correlation with cave structure and style

4.1.1 Geographical distribution of cave temples and their architectural history

In this section, we review prior research on cave types and painting style, and discuss the correlation between the two. Caves referred to here are man-made chambers that have been carved into a cliff or rock – they are not strictly architectural structures built from components. However, the sculpted form of the caves often contains elements that mimic wooden or stone structures, such as beams, ceilings and walls. For this reason, many studies have been carried out on the structure and form of caves from architectural and historical standpoints.

Bamiyan contains about 750 caves in its cliffs and hillsides that were used as shrines for meditation as well as for residential quarters for Buddhist monks. In fact, the grounds inclusive of the monastic complex and the Giant Buddha statues are believed to have functioned as a Buddhist monastery. Although the connections between caves are now largely lost due to the collapse of the cliff front, they probably served as entrance facilities with stairs and passages when the caves were created. Bamiyan's caves are connected by passages that loop around the two Giant Buddhas, some of which are interconnected while others are independent. The structure, form and size of the caves are diverse and include square, octagonal and circular shapes and various styles of ceilings such as domed (Laternendecke) flat and cross-vaulted. Previous studies on cave structure and form were detailed by Itsuji Yoshikawa (1948), Takehisa Kodera (1972), Takayasu Higuchi (1980) and Shigeru Kubodera and Mayu Iwade (2008).

Takayasu Higuchi of Kyoto University has described the geographical distribution of cave temples in Central Asia. Of the many cave temples across India and around the Tarim Basin, Bamiyan is probably the farthest west. Numerous large cave monasteries are found across India such as the caves at Ajanta, Ellora, Kondivite, Bhaja, Karla, Bedse, Kondana, Pitalkhora and Kanheri and the Nāsik Temple. The Kondana, Pitalkhora and Kanheri caves are located in the west of India, where relatively soft limestone and basalt cliffs lie. Their interiors often contain magnificent sculptures and wall paintings deemed sacred in their time. Afghanistan's cave monasteries are concentrated in Jelalabad, Aybak and Bamiyan. The Bamiyan Valley has smaller cave monasteries tucked into the surrounding Kakrak, Foladi and Qol-e Jalal valleys but compared to India's cave temples in basalt or limestone rock beds, the Afghan cave temples tend to be smaller in size, probably due to Bamiyan's fragile conglomerate rock. Few cave monasteries are found in Gandhara, Pakistan, where mountain temples such as Takht Bahi were built instead, possibly due to a lack of malleable rock for constructing caves in the cliffs (Higuchi 1980: 98).

Beyond the Pamir Plateau, numerous cave monasteries are located along the northern road of the Tarim Basin in Kucha, Kashgar and Turfan. The Kizil grottoes and the Kumtura, Bezeklik and Simsim caves are archetypal examples. Many caves around the Tarim Basin are also carved into fragile conglomerate or sandstone cliffs, as in Bamiyan. However, there are also examples of mud-brick structures built in front of cliffs and used as caves, especially during the later period, as seen in Bezeklik and the Simsim caves. In addition, the caves around the Tarim Basin are similar to the Bamiyan caves in terms of their style and form including their ceilings. The caves stretch to Gansu Province in the west where the Mogao caves of Dunhuang and the Maijishan grottoes lie. In this span, but to the east, lie scattered monasteries such as at Longmen and Yungang. The cave monasteries around the Tarim Basin and up to Gansu Province all contain wall paintings.

Thus, the cave temples appear to be distributed from northwest India to the mountains of Afghanistan, and concentrate along the northern road of the Tarim Basin. These are mountainous areas of relatively soft rock such as limestone, sandstone and conglomerate, with rivers and valleys close to cliffs and massifs, and relatively open surroundings. Wooden Buddhist temples were also built in areas without cliffs, such as the site of Dandan Oilik

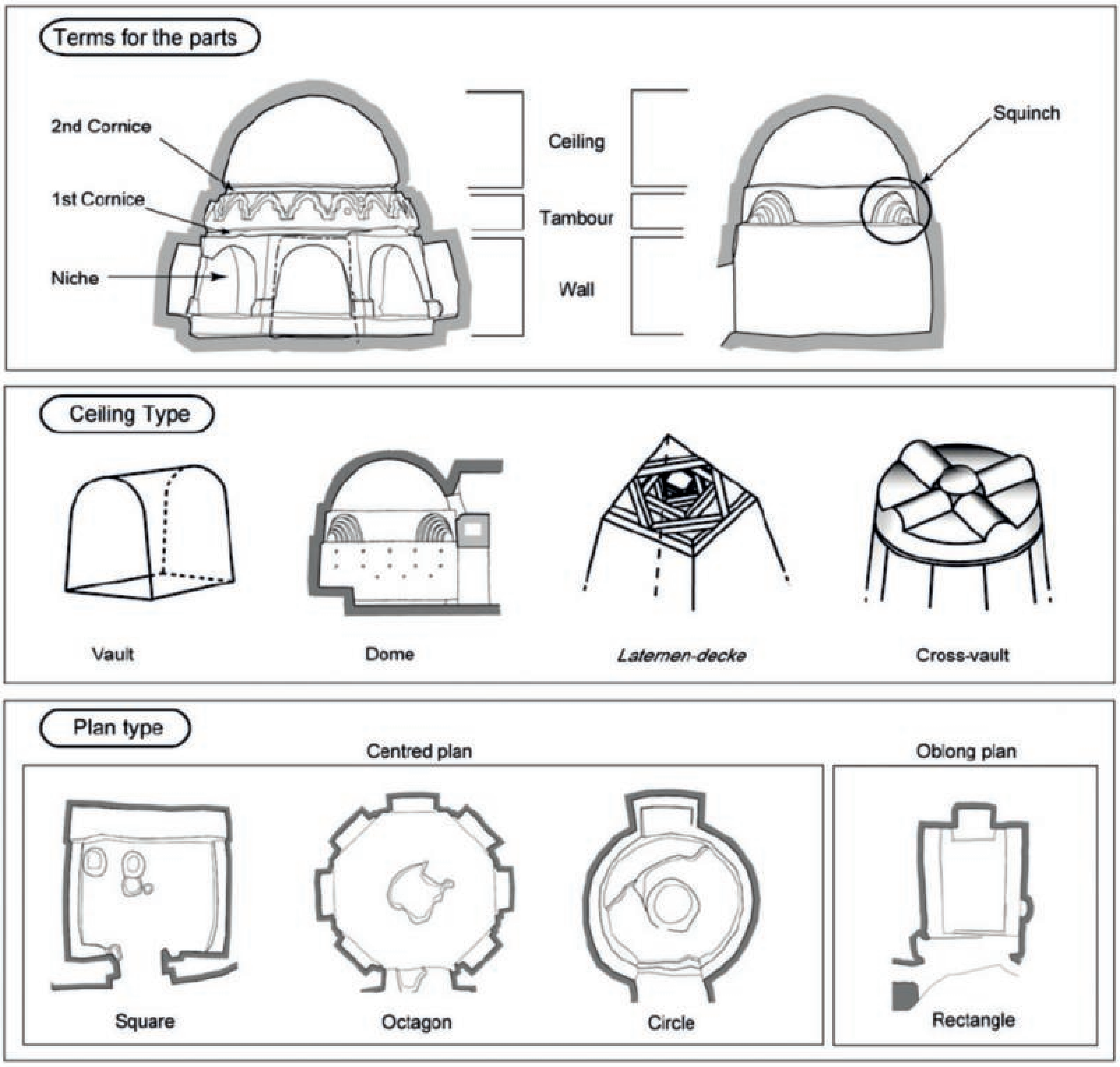


Figure 4.1 Terms used for each part, ceiling and plan (from Iwade and Kubodera 2013: 4, fig. 3).

in the south of the Tarim Basin. Correspondingly, cave temples are thought to exist as an alternative to wooden or stone temples. As is common with Buddhist temples, the caves were used for monks to train and therefore needed to be secluded; this rationale may explain the proliferation of small cave temples deep inside the valley.

4.1.2 Cave styles at the Bamiyan site

The cliffs and slopes at Bamiyan consist of conglomerate rock with very weak, silty layers. This bedrock is composed of red sand, clay and white and brown strata. Bamiyan receives a small amount of precipitation during the cold winter months, which affects the fragile rock formations. The cliffs have been gradually collapsing due to the annual ingress of snow run-off from the mountain tops. Much of the cliff face has fallen, and many entrance facilities such as staircases and passages have been lost. It has therefore been difficult to reconstruct how the caves, with their various functions, were once interrelated to form a complex. For this reason, we have conducted research on the style and structure of the surviving main chambers.

Research on the chronology of changes in the caves, their morphological style and design dates back to the 1920s. For example, the architects Andre and Yedda Godard of the French archaeological delegation in Afghanistan, DAFA (Délégation archéologique française en Afghanistan) measured and sketched Bamiyan's caves. After classifying various styles as having served as shrines for meditation, lecture halls and monasteries, they believed the caves acted as a complex connected by passages (Godard

Table 4.1 Typology of Bamiyan Buddhist caves (after Iwade and Kubodera 2013: 10, table 1).

Plan type		Ceiling type	Features	Examples of caves
Centred plan	Square	Dome	Flat structure remains in the corners	53-IV, 53-VI, 35-III
			Trapezoid form of corbels in the corners	137
			Squinch arches: supporting the dome structure	A Lower (a), B(a), 35-IV, V
			Squinch arches: curved on the dome structure	F1(a), G
			Particular type a	A Upper (c), B1(c), T
			Particular type b	U
		Laternendecke	Imitation of flat wooden structure	F3(f)
			Imitation of wooden structure of joists:	F(c), H, N
			Walls without decoration	53-V, XIII, XV, XVI
			Walls decorated with arcades and pilasters	S(f)
			Irregular type	
		Flat	Section form is almost rectangular	F3(a)
			Section form is trapezoid	E(h)
	Octagon	Dome	Without arcade	53-III, 35-VII
			With single tier arcade	F(a), D1
			With two-tier arcade	53-I, 53-II, XI
		Laternendecke	Normal type	35I, A Upper (a), L, R(a)
			Exceptional type	O(a)
		Cross-vault	Normal type	35-II, D
	Circle	Dome		C, XII(d), 35-IV
Oblong plan	Rectangle	Flat	Section form is almost rectangular	XII(c), XIII(a), B1(a)
			Section form is trapezoid	XVI(a), E(k)
			Section form is trapezoid with steps	O(b), Q(b)
		Vaulted	Barrel vault	XIII(b), N(f)
			Barrel vault with steps	F3(b), East I(b)
		Cross-vault	Cross-vaulted ceiling with a quarter line section	B(d)

et al. 1928). In addition, the Godards attempted to devise a timeframe for the construction of the caves and cave groups based on their style and design. They also suggested that the cave styles were related to the above-ground structures such as the wooden temple complex that would have existed at the time. Kodera (1972) further argued that styles of worship differed according to whether a stupa or a Buddha image was present in the cave, and that a cave's style and the position of a complex on a cliff indicate the time of their creation. Iwade and Kubodera (2013: 237) studied the passages and entrance facilities attached to the caves, and also examined how the caves were connected to the original stairways. They considered the relationship between all the caves located in the upper and lower cliffs in a three-dimensional manner, and concluded that the schematic organisation of the caves provided by the French and Nagoya University teams must be reconsidered.

In both prior studies, Bamiyan's caves were basically organised according to the floor plan and ceiling style (Kodera 1972). However, the structural logistics of connecting a square, block-shaped cave to a circular dome ceiling, for example, is inherently quite complex. Therefore, Kubodera and Iwade (2008) added consideration of the architecture used in construction, such as the adoption of cornices, tambours and squinches, as seen in the transitional space between the walls and ceiling. They deemed this transitional architecture a key classification criterion for determining cave style (Table 4.1).

Figure 4.1 shows the floor plans for the caves divided into two main types: centripetal and long hall. None of the caves conform to Chaitya (prayer hall) or Vihara (monastery) plans, as seen in Indian cave temples. The centripetal plan is further divided into three categories: square, octagonal and circular, while the long hall plan is divided into rectangular styles. Ceiling styles in the caves also relate to floor plans. Ceilings in most centripetal caves are domed, flat, Laternendecke or cross-vaulted, while oblong, long hall caves are vaulted or flat. Most caves in Bamiyan are square or octagonal with domed ceilings. When the floor plan of a cave differs from its ceiling plan, some kind of transitional device is required to connect the two different styles.

The niches of the Eastern and Western Giant Buddhas, as well as those of Caves I and H and Kakrak, are carved into the cliffs to house standing or seated Buddha figures. The ceilings of such niches are vaulted. In the main cliffs at Bamiyan and the Foladi and Kakrak caves, wall paintings and reliefs

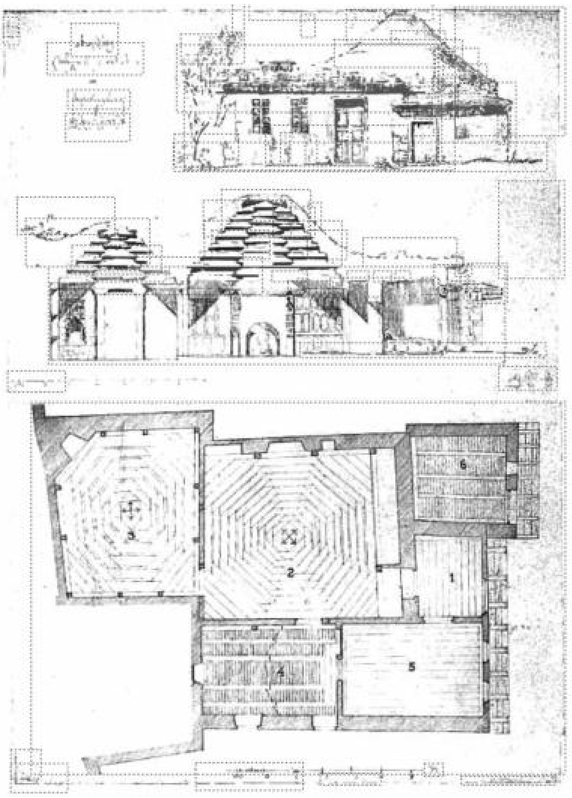


Figure 4.2 An example of Laternendecke in Akhaltsikhe, Georgia (after Lordknpanidae 1960: pl. 34).

Figure 4.3 An example of Laternendecke in Akhaltsikhe, Georgia (after Lordknpanidae 1960: pl. 51).

are found mainly in the relatively small centripetal caves. These caves are square, octagonal and circular and many have domed or square-vaulted ceilings. In contrast, with the exception of Cave K_3, rectangular caves have few wall paintings or other decorations. For this reason, it is assumed that centripetal caves functioned as shrines and ceremonial spaces, while rectangular caves had a more mundane function as storage units or housing.

The origins of cave styles have also been the subject of much discussion. The genesis of cave monasteries is often traced to India but Itsuji Yoshikawa, for example, links the origins of octagonal and circular centripetal caves to the monumental architecture of the Mediterranean world, and considers the Laternendecke (so-called due to its lantern-like shape) ceiling to be an imitation of beam structures seen in Central Asian houses. Higuchi also noted the distinctive structure of domes, vaults, squinch arches and Laternendecke ceilings found in Bamiyan, and traced these to Western influences rather than associating them with Indian cave temples. In particular, he makes a connection between the domed ceiling of the square caves and the Byzantine and Sasanian palace styles (Higuchi 1980: 111). Iwade and Kubodera (2013: 231) also noted that the ceiling styles and decorations were designed based on realistic architectural structures and components, as well as the influence of masonry found in Mediterranean architecture as seen in the cornices of transitional sections in Greek architecture. The origins of the cave styles are as complex as the wall painting motifs owing to Bamiyan's diverse historical and geographical ties to the broader region.

4.1.3 Square-vaulted Laternendecke ceiling

Several of Bamiyan's caves have distinctive ceiling styles, one being the style known as the Laternendecke. This has ceiling beams that appear to imitate wooden ones, which are assembled in a square, turned 90 degrees and then piled over. It is a triangular corner truss-bracket ceiling. Designs using such structures are believed to have originated in Asia Minor and spread to Armenia and other parts of the Caucasus. They are widely visible across Eurasia. According to Itsuji Yoshikawa (1948), the Laternendecke style originated in the mountainous region of South Central Asia, along the Wakhan Corridor of Pamir and northern Badakhshan. It can be seen in Kashmir's stone temple of Pandrethan (9th century, Hindu temple), the private houses of Hunza, and the palace of Nisa (250 BC–AD 226, Parthian period) (Higuchi 1980: 108). It can also be observed in Ashkhabad, Turkmenistan (3rd century BC) as well as Bulgaria's Thracian era rock chamber tombs (2nd century BC) and North Korea's Koguryo mural tombs (6th century AD) (Higuchi and Barnes 1995).

Scholars have also noted the Laternendecke style in Qutub Minar (New Delhi, India), Akhaltsikhe in Georgia (Lordknpanidae 1960: pls 34 and 51) (Figs 4.2 and 4.3), and Tajikstan's West Pamirs (19th century) (Mukimov and Mamajanova 1990: 121) (Fig. 4.4).

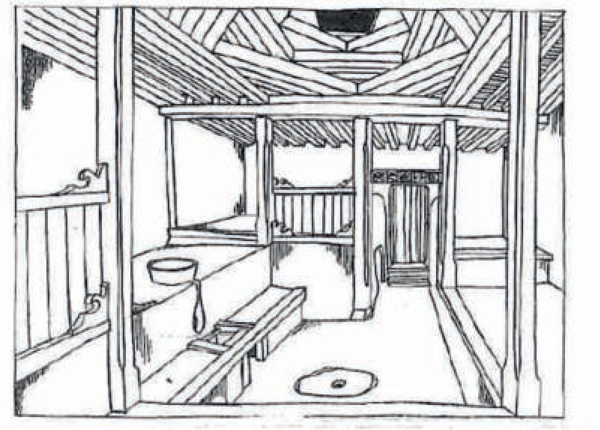


Figure 4.4 An example of a 19th-century Laternendecke in West Pamirs, Tajikistan (after Mukimov 1990: 121, pl. 97).

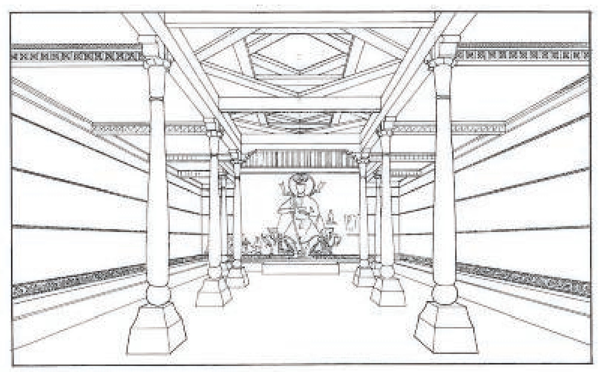


Figure 4.5 The Blue Hall of Varakhsha in the Sogdian Palace of Bukhara, Uzbekistan is also believed to have a Laternendecke ceiling style (after Maršak 2000: 159, fig. 7).

Figure 4.6 The Laternendecke ceiling of the Great Entrance Stupa of Alchi in the Leh district, India (12th century). Photo: G. Kozicz, 2009.

Figure 4.7 The Laternendecke ceiling of the inner stupa of Stupa 3 of Alchi in the Leh district, India (13th century). Photo: N.J. Chunka, 2018.

The Blue and Red Halls of Varakhsha at the Sogdian Palace in Bukhara are also believed to have this ceiling style (Maršak 2000) (Fig. 4.5), while the Alchi monastery in Leh, Ladakh, has a Laternendecke ceiling with painted wooden beams and mandala patterns (Figs 4.6 and 4.7). The constituent materials should be further studied in order to compare drying oils in wall paintings to those on Laternendecke structures.

Bamiyan's square-vaulted Laternendecke ceiling is probably a design adaptation mimicking the bracing of beams in traditional wooden buildings (Godard *et al.* 1928; Kubodera and Iwade 2006: 69–70). However, similarly designed ceilings were also found mainly in the Tarim Basin by the 5th and 6th centuries (Kizil Caves 131, 132, 156, 165, 166, 167, 169, 207 and 227) (Howard and Vignato 2015: 33, 35), Kizil-Qargha (Cave 32) (Howard and Vignato 2015: 10) and the Korean Peninsula (Complex of Koguryo Tombs). Bamiyan Cave XV is a square cave with a high Laternendecke ceiling and pillars carved in relief on the inward-leaning surface above the side walls. Higuchi, who conducted the survey, noted that some of the columns and the beams on the square-mounted ceiling are painted yellow, 'as if depicting a timber-framed building' (Higuchi 1980: 82). Its origin is not clear but, as already mentioned, the unique square-vaulted form of the Laternendecke style is closely related to wooden structures and is said to imitate them.

4.1.4 The relationship between cave styles and chronology

Radiocarbon dating of straw fibre from rendering in the caves makes clear that there is a correlation between a cave's type and its construction date (Fig. 4.8) (Iwade and Kubodera 2013: 250). Caves with square plans and domed or flat ceilings are the oldest, followed by circular caves with domed ceilings and, from the mid-7th century onwards, caves with square-vaulted or cross-vaulted Laternendecke ceilings. Bamiyan's small-scale caves with square vaults and cross-vaulted ceilings that closely resemble wooden structures emerged at least a hundred

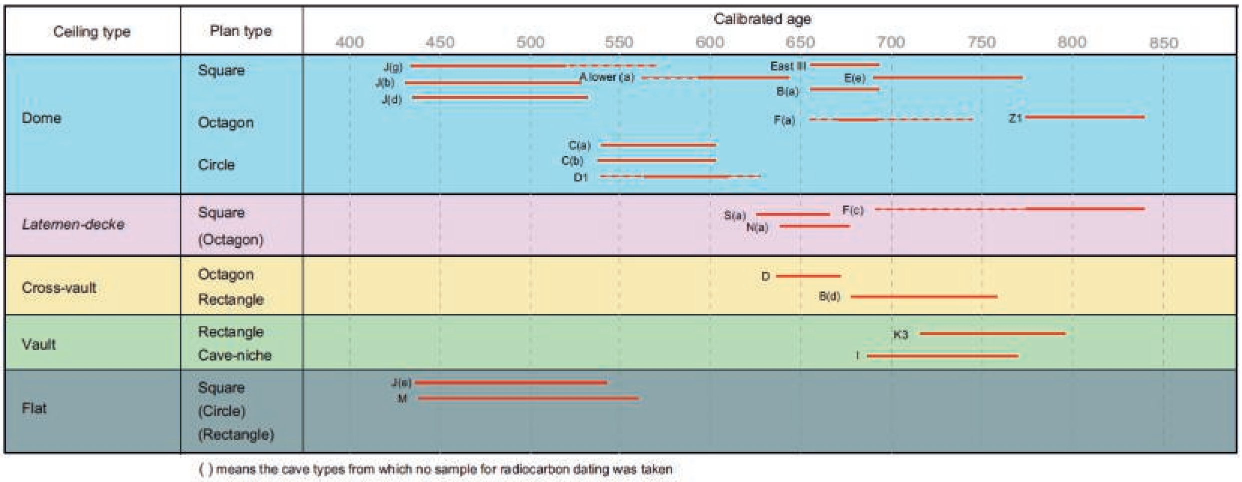


Figure 4.8 Comparison between the ceiling types of the cave and the results of dating (after Iwade and Kubodera 2013: 252, table 9).

years after the local arrival of Buddhism. Much more could be learned through further examination across a variety of research fields.

4.1.5 The relationship between painting composition and cave style

It is interesting to examine the relationship between wall paintings – particularly those made with drying oil (Bamiyan F(c), S(a), N(a), B(d), L, Foladi 2, 3, 4, 6, Kakrak 43, 44, Qol-e Jalal) – and cave style. Some fatty acid was detected from wall paintings in Caves E(e), J(c), K_3 and M, but the amount is low (0.1–1%) and P/S and A/P values are too low to be confirmed as drying oils. Therefore, we can assume that these samples have different sources of fatty acid relative to the samples from the 12 caves that contain high values of fatty acid. Figures 4.9, 4.10 and Table 4.2 summarise the caves and cave types containing the wall paintings in this study. Caves in which drying oil was identified as a binding medium are shown in Table 4.2 with their names in bold and underlined. The other caves were painted with water-soluble binding media such as proteins or plant gums.

In Bamiyan, the majority of caves are square or octagonal in plan with domed ceilings. However, in the particular case of caves in which oil painting techniques have been identified, seven are square or octagonal in plan with Laternendecke ceilings: Bamiyan Caves F(c), L, N(a) and S(a), Foladi Caves 4 and 6, Kakrak Cave 43 and Qol-e Jalal. Domed or cross-vaulted ceilings are only found in Kakrak Cave 43 and Bamiyan Cave B(d). Rectangular caves with vaulted ceilings are located in Foladi Cave 3 and Kakrak Cave 44. These caves are adjacent to Foladi Cave 4 and Kakrak Cave 43, both of which have Laternendecke ceilings and are therefore expected to be functionally interrelated.

There are several similarities in technique connecting the 12 wall paintings containing drying oil other than the fact that poppy seed or walnut oil served as their binding media. Firstly, all 12 paintings have a very smooth surface, perhaps because the walls were polished when they were in a semi-dried condition. Secondly, they are made up of layers of painting materials containing various organic substances, such as resins and proteins. These layers include materials applied as sizing, ground, paint or glaze, as well as the use of lead white as a white ground and metal leaf such as gold or tin. There are also similarities in technique in the composition of the paintings. For example, some resemble a mandala with a Thousand Buddhas while others use vivid shading techniques on the nimbi and lotus flowers. Additionally, in some paintings, fine iron-wire strokes were used to outline and detail figures and patterns.

The radiocarbon dates of the oil wall paintings correspond to Phase II (2) to Phase III (mid-7th–late 8th century), as discussed in Chapter 1. This timeline aligns with the results of the previous section, which showed that small-scale caves with square- or cross-vaulted Laternendecke ceilings appeared in Bamiyan from the early 7th century onwards (Fig. 4.8). What, then, is the significance of the relationship between the cave style and materials used in the wall paintings? As discussed in the previous section, caves with square-vaulted Laternendecke ceilings were probably modelled on wooden structures, such as beams. This suggests that the drying oil painting technique was originally used to paint

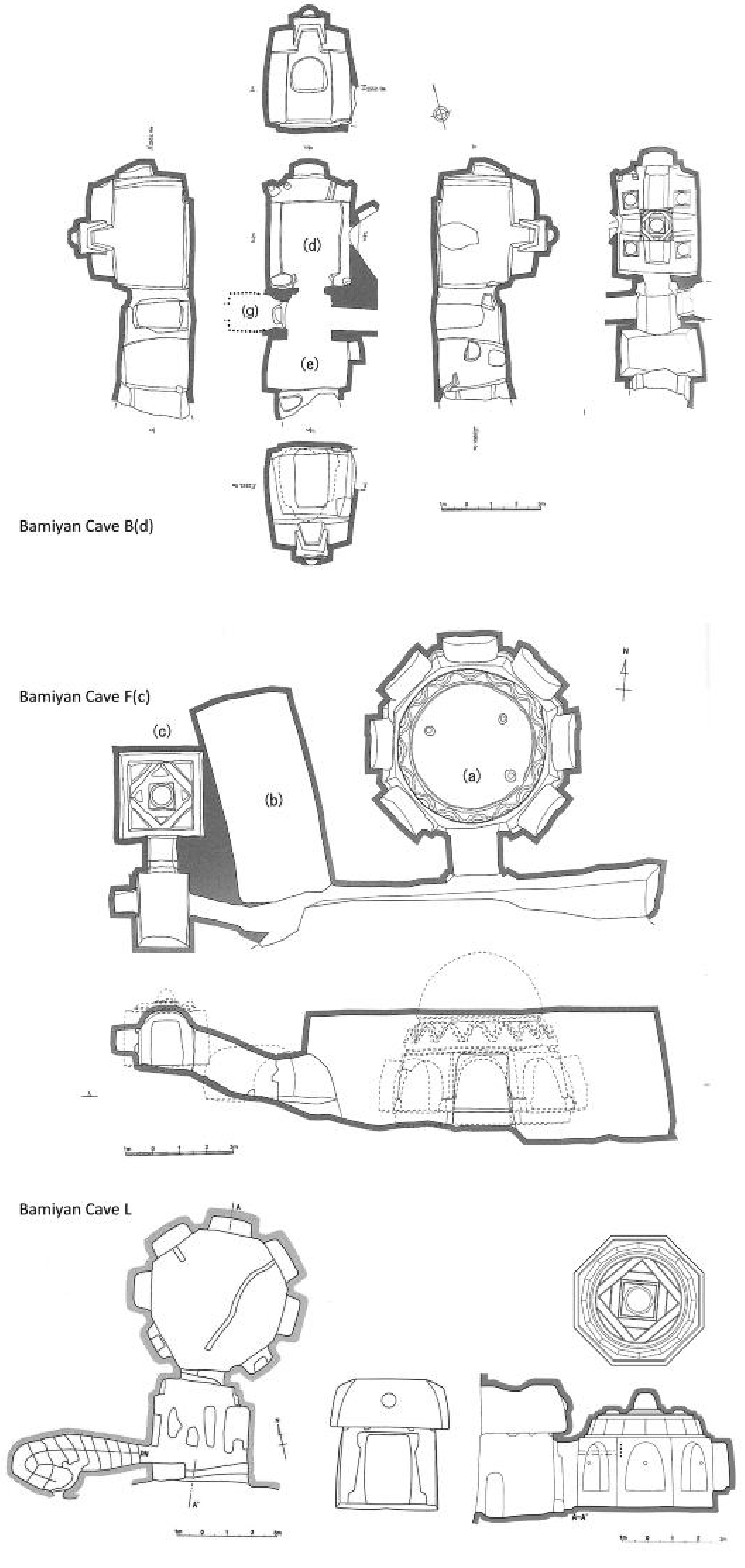


Figure 4.9 Caves with Laternendecke ceilings in Bamiyan: B(d), F(c) and L (after Iwade and Kubodera 2013: 108, fig. 272; 67, fig. 144; 81, fig. 189).

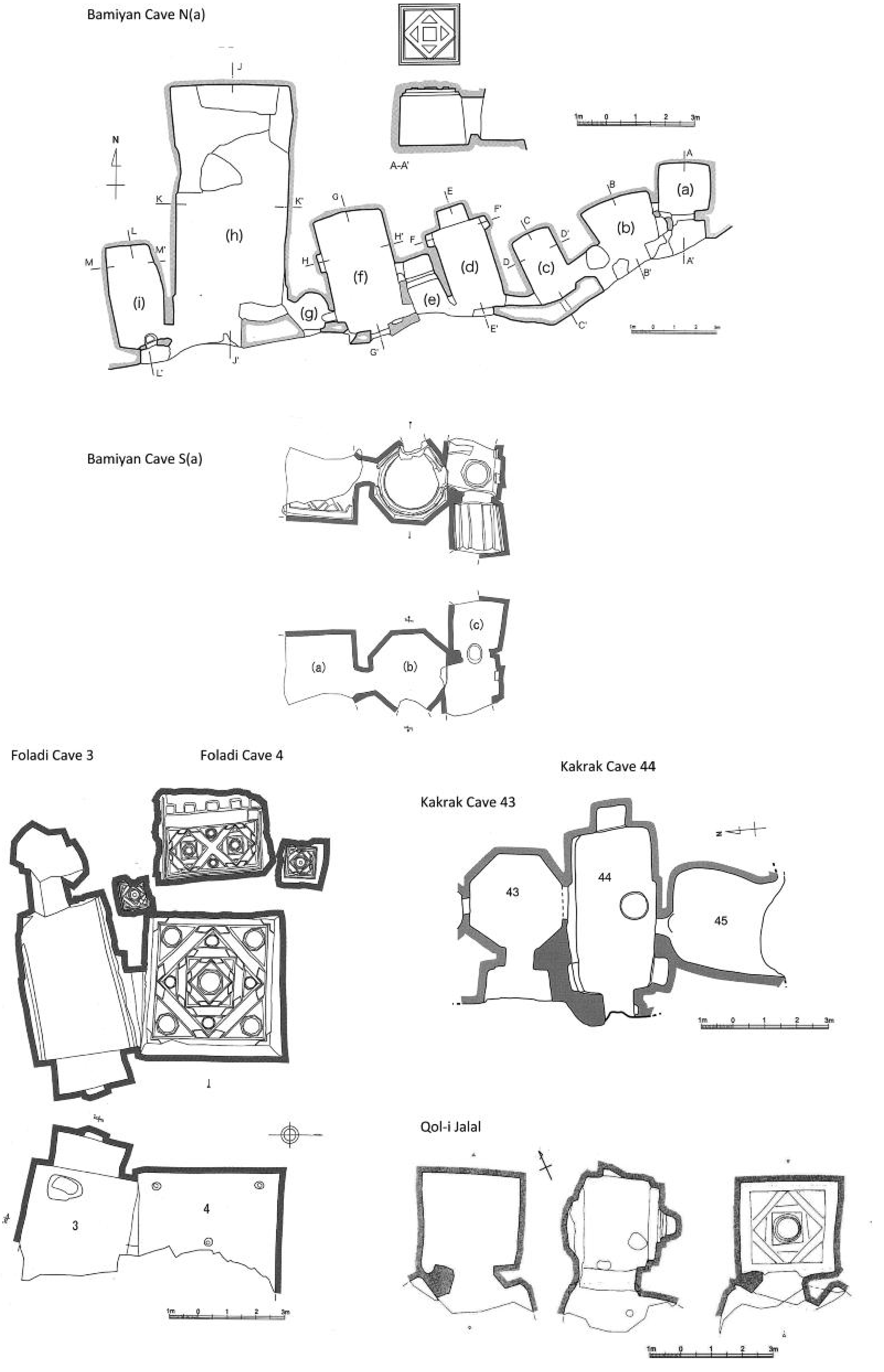


Figure 4.10 Caves with Laternendecke ceilings in Bamiyan: N(a) and S(a), Foladi Caves 3 and 4, Kakrak Caves 43 and 44, and Qol-e Jalal (after Iwade and Kubodera 2013: 166, fig. 457; 141, fig. 376; 267, fig. 592; 215, fig. 614; Kubodera and Iwade 2006: fig. 1).

Table 4.2 Styles of caves with wall paintings. Bold/underlined text indicates that drying oil was identified as a binding medium (after Iwade and Kubodera 2013: 249, table 7).

Plan type		*Ceiling type*	*Transition part*	*Example of caves*
Centred plan	Square	Dome	Only cornice	East III
			Squinch (in tambour)	A lower salle, E(e), J(g)
			Squinch (in dome)	G
			Corbel	J(b), J(c), J(d)
			None	E(d)
		Lanterndecke	None	**F(c)**, H(b), **N(a)**, **S(a)**, **Foladi 4**, 5, **6**, **Qol-e Jalal**
		Flat	None	J (f), M
	Octagon	Dome	Only cornice	**Kakrak 43**
		Lanterndecke	Polygonal tambour	**L**
		Cross-vault	Cornice (+ tambour)	D
	Circle	Dome	Cornice (+ tambour)	C(a), C(b)
	Unknown	Dome	Unknown	East detached
Oblong plan	Rectangle	Vault	None	K, **Kakrak 44**, **Foladi 3**
		Cross-vault	None	**B(d)**
Cave niche				I, H(a), E(c)

wooden structures. Even today, in China and other areas of East Asia, oil-based paints such as tung oil have historically been used to protect the colours and wood of external wooden structures from the elements. It is likely that the same materials and techniques used to paint wooden Buddhist temples were also employed to paint the caves as a mode of imitation. Currently there is no evidence to support this hypothesis as no painted wooden materials have been excavated from surrounding monuments of the same period. Nevertheless, there is more to learn as excavations of above-ground temples with painted wooden structures are likely to take place in the future in the Tarim Basin, Kashmir and their surrounding areas.

Caves containing oil wall paintings are located across the central and eastern cliffs of the Bamiyan main cliffs but there does not appear to be any pattern in their distribution. Similar caves are also found in the Foladi, Kakrak and Qol-e Jalal valleys, a little farther away. The distance of the caves from the Bamiyan main cliffs suggests that these Buddhist temples may have been deliberately placed in remote locations. The oil wall paintings appear to be crafted in a detailed, time-consuming manner, and would have taken a relatively long time to complete. The process of making Buddhist paintings may also have been part of a religious practice, and the long hours spent on painting these mandala-like images in the small caves would probably have required a tranquil environment.

It is possible that those responsible for these wall paintings had professional experience in painting wooden structures. It is difficult to conduct a comparative study of the monastery complexes of the period as they have all been destroyed, but their relationship to the traditions and techniques of the mountainous regions of Kashmir and northern India, where wooden temple buildings were common, can be examined.

4.2 Buddhism and paintings in Eurasia: artisans, trade and technology

4.2.1 Trade in painting materials

This study found that some Buddhist wall paintings at Bamiyan, especially those in oil, are secco paintings with a very sophisticated multilayered structure and a variety of pigments. The relationship between regional exchange and the transportation of painting styles can be verified by the physical evidence of trade in painting materials. In particular, precious pigments with unusual tones could have been acquired through long-distance trade. The type of pigment needed depended on the type and characteristics of the painting itself, but the artist's origin may have influenced the choice of painting materials. The quality of materials chosen would also have been impacted by the economic circumstances of the donor.

The Pamir Plateau in Central Asia is rich in metal resources such as gold, silver, zinc and lead. Sogdian silverware, for example, is a product of silver's abundance in the surrounding area. The profusion of metals also means that the region has plenty of natural minerals in various colours, derived from transition metals such as mercury and copper. The exchange of goods between the East and the West between the 5th and 9th centuries, when Bamiyan's wall paintings flourished, is not well documented. As a result, there are many unanswered questions about specific goods and their origins. However, an overview of the provenance of painting materials used in Central Asia and the Mediterranean world would be helpful.

Information on the origin of minerals used to produce certain precious pigments, as well as the raw materials needed to synthesise them, has been recorded by many scholars and writers. For example, Theophrastus (4th century BC), in his treatise *On Stones*, mentions lapis lazuli from Afghanistan, cinnabar from Iberia and Colchis, and red and yellow ochre from Cappadocia (Caley and Richards 1956). He repeatedly mentions the scarcity of blue and devotes a large part of his article to *kyanos* blue (probably Egyptian blue), which was synthesised in Egypt. He also mentions the natural minerals *sandaraki* (realgar), *arrenikon* (orpiment) and chrysocolla (copper silicate) (Caley and Richards 1956: 53), all of which were widely used in Greece and Egypt. Lastly, the artificial production of lead white and verdigris (copper acetate) is also described.

On the subject of chrysocolla, Dioscorides (1st century BC) states that the best are those (*chrusokolla*) from Armenia followed by those from Macedonia and Cyprus (*De materia medica* 5: 104) (Osbaldeston 2000: 797). Chrysocolla may also be the same material that Theophrustus calls *tanoi*, Bactrian stone and green turquoise, describing it as from Cyprus and an island lying off the ancient town of Chalcedon (Caley and Richards 1956: 50, 102–103). Looking at examples from the Hellenistic period, at Delos and Persepolis, orpiment, realgar, sulphur flowers and other pigments were found in earthenware bowls (Kakoulli 2002; Stoduski *et al*. 1984). These brightly coloured, hard-to-find pigments are believed to have been imported from specific regions by painters or specialist artisans (Kakoulli 2007).

When considering the movement of goods and trade in this region, it is important to refer to the various trade networks that existed between the Mediterranean world and India, Africa and Persia in the Greek and Roman periods. One source of information on trade is the *Periplus Maris Erythraei* (*Periplus of the Erythraean Sea*) (Murakawa 1993; Shitomi 2016a,b), a commercial guide written by an unknown author who describes in detail the kinds of goods exchanged in each port city along the coast of the Erythraean Sea (the Red Sea, Arabian Sea, Persian Gulf, Indian Ocean and the Bay of Bengal), and what goods were relayed at each port. These include not only the raw materials for luxury jewellery, but also pigments, resins and other materials that could serve as painting materials.

At that time, trade between the Roman Empire and India was carried out by means of a pelagic route between the southern coast of the Arabian Peninsula and the western coast of the Indian subcontinent. The monsoon winds, known as the Hippalus (the southwest monsoon wind), were used to connect the southern Arabian Peninsula with the southwest coast of India. The Indian Ocean has southwesterly winds in summer and northeasterly winds in winter. As a result, 'a network of shipping and trading activities was established linking the trading ports of the Indian Ocean and transporting people, goods, culture and information safely, reliably, regularly and therefore inexpensively between the continents' (Yajima 1993: 103–104).

One of the reasons for the increase in trade between Rome and the East was the growing demand for luxury goods such as perfumes and silk following the unification of the Mediterranean world by Rome in 27 BC. At the same time in the East, the Han Empire was trading goods from the South China Sea to the Indian Ocean. These goods were brought to the Mediterranean world from India, while commodities also travelled in the opposite direction. Thus, the Mediterranean was closely linked to the Indochina Peninsula and China.

The Parthians interfered with trade between China and Rome between 247 BC and AD 224. At this time, the Kushan dynasty, which ruled the north and south of the Hindu Kush Mountains, sought a trade route to the Indus River in the south. Once they found it, silk cloth from China (Ceres) and goods from Central Asia were shipped down the Indus River from Gandhara and Taxila, and from Barbaricon (Karachi) to Rome by sea (Machida 1992: 163). The *Periplus of the Erythraean Sea* describes trade between Rome and the Indian Ocean in the first century AD, when various goods were bought and sold by merchants from the East. These included Indian pepper, sesame oil, *nardos*, *lycium* and various other incenses, vegetable spices and medicines; ivory, pearls, precious stones (agate, etc.), tortoiseshell, black indigo, *kinnabari* (dragon's blood), lac (sticklac) and other pigments; iron and steel; and *sappeiros* (lapis lazuli) from Central Asia, *kalyāṇa* (Callaina),[1] and silk and cotton cloth from Ceres.

Figure 4.11 Location of the lapis lazuli mine, Badakhshan, Afghanistan.

These were shipped from Barbarikon at the estuary of the Indus River in northern India.

In exchange for these valuable goods from the East, Rome brought to India large quantities of gold and silver coins, glassware, silverware, wine, frankincense, coral, copper, tin, lead and other minerals, *sandalaki* (realgar) and flax cloth (Murakawa 1993: 65–70). At that time, tin and lead were 'extracted with great labour in Spain, and throughout all the Gallic provinces' (*Naturalis historia* XXXIV, ch. 49) and as stated by Pliny (and also in the *Periplus of the Erythraean Sea*), 'India has neither copper nor lead, but she procures them in exchange for her precious stones and pearls' (*Naturalis historia* XXXIV, ch. 48).

There is a theory that direct trade between the Mediterranean world and India declined after the 3rd century with the rise of the Sasanian dynasty and the growing influence of Persian merchants and the Ethiopian kingdom of Aksum in the Indian Ocean (Shitomi 2016a, b). However, it is natural to assume that the trade routes once opened continued to exist and that Persian and Arab merchants continued to exchange goods including various painting materials such as lapis lazuli.

From the end of the 6th to the end of the 8th century, trade between the East and the West was active in Central Asia under the rule of the Gokturks. The powerful Tang dynasty was at the centre of economic and cultural trade in the East. A study of exotic items brought to the Tang by the East–West trade is detailed in Schafer (1963). Trade routes at this time crossed the Indian Ocean and overland along the Silk Road. It is often unclear which items were transported from which regions, but the port city of Seerah (now Bandar Taheri) on the eastern shore of the Persian Gulf, near the Abbasid capital of Baghdad, was the centre of trade in the East, using the monsoon winds to conduct maritime trade via the Persian Gulf, the Indian Ocean and the South China Sea (Schafer 1963: 11–13). Incenses, natural resins and minerals from Central and West Asia

Figure 4.12 Processed lapis lazuli as a pigment from the Mogao caves, Dunhuang, China (after Mizuno *et al.* 1996).

Table 4.3 List of pigments found in Bamiyan's wall paintings.

Colour	*Pigment/colorant*	*Mineral name*	*Chemical formula*
Whites	Gypsum	Gypsum	$CaSO_4.2H_2O$
	Anhydrite		$CaSO_4$
	Chalk	Chalk	$CaCO_3$
	Lead white	Hydrocerussite/cerussite	$2PbCO_3.Pb(OH)_2$/ $(PbCO_3)$
		Laurionite	$PbCl(OH)$
		Phosgenite	$Pb_2Cl_2CO_3$
		Anglesite	$PbSO_4$
		Susannite	$Pb_4SO_4(CO_3)_2(OH)_2$
		Palmierite	$(K,Na)_2Pb(SO_4)_2$
		Leadhillite	$Pb_4SO_4(CO_3)_2(OH)_2$
		Blixite	$Pb_8O_5(OH)_2Cl$
		Cotunnite	$PbCl_2$
	Kaolinite	Kaolinite	$Al_2Si_2O_5(OH)_4$
Yellows	Goethite (yellow ochre)	Goethite	α-$FeO(OH)$
	Orpiment	Orpiment	As_2S_3
	Massicot	Massicot	β-PbO
	Litharge	Litharge	α-PbO
	Gamboge		natural resin from Guttiferae trees
Reds	Minium (lead red)	Minium	Pb_3O_4
	Haematite (red ochre)	Haematite	α-Fe_2O_3
	Cinnabar	Cinnabar	HgS
	Vermilion	Cinnabar	HgS
	Realgar	Realgar	α-As_4S_4
	Red lac resin		resinous secretion of lac insects (*Kerria lacca*). Laccaic Acid A
	Madder lake		$C_{14}H_8O_4$
Blues	Lapis lazuli (ultramarine)	Lazurite (calcite, pyrite)	$Na_7Ca(Al_6Si_6O_{24})(SO_4)(S_3) \cdot H_2O$
	Azurite	Azurite	$2CuCO_3.Cu(OH)_2$
	Indigo		$C_{16}H_{10}N_2O_2$
Greens	Malachite	Malachite	$CuCO_3.Cu(OH)_2$
	Atacamite(/paratacamite)	Atacamite/paratacamite	$Cu_2Cl(OH)_3$／$Cu_3(Cu,Zn)(OH)_6Cl_2$
	Verdigris (copper acetate)		i.e. $Cu(CH_3COO)_2 \cdot H_2O$
	Verdigris (copper resinate)		$Cu(C_{19}H_{29}COO)_2$
	Chrysocolla	Chrysocolla	$Cu_{2-x}Al_x(H_{2-x}Si_2O_5)(OH)_4 \cdot nH_2O$, $x < 1$
	Celadonite (green earth)	Celadonite	$K(Mg,Fe^{2+})(Fe^{3+}Al)Si_4O_{10}(OH)_2$
	Glauconite (green earth)	Glauconite	$(K,Na)(Fe^{3+},Al,Mg)_2(Si,Al)_4O_{10}(OH)_2$
Blacks	Bone black		$C + Ca_3(PO_4)_2$
	Charcoal black (carbon black)		C
	Lamp black		C
	Magnetite	Magnetite	Fe_3O_4
Metal foils	Gold leaf	Gold	Au
	Tin leaf	Tin	$Sn + Pb$ (impurity)

were imported to the Tang dynasty as medicines, as mentioned in various Chinese texts.

The Batakhshan region, where lapis lazuli is mined, is about 500 km northeast of Bamiyan (Fig. 4.11), which suggests that it was relatively easy to obtain lapis lazuli. In fact, many lapis lazuli gemstones can be seen in the bazaars of Bamiyan today. However, the quality of refined lapis lazuli pigment appears second grade when compared to the lapis lazuli blue in Cave 224 of the Kizil grottoes for example. Lapis lazuli, on the other hand, was a major trading commodity, sought from as far away as the Mediterranean world. It also migrated to the East: a lump of pigment made from gypsum, lapis lazuli powder and glue was found in front of Caves 53–55 at the Mogao caves, Dunhuang, and attributed to the

Five Dynasties (Fig. 4.12). This was a so-called ready-made paint that could be used for painting as soon as water was added. In the Dunhuang Testament S3553, a letter written by a camel driver to a monk mentions the 'golden blue' (lapis lazuli) brought by the camel driver to the Mogao caves (Mizuno *et al.* 1996). Thus it is possible that painting materials were not transported in their original mineral state, but rather in processed and ready-made forms.

It is also likely that the painters of the time had a good knowledge of the geological characteristics and vegetation of each region, as well as the specific materials available in each production area. A wide network may have been in place to facilitate the procurement of pigments from different regions, depending on the economic circumstances of the artist in question. Table 4.3 reviews the pigments found that were possibly in use in Bamiyan: of these, gypsum, lime, red ochre, yellow ochre and carbon black/lampblack were probably the most readily available materials in and around Bamiyan. Lapis lazuli was also available in Bamiyan, some of which would have undergone a refining process to extract the blue mineral lazurite, which was processed into pigment by a rather sophisticated professional method. Similarly, gold leaf, tin leaf, lead white, red lead, vermilion and possibly atacamite are artificially produced by specialised processes.

4.2.2 Preliminary discussion of the origin of lead-based pigments based on lead isotope analysis

Because walnut trees and poppy plants grow widely in Central and West Asia, the presence of their oils in oil paintings provides no clues as to the origin of the raw materials. When it comes to lead, however, we may identify the origin or source of lead pigments based on the analysis of lead isotope ratios. Lead allows us to determine a likely provenance because isotope ratios offer information about the mine of origin.

Minium and lead white are synthetic pigments produced by artificial oxidisation and carbonation. Strips of metallic lead are place in earthenware pots and exposed to acetic acid (vinegar). After some months, the acetic and carbonic acid reacts with the lead forming a white crust – lead white. Red lead can be produced from metal lead, heated to 600 °C with metal lead to become PbO; PbO_2 can then be produced with further heating to 400–600 °C. In the Mediterranean world, for example, lead is obtained in large quantities from mines such as the Laurion mine near Athens, but there is no documentary evidence to demonstrate whether lead from these mines was used in the Greek and Roman periods or in the Hellenistic period (Kakoulli 2009: 44).

Numerous provenance studies using lead isotope analysis have been conducted since the 1960s, often on archaeological finds such as bronzeware. However, very few have been carried out on lead-based pigments. One study of the use of lead white in 17th-century oil paintings by artists such as Rembrandt, Van Eyck and Rubens analysed over 200 samples and found that the sources of lead were mines in England and Germany (Fabian and Fortunato 2004).

Robert H. Brill, a well-known glass researcher, conducted similar analyses on pigments used in wall paintings at the Mogao caves (Brill *et al.* 1997). His analysis also covered materials held by the Fogg Art Museum at Harvard University, which included wall paintings originally found near the ceiling of the niche surrounding the Western Giant Buddha, the relief decoration of the Tianlong Mountain caves and the painting of stone statues in Cave 6 of the Yungang caves, all initially collected by Benjamin Rowland. Brill also analysed the lead isotope ratios of ingots from the Shahr-i Sokhta (Iran) site in Central Asia, modern lead sulfate from Herat (Afghanistan), and metal artefacts from the Farinjal (Afghanistan) metal-production site in an attempt to compile basic data on lead isotopic ratios in Central Asia.

Another example was carried out as part of a joint research project between the Conservation Institute of Dunhuang Academy and the National Research Institute for Cultural Properties, Tokyo (Zhao *et al.* 2007). For the Mogao caves, pigment samples were taken from 18 caves spanning the Northern Wei, Western Wei, Northern Zhou, Five Dynasties, Song, Western Xia and Yuan dynasties (4th–14th century): Caves 5, 25, 40, 95, 249, 259, 260, 265, 272 (2 samples), 275, 285 (2 samples), 290, 296, 334, 352, 365 and 477. Most of the lead-pigment samples were blackened. In addition, plattnerite, a discoloured product of minium, was detected.

Figure 4.13 shows a re-plotting of data from the analysis by Brill *et al.* (1997) for the area around Afghanistan. The lead isotope ratios of metal artefacts and deposits from the Farinjal site are also plotted in this figure. At a glance, it can be seen that the lead isotope ratios of pottery from the same Farinjal site appear close the plot of Brill's samples. Since these pottery sherds are thought to have been produced in

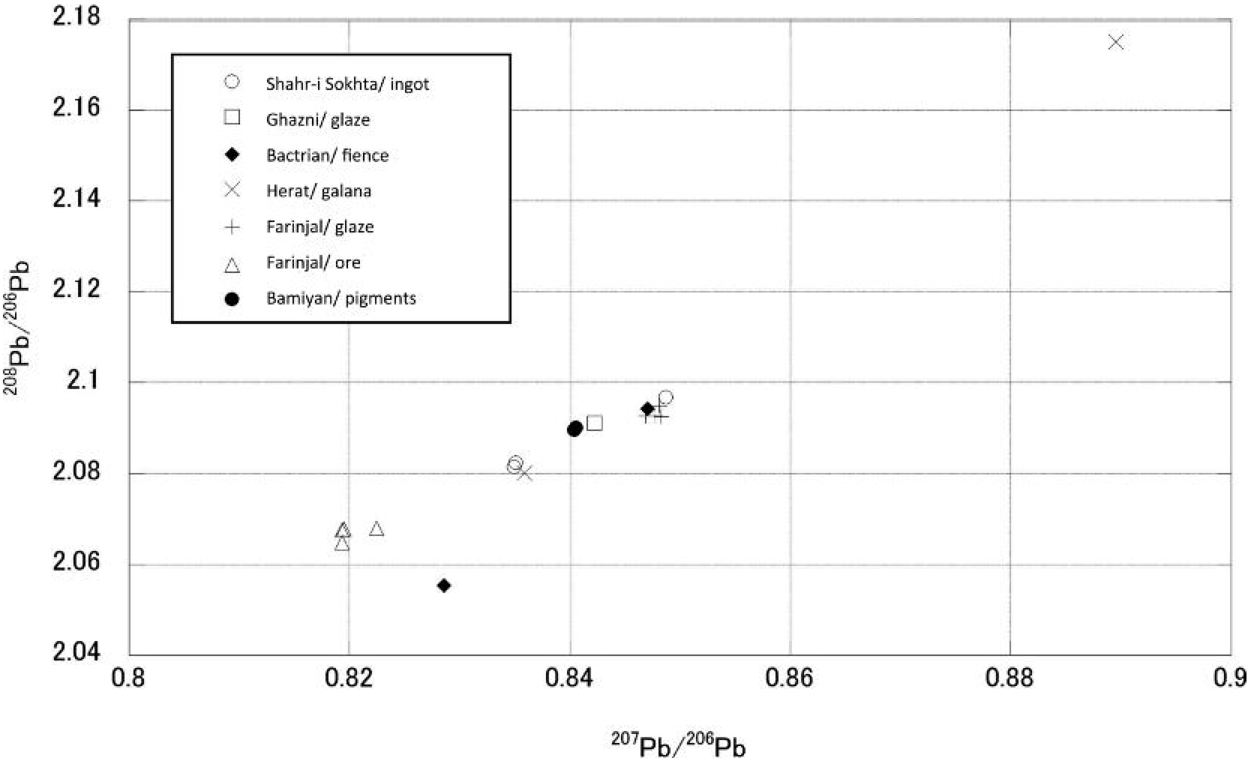


Figure 4.13 Lead isotope ratios of lead-containing materials from Central Asia (based on data from Brill *et al.* 1997).

Iran, it makes sense that they might not be related to those of the other items from the site. Similarly, in the same area, a concentration of lead isotope ratios in the glaze on blue beads of an unknown date excavated from Bactria can be seen, relative to those of lead sulfate ore (a raw mineral component of lead) dating from 2200–1800 BC excavated from the Shahr-i Sohta site in Iran. This suggests that glazes on Iranian pottery from Farinjal may have been produced using a raw material similar to lead sulfate, which was available in the vicinity of Shahr-i Sohta.

Next, the isotopic ratios of two pigment samples from the Bamiyan site are plotted in very close proximity to each other. Lead-based pigments used at Bamiyan show lead isotope ratios that are far from those of the metal deposits at Farinjal, but still close to the isotope ratios of the lead glaze on the glazed animal statues purchased by Brill at Ghazni. However, since pigments, unlike lead ore, can travel long distances, it would be premature to draw any conclusions from these limited results.

Figure 4.14 plots the lead isotope ratios of lead pigments from the Bamiyan wall paintings by Brill overlaid on top of lead isotope ratios from Central Asian wall paintings including Dunhuang's Mogao caves. In broad terms, the wall paintings from the Mogao, Tianlongshan, Yungang and Bamiyan caves seem to cluster together, but the overall isotopic variation is extremely high. As these sites are thousands of kilometres apart, it is natural that lead of different origins would have been used. In any case, an absolute lack of data on lead isotope ratios in Central Asia as a basis for comparison makes it difficult to directly link the results obtained to places of origin. However, if there is a cohesion between periods and sites, this may reflect the selective use of certain pigments by networks of painters or explain how specific pigments could have been made in manufacturing workshops.

For the wall paintings at the Mogao caves, the lead isotope ratios of Northern Wei and Western Wei are plotted adjacent to each other. It appears that a particular type of red lead from one of these regions was used at that time. On the other hand, lead isotope ratios from the Yuan dynasty are plotted at widely divergent points. Ratios from the two Bamiyan samples appear in very close proximity to each other, suggesting that they are almost identical. As both samples were taken from the blue and red parts of the niche surrounding the Western Giant Buddha, it is clear that they were both painted by the same group of painters at the same time. Therefore, it is not surprising that the lead – either lead white or red lead – is of the same origin.

Central Asia's geology, as evidenced by the Pamir Plateau and Batakhshan, provided access to a wide variety of metal resources. These may have been transformed into many kinds of pigments used

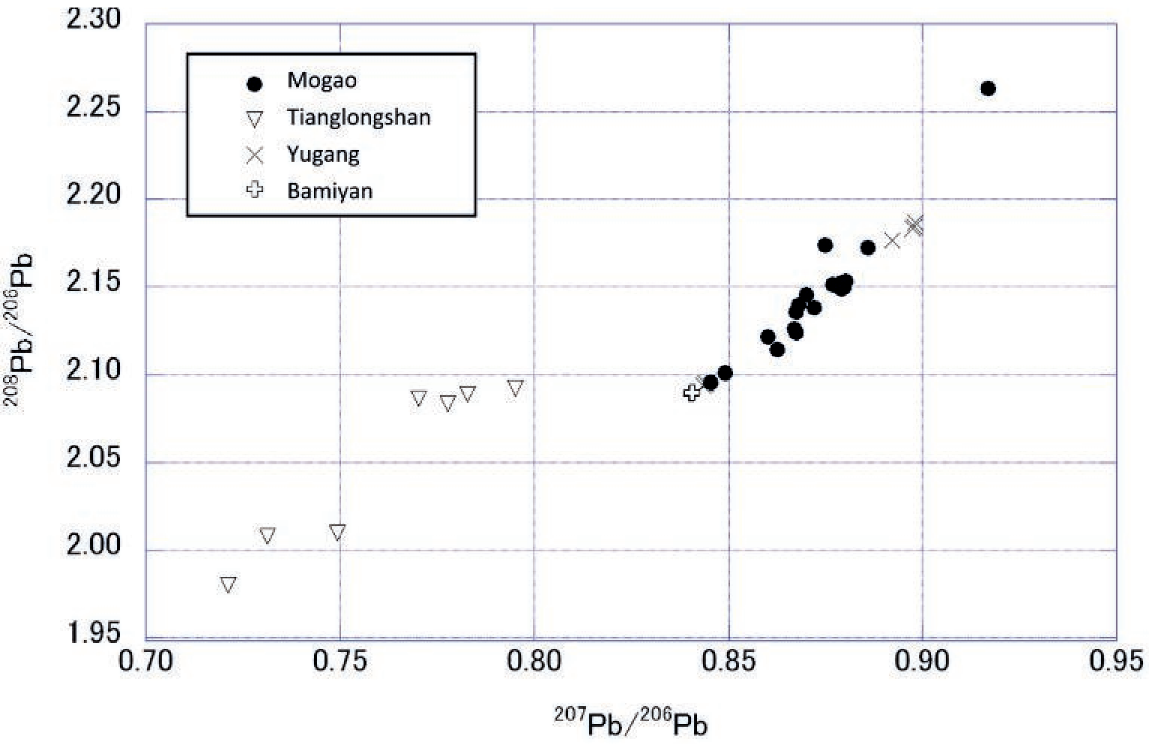


Figure 4.14 Lead isotope ratios of lead-based pigments used for painting materials along the Silk Road.

widely along the Silk Road. Considering the limited number of previous studies, the use of lead isotope ratios may prove to be very useful for understanding the trade and transportation of goods such as lead-based pigments. As a foundation, more basic data should be accumulated for reference on the surrounding areas and lead-based pigments.

4.3 The history of oil painting: a review of its origins and development

4.3.1 Drying oil as a binding medium

Findings thus far in this book have shown that a significant number of Bamiyan's Buddhist wall paintings did indeed use drying oil as a binding medium. These wall paintings were probably painted between the mid-7th and the 9th century. The other earliest known example of an oil painting that contained drying oil as a binding medium is a late 12th-century painted statue in Hemse Church in Gotland, Sweden (Tångeberg 2006). It was once generally believed that the earliest oil painting techniques appeared on the Scandinavian peninsula, and that the Flemish school established oil painting in the 15th century.

Outside of Bamiyan, there are no known examples of the use of drying oils in painting prior to the 12th century, which means that more than 400 years separate their use in each of Central Asia and Northern Europe. The correlation between these two important points in history has not been made clear in art historical and technical historical studies to date. The purpose of this section is to explore the origins and development of painting techniques involving drying oil.

Oils are usually classified as drying, non-drying or semi-drying. As linseed, poppy seed and walnut oil all gradually begin to dry when exposed to air, they are called drying oils. Conversely, oils that remain liquid and do not dry out, such as olive and salad oil, are called non-drying oils. Unsaturated fatty acids, a main component of drying oils, have several double bonds in their molecules which gradually combine with oxygen in the air and become oxidised, producing radicals and peroxides. As the polymerisation reaction between double bonds progresses, the oil molecules bond to each other to form a polymer with a large molecular weight. As a result, the oil eventually loses its fluidity and the oil coating becomes solid and dry.

Oil paints are made by mixing powdered pigments with a binding medium made primarily of drying oil. Moreover, the technique of painting with oil paints is referred to as oil painting. When the oil is fresh and the double bonds have not yet been polymerised (oxidised), the paint retains good fluidity: the oil holds the pigment particles together and helps them to settle on the canvas. The thickness of paint layers and shades of colour can be adjusted by adding natural resins or essential oils to the paint. Once the paint has been applied, the drying oil absorbs oxygen and hardens, holding the pigments in place. Once fixed, the pigment particles do not dissolve again.

However, the mechanisms work differently when animal glues or plant gums are used as binding media: they are water-soluble and their solutions are viscous. When pigments are mixed with these binding media and applied to the surface, the liquid gradually evaporates (or is absorbed) and dries. Finally, any remaining glues or plant gums trap the pigment particles and fix them to the surface. Despite their hardened surface, however, the pigment particles may become loose and the paint may dissolve if water is added once again.

Analysis of the aforementioned late 12th-century painted wooden statue at Hemse Church in Sweden detected drying oil with a P/S ratio of 2.2 to 2.6 (Plahter 2006). In this report, linseed oil is suggested as the drying oil, but not distinctly identified. The P/S value of linseed oil is usually around 1.2, so the drying oil detected here may not have been linseed oil, but perhaps another kind of drying oil such as soya. The woodwork of the church was painted with a white base comprised of drying oil and lead white.

The Flemish school, represented by the Van Eyck brothers in the 15th century, began to use drying oil as an adhesive for painting in earnest, and it is said to have been used in Italy from the mid-15th century onwards, replacing the egg tempera technique. The Flemish school was characterised by its use of a white ground layer and a thin coat of paint made from linseed oil mixed with resin as a binding medium. However, this is because the records of Giorgio Vasari have been passed down unverified (Eastlake 2001: 183). Literature in modern history had, in fact, recognised the Flemish school as the first to systematically establish the use of drying oils in painting.

The origins of oil painting techniques are an important historical subject that should be re-examined. For the time being, however, some examples of the uses of drying oil can be extracted from extant literature. Drying oils have been known at least as far back as Greek and Roman times. The very earliest known drying oils appeared in texts as medicines. Dioscorides (1st century) describes various vegetable oils as medicines in his *De materia medica*, including walnut (1-178) and poppy seed oil (4-65) (Osbaldeston 2000: 176, 607). Pliny (*Naturalis historia* XV, ch. 7) also mentions a number of vegetable oils – besides the main oils of the time pressed from olives and castor beans – such as oils from bitter almonds (*Amygdalinum*) and walnuts (*Caryinum*) (Otsuki 2009a: 165–168). However, these oils were intended for medicinal purposes, such as cleaning blood and improving its colour (*Naturalis historia* XV, ch. 33) (Otsuki 2009b: 212).

The earliest known example of drying oils used in painting and crafts is thought to be in the medical writings of the Greek physician Aetius Amidenus (late 5th–6th century) (Eastlake 2001: 19–20). In addition to its medicinal uses, walnut oil, obtained by crushing or pressing walnuts, was used to protect gilding and encaustic painting. Thus, it appears that by this time, walnut oil was known to have drying properties (oxidative polymerisation) and that this quality could be used to develop a protective layer for art and craftwork such as gold leaf and encaustic painting. This kind of protective varnish could also be made into a gold-coloured varnish by adding yellow or red dyes or resins, similar to the '*mecca*' technique used in medieval Europe at least by the 8th century to make tin or silver leaf resemble gold leaf.

As mentioned in the analysis of patterns on the ceiling of Cave N(a) (Chapter 3, section 3.1), the materials and techniques used in golden varnishes have been described in Latin and Greek texts. For instance, the date and authorship of the original work is unknown, but the *Codex Lucensis* (Lucca Manuscript), thought to date from the 8th century, contains instructions for mixing linseed oil, galbanum, turpentine, Spanish pitch and saffron, frankincense (olibanum frankincense), myrrh, mastic, fir resin (balsam), maturing poplar bloom and betney (cuckoo cherry) to produce a bright varnish for painting (Burnam 1920: 101–102). The text also includes reference to other mixtures for making varnishes including one of linseed oil, turpentine, lac resin, frankincense, myrrh, mastic, betony, cherry gum, poplar flowers, almond gum and fir resin (Burnam 1920: 104). In addition to plant gums and various resins, the text also introduces linseed oil as a drying oil.

Eraclius, in *De coloribus et artibus Romanorum* (Merrifield 1967b: 166), describes a process of making a golden varnish called *auripetrum* by heating a mixture of 'linseed oil, the bark (of vesprum), myrrh, aloes, sandalac or amber' (Merrifield 1967a: 114). The common feature is that the drying oil is not used as a single substance but mixed with plant resins and gums. One reason why analysis of organic matter in Bamiyan's wall paintings has not led to clear findings may derive from this simultaneous use of a variety of organic materials (Taniguchi *et al.* 2022).

References to drying oil as not only a golden varnish, but also a binding medium for painting, can be found in various medieval European texts, including the *Coloribus et artibus Romanorum* as well as *De diversis artibus* by Theophilus (11th–12th century) (Nicolaus 1979 [1985]). Another representative example is found in *Il libro dell'arte* by Cennini (Thompson 1960), who describes how to prepare linseed oil, mix pigments with linseed oil and apply an oil-based glaze on egg tempera to finish a painting. However, there are no known examples of Asian texts describing the use of drying oil in painting. Some pertinent other texts include the Indian *Citrasutra*, which mentions a 'decoction of animal skins (animal hide glue) [or] *bakula* tree resin' (ch. 40, verse 29) as a material for adhering pigments (see Appendix I). It is not clear to what the *bakra* tree refers, but the resin obtained from it may be a plant gum or resin. There are also references to sesame oil and safflower oil as organic substances that can be added when rendering and sizing walls, but as these oils are non-drying they do not oxidise and polymerise to form a coating.

In the medieval Persian and Asian regions, however, the fact that oil can be obtained by crushing walnuts has long been known, as demonstrated by the reference to 'walnut oil' in the *Sino-Iranica* (Laufer 1967: 266–267). This oil in particular was recognised for its use as a lamp oil and cooking oil but is not discussed in connection with painting. Walnuts, known as 'hu' peaches in Chinese ('hu' refers to Persians), grow wild in much of present-day Iran and Central Asia. Similarly, flax and poppy were also readily available plants growing wild in Central Asia and India. It is not known whether this technique of using drying oil existed in South and Central Asia outside Bamiyan, as only a few chemical analyses have been carried out. However, some artefacts among the Western treasures in the Shōsōin Repository[2] in Nara, Japan, are known to have been made using drying oil as discussed in the next section.

The absence of the use of drying oil as a binding medium in South Asian sutras does not mean that drying oil was not used in the Asian world since raw materials for drying oil, such as perilla or tung, were available in Central and South Asia. There is currently no evidence to link the oil painting techniques of Bamiyan's wall paintings directly to drying oils used in Greek and Roman arts and medicines. However, future research and analysis of wall paintings, artefacts and wooden buildings from Kashmir, Ladakh and the Tarim Basin should reveal positive links between techniques in the northern Indian region and elsewhere. Usually, the surfaces of early European oil paintings appear stiff and cracked due to dehydration and shrinkage. Although similar cracks can be observed in Bamiyan's paintings, the paint layers are somewhat softer. Their multilayered structure containing resins and glues appears to have protected them against physical deformation in the paint films. In this case, it would seem that this knowledge and technique was not transferred to medieval Europe.

4.4 Correlation with cave style

4.4.1 Mitsuda-e and Yūshoku

Among the Shōsōin treasures is a group of artefacts known as *mitsuda-e.* This term is now commonly used to refer to objects painted with a mixture of oil and pigments but the origin of the concept or the name is not clear. Today's state of research on *mitsuda-e* owes much to pioneering work carried out between 1950 and 1953 by Rokuro Uemura, Tsutomu Kameda, Koichi Kimura, Daitsu Kitamura, Kazuo Yamasaki and other lacquer specialists and chemists, using literature review and chemical analysis. In addition, the Tamamushi-no-zushi Shrine (the Lacquered Chest Decorated with Dragon and Tiger) in Shōsōin was examined by Gonroku Matsuda, Jō Okada, Saburō Mizoguchi, Daitsu Kitamura, Hirokazu Arakawa, Shinryu Sekine and Norimitsu Kimura in 1970 using a UV light (Matsuda 1975). These studies are extremely important as they are probably the only comprehensive studies of the Shōsōin *mitsuda-e* using historical and scientific methods.

According to Uemura's research, the name '*mitsuda-e*' may have come into use based on knowledge of 19th-century artefacts, i.e. similarities between some Shōsōin treasures and *mitsuda-e*, which were made by mixing lead monoxide (PbO), led to a generalisation of the latter term (Uemura *et al.* 1954a,b). These lacquered pieces were originally made by mixing perilla[3] oil and pigments instead of lacquer. Yellow powdered *mitsuda-sou* (PbO) acted as a drier when added to perilla oil. While boiling the mixture, the lead oxide powder dissolves in the oil, leading it to quickly dry and solidify. The method of boiling tung or perilla oil with *mitsuda-sou* was described in detail as early as 1660 in the *Buryozappitsu* (Uemura *et al.* 1954a). From these documents, Uemura concludes that from the end of the Edo period to the beginning of the Meiji period (1868–1912), crafts using oil containing *mitsuda-sou* came to be known as *mitsuda-e*.

Tadashi Sekino described *mitsuda-e* as 'a type of lacquer painting in which pictorial designs of flowers, plants and animals are applied to a lacquered surface using a mixture of oil, *mitsuda-sou* (as a siccative) and pigments like vermilion, minium, ochre and malachite' and noted that it appeared from the Asuka period onwards (Sekino 1947). He also recorded that it is found on both sides of the door of the *Tachibana fujin nenjibutsu zushi* (miniature) of Hōryū-ji Temple, as well as in the treasures of the Shōsōin and Tōdai-ji collections. However, Sekino did not have the means at that time to determine the exact type of material used in the Shōsōin treasures nor whether drying oil or *mitsuda-sou* was used, so he probably argued the above points on the basis of similarities in appearance with early modern *mitsuda-e*.

Apart from this, Uemura also examined the technique of applying coloured oil on painted or metal surfaces, called *yūshoku* (oil colouring). *Yūshoku* is a painting technique whereby an artist applies a transparent oil layer over black lacquer or a painted surface. It was introduced to Japan from the Tang Empire (710–794) and is said to have been exemplified by the *Kachosaieyūshokubako* (an oil coated lacquer box with a flower and bird design) at Tōdai-ji Temple. Uemura assumed that the *yūshoku* technique would have involved the application of colourless or coloured oil. He pointed out that, at least in the Nara period (710–794), there may have been two main techniques – oil and pigment painting – or to phrase it differently, the 19th-century *mitsuda-e* technique from the literature and *yūshoku*, which involves applying a layer of oil only to the surface (Uemura 1954a). In a description of his success replicating the *mitsuda-e* technique, Hosyun Yoshida states that it is indeed an oil painting technique that mixes pigments and tung oil (Yamauchi 1946).

Matsuda (1975) describes one form of Japan's early oil paintings known as *Tōyu-makie* with an example found in the panels of the Edo-era Yōmei-mon gate in the Nikkō Tōshōgū Temple (17th–18th century). However, this oil painting technique is thought to be rooted in Western influence, carried through Jesuit and Franciscan missionary activities. The Kano school of painters belonging to the Franciscans was closed to the Tokunaga shogunate, who imported oil paintings and their techniques from abroad (Nakau 2019). This technique should therefore be considered as separate from the *mitsuda-e* technique.

4.4.2 Mitsuda-sou as a drying accelerant and siccative

The Japanese term *mitsuda-sou* refers to a pale yellow powder with a chemical composition of lead monoxide. It may have been obtained as a natural mineral or possibly produced by oxidising lead, as done in the synthesis of red lead. There are two types of lead oxide: the tetragonal α-type and the orthorhombic β-type, known as litharge and massicot, respectively. Both have been used as pigments although their histories as pigments are largely unknown (Eastaugh *et al.* 2004). It is important to note that the use of *mitsuda-sou* – not only as a pigment but also as an oil siccative – may suggest the possibility that 'drying oil' and '*mitsuda-sou*' might have originated in the Iranian world with the same meaning.

Because of its high reactivity, when lead monoxide is added to drying oils with unsaturated fatty acids and heated, the oil saponifies to form a lead soap. Moreover, the process accelerates the formation of a dried oil coating. For this reason, lead compounds such as *mitsuda-sou* have until recently been used as drying accelerants (siccatives, driers) in oil-based paints and inks, apart from their use as yellow pigments that easily alter colour. Because of the small amount of lead required for a siccative effect and due to its high reactivity with oil, PbO does not significantly colour the cooked oil depending on the ratio of PbO to oil. It is worth noting that the name of the yellow lead monoxide powder was known in ancient China and Japan as '*mitsuda-sou*', a word of Persian origin. According to Laufer, a scholar in Asian Studies who conducted a comparative study of Chinese and Iranian vocabulary, the word '*mirdäsaṅg*' (equivalent to *mitsuda-sou*) did not emerge until the Tang dynasty. However, the term '*mirdäsaṅg*' (*m'it (m'ir)-dasaṅ*) is the same as '*murdäsaṅg*' (*mut(mur)-ta-saṅ*) (*Zhèng lèi běn cǎo*).

Su Gong, the compiler of the Tang dynasty textbook *Chinese Material Medica*, states that both '密陀' and '没多' are 'from Persia and [resemble] dragon's teeth in shape but [are] stronger and heavier'.

He also observes that some *mitsuda-sou* are white with stone crests, like those found on Yunnan marble (Laufer 1967: 508–509). Furthermore, according to Su Song of the Song dynasty (960–1279), *mitsuda-sou* was also found in the silver and copper foundries of Guangdong and Fujian; the book of *Ben cao yan yi* states that the best ones are those whose colour resembles gold (Laufer 1967: 508–509). This powder was also used as a drier for oil-based paints and as an ingredient in ointments for the treatment of wounds caused by metal weapons (Schafer 1985: 220).

4.4.3 Oil painting technique of the Shōsōin treasures and the Tamamushi zushi of Hōryū-ji Temple

As mentioned earlier, Japanese scientists Rokuro Uemura, Kazuo Yamasaki and their colleagues conducted scientific research on the Shōsōin treasures in the 1950s. They examined 27 Shōsōin objects with the aim of identifying their manufacturing techniques and materials for the benefit of art historical, pharmacological and scientific knowledge (Uemura *et al.* 1954a,b; Yamasaki 1999). Among these, the pharmacologist Kimura Koichi confirmed the presence of oils in the Shōsōin artefacts based on the fact that organic materials emit different fluorescence under UV light. Animal glue, for example, is revealed by a strong blue-white fluorescence, oil of perilla (or *mitsuda* oil) by a yellow fluorescence and lac, an animal resin, by a strong orange fluorescence.

The chemist Kazuo Yamasaki conducted a spectroscopic analysis on part of a sutra scroll (known as *Jingo-Keiun ni-nen Gogankyo* or the Scrolls of Shogozo Repository) dating from AD 768. The analysis confirmed the presence of lead in its oil. Yamasaki (1999) suggests that *mitsuda-sou* served as a drying accelerant in the preparation of this oil. Naruse and Iida (2005) conducted a similar examination of the same scrolls using XRD and XRF analysis. White, red and green colours covered by an oily coating can be seen on the scrolls' shafts. Lead-based white pigments (lead white), such as lead carbonate and lead chloride hydroxide, were detected in the white part of the shafts, known as the 'white *mitsuda*'. Similarly, red pigments made from minium and iron oxide were identified in the red part of the shafts, known as the 'red *mitsuda*' (Yamasaki 1999). Lead was also detected in areas coloured by the iron red pigments. Malachite and lead elements were present in the green areas.

These findings support the hypothesis that lead detected in the red iron oxide areas and the green areas was added to the oil as a drier. When lead white or minium is employed as a pigment, it is harder to determine if lead siccative has been used because of the lead in the pigment itself. Therefore, if lead is detected where the oil and the iron- or copper-based components were used, it must have been present in the oil; this is critical evidence that the oil included lead-based driers. The survey by Yamasaki *et al*. used the most advanced analytical methods available in the 1950s, but there were methodological limitations in determining the types of oil, therefore it is not known whether the oil detected here is perilla oil, tung oil or a completely different type of drying oil.

It is also unclear whether the lead in the oil comes from *mitsuda-sou* or from another lead compound such as lead white or red lead. Our distinction between *mitsuda-e* and *yūshoku* here is based on UV fluorescence reactions bearing similarities to oil and the detection of the element lead (usually by SEM-EDS), as well as an overall judgement in light of historical documents and archival research. A more detailed chemical verification has long been anticipated but analysis of organic substances is difficult because of the need for samples which contravenes the current mainstream preference for non-destructive analysis. Yamasaki *et al*'s survey of 27 objects revealed that seven were made using a technique similar to early modern *mitsuda-e*, in which oil and pigments were used to make paint. In addition, 17 pieces were identified as having been painted with oil or oil with lacquer, a technique known as *yūshoku*.

These 17 pieces and a few fragments comprise the Mitsuda-e Tray, which is preserved in the South storage of the Shōsōin Repository. They are executed in the style of the Tang dynasty in terms of their iconography, including figures under trees. It is thought to have been wheel-thrown and shaped from zelkova wood, then coated with black lacquer on both the inside and outside. After the lacquer, lead white was applied to the inside and painted with yellow pigment, then covered with a thin oil-based coating. On the outer surface, four pieces are painted with oil kneaded into the pigment using the *mitsuda-e* technique, while the rest are painted with the *yūshoku* technique (Fig. 4.15) (Nara National Museum 2007).

According to the observations of the lacquer-master Hoshun Yoshida, the differences in the brushwork and the presence or absence of cloth covering in the patterns on these 17 pieces of the Mitsuda-e Tray suggest that they may not have been made by the same artisan (Yoshida 1941). Thus, it may be said that both techniques were adopted selectively

Figure 4.15 Shōsōin Treasures, Mitsuda-e Tray (No. 14) (after Nara National Museum 2007).

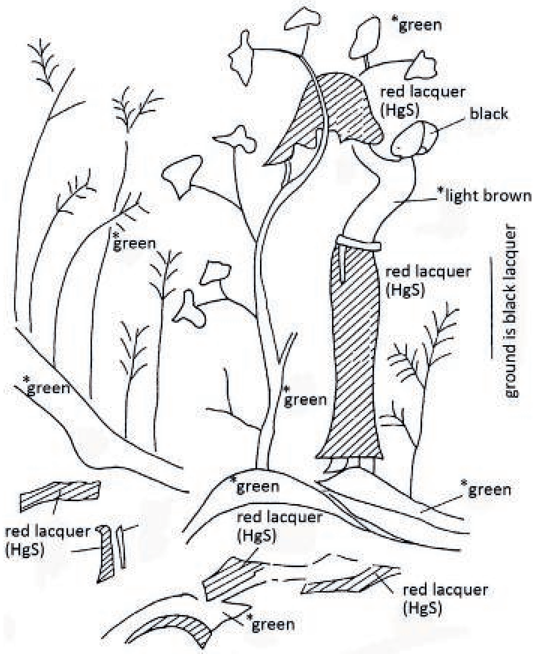


Figure 4.16 Sketch of the *Story of Prince Sattva's Self-Sacrifice to the Tiger*, painted on the Tamamushi Shrine (*Tamamushi no zushi*), Hōryū-ji Temple showing the colours used. The colours that fluoresced under UV light are indicated by * (after Yamasaki 1999).

according to the intended use and the artist's preferred techniques.

Yamasaki also examined the *Story of Prince Sattva's Self-Sacrifice to Tiger* painted on the seat of the Tamamushi Shrine (*Tamamushi no zushi*), Hōryū-ji Temple, using UV light (Yamasaki 1985, 1999). It is painted in red, green and yellow (changing to light brown) on a black lacquer ground; the green and yellow areas show localised fluorescence (Fig. 4.16). It is assumed that the fluorescent areas were painted with some kind of oil and pigment mixture. The colours of the Tamamushi Shrine are very similar to those of the *mitsuda-e* paintings in the Shōsōin treasures. Another similarity between the two is that a black lacquer was applied directly to the wooden vessel before decorations were painted on top.

Based on these results, Uemura suggested that the name *mitsuda-e* used for the Shōsōin treasures was not appropriate, since *mitsuda-sou* was probably used only in the early modern period. He also asserted that the use of oil paint was more important than whether or not the drying accelerant *mitsuda-sou* was used. Therefore, he concluded that it would be more appropriate to call the technique in this work *yūshoku* (Uemura *et al.* 1954a,b). Based on the results of their research into the Tamamushi Shrine and the Mitsuda-e Tray pieces, Uemura and his colleagues have confirmed the existence of East Asia's earliest oil paintings, which are older than those in the West. Ultimately, they trace the origin of the use of oils to the black pottery technique of painting with a mixture of oil and carbon, as seen in China's Shang, Qin and Han dynasties. In *Yu fu zhi* of the *Book of the Later Han* (AD 25–220), and the *Book of Jin*, there are references to red oil, green oil, blue oil and other oil colours, suggesting that *mitsuda-e* and *yūshoku* originated in ancient China (Uemura *et al.* 1954a). *Mitsuda-sou* was created in China to serve as a drier in drying oil. Because its Chinese name is a transliteration from the Persian language, it was probably introduced to Japan during the Asuka period, or at least before the Nara period, together with actual objects such as those represented in the Shōsōin treasures. While these techniques are certainly oil painting techniques in that they use pigments mixed with drying oil as paint, they are only localised and do not have the structure of a drying oil base or layered colouring. In this respect they may be differentiated from the oil painting techniques found in Bamiyan and post-medieval Europe.

4.4.4 Bamiyan's oil painting technique, the 'mecca' technique and the mitsuda-e

The use of drying oil is common to many techniques including the Bamiyan oil painting technique described in this chapter, the *mitsuda-e* or *yūshoku* technique introduced to Japan, the technique for oil paintings disseminated in northern Europe, and techniques that use organic substances mixed with drying oil to paint, as seen in Greek and Roman texts on procedures for making golden varnish. Despite similarities in their use, there is no evidence of any direct links between them.

A clear yellow resin, similar to frankincense, has been found on some of the tin foil on the ceiling of Cave N(a), producing a golden effect that makes patterns and images glow. A comparison of this decorative technique with the '*mecca*' technique confirms that both involve applying a yellow or red coloured material made from natural resin or oil, along with some organic substances, to the tin foil to give the appearance of gold leaf. As mentioned above, the 8th-century *Codex Lucensis* was considered to contain the first evidence of this technique, but even older references to the use of drying oil as a protective layer exist such as the *Sixteen Books on Medicine* by the Greek physician Aetius Amidenus. In this text, he describes using beeswax in encaustic painting as a binding medium and walnut oil as a protective layer over gold leaf. Bamiyan's oil painting techniques exhibit a clear similarity to Mediterranean artistic practices with regard to the use of walnut oil and tin foil. However, the oil used in East Asian arts was probably perilla oil or tung oil, rather than linseed, walnut or poppy seed oil. Moreover, the kinds of drying oils commonly used in East Asia have not yet been identified owing to the paucity of GC-MS analysis on these materials to date. Thus, a direct comparison with Mediterranean and Central Asian examples is not possible.

One key difference in techniques across different geographical areas can be seen in how the Shōsōin treasures, such as the Tamamushi Shrine and the Mitsuda-e Tray, are painted over a black lacquer base, which is very different from the characteristic lead-white ground found in Bamiyan's wall paintings. The origin of the black lacquer technique can be traced to mainland China, as seen in the colouring of terracotta warriors and bronze mirrors. Another difference is in the method of oil painting in *mitsuda-e*. i.e. in *mitsuda-e*, pigments are mixed with drying oil and used as paint that is employed only in specific limited places, whereas both Bamiyan and post-medieval European techniques involve the use of alternating layers of drying oil and paint, applied over an entire surface. At the same time, both Bamiyan wall paintings and *mitsuda-e* do use a mixture of drying oil and pigments to make paints. Drying oil also had uses as an adhesive (mordant) for tin foil and was mixed with lead white as part of the white ground, as seen in the animal pattern on the ceiling of Cave N(a).

In other painting layers, lead white was mixed with other pigments (atacamite, etc.). Lead-based pigments are also used in *mitsuda-e* paintings, such as lead white for white areas and minium for red ones. In *mitsuda-e* painting, a small amount of massicot (lead oxide) or some lead-based substance is used as an oil drier. The process of making *mitsuda* oil involves adding a small amount of *mitsuda-sou* (lead oxide) to drying oil, heating it and then filtering the oil. It is therefore difficult to determine the origin of the lead in the oil as often no particles of lead oxide remain after it has been heated (Cotte *et al.* 2006b).

Although no data exist that definitively indicate the use of lead drier in oil in Bamiyan's wall paintings, there is a high content of lead in the oil. Moreover, the presence of lead soaps where oil is found is also noteworthy. However, as most of the oil paintings are made using lead white or minium, it is not clear whether the lead soap is the result of mixing lead white with drying oil or heating oil with *mitsuda-sou*. When lead white or red lead is used as a pigment, it is difficult to determine by analysis whether or not any lead derived from *mitsuda-sou.* It is also unclear whether the use of lead white or red lead would have negated the need to add *mitsuda-sou* to the oil, as these too have the siccative function of drying oil. Whether the lead in Bamiyan's wall paintings derived from lead white or minium, or if it was the result of the deliberate addition of driers such as *mitsuda-sou*, the oil paints used in Bamiyan were highly suitable for painting as they hardened readily.

One additional feature of the use of lead white in ground and painting layers is that only a small amount of oil is required when mixing lead white with oil to make paint. This is because, unlike calcium-based white pigments such as lime or gypsum, lead white has a low oil absorption capacity. Even if lime or gypsum is mixed with oil, it will appear transparent and will not function as a white pigment due to its refractive index. Lead white also has an advantage in that its coating adheres better to other layers and cracks less. In consideration of these material attributes, a combination of lead white and drying oil would have been an inevitable choice. At present, no other wall paintings using lead soaps have been found in Central, East or South Asia, but few studies have applied the analytical methods necessary to detect these compounds. New analyses of the surrounding regions would be beneficial.

As mentioned above, the most pronounced commonality between Bamiyan's wall paintings and the *mitsuda-e* found in the Shōsōin treasures is the use of drying oil and lead-based pigments such as lead white and red lead, both of which are inorganic compounds obtained by producing a reaction in metallic lead. The refinement of these inorganic compounds, combined with the use of drying oils, led to the development of their respective oil painting techniques. At the same time, the use of walnut oil and coloured tin foil share similarities with Mediterranean practices. All of these examples date to between the 6th and 8th century, therefore it is difficult to say with any certainty which oil painting technique is the earliest. The practice of mixing vegetable oils with artificially created lead-based pigments enable any oil to be transformed into a 'drying' oil, thereby making the technique materially viable.

The history of the use of drying oils for medicinal purposes and the use of lead white suggests that these practices can be traced to the Mediterranean world. However, the richness of organic materials and the use of multilayered structures, as seen in Bamiyan's wall paintings, could not have been achieved without using materials and techniques from northern India and the Iranian world.

Previous literature has discussed the Kuchean manufacture of vegetable oils in Kucha using unknown seeds ('mlyokotau') which could be jute batching oil (*Corchorus capsularis*) but not sesame oil (Ching 2017: 102–105, 158–159, 166–167, 366–377), but painting materials using drying oils have not been identified in the historical texts. Little research has been done on this history of painting, and evidence to support or undermine these theories regarding geographical origin has not yet been found. There is also no evidence of oil painting in Iran, where the drying oil originated. The search for the material and technical roots of oil painting across the Eurasian and African continents is therefore an ambitious challenge that needs to be addressed.

4.5 Relationship with Buddhism

Unfortunately, Xuanzang's *Great Tang Records on the Western Regions* (AD 646) does not mention anything specific about how Bamiyan's temples or wall paintings were made. However, he does describe the 'standing image of the Buddha', thought to be the Western Giant Buddha, and the 'standing image of Buddha Shakamuni made of brass', probably the Eastern Giant Buddha (Kuwayama 2002: 154) (as discussed in Chapter 3, section 3.5). However, apparently, both figures were carved out of rock and then detailed by adding a thick kneaded clay to the exterior, which was then shaped and painted. The Western Giant Buddha was painted red and the Eastern Giant Buddha red, blue and yellow. However, in his text, Xuanzang stated that the Western Giant Buddha was golden and the Eastern Giant Buddha was a cast bronze. Nevertheless, the records do suggest that Bamiyan was somewhere traders passed through regularly, and that the king and nobility, inspired by their deep faith in Theravada Buddhism, used the riches gained from trade to make donations to the monks. It is likely that they used their wealth to construct temples and monasteries, recruit artisans to paint Buddhist works in Bamiyan, and procure materials for the wall paintings.

Huichao also visited Bāmiyan around AD 726–727 and left his account in *Huichao's Wang Wu-Tianzhuguo Zhuan Record of Travels in Five Indic Regions*, one of the Dunhuang documents (Yang *et al.* 1984: 51–52; Kuwayama 1998). However, the king of Bamiyan at that time was a Buddhist 'Hu' or of Iranian descent and, even 100 years after Xuanzang's visit, many monasteries and temples remained, suggesting that Buddhism was flourishing there. It is noteworthy that Huichao's account mentions the existence of both Theravada and Mahayana Buddhism. This raises the possibility that there may have been numerous artisans in the same Buddhist city, with different origins and backgrounds from different Buddhist schools and sects. Analysis to date has revealed the existence of at least three phases of wall painting at Bamiyan, and two very different kinds of painting techniques.

The initial phase begins in the 5th and 6th centuries, when Buddhist caves were first being created in Bamiyan. Artisans influenced by Greek and Roman art and the Sasanid Empire produced wall paintings in the niche surrounding the Eastern Giant Buddha. As can be seen in Caves C, M and J, there was a tendency to leave the uneven surface

Table 4.4 Phases of wall paintings, ground and binding media.

Phase	*Date*	*Ground layer and binding media*	*Caves*
Phase I	5th–late 6th century	Gypsum ground and water-soluble binding media	EGB, Caves J(b), J(d), J(g), M
Phase II	early 6th–mid-7th century	Gypsum ground and water-soluble binding media	Caves C(a), C(b), D1, S(a), A lower salle, H(a), WGB
		Gypsum ground and water-soluble binding media	Caves E, K_3, I
Phase III	mid-7th–late 8th century	Lead white with drying oil as a binding media	Caves B(d), F(c), N(a), S(a), L, Foladi Caves 4, 6, Kakrak Cave 43

* Dating indicated here is based on the ^{14}C result of Nagoya University (Nakamura 2006), however, there are ongoing debates in history and art history over the date of the earliest phase of Bamiyan.

of the render as it is, unpolished. There is no clear correlation between the painting techniques used in Bamiyan at this time and those in India such as Ajanta. The techniques of the Gupta dynasty incorporated a wide variety of organic materials in the creation of wall surfaces and in the production of a wide range of colours.

It is difficult to compare Sasanid wall paintings with those of Bamiyan because so few examples of wall paintings exist in Iran. However, some motifs in Bamiyan's wall paintings, such as the pearl roundels, do appear similar to Sasanid motifs. The few Sasanid wall paintings we have today, such as those at Kuh-e Khwaja, show strong dynamic lines, very different from the fine, iron-wire lines of Bamiyan's wall paintings. Few studies exist on materials used in Iranian wall painting so much is unknown today, but it is likely that the *a secco* technique and a limited range of ochre colours were used for paintings. The earthen renders of Sasanid wall paintings are similar to those of Bamiyan's first phase, where the surface was not smoothed, but painted in a rustic style. It is not clear how Buddhism spread to the peripheral regions of Bamiyan during this first phase, but it is possible that the rise of minorities such as Haphthalites and the fall of Gandhara were the main catalysts.

Later, in the second phase, wall paintings seem to exhibit diverse influences from a variety of cultures and regions, such as Gupta, Gandhara, Sasanian, Central Asia and the Tarim Basin, as seen in the niche surrounding the Western Giant Buddha and Cave H. In particular, they strongly reflect the style of late Gupta art, which was a revival of the Hindu spirit in India. The wall paintings of this period also use gypsum as a white ground and water-soluble binding media made from proteins and plant gums. No significant difference in materials or techniques exists between the two styles of painting. Late Gupta wall paintings share much in common with Bamiyan's wall paintings from the second phase onwards, such as attention to three-dimensional depictions and foliated scroll patterns enmeshed with animals.

Most art found in Pakistan, especially in the region of Gandhara, has taken the form of stone sculptures and stucco work with few examples of wall paintings. Those that have been found appear to have used the *a secco* method, similar to Ajanta rather than Bamiyan. On the other hand, the wall paintings of Sogdiana in Central Asia were not painted in a religious context such as a Buddhist temple, so a direct comparison with Bamiyan's wall paintings requires caution. However, several techniques are reminiscent of those found in Bamiyan's first and second phases, as seen in the niches of both Giant Buddhas and the caves around the Eastern Giant Buddha. These include the monotony of the colours, the use of Sasanid motifs, the iron-wire strokes, the use of gypsum as a white ground layer, and the application of a black layer underneath a blue layer of lapis lazuli.

The Tarim Basin area also has wall paintings that share similarities with techniques used during Bamiyan's second, third and fourth phases such as iron-wire lines. Specifically, this can be seen in the Buddhist monuments on South Tianshan Road and wall paintings around Kucha, such as those at Kizil, Kizilgaha and Simsim. The Bamiyan wall paintings of this period reflect a technical and material connection with Central Asia and the region around the Tarim Basin, including with the Sasanian empire and Sogdiana. This second phase coincides with AD 630 when Xuanzang is said to have visited Bamiyan. According to his account in the *Great Tang Records on the Western Regions*, there were several thousand Buddhist monks in Bamiyan who studied the Lokottaravāda or Theravada in dozens of monasteries (Kuwayama 1987: 20). *A Biography of the Tripitaka Master of the Great Ci'en Monastery of the Great Tang Dynasty* also mentions that Mahāsāṃghikavinaya monks stayed at Bamiyan (Nagasawa 1988: 81). It seems likely that there was at least some relationship between the sect and the choice of subject matter, but a more detailed iconographical analysis is necessary to confirm this.

As the period progressed from the mid-7th to the 8th century, the motifs of Greek and Roman Sun Gods and princes in Central Asian dress disappeared. Then, in the third phase, a group of wall paintings with similarities in their lines and seated Buddha compositions appeared, as seen in Bamiyan Caves B(d) and F(c) and the Foladi and Kakrak caves. The wall paintings of this third phase are characterised by the introduction of oil painting techniques and, as found in the second phase, the use of water-soluble binding media, in common with Central Asian wall paintings. These wall paintings also have square-vaulted Laternendecke ceilings, lead white grounds, multilayered structures and the use of metal leaf (‘*mecca*’ technique). They are also characterised by mandala-like compositions with Thousand Buddha arrangements, very precise brushwork and the use of rich colours. It is believed that these artists who painted seated Buddhas in extremely colourful mandala-like compositions travelled throughout the Bamiyan Valley and its surrounding valleys to offer their knowledge of techniques and materials for painting wooden structures to the small-scale cave monasteries.

Bamiyan’s paintings from this era differ starkly from those of the late Gupta Empire, represented by the *Citrasutra*, which did not use vegetable drying oils such as poppy seed or walnut. Instead, paintings of the late Gupta dynasty used paints such as watercolours and therefore do not seem to be connected to oil painting. However, it is difficult to state from where these artisans who painted such exquisite mandala-like wall paintings came as there are no other clear examples of their work. For instance, the wall paintings around Khotan along the Southwest Passage are considered to be a link between Sasanid art, Kashmir and Tibetan Buddhism and the techniques of the late Gupta dynasty. Looking again at the contemporaneous records, Huichao, who visited Bamiyan in around AD 726 or 727, wrote in *Wang Wu-Tianzhuguo Zhuan Record of Travels in Five Indic Regions* that two schools of Buddhism existed, Mahayana and Hinayana, as did ‘customary rules similar to those in Kapisi, despite the language being isolated in Bamiyan’ (Kuwayama 2002: 157). Interestingly, Bamiyan’s king at that time was of Hu or Iranian descent.

Theravada Buddhism had flourished under the Kushan dynasty in Kashmir and probably Ladakh (Devers 2020).[4] Buddhism was introduced to Tibet from northern India in the 7th century. Tibet’s first king, Songtsen Gampo (AD 581–649), introduced Buddhism by inviting Buddhist monks from India. However, at that time, esoteric Buddhism was thriving in northern India so naturally the Indian monks he invited were also esoteric. Tibetan Buddhism was probably strongly influenced by Mahayana Buddhism.

Wall paintings from Bamiyan’s third phase share a number of common features such as their use of lead white and drying oil, graduated colouring, multi-layered structures, Laternendecke ceilings and the use of mandala-like compositions. These qualities are very different from earlier Bamiyan traditions, and also differ from the traditions of Central Asia, India, the Tarim Basin and the Persian world. As Klimburg-Salter points out, they may be related to the emergence of Shāhī art from northwest India in the foothills of the Hindukush (Klimburg-Salter 1989: 138).

We do not know exactly what kind of Buddhism Huichao saw: Mahayana or Theravada. However, it is possible that the Thousand-Buddha designs that he observed in Bamiyan’s Laternendecke ceilings looked like esoteric Buddhism to him. There is no definitive evidence of esoteric iconography or other elements in Bamiyan’s wall paintings or scriptures. However, as more is learned about Buddhism in Bamiyan, the materials used for these wall paintings and the origins of the oil painting techniques will become clearer.

Notes

1. Probably turquoise, a light greenish stone from Bactria.
2. The Shōsōin Repository is a wooden house at Tōdai-ji Temple. It houses arts and artefacts of the Nara era and gifted materials from the Silk Road via the Tang Empire.
3. *Perilla frutescens var. frutescens* is an annual plant of the Perillaceae family and is different from sesame, *Sesamum indicum.*
4. Mahayana was probably also practised in Kashmir. At least from the era following the Kushans, Mahayana dominated. In this paper, Devers analysed archaeological remains of Buddhist monasteries of the vihara type in the region of modern Lower Ladakh (Purig) and Zangskar (part of modern Ladakh). The ruins can be dated to the Kushan period.

Chapter 5: Conclusion

This study was originally launched as part of an urgent effort to preserve Bamiyan's wall paintings, following damage and theft. Over the course of investigating which methods and supplies might be the most appropriate for cleaning and reinforcing the wall paintings, it became necessary to identify the original materials and techniques used to create them. Local conditions made it impractical to carry a portable XRF spectrometer and other equipment for *in-situ* analysis so we decided to collect about 300 microsamples for analysis in laboratories in Japan, Europe and the United States. Using as many different scientific methods as possible, we discovered that the Bamiyan wall paintings were created through the use of a wide variety of colours and extremely complex techniques. A very small collection of fragments provided a great deal of valuable information at a time when the archaeological sites were threatened by human destruction. They also provided a foundation for the reconstruction of cultural identity in Afghanistan. However, the information obtained from the samples is so voluminous that it would require years to analyse it all.

The wall paintings are made from materials available in the region, such as lime for the frescoes and earth for the walls. The colours applied to the walls – a combination of materials, techniques and skills – were available both locally and transmitted along trade routes. Artistic ideas, skills and painting materials were evident in the wall paintings created by a mobile network of professional artisans such as the *eboshi*, or Buddhist monks, who specialised in painting, and others spread throughout the region. This reflects a technical background that differs slightly from that of worshippers or religious sects and tribes.

For this study, we conducted a comprehensive survey of Buddhist wall paintings from four cave complexes in the Bamiyan Valley using analytical methods designed to clarify the painting materials and manufacturing techniques used. Over the course of this text, we have discussed our findings from a perspective that makes explicit the multidirectional exchanges that have taken place over time between the so-called East and West. Bamiyan's Buddhist wall paintings grew in number over a period of approximately 500 years. However, its cave monasteries took shape in varying ways at different points in time influenced by shifts in Buddhism and the different dynamics of trade and exchange. Therefore, it came as no surprise that dominant techniques and practices also changed.

Bamiyan's wall paintings were dated using a combination of radiocarbon dating and analysis of the technical styles and materials used. We discovered that Bamiyan's wall paintings were created between the 5th century and the late 9th century, a span that can be divided into four phases. The first phase, from the 5th to the late 6th century, includes wall paintings in the niche surrounding the Eastern Giant Buddha, which were made by artisans influenced by Hellenistic, Roman and Sasanian arts. If images in the niche surrounding the Eastern Giant Buddha are somehow related to the Giant Buddha's opening ritual, then they are extremely important. The second phase was marked by a blending of influences from various regions. The third phase, between the mid-7th and late 8th century, witnessed the disappearance of the bold, free-flowing brushstrokes of the Greek and Roman Sun Gods and the Central Asian costumes of the princes, and the emergence of disparate groups of artists who painted seated Buddhas using the style of mandalas with delicate brushstrokes and vivid colours. The fourth phase lasted until the end of the 9th century, by which time the number of wall paintings had declined dramatically. We have shown that since the 5th century, when Buddhist temples were first established in Bamiyan, a variety of wall paintings and cave temples emerged and were shaped by the diverse influences of the Sasanian dynasty, which included techniques and practices from the area around the Tarim Basin and West, Central and South Asia.

In our examination of techniques and materials, we noted many regional changes over time. However, no written records of painting techniques in Bamiyan or the Tarim Basin exist and without such historical texts describing the methods or techniques of the time, it is difficult to substantiate how painters and artisans produced their work. Therefore, in order to learn more about the techniques and materials utilised, we referred to classical texts on painting methods in the Mediterranean world, South Asia and the surrounding areas, while also conducting a direct and comprehensive material analysis of the actual wall paintings.

We introduced prior studies, discussed techniques used in creating wall paintings across Central Asia and conducted physical and chemical analyses of samples collected from Bamiyan. Radiocarbon dating of wall painting renders and a comparative analysis of cave styles were carried out. For physical and chemical analysis of the minute samples taken from the wall paintings, it was possible to detect both organic and inorganic components by using a combination of different analytical techniques. In particular, SR-μXRF, SR-μXRD and SR-μFTIR enabled assessment of small areas and, consequently, a layer-by-layer analysis of paints sitting within very thin layers of only a few micrometres. In addition, the use of GC-MS and LC-MS, as well as the ELISA method, strengthened analysis of organic materials and made it possible to identify specific types of binding material and animals. Our aim was to extract as much information as possible using the most up-to-date methods available in the field of cultural heritage science (Taniguchi *et al.* 2022).

Based on our findings, we developed a genealogy of Bamiyan's wall paintings of each period. Results show that the techniques and materials used reflect two main types of technology. The first involves the use of white gypsum with a water-soluble adhesive such as animal glue or plant gums. Wall paintings that incorporate this feature include the simple colours of the Mediterranean world, the Sasanid dynasty, Sogdiana, the Tarim Basin area and the late Gupta dynasty. These include the polychromies of both Giant Buddhas and the wall paintings on Bamiyan's main cliff.

The second type involves the use of a white ground made of lead white and partially saponified drying oil as a binding medium, with mandala-style compositions. These were painted in the rock-cut caves in the Foladi, Kakrak and Qol-e Jalal valleys, as well as on Bamiyan's main cliff. Interestingly, mineral phases of lead white differed within the layers. We noted the detection of an unusual lead white – hydrocerussite and susannite – and that it was sometimes associated in the same painting but in a different layer with the standard hydrocerussite/cerussite lead white.

This study has confirmed, for the first time, the existence of Buddhist wall paintings that feature white grounds made of lead white and drying oils used as binding media. It also found that the drying oil, with a composition similar to walnut or poppy seed oil, was partially saponified into lead soaps. The latter type of wall paintings was executed using very sophisticated techniques for applying colour, such as the building of layers of different organic substances (natural resins, plant gums, proteins, etc.), and the use of drying oil as a binding medium. This multilayered technique is based on a different principle from that of mixing pigments of different shades, and produces more optically sophisticated colours.

The first appearance of this oil painting technique in Bamiyan is thought to be in the mid-7th century, so these Central Asian wall paintings on earthen plaster represent the oldest examples of oil paintings in the world. We also examined how and why Bamiyan's oil paintings emerged historically by gathering information on their composition, location and ceiling styles.

Many of the paintings are characterised by their association with square-vaulted Laternendecke ceilings, suggesting that they were housed in a 'different' style of cave possibly imitating a wooden structure. The relationship between the oil painting technique and the oil coating of wooden buildings in Asia and elsewhere is one aspect of architectural history that needs to be examined further.

What lay behind the emergence of the unique artistic nature of the caves in the mid-7th century with their oil painting techniques and Laternendecke ceilings? Bamiyan's wall paintings have been interpreted as being 'rooted in a blend of Indo-Iranian Buddhism, esoteric Buddhism and Hindu elements that developed under the power of the Kushano-Sassan dynasty and the Western Turkic Khaganate' (Yasuda 2020). In addition to South Asian, Central Asian and Iranian influences, the influx of Buddhism with early esoteric elements from northern India, such as Kashmir, from the mid-7th century onwards may have had an impact on this period. Buddhist wall (including ceilings) paintings in Ladakh and Kashmir and painted wooden structures such as Buddhist temples in mountainous regions could be studied as comparable examples in future. In order to test our theory and to clarify the origins of the oil painting technique, more research is crucial. We believe that the oil wall paintings identified in this study are not the result of an accidental use of drying oil as a binding medium.

Bamiyan's wall paintings can be differentiated from other Buddhist wall paintings by the materials and painting techniques used. Their sophisticated multilayered structure and the employment of a variety of organic materials appear similar to painting techniques found in the medieval paintings of Northern Europe from the 12th century onwards. We believe that this is a strong reminder of a correlation between the origins of oil painting that cannot be explained by accidental similarities. The techniques of Central Asian, northern Indian and medieval European regions probably co-produced each other.

However, in both the Mediterranean world and Europe, there are no examples of drying oil paintings with a multilayered structure dating as far back as the mid-7th century, although drying oil was used in ancient Rome and Egypt in cosmetics, medicines and to paint wooden ships. The origins of oil painting in the European region are not clear at present, as few analyses have been conducted of pre-medieval paintings. It may be necessary to re-examine the formation of painting techniques and materials in medieval Europe with an eye to Central Asia, northern India and the Iranian world.

Looking towards the East, we also studied painting techniques using drying oil in the Shōsōin treasures of East Asia. One of the oil painting techniques used to paint artefacts, called *mitsuda-e*, is thought to have been used in a wide variety of objects brought to East Asia by Sogdian merchants, probably from the Iranian world, via the Silk Road. The Shōsōin treasures in Japan are one such example. It is known that the term *mitsuda-e* is related to the Persian world '*mirdäsaṅg*' (lead monoxide), which is used as a drying agent for oil. However, the substance to which it refers is unknown because until now, few chemical analyses of *mitsuda-e* artefacts are available, making a comparison impossible.

We would like to assume that oil painting techniques were transmitted both westwards and eastwards from Central Asia, northern India and the Iranian world, but in order to show this empirically, we need more research on painting techniques and materials of the same period across the Mediterranean region, Ladakh, Kashmir, northern India, Central Asia, Western China and surrounding areas. The accumulation of such research may lead to a new understanding of the origin and genealogy of oil painting techniques, specifically regarding where and how these techniques and practices emerged and spread.

This study is the first to detect drying oil in Buddhist wall paintings from Central Asia. Moreover, a series of exhaustive analyses has made it possible to provide a comprehensive picture of how these wall paintings were structured. Bamiyan's oil paintings not only used drying oil as a binding medium, but also involved the sophisticated use of various organic materials, such as resins, plant gums and proteins as animal glues/casein, in multiple layers that included glazing and sizing. The use of lead white, an artificially processed white substance made from lead, together with drying oil, succeeded in achieving a distinctive white colour and also allowed the drying oil to polymerise quickly due to the lead white's role as a drying agent. The technique of applying natural resin on top of tin leaf to mimic gold leaf is also very similar to the medieval '*mecca*' technique. These similarities in materials and techniques are more than accidental or rudimentary – they are historically important evidence of a positive correlation between Central Asia and medieval Northern Europe, where oil paintings were once believed to have originated.

This discovery shows that the history of oil painting can be traced back over a millennium, to at least the mid-7th century in Central Asia. In particular, the fact that the technique was adopted in a Buddhist context in Central Asia raises novel questions for art history, archaeology and history.

Appendix I: The Citrasutra

Ch. 27: S. Kramrisch, 1928, *The Vishnudharmottara (Part III): A Treatise on Indian Painting and Image-making*, Calcutta, Calcutta University Press

Ch. 40 and [] parts: C. Sivaramamurti, 1978, *Chitrasutra of the Vishnudharmottara*, New Delhi, Kanak Publications

Part III, Ch. 27, Verses 7–26.

(Oh) king, I shall now speak to you about the preparation of the principal colours. (Oh) best of kings, there are five principal colours, viz., white (śveta), red (rakta), yellow (pīta), black (kṛshṇa) and green (harit). It would be impossible to enumerate the mixed colours in this world (which are produced by) the mixture of two or three (primary colours) and through invention of various states or conditions (i.e., shades or tones). (Oh) best of kings, now I shall speak to you about the division of dark (śyāma) and white [light colours] (gaura), which is due to the great suitability for getting mixed, of different colours of this world, from which the twofold colour of all is explained (i.e., the light and dark shade of every colour).

Among these (colours), the white (i.e., the light shade) should be of five kinds and the dark of twelve kinds. Bright (gold), light (white), tooth-white, pure-sandal-white, autumn-cloud-white and autumn-moon-white these five traditionally are called the fivefold white (light shade).

(The varieties of śyāma) should be: reddish-dark, mudga (brownish) dark, dūrvā sprout (greenish) dark and grayish dark too, (oh) king, tawny dark and topaz dark, Priyangu-creeper dark and monkey dark. Then come blue-lotus (nīlôtpala) dark and blue as the nīlakaṇṭha bird and purple-lotus (raktôtpala) dark and cloud-dark. Their application is said to be in accordance with the colours of (the respective) objects and they gain in beauty by intermixture of colours.

Having ascertained with precision the colours of deities, I shall speak now about them. Among them, all those of whom I shall not say anything, should be painted white. Vāsuki 1) should be of śyāma (colour), the nāgas should be white in the dvāpara (age), and the daityas, danavas, rākshasas, guhyakas, pisāchas are of the colour of water, without any glow (lit. unglowing by colour). People in the six islands should be of golden colour in the continent of Jambudvīpa, excepting one only, (namely), Bhārata, (oh) king. In Bhārata, (people) born in many countries should be painted. Pulindas and the people of the Deccan are mostly dark by colour, (while) the Śakas, Yavanas, Pallavas and those who are the Vālhikas born in Uttarāpatha should be predominantly white; Pāñchālas, Śūrasenas and those who are of Magadha, Aṅga, Vań ga and Kaliṅga are mostly dark. Twice-born (ones) should be painted of the colour of the moon and the Kshatriyas of the colour of the padma (white lotus). Vaiśyas again should be (only) slightly light in colour, and Śūdras dark. Gandharvas and Apsarās are traditionally said to be and were (actually painted) in many colours. Kings and prosperous people are of the colour of the padma. The sick, the evil-doers, those who are oppressed by evil stars, or have taken shelter in penance, and all family men engaged in toilsome work should also be dark.

The colour of things seen, should be painted resembling (their natural colouring).

1) Here follows a lengthy account of the art of singing, its history and origin. III. Ch. 27,

Ch. 40

Mārkaṇḍeya spoke:

1–3.
Brick powder of three varieties (i.e. smooth, middling and coarse) should be mixed with clay, a third of it in proportion. To this is added fragrant gum resin,[1] bee's wax, honey, kundara grass (liquorice), molasses, safflower soaked in oil, all in equal proportions. To these two parts already composed is added powder of lime three fourths burnt, with bel fruit[2] (*Feronia elephantum*) pulp and lampblack. The rest

or the remaining fourth part is an addition of sand (a little more or less) according to the experience of the skilful artist.

4. Then it is soaked in water stored in a pot so as to get lubricous and is kept so for a month.

5. When after a month it becomes a very soft paste, it has to be carefully taken out and a coat applied by the skilled artist on the wall after testing that it is quite dry.

6. The coating should be smooth, even, firm, free from uneven patches, neither too thick nor too thin.

7–8. When the wall is dry after this coat and is still not quite smooth, it should be smoothened by an application of the clay bereft of *sarjarasa* [Śāla-tree (Shorea robusta)?] and oil by coats of lamp black and frequent wetting of the surface with milk and rubbing, all with a great effort.

9. The wall dries up very soon and does not perish even after a hundred years.

10. In this same manner a variety of mosaic floors can be made in picturesque fashion by the use of two or more different colours.

11–13. When the wall is dry, on a good day with an excellent constellation that is appropriate to the *gaṇa* (*deva* in preference to *manushya* or rākshana), specially suited for starting a picture (like *Punarvasu* for Rāma, Ārdra for *Naṭarāja*, *Rohiṇī* for *Kṛishṇa, Mṛigaśira* for Śiva, etc.), the painter, dressed in immaculate white, pure in mind and body, having adored Vedic seers and uttered auspicious hymns (*svastivākya*), and having bowed to the learned in the art and the Masters in their order, and with great affection for Masters, facing the east, contemplating on the deity to be depicted, should start his work of painting.

14–15. The wise artist should draw and fixed up the proportions and positions of the figures. Then he should colour the painting with colours appropriate in their different situations. Darker and lighter shades should be shown as they occur. This chiaroscuro has been earlier explained by me at length.

16. The primary colours are five, white, yellow, red, black, blue[3] with hundreds of intermediate tones.

17. First the colour scheme is arrived at by separating them and according to the artist's knowledge and capacity for creating the atmosphere in the picture, there should be produced hundreds and thousands of colour tones.

18. Blue and yellow mixed produce green. It may be pale with a greater modicum of white or deeper with blue.

19. According to the colours used quantitatively there is a predominance of a colour, lighter, darker or in equal proportion, making it threefold.

20–21. With one tone predominant many tints are produced. Thus there is the somewhat yellowish green of dūrvā grass,[4] light wood-apple green, green like green pulse[5] and so on, which can all be produced.

20–21. Blue mixed with white is a tertiary colour which again is manifold by the predominance, diminution, equal or lesser proportion of one or the other. Thus is formed the tint like that of the blue lily, of the dark *chāsha* bird.

23–24. Beautiful tints are produced by mixing in calculated proportions. Red *lākshā* tint mixed with white like the *lodhra* flower becomes red like the red lotus, a colour so charming. This again produces several other varying tints.

25–26. The materials for colours are gold, silver, copper, mica, lapis lazuli, red lead, yellow orpiment, lime, red lac, vermillion, indigo, and several more manifold.

27. There are all these in every country and have to be prepared with one colour or another predominant. Metal colours are to be laid in delicately thin sheets or by liquefying them by chemical methods.

28. Mica becomes the solvent liquefier when added to iron. Thus when metal colouring is to be done they have to be suitably prepared.

29. The liquefier of mica is mercury. Hide glue and *bakula* resin glue act to fix and strengthen colours for all of which vermillion juice is also used.

30. The picture painted with brushes of high quality hair and with colours strengthened by the glue of elephant hide, the juice of *dūrvā* and bark resin cannot be destroyed even if washed with water it has a prolonged life for several years.

Here ends the 40th chapter titled 'Colour Composition', a conversation between *Mārkaṇḍeya* and *Vajra* in the *Vishṇudhamotara*.

Notes

Sadakane 1988: 56–58
1. Extracted from *amyris agallochum*.
2. *Aegle marmelos*.
3. In the 8th verse of Chapter 27 of this Purana (the 3rd section on a dance sutra), green is mentioned instead of blue.
4. *Panicum dactylon*.
5. Kapittha: *feronia elophantum*.

Appendix II: Materials, preparation and methodologies

II.1 Visual observation and documentation at the Bamiyan site

During the 2005–2007 Bamiyan Buddhist Wall Painting Conservation Project, a comprehensive survey of the caves and wall paintings at each of the four sites – Bamiyan, Foladi, Kakrak and Qol-e Jalal – was undertaken. As part of this survey, in total 10,000 wall painting fragments were classified and recorded in an inventory. Across all the sites, two semi-open caves that once housed seated Buddha figures as a kind of Buddhist niche – Caves I and H(a) – contained wall paintings that were weathered, discoloured and faded due to exposure to sunlight, wind and dust. Caves with collapsed walls such as Foladi Caves 4 and 6 had wall paintings similarly deteriorated due to identical circumstances.

Caves created in relatively low-lying areas of the cliffs, such as Bamiyan Caves E, N and M, as well as caves around the Giant Buddhas, are easily accessible from the ground and had been used repeatedly over time as spaces for storage and habitation. As a result, wall paintings inside are scratched, stained with soot and in some instances partially cut and looted, all of which make observation difficult. Conversely, wall paintings in caves that are difficult to access, such as those where adjacent stairs and passages have been lost, are in relatively good condition, as seen with Cave K_3. Caves around the Eastern Giant Buddha (Caves A, B, C and D) were subject to conservation and restoration by French and Indo-Afghan teams in the 20th century. Therefore, it is highly likely that their walls were cleaned and strengthened with synthetic resin, which also means that they are no longer in their original state.

During a visual survey of the field, we observed and collected data on the following:

- how rendering layers adhered to rock;
- the number of rendering layers;
- types of materials (straw fibre, animal fibre, etc.) in renders;
- how renders were applied;
- the colour and thickness of underdrawings;
- how lines were drawn;
- variations in colour;
- the presence or absence of metal foils;
- adherence in pigment particles;
- cracks and missing parts in each work;
- the condition of painting layers.

When observing painting layers, we also noted any discoloration or fading in pigments. A portable stereomicroscope (Peak pocket microscope) with 50× and 100× magnification was used for *in-situ* observation. As Caves I, K_3 and N(a) were in need of conservation, we conducted detailed wall-by-wall surveys of their condition. Wall painting fragments recovered from the cave floor were transferred to the Bamiyan Cultural Center, then sorted by cave and classified according to the presence or absence of pigments. In addition to their colour, size and weight, we also recorded the structure of each fragment's cross-section, types of materials found in its renders and its state of deterioration. The condition of its surface and pigment particles were observed using a Yashima shop measuring microscope (YMM-SK) and a digital stereomicroscope with USB connection (YDU-2N). Fragments proved to be more useful than intact paintings on the wall for understanding the structure of wall paintings because underdrawings and the back of the wall paintings were more visible and could be compared across locations.

II.2 Sample collection

Small samples were taken for detailed analysis of technique, materials and surface deposits. Each sample was brought back to Japan with the permission of Afghanistan's Ministry of Information and Culture to be observed under a stereomicroscope in a laboratory at NRICPT. Next, samples were also partly encapsulated in polyester resin to create polished sections. The surfaces were polished and the layer structure observed for analysis of inorganic and organic substances.

From approximately 50 caves with wall paintings in Bamiyan, Foladi, Kakrak and Qol-e Jalal, only a total of 31 (23 caves in Bamiyan, 5 in Foladi,

Table A.II. 1 List of samples from different caves in Bamiyan.

Bamiyan			*Foladi*			*Kakrak*			*Qol-e Jalal*		
Cave	Sample number	*In-situ* sample	Cave	Sample number	*In-situ* sample	Cave	Sample number	*In-situ* sample	Cave	Sample number	*In-situ* sample
A lower salle	5		2	1		Cave 43	6		Qol-e Jalal	7	7
B(d)	4		3	7		Cave 44	5	5			
C(a)	6	1	4	31	17						
C(b)	1		5	7	7						
D	2		6	14	13						
D door front	1										
E(c)	3		Total	60		Total	11		Total	7	
E(d)	7										
F(c)	9										
G	4	4									
H(a)	3										
H(b)	1										
I niche	30	26									
I pradaksina	4										
J(b)	9										
J(c)	6										
J(d)	5										
J(f)	3	3									
K	25	23									
L	2	2									
M	9	7									
N(a)	36	17									
S(a)	6										
East III	3										
East Displaced	7										
EGB*	7										
WGB*	3										
Total	201										

* provided by ICOMOS Germany (E. Meltzl)

2 in Kakrak, and 1 in Qol-e Jalal) had clearly legible painting elements (including in recovered wall painting fragments). Of these, the anterior chamber of Cave D, which may have been painted at a different time, the niche surrounding the Eastern Giant Buddha and the *pradakshinapata* (ritual passage) of Cave I were all sampled separately. The total number of samples taken was 279: 201 from the Bamiyan Caves and both Giant Buddhas, 60 from the Foladi Caves, 11 from the Kakrak Caves and 7 from the Qol-e Jalal Caves (Table A.II.1). Samples were also taken to assess the condition of wall paintings for conservation in Caves I, K_3 and N(a). In addition to the samples, black deposits on walls and fibre fragments from renders were also collected but are not included in the samples listed here.

As most samples were collected from small fragments recovered on cave floors, it was difficult to ascertain their original position therefore when taking the inventory, these fragments were classified and recorded according to colour. In addition, coloured fragments from the surfaces of each Giant Buddha were provided by the ICOMOS German Committee. These samples were recovered from debris of exploded rocks during conservation work on the two Giant Buddhas. Both Buddhas are believed to have been made by applying kneaded clay to the surface of conglomerate rock, shaping the clay to form distinct patterns and folds of clothing, and then painting the completed statues (Chapter 3, section 3.5). However, due to extensive damage in 2001, it is no longer possible to ascertain whether any given fragment found around the Giant Buddhas originally part of their painted surfaces, part of a wall painting in their niches, or part of a wall painting from a cave at their feet. Therefore, we decided to analyse the binding media in these specimens that may have originally come from the Giant Buddhas for reference purposes only. The pigments and binding media of painted samples taken from the two Giant Buddhas were also analysed by ICOMOS Germany, the Technical University of Munich (Germany) and the University of Pisa (Italy) (Blänsdorf *et al.* 2009b; Blänsdorf 2016, 2021).

II.3 Observation of samples and cross-sections with UV and visible light microscopy

II.3.1 Stereomicroscopic observations in normal light and UV fluorescence

Each microsample was observed and photographed under a stereomicroscope (Olympus BX51) using normal light and UV fluorescence. Their characteristics were then noted in the observation table. Observations were conducted at magnifications ranging from 40 to 100× with particular attention paid to the appearance of cracks in coloured surfaces, the colour and texture of materials in the ground and painting layers, and the presence or absence of inclusions or additives in the rendering layer.

II.3.2 Preparing cross-sections and observations with UV and visible light microscopy

Each microsample was observed under a stereomicroscope, noted in the observation table and then mounted in polyester resin (Struers cold mounting resin No. 105). Because samples may contain water-soluble binding media and moisture-sensitive pigments, the surfaces of the 4,000–12,000 mesh samples were dry polished using Micro-Mesh (Micro-Surface). Cross-sections were observed, described and photographed in the polarised light mode of a stereomicroscope (Olympus BX51) using normal and UV fluorescence as light sources. The method of observation was adapted from Plesters (1956).

II.4 Synchrotron radiation-based microanalytical techniques

II.4.1 Principles of synchrotron radiation

Synchrotron radiation (SR) is produced when charged particles (e.g. electrons) travelling at the speed of light are redirected by a magnetic field. Synchrotron radiation is characterised by the concentration of light in the tangential direction of particle beams, resulting in a highly directional and intense light. Compared with a standard X-ray source, SR X-ray beams are more intense and collimated. At the European Synchrotron Radiation Facility (ESRF) in Grenoble, France, the SR light emitted by the 6GeV electron beam is mainly in the hard X-ray range but IR emissions were also used in the present study. The emission spectrum is quasi-continuous over large energy ranges. The high signal-to-noise ratio of the SR techniques, even when the beam is focused on a small area, makes them very useful for high-precision and high-sensitivity analysis of small and heterogeneous samples such as fragments from wall paintings (Cotte *et al.* 2006b). For example, SR micro-X-ray fluorescence (SR-μXRF), SR micro-X-ray diffraction (SR-μXRD) and SR micro-Fourier transform infrared (SR-μFTIR) enable the detection of elements, crystalline phases and molecular groups, respectively, at a high speed and high resolution with greater sensitivity than a conventional X-ray or IR source could provide.

In this study, SR- μFTIR, μXRF (ID21) and μXRF and μXRD (ID18F and more recently at ID13) were combined to identify organic and inorganic materials in a total of 51 samples, as part of the research project entitled 'Combination of Micro-X-ray Diffraction and Micro-Infrared Spectroscopy for the Study of Multi-Layered Buddhist Wall Paintings from Bamiyan (EC101 Y. Taniguchi, M. Cotte, E. Checroun)'.

II.4.2 SR-μFTIR at ID21, ESRF

The SR-μFTIR analyses were carried out at the former IR end-station at the ID21 beamline. We used the Nexus IR spectrometer coupled with the Continuμm microscope from Thermo Nicolet. Spectra were measured in transmission mode. The beam size was reduced to 12×12μm^2 or 8×8μm^2. 2D maps were acquired through one FTIR spectrum (a sum of 25 scans in the range of 4000–800 cm^{-1} and with a resolution of 8 cm^{-1}) at every pixel, and by raster scanning the sample with a step size of 8–12μm. Data acquisition and analysis were carried out using Omnic software from Nicolet and with the ROI Imaging tool from PyMca software (Cotte *et al.* 2016).

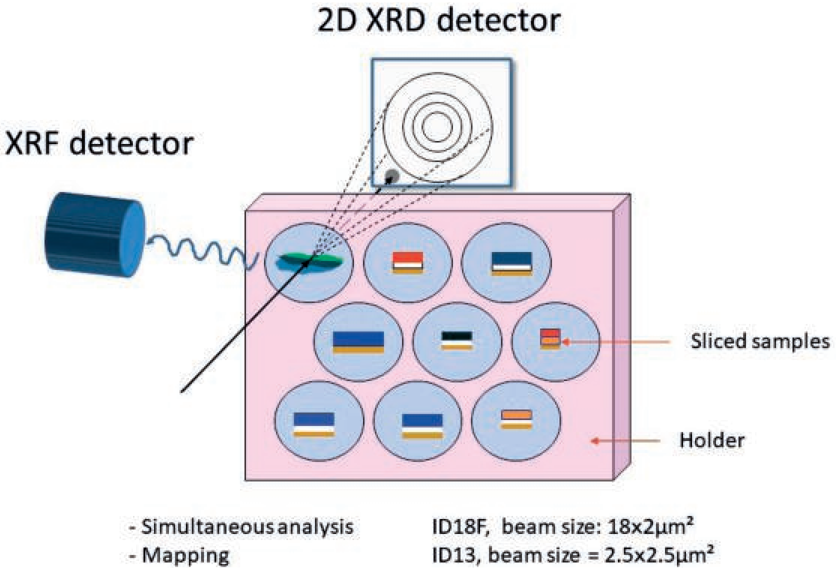


Figure A.II.1 Setting up a simultaneous μXRD/μXRF measurement using synchrotron radiation.

II.4.3 SR-μXRF at ID21, ESRF

Elemental maps were obtained by 2D μXRF mapping using the ID21 scanning X-ray microscope. The beam energy was selected thanks to a fixed-exit, double crystal Si (111) monochromator, located upstream of the microscope. The beam size was reduced to 0.8×0.2 μm^2 (hor. × ver.) thanks to Fresnel zone plates in 2006 and to Kirkpatrick Baez mirrors in 2022. The micro fluorescence signal was collected in the horizontal plane perpendicular to the incident beam direction by using an HPGe solid-state in 2006 and a Silicon Drift Detector in 2022, both being energy-dispersive detectors. Measurements were performed under a vacuum. μXRF maps were batch fitted using the software package PyMca (Sole *et al.* 2007).

II.4.4 SR-μXRD/μXRF at ID18F, ESRF

In 2006, the first SR-μXRD/μXRF analyses were performed at the former D18F beamline, at the ESRF (Fig. A.II.1). The excitation energy was fixed at 28 keV and the beam was focused to 15×1 mm^2. Both XRD and XRF were collected simultaneously at every pixel of the 2D maps. Because of the high duration of each acquisition at the time (3–5 seconds for data acquisition and 3–5 seconds for data saving per pixel), and the large horizontal beam size, maps were reduced to 3 columns (covering 45 μm) of 100 μm height scanned with 1 μm step ending into ~300 points per sample. XRD measurements were carried out in transmission mode.

II.4.5 SR-μXRD/μXRF at ID13, ESRF

The above SR analyses were carried out primarily in December 2006. Since then, ESRF beamlines have dramatically evolved with improved state-of-the-art optics, mechanics, detectors, electronics and software. More importantly, the ESRF storage ring was fully dismounted in 2019 and a new Extremely Brilliant Source (EBS) machine capable of higher performance was installed. In order to take advantage of these new capabilities, the same samples were re-analysed in 2021 by μXRD and μXRF at the micro-branch of the ID13 beamline at the ESRF. The beam size was much smaller (2.5×2.5 μm^2) and data acquisition much faster (only 10 ms per pixel). This allowed us to acquire much larger maps (up to 700×500 μm^2) with better resolution (step size was set to 1 μm), all in a reasonable amount of time (less than one hour per map).

II.4.6 Sample preparation for SR microanalyses

For SR-μFTIR analysis, one part of the microsample obtained from the wall painting was manually sliced under a binocular stereomicroscope, placed in a direction to preserve the layered structure and compressed between diamond cells (Fig. A.II.2). Measurements were also carried out on thin sections from resin embedded samples, but the signal of resin was usually strongly overlapping with the signal from the paint materials. For SR-μXRD analysis, thin sections (5–50μm) were cut from samples encased in resin using a microtome. Sections were

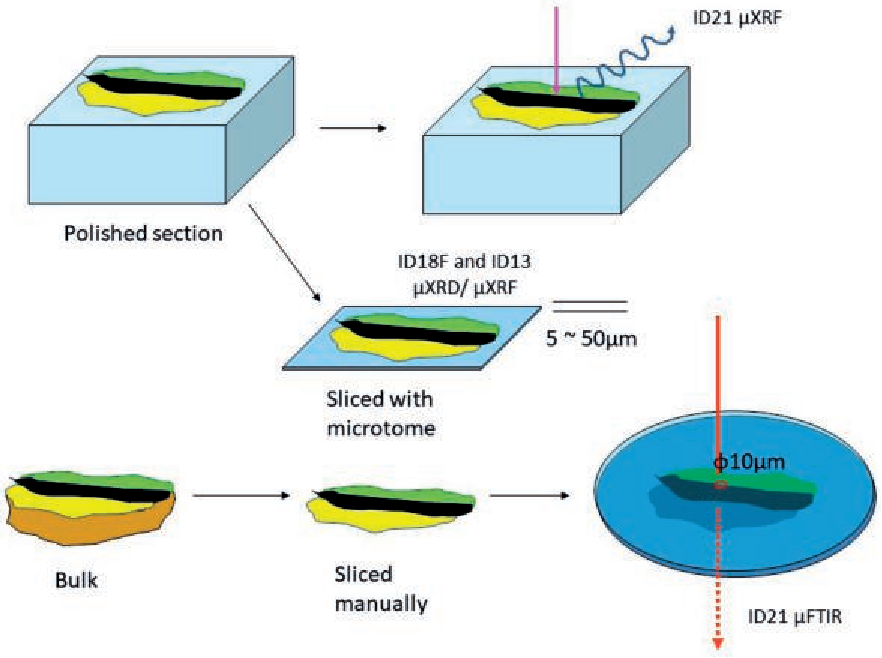


Figure A.II.2 Prepared samples for analysis using synchrotron radiation.

then sandwiched between two thin polyethylene sheets. SR-μXRF analysis was carried out on both thin sections and polished sections. Each specimen was collected and stored for future analysis and investigation by other methods.

II.5 Gas chromatography-mass spectrometry

SR-μFTIR, SR-μXRD and SR-μXRF analyses at the ESRF revealed that the Central Asian wall paintings have a complex multilayered structure, with each layer containing a different variety of organic matter such as oils, resins, proteins and polysaccharides. However, as these methods do not allow for the precise identification of organic species, both GC-MS (three methods for fatty acid, amino acid and plant gum identification) and ELISA (enzyme-linked immunosorbent assay) methods were used (Mazurek 2006). The analysis was carried out in collaboration with the Getty Conservation Institute in the USA.

II.5.1 Principles of GC-MS

GC-MS is a column chromatographic method that uses a gas sample as mobile phase for qualitative and quantitative study of multi-component mixtures. The amount of sample introduced may be as little as a few μg and is fed into a column that is gaseous and capable of separating the individual components. As the sample passes through the column, the components are separated by the difference in affinity between them and the stationary phase in the column. The retention time is specific to the compound under certain conditions and thus allows identification of components in the gas. The area of the MS peak corresponds to the amount of the component, thus enabling quantitative analysis.

II.5.2 Analysis of oils, beeswax and resins

II.5.2.1 Sample preparation

Tens to hundreds of μg of each sample were weighed on a Cahn Ultra Microbalance, placed in a screwtop conical vial and mixed with toluene and Alltech Associates Meth Prep II (TMTFTH: m-(trifluoromethyl)phenyltrimethylammonium-hydroxide, methanol 0.2 M; 2ml:1ml) for saponification and methylation (Sutherland 2007). The vials were warmed on a hot plate at 60 °C for 1 hour and then cooled to room temperature. They were then mixed by a homogeniser and 1 μl of the supernatant liquid was injected into the gas chromatograph.

II.5.2.2 Analytical conditions

The analysis was carried out on a Hewlett Packard 5972 GC/MS. A DB-5MS capillary column (30 m × 0.25 mm × 1 μm) was used as separation column and helium as a carrier gas (constant pressure 12. 67 P/Si). The sample was injected using the splitless method, with a purge time of 60 s and its inlet temperature set at 300 °C. The transfer line temperature was set at 280 °C. The oven temperature was held at 50 °C for 2 min, then increased to 320 °C at 10 °C/min and held there for 20 min. The mass spectrometer was set to total ion monitoring (TIM) mode. The ionisation method is electron ion impact (EI) with an ionisation voltage of 70 eV. The interface temperature was set at 230 °C. A mixture of pimelic, suberic, lauric, azelaic, sebacic, myristic, palmitic, stearic, arachidic and oleic acids was used for calibration to create a calibration curve for quantification. ChemStation was used for the analysis.

II.5.3 Protein analysis

II.5.3.1 Sample preparation

After the above analysis, the remaining sample was dried in the vial and the amino acids were silylated and subjected to GC-MS. 100 µl of hydrochloric acid (6.0 N) was added to the vial, purged with nitrogen gas for 20 seconds, and sealed. The vial was evaporated to dryness by passing nitrogen gas through it while it was warmed to 60 °C. Then 30 µl of ultrapure water was added to the sample before the vial was centrifuged and dried in the same way as before. The contents were completely dried before rinsing with ethanol (99.5 v/v%) and silylation.

The solvent of the mixture was added to the vial and the lid of the vial was replaced. The solvent for silylation was a mixture of silylated pyridine and the silylating agent was Pierce Chemical's MTBSTFA + 1% TBDMCS (N-methyl-N-(tert-butyl-dimethylsilyl)-trifluoroacetamide + tert- butyl-dimethylchlorosilane) (Simek *et al.* 1994). The mixture for silylation was 3 ml of a mixture of 30% MTBSTFA/TBDMCS and 70% pyridine. On a hot plate, the vials were warmed at 60 °C for 30 min and then heated in an oven at 105 °C for 5h. After cooling in the air, the vials were centrifuged and 1 µl of the supernatant liquid was injected into the gas chromatograph.

II.5.3.2 Analytical conditions

A quantitative analysis of TBDMS derivatives was performed on a Hewlett Packard 5972 GC-MS using a DB-5MS capillary column (30 m × 0.25 mm × 1 µm). The carrier gas was helium at a flow rate of 45 cm/sec (constant flow rate). The sample was injected using the splitless method at an inlet temperature of 260 °C with a purge time of 60 sec. The transfer line temperature was set at 280 °C. The oven temperature was held at 105 °C for 1 min, then increased to 320 °C at 20 °C/min and held there for 3 min. In order to avoid saturation of the detector with solvent peaks, the mass spectrometer was set to detect from 4 min onwards in the retention time. The mass spectrometer was set to selective ion monitoring (SIM) mode. The ionisation method is electron ion impact (EI) with an ionisation voltage of 70 eV. The interface temperature was set at 230 °C. Mixtures of amino acids, fatty acids and glycerol were used as calibration curves for quantification. ChemStation was used for the analysis.

II.5.4 Analysis of polysaccharides

II.5.4.1 Sample preparation

The method of analysis followed Mawhinney *et al.* (1980). Tens to hundreds of µg of sample were weighed on a Cahn Ultra Microbalance, placed in a screwtop conical vial and the allose solution was added to a final concentration of 20 ppm. To hydrolyse the polysaccharide, 100 µl of trifluoroacetic acid (1.2 N) was added. The vial was purged with nitrogen gas for 30 sec and sealed. It was then warmed at 125 °C for 1h and subsequently cooled to room temperature. Next, the vial was centrifuged and the supernatant liquid was transferred to a 20 ml vial for analysis, which was warmed up to 50 °C and evaporated to dryness using nitrogen gas. The vial was then rinsed with distilled water for GC-MS analysis and dried before being rinsed once more with anhydrous ethanol to dry the contents.

200 µl of O-methylhydroxylamine hydrochloride solution (2 ml/1 ml/300 mg) dissolved in pyridine and methanol was added to the vial for analysis and its lid replaced. The vial was then warmed at 70 °C for 20 min and allowed to cool naturally to room temperature. The mixture was slowly evaporated for 10 min until it became syrupy. 400 µl of pyridine plus acetic anhydride (1 ml/3 ml) was added, the lid was replaced and the vial was kept warm at 70 °C for 20 min. Vials were allowed to cool naturally to room temperature and dried using nitrogen gas until their contents were reduced to a syrupy or dry solid. The contents were liquefied by adding 400 µl of chloroform.

In order to remove salts produced during the derivatisation process, the sample was rinsed with 500 µl of hydrochloric acid (1.0N) and again with 500 μl of ion-exchanged water. The supernatant layer was carefully removed using a pipette and the bottom layer dissolved in chloroform was used. 200 μl of the sample solution dissolved in chloroform was taken and evaporated into dry solids. Then 50 μl of chloroform was added and 1 μl of this solution was injected into the gas chromatograph.

II.5.4.2 Analytical conditions

Analysis was performed on a Hewlett Packard 5972 GC-MS using a J&W Scientific DB-WAX capillary column (15 m × 0.25 mm × 0.25 μm). The carrier gas was helium at a linear velocity of 60 cm/sec. The sample was injected using the splitless method, with an inlet temperature of 240 °C and a purge time of 60 sec. The transfer line temperature was set at 240 °C. The oven temperature was kept at 105 °C for 1 min, then increased to 180 °C at 30 °C/min and to 240 °C at 5 °C/min, and kept there for 2 min. ChemStation was used for the analysis (Schilling 2005).

II.6 Analysis of binding media materials using the ELISA method

II.6.1 Principles of the ELISA method

The ELISA method is an analytical method used in the biological field. This method can be used to detect the type and concentration of antibodies and antigens in a sample. ELISA uses a highly specific antigen-antibody reaction, based on an enzymatic colour reaction, and is therefore an effective method for the detection and quantification of specific proteins in small quantities, even in the parts per trillion (ppt) order, where a variety of proteins is present in the same sample. In recent years, attempts have been made to simultaneously measure proteins of mammalian origin (such as collagen from animal glues, casein from dairy products, egg white albumin from egg whites and phosvitin from egg yolks) and plant origin (polysaccharides from plant gums), since organic matter containing many different proteins may be present in a coloured sample (Heginbotham *et al.* 2006; Mazurek *et al.* 2008). Here, a two-step antibody reaction technique called the indirect method was used. However, it is often very complicated to analyse painted samples due to the presence of organic substances such as drying oils, beeswax, resins and pigments that have deteriorated over the years.

Table A.II.2 Antibodies used in ELISA at GCI (as at 2007).

Primary Antibodies	*Secondary Antibodies*
Collagen I #ab6577	Rabbit IgG #AP123A
Collagen I #ab19811	Goat IgG #ab6742
Fish collagen I #T89171R	Rabbit IgG #AP132A
Casein #RCAS-10A	Rabbit IgG #AP132A
Ovalbumin #ab1225	Rabbit IgG #AP132A
Phosvitin #sc-46681	Mouse IgG #AP124A
Plant gum #JIM13	Rat IgM KPL#05-16-03
Gum tragacanth #MAC265	Rat IgG #AB6846

Table A.II.3 Antibodies used in NMWA at GCI (as at 2007).

Primary Antibodies	*Secondary Antibodies*
Collagen I #ab34710	Rabbit IgG #AP132A
Collagen I #ab19811	Goat IgG #ab6742
Fish collagen I #T89171R	Rabbit IgG #AP132A
Casein #bs-0813R	Rabbit IgG #AP132A
Ovalbumin #ab1225	Rabbit IgG #AP132A
Phosvitin #sc-46681	Mouse IgG #AP124A
Plant gum #JIM13	Rat IgM #A110-100AP
Gum tragacanth #MAC265	Rat IgG #A8438

Table A.II.4 The mean of the 24 absorbance readings at OD_{405} for each antibody at GCI.

Antibody	*Mean OD_{405}*	*Std Dev*	*+ 3 sigma window*
Collagen I #ab6577	0.10	0.02	0.15
Fish Collagen I #T89171R	0.12	0.02	0.16
Casein #RCAS-10A	0.18	0.01	0.22
Ovalbumin #ab1225	0.13	0.03	0.21
Phosvitin #sc-46681	0.16	0.03	0.26
Plant gum #JIM13	0.10	0.01	0.14
Gum Tragacanth #MAC265	0.12	0.02	0.18

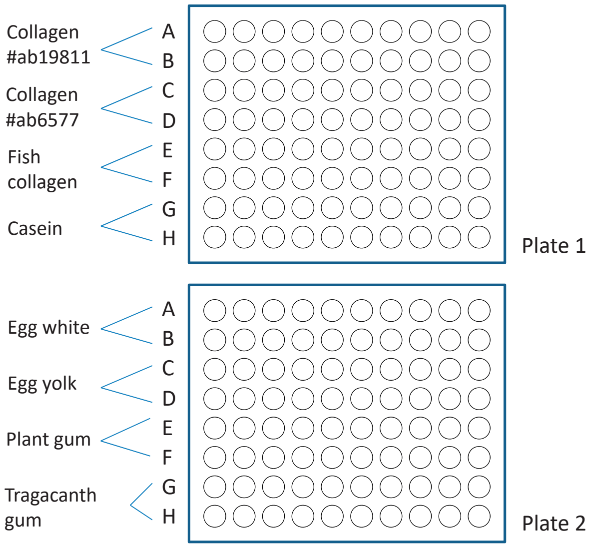


Figure A.II. 3 Microplates used in the ELISA method at the GCI. One test per vertical column.

II.6.2 Sample preparation and analytical methods

Sample preparation and analysis were conducted in accordance with previous studies (Mazurek *et al.* 2008). Samples were analysed with ELISA both at the Getty Conservation Institute (GCI) in the USA and the National Museum of Western Art (NMWA) in Japan. The primary and secondary antibodies used are listed in Table A.II.2 (GCI) and Table A.II.3 (NMWA). Skimmed milk is usually used to block the solid phase but, because casein may be identified in the analysed samples, Sea Block buffer was used as an alternative.

At the GCI, ELISA analyses was carried out as follows: the absorbance of the samples was measured at 405 nm using p-nitrophenyl phosphate (pNPP) as a chromogenic reagent in a microplate spectrophotometer (Fig. A.II.3). The absorbance of each sample was measured twice. 24 wells were analysed by ELISA using each of the specific antibodies listed in column 1 (Table A.II.4). The wells did not contain added antigen (blanks), and the mean of the 24 absorbance readings at OD_{405} are reported for each antibody in column 2 (Table A.II.4). The standard deviation was calculated based on the readings and multiplied by 3 sigma. This gives the limit of detection using a 99% confidence level. Data are lacking for Collagen I #ab19811. Based on these results, a minimum OD_{405} value of 0.3 was routinely used as the threshold for all ELISA positive tests.

At the NMWA, the following methods were used based on the GCI's procedure (GCI 2015) with modifications: dilutions of samples are used as 40, 20, 20, 10 µL for each well, and antibodies were modified as shown in Table A.II.3. As blocking buffers, Sea Blocking buffer and Blocker BOA in PBS (for anti-fish collagen antibody, anti-ovalbumin antibody and anti-plant gum antibody) were selected. Measurements were taken at OD_{405} and OD_{630} at the same time in order to avoid too high absorbance values. Average values of blank +3SD were used as the threshold, and more than 3 positives out of 4 wells of different dilutions were considered as positive.

Appendix III: Results of SR-µXRF/µXRD/µFTIR analysis

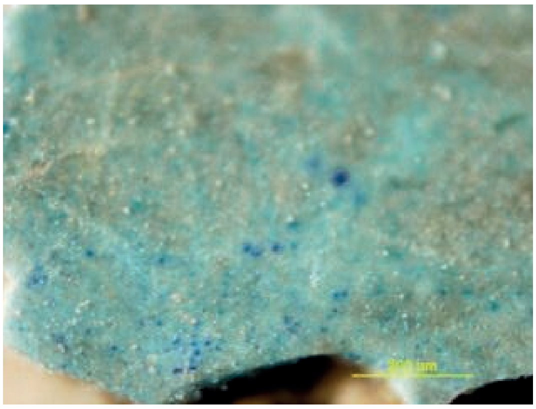


Figure A.III.1 BMM001: stereomicrograph image.

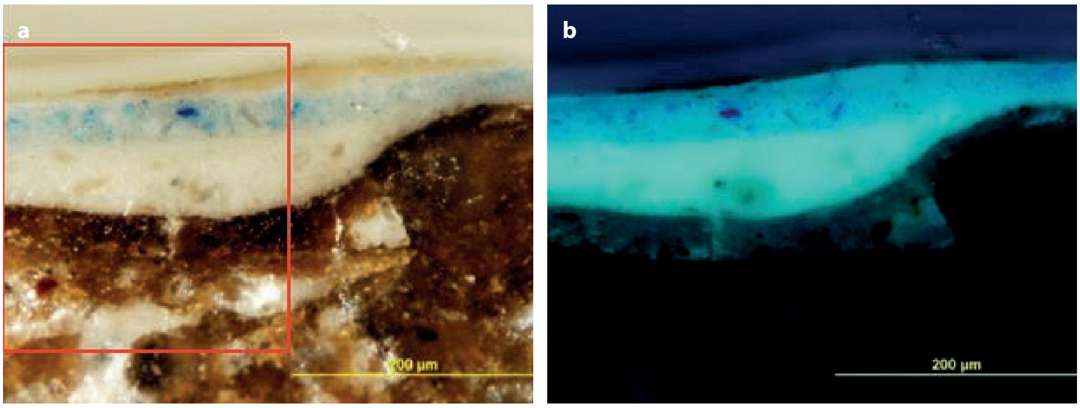


Figure A.III.2 BMM001: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light. The red frame is the imaging area.

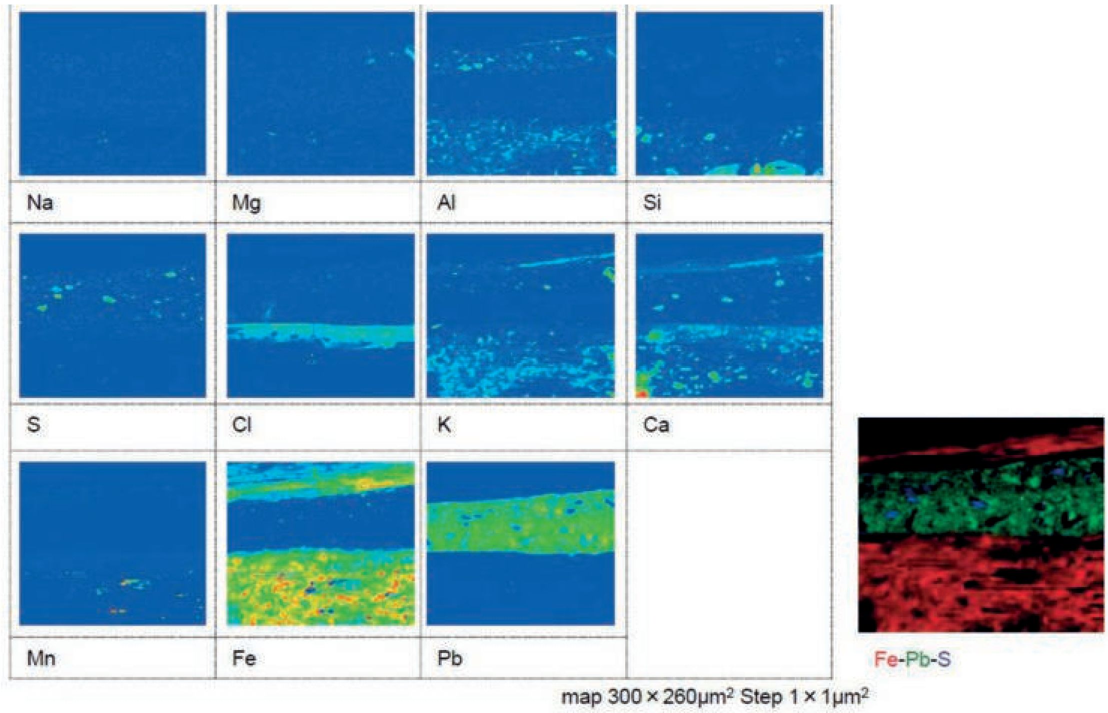


Figure A.III.3 BMM001: µXRF imaging (PyMCA: ID21.

Figure A.III.4 BMM009: stereomicrograph image.

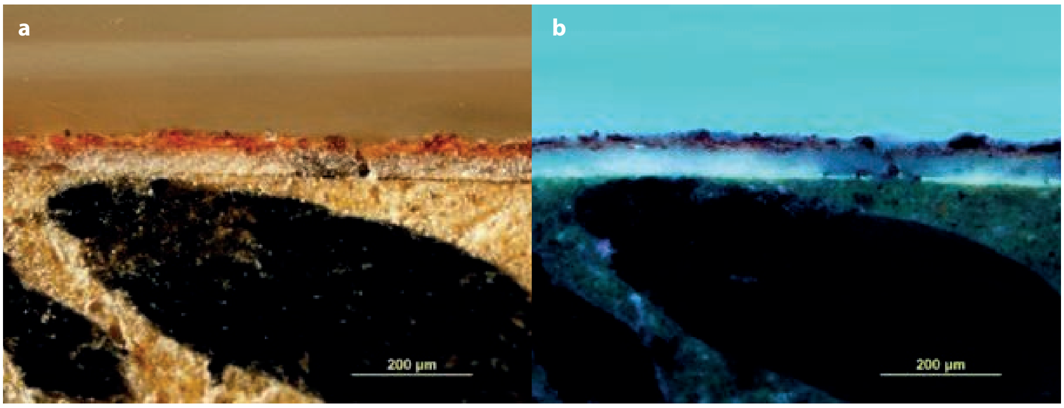


Figure A.III.5 BMM009: photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

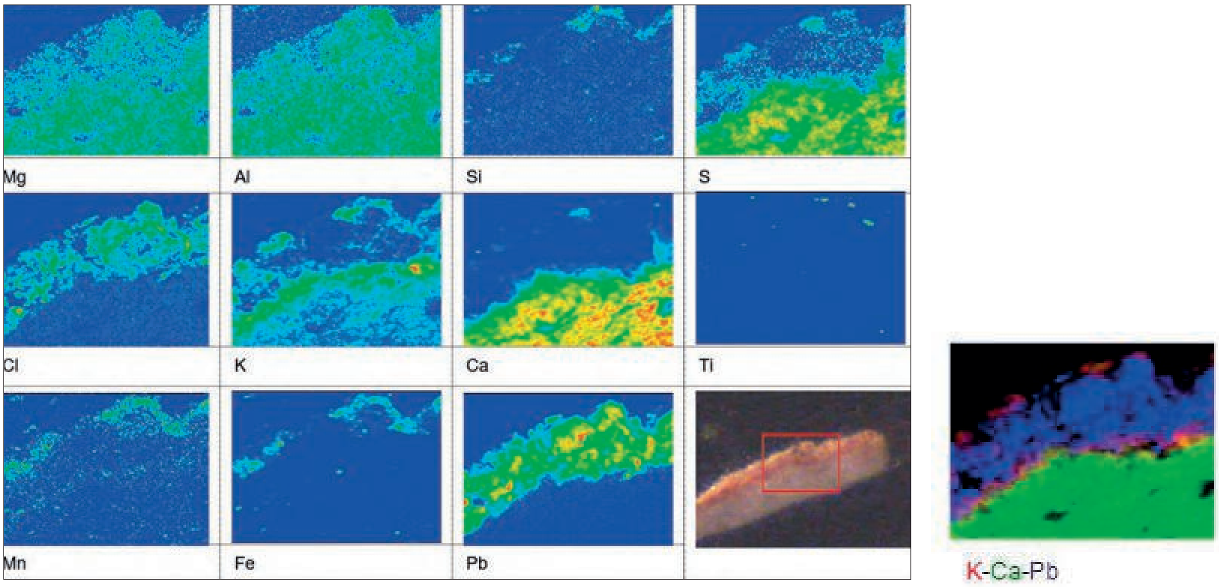


Figure A.III.6 BMM009: µXRF imaging (PyMCA: ID21).

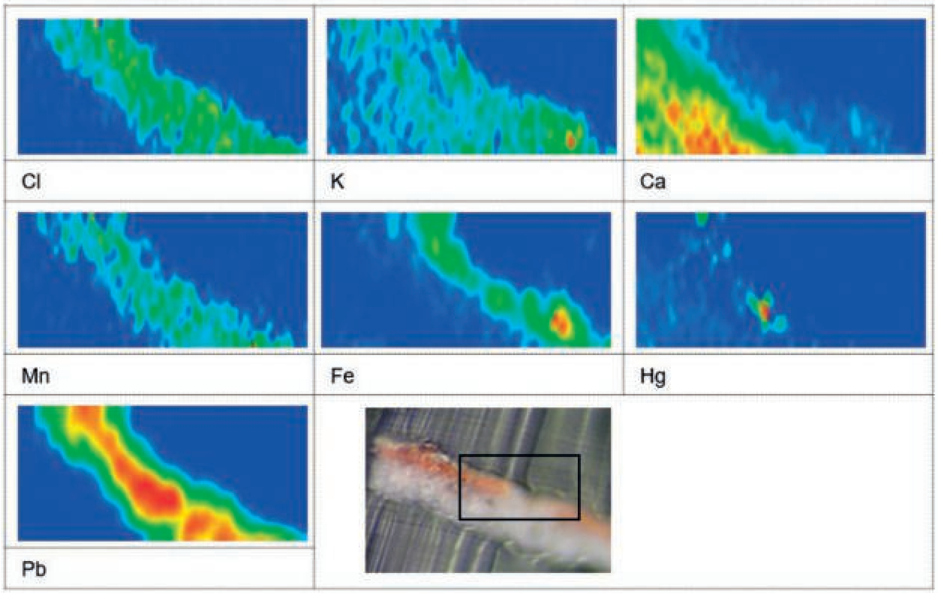


Figure A.III.7 BMM009: µXRF image (PyMCA: ID18).

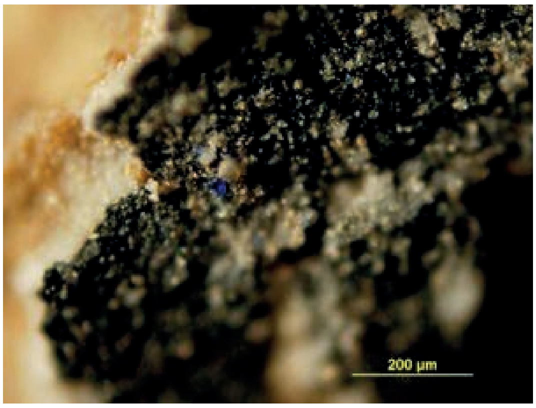


Figure A.III.8 BMM033: stereomicrograph image.

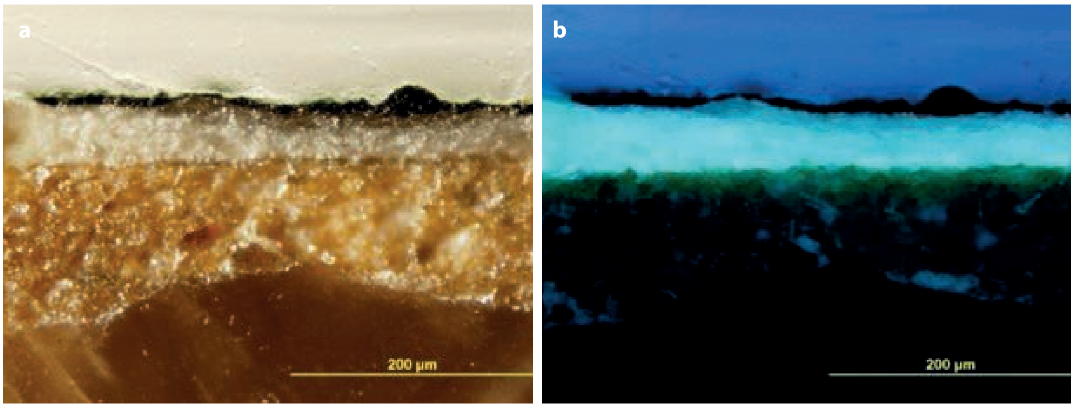


Figure A.III.9 BMM033: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

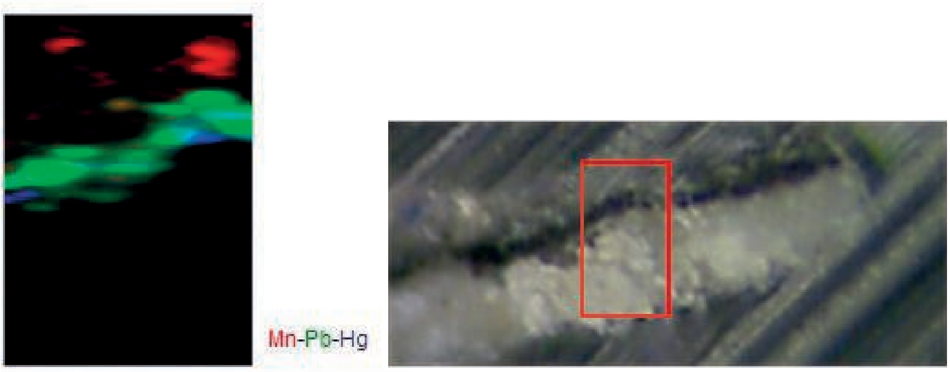


Figure A.III.10 BMM033: µXRF imaging (PyMCA: ID21).

Figure A.III.11 BMM033: µXRF image (PyMCA: ID21).

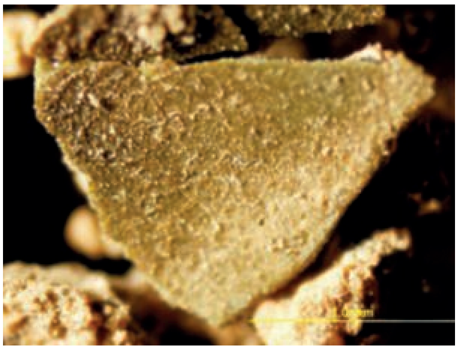


Figure A.III.12 BMM035: stereomicrograph image.

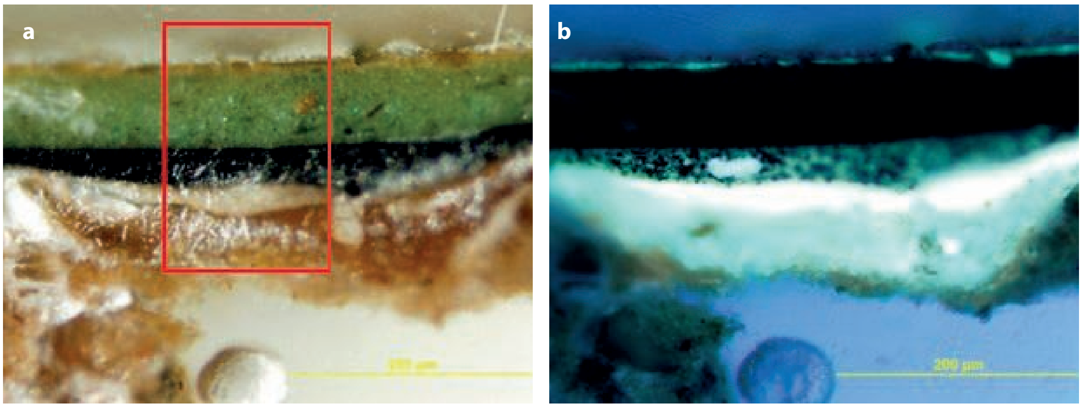


Figure A.III.13 BMM035: PLM photomicrograph of cross-section in (a) normal diffused light and (b) UV light. The red frame is the imaging area.

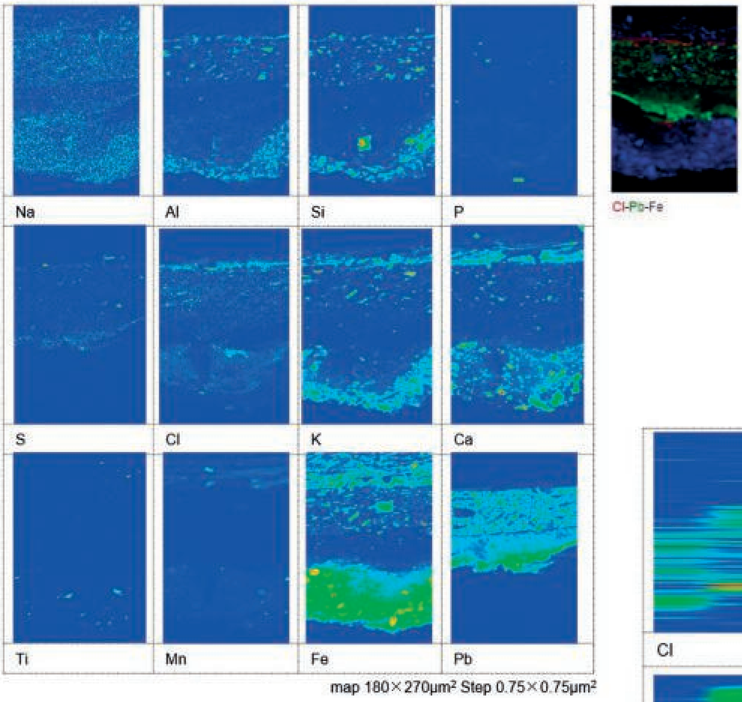


Figure A.III.14 BMM035: µXRF imaging (PyMCA: ID21).

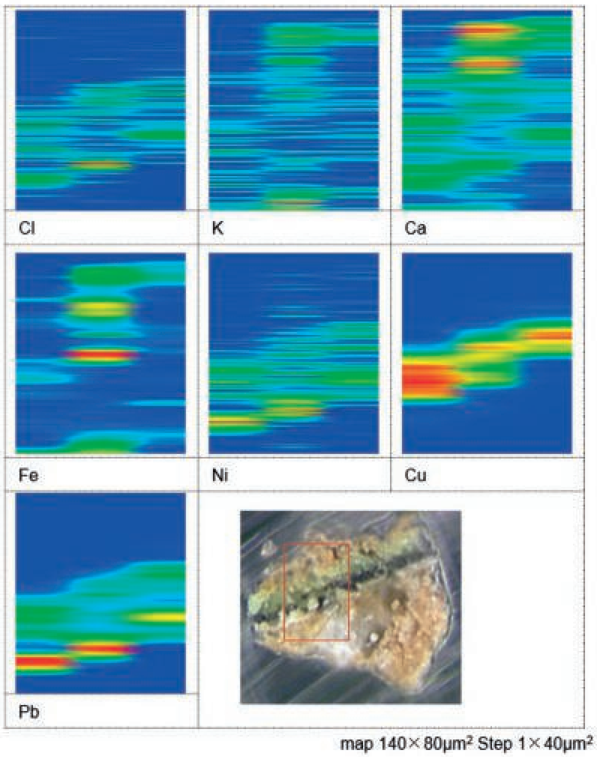


Figure A.III.15 BMM035: µXRF image (PyMCA: ID18).

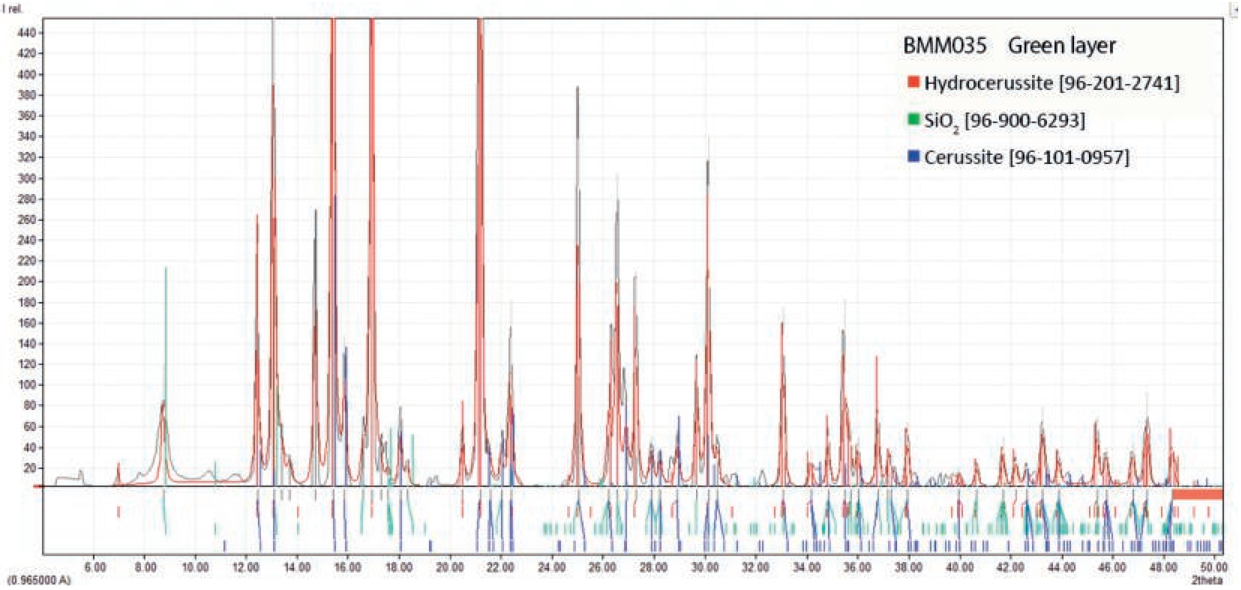


Figure A.III.16 BMM035: X-ray diffraction pattern of green (painting layer).

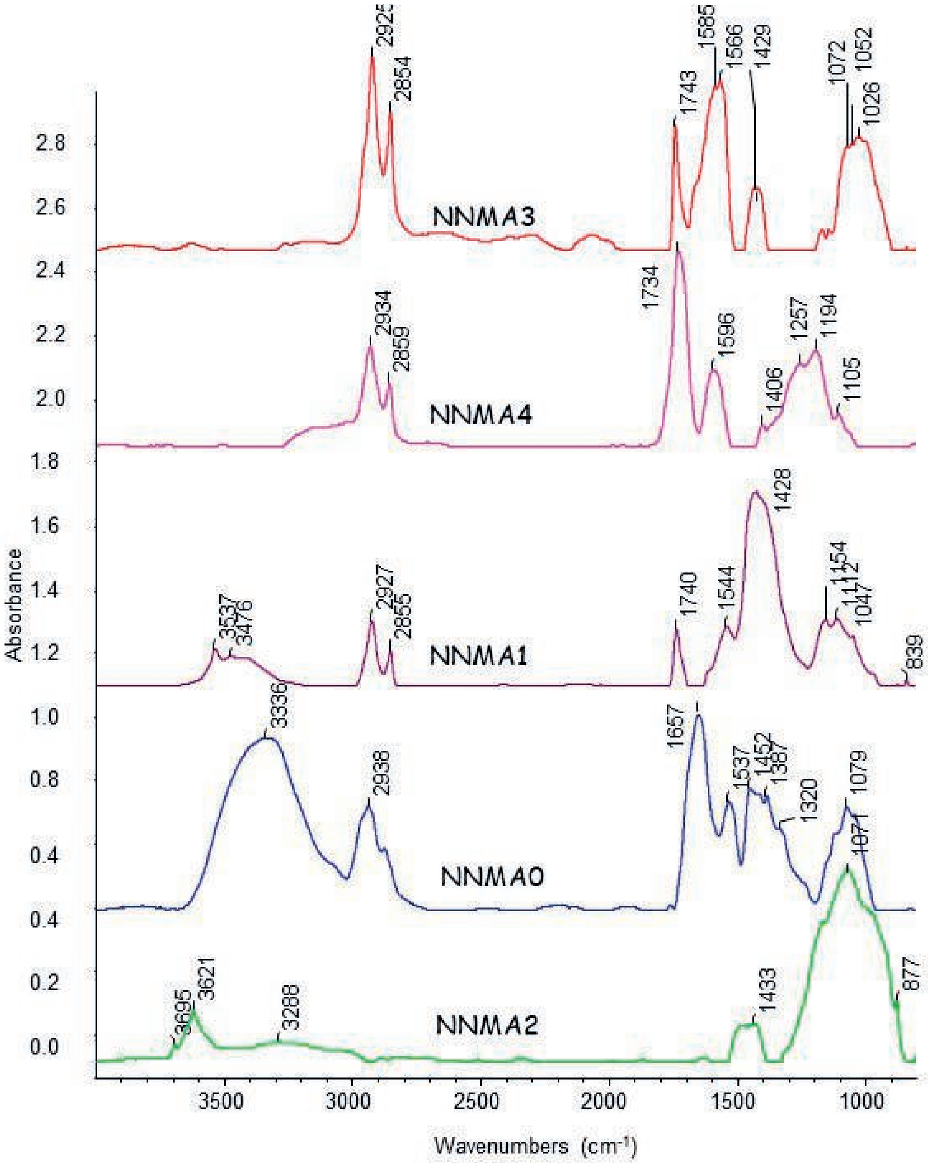


Figure A.III.17 NNMA FTIR components obtained from the analysis of a map acquired on the BMM035 fragment. Maps are shown in Figure A.III.18.

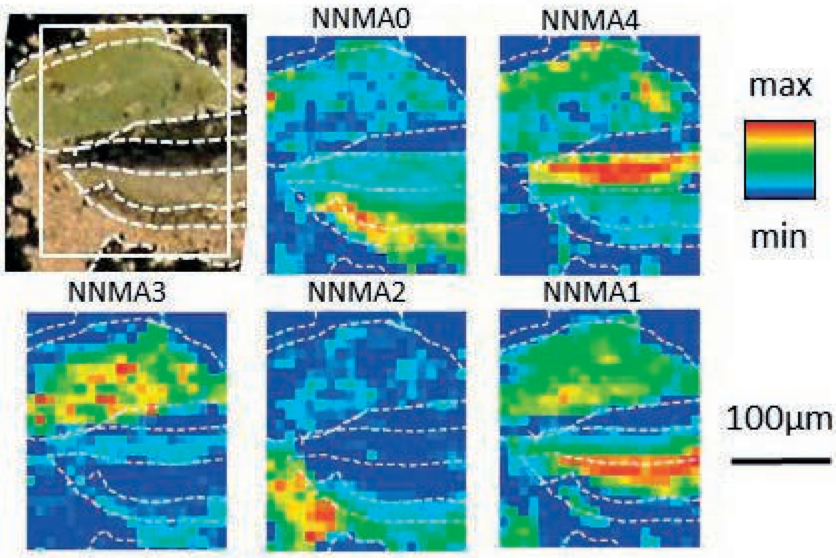


Figure A.III.18 NNMA FTIR maps obtained from the analysis of a map acquired on the BMM035 fragment. NNMA vectors are shown in Figure A.III.17.

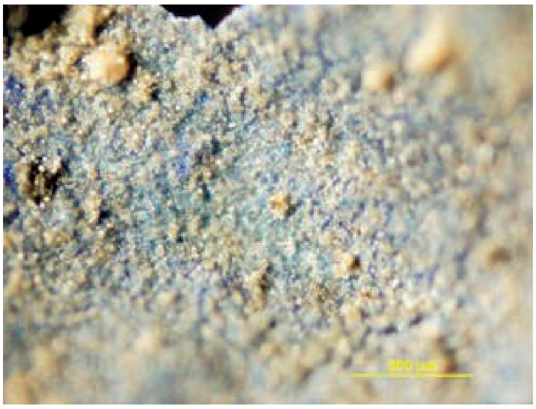


Figure A.III.19 BMM039: stereomicrograph image.

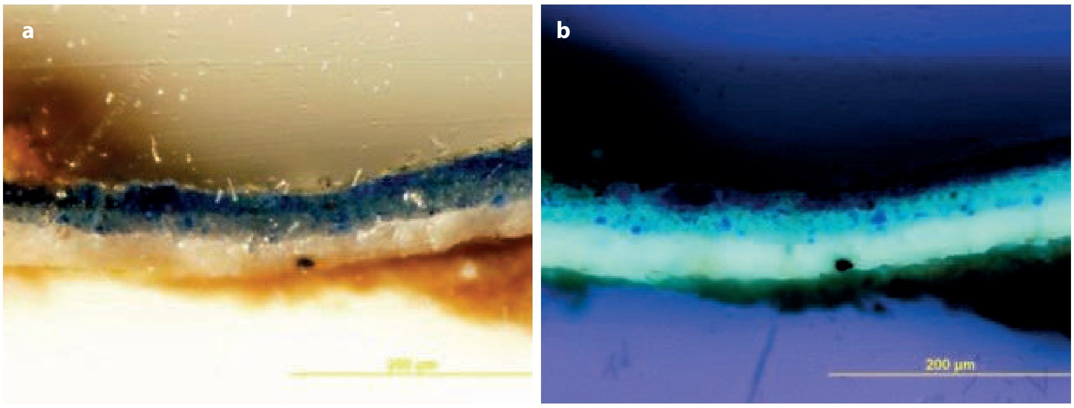


Figure A.III.20 BMM039: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

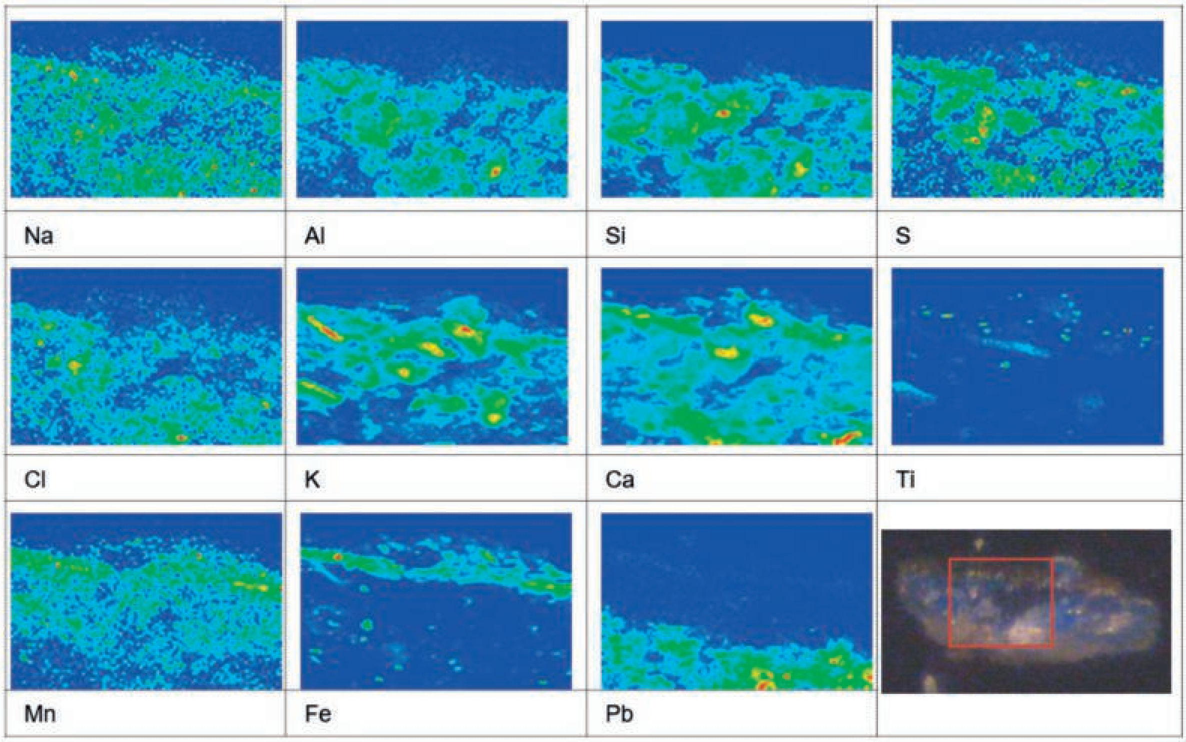


Figure A.III.21 BMM039: µXRF image (PyMCA: ID21).

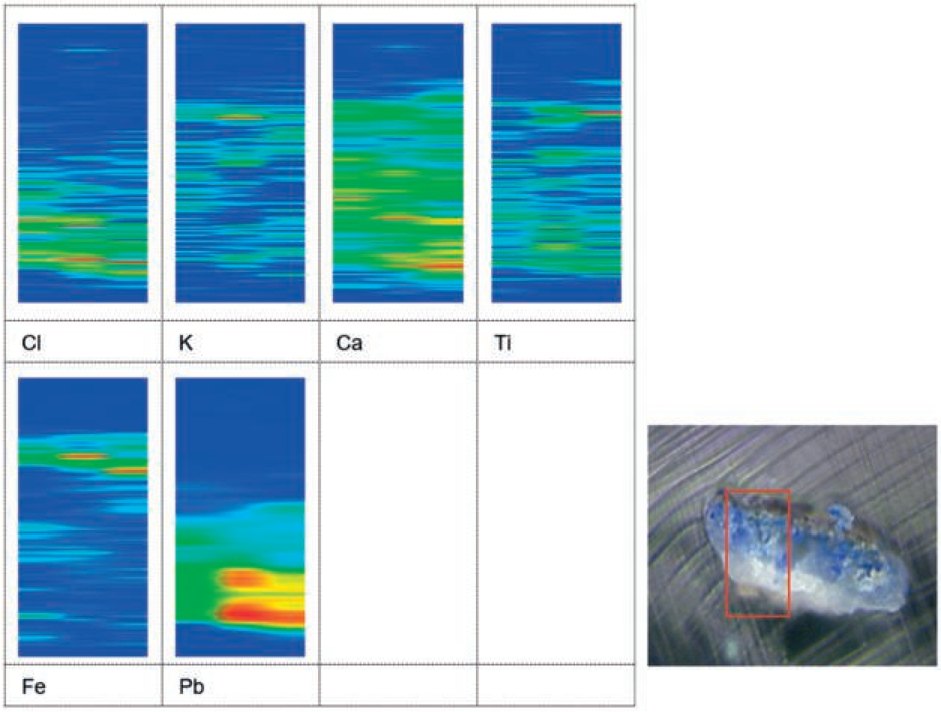


Figure A.III.22 BMM039: µXRF image (PyMCA: ID18).

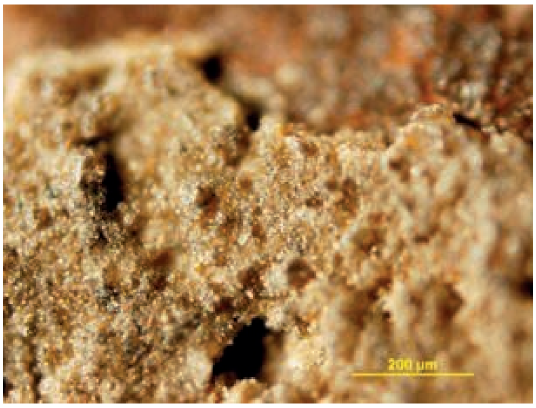


Figure A.III.23 BMM040: stereomicrograph image.

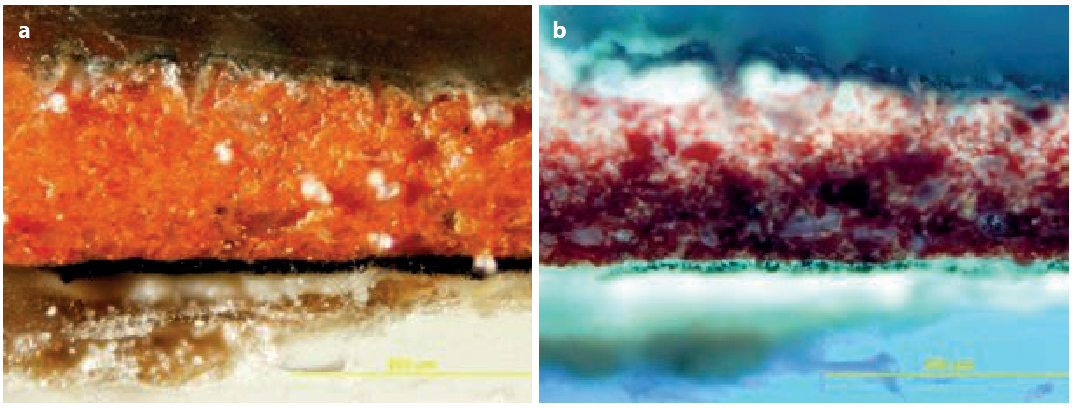


Figure A.III.24 BMM040: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

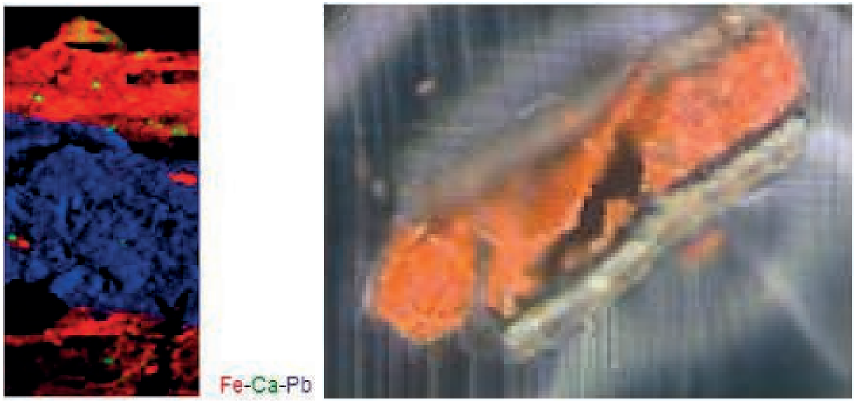


Figure A.III.25 BMM040: µXRF imaging (PyMCA: ID21).

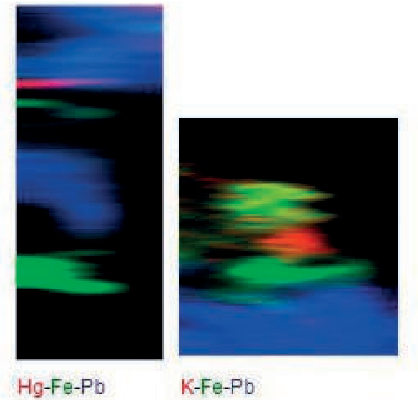


Figure A.III.26 BMM040: µXRF imaging (PyMCA: ID18).

Figure A.III.27 BMM045: stereomicrograph image.

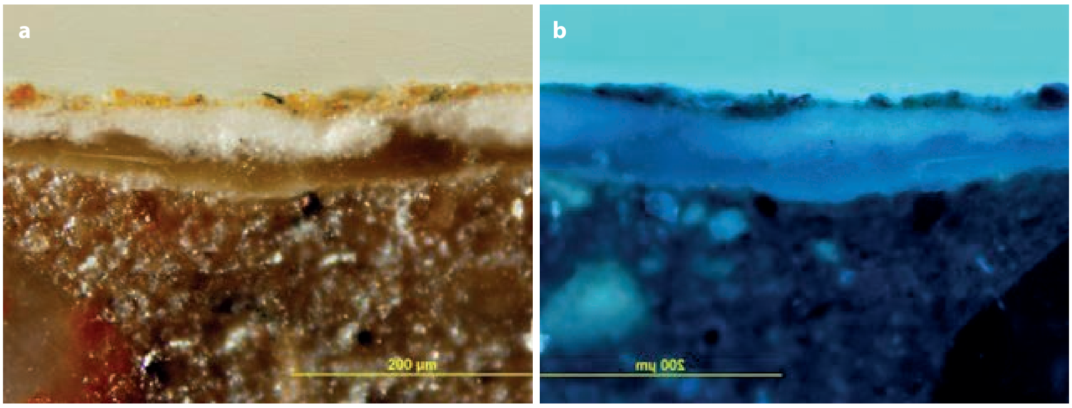


Figure A.III.28 BMM045: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

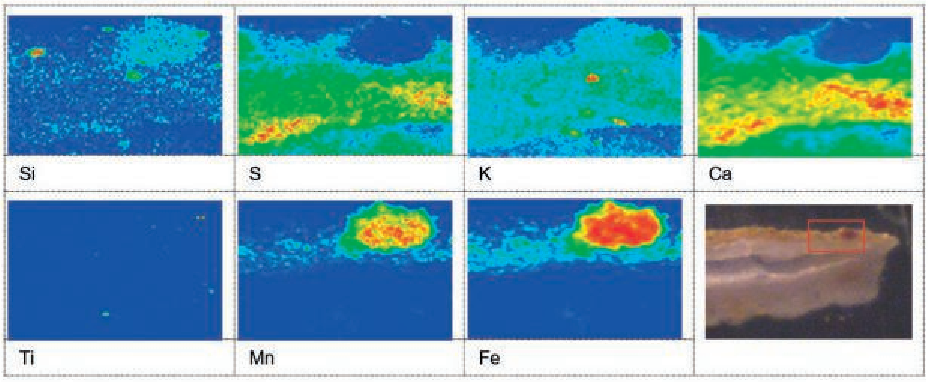


Figure A.III.29 BMM045: µXRF image (PyMCA: ID21).

Figure A.III.30 BMM055: stereomicrograph image.

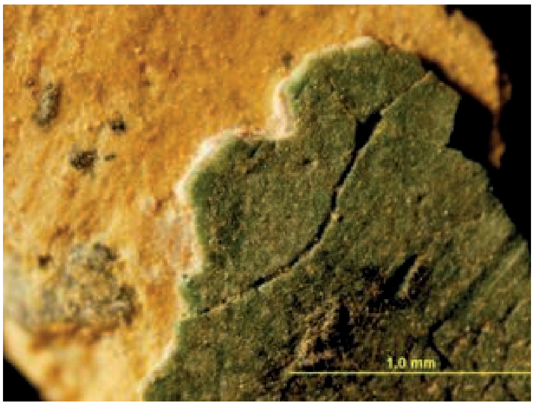


Figure A.III.33 BMM068: stereomicrograph image.

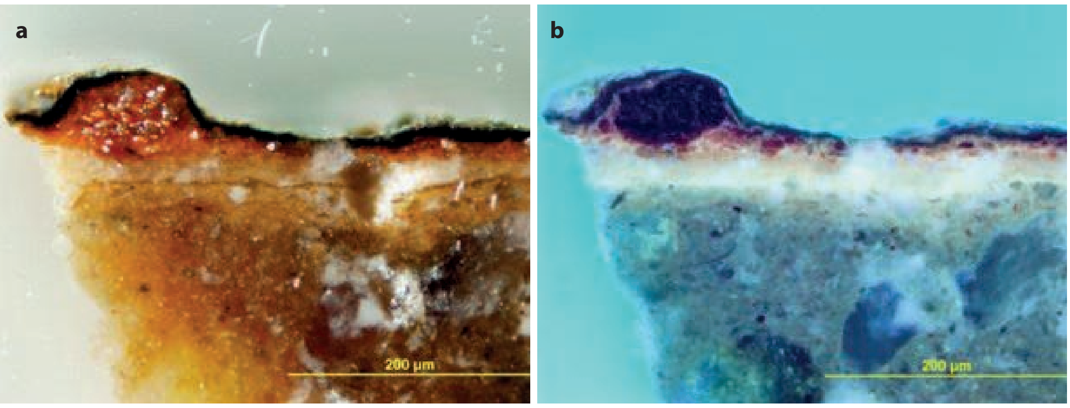


Figure A.III.31 BMM055: PLM photomicrograph of cross-section (a) in normal diffused light and (b) UV light.

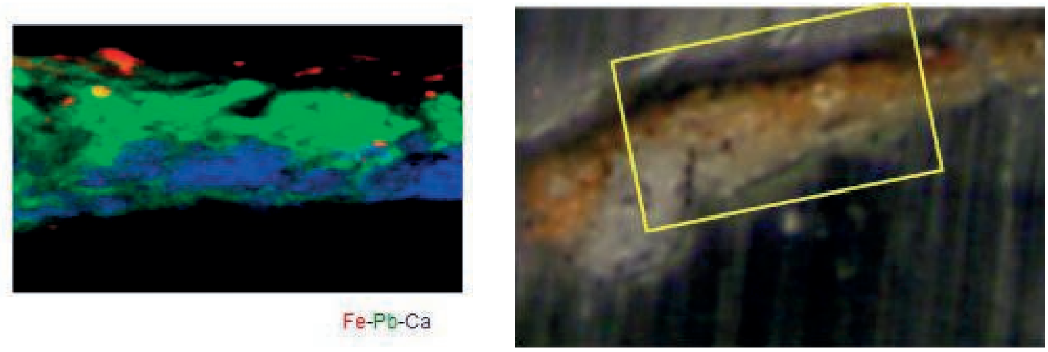


Figure A.III.32 BMM055: µXRF imaging (PyMCA: ID21).

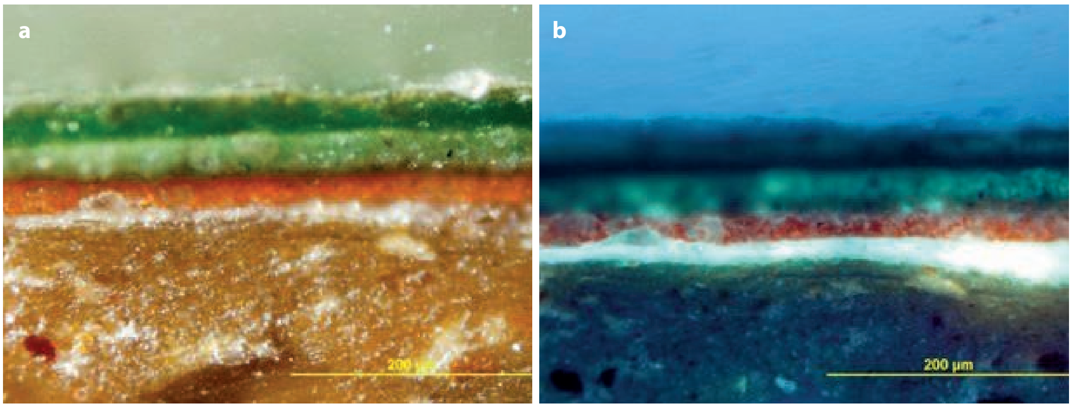


Figure A.III.34 BMM068: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

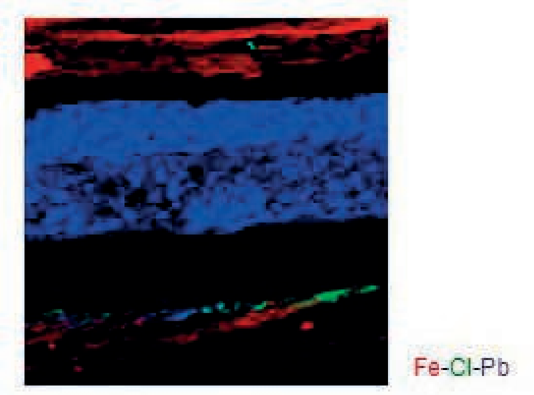


Figure A.III.35 BMM068: μXRF image (PyMCA: ID21).

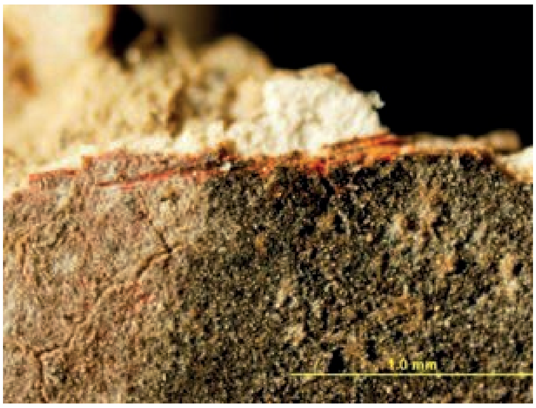


Figure A.III.36 BMM081: stereomicrograph image.

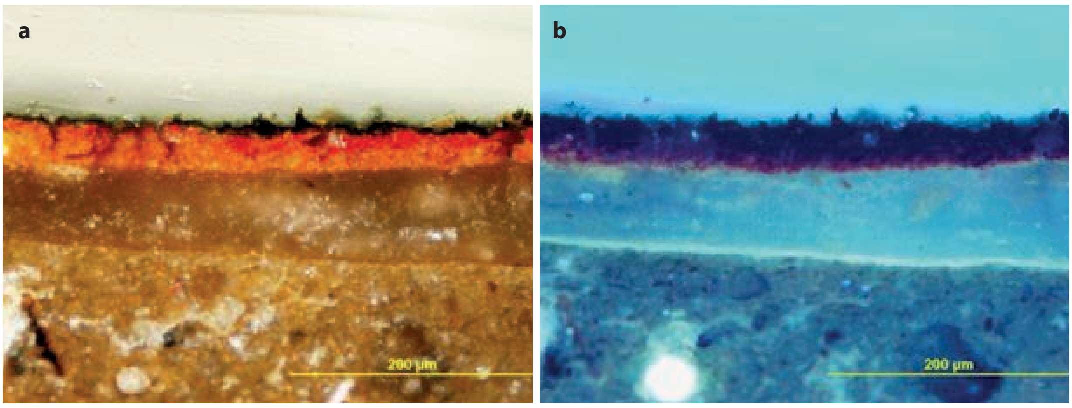


Figure A.III.37 BMM081: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

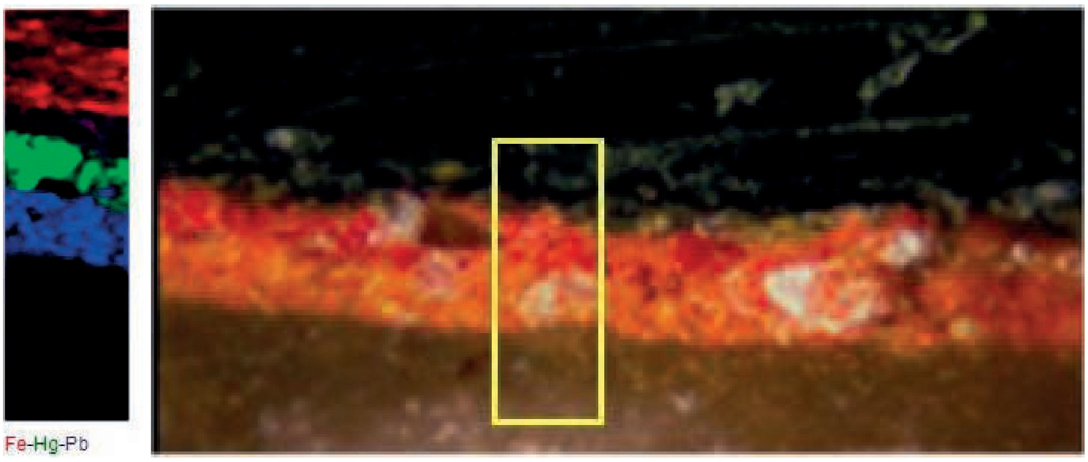


Figure A.III.38 BMM081: μXRF imaging (PyMCA: ID21).

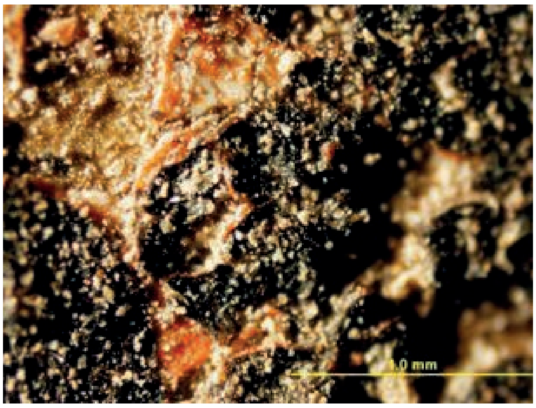


Figure A.III.39 BMM082: stereomicrograph image.

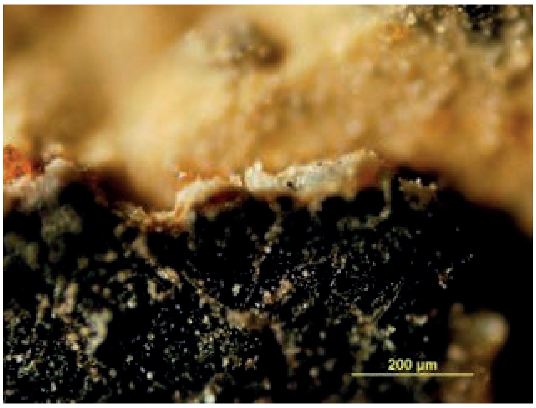


Figure A.III.43 BMM083: stereomicrograph image.

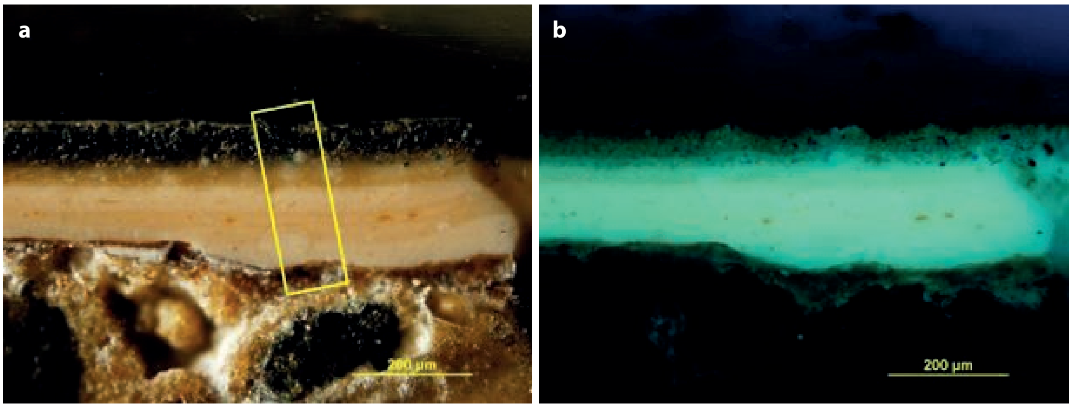


Figure A.III.40 BMM082: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

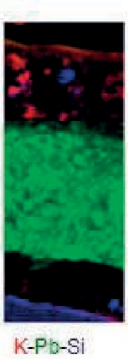


Figure A.III.41 BMM082: µXRF image (PyMCA: ID21).

Figure A.III.42 BMM082: µXRF image (PyMCA: ID18).

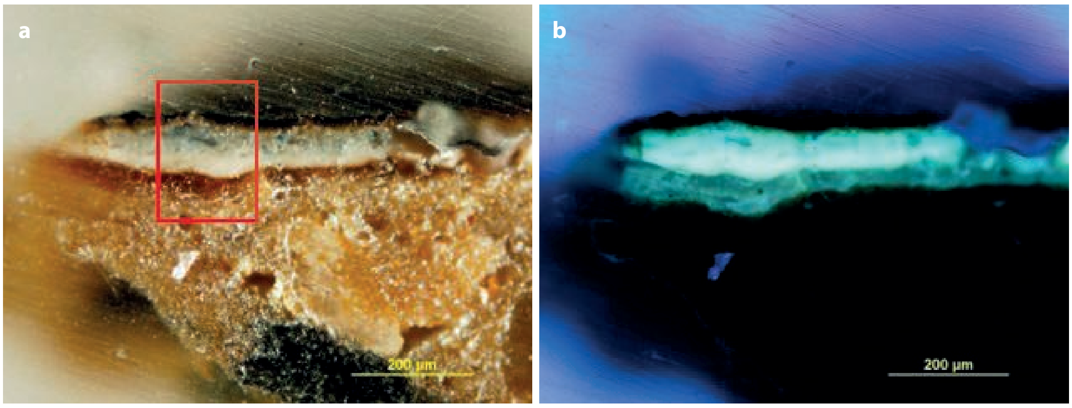


Figure A.III.44 BMM083: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light. The red frame is the imaging area.

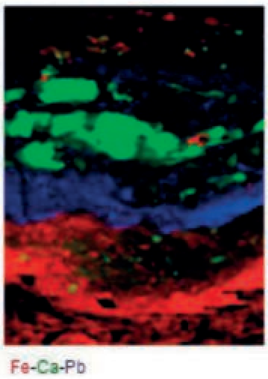


Figure A.III.45 BMM083: µXRF image (PyMCA: ID21).

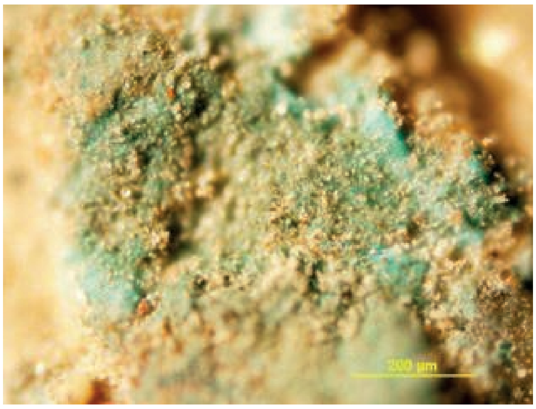


Figure A.III.46 BMM091: stereomicrograph image.

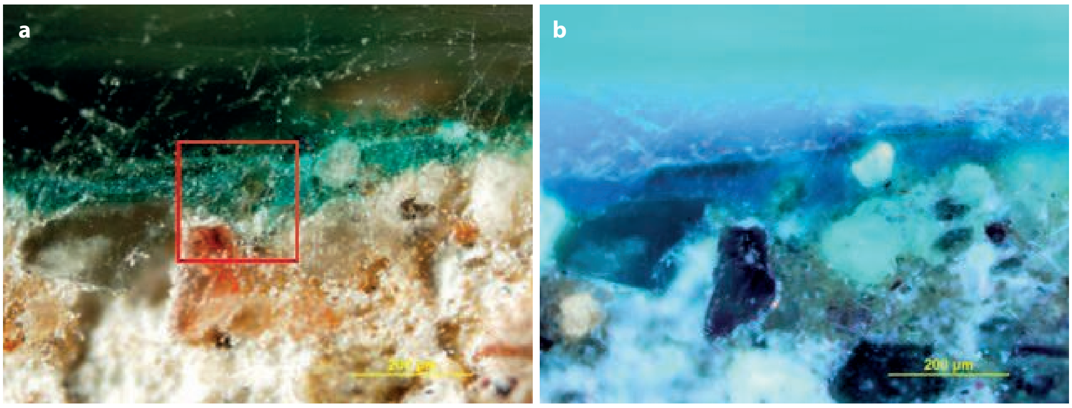


Figure A.III.47 BMM091: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light. The red frame is the imaging area.

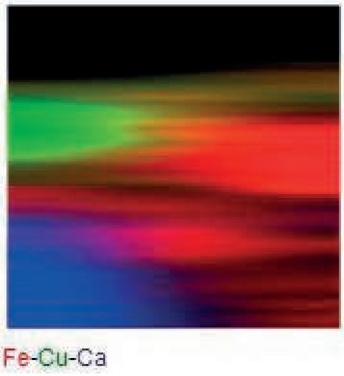


Figure A.III.48 BMM091: µXRF image (PyMCA: ID18).

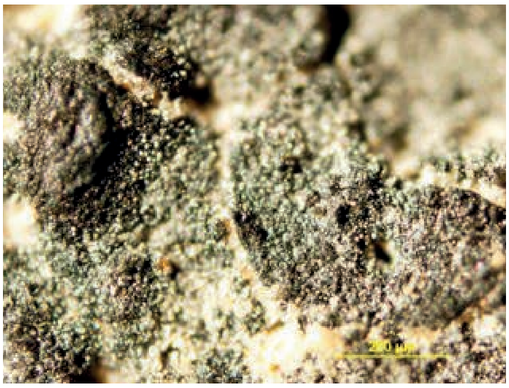


Figure A.III.49 BMM101: stereomicrograph image.

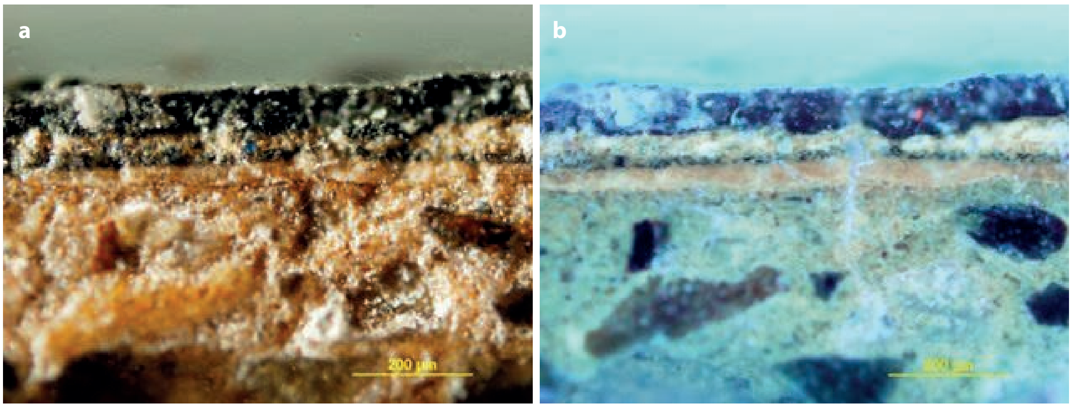


Figure A.III.50 BMM101: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

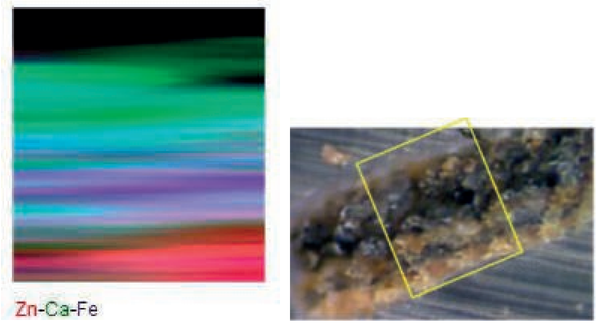


Figure A.III.51 BMM101: µXRF imaging (PyMCA: ID18).

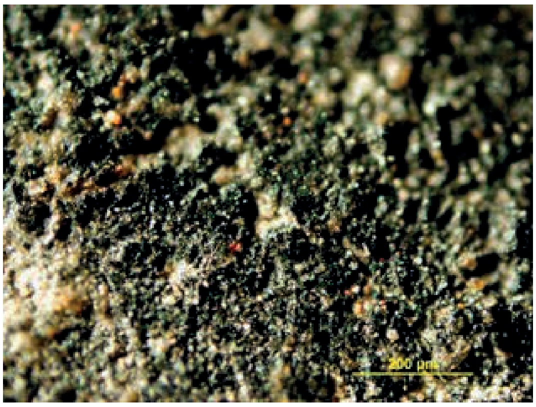


Figure A.III.52 BMM111: stereomicrograph image.

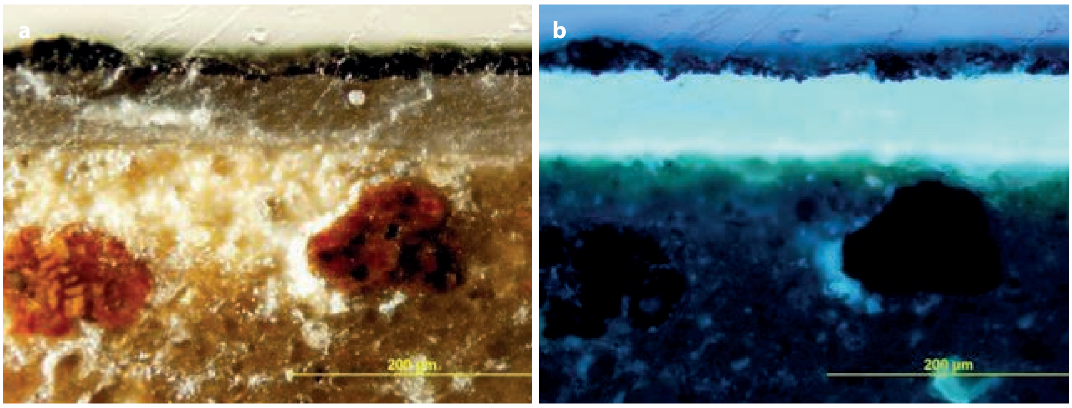


Figure A.III.53 BMM111: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

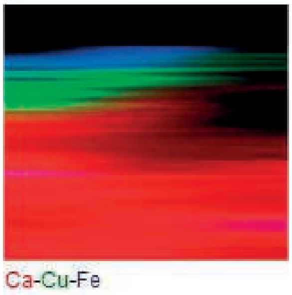


Figure A.III.54 BMM111: µXRF image (PyMCA: ID18).

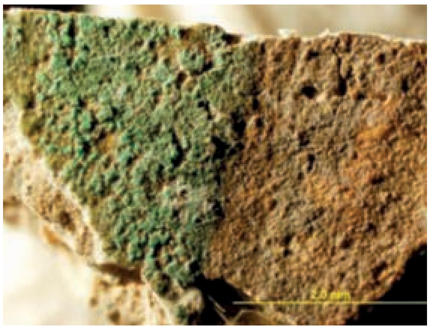


Figure A.III.55 BMM125: stereomicrograph image.

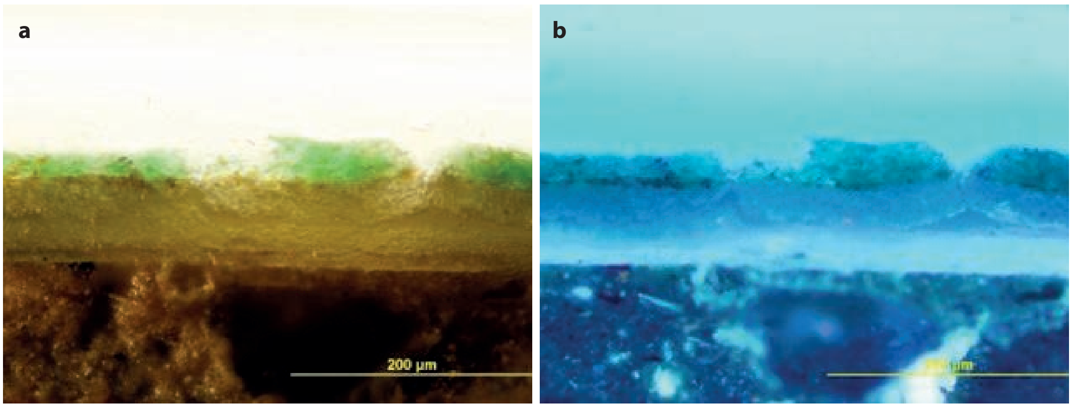


Figure A.III.56 BMM125: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

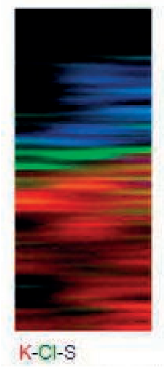


Figure A.III.57 BMM125: µXRF image (PyMCA: ID18).

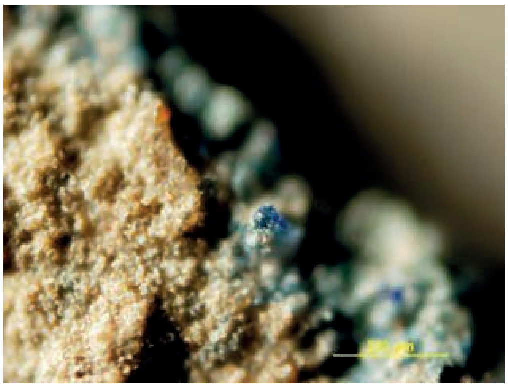


Figure A.III.58 BMM128: stereomicrograph image.

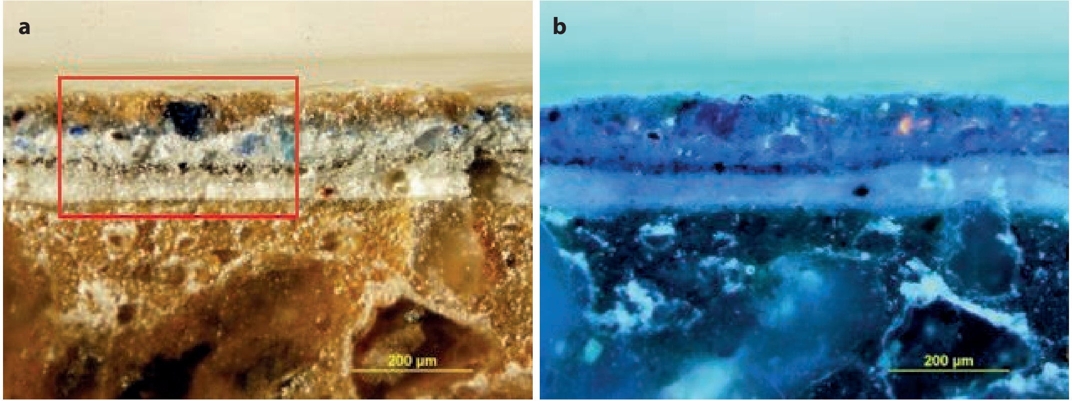


Figure A.III.59 BMM128: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light. The red frame is the imaging area.

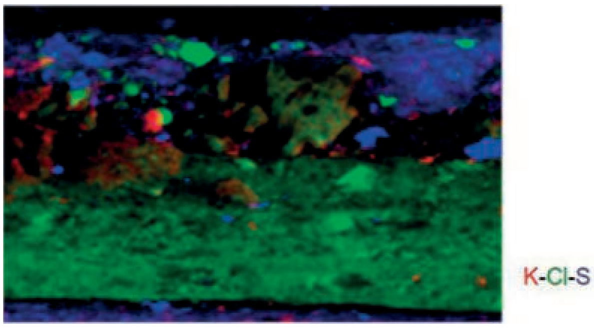


Figure A.III.60 BMM128: µXRF image (PyMCA: ID21).

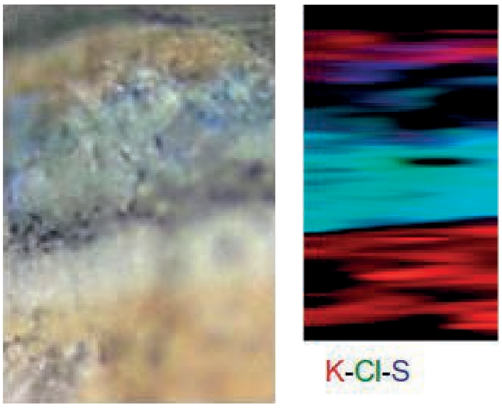


Figure A.III.61 BMM128: µXRF imaging (PyMCA: ID18).

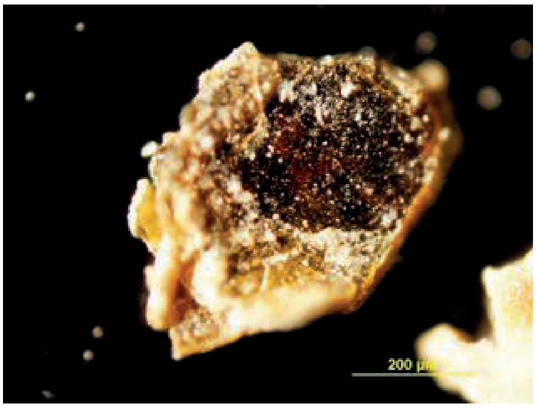


Figure A.III.62 BMM145: stereomicrograph image.

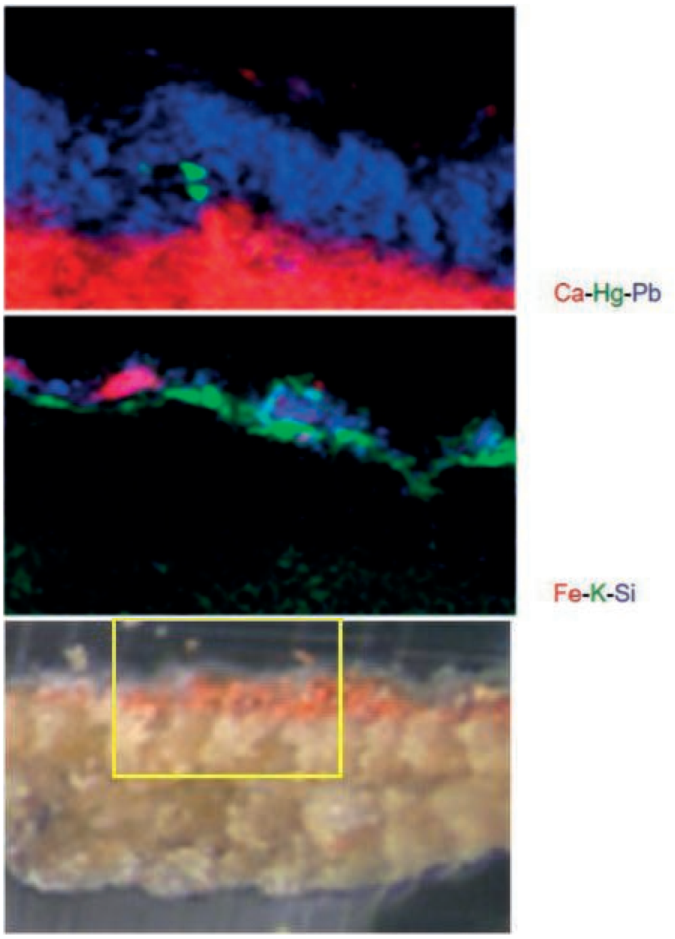


Figure A.III.63 BMM145: µXRF image (PyMCA: ID21).

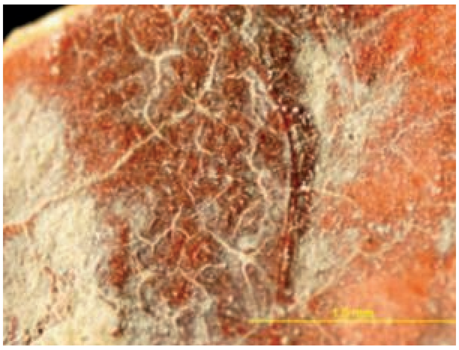


Figure A.III.64 BMM181: stereomicrograph image.

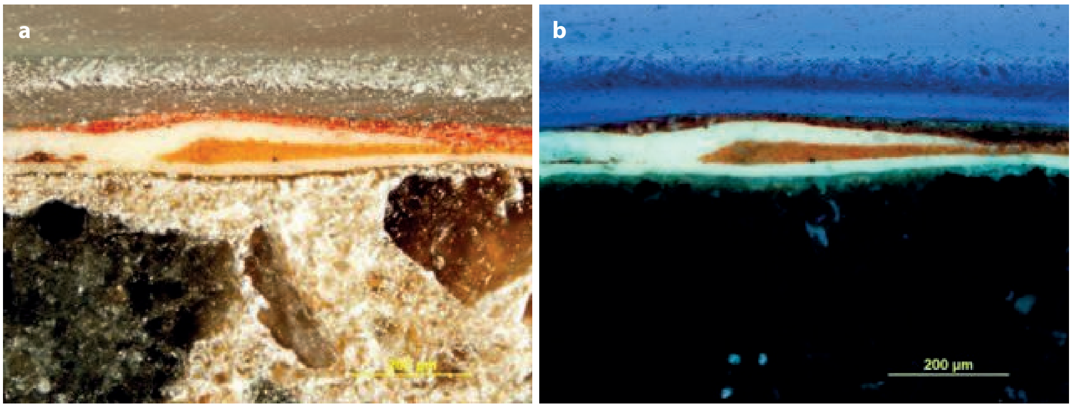


Figure A.III.65 BMM181: PLM photomicrograph of cross-section (a) in normal diffused light and (b) UV light.

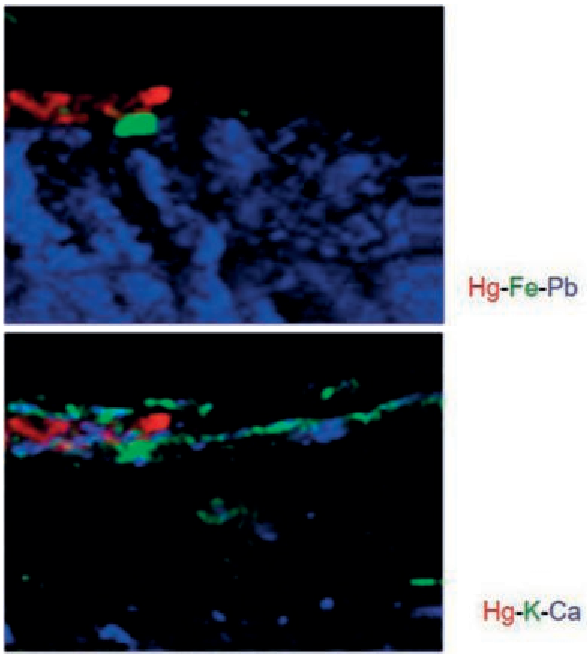


Figure A.III.66 BMM181: µXRF imaging (PyMCA: ID21)

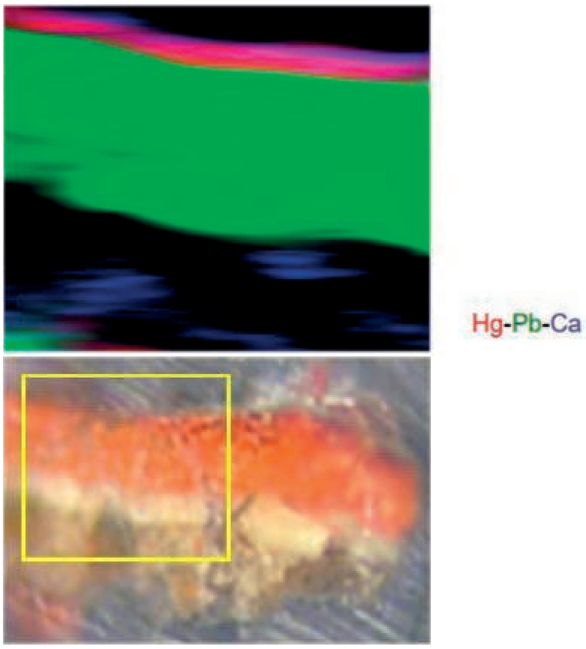


Figure A.III.67 BMM181: µXRF imaging (PyMCA: ID18).

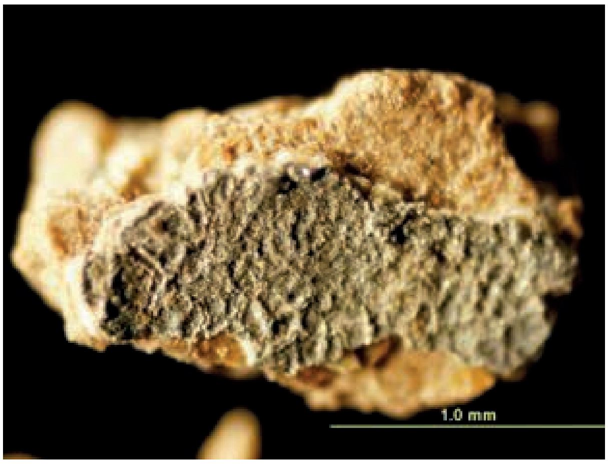


Figure A.III.68 BMM177: stereomicrograph image.

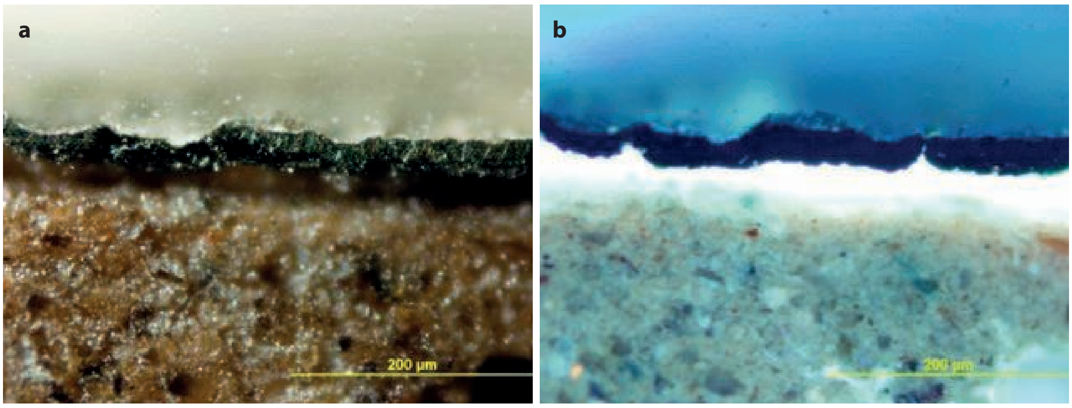


Figure A.III.69 BMM177: PLM photomicrograph of cross-section (a) in normal diffused light and (b) UV light.

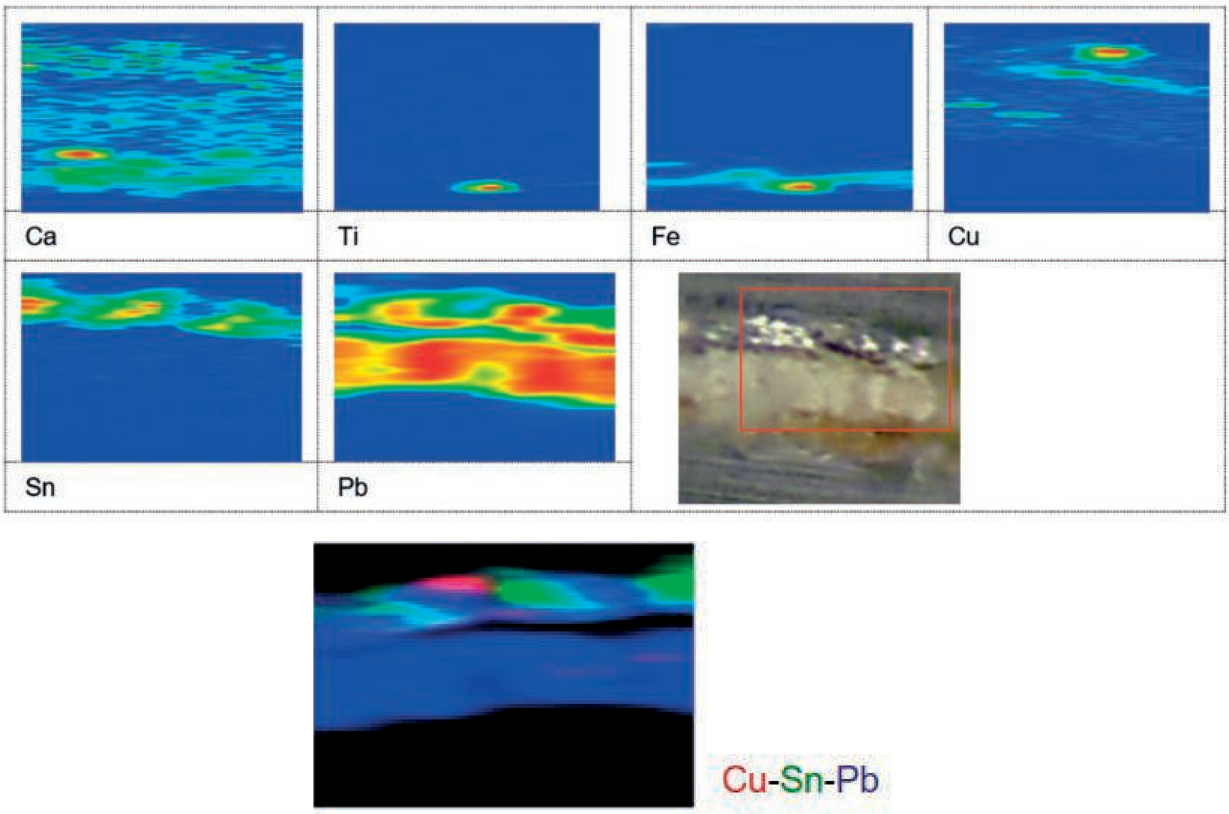


Figure A.III.70 BMM177: µXRF imaging (PyMCA: ID18).

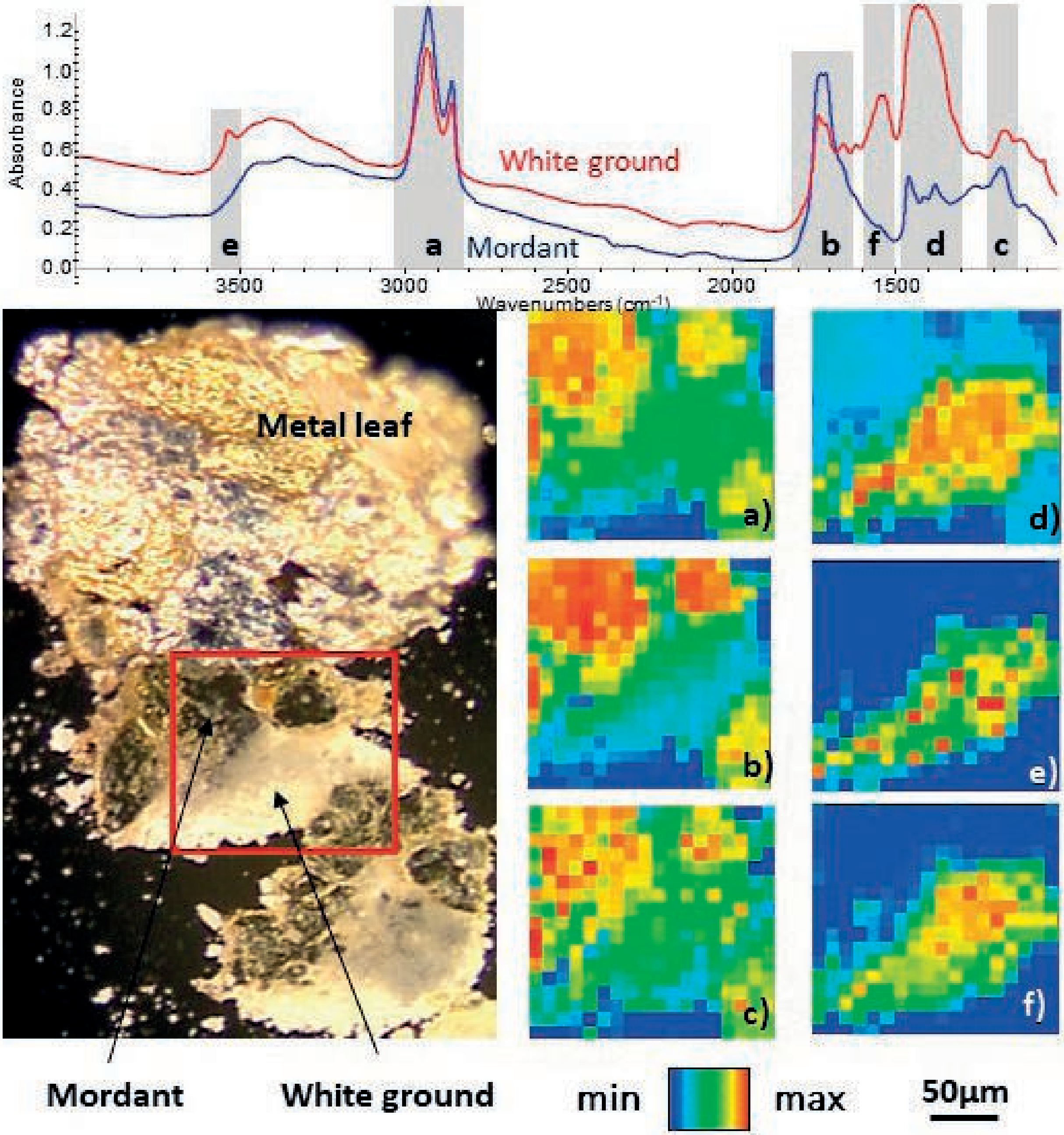


Figure A.III.71 µFTIR analysis of a fragment from sample BMM178. This sample is composed of a white ground, a mordant and a metal leaf. ROI analysis of the FTIR shows the presence of an oil based mordant: ((a) v(CH), (b) ester v (CO), (c), δ(C-H), v(C-O) (c)), and a white ground layer made of lead white ((d) carbonate v3(C-O) and (e) lead hydroxide v(O-H)) and saponified oil ((f) lead carboxylate vAS (CO)). Spectra shown are averages calculated over the two layers. The grey rectangles represent the ROIs used for the calculation of the maps.

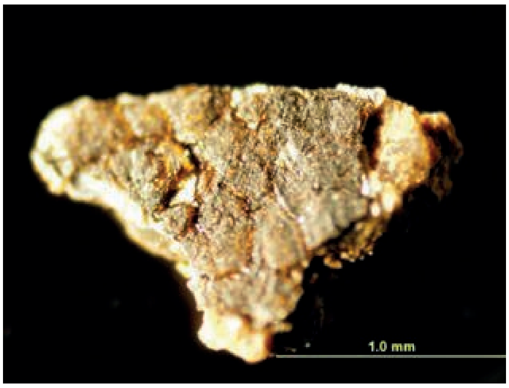


Figure A.III.72 BMM186: stereomicrograph image.

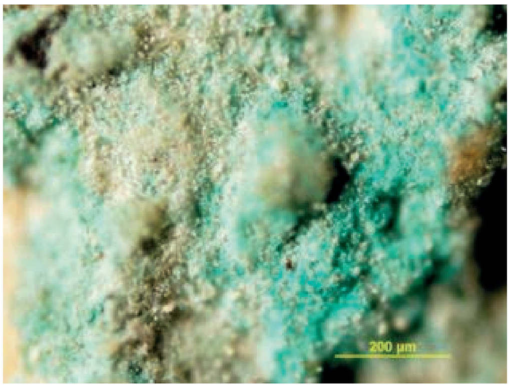


Figure A.III.75 BMM210: stereomicrograph image.

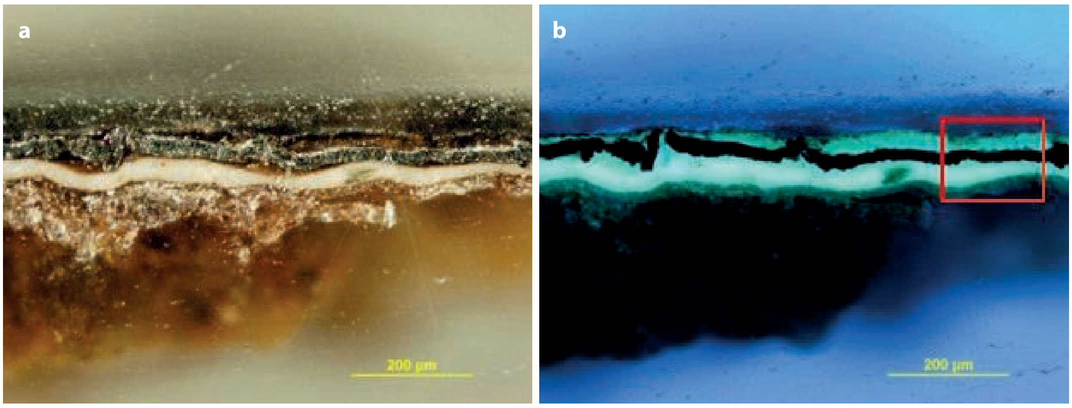


Figure A.III.73 BMM186: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light. The red frame is the imaging area.

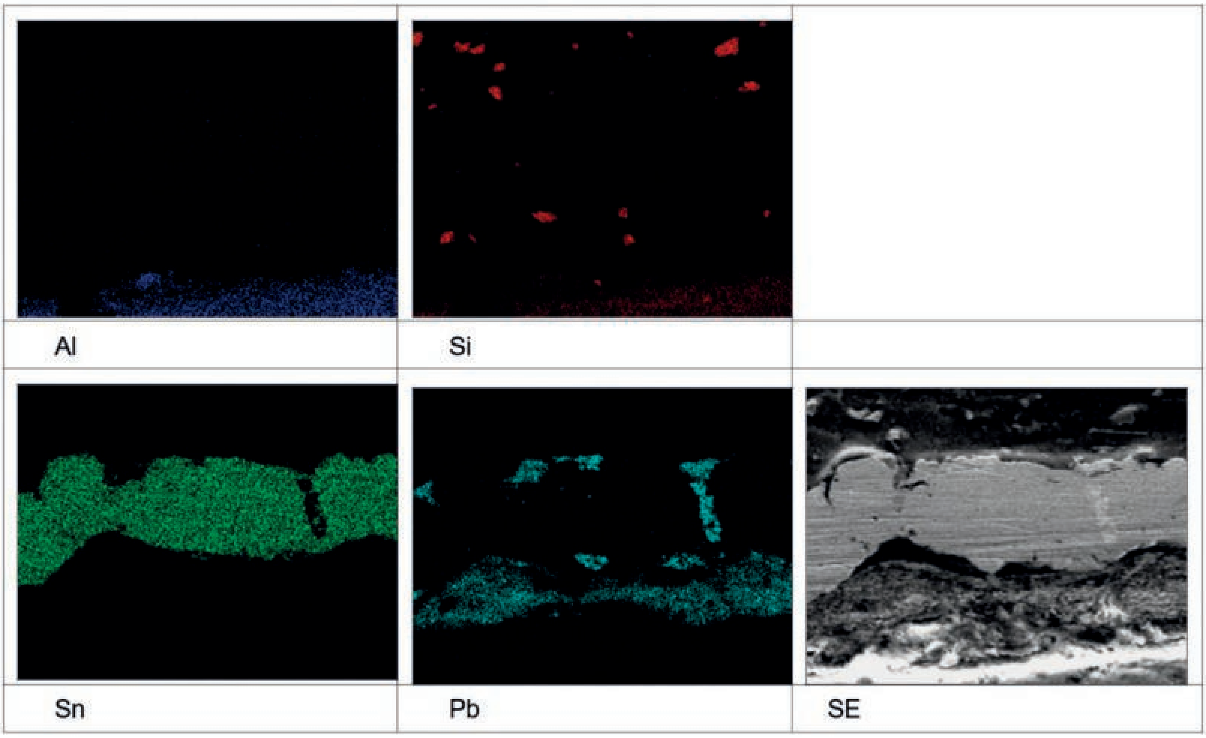


Figure A.III.74 BMM186: mapping using SEM-EDS.

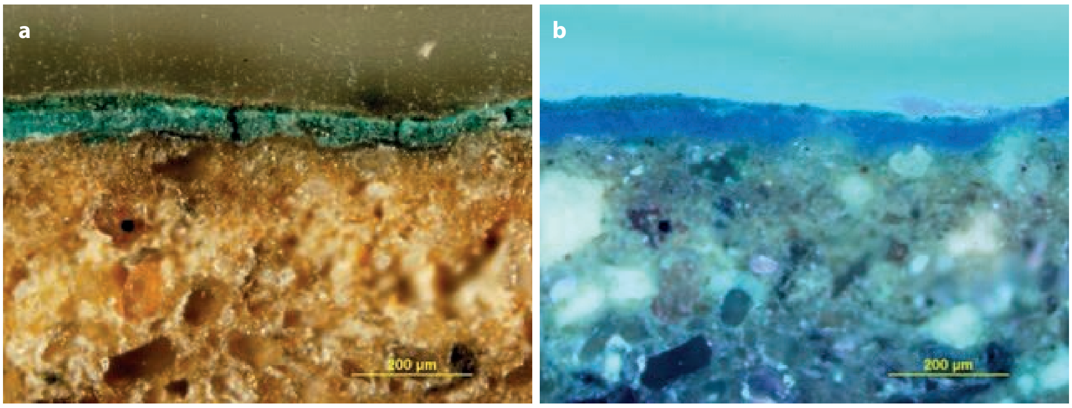


Figure A.III.76 BMM210: PLM photomicrograph of cross-section in (a) normal diffused light and (b) UV light.

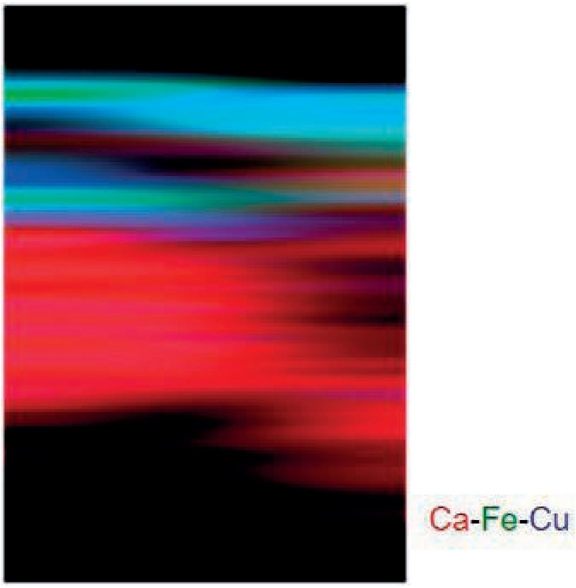


Figure A.III.77 BMM210: µXRF image (PyMCA: ID18).

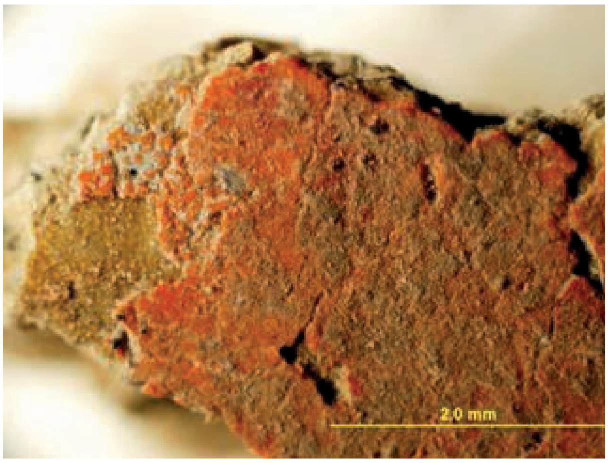


Figure A.III.78 FDM059: stereomicrograph image.

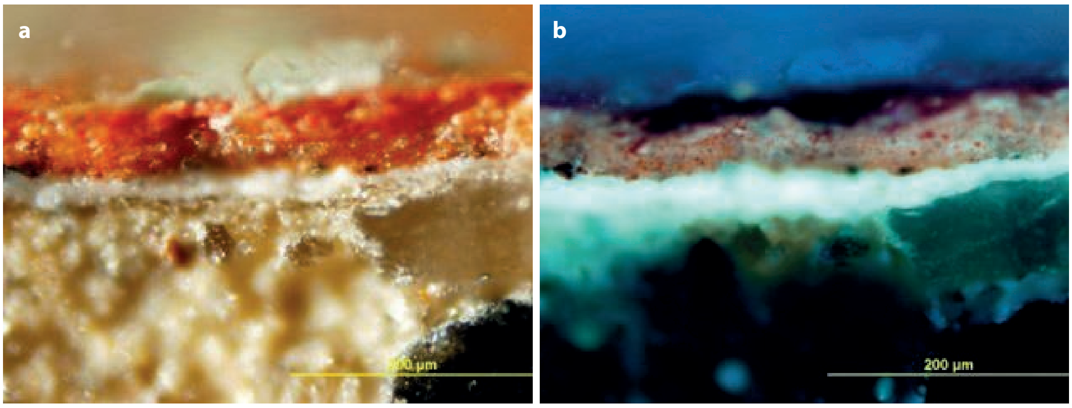


Figure A.III.79 FDM059: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

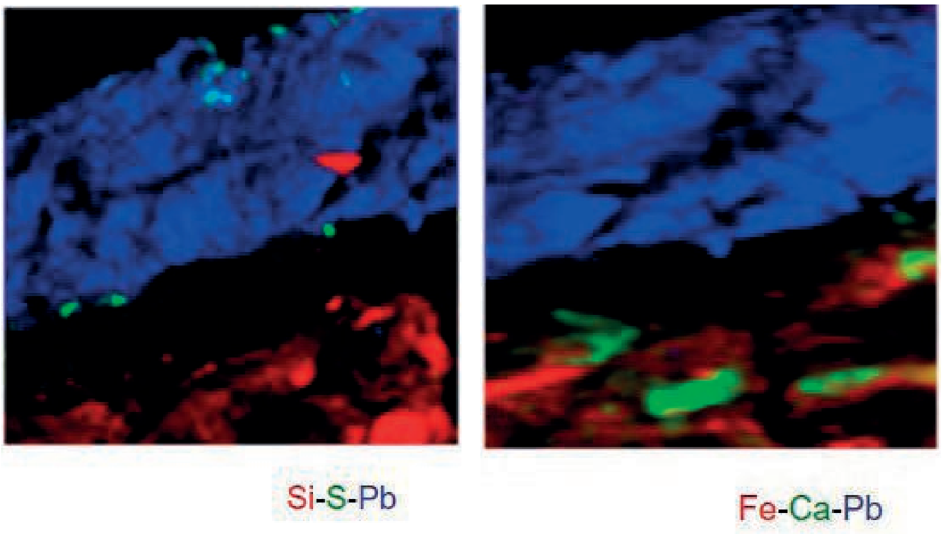


Figure A.III.80 FDM059: µXRF imaging (PyMCA: ID21).

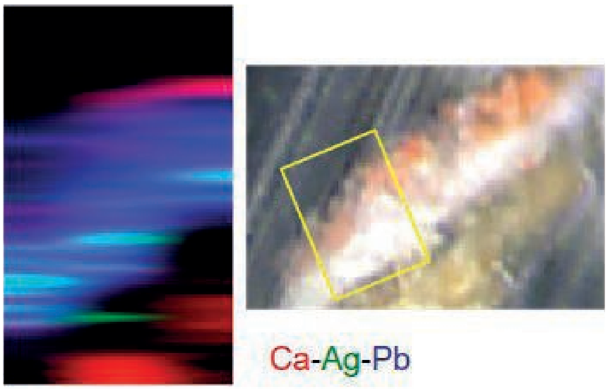


Figure A.III.81 FDM059: µXRF image (PyMCA: ID18).

Figure A.III.82 FDM011: stereomicrograph image.

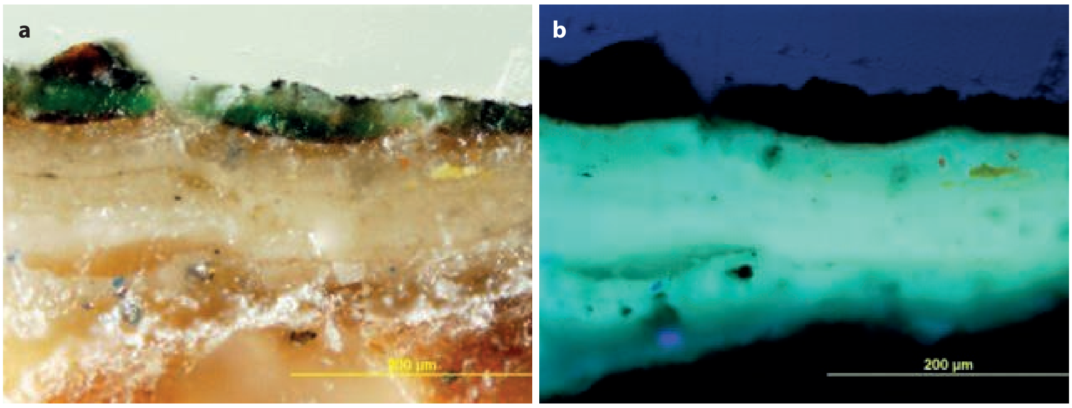


Figure A.III.83 FDM011: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

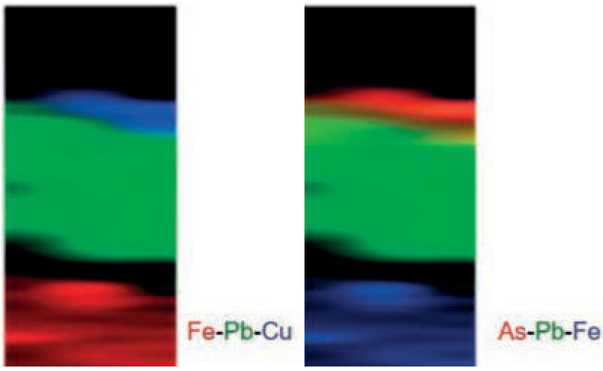


Figure A.III.84 FDM011: µXRF imaging (PyMCA: ID18).

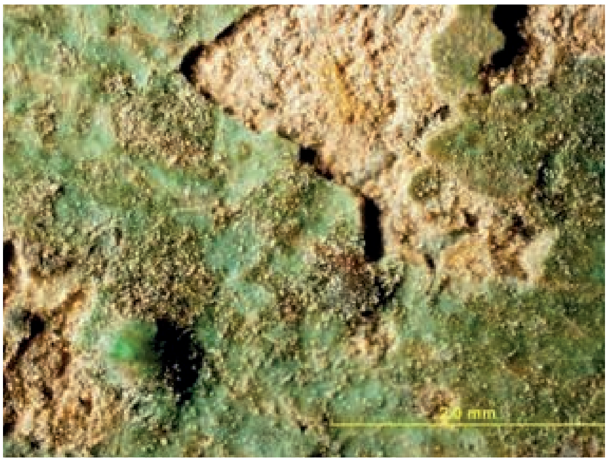


Figure A.III.85 FDM053: stereomicrograph image.

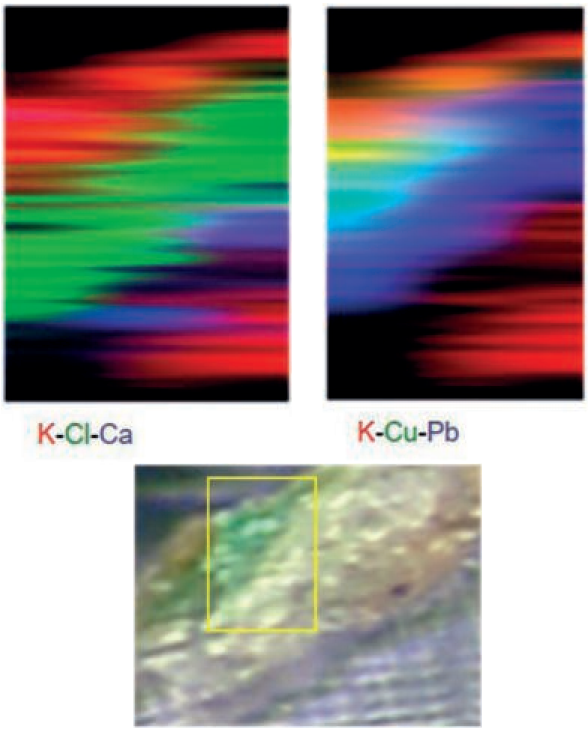


Figure A.III.86 FDM053: µXRF imaging (PyMCA: ID18).

Figure A.III.87 FDM033: stereomicrograph image.

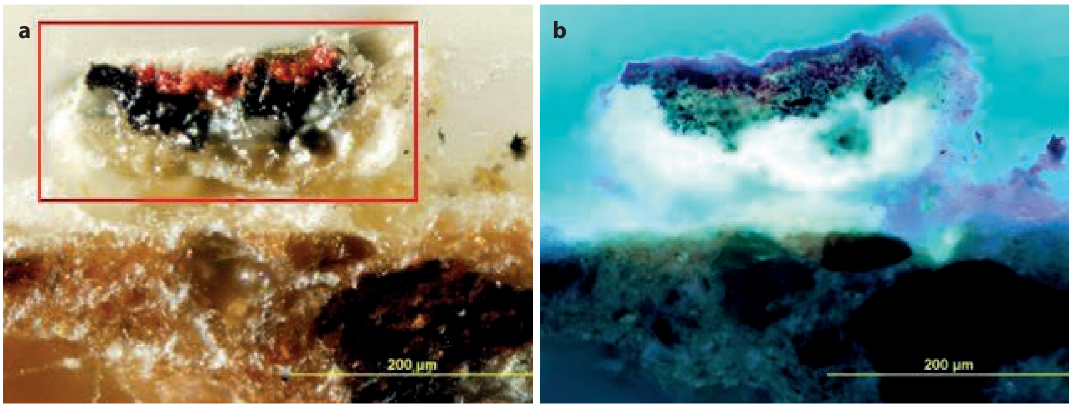


Figure A.III.88 FDM033: PLM photomicrograph of cross-section in (a) normal diffused light and (b) in UV light. The red frame is the imaging area.

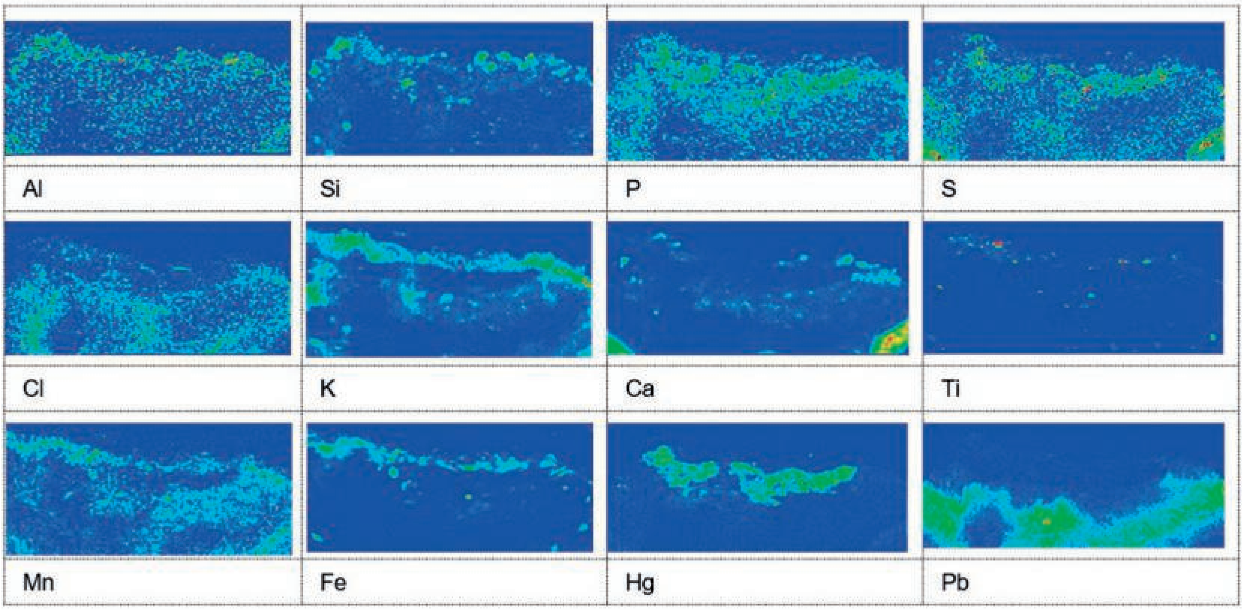


Figure A.III.89 FDM033: µXRF image (PyMCA: ID21).

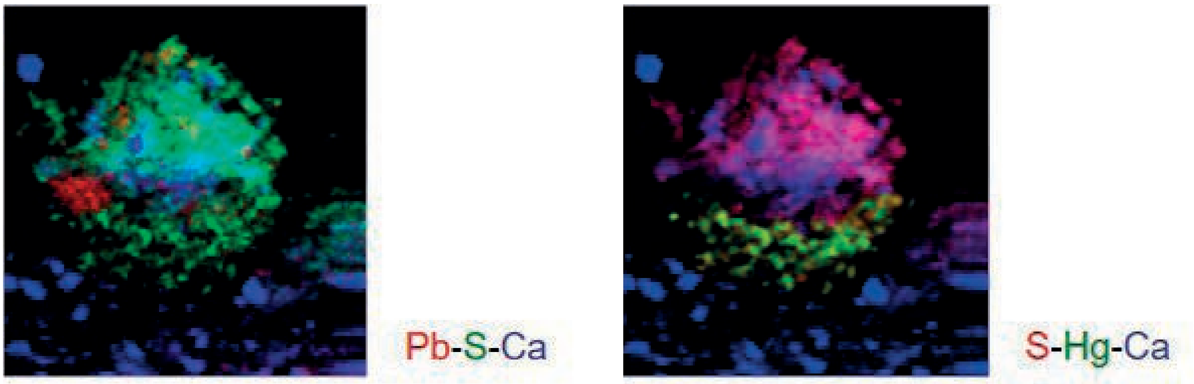


Figure A.III.90 FDM033: µXRF image (PyMCA: ID21).

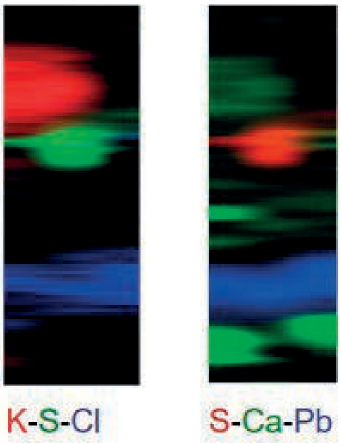


Figure A.III.91 FDM033: µXRF image (PyMCA: ID18).

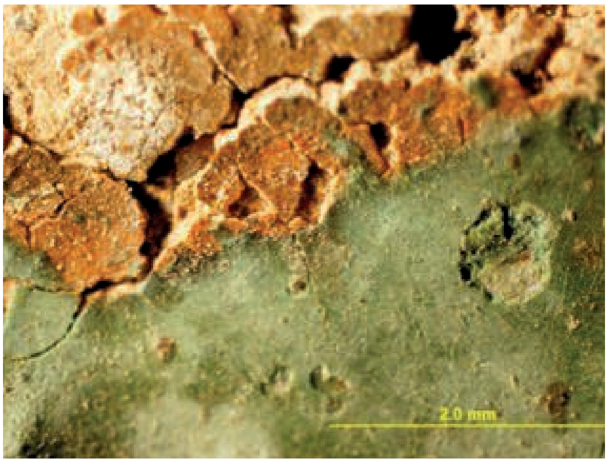


Figure A.III.92 FDM055: stereomicrograph image.

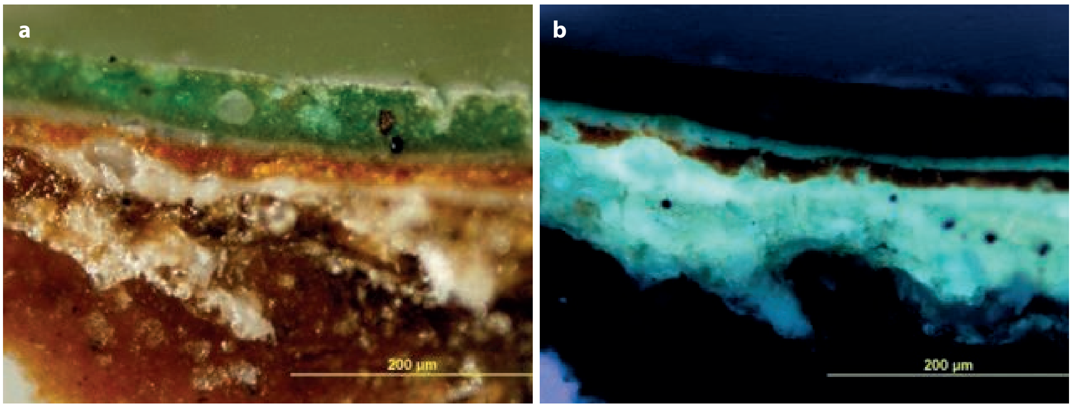


Figure A.III.93 FDM055: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

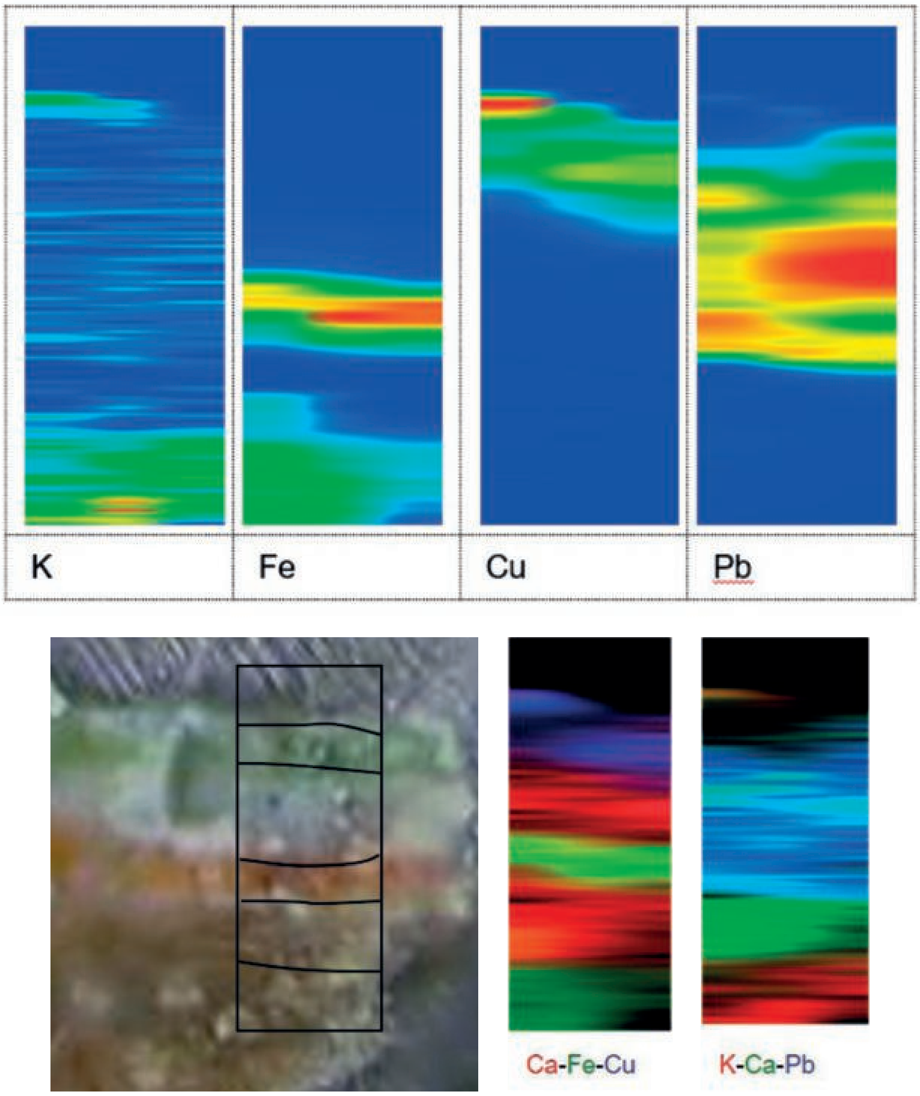


Figure A.III.94 FDM055: µXRF image (PyMCA: ID18).

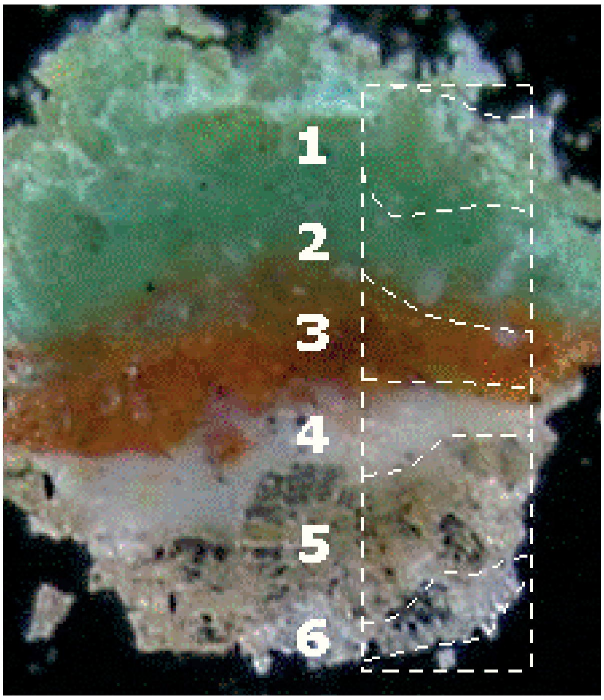


Figure A.III.95 FDM055: observable multilayered structure when the sample is compressed between diamond cells.

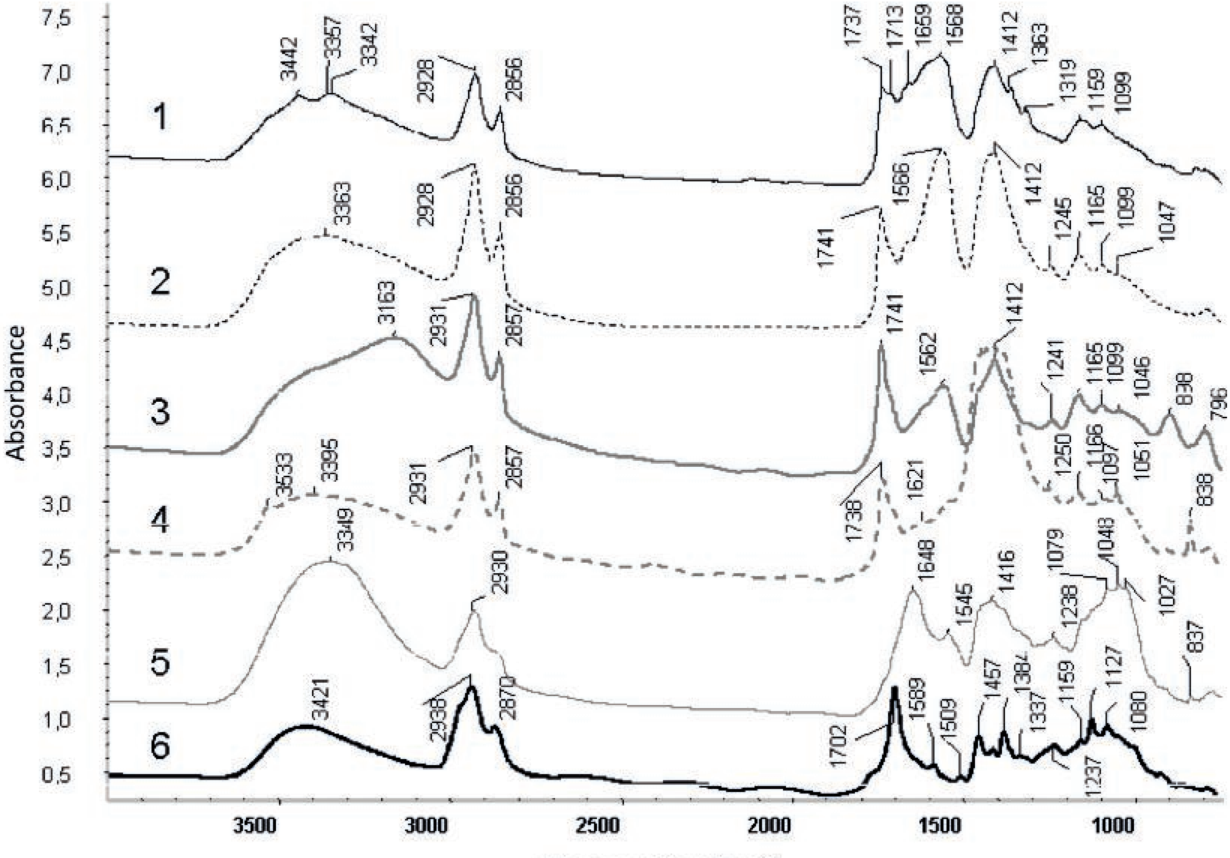


Figure A.III.96 FDM055: spectra of each layer obtained by µFTIR (corresponding to layer numbers in the figure at left).

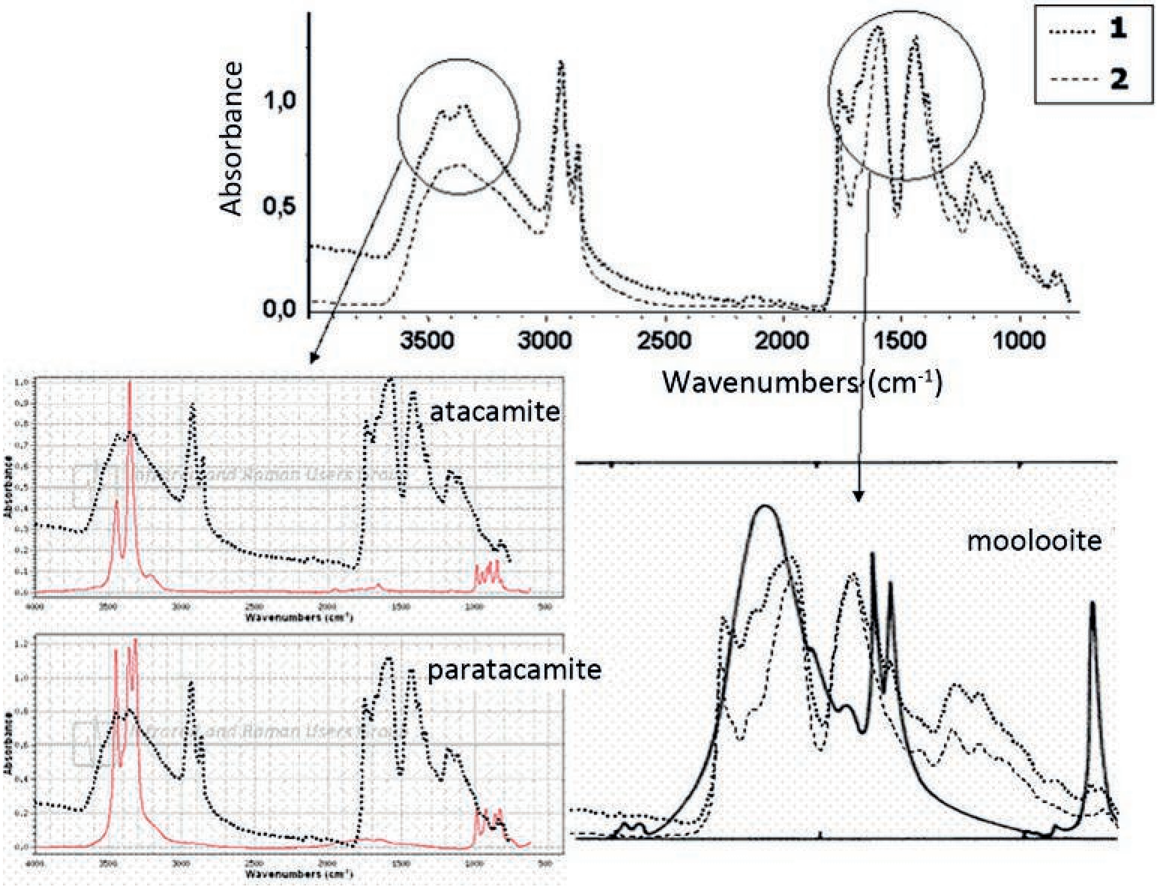


Figure A.III.97 FDM055: comparison of the bright green portion of the painted surface (1) and the green layer (2) with the spectra of atacamite and moolooite.

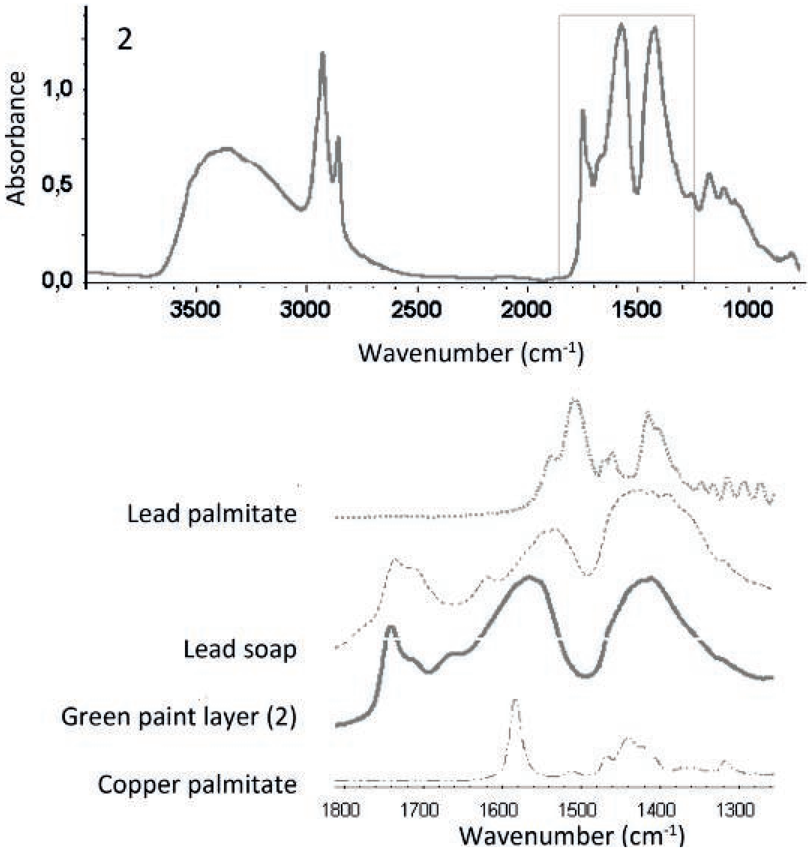


Figure A.III.98 FDM055: comparison of the spectra of the green-coloured layer (2) with the organometallic salts produced by the reaction of copper and lead to pigments in the oil.

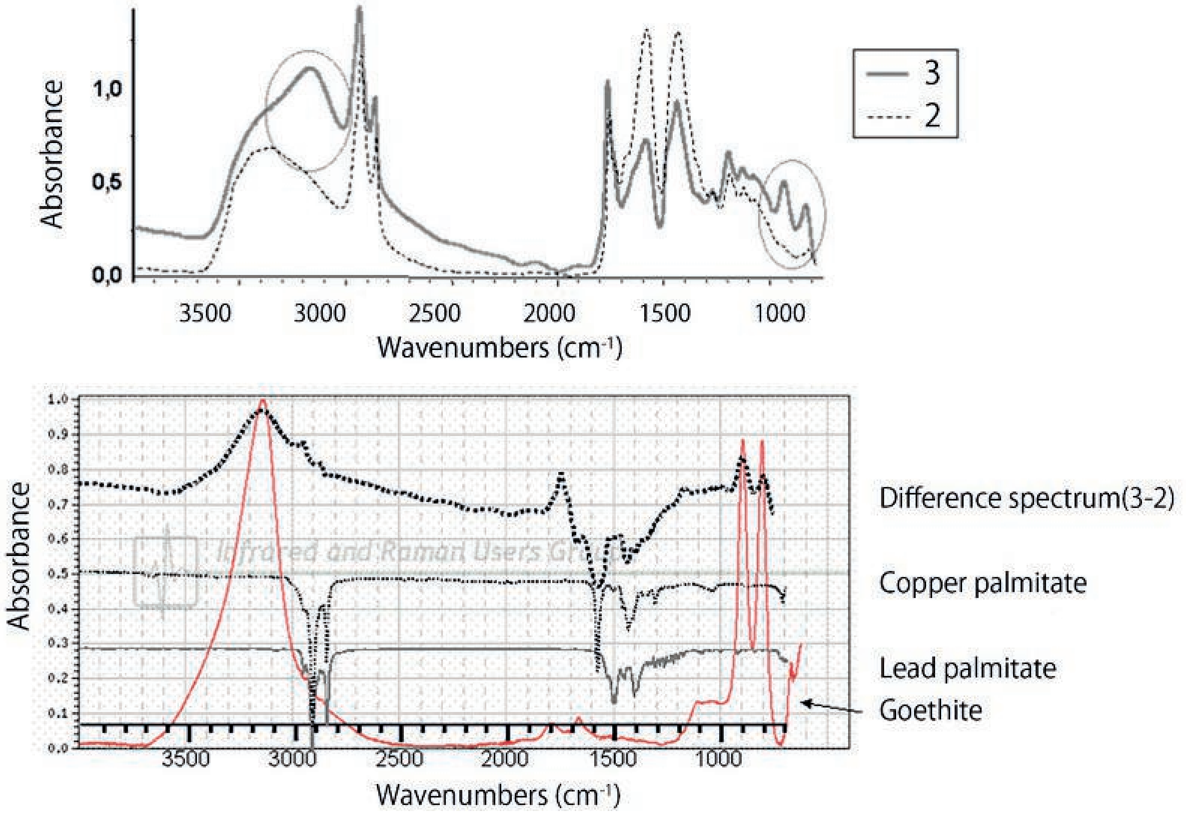


Figure A.III.99 FDM055: comparison of the different spectra of the red-coloured layer (3) and the green-coloured layer (2) with the spectra of goethite and organometallic salts.

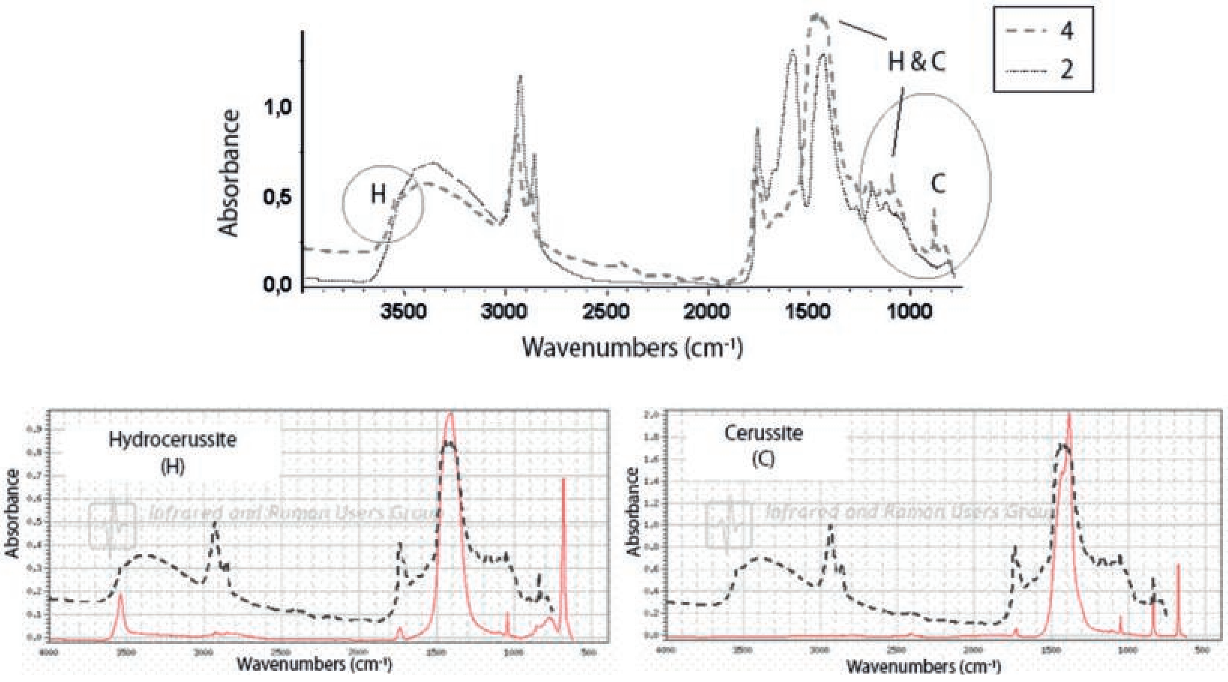


Figure A.III.100 FDM055: comparison of the spectra of the white ground layer (4) with those of the green painting layer (2) and lead white (hydrocerussite and cerussite).

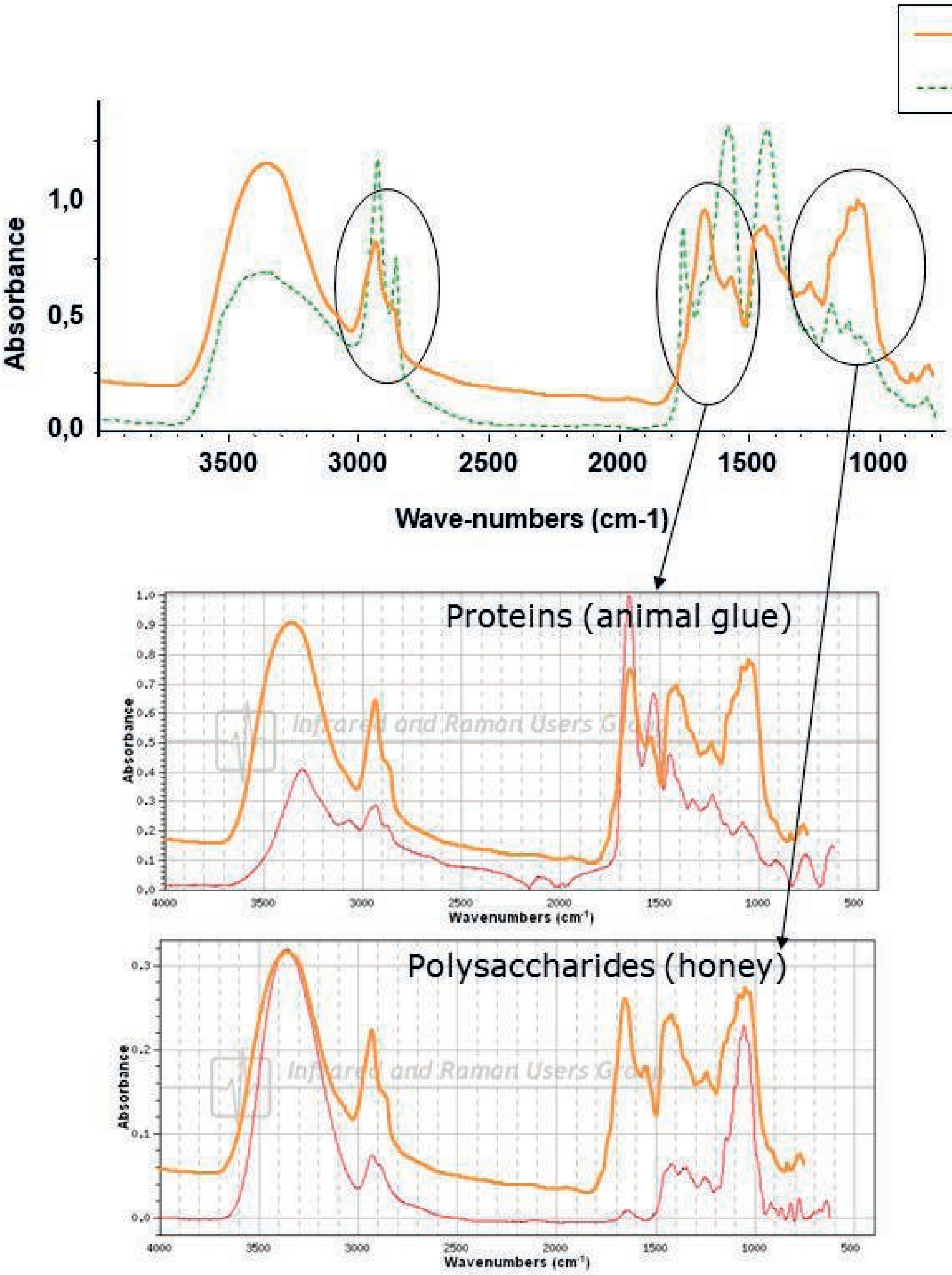


Figure A.III.101 FDM055: comparison of the spectra of the yellow transparent sizing layer (5) with proteins (reference: animal glue) and polysaccharides (reference: honey).

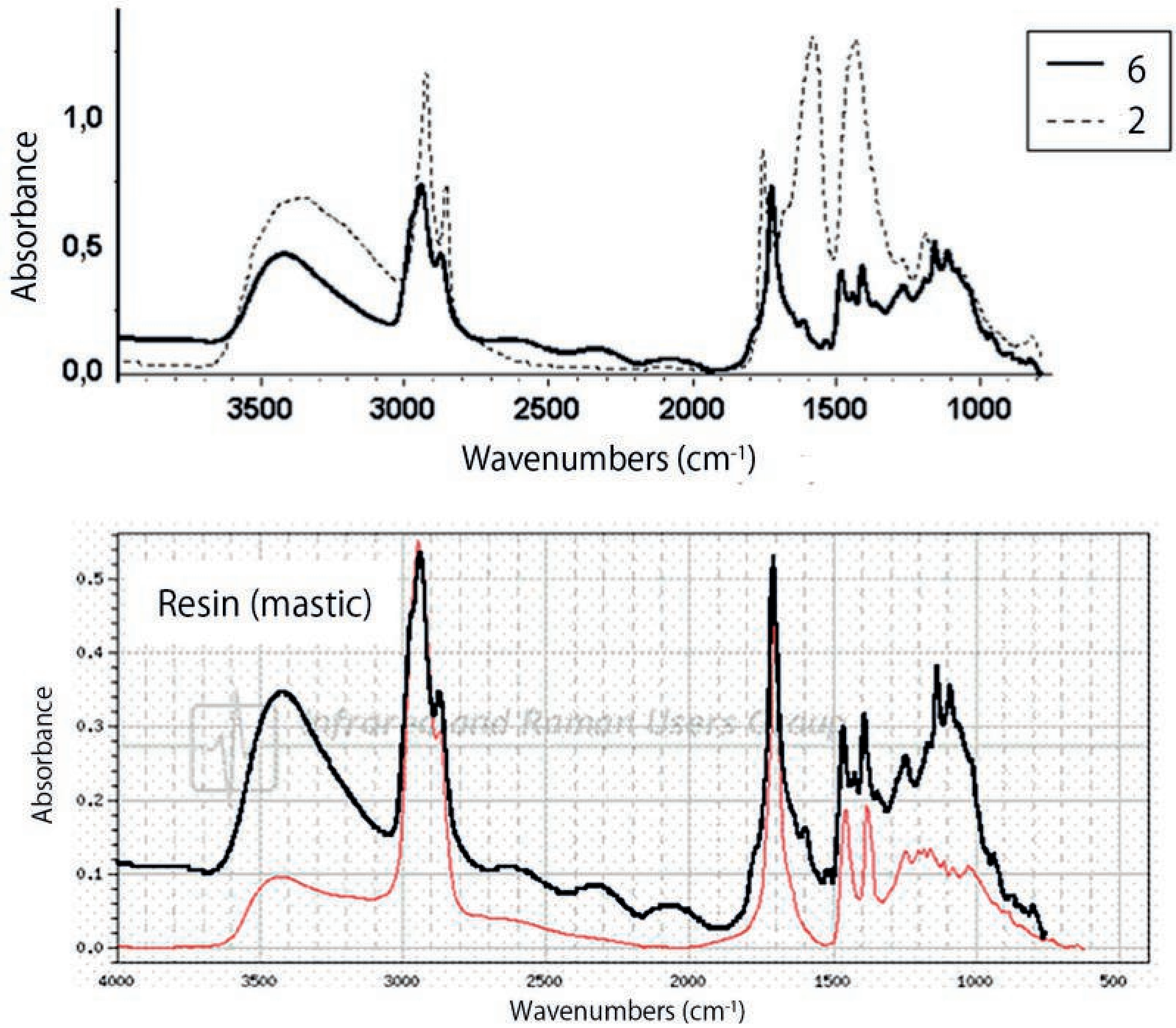


Figure A.III.102 FDM055: comparison of the spectra of the transparent sizing layer (6) with that of a natural resin (reference: mastic resin).

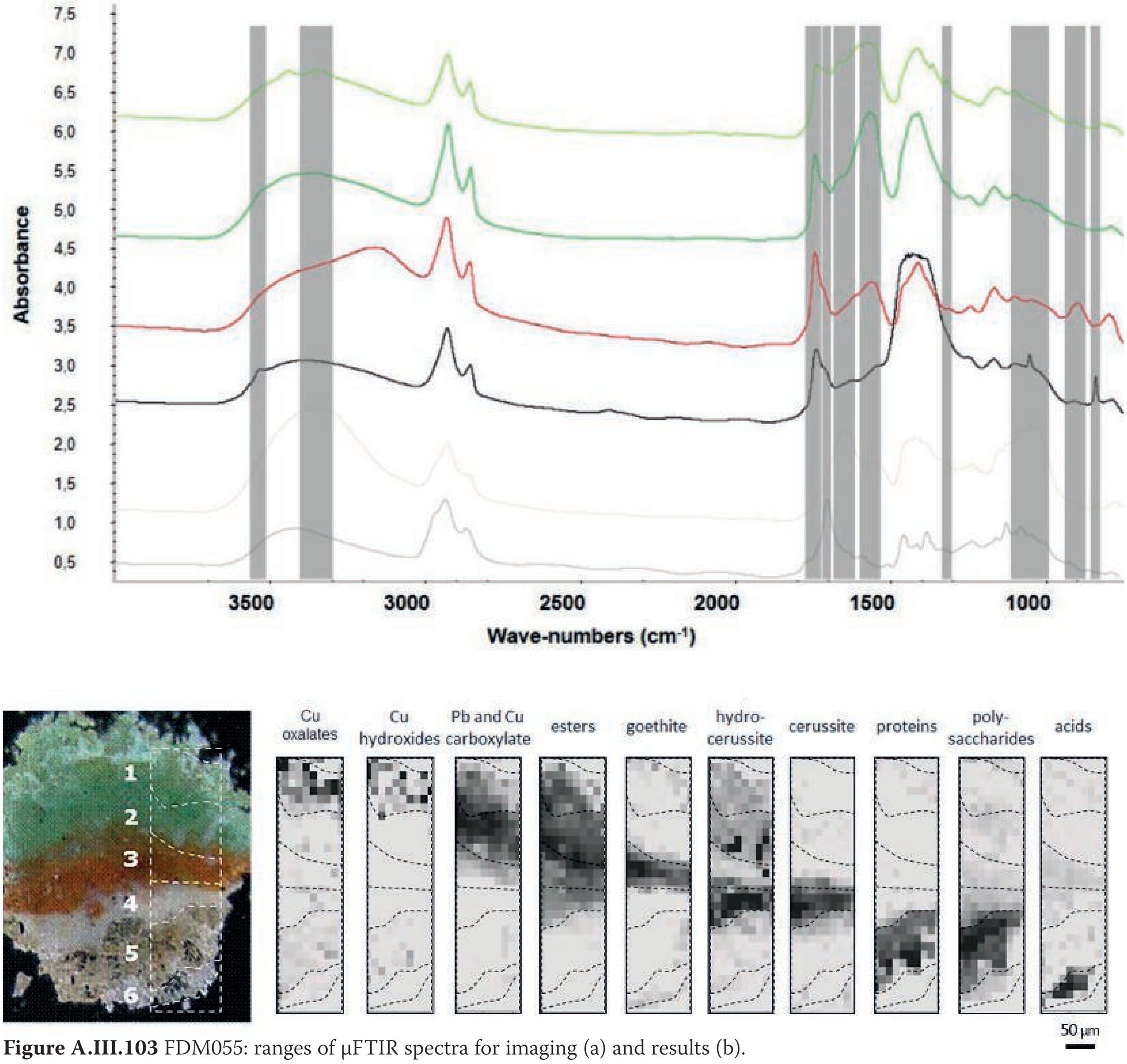


Figure A.III.103 FDM055: ranges of μFTIR spectra for imaging (a) and results (b).

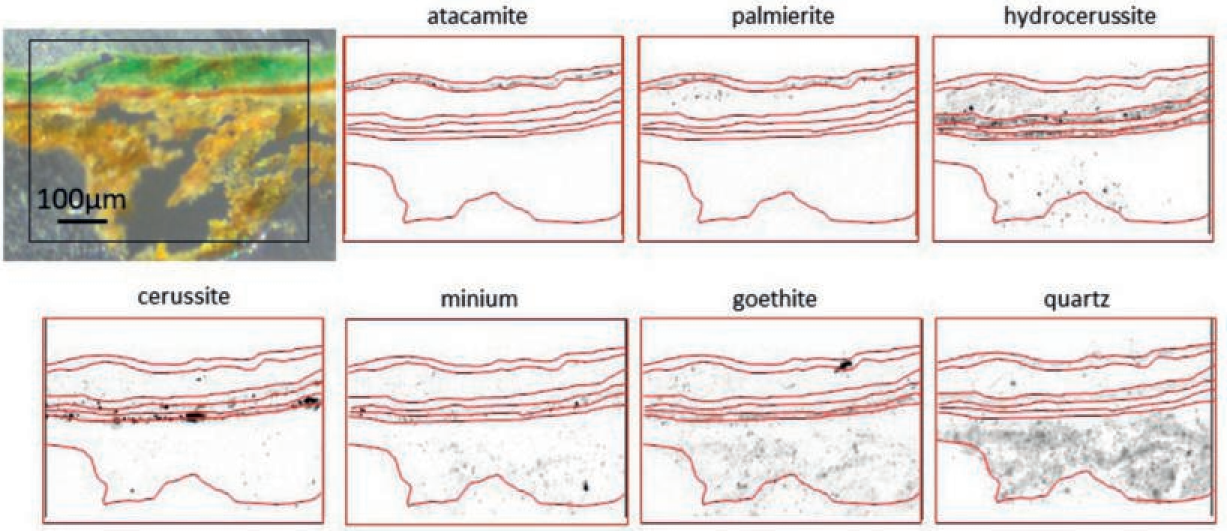


Figure A.III.104 FDM055: µXRD crystalline phase maps of atacamite, palmierite, hydrocerussite, cerussite, minium, goethite and quartz on a thin sample (ID13).

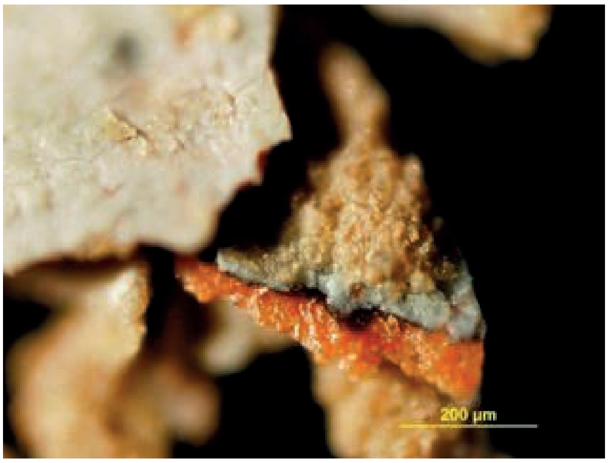


Figure A.III.105 FDM037: stereomicrograph image.

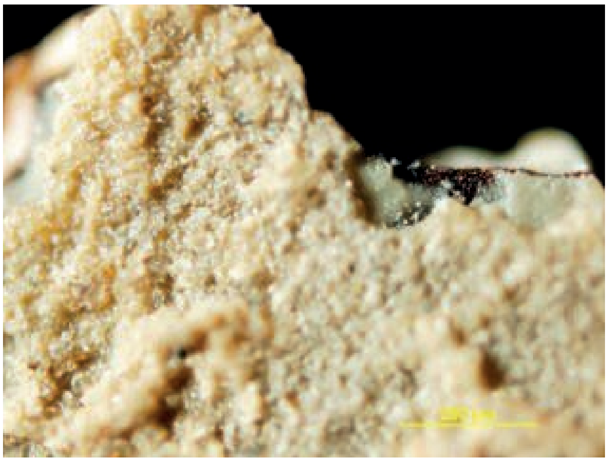


Figure A.III.109 KAK03: stereomicrograph image.

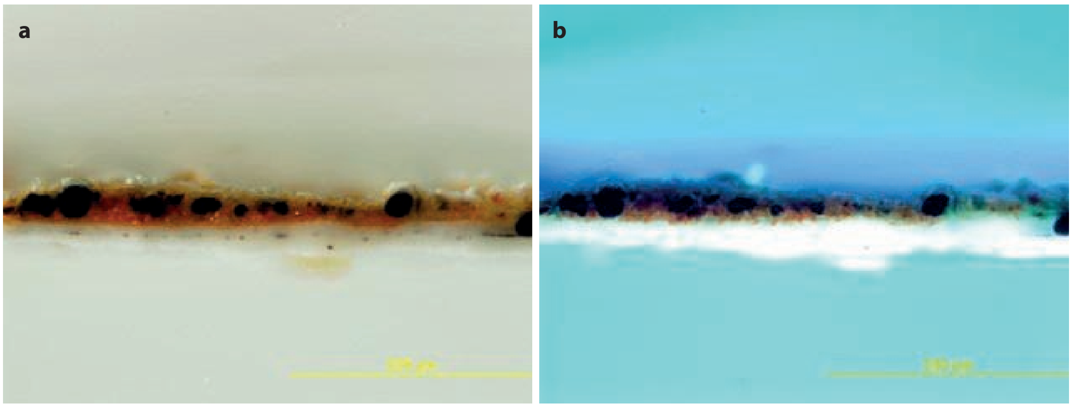


Figure A.III.106 FDM037: PLM photomicrograph of cross-section (a) in normal diffused light and (b) in UV light.

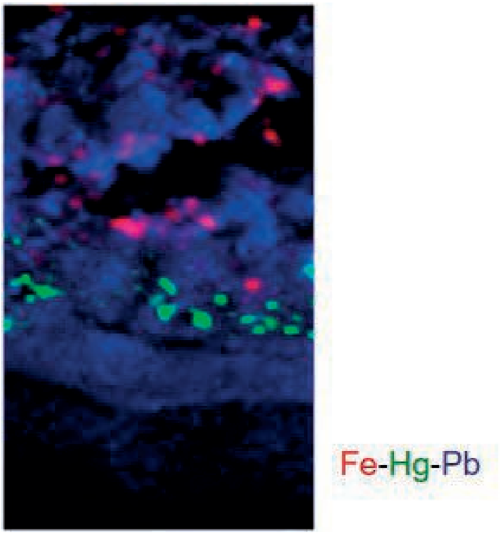


Figure A.III.107 FDM037: µXRF image (PyMCA: ID21).

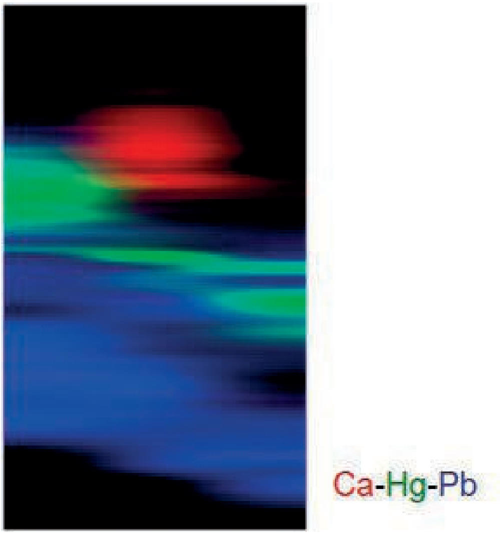


Figure A.III.108 FDM037: µXRF image (PyMCA: ID18).

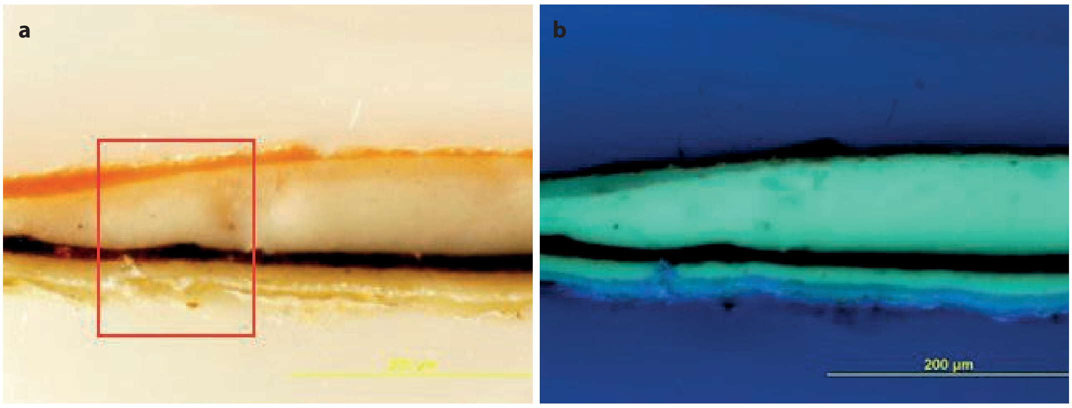


Figure A.III.110 KAK03: PLM photomicrograph of cross-section in (a) normal diffused light and (b) in UV light. The red frame is the imaging area.

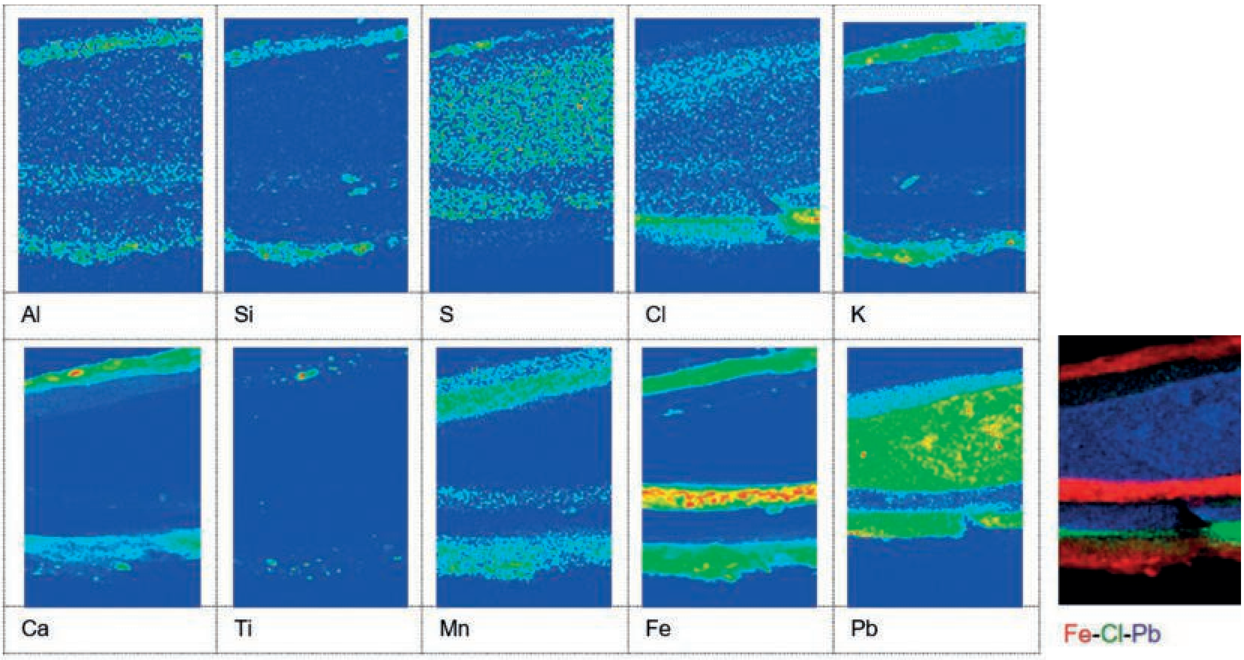


Figure A.III.111 KAK03: µXRF imaging (PyMCA: ID21).

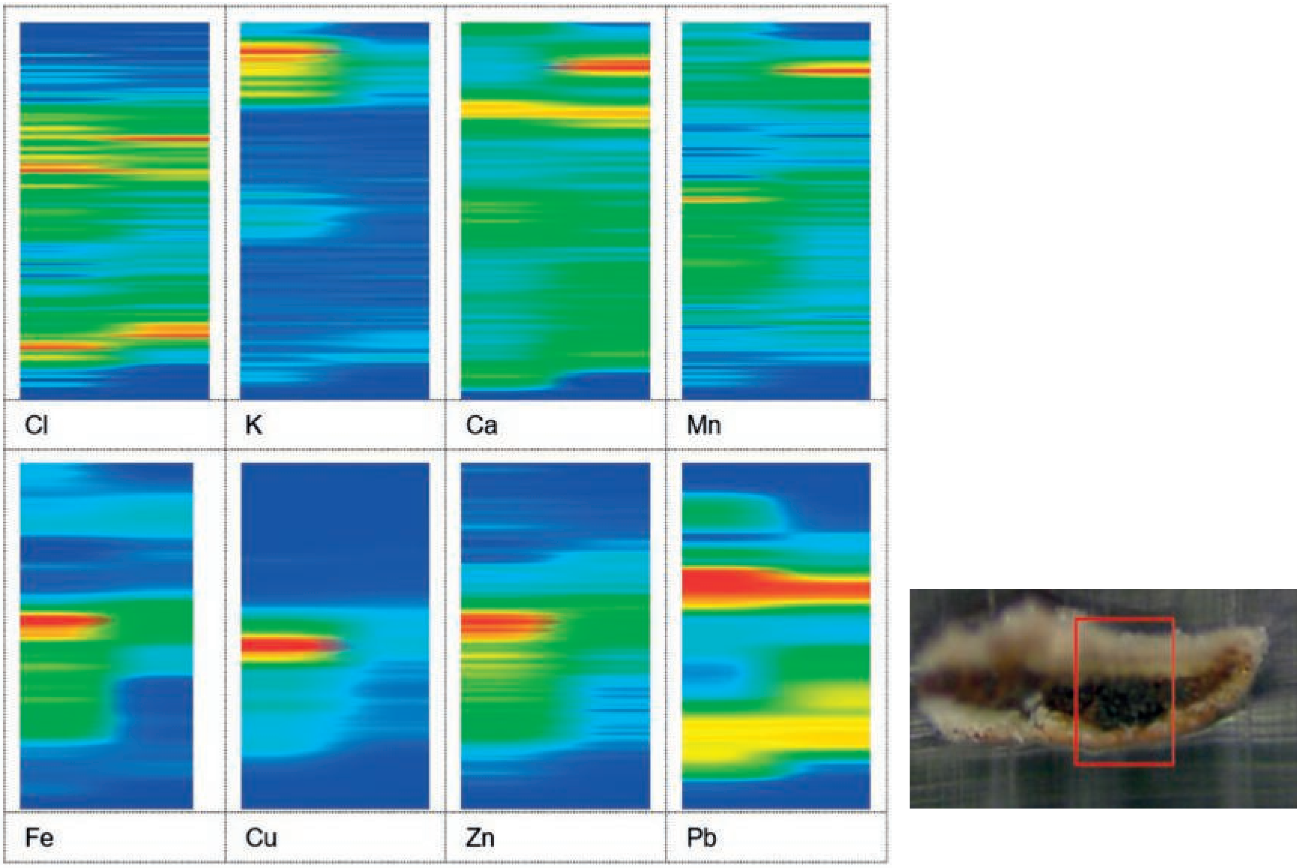


Figure A.III.112 KAK03: µXRF imaging (PyMCA: ID18).

Figure A.III.113 QJM05: stereomicrograph image.

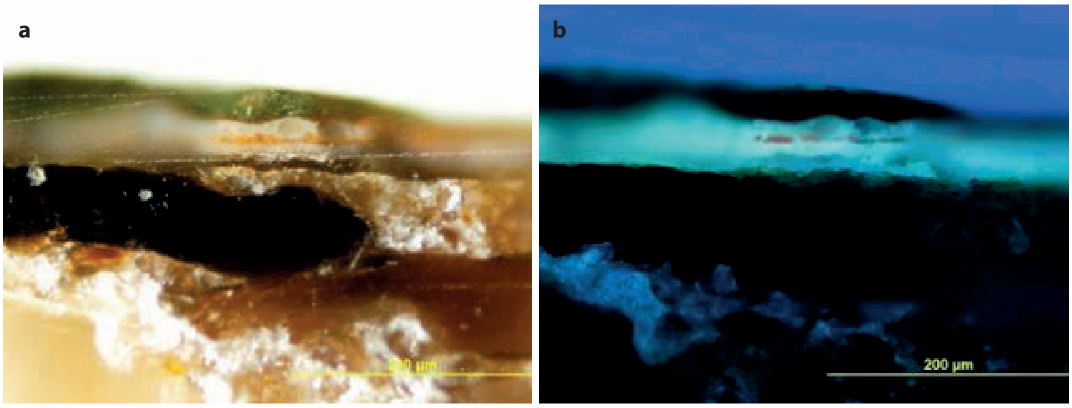


Figure A.III.114 QJM05: PLM photomicrograph of cross-section in (a) normal diffused light and (b) in UV light.

Figure A.III.115 QJM05: µXRF image (PyMCA: ID18).

Figure A.III.116 QJM06: stereomicrograph image.

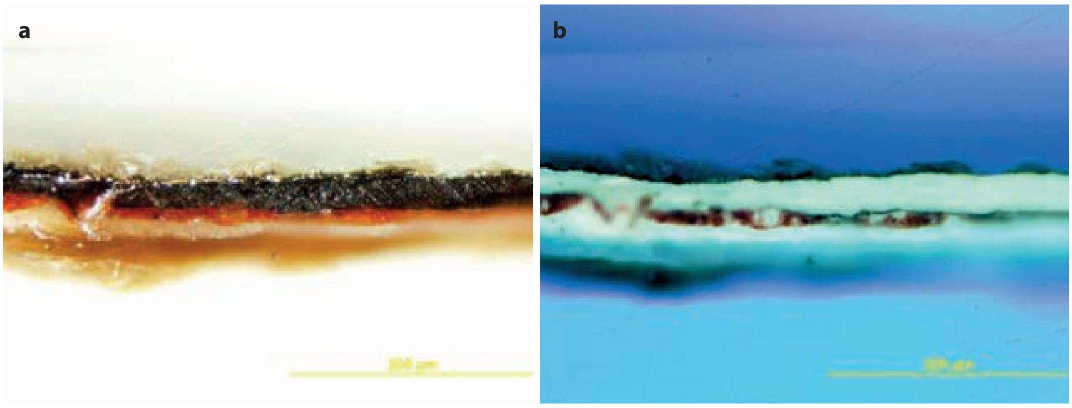


Figure A.III.117 QJM06: PLM photomicrograph of cross-section in (a) normal diffused light and (b) UV light.

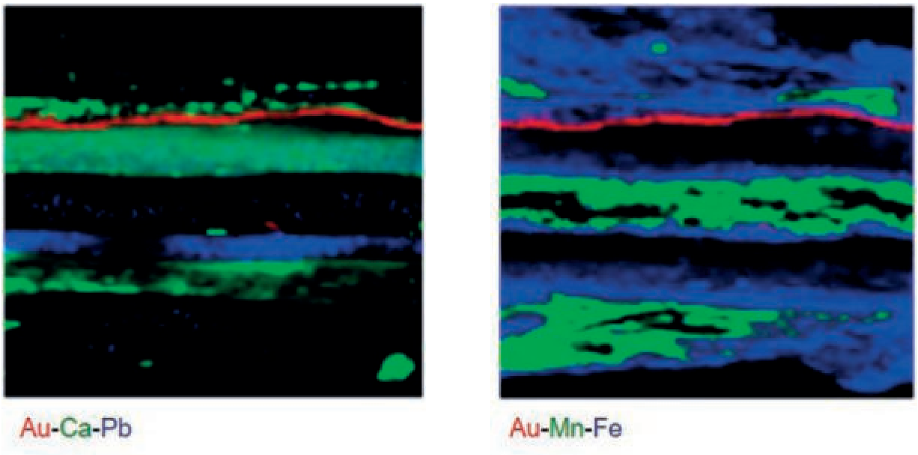


Figure A.III.118 QJM06: µXRF image (PyMCA: ID21).

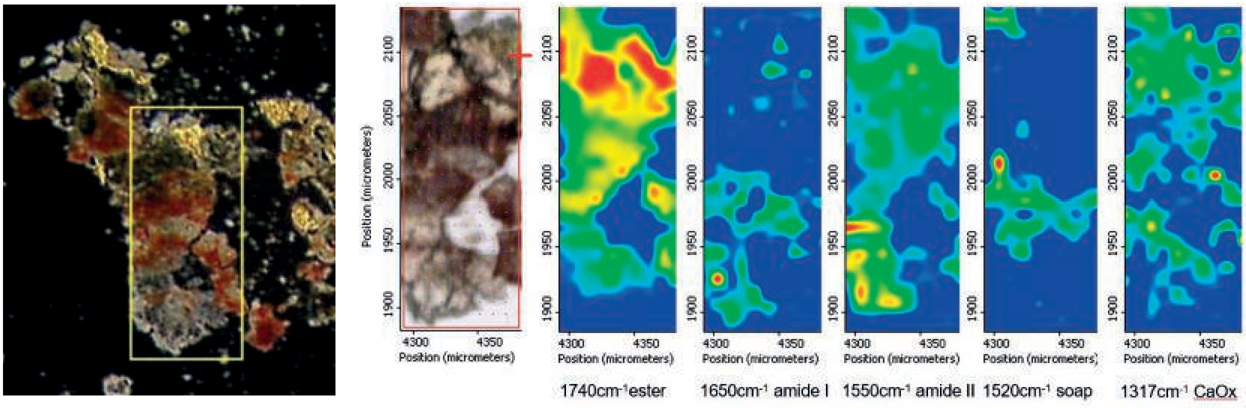


Figure A.III.119 QJM06: ranges of μFTIR spectra for imaging and results (ID21).

Table A.III.1 BMM001: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Glaze	Transparent yellow	Fe, K, Ca	
2	Painting layer	Pale blue	Pb, Fe, Al, S, K, Ca, Na	Lapis lazuli, lead white, gypsum
3	Ground layer	White	Pb, S, Ca	Lead white, gypsum
4	Sizing layer			Organic substance
5	Rendering layer	Brown	Cl/Fe, Al, Si, K, Ca, Mn	Earthen material, salts (Cl)

Table A.III.2 BMM009: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Pale yellow	Fe, K, Mn	
2	Painting layer	Red	Pb, Cl, Mn, Hg, Fe	Cinnabar/ vermilion, minium, cotunnite, laurionite, anglesite
3	Ground layer	White	S, Ca, K	Gypsum

Table A.III.3 BMM033: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layer (alteration)	Black	Fe, Mn	
2		Black	Pb, Cu, Hg	Cinnabar/vermilion, lead-based pigment, copper-based pigment (alteration)
3	Ground layer	White	Ca, S, Sr	Gypsum

Table A.III.4 BMM035: elements detected in each layer and possible pigments and organic components.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Pale yellow	Ca, K	Calcium oxalate
2	Glaze	Transparent yellow	Al, Ca, Fe, K, Mn, Si, Cl	Organic substance (resin)
3	Painting layers	Green	Cu, Cl, Pb, Fe, Si	Copper-based green pigment, lead white (hydrocerussite, cerussite), yellow ochre, drying oil
4		Black	Pb	Lead white, carbon black, resin, drying oil
5	Ground layer	White	Pb, Cl	Lead white (hydrocerussite and susannite), lead soaps
6	Sizing layer	Brown	Fe, K, Mn, Al, Si, Ca	Organic substance (protein), calcium oxalate

Table A.III.5 BMM039: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Whitish grey	Fe, Ca, K, S, Si	
3	Painting layers	Deep blue	Ca, K, Fe, S, Si	Lapis lazuli, gypsum, calcium oxalate, unknown iron-based material
4		Greyish blue	Ca, K, S, Si, Pb	Lapis lazuli, sodalite, lead soap, lead white (hydrocerussite>cerussite), gypsum, calcium oxalate
5	Ground layer	White	Pb, Ca, Si	Lead white (hydrocerussite and susannite), lead soaps
6	Sizing layer	Brown		Organic substances (protein), calcium oxalate

Table A.III.6 BMM040: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	Fe, Ca	
2	Painting layer	Red	Pb, Hg	Minium, cinnabar/ vermilion
3	Mordant?	Black	Fe	
4	Ground layer	White	Pb, Fe, Ca, Ti, K	lead white (hydrocerussite and susannite), lead soaps
5	Sizing layer	Brown	Fe, Ca, Ti, K	

Table A.III.7 BMM045: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layer	Yellow	Fe, Si, K, Mn	Yellow ochre
2	Ground layer	White	Ca, S, K	Gypsum
3	Sizing layer	Brown		

Table A.III.8 BMM055: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration	Black	Fe, K	
2	Painting layers	Orange	Hg	Cinnabar/vermilion
3		Red	Pb, Cl, Co, Ni, Cu	Minium
4	Ground layer	White	Ca, S	Gypsum
5	Sizing layer	Brown		

Table A.III.9 BMM068: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposits	Light brown	Al, Fe, K, Cr, Mg/P	
2		White	Ca, S	Gypsum
3	Painting layers	Green	Pb, S	Lead white + ?
4		Whitish green	Pb, S	Lead white + ?
5		Orange		
6	Ground layer	White	Pb, Cl, K, Ca, S	Lead white, gypsum
7	Sizing layer	Brown	Fe, Ti	Earthen clay

Table A.III.10 BMM081: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration	Black	Si, K, Ca, Mn, Fe/Cl	
2	Painting layers	Red	Hg	Cinnabar/vermilion
3		Orange	Pb>Cl	Minium
4	Ground layer	White	Ca, S, K, Mn	Gypsum
5	Sizing layer	Brown		

Table A.III.11 BMM082: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration	Black	Hg, Ca, K, Si, Al	Cinnabar/vermilion, lapis lazuli
2	Painting layers	Creme white	Pb	Lead white
3		Creme white	Pb	Lead white
4	Ground layer	White	Pb	Lead white
5	Sizing layer	Transparent yellow	Fe, Ca, K, Si, Al	

Table A.III.12 BMM083: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration	Black brown	Cu, K, Fe, S, Ca	Cu-based alteration product
2	Painting layer	White + blue particles	Pb, Si, Ca, Cl, K	Lead white, Ca-contaning particles, lapis lazuli
3	Ground layer	White + blue particles	Pb, Si, Ca, Cl, K	Lead white
4	Sizing layer	Transparent yellow	Fe, Mn, Fe, Ca	

Table A.III.13 BMM091: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown		
2	Painting layers	Green	(Cu, S), (Fe, K, Zn) + spot As	Cu-based green pigment, As-based pigment
3		Bluish green	(Cu, S), (Fe, K, Zn) , Ca, Sr, Cl + spot As	Cu-based pigment, lime white, As-based pigment

Table A.III.14 BMM101: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration/ surface deposit	Dark brown	Ca, Fe, K	
2	Painting layers	Brown	Cu, Zn, Rb, K	Cu/Zn-containing pigment
3		Blue	Cu, Zn, Rb, K	Cu/Zn-containing pigment
4	Ground layer	Light brown	Ca, S	Gypsum

Table A.III.15 BMM111: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Dark brown	Fe, K	
2	Painting layer	Green (blackened)	Cu, K, As	Cu-based green pigment + ?
3	Ground layer	Light brown	Ca, S	Gypsum

Table A.III.16 BMM125: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	White	Ca, S	Gypsum
2	Painting layer	Whitish green	Cl, Sr, Cu	Cu-based green pigment (atacamite)
3	Ground layer	White	K, Ti, Cr, Mn, Fe	?

Table A.III.17 BMM128: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	Fe, Si, Co, Cr, Cu, Ag, As, Cl, Zn	
2	Painting layers	Blue	Ca, Ci, Cu, S	Cu-based blue pigment (azurite?), lapis lazuli? Gypsum
3		Black	S, Cl	
4	Ground layer	White	Ca, S	Gypsum
5	Rendering layer	Brown	Fe, Si, Co, Cr	Earthen clay

Table A.III.18 BMM145: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Grey	Fe, Cr, Mn, K, Si	
2	Painting layer	Pale red	Pb, Ag, Co, Cu, Ni, Mn, Ti, Zn, Hg, Si	Minium, cinnabar/vermilion
3	Ground layer	Light brown	Ca	Gypsum

Table A.III.19 BMM181: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	K, Ca	
2	Painting layers	Red	Hg, S, spot Fe, Cr	Cinnabar/vermilion
3		Orange	Pb	Minium
4	Ground layer	White	Pb	Lead white
5	Sizing layer	Light brown	Ca, Fe	

Table A.III.20 BMM177: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Glaze	Transparent bluish yellow	Cu, K, Ca, Cl	Copper chloride/ copper oxalate/ calcium oxalate
2	Painting layer	Silver	Sn, Pb	Tin leaf (with lead)
3	Mordant	Black		
4	Ground layer	White	Pb, Ca	Lead white (hydrocerussite and susannite), lazurite
5	Sizing layer	Brown	Ca	Quartz, calcite

Table A.III.21 BMM178: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*		*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layer	Silver		Tin leaf (with lead)
2	Mordant	Black		Drying oil
3	Ground layer	White		Lead white, lead soap, drying oil
4	Organic layer	Brown		Drying oil
5	Sizing layer	Transparent		Protein

Table A.III.22 BMM210: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	Cu, Fe, Zn + spot As, Pb	Cu-based green pigment
2	Painting layers	Green	Cu, Fe, Zn + spot As, Pb	Cu-based green pigment
3		Bluish green	Ca, Sr > Cu	Cu-based green pigment + Ca-containing pigment
4	Sizing layer	Light brown		

Table A.III.23 FDM059: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layers	Whitish green	Ca, Fe	?
2		Orange	Ca, Cl, Cu, Hg, K, Mn, Ni, Pb, Zn	Minium, cinnabar/vermilion
3		Black particles	Ca, Cl, Cu, Hg, K, Mn, Ni, Pb, Zn	Cu-based pigment
4	Ground layer	White	Pb	Lead white
5	Sizing layer	Light brown	Si	
6	Rendering layer	Brown	Fe, K, Ca, Al, Si, Cr, Ca, S, Si	Earthen clay, gypsum

Table A.III.24 FDM011: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layers	Green/ brown	Cu, As	Cu-based green pigment, orpiment
2		White + yellow particles	Pb, (As)	Lead white, (orpiment)
3	Ground layer	White	Pb, Cl, Cu	Lead white
4		White + blue particles	Pb, Cl, Cu	Lead white
5	Sizing layer	Light brown	Fe, Ca	

Table A.III.25 FDM053: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	K	
2	Painting layer	Green	Cu, Cl, Pb, Zn, Co	Atacamite
3	Ground layer	White	Ca, Fe, K	
4	Sizing layer	Light brown		

Table A.III.26 FDM033: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	S, Ca, K, Ti	Gypsum
2	Painting layers	Red/ white	Hg, S	Cinnabar/vermilion
3		Black	Ca, Pb	Minium (blackened?)
4	Ground layer	White	Pb, Cl, Cu, Ni, Ag	Lead white
5	Sizing layer	Light brown	Ca	

Table A.III.27 FDM055: elements detected in each layer and possible pigments, organic components and binding media.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Alteration	Whitish green	Cu, Cl, Ag, (K)	Copper oxalate (moolooite)
2	Painting layers	Green		Atacamite, copper/ lead carboxylates, drying oil
3		White	Pb, Cl, Ca, Mn, As	Lead white (hydrocerussite>cerussite), orpiment, lead carboxylate, drying oil
4		Red	Pb, Ca, Fe, Co, As	Minium, red ochre (goethite), lead carboxylate, drying oil
5	Ground layer	White		Lead white (hydrocerussite>cerussite), lead carboxylate
6	Sizing layers	Transparent yellow		Protein, polysaccharide
7		Transparent pale yellow		Resin
8	Rendering layer	Brown	K, Mn, Ti, Fe	Earthen clay, quartz

Table A.III.28 FDM037: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Painting layers	Orange + deep red particles	Ca, Pb, Fe	Minium, red ochre
2		Orange	Pb, Hg	Minium, cinnabar/vermilion
3	Ground layer	White	Pb, Ti, Mn, V, Fe	Lead white
4	Sizing layer	Light brown		

Table A.III.29 KAK03: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Grey	Al, Si, S, K, Ca, Fe, Mn, Ti	
2	Glaze	Transparent yellow		
3	Painting layer	White	Pb, S	Lead white
4	Mordant?	Black	Fe, Mn, Al, Cu	
5	Ground layer	White	Pb, S	Lead white
6	Sizing layer	Brown	Fe, Mn, K, Ca, Ti, SI, Al	

Table A.III.30 QJM05: elements detected in each layer and possible pigments.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	Fe, Ca, spot As	
2	Painting layer	Green	Cu, Ag	Copper-based green pigment
3	Ground layer	White + yellow	Ca, Pb	Lead white
4	Sizing layer	Light brown		

Table A.III.31 QJM06: elements detected in each layer and possible pigments and binding media.

Layer number	*Layer*	*Colour*	*Detected major elements*	*Estimated pigments/colorants, alteration products, inclusions*
1	Surface deposit	Light brown	Fe, Ca	Calcium oxalate
2	Gold leaf	Gold	Au	Gold
3	Mordant	Dark brown	Ca/Mn	Drying oil, lead soap
4	Bole	Red	Mn, Pb, Fe	Red ochre, minium
5	Ground layer	White	Pb	Lead white
6	Sizing layer	Light brown	Fe, Ca/Mn	Protein

Table A.III.32 Results of fatty acids detected by GC-MS using the methprep II method at the GCI.

Sample number	Cave	Description	Sample Weight ug	Final Volume ul	Pimelic acid	Suberic acid	Lauric acid	Azelaic acid	Sebacic acid	Myristic acid	Palmitic acid	Stearic acid	Eicosanoic acid	Oleic acid	P/S	A/P	identified
BMM 063	B(d)	red, white ground	76	15	6.97	25.7	0.0	123.2	10.7	1.2	52.4	17.0	0.0	0.0	3.1	2.4	drying oil
BMM 091	C(a)	unusual light blue	20	20	0	0.0	0.0	0.0	0.0	0.0	0.3	0.4	0.0	0.0	0.8	0.0	ND
BMM 209	C(b)	green, white ground	219	20	0	0.0	0.0	0.0	0.0	0.0	6.9	5.3	0.0	2.3	1.3	0.0	ND
BMM 099	D	red, white ground	41	20	0	0.0	0.0	0.0	0.0	0.4	3.4	4.5	0.0	0.5	0.8	0.0	ND
BMM 067	East III	yellow, white ground	198	20	0	0.0	0.0	0.6	0.0	0.0	4.3	3.7	0.0	2.7	1.1	0.1	ND
BMM 108	E(c)	ground and soot deposit	191	20	0.42	0.5	0.0	0.7	0.2	0.5	2.8	1.8	0.1	0.4	1.5	0.2	ND
BMM 101	E(e)	darkened surface, later soot?	248	15	2.23	2.4	0.0	4.9	1.3	2.9	15.0	9.3	1.0	0.0	1.6	0.3	oil
BMM 083-1	F(c)	translucent yellow glaze and render	556	15	0	1.0	0.0	4.4	0.3	2.8	6.8	2.0	0.0	1.3	3.4	0.6	ND
BMM 083-2	F(c)	black darkened layer, yellow glaze, white ground	81	15	4.96	17.1	0.0	74.7	6.0	1.6	30.5	11.2	0.4	0.0	2.7	2.4	drying oil
BMM 045	G	yellow, white ground	84	20	0	0.0	0.0	0.1	0.0	0.0	1.0	0.6	0.0	0.0	1.6	0.1	ND
BMM 212	H(a)	yellow, white ground	309	20	0.18	0.1	0.0	0.3	0.0	0.2	1.4	0.9	0.0	0.0	1.6	0.2	ND
BMM 211-2	H(b)	red and white ground	416	20	0	0.0	0.0	0.5	0.0	0.0	2.2	1.2	0.0	0.0	1.9	0.2	ND
BMM 009	I	red, white ground	53	20	0	0.1	0.0	0.2	0.0	0.0	0.5	0.3	0.0	0.0	1.6	0.4	ND
BMM 073	I	blue, black, white ground	408	20	0	0.3	0.0	0.5	0.0	0.9	6.2	3.8	0.1	0.3	1.6	0.1	ND
BMM 128	J(b)	blue/black, white ground	218	20	0.34	0.5	0.0	1.2	0.2	1.0	4.6	3.2	0.0	0.0	1.5	0.3	ND
BMM 112	J(c)	green on white ground	117	20	2.74	3.9	0.9	8.8	1.6	4.8	20.9	15.6	0.4	0.2	1.3	0.4	oil (unknown)
BMM 113	J(c)	green on white ground	276	20	1.73	3.1	1.5	8.5	1.1	6.9	29.0	19.4	0.6	0.6	1.5	0.3	oil (unknown)
BMM 120	J(d)	blue, pink ground	183	20	0	0.3	0.0	0.6	0.0	0.5	4.1	3.3	0.0	0.3	1.2	0.1	ND
BMM 169	J(f)	red, render	131	20	0	0.0	0.0	0.2	0.0	0.0	1.5	0.9	0.0	0.0	1.7	0.1	ND
BMM 080	K	flesh colour, white ground	1154	20	0.81	0.3	0.5	0.6	0.2	2.7	14.0	7.5	0.3	0.7	1.9	0.0	ND
BMM 134	K	brown deposit, red, white ground	451	20	1.12	1.6	0.3	2.6	0.3	0.8	5.4	3.7	2.1	0.3	1.5	0.5	ND
BMM 135	K	brown deposit on chaff	156	20	0.34	77.0	0.0	1.7	0.2	0.7	16.5	41.4	1.5	0.3	0.4	0.1	oil
BMM 203	L	green, white ground, yellow size	37	20	1.94	5.6	0.0	25.1	1.8	0.5	11.3	3.9	0.1	0.2	2.9	2.2	drying oil
BMM 204-2	L	black deposit	966	20	0	0.9	0.0	4.0	0.6	4.6	23.6	13.1	3.4	1.8	1.8	0.2	oil
BMM 053	M	darkened surface, later soot?	146	15	0	1.7	0.0	4.2	0.7	0.9	7.8	5.3	0.3	0.0	1.5	0.5	oil
BMM 054	M	red, white ground	223	20	5.22	10.7	0.7	32.3	4.7	4.0	25.2	13.3	1.0	0.1	1.9	1.3	drying oil
BMM 035	N(a)	resin, green, black oils, white ground, yellow size	54	15	6.32	20.3	0.0	91.0	5.3	1.9	51.4	16.7	0.5	8.9	3.1	1.8	dyring oil
BMM 040	N(a)	red/oily red, white ground	104	15	6.52	23.6	0.0	123.2	8.8	1.7	51.8	15.5	0.4	1.5	3.3	2.4	drying oil
BMM 183	N(a)	deep red glaze on orange	17	15	0	1.4	0.0	4.5	0.0	0.2	2.0	0.7	0.0	0.0	2.7	2.2	drying oil
BMM 184-1	N(a)	yellow resin on tin leaf	20	15	0	0.9	1.0	2.2	0.3	0.0	0.6	0.4	0.0	0.0	1.5	3.4	ND
BMM 184-2	N(a)	tin leaf and white ground	20	15	0.24	0.7	0.0	3.0	0.3	0.0	1.9	1.0	0.0	0.0	2.0	1.6	ND
BMM 163	S(a)	grey/green, white ground, size	1438	20	26.07	90.9	2.3	349.2	31.2	10.7	230.4	93.2	2.8	0.0	2.5	1.5	drying oil
BMM 157-2	East Displaced	blue and render	953	20	0	0.0	0.0	1.3	0.0	0.0	2.4	1.2	0.0	0.0	1.9	0.6	ND
BMM 189	EGB	blue in white, white ground	2412	20	0.38	0.2	0.0	0.5	0.1	0.7	4.3	2.5	0.2	1.6	1.7	0.1	ND
BMM 190	EGB	red, white ground, render	1162	20	0	0.0	0.0	0.2	0.0	0.0	1.7	1.2	0.0	0.9	1.4	0.1	ND

BMM 191	EGB	blue in white, white ground	155	20	0.05	0.3	0.0	0.5	0.0	1.0	7.0	4.1	0.2	0.0	1.7	0.1	ND
BMM 199	WGB	red	841	20	0	0.2	0.0	0.5	0.0	0.0	2.8	3.8	0.0	0.0	0.7	0.2	ND
BMM 201	WGB	deep red	401	20	0	0.0	0.0	0.7	0.0	0.0	3.3	4.4	0.0	0.0	0.7	0.2	ND
FDM 059	Foladi 2	red and white ground	26	15	1.42	5.0	0.0	21.0	1.6	0.0	8.1	2.6	0.0	0.0	3.1	2.6	drying oil
FDM 043-1	Foladi 3	yellow translucent size	14	20	0.31	1.1	0.0	3.8	0.2	0.2	2.1	1.1	0.0	0.0	1.9	1.8	drying oil
FDM 043-2	Foladi 3	white, white ground, size	263	20	13.53	40.6	0.5	161.6	12.2	1.2	47.8	17.4	0.4	23.0	2.7	3.4	drying oil
FDM 014	Foladi 4	salmon pink, yellow, white ground, size	330	20	10.63	32.0	0.0	153.0	10.0	0.6	30.2	13.1	0.4	0.2	2.3	5.1	drying oil
FDM 055-1	Foladi 4	green, white ground, size, sandy render	1096	20	21.83	86.7	2.3	399.4	23.6	7.8	217.6	76.7	1.9	18.9	2.8	1.8	drying oil
FDM 055-2	Foladi 4	reddish sandy upper render	3533	20	0	0.0	0.4	0.3	0.0	0.7	2.8	1.1	0.0	0.4	2.5	0.1	ND
FDM 023	Foladi 4B	white, orange, white ground, size	330	20	24.51	66.7	3.2	244.8	20.0	10.9	153.5	56.7	1.4	0.6	2.7	1.6	drying oil
FDM 003	Foladi 5	red, white ground, size	147	20	0.07	0.3	0.0	0.7	0.1	0.3	2.5	1.5	0.0	0.0	1.6	0.3	ND
FDM 026	Foladi 6	black (green), white ground, white size	80	20	2.2	6.2	0.0	22.1	1.8	0.0	6.0	2.3	0.1	0.0	2.6	3.7	drying oil
FDM 060	Foladi 6	brown green, red, white ground, size	406	20	10.3	26.4	0.0	100.9	7.2	1.0	26.4	14.2	0.7	0.0	1.9	3.8	drying oil
KAK 03	Kakrak 43	red, black organic, white ground	54	15	5.17	16.9	0.0	70.1	5.5	1.6	40.9	12.8	0.0	0.0	3.2	1.7	drying oil
KAK 10	Kakrak 44	white, blue, white ground	427	20	19.06	75.2	0.0	348.9	25.4	5.3	223.3	81.2	2.0	6.1	2.7	1.6	drying oil
QJM 06	Qol-e Jalal	gold leaf and mordant	20	20	5.03	15.7	0.0	73.4	5.5	1.0	18.4	6.5	0.0	0.0	2.8	4.0	drying oil
QJM 07	Qol-e Jalal	brownish colour, whole paint layer	176	15	3.76	13.3	0.0	62.5	5.4	0.0	21.3	8.3	0.0	0.0	2.6	2.9	drying oil
Reference Oils																	
Plane tree gum (modern ref, Bamiyan)						0.38	0.9	0.0	2.0	0.2	0.4	3.8	1.5	1.7	1.3	2.5	
Almond oil, Fowler (film)						0.53	3.5	0.0	15.9	0.7	0.0	4.6	4.2	0.4	1.5	1.1	
Linseed oil refined, Winsor & Newton						4.93	35.5	0.0	365.2	14.5	0.6	165.0	134.7	3.4	25.1	1.2	
Poppy seed oil, Grumbacher						7.02	70.2	0.0	655.7	20.9	0.6	365.7	116.6	4.4	18.7	3.1	
Poppy seed oil cold pressed, Grumbacher						6.77	72.2	0.0	591.3	19.6	0.8	414.0	117.8	4.1	107.0	3.5	
Poppy seed oil sunbleached						5.05	46.2	0.0	414.7	12.4	0.4	328.6	74.1	3.1	5.2	4.4	
Sesame oil, Arrowhead Mills						1.11	7.5	0.0	56.5	1.9	0.3	105.4	89.5	6.6	227.9	1.2	
Soy oil, Spectrum Naturals						3.44	26.0	0.0	201.2	9.7	0.6	252.8	116.3	7.1	21.7	2.2	
Sunflower oil cold pressed, Schminke						0.4	2.9	0.0	21.2	1.1	0.0	6.3	12.2	0.7	2.4	0.5	
Tung oil, China						0.22	2.7	0.0	31.8	0.4	0.8	102.0	109.9	6.1	325.0	0.9	
Walnut oil, Spectrum Naturals						5.37	40.2	0.0	363.5	11.6	0.4	117.5	37.3	1.6	2.5	3.1	
Walnut oil, Rougie						4.54	35.4	0.0	339.9	12.4	0.5	324.5	142.3	4.1	12.5	2.3	
Safflower						4.81	38.0	0.0	286.2	12.0	0.8	147.8	112.6	12.5	16.3	1.3	

* All the oil references except for the plane tree gum are from M. Schilling 1989

Table A.III.33 Results of amino acids detected by GC-MS at the GCI.

Sample Number	Cave	Description	Sample Weight ug	Final Volume ul	Oil %	Protein %	Alanine	Valine	Isoleucine	Leucine	Glycine	Proline	Hydoroxyproline	Candidates/ correlation coefficient	results
BMM 063	B(d)	red, white ground	76	30	13.2	5.3	20.7	3.8	2.8	7.4	36.1	15.0	14.3	glue, 0.97	glue
BMM 091	C(a)	unusual light blue	20	30	1.4	0.0									
BMM 099	D	red, white ground	41	30	1.7	1.4	10.7	8.7	5.7	15.6	27.7	14.9	16.7	no match	
BMM 067	E III	yellow, white ground		30	0.6	0.2	13.0	10.8	8.0	15.7	28.6	17.5	6.5		
BMM 083-2	F(c)	black darkened layer, yellow glaze, white ground	81	30	8.3	1.0	16.9	13.8	8.3	15.8	19.4	11.8	13.2	no match	
BMM 045	G	yellow, white ground	84	30	1.2	1.0	13.8	20.7	9.3	19.4	8.0	25.7	3.0	casein, 0.95	casein
BMM 212	H(a)	yellow, white ground	309	30	0.2	0.1	15.8	10.2	8.2	17.0	33.3	12.2	3.3	no match	
BMM 211-2	H(b)	red and white ground	416	30	0.2	0.4	18.2	16.0	10.6	18.8	19.6	15.9	1.0	egg, 0.91	egg
BMM 009	I	red, white ground	53	30	1.9	0.0									
BMM 128	J(b)	blue/black, white ground	218	30	0.8	2.2	20.1	13.7	7.5	17.7	25.8	13.3	1.7	no match	
BMM 120	J(d)	blue, pink ground	183	30	0.6	0.7	19.0	14.1	8.7	20.1	17.0	19.9	1.2	no match	
BMM 169	J(f)	red, render	131	30	1.0	1.1	11.3	18.1	8.0	26.8	9.3	23.7	2.9	casein 0.92	casein
BMM 134	K	brown deposit, red, white ground	451	30	0.3	1.3	26.7	8.2	6.0	11.4	35.4	10.9	0.0	no match	
BMM 135	K	brown deposit on chaff	156	30	2.3	4.8	28.8	10.8	7.5	14.6	30.8	7.5	0.0	no match	
BMM 203	L	green, white ground, yellow size	37	30	24.8	3.5	15.2	10.2	7.1	16.2	32.1	8.4	10.6	no match	
BMM 053	M	darkened surface, later soot?	146	30	1.0	0.7	16.7	23.4	11.5	22.4	12.1	12.0	1.9	egg, 0.92	egg
BMM 054	M	red, white ground	223	30	2.4	2.0	19.5	16.6	11.7	23.4	19.8	8.9	0.0	egg, 0.96	egg
BMM 035	N(a)	resin, green, black oils, white ground, yellow size	54	30	8.4	1.3	19.3	16.3	9.9	17.6	20.1	16.8	0.0	no match	
BMM 183	N(a)	deep red glaze on orange	17	30	6.5	0.0									
BMM 191	EGB	blue in white, white ground	155	30	1.3	3.2	14.0	14.8	10.9	24.2	19.0	17.0	0.0	no match	
FDM 059	Foladi 2	red and white ground	26	30	9.6	0.0									
FDM 043-1	Foladi 3	yellow translucent size	14	30	18.0	14.7	15.2	10.2	8.1	16.8	23.7	15.3	10.7	no match	
FDM 043-2	Foladi 3	white, white ground, size	263	30	6.1	2.6	19.3	10.2	8.3	15.4	25.0	12.0	9.8	no match	
FDM 014	Foladi 4	salmon pink, yellow, white ground, size	330	30	8.9	1.9	20.0	8.5	6.8	13.8	33.9	8.2	8.8	glue, 0.91	glue
FDM 003	Foladi 5	red, white ground, size	147	30	1.7	1.6	21.9	13.5	10.6	20.4	20.6	11.0	2.0	egg, 0.96	egg
FDM 060	Foladi 6	brown green, red, white ground, size	406	30	1.5	0.4	21.8	4.4	2.3	5.5	43.0	12.0	11.0	glue, 0.99	glue
KAK 03	Kakrak 43	red, black organic, white ground	54	30	9.7	0.3	19.2	5.4	4.4	8.3	41.3	8.9	12.6	glue, 0.98	glue
KAK 10	Kakrak 44	white, blue, white ground	427	30	6.4	2.2	25.1	14.8	10.2	18.4	19.8	11.7	0.0	egg, 0.99	egg
QJM 07	Qol-e Jalal	brownish colour, whole paint layer	176	30	6.6	1.3	16.4	6.8	5.6	13.0	26.3	13.3	18.6	no match	
Reference Proteins															
isinglass (Zecchi)							18.1	3.4	2.2	4.1	46.9	15.0	10.3		
collagen & gelatine (mean)							15.7	3.0	1.8	3.7	46.7	16.7	12.4		
egg white (mean)							22.0	18.3	12.8	21.6	14.8	10.5	0.0		
whole egg (mean)							22.0	17.4	13.2	22.0	14.5	10.9	0.0		
egg yolk (mean)							20.9	16.6	13.3	23.2	14.1	11.9	0.0		
casein (mean)							10.9	16.9	12.8	22.0	8.6	28.8	0.0		

Table A.III.34 Results of polysaccharides detected by GC-MS at the GCI.

Sample Number	Cave	Description	Weight (ug)	Volume (ml)	Rrhamnose	Ffructose	Arabinose	Xylose	Mannose	Fructose	Glucose	Galactose	Identification	%Gum
BMM063	B(d)	red, white ground	5717	0.05	0.3	0.3	5.7	1.0	0.0	3.5	15.9	1.5	Gum	0.02
BMM094	C(a)	pink ground	168	0.05	0.0	0.0	0.0	0.0	0.0	0.0	3.3	0.0	Sugar	0.10
BMM209	C(b)	green, white ground	360	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM098	D	blue, white ground, render	235	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM107	E(c)	black deposit, red, (all paint layer)	90	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM109	E(c)	brown paint	1041	0.03	0.0	0.0	0.0	1.0	0.0	0.0	3.5	0.0	Sugar	0.01
BMM101	E(e)	darkened surface, later soot?	743	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM157_1	East Displaced	blue and render	1953	0.05	0.0	0.0	0.0	0.0	0.0	0.0	2.7	0.0	Sugar	0.01
BMM158	East Displaced	red no ground flakes, nimbus	80	0.05	0.0	0.0	0.0	0.0	0.0	0.0	3.0	1.4	Sugar	0.28
BMM083_1	F(c)	translucent yellow glaze and render	19	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM082	F(c)	black red yellow, resinous material	817	0.03	0.0	1.4	36.1	4.6	2.8	0.0	12.9	12.3	Gum	0.26
BMM045	G	yellow, white ground	54	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM212	H(a)	yellow, white ground	464	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM071	I	white on black	251	0.03	0	0	0	0	0	0	0	0	ND	0.00
BMM128	J(b)	blue/black, white ground	66	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM111	J(c)	black (blackened surface), white ground, render	152	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM113	J(c)	black (green) + orange yellow	157	0.03	6.5	2.5	33.9	54.0	2.9	0.0	25.2	31.1	Gum	2.98
BMM120	J(d)	blue, pink ground	808	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM169	J(f)	red, render	3547	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM080	K3	flesh colour, white ground	19	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM134	K3	brown deposit, red, white ground	138	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM203	L	green, white ground, yellow size	20	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM204_1	L	black deposit	492	0.05	0.0	0.0	1.9	1.4	0.0	0.0	1.9	0.0	Sugar	0.05
BMM054	M	red, white ground	91	0.05	0.0	0.0	0.6	3.0	1.4	0.0	16.0	23.0	carbohydrates (galactose)	2.42
BMM053	M	darkened green	201	0.03	0.0	0.0	0.0	0.0	0.0	0.0	3.3	1.2	Sugar	0.07
BMM035	N(a)	resin, green, black oils, white ground, yellow size	160	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM040	N(a)	red/oily red, white ground	265	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM184	N(a)	red with black	487	0.03	0.0	0.0	17.9	2.1	1.7	0.0	5.7	7.4	Gum	0.21
BMM184_3	N(a)	yellow translucent size	208	0.05	0.0	0.0	0.0	3.5	0.0	0.0	1.0	0.0	Sugar	0.11
BMM181	N(a)	dark red	247	0.03	0	0	0	0	0	0	0	0	ND	0.00
BMM183	N(a)	reds	285	0.05	0.0	0.7	10.3	1.3	1.8	0.0	8.8	14.3	Gum	0.65
BMM162	S(a)	red, whole paint layer	213	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM163	S(a)	grey/green, white ground, size	1098	0.03	0.0	0.0	3.3	1.3	0.0	0.0	3.9	1.9	Gum	0.03
FDM059	Foladi 2	red and white ground	34	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
FDM055 Lower	Foladi 4	lower rendering	13969	0.05	2.2	0.0	1.8	0.0	1.4	0.7	1.1	0.9	ND	0.00
FDM055 Upper	Foladi 4	upper rendering	14538	0.05	0.0	0.0	0.0	0.0	0.0	0.0	1.4	0.0	ND	0.00
FDM051	Foladi 4	red on white ground	659	0.03	0.0	1.2	41.3	2.3	2.8	0.0	42.9	19.5	Gum	0.50
FDM054	Foladi 4	green on white ground	806	0.05	0.5	2.1	41.7	4.4	6.6	0.0	55.0	73.0	Gum	1.14
FDM023	Foladi 4B	white, orange, white ground, size	139	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
FDM026	Foladi 6	blackened green, white ground, size	63	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
FDM060	Foladi 6	brown green, red, white ground, size	207	0.05	1.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
KAK01	Kakrak 43	red, render	1212	0.05	0	0	0	0	0	0	0	0	ND	0.00
KAK05	Kakrak 43	yellow on white ground	506	0.03	0.0	0.0	3.7	2.8	0.0	0.0	9.1	4.3	Gum	0.12
KAK10	Kakrak 44	white and ground	119	0.05	0	0	0	0	0	0	0	0	ND	0.00
QJM07	Qol-e Jalal	brownish colour, whole paint layer	34	0.05	0	0	0	0	0	0	0	0	ND	0.00
BMM191_1	EGB	blue particles in white, white ground	136	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM191_2	EGB	black	435	0.03	0.0	0.0	0.0	0.0	0.0	0.0	1.9	1.2	Sugar	0.02
BMM189	EGB	yellow on white ground	222	0.03	0.0	0.0	1.4	4.7	0.0	0.0	0.0	0.0	Sugar	0.08
BMM190	EGB	red on render	637	0.03	0	0	0	0	0	0	0	0	ND	0.00
BMM191	EGB	blue particles in white, white ground	6429	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM199	WGB	red	159	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00
BMM201	WGB	deep red	377	0.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	ND	0.00

Table A.III.35a Results of ELISA at the GCI.

Sample Number	Cave	Description	PLATE 1								PLATE 2								Identified
			Collagen #ab19811 (Goat)		Collagen #ab6577 (Rabbit)		Fish collagen		Casein		Egg white		Egg Yolk		Plant gum		Tragacanth		
			A	B	C	D	E	F	G	H	A	B	C	D	E	F	G	H	
BMM 059	A	green brown? slightly yellow	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 063	B(d)	red, white ground	0.194	0.166	0.097	0.102	0.201	0.129	failed	failed	0.019	0.186	0.257	0.204	0.092	0.089	0.088	0.087	
BMM 091	C(a)	unusual light blue	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 209	C(b)	green, white ground	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 101	E(e)	darkened surface, later soot?	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 083-1	F(c)	translucent yellow glaze and render	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 045	G	yellow, white ground	0.142	0.136	0.086	0.074	0.091	0.088	failed	failed	0.137	0.121	0.075	0.072	0.074	0.069	0.078	0.077	
BMM 212	H(a)	yellow, white ground	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 009	I	red, white ground	0.137	0.146	0.080	0.081	0.089	0.100	failed	failed	0.127	0.120	0.074	0.074	0.070	0.071	0.076	0.073	
BMM 053	M	darkened surface, later soot?	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 053-2	M	darkened surface, later soot?	0.139	0.125	0.084	0.089	0.096	0.099	failed	failed	0.184	0.170	0.082	0.075	0.077	0.075	0.079	0.087	
BMM 035	N(a)	resin, green, black oils, white ground, yellow size	0.134	0.124	0.085	0.076	0.117	0.091	failed	failed	0.141	0.158	0.071	0.075	0.071	0.075	0.081	0.078	
BMM 040	N(a)	red/oily red, white ground	0.145	0.155	0.102	0.098	0.096	0.090	failed	failed	0.753	1.037	0.117	0.560	0.084	0.072	0.075	0.078	egg white
BMM 157-2	East Displaced	blue and render	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
BMM 201	WGB	deep red	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	
FDM043-1	Foladi 3	yellow translucent size	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	0.140	0.140	0.078	0.088	0.099	0.073	0.081	0.113	
FDM 055-1	Foladi 4	green, white ground, size, sandy render	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	0.134	0.131	0.101	0.126	0.085	0.082	0.082	0.083	
FDM 055-2	Foladi 4	reddish sandy upper render	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	0.147	0.140	0.203	0.291	0.074	0.129	0.101	0.096	
FDM 003	Foladi 5	red, white ground, size	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	0.122	0.126	0.083	0.113	0.080	0.069	0.091	0.083	
FDM 060	Foladi 6	brown green, red, white ground, size	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	0.174	0.218	0.117	0.137	0.086	0.080	0.094	0.099	
KAK 01	Kakrak 43	ochre red	0.174	0.175	0.090	0.081	0.205	0.131	failed	failed	0.159	0.158	0.212	0.204	0.079	0.084	0.081	0.084	
KAK 03	Kakrak 43	red, black organic, white ground	0.142	0.123	0.086	0.081	0.099	0.090	failed	failed	0.136	0.113	0.072	0.070	0.072	0.072	0.072	0.077	
QJM 07	Qol-e Jalal	brownish colour, whole paint layer	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	n.d	

Table A.III.35b Results of ELISA (BMM082: Cave F(c)) at the GCI.

sample	cave	Egg White			Animal Glue (anti-collagen#ab19811)			Animal Glue (anti-collagen#ab6577)			Casein			Identified
		40µL	40µL	average+3SD	40µL	40µL	average+3SD	60µL	60µL	average+3SD	60µL	60µL	average+3SD	
BMM082	F(c)	0.220	0.205		**0.509**	**0.516**		**0.973**	**0.970**		**1.373**	**1.351**		Casein, animal glue

Table A.III.36 Results of ELISA at the NMWA. Numbers in bold indicate values above the threshold.

Sample	Cave	Egg white					Animal glue (anti-collagen#ab19811)					Animal glue (anti-collagen#ab34710)				
		40 µl	20 µL	20 µL	10 µL	average values of blank + 3SD	40 µL	20 µl	20 µL	10 µL	average values of blank + 3SD	40 µl	20 µL	20 µl	10 µl	average values of blank + 3SD
BMM191	EGB	**0.345**	0.238	0.213	0.244	0.334	0.105	0.109	0.113	0.107	0.118	0.144	**0.168**	**0.165**	**0.154**	0.145
BMM094	C(a)	0.237	0.347	0.296	0.303	0.379	0.106	0.117	0.098	0.117	0.117	0.106	0.123	0.120	0.133	0.194
BMM096	C(a)	0.286	0.194	0.228	0.366	0.379	0.111	0.117	0.112	0.114	0.117	0.106	0.129	0.152	0.143	0.194
BMM210	C(a)	**0.457**	**0.460**	**0.385**	0.344	0.379	**0.174**	**0.145**	**0.155**	**0.143**	0.117	0.169	0.156	0.095	0.141	0.194
BMM209	C(b)	0.330	0.249	0.269	0.275	0.334	**0.126**	0.115	**0.120**	0.114	**0.118**	0.135	0.125	0.137	0.116	0.145
BMM099	D	**0.188**	0.135	0.082	0.129	0.179	0.063	0.053	0.058	0.050	0.075	0.237	0.258	0.266	0.288	0.388
BMM082	F(c)	0.310	0.386	0.449	**0.670**	0.615	0.061	0.075	0.077	0.075	0.091	**0.723**	0.402	0.409	0.483	0.525
BMM45	G	0.439	0.431	0.427	0.431	0.463	0.083	0.087	0.085	0.084	0.128	0.095	0.095	0.104	0.104	0.126
BMM212	H(a)	**0.196**	0.149	0.141	0.161	0.179	0.064	0.056	0.058	0.058	0.075	0.230	0.265	0.310	0.250	0.388
BMM129	J(b)	0.412	0.428	0.428	0.424	0.463	0.086	0.088	0.085	0.085	0.128	0.109	0.123	0.121	0.124	0.126
BMM113	J(c)	0.360	0.413	0.399	0.454	0.463	0.097	0.095	0.084	0.079	0.128	0.067	0.111	0.116	0.106	0.126
BMM115	J(c)	0.384	0.410	0.458	0.440	0.463	0.087	0.091	0.080	0.066	0.128	0.105	0.111	0.114	0.103	0.126
BMM120	J(d)	**0.492**	**0.501**	0.068	0.447	0.463	0.061	0.078	0.086	0.081	0.128	0.102	0.103	0.108	0.101	0.126
BMM169	J(f)	**0.402**	0.344	0.246	0.356	0.379	0.099	0.116	**0.134**	**0.119**	0.117	0.125	0.140	0.090	0.141	0.194
BMM137	K_3	**0.216**	0.127	0.138	0.136	0.179	0.070	0.061	0.073	0.065	0.075	0.245	0.271	0.245	0.326	0.388
BMM203	L	**0.206**	0.132	0.124	0.149	0.179	0.059	0.054	0.059	0.058	0.075	0.187	0.264	0.271	0.239	0.388
BMM204	L	0.313	0.362	0.441	0.531	0.615	0.065	0.067	0.067	0.057	0.091	0.432	0.287	0.336	0.376	0.525
BMM053	M	**0.096**	0.075	0.070	**0.082**	0.081	0.059	0.056	**0.071**	0.054	0.060	0.202	0.227	0.260	0.238	0.383
BMM183	N(a)	**0.125**	0.080	0.071	**0.085**	0.081	0.057	0.056	**0.067**	**0.066**	0.060	**0.307**	0.286	0.336	0.292	0.383
BMM035	N(a)	0.241	0.123	0.170	0.162	0.259	0.050	0.045	0.047	0.053	0.062	0.255	0.233	0.224	0.261	0.381
FDM003	Foladi 5	**0.137**	**0.103**	**0.098**	**0.109**	0.081	**0.068**	0.056	0.043	0.059	0.060	0.220	0.255	0.241	0.276	0.383
FDM019	Foladi 4	**0.209**	**0.196**	**0.214**	0.164	0.179	0.072	0.056	0.068	**0.078**	0.075	0.310	0.309	0.303	0.345	0.388
FDM043	Foladi 3	0.156	0.123	0.139	0.137	0.179	0.058	0.054	0.057	0.058	0.075	0.232	0.195	0.211	0.230	0.388
KAK01	Kakrak 43	0.242	0.124	0.159	0.182	0.259	0.056	**0.062**	0.058	0.055	0.060	0.235	0.240	0.213	0.241	0.383
KAK05	Kakrak 44	**0.095**	**0.082**	0.067	0.077	0.081	0.059	**0.061**	0.057	0.059	0.060	0.248	0.301	0.236	0.242	0.383

Sample	Cave	Casein					Plant gum					Tragacanth					Identified
		40 µl	20 µl	20 µl	10 µl	average values of blank + 3SD	40 µl	20 µl	20 µl	10 µl	average values of blank + 3SD	40 µl	20 µl	20 µl	10 µl	average values of blank + 3SD	
BMM191	EGB	**0.506**	**0.282**	**0.340**	**0.240**	0.102	0.024	0.173	0.189	0.126	0.564	**0.076**	**0.083**	**0.078**	**0.094**	0.070	animal glue, casein, tragacanth
BMM094	C(a)	**0.125**	0.097	**0.105**	**0.105**	0.104	0.133	0.165	0.164	0.143	0.183	0.069	0.066	0.071	0.080	0.103	casein
BMM096	C(a)	**1.386**	**0.910**	**0.788**	**0.539**	0.104	**0.196**	**0.283**	**0.276**	**0.250**	0.183	0.070	0.069	0.069	0.075	0.103	casein, plant gums
BMM210	C(a)	**0.186**	**0.130**	**0.138**	**0.139**	0.104	**0.193**	**0.197**	0.137	0.128	0.183	0.076	0.077	0.079	0.074	0.103	egg white, animal glue, casein
BMM209	C(b)	**1.212**	**0.910**	**0.927**	**0.489**	0.102	0.084	0.073	0.070	0.050	0.564	**0.091**	**0.086**	**0.078**	**0.094**	0.070	casein, tragacanth
BMM099	D	0.057	0.045	0.051	0.059	0.078	0.121	0.100	0.108	0.088	0.156	0.050	0.054	0.054	0.059	0.122	
BMM082	F(c)	**0.245**	**0.230**	**0.230**	**0.225**	0.074	**0.304**	**0.323**	**0.298**	**0.311**	0.171	0.053	0.053	0.053	0.086	0.079	casein, plant gums
BMM45	G	**0.192**	0.088	0.089	0.087	0.112	0.104	0.099	0.103	0.084	0.198	0.060	0.062	0.065	0.078	0.084	
BMM212	H(a)	0.071	0.049	0.052	0.062	0.078	0.130	0.126	0.107	0.094	0.156	0.050	0.054	0.055	0.070	0.122	
BMM129	J(b)	**0.615**	**0.496**	**0.480**	**0.466**	0.112	0.108	0.146	0.149	0.157	0.198	0.068	0.071	0.074	0.071	0.084	casein
BMM113	J(c)	**2.340**	**1.063**	**1.212**	**0.755**	0.112	0.159	0.110	0.120	0.122	0.198	0.068	0.070	0.060	0.066	0.084	casein
BMM115	J(c)	**0.563**	**0.310**	**0.362**	**0.220**	0.112	**0.460**	**1.610**	**1.512**	**1.697**	0.198	0.075	0.071	0.074	0.073	0.084	casein, plant gums
BMM120	J(d)	**0.575**	**0.306**	**0.304**	**0.228**	0.112	**0.336**	**0.694**	**0.721**	**0.676**	0.198	0.072	0.071	0.068	0.072	0.084	casein, plant gums
BMM169	J(f)	**1.119**	**0.914**	**1.085**	**0.668**	0.104	**0.223**	**0.203**	0.151	0.166	0.183	0.072	0.071	0.072	0.075	0.103	casein
BMM137	K_3	**0.253**	**0.237**	**0.271**	**0.242**	0.078	0.123	0.065	0.087	0.098	0.156	0.054	0.054	0.051	0.048	0.122	casein
BMM203	L	**1.673**	**0.989**	**1.208**	**1.151**	0.078	**0.202**	0.095	**0.160**	**0.184**	0.156	0.049	0.045	0.047	0.043	0.122	casein, plant gums
BMM204	L	0.057	0.034	0.057	0.066	0.082	**0.184**	0.162	0.151	0.132	0.171	0.052	0.051	0.051	0.051	0.079	
BMM053	M	0.050	0.049	0.053	0.051	0.083	0.088	0.104	0.111	0.106	0.244	**0.057**	0.050	0.056	0.051	0.056	
BMM183	N(a)	0.068	0.060	0.063	0.058	0.083	0.165	0.158	0.211	0.127	0.244	0.050	0.049	0.045	0.041	0.056	
BMM035	N(a)	0.042	0.042	0.040	0.039	0.077	0.091	0.087	0.087	0.079	0.099	0.049	0.050	0.047	0.045	0.050	
FDM003	Foladi 5	0.065	0.047	0.044	0.042	0.083	0.115	0.097	0.120	0.079	0.244	0.046	0.056	0.046	**0.057**	0.056	egg white
FDM019	Foladi 4	0.055	0.053	0.053	0.059	0.078	0.104	0.106	0.094	0.109	0.156	0.051	0.049	0.049	0.041	0.122	egg white
FDM043	Foladi 3	0.058	0.049	0.052	0.063	0.078	0.111	0.095	0.097	0.093	0.156	0.050	0.049	0.052	0.062	0.122	
KAK01	Kakrak 43	0.064	0.048	0.047	0.046	0.083	0.117	0.132	0.151	0.100	0.244	0.050	0.049	0.048	0.056	0.056	
KAK05	Kakrak 44	**0.241**	**0.201**	**0.154**	**0.296**	0.092	0.076	0.110	0.182	0.072	0.244	0.040	0.060	0.046	0.033	0.056	casein

Appendix IV: Aspects and techniques of wall paintings found in neighbouring regions

This appendix is an overview of wall paintings outside of the Bamiyan area and the techniques used to create them. Wall paintings dating between the 2nd and 9th century are found in caves throughout Central Asia, where paintings were made using the *a secco* technique on earthen plaster walls. In particular, many similar to those found in Bamiyan exist around the Tarim Basin and in India (Ch. 1, Fig. 1.36). Large wall paintings are also known to adorn the walls of palaces and residences rather than caves in Tajikistan and Uzbekistan, which once flourished as Sogdiana. In addition, there are abundant examples of paintings on the walls of temples and Buddhist niches in southern Afghanistan, south of the Hindu Kush. In Gandhara and the rest of the Iranian world, images of the Buddha and decorations such as stucco are well known but there are very few examples of wall paintings. In order to gain a better relational understanding of the Bamiyan wall paintings, examples of wall paintings in these regions are provided.

IV.1 Wall paintings in India

The wall paintings of ancient and medieval India have been comprehensively summarised by Osamu Takata (1971). While stone statues are common in India, wall paintings are harder to find. Apart from Ajanta and Pitalkhora, only Bagh has wall paintings dating back to between the late 5th and early 7th century. This section provides an overview of Gupta wall paintings in India, particularly in terms of the techniques and materials used starting with the well-preserved wall paintings in the Ajanta caves. In the left corridor of Caves 9 and 10, for example, it is possible to see monochromatic paintings from the 2nd and 3rd centuries that do not depict an image of the Buddha. Later, during the Gupta dynasty (from the 4th to the end of the 6th century), 'the Buddha image became an important part of wall paintings … the variety of colours became richer, the emphasis on three-dimensionality was added through the use of roundels, and the refinement of the painting technique in general became more pronounced, marking a golden age in the history of Indian painting' (Takata 1971: 151).

Notably, despite the fact that the production of Buddha sculptures became commonplace during this period with the main focus on the Buddha himself, many of the wall paintings were based on Buddhist narratives such as the Jatakas, which depict the story of the Buddha's previous life as well as his biography. These narrative wall paintings, which often run along cave walls, feature three-dimensional decorations as seen in the neck ornaments crafted in raised white, and in the highlighting of eyebrows, eyes, noses and other elevated components in white on Bodhisattvas and offerings (Fig. A.IV.1). Ajanta wall paintings also include many decorative patterns of flowers and plants such as arabesques, as well as representations of human figures, birds and animals. The arabesque pattern on the ceiling of Ajanta Cave 17 (Chapter. 3, Fig. 3.5), enmeshed with animal patterns on a red background, can also be seen on the side of a ceiling beam in Bamiyan's Cave N (a), as described in Chapter 3 (section 3.1.)

The techniques and materials used in the Ajanta wall paintings have been investigated by the Archaeological Survey of India (ASI), the Istituto Centrale per il Restauro (ICR) (currently, the Istituto Superiore per la Conservazione ed il Restauro: ISCR) in Rome and the National Research Institute for Cultural Properties, Tokyo (NRICPT). The caves are made of basalt containing green iron sulfide minerals. After the caves were hollowed out, the walls were chiselled to facilitate adhesion of the wall paintings. According to Nayar *et al.* (1999), one method for making a white ground is to mix lime with boiled ripe kadukka (*Terminalia chebula* (Combretaceae)) although it is not clear how this was deduced. The pigments used include lime, red ochre, yellow ochre, green earth, azurite, lapis lazuli, lampblack (Takata 1971), indigo as blue, a mixture of gamboge and indigo as green and plant gum from vepu (*Azadirachta indica A. Jass* (Meliaceae)) as a binding

Figure A.IV.1 Buddhist wall painting in Ajanta Cave 2. Photo: Y. Taniguchi, 2006.

medium. The gum darkens with age, so it is used for black and blue.

A similar palette has been studied by ASI conservation scientists. Sharma, for example, states that a limited number of mineral-derived pigments were used at Ajanta, namely lampblack, red ochre, yellow ochre, lime/gypsum/white clay, green earth and lapis lazuli (Sharma 2007: 102). The earth pigments, ochre and green earth, are relatively readily available in and around Ajanta. As mentioned above, ASI's analyses mainly concern pigments of mineral origin therefore there are few reports regarding the use of arsenic sulfide pigments (e.g. realgar and orpiment) or synthetic inorganic pigments such as lead white and red lead.

In Cave 17, which was investigated by the ICR, the white ground comprised mainly of kaolinite with some calcium white pigments (calcium carbonate, etc.) while, in rare cases, lead white was found. The analysis also confirmed the use of lapis lazuli as blue; minium, realgar, red ochre and sometimes cinnabar as red; yellow ochre, orpiment and litharge as yellow; and green earth as green (Artioli *et al.* 2007). Results of this study suggest that the Ajanta wall paintings, which are thought to have been painted with relatively simple iron-based pigments (ochres) and lime white, may have been painted with pigments produced by professional groups. Such synthetic pigments may not have originated from the Ajanta area but obtained through broader channels.

Findings of the field survey in Ajanta showed that clay renders were thin and robust and contained crushed plant fibres, probably derived from the excrement of cows or other animals. At times, plant seeds and finely mashed fibre fragments were also observed. The solidity of the walls suggests that some kind of glue was used to apply the render. According to the ASI, the viscous liquids (containing plant gums) obtained by soaking plants such as *Siraitia grosvenorii* (also known as monkfruit or luohan guo), linseeds and hilda seeds (*Terminalia chebula*, commonly known as black or chebulic myrobalan) in water may have been used traditionally for the Ajanta wall paintings. However, no conclusions can be reached as currently there are no analytical results to support this hypothesis. The Gupta wall paintings were painted with materials made of organic substances, but do not include vegetable drying oils such as poppy seed or walnut oil.

The 7th-century wall paintings of the Sittannavāśál Cave, a Jain temple in Tamil Nadu, appear on lime walls. The pigments used include lime white for white, yellow ochre for yellow, red ochre for red, natural ultramarine for blue, green earth for green, charcoal or lampblack for black, and in some places a mixture of natural ultramarine and green earth (Paramasivan 1938). The pigments were basically fixed with lime, as in the *a fresco* technique, but vegetable gums were used in some areas, such as the black parts. The materials and techniques demonstrate a strong mix of Gupta dynasty techniques and those of the Mediterranean world.

The *Citrasūtra* (see Appendix I) is a useful reference for painting techniques and materials used in Gupta wall painting (Sivaramamurti 1978; Kramrisch 1928). Walls were first prepared with a mixture of three kinds of brick powder and clay, mixed with fragrant gum resin, beeswax, honey, *kundara* grass (liquorice), molasses, safflower soaked in oil, lime, bel fruit (*Feronia elephantum*) pulp, lampblack and sand. After soaking the material in water for a month, the paste (earthen render) was applied to the wall and *sarjarasa* (gum of the Śāla tree, white dammar) spread on the surface. There is no mention of the use of animal excrement in the *Citrasūtra*, but various organic substances such as resins, polysaccharides, oils, beeswax and lime are mentioned as sizing agents. After drying, the walls were sized with resin and oil and then polished to a smooth surface. Milk, which contains casein and proteins, may also have been used to size the surface of the wall.

Underdrawings were done in white, reddish-brown or black with five basic colours (white, red, yellow, black and green) used for painting. Pigments included gold, silver, copper, mica, lapis lazuli, lead red, orpiment, lime, lac, cinnabar/vermilion, indigo and metal leaf. There are also references to the use of layering techniques, such as painting over a white and lac mixture with lac and *lodhra* flower red dyes. Animal glue and *bakula* resin glue are also mentioned as binding materials.

Red lac resin has been identified in red in Ajanta Cave 2 by the NRICPT (Shimadzu 2021). If the lac pigment is mainly composed of laccaic acid, which is a red dye derived from stick lac, then red lac resin is a transparent red glaze made by treating both shellac and laccaic acid with alkali and heat (Taniguchi *et al.* 2014). In recent years, the use of this red-coloured material has been verified as a mordant for tin foil in Cave 171 of the Kizil grottoes in Kucha (Zhou *et al.* 2020a,b; 2021). This is noteworthy because it may have been used for red in a wide range of regions from Central to East Asia. The raw material, stick lac, is only produced in tropical regions such as India and Thailand thus making it one of the most important colorants, judging by its geographical distribution. Lac is also mentioned in the *Citrasūtra* (Ch. 40: 25–26).

In the *Citrasūtra*, the names of materials used as binding media with pigments include hide glue (animal skin glue), *bakula* resin and the yellowish-green juice of *dūrvā* grass. It is not clear whether the Sanskrit translations correspond to the correct modern names of plants and pigments, but it can be inferred that a variety of materials, including animal glue and plant gums (or resins), were used as binders. As can be seen in the *Citrasūtra*, a wide variety of organic materials was used to create walls, and various colours were obtained through the complex blending of materials. The Gupta wall paintings have many elements in common with Bamiyan's wall paintings, such as three-dimensional depictions and arabesque patterns together with animals.

IV.2 Wall paintings around the Tarim Basin

In Xinjiang, along the Tarim Basin in the Taklamaken Desert, there are many Buddhist monasteries and Buddhist caves with wall paintings (Chapter 1, Fig. 1.36). Farther to the east, in what is now Gansu Province, there are cave temples in sites such as the Mogao caves, Dunhuang and the Yulin caves. The major archaeological sites in and around the Tarim Basin are spread along the northern and southern fringes of the Taklamaken Desert, including Turfan, Kucha and Kashgar along the Tien Shan South Road (north of the Taklamaken Desert and south of the Tien Shan Mountains) and Khotan, an oasis state, along the South-West Road (south of the Taklamaken Desert and north of the Kunlun Mountains).

Representative sites with wall paintings include the Bezeklik grottoes (from the 6th century) and the Toyok caves in Turfan; the Kizil grottoes (3rd–9th century); the Simsim (4th century); the Kumtura; the Kizilgaha caves, Duldur-Akur in Kucha; the Tomshuk (6th–7th century) (Stein 1980: 1309; 1928: 77); and the Karadon (3rd–4th century) (Ito 2003). In Khotan along the South-West Road, the sites of Dandan Uilik, Mīrān and Niya (Hansen 2004) are also known. As no texts detailing wall painting materials or techniques from this period exist, it is necessary to analyse the wall paintings in order to identify the specific names of the painting materials used.

Many archaeological sites around the Tarim Basin were surveyed by the great powers during their Central Asian expeditions at the end of the 19th and into the 20th century. At this time, many fragments were taken from the caves and transported to Germany, Britain, France, India, Russia, Japan and other countries. During this period, Russia also dispatched a number of expeditions to Central Asia. An overview of analyses conducted at this time is given in Chapter 1 (section 1.3).

In Xinjiang, there are no texts detailing painting techniques or procedures, but valuable records exist of painters from the Tarim Basin in the same period. Examples of such texts include *Famous Paintings through History* 'Tang Dynasty, first volume' by Zhang Yanyuan (9th century) and *On Famous Paintings of the Tang Period* by Zhu Jingxuan (9th century), both of which include descriptions of Yuchi Bazhina (Yü-ch'ih Pa-chih-na), who served in the Sui Dynasty, and his son, Yuchi Yiseng (Yuchi Yiseng/ Viśa Īrasangä / Yü-eh'ih I-sêng), who served in the Tang dynasty (Schafer 1963: 32). They are described as painters from the royal family of the Khotan (Nagahiro 1954).

Apart from the painting at the Daci'en Temple located in Xi'an (Kobayashii 1953), it is not certain which works were actually painted by them. However, Īrasangä is known to have been an excellent painter of Buddhist and foreign portraits. He came to Luòyáng, China in the mid-7th century to introduce a new painting style of Iranian origin. He is credited as having brought the Western 'iron-wire' technique for drawing lines, which involves using a line of unvarying thickness to outline figures. This was drastically different from the painting traditions of the Tang dynasty. The outstanding iron-wire strokes in wall paintings in Buddhist temples at Dandan Uilik and Domoko (discussed later) reflect the painting techniques of this school. The origin of the iron-wire strokes at Bamiyan may be linked to the painting techniques of the Khotan area, which hold the key to connecting the Iranian world of the Sasanian dynasty with Tibetan Buddhism and post-Gupta dynasty techniques. Bussagli discussed these stylistic similarities between wall paintings in Khotan (Farhād Bēg-yailaki and Balawaste in Domoko) and Bamiyan (Caves K_3 or Caves E), each a fusion of Ghandharan techniques with Sasanian Iranian elements (Bussagli 1979: 58).

The Bezeklik caves were built over a period of seven centuries, from the late Northern and Southern dynasties to the Tang, Five dynasties, Song and Yuan dynasties. Their wall paintings start with reddish-brown underdrawings over a white ground, which are then marked in black using lines with varying degrees of detail. The deteriorated state of these paintings – which includes darkening in red layers and the appearance of 1–2 cm cracks and peeling (Caves 20 and 26) – is similar to those in the Mogao caves. Cracks and delamination in the Bezeklik caves may have been caused by the binding media used. Although the wall paintings are of a later date, there are some areas where the gilding seems to have been removed (Cave 39). Compared to Bamiyan, the wall paintings in the Bezeklik caves appear to have a drawing style more closely aligned with Eastern influences, and deterioration that manifests as a more prominent curling and scaling of the surface, thought to be due to the binding medium.

The Kizil grottoes – Buddhist monasteries probably created in a period[1] earlier than Bamiyan – consist of approximately 230 caves. One style of cave is known as the 'central pillar cave', a style that can be also seen at other cave monasteries such as the Mogao caves in Dunhuang. There are also other cave styles such as those with domed ceilings, vaulted and square-vaulted ceilings, which share the strongest similarities with Bamiyan.

At the Kizil grottoes, the caves are carved into cliffs of conglomerate and sandstone, which are considerably softer than Bamiyan's conglomerate cliffs. Differences in the construction of walls at the two sites include the use of keying[2] on soft sandstone, the application of a 1 cm layer of fine earthen render (Cave 38) and of a white ground directly on a sandstone surface (Cave 48) at the Kizil grottoes. A thick layer of paint was applied to the white ground, and it appears that the yellows have whitened or blackened and the reds have darkened (Caves 38, 69 and New Cave 1). This gives the impression that only blue, green and white colours have survived unscathed. In addition, shading that incorporates hues ranging from pink to dark brown was used to create a three-dimensional representation of human figures.

The 7th-century wall paintings include images such as offerings in Central Asian clothing (e.g. Cave 192 and New Cave 1), seated Buddhas that reinforce a Thousand Buddha motif on the dome ceiling (Cave 189), and whitish-green figures depicted in a thick painting layer with fine scaly cracks and gold leaf decorations on vestments (e.g. Cave 69). In our opinion, these bear a high degree of similarity with wall paintings of Bamiyan's second phase, i.e. from the mid-7th century onwards. However, they differ greatly from Bamiyan in that the *pradakshinapata* is often accompanied by a clay sculpture of a reclining Buddha.

The Kizilgaha grottoes, like Kizil, are a group of caves carved into the slightly soft grey sandstone conglomerate cliffs. There are caves with high domed ceilings and with a ritual circuit for *pradaksina* (circumambulation), caves with square-vaulted ceilings and niches with standing Buddhas. The cave forms are similar to those of both Kizil and Bamiyan. Some of the wall paintings share a similarity with those of the Foladi caves, with keying on grey sandstone followed by two layers of reddish-brown sandy render and white render (Cave 11). There are also statues of donors wearing Central Asian tunics (Caves 14, 30 and others) and richly gilded decorations (Caves 14, 16, 23, 30, 32 and others), although the latter have now been removed. The white ground and painted layers are finely fissured and scaled (e.g. Caves 16 and 32), a surface condition common to wall paintings of the second, third and fourth phases at Kizil and Bamiyan.

There is a tendency for the red to blacken, which makes the remaining green, white and blue colours appear more prominent (Cave 11), as well as a pronounced shading of the human figures (Cave 23). In Kizilgaha, there is also a Thousand Buddha motif and a wall painting in which the blue colour, possibly lapis lazuli, is greyish-white (Cave 21). This is also thought to be strongly related to Bamiyan's wall paintings of the second and third phases. Many of the caves at Kizilgaha, like Kizil, have a reclining Buddha. However, the large number of reclining Buddhas within each cave is a characteristic that differs significantly from Bamiyan. The Simsim caves also share similarities with Bamiyan's caves of the second, third and fourth phases, particularly in relation to the painting techniques. Buddhist monuments along the South Road of Tianshan, in the vicinity of Kucha (including Kizil, Kizilgaha and Simsim), each show a strong correlation with the Bamiyan wall paintings from the 7th century onwards.

Located in the desert northeast of Khotan, Dandan Uilik is a Buddhist site that flourished in the 7th and 8th centuries. It was first surveyed by Sven Anders Hedin of Sweden in 1896, and later by British archaeologist Marc Aurel Stein (1900–1901), German geographer Emil Trinkler and Swiss botanist Walter Bosshard, among others. The Xinjiang Archaeological Research Institute visited Dandan-Uiliq in 1996 and in 1998, Christoph Baumer from Switzerland also led an expedition. In 2002, a joint Chinese-Japanese expedition (comprised of the Xinjang Cultural Relics Bureau, the Xinjiang Archaeological Research Institute Bureau, the Xinjiang Archaeological Research Institute and the Niya Research Institute of Bukkyo University, Japan) discovered fragments of wall paintings protruding from the site of a wooden Buddhist temple (Zhang *et al.* 2009; Kojima *et al.* 2007).

Several features similar to Bamiyan's Cave K_3 were identified such as the reddish-brown underdrawings, red outlines of uniform thickness (using iron-wire strokes), black lines with inflection to illustrate the folds of cloth, and standardised seated Buddhas. Moreover, scaly cracks in the white ground layer and the paintwork also resembled those of Cave N(a) and the Foladi wall paintings.

According to the 2007 report by the Chinese-Japanese expedition, the wall paintings at Dandan Uilik seem to consist of renders of sand, mud, lime[3] and with some fibres, a white ground and painting layers (Kojima *et al.* 2007). Due to the fragmentary nature of the analysis, it is difficult to get a complete picture, but it seems that the white is gypsum[4] and the red pigment is iron-based. Unfortunately, the texture on the surface of the painting is not visible due to the synthetic resin that was applied after its excavation in 2002, making it difficult to observe the painting technique. The colours used include red (vermilion, red lead), blue, blue-grey, yellow and black, but the names of the pigments used are not known. Motifs include a figure with a beast's head, which may be a boar or a wolf, as well as Kishimojin (the goddess of childbirth), a seated figure with four arms and a crown, male and female earth deities, and a deformed deity with arms.

Gu Libiya of the Academy of Fine Arts pointed out one similarity between the Amitāyus-Buddha of Tibetan Buddhism and the seated Maitreya Bodhisattva statue (Gu Libiya 2007, CD 4:18), the latter of which has jewel vases in both hands and wears jewel necklaces, *muktāhāra*, bracelets and arm rings (Gu Libiya 2007). Usually, Maitreya Bodhisattva holds a vase in his left hand only but in the case of the Amitāyus-Buddha in Tibetan Buddhism, figures hold vases in both hands, thus there are similarities. It is fascinating to see similarities to the Bamiyan's Cave K_3, particularly as they relate to the overall style of expression and the Bodhisattva iconography. The wall paintings at Dandan Uilik are considered to be strongly influenced by later post-Gupta elements, such as those of Tibetan Buddhism and Hinduism.

Similarly, fragments of wall paintings associated with a wooden Buddhist temple have been excavated from the site at Topraktong 1, Domoko near Khotan. The fragments are probably from wall paintings

Figure A.IV.2 Winged angels from the Mīrān site (after Stein 1981: pl. XLI, M. III. v).

Figure A.IV.3 Shakyamuni (Gautama Buddha) statues and monks from the Mīrān site (after Stein 1981: pl. XLII, M. III. 003).

depicted on earthen walls with a white ground, but the details are not clear. The painting seems to have been done in white, red, green and other colours, using gradation techniques. One figure of a male warrior, possibly Vaiśravaṇa, with an arabesque pattern in green and white on the collar of his upper garment painted in iron-wire strokes – similar to that seen at Bamiyan – was excavated from the south wall (Chapter 1, Fig. 1.37). The wall paintings from the Dandan Uilik site have some elements in common with those from Bamiyan's second, third and fourth phases, such as the underdrawings, arabesque motifs and gradation techniques, but the greatest difference is the use of gypsum as the white ground. It is hoped that future comparative studies will reveal the detailed painting structures and

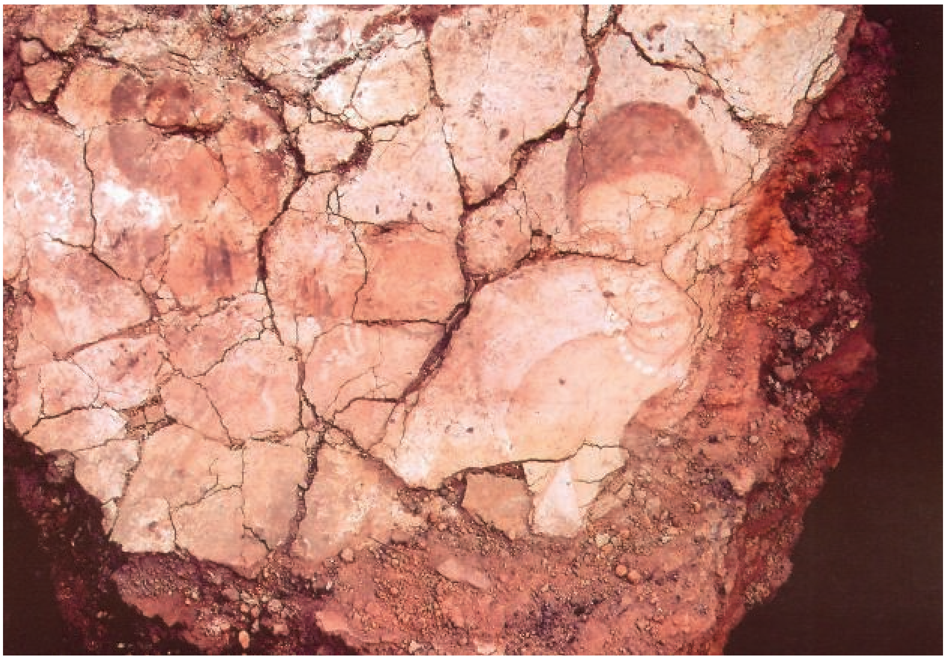


Figure A.IV.4 Wall painting fragment from Jinan Wali Dheri (after Bhatti 2006: 95).

binding media used in wall paintings around the South-Western provinces.

The Karadong site, located to the southwest of the Taklamakan Desert, has two Buddhist monasteries: Temples N61 and N62. Both wooden structures had wall paintings which, according to Genzo Ito, were similar to those at Niya and Dandan Uilik and had several unique characteristics such as the use of white highlighting on faces (Ito 2003). While the Chinese-French mission dated the monasteries as belonging to the 3rd– 4th century AD, and the Japanese mission as the 3rd–5th century AD (Ito 2003), radiocarbon dating with the IntCal20 suggested 195 BC–AD 410 (1σ), 516 BC– AD 645 (2σ) for Temple N61, and AD 213–328 (1σ), AD 129–352 (2σ) for Temple N62.

The site of Mīrān is located far to the east, near the outlet of the Taklamakan Desert. The most famous wall painting there had Greco-Buddhist style winged angels found near the third stupa, now in the collections of the National Museum, New Delhi and the Tokyo National Museum. The red robes of a monk and inflected brushstrokes are the distinguishing characteristics of this piece. The pronounced nostrils and brushstrokes are fairly similar to those of Parthian and Sasanian wall paintings (Figs A.IV.2 and A.IV.3).

The wall paintings at Mīrān were painted using an *a secco* technique after plaster had been applied as a white ground to a clay render. Stein, who discovered the wall paintings, described them as being 'all painted in tempera' (Stein 1921: 499). Sir Arthur Church, who analysed a sample of the wall paintings at Mīrān, identified an earth render reinforced with reed leaves and stems[5] (Church 1921). The gypsum ground is coloured pink by iron oxide, probably red ochre. Church concluded that wall paintings at Mīrān were painted using unique methods because gypsum, not lime mortar, was used for the ground, which fixed the pigments to the wall through the formation of calcium carbonate as per European *a fresco* techniques. The pigments at Mīrān were identified as iron oxide, malachite, low-purity yellow ochre, brown earth and carbon black, the last of which was probably used as ink. Judging from motifs, substrates and how lines were drawn, the wall paintings of Mīrān are thought to have incorporated more Western techniques and materials than other wall paintings around the Tarim Basin. There is also the name Tita, which seems to be the name of a painter of Western origin (Titus) adapted to Sanskrit or Prākrit. One wall painting has an inscription in the Kalośūti script, which points to a connection with the Gandhara region (Stein 1921: 530).

IV.3 Wall paintings in Pakistan

While a number of stone statues and stuccoes have been found in Gandhara, wall paintings are a rarity. One wall painting was found at Jinnan Wali Dheri (Fig. A.IV.4), a Buddhist site located 10 km north-east of Taxila. This site, which dates to between the 3rd and the 8th century, contains a stupa and a monastery. Some 2,000 fragments of wall paintings depicting Buddha and Bodhisattvas, probably dating back to the 5th century, were found on both sides of the monastery entrance during excavations in 2002 (Bhatti 2006). Khan (2020) dates the wall paintings to the 4th and 5th centuries based on Kharoṣṭhī inscriptions, while Fukuyama believes them to be late 5th century by comparing them with wall paintings in other caves at Ajanta, Foladi and Bamiyan (Fukuyama 2014: 383–392). The entrance and painting fragments had been severely burnt, suggesting exposure to fire. These are the only wall paintings found to date. The use of white to represent a necklace, the technique of highlighting the eyebrows, eyes, nose and elevated areas with white, and the representation of the body are strongly reminiscent of the Ajanta wall paintings.

The materials used have not been fully identified. According to visual observations (in July 2006), the wall paintings consist of at least two layers of earthen render with a white ground layer and a painting layer on top. The nature of the original clay is unknown as the earthen render has been darkened by fire. The upper layer of the render contained many white particles, something not observed in wall paintings in India or Afghanistan. According to Fukuyama's observations, stone masonry is covered with a layer of plant fibres mixed with mud, about 2 cm thick, which is then covered with one layer of white ground: fine-grained earth mixed with lime at a thickness of about 4 mm. Underdrawings were made with a brownish colour and the painting has some lapis lazuli blue (Fukuyama 2014: 381). According to a report by Khan and Mahamood-ul-Hussan (2008: 305), the black, white and red pigments used in this wall painting could be obtained from the Khari Murit hills, about 25 km west of the site, and the blue pigments are believed to be made from lapis lazuli. This blue pigment remained blue even after exposure to fire. Very few examples of wall paintings exist in Pakistan, especially in the region of Gandhara. Those found were probably painted in the *a secco* technique, very similar to Ajanta's wall paintings.

IV.4 Wall paintings in Iran

In Iran also, stucco reliefs are common and very few examples of wall paintings exist. To date, only two examples of wall paintings are known. The Iranian wall paintings appear to be secco works painted on earthen walls, although the details are not clear. The site of Kuh-e Khwaja, located in the easternmost province of Sistan-Baluchestan, is a Parthian fort site and a Zoroastrian temple dating back to between the 1st and 3rd century. The wall paintings, which contained Greek and Iranian heads and paintings of Iranian royalty, have been associated with a 1st-century structure (Kawami 1987a,b; Yamauchi 1997) (Fig. A.IV.5). The wall paintings were detached by Sir Aurel Stein and Ernst Herzfeld. The Stein collection is now in the National Museum, New Delhi, while the Herzfeld collections are held in the Metropolitan Museum of Art, New York, and Tehran.

According to Herzfeld (1941), the wall painting with purple, purplish-red and orange colours is a 'medieval' painting in the Greco-Bactrian style. Wet chemistry and SEM-EDS detected gypsum used as a white ground, carbon black (not ivory or bone black) for black, copper-based pigments for green and iron oxide for red and flesh colours. No mercury sulfide was identified in red by Lawrence Becker and Robert Koestler of the Metropolitan Museum of Art (Kawami 1987b). A visual examination of the collection at the museum suggests that the painting was executed on a render made of clay containing fine straw fibres. The surface of the render was minimally prepared and remains highly uneven. A white ground was applied on top of the render, and black lines (approximately 2–4 mm in width and inflected) were drawn with simple brushstrokes and painted with yellowish-brown and light brown colours, probably using the *a secco* technique. These lines were composed of natural earth pigments. The bright colours of purple and scarlet described by Herzfeld could not be observed.

Another example can be found in a wall painting in a Sasanian period manor house at Hājiābād in southern Iran, some 280 km east of Shiraz, believed to date back to the 4th century. Portraits of gods, kings and heroes are painted in a circle. The details of the technique are not clear, as there are few references to it, but it is reported to have been done on a clay wall mixed with plant fibres, with a red ochre underdrawing, and final outlines and details in black (Azarnoush 1994). The secco wall painting probably used a limited range of colours: milky white, blue, black, red, brown and yellow. In one case, the pattern was painted in white on a dark grey-blue background. The strong black outlines and lines for detailing, which are common to all these sources, are also considered to be characteristic of Sasanian painting.

Figure A.IV.5 Fragment of a wall painting from the Kuh-e Khwaja site in a collection at the Metropolitan Museum of Art, New York. Photo: Y. Taniguchi, 2009.

IV.5 Wall painting paintings south of the Hindu Kush, Afghanistan

Despite the Greco-Roman motifs, the use of gypsum and clay to create stuccoes is common in wall paintings south of the Hindu Kush Mountains, as seen at Hadda (Kushano-Sasanid dynasty, 2nd–5th century). This technique is thought to be similar to that used in the wall paintings of Iran and Mīrān. The Hadda site is a Buddhist monastery located near the Khyber Pass on the border between Afghanistan and Pakistan. It shows a blending of Greek and Buddhist styles, as seen at the temple with a stupa where a statue of Hercules with a *vajra* pestle stands next to a stucco sculpture of Buddha rather than the *vajra* god. Strong Greco-Buddhist influences can be seen. One example of the styles and materials used in Hadda is a wall painting, now housed in the Musée national des arts asiatiques-Guimet in Paris, that shows a pair of winged angels holding flower cords decorating a Buddha image in a Buddhist niche (see Fig. 2 in the Introduction).

An analysis conducted in Italy of the materials in the Hadda fragments found that not only was the *a secco* technique applied, but also that some fragments – such as one depicting a standing Buddha (MG 17451, Musée national des arts asiatiques-Guimet) – were painted in an *a fresco* technique using lapis lazuli and red ochre as pigments and lime as a ground (Figs A.IV.6 and A.IV.7) (Cambon 2004). This is an extremely rare example of a Greco-Buddhist wall painting exhibiting a strong Hellenistic influence, mixing both the *a fresco* technique using lime and the *a secco* technique using gypsum.

The Hadda site, located south of the Hindu Kush Mountains, is a foothold between the Mediterranean and Eastern worlds, influenced by Hellenistic culture. In terms of iconography and materials, it can be considered an eastern limit of the *a fresco* technique, showing both Greek and Roman as well as Indian Buddhist elements. However, in terms of painting techniques and materials, it is unlikely to be related to the Bamiyan wall paintings. In 2016–2020, the Tokyo National University of Fine Arts and Music (now the Tokyo University of the Arts) carried out a conservation project on the wall paintings from Mes Aynak (Kijima 2021; Yasuda *et al.* 2020), a Buddhist town that flourished between the 3rd and 7th century. Four

Figure A.IV.6 Wall painting fragment from Hadda (MG17451) (after Cambon 2004: 162, fig. 70).

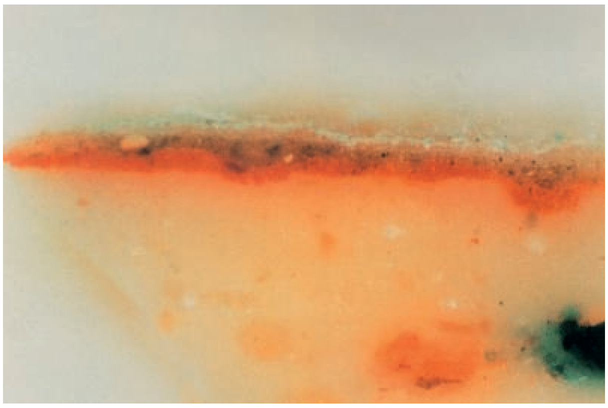


Figure A.IV.7 Cross-section of a wall painting fragment from Hadda (MG17451), painted with the *a fresco* technique (after Cambon 2004: 166, fig. 82).

Buddhist monasteries, a stupa and a Zoroastrian fire temple are known at the site, where secco wall paintings and clay sculptures were excavated. Moreover, rich underground copper deposits have left traces of bronze metallurgy dating from the Bronze Age. The China Metallurgical Group Corporation (MCC) has purchased the mining rights to the copper therefore the National Museum of Afghanistan has worked hard to relocate and house the stupas, wall paintings and statues through archaeological research before they are lost (Bloch 2015).

IV.6 Wall paintings in Afghanistan, Tajikistan and Uzbekistan, north of the Hindu Kush

North of Bamiyan, in what is now Tajikistan and Uzbekistan, are a number of sites with Buddhist monuments and wall paintings associated with the Sogdians:

- Kara-tepe (late 1st–2nd century) (Mkrtichev 2002);
- Fiyaz-tepe (2nd–4th century);
- Balalyk-tepe;
- Ajina-tepe (7th–8th century) (Babadjanova 2017; Shahi 2019);
- Kafir-Kala (Litvinsky and Zeymal 1971);
- Kala-i Kafirnigan, Penjikent (Marshak and Raspopova 1990, 1998; Maršak 2000);
- Dokhtar-i Noshirvan (Nigār) (early 8th century) (Klimburg-Salter 1993; Mode 1992);
- Tabqa.

At the site of Kala-tepe, a wall painting of a seated Buddha has been found on an earthen rendering wall (Mkrtichev 2002; Yasuda 2020). Tavqa, a fortified site 4 km north of the town of Sherabad, Uzbekistan, is said to have been painted in the Toharistani tradition, with two layers of paint (Rahmonov 2001: 139). Among them are the wall paintings of Ajina-tepe and Kala-i Kafirnigan. Much of the analytical work on wall painting techniques in this region has been carried out by Russian scholars (Litvinskij 1981) since Uzbekistan and Tajikistan were formerly part of the Soviet Union. Alexander Kossolapov and Boris Marshak of the State Hermitage Museum in St Petersburg collected examples of Russian analyses of wall paintings; these have been verified recently with new data (Kossolapov and Marshak 1999; Kossolapov and Kalinina 2007). In addition, since 2008, NRICPT has been conducting research on the wall paintings discovered by the excavation team of the former Soviet Union in the collection of the National Museum of Antiquities of Tajikistan. As part of this work, it has analysed wall paintings from Kala-i Kakhkakha, Penjikent, Kara-i Kafirnigan, Ajina-tepe and other sites (Shimadzu 2009).

These wall painting fragments are also painted using an *a secco* technique consisting of kneaded earthen clay with chaff tempers. The best-known wall painting fragment from the site of Kala-i Kafirnigan is a group of worshippers wearing Central Asian tunics with Sasanid prayer beads in the collection of the

Figure A.IV.8 Wall painting fragment from the Kala-i Kafirnigan site in the collection at the National Museum of Antiquities of Tajikistan. Photo: Y. Taniguchi, courtesy of NRICPT, 2007.

National Museum of Antiquities of Tajikistan. The red background, reddish-brown lines for underdrawings and iron-wire strokes are representative of the location and era (Fig. A.IV.8). The colours are dull and the work lacks shading and gradated hues. The surface of the wall and the ground layer are uneven, and the wall seems to have been minimally adjusted relative to later Bamiyan wall paintings. NRICPT analysis confirmed the presence of ochres in yellow and red areas, and lead-containing pigments in paint layers, although they are now pale red or grey. Sulfur, probably gypsum, has been detected in the white ground of the wall paintings. No green or blue was found but cinnabar/vermilion or mercury-containing reds were identified from Ajina-tepe (Shimadzu 2009, 2010: 111).

According to Russian analysis of the wall paintings from Ajina-tepe, the substrate coexists with gypsum or lime, and yellow ochre was used in yellow, cinnabar/vermilion (artificial or natural) in red, and lapis lazuli in blue. In an unusual case, green earth (ceradonite) was identified in green (Kossolapov and Marshak 1999). The geological setting of Tajikistan suggests that green earth is available in the vicinity but its use is not found in other Buddhist wall paintings in Central Asia.

Although the Buddhist wall paintings north of the Hindu Kush date from the 7th or 8th century, their techniques and materials are relatively similar to Bamiyan's wall paintings. These wall paintings are not related to Buddhist monasteries, but were painted to decorate the private residences of Sogdian lords, therefore they are not the subject of this book. However, because of their possible relationship to Sogdiana in terms of region, period and trade, they are interesting. Samarkand was formerly called Maracanda, and the central capital of the Sogdians was Afrasiab. Many wall paintings have been excavated from the capital, including the well-known Afrasiab series of wall paintings from the Paintings of the Ambassadors (7th century) in the private residence of a nobleman. (Fig. A.IV.9). A long procession of people of different origins and a large number of Turks escorting delegations from China and Tibet are depicted on the 7th-century wall paintings. Envoys from the Central Asian principalities of Chach (now part of Tashkent) and Chaghaniyan (between Samarkand

Figure A.IV.9 Wall painting of the Hall of the Ambassadors from the Afrasiab (Samarkand, Uzbekistan). Photo: Y. Taniguchi, courtesy of NRICPT, 2005.

and Termez) are shown wearing silk fabrics with the typical Sasanid pearl roundel motif featuring an animal (Grenet 2007). In addition to lead white and gypsum, lapis lazuli blue, orpiment and vermilion were identified in Afrasiab wall paintings (Kossolapov and Marshak 1999).

The site of Penjikent, near Samarkand, was once the capital and numerous wall paintings dating back to between the 5th and 8th century have been found in local palaces, temples and private residences (Marshak *et al.* 1990). The site of Shahristan (Kara-i Kafkaha), also in the north of Tajikistan, contains many wall paintings (c. AD 740) (Kageyama 2013), including a large group of battle scenes.

Wall paintings from the 6th and 7th centuries found at the Varakhsha site in Bukhara are also important. Their red background depicts human figures on elephants (interpreted as the deity Adbag/Indra) (Tanabe and Maeda 1999: 215) and a hunting scene in which the deity is pursuing a large leopard (Shishkin 1963). The ceiling appears to be Laternendecke (Maršak 2000). In 2012, the NRICPT conducted an X-ray fluorescence analysis of wall painting fragments from the Varakhsha site, which are housed in the National Museum of Natural History of Uzbekistan. It reported that the red fragments are iron-based, the white fragments are calcium-based (gypsum or lime), and the painted areas are lead white (Hayakawa *et al.* 2013).

Other wall painting fragments from the site of Fayaz-tepe from the 2nd–4th century (Al'baum 1991) and the late 4th century (Muzio 2008) have also been analysed. Iron, lead and mercury were identified in the red (Hayakawa *et al.* 2013), each of which indicates red ochre, lead red and cinnabar/vermilion, respectively, while calcium-based white (gypsum or lime) was used for the ground. From 2016 to 2020, wall painting fragments from the Fayez-tepe site were researched and analysed by the Japanese mission; they identified gypsum and iron oxide (Tamura *et al.* 2021). The date of the wall paintings, however, should be revised from recent studies on styles of inscriptions and ornaments (Kageyama 2022) as it could be earlier.

Russian scholars have conducted in-depth research on painting techniques in the Sogdiana wall paintings, as well as on the situation of wall paintings north of the Hindu Kush. The results have been published by Kossolapov and Marshak (1999) and Kossolapov and Kalinina (2007). In the territory of Sogdiana, the NRICPT conducted a survey of wall paintings from the site of Kala-i Kakhkha[6] as part of a broader survey of the National Museum of Antiquities of Tajikistan's collection (8th century) (Shimadzu 2009, 2013). The wall paintings of Sogdiana, like those of the surrounding area, are painted on earthen walls using an *a secco* technique. However, instead of cave walls, they are painted on the walls of mud-brick buildings.

Russian analysis of wall paintings from Penjikent indicates the use of gypsum or lime as a white ground. In addition, lapis lazuli has been identified in blue, red ochre in red, and yellow ochre, litharge and orpiment in yellow (Kossolapov and Marshak 1999: 74; Shimadzu 2013). According to Kossolapov and Marshak, no green pigment is recognised in the wall paintings in Sogdiana but malachite is sometimes found as a green colour at the 6th–late 9th-century site of Shahristan (the formal capital city of Ustrushana, Kala-i Kafkaha I) (Kossolapov and Marshak 1999: 52, 56, 78). A layer of black comprised mainly of carbon was applied underneath the lapis lazuli which has produced a darker blue colour. Shimadzu's analysis accords with these results, but also highlights the presence of some lead-based pigments in the Shahristan wall paintings. The detection of copper in black areas suggests that, as with Bamiyan's wall paintings, a green colour consisting mainly of a copper compound was used.

Wall paintings at Afrasiyab, unlike those at Penjikent and Shahristan, were characterised by the use of lead white, gypsum and cinnabar/vermilion in red. A defining feature of the Sogdiana wall paintings is that the plaster used for white grounds is a highly pure gypsum with no traces of anhydrite (Kostrov 1959, 1963). Kossolapov and Marshak believe that the Sogdiana wall paintings were basically produced with pigments and binding media in the vicinity of Sogdiana. However, they also suggest that materials obtained through trade, such as lapis lazuli from Batakhshan as well as lead white and cinnabar/vermilion, may also have been used (Kossolapov and Kalinina 2007). The word *spytc* (meaning 'white stuff'), assumed to be lead white, is listed as a trade item in the 'Sogdian Ancient Letter V' written by a Sogdian merchant. The Sogdians are known to have traded in lead white as *spytc* as early as the 6th century (de La Vaissière 2016 [2019]: 41, n.15).

Russian scientists Vadim Birstein (1975, 1977) and Camilla Kalinina of the State Hermitage Museum have also analysed binding media (Kossolapov and Kalinina 2007). Birstein concluded that the gums of fruit trees such as cherry and apricot would have been used as binding media in Central Asian wall paintings (Kossolapov and Marshak 1999). Kalinina used GC-MS analysis to detect polysaccharides in wall paintings from the Penjikent site, including a large amount of galactose and mannose, and small amounts of arabinose and xylose. It suggests that the gum from leguminous species such as guar and shoofly (*Biancaea decapetala*), both of which contain high levels of galactose, may have been used as a binding medium when mixed with small amounts of gum from fruit trees such as peach and apricot (Kossolapov and Kalinina 2007: 92).

These wall paintings rest on a white ground on earthen plaster. They have underdrawings with red lines using ochre and are painted in various colours and shades with details marked in black or ochre strokes; the painting has neither gradated hues nor three-dimensional shading techniques. Few greens remain, many of which are thought to have turned black, but the green colour covers a smaller area compared with blue, yellow and red. There is no evidence of gilding or other metallic decoration. With regard to technique and material, the wall paintings of Sogdiana have much in common with those of the Ajina-tepe and Kala-i Kafirnigan sites, both north of the Hindu Kush. However, the use of Sasanid Persian motifs, iron-wire strokes, gypsum as a white ground, and the application of a black layer under a blue layer of lapis lazuli are reminiscent of painting techniques used in the first and second phases of the Bamiyan wall paintings, such as those found in the niches of both Giant Buddhas and in the caves around the Eastern Giant Buddha. Old wall paintings are also known in Afghanistan, north of the Hindu Kush: specifically, 4th–early 5th-century wall paintings found in Ghulbiyan, Bal Chirāgh within Faryab Province. These were viewed as early Sasanid period paintings with iconography of the Iranian god Sirius, Tištrya, and two groups of worshippers (Lee and Grenet 1998). Technical and material information was not reported.

Notes

1. Kizil's date of the excavation was determined at the beginning of the 20th century by Waldschmidt, who classified the caves into three styles from the beginning of the 6th century to the middle of the 7th century. Dating was based on the typeface of the Brahmi script on the wall paintings. The Chinese researcher Su Bai classified the caves into three styles from the 4th to

the 7th century based on cave structures and the results of radiocarbon dating by Peking University. However, there are problems with the methodology for dating, and the chronology pertaining to Kizil should be re-examined (Iwai 2008).

2. A technique for cutting into walls to improve the adhesion of the upper layer to the lower surface.
3. Described as 'calcium carbonate' in the report.
4. It is stated that lead dihydrate sulfate was detected by laser Raman spectroscopy, but the Raman spectrum is shown. It is more likely that calcium dihydrate sulfate (gypsum) was incorrectly stated as lead dihydrate sulfate.
5. This probably refers to straw fibre.
6. The Kala-i Kakhkakha site is located in Bunjikat village in the Shahriston District.

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The site of Bamiyan in Afghanistan site has suffered numerous disasters throughout its history which have resulted in damage to many of its cultural treasures including the Great Buddha statues and innumerable Buddhist wall paintings. Despite these losses, the authors of this volume discovered the world's oldest example of an oil painting, dating back to the mid-7th century, as well as a myriad of other clues to the historical painting techniques of this region.

This book presents invaluable data and scientific analyses relating to the identification of the materials and techniques used in Bamiyan's Buddhist wall paintings. The results are discussed from a global perspective, addressing the fluidity of cultural interactions between the geographical east and west across the Eurasian continent.

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