

Article

Yellow Dyes of Historical Importance: A Handful of Weld Yellows from the 18th-Century Recipe Books of French Master Dyers Antoine Janot and Paul Gout

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Abstract: Antoine Janot and Paul Gout were 18th-century master dyers who specialised in the piece-dyeing of fine wool broadcloths manufactured in Languedoc (southern France) for exportation to the Levant. They wrote treatises, similarly entitled *Mémoires de Teinture* (Memoirs on Dyeing), illustrated with dozens of dyed textile samples. Janot's is dated 1744, and Gout's is dated 1763. These books are full of extremely valuable information, which is unique to its time: the books are composed of carefully described recipes for every colour, each originally preceded by a sample swatch. The yellows of both master dyers made from weld (*Reseda luteola* L.), belonging to recipes described as Jaune (yellow), were reproduced. The influence of the ingredients, in both the mordanting and dyeing baths, was assessed, and the timings for both baths, which in many recipes are not clear, were tested. This provided key knowledge on the technological processes for dyeing with weld from these 18th c. French masters. The resulting reference samples were preliminarily analysed by a multi-analytical approach. Their chromatic specifications, expressed in the CIEL*a*b*, were compared with those in the *Mémoires*. Moreover, the influence of the recipe on the percentages of the chromophores was assessed by HPLC-DAD. It was found that the bran and tartar in the mordanting bath influence the final obtained colour and that lime is essential to obtain a bright yellow.

Keywords: dyed textiles; weld; *Reseda luteola*; tartar; lime; master dyers



Citation: Santo, M.; Cardon, D.; Teixeira, N.; Nabais, P. Yellow Dyes of Historical Importance: A Handful of Weld Yellows from the 18th-Century Recipe Books of French Master Dyers Antoine Janot and Paul Gout. *Heritage* **2023**, *6*, 7466–7481. <https://doi.org/10.3390/heritage6120391>

Academic Editors: Francesca Volpi, Chiara Anselmi and Manuela Vagnini

Received: 17 October 2023
Revised: 21 November 2023
Accepted: 23 November 2023
Published: 29 November 2023



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1. Introduction

Yellow dyes have been used throughout the millennia as a part of cultural heritage, up until the advent of synthetic colourants in the 19th century. These were used for yellows and also for oranges and greens, in combination with red and blue dyeings, respectively. Because they were often locally grown and rarely imported, unlike indigo and cochineal, for example, yellow dyes can provide crucial information on an artwork's production, in terms of provenance and manufacture [1]. Despite the wide range of plants that can provide an array of yellow hues, the preferred source of yellow in Europe was weld [1].

"Weld for yellows is equivalent to Dyer's Madder for red" [1]. Weld (*Reseda luteola* L. Resedaceae), as shown in Figure 1, or dyer's mignonette (*gaude* in French), can grow in sandy and rocky soils, making it extensively available in wild fields in Europe. It is rich in luteolin-based chromophores, making it very lightfast. The water extracts of the plant are rich in luteolin-7-*O*-glucoside (Lut-7-*O*-glc), in higher amounts, followed by luteolin-3',7-*O*-glucoside (Lut-3',7-*O*-glc), luteolin (Lut), apigenin-7-*O*-glucoside (Api-7-*O*-glc), chrysoeriol glycoside (Chry-glc), luteolin-4'-*O*-glucoside (Lut-4'-*O*-glc), apigenin (Api), apigenin-6,8-di-*C*-glucoside (Api-6,8-*C*-glc), and luteolin di-*O*-glucoside (Lut-di-*O*-glc) [2–6].

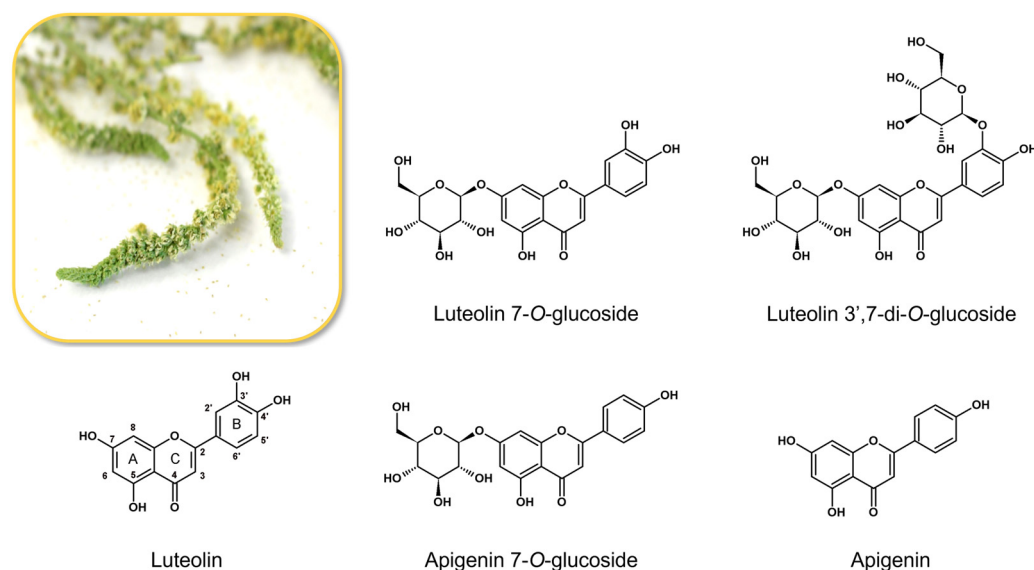


Figure 1. Weld (*Reseda luteola* L. Resedaceae), collected for experiments, and the main chromophores present: luteolin 7-*O*-glucoside (Lut-7-*O*-glc), luteolin 3',7-*O*-glucoside (Lut-3',7-*O*-glc), luteolin (Lut), apigenin 7-*O*-glucoside (Api-7-*O*-glc), and apigenin (Api).

Even though weld largely ceased to be used after the 19th century, its importance in textile dyeing and painting must be taken into consideration when studying historical documents [7]. As a source of yellow, which is one of the primary colours recognised among dyers, weld was also one of the bases of an extensive colour gamut obtained from yellow dyes, such as greens, oranges, light browns, and even top-quality blacks.

Although the flavonoids in weld possess an integrated photo-protective mechanism (named the excited-state intramolecular proton transfer (ESIPT)), depending on their structure, it can be more or less efficient, which results in different colour alterations. Their stability also depends on the surrounding environment, pH, and temperature, among others [8–11]. Recent studies by our group unveiled the quantum yields of degradation for a group of flavonoid dyes, where a stability scale was proposed for luteolin, quercetin, and kaempferol, as well as their glycosides [12]. In this study, it was proven that by using irradiation wavelengths superior to 300 nm, it is possible to simulate degradation conditions. The calculated quantum yields of degradation (Φ_R) were used to determine the stability of the molecules in solution, proving that luteolin and its glucoside are on the same stability scale as indigo, one of the most stable natural dyes known (5.99×10^{-6} and 8.67×10^{-6} , respectively) [12]. Nevertheless, when studying historical textiles, the complexation with alum becomes impossible to ignore, since the stability of these molecules depends on the environment surrounding them. Villela et al. explored the photostability of luteolin in the presence of Al^{3+} , testing different percentages of this ion and concluding that the stability decreases as the concentration of alum increases [13]. After the complexation of luteolin with Al^{3+} , the ESIPT is less likely to occur [13].

Considering this, historically accurate reconstructions can be very important to provide new insights into the formulations that master dyers used to produce stable yellows, as well as to understand their degradation mechanisms. Since their stability is so dependent on the environment, the formulation, i.e., the recipe, has a great impact on their lightfastness.

1.1. Master Dyers: Antoine Janot and Paul Gout

To better understand the use and preparation of yellows in textile production, one must resort to historical sources. The most useful preserved documents are those that describe the dyeing practices of a particular time and illustrate them with dyed textile samples. For this study, we focused on dye recipe books produced by two master dyers

from Languedoc, in southern France, Antoine Janot and Paul Gout, during two successive periods of the 18th century.

Antoine Janot (1700–1778) wrote three *Mémoires* on dyeing. The first, titled “*Mémoire—on trouvera les opérations de la teinture du grand et bon teint des couleurs qui se consomment en Levant avec la quantité et qualité des drogues qui les composent*” (Memoir—on the processes of dyeing the colours that are consumed in the Levant in the good and fast mode of dyeing, with the quantities and qualities of the drugs from which they are obtained) is dated 31 May 1744 and describes the processes to obtain 65 colours. It is illustrated by 61 preserved dyed samples. The second manuscript, entitled “*Mémoire sur les opérations que nous avons faites sur les trois couleurs*” (Memoir on the processes we have used for the three colours herein), is also dated 1744, and the author describes optimised processes for three colours that were very fashionable and in high demand at the time: crimson, wine soup, and celadon green. The third manuscript, entitled “*Mémoire pour les rouges de garance*” (Memoir for madder reds), is dated 1747 and corresponds to a period of high competition between French and English dyers to obtain the brightest reds possible with madder [14,15].

Janot was born in Carcassonne and learned the art of dyeing from his father, Jacques Janot, who then was the master dyer of a Royal Manufacture of fine wool broadcloth located in Bize, a big village to the northwest of Béziers. Later on, Antoine and his father established their own dye workshop in the nearby broadcloth-producing town of Saint-Chinian, about 15 km northeast of Bize, and this became the place where Antoine completed most of his dyeing activities. At the apex of Janot’s career, a powerful family of businessmen and clothiers from Carcassonne with international connections, the Pinels, rented the Royal Manufacture of Les Ayres next door to Janot’s own dyehouse in Saint-Chinian. As a result, Janot became acquainted with François Pinel, who managed the Royal Manufacture, and with Paul Gout, his apprentice and aide. Gout was certainly inspired by Janot’s first *Mémoire* to create his own recipe book with dyed samples nearly 20 years later, when he became the manager and then co-owner of the Royal Manufacture of Bize [16,17]. Their respective perspectives and contributions to the history of dyeing techniques do not overlap but rather complete each other. In his dye house, Antoine Janot dyed the cloths of all the independent clothiers of Saint-Chinian, a small town, which, nevertheless, was, at the time, one of the most important wool-broadcloth-producing centres in France. The whole region of Languedoc counted many centres of production of fine wool broadcloths, specially designed for exportation from France to the Ports of the Levant and onward to all parts of the Ottoman Empire. Tens of thousands of pieces were shipped from Marseille each year. Each piece measured 18–20 m in length per 1.39 m in width and weighed about 10.35 kg when dry. In his dye house, Janot had to dye an average of 12–13 such pieces of cloth per day in the range of colours ordered by his customers, the local clothiers, according to the evolution of colour fashions in the customers’ countries. Moreover, like all his colleagues in France, he had to strictly respect the royal regulations on wool and wool cloth dyeing that had been established in 1669 under Louis the XIV by his Minister Jean-Baptiste Colbert. This meant that he was only allowed to use high-quality ingredients, recognised as giving the fastest dyes in amounts and proportions that had been fixed by an agreement with the best dyers of the kingdom [14,15].

Paul Gout’s situation was slightly different when, in 1763, he wrote his own manuscript entitled “*Mémoires de teinture*” (Memoirs on dyeing) [16,17].

This was, firstly, because, as the director of a Royal Manufacture, he had to manage all the aspects of production, from the selection of wools to the checking and packing of the broadcloth pieces. Dyeing and finishing processes were the only operations completed in-house. Therefore, not as many cloth pieces were dyed each day in the Royal Manufacture as in Janot’s specialised dye house. The director of a Royal Manufacture had to either employ a master dyer to supervise the dyeing processes or be trained as a master dyer himself, which was Paul Gout’s case. He, consequently, had both the technical knowledge and the freedom to choose what colours he was going to produce each day, according to

his knowledge of the evolution of fashions among his distant customers in the Eastern Mediterranean countries. He was also free to propose new shades, which could allow him to optimise the expense and the use of his high-quality colourants. He was also freer than Janot to devise new colours and shades, where he introduced small proportions of colourants that had been prohibited by the royal regulations on dyeing in fast dyes. This was because the liberal ideas of the Age of Enlightenment had made considerable progress among French political elites during the 20 years that had passed since Janot had written his own *Mémoires*. Experiments in new technical processes had become less risky or were even encouraged [16,17].

When analysing Janot's manuscript, one cannot help but notice the considerable percentage of recipes for yellows and light browns: almost a fourth of the total recipes (Figure 2). Most greens and some oranges were also produced by top-dyeing blue or red with a yellow dyebath. In Gout's book, weld figures in 33% of the 157 recipes illustrated with samples. Hence, yellow dyes are a striking presence in these books [14–17]. Moreover, these manuscripts are full of extremely valuable information, which is unique to its time. They are composed of carefully described recipes, each originally preceded by a sample swatch, as shown in Figure 3. This gives us the opportunity to reproduce the colours from these books and compare the analytical results to the original swatch. The books are, at the same time, a written record and an artwork prepared by extremely knowledgeable master dyers, who took a lifetime to perfect these recipes. Therefore, these books are the perfect place to start to uncover the hidden secrets of lost yellow colours.

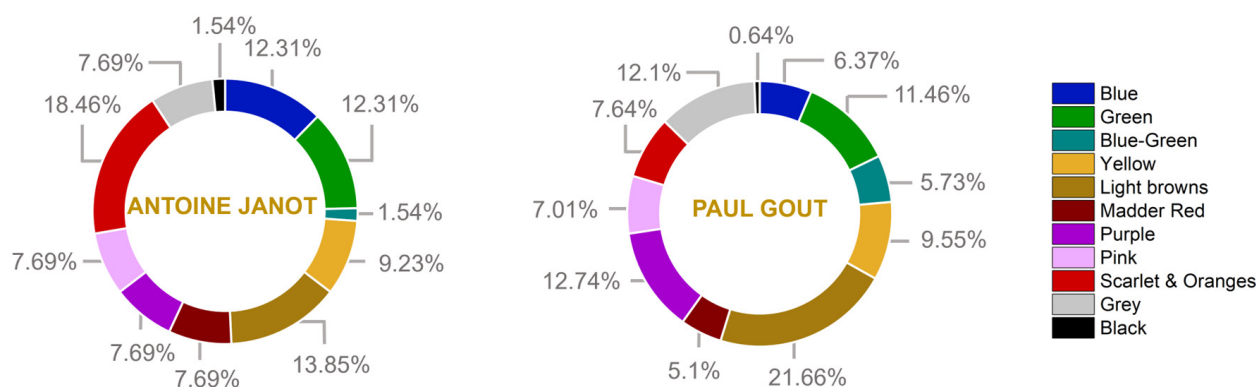


Figure 2. Percentage of each colour recipe present in the *Mémoire de Teinture* of both dye masters, Antoine Janot and Paul Gout. It is clear that the yellows and light browns represent more than a fourth of the total recipes. Moreover, they were also used to prepare oranges, greens, and some reds.



ANTOINE JANOT, 1744



PAUL GOUT, 1763

Figure 3. Yellow swatches from the *Mémoires de Teinture* of both dye masters, Antoine Janot and Paul Gout. The Antoine Janot manuscript is preserved at the Archives Départementales de l'Hérault in Montpellier, France, with the reference C/5569.

1.2. Characterisation of Organic Colourants

The characterisation of organic colourants is still a challenge to this day. Most in situ analytical techniques present several disadvantages for the identification of dyes in artwork. The infrared signals of the dye are difficult to identify due to the presence of other materials, such as the fibres, while, with Raman spectroscopy, the organic dyes usually present strong fluorescence signals, masking the chromophore's fingerprint. Nevertheless, this can be overcome by surface-enhanced Raman spectroscopy (SERS), which provides a fingerprint identification of the chromophore that is present [18,19]. Fibre optic reflectance spectroscopy, although is a promising tool for a preliminary scan of the possible materials present, it does not allow for the identification of specific dye sources belonging to the same molecular family (such as anthraquinones or flavonoids) [20]. Molecular fluorescence in the visible proved to be a powerful technique for the identification of the dye source and also of the formulation [21]. Nevertheless, it requires an extensive database of references and is usually used in synergy with other techniques, such as SERS or HPLC.

Not many techniques provide as many insights into each chromophore as high-performance liquid chromatography (HPLC). Coupled with diode-array detection and mass spectrometry, it is a very robust technique for the identification of organic colourants [22] and is still the most used technique for the study of historical textiles. It allows for the characterisation of all the chromophores present and of new compounds (with mass spectrometry), enabling the identification of the source material. Because of the possibility of the quantification of the chromophores, HPLC could help us understand the influence of certain ingredients on the final composition of the swatches. Therefore, for this study, we decided to ally the study of colour, through colourimetry, with the characterisation of the final molecular composition, through HPLC-DAD.

This study is being conducted within the REVIVE project, "The threads of the past weaving the future: The colours from the Royal Textile Factory of Covilhã, 1764–1850". This project focuses on the history of the Portuguese woollen industry and, in particular, on the study of the dyeing and production processes used from the 18th c. onwards. The Royal Textile Factory of Covilhã, active between 1764 and 1850, was established by the Marquis of Pombal within his policy of promoting the modernisation of national wool production. During this period, the Marquis hired many foreign masters, including French master dyers, to work in the Royal Manufacture. Addressing and understanding the recipes from French master dyers from the 18th century gives us a broader view into French production and how it contributed to creating the unique identity of the Royal Textile Factory of Covilhã.

2. Materials and Methods

2.1. Manuscripts

Antoine Janot's manuscript is preserved in the Archives Départementales de l'Hérault in Montpellier, France, with reference C/5569. Paul Gout's manuscript is preserved in a private collection and is not accessible to the general public.

2.2. Materials

All solvents that were used were HPLC grade. For all chromatographic studies, as well as dye extraction, Millipore ultrapure water was used. Potassium aluminium sulfate dodecahydrate ($\text{AlK}(\text{SO}_4)_2 \cdot 12\text{H}_2\text{O}$) and calcium oxide (CaO, mentioned as lime in the recipes) were purchased from Sigma-Aldrich, while organic bran was purchased from the local supermarket. Crude red tartar ($\text{KC}_4\text{H}_5\text{O}_6$) was collected from barrels of organic red wine produced by Patrick and Alexis Maurel's winery Terres du Pic, Mas de La Liquière, 34380 Mas-de-Londres, France.

Flowering branches of *Reseda luteola* were collected in May 2021 and 2022 by Paula Nabais, from wild populations near Bucelas, north of Lisbon, Portugal (38°54'021" N–9°60'30" W). In May 2023, they were collected by Mara Santo, from wild populations near Caparica, south of Lisbon, Portugal (38°65'9597" N–9°20'4133" W). The plant material was spread in a tray and air-dried in the dark, in a ventilated area at 20 °C. We dried freshly collected and

dried was used except for Gout's recipe with "old weld", in which dry weld collected the previous year was used.


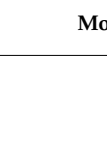













The wool is 100% pure wool broadcloth woven from the Ile-de-France breed of sheep at Eric Carlier's workshop: Le Passe-trame, rue du Moulin Gau, 81660 Payrin-Augmontel, France.

2.3. Weld Dyed Textiles

The recipes are described in the books by Dominique Cardon [14–17]. Antoine Janot only used weld (*Reseda luteola*) as the plant source for the formulation of his yellows, while Paul Gout used other sources of yellows, besides weld, such as flax-leaved daphne (*Daphne gnidium*), young fustic (*Cotinus coggygria*), and old fustic (*Maclura tinctoria*). But one thing to take note of is that both masters used weld to produce the colour they named *Jaune* (yellow), indicating that they associated weld with the colour yellow. However, a combination of weld and flax-leaved daphne was used for one of Gout's samples of *Limon*, another yellow shade.

For Antoine Janot, the colours *Jaune*, *Jaune citron* (lemon yellow), *Jaune soufré* (sulphur yellow), and *Jaune paille* (straw yellow) were prepared using the same dyebath, but by dipping the cloths one after the other, obtaining gradually lighter shades of yellows due to the decrease in the concentration of flavonoids in the bath. Gout describes a yellow *Jaune* and a lighter version of it, *Limon*, which uses 48% weld or flax-leaved daphne. Because of the possible use of another yellow source, *Limon* was left out of this first attempt at reconstructing Gout's yellows. In the second part of his *Mémoires*, Gout describes the preparation of a *Jaune* with "old weld", so this recipe was also prepared with dry weld collected during the previous year to understand the differences. See Table 1 for the recipes described in the *Mémoires* of both masters.

Table 1. Recipes of the yellows of Antoine Janot and Paul Gout. Colourimetry measured by Dominique Cardon. Colour of the samples present in the *Mémoires* (pp. 74–75 in [15]; pp. 79 and 126 in [17]). The percentage of the materials is represented for dried WOF (weight of fibre).

	Recipe	CIE L*a*b* Data	Mordanting Bath			Dyeing Bath (Gaudage)	
Antoine Janot	<i>Jaune</i>	L* = 55.49 a* = 14.20 b* = 60.34					
	<i>Jaune citron</i>	L* = 63.88 a* = 13.11 b* = 66.00					
	<i>Jaune soufré</i>	L* = 70.04 a* = 6.15 b* = 60.67	 Alum 16%	 Bran 5.5%	 Tartar 2%	 Weld 80%	 Lime 1%
	<i>Jaune paille</i>	L* = 66.59 a* = 3.74 b* = 41.26					
Paul Gout	<i>Jaune</i>	L* = 66.93 a* = 20.36 b* = 63.28	 Alum 16%	 Tartar 8%	 Weld 64–68%	 Lime 1%	
	<i>Jaune (old weld)</i>	L* = 64.71 a* = 10.11 b* = 72.70	 Alum 12%	 Tartar 4%	 Weld 68	 Lime 1%	

All proportions were based on the dry weight of the textile. Then, 10 × 10 cm² of unbleached woven wool fabric was pre-mordanted with alum (AlK(SO₄)₂·12H₂O), crude red tartar (KC₄H₅O₆), and wheat bran, in the proportions mentioned in the manuscripts. The dyeing was done by preparing an extraction of weld in water and then adding lime

(CaO) for some experiments. We first started our experiments without the addition of lime, so we used fresh plant material.

The mordanting bath began with the weighting of each ingredient for each sample. After this, the ingredients can be added to the beakers with about 500 mL of Mili-Q water. For each sample, a different bath was prepared. Glass beakers with water and all ingredients were heated under magnetic agitation until complete dissolution of alum, which was around 40 °C. After this, the wool broadcloth was added. The wool stood in the beaker until the water reached 90 °C. Once this temperature was reached, the sample stayed under the same conditions for 30 min. After this time, the wool was taken out of the beaker and was left to dry for at least 24 h. After this, the samples were washed with Mili-Q water and left to dry once again until they were ready to dye. It is hard to determine the purity of the water used by the master dyers in the 18th century. For this reason, we decided to use ultrapure water (Mili-Q) in our reconstructions, ensuring that no other impurities affected the accuracy of our results.

For the dyeing bath, the weld plant was put into tea bags to prevent the plant material from attaching to the broadcloth. The tea bags are made of abaca pulp and cellulose and are free from chlorine, glues, and whitening agents. The beakers were filled with 500 mL of Mili-Q water and heated with the weld and lime (when used). When the temperature reaches 60 °C, the water starts to turn yellow, meaning that the chromophores are being successfully extracted. After the bath reaches a temperature of 90 °C, the mordanted wool can be added. The samples stayed in the dyeing bath for either 1, 8, 10, or 30 min, and lime was only used for the baths with a duration of 30 min. For each variation in the recipe, individual dyeing baths were prepared to guarantee that the weld concentration stayed the same.

The subsequent yellows were prepared by adding more wool broadcloths, one at a time, and removing them after 30 min each. In Janot's yellows, for *Jaune*, a dyeing bath of 30 min was performed; for *Jaune citron*, the same bath was used for 30 min, the same bath was used for the next 30 min to obtain *Jaune soufré*, and, lastly, for *Jaune paille*, the same bath was used for another 30 min. See Figure 4 for the general scheme.

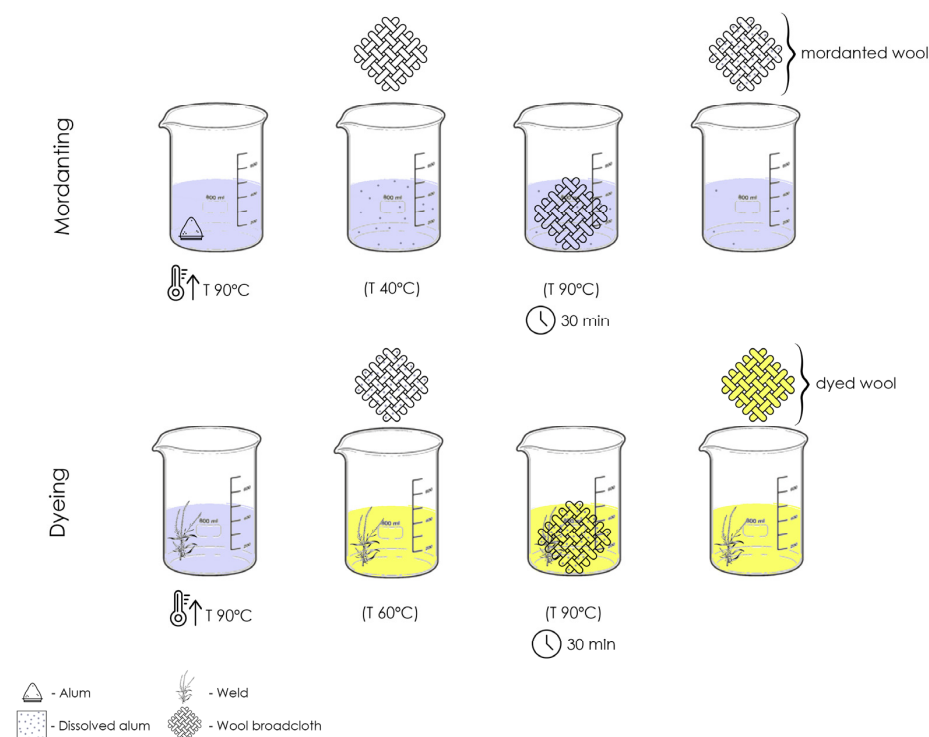


Figure 4. General scheme for the mordanting and dyeing baths used to prepare the reconstructions. When required by the recipe, red tartar and bran were also added to the mordanting bath, along with alum, while lime was added to the dyeing bath before the addition of the wool broadcloth.

2.4. Analytical Techniques

2.4.1. Colourimetry

For measuring colour, a handheld spectrophotometer Lovibond TR 520 with a diffused illumination system, 8° viewing angle, and 48 mm integrating sphere was used. The measuring aperture was 8 mm in diameter. Equipment calibration was performed with white and black references. Colour coordinates were calculated defining the D65 illuminant and the 10° observer. The colour data are presented in the CIE-Lab 1976 system. In the Lab Cartesian system, L^* , relative brightness, is represented by the z-axis. Variations in relative brightness range from white ($L^* = 100$) to black ($L^* = 0$). The (a^* , b^*) pair represents the hue of the object. The red/green y -axis plots a^* , ranging from negative values (green) to positive values (red). The yellow/blue x -axis reports b^* going from negative numbers (blue) to positive numbers (yellow).

2.4.2. High-Performance Liquid Chromatography with a Diode Array Detector (HPLC-DAD)

Approximately 1.0 mg of each dyed sample was extracted by placing it in a microtube with 1 mL solution of oxalic acid (0.2 M) in methanol:acetone:water (0.1:3:3:4, $v:v$), with magnetic agitation, at 60 °C for 1 h. Each solution was filtered with a 0.45 µL PTFE syringe filter prior to HPLC-DAD analysis.

All samples' extracts were analysed in triplicate using a Merck-Hitachi Elite LaChrom HPLC-DAD using an Agilent Poroshell 120 CS-C18 column 150 × 4.6 mm i.d., with 2.7 µm pore size column, thermostated at 25 °C (Merck-Hitachi Column Oven L-2300). Detection was carried out at 350 nm using a diode array detector (Merck-Hitachi Diode Array Detector L-2455). The selected samples were the final versions of all variations, with 30 min of dyeing.

The mobile phases were composed of solvent A, 100% (v/v) methanol, and solvent B, 0.1% (v/v) perchloric acid in water [2]. The flow rate was 0.60 mL/min, the injection volume was 25 µL, and the gradient method started with a two-minute isocratic 7% A gradient, followed by a linear gradient ranging from 7% A to 15% A in 8 min and then reaching 75% A in 25 min, 80% A in 2 min, and 100% A in 2 min.

Two calibration curves for luteolin and luteolin-7-*O*-glucoside were obtained using a concentration range of 5.00×10^{-4} – 9.99×10^{-7} M and 4.38×10^{-4} – 5.84×10^{-7} M, respectively. Each standard was prepared in duplicate, and then each solution was analysed in duplicate as well, giving four measurements per standard. Unknown concentrations were determined from the regression equations $yy = 1.33 \times 10^{11}xx - 9.98 \times 10^5$ and $r^2 = 0.9939$ for Luteolin-like aglycone compounds and $yy = 1.70 \times 10^{11}xx - 1.23 \times 10^6$ and $r^2 = 0.9810$ for Luteolin-7-*O*-glucoside-like compounds.

All results were expressed as mean ± standard deviation and presented as molar equivalents of the chromophore.

2.4.3. Statistical Analysis

Values are expressed as the arithmetic means ± standard deviation. Statistical significance was performed by two-way analysis of variance (ANOVA) followed by Šidák's and Tukey's multiple comparisons test, with a significant difference with 95% confidence interval ($p < 0.05$) and GP: $p > 0.05$ (ns), $p \leq 0.05$ (*), $p \leq 0.01$ (**), $p \leq 0.001$ (***), $p \leq 0.0001$ (****), using GraphPad Prism 9.0.0 software (San Diego, CA, USA).





















3. Results

3.1. Gaudage—Dyeing with Weld

The process for the recipes of both Antoine Janot and Paul Gout starts with a mordanting bath. The ingredients for each recipe of both masters can be seen in Table 1. Variations of these recipes were explored in order to see the influence of each ingredient individually present in the mordanting and dyeing baths. Because the indication of the time for dyeing was based on the turns the textiles made around the cauldron [14–17], the dyeing bath

time was also studied. The bath times that were used were 1, 8, and 10 min. Because the colourimetry showed that more time in the dyeing bath was needed to approach the original's colour, we, finally, tested for 30 min with the addition of lime at 1%. See Table 2 for the variations in each recipe.

Table 2. Variations in each recipe, to understand the influence of the ingredients and of the time of extraction. The pHs for the mordanting and dyeing baths are also described. Lime was added to all variations for 30 min in the dyeing bath.

Recipe	Mordanting Bath	pH of the Mordanting Bath	Dyeing Bath	pH of the Dyeing Bath
Antoine Janot				
<i>Jaune</i>	A.J.1  Alum 16%	3.72		
	A.J.2  Alum 16%  Tartar 2%	2.85		
	A.J.3  Alum 16%  Bran 5.5%	3.57	 Weld 80% fresh	6.36 pH _{lime} = 8.93
	A.J.4			
<i>Jaune citron</i>	A.J.5  Alum 16%  Bran 5.5%  Tartar 2%	2.90		
<i>Jaune soufré</i>	A.J.6			
<i>Jaune paille</i>	A.J.7			
Paul Gout				
<i>Jaune</i>	P.G.1  Alum 16%	3.72	 Weld 64% fresh	6.34 pH _{lime} = 10.38
	P.G.2  Alum 16%  Tartar 8%	2.67		
<i>Jaune (old weld)</i>	P.G.3  Alum 16%	3.72		
	P.G.4  Alum 16%  Tartar 8%	2.67	 Weld 68% old	6.14 pH _{lime} = 7.81
	P.G.5  Alum 12%	3.66		
	P.G.6  Alum 12%  Tartar 4%	2.48		

All variations were completed with 1, 8, and 10 min of the broadcloth in the dyeing bath. Variations A.J.4-7, P.G.2, and P.G.6 were also prepared for 30 min in the dyeing bath, using 1% lime, as described in the recipes.

The main difference between the mordanting baths of both dye masters is the presence of bran in Antoine Janot's recipes. Bran is the hard outer layer of the wheat kernel.

Tartar, the crystalline salt deposit formed in wine barrels during wine fermentation (potassium hydrogen tartrate mixed with calcium tartrate and anthocyanins from the wine), was used as an agent to improve colour and stability. Its stability role, however, needs to be

accessed by performing artificial ageing experiments in the future. The percentages vary within the recipes: Antoine Janot uses 2% red tartar, while Gout proposes 8% when using fresh weld and 4% when using old weld. This most likely has an influence on the final colour, as we ascertained with colourimetry. Gout also uses less plant material, 64–68%, while Janot uses close to the weight of the textile, 80%. Gout was known for reusing baths as much as possible and avoiding wasting any material. Therefore, it is interesting to compare the yellows from both dye masters to understand if a more than 20% difference in the quantity of the plant material would influence the final obtained colour. In this sense, Gout also proposes the use of 12% alum with the old weld, in contrast with the 16% for the *Jaune* by both dye masters.

It is also important to consider the influence of the pH. The optimum pH for mordanting wool with Al^{3+} is in the range of 3–4 [23], since above pH 6 aluminium compounds start to show a low solubility, with the creation of aluminium coagulants [24]. According to Edgar and Simpson, the absorption of Al^{3+} by the wool increases the pH of the bath [23]. The addition of tartar might also be to lower the pH of the mordanting bath. If the pH of the water used by both master dyers was higher, it would make sense to add tartar to optimise the absorption of alum by the wool, thus considerably increasing the percentage of colourants that become fixed onto the wool [25]. This was mentioned by Gout in his treatise “White tartar and red tartar have the same properties in our dyes, where they are of great use to prepare the cloth to take up the colour” (p. 50 in [16]). Moreover, Valery Golikov [26] found that the use of tartar and polyphenolic compounds such as gallotannins allows for the use of water of any quality. This prevents the precipitation of unwanted compounds (such as other salts) on the fibre surfaces, allowing for more homogeneous mordanting and dyeing. In this case, the role of bran is also clear, since bran is known to have a substantial number of polyphenolic compounds [27]. In Antoine Janot’s recipe, bran would prevent the precipitation of impurities, such as iron salts, onto the wool fibres.

The pH of the dyeing bath becomes alkaline with the addition of lime (CaO). Favaro et al. studied the various luteolin species through fluorimetric titration [28]. They found that there are four species within the pH range of 2–12: neutral form (pH < 5), mono-anion (pH ~7), di-anion (pH ~9), and tri-anion (pH ~12). They also described the order of deprotonations: 7-OH; 4-OH; 3-OH and/or 5-OH [28]. The addition of dried plant material would slightly lower the pH, justifying the addition of lime to increase it, optimising the conditions in which it is more likely for metal chelation to occur when the mordanted textile is added to the dyeing bath. This occurs through the OH at C5, only deprotonated at pH \approx 10.3, and the carbonyl at C4 [28]. The use of additives to increase the pH of the extraction bath to optimise metal chelation was already reported in weld lake pigment recipes of the 19th century [2].

Moreover, Gout mentions that lime aids in the extraction of the colour, similar to what was described when potash is added: “. . . put a small amount of quicklime, with ashes, in the bottom, on the first bundle of weld; this is what makes it give out its sap more easily” [16,29]. Kirby et al. described that the addition of potash to a dyeing bath can result in the greater solubility of the dyestuff, and, in the case of weld, it aids in preserving the glycosides in the colouring matter, which may degrade due to the temperatures of the dyeing bath [29].

3.2. Colourimetry

Colourimetry was performed to determine the accuracy of the reconstructions, comparing the results to the originally dyed samples of both Antoine Janot and Paul Gout. The colourimetry of the original samples can be found in Table 1. Colourimetry also allows us to better understand which ingredients have the highest influence on the final colour and in what way.

After the analysis of all the samples, it was found that the ingredients that influenced the colour the most were red tartar and lime, used by both Janot and Gout in different

amounts (see Tables 1 and 2 and Figure 5 and Supplementary Materials Figure S1 and Tables S1 and S2).

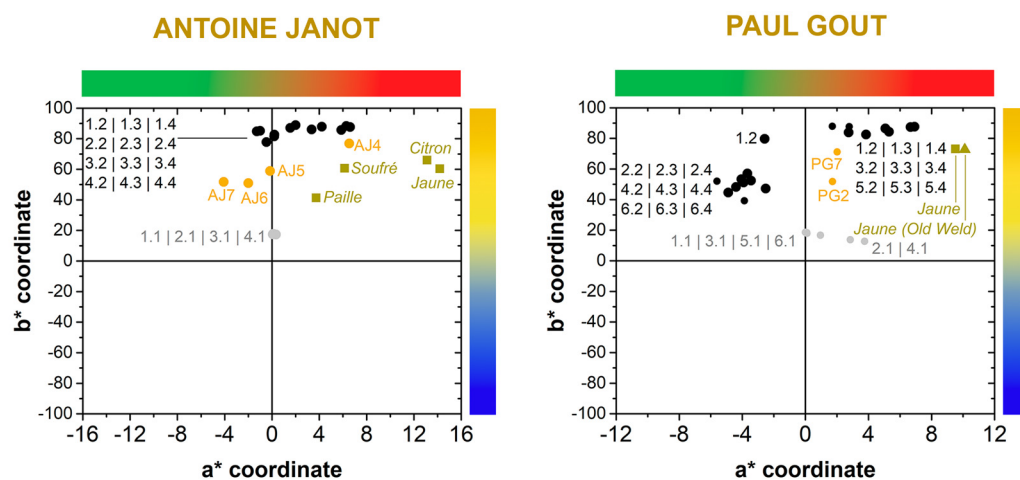


Figure 5. Representation of the a^* and b^* coordinates of all the variations of the recipes (*black* and *bright yellow* for the samples with lime), plotted with the original colours of both dye masters (*dull yellow*).

The reconstruction of the yellow recipes for both dye masters showed that red tartar influences all the coordinates, and the higher the amount of tartar is, the bigger the difference. It was also clear that lime impacts both the L^* and the a^* coordinates. Finally, when looking at dyeing times, it is clear that longer dyeing baths lead to darker tones (a lower L^*) and a more reddish hue (a higher a^*). The 30 min of dyeing variations with lime and dipping successive cloths in the same dyebath leads to lighter colours (a higher L^*), less red (a lower a^*), and also less yellow (a lower b^*), indicating the exhaustion of the yellow colourants in the dyebath, as is expected. For Gout, there is a clear separation of the samples using alum from those using alum and tartar, as seen in Figure 5.

We will address, in future studies, the influence of different sources of red tartar and also the influence of the natural ageing of the historical swatches. It is important to note that the original colours of the 18th-century swatches might have changed due to natural ageing. This might also justify the difference in colours between the original swatches and the reconstructions. Therefore, it becomes necessary to artificially age the reconstructions and begin to understand the conservation condition of the historical samples.

In conclusion, when analysing the colourimetry of the original swatch, where old weld was used by Paul Gout, it appears that there is minimal variation in both hue and brightness, in comparison with the other *Jaune*. This suggests that Gout was able to develop a method that demanded a lower amount of weld, tartar, and alum than the first yellow recipe (*Jaune*), while producing nearly the same colour. This was also the case for our reconstructions. Therefore, we consider that exploring the use of an older weld (aged for more than a year) could be a promising avenue for future investigations.

3.3. HPLC-DAD Analysis

3.3.1. Dyeing Bath

To better understand these shifts in the colour, HPLC-DAD of selected samples was performed. Figure 6 illustrates the results of each chromophore, quantified as equivalents of Lut-7-O-glc or the aglycone form (Lut).

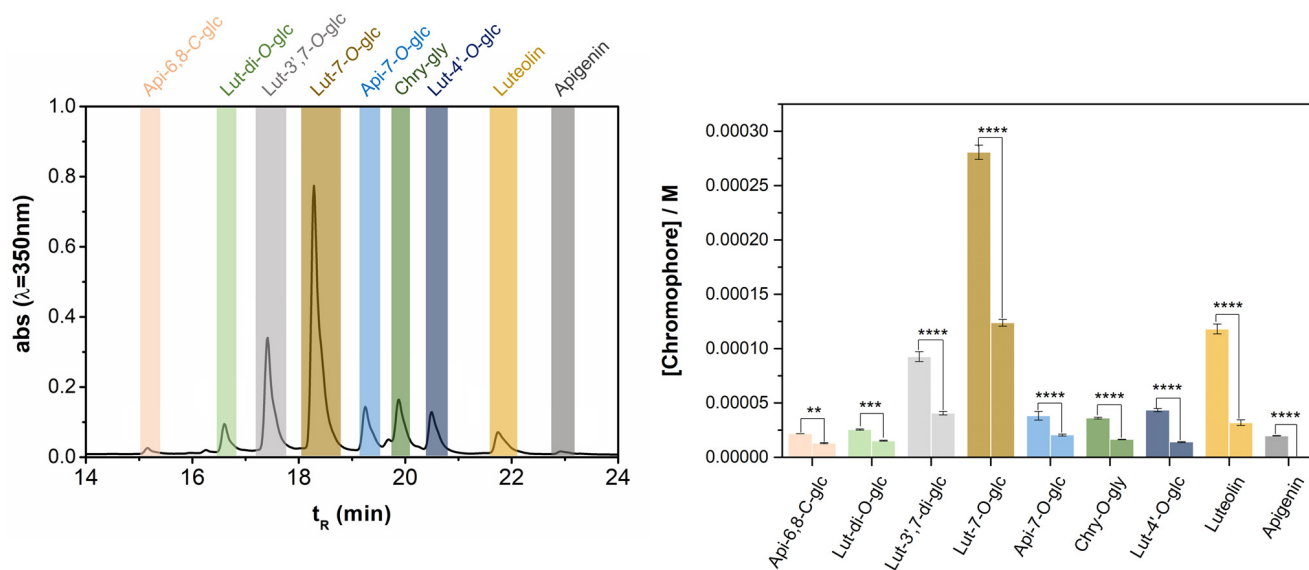


Figure 6. Chromatogram ($\lambda = 350$ nm) of a weld extract (**left**), with the quantification of each chromophore identified in the initial weld dyeing bath (**right**, first column), following Antoine Janot's recipe, and, in the final dyeing bath (**right**, second column), after the dip of successive samples (AJ.4-AJ.7). Values expressed in M of equivalents of Lut-7-O-glc (glucoside forms) or Lut (aglycone forms). Statistical significance was $p < 0.05$, GP: $p \leq 0.01$ (**), $p \leq 0.001$ (***), $p \leq 0.0001$ (****).

All compounds show a concentration decrease. Apigenin is the only compound that was not detected in the final dyeing bath. Lut-7-O-glc is, without a doubt, the compound present in a higher concentration, followed by Lut.

Apart from this, Lut is the compound with the highest reduction (73%). This fact can be an indication that this compound may have a higher binding affinity to the mordanted fibres.

3.3.2. Wool Broadcloth Dyed Samples

Each dyed sample was submitted to the already described extraction process and analysed by HPLC-DAD. Figure 7 shows the quantification results for Antoine Janot's samples, while Figure 8 shows the quantification results for Paul Gout's samples. Tables S3 and S4 in the Supplementary Materials indicate the concentrations of all the detected compounds.

The broadcloth dyed extracts' quantifications were performed in order to compare all the prepared recipes. For this reason, all dyed broadcloths were extracted following the exact same procedure. By comparing Figures 6–8, it was possible to identify the same nine compounds in the weld dyeing bath and after the dye extraction process, using both Antoine Janot and Paul Gout's recipes.

When analysing the HPLC results from Figures 7 and 8, one must remember that a compound that presents a lower concentration may indicate that this compound is able to strongly bind to the textile, which is more difficult to extract. For instance, comparing the major compound present in the dyeing bath (Lut-7-O-glc) and the major compound extracted in both Paul Gout and Antoine Janot's reproductions (Lut) can indicate that Luteolin binds less to the textile than Luteolin-7-O-glucoside. Since we are using a mild extraction procedure [30] and knowing that flavonoid glucoside forms may be converted into their aglycone forms upon using temperatures higher than 60 °C and acids stronger than oxalic acid 0.2 M [31], we do not believe that we are promoting the hydrolysis of Lut-7-O-glc to Lut while extracting the dyed broadcloths.

Nevertheless, Lut-7-O-glc and Lut are the major detected compounds, with Lut being the compound extracted in higher amounts, particularly in Antoine Janot's recipes AJ.1, AJ.2, and AJ.4 (Figure 7). This may indicate that Paul Gout's variations are stronger in terms of the colour compound fixation, since less chromophore is extracted from the dyed samples (Figure 8).

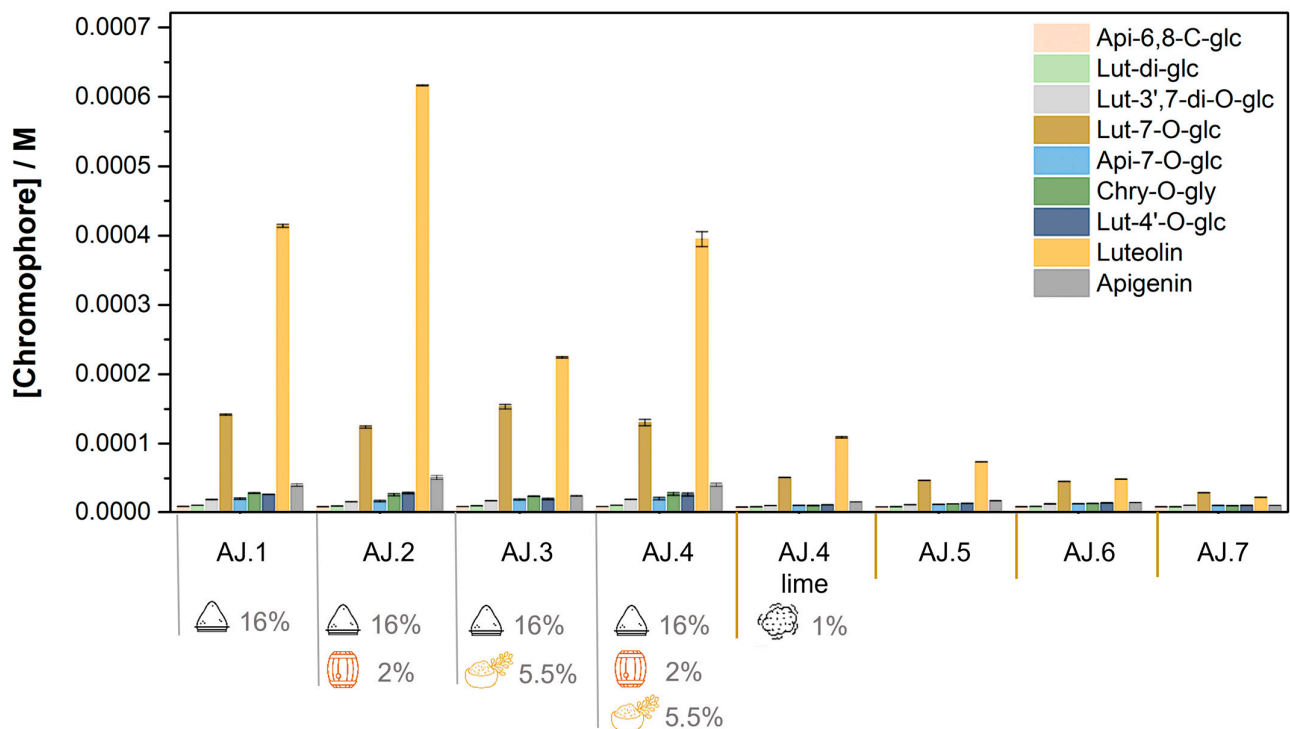


Figure 7. Quantification of each chromophore identified in Antoine Janot's samples. Values expressed in M of equivalents of Lut-7-O-glc (glucoside forms) or Lut (aglycone forms).

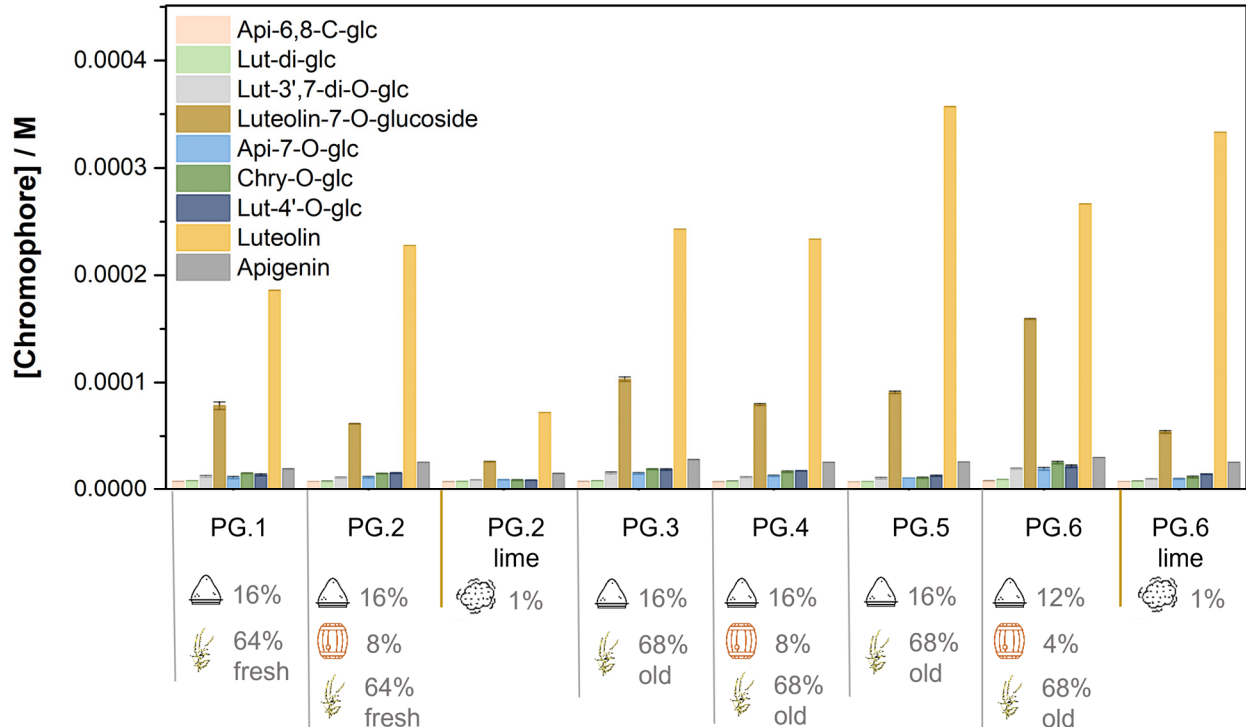


Figure 8. Quantification of each chromophore identified in Paul Gout's samples. Values expressed in M of equivalents of Lut-7-O-glc (glucoside forms) or Lut (aglycone forms).

Particularly for Antoine Janot, the variation with alum and red tartar (AJ.2) is the sample with a higher amount of extracted Luteolin (69% total amount of chromophores), with a 1:0.20 ratio of Lut:Lut-7-O-glc. In contrast, the variation with alum and bran (AJ.3)

has a low amount of Luteolin (15% total amount of chromophores), with a 1:0.44 ratio of Lut:Lut-7-O-glc. By comparing these results with the CIELab analysis (Figure 5), AJ.3 shows a strong yellow tone and the lowest lightness (L^* coordinate). This shows that, by extracting a lower amount of Lut with the oxalic acid (0.2 M) solution, the chromophore strongly binds to the textile using this recipe.

Looking at the last four samples from Antoine Janot's recipes, AJ.4 *Jaune*, AJ.5 *Citron*, AJ.6 *Soufré*, and AJ.7 *Paille*, these samples were prepared by successively dipping the cloths in the dyeing bath, with 30' dyeing each, with the addition of lime. The HPLC-DAD results showed that these are the samples have a less extractable chromophore concentration, particularly Lut and Lut-7-O-glc. This indicated that the higher pH of the dyeing bath, due to the addition of lime, improves the efficacy of the binding.

In contrast, the use of red tartar diminishes the yellow's intensity in Paul Gout's yellows. These recipes show a higher extraction of Lut, which may indicate that these may be less strongly bound complexes.

Apart from PG.6 *Jaune*, the variations with 30 min and lime appear to extract less than the previous ones, which may indicate that the dye is more strongly linked to the textile. This is significant for the chromophores extracted in PG.6, when compared with PG.2, both of which include lime. In fact, the PG.6 lime extracts more chromophores than that of PG.2, indicating the importance of the pH in the dyeing bath. There is no significant difference between using recently collected and dried or old weld.

4. Conclusions

This preliminary approach to the reconstructions of the yellow recipes of both dye masters allowed a closer look at the 18th-century manufacture of dyed broadcloth using weld. When analysing Janot's manuscript, one cannot help but notice the considerable percentage of recipes for yellows and light browns: almost a fourth of the total recipes. In Gout's book, weld figures in 33% of the 157 recipes illustrated with samples. Hence, yellow dyes are a striking presence in these books.

It was found that red tartar has an immense influence on all the coordinates of the CIEL*a*b* system, and the higher the amount of tartar is, the bigger the difference is. Moreover, by using 5.5% bran, Antoine Janot could improve the dyeing recipe by achieving a stronger yellow tone than by just using alum, as shown by HPLC. Both these ingredients were shown to have an immense influence on the final obtained tonality and also on the strength of the complex with alum. The latter needs to be further explored.

The percentage of weld had less influence than expected. In fact, Gout's recipe for *Jaune* using 68% old weld showed to be as yellow as Janot's *Jaune* using 80% recent weld. That being said, in terms of dyeing efficiency, Paul Gout's recipes seem to be more promising, for they use a lower amount of weld, and the chromophores responsible for the yellow dyeing bind more strongly to the textile than Antoine Janot's recipes.

In the dyeing bath, lime is the decisive factor: it not only influences both the L^* and the a^* coordinates (red–green), while having little effect on the b^* coordinate (yellow–blue), but also impacts the strength of the complex. All the samples with lime, with the exception of PG.6, were shown to extract less, which is an indication that the dyes are strongly bound to the textile, considering the intensity of the colours measured by colourimetry. These results directly corroborate the need for a more alkaline dyeing bath, using lime, which would optimise the conditions in which it is more likely for metal chelation to occur: lime is essential to obtain a bright and stable yellow.

Finally, we consider that exploring the use of older weld (left to dry and kept for more than a year) could be a promising avenue for future investigations. Moreover, considering that the historical reconstructions are still not a match with the originals colourimetry-wise, it would be important to explore other sources of red tartar to better understand the influence of different percentages of lime, i.e., a further increase of the pH of the dyeing bath. The analysis of the original swatches would also provide greater insight into these recipes.

These results contributed to technological source research, by undertaking a thorough study of the manufacturing processes of weld yellows by 18th-century master dyers. These reconstructions were accomplished with as much historical accuracy as possible, which allows for the further study of degradation mechanisms and the development of analytical techniques that will permit their identification in aged artwork. Moreover, they were a first step in understanding the specific formulations of French dyers, so we can further address their influence on the production processes and dyeing of wool broadcloths in the Portuguese Royal Manufacture of the 18th century.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/heritage6120391/s1>. Figure S1: Colourimetry analysis for every recipe variation for both dye masters. In grey are the undyed mordanted cloths; in yellow are the final versions for each recipe, according to Table 2. Per variation, the dyeing times tested were 1, 8 and 10 minutes. The final version, 30 minutes in the dyeing bath, also has the addition of lime.; Table S1: Colourimetry data for Antoine Janot samples with the average and the percentage for standard deviation. In grey are the undyed samples, in yellow are the final versions; Table S2: Colourimetry data for Paul Gout samples with the average and the percentage for standard deviation. In grey are the undyed samples, in yellow are the final versions, Table S3: Concentration of all chromophores extracted from all samples prepared following Antoine Janot's recipes, expressed in M of equivalents of Luteolin-7-O-glucoside (glucoside forms) or Luteolin (aglycone forms). Same superscripts within each column (a–k) indicate not significant differences ($p > 0.05$); Table S4: Concentration of all chromophores extracted from all samples prepared following Paul Gout's recipes, expressed in M of equivalents of Luteolin-7-O-glucoside (glucoside forms) or Luteolin (aglycone forms). Same superscripts within each column (a–j) indicate not significant differences ($p > 0.05$).

Author Contributions: Conceptualisation, P.N. and D.C.; methodology, P.N., D.C. and M.S.; preparation of references, M.S.; analysis, M.S., P.N. and N.T.; writing—original draft preparation, P.N. and M.S.; writing—review and editing, all authors. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Portuguese Foundation for Science and Technology (FCT/MCTES), through the funded REVIVE project “The threads of the past weaving the future: The colors from the Royal Textile Factory of Covilhã, 1764–1850” (2022.01243; <https://doi.org/10.54499/2022.01243.PTDC> (accessed on 15 August 2023)), and CEEC junior contracts awarded to Paula Nabais (2021.01344.CEECIND) and Natércia Teixeira (2021.01344.CEECIND). This work was also supported through projects UIDB/50006/2020 and UIDP/50006/2020, funded by FCT/MCTES through national funds.

Data Availability Statement: All data can be shared by contacting the authors.

Acknowledgments: The authors would like to acknowledge the Archives Départementales de l'Hérault in Montpellier, France, and the private owners of Paul Gout's manuscript, which allowed access to both recipe books.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Cardon, D. *Natural Dyes: Sources, Tradition, Technology and Science*; Archetype Publications: London, UK, 2007.
2. Veneno, M.; Nabais, P.; Otero, V.; Clemente, A.; Oliveira, M.C.; Melo, M.J. Yellow Lake Pigments from Weld in Art: Investigating the Winsor & Newton 19th Century Archive. *Heritage* **2021**, *4*, 422–436.
3. Marques, R.; Sousa, M.M.; Oliveira, M.C.; Melo, M.J. Characterization of Weld (*Reseda luteola* L.) and spurge flax (*Daphne gnidium* L.) by high-performance liquid chromatography-diode array detection-mass spectrometry in Arraiolos historical textiles. *J. Chromatog. A* **2009**, *1216*, 1395–1402. [[CrossRef](#)] [[PubMed](#)]
4. Moiteiro, C.; Gaspar, H.; Rodrigues, A.I.; Lopes, J.F.; Carnide, V. HPLC quantification of dye flavonoids in *Reseda luteola* L. from Portugal. *J. Sep. Sci.* **2008**, *31*, 3683–3687. [[CrossRef](#)] [[PubMed](#)]
5. Cristea, D.; Bareau, I.; Vilarem, G. Identification and quantitative HPLC analysis of the main flavonoids present in Weld (*Reseda luteola* L.). *Dyes Pigment.* **2003**, *57*, 267–272. [[CrossRef](#)]
6. Ferreira, E. New Approaches towards the Identification of Yellow Dyes in Ancient Textiles. Ph.D. Thesis, University of Edinburgh, Edinburgh, UK, 2002.

7. Melo, M.J.; Nabais, P.; Vieira, M.; Araújo, R.; Otero, V.; Lopes, J.; Martín, L. Between past and future: Advanced studies of ancient colours to safeguard cultural heritage and new sustainable applications. *Dyes Pigment.* **2022**, *208*, 110815. [[CrossRef](#)]
8. Sengupta, P.K.; Kasha, M. Excited State Proton-Transfer Spectroscopy of 3-Hydroxyflavone and Quercetin. *Chem. Phys. Lett.* **1979**, *68*, 382–385. [[CrossRef](#)]
9. Chou, P.T.; Chen, Y.C.; Yu, W.S.; Cheng, Y.M. Spectroscopy and dynamics of excited-state intramolecular proton-transfer reaction in 5-hydroxyflavone. *Chem. Phys. Lett.* **2001**, *25*, 89–97. [[CrossRef](#)]
10. Yang, Y.; Zhao, J.; Li, Y. Theoretical study of the ESIPT process for a new natural product quercetin. *Sci. Rep.* **2016**, *30*, 32152. [[CrossRef](#)] [[PubMed](#)]
11. Jørgensen, L.V.; Cornett, C.; Justesen, U.; Skibsted, L.H.; Dragsted, L.O. Two-electron electrochemical oxidation of quercetin and kaempferol changes only the flavonoid C-ring. *Free Radic. Res.* **1998**, *29*, 339–350. [[CrossRef](#)]
12. Sharif, S.; Nabais, P.; Melo, M.J.; Pina, F.; Oliveira, M.C. Photoreactivity and stability of flavonoid yellows used in cultural heritage. *Dye. Pigment.* **2022**, *199*, 110051. [[CrossRef](#)]
13. Villela, A.; van Vuuren, M.S.; Willemsen, H.M.; Derksen, G.C.; van Beek, T.A. Photo-stability of a flavonoid dye in presence of aluminium ions. *Dye. Pigment.* **2019**, *162*, 222–231. [[CrossRef](#)]
14. Cardon, D. *Des Couleurs Pour Les Lumières—Antoine Janot, Teinturier Occitan, 1700–1778*; CNRS Editions: Paris, France, 2019.
15. Cardon, D.; Brémaud, I. *Le Cahier de Couleurs d'Antoine Janot/Workbook, Antoine Janot's Colours*; CNRS Editions: Paris, France, 2020.
16. Cardon, D. *The Dyer's Handbook: Memoirs of an 18th-Century Master Colourist*; Oxbow Books: Oxford, UK; Philadelphia, PA, USA, 2020.
17. Cardon, D.; Brémaud, I. *Les 157 Couleurs de Paul Gout/Paul Gout's 157 Colours; Vieilles Racines et Jeunes Pousses*: Mercin, France, 2022.
18. Leona, M. Microanalysis of organic pigments and glazes in polychrome works of art by surface-enhanced resonance Raman scattering. *Proc. Natl. Acad. Sci. USA* **2009**, *106*, 14757–14762. [[CrossRef](#)] [[PubMed](#)]
19. Pozzi, F. Development of Innovative Analytical Procedures for the Identification of Organic Colorants of Interest in Art and Archaeology. Ph.D. Dissertation, Università degli Studi di Milano, Milano, France, 2011.
20. Picollo, M.; Aceto, M.; Vitorino, T. UV-Vis spectroscopy. In *Physical Sciences Reviews*; Giamberini, M., Ja-strzab, R., Liou, J.J., Luque, R., Nawab, Y., Saha, B., Tylkowski, B., Xu, C.P., Cerruti, P., Ambrogi, V., et al., Eds.; De Gruyter GmbH: Berlin, Germany, 2018.
21. Nabais, P.; Melo, M.J.; Lopes, J.A.; Vieira, M.; Castro, R.; Romani, A. Organic colorants based on lac dye and brazilwood as markers for a chronology and geography of medieval scriptoria: A chemometrics approach. *Herit. Sci.* **2021**, *9*, 32. [[CrossRef](#)]
22. Degano, I. Liquid chromatography: Current applications in heritage science and recent developments. In *Physical Sciences Reviews*; Giamberini, M., Ja-strzab, R., Liou, J.J., Luque, R., Nawab, Y., Saha, B., Tylkowski, B., Xu, C.P., Cerruti, P., Ambrogi, V., et al., Eds.; De Gruyter GmbH: Berlin, Germany, 2018.
23. Edgar, J.S.; Simpson, W.S. The absorption of aluminum by wool from water/alcohol mixtures. *Text. Res. J.* **1975**, *45*, 281–284. [[CrossRef](#)]
24. Krupinska, I. Aluminium Drinking Water Treatment Residuals and Their Toxic Impact on Human Health. *Molecules* **2020**, *25*, 641. [[CrossRef](#)] [[PubMed](#)]
25. Wouters, J.; Verhecken, A. The coccid insect dyes: HPLC and computerized diode-array analysis of dyed yarns. *Stud. Cons.* **1989**, *34*, 189–200.
26. Golikov, V. The technology of silk dyeing by cochineal. III. The experimental investigation of the influences of pH, water quality, cream of tartar and oak galls. In *Dyes in History and Archaeology 16/17, Proceedings of the 16th Meeting of Dyes in History and Archaeology, Lyons, France, 11–12 December 1997 and 17th Meeting of Dyes in History and Archaeology, Greenwich, UK, 26–27 November 1998*; Archetype Publications: London, UK, 2001.
27. Vijayalaxmi, S.; Jayalakshmi, S.K.; Sreeramulu, K. Polyphenols from different agricultural residues: Extraction, identification and their antioxidant properties. *J. Food Sci. Tech.* **2015**, *52*, 2761–2769. [[CrossRef](#)] [[PubMed](#)]
28. Favaro, G.; Clementi, C.; Romani, A.; Vickackaite, V. Acidichromism and ionochromism of luteolin and apigenin, the main components of the naturally occurring yellow weld: A spectrophotometric and fluorimetric study. *J. Fluoresc.* **2007**, *17*, 707–714. [[CrossRef](#)] [[PubMed](#)]
29. Kirby, J.; van Bommel, M.; Verhecken, A. *Natural Colorants for Dyeing and Lake Pigments*; Archetype Publications: London, UK, 2014.
30. Zhang, X.; Laursen, R. Development of mild extraction methods for the analysis of natural dyes in textiles of historical interest using LC-diode array detector-MS. *Anal. Chem.* **2005**, *77*, 2022–2025. [[CrossRef](#)] [[PubMed](#)]
31. Ichianagi, T.; Oikawa, K.; Tateyama, C.; Konishi, T. Acid mediated hydrolysis of blueberry anthocyanins. *Chem. Pharm. Bull.* **2001**, *49*, 114–117. [[CrossRef](#)] [[PubMed](#)]

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