Acta Scientiarum
http://periodicos.uem.br/ojs/acta
ISSN on-line: 1807-863X
Doi: 10.4025/actascibiolsci.v42i1.51639

Antioxidant and biological activities of essential oil from Colombian *Swinglea glutinosa* (Blanco) Merr fruit

Beatriz Eugenia Jaramillo-Colorado*, Flor María Palacio-Herrera and Edisson Duarte-Restrepo

Agrochemical Research Group, Chemistry Program, School of Exact and Natural Sciences, University of Cartagena, San Pablo Campus, Carrera 50, #24120, Cartagena, Bolívar, Colombia. *Author for correspondence. E-mail: bjaramilloc@unicartagena.edu.co; beatrizjaramilloc@yahoo.com

ABSTRACT. The objectives of this work were the study of the volatile chemical composition of essential oils (EO's) from *Swinglea glutinosa*, as well as to evaluate their antioxidant, repellent and fumigant properties. The EO was obtained by hydrodistillation from the peel of the fruit, gathered in the city of Cartagena, Bolívar (Colombia). The volatile composition was analyzed by gas chromatography coupled to mass spectrometry (GC-MS). The major compounds found in *S. glutinosa* were germacrene D (4.8%), limonene (5.2%), α-terpineol (6.5%), β-pinene (8.5%), nerolidyl acetate (9.8%), and *trans*-nerolidol (34.6%). *S. glutinosa* showed antioxidant potential (85.8%) (IC_{50}=142.49 μg mL^{-1}). The EO deployed repellent activity against the *Tribolium castaneum* weevil at a concentration of 15.73 nL cm^{-1} at 2 hours of exposure (72%), while the result for the commercial repellent was 50% at the same concentration. EO from *S. glutinosa* displayed the best fumigant activity with LC_{50} of 153.4 μg mL^{-1} air. The essential oil from *S. glutinosa* can be considerated as a natural source of biocides and antioxidants.

Keywords: repellent; fumigant; *Tribolium castaneum* Herbst; gas chromatography.

Received on December 28, 2019.
Accepted on March 13, 2020.

Introduction

The presence of insects in stored grain requires the development and implementation of new naturally occurring agrochemicals for the control and eradication of these harmful pests (Nenaah, 2014). One of the main groups of plague insects, economically important, that affect post-harvest products are Coleoptera beetles. Among them are the red flour beetles, *Tribolium castaneum* (Herbst) (Coleoptera: Tenebrionidae); these beetles act as secondary pests attacking already damaged grains or grain products. They can be found in almost all storage containers of cereals or cereal products, especially in tropical and subtropical climates; they attack corn, wheat, and flour, among others (Jaya, Singh, Prakash, & Dubey, 2014).

Studies of different biological activities, such as repellent, insecticide, antioxidant, antibacterial, and others, show that EOs (essential oils) are useful for controlling pests and other organisms during food storage, due to their high volatility and toxicity (Ukeh & Umoetok, 2011; Kim & Lee, 2014; Celano et al., 2017; Koutsaviti et al., 2018). Compared to some synthetic pesticides, EOs are characterized by minimal effects on human health and the environment, being an extraordinary tool for the agricultural industry (González, Gutiérrez, Ferrero, & Band, 2014).

Pesticides that are used indiscriminately become environmental contaminants causing toxic health effects and pests develop resistance mechanisms against these chemicals (Reis et al., 2016). Due to their bioactive properties, EOs have been considered as an alternative to control insects in stored foods. They are a notable source for developing and implementing new agrochemicals because they are rich in monoterpenes (González et al., 2014; Reis et al., 2016).

EOs are volatile, oily liquids, product of the secondary metabolism of plants (flowers, buds, seeds, leaves, bark, herbs, wood, fruits, and roots). They are comprised of complex mixtures of monoterpenes, sesquiterpenes, phenylpropanoids and other volatile compounds (Sawamura, 2013). They are obtained through distillation by stripping plants through steam or hydro-distillation. They are particularly abundant in some plant families: Conifers, Rutaceae, Umbelliferae, Myrtaceae and Labiatae, and are often localized in specialized histological structures (Koutsaviti et al., 2018; Lee, Annis, Turmaalii, & Choi, 2004). Some EOs have been widely applied in the fields of pharmaceutical, agricultural, sanitary, and cosmetic industries (Silva et al., 2014). They have properties used to kill insects, control pests, and protect stored food crops (Jaya et al., 2014). Furthermore, they are known for their antimicrobial and antioxidant properties, and their use in the

food industry has been widely described (Duarte, Luís, Oleastro, & Domingues, 2016; Silva et al., 2014).

Numerous investigations report that monoterpenes and sesquiterpenes had repellent and insecticidal activities (Ukeh & Umoetok, 2011; Kim et al., 2010; Giner et al., 2013). For example, eucalyptol or 1,8-cineole, monoterpenoid present in the *Eucalyptus* species, was used to control stored grain against *Sitophilus oryzae, T. castaneum* and *Rhyzopertha dominica* (Lee et al., 2004) as well as eugenol, citronellal, and geranial on *Callosobruchus maculatus* and *Sitophilus zeamais* (Reis et al., 2016).

Swinglea glutinosa is a small shrub belonging to the Rutaceae family; it is native of Asia and is characterized by its abundant foliage and alternate trefoil leaves and inedible fruit, with a strong citrus scent. In Colombia, it is used as hedge in rural and urban areas. The plants and fruits of this species are used in traditional medicine because they deploy several biological properties including antimalarial (Weniger et al., 2001a), antiprotozoal (Weniger et al., 2001b), anti-tubercular (Bueno-Sánchez, Martínez-Morales, Stashenko, & Ribón, 2009), antileishmanial (Rocha, Almeida, Macedo, & Barbosa-Filho, 2005) and insecticidal (Koutsaviti et al., 2018).

One of the important products obtained from *S. glutinosa* fruit wastes is the EO isolated from citrus peels. The major compounds found in EO are not always responsible for the biological activities; for this reason, identification and detection of all compounds, both majority and minority, becomes necessary. The analytical technique of choice used for this identification is gas chromatography-mass spectrometry (GC-MS) (Stashenko, Jaramillo, & Martínez, 2004).

In this study, we investigated the volatile chemical composition and antioxidant property of EOs isolated from the peel of *S. glutinosa* grown in Colombia, as well as their repellent and fumigant activities against the *T. castaneum* using *in vitro* bioassays. Also, the repellent and fumigant activities of four pure terpenes present in the EOs of *S. glutinosa* were evaluated.

Material and methods

Reagents

α-Pinene, ascorbic acid (standard substance) and ethanol were purchased from Merck (Darmstadt, Germany); acetone from AppliChem Panreac (Darmstadt, Germany); β-pinene, myrcene, R-limonene, DPPH (1,1-diphenyl-2-picrylhydrazyl) radicals, and C7–C30 n-alkanes were purchased from Sigma-Aldrich (St. Louis, MO, USA); Pirilan from Syngenta S.A. (Colombia) and filter paper from GE Healthcare (Hangzhou, China).

Plant material

The fruits of *S. glutinosa* (Blanco) Merr were collected in its flowering season in the city of Cartagena, Bolivar, Colombia (Figure 1). The taxonomic characterization of the plant was carried out at the Institute of Biology, School of Natural Sciences, University of Antioquia, Medellin, Colombia by Dr. Francisco J. Roldan Palacios. The vouchers of each plant were deposited in the herbarium as a permanent sample; the identification and code of the plant studied are *S. glutinosa,* HUA 185262.

Figure 1. Fruits of *Swinglea glutinosa* (Blanco) Merr. (author's own source)

Extraction of the essential oil

The Extraction was performed in a Clevenger-type apparatus, according to Jaramillo-Colorado, Martelo, and Duarte (2012). Were used 500 g of fruit peels from *S. glutinosa*, finely chopped and submerged in boiling water by using conventional heating for two hours. The EO was separated by decantation and then anhydrous Na_2SO_4 was added to the oil. One EO aliquot (30 µL) was diluted in one mL of dichloromethane for gas chromatography analysis.

Chromatography analysis

The EO was analyzed in an Agilent Technologies GC-MS system model 7890A Network GC coupled to a mass selective detector model 5975 (Palo Alto, California, USA) equipped with a split/split-less injection port (230 °C, split ratio 20:1). The mass spectra were obtained by electron-impact ionization at 70 eV energy. GC conditions were as follows: A HP-5MS capillary column (30m x 0.25mm id x 0.25µm df) with 5% phenyl-poly (methyl siloxane) stationary phase was used for the separation of mixtures. The initial oven temperature was 50 °C for 2 min., and then resumed at a rate of 5 °C min^{-1}. up to 250 °C (5 min.). The carrier gas was helium, with an inlet pressure at the head of the column of 12.667 psi at a rate of 1.172 mL $min.^{-1}$, at 50 °C. The mass spectra and Kovàts retention indexes obtained were compared with those reported in the literature (Adams, 2007).

Antioxidant activity

Antioxidant activity was evaluated as a measure of the ability to scavenge radicals by reacting with $DPPH^{\cdot}$ (1,1-diphenyl-2-picrylhydrazyl) radicals, potential antioxidants (EO) and ascorbic acid (standard substance). Two milliliters of a 3.6×10^{-5} M ethanolic solution of DPPH was added to a solution of the tested samples at different concentrations (300, 200, 150, 100, 50, and 10 µg mL^{-1}). The decrease in absorbance at 517 nm was recorded in an UV–Vis spectrophotometer for 16 min. Antioxidants scavenge the DPPH radical through the donation of hydrogen, which forms the reduced compound, DPPH–H. The color changes from purple to yellow after reduction (a product known as diphenyl picryl hydrazine). Antioxidant activity is expressed as an inhibition percentage, which corresponds to the amount of radical $DPPH^{\cdot}$ offset by the EO, (inhibition percentage of $DPPH^{\cdot}$ radical, % I DPPH.), according to the following equation (Jaramillo-Colorado et al., 2012):

$$\% \, I \, DPPH = \left[\frac{Abs_0 - Abs_1}{Abs_0}\right] \times 100$$

Where Abs_0 is the absorbance of control (without test sample), and Abs_1 is the absorbance of the test samples at different concentrations. IC_{50} (the concentration required to exert 50% of the antioxidant activity) was calculated by linear regression from the percentages of DPPH inhibition. The IC_{50} results were compared according to Tukey's statistical method ($p < 0.05$). Four replications were used for each concentration.

Insects and bioassays

Adults of *T. castaneum* used in the experiments were collected seven days after hatching. Bioassays were carried out in the dark in incubators at 28–30 ºC and 70–80% relative humidity (r.h) at the Agrochemical Research Laboratory of the University of Cartagena. Oat (*Avena sativa*) was employed to feed *T. castaneum.*

Repellent activity

The repellent activity was performed according to (Zhang et al., 2011). The experimental procedure was evaluated by using the area preference method. The EO of *S. glutinosa* and monoterpenes were dissolved in acetone (0.13, 0.63, 3.15, 15.73, and 78.63 nL cm^{-2}). Filter paper 9 cm in diameter was cut in half and 500 µL of each concentration was applied separately to half of the filter paper as uniformly as possible with a micropipette. The other half (control) was treated with 500 µL of acetone.

The treated and control half discs were dried at room temperature to allow evaporation of the solvent. Treated and untreated halves were attached using adhesive tape and placed in Petri dishes. Twenty adults (5–7 day old) of *T. castaneum* were released separately at the center of each filter paper disc. The dishes were then covered and transferred to an incubator at room temperature. Five replications were used for each concentration.

Fumigant activity

The toxic effect from *S. glutinosa* EO and terpenes were tested on *T. castaneum*. Filter paper discs (Whatman No. 1, 2-cm in diameter), deposited at the bottom of Petri dish covers (90 x 15 mm), were used. These were impregnated with oil at doses calculated to provide equivalent fumigant concentrations of 500, 350, 250, 150, 50 µg of oil mL^{-1} air, respectively. Twenty adult insects (1 to 10 days old) were introduced and tightly capped (replicated four times for each concentration). Pirilan, a commercial pesticide containing methyl pirimiphos (organophosphorus pesticide, 300 µg. mL^{-1} air) as an active ingredient, was used as positive control. The mortality percentage was determined after 24 hours from the start of exposure (Prieto, Patiño, Delgado, Moreno, & Cuca, 2011).

Statistical analysis

The results were converted into fumigant percentage and analyzed by ANOVA and Student t tests. Mortality rates were calculated using the statistical formulas of *Abbott* and *Probit* to determine the LC_{50}, chi-square values and related parameters (Jaramillo-Colorado, Suarez-López, & Marrugo-Santander, 2019). Biostat a statistical software (Analyst Soft Robust Business Solutions, BioStat Version 2009) was used, with a confidence level of 5%. Four replicates for each analysis were performed.

Results and discussion

Chemical composition of essential oil by gas chromatography analysis

The yield of EO from *S. glutinosa* was 0.53% (w w^{-1}). In the GC-MS analysis, 89.5% of the volatile composition could be identified (Table 1). Figure 2 shows a typical chromatogram from *S. glutinosa* essential oil. Peak identification can be seen in Table 1.

Table 1. Volatile chemical composition of essential oil from *Swinglea glutinosa* (Blanco) Merr., obtained by hydrodistillation.

Peak No	Compound	Type	RI_C HP-5	RI_t	Relative area, %
1	α–Pinene	M	941	939	3.3
2	Camphene	M	956	953	0.5
3	β-Pinene	M	984	980	8.5
4	β-Myrcene	M	993	991	2.8
5	Limonene	M	1032	1031	5.2
6	1,8 Cineole (Eucalyptol)	MO	1035	1033	2.0
7	γ-Terpinene	M	1060	1062	1.7
8	α-Terpinolene	M	1076	1088	2,4
9	Linalool	MO	1088	1098	1,8
10	α-Terpineol	MO	1185	1189	6.5
11	β-Caryophyllene	S	1418	1418	3.3
12	Germacrene D	S	1478	1480	4.8
13	Germacrene B	S	1566	1562	2.0
14	*trans*-Nerolidol	S	1570	1568	34.6
15	Germacrene-D-4-ol	S	1572	1572	1.0
16	Spathulenol	S	1580	1576	1.5
17	Caryophyllene oxide	SO	1586	1580	1.2
18	Nerolidyl acetate	SO	1670	1676	9.8
19	Germacrone	SO	1697	1693	1.0
20	NI	SO	1718	-	1.0
21	Isobicyclogermacrenal	SO	1730	1733	1.0
22	NI	SO	1745	-	1.0

a) Peak number in Figure 2; b) Identification made by mass spectrometry (EI: electron impact ionization, 70 eV; peak matching >90%) and LRIs. Spectral databases wiley8, NIST08; c) Experimentally RI on the HP-5; d) Averages of three independent extractions; *Tentative identification based in LRIs on HP-5 column; RI – Linear retention indices relative to C_7–C_{30} n-alkanes; RI_c – Calculated RI; RI_t – Theoretical RI (Adams, 2007).

The main components found in the EO from *S. glutinosa*, were sesquiterpenes: germacrene D (4.8%), nerolidyl acetate (9.8%), and *trans*-nerolidol (34.6%), and monoterpenes: limonene (5.2%), α-terpineol (6.5%), and β-pinene (8.5%).

These results are different from those found in EO obtained from other regions in Colombia. In the S. glutinosa EO collected in the Cordoba province, the principal components isolated were β-cubebene (26-28%), β-pinene (24-27%) and elixene (10-11%) (Díaz, Arrázola Paternina, Ortega, & Gaviria, 2005). For oil obtained in the city of Bucaramanga, the predominant compounds were β-pinene (49.6%), α-pinene (12%) and β-sabinene (11%)

(Bueno-Sánchez et al., 2009). The major constituents of oil derived from fruits collected in Cuba were reported as E-nerolidol (41.3%) and caryophyllene oxide (20.9%) (Pino, Marbot, & Fuentes, 2006).

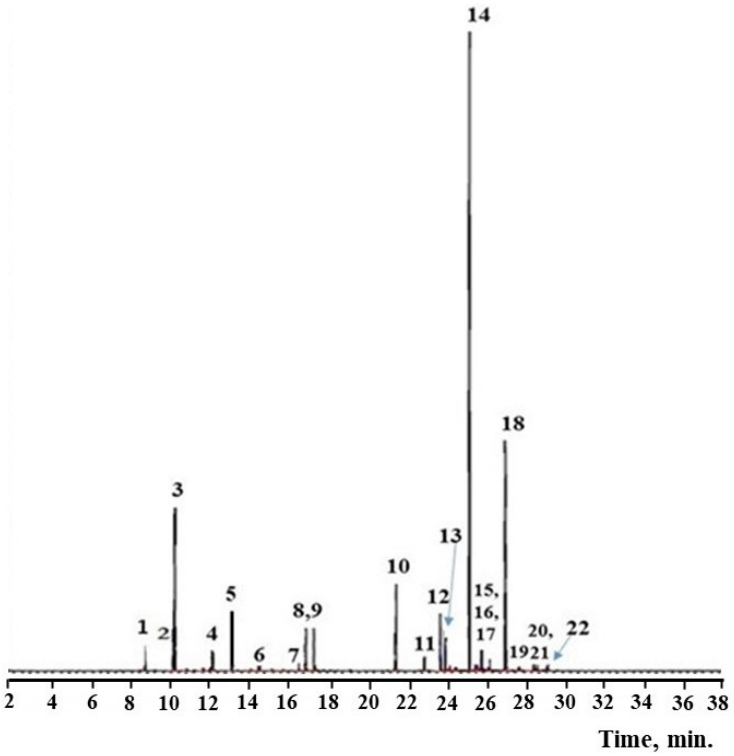

Figure 2. Typical chromatogram obtained from Colombian *Swinglea glutinosa* isolated by hydrodistillation and analyzed by gas chromatography coupled with a flame ionization detector (FID) and mass spectrometry (MS) (See Table 1).

The considerable variability in the composition of the EO is probably due to differences in the ecological and climatic conditions, effect of light intensity, altitudes, crop and soil which are directly related to the production of secondary metabolites (Benyelles et al., 2017; Estell, Fredrickson, & James, 2016; Spitaler et al., 2006). Many investigations indicate the existence of the morphological and chemical variability of plant chemotypes, and their vast geographical range, which suggest that species have adapted to new combinations of environmental factors through permanent changes in the genotype, as well as phenotype plasticity (Souza et al., 2018; Andrade et al., 2016; Barra, 2009; Ricciardi et al., 2009)

Antioxidant activity

Radical DPPH· was neutralized by the EO from *S. glutinosa*, and the maximum inhibition percent of DPPH· was 85.8% (2.5 μg mL^{-1}). A comparison was made with ascorbic acid (a substance used as a reference antioxidant), where the percentage of inhibition against the DPPH· radical was 92.9%. Figure 3 shows that the antioxidant potential of the oil, based on the scavenging activity of DPPH, revealed that a concentration above 142 μg mL^{-1} was necessary to reduce 50% of DPPH in the reaction medium.

Wojtunik, Ciesla, and Hajnos (2014) indicated that π bonds are responsible for the chain-breaking antioxidant activity of monoterpenes. They proved that blocking of conjugated double bonds leads to a decrease of the antioxidant property of monoterpenes. Several investigations have reported strong scavenging activity of terpenes found in citrus peel EO as α-pinene, linalool, citronellol, myrcene, γ-terpinene and limonene, among others (Behrendorff, Vickers, Chrysanthopoulos, & Nielsen, 2013; Sawamura, 2013; Singh et al., 2010). As antioxidant compounds donate a proton to the DPPH radical, greater weighting may be given to double bond positions that increase the availability of allylic protons (due to the weaker C-H bond at allyl groups) (Behrendorff et al., 2013).

Repellent and Fumigant activities

EO from *S. glutinosa* displayed the best repellent activity in higher concentration (15.73 nL cm^{-2}) after both exposure times, with a repellency percentage of 72±2 % at two hours and 67±2 % at four hours. This

was compared to a commercial repellent DEET (N, N-diethyl-toluamide) at the same concentration with an activity of 50±5% for the first two hours and 60±13 % after four hours (Table 2). However, the repellent activity of DEET was highest at 78.63 nL cm^{-2} (76% at 2 hours). These results show that the EO of *S. glutinosa* exhibited a repellent activity close to that of the commercial repellent.

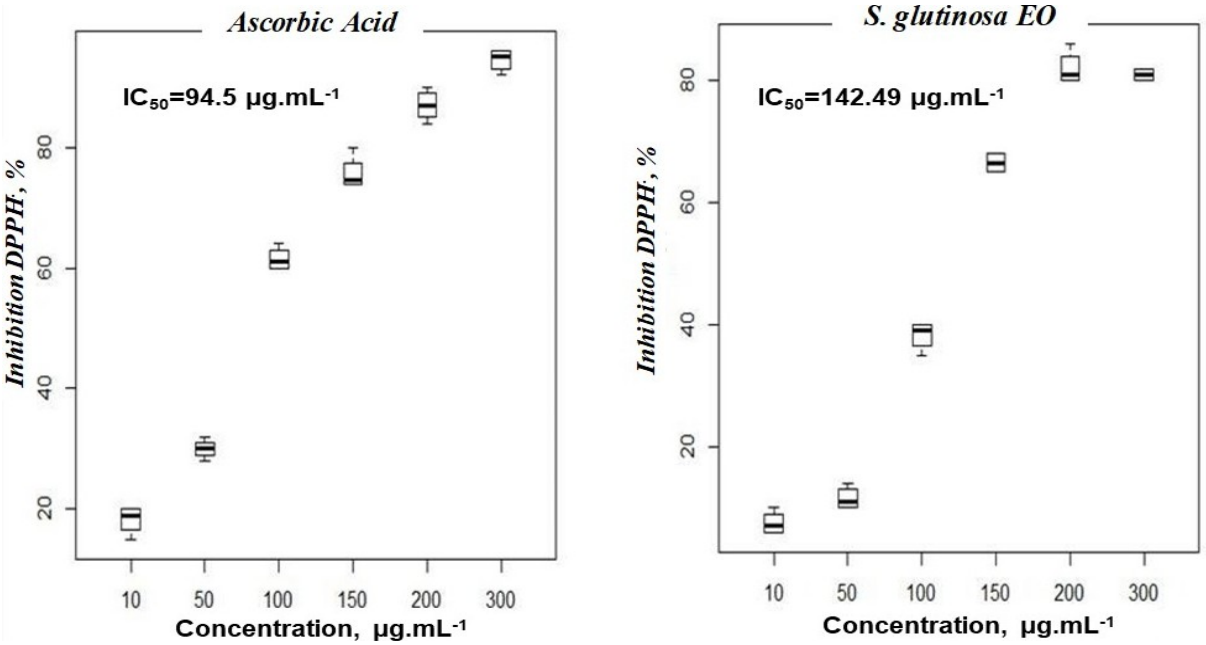

Figure 3. Antioxidant activity from *Swinglea glutinosa* essential oil measured by the DPPH· method compared to ascorbic acid.

Table 2. Repellent activity of essential oil from *Swinglea glutinosa* and terpenes against *Tribolium castaneum*.

Sample	Concentration (nL cm^{-2})	Repellency, %	
		Exposure time 2 hours	Exposure time 4 hours
S. glutinosa EO	0.13	23±2	13±2
	0.63	44±4	39±4
	3.15	57±3	49±2
	15.73	72±2	67±2
	78.63	69±3	58±2
Limonene	0.13	35±5	30±10
	0.63	58±2	50±2
	3.15	63±2	58±6
	15.73	90±5	86±5
	78.63	88±2	88±2
α-Pinene	0.13	70±5	55±5
	0.63	80±2	70±5
	3.15	88±4	68±2
	15.73	82±6	68±2
	78.63	80±5	60±5
β-Pinene	0.13	23±6	25±5
	0.63	30±10	36±7
	3.15	35±7	42±8
	15.73	83±6	74±6
	78.63	67±6	70±5
Myrcene	0.13	17±6	27±9
	0.63	35±6	43±7
	3.15	42±4	50±13
	15.73	60±10	73±5
	78.63	87±6	72±5
N,N-diethyl-toluamide (DEET)	0.13	10±2	16±4
	0.63	16±9	18±6
	3.15	40±11	54±12
	15.73	50±5	60±13
	78.63	76±6	78±5

[a]Repellent values = mean ± SE of the four replicates (SE = standard error). Paired *t* test ($p < 0.05$). [a] %R = [(Nc-Nt)/(Nc + Nt)] *100; Nc: No insects on control; Nt: No insects on treated area.

α-pinene exhibited the highest repellent activity against *T. castaneum* (88%) at 3.15 nL cm^{-2} after two hours of exposure and R-limonene deployed a higher repellent activity than those of α-pinene, β-pinene, and myrcene at 15.73 nL cm^{-2} (Table 2).

The fumigant toxicities of the EO and four monoterpenes were evaluated against adults of *T. castaneum*. The results of a data probit analysis, according to the lack of overlap in 95% fiducial limits, showed that pirimiphos- methyl (positive control, LC_{50}=87.4 μg mL^{-1} air) was significantly more toxic than *S. glutinosa* oil (LC_{50}=153.4 μg mL^{-1} air) as well as other individual monoterpenes assayed (Table 3). The mean lethal concentrations obtained were as follows: myrcene (LC_{50}=177.8 μg mL^{-1} air), (R) - limonene (LC_{50}=189.6 μg mL^{-1} air), α-pinene (213.1 μg mL^{-1} air), and β-pinene (LC_{50}=227.1 μg mL^{-1} air). However, the terpenes exhibited a weaker toxicity compared with pirimiphos- methyl.

Table 3. Fumigant toxicity from *Swinglea glutinosa* essential oil and its constituents against *Tribolium castaneum*.

Treatments	[a]95% FL	LC_{50}	$\chi^2(df)$	Slope ±SE
α-Pinene	[192.751; 233.492]	213.1	171.231	0.014 ± 0.001
β-Pinene	[69.350; 239.858]	227.1	254.862	0.0712 ± 0.032
Myrcene	[138.231; 211.942]	177.8	71.165	0.0059 ± 0.0008
R-Limonene	[170.428; 208.754]	189.6	182.176	0.015 ± 0.002
EO *S. glutinosa*	[140.732; 166.068]	153.4	2.3192	0.0002 ± 0.0009
Commercial insecticide (methyl pirimiphos)	[78.072; 96.718]	87.4	0.183	0.0169 ± 0.0015

[a] 95% lower and upper fiducial limits are shown in parenthesis.

Swinglea glutinosa EO, α-pinene, β-pinene, R-limonene and myrcene exhibited high fumigant activity (≥95%) at 350 μg mL^{-1} of air, after two hours of exposure, as shown in Figure 4. Significant differences were observed in the number of *T. castaneum* dead after treatment with *S. glutinosa* EO (LC_{50} =153.4 μg mL^{-1} air), [F(6.60) =89.54; $p < 0.001$] for all the doses tested when compared to the pirimiphos control group (LC_{50} =87.4 μg mL^{-1} air), [F(6.60) =12.66; $p < 0.001$] (Figure 4). The same was observed for the treatments with α-pinene (LC_{50} =213.1 μg mL^{-1} air) [F(6.60) = 57.14; $p < 0.001$]; β-pinene (LC_{50} =227.1 μg mL^{-1} air L) [F(6.60) = 52.72; $p < 0.001$]; myrcene (LC_{50} =177.8 μg mL^{-1} air) [F(6.60) =66.64; $p < 0.001$]; R-limonene (LC_{50} =189.6 μg mL^{-1} air L) [F(6.60) = 36.56; $p < 0.001$].

Several studies show that the repellent and fumigant properties of EO are associated with the presence of mono and sesquiterpene compounds such as carvacrol, *p*-cymene, thymol, α-pinene, myrcene (Kim et al. 2010), allyl cinnamate and allyl 2-furoate (Giner et al., 2013), 1,8-cineol, linalool, 2-heptyl acetate, 2-heptanol, citral (Ukeh & Umoetok, 2011), caryophyllene oxide, and caryophyllene (Kiran & Devi, 2007; Kim et al., 2010). The repellent action of some terpenes is similar to those of organophosphorus and carbamate insecticides, which are acetylcholinesterase inhibitors, causing rapid death by respiratory depression (Abdelgaleil, Mohamed, Badawy, & El-arami, 2009; López & Pascual-Villalobos, 2015). For example, limonene is a constituent of citrus EO recommended for the control of scale insects on ornamental plants and agricultural activities in the United States (Hollingsworth, 2005). Terpinen-4-ol, 1,8-cineole, linalool, R-(+)-limonene and geraniol were tested in vapor phase against different stages *of Tribolium confusum* (Stamopoulos, Damos, & Karagianidou, 2007). Malacrinò, Campolo, Laudani, and Palmeri (2016) reported that R (+)-limonene was able to reach 100% of efficacy or repellent activity on *T. castaneum* at a concentration of 85 mg L^{-1} air. Chaubey (2012) reported that inhibition of acetylcholinesterase enzyme (AchE) activity was observed in *S. oryzae* adults when fumigated with sublethal concentrations of *Zingiber officinale* and *Piper cubeba* EO (α-pinene and β-caryophyllene, respectively) alone or in binary combinations. Kim, Kang, and Park (2013) presented α-pinene as the strongest AchE activity inhibitor followed by β-pinene and limonene. The synergistic or complementary activities of different compounds present in the same oil play a vital role in the final insecticidal and/or repellent activity (Reis et al., 2016).

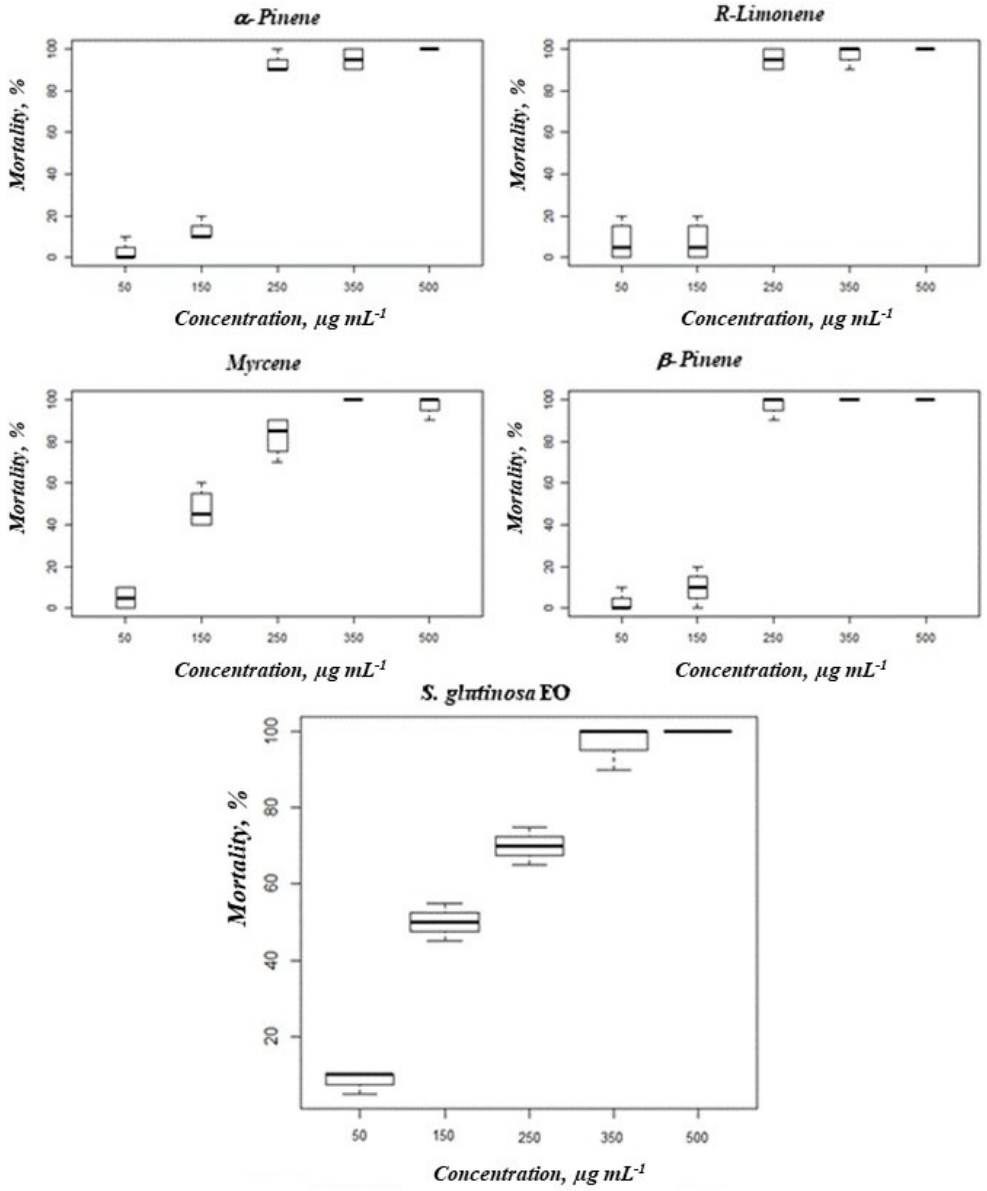

Figure 4. Fumigant activity from *Swinglea glutinosa* essential oil and four constituents against *Tribolium castaneum* at 24 hours after exposure.

Conclusion

We found high antioxidant, repellent and fumigant *in vitro* activities exhibited by the EO from *S. glutinosa* (Blanco) Merr., thus increasing interest in the possible application of essential oils as biocides in the control of insects and as protection of products against oxidation. The essential oil from *S. glutinosa* can be considered a natural source of biocides and antioxidants.

Acknowledgements

The authors acknowledge the support from the Research Group Support Program sponsored by the Vice-Presidency for Research at University of Cartagena; we would also like to thank Jasser Martinez for his technical support.

References

Abdelgaleil, S. A. M., Mohamed, M. I. E., Badawy, M. E. I., & El-arami, S. A. A. (2009). Fumigant and contact toxicities of monoterpenes to *Sitophilus oryzae* (L.) and *Tribolium castaneum* (Herbst) and their inhibitory effects on acetylcholinesterase activity. *Journal of Chemical Ecology, 35*, 518-525. doi: 10.1007/s10886-009-9635-3

Adams, R. P. (2007). Identification of essential oil components by gas chromatography/ mass spectrometry (4th ed.). Carol Stream, IL: Allured Publishing.

Biostat. (2009). *A statistical software BioStat, Version 2009*. Walnut, CA: Analyst Soft Robust Business Solutions.

Andrade, M. S., Ribeiro, L. P., Borgoni, P. C., Silva, M. F. G. F., Forim, M. R., Fernandes, J. B., ... Machado, M. A. (2016). Essential oil variation from twenty-two genotypes of citrus in Brazil-chemometric approach and repellency against *Diaphorina citri* Kuwayama. *Molecules, 21*(6), 814-823. doi: 10.3390/molecules21060814

Barra, A. (2009). Factors affecting chemical variability of essential oils: a review of recent developments. *Natural Product Communications, 4*(8), 1147-1154. doi: 10.1177/1934578x0900400827

Behrendorff, J. B. Y. H., Vickers, C. E., Chrysanthopoulos, P., & Nielsen, L. K. (2013). 2,2-Diphenyl-1-picrylhydrazyl as a screening tool for recombinant monoterpene biosynthesis. *Microbial Cell Factories, 12*(1), 76. doi: 10.1186/1475-2859-12-76

Benyelles, B., Allali, H., Dib, M. E. A., Djabou, N., Paolini, J., & Costa, J. (2017). Chemical composition variability of essential oils of *Daucus gracilis* Steinh. from Algeria. *Chemistry & Biodiversity, 14*(6), e1600490. doi: 10.1002/cbdv.201600490

Bueno-Sánchez, J. G., Martínez-Morales, J. R., Stashenko, E. E., & Ribón, W. (2009). Anti-tubercular activity of eleven aromatic and medicinal plants occurring in Colombia. *Biomédica – Revista del Instituto Nacional de Salud, 29*(1), 51-60. ISSN 0120-4157

Celano, R., Piccinelli, A. L., Pagano, I., Roscigno, G., Campone, L., De Falco, E., ... Rastrelli, L. (2017). Oil distillation wastewaters from aromatic herbs as new natural source of antioxidant compounds. *Food Research International, 99*(1), 298-307. doi: 10.1016/j.foodres.2017.05.036

Chaubey, M. K. (2012). Fumigant toxicity of essential oils and pure compounds against *Sitophilus oryzae* L. (Coleoptera: Curculionidae). *Biological Agriculture and Horticulture, 28*(2), 111-119. doi: 10.1080/01448765.2012.681352

Díaz, C., Arrázola Paternina, G., Ortega, F., & Gaviria, J. (2005). Caracterización del aceite esencial en la corteza del limón Swinglea (*Swinglea glutinosa*) por CG/EM. *Temas Agrarios, 10*(1), 22-28. doi: 10.21897/rta.v10i1.628

Duarte, A., Luís, A., Oleastro, M., & Domingues, F. C. (2016). Antioxidant properties of coriander essential oil and linalool and their potential to control *Campylobacter* spp. *Food Control, 61*, 115–122. doi: 10.1016/j.foodcont.2015.09.033

Estell, R. E., Fredrickson, E. L., & James, D. K. (2016). Effect of light intensity and wavelength on concentration of plant secondary metabolites in the leaves of *Flourensia cernua*. *Biochemical Systematics and Ecology, 65*,108–114. doi: 10.1016/j.bse.2016.02.019

Giner, M., Avilla, J., De Zutter, N., Ameye, M., Balcells, M., & Smagghe, G. (2013). Insecticidal and repellent action of allyl esters against *Acyrthosiphon pisum* (Hemiptera: Aphididae) and *Tribolium castaneum* (Coleoptera: Tenebrionidae). *Industrial Crops and Products, 47*, 63–68. doi: 10.1016/J.Indcrop.2013.02.019

González, J. O. W., Gutiérrez, M. M., Ferrero, A. A., & Band, B. F. (2014). Essential oils nanoformulations for stored product pest control – characterization and biological properties. *Chemosphere, 100*, 130-138. doi: 10.1016/j.chemosphere.2013.11.056

Hollingsworth, R. G. (2005). Limonene, a citrus extract, for control of mealybugs and scale insects. *Journal of Economic Entomology, 98*(3), 772-779. doi: 10.1603/0022-0493-98.3.772

Jaya, Singh, P., Prakash, B., & Dubey, N. K. (2014). Insecticidal activity *Ageratum conyzoides* L., *Coleus aromaticus* Benth. and *Hyptis suaveolens* (L.) poit essential oils as fumigant against storage grain insect *Tribolium castaneum* Herbst. *Journal Food Science and Technology, 51*(9), 2210-2215. doi: 10.1007/s13197-012-0698-8

Jaramillo-Colorado, B. E., Martelo, I. P., & Duarte, E. (2012). Antioxidant and repellent activities of the essential oil from Colombian *Triphasia trifolia* (Burm. f.) P. Wilson. *Journal of Agricultural and Food Chemistry, 60*(25), 6364-6368. doi: 10.1021/jf300461k

Jaramillo-Colorado, B. E., Suarez-López, S., & Marrugo-Santander, V. (2019). Volatile chemical composition of essential oil from *Bursera graveolens* (Kunth) Triana & Planch and their fumigant and repellent activities. *Acta Scientiarum. Biological Sciences, 41*, e46822. doi: 10.4025/actascibiolsci.v41i1.46822

Kim, S.-I., & Lee, D.-W. (2014). Toxicity of basil and orange essential oils and their components against two coleopteran stored products insect pests. *Journal of Asia-Pacific Entomology, 17*(1), 13-17. doi: 10.1016/j.aspen.2013.09.002

Kim, S.-W., Kang, J., & Park, I.-K. (2013). Fumigant toxicity of Apiaceae essential oils and their constituents against *Sitophilus oryzae* and their acetylcholinesterase inhibitory activity. *Journal of Asia-Pacific Entomology,16*(4), 443-448. doi: 10.1016/j.aspen.2013.07.002

Kim, S.-I., Yoon, J.-S., Jung, J. W., Hong, K.-B., Ahn, Y.-J., & Kwon, H. W. (2010). Toxicity and repellency of origanum essential oil and its components against *Tribolium castaneum* (Coleoptera: Tenebrionidae) adults. *Journal of Asia-Pacific Entomology, 13*(4), 369-373. doi: 10.1016/j.aspen.2010.06.011

Kiran, S. R., & Devi, P. S. (2007). Evaluation of mosquitocidal activity of essential oil and sesquiterpenes from leaves of *Chloroxylon swietenia* DC. *Parasitology Research, 101*, 413-418. doi: 10.1007/s00436-007-0485-z

Koutsaviti, A., Antonopoulou, V., Vlassi, A., Antonatos, S., Michaelakis, A., Papachristos, D. P., & Tzakou, O. (2018). Chemical composition and fumigant activity of essential oils from six plant families against *Sitophilus oryzae* (Col: Curculionidae). *Journal of Pest Science, 91*, 873-88. doi: 10.1007/s10340-017-0934-0

Lee, B.-H., Annis, P. C., Turmaalii, F., & Choi, W.-S. (2004). Fumigant toxicity of essential oils from the *Myrtaceae* family and 1,8-cineole against 3 major stored-grain insects. *Journal of Stored Products Research, 40*(5), 553-564. doi: 10.1016/j.jspr.2003.09.001

López, M. D., & Pascual-Villalobos, M. J. (2015). Are monoterpenoids and phenylpropanoids efficient inhibitors of acetylcholinesterase from stored product insect strains? *Flavour and Fragrance Journal, 30*(1), 108–112. doi: 10.1002/ffj.3220

Malacrinò, A., Campolo, O., Laudani, F., & Palmeri, V. (2016). Fumigant and repellent activity of limonene enantiomers against *Tribolium confusum* du Val. *Neotropical Entomology, 45*(5), 597-603. doi: 10.1007/s13744-016-0402-1

Nenaah, G. E. (2014). Bioactivity of powders and essential oils of three Asteraceae plants as post-harvest grain protectants against three major coleopteran pests. *Journal of Asia-Pacific Entomology, 17*, 701-709. doi: 10.1016/j.aspen.2014.07.003

Pino, J. A., Marbot, R., & Fuentes, V. (2006). Aromatic plants from Western Cuba IV. Composition of the leaf oils of *Clausena lansium* (Lour.) skeels and *Swinglea glutinosa* (Blanco) Merr. *The Journal of Essential Oil Research, 18*(2), 139-41. doi: 10.1080/10412905.2006.9699044

Prieto, J. A., Patiño, O. J., Delgado, W. A., Moreno, J. P., & Cuca, L. E. (2011). Chemical composition, insecticidal, and antifungal activities of fruit essential oils of three Colombian *Zanthoxylum* species. *Chilean Journal of Agricultural Research, 71*(1), 73-82. doi: 10.4067/S0718-58392011000100009

Reis, S. L., Mantello, A. G., Macedo, J. M., Gelfuso, E. A., Silva, C. P., Fachin, A. L., ... Beleboni, R. O. (2016). Typical monoterpenes as insecticides and repellents against stored grain pests. *Molecules, 21*(3), 258. doi: 10.3390/molecules21030258

Ricciardi, G., Cicció, J. F., Ocampo, R., Lorenzo, D., Ricciardi, A., Bandoni, A., & Dellacassa, E. (2009). Chemical variability of essential oils of *Lippia alba* (Miller) N. E. Brown Growing in Costa Rica and Argentina. *Natural Product Communications*, *6*, 853-858. doi: 10.1177/1934578X0900400623

Rocha, L. G., Almeida, J. R. G. S., Macedo, R. O., & Barbosa-Filho, J. M. (2005). A review of natural products with antileishmanial activity. *Phytomedicine, 12*(6-7), 514–535. doi: 10.1016/j.phymed.2003.10.006

Sawamura, M. (Ed.). (2013). *Citrus essential oils: flavor and fragrance*. Hoboken, NJ: John Wiley & Sons.

Silva, A. S., Costa, D., Albuquerque, T. G., Buonocore, G. G., Ramos, F., Castilho, M. C. ... Costa, H. S. (2014). Trends in the use of natural antioxidants in active food packaging: a review. *Food Additives & Contaminants - Part A, 31*(3), 374–395. doi: 10.1080/19440049.2013.879215

Singh, P., Shukla, R., Prakash, B., Kumar, A., Singh, S., Mishra, P. K., & Dubey, N. K. (2010). Chemical profile, antifungal, antiaflatoxigenic and antioxidant activity of *Citrus maxima* Burm. and *Citrus sinensis* (L.) Osbeck essential oils and their cyclic monoterpene, DL-limonene. *Food and Chemical Toxicology, 48*(6), 1734-1740. doi: 10.1016/j.fct.2010.04.001

Souza, T. D. S., Ferreira, M. F. S., Menini, L., Souza, J. R. C. L., Bernardes, C. O., & Ferreira, A. (2018). Chemotype diversity of *Psidium guajava* L. *Phytochemistry, 153*,129-137. doi: 10.1016/j.phytochem.2018.06.006

Spitaler, R., Schlorhaufer, P. D., Ellmerer, E. P., Merfort, I., Bortenschlager, S., Stuppner, H, & Zidorn, C. (2006). Altitudinal variation of secondary metabolite profiles in flowering heads of *Arnica montana* cv. ARBO. *Phytochemistry, 67*(4), 409-417. doi: 10.1016/j.phytochem.2005.11.018

Stamopoulos, D. C., Damos, P., & Karagianidou, G. (2007). Bioactivity of five monoterpenoid vapours to *Tribolium confusum* (du Val) (Coleoptera: Tenebrionidae). *Journal of Stored Products Research, 43*(4), 571–7. doi: 10.1016/j.jspr.2007.03.007

Stashenko, E. E., Jaramillo, B. E., & Martínez, J. R. (2004). Comparison of different extraction methods for the analysis of volatile secondary metabolites of *Lippia alba* (Mill.) N.E. Brown, grown in Colombia, and evaluation of its *in vitro* antioxidant activity. *Journal of Chromatography A., 1025*(1), 93-103. doi: 10.1016/j.chroma.2003.10.058

Ukeh, D. A., & Umoetok, S. B. A. (2011). Repellent effects of five monoterpenoid odours against *Tribolium castaneum* (Herbst) and *Rhyzopertha dominica* (F.) in Calabar, Nigeria. *Crop Protection, 30*(10), 1351-1355. doi: 10.1016/j.cropro.2011.05.016

Weniger, B., Um, B.-H., Valentin, A., Estrada, A., Lobstein, A., Anton, R., ... Sauvain, M. (2001a). Bioactive acridone alkaloids from *Swinglea glutinosa*. *Journal of Natural Products, 64*(9), 1221–1223. doi: 10.1021/np0005762

Weniger, B., Robledo, S., Arango, G. J., Deharo, E., Aragón, R., Muñoz, V., ... Anton, R. (2001b). Antiprotozoal activities of Colombian plants. *Journal of Ethnopharmacology, 78*,193–200. doi: 10.1016/s0378-8741(01)00346-4

Wojtunik, K. A., Ciesla, L. M., & Hajnos, M. W. (2014). Model studies on the antioxidant activity of common terpenoid constituents of essential oils by means of the 2,2-diphenyl-1-picrylhydrazyl method. *Journal of Agricultural and Food Chemistry, 62*(37), 9088–9094. doi: 10.1021/jf502857s

Zhang, J. S., Zhao, N. N., Liu, Q. Z., Liu, Z. L., Du, S. S., Zhou, L., & Deng, Z. W. (2011). Repellent constituents of essential oil of *Cymbopogon distans* aerial parts against two stored-product insects. *Journal of Agricultural and Food Chemistry, 59*(18), 9910–9915. doi: 10.1021/jf202266n