A methodology for timing interventions made on the polychrome decorations of the façade of the Palace of King Peter I, the Royal Alcázar of Seville, Spain

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ABSTRACT

A chronostratigraphic approach was used to the study of pictorial layers from the polychrome decorations on the façade of the Palace of King Peter I (the Royal Alcázar of Seville, Spain), to identify the timeline of successive decorative interventions made over time. Stratigraphic and mineralogical studies enabled the identification of modifications made to the artwork, either due to deterioration or to the various interventions made for maintenance purposes. The study of documentary references was accompanied by painstaking fieldwork and diverse laboratory techniques to establish stratigraphic correlations between the different areas and decorative elements of the façade. Future restorations oriented to the long-term conservation of this building (both preventive or of remedial nature) should take into account that the general appearance of the façade at present is close to that of medieval and modern times with the exception of minor modifications made during the intermediate interventions as result of an erroneous interpretation of previous chromatic alteration in specific areas of the façade.

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1. Introduction

Analytical studies today are a common, even indispensable, complement to any research concerning heritage. Most of the studies of polychrome decorations on façades or wall paintings focus on future restoration or preventive conservation and consist of a multidisciplinary approach, including the use of diverse instrumental techniques, to identify the different materials and techniques employed [1–3]. However, an accurate interpretation of the results of the analytical and stratigraphic studies requires not only an understanding of the technical procedures involved in making the pictorial decorations but also the casuistry regarding the execution process. This is important because a purely literal reading of a given sequence can sometimes lead to errors. In particular, the classification and organization of pictorial layers according to relative age can be achieved successfully only after performing stratigraphic correlations between the layers observed in the stratigraphic cross sections of many paint samples.

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The present research was undertaken on the polychrome decorations on the façade of the Palace of King Peter I (the Royal Alcázar of Seville, Spain; Fig. 1A), inhabited since it was built in 1366, to the present day. The research revealed the enormous number of such decorations repeatedly applied to the façade throughout history, some of which have documentary references. The layers upon layers of interventions are proof of the on-going concern for the state of conservation of the building, resulting from the fact that this is the oldest Spanish royal palace still in use today. This work describes the methodology followed in a chronostratigraphic study of the polychrome decorations on the Palace of Peter I at the Royal Alcázar of Seville, which provided comprehensive and verified data.

2. The façade

The façade of the King Pedro I Palace consists of three parts: the central part (portal or front façade) and two apparently symmetric lateral-parts (Fig. 1B). Front façade appears delimited by two slender columns of marble with caliphate capitals and large brick pilasters above them, and finished by two large wood corbels that support the monumental eave made of polychromed wood (Fig. 1C). This is constituted by thin, richly decorated wooden corbels which protect the arrocaste or ornate frieze (Fig. 1D). Three ajimez windows with one or two slender central columns and multilobulated arches occur in the second floor of the front façade, covered by a blue and white ornamental tile with Islamic epigraphy. The lower part of portal is constructed in carved stone with intricate ornamentation of atauriques (stylized plant motifs) that must originally have been polychromed. Lateral parts of the façade have smaller eaves than those of the central part with similar polychrome decorations. The most commonly used building materials are brick and gypsum plaster panels in the upper levels.

3. Stylistic and historical framework

During the 15th century, many luxury royal palaces were built in Spain as a result of the emergence of a new cultural and artistic style called Mudéjar, a mixture of Islamic and Christian architectural elements. This derived partly from admiration for the Alhambra palaces in the court of the Catholic Monarchs Enrique II and Pedro I [4]. After the Reconquest of Spain, the Islamic legacy persisted in the Iberian Peninsula during the next century, sustained by the many Muslim artisans remaining in the conquered territories. These artisans applied their decorative artistic techniques to the edifices built according to the new European styles (Gothic and Renaissance), which were ornamented with finishes in Mudéjar style.

A particular case of a Christian monument having Islamic architectural and ornamental elements is the Palace of Don Pedro I de Castilla. The architecture of the façade imitates Islamic palaces and was constructed following Byzantine models that conceived of the façade of the palaces as a “gate of justice” [5]. The façade was decorated by artisans from the Kingdom of Granada, according to an inscription reading “only God is the victor”, which also appears as a major epigraph in the Alhambra palaces [6]. However, in the façade and throughout the palace of Peter I Contreras [7] recognized the work of numerous artisans with diverse religions and beliefs.

The façade of the King Pedro I Palace has been considered the best example of Islamic civil architecture built in Christian territory, as well as the main monument of the Mudéjar style in Spain [8]. A measure of its magnificence is that, during his visit to Seville, the Nasrid King Muhammad V was so impressed that he ordered a similar construction for the façade of the Comares palace in the Alhambra of Granada [9].

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![Fig. 1. A. Google Earth image of the Royal Palace complex (arrow) and surrounding area in the city of Seville (Spain). B. General view of the façade of the Palace of King Peter I consisting of a central part and two apparently symmetric lateral parts. C. Detail of the portal (or central section) of the façade of this palace. D. State of the monumental eave made of polychromed wood of the central portal during a restoration of the façade (2008–2011).](image-url)
4. Methodology

Generally, polychrome works are relatively simple in the sense that the diverse decorative elements appear coated with a very limited range of colours applied during a specific historical period. However, in the present case study, successive polychrome decorations were applied over different historical periods [10]. Chronostratigraphy consists of a specific methodology for the timing of interventions in polychrome decorations. The ultimate goal of the present study is to establish a time scale applicable to polychrome decorations affected by repeated interventions, regardless of the support and its location. Establishing the ages of the pictorial elements

Fig. 2. A. Architecture sketch of the façade, modified from Almagro [16], and location of the polychrome painted decorations (areas shown in B and C). B. Detail of upper levels of the portal including the monumental eave made of polychromed wood with indication of sampling sites. C. Approximate location of sampling points at the eave and yeserías of the second floor of the lateral sections. The label of the samples consists of the abbreviations PIF corresponding to Peter I façade, followed by the location of the samples in the diverse sectors or sections (P: portal; RH: right hand of the observer; LH: left hand of the observer), in some cases by the type of architectural or decorative elements (E: eave; A: arrocabe or ornate frieze; MO: mocarabes, array of vertical prisms with stalactite-like appearance; T: tabica or riser; C: corbel) of the façade, the type of support (W: polychromed wood; Y: yeserías, decorative gypsum plasterwork; S: carved stone with ornamentation of atauriques, stylized plant motifs) and numbers.
strata in the different areas or decorative elements of an architectural monument involves the correlation of pictorial strata from place to place. Specifically, stratigraphic correlation tries to classify and organize pictorial layers according to relative age.

For the organization of pictorial layers into temporal units, a diverse range of procedures was used in this study, generally sequentially. It began with historical documentation, followed by fieldwork and the study of materials and their alterations by means of instrumental analysis techniques. Later, stratigraphic cross-sections of paint samples were correlated in order to establish the correspondence between the different polychrome levels. Furthermore, the timing of the interventions made on the polychrome decorations deduced from our analytical results was cross-checked against documentary sources. In this way, we established the number of interventions and chronologically classified them. In light of the above, we also identified the overall chromatic changes undergone by the façade and the individual changes of each element over the course of history. This methodology may be useful not only in future studies on the Seville Alcázar but also in the case of other buildings presenting a similar range of challenges.

4.1. Documentary study

An in-depth bibliographic review of the wide-ranging interventions carried out on the Alcázar over the centuries was conducted, and even where there was no explicit mention of the polychrome decorations, references indicating the need for such interventions were taken into account. The review revealed the scarcity of information on the topic, particularly from the Middle Ages, and although in later periods, more data refer to the façade, only a few specific references to polychrome decorations are available. The oldest descriptions of the Alcázar were found in works from the 17th century, among the different descriptions of monuments written by Rodrigo Caro in 1634 [11]. Regarding the façade of this particular palace, he alludes to its colours at that time, enabling comparisons to be drawn with its appearance in subsequent centuries.

The many restorations undertaken in the 19th century were hardly described in suitable documentary studies. One exception involves the significant descriptions of the Royal Alcázar and other monuments in Seville written by José Amador de los Ríos and Francisco María Tubino [6,12], towards the mid- and late 19th century, respectively. Also significant was the extensive documentation by José Gestoso Pérez, preserved in the archives of the Alcázar, which he made available to other researchers [13,14].

The 20th century saw a surge in research on the Alcázar, prompted largely by the advances in archaeological studies by, among others, Rafael Manzano Martos, Miguel Ángel Tabales, and Antonio Almagro Gorbea. The latter carried out a comprehensive planimetry of the entire Alcázar, providing the basis for subsequent studies [9,15–17].

Outstanding publications on the polychrome decorations include those of Ana Marín Fidalgo and Rosario Chaves. The former provides valuable data on the Alcázar in the 16th and 17th centuries, while the latter offers an overview of the interventions made during the 19th century [4,18].

4.2. Fieldwork

It bears highlighting certain considerations regarding the colour and other visual aspects of objects. In the case of polychrome decorations in particular, identifying their “real” colour from the surface presents difficulties due to the accumulated dirt and the chromatic alterations that materials typically present. The study of chromatic changes affecting the general appearance of the façade in the past may influence future interventions on the façade and has a practical application for the long-term conservation of the palace.

Macroscopic examination of the materials and their alteration provided the basis for selecting the points of prime interest for the subsequent sampling stage. In this phase, selected areas were documented photographically, as a whole and also at the level of individual details, including the materials used, the finishes, remains of colour alterations, chromatic changes, etc.

The sampling method consisted of dividing the decorated surface of each of the three main sections of the façade into three vertical bands. Within each of these bands, 14 levels were differentiated, each coinciding across the three sections. These levels were structured by taking as a reference point the ornamental elements and different colours (Figs. 2 and 3). A total of some 242 samples were taken (94 from the central portal and 74 from each of the side sections). This large number of samples was necessary, together with a systematized approach to collecting them, in order to achieve a more complete register of the different polychrome decorations and colour changes.

The first samples, taken from a zone of the central portal of the façade, were used to carry out the material study prior to the works undertaken to conserve and restore the entire façade during the period 2008–2011. All decorative elements of the portal were represented in these samples. The scaffolding required by these interventions enabled close-up examinations of the materials (particularly deterioration and previous interventions), and to take a considerable number of samples – more than is typical in studies of the polychrome material used on monuments.

By the time the samples from the two side sectors or lateral parts of the façade were taken (2009 and 2010), the study of the central portal was already at an advanced stage. With the same systematization process, the two sectors were comparatively studied, revealing that the sector to the right hand of the observer presented greater cohesion in the polychrome decorations, while the decorations on the left-hand sector had suffered greater deterioration (being cracked and powdery). This made the sampling more complex, as in subsequent work performed in this area. In general, these latter samples proved challenging in terms of establishing the sequence of the different layers, as in some cases, the strata
had become separated, destroying the original sequence. However, comparisons with the other façade provided an adequate level of information, as the strata from equivalent levels presented practically identical sequences in most cases. Comparisons between successive layers from different sectors made it possible to reconstruct deteriorated stratigraphic sequences.

### 4.3. Analytical study of painting materials

The analysis of microsamples provided insights into the materials used and the alterations in the artworks under study. The resulting data, which were essential for an accurate determine the techniques used in creating the polychrome decorations, also

![Image](image-url)

**Fig. 4.** Reflected-light image made with crossed polarizers (A) and one polarizer (B) of a pictorial sample containing up to five gilding, which indicate the occurrence of five polychromes labelled from bottom to top as P1 to P5. Gold leaf of the gilding layers is appreciated only in the image with one polarizer. In the case of polychromes P1 and P3, gold leaf is not discerned in this partial image of the entire cross section, but in adjacent areas to it. Starting from bottom to top, these polychrome decorations appear to be composed of: P1: base layer of white and red lead, followed by a gold base made up of a mixture of oil and resins plus yellow pigments (layer of siso) and gold layer; P2: blue pictorial layer constituted by natural azurite with white lead, and an upper gold base (linseed oil and colophony resin) and gold; P3: idem as P2; P4: base layer of white lead, a yellow layer used as the gold base composed by white lead, litharge and iron oxy-hydroxides, and the gold layer; P5: base layer of white lead, yellow layer used as the base layer for the gold and simulating a yellow tone analogous to gold (white lead, chrome yellow linseed oil and colophony resin), and the gold layer of the uppermost polychrome decoration.

![Image](image-url)

**Fig. 5.** Stratigraphic correlation among the samples studied from the arroche of the portal (Fig. 3A). Lines between the stratigraphic sections connect pictorial strata of similar composition. A guide-stratum composed of white lead and calcite is observed in the upper part of the study sections. Layers of stratigraphic sections are also grouped in diverse polychromes (from A to I) resulting from the diverse interventions (see text).
helped to date the work. This was particularly true in the case of materials that are characteristic of a particular period, such as that of pigments from the industrial age used from the 18th century onwards.

In other cases, the material could not be used as a differentiating criterion because pigments and other substances were used throughout a number of periods. However, analytical studies made it possible to associate the different strata within one single intervention by focusing on similarities in the crystal morphology. Such studies can also help in identifying the number of interventions made and in defining a relative chronology for these, by determining the order in which the different strata were superimposed.

In most samples, this sequencing was incomplete, hampering the interpretation of the results and the drawing of reliable conclusions. However, the decorations on the façade are modular in nature and follow a repetitive pattern of elements and chromatic “rhythms”. Therefore, the management of a large number of samples corresponding to different areas of the façade made it possible to compare the analytical results and extrapolate the data.

In a preliminary scientific-technical study [19], we described in detail the different pictorial materials (pigments and inorganic materials providing body to blenders) used in the polychrome decorations of the Peter I Palace façade. We established three main groups of polychrome decorations characterized by the use of specific materials:

- white lead, red lead, cinnabar, malachite, natural azurite, organic black, and gold were used in the bottom layers;
- artificial azurite in the intermediate layers;
- copper arsenates (Scheele’s green), synthetic ultramarine blue, emerald green, and chrome yellow in upper layers.

The reasons for choosing each analytical technique employed here, together with a detailed description of the analytical procedures followed in the application of instruments to the study of the polychrome decorations, are also given in this preliminary study. In addition, a detailed description of the analytical procedures is provided as supplementary material. Most of the technical analyses of paint materials, with the exception of the characterization of the lipid binders and proteinaceous media made on untreated samples by gas chromatography and high-pressure liquid chromatography, respectively, were performed on cross-sections obtained from the

![Image](image_file)

**Fig. 6.** Example of file showing the stratigraphic sequence observed in a sample. A. Interpretative table in which different colours are assigned to pigments. B. General image of the complete sequence of pictorial layers (labelled from L1 to L9) made by using reflected light under crossed nicols. C. BSE image of spherulites of the emerald green corresponding to layer L7. D and E. EDX and Raman spectra of emerald green, respectively.
small pieces of paintings. For the textural and mineralogical characterisation of the pictorial layers and to compare the samples in order to establish the correspondence between the different polychrome levels, samples prepared as polished thin sections were studied by optical and analytical scanning electron microscopy. Single-point Raman microanalyses were also performed on the polished sections for the identification of pigments and their alteration products.

5. Results and discussion

5.1. Stratigraphic correlations

The presence of a considerable number of strata in most of the samples indicated the existence of several interventions on the polychrome decorations which could have comprised a varying number of strata. One of the most complex tasks of the present work was to establish the relationships between a high number of samples (242), each bearing numerous strata (from 3 to 24).

Therefore, it was essential to ascertain the similarities between the composition and chronology of the diverse layers in the polychrome, enabling us to draw conclusions on the strata belonging to each polychrome. The methodology for this phase of the work consisted of a rigorous identification of the materials making up each of the strata. Comparisons based on their nature and composition enabled us to discern a series of guide-strata, which in turn helped to establish guidelines for correspondence and specific dating milestones. For example, the presence of different layers of gilding provided clear indications to help differentiate superimposed polychrome decorations. Some samples contained up to five different strata of gold, which would indicate five different polychrome decorations (Fig. 4). Elsewhere, one thick white layer with distinctive compositional features has served as a guide-stratum when correlating the different polychromes (Fig. 5).

The use of different microscopic and analytical techniques enabled us to establish beyond doubt the number of strata within each polychrome and the number of polychromes in each decorative element. In the case of samples in which some of the strata had been lost or had not been applied, the information gained from samples of equivalent elements was used to establish the level of the conserved strata.

The analytical investigation determined not only the main components of the layers but also the components that were found only in minute proportions. Although these were not relevant in aesthetic or behavioural terms, they were crucial in differentiating between strata corresponding to different interventions. In this context, large numbers of layers were composed primarily of white lead, which could have been considered to belong to a single intervention. However, the fact that some of these contained minor components such as calcium carbonate, calcium sulphate or barite indicated that, in fact, they corresponded to different interventions. Similarly, the use of other materials such as natural or synthetic

![Diagram](image)

**Fig. 7.** Example of stratigraphic correlation among different samples from the lateral sections of the portal corresponding to the same decorative element (green palms; red arrow). A to I refer to the diverse interventions of Table 1.
azurite also provided relevant information to help differentiate apparently contemporary strata.

For easier viewing of the analytical information and identification of the relationships between the pictorial layers, the stratigraphic sequences found in the samples were transferred to a series of tables in which a colour code was assigned to each pigment. A file was created for each sample containing the colour-code table together with all the analytical information corresponding to the diverse pictorial materials of the layers (Fig. 6). Afterwards, the files were used to compare samples, make stratigraphic correlations, and to establish the relative chronology. In addition, the individual files were grouped firstly by type of support, secondly by area, and finally by the decorative elements (Fig. 7).

5.2. Timing of the interventions

Once the strata each polychrome had been distinguished and the relationships between the different samples established, the next step was to relate them to the documented interventions and restorations (Table 1). This was possible thanks to the chronological data which, in most cases, indicated the pigments themselves, as some pictorial materials came into use only at a certain point in history. The nine polychromes corresponding to different temporal interventions can be grouped as follows:

- interventions referred to as A, B, C, and D can be considered original or historical interventions, in the sense that their respective polychrome decorations are located deep in the stratigraphic sequence and contain ancient pigments. Polychromes A and B may be considered chronostratigraphically as a single intervention (original intervention) performed according to the traditional techniques of the Islamic painting. Thus, the first polychrome decoration (A) usually contains the following superimposed strata: a preparatory layer and a base layer of red lead which extend along the overall surface of the wooden support. In the areas with red surface colour, the base layer was covered by pictorial strata of cinnabar (e.g.: sample PL-PI-7-W-46 in Fig. 8). On the other hand, a new polychrome B was applied to

<table>
<thead>
<tr>
<th>Polychrome</th>
<th>Type of intervention</th>
<th>Chronology based in pigments</th>
<th>Chronology based in documented intervention</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>Original and historical interventions</td>
<td>From Medieval times to 20th century (red lead, cinnabar, organic black, azurite, malachite, white lead, iron oxides, calcite)</td>
<td>1364 (referenced in the epigraphy of the front façade)</td>
</tr>
<tr>
<td>B</td>
<td>Intermediate interventions</td>
<td>Synthetic azurite (used from 16th century)</td>
<td>Around 1477 (related to construction of the lateral sections of the portal by the Catholic Monarchs [20])</td>
</tr>
<tr>
<td>C</td>
<td>Recent interventions</td>
<td>Modern synthetic pigments associated with the industrial revolution (Scheele’s green, emerald green, chrome yellow, blanc fixe and ultramarine blue, that were first synthesised from 1778 to 1828)</td>
<td>1805 (internal report by Manuel Zintora about restoration actions on the façade [18])</td>
</tr>
<tr>
<td>D</td>
<td>Synthetic pigments</td>
<td>From Medieval times to 20th century (red lead, cinnabar, organic black, azurite, malachite, white lead, iron oxides, calcite)</td>
<td>1874 (restoration intervention in the caves of the lateral sections of the portal [18])</td>
</tr>
<tr>
<td>E</td>
<td>Synthetic pigments</td>
<td>From Medieval times to 20th century (red lead, cinnabar, organic black, azurite, malachite, white lead, iron oxides, calcite)</td>
<td>1805–1898 (generalized interventions on the polychrome decorations of the entire façade under the direction of the Academia de San Fernando [18,21])</td>
</tr>
</tbody>
</table>

Fig. 8. Representative table showing the organization of the pictorial layers in samples from the tabica (riser) of the central portal of the façade into nine chronostratigraphic units corresponding to interventions. These units are bounded by synchronous horizons which allow age correlations between stratigraphic layers of different samples.
those areas with colours other than red, where an intermediate base layer of white lead with gypsum occurs between a subjacent layer of red lead and the green pictorial layers made of malachite or blue strata of azurite (sample PIF-P-T-W-50 in Fig. 8). Intervention D was made in the times of the Catholic Monarchs also following the Nazari style. It constitutes the first polychrome of the yeserías (decorative gypsum plasterwork) of the lateral sections of the portal, which bears the same chromatic finishes as used on the wood of the rest of the façade:

- intermediate interventions (E and F) coincide with the appearance of the synthetic pigments, in particular synthetic azurite, used massively since the 16th century and according to the documentary research correlates with a repair performed on the decorative gypsum plasterwork of the lateral sections of the portal (Table 1). In the case of polychrome decoration referred as E, a large variety of colours were used in non-gilded areas. The stratigraphic analyses evidence a broad use of pigments (azurite, vermillion, red lead, and organic black, among others) at this level. Gilded areas were not retouched. This constitutes a drastic chromatic change in the façade, which was partially reversed with intervention F, here considered a proper gilding restoration consisting of re-touching areas of loss with “gold” paint;
- recent polychrome decorations (G, H and I; Table 1) consisted of more or less generalized interventions on the façade characterized by the use of modern synthetic pigments as Scheele’s green, emerald green, chrome yellow, blanc fixe or synthetic ultramarine blue. They are very well-documented (Table 1), as is the case of the major renovations related to the use of the palace as the residence of the dukes of Montpensier or a major restoration of the façade under the direction of the Academia de San Fernando [18,21].

5.3. Chromatic evolution

After the study of materials and the identification of their correlations via historical profiles, the chromatic evolution of the different decorative elements of the façade was considered. As a result of successive interventions on the decorations over the years, the image of the entire façade could have been changed. To assess this possibility, we prepared a set of tables (Figs. 7 and 8) in which all of the information gathered from all the samples from each individual element (144 tables) or area (28 tables) in the façade was pooled and compared with the chromatic data from the immediate surrounding area. The main results derived from the individualized and comprehensive study performed for each decorative element indicate that minor chromatic changes in the general aspect of the façade derive from an erroneous interpretation of intermediate restoration interventions due to alteration processes that affect specific pigments. In this sense, we confirmed an evolution in many decorative elements, from an early predominance of blue colours (synthetic azurite) to a generalized use of emerald green in those. The introduction of these green copper-based pigments in recent interventions resulted from chromatic alterations of the blue pigments as azurite to light green compounds (atacamite type minerals). With respect to gilded elements, it should be emphasized that most of these decorative elements of the façade, in particular the mocarabes (honeycomb stalactite ornamentation) and to a lesser extend the tabica (riser) and the arrocoe of the monumental eave made of polychromed wood, were invariably gilded (Fig. 9).

Similarly, there was also a permanency of red and black colours in the few decorative elements where they were used. Red and black, and a minor amount of green, were broadly used in the coloured backgrounds over which decorative elements of the polychrome wood were located. Black backgrounds have not change over time, nor has the red background of the epigraphy located in the arrocoe of the central section of the façade. However, that corresponding to

epigraphy of lateral sections was modified repeatedly from green or blue backgrounds (first with malachite green, followed by blue azurite and later green emerald) to finally red background.

6. Conclusions

The results of the present work underline the effectiveness of the methodology implemented during the course of the study. These methods are also being applied, with good results, in the context of other aspects of the palace, such as in the Patio de las Doncellas, in plasterwork, and in alfarjes (wooden decorated ceilings). The fact that it has been possible to relate documentary and historical aspects to materials and data derived from an exhaustive analytical study demonstrates the importance of a multidisciplinary approach in drawing accurate conclusions.

The present methodology enabled us to cross-check all of the information compiled in the study, establish differential and reference data, and identify the complete stratigraphic sequence. This was particularly complex since none of the samples contained the complete sequence and, in general, the number of polychromes
included in a single sample was small, either due to the loss of one or more levels of polychrome or the good condition of the colour when a restoration intervention was performed.

By comparing this sequence of superimposition of strata with documentary references to the different interventions undergone by the façade over the centuries, it was possible to correlate samples by areas. This, in turn, enabled us to associate with a high degree of accuracy the different levels of polychrome and the documented interventions.

The individualized study of each decorative element – drawing on samples from similar decorations in different areas – provided a clear, definitive inventory of the interventions made, based on a comparison between each element and the particular setting in which it was found. A conclusion from this is that the most significant modifications in the façade were introduced during the intermediate interventions.

Acknowledgments

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.culher.2016.01.007.

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