Hues of Art: Pigment Analysis of Unprovenanced Wall Painting Fragment from Pompeii

Tuuli Kasso

This study uses a non-destructive approach on fresco fragments from Pompeii, focusing on the artist's pigment palette used in the 1st century A.D. in Roman wall paintings. The Tukkila fragments were brought to Finland from Pompeii in 1947 in the aftermath of World War II, and the precise provenance (house, insula) is not known. However, objects, 'things', and even fragments, provide us with knowledge through the materials they were made from, linking to the choice and intention by the maker. Pigments can be identified by their chemical composition in addition to wall painting techniques. The identification of the pigments was conducted with a two-phase non-destructive method pXRF (portable X-ray Fluorescence), directing further analysis with a micro-destructive method SEM-EDX (Scanning Electron Microscopy with Energy Dispersive X-ray Spectroscopy). The results were compared to known compositions of historical pigments, tradition of fresco painting, and classic literature. The results represent the ideal Pompeian pigment palette of alkaline resistant colors (green earth, yellow ochre, cinnabar, Egyptian blue), agreeing with the craft of Roman wall painting, further supported by the literary evidence from Pliny the Elder and Vitruvius.

Introduction

The impact of the devastation caused by the catastrophic eruption of Vesuvius in A.D. 79 is most evident throughout the settlements and cities it destroyed. The most famous, Pompeii, was re-discovered in 1748, and since has greatly influenced arts and been a continuous target of studies.[1] Despite the long history of researching Pompeii, there are still significant gaps in our knowledge about daily Roman life, represented not by the literal evidence, but by the material culture left behind.[2] In the late 1990s it was resolved that no more excavations should be carried out in Pompeii, and the focus of the research would be targeted to the conservation and documentation of the areas already excavated.[3] Currently, new excavations are implemented again. The choice to pertain from excavation was influenced due to the reason that the wall paintings discovered earlier from the ancient city are under a grave risk of disappearing completely. Current estimation is that nearly 80 percent of the wall paintings excavated after 1748 have been destroyed by deterioration, caused by light exposure, weathering and environmental conditions. For instance, sulphur dioxide (SO_2) in present-day pollution cause the calcium carbonate ($CaCO_3$) in the plaster to turn to gypsum ($CaSO_4.2H_2O$), causing the flaking of the surface, further predisposing the wall paintings to decay. Documentation plays a substantial role in preserving what is still left, but much knowledge is already lost with the paintings faded beyond recognition.[4]

Despite the discrepancy of excavating Pompeii contributing to the loss of evidence, other human events such as the coming of war to Pompeii brought new findings, albeit at a great price. During World War II, the Allies bombed Pompeii on several occasions, resulting in extensive damage to the city. However, bomb pits outside the city walls exposed the location of the Villa Imperiale, a previously unknown building with rich and elaborate murals. After the war ended, Pompeii was in a chaotic state. Photographs show the crumbling *villae*, and the surrounding mayhem. Streets were scattered with pieces of wall paintings.[5] Finnish architect Iiro Tukkila visited Pompeii in 1948, and brought back with him two wall painting fragments, displayed in Figures 1 and 2. In 2006 Tukkila's widow donated the fragments to EPUH (EXPEDITO POMPEIANA UNIVERSITATIS HELSINGIENSIS), The Pompeii Project by the University of Helsinki. [6] The provenance of these fragments, which are still in excellent

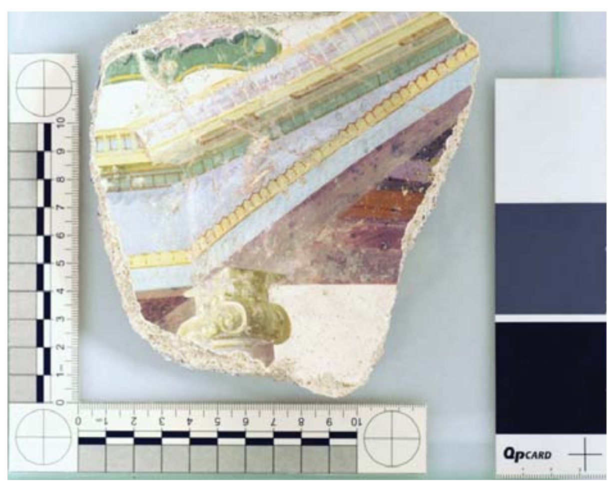

Fig. 1 and 2. The "Tukkila fresco fragments". Referredto as "Fragment A" (on the top) and "Fragment B" (on the bottom). The results of the analysis of Fragment A are presented in this study.

condition, is unknown including which *regio*, *insula*, house or room they originated. Research into the provenance has yet to be done, as such comprehensive work *in situ* has not been possible to implement.

The Significance of Pompeii for the Study of Roman Art

German scholar August Mau classified wall painting styles in Pompeii into the four Pompeian styles in 1899. This categorization of the styles does not only apply to Pompeii, but also to painting styles throughout the Roman world. Although several scholars before Mau had published their own classifications, Mau's version is the most recognized due to its coherence and clarity.[7] According to Docent Antero Tammisto, the Tukkila fresco fragments are identified as belonging to the late Third or early Fourth Style.[8] The Third Style first appeared around 15 B.C., continuing until A.D. 50 when the Fourth Style formed.[9] This gives the fragments an approximate dating from 15 B.C. to A.D. 79.

In addition to styles, the pigments used in Roman wall paintings have been a target of interest since the early days of researching Pompeii. Chemists Jean-Antoine Chaptalin and Sir Humphry David conducted extensive research in the 19th century, and in 1967 Selim August published the results of his 18 year investigation in *I colori Pompeiani*.[10] The current international groups working with multianalytical methods in Pompeii mark an entirely new era of researching the pigments in Pompeii, benefitting from the hand-held instrumentation e.g. portable Raman- and infrared spectroscopy, allowing non-destructive measurements of remains *in situ*.[11] The materials tell us about the quality of the wall paintings, their makers, commissioners and purpose of the wall paintings through the choice of materials. The 1,500 buildings at Pompeii were painted inside and outside with around 20,000 m^2 of wall paintings covering Pompeii on every level: from the advertisement of gladiatorial shows and the assortment of a wine-serving *thermopolium* to the imperial villas and public temples.[12]

Although several names of painters, such as Aristomenes of Thasos, Andron of Ephesus and Polycles mentioned by Vitruvius have survived, the majority of wall painters remain unidentified. Yet, this is not necessarily the result of poor preservation, rather it is more likely that many paintings were unsigned as most artists in ancient Rome were anonymous, and considered to be low-status workers with an unfavorable position in the society.[13] Working together, painters' groups formed and worked as local workshops or mobile groups. The styles of these groups can be identified based on the conservative repertory of Roman art. Groups can be distinguished from one another by their technique, but this is only just a new area in the study of Roman wall paintings.[14]

Questions relating to perceptions and value of Roman wall paintings in the Roman world have been raised by modern scholars. Umberto Pappalardo mentions a relationship between the decorations and homeowners. According to Pappalardo, this bond must have been more profound than expected, as fixed art, such as mosaics and wall paintings were hard to move compared to the modern-day pictures on our walls. Nowadays we experience the Roman wall paintings as art quite decidedly. The perception of art differed in the ancient period in a way that we, who have developed our sense of art and aesthetics through the 19th century romanticism, might never truly grasp.[15] What we need to attain, is the Roman thought on the difference between mere decoration and art – perhaps this separation is dispensable. Despite many authors claim that *l'art pour l'art* or aestheticism were more or less unfamiliar concepts for the Romans, a so-called aesthetic approach must have been present.[16]

The Technique of Roman Wall Paintings

Artists employed various techniques of painting that were specific to the types of materials used, the scale of the painting, and the level of detail required. Most paintings began *a fresco*, painted with high alkaline-tolerant pigments mixed in water on wet plaster. From this, details and the finishing touches were added *a secco*, on dry plaster. This was done using binding agents such as saponified lime, animal or plant-based glues, with low alkaline-tolerant pigments.[17] Therefore, the term *fresco* should be carefully used, as technically it is a false expression of the entity of Roman wall paintings. In addition to technical matters, economic and social standards influenced the artist's palette. Certainly, the taste of the artist could not map out the choosing of the materials completely, since those who commissioned the paintings must have been well aware of the difference between using rare and precious materials over easily accessed ones as common earth pigments widely available, and cheap.

Concerning the technique of Roman wall painting, there are two matters about the technique of frescoes that must be addressed that affected the choice of materials as well. As the lime plaster is highly alkaline, alkaline-tolerant pigments were preferred. Ancient authors, such as Vitruvius in his *De architectura* (25 B.C.) and Pliny the Elder in *Naturalis Historia* (1st century A.D.), describe the fresco technique quite meticulously, and discuss the best pigments to be used in frescoes. The poor durability of cinnabar in frescoes when exposed to sunlight and moonlight was known in ancient Rome, the red color turning dark over time.[18] Vitruvius comments on treating cinnabar-containing wall paintings with Punic wax, made by bleaching beeswax in the sun. This helped to preserve the color; the dry painting would be brushed with hot Punic wax, then smoothed down with a hot tool. A final finish was made with polishing the surface with linen cloths.[19]

The second issue which affects the fresco technique is the dryness level of the plaster. It is fundamental that the painting is conducted on the plaster at the optimum state. If too wet, the brush paws the surface. If too dry, the pigments will not become fixed to the plaster. The pigment particles are only sealed when calcium carbonate from the lime travels to the surface with

Fig. 3. South wall and part of the eastern wall of the oecus in Casa dei pittori al lavoro. On the left the centrepiece of the wall is unfinished, with a lot of the background is missing, including the socle of the walls.

evaporating water, forming a layer of crystallized calcium carbonate on top of the paint.[20]

Due to these processes the painters had to prepare timing for the work depending on its scale, all together the day's work, *giornata*. The wet plaster *intonaco*, was applied over a rougher grounding layer, *arriccio*, only a controllable area at a time. First the *parietarii* prepared the background, and the work was conducted piece by piece culminating in the centrepiece, which was painted by the *imaginarii*, figure painters. Of course, the groups had members to perform the less artistic tasks, such as grinding the lime. The designer and master painter *redemptor*, was in charge, and the most appreciated member of the painting group, but not even the master signed the works that had been so systematically made.[21] It is possible that the technique itself had an influence on the style of the wall paintings, as the tripartite horizontal division remained as a dominant feature in the wall paintings from the First to the Fourth Style.[22] Archaeological evidence supports the manufacturing of the frescoes in addition to the exact roles of the painter groups. *Casa dei pittori al lavoro* (fig. 3), or The House of the Painters was given its name from the unfinished frescoes and working tools found in the *oecus*, the main hall.

The tradition of wall painting and the ancient literature give us some understanding of the pigments that may have been chosen for the frescoes, but the precise identification of the historical pigments can be made through the analysis of their chemical composition. Pigments have significant differences in their capability to cover and dye, to tolerate light, to react and to absorb oil, not forgetting the differences in particle size, density and toxicity. Density of the color has much to do with the elements of the pigments, for instance, lead (Pb) has a high density and is therefore highly opaque, for which reason it was favored through history.[23] Considering the fact that the choice of materials could vary greatly depending on the region, time and possibly owing to the artist as well, distinct study of the pigments is important. Ulla Knuutinen also points out that accurate analysis is needed, as the terminology and nomenclature of pigments can be unclear due to the variety of the chemical composition of a color carrying the same name, for example Pompeian red (figs. 4 and 5). Cinnabar (mercury (II) sulfide, HgS), minium/red lead (Pb_3O_4) and iron (III) oxides such as hematite and red ochre have all been called Pompeian red but their consistency is dissimilar to one another.[24]

Fig. 4. Detail of a psychedelic female figure on a "Pompeian red" background from the east wall in the triclinium in Casa dei casti amanti.

Fig. 5. Modern day industrial red pigment sold as Pompeian red.

The majority of historical pigments are of inorganic origin and have metal elements in their composition. However, some organic pigments can also have metal elements in the form of salts, including magnesium (Mg) and calcium (Ca). In some cases, a specific key element only appears in certain pigments or produces a certain color.[25] In spite of this, identifying the historical pigments is never simple, since the object studied may have been contaminated, transformed due to aging and weathering or the original paint can consist of a mixture of several pigments that complicate the study substantively. Besides the chemical composition, pigments form crystal structures that are identifiable as well. Typically, iron oxides have an octahedron structure, but in contrast cinnabar form trigonal crystals.[26]

Pigment Analysis of Tukkila Fragment A: Two-phase Approach

The pigments and painting technique of the Tukkila fragments were studied using non- or micro-destructive methods due to the high historical value of the fragments, and the results of Fragment A are presented in this article. The sequence of the paint layers was examined from the surface with optical microscopy, as making cross-sections was found to be too damaging for the fragments. The spectroscopic study of the pigments was conducted with a multianalytical two-phase method. Starting with a non-invasive method, the fresco fragments were first studied to determine the need for additional research, and then continuing using a micro-invasive method to finalize the analysis. Firstly, the fresco fragments were examined with XRF (X-ray fluorescence), resulting in mostly very indefinable outcome of the pigments. Secondly, based on the initial results of XRF, the study was further carried out using a more accurate method, in this case the SEM-EDX (scanning electron microscopy with energy dispersive X-ray spectroscopy), which would provide more detailed information about the chemical compounds of the pigments.

The XRF analysis was conducted with a Bruker S1 Titan portable hand-held energy dispersive X-ray fluorescence spectrometry (pXRF), with an 8 mm spot size, at the Department of Archaeology of the University of Helsinki. Using the calibration application Geochem and GeoGhem Trace method, the sample areas were measured with this mode accordingly: Phase 1 (heavy elements): 45 kV/8,9 uA (with TiAl-filter), Phase 2 (light elements): 15 kV/30 uA (no filter). The measurement time for each phase was 60 seconds, altogether 120 seconds for each spot. Only key elements affiliated with pigments are reported in this study, and the lowest levels under 1% or under limit of detection have been omitted.

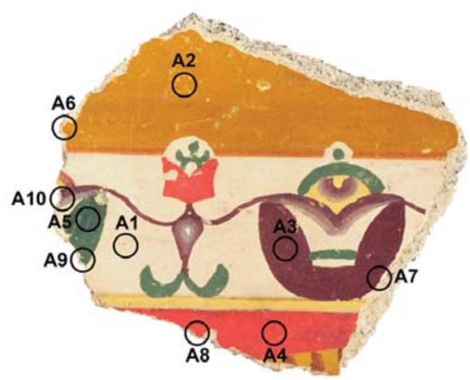

Fig. 6. Measurements and sampling areas from Fragment A.

pXRF-measurements:

A1 White

A2 Yellow

A3 Purple

A4 Red

A5 Green

SEM-EDX sampling areas:

A6 Yellow

A7 Purple

A8 Red

A9 Green

A10 Blue

Analyses with the SEM-EDX were made in the Nanomicroscopy Center at Aalto University with Dr. Krista Vajanto, using analytical high-resolution SEM, JEOL JSM-7500FA. Only the edges of the fragments and already damaged areas were chosen for the samples, sampling carefully 1-2 mm^2 areas for the samples. In this case, the samples were prepared on aluminum stubs with double-sided carbon tape, and no coating was used. Measurements were taken with COMPO mode, with the acceleration voltage 15 kV, emission current 20 kV, and probe current 20 kV in 8 mm working distance due to the backscattering detector. Otherwise an SEI detector was used for the scanning. Each of the micro samples was scanned thoroughly, choosing particles for measurement that had most the appearance of a pigment particle. The results were also compared with ancient Roman literature and related research.

During the inspection of Fragment A with the optical microscope, some areas of the fragment turned out to be surprisingly interesting in contrast to its simple style. Figure 7 shows the presence of some very large, square shaped blue crystals within the violet and blue gradient areas. This would indicate already that the blue pigment here might be Egyptian blue, as it generally has a very well-known cubical crystal form. Figure 8 depicts an interesting phenomenon on the border of yellow and red. On the areas where the top layer of the red paint layer has vanished, some blackening process is clearly noticeable. This might be due to the aging process of cinnabar, which results in a greyish or black layer. Cinnabar is a precious pigment, not used for secondary paintings. To conclude, Fragment A seems to be painted *a fresco*, with remarkably thick and solid colors. The painting order can be observed as well with the microscope: top and bottom strips were prepared first, followed by the abstract floral pattern in the middle.

The results of the XRF-measurements are displayed in Table 1. The white color of the fresco is most likely just lime white, a calcium carbonate ($CaCO_3$) with some magnesium present in this sample A1 White. It might be *paraetonium white*, which was a favored white color for frescoes.[27] The strong presence of mercury (Hg) in A4 Red indicates cinnabar (HgS) as the red pigment.[28] As a preliminary study, no exact results were assumed with the pXRF, knowing the directive quality of this method. A2 Yellow, A3 Purple and A5 Green might be mixtures and/or of earth colors of high iron and silicate content, but no specific pigments could be identified based on this. Hence it was no surprise that the results pointed directly to the need for further analysis. As the blue details of the

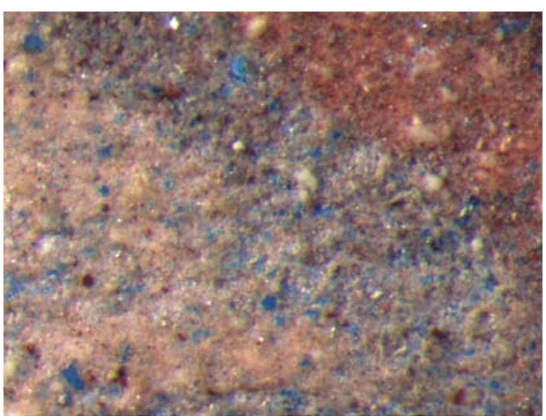

Fig. 7. Blue crystals on Fragment A.

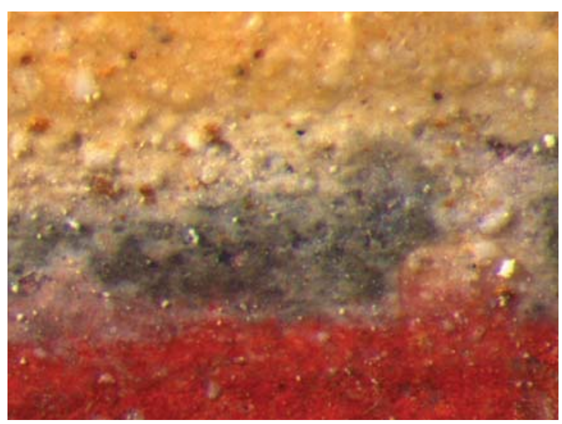

Fig. 8. Darkening of cinnabar in Fragment A.

Sample ID	%MgO	$\%Al_2O_3$	%CaO	$\%SiO_2$	%Fe	%Cu	$\%K_2O$	$\%P_205$	%Hg
A1 White	9.3323	2.9306	56.0835	1.5561					
A2 Yellow	6.178	1.1733	41.6878		13.405				
A3 Purple	6.1246	3.0135	40.178	16.111	8.2069				
A4 Red	11.2077	2.9884	15.0837	4.2694		1.385		1.8032	19.393
A5 Green	5.8612	1.1658	42.8942	16.8111	3.709				

Table 1. The results from Fragment A with pXRF with key elements.

fresco were too small to be studied due to the small measurement area of the pXRF, the blue color was added for SEM-EDX analysis to argue if Egyptian blue was *de facto* used in this fragment. Displayed in Table 2, examination of the pigments with SEM-EDX gave fairly comparative results to form an understanding of the pigments used in Fragment A.

With the combination of pXRF and SEM-EDX the pigment palette used in Fragment A could be identified. Painting *a fresco* and thick mixtures of paint with good quality pigments, the artist first used lime white before painting the top yellow strip with yellow ochre and the bottom strip with precious cinnabar. Green earth, lime white, cinnabar and Egyptian blue were used for the abstract floral pattern. The violet color, was accomplished by mixing a variety of pigments, in this case cinnabar with Egyptian blue and possibly darkened with black iron oxide and/or another black pigment. The presence of carbon (C) does indicate a carbon-based black pigment, and the prominent content of iron (8,2069 %) might mark the presence of an iron oxide pigment, such as red ochre (Fe2O3). All the colors mentioned are commonly known pigments used in Pompeii.[29] If further analyses can be made, studying the light-yellow color used on a few places in the fresco could be interesting, as it has a cooler tone compared to the rich yellow ochre used so generously.

Conclusion

Fragment A is a 'textbook' example of the Pompeian pigment palette used in the 1st century A.D., painted using good quality pigments common to the area and period. Though simple in style, Fragment A might be from a border lining to a more elaborate centerpiece in a room

Sample ID	Key elements	Pigment(s)
A6 Yellow	Fe, Si, O	Yellow ochre $(Fe_2O3)\cdot nH_2O$
A7 Purple	Hg, Si, Fe, Mg, Cu, O	Mixture (?) including:
		Cinnabar (HgS)
		Egyptian blue $(CaOxCuOx_4SiO_2)$
		Black iron oxide $(FeOxFe_2O_3)$
		Charcoal black (C), Graphite (C), Lamp black (C)
A8 Red	Hg	Cinnabar (HgS)
A4 Green	Fe, Si, K, Mg, Al	Green earth (Fe-Mg-Al-K-hydrosilicate)
A5 Blue	Cu, Si, O	Egyptian blue $(CaOxCuOx_4SiO_2)$

Table 2. Summary of the key elements detected with SEM-EDX and results.

important to residents, as cinnabar was an expensive pigment compared to other reds.[30] As such, the pigment analysis gives us information beyond the image and style. However, studying historical pigments – or even trying to identify them on a primary level – is definitely not in any case a straightforward or an easy task. Results depend greatly on the samples, that can include contamination from past conservation and restoration treatments. In addition, scientist should collaborate with art historians and archaeologists for a coherent picture of the human past, ever increasing the current interdisciplinary approach. Thorough knowledge about the subject and knowing the instrumentation are both matters of great importance, as no measurement is a direct result, but subject to interpretation. Nevertheless, objects can tell us more than is visible to the naked eye through their materials. With the study of materials, we can further understand the craftsmen and the Pompeian pigment palette, casting light on the ancient art and people who conducted it.

Acknowledgements

"There are these Pompeian fragments in a drawer that we do not know much about..." I want to thank the director of EPUH, Docent Antero Tammisto for allowing me to work with the Tukkila fragments for my research, and the ever-lasting spark for Pompeii. Using the state-of-the-art SEM-EDX methods and instrumentation was only possible with the generous opportunity given by Professor Janne Ruokolainen at the Nanomicroscopy Center in Aalto Unviersity, and the guidance of Krista Vajanto, PhD. I want to thank Elisabeth Holmqvist, PhD for letting me work with the pXRF at the University of Helsinki. I am grateful for the support during my work and the comments given on the early version of the manuscript by friends and colleagues. Without it, the Pompeian pigment palette in this case could have stayed in the drawer for all time.

Endnotes:

1. Foss 2007, 28.
2. Savunen 1998, 19.
3. Mannerheimo 2014, 3.
4. Castro et al. 2012, 9; Hornytzkyj et al. 2007, 7; 12 & 20; Tammisto 2009, 28.
5. García and García 2006, 19-21 & 26-29.
6. Castrén and Tammisto 2016 pers. comm.
7. Foss 2007, 35; Pappalardo 2009, 9.
8. Tammisto 2016. pers. comm.
9. Ling 1991, 52; Pappalardo 2009, 10.
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11. Castro et al. 2012, 9; Maguregui et al. 2010, 1400-1401.
12. Dunkle 2008, 67; Ling 1991, 1; Mannerheimo 2014, 10.
13. Gassiot-Talabot 1971, 13 & 183-184; Hardwick et al. 2000, 117-118; Vitruvius Book III: Introduction, 70.
14. Hardwick et al. 2000, 118.
15. Pappalardo 2009: 7; Kajava et al. 2009, 348-349.
16. Kajava et al. 2009, 348.
17. Fuga 2006, 92-95; Cuní 2016, 1.
18. Mannerheimo 2014, 21; Knuutinen & Mannerheimo 2009, 187-188; Pappalardo 2009, 9; Vitruvius Book VII: Chapter X: 218 & Chapter XIV: 221.
19. Nöller 2015, 46-47.
20. Bóna 2012.
21. Harwick et al. 2000, 117; Pappalardo 2009, 9.
22. Tammisto 1998, 98.
23. Knuutinen 1996, 1 & 9.
24. Knuutinen and Mannerheimo 2006; Knuutinen and Mannerheimo 2009, 40.
25. Knuutinen and Mannerheimo 2006.
26. Berrie et al. 2007, 58-59; Knuutinen and Mannerheimo 2006.
27. Eastaugh et.al. 2008, 296; Pliny XXXV: xviii.)
28. Eastaugh et. al. 2008, 239 & 105; Knuutinen 1996, 35.)
29. Knuutinen and Mannerheimo 2006; Mannerheimo 2014, 23; Pliny XXXV: xxv.)
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