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Review article

Chromatic and medicinal properties of six natural textile dyes: A review of eucalyptus, weld, madder, annatto, indigo and woad

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ABSTRACT

The environmental and health-related impacts of synthetic dyes have led to growing interest in natural dyes as sustainable and eco-friendly alternatives. However, natural dyes have been used to dye textiles with limited color shade and poor fastness properties, and little research has been conducted in this field. Additionally, natural dyes also have the potential to provide added functionalities to textiles, such as antibacterial and anti-UV properties. A systematic literature review of 38 studies was conducted to analyze the use of six natural dyes derived from eucalyptus (*Eucalyptus globulus Labill.*), weld (*Reseda luteola* L.), madder (*Rubia tinctorum* L.), annatto (*Bixa Orellana* L.), true indigo (*Indigofera tinctoria* L.) and woad (*Isatis tinctoria* L.). These dyes were selected after a preliminary analysis of studies on plant-based natural dyes with primary colors, considering their chromatic and potential medicinal properties. This study explores the influence of different dyeing parameters and auxiliary products in these properties. The research discussed how the chromatic and medicinal properties of natural dyes can be affected by various factors and provides a summary table with the chromatic palette possibilities according to the different materials and processes relationships. Exploring the combination of natural dyes with environmentally friendly auxiliary products can be a promising development area for creating a wide range of color shades. Further research is also needed to optimize the dyeing processes with natural dyes, towards more sustainable textile dyeing possibilities.

1. Introduction

Pollution from the textile industry is a severe global problem [1]. It is responsible for the release of toxic chemicals into the environment, which affects ecosystem preservation and has harmful effects on human health [2].

Synthetic materials and toxic products remain prevalent in the textile industry because of their convenience, efficiency and durability, particularly in dyeing processes with synthetic dyes [3]. Heavy metals and chemical auxiliaries, such as acids, promoters, alkalis, oils, and softeners are sometimes utilized in dyeing processes to ensure good color strength (K/S) and color fastness properties [4].

Color strength is a parameter that indicates the amount of light absorbed by a dyed material compared to an untreated or undyed material. By using a straight-line relationship between the reflectance and concentration of dye on the fiber, K/S can be calculated. Kubelka-Munk equation (a) relates a mathematical function of reflectance to the absorption and scattering values, where R is the percentage of reflectance of a light source with a known wavelength on an opaque layer of material, K is the absorption coefficient, and

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S is the scattering coefficient [5]. A higher K/S value indicates that more dye has been absorbed by the fabric, resulting in a more intense color [6].

$$\frac{K}{S} = \frac{(1-R)^2}{2R} \tag{1}$$

Color fastness is the ability of a material to retain its color over time and/or to resist the transfer of its colorants to adjacent materials. Textile color can be affected by various factors such as lighting, washing, rubbing and perspiration, which can alter quality and appearance [7]. Color fastness properties can be assessed qualitatively and quantitatively, through direct observations and color measurement. Color change is comparatively analyzed with samples previous to and after exposure to a specific condition. Color transfer is expressed as staining and it is analyzed through color change of adjacent fabrics [8,9].

There are different types of measurement standards that use scales to evaluate specifics conditions for different purposes and applications. The most commonly referred organizations that develop standards in textile fastness tests are: International Standards Organization (ISO); American Association of Textile Chemists and Colorists (AATCC); and European Committee for Standardization (CEN). These organizations developed standards like ISO 105, AATCC 16 or EN ISO 105, which use different scales, such as: the gray scale, which contains a rating from 1 to 5, where 1 represents 'very poor' and 5 'excellent' [5]; the blue scale, used to assess light-fastness, rated between 1 and 8, 1 being 'very poor' and 7 and 8 being 'excellent'; and the CIE Lab color system, which is based on calculating the reflectance values of the test sample and the standard sample at different wavelengths using a spectrophotometer. The color difference in the CIE Lab system is calculated by the formula:

$$\Delta E^*_{ab} = \left[(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2\right]^{1/2}$$

The Δ symbol means "difference in", L^* value indicates lightness (vertical axis on the color space diagram with values from 0 to 100, black to white), a^* value indicates red-green component of a color (+a* being red and -a* being green) and b* value indicates yellow-blue axis (+b* being yellow and -b* being blue) [5,10].

In addition to products used to improve K/S values and fastness properties, after being dyed, many textiles are subjected to other finishings, such as waterproofing, abrasion resistance and microbial protection [4], for which chemical products are also used. The absorption and accumulation of synthetic dyes and certain auxiliary products in the human organism may result in chronic health complications [11], such as dermatitis, pulmonary disorders and carcinogenesis [12,13].

Exposure to these products through clothing begins in the early hours of life and continues throughout life, thereby increasing the risk of health problems. The skin is the main organ affected by such exposure [14]. Most of these chemical products are absorbed by the human body through the skin, especially in body regions where there is greater sweating and friction, which has detrimental dermatological effects [11,12].

However, with the increasing demand for sustainable practices and the search for greater comfort and well-being, the textile industry has been forced to shift to more sustainable dyeing practices [15]. As a result, the use of natural products, such as plant extracts, has become increasingly sought after in textile dyeing for ecological purposes [3,16], being also considered a safer alternative to human health, comparatively to synthetic dyes [17,18]. Plant-based natural dyes have been used for centuries for dyeing, offering a variety of health advantages and novel qualities [19]. In the same way that the skin absorbs harmful substances, it also absorbs substances of natural origin with beneficial health properties [20].

Two of the most important properties to clothing with regard to well-being described in the literature are antimicrobial activity and ultraviolet (UV) protection [21]. An antimicrobial finishing is important for controlling microbial infestation and reducing odor. The properties of the fiber itself, the structure of the substrate, the chemical processes used, and the environment to which the textile is exposed, such as temperature and humidity, favor the growth of fungi and bacteria [3]. Proximity to the human body also offers an optimal medium for adherence, transfer and propagation of infections caused by microbial species [19]. For this reason, unpleasant odors, stains, discoloration, loss of properties, dermal infections and other related diseases are expected consequences of microbial infestation and can be avoided by antimicrobial finishing [22].

According to the literature, protection from UV radiation is also essential for maintaining well-being. UV rays are harmful to humans and can cause skin damage [18]. There are chemicals in the industry that are supposed to improve this protection, such as phenyl salicylates, benzophenones and acids, but they are not environmentally friendly [3]. Some studies report the presence of UV protection in some natural dyes, such as madder and indigo [18,23,24].

Aesthetic qualities are also important. Color is one of the main characteristics in the textile industry, which emphasizes the importance of colorants and dyeing processes [25]. In addition to the presence of color in nature, it is also introduced into clothing, furniture, cosmetics and other objects through substances known as dyes and pigments. The main difference between the two types of colorants is the solubility; pigments are insoluble chemical compounds, which are chemically and physically unaffected by the substrate on which they are applied; dyes are mostly soluble in aqueous solutions (or undergo processes that voluntarily destroy any crystalline structure) and are affected by the substrate on which they are applied [8]. In this sense, different types of dyes are used, according to the textile fiber to be dyed.

Since the beginning of humanity, animals and plants have been sought after for the development of dyes and pigments [26] capable of coloring diverse materials [27]. However, natural textile colorants have some drawbacks that hinder their widespread application. These include high costs, low variability in the quality and quantity of raw materials, complex and time-consuming extraction and purification methods, poor fastness properties, and absence of standardized procedures and formulations for color production, like dyeing conditions and mordanting agents, which restrict their use in high-end value-added products [28]. Exploiting the full potential of natural dyes is essential for generating a wide range of natural shades. Therefore, it is crucial to explore the potential of natural dyes, particularly those that can act as primary colors in the formulation of secondary shades [24].

Primary colors are colors that can be combined to produce other colors but cannot be obtained by mixing any other colors [8,29]. There are two types of color mixing: additive (light colors) and subtractive (pigment colors). Additive mixing is created by light sources, where the primary colors are red, green, and blue. The union of these three colors produces white light. In subtractive mixing, the primary colors are magenta, cyan and yellow [10,29]. However, it is difficult to find magenta and cyan in nature because of the limited availability of natural sources. This led color theorists to use the primary colors of yellow, red and blue for subtractive synthesis before advent of synthetic dyes [30]. In the subtractive process, the mixture of pigments produces less luminous colors, so that the combination of primary colors becomes darker [8,10,29].

In the textile industry, mixing primary colors of conventional colorants to create various shades is standard procedure. Using primary colors of natural dyes provides an opportunity to obtain a broad spectrum of shades and expand the color palette of textiles dyed with natural colorants [24]. However, increasing the dyeability of natural dyes remains challenging [31]. The dyeability of natural dyes depends on various factors, such as the chemical structure and molecular weight of the dye molecules, the pH and temperature of the dyeing solution, the type and concentration of mordants and the type and structure of the textile fiber [32]. Substantivity describes the degree of attraction or affinity between a dye and a fiber. Most natural dyes exhibit low substantivity because they lack active groups that can form chemical bonds with textile fibers, leading to poor fastness properties of dyed textiles [28]. To improve these properties, auxiliaries are used to bind the dye to the fiber, called mordants, many of which are toxic [33].

The use of these toxic substances contributes to non-ecological technology because the concentration of metal-based mordants is sometimes relatively high, posing health and environmental risks [34]. Mordants, as well as the dyeing process, considerably influence colorimetric parameters such as K/S and color fastness [35]. However, vat dyes are a class of dyes that do not require mordants to obtain good color properties [36]. These dyes, such as true indigo and woad, are insoluble in water but become soluble when they are reduced by a chemical agent to form a soluble compound. This compound, also known as a *leuco* dye, can impregnate the fiber and revert to an insoluble state by oxidation through air or another oxidizing agent, dispensing the use of mordants [7].

After a preliminary analysis of published studies on existing plant-based natural dyes with primary colors – yellow, red and blue - six dyes were selected for this research because of their color properties and medicinal properties potential: dyes from eucalyptus (*Eucalyptus globulus Labill.* (EG)) and weld (*Reseda luteola* L.), known to dye textiles with yellow color; madder (*Rubia tinctorum* L.) and annatto (*Bixa Orellana* L.), with red color; and true indigo (*Indigofera tinctoria* L.) and woad (*Isatis tinctoria* L.), with blue color. True indigo and woad are two different species of indigo plant. In this study, the term 'indigo' refers specifically to the true indigo species.

This study analyzes existing research on the medicinal and coloristic properties of the textiles dyed with natural dyes: eucalyptus, weld, madder, annatto, true indigo and woad, through a systematic literature review. This work understands the relationship between the different dyeing parameters (such as, dyeing methods, dye concentration, dyeing temperature and time, type of fiber, type of mordants and treatments) and their influence on the chromatic results and in the medicinal properties of the dyed fabrics. The literature exposed a lack of research on the latest advancements in the field. For this reason, a cross-disciplinary overview of the intersection of chromatic and medicinal concerns was considered pertinent. Additionally, this work provides fundamental support for

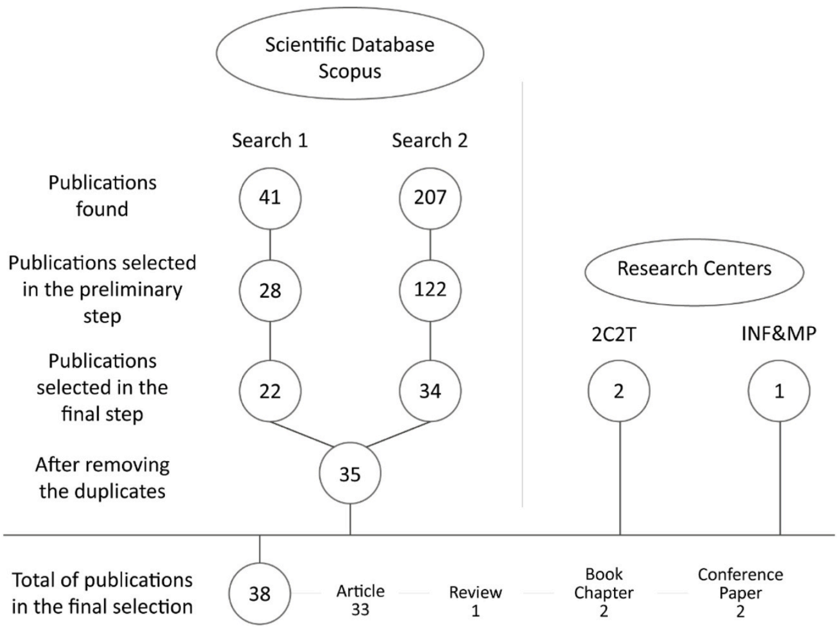

Fig. 1. Search steps in the Scopus database and in the Centre for Textile Science and Technology (2C2T) and Institute of Natural Fibers and Medicinal Plants (INF&MP) research centers.

the author's research project.

2. Methods

A systematic review of the literature was conducted based on the Torraco method [37]. The work focused on six natural textile dyes, eucalyptus, weld, madder, annatto, indigo and woad, studying their: i) main chromatic and medicinal properties; ii) dyeing and mordanting processes; iii) treatments and auxiliary products; iv) color strength and v) color fastness properties.

The selection of the body of scientific publications analyzed in this review comprised three main stages. The first two steps involved searching the Scopus database for two queries. Subsequently, a search was conducted on the publications of the two research centers with expertise in the area. All the publications were analyzed, and 38 were selected for the final review (Fig. 1).

For data collection, the Scopus database was searched using the following combination of keywords:

(eucalyptus OR weld OR "reseda luteola" OR madder OR "rubia tinctorum" OR annatto OR achiote OR "bixa orellana" OR indigo OR "indigofera tinctoria" OR woad OR glastum OR "isatis tinctoria" AND textile* OR fashion AND color* OR colour* OR dye* AND medicinal OR antimicrobial OR antibacterial OR anti-inflammatory OR anti-uv OR antioxidant)

Only articles, reviews, book chapters and conference papers were included in the research. The research was conducted in February 2023, with publication dates restricted to the previous 8 years (2015–2023).

Based on these criteria, 41 publications were included in the analysis. Titles, keywords, and abstracts were reviewed as the first filters in the preliminary selection step. Publications that met the requirements for discussing the chosen plants and their properties proceeded to the final selection process. In the previous stage, 28 publications were pre-selected. These publications were analyzed through an in-depth reading. New criteria were included in this phase in order to focus the research: English language, full-text access, focus specifically on the dye extracted from the plant species under study and being related to the textile dyeing processes. Of the 28 publications, 22 that met these criteria were selected.

Because the query was restrictive and several articles that were already known to the authors did not appear, an update was made to the query. The new search used the same research parameters as the previous search, changing only one of the Boolean operators AND to OR:

(eucalyptus OR weld OR "reseda luteola" OR madder OR "rubia tinctorum" OR annatto OR achiote OR "bixa orellana" OR indigo OR "indigofera tinctoria" OR woad OR glastum OR "isatis tinctoria" AND textile* OR fashion **OR** color* OR colour* OR dye* AND medicinal OR antimicrobial OR antibacterial OR anti-inflammatory OR anti-uv OR antioxidant)

A total of 207 publications were found, 36 of which were repeated in the previous search. The same steps were used for the respective filters of the previous query. Considering the publications already reviewed and after reading the titles, abstracts and keywords of the remaining publications, 122 publications were preselected. By applying the selection criteria of the last step, 34 of the 122 publications were selected. Only 1 paper at the intersection of the two queries was absent from this query. Therefore, these two queries were combined, and 35 publications were found to be eligible for the study.

Additionally, the work carried out by two specific research centers was also examined, even though the publications were not Scopus indexed. These institutions are the authors' research center, the Centre for Textile Science and Technology (2C2T), Portugal, and the Institute of Natural Fibers and Medicinal Plants (INF&MP), Poland, due to the growing collaboration between the two centers that have been working on this subject.

Applying the same inclusion criteria for queries in the database, 6 publications were pre-selected in the preliminary selection step. In the final selection process, only three publications met the selection criteria: two conference articles from 2C2T and one journal article from the Institute of Poland. A total of 38 papers were selected for this study after the database searches and the work of the research centers were merged.

After the final selection, the publications were organized by title, authors, type of publication (journal or conference), name of the journal or conference and year of publication. A narrative synthesis of the studies was conducted, and the main conclusions of each publication were divided by dye and summarized in Table 1 for discussion purposes.

3. Results and discussion

A literature review was conducted to examine the use and properties of six selected natural dyes in textile applications. These dyes are eucalyptus, weld, annatto, madder, woad and indigo. The review considered the following aspects: frequency of citation, medicinal benefits, color fastness properties, K/S values, fabric type, auxiliary products, and mordanting treatments. Table 1 resumes the main findings of this work associated with each natural dye.

The results showed that madder was the most frequently cited natural dye, appearing in 16 publications, while weld and woad dyes were the least frequently mentioned. All studies followed standardized testing methods. The common medicinal benefits of the selected natural dyes were antibacterial activity and UV protection, except for woad, which lacked evidence for the latter. The antibacterial action against gram-positive bacteria is more incident than against gram-negative.[1]

Some dyes also demonstrated other medicinal effects, such as anti-inflammatory, antioxidant, and anticancer properties. Moreover,

[1] Gram-positive and gram-negative bacteria are two groups of bacteria with different cell wall structure and health implications. Gram-positive bacteria, such as *Staphylococcus* species, have a thick cell wall and can cause infections like skin infections. Gram-negative bacteria, such as *Escherichia coli*, have a thin cell wall and can cause infections like pneumonia and urinary tract infections.

Table 1
Main findings of the research.

Natural Dye	Source/Authors/Year	Main findings
Eucalyptus ***(Eucalyptus globulus Labill.)***	[19] Das & Das, 2022	Woven cotton textiles were dyed with extracts of *Eucalyptus Globulus Labill.* (EG). The pre-mordanted fabric with alum had a greater color strength (K/S) than the original fabric. Dyed samples became darker due to the increase in EG-concentration. The antibacterial effect was more potent against gram-positive bacteria. K/S values and the antibacterial effect increased with an increase in the dye concentration. However, this property decreased after 20 washing cycles.
	[38,39] Endris & Govindan, 2021, 2022	Cotton fabrics dyed with EG leaf extracts were meta-mordanted with aluminum sulfate and post-treated with EG essential oil. A yellow color was obtained in the samples and the optimized samples (dyed and treated) showed excellent K/S and very good color fastness to lighting, washing, and rubbing. Essential oil treatment improved the antibacterial properties of the fabrics compared to dyed fabrics.
	[40] Yılmaz, 2021	Wool fabrics were EG-dyed without and with different mordants. Mordanted fabrics with alum gave a golden yellow color, with tin chloride a bright yellow color, with iron sulfate beige and with copper sulfate brown. Copper sulfate mordanted fabrics had the higher K/S values and alum mordanted fabrics had the lowest. All the samples showed good light and washing fastness. Non-mordanted EG-dyed fabrics showed antibacterial activity against *Staphylococcus aureus* (S. aureus). The addition of mordants also resulted in antibacterial activity against *Escherichia coli* (E. coli) as well.
	[41] Maqbool et al., 2019	Cotton fabrics dyed with extracts from seven plants, including EG, demonstrated excellent anti-ultraviolet (UV) properties, reasonable insect repellency, and a reasonable positive control against gram-positive and gram-negative bacteria. The fabrics also had good fastness properties but moderate K/S values.
	[42] Jajpura et al., 2019	Chitosan is one of the most abundant biodegradables, renewable, and nontoxic biopolymers available in nature. This makes it a suitable replacement for some synthetic polymers. EG-dyed fabrics are reported to demonstrate a wide scope of brown shades in the yellow-red color coordinates.
	[43] Silva et al., 2018	The EG-dyed cotton fabrics were pre-mordanted with chitosan. The absorption of the dye increased with increasing temperature, producing greater K/S values. Chitosan pre-mordanting also improved the fastness properties and became a more sustainable process by eliminating the use of electrolytes. These fabrics exhibited good antimicrobial and anti-UV properties.
Weld ***(Reseda luteola* L.)**	[44] Schmidt-Przewozna & Zajaczek, 2022	Linen fabrics pre-mordanted with alum and goal oak (*Quercus infectoria G.Olivier*) were dyed and meta-mordanted with 5 mordants. Meta-mordanting with alum resulted in lemon yellow, sodium carbonate in bright yellow tones, citric acid in beige, copper sulfate in olive green, and iron sulfate in brown. The dyed fabrics showed antimicrobial activity, positive effects on skin hydration, and excellent anti-UV properties.
	[45] Dumitrescu et al., 2018	Cotton weld-dyed fabrics were mordanted with mimosa or with a combination of mimosa and alum. All samples were yellow in color and showed excellent anti-UV properties and moderate antibacterial activity compared to the non-mordanted samples. The color fastness to washing and acid perspiration was very poor. Fastness to lighting and alkaline perspiration were moderate, with the latter improving with the greater use of mordants.
	[46] Sadeghi-Kiakhani et al., 2018	Cotton fabrics were pre-treated with a Chitosan–Cyanuric Chloride hybrid (Ch–Cy) or pre-mordanted with aluminum sulfate. All the samples were yellow in color. The K/S values and microbial reduction of the pre-mordanted samples were lower than those of the pretreated samples. K/S values increased with increasing Ch-Cy concentration. The color fastness was good for both dyed fabrics.
Madder ***(Rubia tinctorum* L.)**	[13] Elmaaty et al., 2023	Wool fabrics were madder-dyed and pre-mordanted with chemical and bio-mordants. Pre-mordanting with aluminum sulfate yielded reddish-brown hues, *Punica granatum* (pomegranate) produced heavily reddish orange, *Clonorchis sinensis* (C. sinensis) produced brown, and *Rhus coriaria* (R. coriaria) produced bright orange. The K/S values increased in mordanted samples. Color fastness were very good to excellent in all samples. Dyed fabrics showed antibacterial activity compared to un-dyed fabrics.
	[47] Sadeghi-Kiakhani et al., 2022	Wool yarns were pre-treated with chitosan-poly (amidoamine) dendrimer (Ch-PAMAM) hybrid, pre-mordanted with alum and dyed with cochineal or madder. The treated wool samples required 50 % less dye and dyeing time than raw wool samples and showed excellent antioxidant and antimicrobial properties.
	[48] Fernandes et al., 2022	The effect of plasma and cationization pre-treatments on the fastness and UV protection of cotton fabrics madder-dyed was investigated. The pH value changed the color of the fabric, when the pH of the solution increased, the color changed from yellow to purple. Cationization improved the dye uptake onto cotton, but with poor fastness. Simultaneous pre-treatment with metallic mordants significantly improved the fastness properties.
	[49] Alves et al., 2022	The madder-dyed cotton fabrics were pre-mordanted with two bio-mordants, quebracho or laccase and in mixtures with metal mordants. Pre-treatment with quebracho increased the K/S value, washing and UV light fastness, and UV protection, whereas laccase only showed an increase in UV light fastness. The samples treated with laccase and aluminum-potassium sulfate hydrate and iron sulfate heptahydrate showed a higher K/S than those treated with quebracho. Greater UV protection factors were obtained for the pre-treated samples.
	[50] Cai et al., 2021	Cotton fabrics were dyed with madder and alum, or with compounds of citric acid (CA) or a mixture of CA and dicarboxylic acids as cross-linking agents. Mordanted samples with alum

(*continued on next page*)

Table 1 (*continued*)

Natural Dye	Source/Authors/Year	Main findings
		had an orange color, with CA dark orange hue, and with CA and carboxylic acids orange-brown hues. The K/S values, wash, rub and light fastness, and medicinal properties increased and improved with cross-linking agents. This treatment also showed excellent anti-wrinkle and anti-UV properties.
	[51] Sadeghi-Kiakhani et al., 2021	Madder-dyed wool yarns and pre-treated with Sodium Alginate- Silver nanoparticles (SA-Ag NPs) demonstrated a significant increase in K/S values compared to untreated yarns. The pre-treated samples showed very high level of antimicrobial and antioxidant efficiency. The color fastness properties of the samples were generally very good.
	[22] Güzel et al., 2020	Silk fabrics were dyed with madder and gall oak (*Quercus infectoria G.Olivier)* and pre-mordanted with alum. Dark red colors were observed in the fabrics with gall oak and light red in the fabrics with only the madder. The antimicrobial activity against S.aureus bacterium increased in the silk fabric dyed with only madder. A combination of madder and 50 % gall oak affected both S. aureus and E. coli. Fabrics retained nearly 100 % activity up to 5 launderings against S.aureus and E.coli. No color change was observed after washing, demonstrating good K/S values.
	[52] Pour et al., 2020	The madder-dyed wool fabrics treated with bio-nano-mordant composed of microbial transglutaminase (*m*-TGase) and bentonite showed highest K/S values and antibacterial activity for comparing with untreated. The treated samples resulted in a change in hue from pink to red and a lighter red hue was obtained with higher *m*-TGase concentrations. The washing fastness was good, but the light fastness was not satisfactory.
	[53] Yadav et al., 2019	Pre-mordanted with chitosan and madder-dyed woolen fabrics showed increased K/S values with increasing chitosan and dyeing concentrations. The antibacterial activity was high in the mordanted samples and increased with increasing dye concentration. Wash and rub fastnesses were excellent.
	[54] Schmidt-Przewoźna et al., 2018	Madder-dyed organic cotton and knitted linen were pre-mordanted with alum and chebulic myrobalan (*Terminalia chebula Retz*), gall oak or oak bark (*Ouercus cortex*). The madder extracts had positive effect on affected skin. Madder-dyed samples showed reducing bacteria activity. The skin of women with dermatoses improved during the wearing of the tested clothes.
	[3] Basak et al., 2018	Madder is a natural dye that can provide a red color to cotton fabric. The color of the dyed fabric can be changed by changing the mordant. It has been reported that dyed fabrics exhibit adequate light and wash fastness.
	[46] Sadeghi-Kiakhani et al., 2018	Cotton fabrics were pre-treated with a Ch–Cy or pre-mordanted with aluminum sulfate. All the samples had reddish hues. The K/S values and microbial reduction of the pre-mordanted samples were lower than those of the pretreated samples. K/S values increased with increasing Ch-Cy concentration. The color fastness was good for both dyeing.
	[55] Dumitrescu et al., 2018	Cotton knits were dyed with madder extracts mordanted with mimosa and with the combination of mimosa and alum. Pre-mordanting with the combination of 2 % mimosa/4 % alum provided red-orange hues and 8 % mimosa/15 % alum orange hues. The color fastness to washing and lighting were poor and the fastness to perspiration and rubbing was moderate. The dyed fabrics demonstrate an excellent anti-UV property and moderate antibacterial activity.
	[56] Agnhage et al., 2017	Madder-dyed polyester fabrics showed a yellow/orange color. The dye improved both the anti-UV and the antibacterial properties of the fabrics. Moderate to good light fastness and good to excellent fastness to washing and rubbing.
	[57] Barani et al., 2017	Wool fabrics were pre-mordanted with silver nanoparticles (AgNPs) and dyed with madder. Pre-mordanted dyed fabrics had higher K/S values and better wash fastness compared to non-mordanted dyed fabrics. The samples presented good antibacterial activity against gram-positive bacteria.
	[58] Mehrparvar et al., 2016	Wool fabrics modified with a chitosan-polypropylene imine dendrimer (CS-PPI) were dyed with madder. This treatment showed improvements in K/S values, even when compared to aluminum sulfate pre-mordanting. Good color fastness values were also obtained.
Annatto ***(Bixa orellana* L.)**	[42] Jajpura et al., 2019	Annatto is a carotenoid-based natural colorant consisting of bixin and norbixin chemical constituents and has been reported to exhibit yellow to red shades on textile materials.
	[3] Basak et al., 2018	Annatto dye has been reported to produce orange-yellow color on textiles. However, the fastness properties of annatto dye are questionable.
	[59–61] Moses & Venkataraman, 2016, 2016a, 2017	Cotton fabrics were pre-treated with sodium hydroxide, morpholine or cellulase enzyme and dyed with annatto or other dyes. The treated annatto-dyed fabrics showed excellent light fastness, satisfactory wash fastness and poor rub fastness. The treated fabrics showed better antibacterial and UV protection than un-treated fabrics.
	[62] Shahid-ul-Islam et al., 2016	Studies have reported that annatto is a natural dye with huge potential in textile and leather sectors. Annatto colorants have demonstrated better results when applied to natural fibers. Most studies have shown that annatto extracts have potent biological activities.
	[63] Chattopadhyay et al., 2015	Jute fabrics were dyed with annatto and pre-mordanted with myrobalan or pomegranate, and inorganic mordants (iron sulfate or potash alum). All fabrics mordanted with iron sulfate produced dark red shades and with posh alum produced lighter and yellower shades. There was a substantial improvement in color yield, levelness of dyeing, and wash fastness properties when bio and inorganic mordants were used. Lightfastness was moderate and rubbing fastness was very good. Very good UV protection and antimicrobial activity were also observed.

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Table 1 (*continued*)

Natural Dye	Source/Authors/Year	Main findings
True Indigo (***Indigofera tinctoria*** L.)	[64] Reningtyas et al., 2021	Cotton fabrics dyed with natural indigo and coated with nano chitosan and zinc oxide nanoparticles (ZnONPs) showed increased light fastness. ZnONPs demonstrated ability as an anti-UV agent to protect the color of dyed cotton.
	[24] Tambi et al., 2021	Polyester fabrics were dyed with indigo and more two natural dyes (pomegranate and kumkum). The combination of natural dyes was used as primary colors to produce a variety of shades of blue, gray and green, e.g., 50:50 mix of kumkum and indigo dye produced a shade of green, while a 75:25 mix of indigo and pomegranate produced a shade of gray. According to the authors, the fabrics showed good K/S values (K/S > 1.7). The dyed samples displayed antibacterial activity and UV protection, but poor antioxidant activity.
	[65] Barani, 2020	AgNPs were synthesized through two methods: silver pretreatment and simultaneous silver treatment during indigo dyeing. Indigo-dyed wool samples without any silver treatment had a blue shade, the silver simultaneous and pre-treated undyed-wool samples had a pale yellow and brown-yellowish shade, AgNPs on the wool fibers surface had brown-yellowish shade, AgNPs pre-treatment resulted in a dark blue shade and low brightness, and simultaneous AgNPs treatment resulted in a blue shade. AgNPs increased K/S values of all samples.
	[3] Basak et al., 2018	Indigo is a well-known natural dye that produces a blue color. It has been reported that indigo dye has good fastness properties and is mainly used for the coloration and production of cotton denim.
	[59–61] Moses & Venkataraman, 2016, 2016a, 2017	Cotton fabrics were pre-treated with sodium hydroxide, morpholine or cellulase enzyme and dyed with indigo or other dyes. The treated indigo-dyed fabrics showed excellent light and wash fastnesses, but poor rub fastness. The indigo dye showed the highest antibacterial properties of all natural dyes analyzed in this study and excellent UV protection.
	[66,67] Jeyaraj et al., 2015, 2016	Silk fabric and silk-polyester-lyocell blends were dyed with indigo and pre-treated with hydrochloric acid. Both materials showed good K/S values. The wash and light fastness of both dyed materials were good, but the rub fastness properties were poor. All samples showed excellent antimicrobial property and very good UV protection.
Woad ***(Isatis tinctoria* L.)**	[3] Basak et al., 2018	Wood-dye has been reported to produce a blue color onto cotton and jute fabrics with good fastness properties.
	[36] Bektaş et al., 2016	Cotton and wool yarns were dyed with extracts of woad. Isatis leaves removed the toxic effects of sodium dithionite (a reducing agent) and showed antigenotoxic effect. Dark blue color was observed in wool yarns, while lighter blue color was obtained from cotton yarns.
	[21] Alkan et al., 2015	Silk fabrics were dyed separately with weld, gall oak, or a combination of weld and gall oak in different percentages to produce green colors. The results showed that the dark-colored fabrics had a greater reduction in the survival of S. aureus.

some dyes showed additional advantages, such as insect repellency, pleasant odor, and anti-wrinkle effects.

All dyes exhibited reasonable color fastness properties, although only one study assessed the color fastness of woad. Cotton fabric was the most widely used material for dyeing, followed by wool fabrics. Pre-mordanting was the most used treatment, with the best results for most of the selected dyes. Alum and chitosan were widely used mordants.

Alum is a safe and effective chemical compound, typically a hydrated double sulfate salt of aluminum and other metal, such as potassium, ammonium or sodium. Potassium alum, which has the formula $KAl(SO_4)_2 \cdot 12\ H_2O$, is the most common form of alum and is used in the selected publications. Islam et al. [68] reported that chitosan, a biopolymer obtained from the deacetylation of chitin, present predominantly in the exoskeletons of crustaceans and the cell walls of some fungi, has the potential to substitute some synthetic polymers due to its biodegradability, renewability and nontoxicity. This substance can be produced using waste from the seafood industry and can replace numerous harsh chemicals in the chemical and textile industries. Due to its ecologically and environmentally friendly properties, its production and applications are expected to increase significantly in the future [42].

- **Eucalyptus dye**

Eucalyptus plants are widely used as a source of essential oils in various fields such as medicine, perfumery, and coloration [69]. These plants have a high lignin content, which confers resistance to microbial and enzymatic action, as well as several medicinal properties, including anti-inflammatory, anticancer, antibacterial, antiseptic, and astringent properties [40,69]. Most previous studies on the application of eucalyptus in textiles have focused on the use of the essential oil extracted from the leaves of this plant. However, this study showed the potential of using eucalyptus as a textile dye.

Seven publications that discussed this dye were selected, including one review. The results showed that all studies used eucalyptus leaves or extracts as a natural dye for cotton or wool fabrics. According to the selected literature, the colors obtained with eucalyptus dyeing varied between yellow and brown, depending on the dye concentration, dyeing process, and mordant choice.

According to Yılmaz [40], EG-dyed wool fabrics mordanted with alum gave a golden yellow color, with tin chloride gave a bright yellow color, with iron sulfate beige and with copper sulfate brown. The latter dyeing experiment obtained the higher K/S values and the dyeing experiment with alum obtained the lowest. The mordant positively correlated the K/S value and influence the color of the fabric.

Regarding other dyeing process conditions, other studies found that increasing the dye concentration at the same temperature and time resulted in a higher K/S value [19,38]. According to Endris and Govindan [39], when the temperature was increased in cotton fabric dyeing process, the K/S value was not affected, suggesting that dyeing at lower temperatures is preferable for better results and a more efficient dyeing process. In contrast, according to Silva et al. [43], the absorption of the dye in the dyeing process of knitted cotton fabric pre-mordanted with chitosan increased with temperature, producing greater K/S values in the samples. This divergence may be due to the use of different dyeing methods. The first study used the padding method, whereas the second used the exhaustion method. In padding, the dye is impregnated into the fiber by pressure; therefore, temperature is not significant in dyeing bath. In contrast, during exhaustion, the dye is transferred by diffusion; therefore, at higher temperatures, the dye molecules enter the fabric faster. Other factors may have influenced these results, such as the different characteristics of the cotton samples or the mordant used.

Several processes were employed to enhance the K/S and color fastness properties of dyed fabrics. The most common processes were pre-mordanting or meta-mordanting with alum [19,40] or meta-mordanting with aluminum sulfate ($Al_2(SO_4)_3$) [38,39].

All articles that presented tests of fastness to lighting, washing, and rubbing (wet and dry) showed reasonable results [38–41,43]. This can be explained by the tannins in the eucalyptus extract, which function as mordants and fix the colors in the fibers [43]. Thus, eucalyptus extract is simultaneously a dye and a mordant. Additionally, the use of other mordants or bio-mordants improve dyeing properties, increasing, in most cases, 1 value for all fastness tests (lighting, washing, and rubbing), on a gray scale. For example, a study that pre-mordanted samples with chitosan showed improvements in wash and rub fastness (dry and wet), ranging from 3 to 4–5, from 4 to 5 to 5 and from 4 to 4–5, respectively. These results can be explained by the crosslinking between the polymer and fiber, resulting in the formation of positive groups on the cotton surface. These groups can interact with the negative groups of natural dyes, thereby increasing dye uptake and improving color fastness [43]. In other study that dyed fabric with and without aluminum sulfate mordant with same dyeing conditions, wash fastness values increased from 1 to 2 to 3–4, rub fastness from 4 to 4–5 (dry) and from 2 to 3 to 3–4 (wet), and light fastness from 3 to 4 [38]. However, Yilmaz [40] obtained a value of 5 for all woolen fabrics dyed without mordant or pre-mordanted with alum, tin chloride, or copper sulfate, except for iron sulfate, where the fastness value to washing was lowered to 4–5. This decline may have occurred because iron sulfate can react with dye molecules and form an insoluble solid, making the dye less soluble in water and more likely to fade.

All the selected articles that discussed the natural dye extracted from eucalyptus showed antibacterial action, with greater emphasis on gram-positive bacteria. A study conducted by Das and Das [19] reported antibacterial action against both types of bacteria; the higher dye concentration, the larger the diameter of inhibition. The authors also indicated that the antibacterial effect decreased after 20 washing cycles, yet it can still retain 56.2 %–88.5 % of its initial antibacterial property.

The auxiliary products and processes used may also interfere with medicinal properties. In the study conducted by Yilmaz [40], samples of wool-dyed with EG without mordant showed 99.99 % antibacterial effect values against *Staphylococcus aureus* (S. aureus) and no antibacterial effect against *Escherichia coli* (E. coli), however, when any of the mordants used in the study were applied (alum, iron sulfate, tin chloride or copper sulfate) the samples had a 99,99 % antibacterial effect against both bacteria.

Additionally, another study conducted by Endris and Govindan [38] reported that dyed-cotton fabrics mordanted with aluminum sulfate did not demonstrate a bacterial reduction for E. coli but showed a 50 % bacterial reduction for S. aureus. However, the application of EG leaf essential oil pos dye improved antibacterial properties, totally killing and removing both bacteria, even after 5 washes. After 10 washes, the treated samples showed 100 % reduction in gram-positive bacteria and 74 % reduction in gram-negative bacteria. This information is consistent with the work of Maqbool et al. [41] and Silva et al. [43], who observed a greater inhibitory effect against S. aureus bacteria in EG-dyed cotton samples, in first study without mordants, and in the second, pre-mordanted with chitosan. This last treatment improved the antibacterial action against S. aureus, but had no effect on E.coli. However, a larger bacterial inhibition zone (BIZ) was obtained for samples with higher dye concentrations, indicating that a higher dye concentration resulted in an increase in antibacterial activity.

The same applies to anti-UV properties; it is generally observed that a higher K/S and darker shades of dyed fabrics led to a higher Ultraviolet Protection Factor (UPF). Nevertheless, in a study conducted by Maqbool [41], some cotton fabrics with low K/S values exhibited maximum mean UPF and belonged to the excellent class of UV protection. Undyed fabric obtained an average value of 3.8 UPF, which does not offer protection against UV rays, according to the authors. This is attributable to the fact that mean UPF values are influenced not only by K/S values but also by the chemistry of the dye molecules and principal constituents of the dye mixture. In addition to this study, another study by Silva et al. [43] also reported this property in dyed cotton fabrics. According to the authors, UPF values increase with increasing concentration of the extract in the dyeing, which was also improved by pre-treatment with chitosan. For example, a fabric dyed with 10 g per liter (gL^{-1}) increased mean UPF values from approximately 16 to 25 compared to fabrics dyed with 50 gL^{-1}. Consecutively, the same samples pre-mordanted with chitosan increased, approximately, from 34 to 60 UPF values. As previously mentioned, samples pretreated with chitosan exhibited increased extract uptake, which led to enhanced absorption of some organic constituents, such as flavonoids. Therefore, it is probable that these substances are responsible for the UPF ratings of colored materials.

In addition to its medicinal properties, this natural dye exhibits insect repellency and pleasant odor as prominent features, which have been positively assessed in 4 out of seven publications [19,38,39,41].

- **Weld dye**

Weld plants have health benefits and usually show good results in terms of textile coloration [26]. Studies that evaluate the natural dyeing of the yellow color reveal the potential of the weld dye to obtain dyeing with good fastness to washing and resistance to lighting. These studies also demonstrated antimicrobial activity, which reverberated the positive effects on the skin, such as increasing hydration and protection against UV radiation [26,70].

Only three publications discussing this dye were selected. The results showed that two of the publications performed dyeing on cotton fabrics, while the other carried out dyeing on linen fabrics. All dyeing resulted in colors ranging from yellow to brown [44–46].

Different mordants were used to obtain different shades and tones. In a study carried out by Schmidt-Przewozna and Zajaczek [44], pre-mordanted fabrics with alum and gall oak (*Quercus infectoria G.Olivier*) and meta-mordanted with: a) alum resulted in lemon yellow; b) sodium carbonate in bright yellow tones; c) citric acid in beige; d) copper sulfate in olive green; and e) iron sulfate in brown. However, some of these mordants, such as copper sulfate and iron sulfate, have toxic effects that limit their use.

The use of mordants to improve color properties was common in publications on this dye, especially the use of alum, either alone or in combination with bio-mordants [44,45]. Treatment with crosslinked chitosan–Cyanuric Chloride hybrid (Ch–Cy) was also reported. According to Sadeghi-Kiakhani et al. [46], the K/S values of the cotton samples dyed and pre-mordanted with aluminum sulfate were lower than those of the samples pretreated with Ch-Cy. It was observed that the K/S values increased with increasing Ch-Cy concentration. According to the authors, this treatment was primarily responsible for enhanced dye absorption. The treatment of cotton with Ch–Cy suggests that it can be dyed more deeply without the need for aluminum sulfate mordanting.

With this dye it was possible to observe an improvement in color fastness with the use of mordants. The pre-mordanting with alum and gall oak improved the fastness to washing and lighting, going from a value of 4–5 compared to the non-mordanted dyed samples in both fastness, but resulted in no difference in rubbing, perspiration, and hot-pressing fastness with already good results, with value of 4 [44].

However, another study reported that fabrics had very poor color fastness to washing and acid perspiration (with values between 1 and 2), with the worst results observed for fabrics mordanted with both mimosa and alum compared to those mordanted with only mimosa. On the other hand, fabrics pre-mordanted with larger amounts of mordants, especially with 8 % mimosa +15 % alum, showed better color fastness to alkaline perspiration (3–4). The color fastness for wet rubbing was primarily good for fabrics pre-mordanted with 2 and 8 % mimosa tannin (3–4), whereas the dry rub fastness had very interesting results (4–5). The light fastness of all fabrics was moderate (3) according to gray scale for both 7 h and 26 h of exposure, regardless of the percentages of mordants used [45].

The medicinal properties of weld natural dye for textile applications were also examined in the literature review. The results showed that the weld dye had excellent anti-UV properties in samples pre- or meta-mordanted, according to two of the selected articles [44,45]. According to Schmidt-Przewozna and Zajaczek [44], linen samples that were pre-treated with alum and gall oak and dyed with weld and soda mordant had very good protection against UV rays, with UPF 25 based on the Australian Standard. Samples mordanted with copper and iron showed even better results, achieving excellent protection (UPF 50), as did samples pre-mordanted with alum and mimosa or with mimosa alone [45]. The mordant seems to have a significant influence on the anti-UV property of dyed fabrics, possibly because it helps to fix the dye in the fiber and increase the resistance to UV rays.

Moreover, all studies showed antibacterial action in dyed samples, particularly against gram-positive bacteria. According to Sadeghi-Kiakhani et al. [46], the antibacterial property was maintained even after 10 washing and drying cycles, with a microbial reduction of 99.96 %–91.14 % against E. coli and 98.61 %–85.61 % against S. aureus. This study also showed the effectiveness of treatment with chitosan–Cyanuric Chloride hybrid (Ch–Cy) in improving the antibacterial activity. Another study reported an antibacterial effect against S. aureus on cotton-dyed fabrics pre-mordanted with mimosa and alum. However, low dyestuff content in fabrics reduced the effectiveness of weld-dyed textiles against bacteria [45].

In addition to its medicinal properties, this natural dye also exhibited anti-wrinkle properties.

- **Madder dye**

Madder was the most frequently cited natural dye in the selected literature, appearing in 16 publications, including one review article and one book chapter. All studies used madder extracts as a natural dye for cotton or wool fabrics or yarns, except for two studies that used silk and polyester fabrics. The madder plant also performed is reported to perform well in combination with other natural dyes and different mordants, allowing a wide spectrum of colors resulting from these combinations [70,71]. However, in selected publications, only the combination with different mordants was explored.

According to the selected literature, the colors obtained with madder dyeing vary between red, pink, orange and yellow. The choice of mordant had a significant effect on the resulting color: aluminum sulfate yielded reddish-brown hues, *Punica granatum* (pomegranate) produced heavily reddish orange, *Clonorchis sinensis* (C. sinensis) produced brown, and *Rhus coriaria* (R. coriaria) produced bright orange [13].

In another study conducted by Dumitrescu et al. [55], alum gave lighter shades to the fabrics. It was also possible to obtain a red color in fabrics mordanted with mimosa tannin and dyed with the madder extract. The combination of alum and mimosa in pre-mordanting provided red-orange and orange shades with 2 % mimosa/4 % alum and 8 % mimosa/15 % alum, respectively [55]. Also, according to Fernandes et al. [48], the pH value changed the color of the fabric, when the pH of the solution increased, the color changed from yellow to purple.

Treatment of wool with bio-nano mordant resulted in a change in hue from pink to red. The concentration of microbial transglutaminase (*m*-TGase), an enzyme produced by the *Moraxella osloensis* bacteria, was found to affect the color of the dyed wool, with higher concentrations resulting in a lighter red hue. Treatment with 5 % *m*-TGase and 5 % bentonite by weight of fiber (WOF) promoted dyeability and increased madder dye absorption. Bentonite is a clay that is created by the weathering of volcanic ash. Both materials are considered good candidates for the textile industry because of their environmentally friendly properties [52].

The literature review also examined the use and properties of different processes to enhance K/S, color fastness, and medicinal properties of fabrics dyed with madder. The results showed that the application of chitosan and chitosan nanoparticles was one of the most common treatments, as well as the pretreatment of samples with cationization, plasma and silver nanoparticles (AgNPs). The chemical process of cationization changed the cellulose molecule, making it strongly cationic. As a result, the cotton fiber developed positive surface charges, which aided in its ability to interact with color. Cotton cationization allowed salt-free dyeing and improved substrate dyeability [72]. In other study that used plasma treatment, no variations in saturation were observed on dyed cotton fabrics; however, cationization significantly increased the K/S [48].

In addition to AgNPs, treatment with Sodium Alginate-Silver nanoparticles (SA-AgNPs) was also used to enhance antibacterial and color properties of dyed fabrics. Both methods can increase the K/S and durability of colored materials while reducing the amount of water, energy, and chemicals used in the dyeing process. Wool yarns dyed with madder and pretreated with SA-AgNPs demonstrated a significant increase in K/S values compared to untreated yarns [51]. However, due to the potential release of silver ions into the soil or wastewater, treatment with AgNPs may also pose certain environmental hazards [73].

The findings revealed that alum and aluminum sulfate were the most frequently used mordants, individually and in combination with some of the above-mentioned treatments, including chitosan or chitosan modifications. According to Cai et al. [50], alum mordant dyeing enhanced the K/S of the madder dye on the cotton fabric because the K/S value of the mordant dyed fabric was approximately 85 % higher than that of the directly dyed fabric.

A study conducted by Sadeghi-Kiakhani et al. [47] indicated that a chitosan-poly(amidoamine) hybrid (chitosan-PAMAM) can increase dye absorption by almost 10 % in the exhaustion process of natural dyes compared to chitosan. In addition, other study reported that treatment with a chitosan-polypropylene imine dendrimer (CS-PPI) showed improvements in K/S values, even when compared to aluminum sulfate pre-mordanting [58].

Studies that used pre-mordanting with alum and chitosan in conjunction with another treatment tended to have better color fastness and K/S values results [46,47,53]. However, the exclusive application of other bio-mordants such as mimosa, quebracho, and laccase did not show satisfactory results in terms of fastness properties [49]. But this does not happen with all bio-mordents, in other study, pre-mordanting with pomegranate, R. coriaria, and C. sinensis, showed improvements in K/S values, which can be explained by these mordants being rich in tannins, components that improve the absorption of alizarin (the main madder pigment) into the fabric [13].

The dyeing process time and temperature both have an impact on the K/S values. In a study by Sadeghi-Kiakhani et al. [51], K/S values increased slightly when the dyeing process was extended from 30 to 60 min, but then leveled off. The temperature of the dyeing bath did not show the same results. At 70 °C, there was greater absorption of the dye than at 90 °C. This can lead to optimization of costs and energy consumption associated with natural dyeing of wool fibers. The treatments mentioned above improve K/S, fastness, and medicinal properties.

Madder dyeing of polyester fabric at high temperature without mordants achieves a moderate to good (3–4) light fastness and a good to excellent (4–5) washing and rubbing fastness [56]. In contrast, in a study conducted by Elmaaty et al. [13], there was no difference in fastness to lighting, rubbing, and washing between wool fabrics dyed without a mordant and those dyed with bio-mordants or aluminum sulfate. All samples of this study had satisfactory results, with fastness rating of between 4 and 5, even after 5 washing cycles. In wool fabrics treated with bio-nano mordants, the fastness to washing showed better results compared to untreated fabrics, although it is not satisfactory (2–3) [52].

However, a study by Schmidt-Przewoźna et al. [54] reported that dyeing of samples pre-mordanted with a combination of alum and two different bio-mordants, Chebulic Myrobalan (*Terminalia chebula Retz*) and gall oak, showed a significant improvement in fastness to washing. On the linen/elastane fabric, the fastness to washing increased from 3 to 4 to 5; on linen, it increased from 2 to 3 to 5; and on organic cotton, it increased from 4 to 4–5. All the washing tests were conducted according to the same standards.

Another study showed that pre-treatment of wool samples with chitosan obtained excellent results for fastness to washing and rubbing, with a rating of 4–5 and 5 [53]. This is supported by another study, which found that both pre-treatment with Ch-Cy or pre-mordanting with aluminum sulfate were effective in improving fastness to washing, lighting, and rubbing, with ratings changing from 2 to 3 to 4–5; 3–4 to 5; and 3 to 4, respectively. Both these studies suggested that the amino groups on chitosan molecules are likely responsible for the improved bonding with dye molecules on cotton fabrics [46].

The pre-treated wool fabrics with AgNPs also showed promise, increasing the fastness to washing of the samples from 4 to 4–5 and showing higher K/S values [57]. However, according to Dumitrecu et al. [55], mimosa mordanting, either alone or in combination with alum, demonstrated a very low fastness to washing and lighting (1 and 3, respectively). This is likely because mimosa mordanting does not form strong bonds with dye molecules. Mimosa mordanting is not as effective, but it can still improve the rubbing fastness.

Overall, the results of these studies suggest that the amine groups on chitosan molecules are the most effective way to improve the wash and rub fastness of the madder-dyed textiles.

Medicinal properties were addressed in thirteen of the selected articles. These articles showed an antibacterial effect with a greater emphasis on gram-positive bacteria. A study by Elmaaty et al. [13] found that samples pre-mordanted with bio-mordants showed a high antibacterial effect against gram-negative bacteria, with BIZ between 15 and 25 mm. However, the same study found that the same samples had a low effect against gram-positive bacteria, with a diameter BIZ of only 3 mm. This antibacterial effect may be due to the release of alizarin molecules from the dyed fabric, which disrupts the bacterial cell wall and inhibits bacterial growth. It was also possible to verify dyed fabrics with antibacterial activity against both groups of bacteria. This was supported by a study by Agnhage et al. [56] which found that madder-dyed polyester fabric had an antibacterial activity of 86 % against both types of bacteria tested, while the undyed fabric had an antibacterial activity less than 17 %.

Another study showed that dyeing only with madder and pre-mordanted with alum increased the antibacterial action against S. aureus, but not against E. coli. When this dyeing was combined with 50 % gall oak, an inhibition rate of more than 99 % was obtained against both bacteria, a property that was maintained after 5 washes [22].

Wool yarns that were not treated with any products had weak antimicrobial activity of 20 %. However, when the yarns were treated with SA-AgNPs, the antimicrobial activity increased to 74 %. The antimicrobial activity was further improved to 80–85 % when the yarns were dyed with madder after SA-AgNPs treatment. The treated and dyed yarns also had very good durability, with antimicrobial activity against E. coli and S. aureus of more than 70 % even after 10 washing cycles [51].

According to Pour et al. [52] the pre-mordant with *m*-TGase and bentonite resulted in increased antibacterial activity in madder-dyed wool fabrics. The antibacterial properties of these bio-mordants can explain their effectiveness in textile dyeing. They improve the fixation of dyes in fibers and reinforce the dye activity.

In cotton fabric, the antibacterial property was significantly enhanced by dyeing with madder. The SUA + CA (succinic acid + citric acid) cross-linking of dyed cotton fabric further improved its antibacterial performance by more than 60 % compared to untreated cotton fabric. This was mainly attributed to the anthraquinone structure of madder dye, which could interact with bacterial proteins and inhibit their functions. This process affected both Gram-positive and Gram-negative bacteria. Cross-linked dyed cotton fabric had the best antibacterial property, around 70 %, due to its high color yield [50].

Another study showed that non-mordanted dyed fabrics had good antibacterial activity against S. aureus and E. coli, with bacterial reductions of 95.3 % and 96.2 %, respectively. This could be attributed to the linkage of chromophores to glycosides in natural dyes. According to the authors, he antibacterial activity of the dyed samples showed significant increases after pre-mordanting with chitosan and the dye concentration increased. With the same dye concentration, mordanting with chitosan increased the bacterial reduction of S. aureus and E. coli to 99.2 % and 99.1 %, respectively. When the dye concentration increased, bacterial reduction also increased, demonstrating the antibacterial property of the madder natural dye [53].

Furthermore, a study conducted by Sadeghi-Kiakhani et al. [46] found that bacterial reduction was below 90 % in samples treated with chitosan. However, in samples treated with Ch-Cy, bacterial reductions increased to 99.96 % and 98.61 % for E. coli and S. aureus, respectively. These percentages decreased after ten washes to 91.14 % and 85.61 %.

In addition to antibacterial activity, dyed fabrics have also been shown to offer UV protection. The study conducted by Dumitrescu et al. [55], found that cotton samples mordanted with mimosa or a combination of mimosa and alum had excellent UV protection results (50+ UPF). The same was true for the polyester fabric, which had an undyed samples UPF value of 65. According to authors, this value is already high, nevertheless, when the fabric was dyed with 3 % and 5 % WOF of madder dye, the UV protection increased to 106 and 112 UPF, respectively [56].

The antioxidant properties were also mentioned in selected publications [48,51,54]. A study by Schmidt-Przewoźna et al. [54], found that fabrics dyed with madder and pre-mordanted with three different bio-mordants, Chebulic Myrobalan, gall oak or oak bark (*Ouercus cortex*), and alum had positive effects on the skin of people with different types of dermatoses. The fabrics were found to reduce skin aging, strengthen and tone blood vessels, increase skin hydration, and reduce skin lesions.

Madder dye has also been shown to have anti-wrinkle features, as assessed in two publications [46,50].

- **Annatto dye**

Annatto plants have numerous applications in traditional medicine. Several studies point to beneficial properties, such as antibacterial, analgesic, healing, antioxidant, and astringent properties, which are also used for the treatment of skin diseases [74,75].

Seven publications related to this dye were selected, one of which corresponds to a review section and two to a book section. All articles used annatto extracts as natural dyes for cotton and jute fabrics. According to the literature, the colors obtained with annatto dyeing vary between yellow, orange, and red. According to Basak et al. [3], the use of mordants for this natural dye is essential, and the color of the fabric can be influenced by the use of these mordants.

All articles mentioned pre-treatment fabrics, three with sodium hydroxide, morpholine, and cellulase enzyme [59–61] and one with bio-mordants–myrobalan, pomegranate, and inorganic mordants (ferrous sulfate or potash alum) [63]. The latter study obtained more satisfactory results with regard to the color fastness properties and K/S values compared to the three treatments used in the first articles. Nevertheless, in this study, the double mordant of one of the bio-mordants with one of the metallic mordants resulted in a substantial improvement in the uniformity and levelness of the dyed jute fabric. For two-fold mordanting, K/S values increased for bio-mordant plus subsequent ferrous sulfate treatment due to supporting and additive color interactions, but K/S values declined for bio-mordant plus subsequent alum treatment due to complementary or opposite color interactions.

In the study conducted by Chattopadhyay et al. [63], it was also possible to observe low fastness values to washing in jute fabrics dyed with annatto with different pH values, ranging between 1 and 2 and 2. However, these values improve when pre-mordanting is applied to the samples (between 3 and 3–4), either with the myrobolan bio-mordant with iron sulfate or alum, or with the pomegranate bio-mordent with iron sulfate or alum. Fastness to light and friction is also improved, although not as markedly.

There is no significant improvement in the wash fastness of the woven and knitted cotton fabric (sodium hydroxide treated, morpholine treated, cellulase treated and untreated) dyed with annatto [61].

All selected articles that discussed the natural dye extracted from annatto showed good UV protection properties and satisfactory antimicrobial activity against gram-positive bacteria. According to Moses and Venkataraman [61], fabrics dyed with untreated cotton had a UPF value of 34, which is considered good protection against UV rays. Fabrics treated with sodium hydroxide, morpholine or cellulase enzyme had UPF values between 37 and 40, which is a slight improvement. However, in other study, a greater improvement was observed in jute fabrics pre-treated with double mordant compared to non-mordanted fabrics (24.14 UPF). UPF values rose to 38.55, 35.50, 37.29 and 36.19 for fabrics mordant with myrobolan and iron sulfate, myrobolan and alum, pomegranate and iron sulfate and pomegranate and alum, respectively [63].

In addition to its medicinal properties, anti-odor and stain resistance were evaluated as prominent features with good results [59–61].

- **True Indigo dye**

Indigo plants have been used to produce blue dyes since prehistoric times. These dyes are free of mordants and have anti-UV, anticancer, cytotoxic, antiseptic, and healing properties. They can also contribute to the treatment of infections and dermatological, respiratory, and infectious diseases [76,77].

Nine publications were selected for this study, one of which was a book chapter. Cotton was the most common material used for dyeing with indigo leaves or extract, followed by polyester, silk, a mixture of silk with polyester, and lyocell. According to the literature, the colors obtained with eucalyptus dyeing vary between blue and gray. However, the study conducted by Tambi et al. [24] showed that indigo can be combined with two other natural dyes (pomegranate and kumkum) to obtain secondary shades by mixing the primary colors. Beyond blue and gray, the authors reported that it was possible to obtain shades of green by changing the percentages of the combined dyes. For example, a 50:50 mix of kumkum and indigo dye produced a shade of green, while a 75:25 mix of indigo and pomegranate produced a shade of gray.

The color also varies depending on the treatment. In the study conducted by Barani [65], indigo-dyed wool samples without any silver treatment presented a blue shade. Moreover, the undyed-samples had a pale yellow and brown-yellowish shades with silver simultaneous treatment and silver pre-treatment, respectively. AgNPs on the wool fibers surface resulted in brown-yellowish shade. Therefore, pre-treated and dyed wool samples with silver ions resulted in a dark blue shade and low brightness. However, the simultaneous silver treatment produced a blue color with higher chromaticity values than the silver pre-treatment.

Natural dyes are difficult to extract and produce and can be difficult to work with due to lack of knowledge and standardization in procedures and materials used. However, there are potential solutions to these challenges that are worth exploring. A field of potential development can be the production of different shades using the primary colors of natural dyes. This can be achieved by mixing different dyes together or by using different mordants. This would allow for the use of only three primary dyes, which would simplify the dyeing process and make it more accessible to the textile industry [28].

According to Tambi et al. [24], the blending of dyes produced different hues with varying color intensities. In this study, the dye present in a higher proportion dominated the color of the dyed fabric in most secondary shades. Variations in tone and lightness of shade were caused by variations in dye concentration in binary mixtures.

Several processes have been employed to enhance the K/S and color fastness properties of dyed fabrics. All tests in the selected literature performed pretreatments on the samples. The products used for this pretreatment varied between AgNPs, chitosan and ZnONPs, sodium hydroxide, morpholine, cellulase enzyme, and hydrochloric acid.

In general, it was possible to observe good levels of K/S. According to Tambi et al. [24], the K/S values changed as the dye concentrations changed. Pre-treatment with AgNPs also influenced K/S of dyed fabrics. Indigo-dyed wool samples without silver treatment presented lower K/S values at all visible wavelengths compared with the silver-treated indigo-dyed samples [65]. In other study, silk fabrics and silk-polyester-lyocell blends dyed with indigo and pre-treated with hydrochloric acid showed good fastness to lighting and washing. However, the rub fastness was poor [66]. Compared to other dye classes, indigo-dyed fabrics typically have high wash fastness properties. This is due to the insoluble characteristic of indigo dyes which forms a covalent bond with the cellulose fibers of the fabric, improving color fastness. Therefore, all indigo-dyed woolen yarn samples showed an excellent washing fastness. Additionally, simultaneous silver treatment during indigo dyeing process improved the light fastness (from 6 to 7 to 7–8 on the blue scale) and maintained the wash fastness (4–5 on the gray scale) of dyed fabrics compared to pre-treatment with silver. In these processes, sodium dithionite was used as the reducing agent [65].

Cotton fabric treated with sodium hydroxide and dyed showed improved wash fastness (4–5) compared to morpholine treated, cellulase treated, and untreated cotton fabrics (3–4 and 4). Sodium hydroxide also showed higher light and rubbing fastness values. These findings suggest that sodium hydroxide treatment can improve the durability of dyed cotton fabrics [61].

The study conducted by Reningtyas et al. [64] used nanochitosan and ZnONPs as anti-UV agents to protect the color of natural indigo dyed cotton. The authors found that applying a layer of nanochitosan before coating cotton with ZnONPs increased the amount of ZnONPs absorbed by the cotton by up to four times. This resulted in improved light fastness properties compared to untreated cotton. In this study, sodium dithionite ($Na_2S_2O_4$) and sodium carbonate (Na_2CO_3) were used as reduction agents together with a natural dye in the dyeing process.

The results showed that the polyester fabric dyed using high-temperature high-pressure (HTHP) dyeing process had satisfactory color fastness to lighting (5–6), washing (4) and rubbing (3–4). This is likely due to the mechanism of dyeing polyester using the HTPH process, showing the influence of the temperature [24]. However, it is important to note that the fastness to rubbing of indigo dyed fabrics is generally low. This is supported by the study by Jeyaraj et al. [66], which demonstrated good fastness to washing and lighting (4) and low fastness to rubbing (between 1 and 2 and 2).

Regarding medicinal properties, six publications showed antibacterial properties and 4 anti-UV properties. However, only one study has performed quantitative antibacterial tests, which showed considerable antibacterial properties with a bacterial colony reduction of >83 % [24]. In the remaining studies, qualitative tests were used to measure the BIZ. The BIZ of dyed chitosan applied cotton fabrics varied between 26 and 40 mm [60] while the BIZ of dyed silk and its mixed fabric varied between 36 and 42 mm [66,67]. In all tests, the antibacterial action was superior against S. aureus compared to E. coli.

According to Moses e Venkataraman [61], fabrics dyed with untreated cotton obtained good protection against UV rays with a UPF value of 33. However, there was no significant improvement in the UPF value of fabrics treated with sodium hydroxide, morpholine or cellulase enzyme, with values ranging from 36 to 38 of UPF.

Indigo-dyed polyester fabrics showed significantly higher UPF values than undyed polyester fabrics. The UPF value of an undyed polyester fabric is 42.97, while the UPF value of an indigo-dyed polyester fabric is 198.40. This difference is likely due to the fact that

Table 2
Summary of colors obtained for different dyes as a function of different auxiliary products and treatments.

Auxiliary products and treatments	Natural dyes					
	Eucalyptus	Weld	Madder	Annatto	True indigo	Woad
None	Yellowish-brown [40]; "brown shades" [42]	Light yellow [44]; Beige [46]	Red [3]; "yellow/orange" [56]; Pink [46,48,52]; Brown, reddish-brown and dark brown [13]; Beige [49]; Orange [50]	"Orange-yellow" [3, 62]; "Yellow to red" [42]; Orange [59–61]	Blue [3,24, 59–61,65]; Gray [24]	Blue [3,36]; Dark and light blue [36]
Alum	Golden yellow [40]; "darker" [19]	Yellow [44]	Orange [50]; Pink [48,49]; Light red [22]			
Aluminum sulfate	Yellow [38,39]	Light yellow [46]	Reddish-brown [13]; Red [46]			
Tin chloride	Bright yellow [40]					
Iron sulfate	Beige [40]	Brown [44]	Gray [48,49]			
Copper sulfate	Brown [40]	Olive green [44]				
Sodium carbonate		"Light lemony" [44]				
Citric acid		"Light creamy" [44]	Orangish brown [50]			
Mimosa		Yellow [45]	Red [55]			
Pomegranate			Reddish orange [13]		Gray, blue, dark blue olive green [24]	
Clonorchis sinensis			Brown [13]			
Rhus coriaria			Bright orange [13]			
Kumkum					Green, dark blue, bright blue [24]	
Quebracho			Beige [49]			
Laccase			Beige [49]			
AgNPs					Blue and dark blue [65]	
Ch–Cy		Yellow [46]	Red [46]			
Plasma			Beige and pink [48]			
Cationization			Pink and purple [48]			
Gall oak						Green [21]
Gall oak + alum		"Creamy yellow" [44]	Dark red [22]			
m-Tgase + bentonite			"Pink to red" [52]			
Alum + mimosa		Yellow and dark yellow [45]	Reddish orange; Orange [55]			
Citric acid + carboxylic acids			Orangish brown [50]			
Plasma + alum			Pink [48]			
Cationization + alum			Pink and purple [48]			
Plasma + iron sulfate			Gray and beige [48]			
Cationization + iron sulfate			Purplish gray [48]			
Quebracho + alum			Pink [49]			
Quebracho + iron sulfate			Gray [49]			
Laccase + alum			Pink [49]			
Laccase + iron sulfate			Dark gray [49]			
Myrobalan or pomegranate + iron sulfate				Dark red [63]		
Myrobalan or pomegranate + potash alum				Light and yellowish red [63]		

indigo dye molecules can penetrate the polyester fibers, blocking UV radiation from passing through the fabric [24].

In addition to its medicinal properties, indigo dye also exhibited anti-odor and stain resistance. These properties have been positively assessed in three of seven publications [59–61]. However, indigo-dyed polyester showed significantly lower antioxidant activity than other dyed samples [24].

- **Woad dye**

Three publications were selected for this study, one of which corresponded to a book chapter. Several samples were used to test dyeing with woad dye; the most used was cotton, followed by wool, silk, and jute. According to the selected literature, the textile colors obtained with this dye can vary between blue and gray and these dyes can be used together with other natural dyes to provide other colors. The combination of woad with weld and gall oak is one such case that provided a green color [21]. According to the authors, dyed-woad silk fabrics were mordanted with alum and then, fabrics were dyed separately with weld and gall oak and together weld and gall oak in different percentages to obtain green color. The color also varies depending on the fiber used. According to Bektaş et al. [36], woad-dyed woolen yarns obtained a darker blue color than cotton yarns. Though, conforming to Basak et al. [3] it has been reported that woad-dye produces a blue color on both cotton and jute fabrics with good fastness properties. However, none of the selected articles performed color fastness tests for dyeing with woad.

According to Bektaş et al. [36], sodium dithionite has been considered the best reducing agent in traditional and industrial dyeing. However, this reducing agent and its derivatives are major pollutants in the textile industry and subsequently have hazardous effects on public health, showing genotoxicity in human lymphocytes. In accordance with the author, when the product is used in dyeing with woad, this dye removed the genotoxic effects of sodium dithionite due to its antigenotoxic properties.

The antibacterial action was only evaluated in a study that analyzed the possibility of green color with the overlapping of different natural dyes and the alum mordant. The results of the counting test showed a greater reduction in the survival of S. aureus in dark-colored fabrics. Bacterial reduction was 91–99 % in samples dyed with the three natural dyes (woad, gall oak, and weld). However, in samples dyed only with woad and weld, no bacterial reduction was identified, demonstrating that this property originates from the natural dye gall oak and not from the woad [21].

3.1. Final discussion

Many mordants and treatments were used in textile dyeing to improve the coloristic properties and obtain different colors and shades in the fabrics used, as shown in Table 2. It can be observed that eucalyptus and weld dyes can produce a yellow color with alum, aluminum sulfate or without mordants, for example. When EG-dyed fabrics are mordanted with copper sulfate, the color becomes brown; however, weld-dyed fabrics mordanted with the same mordant produce olive-green. Both madder and annatto dyes, produce orange and red colors in un-mordanted fabrics; however, the literature showed that madder can be used with a variety of auxiliary products and treatments to obtain other colors, such as pink, gray, brown, beige, and purple. The dyes extracted from indigo plants can produce blue and gray colors and when combined with other dyes, such as pomegranate, kumkum or gall oak, can also produce green.

However, the variation in shades does not depend only on the mordants and treatments used. In the documents analyzed, it was possible to observe that the chromatic properties were also affected by i) the dyeing methods, ii) the concentration of the dye and mordants, iii) the dyeing temperature and time and vi) the type of fiber.

The exhaustion method is the most used in the dyeing process of all selected natural dyes. Despite being more time-consuming than other methods, the preference for the exhaustion method may be related to the production of more consistent results by controlling the temperature, time and amount of dye and auxiliary products to be used.

All dyes showed antibacterial properties, but the antibacterial activity was likely to decrease over time and with washing. However, the use of mordants and treatments, such as plasma treatment or the application of essential oils, can delay this decrease and improve the antibacterial action. Many mordants used in textile dyeing with natural dyes also possess medicinal properties. This means that, in addition to helping a higher percentage of dye to bind with the fiber, they will also reinforce the antibacterial activity present in the dye, leading to an increase of antibacterial activity in the dyed fabric.

The fashion industry relies on changes and novelty to drive sales and consumer interest. Although some colors are consistently popular and repeated in fashion trends, the need for change underscores the importance of a diverse color palette in textile dyeing. This study analyzed the potential that natural dyes to create a wide range of colors. The table systematized the colors of the dyes studied in order to provide a better overview of the chromatic palette of each dye, which is not immediately apparent in isolation in the publications. However, each dye has unique characteristics that require specific approaches in relation to the fiber that will interfere with the color obtained. The complexity of the different variables in the dyeing process makes it difficult to establish standards for natural dyes, which leads to the need to optimize the processes, as it is difficult to obtain the same color over different baths or reproduce a shade.

The information available is often experimental and vague. There is a growing concern about finding more environmentally friendly alternatives to synthetic dyes, while also allowing designers to explore fashion from a chromatic point of view, without the restriction of a small palette.

4. Conclusion

Color is one of the most important elements of the fashion and textile industry and is a key factor in consumers purchasing decisions. As consumers become more aware of the environmental and health impacts of synthetic dyes, demand for natural dyes is expected to increase.

Natural dyes can be difficult to extract and produce and they are usually reported to dye textiles with limit color shades. However, there are ways to try to overcome these challenges. One promising development area is the creation of different shades using primary colors of natural dyes and different mordants. This would allow the use of only three primary dyes, which would simplify the dyeing process and concentrate the research work on three colorants.

This study showed that the chromatic properties and antibacterial activity of six natural dyes derived from eucalyptus (*Eucalyptus globulus Labill.*), weld (*Reseda luteola* L.), madder (*Rubia tinctorum* L.), annatto (*Bixa Orellana* L.), true indigo (*Indigofera tinctoria* L.) and woad (*Isatis tinctoria* L.) can be affected by a variety of factors. The use of mordants and treatments can help to improve the color-fastness and antibacterial properties of natural dyes, making them a more sustainable and environmentally friendly alternative to synthetic dyes.

The medicinal properties of natural dyes have become increasingly important in textile applications, emphasizing the need to refine dyeing processes to preserve these beneficial attributes in dyed fabrics. Antibacterial action and UV protection are the main medicinal properties of the selective natural dyes. Despite the potential of natural dyes as healthier alternatives for the textile industry, there are noticeable gaps in the literature, particularly concerning weld and woad dyes. Therefore, it is imperative to cultivate interdisciplinary research on the potential synergies between chromatic and medicinal properties, as well as the aesthetic aspects of textiles dyed with natural dyes. This review facilitates the exploration and optimization of the use of new materials and colors through more sustainable approaches, thereby empowering designers to make enlightened decisions about incorporating natural colorants with medicinal properties into their work. By addressing these knowledge gaps and improving dyeing processes, natural dyes can emerge as a viable and health-conscious alternatives for the textile industry aligning with the principles of sustainability and well-being concerns.

Data availability statement

Data included in article/supp. material/referenced in article.

CRediT authorship contribution statement

Diana Santiago: Investigation, Writing – original draft. **Joana Cunha:** Supervision, Writing – review & editing. **Isabel Cabral:** Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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