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REVIEW ARTICLE

Chemical Constituents and Uses of *Calotropis Procera* and *Calotropis Gigantea* – A Review (Part I – The Plants as Material and Energy Resources)

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Abstract: The traditional and current use of *Calotropis procera* and *C. gigantea*, two soft-wooded, xerophytic shrubs of the family *Apocynaceae*, are reviewed against the background of the plants' chemical constituents and their biological properties. The focus is on the usage of the plants for building materials, natural pesticides, animal feed and bioremediative purposes.

Keywords: Biomaterial, Biomass, Petrocrop, Natural pesticides, Bioremediation, *Calotropis procera*.

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1. INTRODUCTION

Plants growing in arid regions have elicited increased attention, because the hostile environment, in which these plants survive, forces them to develop chemical protective systems rarely found in vegetation of other ecosystems. Furthermore, many of the plants grow in areas, where the dependence on traditional, plant-based medicines over industrially produced pharmaceuticals persists to this day. The two plants, *Calotopris procera* (giant milkweed, also named *C. persica*) and *Calotropis gigantea* (crown flower), have been used widely in traditional medicine in North Africa, the Middle East, and South and South-East Asia. This has led to extensive research on the chemical constituents of the plants. Both plants are known to be sources of cardenolides, and newer research has yielded a number of interesting cancer-active constituents. In addition, extracts of both plants have remarkable nematocidal, molluscidal and insecticidal activities. In many regions, the wood of *Calotropis* plants has been used as a building material and as a source of fuel. In addition, certain parts of the plants have been used as feed for livestock. In other regions, *Calotropis* plants are seen as invasive species that threaten local plant life and that due to their toxicity also pose a threat to grazing field animals. The complexity of the plants' properties and chemical constituents combined with the wide geographic distribution and regional use of *C. procera* and *C. gigantea* has led to a fast-growing body of research on the two plants. For *C. gigantea* alone, approximately 120 research entries have appeared in 2006 (Australian New Crop Website), while in 2018, the database *Web of Knowledge* listed 55 research papers for *C. procera* and 30 research papers for *C. gigantea*. Short reviews on the two plants have appeared previously [1 - 9]. Two more comprehensive reviews have been given on ethnopharmaceutical aspects of *C. gigantea* [10, 11]. The current review provides a comprehensive, up-to-date picture of the uses and chemical constituents of both plants. The review comes in two parts, where the current part focuses on the usage of the plants for building materials, natural pesticides, animal feed and bioremediative purposes.

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1.1. Geographic Distribution and Habitat

Calotropis procera (Ait.) R. Br. and *Calotropis gigantea* R. Br. are two species of soft-wooded, evergreen, perennial shrubs of the family *Apocynaceae*, and subfamily *Asclepiadoideae* (milkweeds). Traditionally, the two plants have developed in two separate regions of the world, one, *C. procera*, predominately in Africa and in the Middle East, the other, *C. gigantea*, in Asia. Due to rapid expansion, often with human help, today they share some of the same habitats in the same regions.

C. gigantea is native to much of South Asia, such as to Iran, Pakistan, Nepal, Sri Lanka and India, and much of South East Asia such as to Malaysia, Myanmar, the Philippines, Indonesia, China, Laos, Thailand, and Vietnam. *C. gigantea* has been introduced to many of the Pacific Islands, including the Hawaiian Islands and New Guinea, whereas in some areas, such as in Palau [12] and on the Solomon Islands [13], it has been brought under cultivation. Also, in northern Australia, it

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has been introduced and cultivated. In the Americas, it has also been brought under cultivation in Barbados [14] and has established itself in Cuba [15] as well as in Brazil, where it is said to be an aggressive invader of the Caatinga ecoregion [16]. With Indian immigrants, *C. gigantea* has also been introduced to Africa, including the Seychelles and Mauritius.

As a native plant, *C. procera* is found in many parts of tropical Africa, including Madagascar and in far south as Angola, in arid zones of Northern Africa and the Middle East. It has spread to South Asia and South-Eastern Asia. It has been introduced into parts of South America [6, 17] and Australia, where it is considered to be an undesirable weed. Also, it now occurs on the Caribbean and Hawaiian Islands, in Central America, South Africa as well as on the Spanish Canary Islands. In the Philippines, it has been reported as a relative new-comer. *C. procera* is relatively salt-tolerant, although the reduced dry mass of the plant is associated with the salinity of the soil [18, 19], and grows well on degraded lands. The rootstock has been reported to be resistant against fires [20] and temporary flooding. Both plants thrive in an arid environment [21], and *C. procera* can be seen at the edge of the desert in the United Arab Emirates (UAE) (see Fig. 1). In the UAE, it has been estimated that on desert land dominated by *C. procera,* about 0.66 tons of soil organic carbon (SOC) per hectar is sequestered annually [22]. In Mauretania, *C. procera* has been used in sand encroachment management, as it was found to be the first species to colonize sand dunes [23]. In the Indian subcontinent, *C. procera* has been seen at up to 1000 m elevation [24]. As the propagation of *Calotropis* plants is aided by human activity [25], it has been used as an indicator of man-induced historical perturbation of the environment [26]. It is also an indication of overgrazing in many areas.

Fig. (1). Photo of a *Calotropis procera* bush in Al Ain, UAE, showing the size to which *C. procera* can grow.

1.2. General Description

C. gigantea can grow to 4 m high, while *C. procera* is usually smaller, although in the Al Ain area in the United Arab Emirates, the plants can also grow up to 4.5 m in height (Fig. **1**). The stem can be up to 25 cm in diameter [27]. The opposite leaves of *C. procera* are large, up to 18 cm and up to 13 cm broad. Leaves of *C. gigantea* are up to 10 cm long and 8 cm wide [28]. *C. procera* has a profusion of five-petaled, sweet-smelling white flowers, 3.8 to 5.1 cm in size, with a marked purple tip. The equally five-petaled flowers of *C. gigantea* are white to lavender in color, and in contrast to those of *C. procera*, without fragrance. The grey-green fruits of *C. procera* are 8 – 12 cm long, containing 350-500 seeds with tufts of white, silky hair. Similarly, *C. gigantea* exhibits single or paired fruits, 7-10 cm long, with a profusion of white tufted, brown seeds, 2.5 – 3.5 cm in length [29]. The seeds of both plants are distributed by wind and water, but also by birds when the ripe pods burst. Flowering takes place all through the year. The plants propagate through suckering as well. Thus, crowns and roots form suckers, where also broken stems can take root and regenerate. Pollination happens through insects, mostly through bees or butterflies.

1.3. Historical Use of the *Calotropis*

The name "Calotropis" stems from Greek with the meaning of "beautiful boat keel". Historically, *Calotropis* has been of economic interest as many parts of the plant are usable. It is a sacred plant to many Hindi, associated with the observances of the maruts (winds), demigods of the rigvedic god Rudra. In the Arabian world, *Calotropis* was associated with sun-worship in ancient times, and also in Vedic times, the leaves of *Calotropis*, in form of arkapattra (sun-leaf) or arkaparna (lightning leaf), have been associated with sun-worship.

The Roman Jewish historian Titus Flavius Josephus (37-c.100) mentioned *C. procera* as the apple of Sodom. As such it is listed in the Mishnah and the Talmud. Abu Hanifa Dinawari (815-896) included *Calotropis* in his Book of Plants [30]. Also, Ibn Sina (980-1037) has made note of the plant. In Renaissance Europe, the plant has been described by Prosper Alpinus (1553 – 1617) (De Plant. Aegpyti, Venet, 1592, Ch. XXV). Prosper Alpinus had studied the plant in Egypt (1580-1584) [31 - 33].

However, human interaction with the plant dates back to much earlier times. Thus, charcoal remains of *C. procera* have been found at archaeological sites associated with pre-dynastic settlements in the Nile Valley of Upper Egypt [34], although *C. procera* was not a predominant source of firewood in early Egypt [35]. Nevertheless, an increase in the abundance of *C. procera* with an increase of human agricultural activity has been noted [34]. Interestingly, in much later times, charcoal derived from *Calotropis* was utilized in gun powder, especially in Indo-China [36].

In some early civilizations, *C. procera* fibers served in textile making, such as in Cyprus in 2000 BC [37]. According to Kramer, the fibers from the inner bark were used in the manufacture of cloth for princes and nobles. Later, *Calotropis* was used to make string and cord in Oman [38], and in Borneo, seed hairs of the plant were made into threads. In India, the fibers have been called the bow-strings of India and have been used for rug and net-making as well as for sewing threads.

Always, the flowers of the plant have served decorative purposes. This continues to this day, even in countries where

Calotropis has been introduced only recently, such as in Hawaii, where the flowers of *C. gigantea* are included in garlands (leis), and in the Philippines, where the flowers are used to decorate rosaries. In India, the maruts (storm deities) have been greeted with a garland of flowers of *Calotropis* on Saturdays for centuries. Nowadays, *C. gigantea* is sold as a plant for home and garden, specifically as a butterfly attractor in a number of countries [39]. The ornamental potential of *C. procera* has been highlighted in a recent article [40].

In general, the *asclepias* (milkweeds), which apart from the *Calotropis* species, include more than 140 species of plants, have long been included in traditional medicine and have been named after the Greek god of healing. The Greek-Arabian traditional medicine knows the *Calotropis* plant under the name of Madar and Ushar, where from ancient times, extracts and powders of different parts of *Calotropis* have been used. Both *C. procera* [under the name of Raktha Arka] and *C. gigantea* [under the name of Arka kalpna and Sweta Arka] have been described in the Aryuvedic classics [41]. *C. procera* was used medically by the ancient Egyptians [42], seemingly going back as far as the Neolithic period [43], and the plant can also be found in the Sudanese traditional medicine [44] as well as in the traditional medicine of North and Central Africa and the Middle East in general and of Central Asia.

Lastly, in Africa, arrow poisons have been derived from the plants. It was this fact that led to the first modern, extensive investigations (Hesse) on the chemical constituents of *C. procera*, carried out in order to identify the chemical structures of the poisonous components, specifically the cardiac glucoside components.

2. CURRENT NON-MEDICINAL USE OF THE PLANT

2.1. Use as a Building Material

The *Calotropis* plant has been used for materials, where, when cultivated at 1 to 1.5 m spacing as done in certain regions in South America and the Caribbean, annual yields of up to 500 kg fibre production per ha can be expected [45]. At 525 trees per ha, Nasser *et al*. have calculated an annual branch yield of even up to 5 tons per ha [46]. The wood is light-weight with a typical air-dried wood density of 0.39 g/cm^3 [46]. The stem of the plant is used for roof-making. The use of the stem fibre for paper, nags and nets has also been reported [47]. It has been found that chemomechanical pulping of *C. procera* leads to a high yield of pulp, suitable for paperboard [48]. Currently, the fibers are still used for rope making [49] in both Africa and South America. Floss (silky hair, akund floss, 2-3 cm long, 12-42 microns wide) from seed stands have been used as stuffing for pillows and mattresses. This use was known in the Acient Egypt and by 1910, *Calotropis procera* was cultivated in Djibouti, producing about 1200 kg seed hair per hectare [31, 33]. Even recently, seed hair from *Calotropi*s has been presented as a potential silk replacer [50]. Seed hair has been tested as a thermal insulating material and has been found comparable to Rockwool mineral fibers in its insulating properties. Tests have also been performed with insulating boards made from seed hair, mixed with either phenol-formaldehyde resin or a cornstarch based binder [51]. Insulating material from a composite of *C. procera* fibres and phenol-formaldehyde resins has been shown to have high water repellency [52]. *C. procera* flax has been forwarded as a binding material for the improvement of acoustic plaster [53]. Lately, the use of fibers from *C. procera* as a reinforcement material for thermoplastic composites was suggested, with polypropylene (PP) as matrix polymer and maleated polypropylene Epolene G-3003 as coupling agent [54]. *C. procera* fibers were tested as reinforcement material in an epoxy matrix, too [55]. Fibers of *C. gigantea* have gained attention as well, with studies on purifying the fibers for mass production [56], and on new bleaching [57] and dyeing techniques [58, 59] to make them commercial products, although dye uptake and dye fixation on the fibres remains a challenge [58]. *C. gigantea* fibers from bark and seeds have been promoted as promising raw materials for fiber-reinforced composites [60 - 63]. Polylactic acid (PLA) and polyester have been used as matrix polymers in combination with *C. gigantea* fibres as reinforcement material [64, 65]. *C. gigantea* fibres exhibit a high degree of tubular hollowness (80–90%). Along the fiber, no natural twist exists. In one study, the crystallinity of *C. gigantea* fibres was found to be 42.5% with a crystallinity orientation index of 85.4% [58]. Bast fibres of *C. gigantea* have been noted to have increased crystallinity upon treatment with 5w% alkali solution, leading to higher tensile strength of the material [66].

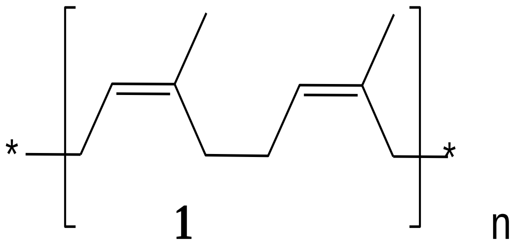

Fig. (2). Polyisoprene (**1**).

During World War 1, rubber for tyres was produced from the *C. procera*'s latex [33]. Although, the composition of the latex of the plant is very complex, it can consist of up to 25-35% of natural rubber [poly(cis-1,4-isoprene, **1**), Fig. **2**] [67]. This polyisoprene has coagulative-like properties that lead to an increased adhesiveness of the latex, *e.g*., when secreted as a response to herbivory [68].

2.2. Use as Adsorbent

The water-resistance and hydrophobicity of *Calotropis* fibers, treated and untreated, lend them the potential to be used as adsorbents for hydrocarbons, such as for oil [69, 70]. Typically, the derivatization of the fibers with hydrophobic alkylsilyl and perfluoroalkylsilyl moieties such as with octadecyltrichlorosilane [71] or with 1*H*,1*H*,2*H*,2H-perfluorooctyltriethoxysilane [70] enhances their oil adsorbent properties. Cao *et al*. fabricated a roughened *Calotropis gigantea* fiber by immobilizing copper and nickel nanoparticles on the original fiber by the impregnation method, using $NaBH_4$ as reductant. The treated fibers were noted to readily adsorb oils from model oil–water mixtures [72]. Excellent and selective oil sorption behavior properties of *C. procera* and *C. gigantea* blended with cotton and polypropylene fibers were observed by Thilagavathi *et al*. [73]. The maximum oil sorption capacity of the developed thermally-bonded nonwovens was 40.2 g/g for high density (HD) oil and 23.0 g/g for diesel oil. Pyrolysis of *C. gigantea* fibers leads to carbon fibers that have a take-up capacity of 130 g/g for model oils in water-oil mixtures [74]. Sheets of *C. gigantea* fibers are also sandwiched between thin polyether sulfone layers leading to a shapable construct which exhibits an oil sorption capacity of 4.3 – 9.3 g oil/ g sorbent [75].

The surface of *C. gigantea* fibers can be functionalized such as with poly(m-phenylenediamine) (**2**) to be used as selective adsorbents. In the case of poly(m-phenylenediamine) functionalized *C. gigantea* fibers, these can be prepared easily by oxidative polymerization of the diamine on the surface of the fibers by immersing the fibers into an aqueous solution of *m*-phenylenediamine in the presence of ammonium persulfate. The functionalized fibers are excellent reductive adsorbents of Cr(VI) [76]. Poly(*m*-phenylenediamine) **2** functionalized *C. gigantea* fibers have also been used for the adsorption of ciprofloxacin (**4**), a fluoroquinolone antibiotic, which is a major pharmaceutical effluent, from wastewater [77]. Treating *C. gigantea* fibers with aq. $NaClO_2$ and doping them subsequently with polydopamine **3** allows for the adsorption of ciprofloxacin (**4**) and norfloxacin (**5**), a further fluoroquinolone antibiotic [78] (Fig. **2**). Dried leaf powder of *C. procera* has been used successfully as adsorbent for the dyes malachite green (**6**) [79] and Congo red (**7**) [80] from aqueous solutions (Fig. **3**).

2.3. Usage as Fuel

Within limits, the wood of *C. procera* is utilized as cooking fuel in some areas [81, 82]. Also, the plant is being studied extensively as a source of biofuel [83] with the gross heat content of the plant estimated at 6.1 kcal/g [84]. Erdman *et al*. give a heat value of the whole plant of 4.2 kcal/g [85], Radhaboy *et al*. put it at 5.2 kcal/g [86]. The heat values of whole plant fractions extracted with benzene, with petroleum ether and with ethyl acetate have been given as 9.6 kcal/g, 13.7 kcal/g, and 7.4 kcal/g, respectively [87]. Both the seeds of *C. procera* and *C. gigantea* are judged as having the potential of providing biodiesel conforming to European and ASTM standards [88] and have a relatively high oil-content (*C. gigantea*: 31%; *C. procera*: 26%) [89]. In all published seed oil analyses [89 - 91] of both *C. procera* and *C. gigantea*, oleic acid (**10**), palmitic acid (**12**), linoleic acid (**9**) and stearic acid (**13**) were the main constituents. Seed oil from *C. procera* of North Eastern Brazil has been shown to have an oil content of 21%, with a range between 19.7% and 24.0% [90], with constituent unsaturated palmitoleic (1.7%, **8**), linoleic (35.3%, **9**), oleic (33.3%, **10**) and elaidic (4.2%, **11**) acids as well as saturated palmitic (15.8%, **12**) and stearic (9.5%, **13**) acids [92, 93], some bound in triglycerides [92]. Seeds of *C. gigantea*, collected in Shwebo District, Myanmar, were equally analyzed to have 33.3 w% oil content with palmitic (15.5%, **12**), linoleic (36.3%, **9**), oleic (30.3%, **10**) and stearic (10.5%, **13**) acids along with palmitoleic (0.3%, **8**), asclepic (0.8%, **14**), linolenic (0.8%, **15**), arachidic (0.6%, **16**), behenic (0.1%, **17**), and lignoceric (0.4%, **18**) acids as minor constituents [91] (Fig. **4**).

The use of dry biomass of *C. procera* and *C. gigantea* to produce biogas has been studied closely since 1992. The fermentation of a suspension of 4% (w/v) of dried leaves of *C. procera* in water at an initial pH of 7.5 has been found to give 2·9 to 3·6 litres of biogas day^{-1} litre^{-1}. The fermentation was found to be fast with 66% of dry material loaded being degraded during the first 2 days of incubation at 30°C with the resulting biogas containing 56–59% (v/v) methane [93]. P. Gourdon *et al*. record 280 mL volatiles /g solid of *C. procera* leaves [94]. Biogas production with a mixture of fresh, chopped leafy biomass of both *C. procera* and *C. gigantea* with cow and buffalo dung was tried, also [95, 96]. This co-digestion led to an increased volume of biogas produced, albeit with a lower methane and higher carbon dioxide content [96].

Also, *C. procera* and *C. gigantea* have been studied as potential petrocrops. Extraction of the plant itself, especially with hexane and heptane, has led to high-yielding hydrocarbon fractions, which have been forwarded as potentially useful chemical feedstock [83, 85, 97 - 99]. Within this context, transgenic *C. gigantea* plants have been grown with alkane contents that were up to 30% higher than in the wild-form of *C. gigantea* [100]. The biocrude thus obtained was subjected to thermal and catalytic cracking to deliver gas and liquid fuels containing mono- and diaromatics, olefins and saturated alkanes [83]. The gas products from the catalytic cracking process were seen to be propene (22.0% – 26.7%), isobutene (8.1w% - 15.0w%) and pentanes/pentenes (22.7w% - 28.9w%) at temperatures between 460 °C and 520 °C [83].

Pyrolysis of *C. procera* derived biomass has been attempted, also. Thus, co-cracking of petroleum vacuum residue with polypropylene and *C. procera* derived biomass has been found to lead to a lowering of the activation energy as compared to cracking petroleum vacuum residue alone [101, 102].

2.4. Animal Feed

In many regions, livestock feeding on Calotropis plants is limited, and toxicity associated with animal feeding on the plants is a constant problem [103 - 105], with sheep fatalities having been reported at 5-10 g of latex feed per kg of bodyweight. Wildlife grazing of the plant is scant [106], but browsing of leaves and flowers by gazelles has been seen, especially in times of drought [107]. The grazing of leaf and flower was observed to some extent also with goats, sheep and cattle in the arid region of Cholistan rangelands in Pakistan [108]. It has been suggested that the toxicity of the plants for animals may depend on biotypes and on the environmental conditions during the plants' growth [109, 110]. It has also been noted that drying may result in the loss of some of the more toxic components such as flavonoid and cardiotonic glycosides of the plant material [111, 112]. Additionally, there have been recent efforts to find safe limits to include the plants in hay and silage as an alternative feed in arid and semi-arid regions [111, 113]. In NW Brazil [114], and to a smaller extent in parts of India [115], silk flower hay (SHF, *C. procera* SW.) has been reported to be suitable for feeding goats, sheep, and camels. In sheep and goat farming, it was noted that substitution of corn and soybean by silk flower hay of up to 30% did not decrease the nutrient uptake by Morada Nova lambs [116] and did not decrease the sensory attributes of their meat [117] or important attributes of the milk of the goats [118].

There has been a study carried out with *C. procera* grown in the southern coastal region of Puerto Rico, combining the use of *C. procera* as a biofuel and the use of the processed residues as animal feed [97]. It was found that the crude protein content of unextracted and extracted *C. procera* was comparable to that of *Euphorbia*, and was found to be of sufficient level for most goat, sheep, and cattle maintenance.

For human consumption, it has been reported that nectar crystals from dried flowers of *C. procera* have been used as a sugar substitute. Thus, in Java, the central part of the flower has been processed into a sweetmeat named Chinese candy [119]. On the other hand, it must be recognized that the nectar can contain poisonous components as well. Cases of fatal poisonings of humans with both *C. procera* and *C. gigantea* [120] are known. In general, the cardenolides found in C. *procera* are toxic to vertebrates.

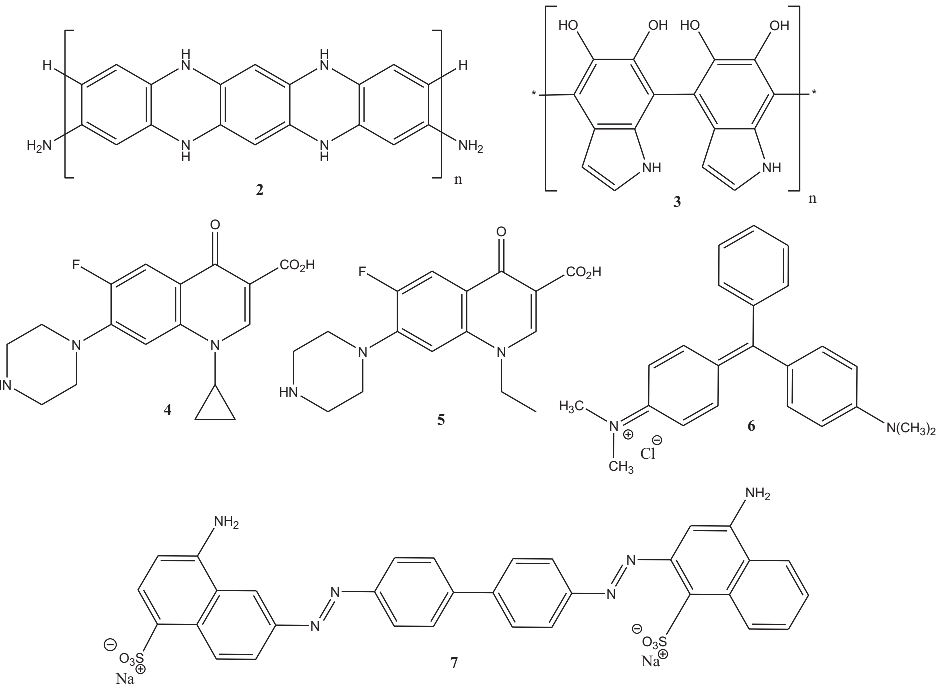

Fig. (3). Dried leaf powder of C. procera has been used successfully as adsorbents for the dyes malachite green (6) and Congo red (7) from aqueous solutions.

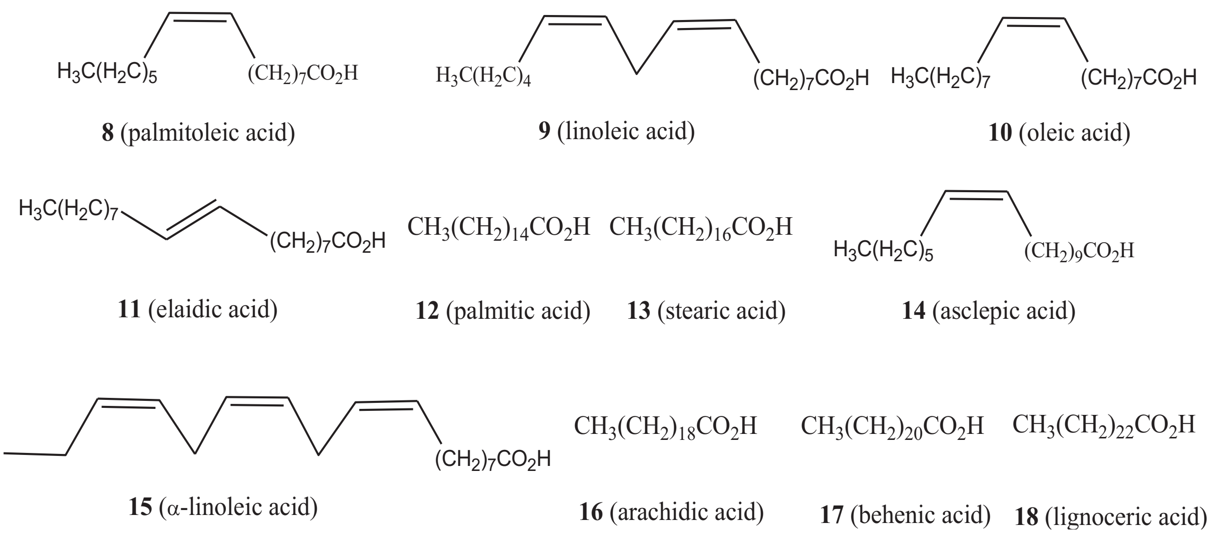

Fig. (4). Carboxylic acid content of *C. procera* and *C. gigantea* seed oils.

2.5. Pesticidal Use of the Plant

The reports on the bioactivity of extracts from both species are numerous, where the extracts have been utilized as herbicide [121], fungicide [122], insecticide [123 - 128], nematicide [129 - 133], acaricide [134], and as molluscicide [122, 135, 136]. Traditionally, extracts from *C. procera* have been used alone or in combination with that of other plants, such as in a combination of the extract of *C. procera* flowers, *Azadirachta indica* and *Nicotiana tabacum* leaves, and *Trachyspermum ammi* seeds used against the common cattle tick *Rhipicephalus microplus* (Boophilus) [137].

2.6. Insecticidal Use – Insects Associated with *Calotropis plants*

The larvicidal role of *C. procera* in mosquito control was reported already more than 25 years ago [127]. The latex from the green parts of the plant, which in effect represents a system of defense of the plant itself against insects [68, 122, 138], severely affects larvae development and mortality and suppresses egg hatching [139] in the mosquito *Aedes aegypti*, which is a vector of the dengue virus. At the same time, mixing water with aqueous latex extract leads to reduced ovipositioning of gravid *A. aegypti* females [140]. Laboratory experiments using water without and with various concentrations of larvicidal latex extract seem to indicate that the ovipositioning female can distinguish among the extract concentrations to lay its eggs in the medium with the least larvicidal concentration [140]. Aqueous latex extracts, but not those of the flowers, were shown to also have a larvacidal activity in the mosquito *Anopheles labranchiae* [141] and *Culex qinquefasciatus* [142, 143]. Aqueous leaf extracts were found to have larvacidal activity against *Anopheles arabiensis* [144]. Also, the extracts exhibited oviposition deterrent activity in the mosquitoes *C. qinquefasciatus* and *A. arabiensis* [144]. Moderate larvacidal activity of aq. extracts of *C. procera* was found against *C. quinquefasciatus* Say [142, 144, 145] and *Anopholes stephensi* [142]. Methanolic latex extracts of *C. procera* have been proven to be the most effective, however, as larvacide against such dengue vectors as *A. aegypti*. Thus, Singhi *et al*. have carried out a field study in selected areas of Jodphur City, India, dispersing an aq. solution of a dried methanolic extract of *C. procera* latex in water tanks and containers that normally function as breeding areas of different mosquito species such as *C. quinquefasciatus*, *A. stephensi*, and *A. aegypti*. A 100% larval mortality was found throughout when applying the solution at a concentration of 100 ppm [146]. Also, methanolic extracts of *C. gigantea* leaves have larvicidal (1^{st}-4^{th} instar tested) and pupicidal effect on *C. quinquefasciatus*, *A. stephensi*, and *A. aegypti* [147].

The extracts of *C. procera* leaves have been shown to have a profound effect on a diminished survival of the fifth instar larvae of the desert locust *Schistocerca gregaria*. Additionally, the arrest of ovarian growth and the absence of sexual maturity in adult *Schistocerca gregaria* were noted [148]. Also, latex extracts of *C. procera* are effective against *S. gregaria* [149]. Interestingly, the extracts of *C. gigantea* have been found to be active against *S. gregaria* as well, with an isolated plant component, a non-protein amino acid, forwarded as an antifeedant [150], thought to be the active principle. Later, the structure of this amino acid could not be confirmed [151]. Leaf extracts of *C. procera* were found to be effective against the flesh fly, *Sarcophaga haemorrhoidales fallen* [126] and have been advocated as an insect control against the two moths *Clostera cupreata* (Noctuidae) and *Plecoptera reflexa* (Noctuidae), where the 3^{rd} instar larvae were targeted [152]. *Calotropis procera* has been used for the control of the ladybird beetle *Henosepilachna elaterii* [123], the painted grasshopper *Poecilocerus pictus* Fab [153], the lesser grain borer *Rhizoperta dominica* [124, 154, 155], the rice weevil *Sitophilus oryzae* and the silver leaf whitefly *Bemisia tabaci* Gennadius [156]. The root bark of *C. gigantea* [157] and aq. leaf extracts of *C. procera* [158] show insecticidal activity against the red flour beetle *Triboleum castaneum*, while the root bark of *C. procera* is active against the confused flour beetle *Triboleum confusum* [159]. While mixed extacts from *C. procera* and other plants have been found most effective against the common cattle tick *Rhipicephalus microplus* (Boophilus) [137, 160] (see above), leaf extracts and whole plant extracts of *C. procera* alone are successful against the common cattle tick [160, 161] and the Asian blue tick (*Rhipcephalus microplus*), affecting both the oviposition of female ticks and the larval mortality rate [161, 162]. Cardiac glycosides in the extracts of *C. procera* can be seen as one of the active principles according to the work by D. H. Al-Rajhy *et al*. on the efficacy of the extracts on the camel tick *Hyalomma dromedarii* (Acari: *Ixodidae*) [163].

Nevertheless, by far, not all insects are affected negatively by the plant. The African monarch, *Danaus chrysippus*, a butterfly common to many areas in Africa and Asia, thrives on the plant and its caterpillars (1^{st} – 5^{th} instar) utilize some of the latex proteins in their diet. It is suggested that the caterpillars' proteolytic digestive system destroys the toxic proteins of the *Calotropis* latex, including the peptidases, and thus makes them immune to the toxic principles of the plant [68, 164]. There also seems to be a fine balance in the feeding behavior of the *Danaus chrysippus* caterpillars, notably of the younger instars, where the need for nourishment is off-set by the exposure to *de facto* poisonous cardenolides in the latex, where caterpillars cut the leaves, wait for the exuded latex to dry, and progress on feeding off the leaves while avoiding the latex for the most part [68, 164].

C. procera and *C. gigantea* are host to a larger number of other insects such as the cossid moth, *Semitocossus johannes* (Staudinger) [165], the larvae of which can cause severe damage to the plant. Especially, the nectar of the flowers attracts insects, including ants. The large Arabian carpenter bee, *Xylocopa sulcatipes* Maa. is noted to build its nests in dry stems of *C. procera* [166]. Here, even a *X. sulcatipes* – *C. procera* coevolutionary pattern has been suggested with *X. sulcatipes* as the natural co-adapted pollinator [167]. It is known that many species of *Asclepias* plants, in general, attract insects such as butterflies [168, 169] and milkweed bugs [170]. Ten insect species were recorded in India alone to feed on *C. gigantea* [171, 172]. Altogether, 65 species of insects and 5 species of mites have been documented on *C. procera* and *C. gigantea* [173]. In the United Arab Emirates, the black carpenter bee (*Xylocopa ctenoxylocopa fenetrata*), canary carpenter bee (*Xylocopa koptortosoma aestuans*), oriental wasp (*Vespa orientalis*), and scoliid wasp (*Vobalayca flavifrons*) are typical insects associated with *C. procera*. Butterflies will lay eggs on milkweeds as will moths [165], and it is the caterpillars which incorporate the sequestered cardenolides from the plants in their defense systems. Within this context, the cardenolide fingerprint of monarch butterflies, *Danaus plexippus* L., reared on different milkweeds has been studied [174 - 176]. The transfer of cardiac toxins from the caterpillar feeding on *C. procera* to the tissue of the adult butterfly has also been noted in the common tiger butterfly, *Danaus chrysippus*. The grasshopper *Poecilocerus pictus* feeds on *C. gigantea*. Again, it was found that cardiac glycosides are taken up by the insect [177, 178]. Here, however, a comparative characterization of the cardiac glycosides of *C. gigantea* and the feeding *P. pictus* revealed partial metabolisation of some of the substances in the insect extract. It was found that the insect extract reduces the viability of A549 (carcinomic human a lveolar basal epithelial) and COLO 205 (human caucasian colon adenocarcinoma) cells, inducing apoptosis in COLO 205, where the extract distinguishes between normal human cells and cancer cells [179, 180]. The unpalatability of insects reared on cardiac glycoside containing milkweeds was demonstrated in feeding experiments of *Asclepias currasavica* L. reared monarchs to the American blue jay *Cyanocitta cristata bromia* [181]. The aphid *Aphis nerii* is another insect that thrives on *C. gigantea*. Aphids feeding on *C. gigantea* leaves were found to have longer life-spans, with higher fecundity of the females, while the lady-beetle *Menochilus sexmaculatus*, the main predator of *Aphis nerii*, exhibited a shorter life-span when on a diet of *C. gigantea* fed aphids [182]. The *Calotropis* plants are not only hosts to insects, but to birds such as sunbirds as well.

On the other hand, insects lead also to possible ways for a classical biological control of *Calotropis*. It has been noted that three pre-dispersal seed predators, the Aak weevil *Paramecops farinosus*, the Aak fruit fly *Dacus persicus* in the Indian subcontinent and the Sodom apple fruit fly *Dacus longistylus* in the Middle East can be seen as prospective biological control agents of the plant [173].

2.7. Nematicidal/Schistosomicidal/Antihelminthic Activity

Calotropis procera has been used for the control of plant-pathogenic nematodes *Meloidogyne javanica* [183, 184] and *Meloidogyne incognita* [132, 185, 186]. It must be remarked that the mortality rates of *Meloidogyne incognita* juveniles induced by the leaf extracts of *C. procera* are not as high as that induced by the extracts of some other plants such as *Acacia nilotica*, *Aristilochia bracteolate* or *Chenopodium album* [187]. Latex extracts of *C. gigantea* have also been used against *Meloidogyne incognita* as well as against the cowpea cyst nematode *Heterodera cajani* [188]. Interestingly, the methanolic extracts of *Calotopis procera* flowers also exhibit schistosomicidal activity [189]. Aqueous and methanolic extracts of dried flowers of *C. procera* have antihelmintic effect as studied on live *Haemonchus contortas* [190]. Sheep infected with gastrointestinal nematodes were treated with the extracts and the egg counts in treated sheep decreased significantly [190] although the activity was found to be lower than that of levamisole. Also, the latex of *C. procera* has been shown to have antihelmintic effect in sheep [191, 192]. Ethyl acetate solutions of lyophilized *C. procera* latex inhibited egg hatching of *H. contortas*, showed a pronounced larvicidal effect (EC_{50} = 0.22 mg/mL) and inhibited the motility of the parasites by over 80% after 6h contact time [193].

2.8. Molluscicidal Activity

Much of the here-mentioned activity can be attributed to the cardenolide content of the extracts as could also be shown by the molluscicidal activity of cardenolide extracts from *C. procera* against the land snail *Monacha cantania* [136] and single-compound toxicity studies of uscharin in the snail *Thepa pisana* [135]. Even fresh leaves of *C. gigantea* at 200 g/ha have been used to control the golden apple snail, *Pomacea canaliculata*, in rice [194]. Alcoholic extracts of fruits, leaves, stems, and roots of *C. procera* were found to show molluscicidal activity against *Biomphalaria arabica* [195], the intermediate host of the trematode *Schistsoma mansoni* in Saudi Arabia.

2.9. Plant Fungicidal Use

Different aerial parts of *C. gigantea*, such as the leaves, have been used as anti-fungicide, especially against *Phyllactinia corylea* (powdery mildew), *Peridiopsora mori* (brown rust), *Pseudocercospora mori* (black leaf spot), *Myrothecium roridum* (brown leaf spot), *Colletotrichum graminicola*, *Drechslera sorokiniana* (Drechslera leaf blight), *Fusarium solani*, *Macrophomina phaseolina*, and *Phomopsis sojae* (soybean stem blight). Mixtures of extracts of *C. gigantea*, *Azadirachta indica* (neem), *Datura stramonium* and cow manure have been seen to be effective against *Fusarim mangiferae*, a fungus leading to floral malformation in mango [196]. Extracts of *C. procera* were found to inhibit growth, sporulation, and the conidial germination of *Drechslera biseptata* and *Fusarium solani* [197]. Others have reported the fungi toxic properties of extracts of the plant [198], especially also against seed-borne mycoflora of wheat, which includes *Altemaria alternata*, *A. clamydophor*, *Aspergillus niger*, *A. flavus*, *Rhizopus oryzae*, *Mucor* spp., *Fusarium* spp., *Drechslera australiensis*, *Penicillium* spp., *Curvularia lunata* and *Cladosporium* [199]. Recently, it has been found that methanolic *C. procera* leaf extracts are active against *Macrophomina phaseolina*, a fungus prevalent in arid tropical or semitropical regions causing stalk and fruit rot in many crop plants [200]. Extracts of *Calotropis procera* were found to be active against the fungal pathogen *Alternaria solani*, which causes early blight disease in tomatoes [201].

2.10. Allelopathic Activity

Aqueous extracts of dried leaves have a strong adverse effect on the germination of wheat (*Triticum aestivum*, Graminaea) [121, 202, 203] and influence the germination of barley (*Hordeum vulgare,* Graminaea), cucumber (*Cucumis satires*, Curcurbitaceae), radish (*Raphanus sativus*) [202], and Fenugreek (*Trigonella foenumgraecum*, Leguminosae) to a lesser degree [121]. Also, aqueous leaf extracts were reported to exhibit an allelopathic effect on the germination and seedling growth of maize, where inhibition of radicle and plumule growth of four maize cultivars were noted [204].

Flower extracts of *C. procera* were found to have an effect on the germination of wheat and canola (*Brassica napus*), not only delaying the germination but also leading to a reduction of root and shoot lengths of the plants. Also, the germination, seedling growth and biomass yield of acacia species such as of *Acacia tortilis* (Forssk.) are detrimentally influenced by aq. *C. procera* leaf extract [205]. Aq. whole plant extract mixtures of *Datura stramonium* and *C. procera* have been advocated for the management of the invasive parthenium weed *Parthenium hysterophorus L* [206]. In this case, the allelopathic properties of the extracts were attributed to quercetin (**19**), *p*-coumaric acid (**20**), gallic acid (**21**), sinapinic acid (**22**), and chlorogenic acid (**23**) (Fig. **5**). In another study, caffeic acid (**24**), gentisic acid (**25**), catechol (**26**), syringic acid (**27**), ellagic acid (**28**), resorcinol (**29**), and *p*-hydroxybenzoic acid (**30**) in addition to gallic and *p*-coumaric acid, **20** and **21**, were the isolated compounds made responsible for the allelopathic behavior of aq. leaf extracts of *C. procera*, this time on *Cassia sophera* and *Allium cepa* L. (onion) (Fig. **6**) [207]. Nevertheless, even in the arid wasteland, other plants can be seen growing in close proximity to *C. procera*. Reversely, it has been noted that buffel grass (*Cenchrus ciliaris*) has allelopathic toxicity towards the roots of *Calotropis* [208].

2.11. *Calotropis* in Environmental Monitoring and Bioremediation

Calotropis plants can grow under adverse conditions and have been found to be tolerant of pollution. This suggests that the species can be grown in the neighborhood of polluted sites as a remedial measure [209]. Thus, the uptake of metals by *C. procera*, both from the soil *via* the roots and from the air *via* the leaves [210], has received some interest. The results have been found to vary, where both the conditions on site and the matrix incorporating the heavy metals may well play a role. *C. procera* has been used for the monitoring of Pb in automobile-

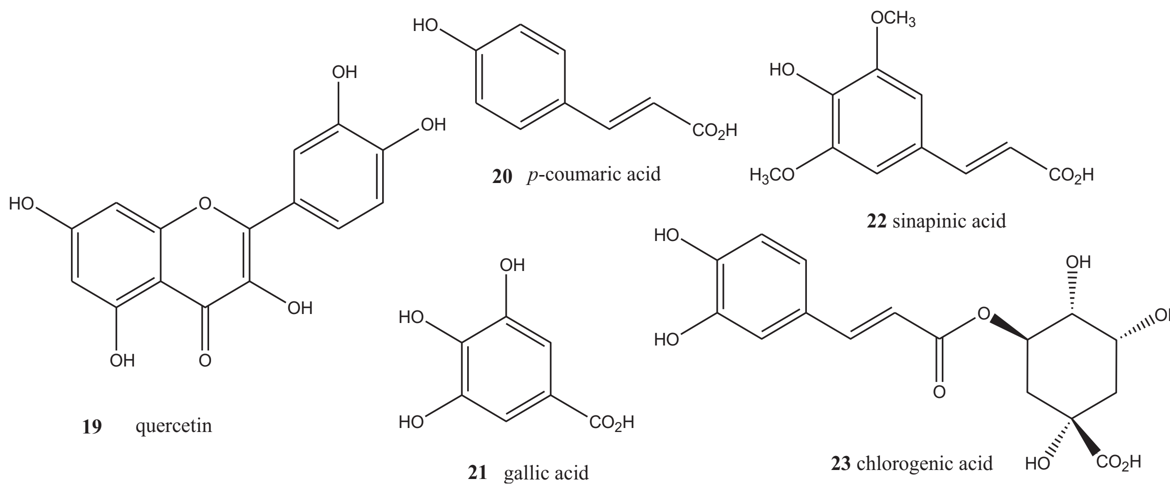

Fig. (5). Allelopathic compounds of aq. whole plant extracts of *C. procera* [206].

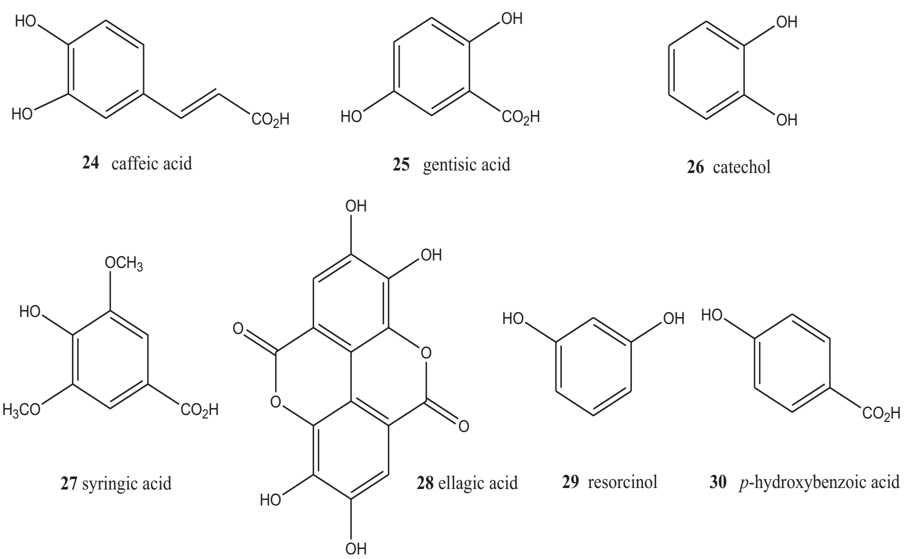

Fig. (6). Allelopathic compounds of aq. leaf extracts of *C. procera* [207].

exhaust pollution [211, 212]. *C. procera* has been under discussion as a phytoremediator of soils polluted with metal wastes [213 - 215]. In some cases, it could be shown that *C. procera* shows a high transfer factor for Cd, Ni and Pb from the soil to the plant, with the highest concentrations of Pb and Cd found in the fruits, and of Ni in the stem of the plant [216]. The accumulation of Cd in the leaves has also been demonstrated with plants growing on contaminated soil near Riyadh, Saudi Arabia [217] and on contaminated soil near Agra in the urban North Central area of India [218]. In the same general region, *C. procera* has been found to have a good bioconcentration factor for Zn, Mn, Cd and Cu, but a low translocation factor, where concentrations of these heavy metals were higher in the roots than in the leaves of the plant [219]. This study is juxtaposed by the findings of M. Singh *et al.*, who showed that *C. procera* has a moderate bioaccumulation capacity for Co, Cr, Cu, Pb, and Zn with a high translocation capacity of these metals to the shoots of the plant [220]. On the other hand, a third study has found no accumulation at all of Zn and Ni in leaves, shoots or roots from contaminated soils in the Unnao, Uttar Pradeh area [221]. The results delineating the Cd-uptake of the plant are also not clear-cut. Investigations have shown that Cd(II) salts are readily adsorbed by *C. procera* at both pH 5.0 and pH 8.0, with a maximum biosorptive capacity of 40 – 50 mg Cd(II) / g of plant material [222]. Interestingly, though, a study of the metal accumulation in *C. procera* in polluted areas within Lucknow, India, showed little uptake of Cd [223] and the use of *C. procera* as indicator plants in heavily contaminated soils surrounding the Mahad AD'Dahab mine, Saudi Arabia, led to no accumulation of Cd in the plants at all, though high concentrations of Cd could be observed in neighboring *Pergularia tomentosa* and *Salsola sp.* plants. In this case, however, *C. procera* could be used as an indicator of As, Pb, Zn (also ref. 224), and Cu [225]. The use of *C. procera* for the phytostabilisation of Cu and Zn was also suggested by Al-Qahtani [217] and Dan-Badjo *et al.* have remarked on the significant uptake by *C. procera* of both As and Zn from the soil of a gold mining area in Niger [226]. From a location in Punjab, a significant correlation was reported between the Cr and Pb content in the soil and the content of these metals in *C. gigantea* plants growing in the area [227]. Pb and Cd at levels of 32 to 80 mg/kg (Pb) and of 2.8 to 3.6 mg/kg (Cd) in the leaves of *C. procera* were found to exhibit toxicity to the plant as evidenced by lower chlorophyll contents in the leaves [218], leading to reduced photosynthesis of the plant. It must be noted that some additional stress factors may have been contributing, such as the proximity of the plants to heavy road traffic resulting in exposure to various other pollutants. There is some concern regarding the good metal-extraction capability of the plant that often also serves medicinal purposes as the effect of the metal concentration in the plant on the plant's medicinal extracts is not known. Also, the use of *Calotropis* as fodder in more highly contaminated areas should be avoided [225].

Processed parts of *C. procera* have also been used in environmental remediation. Thus, *C. procera* roots have been utilized in the removal of Cr(III), As, and Cu from water, incl. waste-water [228 - 231]. Oven dried, ground and sieved *C. procera* roots of 100 mesh size in fixed-bed columns have been used to remove Pb(II) from water [231]. The results obtained when using *C. procera* latex in the purification of domestic and industrial wastewater in Nigeria, on the reduction of turbidity, color, odor, microbial load and total coliforms were comparable to those using already proved coagulants such as *Moringa oleifera*, *Jatropha curcas*, $FeCl_3$, and aluminum sulfate (alum) [232]. Simple carbonization of *C. procera* leaves in the presence of sulfuric acid at 150°C left charcoal of 40-60 mesh, which was used to remove Zn(II) from water [233]. Also, polyvinyl-coated activated carbon prepared from *C. procera* leaves was used for the removal of Zn(II) from synthetic wastewater [233]. Cut *C. procera* leaves have been studied in water purification in Nigeria, where it was found that treatment significantly reduced the water's turbidity and coliform count [234]. Biosorption studies of Cd(II) on leaf biomass have also been carried out [235]. Polyvinyl coated activated carbon prepared from *C. procera* leaves was used for the removal of Cd(II) from synthetic wastewater [236].

CONCLUSION

Calotropis procera and *Calotropis gigantea* are undemanding plants that, while being undesirable weeds in some regions, have economic potential as a source of fuel and chemical feedstock. The review, which is seen as part 1 of a two-part series, showed their use in providing construction materials and, in limitations, their utilization as animal feed. Extracts of *C. procera* and *C. gigantea* are employed widely as natural pesticides. The application of the two plants in bioremediation efforts, including in the monitoring of environmental pollutants in the soil was discussed, also.

CONSENT FOR PUBLICATION

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CONFLICT OF INTEREST

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