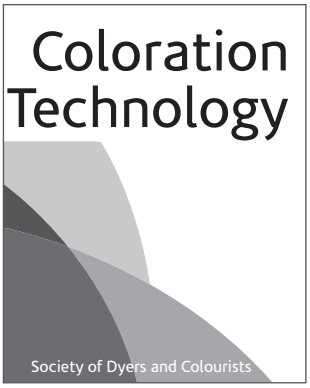
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Comparison of madder (*Rubia tinctorum* L.) and weld (*Reseda luteola* L.) total extracts and their individual dye compounds with regard to their dyeing behaviour, colour, and stability towards light



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This paper reports on the colorimetric properties of natural dyes from madder and weld plants. For both crops, a comparison is made among several individual dye compounds and the total plant extracts. Dyeing properties, colour properties and colour stability have been studied. Insight has been gained into the role of the major dye compounds present in the plant. For madder as well as for weld, the use of the total plant extract is considered to be advantageous over the use of individual dye compounds in terms of solubility, colour tone and stability towards light, and non-coloured components present in the plant extract are considered to be responsible for increasing the solubility and the stability of the dye compounds.

Introduction

In the current ecologically driven revival of natural dyes, there has been significant interest in the cultivation of specific dye crops and also in the production of plant-derived dyes from waste streams. Many dye crops are known and described, with the emphasis on the dyeing properties of the obtained dye. Numerous studies report the colour and fastness of various natural dyes extracted from different plant sources. Reports generally consider the total plant extract, obtained after limited purification [1–7].

For most common natural dyes, the main dyeing compounds have been characterised and described. Knowledge of the main compounds and their degradation products is particularly useful in the field of historical textiles and their preservation [8–12]. Although a focus on the main compounds is a practical approach for analytical purposes, this is too limited for the assessment of dyeing properties. The aim of this research is to gain more insight into the dyeing behaviour of individual dye compounds in comparison with the total plant extract. In doing so, information is acquired on the role of non-colouring plant compounds. This is the first time that such an elaborate comparison has been made with more than two individual dye compounds.

Most plant extracts that are used as dyes contain several compounds, which together are responsible for the final colour. In a natural dye source such as madder root, alizarin and purpurin are believed to account for the red colour. In fact, madder roots contain several dozen different anthraquinones, of which the 30 most prominent [11,13] all contribute to the final colour hue. Furthermore, the presence of non-colouring plant compounds may play an important role in terms of other aspects such as rate of solubility or stability of the colour. The literature provides several examples of non-anthraquinone colorants that may be applicable. The presence of reducing sugars may accelerate degradation of anthocyanins in cabbage juice [14]. A case has been reported in which extract clean-up increased the light stability of a carotenoid [15]. The antioxidant properties of phenolic compounds, present in all plants, are well recognised. In the food industry, it has been reported that the interaction with such compounds can increase colour stability for anthocyanins, known as copigmentation [16]. Applying similar techniques to the extraction of anthraquinonoid glucosides, an extraction method for textile artefacts was reported, in which the presence of glucose diminished degradation of the colouring compounds [17]. Because of the complexity of plant extracts, the influence of the various coloured and non-coloured compounds is not yet fully understood.

One of the challenges for industrial use of dye crops is that their performance, in terms of fastness properties, is considered poorer than that of synthetic dyes. Synthetic dyes are mostly pure, well-characterised compounds, whose behaviour has been thoroughly studied for the past century. In contrast, the scientific expertise on natural dyes is still growing. With regard to natural dyes, there are sparse reports about the influence of the individual dye compounds present in the plant on the overall dyeing behaviour and fastness properties of the plant extract. Sakata and Katayama reported the results of dyeing silk with three individual anthraquinones from madder [18]. Gupta *et al.* [19] used two individual anthraquinones from madder for dyeing nylon. Clementi *et al.* [20] compared spectroscopic and chromatographic properties of wool dyed with alizarin and purpurin with madder-dyed wool. Ford *et al.* [21] reported the sorption behaviour of pure alizarin and a mixture of lucidin primeveroside and ruberythric acid on

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wool. It is our belief that detailed comprehension of the colorimetric behaviour of individual dye compounds leads to more insight into the behaviour of the total plant extract. At the same time, the role of non-coloured compounds in the total plant extract can become clearer. This opens up new possibilities for improving the colour, stability and colour fastness of textiles dyed with plant extracts. The aim of the research on two major natural dyes, madder (*Rubia tinctorum* L.) and weld (*Reseda luteola* L.), presented in this paper, is to improve understanding of the influence of individual dye compounds on dyeing properties of the total plant extract.

Experimental

Materials

The following substances were commercially available: alizarin (**ali**, 97%), purpurin (**pur**, 60%) (Sigma, USA), luteolin (**lut**, 98%) (Extrasynthese, France), aluminum sulphate (Alfa Aesar, USA), tartaric acid (Merck KGaA, USA), formic acid and regular solvents. Commercial dyes were produced by Rubia 100% Natural Colours (The Netherlands). Rubia Yellow (RY) is a dried alcohol/water extract from weld. Rubia Red (RR) and Rubia Red A (RRA) are dried alcohol/water extracts from madder root.

The major constituents from weld and madder were isolated from plant material. Luteolin-7,3′-*O*-diglucoside (**ldg**) and luteolin-7-*O*-monoglucoside (**lmg**), isolated from weld, were kindly provided by E. J. C. van der Klift. Lucidin primeveroside (**lp**) and ruberythric acid (**ra**) were extracted from RR in the following manner: 150 mg of RR was dissolved in 20 ml of a mixture of ethanol and water (20/80 v/v). After centrifugation, the clear supernatant was absorbed on a solid-phase extraction (SPE) column (1000 mg, 6 ml, GracePure C18-Max, 60 Å and 50 μm) and eluted with a gradient (20–60%) of ethanol in water. Fractions containing **lp** and **ra** were dried with a rotary evaporator followed by lyophilisation, yielding a yellow solid. This procedure was repeated several times. The solid was dissolved and **lp** and **ra** were separated on a preparative high performance liquid chromatography (HPLC) system (Shimadzu, Japan) equipped with a 22 × 250 mm Alltima C18 column (5 μm; Alltech, USA), with ultraviolet (UV) detection at 260 nm. Multiple runs were performed isocratically (20 ml/min) with water: acetonitrile (73:27). The yield was *ca*. 60 mg of each pure compound. Pseudopurpurin (**pp**) and munjistin (**mun**) were obtained from RRA in the following manner: 0.65 g of the dye was dissolved in 100 ml of water followed by liquid-liquid extraction (LLE) with 3 × 25 ml of ethyl acetate to remove water-insoluble components. To the aqueous fraction, sodium chloride was added to a concentration of *ca*. 100 g/l. This was followed by a second LLE step with ethyl acetate. The organic layer was collected and back-extracted with 50 ml of aqueous sodium hydroxide of pH 9. The aqueous layer thus obtained contained both **pp** and **mun**. The remaining sodium chloride was removed by transferring the aqueous layer to a beaker and mixing it with 0.5–1 g of C18 resin material, which absorbed only the dye compounds. Afterwards, the resin material was eluted with ethanol/water (70/30 v/v). This procedure was repeated several times. Next, **pp** and **mun** were separated by column chromatography on a Sephadex LH20 column (8 cm height × 1.2 cm internal diameter (i.d.)) using water as the eluent. The yield was *ca*. 30 mg of each pure compound. The identities of purified compounds were confirmed using proton nuclear magnetic resonance (^{1}H NMR) and HPLC mass spectrometry (MS) measurements.

The absolute purity of the compounds was determined by means of quantitative ^{1}H NMR measurements using maleic acid (99.8%) as internal standard [22]. The compounds possessed the following purities: **ldg** 86%, **lmg** 90%, **lp** 98%, **ra** 86%, **pp** 68%, and **mun** 68%. Impurities were not fully characterised but were invisible with UV detection between 250 and 300 nm, where the target compounds are detected, so are considered residual water or inorganic impurities.

Dyeing procedure

Because of the small amounts of dye compound available, commercial dyeing apparatus could not be used. Instead, dyeing was performed in a laboratory reflux set-up in a 250 ml round-bottomed flask, allowing visual inspection of the dyeing process. Five g of ready-to-dye wool was soaked in water at room temperature for 10 min and centrifuged. Wool was premordanted in a solution of 0.13 g tartaric acid and 1.0 g aluminium sulphate in 100 ml of water at 95 °C for 1 h. Afterwards, the wool was rinsed twice in water and centrifuged. A dyebath was prepared by adding 450 mg of commercial dye or an amount of each isolated component to 100 ml water. The following amounts of component were individually used: 9.5 mg of **ldg**, 6.8 mg of **lmg**, 4.1 mg of **lut**, 34.6 mg of **lp**, 32.7 mg of **ra**, 10.6 mg of **pp**, 8.0 mg of **mun**, 15.4 mg of **ali**, or 21.2 mg of **pur**. Wool was added to the dyebath, which was then heated and kept at 100 °C for 1 h. Next, the wool was rinsed twice in boiling water. Samples from the dyeing and rinsing baths were combined, filtered and analysed by HPLC, according to methods described earlier by Derksen *et al*. [23] and Villela *et al*. [24]. Calibration curves were used to quantify the amount of each dye present.

Exposure to UV light

Light fastness tests were performed in a QUV Accelerated Weathering Tester (Q-lab, USA) according to the ISO 105-B02 method. Samples were also subjected to prolonged exposure (40 and 60 h for weld and madder, respectively) in order to measure CIE $L^*a^*b^*$ values over time. Furthermore, the amount of dye in the wool was determined upon prolonged exposure to light. To this end, dyed threads of wool were wrapped into an oblong bundle in which each turn of the thread was ordered in a parallel fashion. They were placed on top of a piece of cardboard (5 × 20 cm) and, to avoid moving the threads, each bundle was secured in the middle with a paperclip. The cardboard pieces were placed in the tester. The exposure times were 20 h for **ldg**, **lmg** and **lut**, 30 h for **mun**, 60 h for **pp** and **pur**, 80 h for **lp** and **ra**, and 100 h for **ali**. Threads were extracted before and after exposure.

Extraction method

The extraction method was modified from Zhang *et al.* [25] and Villela *et al.* [26]. From the dyed wool, 0.1 g of threads were taken and placed in 3 ml of extraction liquid, consisting of methanol: water: formic acid in a 80:15:5 v/v/v ratio. An exception was made for wool dyed with munjistin or pseudopurpurin; for these two compounds, the

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extraction liquid consisted of tetrahydrofuran: methanol: water in a 50:25:25 v/v/v ratio. The extraction liquid was kept at 60 °C for 30 min. After 10 s of sonification to ensure maximum solubilisation, aiding extraction, the wool was removed from the liquid, after which the extract was analysed by HPLC.

Colorimetric measurement

A Color-Eye 7000 spectrophotometer (X-rite, former Macbeth, USA) equipped with Lite Quality Control v.1.9 software (Optiview, USA) was used to measure CIE *L*a*b** values from dyed wool before, during and after exposure to light. For each sample, a triple measurement (observer 2°, illuminant D65, DREILL status) in reflectance mode was averaged. The spectrophotometer was calibrated with a white and black standard. CIE *L*a*b** values were determined at fixed intervals. These were 0, 2, 5, 10, 15, 20, 25, 30, 35 and 40 h for extracts and compounds from weld, and 0, 2, 5, 10, 20, 30, 35, 40, 45, 50, 55 and 60 h for extracts and compounds from madder.

Results and Discussion

A comparative study was made between plant extracts from weld or madder and individual dye compounds, respectively flavones and anthraquinones, which were identified in the plant. Extract RY is a powder-dried alcohol/water extract from weld which contains soluble plant compounds including sugars and several flavones, which provide the yellow-greenish colour. The isolated compounds used for dyeing were the three major flavones present in weld: two glucosides, namely luteolin-7,3′-O-diglucoside (**ldg**) and luteolin-7-O-monoglucoside (**lmg**), and one aglycone, namely luteolin (**lut**). From madder, two extracts were used: RR and RRA. Both are powder-dried alcohol/water extracts from madder root containing various soluble plant compounds, such as carbohydrates, and a range of anthraquinones, which give the characteristic red colour. RR contains mostly glycosidic anthraquinones in the form of primeverosides. RRA has a higher content of aglyconic anthraquinones with free carboxylic acid and hydroxyl groups. During the production process of RRA, the primeverosides from madder root are hydrolysed to their corresponding aglycones using endogenous enzymes, as described in the literature [27]. The individual compounds used for dyeing were six major anthraquinones present in madder: two primeverosides, namely lucidin primeveroside (**lp**) and ruberythric acid (**ra**), two aglycones with free carboxylic acid groups, namely pseudopurpurin (**pp**) and munjistin (**mun**), and two aglycones with only hydroxyl groups, namely alizarin (**ali**) and purpurin (**pur**). All of the dye compounds are shown in Figure 1.

Dyeing behaviour of extracts *vs* individual dye compounds

Wool was pretreated by a mordanting step using aluminium sulphate and tartaric acid and was subsequently dyed with an extract or an individual dye compound. After dyeing, residues of dye compound were washed off from the wool in two rinsing baths and analysed. These residual amounts are included in the amounts labelled 'after dyeing' in figures 2 and 3. In Figure 2, the amount (in mmoles/bath) of individual dye compound present in each dyebath before and after dyeing is given. The amount of dye in each dyebath from the total extract before and after dyeing is presented in Figure 3. An experimental challenge was the limited solubility of the dye compounds in water. To make sure the dye uptake was studied without the interference of a solubility effect, the experiment was set up in such a way that the initial molar amount for each individual dye compound was of the same order of magnitude as the sum of dyeing compounds in the corresponding extract. For ease of comparison, this sum is also presented in Figure 3. The rate of solubility at room temperature was higher for the plant extracts than for the individual dye compounds. The latter only dissolved after the dyebath was heated. Evidently the extracts show a higher rate of solubility due to the presence of other plant compounds such as carbohydrates and natural surfactants. As can be seen in Figure 3, the amounts of individual dyeing compounds within each extract vary. This is due to the natural variation in the plant: some dyeing compounds are naturally more abundant than others. In the case of RRA, the natural composition was altered by incorporating a hydrolysis step in the production process.

From figures 2 and 3, it is clear that there is a large variation in dye uptake by the wool among the various compounds. Looking at weld compounds, the glucoside **ldg** has poor dye uptake, probably caused by a diminished bonding ability with the mordant. Both the catechol moiety

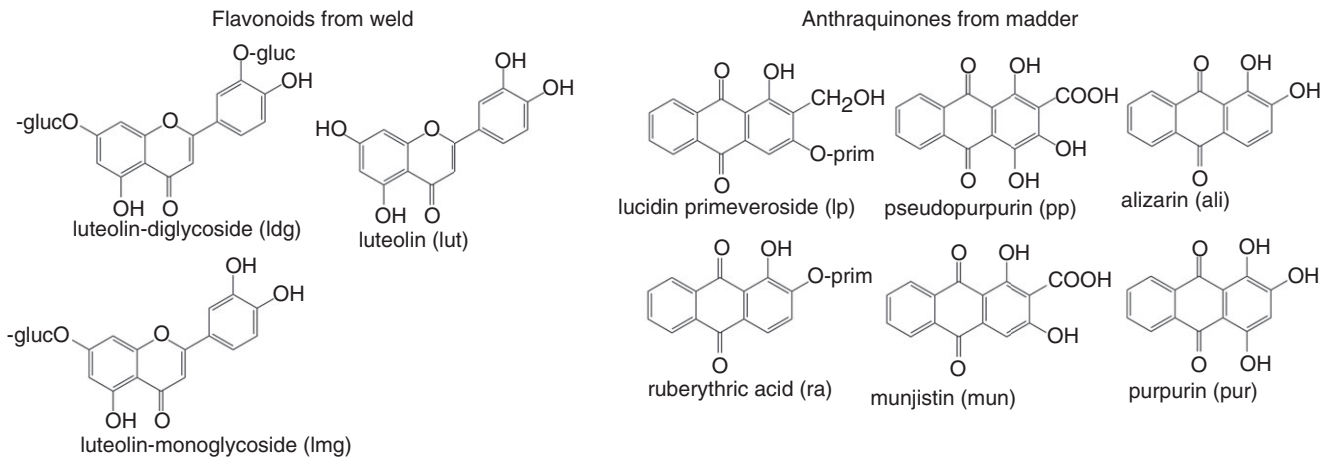


Figure 1 Structures of dye compounds from weld (**ldg**, **lmg**, **lut**) and madder (**lp**, **ra**, **pp**, **mun**, **ali**, **pur**); gluc, glucose; prim, primeverose

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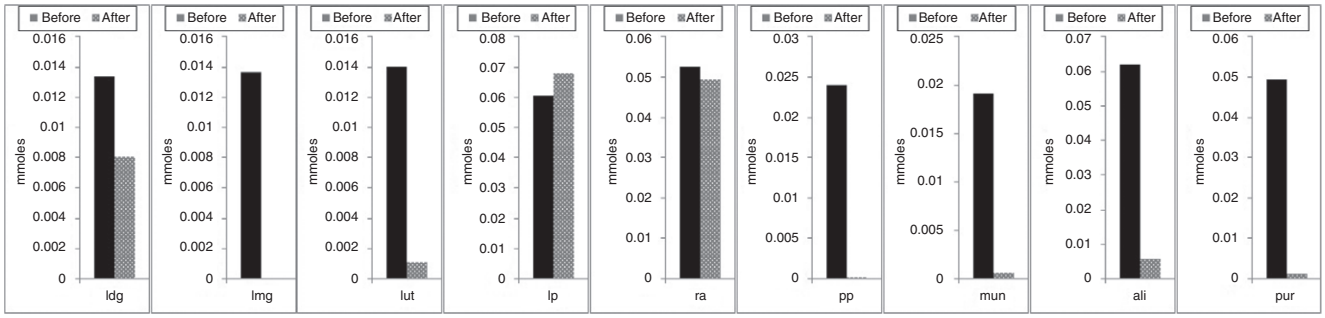


Figure 2 Amount of individual dye compounds (in mole/bath) in each individual dyeing bath before and after dyeing wool for compounds from weld ((**ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin) and from madder (**lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin); solid bars, before dyeing; dotted bars, after dyeing

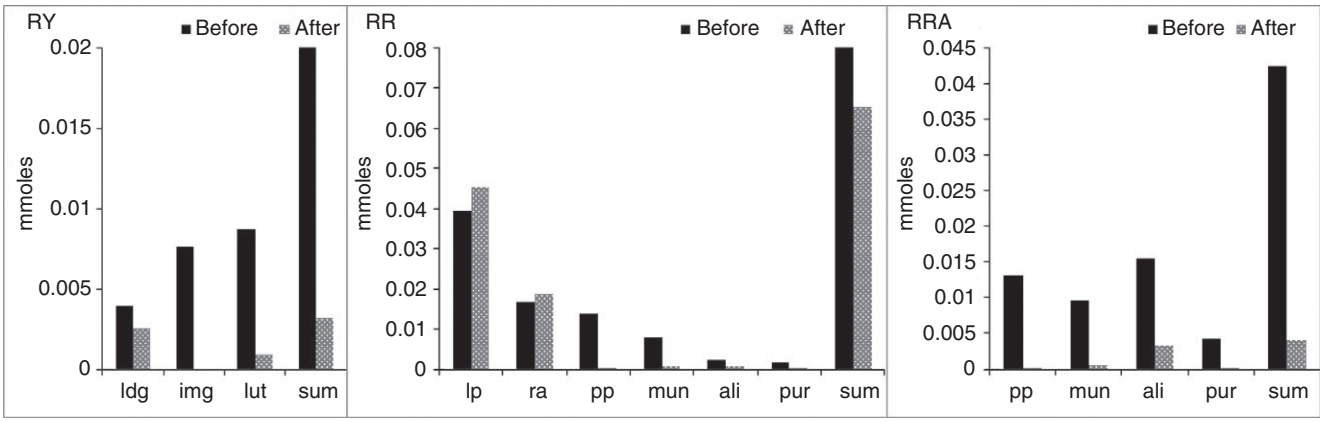


Figure 3 Amount of dye compounds (in mole/bath) in dyebaths before and after dyeing wool for the plant extracts (left) RY from weld, and (middle) RR and (right) RRA from madder; **ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin; **sum**, sum of all compounds; solid bars, before dyeing; dotted bars, after dyeing

and the vicinal carbonyl-hydroxyl moiety in flavones are held responsible for binding to the aluminium mordant and textile [28]. In the case of glucoside **ldg**, one of two vicinal hydroxyl groups is attached to glucose and no longer available for bonding. This is confirmed by glucoside **lmg**, which shows good binding in the extract as well as in pure substance. This molecule still possesses vicinal hydroxyl groups at the 3′- and 4′- positions, because only the hydroxyl group at the 7′- position is glucosylated. The aglycone **lut** is almost completely taken up from the dyebath and fixed onto the wool. A further reason for faster uptake of the aglycone is its more hydrophobic character, preferring to bind to hydrophobic domains in the wool.

Looking at madder compounds, one can see a similarly poor dye uptake of the primeverosides **lp** and **ra**. The seemingly small increase in dye per bath is believed to be an analytical inaccuracy, caused by the challenges of working with such small amounts and the possible loss of dyebath volume. Unfortunately, scarcity of the pure compounds prohibited duplicate measurements. In anthraquinones, the catechol moiety and vicinal carbonyl-hydroxyl moiety are also believed to play an important role in binding ability [29], which is disrupted by the presence of sugar molecules. The aglycones **pp**, **mun**, **ali** and **pur** are almost completely taken up from the dyebath and fixed onto the wool. It is noteworthy that the relative dye uptake for **pp**, **mun**, and **pur** appears as good as for **ali**, which is historically considered the main dye compound. The poor uptake for the glycosides does not match results from Ford *et al.* [21], wherein it was observed that ruberythric acid has preferential sorption properties on wool over the aglycone alizarin. Differences in experimental set-up (dyeing time, concentration, liquor-fibre ratio) may be a reason for this. When comparing Figure 2 with Figure 3, there is very little difference in the dye uptake of individual dye compounds whether they are in a separate bath or part of an extract. So the influence of the non-coloured constituents in the extract on the dye-uptake behaviour of individual compounds is limited.

Colour of extracts *vs* individual dye compounds

For all dyed wool specimens, CIE $L^*a^*b^*$ values were measured (Figure 4). The L^* values for the extracts are lower than for individual compounds. The use of purified dye compounds appears to improve the lightness. For weld, a^*b^* values are quite similar for the extracts and individual dye compounds. Also, to the naked eye the colours are similar. Only the wool dyed with **ldg** has a different colour, as shown most clearly in the b^* value, caused by the low dye uptake. For madder, there is a larger variation in a^*b^* values between the different anthraquinones, as previously demonstrated by Sakata and Katayama [18]. Again, the values for both of the glucosides **lp** and **ra** are influenced by the small amount of dye actually taken up by the fabric. Wool dyed with **mun** is yellow-orange to the naked eye, which is confirmed by the a^*b^* values. The other aglycones in madder have red (**ali**), reddish purple (**pur**) and pink-red (**pp**) colours, in agreement with Schweppe [30]. The colour

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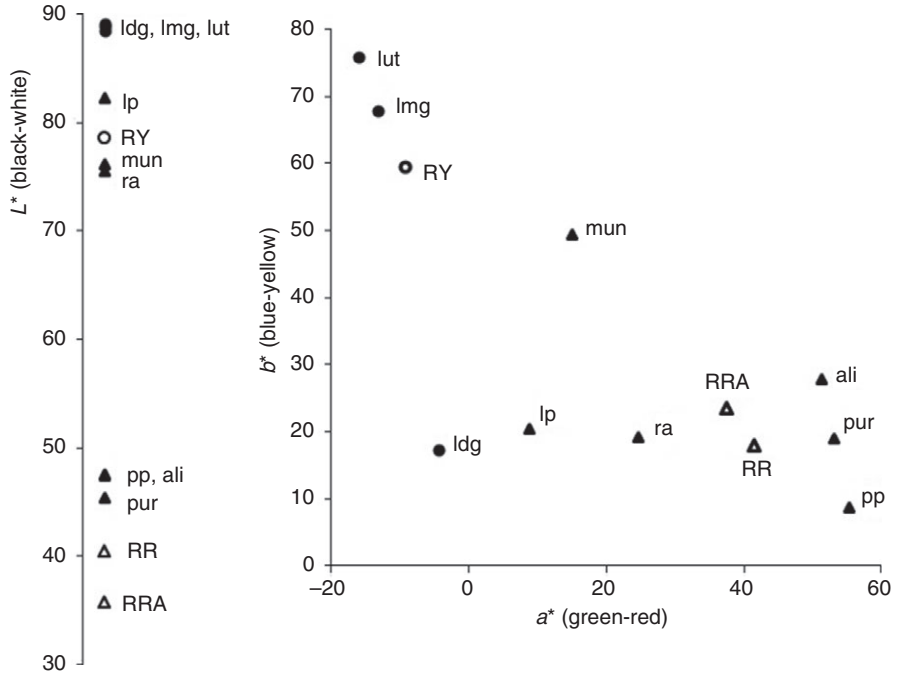


Figure 4 (left) Brightness L^* and (right) colour hue a^*b^* for wool dyed with plant extracts RY from weld, and RR and RRA from madder and with individual dye compounds; open markers are extracts, solid markers are individual compounds, dots are for weld, triangles are for madder; **ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin

of the extracts RR and RRA is positioned between the various colours of the individual dye compounds. The redness of the colour is caused by the predominance of dyes as **ali**, **pur** and **pp**. Figure 4 gives an impression of the varied colour tone of natural dyes.

Stability towards light for extracts *vs* individual dye compounds

For all samples, the light fastness was determined by exposure to UV-A light and by comparing the change in colour to a blue wool-standard [31]. The results are summarised in Table 1. For weld, light fastness is very poor for the extract as well as for the individual dye compounds. The poor light fastness of natural yellow dyes is generally known in the field. However, a value of < 1 appears to be low in comparison with values in the literature (1–6 under various dyeing conditions [3,32,33]), so samples were tested by two independent labs (Textile Lab, The Netherlands and TO_2C, Belgium). This resulted in a similar light fastness value of 1. For madder, the highest light fastness value is given for **ali**. It can be seen that the extracts also have quite high fastness values relative to the individual dye compounds: 4 and 4/5 *vs* values ranging from < 1 (**mun**) to 5 (**ali**).

Table 1 Light fastness values for wool dyed with extracts from madder and weld and with individual dye compounds

Source	Extract/compound	Light fastness
Weld	RY	< 1
	ldg	< 1
	lmg	< 1
	lut	< 1
Madder	RR	4
	RRA	4/5
	lp	4
	ra	4
	pp	3
	mun	< 1
	ali	5
	pur	3/4

ldg, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin.

Extraction from wool for individual dye compounds

The stability of the colour on wool was studied further by investigating the quantitative decrease of the individual dye compounds on the wool. For this purpose, dyed wool threads were exposed to UV light for a prolonged period of time. From samples of non-exposed and exposed threads, individual dye compounds were extracted in a heated mixture of MeOH/H_2O/HCOOH (80:15:5 v/v/v). As an exception, wool dyed with **mun** or **pp** was extracted in THF/MeOH/H_2O (50/25/25 v/v/v). The amount of each dye on wool was determined before and after exposure to UV light. This was done for the individual dye compounds from both plants. The results are displayed in Figure 5, together with the initial amount on wool (which equals the difference in the dyebath before and after dyeing from Figure 3), to give an idea of the extraction efficiency. No additional peaks appeared, indicating the absence of degradation.

Of the three compounds from weld, only **lut** was completely extracted from dyed wool. For **ldg,** the dye uptake as well as extraction are rather poor, which compromises the indicative value of the extraction efficiency. Both **lmg** and **lut** show a considerable decrease in extracted dye amount before and after exposure to light. The light causes photodegradation of the dye compounds on the wool, which explains the observed decrease. Looking

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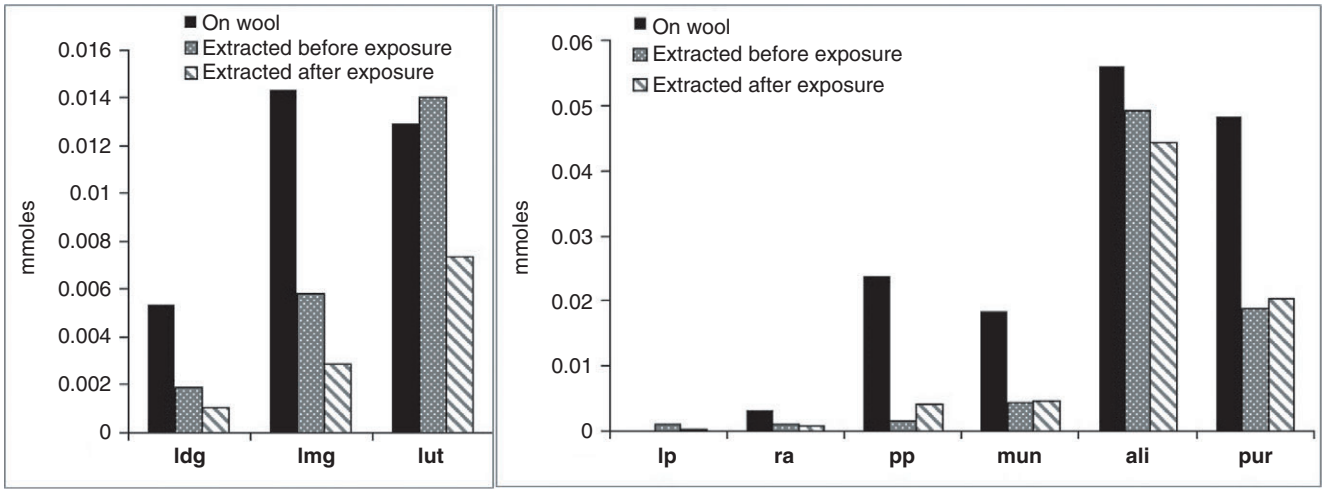


Figure 5 Amount of dye compounds (in mole/cloth) on wool and extracted from wool before and after exposure to light (left) for weld and (right) madder. Exposure times were 20 h for **ldg**, **lmg** and **lut**, 30 h for **mun**, 60 h for **pp** and **pur**, 80 h for **lp** and **ra**, and 100 h for **ali**; **ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin

at madder, the extracted amounts of the glucosides **lp** and **ra** are not particularly meaningful since their dye uptake was very low to start with, and these will not be discussed further. For aglycones **pp** and **mun**, having a carboxylic acid group, several solvent mixtures were tried for extraction, but all showed a poor extractability from the wool. Evidently the presence of a carboxylic acid group causes a stronger binding to the wool, possibly due to electrostatic interactions between the deprotonated carboxylic acid moiety and protonated amine moieties of the protein. The aglycones **ali** and **pur** are highly extractable from wool. The amount of extracted dye is approximately the same before and after exposure to light. This indicates little photodegradation of these dye compounds on wool. Comparing the aglycone **lut** (from weld) with **ali** and **pur** (from madder), it is clear that under the influence of light the amount of **lut** decreases the most and in a shorter period of time. This correlates with the lower light fastness value for **lut** in comparison with those for **ali** and **pur**.

Colour change for extracts *vs* individual dye compounds

In addition to light fastness measurements, $L^*a^*b^*$ values were measured at various time intervals during exposure to UV light. This was done for a prolonged period after the initial colour change was visible to the eye. The alteration in L^* and a^*b^* values is shown in Figure 6. The colour difference during exposure to light is further illustrated in the ΔE value, defined as $\sqrt{((\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2)}$. These ΔE values are shown in Figure 7.

The development of $L^*a^*b^*$ values upon exposure to UV light shows a similar pattern for the individual dye compounds and the extract from weld. The brightness (L^*) shows a large change in the first 2 h, and thereafter a gradual decrease within the measured timeframe of 40 h. A bigger change is observed for the colour hue. The a^*b^* values, and therefore also the ΔE values, keep on changing under the influence of prolonged exposure. The largest increase in ΔE is for the individual dye compounds in weld and to a lesser extent for the extract, indicating a better stability to light for the extract.

For madder, looking at the colour hue, the change in a^*b^* values for the extracts is considerably smaller than that for the individual dye compounds. This can partly be ascribed to a summation effect, since the b^* value changes to more yellow for three dye compounds (**lp**, **ra**, **pur)**, is nearly constant for **pp**, and changes to more blue for **mun** and **ali**. However, for the a^* value this is less credible. Only one dye compound (**mun**) changes to more red, while the five other dye compounds change to more green. There could also be an influence of non-dye components in the extracts, such as sugars or polyphenols, which are naturally present in madder. Unidentified compounds could act as UV-stabilisers. The change in L^* is small for both madder extracts and the individual dye compounds. The largest change seems to occur for the yellow/orange compounds **lp**, **ra** and **mun** in madder. It should be noted that for **lp** and **ra** the degree of change is not particularly relevant because of their low dye uptake. Overall, the total colour change ΔE for the madder extracts is smaller than for the individual dye compounds, similar to weld. For the weld extract as well as for the madder extract, the general pattern is comparable with the fading rate curve for this plant presented by Yoshizumi and Cox Crews [32]. Both plants exhibit a type II fading rate curve [34], in which the initial change for weld is larger than for madder.

Conclusion

This study presents for the first time a comparison of dyeing behaviour between total plant extract and several of their individual dye compounds from two plants. For both the madder and weld plant, the use of a total plant extract as textile dye appears to be advantageous over the use of individual dye compounds isolated from the plant. Dye compounds show a similar dye uptake by wool when applied as individual compounds or when present in a total plant extract. So the influence of non-coloured plant compounds on the uptake is small, but the increased rate of solubility of the compounds in the extract must be considered an advantage. For extracts as well as for pure compounds, it was found that aglycones demonstrate

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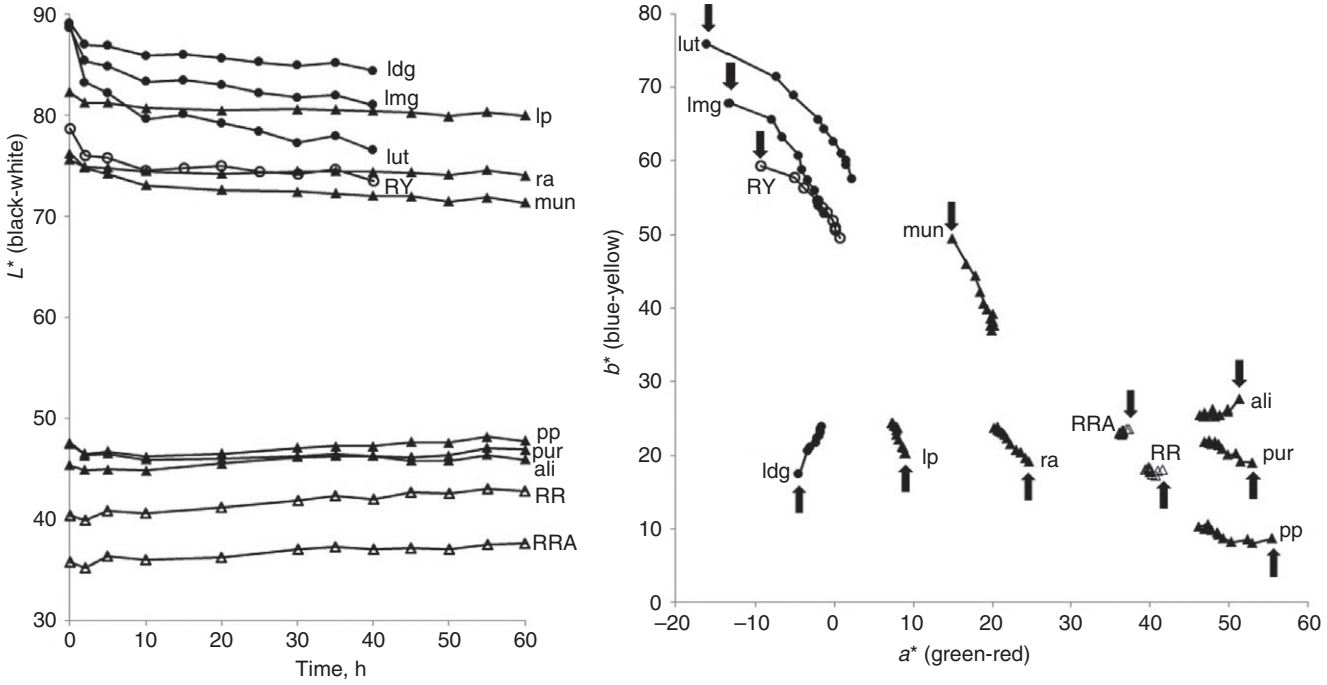


Figure 6 Variation in (left) brightness L^* and (right) colour hue a^*b^* for wool dyed with plant extracts RY from weld, and RR and RRA from madder and with individual dye compounds during exposure to UV light; arrows indicate t = 0; open markers are extracts, solid markers are individual compounds; dots are for weld, which was exposed for 40 h; triangles are for madder, which was exposed for 60 h; **ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin

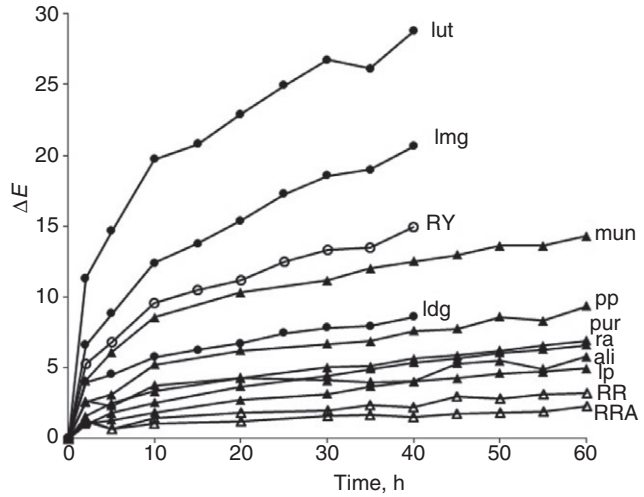


Figure 7 ΔE values for wool dyed with plant extracts RY from weld, and RR and RRA from madder and with individual dye compounds during exposure to UV light; open markers are extracts, solid markers are individual compounds; dots are for weld, triangles are for madder; **ldg**, luteolin diglycoside; **lmg**, luteolin monoglycoside; **lut**, luteolin; **lp**, lucidin primeveroside; **ra**, ruberythric acid; **pp**, pseudopurpurin; **mun**, munjistin; **ali**, alizarin; **pur**, purpurin

higher dye uptake than glycosides. The individual dye compounds exhibit a considerable variation in $L^*a^*b^*$ values, especially in the case of madder. The colour of the extracts is in between the various colours of the individual dye compounds. The presence of different compounds in one plant extract can be considered an incorporated enrichment for the colour tone. For weld, the light fastness is poor for both the extract and the individual dye compounds. The poor light fastness of flavonoids is widely known. For madder, the light fastness of the extracts is high when compared with the individual dye compounds; only that of **ali** is higher. Comparing the behaviour of individual dye compounds in wool is difficult because of differences in dye uptake and extraction recovery. It is clear, however, that the amount of **lut** in wool after exposure to light decreases more than the amount of **ali** and **pur**, and in a shorter period of time. This difference in lifetime between dye compounds from two sources corresponds with the light fastness of the plant extracts (poor for weld, good for madder). Finally, for both plants, the ΔE value upon exposure to light is smaller for extracts than for individual dye compounds, indicating a better colour stability to light for the total extract.

Overall, substantial insight was gained regarding the individual contribution of individual dye compounds on the dyeing properties of the total plant extract. The use of a total plant extract is considered advantageous over the use of individual dye compounds in terms of rate of solubility, colour tone and stability towards light. Obviously, production costs are also more favourable in the case of extracts. The results raise an interesting question about which non-coloured components in the extract are responsible for increasing the rate of solubility and stability of the dye compounds. Further research should focus on these stabilising components in order to increase the performance quality of weld extract in particular, and natural dyes in general.

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