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ARTISTS' PIGMENTS RECONSIDERED: DOES MODERN SCIENCE MATCH THE HISTORICAL CONTEXT?

Christoph Krekel, Ursula Haller and Andreas Burmester

ABSTRACT

An evaluation of old analytical reports and publications about historical painting materials revealed systematic errors in interpreting analytical data, beginning with too narrow a range of expected materials, simplifications in their characterization, and ending with over- and misinterpretations. Most of these errors could be avoided by a systematic consultation of historical documentary sources: performing analyses on historical materials makes sense only when interpreting them in an historical context. This quite banal statement cannot be taken for granted at all, which is shown by the examples *Kesselbraun*, mountain green (*Berggrün*) and *Schiefergrün*. To broaden the range of expected materials and to increase the reliability of analytical results, it is indispensable that scientific and historical disciplines work together very closely. Only crossing boundaries between disciplines will prevent systematic mistakes in the interpretation of analytical findings.

ZUSAMMENFASSUNG

Eine Evaluation alter Analysenberichte und Publikationen ergab gravierende systematische Fehler bei der Interpretation naturwissenschaftlicher Analyseergebnisse von historischen Malmaterialien. Es konnten einerseits starke Vereinfachungen beim Materialspektrum und der Charakterisierung der Materialien, andererseits Über- und Fehlinterpretationen von Analysendaten festgestellt werden. Viele dieser Fehler könnten durch eine systematische Einbeziehung von Quellschriften vermieden werden, denn Materialanalysen können nur innerhalb des historischen Kontexts sinnvoll interpretiert werden. Dass diese an sich banale Aussage leider keine Selbstverständlichkeit ist, wird anhand der Beispiele Kesselbraun, Berggrün und Schiefergrün belegt. Um die Bandbreite der zu erwartenden Materialien zu erweitern und die Zuverlässigkeit der Analyseergebnisse zu erhöhen, ist eine enge Zusammenarbeit zwischen naturwissenschaftlichen und historischen Disziplinen unabdingbar. Nur wenn Grenzen zwischen den Disziplinen überschritten werden, können systematische Fehler bei der Interpretation von Analyseergebnissen vermieden werden.

INTRODUCTION

Since the successful introduction of investigation methods such as scanning electron microscopy with energy-dispersive X-ray analysis (SEM-EDX), Raman microscopy, and others, the analysis of materials used in easel painting, manuscript painting and wall painting seems to be a problem that has been solved. The data produced by these scientific methods are presented in a continuously improving computer-designed format. In many cases, this leads to very straightforward and easy interpretations, where one set of analytical data in the major compounds always leads to the same final result.

Accordingly, in the secondary literature on art technology, a quite narrow set of pigments used in easel painting up to the end of the eighteenth century has been established over the last 40 years and has constantly been found since. Basically, this spectrum of colourants has been known since the fundamental works of Gettens, Plesters, Kühn, and others. Assuming the correctness and completeness of the previous investigations and publications, a mutually reinforcing system has been established in which the interpretation of the analytical data is considered to represent a high certainty of truth.

Of course, the present authors are not the first to discover that the situation in the analysis of painting materials is more complex, and most of the leading working groups have considered the problem. By re-evaluating old scientific reports and publications, six major reasons for mistakes could be identified.

- Firstly, in spite of modern analytical methods, some pigments, mentioned in documentary sources and undoubtedly used frequently by painters, have been overlooked, although they have probably been among the samples analysed.

- Secondly, in certain groups of pigments, for example mineral copper greens, the interpretation is still quite often reduced to simple equations like green + copper + crystalline = malachite, which draws a biased picture of the actual situation.
- Thirdly, secondary conversion products, such as so-called copper resins or copper oleates, have been considered to be commonly-used pigments.
- Fourthly, the attempt to find designations corresponding to increasingly accurate analytical results leads to a modern terminology that very often suggests a higher diversity of materials than was historically offered.
- Fifthly, the attempt to correlate specific historical terms with analytical data carries a danger of over-interpretation, if we are trying to be more exact in the definition of historical materials that occurred in the past.
- Finally, the analysis of historical materials is the analysis of mixtures, which is very often neglected. If we discuss iron vitriol as a historical material for black tanning, we have to know that we are not referring to pure $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$; if we think of lead-tin yellow I, it is not Pb_2SnO_4 , but a mixture with SnO_2 and PbO (which has possibly further reacted with the medium) etc.

In any case the results of the investigations do not match the historical context and there is a "discrepancy between the materials described in past treatises and reports of pigments found on artefacts", as Eastaugh, Walsh, Chaplin and Siddal have already stated in *The Pigment Compendium* [1]. The main reason for this discrepancy seems to be the lack of education of many scientists in using, understanding and trusting in documentary sources. Some of them may regard samples as pure physical matter and not as part of an artefact in its historical context. They are not trained in using old treatises as a tool of the same value as the electron microscope or gas chromatography – mass spectrometry (GC-MS). It is a challenge for the future to help scientists to cross boundaries to historical art technology.

By singling out two pigments, namely *Kesselbraun* and mountain green (*Berggrün*) as examples, this contribution aims to illustrate the problem and to suggest ways of dealing with it.

DOCUMENTARY SOURCES AS A LANDMARK FOR ANALYTICAL INVESTIGATIONS: KESSELBRAUN

Given that the analysis of a sample of a dark brown paint layer from the sixteenth century shows copper and oxygen to be the major constituents along with some particles of charcoal black only, many conservation scientists would assume that the sample contains either converted verdigris or a former black paint with some copper salt as a siccative. The likelihood that the analyst would consider a special copper-containing brown pigment is quite low. The lack of awareness of the existence of such a pigment may deprive the scientist of the possibility of expecting it and looking for it more closely.

From the position of science, it would be more professional always to expect something unexpected from analytical data. This presupposes that we give up all claims to certainty to create an atmosphere in which every little anomaly appears to be strange and astonishing. In a completely different context, Ashley-Smith

[2] has introduced the idea of 'professional uncertainty', which may easily serve as a concept here. However, the structure of human thinking is organised in such a manner that, out of a set of solutions, we automatically choose the one we are aware of, rather than contemplating the high probability that something unexpected may occur. Therefore, recognizing and scrutinizing unusual data becomes easier as the number of possible solutions known to the scientist increases. To illustrate this, think of a rider sitting blindfolded on an animal not knowing that there are other animals which can be ridden apart from horses. The rider would undoubtedly assume that he/she is sitting on a horse, without further consideration. Moreover, he/she would not even question whether it was a horse. Only if he/she knows that riding is possible on mules, camels or even cows, would he/she consider the problem and take these solutions into consideration.

Documentary sources are certainly the most important tool to create an environment of professional uncertainty, because they provide a variety of unsolved questions for which our analytical data may be the answer. Indeed, by thinking of trade sources like account books, inventories [3], apothecaries' *taxae* [4, 5] etc., or of literature concerning mining, technology, alchemy, etc. providing a picture of artists' materials traded at a certain location and at a certain time, the spectrum of possibly-used pigments differs remarkably from the narrow set usually found. In addition, it is astonishing, how many recipes in specific art technological sources are still waiting for an interpretation or re-interpretation.

To give only one example of pigments obviously never found in artefacts, although frequently mentioned in documentary sources, attention can be re-focussed on copper-containing brown paint layers.

In the diary of his journey to the Netherlands in 1520–21, Albrecht Dürer wrote that he bought a pigment which he calls *Kesselbraun* (literal translation: kettle brown) [6]. In trade sources and in artists' treatises, *Kesselbraun* is listed quite frequently. From the fifteenth to the nineteenth centuries, hints as to the nature of *Kesselbraun* can be found in other sources concerning mining, glass production and the production of ceramic glazes and copper processing, because it was used in large quantities to glaze pottery, colour glass and also as a pigment for painting [7]. Not knowing anything about the nature of Dürer's pigment, except for its obviously brown colour, it is definitely a problem to link the historical term to analytical findings. However, significantly high copper contents in samples of brown paint layers analysed on the occasion of the Munich Dürer exhibition in 1998 led to the assumption that a brown copper pigment had been used and must be identical to the material mentioned as *Kesselbraun* [8]. To verify this assumption, a link between the term and the material had to be sought in a documentary source that mentioned *Kesselbraun* and which was dedicated to a specific artefact. It was found in the *Einnahmen- und Ausgabenbuch* (*Purchase and Distribution Book*) of Wolf Pronner [9], who was an official at the Munich court of Duke Wilhelm V and held the title of an 'administrator of painting'. From 1586 to 1590, he listed painting materials purchased and distributed by him to the artists working at court. He not only had to purchase materials of the highest quality wherever he could get them at good prices and distribute them in small quantities to the artists, but he also had to record everything accurately, particularly the purpose of the materials. This provides a direct link between artefact and materials in very many cases. In addition, Pronner had to supervise the ongoing work of the painters. It is therefore possible to reconstruct the manufacturing of selected works of art from the purchasing of the materials to their completion.

In Pronner's notes, the extraordinarily large quantities of *Kesselbraun* attract attention, Fig. 1. No other pigment was

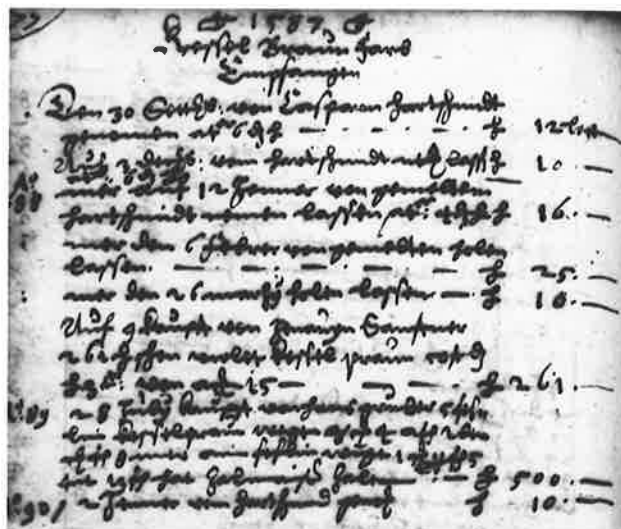


Fig. 1 One of the pages of purchase of *Kesselbraun*, detail. (Wolfgang Pronner, *Purchase- and Distribution Book*, BHStA, HR I, Fasz. 279/4, fol. 77v).

purchased in anywhere near similar quantities. He bought it from Munich merchants, Tyrolean producers (or merchants) or international trading companies that shipped it from Venice. *Kesselbraun* was used as a pigment for paintings in oil and in the fresco technique. In the monumental high altar painting of the court church of St Michael, the *Triumph of Saint Michael* by Christoph Schwarz, it was used to colour the ground. Moreover, large amounts were added to stucco pastes, mortars and screeds.

A closer look at the notes for the side altar painting *Martyrdom of St Andrew* (by Christoph Schwarz and Alessandro Paduano) shows that *Kesselbraun* and *Umbrä* were delivered by Pronner to the painters as brown pigments. An examination of brown samples by SEM-EDX and X-ray diffractometry revealed that the brown paint layers largely consist of a brown copper-containing pigment with a thus far unknown crystal structure, Figs 2 and 3. On the basis of Pronner's notes, it is quite probable that this material is *Kesselbraun*. Although further examinations still have to be carried out and the spectrum of documentary sources to be evaluated has to be extended, it can be postulated that *Kesselbraun* is a mixture of black and red copper oxides, accruing as by-products of copper forging. Copper smiths sold these oxides as fine grades of 'copper hammer blow' (copper oxides formed during the working of copper metal), copper slag or copper ashes [10]. It is also to be found as 'copper brown' (*Kupferbraun*) in documentary sources [11].

However, the following question arises: how is it possible that this obviously ordinary pigment could have been overlooked for so many years? There are two possible explanations: firstly, copper oxides are not always crystalline and, being quite reactive, they might have formed brown and green organic copper compounds with the medium. Secondly, some of the data on the brown paint layers with high copper content may have been explained either as converted verdigris or as a paint layer with a brown organic pigment and a copper salt as siccative, as noted above. Although our investigations are still ongoing, the initial results from the database of the Doerner Institut indicate that high copper contents are quite common in brown paint layers of paintings from the fifteenth to the seventeenth centuries, and this should be kept in mind while interpreting analytical data.

This example shows very clearly that the scientific investigation of painting materials has to be rooted deeply within the context of documentary material and sources. Therefore,



Fig. 2 Alessandro Paduano and Christoph Schwarz, *Martyrdom of St Andrew*, 1587-90, St Michael, Munich, detail.

knowledge of the primary sources is indispensable for the scientist's interpretation of analytical data.

Nevertheless, even if the evaluation of documentary sources in combination with the analysis of paint samples and the re-examination of old reports seems to focus on *Kesselbraun* consisting of copper oxides, we have to consider that, in historical terminology, one name could have been used for different materials of similar colour and consistency. With regard to *Kesselbraun*, for example, the Zedler Encyclopaedia [12] identifies the material as both a copper pigment and a brown earth, while in other sources, predominantly Italian, a strong connection to brown manganese compounds becomes obvious [9, pp. 130-131].

Because there has been no certainty about the identity of materials in the past, we should avoid simulating certainty in our current definitions. We encounter this attitude towards many pigments. In the case of our next example — mountain green — this still leads to avoidable discussions and confusions.



Fig. 3 Cross-section from Fig. 2.

MATCHING MODERN SCIENCE WITH HISTORICAL CONTEXT: MOUNTAIN GREEN

Pigments identified on paintings are designated either with historical names such as 'Naples yellow', invented names such as 'lead-tin yellow', or mineralogically respectable chemical names such as 'malachite' or 'copper acetate'. In some cases, this mix of different sets of nomenclature is misleading or does not match the historical context. Green pigments of a mineralogical origin are a good example of this.

Not so long ago, only five different green pigments were described as having been used in artefacts of the Middle Ages and the Renaissance: verdigris, malachite, green earth, copper resin-ate, and sap green. Although it would be very tempting to discuss the other four terms in the same way, 'malachite' may be the best illustration of how modern analyses and historical context do not match. On the one hand, the interpretation of analytical data draws a very simplified picture of the actual situation; on the other hand, the attempt to find a terminology for increasingly accurate analytical results suggests a higher diversity of materials than was historically offered.

Publications on the history of artists' pigments are usually illustrated with large specimens of the mineral malachite, Fig. 4. They are usually of Moroccan, Congolese, Russian, Peruvian, etc. origin. Apart from the fact that none of these mines "in the Ural mountain region [. . . or . . .] the Catanga district of Zaire" [13, p. 183] are of any historical importance in European painting, the samples are generally of gemstone quality. Analyses of these specimens usually yield quite pure $\text{CuCO}_3 \cdot \text{Cu(OH)}_2$: the first step to misunderstanding of the historical pigment. Consequently, the scientists' expectations are biased, and the analysis of green, copper-containing pigment samples becomes remarkably easy: green + copper + crystalline = malachite. This simple pattern has led to hundreds of 'notable occurrences of malachite' [13, p. 196] in Western paintings, derived from a quick inspection with an optical microscope.

Generally, the term malachite has two different aspects. From the mineralogical point of view, malachite is a basic copper carbonate. Before mineralogy was established as a scientific discipline in the late-eighteenth century, documentary sources define 'malachite' as the green gemstone only. It was used in handicrafts, for jewellery or as a pharmaceutical product, whereas the pigment was indicated by mountain green or related names. (As a possible exception we find *Malchitgrün* mentioned in a list of pigments at the court in Munich (Hoflager) dated 1598). In times before the above-mentioned rich mines in Congo, Russia, Peru or elsewhere existed, it is very unlikely that malachite of a gemstone quality would have been processed into a colourant. Accordingly, German apothecaries' *taxae* list prices for *Lapis malachites* of up to one *Florin* per *lot* (15 g) and it is never mentioned in the chapters on *colores* where we find 'mountain green' or related terms [14]. In summary, the term 'malachite' should be used as exactly as it is in both historical and modern scientific literature,



Fig. 4 Typical specimen of malachite.

and not as a general term for green copper-containing mineral pigments; this is still an unfortunate habit in sections of art technological literature.

An examination of carefully-conducted analyses of paint samples over the last twenty years shows that malachite was not the only green copper mineral which has been used as a pigment: brochantite ($\text{Cu}_4(\text{OH})_6\text{SO}_4$), posnjakite ($\text{Cu}_4(\text{OH})_6\text{SO}_4 \cdot \text{H}_2\text{O}$), atacamite ($\text{Cu}_2(\text{OH})_3\text{Cl}$) and others have been found frequently, quite often in mixtures with malachite and other minerals. All these compounds are often associated in natural deposits. For example, the investigation of green samples from the Wallerfangen pigment mine, one of the most important German mines both for azurite and green copper pigments in the sixteenth century [15], revealed four different green minerals starting with malachite, brochantite and antlerite and ending with the rather exotic strashimirite ($\text{Cu}_8(\text{AsO}_4)_4(\text{OH})_4$). For Schwaz/Tyrol more than 40 green copper minerals have been described [16]. There is no evidence that green minerals were separated mineralogically before being processed and shipped as pigments. Indeed, the analysis of an early nineteenth-century pigment sample from the collection of the Doerner Institut labelled as *Berggrün* (mountain green) (Fig. 5), indicated a mixture of atacamite, paratacamite, malachite and posnjakite, and a nineteenth-century *Berggrün* from the Academy of Fine Arts in Stuttgart (Fig. 5) consisted of brochantite with traces of atacamite, paratacamite and quartz [17].

Painters and tradesmen assessed green pigments by quality and hue. If a certain product was acceptable in these respects, they would not have cared whether it was pure malachite or a mixture of three, five or even seven compounds. Therefore, old designations can definitely not be regarded as 'obsolete' [13, p.183] because no modern terminology can represent this heterogeneous character of historical materials. For this general consideration one can find many other examples apart from mountain greens. The careful re-introduction of historical names where

suitable and where this terminology is unambiguous is very desirable.

However, correlating specific historical terms with analytical data goes along with the danger of over-interpretation, especially if designations for special qualities are used, and not general terms which include specific qualities. Again, mountain green can help as an example.

An evaluation of apothecaries' *taxae*, art technological treatises as well as mining or trading literature shows that copper-containing greens of different hues and qualities were on the European market in the fifteenth to seventeenth centuries. The general terms were *Chrysocolla nativa*, *Berggrün*, *mountain green*, *verde azzurro*, *verdetto della magna*, etc. Quite often, the origin of the products was added, of which Hungary (Neusohl/Banská Bystrica or Herrengrund/Špania Dolina) and Tyrol (Schwaz) are most frequently mentioned. A general analysis of names for green copper-containing minerals has been published elsewhere [17].

Apart from these general terms, particularly in Germany, certain qualities or hues were denominated as *Schiefergrün* (*Schifergrün*, *Schiffergrün*). In two recent papers, which have the benefit of bringing together the observation of spherulitic malachite as a pigment and the formation of green pigments as precipitation products of acidic copper-containing tunnel waters on calcite and dolomite, Heydenreich *et al.* [18–19] assumed that *Schiefergrün* was identical to the above-mentioned precipitation product. But even if a precipitated spherulitic malachite from Hungary may be traded under the name of *Schiefergrün*, it would be an over-interpretation to reduce the historical term to this, and labelling analyses of spherulitic malachite as *Schiefergrün* would be highly speculative.

Apart from the fact that it has clearly been shown in an earlier paper [17] (which was not taken into consideration by Heydenreich *et al.*) that *Berggrün* and *Schiefergrün* could even have been used as synonyms in many sources, such as German apothecaries' *taxae*, *Schiefergrün* was also a product of the Tyrolian copper mines [9, pp. 138–140]. Because the Tyrolian ores (*Fahlerz*, basically tetraedrite and tennantite), of which the mountain greens are formed as secondary products, do not produce acidic copper-containing tunnel waters (mostly vitriols), it is impossible that the Tyrolean *Schiefergrün* is precipitated spherulitic malachite.

Although the use of historical terminology for the designation of analytical results is desirable in some cases, this short example may show that the interpretation of historical terminology should be conducted very carefully. Consulting and interpreting specific documentary sources is only possible in a broader historical context. Again, because historical terms very rarely describe a specific compound or a mineralogical phase, we should avoid trying to be more exact than was possible in the past.

CONCLUSION

In recent years, the study of documentary sources in art and archaeology has made remarkable progress. Milestones include the *Cologne Database for Painting Materials and Reconstructions* by Oltrogge [20], the publication of mediaeval recipe books for painters by Clarke [21], the foundation of a working group on Art Technological Source Research (ATSR) in 2001, and the conference *European Trade in Painters' Materials to 1700* in London in 2005.

In spite of our high-performance analysis, we should never forget that our interpretations have to be rooted in a historical context. In understanding documentary sources as a tool in the scientific analysis of artists' materials, we have to consider that this tool has been sharpened remarkably in recent years. In order to match the results of both analytical and historical findings, a



Fig. 5 Nineteenth-century *Berggrün* sample. Collection of the Academy of Fine Arts, Stuttgart.

closer collaboration and cross-linking between historical and scientific disciplines is indispensable. Only crossing boundaries between disciplines will prevent systematic mistakes in the interpretation of analytical findings.

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