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Dialium guineense Willd. *Parkia biglobosa* (Jacq.) R. Br. Ex Benth. and *Tamarindus indica* L.: Review of known and synergetic bioactive compounds

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Abstract
Native from Africa, *Dialium guineense* Willd. *Parkia biglobosa* R. Br. Ex Benth. And *Tamarindus indica L.* grow in tropical regions, and are very widespread in West Africa, particularly in Benin. They are simultaneously used in the traditional healing of infectious diseases but there is a lack on their bioactive compounds description. This work places a special emphasis on inventory of known bioactives compounds from the three plants and their mechanism of action in order to identify the need for further research. It emerges from this work that numerous studies confirm the biological activities of the plants extracts. Nevertheless, few bioactive molecules are described as well as the mode of action of the active extracts.

Keywords: *Dialium guineense* Willd. *Parkia biglobosa* (Jacq.) R. Br ex Benth. *Tamarindus indica* L. pharmacology, bioactive compound

1. Introduction
Because of their diet and medical care, humans are largely dependent on plants (Uusiku *et al.* 2010) [76]. In Africa, for example, more than eighty percent of the population relies exclusively on plants for healing (World Health Organization, 2013) [79]. While it is true that this practice has existed for thousands of years on the whole surface of the globe, it persists in Benin. This mainly because of the satisfaction of the populations resorting to medicinal plants. Therefore, there is a real culture of traditional medicine in this country, the first resort of eighty percent of the population (Mosnier *et al.* 2006) [53]. The low income of the majority of the population limiting access to modern care (Minot and Daniels, 2005) [50], the lack of social security system and scarcity of hospital facilities or medical staff (especially in rural areas) largely contribute to this situation. Nevertheless, the absence of side effects and low toxicity are worldwide arguments in favor of herbal medicine (Cowan, 1999) [18]; (Iwu *et al.* 1999) [36]. In this context, the Ministry of Health of this country, like many others in Africa, has assigned itself the task of training, promoting and integrating traditional healers into its national health system. At the same time, antibiotic resistance has become a public health problem worldwide (Ventola, 2015) [77], so finding new bioactive molecules has become essential. Plants play an important role in this new battle because their secondary metabolites (papaverine, berberine, curcumin, etc.) have already shown interesting activity both in anti-infectious therapy, and against metabolic or cardiovascular pathologies (Nokta *et al.* 1993) [54]; (Hayashi *et al.* 2007) [31]; (Wang *et al.* 2010) [78]; (Kamatou *et al.* 2006) [38].
Our ethnobotanical inquiry in three medicinal plants markets, and with ten traditional healers revealed a recurrence of *Tamarindus indica* L. *Parkia biglobosa* (Jacq.) R. Br. Ex Benth. And Dalium guineense Willd. As well-known and often combined traditional medicines against microbial infections. The available knowledge on these plants was searched using the key words *Dialium guineense* Willd. *Parkia biglobosa* (Jacq.) R. Br. Ex Benth., *and Tamarindus indica L.* in 'Google scholar', 'NCBI', 'Springer Link' and 'Web of Science' databases. We formally identified them at the Benin National herbarium where specimen are deposed under Voucher numbers AA 6727/HNB, AA 6728/HNB and AA 6729/HNB respectively.
Their aerial parts are associated in the healing of infectious diseases. This supposed they could contain synergistic compounds and it appears necessary to make the inventory of their known bioactive molecules in order to support this assumption.

Surprisingly, due to their over-exploitation (timber, coal...), or their difficult regeneration linked to a long dormancy (Faustin *et al*. 2013) [25], we also notify that those three plants appear in the top ten plants threatened with extinction in Benin (Lykke, 2000) [44] (Eyog Matig *et al*. 2001) [24] (Meregini, 2005) [47] (Smagadi, 2005) [67] (Ewedje and Tandjiekpon, 2011) [23]. In order to understand why they are combinated by traditionl healers and to contribute to their safeguarding, this work aims to make an inventory of their known molecules and identify the needs for pharmacological and phytochemical research.

2. Botanic aspects of Fabaceae and known antimicrobials from this family

Dialium guineense Willd. *Parkia biglobosa* (Jacq.) R. Br. Ex Benth. And *Tamarindus indica L.* are three plants of the Fabaceae family. Still known as Leguminosae (vegetables), Fabaceae, by the number of species (about 19,500), and genera (751), constitute one of the most represented families of the subphylum Angiosperms or Magnoliophyta (Group and others, 2013) [29]. They have conquered various habitats with varied morphologies ranging from vines to shrubs or trees. An almost exclusive feature of this family is the fixation of atmospheric nitrogen by symbiosis with soil rhizobia. This family identified and described by Adanson then by Jussieu (Adanson, 1763) [1]; (Jussieu, 1789) [37] was subdivided into three subfamilies on the basis of morphological homology criteria: Caesalpinioideae, Mimosoideae, and Papilionoideae. Advances in molecular biology, including sequencing of ribosomal genes (Soltis *et al*. 1997) [68], chloroplasts genes (Savolainen *et al*. 2000) [66], nuclear or mitochondrial genes (Qiu *et al*. 2005) [61] have allowed evolution of taxonomy to become more precise. Thus, a new classification by the 'Legume Phylogeny Working Group' (LPWG) in 2017 reports six subfamilies. Mimosoideae are now divided into four subfamilies: Duparquetioideae, Cercidioideae, Detarioideae (represented by *Tamarindus indica L.*) and Dialioideae of which *Dialium guineense* Willd. Is part (Azani *et al*. 2017) [12]. The phylogenetic tree of the Fabaceae is now as shown below (Fig. 1).

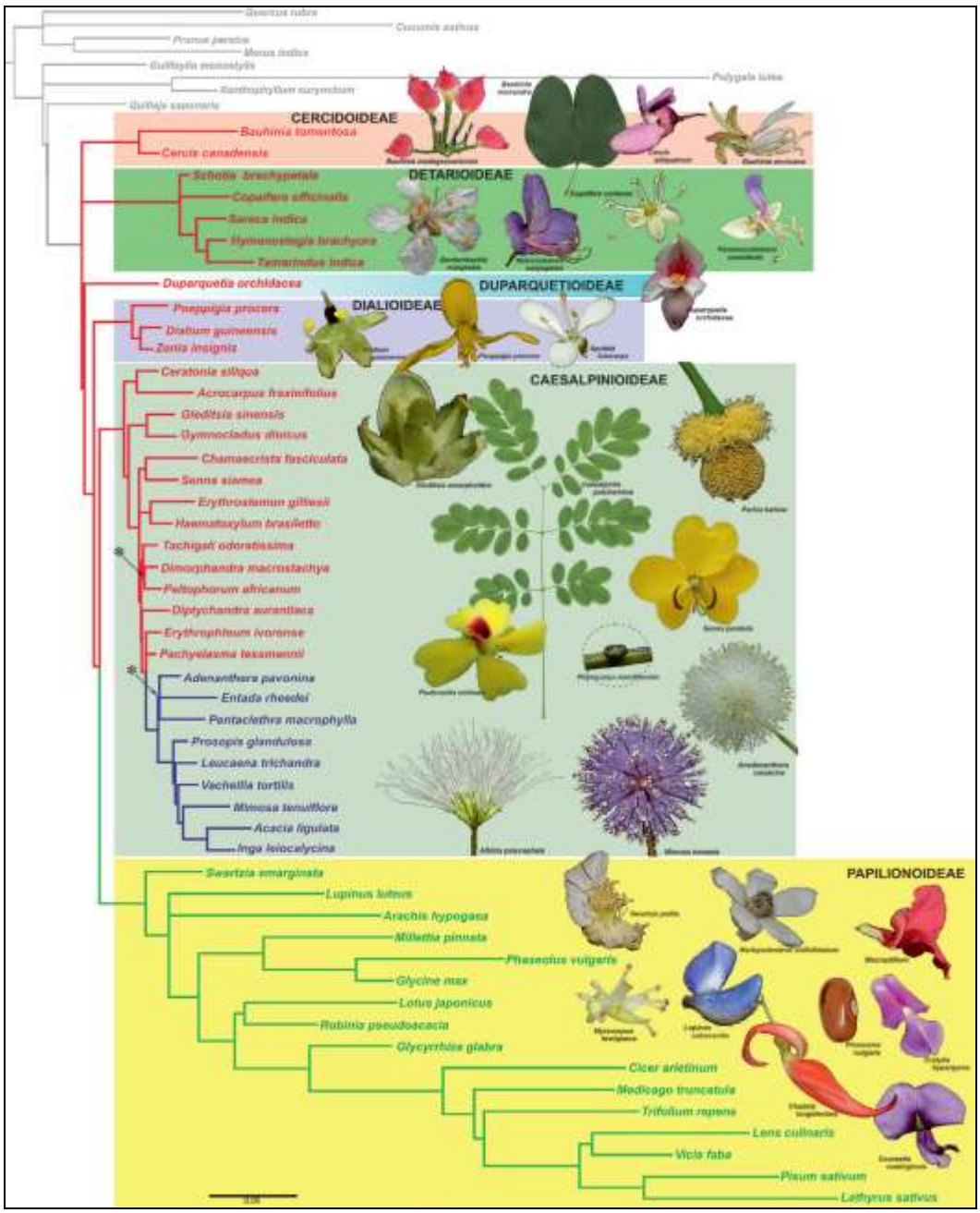

Fig 1: Phylogeny and classification of Fabaceae (Source: LPWG 2017)

Fabaceae are an important source of low-cost protein for many populations in developing countries, as opposed to meat

and fish products (Balogun and Fetuga, 1986) [13]. Their use in traditional medicine is widespread (Rosado-Vallado *et al*. 2000) [64]; (Koné *et al*. 2004) [40]; (Ma *et al*. 2011) [45]; (Rahman and Parvin, 2014) [63]. Furthermore, some antibacterial compounds have already been isolated from this family (Dzoyem *et al*. 2017) [20]. It is then reasonable to expect the same from those three plants.

2.1. Dialium guineense Willd: botanic, uses and bioactive compounds

Dialium guineense Willd. Belongs to the subfamily of Dialioideae. It is a plant from the regions of tropical Africa, but it is exceptionally present in Sao Tome and Principe. It grows on land left fallow and in dry or dense forests, as well as forest galleries, from Senegal to southern Nigeria. It is noteworthy that, in Benin, this plant is present from the coast to the southern region. Its general morphology is illustrated in Figure 2.

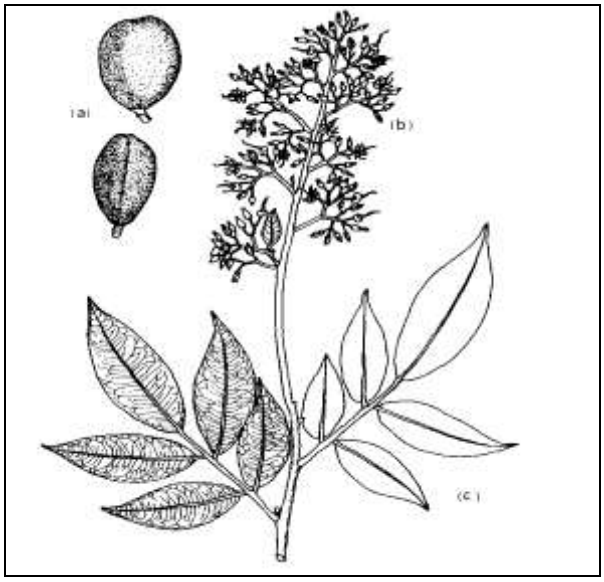

Fig 2: (a) fruit, (b) inflorescence, (c) leaves of *Dialium guineense* Willd. (Drawings by Messrs C.Z. Gbéhou and E.-E. Ewedje, Flore analytique du Bénin).

Roots, leaves and bark of the plant are used in the treatment of malaria, coughs, bronchitis, diarrhea, palpitations, dysmenorrhea, ulcer, anemia, hemorrhoids ("Adjanohoun *et al*. 1989 [4]. Banque de données de médecine traditionnelle et pharmacopée. Paris. 124 p. ISBN: 92-9028-146-4," n.d.) (Odukoya *et al*. 1996) [5] (Bero *et al*. 2009) [16]. They are also used for contraception as well as to regulate menstruation. In Nigeria, women use the leaves against genital infections, and to improve lactation whereas twigs are used as native toothbrushes to protect against tooth decay and dental plaque (Bero *et al*. 2009) [16]; (Okwu and Ekeke, 2003) [56]; (Okwu and Ekeke, 2003) [56]; (Lokonon *et al*. 2013) [43]; (Akinpelu *et al*. 2011) [7].

Rich in nutrients (Arogba *et al*. 1994); (Gnansounou *et al*, 2014); (Ayessou *et al*. 2014), black tamarind (*Dialium guineense* Willd.) fruits are widely consumed in Benin. The tree, by its atmospheric nitrogen fixation, contributes to fertilization of the soil whereas leaves and branches are used in lagoon fish culture as abode and fodder (Ewedje and Tandjiekpon, 2011) [23]. On the other hand, the wood is also used in house or attics constructions, traditional tool handles, and charcoal making or as firewood (Lokonon *et al*. 2013) [43].

Table 1: Summary of known molecules from *Dialium guineense* Willd.

Organs	Extraction Solvent (s)	Biological proven activity	Family/Molecules	Active molecules isolated	Reference (s)
Fruit (pulp)	Water	Molluscicide	Saponins	Glycosides triterpenoids	[30]
	Ethanol	detoxification	Flavonoids, phenolic compounds	N.D.	[77]
Bark (stem or root)	Methanol + Water (maceration)	Antibacterial (*S. aureus*)	Flavonoids, alkaloids, tannins and Saponins	N.D.	[41]
	Ethanol	Antimicrobial	Alkaloids, Flavonoids, tannins and Saponins.	Anthraquinone[a]	[38, 78]
leaves	Hydrodistillation Water (maceration)	Antibacterial (*S. aureus*)	Flavonoids, alkaloids, tannins and Saponins	N.D.	[41]
	Ethanol	Antibacterial (*S. aureus*) Antiviral (Herpes type 1)	Flavonoids, alkaloids, tannins and Saponins	N.D.	[41, 79, 40]
	Methanol/Water	Antimicrobial (vibrio)	Flavonoids, alkaloids, tannins, glycosides cardiac, steroids and Saponins	N.D.	[34]
	Methanol	Antimicrobial/Antioxidant	Compounds phenolic	N.D.	[80]
Seeds	Hydrodistillation	Allatocidal	Precocene I (78.8%) β-caryophyllene (5.3%) Valencene (1.4%) cadalene (1%)	Precocene I[a]	[42]
	Tampon Tris-HCl	hemagglutination	Lectin	Glycoprotein DigL	[43]

N.D = Not determined
a= Structure on Fig. 5 below

Most of work listed in Table 1 above have proven the antimicrobial activity of *Dialium guineense* Willd. Extracts.

However, none of them have isolated the active compound. Concerning anthraquinone found in bark extract (Olajubu *et al*. 2012), this latter is able to form an irreversible complex with nucleophilic amino acids leading to an inactivation of the enzymatic proteins containing these specific amino acids (Stern *et al*. 1996). This property is supposed by the authors to be responsible for the antimicrobial activity, but no experiments were reported to date to support their assumptions. Thus, different teams linked the antimicrobial activity to the presence of chemical entities issued from alkaloids, flavonoids, tannins and saponins ("Agassounon *et al*. 2001. Evaluation des activités cytotoxique, antivirale, antibactérienne et antifongique de six plantes," n.d.)) (Akinpelu *et al*. 2011) [7]; (Orji, 2012) [58] but the exact nature of the active compounds remain unknown. Finally, only two compounds were clearly identified from this plant and out of the claimed antimicrobial domain: precocene I isolated (Essien *et al*. 2007) [22] for its allotocidal effect and a glycoprotein named Dig Lhighlighted (Bari *et al*. 2013) [14] for its hemagglutination effect.

2.2 *Parkia biglobosa* (Jacq.) R. Br. Ex Benth: botanic, uses and bioactive compounds

P. biglobosa (Jacq.) R. Br. Ex Benth. Is a multi-year Mimosoideae (Hopkins, 1983) [33]. It is a tree ten to fifteen meters high with a spreading crown. Globular, the flowers are red or orange (Fig. 3) and bloom from December to January, and from February to March (Akoegninou, 2006) [8]. Concerning its geographical location, this tree is present in tropical Africa between 3° and 15° north.

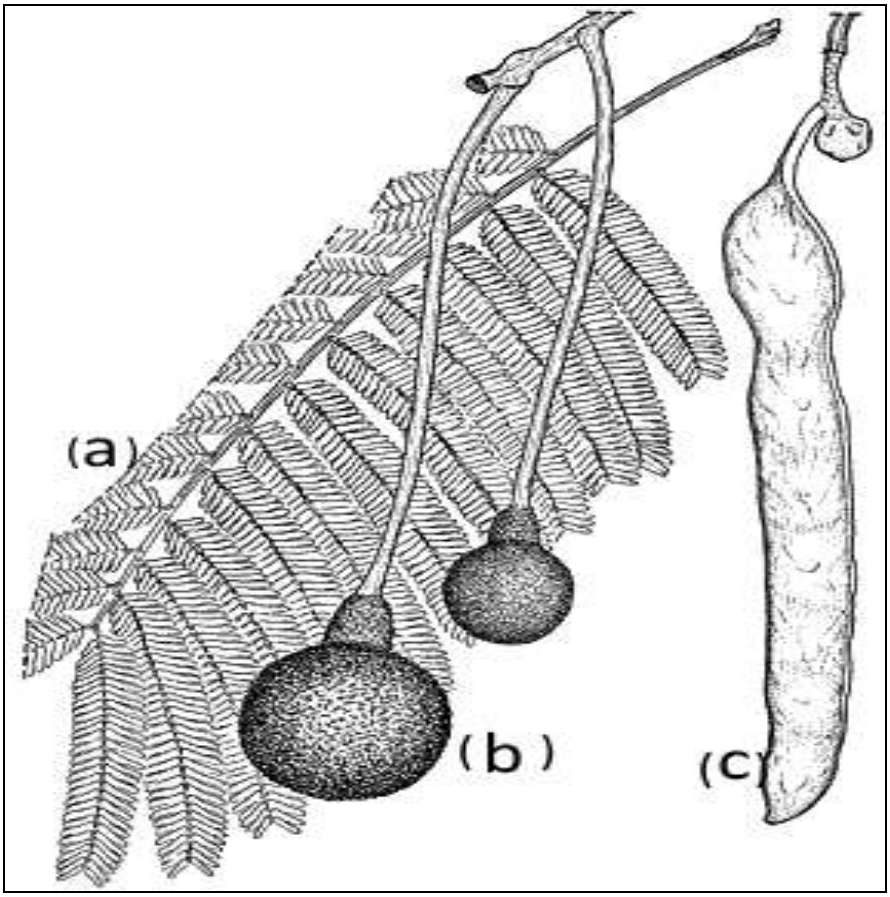

Fig 3: (a) leaves, (b) inflorescence (c) fruit of *Parkia biglobosa* (Jacq.) *R. Br Ex Benth.* (Redrawn and adapted from M.M. Spitteler) / http://database.prota.org/PROTAhtml/Parkia % 20biglobosa_En.htm)

Parkia biglobosa (Jacq.) R. Br. Ex Benth occupies the fifth position among the most used plants in traditional Beninese medicine (Eyog Matig *et al*. 2001) [24]. Its fruits are consumed by humans, and its leaves are used as livestock fodder (Sabiiti and Cobbina, 1992) [65]. The seeds, by fermentation, are used to prepare a very nourishing local mustard used whith different parts of the plant against high blood pressure, hemorrhoids, dermatoses, diabetes, snake venom etc. (Assane *et al*. 1993). For example, the word in Hausa (the language widely spoken in West Africa) for *P. biglobosa* (Jacq.) R. Br. Ex Benth. Is "Dawa Dawa". This word, borrowed from Swahili, refers to the term 'Dawa', used for any plant-based medication ("Swahili-English Dictionary," n.d.).

Like many fruit trees, *P. biglobosa* (Jacq.) R. Br. Ex Benth. Plays an important role in feeding both humans and livestock. It is also used as fuel (Sabiiti and Cobbina, 1992) [65]; (Ræbild *et al*. 2011) [62]. Wild animals, including chimpanzees, feed on them, and disperse seeds (Kunz and Linsenmair, 2007) [42]. The tree is known to farmers to improve soil fertility, and to protect them from erosion (Bayala *et al*. 2007) [15]. Bark and pods are respectively used as dyeing and pottery colorant (Ouédraogo, 1995) [59]. Despite this important role, the regeneration of the species is insufficient (Koura *et al*. 2013) [41].

Table 2: Summary of known molecules from *Parkia biglobosa* (Jacq.) R. Br. Ex Benth.

Organs	Extraction Solvent(s)	Biological proven activity	Family/Molecules	Active molecules isolated	References
Fruit (pulp)	Acetone	Antioxidant	Polyphenols	N.D.	[81]
Bark (root or stem)	Water	Antioxidant Antibacterial Cicatrizing	Polyphenols	N.D. E. E.	[82, 54, 83, 59]
	Ethanol	Antioxidant Antibacterial Cicatrizing	N.D.	N.D.	[56, 59]
	Methanol + Water	Anti- snake venom	N.D.	N.D.	[84]
	Water + ethanol	Antibacterial macrophages stimulation	Sterols, triterpenes, polyphenols Polysaccharides	N.D. Polysaccharides	[54 55, 85]
	Methanol + Dichloromethane	Antioxidant	Proanthocyanins	Procyanidines, prodelphinidines and they glucuronic derivatives	[86]
	Methanol	Hepatoprotective Antibacterial	Polyphenols	N.D. N.D.	[87, 88]
	Methanol + Water	Antioxidant, synergy with antibiotics	Polyphenols	N.D.	[89, 90]
Leaves	Water	Antioxidant Antibacterial Cicatrizing	N.D.	N.D.	[59]
	Butanol	Antidiabetic	-	Lupeol[a]	[57]
	Ethanol	Antioxidant Antibacterial Cicatrizing	N.D.	N.D.	[59]
	Methanol	Neuroprotection	Polyphenols	Catechin[a]	[58]
	Water + ethanol	Vasorelaxation	Procyanidines	N.D.	[91]
Seeds	Water	Antidiabetic, hypoglycemic and antihypertensive	N.D.	N.D.	[92, 93]
	Methanol	Antidiabetic and hypoglycemic	N.D.	N.D.	[92]
	Sulfate ammonium	Antinociceptif Anti-inflammatory	Lectins	Lectin 4MQ0[*]	[94]
	Extraction des proteins	Antioxidant	Protein	E. O.	[95]
	Ethanol/ Ether petroleum	Pesticide	Polyphenols	N.D.	[96]

***** Access code RCSB Protein Databank E.O =Elucidation On going
N.D. = Not determined a= Structure on Fig. 5 below

As summarized in Table 2, molecules identified from *Parkia biglobosa* (Jacq.) R. Br. Ex Benth. Have no relation with its antimicrobial properties assertained by some authors. For instance, (Millogo-Kone *et al.* 2007) [48] (Millogo-Kone *et al*. 2008) [49] (Adetutu *et al.* 2012) [2] concluded that this plant might contain antibacterial molecules but not clearly identified them until now. On the other hand, lupeol isolated from the leaves' butanol extract is known to stimulates β-cell function and increases insulin secretion by inhibiting α-glucosidase and α-amylase in non-competitive and uncompetitive inhibition patterns respectively (Ibrahim *et al*. 2016) [34]. Concerning catechin obtained from leaves methanol extract, this compound is supposed to be involved in mechanisms capable of boosting cellular thiol contents under conditions of oxidative stress (Komolafe *et al*. 2014) [39] (Grønhaug *et al*. 2008) [59].

2.3 *Tamarindus indica* L.

Tamarindus indica L. is a well known Detarioideae native to tropical Africa where it grows wild. Its name (date of India) derives from Arabic (Tamar Hindi) and refers to India from where it was introduced in Arabia. In Woloff (language widely spoken in Senegal), it is called "Dakkhar" and could be origin of the city name ''Dakar'' (Tignokpa *et al.* 1986) [74]. It grows very well in semi-humid or arid regions (Meher *et al*. 2014) [46]. It is a large tree up to 25 meters tall (Tariq *et al*. 2013) [73]. The fruit is a pod containing a seed surrounded by fiber and an acidulous pulp widely used as a condiment (Akoegninou, 2006) [8].

The laxative and carminative effects of *Tamarindus indica L*. fruits are well known. Other organs are used in the treatment of a wide range of pathologies such as toothaches (Tapsoba and Deschamps, 2006) [72], bacterial infections, malaria (Doughari, 2006) [19], snake or insect bites, wound healing (Tignokpa *et al*. 1986) [74], conjunctivitis, diabetes (Buchholz and Melzig, 2016) [17], hypertension, etc. (Havinga *et al*. 2010) [30]. The fruits are also antipyretic (Iwu, 2014) [35], antiscorbutic, hepatoprotective, and used in the treatment of biliary disorder (Morton and Dowling, 1987) [51]. (Tapsoba and Deschamps, 2006) [72] Fruit is both a food (consumed as a drink) and a spice (Hines and Eckman, 1993) [32] (Tsuda *et al*. 1994) [75]. It's subject of intense international trade ("Morton Julia F. 1958 [52] The Tamarind It's food medecinal and industrial uses Florida State Horticultural Society.pdf," n.d.). It is also noteworthy that the tree is useful for the protection of traditional African habitats that are generally fragile typically against wind erosion of soils (Ebifa-Othieno *et al*. 2017) [21].

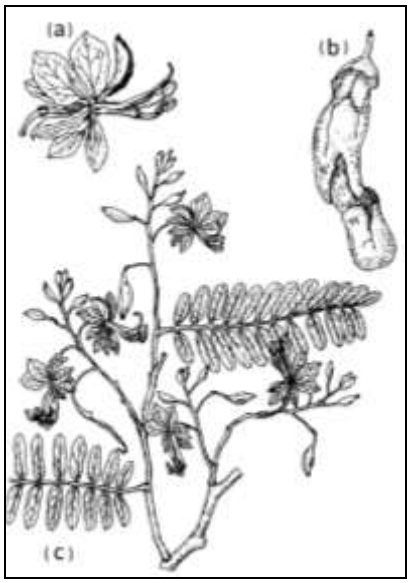

Fig 4: (a) Flower, (b) fruits and (c) leaves of *Tamarindus indica L.* (Illustration: Busson 1965, Flore analytique du Bénin)

Table 3: Summary of known molecules from *Tamarindus indica L.*

Organs	Extraction Solvent(s)	Biological proven activity	Family/Molecules	active molecules isolated	References
Fruit (pulp)	Water	Antibacterial Antioxidant	Flavonoids, Alkaloids, Tannins, Glycosides cyanogenic and Anthraquinones	N.D.	[97, 98, 99, 100]
	Methanol	Antibacterial Anti-inflammatory Lipid-lowering	Polyphenols, sterols, triterpenes, Alkaloids and tannins	N.D.	[101, 102, 103, 104, 100]
	Ethanol	Antidiabetic Antibacterial Hepatoprotective	Flavonoids, Alkaloids, Tannins, Glycosides cyanogenic and Anthraquinones	N.D.	[97, 99, 105, 106]
	Hexane	Antibacterial	Alkaloids and tannins	N.D.	[102]
	Acetone	Antibacterial	Phenols and Flavones	N.D.	[101]
Bark (stem or root)	Water	Antibacterial	Polyphenols, Sesquiterpenes, Alkaloids Phlobatamines Tannins, Glycosides cyanogenic and Anthraquinones	N.D.	[64, 99]
	Methanol	Antibacterial	Polyphenols, Sesquiterpenes, Alkaloids and Phlobatamines	N.D.	[64]
	Acetone	Antibacterial	Polyphenols, Sesquiterpenes, Alkaloids and Phlobatamines	N.D.	[64]
	Ethanol	Antibacterial	Flavonoids, Alkaloids, Tannins, Glycosides cyanogenic and Anthraquinones	N.D.	[99, 107]
Leaves	Water	Antibacterial	Polyphenols, Sesquiterpenes, Alkaloids and Phlobatamines	N.D.	[64, 99, 100]
	Methanol	Antibacterial Antioxidant	Polyphenols, Sesquiterpenes, Alkaloids and Phlobatamines	N.D.	[64, 100, 108]
	Acetone	Antibacterial	Polyphenols, Sesquiterpenes, Alkaloids and Phlobatamines	N.D.	[64]
	Ethanol	Antibacterial	Flavonoids, Alkaloids, Tannins, Glycosides cyanogenic and Anthraquinones	N.D.	[99]
Seeds	Water	Antioxidant Antidiabetic Cicatrizing	Polyphenols Glycosides Sugars	N.D.	[98, 109, 110]
	Ethanol	Growth factor (antibiotic) Antioxidant Cicatrizing	Carbohydrates Compounds phenolic unsaturated fatty acids	Tocopherol[a]	[73, 111, 109]
	Tampon de lyse A	proteinase inhibitor	Protein	Kunitz type proteinase inhibitor	[76, 109]
	Methanol	Cicatrizing	N.D.	N.D.	[109]
	Tampon phosphate saline	Cicatrizing	Alkaloids Tannins Saponins	N.D.	[109]

N.D = Not determined
a=Structure on Fig. 5 below

Table 3 above show that few molecules are well describe from *Tamarindus indica* and have no direct relation whith its antiacterial activity. Alpha tocopherol was isolated from seeds ethanol extract (Aengwanich *et al*. 2009) [5] and is a well-known antioxidant compound ((Gordon, 1990) [27] (Suzuki *et al*. 1993) [70]. A protein isolated from this plant has been identified as a Kunitz type proteinase inhibitor (Patil *et al*. 2009) [60]. The third reported molecule from *Tamarindus indica L.* is tamarindienal or (3*E*)-2, 5-Dioxo-3- hexenal: the bitter principle of the fruits but untit now no interesting biological activity was related to this molecule.

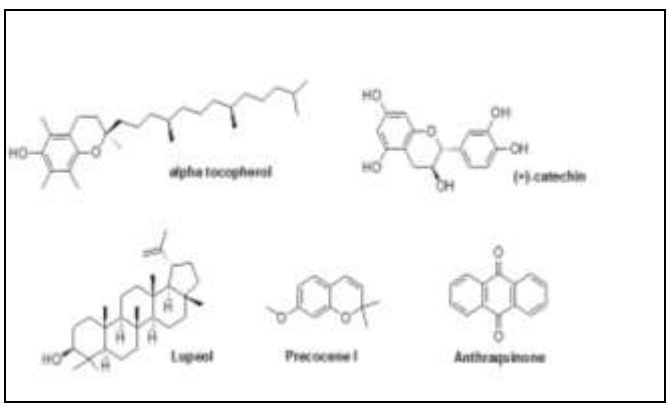

Fig 5: Structures of known molecules from the three plants

Conclusion

Although present work highlights numerous studies on the pharmacological activities of the three plants that have been tested and proven, there is no antimicrobial active molecule described. This lack of knownildge about the bioactive compounds of the three plants don't allow an objective study of their combinated use against infectiuous deseases by Benin traditional healers. We then think it would be interresting to test the extracts together, to select the more efficient combinaison and fractionate it. Antibiotic resistance proving to be a global health priority, this could be a promising way in the search for new antibacterial molecules.

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