

Supplementary file S1. Studies made on the colors of Iberian manuscripts

As the described in point 1.2 of the main text, the techniques allow the establishment of a molecular palette, and this work will compare the data obtained with the data from the color palette of the Ajuda Songbook. This manuscript has been previously associated with the Alfonsi production due to its iconography and high material quality, but one fundamental question about its identity remains obscure: where it was produced. The comparison between both scriptoria might shed light on this question.

The number of works that study the color palette used in manuscripts produced in the Iberian Peninsula is scarce, to the author's knowledge, and is presented on Table S1 and Table S2. Of the works found, the most ancient manuscript is from the 6-7th century, and their location is doubted (between Spain, Northern Africa, or Italy). Nonetheless, at least one third of the studies were made on Portuguese manuscripts, and the color palette found remains very similar in the 12th and 13th centuries. In the Ajuda Songbook, the same palette was uncovered with the most whopping discovery being the identification of brazilwood lake pigments, in contrast to the characteristic lac dye colors found in the Portuguese manuscripts. After the 14th century, there is a slight change in the pigments used, with the higher use of azurite and the introduction of other pigments. In fact, the knowledge of the molecular color palette of each manuscript, whose location and date are known, is indispensable to comprehend and study objects where this information is undefined or absent. By establishing the characteristic materials used through time, from the 12th to the 16th centuries per example, in a specific location, will widen the possibility of complementing missing information, as is the case for the Ajuda Songbook.

Table S1 – Studies made in medieval illuminated manuscripts produced in the Iberian Peninsula, with a de-scription of the date, name or theme of the manuscript and its origin, colorants found divided by applied pure or in the mixture, techniques used to characterize them and respective reference.

Date	Name Theme	Origin	Colorants	Techniques used	Reference
6 th -7 th 9 th (mod.)	<i>Ashburnham Pentateuch</i>	Spain, Northern Africa or Italy*	Pure: Iron-gall ink, indigo, Egyptian blue, carbon black, red ochre, yellow ochre, red lead, green earth, Verdigris ^o , madder, white lead, mixture of madder and Egyptian blue or indigo, orpiment	FORS, XRF, Spectrofluorimetry, Visible Induced Luminescence	[1]
12 th -13 th	<i>Liber Feudorum Maior</i> (cartulary)	Catalonia	Pure: Lapis lazuli, vermilion, lead white, orpiment, organic yellow pigment, lead-based yellow, copper-based pigment, iron-gall ink, organic carbon-based ink	XRF, MA-XRF, Data treatment (PyMCA software)	[2]
			Mixture: Lead white with yellow colourant		
1189	<i>Lorvão Apocalypse</i>	Portugal	Pure: Vermilion, red lead, orpiment, black	MicroEDXRF, microRaman, microFTIR	[3]
			Mixture: Red lead with orpiment, vermilion with red lead or extenders, red lead with white lead		
12 th -13 th	Manuscripts from Santa Cruz de Coimbra, Santa Maria de Alcobaça and São Mamede do Lorvão monasteries	Portugal	Pure: Vermilion, red lead, orpiment, lad dye, lapis lazuli, indigo, white lead, carbon and bone black, bottle green, azurite and malachite	microRaman, microFTIR, microXRF, microXRD	[4]
			Mixture: Indigo with lapis lazuli, vermilion with lead chalk or red lead, organic dye with vermilion		
12 th -13 th	15 manuscripts of Santa Maria de Alcobaça monastery	Portugal	Pure: Lapis lazuli, azurite, bottle-green, orpiment, red lead, yellow and red organic dyes, white lead, indigo	microRaman, microFTIR, microEDXRF	[5]
			Mixture: vermilion and/or organic red; carbon-based pigment with lead white, lapis lazuli with indigo or carbon. Based black pigment, vermilion with calcite, vermilion with lead white		
12 th -13 th	<i>Alcobaça Beatus</i> (Alc.247)	Portugal	Pure: Lapis lazuli, vermilion, red lead, white lead, bottle green, carbon black, lac dye	FORS, microRaman, microFTIR, Spectrofluorimetry	[6]
			Mixture: vermilion and gypsum, lac dye and gypsum		
13 th -14 th	<i>Cancioneiro da Ajuda</i>	Iberian Peninsula	Pure: Lead white, red lead, vermilion, yellow ochre, mosaic gold, orpiment, lapis lazuli, indigo, azurite, carbon black	microXRF, microRaman, FTIR, FORS and microspectrofluorimetry	[7]
			Mixture: brazilwood with calcium carbonate and lead white, brazilwood with gypsum. Lead white mixed with other pigment to control de hue		

*There is doubt to which of the three places is the place of origin. | o occasional use

Table S2 – Studies made in medieval illuminated manuscripts produced in the Iberian Peninsula, with a de-scription of the date, name or theme of the manuscript and its origin, colorants found divided by applied pure or in the mixture, techniques used to characterize them and respective reference.

Date	Name Theme	Origin	Colorants	Techniques used	Reference
14 th	<i>Winter Breviary</i> (Alc.54)	Portugal	Pure: Azurite, vermilion, red lead, yellow ochre, brazilwood lake, orcein purple, carbon black, lead white and silver	FORs, microRaman, microFTIR, microXRF	[8]
14 th	Cantoral	Puentedura, Burgos, Castile and Lion	Pure: Vermilion, lamp black and azurite	FT-RAMAN	[9]
14 th -15 th	Three cantorals	Spain	Pure: Vermilion, azurite, saffron	FT-Raman	[10]
			Mixture: Lamp black with manganese (IV) oxide, azurite with carbon black		
13 th -16 th	<i>Cancioneiro da Ajuda</i> and <i>Lineage book</i> <i>Winter Breviary</i> (Alc.54) <i>Fernão Vaz Dourado Atlas</i>	Iberian Peninsula; Portugal	Pure: brazilwood, orcein	FORs, MICROFTIR, SERS, microspectrofluorimetry, high-resolution mass spectrometry	[11]
			Mixture: Brazilwood with calcium carbonate and lead white, brazilwood with gypsum		
1430-1450 1470	<i>Lisbon's General Chronicle of Spain</i> and <i>Chronicle of D.Duarte de Meneses</i>	Portugal	Pure: copper-based pigment, red lead, malachite, lapis lazuli, azurite, animal anthraquinone, iron-gall ink	Digital microscopy, h-EDXRF, FORs, MA-EDXRF	[12]
			Mixture: Copper-pigment with lead white, dark brown ochre with lead white		
1459	Privilegios rodados (nobility of Gil Fernandez)	Castile and Lion	Pure: Iron-based ink, lead tin yellow type I, gamboge, azurite, indigo, vermilion, red lead, malachite and lead white**	Portable XRD/XRF, Raman spectroscopy, SEM-EDX and capillary electrophoresis (organic compounds)	[13]
			Mixture: Indigo with azurite, lead with other colorants		
1477, 1498**	Islamic judicial documents	Granada	Pure: Iron-gall ink	SEM-EDX	[14]
			Mixture: Iron-gall ink with carbon black		
1512	<i>Charter of Vila Flor</i>	Portugal	Pure: Iron-based ink, vermilion, red lead, iron-gall ink, organic dye, azurite, lead white, malachite	microEDXRF, fluorescence microFTIR	[15]
			Mixture: Azurite with lead white, vermilion with chalk		

**fifteen of the twenty one documents. The other six document don't have date.

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