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Short Report

Chemical Composition and Larvicidal Activity of the Essential Oils of *Cordia leucomalloides* and *Cordia curassavica* from the Northeast of Brazil

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Os óleos essenciais das folhas de *Cordia leucomalloides* e *Cordia curassavica* foram obtidos por hidrodestilação e suas composições químicas determinadas por uma combinação de CG-EM e CG-DIC. Como resultado, vinte e três componentes foram identificados em ambos os óleos, representando 98,6 e 91,2% da composição volátil. O óleo essencial de *C. leucomalloides* foi caracterizado por uma alta percentagem de sesquiterpenos (90,6%), sendo δ-cadineno (17,4%), (*E*)-cariofileno (15,7%), biciclogermacreno (12,5%) e germacreno D (11,2%) os majoritários. Por outro lado, o óleo de *C. curassavica* mostrou proporções similares de monoterpenos (47,3%) e sesquitepenos (43,9%), entre os quais α-pineno (20,5%), β-pineno (13,1%), (*E*)-cariofileno (12,4%) e biciclogermacreno (13,8%) foram os compostos predominantes. O potencial larvicida dos dois óleos foi avaliado contra larvas no terceiro estágio do mosquito *Aedes aegypti*. Os resultados mostraram que ambos os óleos apresentaram atividade biológica significativa, particularmente o óleo essencial de *C. leucomalloides*, o qual foi capaz de matar 98,7% das larvas numa concentração de 100 ppm.

The essential oils obtained from the leaves of *Cordia leucomalloides* and *Cordia curassavica* were obtained by hydrodistillation and their chemical compositions determined by a combination of GC-MS and GC-FID. Twenty-three components were tentatively identified in both oils representing 98.6 and 91.2% of the volatile content. The oil of *C. leucomalloides* was characterized by a large percentage of sesquiterpenes (90.6%), being δ-cadinene (17.4%), (*E*)-caryophyllene (15.7%), bicyclogermacrene (12.5%) and germacrene D (11.2%) the major ones. On the other hand, the oil of *C. curassavica* showed similar proportions of monoterpenes (47.3%) and sesquiterpenes (43.9%) among which α-pinene (20.5%), β-pinene (13.1%), (*E*)-caryophyllene (12.4%) and bicyclogermacrene (13.8%) were the predominant compounds. The larvicidal potential of the two oils were evaluated against the third-instar of *Aedes aegypti* larvae. The results showed that both oils exhibited significant activity, particularly the oil of *C. leucomalloides* which was able to kill 98.7% of the larvae in the concentration of 100 ppm.

Keywords: *Cordia leucomalloides*, *Cordia curassavica*, essential oil, larvicidal activity, terpenoids

Introduction

The pantropical genus *Cordia*, represented by shrubs and trees, is one of the major and most important genus of the family Boraginaceae.[1] Some of its species yield excellent wood naturally resistent to attack of several organisms,[2,3] while others are widely used in traditional medicine in several parts of the world.[4-6] Various phytochemical studies on *Cordia* species have been published, revealing a great number and diversity of secondary metabolites including triterpenes,[4]

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flavonoids,[7] sesquiterpenes,[8] saponins,[9] hydroquinones[3] and quinones.[10] Nevertheless, there are few studies published about the chemical composition of the essential oils of those plants. To the best of our knowledge, only the chemical composition of the essential oils of *C. curassavica* (Jack.) (syn.: *C. verbenacea* D.C.),[1,11] *C. nitida* Vahl.,[12] *C. trichotoma* Vell.[13] and *C. globosa* (Jacq.) H.B.K.[14] have been described. The two later were recently subject of our studies, including their essential oils.

A literature survey revealed that the oil of *C. leucomalloides* has not been reported but *C. curassavica* has been the subject of several studies, probably due its etnobotanical value.[1,5,6,15] *C. curassavica* is a medicinal plant with anti-inflammatory and antimicrobial properties. Indeed, artemetin, a flavonoid with anti-inflammatory activity[5] was isolated from the leaves and its essential oil has exhibited antimicrobial properties.[12] In addition, naphthoquinones with antifungal and larvicidal activities have been isolated from the roots of this plant.[6] As part of our research program on *Cordia* species native to Northeast of Brazil,[8-10,13,14,16-18] the current paper describes the chemical composition of leaf oils of *C. leucomalloides* and *C. curassavica*, as well as their larvicidal effects against the mosquito *Aedes aegypti*, the vector of the yellow fewer and dengue.

Results and Discussion

The list of volatile components and the percentage composition of both oils are shown in Table 1. In the essential oil of *C. leucomalloides* twenty-three constituents were identified, which represent 98.6% of the total composition. The sesquiterpenoids constitute the dominant fraction (90.6%), with (*E*)-caryophyllene (15.7%), germacrene D (11.2%), bicyclogermacrene (12.5%) and δ-cadinene (17.4%) as the major ones. Likewise, twenty-three constituents were also identified for *C. curassavica* accouting for 91.2% of the oil composition. The percentage of monoterpenes (47.3%) and sesquiterpenes (43.9%) were approximately the same. α-Pinene (20.5%) and β-pinene (13.1%) were the most significant constituents identified in the monoterpene fraction, while (*E*)-caryophyllene (12.4%) and bicyclogermacrene (13.8%) were the main sesquiterpenes.

Previous studies on leaf essential oil of *C. curassavica*, from the southeast region of Brazil, showed that the main compounds were α-pinene (29.7%) and (*E*)-caryophyllene (25.3%). Other terpenoids were also found in significant amounts including *allo*-aromadendrene (10.0%), α-humulene (4.6%), β-gurjunene (4.1%), spathulenol (2.8%) and bicyclogermacrene (2.7%).[6] On the other hand, in the oil of *C. curassavica* from Panama, the most prominent constituent was β-terpinene (32.0%) followed by (*E*)-caryophyllene (12.0%), camphene (9.0%), germacrene D (9.0%) and α-pinene (6.0%).[1] Despite the compositional differences detected in the three oils samples of *C. curassavica* from different geographic sites it is important to emphasize the presence of (*E*)-caryophyllene and α-pinene among the main constituents of both oils of Brazilian origin compared to the high content of β-terpinene solely for *C. curassavica* of Panamanian origin. α-Pinene, (*E*)-caryophyllene, β-elemene, α-humulene, bicyclogermacrene and δ-cadinene are common constituents for *C. curassavica* of any origin, while camphene is common just to *C. curassavica* oils from Panama and Northeastern Brazil. It is also important to point out that (*E*)-caryophyllene, germacrene D and bicyclogermacrene are the major

Table 1. Volatile compounds identified in the leaf essential oils of *C. leucomalloides* and *C. curassavica*

		C. leucomalloides	*C. curassavica*
Compounds[a]	KI[b]	Relative area / (%)	
α-Phellandrene	934	-	1.0
α-Pinene	942	1.5	20.5
Camphene	953	-	0.8
Sabinene	976	0.9	5.0
β-Pinene	980	3.3	13.1
Myrcene	992	-	0.9
p-Cymene	1021	-	1.0
Limonene	1031	-	1.5
γ-Terpinene	1060	-	1.2
Linalol	1096	-	1.1
Camphor	1131	1.5	0.6
Borneol	1163	0.8	-
4-Terpineol	1173	-	0.6
α-Copaene	1381	6.0	1.2
β-Bourbonene	1388	0.8	0.9
β-Elemene	1392	-	1.2
Longifolene	1405	0.5	-
(*E*)-Caryophyllene	1420	15.7	12.4
α-Bergamotene	1433	-	0.7
γ-Elemene	1444	2.2	-
α-Humulene	1464	1.0	1.6
α-Amorphene	1484	2.1	-
Germacrene D	1489	11.2	6.7
Bicyclogermacrene	1503	12.5	13.8
Cubebol	1515	1.1	-
δ-Cadinene	1525	17.4	-
Cadina-1,4-diene	1532	0.6	-
Germacrene B	1555	7.1	-
Spathulenol	1565	3.2	2.2
Caryophyllene oxide	1571	2.7	1.6
Globulol	1574	1.8	-
Viridiflorol	1590	1.5	1.6
α-Bisabolol	1652	3.2	-
Monoterpenes		8.0	47.3
Sesquiterpenes		90.6	43.9
Total		98.6	91.2

[a]Constituents listed in order of elution on DB-5 column. [b]KI = kovats retention index according to *n*-alkanes.

components common for oils from *C. leucomalloides* and *C. curassavica* from Northeast of Brazil, but δ-cadinene for *C. leucomalloides* and α- and β-pinene for *C. curassavica* can distinguish them.

The leaf oils from both investigated *Cordia* species exhibited significant activity against the third-instar of *A. aegypti* larvae, showing LC_{50} of approximately 63.1 ppm for *C. leucomalloides* and of approximately 97.7 ppm for *C. curassavica.* The same order of LC_{50} have been observed previously for the essential oils from *Lippia sidoides* (63 ppm), *Cymbopogon citratus* (69 ppm), *Ocimum americanum* (67 ppm) and *O. gratissimum* (60 ppm) all considered of high interest for the control of *A. aegypti*.[19,20] Taking in consideration the LC_{50} of the mentioned oils and the almost two fold higher toxicity of the oil from *C. leucomalloides*, relative to *C. curassavica*, it can be stated that the oil from the former has potential to become a promising natural agent to threat the mosquito life cycle, thus helping in the prevention of dengue fever.

Experimental

Plant material

The leaves of *C. leucomalloides* (No. 34725) and *C. curassavica* (No. 34715) were harvested during the flowering stages in January 2005, from Crato County, Ceará, Brazil. Voucher specimens have been deposited at the Herbário Prisco Bezerra (EAC) of the Departamento de Biologia, Universidade Federal do Ceará.

Extraction of the essential oils

The fresh leaves of *C. leucomalloides* (780 g) were subjected to hydrodistillation in a Clevenger-type apparatus for 2:00 hours to afford 0.07% of a pale yellow oil: $[\alpha]_D^{20}$ - 16° (*c* = 0.05, $CHCl_3$), η^{25} 1.6705. The same procedure were applied to fresh leaves of *C. curassavica* (900 g) to yield 0.13% of also a pale yellow oil: $[\alpha]_D^{20}$ - 30° (*c* = 0.05, $CHCl_3$); η^{25} 1.6633. The yields (m/m) were calculated based on the fresh weight of the plant materials. The isolated oils, after drying over anhydrous sodium sulfate and filtered were stored in sealed glass vials and maintained under refrigeration before analysis.

Chemical analysis

GC-FID. The quantitative analysis was performed on a Shimadzu GC-17A gas chromatograph equipped with a flame ionization detector using a non-polar DB-5 fused silica capillary column (30 m × 0.25 mm i.d., 0.25 μm film thickness). Hydrogen was used as carrier gas at a flow rate of 1 mL min^{-1} and 30 psi inlet pressure; split ratio 1:30; the column temperature was programmed from 35 ºC to 180 ºC at a rate of 4 ºC min^{-1}, then heated at a rate of 17 ºC min^{-1} to 280 ºC and held isothermal for 10 min; both injector temperature and detector temperature were 250 ºC.

GC-MS. The analysis was carried out on a Hewlett-Packard Model 5971 GC/MS using a non-polar DB-5 fused silica capillary column (30 m × 0.25 mm i.d., 0.25 mm film thickness); carrier gas helium, flow rate 1 mL min^{-1} and with split mode. The injector temperature and detector temperature were 250 ºC and 200 ºC, respectively. The column temperature was programmed from 35 °C to 180 ºC at 4 ºC min^{-1} and then 180 °C to 250 ºC at 10 ºC min^{-1}. Mass spectra were recorded from 30 – 450 *m/z*. Individual components were identified by matching their 70 eV mass spectra with those of the spectrometer data base using the Wiley L-built library and two other computer libraries MS searches using retention indices as a preselection routine,[21,22] well as by visual comparison of the fragmentation pattern with those reported in the literature.[23,24]

Larvicidal bioassay

Portions of essential oils (5 to 500 ppm) dissolved in H_2O:DMSO (98.5:1.5) were placed in a beaker (50 mL). 50 instar III larvae of *A. aegypti* were delivered to each beaker. After 24 hours, at room temperature, the number of dead larvae was counted and the lethal percentage calculated. A control using DMSO and water was carried out in parallel. For each sample, three independent experiments were run. The bioassays were performed at Laboratório de Entomologia, Núcleo de Endemias da Secretaria de Saúde do Estado do Ceará, Brazil.

At the concentrations of 500 and 250 ppm there was 100% larvae mortality for both oils. At 100 ppm the observed mortality was 98.7±0.9 and 50.7±2.0, while at 50 ppm the figures were 9.3±1.2 and 16.0±2.0, for *C. leucomalloides* and *C. curassavica*, respectively. This made possible the calculation of the LC_{50} as 63.1±0.5 and 97.7±1.0, for *C. leucomalloides* and *C. curassavica*, respectively.

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Supplementary Information

GC-MS chromatograms of the essential oils from leaves of *C. leucomalloides* and *C. curassavica* are available free of charge at http://jbcs.sbq.org.br, as PDF file.

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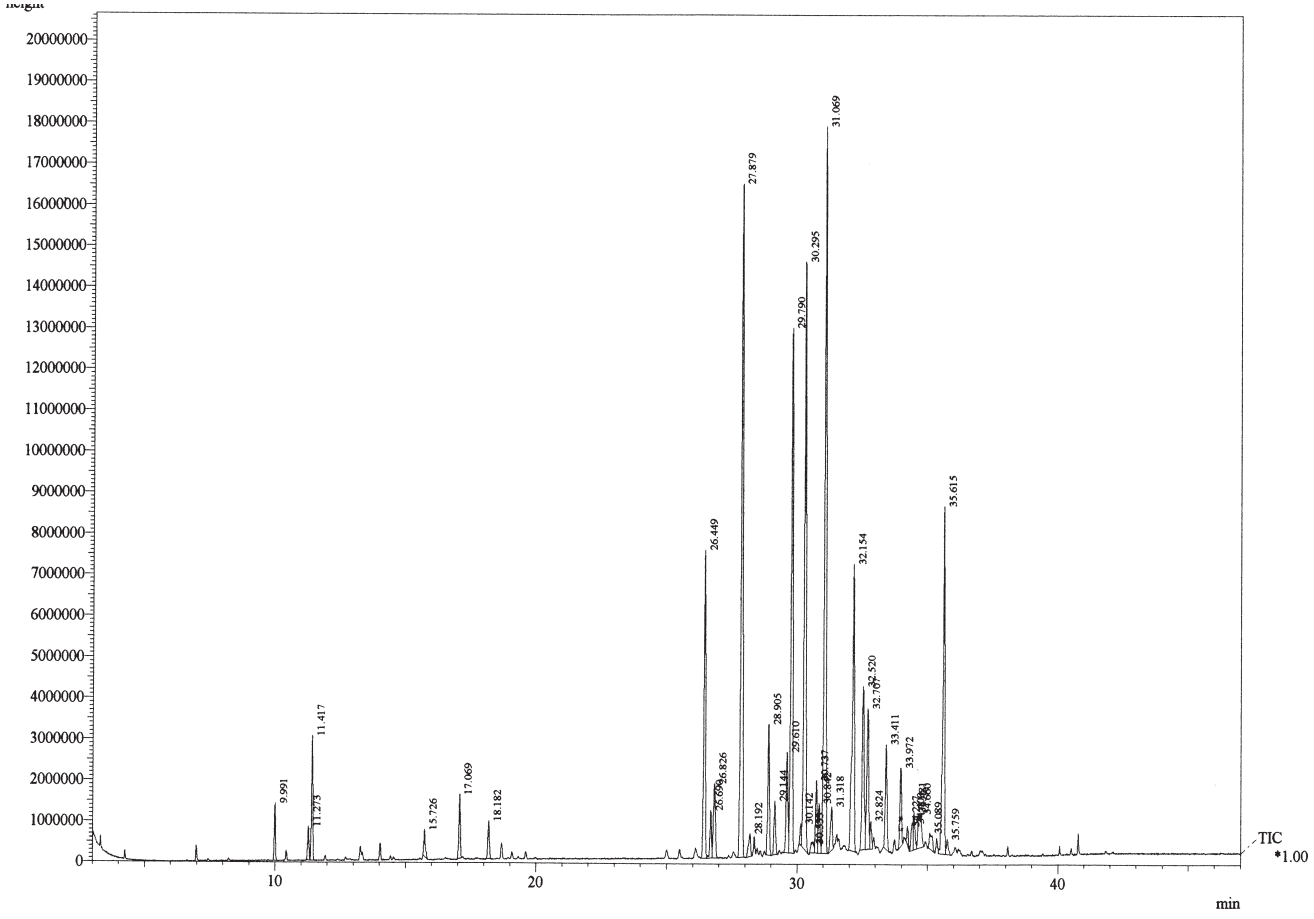

Figure S1. GC-MS chromatogram of the essential oil from leaves of *Cordia leucomalloides.*

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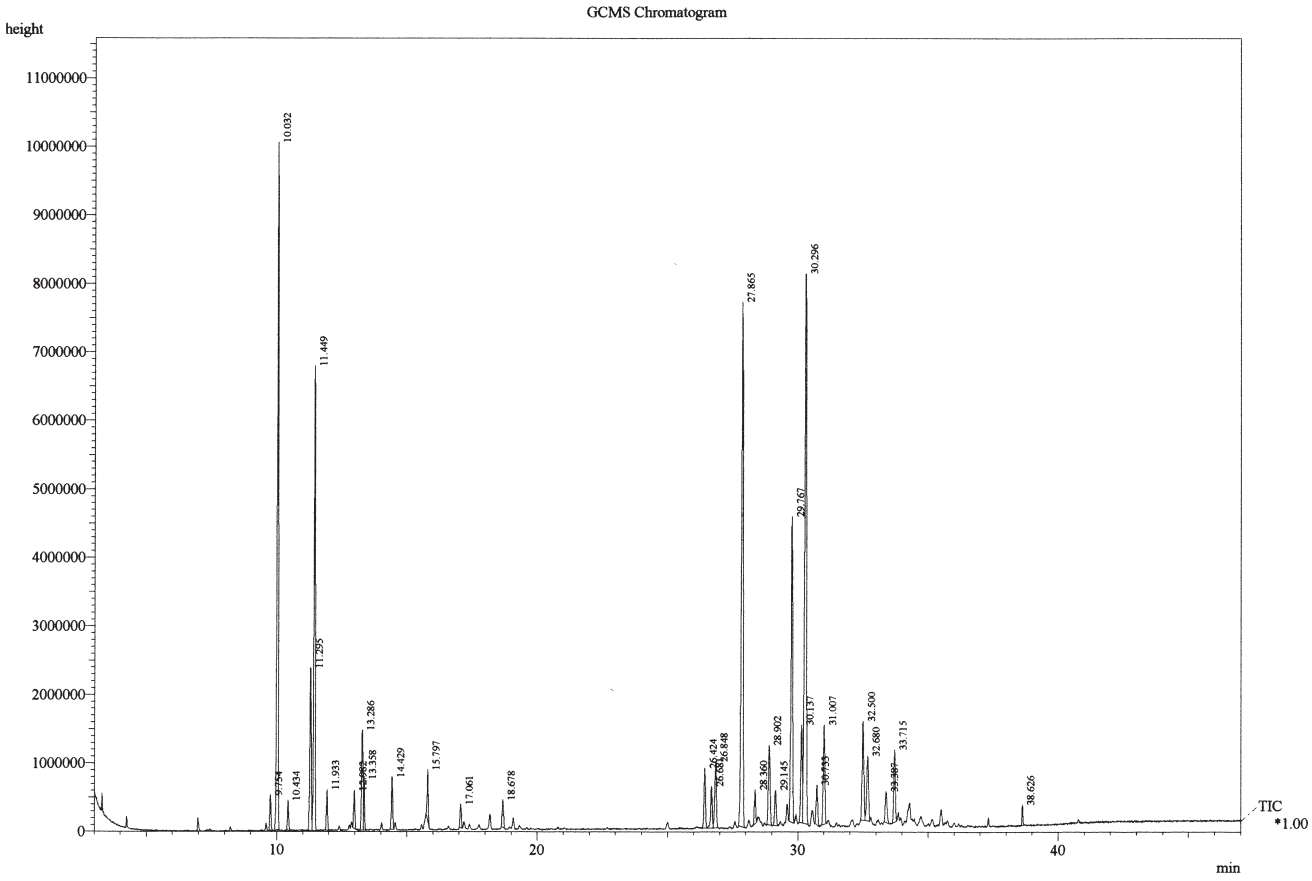

Figure S2. GC-MS chromatogram of the essential oil from leaves of *Cordia curassavica.*