



Reading the materiality of the Oxyrhynchus Papyri: non-invasive analyses to reveal scribal choices

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Abstract

This paper presents the first ever material characterisation of inks used in 23 documentary papyri excavated in Oxyrhynchus, about 160 km south-east of Cairo. These papyri date to between the fourth and seventh centuries CE, and are nowadays preserved at the Art, Archaeology and Ancient World Library in Oxford. The material analyses were carried out with in situ analytical methods, namely infrared reflectography (IRR) and X-ray fluorescence (XRF). Moreover, the data obtained with XRF on the papyrus support were further explored with principal component analysis (PCA). To our knowledge, this study represents the first instance in which PCA was applied to a dataset obtained from ancient papyri with portable, non-invasive analytical equipment. The differences in the material composition highlighted suggest the papyrus used for these documents proceeded from different sources. Furthermore, the investigation of the writing inks confirmed a trend we highlighted during previous material studies of manuscripts from the Byzantine period: carbon-based ink is predominantly used for documentary texts, while iron-gall ink is for literary texts. Finally, the peculiar case of a document bearing the proceedings from the court of Alexandria suggests the existence, in this area, of specific technical and scribal traditions that have not been pointed out before.

Keywords Oxyrhynchus papyri · Material analyses · Writing inks · Carbon ink · Mixed ink

Introduction

In the past few years, humanities and heritage science scholars have frequently addressed the study of ancient Mediterranean writing ink manufacture and developed this field of research remarkably. Information on ingredients and processes has often been recollected through meticulous archival research on surviving treatises and recipes (Zerdoun Bat-Yehuda 1983; Daccache and Desreumaux 2015; Raggetti 2016; Christiansen 2017; Rabin 2017; Colini 2018; 2021). Furthermore, the investigation of these sources has been complemented by an increasing number of material studies that achieved major breakthroughs in the characterisation of ancient inks, focusing largely on Egyptian manuscripts (Brun et al. 2016; Chiappe et al. 2019; Arlt et al. 2019; Christiansen et al. 2020; Maltomini

et al. 2021; Autran et al. 2021; Pereira et al. 2021; Nodar et al. 2022; Cohen 2022). The latest studies in this area consistently reported the use of carbon ink on manuscripts dated between the Pharaonic period and the early Middle Ages (ca. 600–700 CE). In some cases, material studies on original manuscripts revealed that Egyptian inks contained other constituents together with carbon, as evidenced by the presence of different transition metals (Fe, Cu, and Pb) (Chiappe et al. 2019; Arlt et al. 2019; Christiansen et al. 2020; Maltomini et al. 2021; Autran et al. 2021; Pereira et al. 2021) and polyphenols (Nehring et al. 2021; Ghigo and Albarrán Martínez 2021).¹ The presence of iron (with or without other metals found in vitriol²) and/or polyphenols in an ink containing carbon is consistent with existent recipes of mixed inks obtained by blending carbon and iron-gall ink or carbon and vegetal extracts (Zerdoun Bat-Yehuda 1983; Colini 2018a, b). However, the origin of transition metals found in carbon inks with no iron and no polyphenols remains partly unclear.

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¹ A solution of different polyphenolic molecules extracted from vegetal matter is described as “plant ink” in the treatise by Monique Zerdoun Bat-Yehouda.

² A mixture of sulphates containing primarily iron, copper, and zinc used to make iron-gall ink.

Synchrotron-based analyses on small collections of papyri suggested that copper might have been a by-product found in soot proceeding from metallurgy production (Christiansen et al. 2017), while lead might have been added to the inks as a drying agent (Christiansen et al. 2020). Furthermore, material analysis of several parchment and papyrus manuscripts reported several examples of iron-gall ink, with the earliest instances dating to the fourth and fifth centuries (Ghigo 2020, 52–105; Rabin and Krutzsch 2019). The papyrus V of Leiden (third century CE) reports a recipe for iron-gall ink that bears certain similarities with later medieval sources describing its preparation (Ghigo et al. 2020; Preisendanz 1931, 83).

The transition that saw the slow replacement of carbon-based inks with iron-gall inks, very popular in Egypt and Europe during the late Middle Ages (Kolar and Strlič 2006; Ghigo 2020; Cohen 2022), remains somewhat unclear. A recent study suggested that copper-containing carbon inks might have represented the link towards iron-gall inks (Nehring et al. 2021). Regardless, it is evident that both these types of inks were customary in Egypt for several centuries. Recently, different studies on substantial collections of manuscripts from various areas of Egypt pointed out an unexpected correlation between the type of text and type of ink on manuscripts dated between the fourth and the eighth century CE. Such investigations showed that carbon-based inks were primarily used for documentary texts (mainly commercial documents such as orders of payment and receipts, but also private letters and administrative documents), while iron-gall inks were for literary texts, including Christian literature (Ghigo 2020; Ghigo and Albarrán Martínez 2021; Nodar et al. 2022). Although the reasons behind such trends remain unclear, they suggest specific choices or traditions linked to the materials used in manuscripts production. Similarly, an ongoing study on the width of *kollemata* (papyrus sheets that were joined together to form a roll) of over a thousand manuscripts from Oxyrhynchus (first-seventh centuries CE) recently pointed out differences that suggest intentional choices of specific types of support depending on the genre, typology, and production stage of a manuscript.³

The present contribution explores this topic further by presenting the material characterisation of 23 documentary papyri of different types, ranging between private letters and court proceedings. It aims to observe whether the level of formality of these documents correlates to compositional differences in the writing support and ink that might reflect specific scribal traditions or intentional choices of the scribes. The papyri investigated are preserved at the Art, Archaeology and Ancient World Library of the University of Oxford and date to between the fourth and seventh

centuries CE. They were excavated at Oxyrhynchus during the campaigns conducted by B. P. Grenfell and A. S. Hunt around the turn of the twentieth century and were found in the rubbish mounds that surrounded the city, attesting to multiple aspects of everyday life of its citizens. Although they proceed from Oxyrhynchus, their origin might not be in all cases the same: official documents, private letters, or even books containing literary works may have been produced elsewhere and ended up in Oxyrhynchus with their recipients or owners.

The most formal documents selected (P.Oxy.1876, 1878, 1879, 1880, 1942, 2665, 3637, 4381) include letters and reports by public officers and court proceedings that were, in all likelihood, copies from the proceedings kept at the provincial archive (Pedone 2020).⁴ They represent a kind of official, carefully executed document where a high degree of formality is sought. A high degree of formality might be expected as well for the papyri containing official nominations (P.Oxy.3985, 3987). Similarly, a certain degree of formal care is essential for the petitions addressed by private citizens to the administration (P.Oxy.1886, 2268, 3584, 3585, 3586, 4614) or letters addressed to the administration by citizens in a more or less private manner (P.Oxy. 1842, 1869). A less formal group of documents includes letters by and to several figures of the administration (P.Oxy.1830, 1831, 1840). Finally, some documents definitely not belonging to the sphere of the administration have been selected as counter-samples (P.Oxy.4620, 4629). The list of papyri studied is reported in Table 1, along with the analytical results.

Results and discussion

Support

XRF was performed on 20 out of 23 papyri since P.Oxy.1886, 2665, and 4620 were not available for elemental analyses. The XRF data collected on the papyrus support were further explored using PCA which highlighted a correlation between the differences in the elemental composition described by the first three principal components and the place of origin of the papyri studied.

Figure 1 shows the plot of PC1 versus PC2. The measurements acquired on the supporting glass used to manoeuvre the papyri, represented as dots, fall in a different area of the chart, indicating that its elemental composition does

³ Personal communication, Serena Causo (Ghent University), September 2022.

⁴ Also in Pedone, “The contribution of the CLTP to the study of Roman justice” (forthcoming), where consistent evidence is given that copies of court proceedings were issued from the provincial chancery registers and it is convincingly argued that most court proceedings that have reached are in fact such copies.

Table 1 List of the manuscripts studied and results of IRR and XRF analysis

	Degree of formality*	Date	Type of document	IRR	XRF — elements in the ink**
P.Oxy.2665	5	305–306 CE	Report of property registration	Semi-transparent	N.a
P.Oxy.4381 (ink 1)	5	375 CE	Court proceedings (trial in Alexandria)	Semi-transparent	S
P.Oxy.4381 (ink 2)	5	375 CE	Court proceedings (trial in Alexandria)	Opaque	S, Ca
P.Oxy.4381 (ink 3)	5	375 CE	Court proceedings (trial in Alexandria)	Semi-transparent	N.a
P.Oxy.1880	5	427 CE	Abandonment of legal proceedings	Opaque	None
P.Oxy.1879	5	434 CE	Court proceedings	Opaque	S
P.Oxy.1878	5	461 CE	Court proceedings	Opaque	None
P.Oxy.1876	5	480 CE	Court proceedings	Opaque	S, Ti, Fe, Pb
P.Oxy.1942	5	500–599 CE	Letter by a public officer (praeses)	Opaque	S, Fe, Pb
P.Oxy.3637	5	623 CE	Letter by a public officer	Semi-transparent	S, K, Ca, Mn, Fe
P.Oxy.3985	4	473 CE	Appointment	Opaque	S, Ca, Fe, Cu
P.Oxy.3987	4	532 CE	Appointment	Opaque	S, Cu, Pb
P.Oxy.1886	3	472? CE	Petition to a defensor	Opaque	N.a
P.Oxy.3585	3	425–475 CE	Petition to Flavius Strategius (Apions archive)	Opaque	S, Ti, Fe
P.Oxy.2268	3	475–499 CE	Petition to a defensor (written in the Herakleopolites)	Opaque	S, Pb
P.Oxy.4614	3	475–499 CE	Petitions to the riparius (Apions archive)	Opaque	None
P.Oxy.3584	3	400–499 CE	Petition (petitioner from the village of Tampemu) (Apions archive)	Opaque	Fe
P.Oxy.3586	3	400–499 CE	Petition to Flavius Strategius (Apions archive)	Opaque	Fe, Cu
P.Oxy.1842	3	500–599 CE	Letter by a private citizen	Opaque	S, K, Ca, Ti, Pb
P.Oxy.1869	3	500–699 CE	Letter by a public officer	Semi-transparent	S
P.Oxy.1831	2	455–499 CE	Letter by a headman (written in Tholthis)	Opaque	Ti
P.Oxy.1830	2	500–599 CE	Letter by a public officer (written in Takona)	Opaque	S, K, Ti, Fe
P.Oxy.1840	2	500–599 CE	Letter by a public officer	Opaque	S, Pb
P.Oxy.4620	1	475–550 CE	List of offers	Opaque	N.a
P.Oxy.4629	1	500–699 CE	Letter by a private citizen	Opaque	S

*Rated from 2 to 5, with 2 being informal and 5 being highly formal. 1 stands for documents not belonging to the administration

**The elemental content of the ink was established by comparing XRF spectra measured on different spots of ink with those on neighbouring spots of blank papyrus. Only elements whose measured XRF intensity was higher in the ink in the majority of regions investigated are reported in the table. N.a. indicates that no elemental information is available

Fig. 1 PCA score plot of XRF raw peak areas showing PC1 vs PC2, with percentages of variance explained by each component. The plot highlights four papyri whose material composition is different than the rest of the collection. The overlapping loading plot (represented by arrows with element tags) shows the contribution of different elements to each principal component, highlighting material differences across the collection

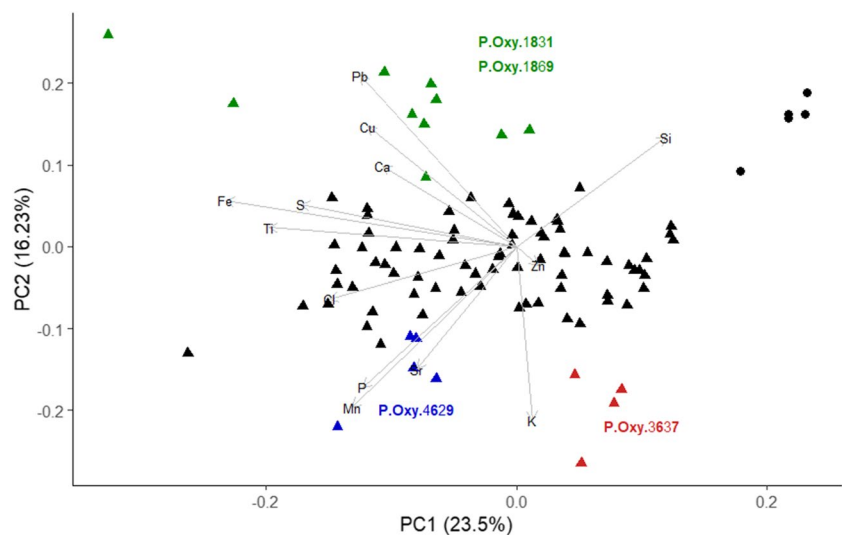
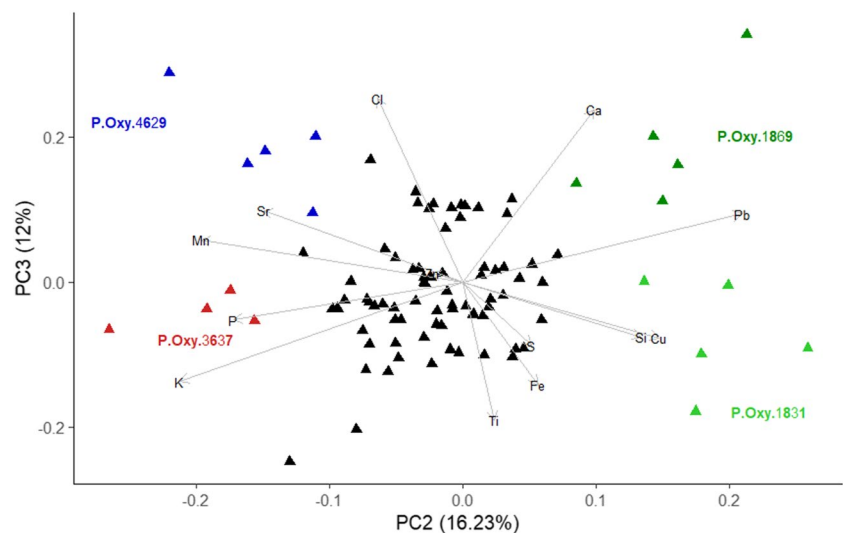


Fig. 2 PCA score plot of XRF raw peak areas showing PC2 vs PC3, with percentages of variance explained by each component. The plot highlights four papyri whose material composition is different than the rest of the collection. The overlapping loading plot (represented by arrows with element tags) shows the contribution of different elements to each principal component, highlighting material differences across the collection



not interfere with the evaluation of the papyri's elemental composition.

Most papyri have a very similar material composition across the range of elements detected with XRF, forming a cluster in the centre of the chart. However, three manuscripts fall out of this area: P.Oxy. 3637 (red), 1831, and 1869 (green). The PCA loadings reveal that XRF analysis on P.Oxy.3637 measured higher intensities of potassium, while for P.Oxy.1831 and 1869, higher intensities of copper and lead were measured. Furthermore, the chart shows that the XRF measurements of P.Oxy.4629 tend to form a separate cluster (blue), which becomes clearly distinguished when plotting PC2 vs PC3 (Fig. 2).

The PCA loadings on this second plot highlight that higher intensities of strontium were measured for P.Oxy.4629. Furthermore, the plot confirms that P.Oxy.3637 is separated from the main group of papyri, as they are P.Oxy.1831 and 1869. In addition, it highlights a compositional difference in these two last manuscripts, the former showing higher intensities for copper and the latter for lead. Normalisation to calcium⁵ was used to observe relative elements concentration for potassium, copper, lead, and strontium and reduce the effect of the papyrus thickness on XRF measurements. Figure 3 shows that the values of K/Ca for P.Oxy.3637 are much higher than those measured

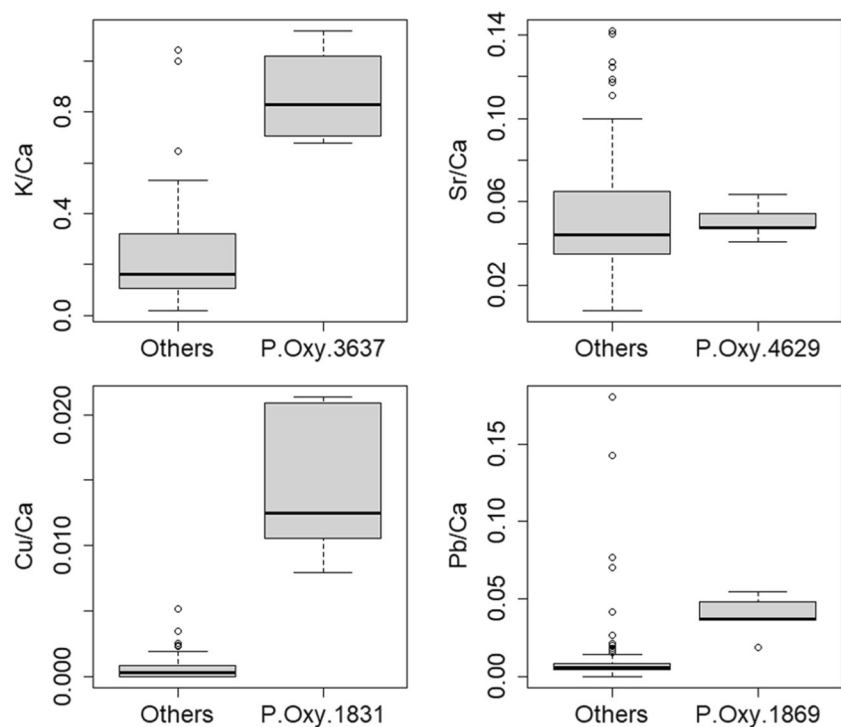
across the rest of the collection, as are the values of Cu/Ca for P.Oxy.1831 and Pb/Ca for P.Oxy.1869. This indicates that the discrepancy highlighted by PCA is due to a compositional difference in the papyrus paper. Copper and lead found in P.Oxy.1831 and 1869, respectively, are not naturally contained in the papyrus plant and most likely result from contamination during manufacturing or storage processes (see files S1 and S2 for representative spectra of the papyrus paper of P.Oxy.1831 and 1869). By contrast, potassium is naturally occurring in the papyrus plant and its abundance in P.Oxy. 3637 might relate to the composition of the plant originally used to manufacture the paper used for this document, which in turn could have reflected specific environmental conditions. Figure 3 shows that in contrast to the values of K/Ca, Cu/Ca, and Pb/Ca, those of Sr/Ca for P.Oxy.4629 are well within the range of those measured across the collection. Therefore, the difference in the intensity of strontium measured by XRF and highlighted by PCA relates, in this case, to a manufacturing procedure that resulted in a thicker sheet of papyrus.

Interestingly, the three papyri showing compositional differences bear letters written by public officers that almost certainly originated outside of Oxyrhynchus. P.Oxy. 1869 and 3637 bear textual evidence they were written elsewhere,⁶ and the heading P.Oxy. 1831 indicates Tholthis (a village not far from the capital) as the place of origin.

⁵ Previous research showed that reference or fundamental parameter methods traditionally used for the quantification of XRF measurements do not deliver satisfactory results on thin-layer systems such as papyrus, parchment, or paper. While a model was developed in the past for the quantification of XRF analysis of inks on paper (Hahn et al. 2004; Malzer, Hahn, and Kanngießner 2004), the intrinsic heterogeneity of parchment and papyrus precludes the possibility to develop standardised models for elemental quantification based on XRF measurements.

⁶ In P.Oxy. 1869, the sender addresses the recipient (presumably staying at Oxyrhynchus, where the letter was found) saying “for you discoursed at length in his praise when staying here” (translation by editors), thus suggesting he is writing from outside Oxyrhynchus. Similarly, P.Oxy. 3637 is an official communication to Marinus (the officer in charge of tax collection for the Persian authorities in the Oxyrhynchus area). Specific adverbs of place suggest the letter was written from outside Oxyrhynchus.

Fig. 3 Box and whisker plot showing the normalisation to calcium of potassium, strontium, copper, and lead measured across the collection



Ink

Infrared reflectography showed that the inks investigated absorb infrared radiation above 1510 nm, remaining visible in the infrared images obtained. This indicates that charcoal or soot was used in their making. In fact, carbon is characterised by a high (and constant) absorbance rate above 1510 nm, where it remains completely opaque.⁷ By contrast, polyphenols stop absorbing radiation above 900 nm, and iron-gall ink stops absorbing between 1200 and 1450 nm, depending on its preservation (Mrusek et al. 1995; Rabin and Binetti 2014; Aceto et al. 2014). Both these types of ink would appear transparent above 1510 nm.

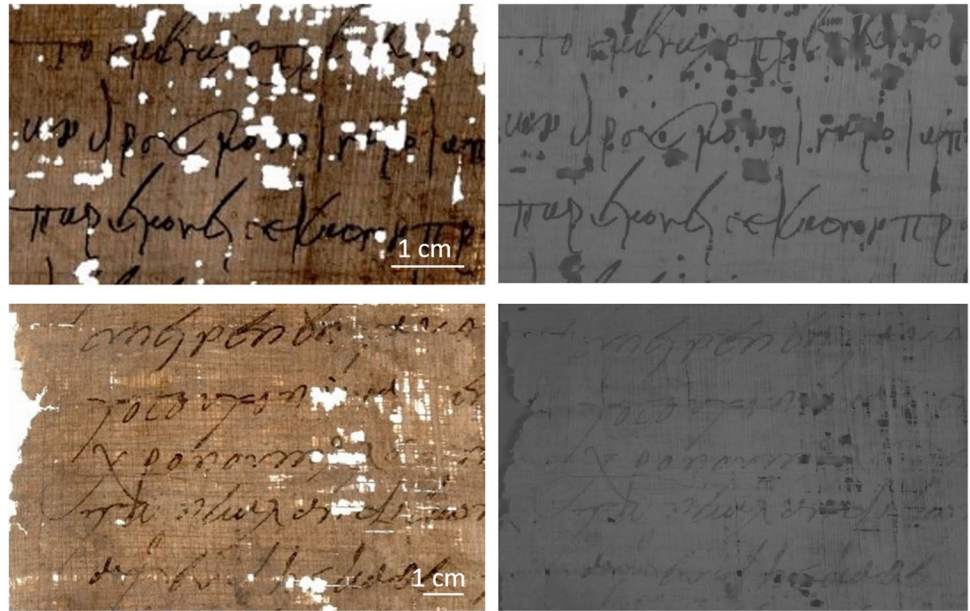
While most of the inks investigated remained completely opaque above 1510 nm, the inks on P.Oxy.1869, 2665, 3637, and 4381 (ink 1) showed a change in opacity. This indicates that soot or charcoal was initially combined with other ingredients, thus resulting in the so-called mixed inks. Figure 4 compares the visible and infrared images of the inks on P.Oxy.2268 and P.Oxy.1869. The ink on P.Oxy.2268 maintains its opacity above 1510 nm, while the ink on P.Oxy.1869 changes it quite dramatically, indicating that carbon is not the only ingredient in this writing medium.

⁷ For a reflectance spectrum of carbon black, see: “Lamp Black” in Cultural Heritage Science Open Source: <https://chsopensource.org/lamp-black-k-47250/>.

XRF analysis on the mixed inks from P.Oxy.1869 and 4381 (ink 1) revealed they did not contain any iron. Therefore, carbon must have been mixed with polyphenols extracted from vegetal matter (such as oak galls) rather than with iron-gall ink. By contrast, Fig. 5 shows that iron was found in the inked areas on P.Oxy.3637. The comparison of XRF spectra collected on an inked area and adjacent blank papyrus in Region 1 shows that sulphur, iron, and manganese peaks are higher in the inked area than in the papyrus. Furthermore, the bar chart shows that the XRF intensity (raw peak areas) of these elements is higher in the ink for most areas investigated. Sulphur, iron, and manganese are commonly found in vitriol and have been reported in Egyptian iron-gall inks from the same period (Ghigo 2020, 119–26). Potassium and calcium, usually associated with the binder or the gallnuts used to produce iron-gall ink, are also consistently higher in the inked areas.

For comparison, Fig. 6 shows XRF results obtained on P.Oxy.1869, written with a mixed ink containing carbon and vegetal extracts. The comparison of XRF spectra collected in Region 5 shows that the one measured in the inked area almost entirely overlaps that of blank papyrus. Furthermore, the bar chart shows that while the intensity of sulphur is consistently higher in the ink, the intensity of iron and manganese is more often higher in the papyrus. Since the investigated regions were chosen where the ink trace looked thicker, the fact that iron and manganese are higher in the inked areas in a minority of cases suggests

Fig. 4 Visible and infra-red images (spectral range 1510–1700 nm) of P.Oxy. 2268 (top) and 1869 (bottom)



that these elements are not contained in the ink and highlights the intrinsic compositional heterogeneity of ancient papyrus paper that has been previously described (Ghigo et al. 2020).

XRF analysis on the inks that remained completely opaque above 1510 nm revealed, in most cases, that the ink

contained different combinations of transition metals such as titanium, manganese, iron, copper, and lead. Although previous studies suggested that a specific element could hint at the provenance of the soot or the intentional use of drying agents (Christiansen et al. 2017; 2020), the presence of a combination of different transition elements in a carbon

Fig. 5 Top: Comparison of XRF spectra of an inked area and blank papyrus measured in Region 1 on P.Oxy.3637. Bottom: bar plot of the XRF intensities (raw peak areas) measured in inked and non-inked areas from different regions (1 to 4)

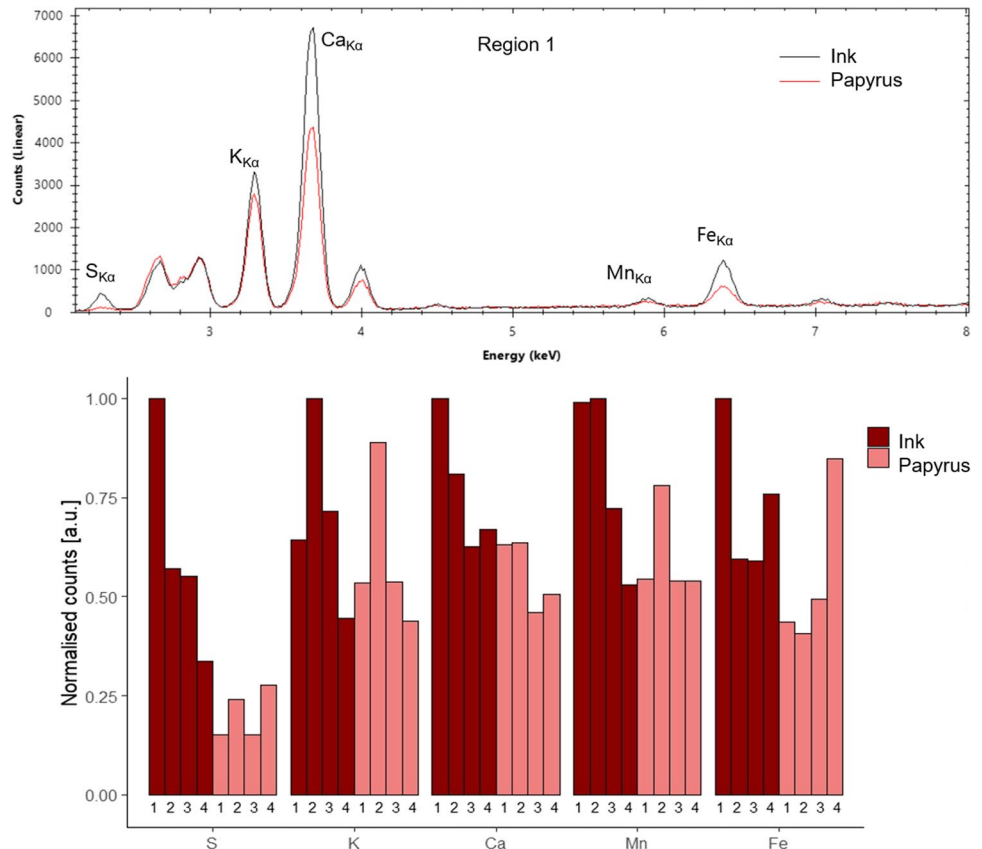
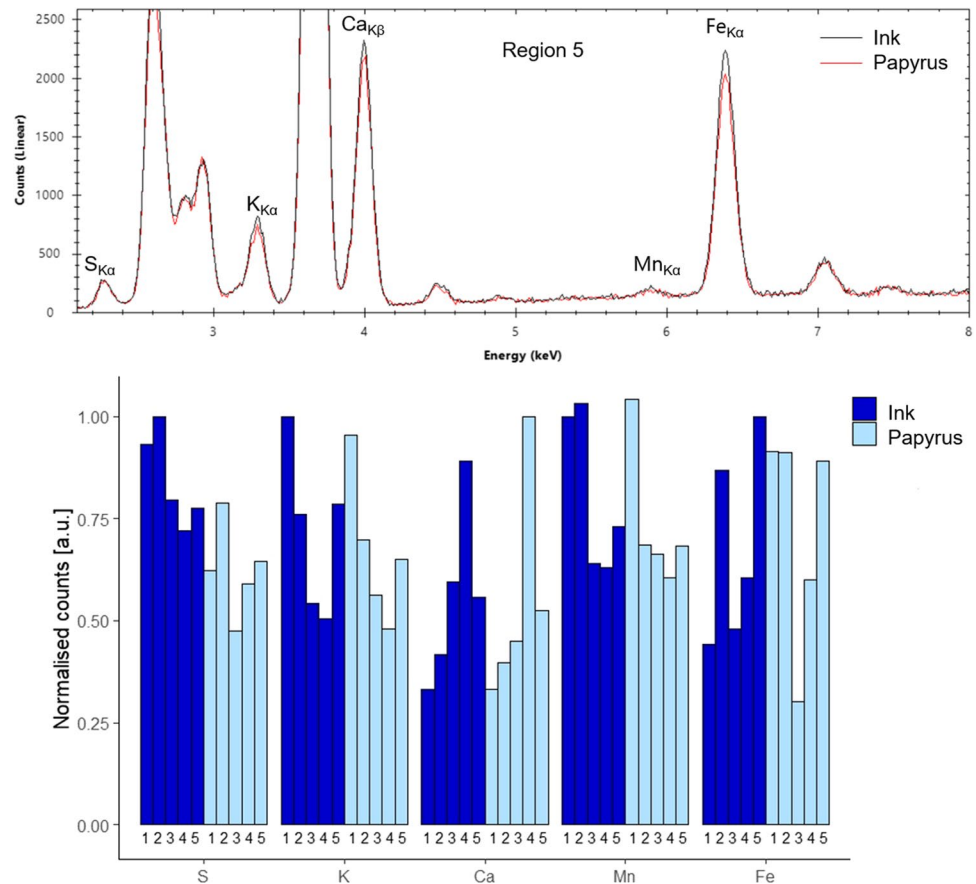


Fig. 6 Top: Comparison of XRF spectra of an inked area and blank papyrus measured in Region 5 on P.Oxy.1869. Bottom: Bar plot of the XRF intensities (raw peak areas) measured in inked and non-inked areas from different regions (1 to 5)



ink suggests more likely that these writing media were less refined, perhaps due to contamination during manufacture, storage, or usage. It might indicate that the writing media were purchased rather than self-prepared through careful and consistent processes. Table 1 summarises the results of IRR and XRF analysis.

P.Oxy. 4381, bearing the proceedings from the court of Alexandria, presented the most interesting case study. This document travelled to Oxyrhynchus together with the soldiers involved in the trial. It is written in Latin and Greek, with a Latin heading reporting date, place, and indicating the issue to be discussed in the trial, followed by a Greek text detailing the plaintiffs and the matter in discussion and a final Latin section with the court's decision. Curiously, these parts are written with different inks, as shown in Fig. 7. The comparison between visible and infrared images of area A shows that the Latin script of the first three lines changes opacity above 1510 nm, while the Greek text from the fourth line onwards remains opaque. Therefore, the first lines were written with a mixed ink (containing carbon and vegetal extracts, as XRF did not detect elements that might be associated with iron-gall ink), while carbon ink was used thereafter.

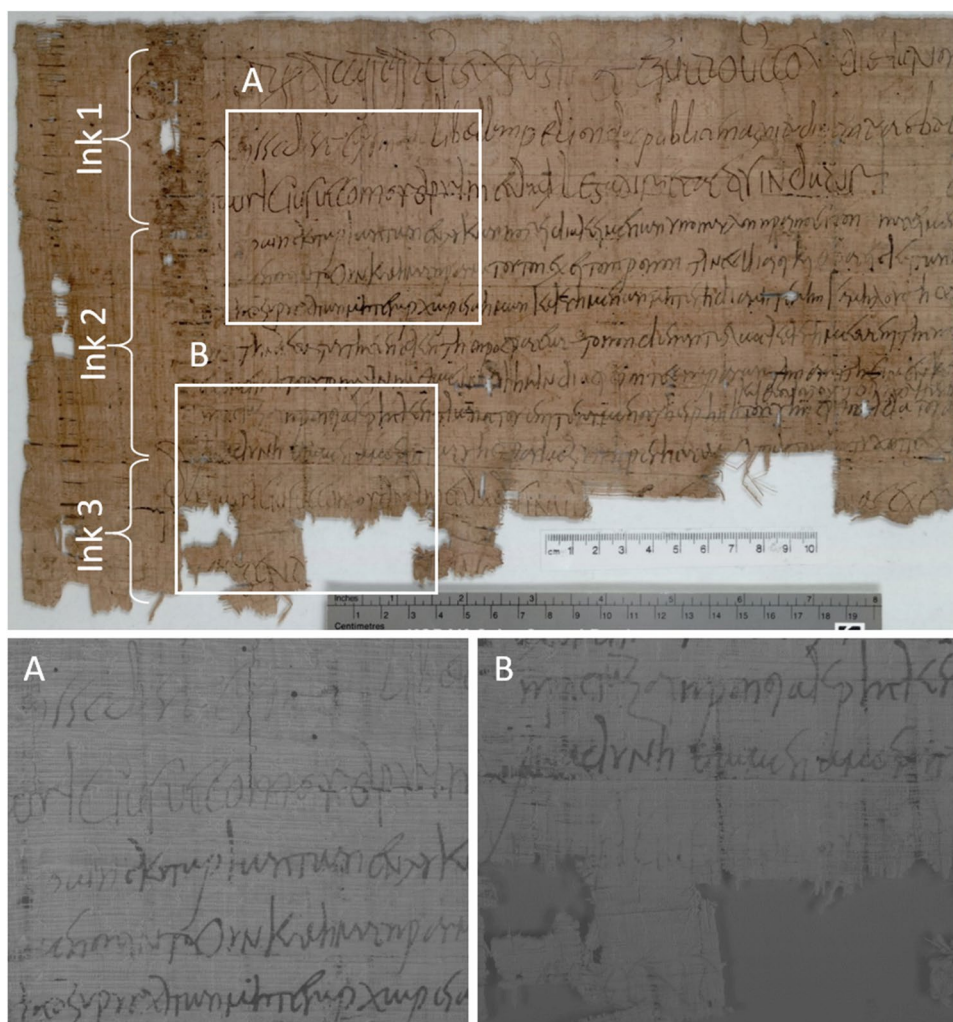
The first change of ink happens at the beginning of line 4, in the middle of the Greek phrase designating the plaintiffs.

This initially led us to think that the ink difference was not related to the different languages and parts of the text. We originally concluded the scribe of this manuscript ran out of ink in the middle of a sentence and started a new batch, unaware of the material difference between the two writing media. However, we reconsidered our interpretation when we observed the ink changes again towards the end of the text, in correspondence with the final part in Latin (Fig. 6, area B).

The last ink used on this document remains only barely visible above 1510 nm. Ongoing research shows that the low contrast between papyrus and ink that characterises this part of the document makes it difficult to classify the writing medium due to the spectral behaviour of plant and iron-gall inks above 1500 nm.⁸ Spectral information on top of the acquired imaging is needed to conclude the nature of this writing medium, whose composition could not be characterised further using XRF since the ink trace in this area is narrower than the smallest collimator available on our XRF spectrometer (0.5 mm) and partly abraded. However, the difference in opacity that can be observed in the infrared

⁸ Personal communication, Ivan Shevchuck (Centre for Manuscript Cultures, University of Hamburg), September 2022.

Fig. 7 Visible and infrared images (spectral range 1510–1700 nm) captured in areas **A** and **B** of P.Oxy.4381



pictures between the first and last part in Latin (Fig. 6, area A top and area B bottom) suggests these were penned with different inks.

We believe that although the first ink change does not correspond precisely with the language change, this correlation between different parts of the text, languages, and inks is too coincidental to result from a scribe unintentionally changing writing materials multiple times during the drafting of one and the same document. While the first ink change in the middle of the sentence seems to exclude that two different hands intervened in the writing of the document; that is, one for the Latin, another one for the Greek sections (Cavallo 2008),⁹ the intentional use of inks of a different colour or

shade¹⁰ to visually highlight different parts of the text might better explain this material difference. In fact, it has been noted in similar documents that palaeographical changes in the scribe's writing, such as enlarging the letter size, emphasising ornamental traits or reducing writing speed (and thus the text's cursiveness), were deployed to make recognisable, at first sight, the different sections in it, particularly the magistrates' interventions in Latin (Pedone 2020, 67–68).

Conclusions

The analysis of this collection showed that all of the manuscripts were written with inks containing carbon. Surprisingly, no pure iron-gall inks were found, despite this writing

⁹ As a matter of fact, it is well known that Diocletian's reforms of the administration caused scribes in the Eastern part of the Empire to write in Latin too, which resulted, among other things, in a script featuring signs used for both languages, though with different phonetic values, i.e. the so-called "koine grafica greco-latina," of which P.Oxy. 4381 is a good example itself.

¹⁰ While carbon ink is pitch black, plant ink comes in different shades of brown depending on the composition of the plant extracts. Adding carbon ink to plant ink would slightly change the colour, resulting in a darker shade of brown.

medium being used in Egypt at that time (fourth–seventh centuries CE). This confirms a previously highlighted trend in the predominant use of carbon-based inks for documentary manuscripts (Ghigo 2020; Ghigo and Albarrán Martínez 2021). However, in very few cases (P.Oxy.1869, 2665, 3637, and 4381), carbon was mixed with plant and iron-gall ink, which seem to have served as “extenders.” No strong correlation was found across the entire collection between the degree of formality of the document and the composition of the writing medium. However, three out of four court proceedings written in Oxyrhynchus (P.Oxy.1878, 1879, 1880) were penned with relatively “clean” carbon ink containing no transition metals that might be attributed to contamination during manufacturing or storage processes. Further investigation is needed to address whether this preliminary result reflects a broader trend resulting from specific technical traditions.

Interestingly, P.Oxy.4381, bearing the proceedings of the court of Alexandria, was found to be written with different inks, which correlate with the languages (Latin and Greek) used in different parts of the text. This pattern was found exclusively in this document, as the bilingual proceedings from the court of Oxyrhynchus were consistently written with the same ink. The combined analysis of palaeographical and material features suggest that the ink change in P.Oxy.4381 resulted from the intentional choice of a scribe aiming to visually highlight the different parts of the text. Therefore, this document might bear evidence of established technical and material traditions that existed at the court of Alexandria and have not been investigated so far. A systematic material characterisation of several court proceedings from the capital might reveal interesting details regarding the traditions that governed the scribal practice in this context.

Finally, by combining XRF and PCA, this investigation highlighted differences in the elemental composition of the papyrus paper of three letters written outside of Oxyrhynchus, as revealed by their text. This result is worth attention considering that to our best knowledge, this is the first time that PCA has been used in combination with a portable, non-invasive method for the study of ancient papyri (previous studies combined PCA with SEM–EDX and Synchrotron-based microanalyses (Chiappe et al. 2019; Christiansen et al. 2020; Pereira et al. 2021; Nodar et al. 2022)). Therefore, the findings here encourage further research along these lines as the combination of XRF and PCA can integrate historical research on the relationship between the central administration and local communities and, more in general, on papyrus provenance and sourcing.

Experimental

Handling of the papyri During the analyses, the manuscripts were placed on a supporting glass to handle and displace them while minimising the papyrus’s physical stress. As shown by the plot of PC1 vs PC2 (Fig. 1), the material composition of the supporting glass was fundamentally different than the one of the papyri and did not interfere with the evaluation of results.

Apollo camera, Opus This infrared camera features a cooled indium gallium arsenide sensor with a spatial resolution of 128×128 pixels and a range of sensitivity between 900 and 1700 nm. The infrared images were acquired by scanning the manuscripts using a long-wave filter that blocked the incoming radiation below 1510 nm.

CRONO, Bruker This XRF spectrometer features a Rhodium target tube and a 50-mm^2 SDD detector with energy resolution < 140 eV for Mn $K\alpha$ with an input count rate of up to 500,000 cps. It allows to detect elements in the range $11 < Z < 92$. The spectra presented in this article were obtained with a 0.5-mm collimator, operating at 40 keV and $80 \mu\text{A}$. The number of investigated regions per manuscript varied between 3 and 5, depending on the size of the papyrus. Per each region, we measured both a blank area of the support and a neighbouring inked area, resulting in 6 to 10 measurements per manuscript. The data were processed using the CRONO software (Bruker) by computing raw peak areas of each detected element.

Principal component analysis (PCA) The XRF’s raw peak areas of the elements detected across the manuscripts studied (Si, P, Cl, K, Ca, Ti, Mn, Fe, Cu, Zn, Pb, and Sr) were imported into RStudio to compute a scaled and centred PCA model using the function `{prcomp()}`.

Supplementary information The online version contains supplementary material available at <https://doi.org/10.1007/s12520-023-01839-9>.

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Author contributions T. G.: conceptualisation, methodology, investigation, formal analysis, visualisation, writing — original draft. A. N. D.: conceptualisation, investigation, writing — review and editing.

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Data availability The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Competing interests The authors declare no competing interests.

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References

- Aceto M, Agostino A, Fenoglio G, Idone A, Gulmini M, Picollo M, Ricciardi P, Delaney JK (2014) Characterisation of colourants on illuminated manuscripts by portable fibre optic UV-visible-NIR reflectance spectrophotometry. *Anal Methods* 6(5):1488–1500
- Arlt T, Mahnke H-E, Siopi T, Menei E, Aibéo C, Pausewein R-R, Reiche I, Manke I, Lepper V (2019) Absorption edge sensitive radiography and tomography of Egyptian Papyri. *J Cult Herit* 39:13–20
- Autran P-O, Dejoie C, Hodeau J-L, Dugand C, Gervason M, Anne M, Martinetto P, Bordet P (2021) Revealing the nature of black pigments used on Ancient Egyptian Papyri from Champollion collection. *Anal Chem* 93:1135–1142
- Brun E, Cotte M, Wright J, Ruat M, Tack P, Vincze L, Ferrero C, Delatre D, Mocella V (2016) Revealing metallic ink in Herculaneum Papyri. *Proc Natl Acad Sci* 113(14):3751–3754
- Cavallo G (2008) *La Scrittura Greca e Latina Dei Papiri: Una Introduzione*. Fabrizio Serra Editore, Pisa, Roma
- Chiappe C, Pomelli CS, Sartini S (2019) Combined use of scanning electron microscopy–energy-dispersive X-ray spectroscopy and Fourier transform infrared imaging coupled with principal component analysis in the study of Ancient Egyptian Papyri. *ACS Omega* 4(26):22041–22047
- Christiansen T (2017) Manufacture of black ink in the Ancient Mediterranean. *Bull Am Soc Papyrol* 54:167–195. <https://doi.org/10.2143/BASP.54.0.3239877>
- Christiansen T, Cotte M, Loredó-Portales R, Lindelof PE, Mortensen K, Ryholt K, Larsen S (2017) The nature of Ancient Egyptian Copper-containing carbon inks is revealed by synchrotron radiation based X-ray microscopy. *Sci Rep* 7:15346. <https://doi.org/10.1038/s41598-017-15652-7>
- Christiansen T, Cotte M, de Nolf W, Mouro E, Reyes-Herrera J, de Meyer S, Vanmeert F et al (2020) Insights into the composition of Ancient Egyptian red and black inks on Papyri achieved by synchrotron-based microanalyses. *Proc Natl Acad Sci* 117(45):27825–27835
- Cohen Z (2022) *Composition analysis of writing materials in Cairo Genizah documents*. Cambridge Genizah Studies, 15. Leiden ; Boston: Brill
- Colini C (2018) *From recipes to material analysis: the Arabic tradition of black inks and papercoatings (9th to 20th Century)*. Hamburg
- _____. (2021) “I tried it and it is really good”: replicating recipes of Arabic black inks. In *Traces of ink experiences of philology and replication*, edited by Lucia Raggetti, 131–53. Brill
- Daccache J, Desreumaux A (2015) ‘Les Textes Des Recettes d’encres En Syriac et En Garshuni’. In *Manuscripta Syriaca. Des Sources de Première Main*, F Chatonnet and M Debié (eds.) 195–246. Paris: Librairie orientaliste Paul Geuthner
- Ghigo T (2020) *A systematic scientific study of coptic inks from the Late Roman period to the Middle Ages*. Sapienza University of Rome, University of Hamburg, Rome - Hamburg
- Ghigo T, Albarrán Martínez MJ (2021) The practice of writing inside an Egyptian monastic settlement: preliminary material characterisation of the inks used on coptic manuscripts from the monastery of Apa Apollo at Bawit. *Heritage Science* 9(1):62. <https://doi.org/10.1186/s40494-021-00541-0>
- Ghigo T, Rabin I, Buzi P (2020) Black Egyptian inks in Late Antiquity: new insights on their manufacture and use. *J Archaeol Anthropol Sci* 12:70. <https://doi.org/10.1007/s12520-019-00977-3>
- Hahn O, Malzer W, Kanngießer B, Beckhoff B (2004) Characterization of iron-gall inks in historical manuscripts and music compositions using X-ray fluorescence spectrometry. *X-Ray Spectrom* 33(4):234–239. <https://doi.org/10.1002/xrs.677>
- Kolar J, Strlič M (eds) (2006) *Iron gall inks: on manufacture, characterisation, degradation and stabilisation*. National and University Library of Slovenia, Ljubljana
- Maltomini F, Ghigo T, Hahn O, Rabin I (2021) Florentine Papyri under examination: the material study of the inks used at the beginning of the common era in the “Family of Kôm Kâssûm” archive (Hermopolis). *Archiv Für Papyrusforschung* 67(1)
- Malzer W, Hahn O, Kanngießer B (2004) A fingerprint model for inhomogeneous ink–paper layer systems measured with micro-X-ray fluorescence analysis. *X-Ray Spectrom* 33(4):229–233. <https://doi.org/10.1002/xrs.676>
- Mrusek R, Fuchs R, Oltrogge D (1995) *Spektrale Fenster zur Vergangenheit: Ein neues Reflektographieverfahren zur Untersuchung von Buchmalerei und historischem Schriftgut*. Naturwissenschaften 82(2):68–79
- Nehring G, Bonnerot O, Gerhardt M, Krutzsch M, Rabin I (2021) Looking for the missing link in the evolution of black inks. *Archaeol Anthropol Sci* 13(4):71
- Nodar A, Pereira FJ, Ferrer N, López R (2022) Ink and support characterization of typologically established Papyrus groups from the Palau-Ribes collection. *Herit Sci* 10(1):107. <https://doi.org/10.1186/s40494-022-00742-1>
- Pedone M (2020) *Apud Acta. Studi Sul Processo Romano Alla Luce Della Documentazione Papirologica (IV-VI Sec. DC)*. Giappichelli, Torino
- Pereira FJ, Lopez R, Brasas M, Alvarez R, Aller AJ (2021) Synergism between SEM/EDX microanalysis and multivariate analysis for a suitable classification of Roman and Byzantine Papyri. *Microchem J* 160:105688

- Preisendanz K (1931) *Papyri Graecae Magicae. Die Griechischen Zauberpapyri*, Herausgegeben Und Übersetzt von Karl Preisendanz. V. 2. Teubner, Berlin
- Rabin I, Binetti M (2014) NIR reflectography reveals ink type: pilot study of 12 Armenian manuscripts of the Staatsbibliothek Zu Berlin'. *Բանբեր Մատենադարան* (Banber Matenadaran) 21: 465–70
- Rabin I, Krutzsch M (2019) The writing surface Papyrus and its materials 1. Can the writing material Papyrus tell us where it was produced? 2. Material study of the inks. In Proceedings of the 28th Congress of Papyrology; 2016 August 1–6; Barcelona. Barcelona: Publicacions de l'Abadia de Montserrat, Universitat Pompeu Fabra; 2019, pp 773–781. Publicacions Abadia de Montserrat
- Rabin I (2017) Building a bridge from the Dead Sea scrolls to Mediaeval Hebrew manuscripts. In *Jewish manuscript cultures: new perspectives*, edited by Irina Wandrey, 310–22. *Studies in Manuscript Cultures* 13. Berlin & Boston: Walter de Gruyter. <https://doi.org/10.1515/9783110546422-012>
- Raggetti L (2016) Cum Grano Salis. Some Arabic ink recipes in their historical and literary context. *J Islam Manusc* 7(3):294–338. <https://doi.org/10.1163/1878464X-00703002>
- Zerdoun Bat-Yehuda M (1983) *Les Encre Noires au Moyen-Âge (jusqu'à 1600)*. Centre National de la Recherche Scientifique, Paris

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