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Towards sustainable *Rubia tinctorum L.* dyeing of woven fabric: How life cycle assessment can contribute

Tove Agnhage [a, b, c, d, *], Anne Perwuelz [a, b], Nemeshwaree Behary [a, b]

[a] ENSAIT, GEMTEX, F-59100, Roubaix, France
[b] Univ Lille Nord de France, F-59000, Lille, France
[c] Univ of Borås, Swedish School of Textiles, S-501 90, Borås, Sweden
[d] Soochow Univ, College of Textile and Clothing Engineering, Suzhou, Jiangsu, 215006, China

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ABSTRACT

Bio-based dyes for textile dyeing have been widely studied on account of their environmentally friendly approach but in order to be considered as an eco-friendly dyeing concept, additional parameters need to be considered. The purpose of this study was to improve a textile (polyester) dyeing process with madder dye (*Rubia tinctorum L.*), from an eco-sustainable point of view. A life cycle assessment (LCA) has been performed to determine the environmental impacts associated with the dyeing process, at research lab-scale. The identified hotspots were: the solvent and energy use for madder dye extraction and the liquor:fabric ratio in the dyeing phase. Results showed that reduced impacts from both hotspots were needed in order to perform the best in all impact categories studied. Indeed, decreased solvent and energy consumption by ultrasound-assisted dye extraction reduced the global warming potential, photochemical ozone creation potential and air acidification, while minimized water consumption in the after-wash of the dyed fabric was a promising option for improvement in water depletion and eutrophication.

1. Introduction

The main environmental issues of concern, associated with the textile production chain, arise from the use of energy, water and chemicals (Allwood et al., 2006; Greenpeace, 2011) and the European Science Foundation's COST Action 628 (Nieminen et al., 2007) has concluded that new emerging cleaner technologies are essential in order to reduce environmental burdens of textile processes. The COST Action also suggests the use of life cycle assessment methodology, in order to focus the development of new technologies properly.

Ozturk et al. (2016a) presents sustainable textile production through cleaner production options, i.e. pollution prevention, such as optimization and minimization of water, energy and chemical consumption and chemical substitution. Chemical substitution involves replacing chemicals having high polluting effect or toxic properties with alternatives having less impact on the environment (Ferrero et al., 2013) and in this context, natural dyes have attracted attention on account of their environmentally friendly approach and additional functional properties. A recent paper (Zhou et al., 2015) presents simultaneous dyeing and functionalization (UV protection ability, antimicrobial and antioxidant activity) of silk with natural dyes. More on natural dyes as antimicrobial agents for textiles can be found in the review by Kasiri and Safapour (2014). Furthermore, a review by Shahid-ul-Islam et al. (2013) highlights textile applications of environmentally friendly plant-based products.

Natural dyes are commonly considered as eco-friendly because these are obtained from renewable resources as compared to synthetic dyes, which are derived from nonrenewable resources. However, the production and use of bio-based dyes are not free from environmental concerns. Accordingly, dyes derived from threatened species or mordant dyes that contain heavy metals as an integral part of the dye molecule are prohibited in the latest (4.0) version of Global Organic Textiles Standards (GOTS), (2014). Further details on sustainability issues related to the production and use of natural dyes, including environmental and social aspects as well as cost considerations, are given by Saxena and Raja (2014).

* Corresponding author. Ecole Nationale Supérieure des Arts et Industrie Textiles (ENSAIT), Laboratoire de Génie et Matériaux Textiles (GEMTEX), 2 Allée Louise et Victor Champier, BP 30329 59056 Roubaix, France.
E-mail address: tove.agnhage@ensait.fr (T. Agnhage).

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The tinctorial plant madder (*Rubia tinctorum L.*) is one of the oldest dye plants used throughout history. The plant is native in Southern and Southeast Europe, in the Mediterranean area, Asia Minor and in the Caucasus and nowadays the madder also grows in China and Japan, among other countries (Derksen, 2001). Madder is recognized as a promising dye plant for large scale cultivation under European soil and climate conditions (Biertümpfel and Wurl, 2009).

The coloring species, which are found in the root of the plant, are grouped together in the Color Index as C.I. Natural Red 8. Alizarin (1,2-dihydroxyanthraquinone: C.I. Mordant Red 11) is the main dye of the root dye extract. Some of the other dyes present in the roots are purpurin, xantho-purpurin, rubiadin, pseudo-purpurin, munjistin and lucidin, Fig. 1. (Cuoco, 2012; Drivas et al., 2011). The lucidin compound may raise toxicity issues, but its content in the madder dye extract is reduced by using appropriate extraction method (Derksen et al., 2003).

Some papers deal with improved dyeing process with madder, with respect to reduced use of water, energy and chemicals (Barani and Maleki, 2011; Guzel and Akgerman, 2000; Montazer et al., 2007; Shams-Nateri, 2011). However, environmental impact assessment of the production and use of madder for textile dyeing has, to our knowledge, not been performed.

Foulet et al. (2015) & Pasquet et al. (2014) showed that life cycle assessment (LCA) is a useful tool to assess process impacts at research stage. The LCA methodology evaluates the overall environmental consequences of a product, process or human activity by quantifying the energy and materials used (inputs), the wastes and emissions released to environment (outputs) and the environmental impacts of those inputs and outputs (Jiménez-González et al., 2000). The broad scope of LCA helps to avoid a narrow view of environmental concerns and reduces the risk for burden shifting (shifting the environmental problem from one impact category to another or one life cycle phase to another) (Roos et al., 2015).

Parisi et al. (2015) performed a life cycle assessment to evaluate the environmental impacts associated with a new dyeing process in comparison to a classical dyeing process. Other LCA studies deal with spin-dyeing versus conventional dyeing (Terinte et al., 2014) as well as pad-dyeing technology (Yuan et al., 2013). Some papers evaluate environmental improvements not through LCA but via comparison of specific consumptions of resources and waste water generation (Long et al., 2014; Ozturk et al., 2016b; Xu et al., 2016). Haddar et al. (2014) evaluated the environmental performance, in terms of polluting effect, of dyeing with natural colorants by measuring the polyphenol concentration, chemical oxygen demand (COD) and biological oxygen demand (BOD).

This study, differently from others, explores LCA driven process optimization of bio-based dyeing of textiles. Specifically, the LCA has a gate-to-gate perspective on dyeing of polyester fabric with madder dye. Noteworthy, in previous paper (Agnhage et al., 2016) we dealt with technical aspects related to madder dyeing of polyester fabric and the process was optimized with respect to quality issues such as durability. This study then focuses on environmental impact assessment of the total dyeing process, which includes madder dye production, to identify hotspots and optimize the process with respect to environmental sustainability.

2. Methods

2.1. Goal and scope definition

A life cycle assessment (LCA) has been performed as a gate-to-gate analysis focused on the main system units composing dyeing of polyester fabric with madder dye at lab-scale, with the goal to improve the environmental sustainability of the dyeing process. The study was carried out according to ISO 14040 (ISO, 2006a) and ISO 14044 (ISO, 2006b) standards. EIME software (Bureau Veritas, CODDE) was used.

2.1.1. Functional unit and geographical scope

The functional unit (FU) chosen was dyeing of 1 kg woven polyester fabric (110 g/m^2) with 3% owf madder dye. The color and the durability (see Section 4.2.2.) remain the same in the different scenarios and correspond to optimized dyeing conditions based on Agnhage et al. (2016).

The geographical scope was France. French electricity mix was used, which is predominantly nuclear (78.5%).

2.1.2. System boundary and allocation

A process-flow diagram of the system studied, i.e. the 'total dyeing process', is shown in Fig. 2. Upstream processes included

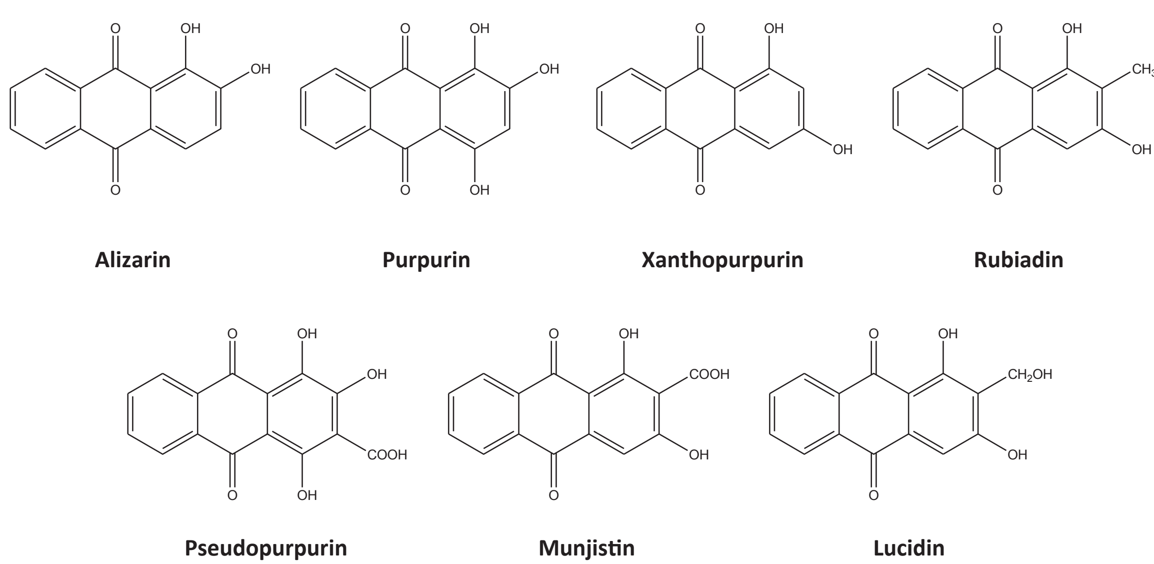


Fig. 1. Structures of anthraquinone aglycones found in madder roots.

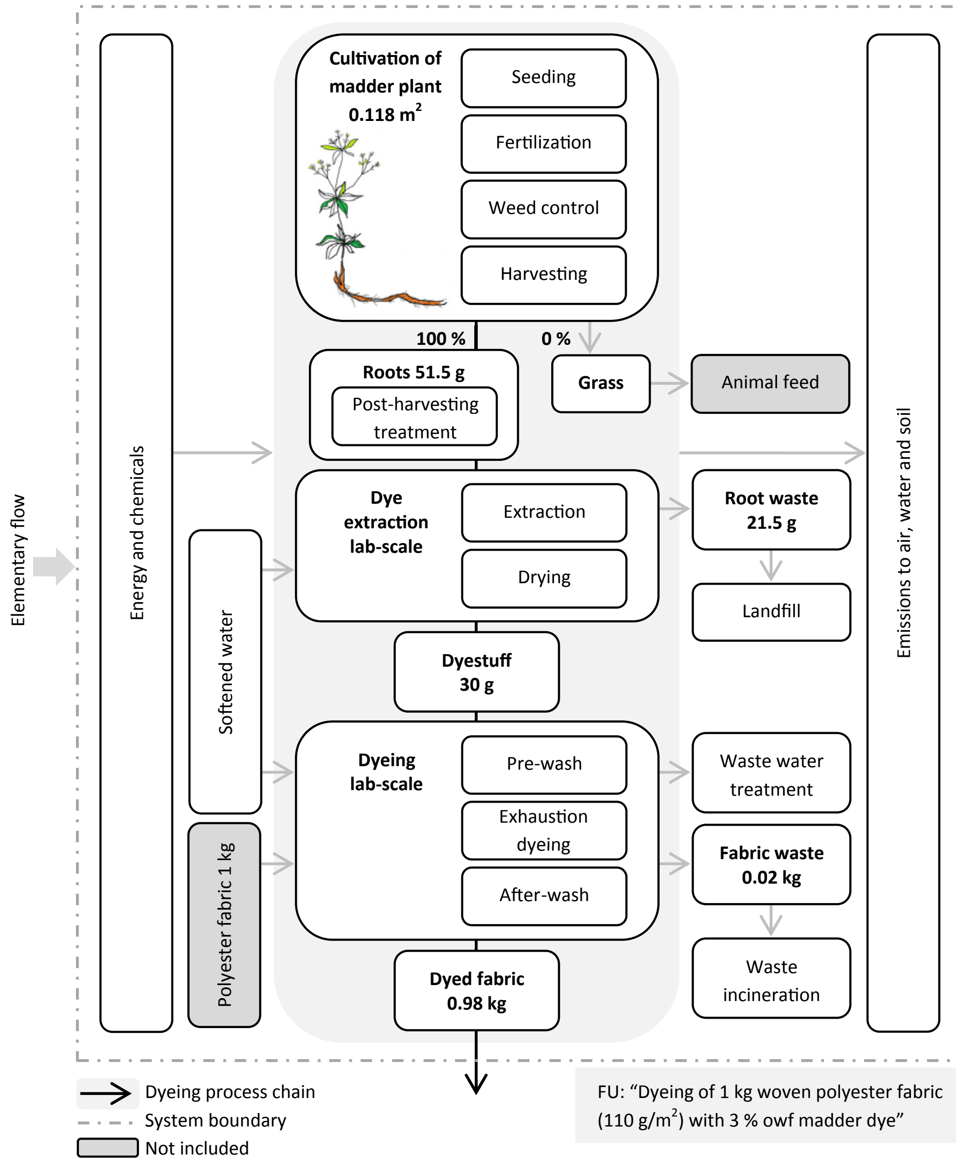


Fig. 2. Process-flow diagram of the system studied. Grey boxes were not considered in the study.

inputs such as water softening treatment, energy generation and chemical production necessary for dyeing 1 kg of polyester. Downstream outputs included wastes and emissions into air, water and soil. Transportation of chemicals and maintenance of machines were not considered.

No allocation was considered because a 'worst-case' scenario was preferred. Thus, 100% of the loadings for cultivation of madder plant were allocated to the roots, since the root part was the main product for dye extraction.

2.2. *Life cycle inventory (LCI) and modeling*

Life cycle inventory calculations were performed using EIME software, Bureau Veritas CODDE (France), with included databases such as ELCD, EcoInvent and CODDE. The databases within the EIME software were the source of the background data upon which materials (energy, softened water, acetic acid, fertilizers, etc.) and processes (waste water treatment, fertilization, etc.) were modeled.

Inventory activity data (see Table 1) were acquired from different sources. Description of the dyeing process chain and data sources are given in the following sections, starting from cultivation of madder plant (Section 2.2.1.) and ending with the dyeing phase (Section 2.2.4.).

2.2.1. *Cultivation of madder plant*

2.2.1.1. Seeding and duration of vegetation. It is preferred to harvest madder after three years of cultivation, because the yield (of both roots and colorants) increases significantly between the first and the third year (Biertümpfel and Wurl, 2009; Cardon, 2007). So, the present study considered a three-year cultivation period. Background data for seeding and harvesting were obtained from EIME database.

Based on Biertümpfel and Wurl (2009), crop rotation was considered. Furthermore, it was assumed that crop rotation had been used for more than 20 years and that no land use change would apply in this case (PE International, 2015). In order to include

Table 1
LCI for the dyeing process reference scenario.

		Provider	Total		
			Agriculture	Dye extraction	Dyeing phase
Materials and location	Soft water (L) RER	CODDE	–	2.57E-01	7.47 E+01
	Electricity mix (MJ/kg) FR	ELCD	–	5.30 E+00	4.34 E+01
	CAN (kg) RER	CODDE	5.67E-03	–	–
	DAP (kg) RER	CODDE	1.41E-03	–	–
	MOP (kg) RER	CODDE	5.31E-03	–	–
	Methanol (kg) RER	CODDE	–	9.12E-02	–
	Acetic acid (kg) RER	CODDE	–	–	8.54E-03
	Detergent (kg) RER	CODDE	–	–	2.67E-01
Processes and location	Seeding (m^2) RER	CODDE	1.18E-01	–	–
	Fertilization (m^2) RER	CODDE	3.55E-01	–	–
	Weed control (m^2) RER	CODDE	1.18E-01	–	–
	Harvesting (m^2) RER	CODDE	1.18E-01	–	–
	Evaporation (kg) RER	CODDE	2.06E-01	1.27 E+00	–
	Waste water treatment (kg) RER	ELCD	–	8.03E-02	1.50 E+01
	Landfill (kg) RER	ELCD	–	2.15E-02	–
	Textile waste incineration (kg) RER[a]	ELCD	–	–	2.00E-02

[a] Energy recovery has been taken into account.

crop rotation effects, a more detailed, site-dependent assessment is necessary.

Whether or not to account for biogenic carbon storage in bio-based products is the subject of ongoing debates (Pawelzik et al., 2013). CO_2 capture from the atmosphere during madder plant growth may be considered as negative emission. Nevertheless, as the current study modeled a 'worst-case' scenario, biogenic carbon was excluded from the inventory analysis.

2.2.1.2. Fertilization and weed control. The root yield not only depends on duration of vegetation but also on the amount of nutrients in the soil. Figures, discussed by Chenciner (2000), show that fertilized rich soil increased the root yield from 1.87 (for unfertilized and dry soil) to 4.35 ton/ha (France). If fertilization is carried out, the yearly amount of N, P and K should be 120–160, 40 and 150 kg/ha, respectively (Biertümpfel and Wurl, 2009).

Our study considered a maximum yearly amount of nutrients (for all three years of vegetation). The dominating N, P and K fertilizers in Europe were modeled namely CAN, DAP and MOP (Hasler et al., 2015) using background data from EIME.

Weed control is important during the first vegetation year, and was therefore taken into account in the study.

Irrigation was not included in the modeling though it may be carried out during the first vegetation year. Therefore, the influence of irrigation was assessed in sensitivity analysis (Section 4.2.4.) (background data from EIME software).

2.2.1.3. Harvesting and root yield. After harvesting of madder, two products are obtained, madder roots and 'grass' (weight ratio root:grass approximately 10:15). The grass can be fed directly to animals or dried to hay (Chenciner, 2000).

The study considered a root yield of 4.35 ton dry roots/ha (France), based on maximum amount of applied fertilizers (see Section 2.2.1.2.). The influence of root yield on the impact assessment results was then studied in sensitivity analysis, with focus on cultivation in France. It should be noted that the results cannot be directly extrapolated to other production regions because the amount of roots/ha varies from one country to another, for example around 3 ton/ha in Germany and 8 ton/ha in Italy (Angelini et al., 1997; Biertümpfel and Wurl, 2009; Cardon, 2007; Chenciner, 2000; Saxena and Raja, 2014).

2.2.2. Post-harvesting treatment

The classical post-harvesting treatment of madder roots consists of two main processes, namely drying and stamping of the roots into powder (Derksen, 2001). Low drying temperature is recommended (40 °C) (Biertümpfel and Wurl, 2009). Under favorable climates the roots may be dried in open air (Cardon, 2007).

The present study considered manually stamping of the roots. Drying was modeled as low temperature grain drying through evaporation.

2.2.3. Dye extraction

The tinctorial plant madder contains coloring species but also other non-coloring plant constituents, starch among others. The dyes must therefore be extracted from the plant material, i.e. separation of the coloring species from the non-coloring ones, in order to prepare dyes of high purity.

Based on information from the supplier of madder dye (Section 2.2.4.1.), the present study considered a hydro-alcohol extraction method. Activity data for the flows originated from madder dye extraction at lab-scale (Cuoco, 2012; Cuoco et al., 2009). Additionally, the modeling was validated by communication with experts in the field (see acknowledgments).

2.2.3.1. Conventional extraction. Based on Cuoco et al. (2009), a LCI for conventional reflux extraction (1 h, methanol/water 80/20 v/v) was created, which considered a dye yield of 58.3%. The inventory was supplemented with energy consumption data for applying reflux extraction (1 h), measured using a power meter (Chauvin Arnoux C.A. 8332B) at our research laboratory. Note that only the working time of 1 h was considered, the energy required to heat the apparatus was not taken into account. The measurements were carried out with 150 mL methanol/water (80/20 v/v). Accordingly, the LCI included 257.3 mL of softened water, 5.3 MJ of electrical energy and 115.3 mL of consumed methanol, for the production of 30 g of dyes and 21.5 g of root waste.

2.2.3.2. Ultrasound-assisted extraction. Compared to conventional extraction, ultrasound-assisted extraction is considered as a cleaner technology due to the reduction of temperature and time which results in lower consumption of energy (Shahid-ul-Islam et al., 2013). Moreover, from Cuoco et al. (2009), the solvent consumption is reduced while at the same time the dye yield is increased. Optimized conditions for ultrasound extraction were considered in this study (18 min, 36 °C, methanol/water 37/63 v/v) with a dye yield of 64.3%. The LCI data included 734.8 mL of softened water, 2.4 MJ of electrical energy and 47.6 mL of consumed methanol, for the production of 30 g of dyes and 16.7 g of root waste.

2.2.3.3. Drying of dyestuff — solvent evaporation. A drying (evaporation) step was included in the process chain in order to convert the dye extracts into powder form. By including drying, reuse of the methanol solvent was taken into account.

For methanol reuse, it was assumed that 90% of the methanol used in the dye extraction solution could be recovered through evaporation/condensation, while all the solvent absorbed by the root waste were lost. The amount of absorbed methanol/water mixture in the root waste was assumed to be 80 wt %.

On these bases, the inventory included evaporation of 1.27 L (of methanol/water 80/20 v/v) and 1.15 L (of methanol/water 37/63 v/v) for the conventional and ultrasound method, respectively.

2.2.4. The dyeing phase

Data acquisition for the dyeing phase was based on foreground information, i.e. data that was newly compiled for the purpose of this study, from laboratory experiments. The data collected within the laboratory regarded each step of the dyeing phase by considering the energy and water consumption, and the quantities of chemicals used. The energy consumption for the exhaustion dyeing and the after-wash was measured using a power meter (Section 2.2.3.1.) whereas the energy consumption for the pre-wash was modeled based on technical document (Pesnel, 2014).

2.2.4.1. Materials. A polyester (polyethylene terephthalate) plain woven fabric of density 110 g/m^2, with 60 warp threads/cm and 34 weft threads/cm, was dyed with GOTS and REACH certified madder dye (*Rubia tinctorum L.*) (C03), used as received from Couleurs de plantes (France). Hydro-alcohol extraction had been used to obtain the dye.

2.2.4.2. Pre-wash. A light scouring ‘pre-wash’ was carried out to prepare the fabric for dyeing, using a conventional wash at 60 °C. The LCI data included 9.7 L of softened water, 0.69 MJ of electrical energy and 16.7 g of detergent, for scouring of 1 kg polyester fabric.

2.2.4.3. Exhaustion dyeing. The experimental set-up for the dyeing was based on optimized conditions previously described by our research group (Agnhage et al., 2016), namely using a dyebath pH of 5 (in presence if aqueous acetic acid) at 130 °C for 45 min. A liquor:fabric ratio (LR) of 15:1, in 200 mL beakers in a laboratory-scale Labomat dyeing machine (Switzerland), was used. Accordingly, the LCA was modeled to include 15 L of softened water, 22.14 MJ of electrical energy and 8.54 g of acetic acid.

2.2.4.4. After-wash. Based on previously cited (Agnhage et al., 2016), initially an after-wash was carried out using the same conditions as for ISO 105:C 10 standard wash fastness test (40 °C, 30 min, LR 50:1, 5 g/L standard ECE detergent). With these conditions, the LCI included 50 L of softened water, 20.6 MJ of electrical energy and 250 g of detergent.

In order to reduce the environmental load from the after-wash, the present paper explored scenarios with lower LR (15:1) and without detergent.

2.2.4.5. Color measurements. Spectral reflectance factors (taken between 400 and 700 nm wavelengths) of the dyed fabric were measured using a Datacolor Spectraflash SF600 reflectance spectrophotometer (Datacolor International) as described in Agnhage et al. (2016). K/S values (representing the color strength) were calculated from reflectance factors R by the software using the Kubelka-Munk Equation (1) (Haddar et al., 2014).

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

2.3. Life cycle impact assessment (LCIA)

Ten impact categories were assessed: Air Acidification (AA)[AE], Air Toxicity (AT)[m^3], Freshwater Ecotoxicity (FWE)[CTUe], Global Warming Potential (GWP)[kg CO_2 eq.], Ozone Depletion Potential (ODP)[kg CFC-11 eq.], Photochemical Ozone Creation Potential (POCP)[kg NMVOC eq.], Raw Material Depletion (RMD)[person reserve], Terrestrial Ecotoxicity (TE)[kg 1,4 − DB eq.], Water Depletion (WD)[dm^3] and Water Eutrophication (WE) [kg PO_4^{3-} eq.]. The potential environmental impacts were expressed at midpoint level.

3. Presentation of the scenarios studied

Firstly a characterization analysis of the reference scenario was performed and hotspots were identified. Subsequently, three additional scenarios were chosen in order to assess opportunities for environmental improvements of the process. The different scenarios were:

Scenario 1 Reference case
Scenario 2 Ultrasound-assisted extraction of madder dye
Scenario 3 After-wash with decreased LR and reduced use of detergent.
Scenario 4 A combination of scenario 2 and scenario 3.

3.1. Scenario 1 (S1)

The first scenario presents the reference case. The reference considered a land area for cultivation of madder plant (0.118 m^2) which corresponded to the amount of dry roots required (51.5 g) to produce 30 g of dyes through conventional reflux extraction. 30 g presents the amount of dyes necessary per functional unit.

The pre-wash (Section 2.2.4.2.) and exhaustion dyeing (Section 2.2.4.3.) were carried out as described in previous sections. The after-wash applied was the same as the one used for standard wash fastness tests (Section 2.2.4.4.).

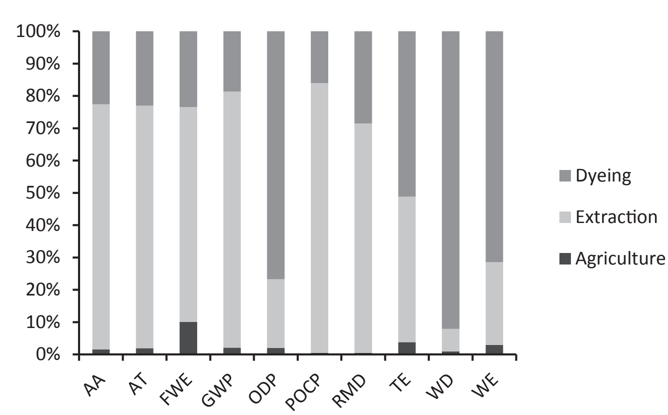


Fig. 3. LCIA results (characterization) associated to the dyeing process reference scenario.

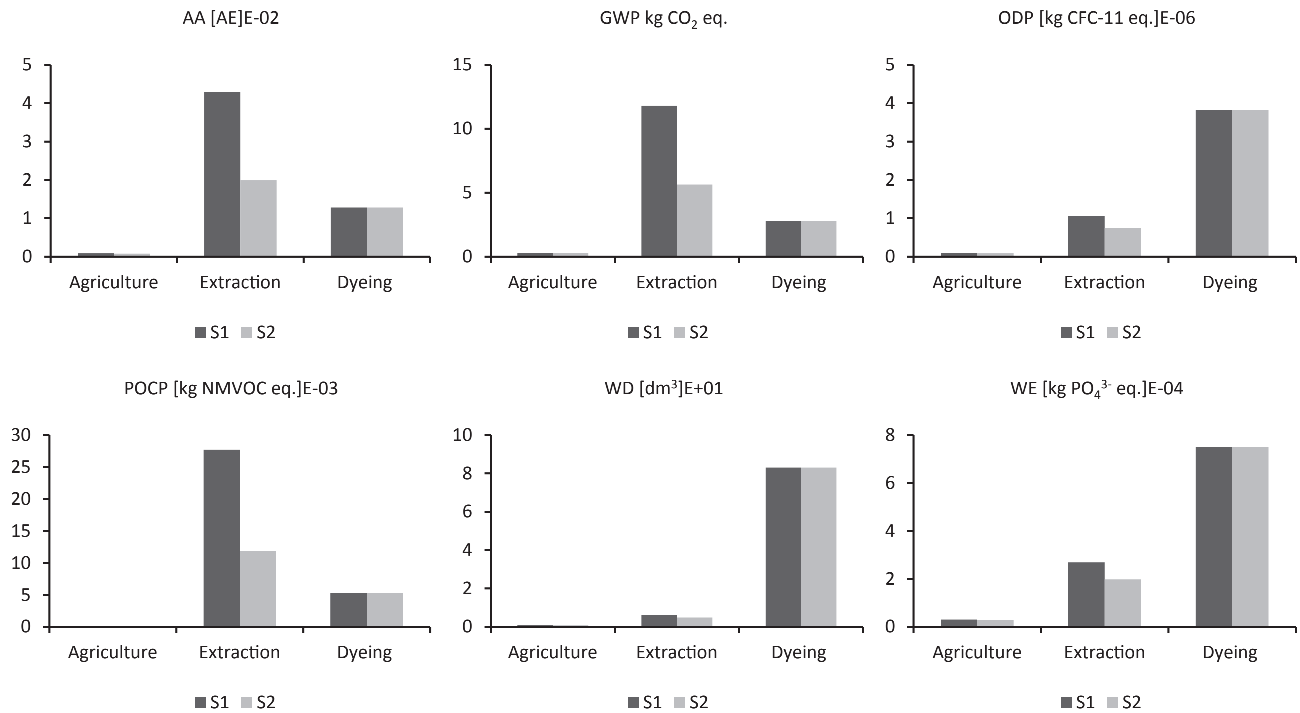


Fig. 4. LCIA results for scenario 1 (S1) and scenario 2 (S2).

3.2. Scenario 2 (S2)

In the second scenario, ultra-sound assisted extraction of madder dye was introduced in the dyeing process. The potential environmental impacts of scenario 2 were assessed and the results compared with the reference scenario.

3.3. Scenario 3 (S3)

The third scenario focused on reduced liquor:fabric ratio (LR) in the after-wash. Scenario 3 (1) considered LR 15:1 maintaining the same concentration of detergent as in the reference case. Moreover, a scenario 3 (2) was created, designed to emphasize the impacts from the use of detergent. For this purpose, S3 (2) excluded detergent in the after-wash, maintaining the same LR as in S3 (1).

3.4. Scenario 4 (S4)

The fourth scenario considered a dyeing process with both scenario 2 and scenario 3 (2).

4. Results and discussion

4.1. Comparison between phases – S1

Contributions from the three different life cycle phases: agriculture (seeding to post-harvesting treatment), dye extraction (extraction and drying) and the dyeing phase (pre-wash to after-wash) to the environmental profile of the total dyeing process are shown in Fig. 3.

The results pointed out that, for dyeing of 1 kg woven polyester fabric with 3% owf madder dye, the dye extraction phase and the dyeing phase play major roles in environmental burden of the process. The sum of these two steps contributed to more than 85% on each impact category.

It can be seen that (Fig. 3), depending on the impact category there was a dominance of one of the two. Namely, the dyeing phase showed the highest contributions for the ODP, TE, WD and WE categories (ranging from 51% to 92%), while the extraction phase was responsible mainly for the contributions to the other 6 impact categories (ranging from 66% to 84%).

The major role of the extraction phase on the overall profile was mainly due to the inputs of energy and methanol (production of the consumed amount and solvent evaporation for reuse), whereas the large role of the dyeing phase was related to the inputs of energy and water. The corresponding LCI is shown in Table 1.

In this context, the present work chose to focus on ways to reduce energy and solvent consumption in dye extraction (scenario 2) and reduce energy and water use in the dyeing phase (scenario 3).

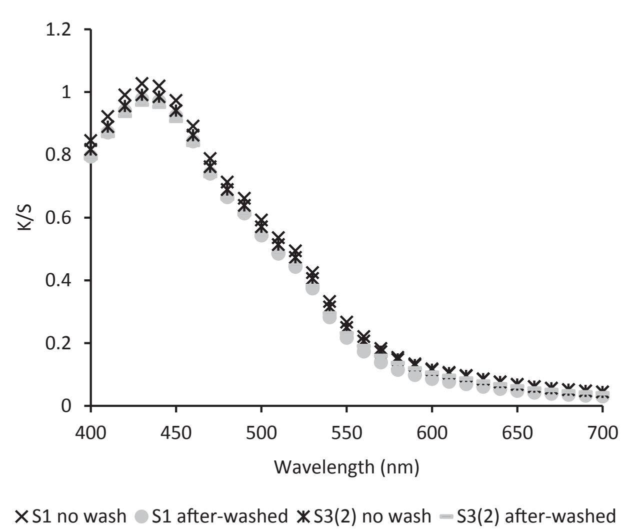
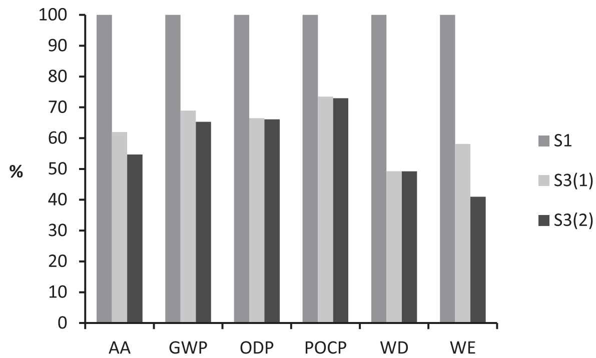


Fig. 5. Effect of after-wash on color yield (K/S).



Note. Absolute values for S1: AA=1.28E-02 AE, GWP=2.78E+00 kg CO_2 eq., ODP=3.82E-06 kg CFC-11 eq., POCP=5.31E-03 kg NMVOC eq., WD=8.30E+01 dm³ and WE=7.50E-04 kg PO_4^{3-} eq.

Fig. 6. LCIA results for after-wash scenario 3 (1) and 3 (2) relative to the reference scenario (S1) dyeing phase.

4.2. Comparison between four scenarios

4.2.1. Ultrasound-assisted extraction

The environmental impacts of scenario 2 and scenario 1 dyeing process were assessed through all the 10 impact categories, whereof results associated to 6 of them are presented in Fig. 4

The impacts from the dyeing phase were independent of any change in the extraction phase. So, the environmental burdens for the dyeing phase were the same for S2 and S1 and there were no reduced impacts (Fig. 4).

The agriculture phase on the other hand was influenced by the extraction method. As the dye yield was higher with ultrasound, a lower quantity of roots were needed and consequently less land area for the production of 30 g madder dyes (0.107 m^2 compared to 0.118 m^2). From Fig. 4, however, the major improvements with respect to reduced environmental impacts were in the extraction phase.

Namely, when the extraction phase of S2 was compared to that of S1, S2 showed lower contribution on each of the impact categories. Clearly, scenario 2 was in favor for AA, GWP and POCP, due to the higher extraction yield and lower consumption of solvent and energy. Evidently, ultrasound-assisted extraction would improve the environmental sustainability of the dyeing process.

4.2.2. Liquor:fabric ratio (LR) and detergent use

In previous work (Agnhage et al., 2016) the after-wash was improved from an eco-point of view by replacing reduction clearing (which uses sodium dithionite and alkali) with a standardized wash for wash fastness tests (ISO 105:C 10) and by doing so, reduce the use of potentially toxic chemicals. Importantly, the standardized wash considered a very high LR (50:1). Lower LR after-wash was therefore explored, namely the same one used as in dyeing (15:1). Additionally, reduced amount of detergent was investigated.

The color yield (K/S) of the dyed fabrics was measured before and after the after-wash. From the different conditions studied, neither LR nor detergent quantity influenced the color performance, Fig. 5. So, the color and color's durability of the dyed fabric remained the same. From Fig. 5 it is also clear that excellent wash-fastness was obtained, since the K/S was almost the same before and after wash.

LCA was conducted for the two after-wash scenarios, 3 (1) and 3 (2). In Fig. 6, the LCIA results, for 6 of 10 impact categories, are presented as relative values to the reference scenario dyeing phase. As seen (Fig. 6), the scenarios with lower LR improved in all 6 impact categories. Their contribution ranged from 73.5% for POCP to 41.0% for WE. Scenario 3 (1) showed lowest impact in WD (49.3%). The two after-was scenarios improved equally much in WD, but scenario 3 (2) presented the lowest impact in WE, due to the fact that no detergent was used. Clearly, implementation of water consumption minimization through reduced liquor:fabric ratio in the after-wash decreased environmental burdens of the dyeing phase.

4.2.3. Optimized scenario

By referring to Section 4.1., solvent and energy minimization in the extraction phase and reduced water consumption in the dyeing phase might be good starting points in order to improve the environmental sustainability of the dyeing process studied. Though, as seen in Fig. 7, scenario 4 clearly presents lowest impacts overall, corroborated by data reported in Table 2. S4 considered the combination of S2 and S3 (2) and was, from the different conditions studied, the best alternative with respect to reduced environmental impacts without altering the final color performance of the dyed fabric.

4.2.4. Sensitivity analyses

The reference scenario considered madder cultivation in France with a maximum amount of nutrients applied, a dry root yield of

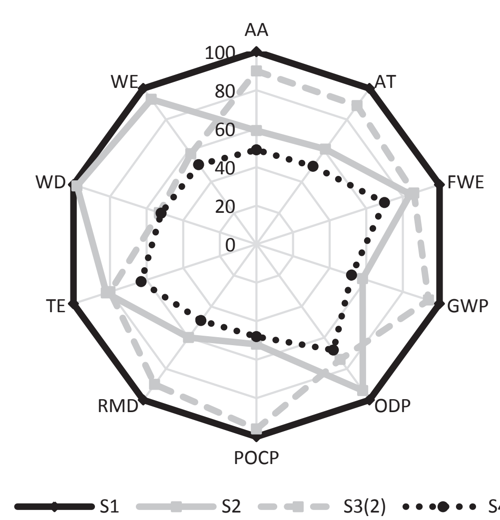


Fig. 7. Comparison of the four scenarios. S1 = Reference; S2 = extraction scenario; S3 (2) = after-wash scenario; S4 = both S2 and S3 (2).

Table 2
Impact assessment results for dyeing process scenarios.

Impact category	Unit	S1	S2	S3 (2)	S4
Air acidification (AA)	AE	5.65E-02	3.34E-02	5.07E-02	2.77E-02
Air toxicity (AT)	m^3	1.90E+06	1.15E+06	1.70E+06	9.48E+05
Freshwater ecotoxicity (FWE)	CTUe	5.74E-01	4.85E-01	4.93E-01	4.04E-01
Global warming potential (GWP)	kg CO_2 eq.	1.49E+01	8.71E+00	1.39E+01	7.74E+00
Ozone depletion potential (ODP)	kg CFC-11 eq.	4.98E-06	4.66E-06	3.69E-06	3.37E-06
Photochemical ozone creation potential (POCP)	kg NMVOC eq.	3.32E-02	1.73E-02	3.17E-02	1.59E-02
Raw material depletion (RMD)	Person reserve	1.76E-03	1.05E-03	1.58E-03	8.67E-04
Terrestrial ecotoxicity (TE)	kg 1,4-DB eq.	2.27E-04	1.87E-04	1.83E-04	1.42E-04
Water depletion (WD)	dm^3	9.02E+01	8.87E+01	4.80E+01	4.65E+01
Water eutrophication (WE)	kg PO_4^{3-} eq.	1.05E-03	9.76E-04	6.08E-04	5.33E-04

4.35 ton/ha, no irrigation of water in cultivation and a methanol reuse of 90% from the dye extraction solution. In order to test possible alternatives to the reference scenario, three sensitivity analyses were performed.

Firstly, the influence of fertilization was tested. Without the application of fertilizers, a lower root yield was considered (1.87 ton/ha). When unfertilized soil was compared to the reference scenario cultivation phase, for the production of the same amount of roots, unfertilized soil with a lower root yield clearly reduced environmental impacts (Supplementary data A). However, when the total dyeing process was considered, the reduced environmental impact corresponded to a reduction less than 1% on each of the impact categories, as seen from Table 3.

Secondly, a sensitivity analysis was carried out on irrigation in madder cultivation. When the cultivation phase from seeding to harvest was considered, it was obvious that irrigation during the first vegetation year increased environmental burden in all ten impact categories (Supplementary data A). The increased environmental impact due to irrigation however, corresponded to an increase less than 1% on each impact category when the entire process chain was taken into account (Table 3).

The third sensitivity analysis was made on methanol reuse in the extraction phase. A reuse of 95% was hypothesized (5% consumption) and compared to the reference scenario. The LCI was revised accordingly and LCIA results are presented in Table 3 and in supplementary data A. Indeed, better dye extraction method with respect to efficient solvent reuse would decrease environmental impacts.

4.3. General discussion

In this part, data quality (representativeness) will be communicated, based on three quality indicators: temporal, geographical and further technological correlation (Huijbregts et al., 2001) (see also Table 4). The indicator ‘temporal correlation’ represents the time correlation between the year of the study and the year of the obtained data. The indicator ‘geographical correlation’ expresses the degree of accordance between the defined area, according to the goal of the study, and the area covered by the obtained data. The indicator ‘technological correlation’ concerns all aspects of correlation other than the temporal and geographical, such as how well the inventory data represent specific processes or materials under study (Weidema and Wesnaes, 1996). It should be emphasized that the scores in Table 4 serve as identification numbers only and should be seen as a representation of a subjective judgment of the data quality (Weidema, 1998). The lower the score number is the better is the data quality.

The uncertainties in this study are expected to occur mainly due to lack of representative data for the agriculture phase. Both the temporal and technological correlation were low (5 and 4, respectively). Here it must be stressed that, it was beyond the scope of this study to perform a full case specific assessment of madder cultivation, therefore literature and consulting expertise were considered. Even if crop rotation, for more than two decades, was considered in this study, thus land use change was not accounted for, land use change could be studied more in detail (Schmidt et al., 2015). Besides land use change, biodiversity is an important aspect that recently has received attention and should be addressed in future work (de Baan et al., 2013). This would allow a more comprehensive insight into the environmental impacts of madder cultivation.

For the dye extraction phase, technological representativeness for solvent evaporation was a critical point. Furthermore, it becomes relevant to address the reliability (reliability indicator not included in Table 4). Namely, in this study the reuse of solvent was a qualified estimate. For future work it will be necessary to verify the data on solvent reuse through measurements.

Indeed, natural dyes need to be extracted from the plant material, possibly through methods that are not entirely eco-friendly. Research priorities should therefore be set to explore cleaner extraction technologies and close LCI data gaps for natural dye extraction. This will open the way to enhance the accuracy of the environmental impact assessment and allow for more robust and credible decision support towards sustainable use of bio-based dyes for textiles.

Finally, it must be pointed out that, assessment of environmental impacts using LCA tool plays a key role when striving towards sustainable bio-based dyeing of textiles and environmental burdens from all three life cycle phases: agriculture, dye extraction and dyeing need to be considered. Otherwise one phase may offset environmental gains achieved through improvements in the other two. Due to the multidisciplinary nature of the field, for future work, close collaboration between relevant research disciplines is encouraged so as to improve the data quality and only then can we truly begin to talk about LCA's contribution towards sustainable bio-based dyeing of textiles.

Table 3
Results from sensitivity analyses (% of the dyeing process reference scenario).

	AA	AT	FWE	GWP	ODP	POCP	RMD	TE	WD	WE
1.87 ton root/ha, without nutrients	99.70	99.90	99.04	99.66	99.98	>99.99	99.97	99.79	99.64	99.68
Irrigation first vegetation year	100.05	100.07	100.34	100.05	100.01	100.02	100.02	100.20	100.11	100.00
5% consumption of methanol	70.69	71.91	92.91	70.31	99.95	65.07	71.26	90.41	99.83	96.32

Table 4
Pedigree matrix with three data quality indicators, according to Weidema (1998).

	Data quality indicator	Score	Explanation
Agriculture	Temporal correlation	5	Age of data more than 15 years of difference
	Geographical correlation	1	Data from the area under study (France)
	Further technological correlation	4	Data on related processess or materials but from same technology
Extraction	Temporal correlation	2	Data cover 6 years difference
	Geographical correlation	2	European average data
	Further technological correlation	1; 3	Data from processes and materials under study (dye extraction); data from materials under study but different technology (solvent evaporation)
Dyeing	Temporal correlation	1	Data cover the year of study
	Geographical correlation	2	European average data
	Further technological correlation	1	Data from processes and materials under study

5. Conclusions and perspectives

This study provides a first attempt to quantify environmental impacts of the production of madder dye and shows LCA driven optimization, of a lab-scale dyeing process, so as to dye polyester fabric with madder dye in a more environmentally friendly way.

In view of current data availability, the initial gate-to-gate LCA (from seeding of madder plant to madder dyed polyester fabric) identified two hotspots for the optimization of the dyeing process:

- Reduced solvent and energy use for extraction of madder dye
- Consideration of the water consumption in the dyeing phase

The environmental impact assessment results showed that improvement in both hotspots was needed in order to perform the best in all impact categories studied.

Importance of the work resides in the approach to contextualize the application of life cycle assessment to the textile sector dealing with the natural dyeing and that it shows how gate-to-gate LCA can help to focus future research and developments in this area correctly. It is envisaged that studies like the present one, will be increasingly important for eco-optimized use of bio-based dyes for textile dyeing.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jclepro.2016.09.183.

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