International Journal of Engineering, Sciences and Technologies
ISSN : 3079-2045
https://www.ij-est.org/
Volume 2, issue 2, https://doi.org/10.70961/GPVU8045

Dye plants of Madagascar, color potential to replace artificial textile dyes

Tsiza Faratiana Andriamihaja[a], Mahery Andriamanantena[b], Edenafanambinantsoa Soamaro Evah Andrianjato[a], Tsanta Antenaina Ratsimbazafy[b], Carys Schutzbach[c], Panja Ramanoelina[b], Achim Ecker[c] , Fanjaniaina Razafimbelo Fawbush[b]

[a]Qualité et Valorisation des Ressources Industrielles, Agricoles and alimentaires (QVRIAA), Ecole doctorale «Génie des Procédés et des Systèmes Industriels, Agricoles and alimentaires», MG-101, Antananarivo, Madagascar
[b]Mention Industries Agricoles and alimentaires (IAA-ESSA), Université d'Antananarivo, MG-101, Antananarivo, Madagascar

[c]Center of Industrial Chemistry and processes, Zurich University of Applied Sciences (ZHAW), Zurich - Switzerland)

Corresponding author : Tsiza Faratiana Andriamihaja

Adress: Ecole Doctorale Génie des Procédés et des Systèmes Industriels, Agricoles and alimentaires, 101 Antananarivo, Madagascar
e-mail : tsizaf@gmail.com
Phone number : +261 34 21 323 32

Abstract

The textile industry is one of the most polluting in the world, mainly due to wastewater discharges from dyeing with petrochemical dyes. In recent years, there has been a trend towards the use of natural fibers and dyes, and a move towards the bioeconomy. Madagascar is home to some one hundred dye plants that also deserve to be valorized in this field. The aim of this study is to assess the potential of a number of Madagascar plants for dyeing natural fibers such as wool and cotton, using different types of mordants. The study will focus on experiments carried out with four different species with potential sources of different colors, including *Psiadia altissima, Eugenia jambos, Passiflora subpeltata* and *Aphloia theiformis*. The extracts obtained by aqueous extraction were tested with five types of mordants: alum, FeSO4, CuSO4, tartaric acid and ash. Dye fastness was assessed by means of wash fastness tests. A color range of blue, green and yellow was obtained. Wash fastness varies according to plant and mordant. Good fastness was observed for cotton dyed directly with *Eugenia jambos* and wool mordanted with CuSO4. More studies on improving fastness are needed, but this new knowledge of Madagascar plants could be a starting point for their application in the textile industry.

Key words: dye plants, fastness, washing, natural dye, textile, Madagascar, natural fiber

1. Introduction

The dyeing of yarns and fabrics with synthetic dyes is responsible for around 20% of global pollution of drinking water, and poses risks to human health [1, 2, 3]. The textile industry has thus become one of the world's most polluting industries, accounting for over 5% of global greenhouse gas emissions [4, 5, 6]. In recent years, there has been a clear trend towards the use of natural fibers and natural dyes in the textile industry [7], and European authorities are pushing in the direction of a bio-economy and circular economy [8,9]. However, industrial dyeing is still based on fossil resources, with a few exceptions, especially on an industrial scale [10,11]. To complement these efforts, more research into available plant resources and their performance is therefore essential.

Madagascar, with its rich biodiversity, is home to at least 128 dye plants, which may be endemic, indigenous or introduced. Available data on the inventory, use and properties of these dye plants is still very limited [12]. As far as the application of these findings to the textile industry in Madagascar is concerned, scientific research is also still very recent. There was research on *Indigofera arrecta* in 2008 [13], followed by wool dyeing *with Lawsonia inermis* L. and *Cassia obovata* Collad in 2022 [14]. In 2023, the characterization of a number of Madagascar dye plant extracts demonstrated their antioxidant and biological properties, as well as their low toxicity [15]. These previous studies highlight the textile color potential of Madagascar's dye plants. Many plants have yet to be studied, including those in the highlands, where dye plants and traditional knowledge are rapidly disappearing [12, 16]. There is also a lack of assessment of color fastness, which is a prerequisite for determining dye quality.

It is therefore interesting to evaluate the color potential of a few plant resources from the Highlands of Madagascar with different mordants on a cellulosic fiber such as cotton and a protein fiber such as wool. The aim is to identify plants frequently used in traditional Malagasy dyeing, which are adaptable and have rare potential colors such as green, or basic colors of interest for textiles. The study also aims to assess color fastness to washing, which is one of the primary criteria for characterizing fabric dyeing [17].

2. Methodology

1.1. Study area

This research was conducted in three (03) regions of the Central Highlands of Madagascar highlighted by the University of Antananarivo and IFPECO in 2017. These regions are Analamanga, Itasy and Amoron'i Mania (**Figure 1**).

1.2. Ethnobotanical survey and plant selection

In order to identify and conserve Malagasy know-how, surveys were carried out among artisan dyers. To complete this information, a bibliographical search on plants and their characteristics was carried out in order to select a few plants for the dyeing tests, according to the criteria set out in Table 1 (**Table 1**).

1.3. Plant collection and preparation

Fresh plant material is collected in each zone, and each plant is then identified by a botanist. The raw materials collected are processed fresh or dried and then ground according to the traditional practices collected for comparison.

1.4.Extraction and dyeing

This involves decoction for 1 hour at 90°C with a 1:10 ratio (plant matter:water) if the plant is used fresh, and a 1:20 ratio if the plant is used directly dried. Cotton and wool fabrics are cut at a rate of 1g per sample tested. Mordanting is carried out simultaneously with dyeing, testing 5 different types of mordant including Alum $KAl(SO_4)2,12H_2O$; iron sulfate $FeSO_4,7H_2O$; copper sulfate $CuSO_4,7H_2O$; tartaric acid and ash.

1.5.Wash fastness

Wash fastness is assessed on a selection of the most interesting plants, in accordance with ISO 105 C06:2010. The fabrics are washed in a soap solution with a concentration of $4g.L^{-1}$ for 30 mn at 40°C, with a ratio of 1:50 (fabric: soapy water). The washed fabrics are then rinsed and dried. The color difference is assessed by calculating the ΔE*ab obtained after 5 washes on a gray scale from 1 to 5 (1: poor, 5: excellent). L*a*b* coordinates are collected before and after washing to evaluate color with a Datacolor colorimeter.

$$\Delta E^*ab = \sqrt{(\Delta L^{*2}) + (\Delta a^{*2}) + (\Delta b^{*2})}$$

With: a*: color intensity in red (+) or in green (-); b*: color intensity in yellow(+) or in blue(-); L*: brightness or lightness (0: black, 100: white)

3. Results

a. Plants surveyed

The natural fibers dyed naturally by the artisan dyers surveyed are raffia and silk. The dyeing process is generally the same, with a decoction lasting 1 to 2 hours. One exception is the dyeing process for *Indigofera spp*. which consists of anaerobic dyeing with oxidation (vat dye). Twenty-two plants are listed and a variety of colors can be obtained, including red, blue, yellow and green (**Table 2**). Approximately 25% of these plants are still largely unexplored in the textile field.

b. Dyeing and fastness tests

Dyeing and stability tests focused on the following four species: *Psiadia altissima, Eugenia jambos, Aphloia theifomis* and *Passiflora subpeltata*.

For dyeing with *Psiadia altissima* (**Table 3**), a color with green (a* negative) is obtained, with a more pronounced green on wool dyed and mordanted with ash. Shades range from green to orange-brown, depending on the mordant. Average wash fastness is 1.6. The best wash fastness is obtained with direct dyeing (Grey scale=3).

For dyeing with *Passiflora subpeltata* (**Table 4**), the shades obtained are shades of yellow depending on the mordant, except for iron sulfate mordanting with a higher red (a*=4.46) which gives orange-brown shade. The most pronounced yellow is found in wool dyed with alum. Average fastness is 1.8. The best wash fastness is obtained with direct dyeing (Grey Scale=3).

For *Eugenia jambos* dyeing (**Table 5**), the hue obtained is a combination of red and yellow (a* and b* positive). A characteristic blue is obtained in the case of cotton dyed with iron sulfate mordant. The color obtained are shades of orange, depending on the mordant. Average wash fastness is 2.1 but good fastness is obtained with direct cotton dyeing and iron sulfate mordanting (Grey scale=4).

For dyeing with *Aphloia theiformis* (**Table 6**), the color obtained is generally a shade of orange (a* and b* positive) but with a more pronounced yellow. The most pronounced yellow is found in wool dyed with alum mordant (a* between 0.03 and 2.03, b* between 28.97 and 32.34). Average fastness is 1.6. The best wash fastness is obtained with iron sulfate mordant stain (Grey scale=2.5).

c. Color difference after drying

The color difference between fabrics dyed with fresh plant and fabrics dyed with dried plant is shown in **Table 7**. An overall average difference is observed.

4. Discussions

a. Blue color

Only one source of blue is known to dyers: the *Indigofera* genus. A number of scientific studies are being carried out on this plant. It is already used worldwide, confirming its potential for natural textile dyeing [18, 19]. Nevertheless, it is a plant that cannot withstand the cold winters of these regions [13] and is becoming rare in these areas. As it is not cultivated and has no specific collection management, its availability is not guaranteed. Cultivation and good collection management outside flowering periods is one solution for its application in Madagascar. The search for other sources of blue is also an alternative. In the literature, plants growing in Madagascar such as *Catharanthus roseus* , *Rocella tinctoria*, *Clotalaria incarna* or *Ravenala madagascariensis* seeds are proposed as other sources of blue, but are still unknown to the artisan dyers surveyed [13, 16, 20]. An in-depth study of these plants and their dyeing potential in the textile field is a step that should be considered to ensure that the blue hue is obtained.

In dyeing tests, *Eugenia jambos* produced a shade of blue on cotton using iron sulfate as a mordant. This shade of blue, approaching black, may be due to the reaction of tannin with iron to form ferrous tannate [21, 22]. The average a* value of -3.47 on iron mordanted cotton remains, however, a small blue value, and the low wash fastness, in contrast to the good fastness results to be enhanced by the tannin-iron complex reported in the silk literature [21, 22], still limits its application as a source of blue. Further studies to accentuate this blue and improve strength are necessary and still in progress, but this plant remains promising.

b. Red and orange color

For red and pink dyeing, the main plant used locally is "Nato" or *Labourdonnaisia madagascariensis* Pierre ex Baill, which is becoming rare according to artisan dyers but is classified as Data Deficient for assessing extinction risk by the IUCN in 2021 [23]. Other potential red dye plants have already been studied in 2023, such as *Paracarphalea kirondron* and *Tectona grandis*. The latter is on the list of plants surveyed and contains tectograndone, a naphthaquinone red dye [15] that may also hold promise for textiles.

In the dye tests carried out, red was not a dominant color. *Aphloia theiformis* can nevertheless offer orange shades with copper sulfate and tartaric acid mordants. The same applies to *Eugenia jambos*, with good strength (rated 4). *Eugenia jambos* leaves contain flavonoids and tannin [24]. It is a plant used by artisan dyers in the regions surveyed, but does

not feature in the list of plants previously identified by Andriamanantena *and al*. in 2019. It is nevertheless an adaptable and non-invasive plant. It can thus be a good source of purplish gray for direct cotton dyeing, or of brown for direct copper mordanting.

c. Green color

Unlike blue, there are several natural sources of green in Madagascar. *Psiadia altissima* is the most widely used to produce green color in raffia and silk. This plant is endemic to Madagascar and its dyeing properties for textiles have not yet been studied. It is an interesting plant because it is adaptable and non-invasive, and can be used as a green manure for the soil [25, 26]. The most pronounced green color was obtained with wool dyed with ash, suggesting that the mineral concentration in the ash favors the green color. The phytochemical study by Kumar *and al* (2009) showed that *Lantana camara* ash is rich in minerals such as calcium and magnesium [27]. Color variation is moderately stable using fresh or dry *Psiadia altissima* leaves. As the green hue is generally given by chlorophyll, we assume that there has been a reconstitution of chlorophyll by the minerals contained in the ash, as it is a tetrapyrol linked to a magnesium ion [28]. Phytochemical research on *Psiadia altissima* leaves has shown that they contain alkaloids, traces of leucoanthocyane and quinone, tannin and polyphenols [29, 30]. It is assumed that these compounds are responsible for the other hues obtained. Although the wash fastness of *Psiadia altissima* green is not high, the final color obtained is still an attractive green that may be of interest to the textile industry, as green is difficult to obtain. Plant sources giving this color directly are rare in the world [21]. Green is generally obtained by combining blue and yellow [16].

d. Yellow color

For yellow, *Curcuma longa* is a plant already renowned as a source of natural pigment with its active ingredient: curcumin (E101) [31]. Nevertheless, there is a risk of competition between the great need for plant materials for textile dyeing and the turmeric food and cosmeceutical industry. Other lesser-used plants include *Aphloia theiformis*, whose main constituent is mangiferin, a xanthonoid dye [20, 32], and *Passiflora subpeltata*, rich in phenolic compounds, tannins and flavonoids [33] are proposed. These plants are other sources to consider, as they are still largely unexplored in this field, and their active compounds are water-soluble.

A yellow peak is obtained with dyeing wool and cotton fabrics simultanous mordanted with Alum with *Passiflora subpeltata*. This peak is even higher than that obtained with *Aphloia theiformis*, traditionally a source of yellow. The presence of flavonoids may account for this

dominant yellow. It is therefore suggested that *Passiflora subpeltata* should be preferred to *Aphloia theiformis* as a source of yellow for cotton and wool.

e. Fastness

Overall wash fastness ranges from 1 to 4, with better fastness with direct dyeing for all plants except *Aphloia theiformis*. Nevertheless, a* and b* values increase and darker colors are obtained. The use of mordants is confirmed as interesting to vary the color and fix more pigments joining the conclusions of Bechtold *and al*. (2007) [21]; Samanta and Agarwal (2009) [34]; Shahid *and al*. (2013) [35] and Rossi *and al*. (2015) [17]. Other strength-enhancing methods are thus to be determined such as pretreatments and other process parameters with regard to these plants to take full advantage of their dyeing potential.

After washing, color changes are observed and the hue becomes lighter. This is the case of the green color of wool dyed with *Psiadia altissima* mordanted with ash, probably due to the change in pH on contact with the more alkaline soap solution. This finding was also reported by Samanta and Agarwal in 2009 [34] on other plants. More studies on dye bath and pH-varying soap solution tests can be carried out to improve initial color and fastness.

5. Conclusion

The scale of pollution caused by synthetic dyes used in textile dyeing is driving the need to find more environmentally-friendly natural substitutes that also meet the demand for color and stability in the textile industry. Madagascar's plants, still little studied in this field, were therefore proposed in view of its rich biodiversity, including dye plants. To this end, an ethnobotanical survey was carried out in 03 regions of Madagascar's Central Highlands, among artisan dyers of raffia and silk. The aim of this survey was to identify and select plants that could be adapted to potentially interesting colors, i.e. those providing basic colors for the textile industry. Dyeing tests with different mordants and a wash fastness test on the selected plants were then carried out to determine the colors obtained from Malagasy dye plants on cotton and wool, and to evaluate these colors.

It was found that Madagascar has a plant source that is already traditionally used in textiles and can offer a range of colors, including primary colors such as blue, yellow and red, as well as colors that are difficult to obtain directly, such as green on silk and raffia. The plants used in the North-West are similar to those used in the Highlands of Madagascar, except for one plant not yet recorded by previous ethnobotanical surveys: *Eugenia jambos*. For cotton and wool, the four plants selected were: *Psiadia altissima, Passiflora subpeltata, Eugenia jambos* and *Aphloia theiformis*. The application of different mordants gives different colors. *Psiadia altissima* is an interesting source of green with ash mordants. *Passiflora subpeltata* is preferred to *Aphloia theiformis* as a source of yellow. *Eugenia jambos* is proposed for its good fastness to obtain grey and brown. As wash fastness is moderate or even low with the application of certain mordants, this result opens the way to further studies to improve the fastness of these colors, which are of interest for their effective application in the textile field. Further research into the biological and toxic properties of these plants, or sustainable cultivation methods to make them available, is also required.

6. Acknowledgements

The authors would like to thank all the artisan dyers who took part in the survey for their hospitality and sharing. We would also like to thank CNARP for drying the plants and Pr Stephan Rakotonandrasana for his botanical support during the collect and the identification. This study is part of the Natural Malagasy Plant Dyes project funded by the Swiss National Science Foundation. The authors would also like to thank them for their funding.

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8. Tables

Table 1 : Selection criteria

Potential color	Part used	Disponibility	Research level
Red	Unused for alimentation	Industrial wastes (agriculture, food or wood industries)	Less research
Blue	Less valorized	Adaptable and not invasive	No research
Yellow			
Green			

Table 2 : List of plants used by dyers

Family	Scientific name	Malagasy vernacular name	Part of the plant used	Mordant	Color obtained	Other use	Research level (*)
Papilionaceae	*Indigofera arrecta, I. tinctoria*	Aika	Fresh leaves	None	Blue, purple, brown	Medicinal plant	+++
Onagraceae	*Ludwigia octovalvis* (Jacq.) P.H. Raven	Volondrano	Fresh leaves	Rusty black clay mud	Black	Medicinal plant	+
Myrtaceae	*Eucalyptus globulus*	Kininina	Dried bark	Rusty black clay mud	Black	Medicinal plant Firewood	
				None	Brown		
Myrtaceae	*Eugenia jambolana* Lam.	Rotra	Fresh bark	None	Brown	Fruit consumed	+++
Sclerodermataceae (Règne Fungi)	*Pisolithus arhizus*	Tainkininina	Whole part	None	Brown, light yellow	Amend ment	
Anacardiaceae	*Mangifera indica*	Manga	Fresh leaves	None	Brown	Fruit consumed	++

Family	Scientific name	Malagasy vernacular name	Part of the plant used	Mordant	Color obtained	Other use	Research level (*)
Myrtaceae	*Psidium guajava* L.	Goavy	Fresh leaves	None	Grey	Medicinal plant Fruit consumed	+
Nymphaeaceae	*Nymphaea stellata*	Voahirana	Fresh leaves	Rusty black clay mud	Grey	Ornemental plant	++
Zingiberaceae	*Curcuma longa*	Tamotamo	Dried rhizome powder	Alum (lighter) or ash (darker)	Yellow	Spice Essential oil	++
Aphloiaceae	*Aphloia theiformis*	Voafotsy	Fresh leaves	None	Yellow	Medicinal plant	+
Hypericaceae	*Harungana madagascariensis*	Harongana	Dried bark	None	Orange	Construction and firewood	+++
Asphodelaceae	*Aloe vaombe*	Vahombe	Fresh leaves	None	Pink	Medicinal plant	+
Verbenaceae	*Tectona grandis*	Kesika	Fresh and dried leaves	None	Pink, red	Medicinal plant Construction	++
Amaryllidaceae	*Agave sisalana*	Taretra	Fresh root	None	Purplish pink	Leaves for rope making	+
Amaryllidaceae	*Allium cepa* L.	Tongolo	Dried peel	None	Pink, yellow	Bulb consumed	++
Sapotaceae	*Labourdonnaisia madagascariensis Pierre ex Baill.*	Nato	Dried bark	None	Red	Dyeing	+

Family	Scientific name	Malagasy vernacular name	Part of the plant used	Mordant	Color obtained	Other use	Research level (*)
Asteraceae	*Psiadia altissima*	Dingadingana	Fresh leaves	Alum (lighter) or ash (darker)	Green	Medicinal plant Essential oil Green fertilizer	+++
Passifloraceae	*Passiflora edulis*	Garana	Fresh leaves	Alum (lighter) or ash (darker)	Green	Medicinal plant Fruit consumed	+
Myrtaceae	*Eugenia jambos*	Zamborozana	Fresh leaves	Alum or ash	Green	Medicinal plant Fruit consumed	+
Passifloraceae	*Passiflora subpeltata*	Garana dia	Fresh leaves	Alum or ash	Green, yellow	Medicinal plant	+++
Moraceae	*Morus alba* L.	Voaroy hazo	Fresh fruit	None	Purple	Medicinal plant	+

(*) Level of study in the textile field: +: not or little studied, ++: moderately studied, +++: much studied.

Table 3 : Characteristics of shades obtained using Psiadia altissima leaves

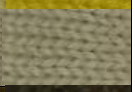
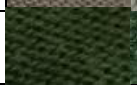
Mordant	Fiber	L*	a*	b*	ΔE	Wash Fastness	Without washing	After 5 washing
Direct	Cotton	77,40	- 1,43	17,00	8,98	2,0		
	Wool	61,73	- 1,87	27,20	3,92	3,0		
Alum	Cotton	79,40	- 1,67	27,33	16,20	1,0		
	Wool	71,37	- 1,47	42,47	26,32	1,0		
$CuSO_4$	Cotton	63,50	2,37	22,27	6,64	2,0		
	Wool	37,80	2,07	23,10	10,96	1,5		
$FeSO_4$	Cotton	42,10	- 2,60	5,50	23,64	1,0		
	Wool	24,00	0,87	1,83	11,20	1,5		
Tartaric acid	Cotton	77,30	0,57	15,83	7,43	2,0		
	Wool	56,73	4,57	27,30	4,20	2,5		
Ash	Cotton	50,59	1,35	10,05	20,10	1,0		
	Wool	46,31	- 5,49	23,62	14,60	1,0		

Table 4 : Characteristics of shades obtained using Passiflora subpeltata leaves

Mordant	Fiber	L*	a*	b*	ΔE	Wash Fastness	Without washing	After washing
Direct	Cotton	78.00	- 0.44	22.23	2.98	3.0		
	Wool	57.72	0.47	37.35	4.91	2.5		
Alum	Cotton	78.27	- 1.42	53.11	25.59	1.0		
	Wool	67.03	1.54	59.00	17.25	1.0		
$CuSO_4$	Cotton	66.96	- 0.30	43.14	9.34	1.5		
	Wool	40.83	2.77	29.57	5.86	2.0		
$FeSO_4$	Cotton	40.45	4.46	15.83	28.35	1.0		
	Wool	31.32	3.14	14.26	5.84	2.0		
Tartaric acid	Cotton	79.42	- 0.95	15.38	5.75	2.5		
	Wool	65.98	0.08	32.94	15.66	1.0		

Table 5 : Characteristics of shades obtained using Eugenia jambos leaves

Mordant	Fiber	L*	a*	b*	ΔE	Wash Fastness	Without washing	After washing
Direct	Cotton	70.37	4.20	11.57	2.03	4.0		
	Wool	56.57	6.87	20.33	14.40	1.0		
Alum	Cotton	72.27	2.07	36.03	13.93	1.0		
	Wool	68.03	0.67	42.47	18.47	1.0		
$CuSO_4$	Cotton	56.40	7.97	26.53	6.49	2.0		
	Wool	33.83	8.37	19.67	1.94	4.0		
$FeSO_4$	Cotton	26.47	0.23	- 3.47	21.74	1.0		
	Wool	22.87	4.10	0.07	7.25	2.0		
Tartaric acid	Cotton	70.00	2.00	15.80	2.74	3.5		
	Wool	48.83	7.53	22.90	12.69	1.0		

Table 6 : Characteristics of shades obtained using Aphloia theiformis leaves

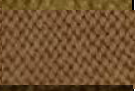
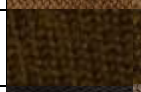
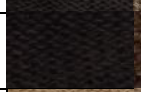
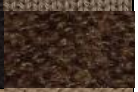
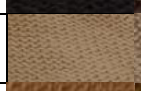
Mordant	Fiber	L*	a*	b*	ΔE	Wash Fastness	Without washing	After 5 washing
Direct	Cotton	74.37	4.18	18.86	7.03	2.0		
	Wool	66.53	4.11	21.22	13.03	1.0		
Alum	Cotton	68.68	0.03	32.34	15.74	1.0		
	Wool	62.66	2.03	28.97	10.58	1.5		
$CuSO_4$	Cotton	50.42	7.52	25.69	7.89	2.0		
	Wool	37.12	4.79	26.61	4.89	2.5		
$FeSO_4$	Cotton	28.82	0.32	5.18	28.91	1.0		
	Wool	28.64	1.27	6.18	8.97	1.5		
Tartaric acid	Cotton	71.86	5.33	20.05	7.87	2.0		
	Wool	58.74	7.75	26.11	18.36	1.0		

Table 7 : Color difference obtained using fresh and dried leaves of Psiadia altissima and Passiflora subpeltata

		Psiadia altissima		***Passiflora subpeltata***	
Mordant	**Fiber**	**ΔE**	**Grey scale**	**ΔE**	**Grey scale**
Direct	Cotton	3.08	3	7.52	2
	Wool	6.73	2	3.90	3
Alum	Cotton	5.34	2.5	14.85	1
	Wool	14.97	1	15.45	1
$CuSO_4$	Cotton	5.05	2.5	12.19	1
	Wool	9.45	1.5	9.56	1.5
$FeSO_4$	Cotton	12.77	1	2.38	3.5
	Wool	8.28	1.5	1.64	4
Tartaric acid	Cotton	6.74	2	13.17	1
	Wool	14.38	1	18.23	1

9. Figure

Figure 1 : Localisation of the study area