2D macro-XRF to reveal redacted sections of French queen Marie-Antoinette secret correspondence with Swedish count Axel von Fersen

Anne Michelin, Fabien Pottier, Christine Andraud

MATERIALS SCIENCE

2D macro-XRF to reveal redacted sections of French queen Marie-Antoinette secret correspondence with Swedish count Axel von Fersen

Anne Michelin*†, Fabien Pottier†, Christine Andraud

During the French Revolution, Marie-Antoinette, queen of France and wife of Louis XVI, maintained a highly secret correspondence with the Swedish count Axel von Fersen, her close friend and rumored lover. An unidentified censor later redacted certain sections of the exchanged letters. This presumably sensitive content has been puzzling historians for almost 150 years. We report on the methodology that successfully unraveled this historical mystery. X-ray fluorescence spectroscopy was used in macroscanning mode on the redacted sections. Specific data processing was applied to improve the legibility of the hidden writings (elemental ratios, statistical data reduction, multimodal images fusion, unmixing procedure, and image treatments). This methodology successfully revealed the redacted contents of eight letters, shedding new light not only on Marie-Antoinette and Fersen relationship but also on the author of the redactions. It will also be of great interest for other historical and forensic cases involving the disentanglement of superimposed multi-elemental materials.

INTRODUCTION

During the French Revolution, between June 1791 and August 1792, the French royal family was kept under close surveillance at the Tuileries Palace. Nonetheless, Marie-Antoinette (abbreviated as MA), wife of Louis XVI and queen of France, managed to maintain a highly secret correspondence with the Swedish count Axel von Fersen (abbreviated as AF), her close friend and rumored lover, who notably helped organize the flight to Varennes, the failed attempt of the royal family at escaping France. The French national archives currently hold some of the exchanged letters, purchased from the Fersen family archives in 1982. Among these documents written by MA and AF (because AF also archived drafts of the letters he sent to MA), certain sections of a few words to a few lines were later redacted by an unidentified censor (see top of Figs. 1 to 4). Whether state secrets, escape plans, or evidence of a royal love affair, this presumably sensitive content has been puzzling historians for almost 150 years.

Revealing hidden, faded, or degraded graphical contents of historical artefacts is an important topic in cultural heritage research (*1*, *2*). Although chemical analysis had been used in the past, today, instrumentations using light/matter interactions in different ranges of the electromagnetic spectrum are preferred as they can noninvasively map multiple physiochemical properties of the studied surfaces at a macroscopic scale. In each object, the nature and combination of materials (inks, coloring materials, substrate, etc.) are unique. The revelation of a useful contrast with a given technique will depend on the chemical nature/physical structure of these diverse materials. Mapping elemental composition [macro–x-ray fluorescence scanning (MA-XRF)] (*3*–*6*), molecular signature [mid-infrared (IR) scanner] (*7*), multi/hyperspectral imaging (*8*–*10*), heat conduction (thermal imaging) (*11*), or far-IR absorption (THz imaging) (*12*), along with additional specific data processing (*1*, *13*–*15*), can offer entirely new readings of a surface and reveal hidden or degraded contents. Up to this day, imaging research in historical manuscripts is mainly focused on discovering the inside text of rolled/folded papyri or parchments with x-ray tomography (*16*, *17*) or revealing the erased original content of palimpsests or faded/degraded documents using diverse imaging techniques (*3*, *6*, *18*, *19*). Few redacted historical documents such as MA and AF letters have been the subject of published work so far (for example, a musical score, the Luigi Cherubini's 1797 opera *Médée*, blacked out by his author with carbon ink) (*20*). MA and AF letters most closely resemble the palimpsests, with a superimposition of two texts that must be disentangled, but the current problem is also different for two reasons. On the bright side, contrary to the palimpsests, the original text of the letters has not been erased in any way, which makes it easier to detect. On the other hand, the two layers of inks are exactly superimposed as redaction is intentional and the redaction ink is often very dark, absorbing in a wide range of visible wavelengths and thus preventing the analysis of the underlying ink with this type of radiation. Different difficulties must therefore be addressed.

The first part of this project was devoted to establishing a critical grid of potentially useful techniques for separating two similar inks in these documents. Different techniques, from the simplest to the most advanced (microscopy, microtopography, reflectance transformation imaging, IR thermography, hyperspectral imaging in the visible and near IR range, etc.), were tested to determine whether there was a difference in the physical or chemical properties of the two inks to distinguish them. In particular, the possibilities of hyperspectral imaging in the visible and near-IR range have been explored given the successful results obtained both on historical documents and in forensic studies (*10*, *19*, *21*, *22*) to find underlying writings or drawings and given the speed of acquisition (an important parameter on a large corpus of documents). Unfortunately, in the visible range, the black redaction ink absorbs almost all light at all wavelengths, preventing the exploitation of an underlying signal. In the short-wave IR range, the redaction and underlying inks are largely transparent and extremely similar, leading to inconclusive results despite the different chemometric data processing attempts.

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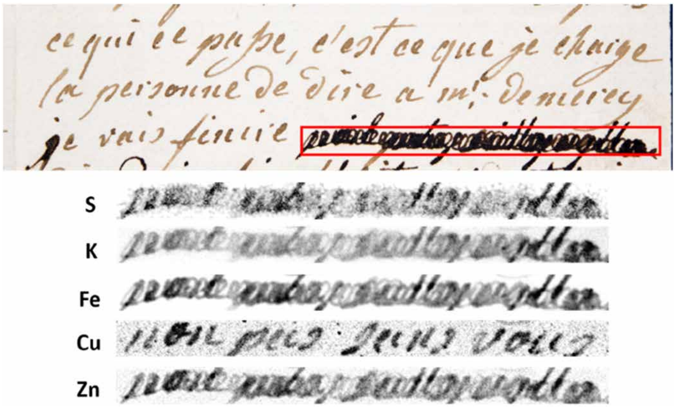

Fig. 1. Recovering the text using elemental map. Details of page 1 of letter 440AP_1_1_4, written by MA, 4 January 1792. Elemental maps corresponding to the red squared area (only the elements composing the inks are shown) (4.0 mm by 54.4 mm; pixel size, 100 μm; dwell time, 100 ms per pixel). Text read thanks to Cu map: "non pas sans vous."

Moreover, the very tight loops of the redaction and the addition of letters by the censor to complicate the reading prevent the use of "joining of the dots" techniques from images where the redaction would have been subtracted. In the end, XRF spectroscopy gave the best results. Preliminary XRF measurements have proved that both the original handwritings and the redaction of all studied letters are made of metal-gall ink, because of the consistent detection of sulfur and iron in conjunction with the IR transparency (*23*). As mentioned by Glaser and Deckers (*24*, *25*), iron is always detected in iron-gall ink, but additional metal elements—that are present as impurities in the vitriol (iron sulfate) used to prepare the ink—are also found in diverse amounts. Sometimes, the mere presence of a specific element can discriminate between two inks (*26*). XRF spectroscopy has been used for many years in the field of cultural heritage to determine the elemental composition of different materials (*27*). The interest of XRF imaging by scanning a large object with a focused beam dates back to the 1990s (*28*, *29*) and has continued ever since (*30*–*32*), especially on historical paintings and illuminated manuscripts, because it is possible to map the distribution of elements at the macroscopic scale and sometimes to highlight underlying drawings or paintings. Initially, applications on cultural heritage objects were carried out on synchrotron source (*5*, *33*), limiting the number of studies because of restricted access to synchrotron. Until a few years ago, a number of mobile MA-XRF instruments using x-ray tubes have been developed in different institutes (*34*–*37*), but the high spatial resolution needed for working on writings of historical document, the long acquisition times, and the small number of equipment available limit the possibility of having such equipment for the type of study conducted in this article. The development and democratization of commercial mobile instruments (*38*, *39*) make it possible to investigate larger corpus of documents and not only on a few exceptional manuscripts.

In MA and AF letters, the inks are always composed of S, K, Mn, Fe, Cu, and Zn in variable proportions. All the redacted parts of MA and AF correspondence were therefore scanned with a Bruker M6 Jetstream XRF scanner (more details in Materials and Methods), with the leading hypothesis that a difference in the chemical composition of the original text and redaction inks would allow discriminating the redacted contents. For this case study, reading the original text often requires customized image processing. The following paragraphs describe each situation that was encountered and how difficulties were addressed. Different strategies have been tested: from the simplest (elemental maps combination) to the more complex [multivariate statistical analyses such as principal components analysis (PCA) and supervised analyses] associated with more classical image processing. The local differences in materials have forced us to adapt the methodology to each line of a letter or even to each word, making it difficult to predict the most efficient strategy upstream. This study also made it possible to obtain information about the author of the redactions.

RESULTS AND DISCUSSION

Recovering the hidden text

The data acquired with an XRF scanner is in the form of a data cube in which each pixel—whose size is determined by the step size (here is chosen equal to the beam size and set between 50 and 180 μm)—contains the XRF spectrum of the analyzed area. Different types of data processing can be envisaged depending on the case: Elemental maps (gray scale of the integrated elemental emission intensity in each pixel) can be extracted, or the data cube can be processed considering all or part of the measured spectrum.

In the best-case scenario, an element is present in the original text in much higher amounts than in the redaction. The map of this element may provide enough contrast to reveal the original text without further data treatment. Figure 1 shows elemental maps of all elements composing the inks of letter 440AP_1_1_4. Copper is obviously the only element of interest to see through the redaction: It is possible to read the original content of the letter in its elemental map. In most cases, however, none of the single element mappings show sufficient contrast between the two inks. In the example in Fig. 2A, no single element, including Cu, reveals the original text. The comparison of the detected abundances in a few spots of the two inks (left-hand side of Fig. 2B) explains this observation: The two elements are present in both inks, and the larger abundance in the redaction prevents any reading of the original text when the two are overlaid. However, the copper-to-iron ratio (right-hand side of Fig. 2B) shows a consistent composition difference between the two inks (0.6 versus 0.2). The mapping of this ratio, binarized with a threshold just below 0.6 (the ratio value for the redaction ink when alone), overlaid with a mask to clear noisy areas where no ink is present, reveals the original text (Fig. 2B). Because the ink composition difference is sometimes very tenuous, these elemental ratio renderings can result in very noisy images even after masking the unwritten areas. Additional image treatments based on outlier detection or filters ("Remove Outliers" and "Smooth" functions in ImageJ software) can enhance the readability of the original text. Because the scanning area for the XRF scanner is restricted to limit the scanning time (for a line of text of about 10 cm by 1 cm, with a step size of 180 μm and a dwell time of 500 ms per pixel, it takes about 4 hours of acquisition, and in some difficult cases, it was sometimes necessary to select a dwell time of 3 s per pixel or a step size of 100 μm, leading to acquisition times of several hours for one word), repositioning the processed images on a photograph of the letter to give context can greatly aid reading, as shown in Fig. 3.

The elemental ratios, even after image processing, do not always produce a result that allows the text to be read, but the XRF data

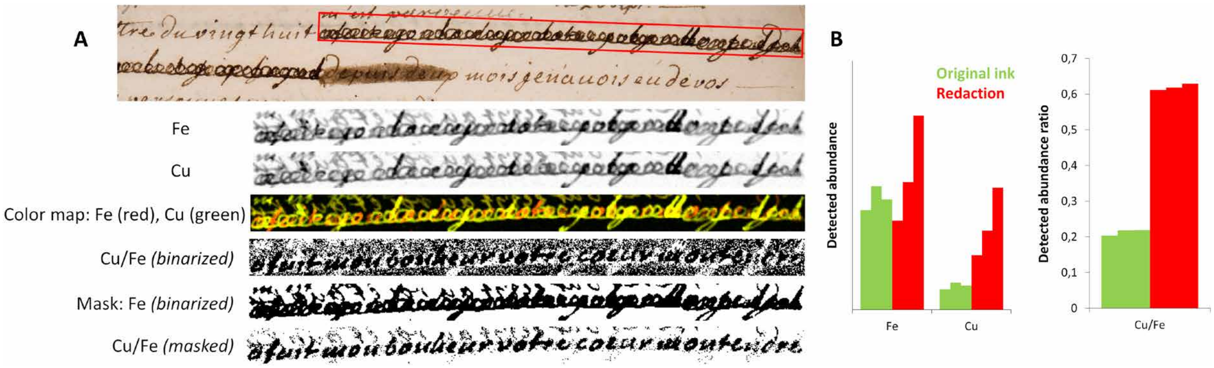

Fig. 2. Recovering the text using ratio of elemental maps. (**A**) Details of page 1 of document 440AP_1_1_8, transcription by AF of a letter written by MA, 26 September 1791. Elemental maps, false color map (Fe map in red and Cu map in green), and combined maps corresponding to the red squared area (7.5 mm by 114 mm; 180 μm; 500 ms per pixel). Read text: "a fait mon bonheur votre coeur mon tendre." (**B**) Detected abundance and abundance ratios of iron and copper in the two inks used in page 1 of document 440AP_1_1_8.

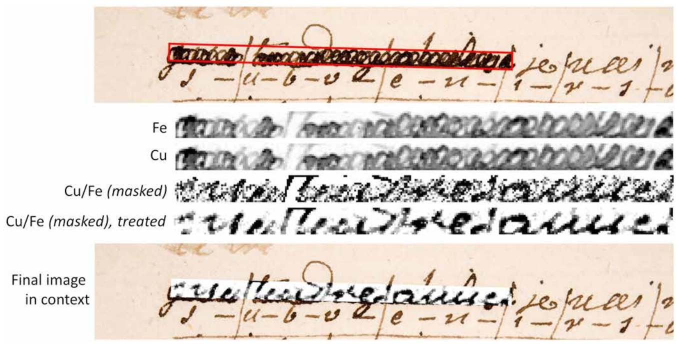

Fig. 3. Recovering the text thank to the context. Details of letter 440AP_1_2_9 written by AF, 13 October 1791. Elemental maps (Fe and Cu) and combined map before and after image treatment (Remove Outliers and Smooth functions in ImageJ software) corresponding to the red squared area (2.7 mm by 50.5 mm; 180 μm; 500 ms per pixel). Read text "ma tendre amie."

cube contains much more information that can be explored in different ways. However, the accumulation of data can sometimes be counterproductive: The difficulty of problems increases rapidly with the number of dimensions leading to poor performance of algorithms on high-dimensional data. The phenomenon is called the curse of dimensionality (*40*). Dimensionality reduction, i.e., transformation of these data into a meaningful representation of reduced dimensionality, can mitigate this curse and facilitate the visualization and the compression while retaining the information content inherent in these data. In our case, this sometimes allows us to concentrate the information about the underlying ink in one image. One of the most well-known techniques used in many fields is the PCA. For this technique, a new coordinate system is calculated from linear combinations of the original dataset. The new axes (principal components) are chosen to maximize the variance of orthogonal projections. They are basically calculated by diagonalizing the covariance matrix (that measures the joint variability of variables): Principal components are obtained by multiplying the matrix of eigenvectors of the covariance, ordering them by eigenvalues (highest to lowest), by the dataset matrix.

PCA, applied to a set of the images of the elemental maps or to all or part of the entire spectrum, can then sometimes give very good results in this study. This is the case for some letters written on both sides of the paper, whose inks (of the original text) on both sides are not identical. The penetration of the x-rays in the light paper matrix will result in the superimposition of data from both sides, and elemental ratios are too noisy to be usable. For example, the verso of page 5 of letter 440AP_1_1_2 (Fig. 4) was written with an ink more concentrated in copper than the ink on the recto side. Moreover, the redaction ink exhibits a larger concentration of Zn. A combination of the three main elements present in the inks (Fe, Cu, and Zn) is hence potentially discriminating. A PCA on these three elemental maps concentrates the information of interest in one single dimension (PC3 in our case), which isolates the original text from the redaction and the verso.

Another multivariate method for the reduction of spectral bands called the minimum noise fraction (MNF) transform, widely used for hyperspectral image processing, was tested (*41*). The MNF transform, described by Green *et al.* (*42*), aims to obtain principal components showing steadily decreasing image quality with increasing component number, which PCA does not produce systematically. In this technique, image quality is measured by the signal-to-noise ratio. In practical terms, instead of choosing new components to maximize variance (corresponding to the information content), as the PCA does, the maximized parameter chosen here is the signal-to-noise ratio. The MNF transform is a linear transformation that consists of two cascaded PCAs to minimize the noise, i.e., to identify noise and segregate it from true information and to retain the useful information into a much smaller set of MNF images.

First, the noise covariance matrix is computed to decorrelate and rescale the noise from the data (a process known as noise whitening), resulting in transformed data in which the noise has unit variance and no band-to-band correlations. The second step is a standard PCA of the noise-whitened data, which orders the bands with respect to the signal-to-noise ratio. Higher eigenvalues (>1) contain more information in the bands, and values near 1 render noisy datasets.

Depending on the letters, or even within a letter, one or the other technique may give a better result (Fig. 5). Different tests by varying the spectral range showed that it is not necessary to take all the data to get the best result. Sometimes, a reduced range or even a selection of a limited number of elemental maps can give similar or even better results (Fig. 5). This is another consequence of the curse of

Fig. 4. Recovering the text using PCA. Details of page 5 of letter 440AP_1_1_2, written by MA, 7 December 1791. Elemental maps, mirrored verso, elemental ratio maps, and PCA of these details (4.8 mm by 116.1 mm; 100 μm; 500 ms per pixel). Read text: "vous que j'aime et aimeroi jusqu'a mon."

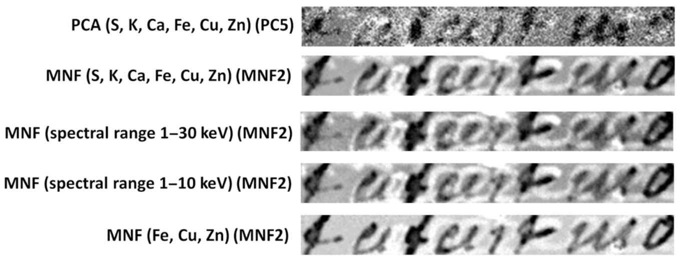

Fig. 5. Recovering the text using MNF. Details of page 1 of document 440AP_1_1_8, transcription by AF of a letter written by MA, 26 September 1791. PCA and MNF of this area (2.88 mm by 26.5 mm; 100 μm; 3000 ms per pixel). Read text: "t a fait mo."

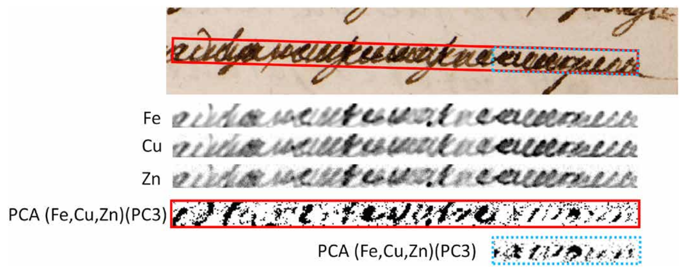

Fig. 6. Recovering the text using PCA on a reduced part of the image. Details of page 1 of document 440AP_1_1_8, transcription by AF of a letter written by MA, 26 September 1791. Elemental map and PCA maps corresponding to the red and blue squared area (3.5 mm by 67 mm; 180 μm; 500 ms per pixel). Read text: "amour."

dimensionality: The information allowing to discriminate the inks being diluted in a too big number of data leads to a nonrepresentativeness of the results for the data reduction algorithms; manually selecting a smaller dataset that is known to be relevant for ink composition helps to achieve better results.

The use of multivariate statistical analysis through PCA or MNF has yielded positive results in many cases. But the quality of the images obtained is not always sufficient to read the letters completely. It is sometimes possible to improve the legibility of certain redacted sections by refinements in the application of these statistical analyses. This statistical analysis generates a linear transformation matrix that is specific for each dataset based on its global variance or signal-to-noise ratio. In certain cases, however, the transformation matrix is not best suited to reveal minor composition differences that are necessary when trying to reveal redacted characters. As a result, if an area of interest remains illegible, it was often useful to run a new PCA/MNF focused on this reduced part of the image. An example is shown in Fig. 6, where the last few characters of the analyzed area are not revealed by the global PCA. Running a new statistical analysis focused on this data subset generates a different transformation that offers a better reading. A marked change in the relative amounts of writing and redaction inks likely explains this divergence in the statistics of the last word of the line. Hence, an alternate elemental projection better reveals the final word of the sentence. It is also possible to perform transformation of dimensionality reduction using the covariance matrix that was calculated from a different area of the same spectral dimensionality. The idea is to use the covariance matrix of one dataset to calculate the principal components of another dataset. In the example in Fig. 7, a classic PCA gives a satisfactory result for area A (area of the document that gives the best results), while the results of PCA on area B remains almost illegible. Here, the variance of area B is probably driven by irrelevant contrast information (possibly due to external material perturbations in material abundances), while in area A, the optimal elemental projection calculated from the PCA is focused on text/redaction elemental contrast. When the covariance matrix of area A is used to transform area B dataset, a much better result is obtained. Additional image processing enhances the final image.

Another methodological approach using supervised data processing sometimes leads to better results. In this case, the XRF data are considered in their entirety: Each pixel of the data cube contains the whole x-ray spectrum. Spectral unmixing process is widely used in the remote sensing field (*43*). In a hyperspectral image, the reflectance at each pixel may be considered as the combination of the reflectance of each material (end-members) present within the pixel (whose size depends on the spatial resolution of the sensor). If the multiple scattering among distinct end-members is negligible and the surface is partitioned according to the fractional abundances, then the spectrum of each pixel is well approximated by a linear mixture of end-member spectra weighted by the corresponding fractional abundances. Spectral unmixing is the process of decomposing the spectral signature of a mixed pixel into a set of end-members and their corresponding abundances. Spectral unmixing results are highly dependent on the input end-members; changing the end-members changes the results. The same approach can be considered for XRF spectra, assuming that the recorded signal is a combination of spectral signature of each material in the stratigraphy (paper, writing, and redaction inks).

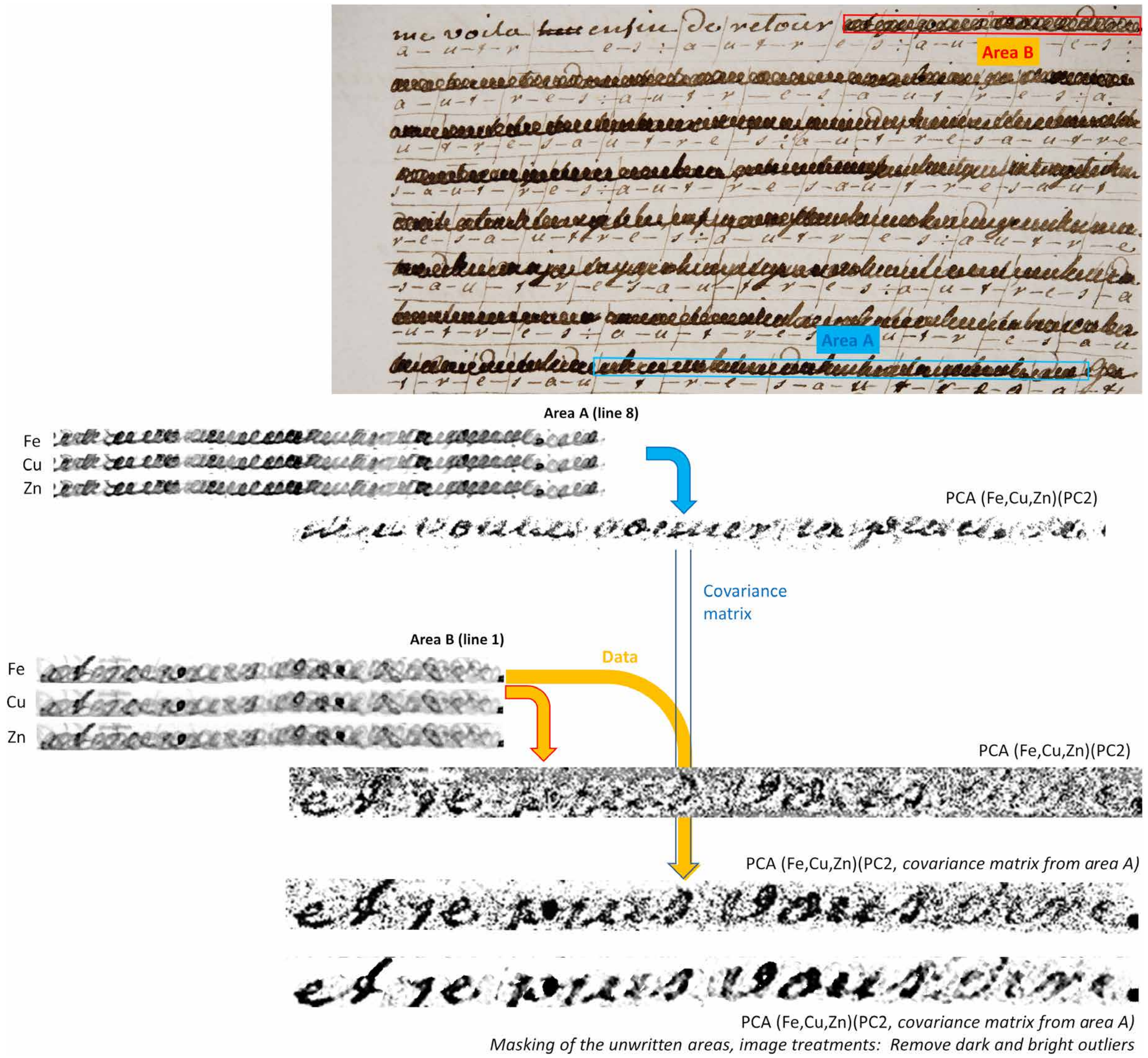

Fig. 7. Recovering the text using PCA with the covariance matrix from a different area. Details of page 1 of document 440AP_1_2_8, written by AF, 10 to 12 October 1791. Elemental map and PCA maps [3.3 mm by 80 mm (line 1) and 2.3 mm by 63 mm (line 8); 100 μm; 750 ms per pixel]. Read text area A: "n'a voulu donner la place de." Read text area B: "et je puis vous dire."

After manually determining the reference spectra of the two inks and the paper in the range 0 to 10 keV, the spectral unmixing procedure on MNF transform data gives the map of the writing ink and other maps (Fig. 8). When the end-members are easily identified (i.e., if there are sufficiently large areas where writing and redaction inks are unquestionably identified) and if the composition of the inks does not vary in the area under study, this treatment is particularly effective. The compositions of the two inks should also not be too similar for this data processing to work.

Fig. 8. Recovering the text using unmixing procedure. Details of page 1 of document 440AP_1_1_8, written by AF, 8 October 1791. Comparison of MNF maps and unmixing procedure. Read text: "et Sophie m'en voudra j'avoue qu'apr(es)."

The data processing methods described above have sometimes produced spectacular results (Figs. 1 to 8). However, for a certain number of redacted passages, the situation is more complex. This is the case for passages written on both sides of the paper. In the example shown in Fig. 4, two different inks are present on both sides of the letter, which is a specific situation: MA actually wrote the two sides of this page with an interval of 2 days. Using a single ink to write on the two sides of a page is much more common. It is the case in the example in Fig. 9: In the elemental and element ratio maps, there is no way to differentiate the writings of the two sides that share exactly the same Fe, Cu, and Zn composition. A merging of optical (standard photograph recolored in blue) and elemental data helps to overpass this issue to some extent by identifying the text written on the verso. To improve readability, areas written only on the back and that do not overlap with the text or redacting

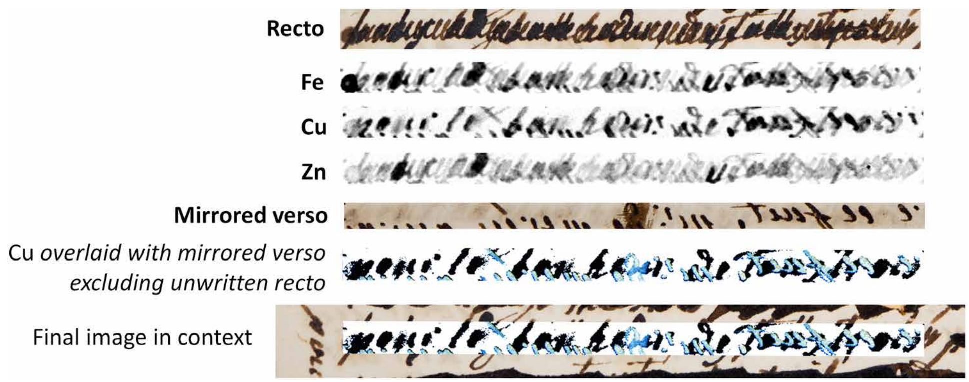

Fig. 9. Recovering the text for letters written on both sides. Details of page 6 of letter 440AP_1_1_2, written by MA, 7 December 1791. Elemental maps and elemental maps overlaid with mirrored verso photograph (4.2 mm by 75.5 mm; 100 μm; 500 ms per pixel). Read text: "pour le bonheur de tous trois."

on the front have been removed. Putting this multimodal image in context helps interpret the redacted content, but it remains difficult to read.

Of the corpus of 15 redacted letters, 8 could be read so far. Regarding the seven remaining documents, all the methodological tools that are presented failed at recovering any of the censored writings. In these cases, both inks (redaction and underlying inks) seem to have very similar compositions, making it impossible to read the redacted passages by the data processing described in this article.

Looking for the censor

In addition to reading the redacted passages, part of the project also consisted of gathering information on the author(s) of this censorship. The most common hypothesis was that redaction was carried out in the second half of the 19th century by the great-nephew of the Count of Fersen, the Baron of Klinckowström, or perhaps by a different member of the Fersen family, before the publication of this correspondence to preserve their reputation.

Point measurements of all inks (writing and redaction) were carried out with the M6 Jetstream (multipoint option) using 200-s dwell times. Their compositions were compared to establish a temporality in the realization of this censorship. The objective was to find out whether some of these redactions were carried out using the same ink (and therefore probably over a limited period of time). It was not possible to use the XRF data cubes already collected, because the analysis conditions were not optimal for this problem; the writing and redaction areas alone were too small, and dwell time was too short. Graphology allows us to affirm that many letters supposedly written by MA were, in fact, copied by AF. This was a common practice at the time: Copies of important letters could be made for political or administrative reasons. The writing ink of these letters is therefore the one used by AF. Focusing only on AF's handwritten letters, it can be seen that there is a clear separation between the ink compositions before and after December 1791. Figure 10 shows the graph of the Zn/Fe ratio versus Cu/Fe ratio because these three elements are the most discriminating. Between August and December 1791, the composition of the inks seemed to vary quite often. For this period, it is difficult to make precise subsets because the limited number of points of analysis does not affirm categorically that the inks belong to the same group. The variability of the composition of the inks within a letter can be important: Perhaps it is due to decantation phenomena in the ink pot, which might induce a slight progressive change in ink composition, as the letter is being written (and also according to the depth in the pot from which the ink was taken), or perhaps it is due to variations in the composition of the support, which always contains a little iron.

In this period, for AF's handwritten letters, approximately three subsets could be identified (Fig. 10)—A: the letter of 13 August 1791, B: letters from 8 to 12 October 1791, and C: letters from 13 to 29 October 1791. After December (D: from 1 December 1791 to 3 May 1792), all the letters seem to have a very similar ink composition. These variations in composition may be the result of ink supply problems, but it is difficult to explain why the composition stabilized from December 1791. This could perhaps be correlated with the arrival of a secretary in the service of the Count between November and December, who might have been in charge of the supply or preparation of the inks. The composition of all redaction inks for MA's and AF's handwritten letters sometimes varies from one letter to another, but it always appears to be comparable to the composition of one of the subsets defined above. For example, the letters 1_1_8 and 1_2_8 (writing ink corresponding to group B) could have been redacted by the ink of group C and the letter 1_1_9 (writing ink corresponding to group C) by the ink of group D. This is unlikely to be a coincidence. Moreover, the redaction ink systematically belongs to one of the posterior subsets (or to the same subset) than that of the writing ink, which is compatible with the temporality of censorship (necessarily posterior to writing). AF would thus be the author of the redactions, which he would have carried out on different occasions shortly after (but not immediately after) having recopied the letters. As the composition of the ink changed quite often in the period from August to December 1791, the writing and redaction ink on the same letter is not necessarily the same, even if applied only a few days after the copying of the letters, making it possible to read the censored sections by the methodology described above.

This seems to be confirmed by the letter 1_1_8, which shows the addition of a text (above the redacted line) from AF's hand with an ink of the same composition as the redaction ink (Fig. 2A). AF would have added these few words above the redacted passage so that the meaning of the sentence remained understandable if the letter had to be shown to a third party for political reasons while avoiding being compromising (initial text: "the letter of the 28th made my happiness"; apparent text: "the letter of the 28th reached me").

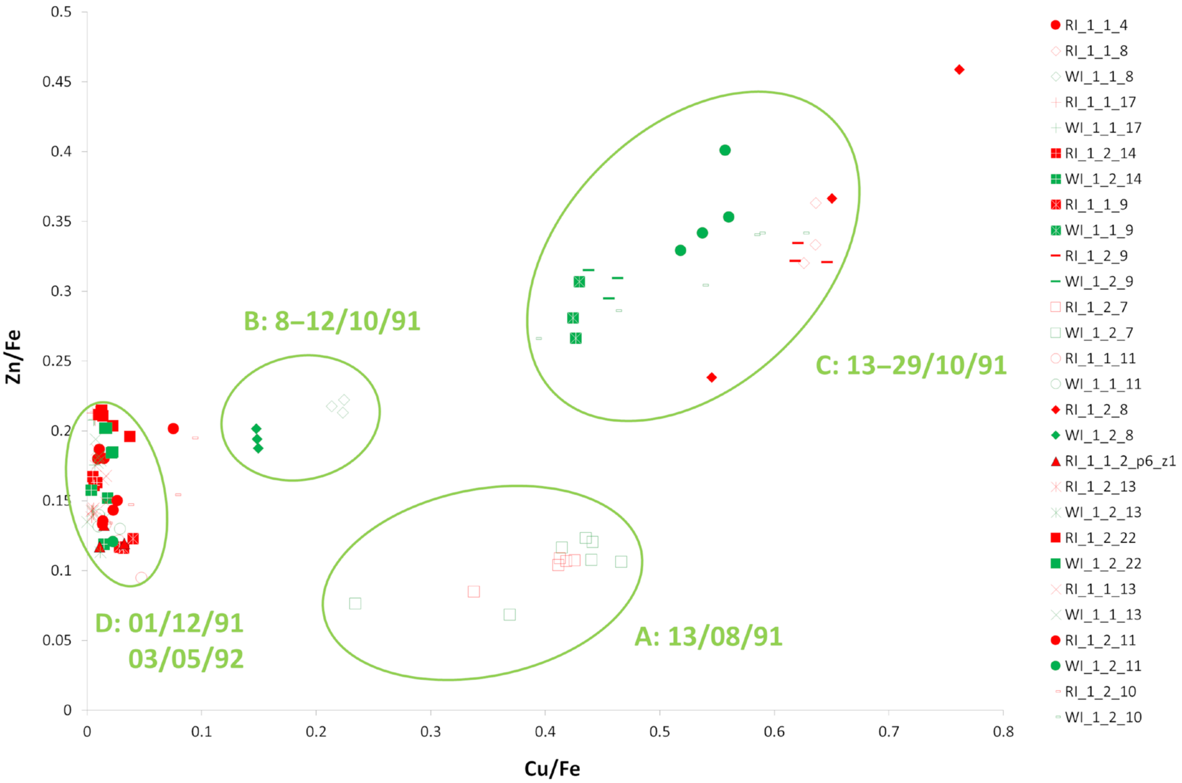

Fig. 10. Looking for the censor. Composition of writing inks (WI) for letters written by AF (in green) and redacting inks (RI) for all letters (in red) (measurement point: 100 μm, 200 s). Graph Zn/Fe versus Cu/Fe.

If our hypothesis is validated, this would explain why the last letters could not be read: If AF is really the censor, as the composition of the ink seems to be the same from December 1791 to May 1792, then the writing and redaction inks are identical, which makes it impossible to distinguish them by XRF spectroscopy.

Historical conclusions

Revealing the hidden fragments of text is less an opportunity to provide information on the relationship between Queen MA and the Count of Fersen than to restore the integrity of this correspondence and to reconstruct the chaotic history of its elaboration and transmission. To read under the censorship does not make it possible to know the truth about the nature of their feelings as the interpretation of texts is always questionable. But for the historian, this correspondence remains a precious testimony of a troubled time and of the way in which the tragic political events influence the transformation of the emotions and the exacerbation of the feelings visible in particular in the personal writings here in these redaction sections. Far from being an exception, this correspondence is rather representative of an experience widely shared by all social actors, which leads them to work through their emotions faced with the collapse of the "old world." However, the choice of vocabulary (beloved, tender friend, adore, madly) attests a particular relationship between MA and Fersen even if there is an influence of the revolutionary torment, which favors a certain emotional intensity.

It is also interesting how intimate aspects and purely political questions are intertwined—the letters mix sentimental effusions (which are redacted), comments on current events, and political considerations. From this point of view, the correspondence of MA and AF illustrates how the revolutionary period reaches paroxysm of the assimilation between love life and political life.

Another interest of the study, by identifying Fersen as the censor, is to see the importance of the letters received and sent to him whether by sentimental attachment or by political strategy. He decided to keep his letters instead of destroying them but redacting some sections, indicating that he wanted to protect the honor of the queen (or maybe also his own interests). In any case, these redactions are a way to identify the passages that he considered to be private. The mystery of these redacted passages that make this correspondence special is perhaps the reason that allowed this correspondence to be spared when the rest was largely destroyed.

Summary

Beyond the specific case of MA and AF correspondence, the development of methodological tools to address this type of unredaction challenge is of great importance for historical and forensic sciences. We showed that macroscopic elemental mapping can be a powerful tool to disentangle iron-gall ink superimposition. We also demonstrated how several data processing tools (sometimes derived from those applied for hyperspectral imaging) and combinations can greatly enhance the legibility of hidden contents, even when the original maps are not initially promising. This methodology successfully revealed the original contents of 8 of 15 redacted documents of the MA/Fersen correspondence that are held at the Archives Nationales and even gave clues as to the author of this censorship. The transcription of the totality of the paragraphs that were successfully unredacted is currently ongoing, supervised by curators and historians. It will be the subject of a different publication. Because the reading of the—sometimes degraded—final images can be difficult and is subject to interpretation, an important avenue for future work regards the automatic transcription of the writings with the help of machine learning (*44–47*).

MATERIALS AND METHODS

Two-dimensional macro-XRF scans of the letters were carried out with the Bruker M6 Jetstream. The instrument consists of a measuring head made of an Rh-target x-ray tube and a 30-mm^2 XFlash silicon drift detector mounted on an XY-motorized stage. The x-rays are focused through polycapillary optics, which allows us to set the beam size by changing the working distance. The pixel size of the elemental maps—determined by the step size—was the same as the beam size and set between 50 and 180 μm, depending on the required resolution. Voltage and current in the x-ray tube were set to 50 kV and 600 μA, respectively; dwell times varied between 100 ms and 3 s per pixel. A first phase of the project consisted of optimizing the acquisition parameters (beam size and dwell time per pixel) to allow the text to be read while minimizing analysis time. For this purpose, one word from each letter was analyzed under different conditions. The redacted passages of each letter were then fully analyzed with this choice of parameters. In certain cases, the initial dataset quality had to be improved from a spatial (pixel size) and spectral (dwell time) standpoint, requiring much longer (up to 30 times) acquisition durations. Ninety-five days of analysis were necessary for this project. Data were collected and treated with the Bruker M6 software. Additional data analyses and image treatments were carried out with ENVI (Excelis) and ImageJ software. Point analyses were performed with the multipoint option of the M6 Jetstream (dwell time of 200 s and beam size of 100 μm).

REFERENCES AND NOTES

1. A. Tonazzini, E. Salerno, Z. A. Abdel-salam, M. Abdel, L. Marras, A. Botto, B. Campanella, S. Legnaioli, S. Pagnotta, F. Poggialini, V. Palleschi, Analytical and mathematical methods for revealing hidden details in ancient manuscripts and paintings: A review. *J. Adv. Res.* **17**, 31–42 (2019).
2. M. Alfeld, L. de Viguerie, Recent developments in spectroscopic imaging techniques for historical paintings—A review. *Spectrochim. Acta Part B At. Spectrosc.* **136**, 81–105 (2017).
3. J. R. Duivenvoorden, A. Käyhkö, E. Kwakkel, J. Dik, Hidden library: Visualizing fragments of medieval manuscripts in early-modern bookbindings with mobile macro-XRF scanner. *Herit. Sci.* **5**, 6 (2017).
4. S. Saverwyns, C. Currie, E. Lamas-Delgado, Macro X-ray fluorescence scanning (MA-XRF) as tool in the authentication of paintings. *Microchem. J.* **137**, 139–147 (2018).
5. U. Bergmann, K. T. Knox, Pseudo-color enhanced x-ray fluorescence imaging of the Archimedes Palimpsest, in *Proceedings of SPIE-IS&T Electronic Imaging, Document Recognition and Retrieval XVI*, K. Berkner, L. Likforman-Sulem, Eds. (SPIE, 2009), vol. 7247, 724702-1-13.
6. E. Pouyet, S. Devine, T. Grafakos, R. Kieckhefer, J. Salvant, L. Smieska, A. Woll, A. Katsaggelos, O. Cossairt, M. Walton, Revealing the biography of a hidden medieval manuscript using synchrotron and conventional imaging techniques. *Anal. Chim. Acta* **982**, 20–30 (2017).
7. F. Gabrieli, K. A. Dooley, J. G. Zeibel, J. D. Howe, J. K. Delaney, Standoff mid-infrared emissive imaging spectroscopy for identification and mapping of materials in polychrome objects. *Angew. Chemie Int. Ed.* **57**, 7341–7345 (2018).
8. R. L. Easton, W. A. Christens-Barry, K. T. Knox, Spectral image processing and analysis of the Archimedes Palimpsest, in *Proceedings of the 2011 19th European Signal Processing Conference* (IEEE, 2011), pp. 1440–1444.
9. K. T. Knox, R. L. Easton, W. Christens-Barry, Image restoration of damaged or erased manuscripts, in *Proceedings of the 16th European Signal Processing Conference* (IEEE, 2008), pp. 1–5.
10. F. Pottier, A. Michelin, S. Kwimang, C. Andraud, F. Goubard, B. Lavédrine, Macroscopic reflectance spectral imaging to reveal multiple and complementary types of information for the non-invasive study of an entire polychromatic manuscript. *J. Cult. Herit.* **35**, 1–15 (2019).
11. F. Mercuri, N. Orazi, I. Industriale, T. Vergata, Pulsed thermography applied to the study of cultural heritage. *Appl. Sci.* **7**, 1010 (2017).
12. M. Picollo, K. Fukunaga, J. Labaune, Obtaining noninvasive stratigraphic details of panel paintings using terahertz time domain spectroscopy imaging system. *J. Cult. Herit.* **16**, 73–80 (2015).
13. A. Giacometti, A. Campagnolo, L. MacDonald, S. Mahony, S. Robson, T. Weyrich, M. Terras, A. Gibson, The value of critical destruction: Evaluating multispectral image processing methods for the analysis of primary historical texts. *Digit. Scholarsh. Humanit.* **32**, 101–122 (2017).
14. A. Tonazzini, Color space transformations for analysis and enhancement of ancient degraded manuscripts. *J. Pattern Recognit. Image Anal.* **20**, 404–417 (2010).
15. M. Alfeld, M. Wahabzada, C. Bauckhage, K. Kersting, G. Wellenreuther, G. Falkenberg, Non-negative factor analysis supporting the interpretation of elemental distribution images acquired by XRF. *J. Phys. Conf. Ser.* **499**, 012013 (2014).
16. D. Stromer, V. Christlein, C. Martindale, P. Zippert, E. Haltenberger, T. Hausotte, A. Maier, Browsing through sealed historical manuscripts by using 3-D computed tomography with low-brilliance X-ray sources. *Sci. Rep.* **8**, 15335 (2018).
17. W. B. Seales, C. S. Parker, M. Segal, E. Tov, P. Shor, Y. Porath, From damage to discovery via virtual unwrapping: Reading the scroll from En-Gedi. *Sci. Adv.* **2**, e1601247 (2016).
18. F. Pottier, A. Michelin, L. Robinet, Recovering illegible writings in fire-damaged medieval manuscripts through data treatment of UV-fluorescence photography. *J. Cult. Herit.* **36**, 183–190 (2019).
19. A. Tournié, K. Fleischer, I. Bukreeva, F. Palermo, M. Perino, A. Cedola, C. Andraud, G. Ranocchia, Ancient Greek text concealed on the back of unrolled papyrus revealed through shortwave-infrared hyperspectral imaging. *Sci. Adv.* **5**, eaav8936 (2019).
20. U. Bergmann, P. Manning, R. Wogelius, Chemical mapping of paleontological and archeological artifacts with synchrotron X-rays. *Annu. Rev. Anal. Chem.* **5**, 361–389 (2012).
21. C. S. Silva, M. F. Pimentel, R. S. Honorato, C. Pasquini, J. M. Prats-Montalbán, A. Ferrer, Near infrared hyperspectral imaging for forensic analysis of document forgery. *Analyst* **139**, 5176–5184 (2014).
22. F. G. France, Advanced spectral imaging for noninvasive microanalysis of cultural heritage materials: Review of application to documents in the U.S. Library of Congress. *Appl Spectrosc.* **65**, 565–574 (2011).
23. A. Stijnman, Iron gall inks in history: Ingredients and production, in *Iron Gall Inks: On Manufacture, Characterisation, Degradation, and Stabilisation*, J. Kolar, M. Strlic, Eds. (National And University Library, 2006), pp. 25–69.
24. L. Glaser, D. Deckers, The basics of fast-scanning XRF element mapping for iron-gall ink palimpsests, in *Proceedings of the Conference on Natural Sciences and Technology in Manuscript Analysis*, C. Brockmann, F. Michael, O. Hahn, B. Neumann, I. Rabin, Eds. (University of Hamburg, 2014), pp. 104–111.
25. O. Hahn, W. Malzer, B. Kanngiesser, B. Beckhoff, Characterization of iron-gall inks in historical manuscripts and music compositions using x-ray fluorescence spectrometry. *X-Ray Spectrom.* **33**, 234–239 (2004).
26. D. Deckers, L. Glaser, Zum Einsatz von Synchrotronstrahlung bei der Wiedergewinnung gelöschter Texte in Palimpsesten mittels Röntgenfluoreszenz, in *Kodikologie und Paläographie im Digitalen Zeitalter 2 / Codicology and Palaeography in the Digital Age 2*, F. Fischer, B. Assmann, Eds. (Norderstedt, Schriften des Instituts für Dokumentologie und Editorik-Band3, 2010), pp. 181–190.
27. M. Mantler, M. Schreiner, X-ray fluorescence spectrometry in art and archaeology. *X-Ray Spectrom.* **29**, 3–17 (2000).
28. M. Mantler, M. Schreiner, F. Weber, R. Ebner, F. Mairinger, An X-ray spectrometer for pixel analysis of art objects. *Adv. X-Ray Anal.* **35**, 987–993 (1991).
29. M. Schreiner, M. Mantler, F. Weber, R. Ebner, F. Mairinger, A new instrument for the energy dispersive X-ray fluorescence analysis of objects of art and archaeology. *Adv. X-Ray Anal.* **35**, 1157–1163 (1992).
30. G. Van der Snickt, K. A. Dooley, J. Sanyova, H. Dubois, J. K. Delaney, E. M. Gifford, S. Legrand, N. Laquiere, K. Janssens, Dual mode standoff imaging spectroscopy documents the painting process of the Lamb of God in the *Ghent Altarpiece* by J. and H. Van Eyck. *Sci. Adv.* **6**, eabb3379 (2020).
31. A. van Loon, P. Noble, D. de Man, M. Alfeld, T. Callewaert, G. Van der Snickt, K. Janssens, J. Dik, The role of smalt in complex pigment mixtures in Rembrandt's Homer 1663: Combining MA-XRF imaging, microanalysis, paint reconstructions and OCT. *Herit. Sci.* **8**, 90 (2020).
32. P. Ricciardi, S. Legrand, G. Bertolotti, K. Janssens, Macro X-ray fluorescence (MA-XRF) scanning of illuminated manuscript fragments: Potentialities and challenges. *Microchem. J.* **124**, 785–791 (2016).
33. J. Dik, K. Janssens, G. Van Der Snickt, L. van der Loeff, K. Rickers, M. Cotte, Visualization of a lost painting by Vincent van Gogh using synchrotron radiation based X-ray fluorescence elemental mapping. *Anal. Chem.* **80**, 6436–6442 (2008).

34. M. Alfeld, G. Van der Snickt, F. Vanmeert, K. Janssens, J. Dik, K. Appel, L. van der Loeff, M. Chavannes, T. Meedendorp, E. Hendriks, Scanning XRF investigation of a Flower Still Life and its underlying composition from the collection of the Kröller–Müller Museum. *Appl. Phys. A* **111**, 165–175 (2013).
35. F. P. Romano, C. Caliri, P. Nicotra, S. Di Martino, L. Pappalardo, F. Rizzo, H. C. Santos, Real-time elemental imaging of large dimension paintings with a novel mobile macro X-ray fluorescence (MA-XRF) scanning technique. *J. Anal. At. Spectrom* **32**, 773–781 (2017).
36. E. Ravaud, L. Pichon, E. Laval, V. Gonzalez, M. Eveno, T. Calligaro, Development of a versatile XRF scanner for the elemental imaging of paintworks. *Appl. Phys. A* **122**, 17 (2016).
37. F.-P. Hocquet, H. Calvo del Castillo, A. Cervera Xicotencatl, C. Bourgeois, C. Oger, A. Marchal, M. Clar, S. Rakkaa, E. Micha, D. Strivay, Elemental 2D imaging of paintings with a mobile EDXRF system. *Anal. Bioanal. Chem.* **399**, 3109–3116 (2011).
38. M. Alfeld, J. C. Vaz Pedroso, M. H. van Eikema Hommes, G. Van der Snickt, G. Tauber, J. Blaas, M. Haschke, K. Erler, J. Dik, K. Janssens, A mobile instrument for in situ scanning macro-XRF investigation of historical paintings. *J. Anal. At. Spectrom.* **28**, 760–767 (2013).
39. M. Alfeld, MA-XRF for historical paintings: State of the art and perspective. *Microsc. Microanal.* **26**, 72–75 (2020).
40. R. Bellman, *Dynamic Programming* (Princeton Univ. Press, 1957).
41. Z. Dabiri, S. Lang, Comparison of independent component analysis, principal component analysis, and minimum noise fraction transformation for tree species classification using APEX hyperspectral imagery. *ISPRS Int. J. Geo-Inf.* **7**, 488 (2018).
42. A. A. Green, M. Berman, P. Switzer, M. D. Craig, A transformation for ordering multispectral data in terms of image quality with implications for noise removal. *IEEE Trans. Geosci. Remote Sens.* **26**, 65–74 (1988).
43. N. Keshava, J. F. Mustard, Spectral unmixing. *IEEE Signal Process. Mag.* **19**, 44–57 (2002).
44. D. Firmani, P. Merialdo, M. Maiorino, E. Nieddu, Towards knowledge discovery from the vatican secret archives. In Codice Ratio - Episode 1: Machine transcription of the manuscripts, in *Proceedings of the 24th ACM SIGKDD International Conference on Knowledge Discovery Data Mining* (Association for Computing Machinery, 2018), pp. 263–272.
45. M. Ibn Khedher, H. Jmila, M. A. El-Yacoubi, Automatic processing of historical arabic documents: A comprehensive survey. *Pattern Recognit.* **100**, 107144 (2020).
46. J. A. Sánchez, V. Romero, A. H. Toselli, M. Villegas, E. Vidal, A set of Benchmarks for handwritten text recognition on historical documents. *Pattern Recognit.* **94**, 122–134 (2019).
47. N. D. Cilia, C. De Stefano, F. Fontanella, M. Molinara, A. Scotto di Freca, What is the minimum training data size to reliably identify writers in medieval manuscripts? *Pattern Recognit. Lett.* **129**, 198–204 (2020).

Acknowledgments: We thank the French national Archives for granting permission to study queen Marie-Antoinette correspondence with count Axel von Fersen. We would like to thank I. Aristide-Hastir, R. Lheureux, and B. Geay at the Archives Nationales for help to carry out the project. Many thanks to P. Lemaigre-Gaffier for historical perspective on this correspondence and, more generally, on the writings during the Revolution. We also thank O. Belhadj-Khlaifi and M. Radepont-Kolin for technical support on the XRF scanner, as well as S. Vaiedelich, C. Marland, and S. Petit for facilitating our experiments at the Cité de la Musique. Many thanks also to F. Kergourlay for the preliminary study of the letters. **Funding:** This project was funded with a post-doctoral fellowship of the Fondation des Sciences du Patrimoine/LabEx PATRIMA (ANR-10-LABX-0094-01). **Author contributions:** A.M. and C.A. were responsible for the project administration and for the funding acquisition. F.P. scanned the letters, and A.M. participated in the experiment. F.P. and A.M. performed the data analysis. Both F.P. and A.M. wrote the manuscript. C.A. revised and finalized it. **Competing interests:** The authors declare that they have no competing interests. **Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data can be accessed at https://datadryad.org/stash/share/7f1P-yeTk_k4jhGV4xYlu0ai5hmOjYytlW1bJA5MTqE.

Submitted 5 January 2021
Accepted 10 August 2021
Published 1 October 2021
10.1126/sciadv.abg4266

Citation: A. Michelin, F. Pottier, C. Andraud, 2D macro-XRF to reveal redacted sections of French queen Marie-Antoinette secret correspondence with Swedish count Axel von Fersen. *Sci. Adv.* **7**, eabg4266 (2021).

Science Advances

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Sci. Adv., 7 (40), eabg4266. • DOI: 10.1126/sciadv.abg4266