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REVIEW **Open Access**

X-ray fluorescence (XRF) on painted heritage objects: a review using topic modeling

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Abstract

This review presents the computational method of topic modeling to identify core topics and time trends in research on X-ray fluorescence (XRF) and its application to cultural heritage. Topic modeling is an approach to text mining based on unsupervised machine learning, which helps to determine core topics within a vast body of text. Due to the large amount of published work on X-ray fluorescence in the area of cultural heritage, traditional literature review has become impractical, inefficient, time-consuming, and potentially less reliable. Therefore, it is important to take stock of which topics have been core to such research and whether specific time trends can be identified within them. Using topic modeling, this review aims to reveal core topics and trends in research on XRF analysis of painted heritage objects by examining 982 articles collected from Web of Science. Within this dataset of articles, ten topics have been identified. The identified topics can be clustered in three main categories: the methods used, the objects studied, and the specific materials studied. In terms of trends in topic share since 2010, it is especially noteworthy to see that the share of articles focused on the identification and study of painting materials and techniques has more than doubled. Similarly, another impressive increase can be observed for articles centered on advanced imaging spectroscopic techniques, such as macro X-ray fluorescence (MA-XRF) and reflectance hyperspectral imaging, for the study of easel paintings. The share of attention within XRF literature given to imaging spectroscopic techniques tripled between 2010 and 2017, though stabilizing in the subsequent years. Conversely, the share of articles which specifically deal with the development and improvement of energy dispersive X-ray fluorescence (ED-XRF) spectroscopic techniques (i.e., portable ED-XRF, confocal micro-XRF, micro-grazing exit XRF) for the elemental analysis (including elemental depth profiling) of painted heritage objects has declined sharply.

Keywords X-Ray fluorescence, Cultural heritage, Painting materials, Painting techniques, topic modeling, Literature review, Computational methods, Machine learning, Text mining, Latent dirichlet allocation

Introduction

The role of techniques from the natural sciences to analyze material aspects of cultural heritage objects is now well established within the fields of archaeology, art history, conservation, and heritage science. Developments of mobile and portable instruments around the turn of the millennium resulted in a substantial growth in the usage of such analytical techniques within these research fields. Consequently, the body of literature on different analytical techniques and their application to cultural heritage increased exponentially. Among the most cited techniques is X-ray fluorescence (XRF), which is commonly used for the elemental characterization of painted heritage objects due to its ability to detect a wide range of painting materials, especially pigments, in a non-destructive, sensitive, and efficient way [1, 2].

The popularity of XRF for the elemental analysis of pictorial layers has also increased, commensurately resulting in a substantial growth in literature on the subject.

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This is, in part, due to the 2013 introduction of the M6 Jetstream, a commercial mobile macro X-ray fluorescence (MA-XRF) scanning instrument by Bruker [3–5]. This non-invasive X-ray fluorescence scanning instrument generates elemental distribution images of entire painted surfaces instead of conventional analytical spectra and graphs; thus providing a more comprehensive basis even for non-experts to acquire insights into the gathered elemental data. More than 210 research articles on applications of MA-XRF scanning in the study of cultural heritage objects alone can be found via Web of Science (WoS).[1] Conclusively, this upward trend in terms of scholarly publications requires an efficient method to review the extensive body of knowledge on XRF techniques in painting materials characterization, which can be expected to keep expanding rapidly in the coming decades. In other words, it is essential to find an efficient and reliable way to address the question: what are core topics and time trends that can be observed in this vast corpus of research? This paper aims to answer this research question by employing topic modeling as a computational method to conduct a review of the XRF literature. Following a systematic data collection process in WoS, 982 articles were identified on the subject. Analyzing all these articles individually would be painstakingly inefficient, require a massive time cost to identify and continuously update code books and labels, and could be influenced by subjective assessments and errors due to human coding [6, 7].

Recent advances in text mining using machine learning have been shown in a range of other fields to be efficient tools to review an extensive corpus of research [8]. Specifically, topic modeling is a computational approach to text mining on a large amount of text using unsupervised machine learning. It is particularly efficient in singling out topics in a large body of literature as well as time trends with regards to the share of attention to those topics [9]. Hence, the method of topic modeling is ideally suited to help answer the proposed research question.

The current paper begins with an explanation of the methods guiding the review, including a discussion of both the data collection process and the topic modeling. Next, the results of the topic modeling are presented, which are the identified core topics and trends in the share of attention to those topics over time. In conclusion, the implications of the findings and the relevance of the review method for the cultural heritage field are discussed.

Methods

Data collection process

The process of data collection consisted of several steps to form the dataset of 982 articles. First, a search was conducted in Web of Science Core Collection by using following search formula (TS = "X-ray fluorescence" AND TS = paint*).[2] The search results were refined by setting the document type filter to article and the language filter to English. The Full Record of the identified articles were subsequently imported into an Excel spreadsheet.[3] Next, the Article Title, Abstract, and Source Title of all articles were checked manually to ensure data were optimal for the topic modeling analysis. Articles were removed from the Excel spreadsheet if they were not published in English or were published in journals irrelevant to heritage science and related fields (e.g., American Journal of Diseases of Children, Environmental Health Perspectives, Forensic Science International). Moreover, articles dealing with an irrelevant topic, or employing other related or non-related spectroscopic and imaging techniques (e.g., Raman spectroscopy, proton-induced X-ray emission (PIXE), X-ray powder diffraction (XRD), diffuse reflectance spectroscopy (DRS), multispectral reflectance imaging), and thus not the analytical technique of XRF (with or without other techniques), were also eliminated from the Excel data file.[4] In some cases, the full article was consulted when the Title, Abstract, and Source Title did not provide enough information to decide whether it should be removed or not. The outcome of this data collection process was an Excel spreadsheet containing 982 articles, as well as their meta-information.

Topic modeling

Topic modeling is employed to analyze the identified 982 articles, specifically, the article abstracts. Latent dirichlet allocation (LDA) is used for this review process. For that reason, the Excel spreadsheet was imported into the general purpose statistical software package Stata (the name

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[1] The search was conducted on 1 October 2023 in the Web of Science Core Collection, which generated 214 results. The used search formula was (TS = MA-XRF OR TS = "MA XRF" OR TS = "macro x-ray fluorescence" OR TS = "macroscopic X-ray fluorescence"). The document type filter was set to articles, and the language filter was set to English.

[2] The search was conducted on 14 August 2023 in the Web of Science Core Collection.

[3] This was done by selecting first the Export Records to Excel option, subsequently fill in the 'Records from … to …' in the Record Options, which enables the export of 1000 records at a time.

[4] During the data refinement process, it became clear that the generated Excel spreadsheet contained about 50 research articles, in which the employed set of instrumentation does not include any XRF-based techniques. About 30 of these articles were included in the generated dataset because the keyword "X-ray fluorescence" was incorporated erroneously in the keywords plus section of Web of Science.

being a combination of statistics and data).[5] The Stata data file has as such 982 observations, each observation (or row) being a specific article and each column representing a specific variable extracted from WoS indicating meta-information about the article.

To prepare the corpus of abstracts for LDA topic modeling, several additional steps of data preparation were undertaken in Stata. First, the variable abstract is considered by Stata as a string-variable, indicating it is filled with text. Subsequently, as a next step, non-alphanumerical characters needed to be removed from the abstracts as these can interfere with LDA's clustering. The characters include: periods (.), exclamation points (!), question marks (?), double points (:), semi-colons (;), colon (,), brackets (()) and blanks (). Now, to eliminate these characters within the text data, several Stata commands were implemented as described by Schwarz [9]. In a following phase, stop words were also removed from the abstracts as these could influence LDA's clustering due to their heavy usage in all text.

A pre-defined list of global stop words was used to remove these words from the text. The list includes following stop words: "a able about across after all almost also am among an and any are as at be because been but by can cannot could dear did do does either else ever every for from get got had has have he her hers him his how however i if in into is it its just least let like likely may me might most must my neither no nor not of off often on only or other our own rather said say says she should since so some than that the their them then there these they this is to too was us wants was we were what when where which while who whom why will with would yet you your". Eliminating the stop words from the article abstracts was the final step in the data preparation process.

The prepared corpus of abstracts was then subjected to LDA in Stata. The reason is that LDA as a machine-learning topic model extracts so-called latent topics from a given corpus of text using unsupervised machine learning. The term latent is used in relation to LDA to convey something that exists in the data but has not yet been made explicit. LDA enables the automatic categorization of large amounts of text into a number of clusters—the number can be chosen by the user—that share a similar content. Such clusters are labeled as topics [10].

While LDA can be applied to various types of textual data, the explicit focus of this article is its capacity to analyze literature. As a literature review tool, LDA has been deployed in a variety of fields, for instance to review studies in medicine [11], public administration [12], economics, [9] and business studies [13]. It has been found that LDA is especially effective as a literature review tool when used for the analysis of a large amount of article abstracts. The topic modeling technique performs particularly well on text that has at least about 50 to 100 words, which abstracts typically do [9]. Text that is shorter in length, like article titles or tweets for instance, is less suitable for LDA as there is not enough information about the co-occurrence of words [14, 15].

Similarly, most LDA-based reviews focus on article abstracts, as is done here, as opposed to full text. There are several reasons for this focus. First, full texts of articles are not always available particularly when they are behind paywalls. Second, full texts especially for older articles are not always in a format that allows text extraction. Third, databases like WoS do not typically allow downloads of large numbers of full texts, which again reduces efficiency as it requires manual processing to make the text suitable for analysis. And, finally, previous research on topic modeling suggests that differences between LDA on abstracts or full texts are especially prominent in smaller datasets samples—the larger the dataset the more similar results become [16–27]. Hence, the choice to concentrate on article abstracts to conduct a review of the XRF literature is based on these observations.

What LDA in essence does to cluster text documents into topics is employing a probabilistic model. The assumption is that text which belongs to the same topic is more likely to use words that are similar to each other, that is, co-occurrence of words. LDA then, as explained earlier by Schwarz, "uses the co-occurrences of words to describe each topic as a probability distribution over words and to describe each document as a probability distribution over topics [9]." The probabilistic model is thus going to explain the text in the documents in the form of a likelihood function. In the next step, LDA utilizes an inference algorithm because maximizing the likelihood function is unfeasible [18].

For this review paper, the Gibbs sampler is employed as an inference algorithm by running LDA in Stata based on the ldagibbs command developed by Schwarz [19]. A Markov Chain Monte Carlo algorithm underlies Gibbs sampler, which is a Bayesian technique, implying that new samples are drawn repeatedly conditional on the other data [18].[6] The default of ten topics was selected to result from the analysis, other parameters

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[5] StataCorp. 2023. Stata Statistical Software: Release 18. College Station, TX: StataCorp LLC.

[6] For further information about mathematical modeling of both the probabilistic model, inference, and Markov Chain Monte Carlo algorithm, we refer to Schwarz C. Ldagibbs: A Command for topic modeling in Stata Using Latent Dirichlet Allocation. The Stata Journal. 2018; 18(1): 101–117.

similarly followed default or standard settings for this type of data:

- alpha (0.25): This value should be between 0 and 1, it indicates the prior for the topic-probability distribution with 0.25 being the default.
- beta (0.1): Similarly, this indicates the prior for the world-probability distribution and should be between 0 and 1, with 0.1 being the default.
- seed (3): This command indicates the seed linked to the random-number generator to ensure results' reproducibility, with 3 being standard.
- burnin_iter (1000): This command indicates the number of iterations that the Gibbs sampler is expected to run as burn-in period, with 1000 being standard.
- samples (10): This command indicates the amount of samples that the algorithm should collect after the burn-in period, with 10 samples needed for robust results.
- sampling_iter (50): This indicates the amount of iterations that the Gibbs sampler should ignore between individual samples, which ensures statistical independence between the samples—the default is 50.
- min_char (5): This command is used to remove short words from the text, in this case words with less than 5 characters as these could interfere with the analysis.

Importantly, the normalize command was also added to make sure that each of the produced variables would contain the probability of an abstract belonging to the specific topic. Similarly, the mat_save command was specified to save, and thus retrieve the word probability matrix. The word probability matrix presents the most frequent words within each of the ten topics, and thus contains indispensable information to label the topic. The analysis resulted in ten new variables, or topics, within the dataset: topic_prob1 to topic_prob10. These variables indicate the probability of an article belonging to the specific topic.

The outcome of the entire process of topic modeling is twofold. First, a probability score for each article on each topic (which are included in the dataset as new variables) is obtained. For instance, if you choose to identify 10 topics, LDA will result in 10 new variables in the dataset and each cell of these variables represent the probability score of an article belonging to that topic. Second, a word probability matrix is ensued, which can be loaded into Stata to identify the most frequent words used within a topic. Both these outputs provide crucial information for naming or labeling the topics and doing additional analyses to identify trends over time within these topics.

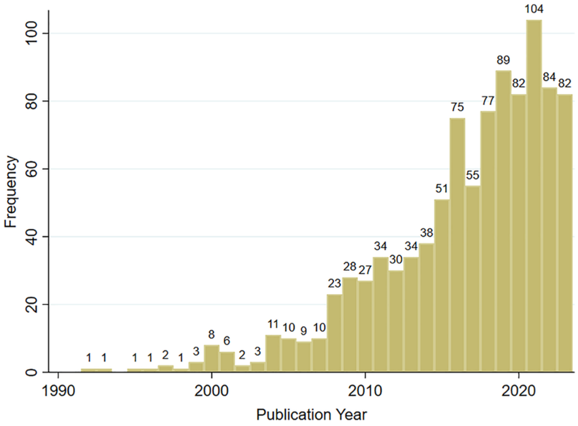

Fig. 1 The number of published articles on XRF analysis of painted heritage objects

Results

The dataset of 982 articles gathered from WoS provides already some basic bibliometric information in terms of the evolution in the number of published articles, the main journals, and thus the different fields contributing to the body of XRF literature. Figure 1 shows the evolution in the number of published articles from 1992 to 2023. From 2004 onwards, the number of publications has been increasing steadily year by year, while from 2010 to 2023 the body of literature enlarged rapidly. Hitherto the highest number of published articles was realized in 2021 with a total of 104 papers. The five scientific journals that have contributed the most to this growing body of literature have been Heritage Science (72 articles), X-ray Spectrometry (68 articles), Microchemical Journal (60 articles), Journal of Cultural Heritage (47 articles) and Applied Physics A: Materials Science and Processing (46 articles). In addition to cultural heritage, the fields of chemistry, physics and materials science significantly add to the research topic of XRF analysis of painted heritage artefacts.

Analysis of the topics

Having produced the topic probability variables, the next step is to make sense of the identified topics and add specific labels to them. This requires human intervention, interpretation and sensemaking of the results. To do so, two outputs are produced. First, using the earlier saved word probability matrix, the five most frequent words within a topic were listed. Table 1 contains this output, and words are ranked based on their word probability score (highest to lowest).

The identified words per topic seem to indicate that the ten topics can be grouped in three broad clusters:

Table 1 Five most frequent words per topic (T)

T1	T2	T3	T4	T5
X-ray (0.0255)	Archaeological (0.0107)	Paints (0.0191)	X-ray (0.0314)	Mural (0.0120)
Analysis (0.0205)	Pottery (0.0105)	Paint (0.0141)	Spectroscopy (0.0284)	Archaeological (0.0083)
Fluorescence (0.0151)	Production (0.0091)	Spectroscopy (0.0090)	Microscopy (0.0280)	Walls (0.0075)
Elemental (0.0086)	Painted (0.0091)	Organic (0.0082)	Electron (0.0211)	Wooden (0.0072)
Method (0.0079)	Study (0.0086)	Paintings (0.0075)	Scanning (0.0200)	Different (0.0066)
T6	**T7**	**T8**	**T9**	**T10**
Pigments (0.0411)	Glass (0.0225)	Techniques (0.0210)	Painting (0.0223)	Paint (0.0212)
White (0.0230)	Century (0.0122)	Painting (0.0188)	Imaging (0.0222)	Degradation (0.0153)
Pigment (0.0203)	Manuscript (0.0095)	Materials (0.0181)	ma-xrf (0.0151)	X-ray (0.0099)
Yellow (0.0192)	Museum (0.0088)	Paintings (0.0149)	Paint (0.0120)	Paints (0.0096)
Green (0.0164)	Medieval (0.0085)	Spectroscopy (0.0150)	Scanning (0.0110)	Surface (0.0085)

Word probability score between brackets

The order of the words is based on their word probability score, from highest to lowest

focused more on the methods used (T1, T4, T9, T10), the objects studied (T2, T5, T7), and the specific materials studied (T3, T6, T8). Within these broad clusters, following initial labels are proposed for the topics solely based on an analysis of the identified words per topic:

- Methods:
  - T1: X-ray fluorescence (XRF) techniques for elemental analysis
  - T4: X-ray spectroscopy techniques (including SEM–EDX)
  - T9: Imaging techniques for the study of paintings (including MA-XRF scanning)
  - T10: X-ray techniques for the study of paint degradation
- Objects
  - T2: Archaeological pottery
  - T5: Different archaeological artefacts (including walls, mural paintings, wooden objects)
  - T7: Glass and illuminated manuscripts
- Materials
  - T3: Paints
  - T6: Pigments
  - T8: Painting materials and techniques

Some preliminary insights can be derived from the ten topics generated by the word probability matrix. The topics comprised in the methods used cluster, particularly topic 4 (X-ray spectroscopy techniques) and topic 9 (imaging techniques for the study of paintings), show that the dataset of 982 articles contains findings from other analytical techniques than XRF. This is to be expected as in the field of heritage science specifically, the research trend of the last decade has been to adopt multi-analytical approaches for the study of painted heritage objects. This implies that XRF is often used together with several other analytical techniques to shed light on different aspects of the material composition and condition of painted heritage objects, and thus to yield complementary or more specific information. More precisely, as is revealed by the identified words of topic 9 (imaging techniques for the study of paintings), these techniques may involve imaging tools directly derived from XRF, like MA-XRF. However, as indicated by the identified words of topic 4 (X-ray spectroscopy techniques), it can also involve X-ray spectroscopy techniques closely related to XRF, such as scanning electron microscopy-energy dispersive X-ray analysis (SEM–EDS).

To further validate the proposed preliminary labels of the topics, a second output was produced. Specifically, using the earlier topic probability scores assigned to each article, the ten articles with the highest scores were listed within each topic. The titles of these articles and their abstracts were analyzed in detail to cross-validate the earlier proposed labels of the ten topics. The Stata output listing the articles has been added in Additional file 1: (SI.1) for full transparency.

In general, the cross-validation indicated that the three broad clusters of methods, objects and materials are largely tenable. Admittedly, this classification of the ten topics may appear too rigid given the interplay between the different topics. This becomes apparent when the aim of the article is, for example, to assess the potential of an analytical methodology for studying the chemical composition of the painted layers of polychrome sculpture in comparison with micro-destructive techniques [117]. As such, this article can fit within either cluster. However, the categorization of the different topics into the three clusters helps to get a general understanding of, on the one hand, the different painted objects, paint materials which have been examined with XRF, and the different approaches adopted in the XRF literature, on the other.

Further analysis of the ten article titles and abstracts per topic revealed that some labels required more refinement. Additionally, this analysis enabled the identification of main research purposes within a topic and simultaneously provided more detailed information about the recent tendency within the cultural heritage field to combine XRF spectroscopy techniques with other analytical tools for the study of painted heritage objects.

Finally, it can be derived from the analysis of the article titles and abstracts that the term ‘painted heritage objects’ covers a broad range of cultural assets. The listed ten articles per topic present findings from research conducted on easel paintings, wall paintings, graffiti, polychrome sculptures, polychrome architectural elements, painted enamels, painted glass windows, illuminated manuscripts, painted handscrolls, painted ceramics, painted lacquerware, colored woodblock prints, dyed leatherwork, historical paint samples, commercial artists’ color charts, colored woodblock prints, pigments, and modern manufactured paint materials (e.g., acrylic paints, ready-mixed house paints). However, most articles are not exclusively concerned with the study of painted heritage objects. Chiefly, when the purpose of the articles is to present a new research methodology, a new technological development, a review of an analytical technique or approach, or to report findings of various types of excavated earthenware or of a measurement campaign conducted in situ, painted artefacts are often discussed along with nonpainted heritage objects. Only two articles do not focus in any way on a painted heritage object but instead on nonpainted clay bricks [75] and bronze sculptures [90]. These two articles are contained in the initial dataset of 982 articles because the words ‘painted’ or ‘painting’ are included in the abstracts or WoS keywords plus. For instance, the abstract of the latter article dealing with XRF analysis of nonpainted bronze sculptures contained the word ‘painted’. The word refers in this instance to the painted, unbaked clay molds from which the studied bronze sculptures were cast.

The results of the analysis are further discussed per cluster in the following paragraphs. The analysis of the titles of the articles and their abstracts explores the question whether earlier proposed labels of the ten topics make sense in relation to the published research on XRF analysis of painted heritage objects.

The methods cluster

The topics of the methods cluster contains articles that focus on XRF techniques for the study of painted heritage objects, whether or not combined with other analytical techniques and with other types of heritage objects (e.g., bronzes, gold objects). The ten articles with the highest probability score under topic 1 (T1) consider XRF techniques for elemental analysis of layered heritage objects by discussing developments of XRF equipment and components [20–22], data acquisition and handling methods [23–26], and instrumentation improvements [27, 28]. Article 10 within this topic, for instance, introduces the silicon drift detector for portable ED-XRF devices [21], while article 9 provides a succinct overview of effective applications of portable ED-XRF for analyzing cultural heritage objects, including both painted and nonpainted artefacts (i.e., easel paintings, murals, bronzes, and metal objects) [26]. Articles 1 to 8, on the other hand, consider technological and methodological progresses of ED-XRF specifically in relation to the issue of poor informative depth of analytical technique for the non-invasive study of multilayered heritage objects (e.g., easel paintings, illuminated manuscripts, Japanese lacquerware, gilded objects). Proposed practical solutions include: the usage of a confocal setup with X-ray optics [27], a combined use of X-ray measurements and Monte Carlo simulations [23, 24], the fusion of XRF instruments (i.e., grazing exit XRF (GE-XRF) and confocal 3D-XRF) [20], and the development of quantification procedures [28]. Apart from these applied solutions, article 1 elaborates on the theoretical background of methods based on the differential attenuation of emitted X-rays to obtain depth-resolved spectroscopic information with XRF [29]. In a similar vein, article 5 provides an overview of different XRF principles and possibilities (e.g., changing the measurement geometry) for multilayer characterization by summarizing their advantages and limitations [30]. All ten articles of T1 were published in spectroscopic or more general physics and analytical chemistry journals, such as: X-ray Spectrometry, Review of scientific instruments, Radiation physics and chemistry and Nuclear Instruments & methods in Physics research section A—Accelerators spectrometers detectors and associated equipment.

Taking this all together, the five most frequent words of T1 are representative for the content of the ten articles. However, the type of heritage objects under study here is not limited to painted heritage objects. Also, in terms of research aim, the ten articles specifically focus on the development and optimalization of ED-XRF instruments and methods for the elemental analysis of multilayered heritage objects and not so much on presenting results from the elemental analysis of this type of objects. Therefore, the label can be more specific regarding the research aim and can be reformulated as follows: The development and optimization of XRF instruments and methods for elemental analysis of layered heritage objects.

T4 clusters articles that also deal with the compositional analysis of multilayered painted heritage objects (i.e., easel paintings, murals, decorative earthen plaster, painted and gilded statues, colored leather objects and fragments, decorated Chinese lacquerware) and paint samples, but the utilized analytical techniques are not limited to non-invasive XRF spectroscopy as is the case in T1. This is already indicated by the inclusion of the words microscopy, electron, and scanning, referring to the technique of SEM–EDS generated by the word probability matrix. Different than XRF, this analytical technique enables not only the study of chemical features but also the morphological characterization of both inorganic and organic materials. Moreover, it provides images of the sample surface at extreme high magnification. The articles of T4 adopt thus a micro-invasive and multi-analytical approach, in which the ensuing elemental data obtained with XRF from objects or samples is further supplemented with more spectroscopic information derived from SEM–EDS. In articles 2, 6, 7 and 10, portable ED-XRF is specifically employed as a diagnostic tool to guide sampling for the more extensive compositional analysis with SEM–EDS [32, 33, 35, 40].

The further analysis of the article titles and abstracts shows that the cluster's initial label 'X-ray spectroscopic techniques (including SEM–EDS)' does not quite cover the load in terms of the methods utilized. Next to spectroscopic techniques, other methods have been adopted in these ten studies to investigate the stratigraphy and microstructure of painted heritage objects in a comprehensive manner. Complementary microscopic and imaging techniques have been used, such as: optical microscopy (OM) [33–38], metallographic microscopy [37], macro photography, infrared photography (IRP) and infrared reflectography (IRR) [31, 32]. In article 3, the tree ring dating technique of dendrochronology is additionally included to corroborate the historical date initially assigned to a wooden support of a sixteenth-century altarpiece [31] while article 4 also makes use of sieve analysis to understanding the composition and properties of an earthen support layer of a fifth-fourteenth century Buddhist mural in China [34]. In addition to XRF and SEM–EDS, molecular spectroscopy techniques are used in all ten articles on microscopic samples. Pigments [38 & 40], minerals [33 & 39], and organic substances (e.g., binding media, dyes, carbon black) [33, 35–37] present in the sample stratigraphy are analyzed by means of Fourier transform infrared spectroscopy (FTIR), micro-Raman spectroscopy and surface-enhanced Raman spectroscopy (μ-SERS).

Finally, even micro-destructive techniques have been employed in five of the ten studies categorized under T4. In particular, pyrolysis gas chromatography mass spectrometry (PY-GC/MS) and high-performance liquid chromatography (HPLC) have been adopted for the characterization of organic media present in easel paintings [31, 32], wall paintings [33], decorative earthen plaster fragments [34] and colored leather objects [35]. In article 2, X-ray powder diffraction (XRD) was added to the set of spectroscopic and microscopic methods as well. This with the aim to identify the mineralogical phases of the preparatory and painting layers of decorative earthen plaster fragments derived from murals in the Petros and Paulos church in Ethiopia [33]. Thus, depending on the required compositional data, as well as the type of multilayered painted heritage object under examination, different sets of complementary analytical and imaging techniques have been utilized. In line with this observation, three different research purposes can be singled out within this group of articles: (1) to comprehensively study degradation processes of materials within the broader stratigraphy [34, 36, 37], (2) to characterize materials (e.g., pigments, binding media) and techniques per stratigraphic layer [31–33, 35], and (3) to analyze the chemical composition and microstructure of pigment present in paint samples [38–40].

Whereas the heritage objects examined in T4 are all painted, the research techniques employed in this group of ten articles are thus not limited to XRF as was the case for T1 (XRF instruments and methods for elemental analysis of layered heritage objects). In other words, the scope of the dataset of 982 articles collected from WoS is in fact broader than XRF analysis on painted heritage objects. As such, the current review simultaneously provides insights into the research tendency of the past ten years in the field of heritage science, and related fields like archaeometry, to adopt a multi-analytical approach to gain a comprehensive understanding of the materials, techniques, stratigraphic structure, micro-structure, and condition of painted heritage objects. Based on these observations, it can be concluded that the initial proposed label based on the five words provided by the topic modeling method is too narrow for T4 in terms of the various methods used in the ten articles, as well as their aims. A more fitting label thus is multi-analytical approaches for stratigraphic analysis of painted heritage objects.

The same applies for T10, which current label is too limited for the same reason. In this instance as well, multi-method analytical approaches are adopted. However, the aim and object focus of these studies are slightly different and more specific, that is, (1) to investigate degradation processes (i.e., photo-degradation, oxidation, migration processes) and alteration products of light-sensitive, inorganic yellow pigments used in oil paintings, as well as possible causes, such as, thermal factors (temperature and humidity), light exposure and the presence of a paint medium [41, 43, 44, 47–50], or (2) to develop or test a novel multi-analytical approach for this type of research [42, 45, 46]. The inorganic pigments examined here are arsenic sulfide pigments (e.g., realgar and orpiment) [41, 43], cadmium sulfide-based yellow pigments (cadmium yellow) [42, 44, 46–48] and lead chromate-based yellow pigments [45, 49, 50].

These studies employ for this purpose various high lateral resolution spectroscopic and imaging methods that make use of synchrotron radiation (SR): micro X-ray fluorescence (μ-XRF), micro X-ray absorption near-edge spectroscopy (μ-XANES), micro X-ray diffraction (μ-XRD), micro Fourier transform infrared (μ-FTIR), tomographic transmission X-ray microscopy, electron paramagnetic resonance (EPR) spectroscopy and 2D full-field X-ray near-edge structure (FF-XANES) imaging. The multi-method approach implemented in the ten studies can be solely based on a combination of these SR-based spectroscopic and imaging methods [41, 43, 47], with or without FTIR spectroscopy [42, 44–45, 48–50], diffuse reflectance ultraviolet–visible spectroscopy (UV–Vis) [44–46], and micro-Raman spectroscopy [49, 50]. For example, article 1 introduces a multi-analytical approach based on the combination of diffuse reflectance UV–Vis, attenuated total reflection (ATR) reflection mode FTIR, SR-based μ-XANES, μ-XRF and EPR spectroscopies for examining the consequences of changing relative humidity conditions before and after light exposure on the reactivity of a series of lead chromate-based pigments [45]. A quite similar multi-method is employed in article 3 [44]. Here UV–Vis and FTIR spectroscopy are used together with SR-based X-ray methods to identify possible causes of the degradation of cadmium yellows in oil paintings and their impact on the overall degradation process of these pictures. Moreover, two articles included in T10, the statistical method of principal component analysis (PCA) was carried out for data reduction [41 & 42]. In article 2, for instance, PCA and subsequent k-means clustering of multi-energy SR μ-XRF maps and μ-XANES were performed to identify the oxidation products of arsenic sulfide pigments and visualize their distribution in the multi-layered paint structure [41]. Finally, in terms of methods adopted in this set of articles, artificially aged model samples, mock-up paints and paintings and dry pigment powder were used instead of samples taken directly from historical oil paintings [43–45, 49]. This was to study and identifying the effects of different environmental conditions or of the paint medium (oil) on various pathways of degradation of inorganic yellow pigments.

The multi-method analytical approaches, which all include SR-based X-ray techniques, employed in the ten articles categorized under T10 allow a comprehensive and detailed analysis of micrometric degradation processes and products of inorganic yellow pigments in oil paintings, and to identify possible causes as well. Hence, the term 'X-ray' included in the word probability matrix does not reflect the various techniques typically adopted to investigate paint degradation processes. Nevertheless, the analysis of the abstracts of the ten articles reveals that the term points to the usage of SR-based X-ray methods, which has gained importance in the past two decades in this area of research. Also, the detailed analysis of the article titles and abstracts shows that the object focus—or more specifically, the material focus—is very specific, namely inorganic yellow pigments used in an oil medium. The terms 'paint' and 'paints' prompted by the word probability matrix again do not cover thus the specific research focus of the articles cluster. Therefore, the final label for T10 is as follows: Multi-analytical approach (including SR-based X-ray methods) for the study of degradation products, processes and causes of inorganic yellow pigments in oil paintings.

Contrary to T4 (multi-analytical approaches for stratigraphic analysis of painted heritage objects).and T10, the five words generated by the word probability matrix give enough information to name T9 correctly as can be concluded from the title and abstract analysis. This topic clusters articles whose research subject is spectroscopic imaging of easel paintings. Within this set of ten article, three different purposes can be singled out.

The first purpose is related to the usage of the imaging techniques on easel paintings, that is, to study their different strata composed of a variety of materials in more detail to identify previous conservation treatments and, above all, to address art historical questions, especially to understand the working methods and materials of artists, and to resolve authenticity matters. For example, the imaging technique of MA-XRF scanning was specifically developed to investigate hidden modifications or compositions (i.e., pentimenti) done by the artists themselves in easel paintings in a non-invasive manner [51]. In total, two articles within T9 use the tool of MA-XRF scanning for this specific purpose, although in combination with other spectral imaging methods, such as element-specific imaging techniques (i.e., neutron activation autoradiography (NAAR)) and hyperspectral imaging spectroscopy (i.e., infrared reflectance imaging spectroscopy) [52, 53]. The non-invasive approach of these imaging techniques notwithstanding, further analysis of cross-sectioned samples was considered desirable to study the stratigraphy in these studies [52, 53]. MA-XRF scanning is used as well in articles 3 and 8 for the chemical identification and mapping of artists' materials [53, 61] but for another reason. More precisely, the aim was to address issues of attribution through a better understanding of working methods and materials employed in the Old Master paintings under study.

Also included in T9 are two review articles that critically assess the application of spectroscopic imaging methods for the study of easel paintings. Article 4 summarizes findings on the application of hyperspectral imaging on paintings to identify and map artists' pigments, improve the visualization of preparatory sketches, and identify non-original material [55]. Article 9 examines dendrochronological potential of synchrotron-sourced X-ray fluorescence mapping to date panel paintings [56]. Other articles headed under T9 are concerned, in turn, with image processing methods. These methods include the development of spectral databases to train a one-dimensional (spectral) convolutional neural network for spectra classification and labeling [57–59], as well as of novel methods for data fusion of various types of technical images of easel paintings, such as multispectral reflective infrared images, X-radiographs, hyperspectral image cubes, and XRF image cubes [60]. In sum, the label defined earlier solely based on the word probability matrix reflects the research scope of the ten articles with the highest probability score particularly well and thus remains unaltered.

The objects cluster

The three topics categorized into the objects cluster give a general overview of the various types of cultural heritage objects that have been examined with XRF, often in combination with other analytical techniques. Conform the five words identified by the word probability matrix, research articles categorized in T2 are concerned with the compositional characterization of excavated painted pottery or pottery shards by using XRF spectroscopic tools, like portable and handheld ED-XRF spectrometer instrumentation [62–64], although in most instances not exclusively. In article 1, for instance, a geochemical assessment of ancient painted ceramics excavated in southern Turkey is performed through portable ED-XRF and neutron activation analysis [65]. This multi-analytical approach generates geochemical data allowing to distinguish between imported and locally produced pottery and offers as such new insights into exchange networks in the Ancient World. The other articles of T2 also concentrate on this issue of ceramic provenance [66], whether or not to also identify historical exchange networks [65–67, 69], to characterize production technology and developments [63, 64, 68, 70, 71], or to address questions of usage as well [62]. In addition to portable ED-XRF and neutron activation analysis, other employed analytical techniques here are: FTIR spectroscopy, micro-Raman spectroscopy, inductively coupled plasma optical emission spectrometry, optical microscopy, SEM–EDS, XRD, and Wavelength Dispersive X-ray fluorescence (WD-XRF) [67–71]. Given the fact that the study object of all ten articles categorized in T2 is excavated painted pottery, and given their similar research objectives, the earlier defined label can be retained.

T5 and T7, as already revealed by the data ensued by the word probability matrix, are less specific in terms of object focus than T2 (archaeological pottery). Both topics contain studies on various heritage objects, both painted and non-painted artefacts: i.e., excavated walls, stone bricks, wall paintings, ancient graffiti, polychrome architectural elements, a polychrome wooden sculpture, rocks, painted enamel objects, illuminated manuscripts, bronze sculptures, glazed red stoneware objects and painted glass windows.

In general, the ten articles with the highest probability score assigned to T5 and to T7 can be characterized as case studies devoted to a specific heritage artefact. The examination of the abstracts of articles included in T5 show that matter is more complicated, however. In this instance, the topic is not defined based on the type of objects investigated but the aim of the analytical studies. The aim is to examine the impact of different climatic factors on the condition of heritage objects. Seven of the ten articles are centered on the analytical study of the conservation state of heritage assets exposed to aggressive outdoor environments [76–79]. The study of the conservation state encompasses the identification of degradation processes and products (e.g., efflorescence, biofilms) [73–76], and, in three cases, the original materials (i.e., pigments, paint medium) [76, 79] and later restoration treatments are characterized as well [78]. The examined objects in these seven articles are archaeological remains of wall paintings [72–74, 76, 78, 79], walls [73–76] and rocks [76]. Three of these seven studies are conducted in situ and employ portable Raman combined with ED-XRF spectrometers to acquire molecular and elemental data [73, 74]. In one study, in turn, the portable instruments are complemented with laboratory techniques on cross sections (i.e., μ-Raman spectroscopy, ion chromatography and soluble salt test) [78]. All three articles are published in the Journal of Raman Spectroscopy. Articles 6 and 9 employ even wider set of analytical techniques, which does not include Raman spectroscopy, to examine degradation processes and products [76, 79]. For example, article 6 makes use of microscopic, molecular, and elemental techniques (i.e., OM, SEM, DNA analyses, Automatic rRNA intergenic spacer analysis (ARISA), XRD and portable ED-XRF) to investigate biofilms present on rocks, walls, and wall paintings in Italian grotto churches and to characterize the pigments of the mural paintings.

Among these seven degradation studies, two articles are specifically dedicated to the introduction a novel analytical methodology. Article 4 presents an in situ analytical method based on portable ED-XRF assisted with the laboratory techniques of μ-ED-XRF and XRD for studying degradation processes that take place in brick walls exposed to aggressive marine environments [75]. The laboratory techniques are adopted here to corroborate and supplement the molecular and elemental data gathered in situ. The second methodological paper, article 10, is dedicated to the development of quantitative Raman imaging methodology to address the issue of pigment degradation of excavated wall paintings [72]. The proposed method combines μ-Raman and μ-XRF spectroscopy with thermal ageing experiments to quantify the thermal transformation degree of Pompeian yellow ochre caused by the 79 AD Mount Vesuvius eruption.

Drawing a conclusion, it can be said T5 covers a much more specific research scope than initially indicated by the five words acquired by the word probability matrix as it deals with the analytical investigation of the impact of different environmental factors on the conservation state of immovable heritage assets, namely wall paintings and building materials. However, it must be added here that three articles included as well in T5 fit less this scope, although they also focus on conservation problems in cultural heritage related to the characteristics of the surrounding environment. One of these articles is concerned with the study insect damage present in a polychromed wooden statue to guide its conservation treatment [77]. In this study, the painting materials (i.e., pigments and medium) together with the adhesive materials utilized for assembling the statue are examined as well. This is done by means ED-XRF and FTIR spectroscopy. Another article focuses on the issue of digital restoration of polychromed wooden churches in Romania that suffer from accelerated degradation due to water infiltration and abandonment, and thus general neglect [81]. A working methodology for the digital preservation of these type of endangered monuments is presented in this paper. The method starts with the analytical identification of the pigments in the painting layers, continues with three-dimensional (3D) digitization of the monument and of its polychromed architectural features, and finishes with a digital restoration of these monuments and painted features. The last article included in T5, which does not really fit the scope, deals with the issue of toxic degradation products in museums [80]. More precisely, this study proposes a novel method to evaluate the performance of Ag species to collect airborne mercury compounds given off by treated heritage objects, such as herbaria and paintings by using Total reflection X-ray fluorescence (TXRF).

Given the common aim to study the conservation state of immovable heritage assets of seven of the ten articles, it can be argued that T5 does not fit really the object cluster but belongs in the methods cluster. Although it could simultaneously be argued that T5 should be moved to the materials cluster given the general focus on the study of degradation processes and products. The preliminary label of T5 thus requires adjustment too. A more suitable label for T5 would be multi-analytical approaches for the study of conservation state of immovable heritage.

T7, the final topic of the object cluster, brings together articles that have different cultural heritage artefacts, including a nonpainted object, as a subject. Nevertheless, the word with the highest probability score is glass, which indicates a focus on painted glass objects. Within the set of articles, four studies can be discerned that center on the investigation of such objects. Among these four studies, one article discusses the outcome of an in situ analysis of a painted glass window dating from the seventeenth century by means of portable ED-XRF spectrometry [82]. The aim of this study is to assess the potential of portable ED-XRF for distinguishing original panes composed of high lime, low alkali (HLLA) glass from later restoration pieces. The other three articles, on the other hand, present findings of chemical analyses of the glass composition and the (glass) coloring agents, like smalt, of painted enamel objects preserved in museum collections. The analyses are executed with portable and/or μ-ED-XRF spectrometers [84]. In two articles, these elemental tools are deployed with a complementary technique. Article 1, for instance, examines a series of painted enamels dating from the sixteenth and nineteenth centuries by means of ED-XRF and electron microprobe analysis. The aim here is to distinguish sixteenth century painted enamel objects from those of the nineteenth century [85]. Article 5 supplements with mobile µ-Raman spectroscopy with portable ED-XRF to confirm the molecular data [83].

In addition to painted glass objects, another type of painted heritage object is well represented within the set of the ten articles with the highest probability score of T7: i.e., illuminated manuscripts. In total, four articles report findings of non-invasive analytical investigation of medieval illuminated manuscripts with a variety of spectroscopic techniques: portable ED-XRF, OM, Raman spectroscopy, spectrofluorimetry and UV–visible diffuse reflectance spectrophotometry with optical fibres (FORS). These analytical campaigns concentrate on the analysis of pigments, dyes, lakes and inks [86–89].

Finally, the two other articles included in T7, provide insights into the chemical composition or manufacturing technology of two other types of heritage objects: i.e., twentieth-century Parisian non-painted bronze sculptures [90] and early Böttger red stoneware made at Meissen with gilded or gold-painted surface finishes [91]. From the five words acquired by the word probability matrix together with analysis of the abstracts of both articles, it can be derived that they were included by the topic modeling analysis in T7 based on the words 'century' and 'museum'. These two words are generic, especially when dealing with literature on the analytical study of painted heritage objects. Therefore, it could be an option to remove them from the abstracts similar to the non-alphanumerical characters and global stop words. Nonetheless, the inclusion of these articles in T7 provides further insights into the various painted heritage objects that have been studied with XRF and related techniques. Additionally, the analysis of the article titles and abstracts sheds a light on various motives behind the analytical study of the different objects included in T7. Besides testing the potential of portable ED-XRF for studying painted glass windows, the reasons motivating the investigation of enameled glassware are (1) to characterize their chemical composition as well as their production technology [84] and (2) to address issues of provenance and authenticity [83, 85]. The former reason also underlies most of the studies on illuminated manuscripts [86, 87, 89] included in T7, as well as, mentioned above, those articles focusing on the twentieth-century Parisian non-painted bronze sculptures [90] and early Böttger red stoneware [91]. While dealing with authenticity issues is also the topic of concern of one of the articles dealing with the chemical analysis of an illuminated manuscript [88]. In sum, the broader focus of T7 in terms of studied objects is valuable for this review because it provides more details about the motives underlying their analytical study. Following this line of reasoning, the initial label of T7 can be broadened. This can be done by including other painted heritage assets, which were not previously contained in the five most frequent words list: i.e., stoneware decorative objects.

The materials cluster

The topics classified under the materials cluster assembles articles concentrating on the analytical study of painting materials. The identified words per topic seem to suggest that the three topics are concerned with research on the same types of materials, namely painting materials (T8), paints (T3), and pigments (T6)—the latter being important constituents in paints. The analysis of the ten articles per topic gives further insights, which help to make sense of these seemingly similar three topics, and accordingly label them better if necessary.

T3 clusters articles that offer insights into the chemical composition of modern manufactured pigments, synthetic organic dyes, fillers and extenders (e.g., barium sulfate), modern paints (e.g., acrylic paints) and paint mixtures, binding media, painting grounds, and varnishes. In general, the aim of these articles is to acquire a better comprehension of the different paints and paint materials, which were made commercially available during the nineteenth and twentieth centuries [97, 98, 100], as well as their specific usage by modern artists, such as Barnett Newman [92], Carmen Herrera [93], Paul Gauguin [94], Alberto Burri [95] and Pablo Picasso [96, 99]. This compositional data is acquired from, on the one hand, paintings and painted prints [92–96], and from manufactured painting materials produced by suppliers as Winsor & Newton, Lefranc and Bocour, on the other [97, 98]. In addition to the analytical investigation of manufactured painting materials, synthetic organic pigments are also studied by means of reproducing modern recipes for making pigments to identify methods of synthesis and characterize the produced coloring material [100]. Finally, information about the chemical composition of synthetic organic pigments and ready-mixed house paints is yield from commercial artists' color charts and historic paint sample cards [99, 101].

For this type of research commonly non-invasive spectroscopic methods, such as ED-XRF or Raman, are combined with chromatographic, colorimetric and/or microscopic techniques, as can be derived from the group of articles included in T3 [93–100]. For instance, article 3 adopts a multi-method approach to study the chemical composition of the first commercial synthetic organic pigments manufacture by Lefranc. This approach is based on the spectroscopic techniques of surface-enhanced Raman, ED-XRF and mid-FTIR in combination with the chromatographic tool of High-performance liquid chromatography with a diode array detector (HPLC–DAD) [98]. In article 5, an even broader set of tools is employed to characterize the binding media of Carmen Herrera's earliest works that date from the late 1940s to early 1950s. With the aid of ED-XRF, FTIR, Raman, SEM–EDX, PY-GC/MS, acrylic binders were observed in one of her early paintings dated to 1949.[7] In other words, the chemical information is generally obtained in a micro-invasive and often even destructive manner. Only one article within this set of ten articles adopted a non-invasive approach by solely conducting ED-XRF spectroscopic measurements on paintings of Barnett Newman [92]. Taken together all these observations about the material focus of the ten articles included in T3, the initial label of this topic can be adjusted from 'Paints' to 'Modern manufactured painting materials.'

The focus of T6 is narrower than of T3 (modern manufactured painting materials) as it represents the analytical study and identification of pigments, both organic and inorganic. However, the research scope is not limited to a specific art historical period as is the case for T3. The aim of most articles categorized into T6 is the identification of pigments present in easel and wall paintings. More precisely, four articles are devoted to easel paintings [102, 107, 109, 110] while five articles have a wall painting as their object of study [103–106, 108]. The remaining article disseminates the findings of a multi-analytical analysis of the composition of earth pigments manufactured by Kremer to assess the company's claim that they are made from raw materials [111].[8] Therefore, this article better fits the scope of the previously discussed topic, namely T3: Modern manufactured painting materials.

In terms of analytical methods used for the pigment identification, differences in approaches can be observed between the research conducted on wall and easel paintings. First, the examination of wall paintings often requires scientists to work in situ as they are dealing with immovable heritage assets. The usage of mobile equipment for pigment identification is therefore essential. As such, portable ED-XRF [103, 106] is used with or without Raman spectrometer for the non-invasive characterization of pigments [108]. Article 7 even introduces a novel method for this type of investigation by using a more exhaustive set of tools [104]. This study aims to establish the advantages and limitations of the combined use of portable ED-XRF, XRD, and UV–Vis-FORS equipment for the non-invasive characterization of pigments from ancient Roman wall paintings from Seville (Spain). An exception to the non-invasive approach here is article 5, which indicates that chemical analyses for pigment identification are executed on samples [105]. In this study, the micro-destructive methods of gas chromatography–mass spectrometry (GC–MS) has been used to also characterize the binding medium, which is here an organic medium as the study object is an encaustic mural painting. Moreover, articles 5 is concerned with identifying and understanding the alteration aspect of the wax medium connected to its aging.

Second, elemental data from easel paintings to identify pigments is preferably obtained through non-invasive spectroscopic methods. Such methods are used in three of the four articles that are concerned with pigment analysis of easel paintings [102, 107, 110] while analysis of cross-sectioned samples was considered desirable in only one study to examine the painting stratigraphy as well [109]. The non-invasive tools employed here are portable ED-XRF [107], with or without portable reflectance FTIR [110] or spectrophotometry [102]. In one of these articles, the spectroscopic study is complemented with principal component analysis to gain profounder insights into the elemental compositions of modern pigments [110]. Given the focus here on modern painting materials, this article can thus be moved to T3.

In sum, the focus of these articles is the identification and characterization of pigments used in historical wall and easel paintings. This common focus notwithstanding, article 10 can be moved to T3. Nevertheless, T6 fits well within the materials cluster, and its initial generated label 'Pigments' can be kept.

T8, the final topic categorized into the materials cluster, represents the application of a multi-analytical approach, which includes ED-XRF analysis, to examine painting materials and techniques of various painted artefacts, both historical and modern ones. Five of the articles included in T8 are devoted to the examination of easel paintings as can be inferred from the abstracts and titles of the ten articles with the highest probability score [112–116]. The five other articles report the results of the examination of other types of painted heritage objects: polychrome terracotta sculptures [117], wall paintings [118, 119], a painted paper handscroll [120], and a polychrome wooden shawabtis [121]. Apart from these differences in studied objects, the general aim of the ten articles with the highest probability score is twofold: to examine both painting materials used to make them, and also the techniques employed by their makers. This is

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[7] This finding shows that modern artists like Herrera employed the acrylic paint medium well before the date of introduction to the European market of the first acrylic-based paints by George Rowney & Sons in 1963.

[8] The analytical methods adopted here are portable ED-XRF, Raman microscopy, XRD and light reflectance spectrophotometry.

Table 2 Overview of the preliminary and final labels per topic

topic	Preliminary labels	Final labels
T1	XRF techniques for elemental analysis	The development and optimization of XRF instruments and methods for elemental analysis of layered heritage objects
T2	Archaeological pottery	Archaeological pottery
T3	Paints	Modern manufactured painting materials
T4	X-ray spectroscopic techniques (including SEM–EDX)	Multi-analytical approaches for stratigraphic analysis of painted heritage objects
T5	Different archaeological artefacts (including walls, mural paintings, wooden objects)	Multi-analytical approaches for studying the conservation state of immovable heritage
T6	Pigments	Pigments
T7	Glass and illuminated manuscripts	Glass-based objects, illuminated manuscripts and stoneware decorative objects
T8	Painting materials and techniques	Painting materials and techniques
T9	Imaging techniques for the study of easel paintings (including MA-XRF scanning)	Imaging techniques for the study of easel paintings (including MA-XRF scanning)
T10	X-ray techniques for the study of paint degradation	Multi-analytical approaches (including SR-based X-ray methods) for the study of degradation products, processes and causes of inorganic yellow pigments in oil paintings

often achieved by studying the paint surface and/or the paint stratigraphy.

For this purpose, different analytical techniques are employed in these studies, which can be divided in two divergent study approaches: (1) the non-invasive approach mainly based on the implementation of mobile single-spot spectroscopic and imaging equipment [112–114, 117, 119–121],[9] and (2) the multi-analytical approach that includes micro-invasive methods [116] with or without micro-destructive chemical analyses of paint samples [116, 118].[10] Moreover, the implemented approaches reflect the specific needs for the study of the materials and techniques used to compose the painted heritage objects.

In terms of specific needs, three groups can be discerned within this set of ten articles. First, two articles specifically focus on exploring the potential of non-invasive analytical techniques for the chemical characterization of the surface layers of painted objects [117, 121]. One of the articles also tried to identify and characterize some previous restoration materials, as well as their distribution on the painted surface [121]. Therefore, a protocol is developed based on non-invasive, multispectral imaging techniques integrated with single-spot spectroscopic techniques (i.e., portable ED-XRF, visible reflectance, FTIR spectroscopy), as well as with OM and XRD analysis. Second, five articles deal with the non-invasive examination of materials and techniques used from the outermost paint layer down to the ground layer [112–114, 119, 120]. Three of these non-invasive studies are conducted in conjunction with detailed assessments of the condition and material history of the painted objects in view of planned conservation treatments [114, 119, 120]. Finally, three articles report the results of multi-analytical investigations of painting materials—including organic materials—painting methods, and the conservation state of easel and wall paintings [115, 116, 118]. In other words, T8 is devoted to the analytical study of the materials and techniques of painted heritage objects, including easel and wall paintings. Eight studies categorized under T8 are also concerned with the investigation and assessment of the condition of the paint layers. However, the previous label 'Painting materials and techniques' can be withheld for T8. The main reason is that the analytical study of painting materials and techniques is very often accompanied by such condition assessments, as well as with a broader art historical evaluation of their maker's working methods and styles.

Based on the above observations, the preliminary labels of five core topics (i.e., T3, T4, T5, T7, T10) required some adjustments. An overview of the preliminary labels and the final labels is shown in Table 2. In addition to these adjustments, T5 was removed from the objects studied cluster and added to the methods used cluster. The final topics per cluster are thus: the methods used (T1, T4, T5, T9, T10), the objects studied (T2, T7), and the specific materials studied (T3, T6, T8).

Analysis of the time trends

Having now answered the first part of the research question—i.e., identifying core topics—we move to the second part of the research question being the time trends in the share of attention to those topics. Using the earlier

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[9] i.e., portable ED-XRF, Raman, FTIR, FORS, XRD, visible reflectance spectroscopy and multi-spectral imaging such as Vis–NIR, ultraviolet induced fluorescence (UVIF) photography and infrared reflectography (IRR).

[10] i.e., Attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR), micro-Raman spectroscopy, SEM–EDS, as well as MS techniques like gas chromatography-mass spectrometry (GC–MS) and direct exposure mass spectrometry (DEMS).

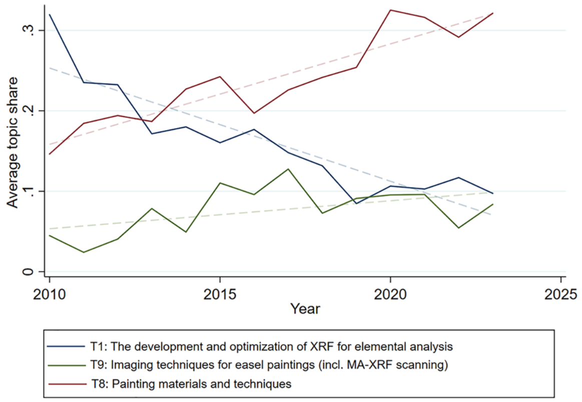

Fig. 2 Time trends of topics 1, 8 and 9 based on average topic share (2010–2023)

calculated topic probability score for each article in each topic as well as the year in which an article was published, indicator variables can be created for the specific topic that has the biggest topic share within each article. Subsequently, the average topic shares can be plotted through time in order to identify time trends. Average topic share means the average share of a specific topic in articles published in a specific year. It can thus be used to compare attention to different topics over time.

As can be seen from Fig. 1, the number of studies about XRF on painted heritage objects has increased exponentially, so a time trend analysis focused on the number of studies in relation to topics would be flawed as it would be confounded by the vast increase in articles in general. By calculating annual topic shares, this issue is addressed as each time point indicates the share a topic has in the total articles published in that year.

In figure SI.1, time trends are shown for all ten topics. Note that the time trend starts in 2010; this choice was made because the absolute number of articles included in the sample was very low before this period (see Fig. 1) which would distort the visual interpretation of the trend. From figure SI.1, two obvious trends especially stand out. Figure 2 is added below to visualize these two trends more clearly. Firstly, T8 (painting materials and techniques) has seen a strong increase in attention between 2010 and 2023, more than doubling in terms of average topic share. This could show the impact of the implementation of mobile and portable non-invasive equipment prior to and during that period, which enabled the in situ study of fragile and valuable painted heritage objects, especially easel, wall paintings and illuminated manuscripts. This development not only enabled scientists to conduct in situ research but it also has introduced mobile instruments with analytical capacities close to or equal to lab-based devices to museums, conservation studios and other cultural centers [122]. Especially, the low relative cost of handheld ED-XRF has rendered spectroscopic analysis more accessible and comprehensible for various stakeholders within the cultural heritage field [123]. Another development that has further contributed to this trend has been the introduction of a commercial mobile MA-XRF scanning instrument (i.e., M6 Jetstream, Bruker) in 2013 [5]. This can be deduced from the time trend of T8 (see Fig. 2). In eight of the ten articles [112–114, 116–120] included in the T8, the research was conducted by museum practitioners and art conservators in collaboration with scientists from universities and national research institutions (e.g., Consiglio Nazionale delle Ricerche (CNR)). As such, the development of mobile and portable non-invasive equipment also brought heritage practitioners and scientists closer together.

Secondly, T1 (the development and optimization of XRF techniques for elemental analysis) has seen a sharp decrease in attention between 2010 and 2023. The downward trend could be explained using the same argument as above: the variety of mobile and portable non-invasive instruments has increased rapidly in the past two decades. Consequently, multi-analytical approaches that combine XRF with various other spectroscopic and imaging tools, often also with laboratory techniques, are commonly adopted for an extensive materials analysis. This evolution has turned XRF spectroscopic devices, such as handheld ED-XRF, into an efficient screening tool, which ensuing elemental data is often used to develop or refine a multi-analytical methodology. In other words, articles that solely report results of XRF analysis on painted heritage objects have decreased in comparison with articles that deal with such multi-analytical research methods. Additionally, XRF is now a well-established method with major technological developments in XRF equipment and components having already taken place. These include, for example, the introduction of the Peltier cooled Si-PIN detector already in 2000 and thus handheld ED-XRF spectrometers [21].

Because of the strong rise and fall of T8 (painting materials and techniques) and of T1 (the development and optimization of XRF techniques for elemental analysis) respectively, Fig. 2 does not provide much visual information about the other topics that might have also experienced an increase and/or decrease but just less pronounced. To assess this, the time trend graphs for each topic are included separately as Additional file 1: (SI). T2, T4, T5, T6 and T7 have all seen ups-and-downs in share of attention over time but the underlying time trend seems to be relatively flat, indicating—if any – only a limited increasing or decreasing trend over time.

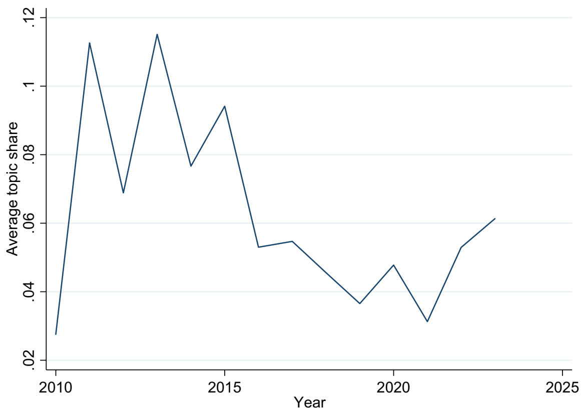

Fig. 3 Time trend of topic 10 (multi-analytical approaches for the study of degradation of inorganic yellow pigments in oil paintings) based on average topic share (2010–2023)

In other words, the scholarly interest in these topics has been rather stable between 2010 and 2023.

The time trends for T9 and T10 show an interesting picture, especially in relation to XRF literature (see figs. 2 and 3). For T9, which centers on imaging techniques for the study of paintings (including MA-XRF scanning), a curvilinear evolution can be observed. Specifically, a sharp increase in topic share between 2010 and 2017, that starts to "flatten out" after this period. This could be related to the development of scanning and full-field imaging systems, most notably: the mobile MA-XRF scanning tool between 2010 and 2013 [3–5]. This imaging scanning technique had initially a strong increase in average topic share probably because it was an innovation, which quickly diffused within the broader heritage field. Hence, the elemental data acquired with this novel imaging technique from easel paintings, as well as developments in instrument components and configuration and in data collection and handling methods saw an initial increased attention in the XRF literature. However, like any innovation, this average topic share has started to flatten out since 2017. A possible explanation for this flattening curve is that MA-XRF, just like ED-XRF, is now often used in combination with other imaging and analytical techniques, and the technique is well developed and thus no major instrumental advances are happening.

T10, which is devoted to multi-analytical study of pigment degradation in oil paintings, shows another curvilinear trend, but an even more extreme one—ranging from an inverted U-shape between 2010 and 2017, which becomes a U-shape between 2017 and 2023. The initial inverted U-shape can be explained perhaps by yet another instrumental trend that manifested itself during the past three decades within the fields of cultural heritage and archaeological science with synchrotron radiation (SR). Synchrotron light-based analysis has become indispensable for the detailed spectroscopic study of the chemical composition and morphology of painting materials and, most importantly, their degradation products, in which the elevated spatial resolution, brightness and energy-selectivity of the primary beam is a distinct benefit. Again, like most innovation diffusions, the shape can be explicated by an initial increase in adoption, that flattened out, even decreased with the emergence of new trends and innovations, but, and this is perhaps less typical of innovations, started to become more popular again recently.

A possible explanation for this divergent time trend is that the accessibility to synchrotron facilities for cultural heritage and archaeological research has notably increased only about half a decade ago, that is around 2018–2019, due to technological progress in detectors and scanning control systems [124].[11] These advances significantly reduced scan times from second to millisecond dwell times. This innovation helped minimizing radiation dose, which is an important safety measurement when studying delicate and precious heritage objects, but also enabled an impressive growth of many techniques for analysis (e.g., XANES imaging) and permitted high-definition imaging of whole objects instead of very small sampling areas. Prior to this technological advancement sample dwell times were of the order of one second per scanning X-ray fluorescence pixel, which entailed that only the analysis of a limited number of minute sampling areas measured at high resolution, or larger areas measured at low resolution, was feasible. These relatively long sample dwell times were thus a substantial limiting factor on scientific output in the past.

Conclusions

This paper set out to answer following research question: what are core topics and time trends that can be observed in research about XRF on painted heritage objects? Topic modeling, an approach to text mining using unsupervised machine learning, was used to identify ten topics which can be clustered into three groups: the methods used, the objects studied, and the specific materials studied. The review yielded detailed information about the various application and developments of XRF on a wide range of painted heritage objects, as well as painting materials. Moreover, it provided additional insights into the

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[11] Other limiting factors before 2018–19 were probably: 1) whether the synchrotron facilities themselves were interested in—or equipped to handle—samples from cultural heritage objects, and 2) the need of users to understand the synchrotron methods, techniques and facilities and to propose good experiments, especially in ways of organizing samples and selecting zones of interest, for cultural heritage and archaeological research.

research trend of the last decade to use XRF in combination with other analytical methods. More precisely, the analysis of topic 2–10 yielded information about the various methods used in combination with XRF for studying archaeological pottery, glass-based objects, illuminated manuscripts, stoneware objects, modern manufactured painting materials, pigments, and painting materials and techniques, and various degradation phenomena. This information can help both heritage scientists and practitioners to select and develop research methods for the analytical study of painted heritage objects.

Time trends were also identified by the review method showing which core topics have seen increased or decreased shares in attention over time. In doing so, a body of research of close to 1000 studies was reviewed and patterns were uncovered that can help inform future research about XRF on painted heritage objects. Topic modeling thus shows great promise for heritage science, especially with an ever-increasing literature base to draw on and make sense of. While the focus of this paper was on topic modeling as a computational approach to literature review, there are several other applications one can think of for heritage science. For instance, to help analyze large sets of documents about specific heritage objects collected in online archives or to analyze reports from heritage and/or conservation professionals with the aim of identifying topics of interest (and trends therein). Furthermore, other text mining methods beyond topic modeling could be used. Sentiment analysis can help to identify the overall sentiment towards a concept in a large number of texts – for example positive or negative sentiment towards cultural heritage as expressed by policymakers in policy documents or by citizens and society at large in social media messages or news articles.

With that said, there are several recommendations to follow when applying topic modeling, and two are emphasized here. First, it is crucial to have content experts involved in the review. Whereas the tool allows the identification of probability scores, topics and words linked to topics in a vast body of literature, it does not offer an actual label to the topic. For this, human interpretation and sensemaking is crucial, and content experts are needed to assess the offered words and highest scoring articles to come up with labels that make sense for the field of study at hand. Similarly, content experts can make sense of the observed trends, placing them within a broader understanding of the field at hand.

Second, like any analysis tool topic modeling is also influenced by the 'garbage in, garbage out' (GIGO) principle. It is vital to embed checks and balances on the data that will be analyzed using topic modeling as flaws in that data can result in flawed analyses. There are several specific things to look at to prepare data (e.g., removing stop words), but the overall quality of the data going into the tool needs careful consideration. For instance, even though WoS was prompted to only include English articles several non-English abstracts were still present in the dataset. Similarly, there were several missing abstracts and/or very short abstracts that could distort the analysis. Some of this may be discovered up front, some of it may be the result of dialogue between a data analyst and content expert when interpreting the results. But it is not unlikely that several iterations could be necessary to ensure an as valid as possible dataset.

In addition to these two important recommendations, there are also limitations (and thus further recommendations) to acknowledge when engaging with topic modeling. First, topic modeling is an approach to doing a scoping or exploratory literature review—identifying topics and trends. It does not provide the depth of a systematic, integrative review, rather its aim is broadness, analyzing a large amount of text that would be unfeasible to analyze using more human-driven review techniques.[12] Second, the focus of the analysis is abstracts, while such abstracts especially in academic journals can be expected to provide enough information for topic identification, the full text could provide more detail and nuance. Both limitations require trade-offs by the researchers and, potentially, combining different review approaches to tackle topic modeling's limitations. In essence, this was also done in the paper at hand by doing a more in-depth qualitative analysis of 10 articles for each topic.

To conclude, topic modeling holds great promise for our field and this study has shown how it can be used to review the large–and ever expanding–body of knowledge about XRF on painted heritage objects.

Abbreviations

ARISA	Automatic rRNA intergenic spacer analysis
ATR	Attenuated total reflection
DRS	Diffuse reflectance spectroscopy
ED-XRF	Energy dispersive X-ray fluorescence
EPR	Electron paramagnetic resonance spectroscopy
FF-XANES	Full-field X-ray near-edge structure imaging
FORS	Fibre optic reflectance spectroscopy
FTIR	Fourier transform infrared spectroscopy
GC–MS	Gas chromatography–mass spectrometry
GE-XRF	Grazing exit XRF
GIGO	Garbage in, garbage out
HPCL	High-performance liquid chromatography
HPLC–DAD	High-performance liquid chromatography with a diode array detector
IRP	Infrared photography

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[12] For an overview and discussion of the different techniques of literature review, see George B, Andersen LB, Hall JL, Pandey SK. Writing Impactful Reviews to Rejuvenate Public Administration: A Framework and Recommendations. Public Administration Review. 2023; 83(6): 1517–1527.

IRR	Infrared reflectography
LDA	Latent dirichlet allocation
MA-XRF	Macro X-ray fluorescence
NAAR	Neutron activation autoradiography
OM	Optical microscopy
PCA	Principal component analysis
PIXE	Proton-induced X-ray emission
PY-GC/MS	Pyrolysis gas chromatography mass spectrometry
SEM–EDS	Scanning electron microscopy-energy dispersive X-ray analysis
SR	Synchrotron radiation
TXRF	Total reflection X-ray fluorescence
UV–Vis	Ultraviolet–visible spectroscopy
WD-XRF	Wavelength dispersive X-ray fluorescence
WoS	Web of Science
XRD	X-ray powder diffraction
XRF	X-ray fluorescence
µ-FTIR	Micro fourier transform infrared spectroscopy
µ-SERS	Micro-Raman spectroscopy and surface-enhanced Raman spectroscopy
µ-XANES	Micro X-ray absorption near-edge spectroscopy
µ-XRD	Micro X-ray diffraction
µ-XRF	Micro X-ray fluorescence

Supplementary Information

The online version contains supplementary material available at https://doi.org/10.1186/s40494-024-01135-2.

Additional file 1: Figure S1. Top ten articles per topic based on topic probability score (Stata output). **Figure S2.** Time trends of the ten topics based on average topic share (2010-2023). **Figure S3.** Time trend of topic 1 (the development and optimization of XRF for elemental analysis) based on average topic share (2010-2023). **Figure S4.** Time trend of topic 2 (archaeological pottery) based on average topic share (2010-2023). **Figure S5.** Time trend of topic 3 (modern manufactured painting materials) based on average topic share (2010-2023). **Figure S6.** Time trend of topic 4 (multi-analytical approaches for stratigraphic analysis of painted heritage objects) based on average topic share (2010-2023). **Figure S7.** Time trend of topic 5 (multi-analytical approaches for studying conservation state of immovable heritage) based on average topic share (2010-2023). **Figure S8.** Time trend of topic 6 (pigments) based on average topic share (2010-2023). **Figure S9.** Time trend of topic 7 (glass-based objects, illuminated manuscripts and stoneware decorative objects) based on average topic share (2010-2023). **Figure S10.** Time trend of topic 8 (painting materials and techniques) based on average topic share (2010-2023).

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Availability of data and materials

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Declarations

Competing interests

The author communicates no competing interests.

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