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Toxicological Activity of Some Plant Essential Oils Against *Tribolium castaneum* and *Culex pipiens* Larvae

Ahmed M. El-Sabrout [1], Mohamed Z. M. Salem [2,*], May Bin-Jumah [3] and Ahmed A. Allam [4]

1 Department of Applied Entomology and Zoology, Faculty of Agriculture (EL-Shatby), Alexandria University, Alexandria 21545, Egypt; elsabroutahmed@alexu.edu.eg
2 Forestry and Wood Technology Department, Faculty of Agriculture (EL-Shatby), Alexandria University, Alexandria 21545, Egypt
3 Biology Department, College of Science, Princess Nourah bint Abdulrahman University, Riyadh 11671, BO. Box 24428, Saudi Arabia; may_binjumah@outlook.com
4 Department of Zoology, Faculty of Science, Beni-suef University, Beni-suef 65211, Egypt; allam1081981@yahoo.com
* Correspondence: zidan_forest@yahoo.com

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Abstract: In the present work, essential oils (EOs) from *Schinus terebinthifolius* (ripe and unripe fruits and leaves), *Origanum majorana* (air-dried aerial parts), and *Psidium guajava* (leaves) were assayed for their insecticidal activity against red flour beetle (*Tribolium castaneum*) and Culex mosquito larvae (*Culex pipiens*). Several components were identified in the EOs using Gas chromatography–mass spectrometry (GC/MS), of which Δ-3-carene (25.9%), γ-terpinene (19.4), and γ-elemene (7.1%) were the major ones in *S. terebinthifolius* ripe fruits, α-pinene (48.9%), germacrene D (12.9%), and α-thujene (7.7%) in *S. terebinthifolius* unripe fruits, γ-elemene (11.7%), spathulenol (10.1%), β-elemene (9.2%), and *p*-cymene (9.1%) in *S. terebinthifolius* leaves, α-pinene (25.5%), (*E*)-caryophyllene (15.7%), (*E*)-nerolidol (16.7%), and cedran-8-ol (8.8%) in *P. guajava* leaves, and terpinen-4-ol (21.7%), γ-terpinene (16.5%), and sabinene (10.1%) in *O. majorana* air-dried aerial parts. The lethal concentration (LC_{50}) was calculated for tested EOs at different time periods (after 6, 12, 24, 48, and 72 h). After 6 h of treatment, the LC_{50} was 33.3 and 6.8 µg/L air for *S. terebinthifolius* ripe and unripe fruits, respectively, and >40 µg/L air for EOs of *S. terebinthifolius* leaves, *O. majorana*air-dried aerial parts, and *P. guajava* leaves. After 24 h of treatment, the LC_{50} was 4.2, <2, 5, >40, and 6.1 µg/L air for EOs of *S. terebinthifolius* ripe fruits and leaves, *O. majorana* leaves, and *P. guajava* leaves, respectively. On the other hand, the LC_{50} values decreased when the exposed period was increased to 72 h, and were <2 µg/L air for EOs of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves along with *P. guajava* leaves, respectively, and 37.912 for EO of *O. majorana* leaves. The LC_{50} value after 24 h of exposure of *S. terebinthifolius* unripe fruit EO was under 2 µg/L air, which means that the EO of *S. terebinthifolius* ripe fruit had a strong effect on adult *T. castaneum* adults compared to other tested EOs using the fumigation method. The present data confirm that the EOs of *O. majorana* leaves and *S. terebinthifolius* unripe fruits and leaves were more effective as larvicide than the EOs of *S. terebinthifolius* ripe fruits and *P. guajava* leaves on *C. pipiens* at a higher concentration (100 mg/L) when applied by the dipping method. EOs from *S. terebinthifolius* unripe or ripe fruits and leaves and *P. guajava* leaves were more effective as adulticide than EO of *O. majorana* leaves against *T. castaneum* when applied by the fumigant method.

Keywords: physiological effects; essential oils; Culex mosquitoes; red flour beetle

1. Introduction

Botanicals are basically secondary metabolites that serve as a defence mechanism for plants to withstand the continuous selection pressure from herbivore predators and other environmental factors.

Plants produce essential oils (EOs), terpenoids, alkaloids, steroids, and phenolics in which many have medicinal, insecticidal, and larvicidal activities [1–10]. Plant EOs, in general, have been recognized as an important natural resource for insecticides [6,8,10–15].

In recent years, and due to the lack of novel insecticides and the high cost of synthetic insecticides, the use of many synthetic insecticides formerly used in mosquito control programs has been limited [5,16,17]. Therefore, the search for alternatives to synthetic chemical insecticides and natural extracts and EOs for vector and pest management that pose little threat to human and environmental health has increased [10,18,19].

Schinus terebinthifolius Raddi (Sapindales: Anacardiaceae) EOs have been reported to have insecticidal properties against *Stegomyia aegypti* Linnaeus in Hasselquist (Diptera: Culicidae), *Anopheles gambiae* sensu lato (Diptera: Culicidae), and *Culex quinquefasciatus* Say (Diptera: Culicidae) [20]. EOs of mature and immature *S. terebinthifolius* showed strong insecticidal activity against *Spodoptera littoralis* Boisduval (Lepidoptera: Noctuidae) and *Phthorimaea operculella* Zeller (Lepidoptera: Gelechiidae) [21]. EO of 1% *S. terebinthifolius* fruit showed great repellency against *Bemisia tabaci* Gennadius (Hemiptera: Aleyrodidae) and potency against *Trialeurodes ricini* Misra (Homoptera: Aleyrodidae) adults [8]. Leaf extract of *S. terebinthifolius* caused damage to the midgut of *Aedes aegypti* Linnaeus in Hasselquist (Diptera: Culicidae) larvae [22]. EOs from fruits and seeds showed mosquitocidal activity against *An. gambiae*, *An. Arabiensis* Patton (Diptera: Culicidae), and *C. quinquefasciatus* [23].

EO obtained from leaves of guava (*Psidium guajava* L., Myrtales: Myrtaceae) has shown promising larvicidal activity against *A. aegypti*, with LC_{50} ranging from 39.48 to 64.25 µg/mL [24] and 24.7 µg/mL [25]. *P. guajava* leaf EO showed notable larvicidal activity against *Chaoborus plumicornis* F. (Diptera: Chaoboridae) and insecticidal activity against *Drosophila melanogaster* Meigen (Diptera: Drosophilidae) [26].

Origanum majorana L. (Lamiales: Lamiaceae) EO is composed of majority constituents, which gives it biological activities [27]. The important larvicidal activity observed by the EO of *O. majorana* could be explained by its chemical composition and the action or effect of the majority compound. Azizi et al. [28] and Pavela [29] reported that *Origunum* species had insecticidal activity against insects. In general, plant EOs have been recognized as an important natural resource for insecticides [5].

Mosquitoes spread serious human diseases such as malaria, yellow fever, dengue, and filariasis [30]. Overall, 212 million cases of malaria and 429,000 deaths were reported worldwide [31]. In urban and rural of Egyptian areas, *C. pipiens* L. (Diptera: Culicidae) is the most common mosquito species that causes health risks to humans. The major insects of stored grains and pulses of many countries such as India, Egypt, and others are rice weevil, *Sitophilus oryzae* L. (Coleoptera: Curculionidae), granary weevil, *S. granaries* L. (Coleoptera L.: Curculionidae), lesser grain borer, *Rhyzopertha dominica* Fabricius (Coleoptera: Bostrichidae), Khapra beetle, *Trogoderma granarium* Everts (Coleoptera: Dermestidae), red flour beetle, *Tribolium castaneum* Herbst (Coleoptera: Tenebrionidae), saw-toothed grain beetle, *Oryzaephilus surinamensis* L. (Coleoptera: Silvanidae), and others [12,13,32].

Many researchers have reported that plant parts, EOs, extracts, or powders mixed with grains reduced insect oviposition, egg hatchability, and postembryonic development, inhibited reproduction, and induced mortality of insect eggs and progeny production of stored product insects [33–39]. Most recently, *Mentha piperita* L. (Lamiales: Lamiaceae) leaf EO at concentrations of 20 and 40 µL/L showed mortality against *T. castaneum* at 65% and 90%, respectively [39], with the fumigation method. Additionally, application of *Taxodium* EOs from different locations in Egypt showed LC_{50} against *T. castaneum* with values of 66.4 and 72.5 µL/L, respectively [13]. EOs of *Ocimum basilicum* L. (Lamiales: Lamiaceae) and *Eucalyptus gomphocephala* DC (Myrtales: Myrtaceae) showed larvicidal activity against mosquitos with LC_{50} values of 22 and 30 mg/L, respectively [10].

The aim of the present study was to evaluate the larvicidal and mosquitocidal activity of five EOs from *Schinus terebinthifolius* (ripe and unripe fruits and leaves), *Origanum majorana* (aerial parts), and *Psidium guajava* (leaves) against *C. pipiens* by the dipping method. An experiment was also

conducted to evaluate adulticide activity of the tested EOs against *T. castaneum* using the fumigant method at several concentrations during different exposure times.

2. Materials and Methods

2.1. Plant Materials and Extraction of Essential Oils

Freshly collected samples (200 g) of ripe and unripe *Schinus terebinthifolius* fruits, *S. terebinthifolius* leaves, *Origanum majorana* air-dried aerial parts, and *Psidium guajava* leaves were cut into small pieces using scissors and hydro-distilled for 3 h using a Clevenger-type apparatus [40]. The collected essential oils (EOs) were dried over anhydrous sodium sulphate (Sigma-Aldrich, Darmstadt, Germany). The EO yields were 3.50, 2.75, 1.15, 3.50, and 0.5 mL/100 g plant material for *S. terebinthifolius* ripe fruits, *S. terebinthifolius* unripe fruits, *S. terebinthifolius* leaves, *O. majorana* air-dried aerial parts, and *P. guajava* leaves, respectively.

2.2. GC-MS Analysis Conditions

Analysis of the EOs was performed using an Agilent 6890 gas chromatograph-mass spectrometer (GC-MS) equipped with an Agilent mass spectrometry detector with a direct capillary interface and HP-5MS fused silica capillary column (30 m × 0.32 mm × 0.25 µm film thickness) (Thermo Scientific, Austin, TX, USA). The program temperature and samples were carried out following previous published works [41,42]. Identification of the constituents was performed based on an mass spectra (MS) library search (National Institute of Standards and Technology (NIST) and Wiley), and by comparing with data in the MS literature [43,44]. The EO compounds were confirmed using the Xcalibur 3.0 data system (3.0, Thermo Fisher Scientific Inc., Austin, TX, USA, 2014) with measuring their Standard Index and Reverse Standard Index [45–48].

2.3. Red Flour Beetle Rearing

Red flour beetle (*T. castaneum*) adults and larvae were reared on wheat flour under laboratory conditions of 27 ± 3 °C and 70 ± 5% relative humidity (RH).

2.4. Mosquito Rearing

A susceptible strain of mosquito larvae, *C. pipiens*, was obtained from the Research Institute of Medical Entomology, Dokki, Egypt. The continuously breeding mosquito colony was maintained in an insectary at 27 ± 2 °C, 75 ± 5% RH at the Department of Applied Entomology and Zoology, Alexandria University, Egypt. The rearing of larvae and feeding of adults were done according to the method of Zahran and Abdelgaleil [30] with some modification.

2.5. Fumigant Assay on Red Flour Beetle

The fumigation experiment was carried out at 26 ± 1 °C and 65 ± 5% RH. Newly emerged adults (1–15 days old) were used in fumigant studies. The fumigant method for the 5 EOs was tested against *T. castaneum* adults. Glass jars (1 L) were used as fumigation chambers (replicates) and filter paper pieces (3 × 3 cm) were joined to the undersurface of the screw caps of the jars. The 5 EOs were applied to the filter paper pieces by 2, 5, 10, 20, and 40 µL/L air. Every jar as a replicate containing 20 insects as treatment and control were repeated 3 times. Filter paper pieces were treated with acetone (Loba Chemie Pvt. Ltd., laboratory reagents & fine chemicals, Mumbai, India) alone as a control. Control insects were kept under the same conditions with acetone. The insect mortality percentage was observed after 6, 24, 48, and 72 h of treatment and the lethal concentration causing 50% mortality (LC_{50}) expressed as mg/L air was calculated from log-concentration mortality regression lines. Insects were considered dead when no leg or antenna movements were recorded. The fumigant method assay was performed as described by Finney [49], El-Bakry et al. [50], and Huang et al. [51].

2.6. Bioassay Toxicity of Mosquitos

The tested EOs of *S. terebinthifolius* ripe and unripe fruits and leaves, *O. majorana* air-dried aerial parts, and *P. guajava* leaves were examined for bioassays [30] on newly second instar larvae of *C. pipiens*. This experiment was conducted by the dipping method using four concentrations of each oil (10, 25, 50, and 100 mg/L). Three replicates for each concentration were prepared. Each replicate including 50 *C. pipiens* larvae was separately put into a 200-mL plastic cup containing 100 mL of distilled water. The tested EO solutions were added to the cups and suspended with 0.05 mL of Tween-20.

The *C. pipiens* larvae were exposed to 10, 25, 50, and 100 mg/L of tested EOs in 100 mL of distilled water. In the control cups, only solvent (absolute acetone) was dissolved in the water. Treated and control larvae were held in the same conditions used for colony rearing. Larval mortality was recorded 24 and 48 h after treatment and continued to the end of the larval stage. Larvae were considered dead when they did not rise to the surface of the solution or when they did not respond to a stimulus. Additionally, pupal and adult mortality was calculated. The longevity parameter was calculated for each development stage of *C. pipiens*.

2.7. Statistical Analysis

The mortality data were subjected to probit analysis to estimate the lethal concentration (LC_{50}) values of tested EOs. Data for the mortality percentage of *T. castaneum* as affected by 3 factors of different concentrations of 5 EOs with different time periods were statistically analyzed using factorial design. To study the significance effects of oil concentration and oil source as well as their interaction as insecticidal activity against *C. pipiens*, two-way analysis of variance (ANOVA) with a two-factor test was used. All analyses were done using the SAS system (Release 8.02, SAS Institute: Cary, NC, USA, 2001) [52]. Comparisons among means were recorded using $LSD_{0.05}$.

3. Results

3.1. Chemical Composition of Essential Oils

Table 1 shows the chemical composition of the essential oils (EOs) from ripe/unripe fruits and leaves of *S. terebinthifolius*. The major components of the oils were Δ-3-carene (25.95%), γ-terpinene (19.45%), γ-elemene (7.07%), α-ylangene (5.34%), *p*-cymene (4.55%), and *D*-limonene (2.94%) in ripe fruits, α-pinene (48.96%), germacrene D (12.95%), α-thujene (7.75%), sabinene (4.96%), and *D*-limonene (4.32%) in unripe fruits, and γ-elemene (11.74%), spathulenol (10.13%), β-elemene (9.24%), *p*-cymene (9.19%), β-phellandrene (5.93%), naphthalene (5.47%), and *D*-limonene (2.22%) in leaves. The main compounds in EO of *P. guajava* leaves were α-pinene (25.51%), (*E*)-caryophyllene (15.74%), (*E*)-nerolidol (16.75%), and cedran-8-ol (8.81%) (Table 2). The main compounds in EO of *O. majorana* air-dried aerial parts were terpinen-4-ol (21.74%), γ-terpinene (16.5%), sabinene (10.14%), γ-terpinyl acetate (6.76%), α-terpinene (6.19%), terpinolene (5.72%), and α-terpineol (5.14%) (Table 3).

Table 1. Chemical composition of essential oils from *S. terebinthifolius* ripe/unripe fruits and leaves.

Compound Name	*S. terebinthifolius* Ripe Fruit Oil	*S. terebinthifolius* Unripe Fruit Oil	*S. terebinthifolius* Leaf Oil
α-Pinene	–	48.9 (696–696)	4.1 (933–933)
Δ-3-Carene	25.9 (675–689)	–	–
β-Pinene	–	–	0.7 (880–888)
Terpinen-4-ol	–	–	0.2 (869–876)
γ-Terpinene	19.4 (657–709)	1.8 (818–821)	–
α-Thujene	–	7.7 (670–736)	0.5 (907–929)
D-Limonene	2.9 (918–919)	4.3 (838–840)	2.2 (921–928)
Sabinene	–	4.9 (861–873)	–
β-Phellandrene	1.2 (868–871)	–	5.9 (833–835)
p-Cymene	4.5 (889–892)	–	9.1 (890–890)
Terpinolene	1.3 (891–894)	1.8 (912–913)	0.4 (892–915)
Cymene	–	2.9 (889–890)	–
Linalool	–	–	0.5 (896–905)
α,2-Dimethyl styrene	–	–	0.4 (897–926)
Carvenone	0.3 (777–788)	0.1 (780–790)	0.7 (821–842)
α-Terpineol	–	–	0.5 (903–919)
trans-Piperitol	–	–	–
cis-Sabinol	0.6 (911–920)	0.3 (904–915)	0.7 (869–890)
p-Cymen-8-ol	–	–	0.2 (863–902)
γ-Terpinyl acetate	–	–	–
Δ-Elemene	2.1 (866–873)	1.8 (871–899)	0.5 (884–888)
Naphthalene	–	–	5.4 (745–847)
γ-Muurolene	0.45 (734–757)	0.21 (728–736)	0.3 (825–837)
β-Elemene	–	–	9.2 (896–899)
Citronellyl acetate	1.1 (807–847)	0.4 (748–805)	–
Aromandendrene	–	–	3.9 (780–789)
Neryl acetate	–	–	–
α-Ylangene	5.3 (853–856)	2.3 (854–859)	0.1 (781–787)
γ-Elemene	7.1 (813–848)	3.7 (830–853)	11.7 (892–895)
IsoGermacrene-D	0.9 (883–896)	0.7 (885–893)	0.6 (911–868)
γ-Cadinene	1.4 (817–837)	0.9 (805–850)	0.3 (848–887)
γ-Selinene	–	–	0.3 (877–879)
Germacrene D	14.7 (893–894)	12.9 (886–889)	3.4 (908–911)
β-Selinene	–	–	1.7 (912–933)
β-Copaene	1.1 (855–867)	0.6 (850–864)	0.2 (868–883)
Valencene	0.3 (831–844)	–	–
(+)-Lepidozene	–	0.2 (830–886)	2.1 (831–898)

Table 1. *Cont.*

Compound Name	*S. terebinthifolius* Ripe Fruit Oil	*S. terebinthifolius* Unripe Fruit Oil	*S. terebinthifolius* Leaf Oil
Δ-Cadinene	0.7 (903–908)	0.2 (869–892)	1.1 (906–914)
Guaiene	–	–	1.6 (827–828)
Selina-3,7(11)-diene	–	–	0.4 (840–868)
Calamenene	–	–	0.3 (793–897)
trans-Sesquisabinene hydrate	–	0.1 (733–831)	–
Globulol	–	–	0.5 (34–863)
Elemoyl acetate	0.2 (830–870)	0.1 (797–817)	–
α-Costol	–	–	0.4 (702–756)
4(15),5,10(14)-Germacratrien-1-ol	0.3 (763–772)	0.2 (816–822)	1.4 (804–816)
α-Calacorene	–	–	0.1 (788–923)
Spathulenol	1.2 (872–895)	0.3 (867–869)	10.1 (832–852)
4(14)-Salvialen-1-one	0.2 (800–851)	–	–
Rosifoliol	–	–	0.7 (782–873)
β-Neoclovene	–	0.1 (788–796)	–
Eudesma-4,11-dien-2-ol	0.5 (794–800)	0.1 (761–767)	0.1 (774–800)
Cubebol	–	–	0.1 (765–822)
Isospathulenol	2.3 (872–876)	1.1 (844–849)	3.5 (865–876)
Isoaromadendrene epoxide	–	0.1 (780–790)	–
β-Caryophyllene oxide	0.3 (777–799)	–	–
11-Hexadecynal	–	–	1.6 (729–760)
α-Cadinol	0.2 (852–863)	–	0.3 (821–852)
Neointermedeol	–	–	1.3 (828–861)
β-Vetivol	2.1 (803–814)	0.1 (810–823)	0.9 (804–823)
α-Costol	–	–	0.8 (816–847)
β-Isonootkatol	0.2 (813–826)	–	–
Aristolene epoxide	–	–	0.5 (763–787)
8-Hydroxy-endo-Cycloisolongifolene	0.2 (770–781)	–	–
Aromadendrene oxide-(2)	–	–	1.0 (823–878)
cis-9-Hexadecenal	–	–	0.1 (721–745)
(Z)-9,17-Octadecadienal	–	–	0.1 (713–805)
Anthracene	–	–	0.2 (908–953)
Viridiflorene	–	–	0.5 (869–870)

Values are relative quantity (%) (standard Index–reverse standard index).

Table 2. Chemical composition of essential oil from *P. guajava* leaves.

Compound Name	Relative Quantity (%)	Standard Index	Reverse Standard Index
α-Pinene	25.5	834	836
Δ-3-Carene	8.8	787	788
β-Pinene	0.5	902	912
Camphene	0.2	871	875
trans-Isolimonene	0.2	864	865
Terpinen-4-ol	0.3	908	910
β-Fenchol	0.5	830	849
L-Bornyl acetate	2.2	929	929
trans-Pinocarvyl acetate	0.2	818	851
Bornylene	0.5	820	829
Bicycloelemene	0.2	776	847
α-Patchoulene	0.3	778	796
Cedrene	0.11	880	898
β-Chamigrene	0.2	895	913
β-Himachalene	0.3	901	910
Thujopsene-(I2)	0.1	890	900
Cuparene	2.6	892	894
(*E*)-Caryophyllene	15.7	872	874
γ-Muurolene	0.2	881	896
(E)-Nerolidol	16.7	870	871
Aristolene epoxide	0.7	757	803
Cedran-8-ol	8.8	878	882
Widdrol	0.6	747	750
Isospathulenol	0.2	815	861
α-Bisabolol	2.1	834	885
Ledene oxide-(II)	1.1	835	837
1,3,3-Trimethyl-2-(2-methyl-cyclopropyl)-cyclohexene	1.4	758	805
8-Hydroxy-endo-Cycloisolongifolene	0.3	812	836
Calarene epoxide	0.1	753	814
Viridiflorene	0.1	771	781
Labda-8(20),12,14-triene	0.1	791	794
13-Epimanool	3.3	778	785

Table 3. Chemical composition of essential oil from *O. majorana* aerial parts.

Compound Name	Relative Quantity (%)	Standard Index	Reverse Standard Index
γ-Terpinene	16.5	877	880
α-Thujene	3.2	915	922
Sabinene	10.1	915	924
α-Terpinene	6.1	895	897
β-Thujene	0.9	910	931
β-Phellandrene	0.7	910	915
p-Cymene	0.8	839	875
Terpinolene	5.7	898	900
γ-Terpineol	5.1	845	847
4-Thujanol	2.6	899	902
trans-4-Thujanol	4.8	916	918
cis-Para-2-menthen-1-ol	0.3	884	886
(*E*)-Caryophyllene	2.5	894	895
Terpinen-4-ol	21.7	876	882
α-Terpineol	5.1	914	925
trans-Piperitol	0.1	833	865
γ-Terpinyl acetate	6.7	830	830
Bornyl acetate	0.3	911	945
Terpinyl propionate	0.6	845	886
Neryl acetate	0.5	891	897
γ-Elemene	1.4	886	906
α-Humulene	0.1	863	880
Germacrene D	1.6	892	894
Spathulenol	0.5	902	903
Isospathulenol	0.2	828	843
β-Caryophyllene oxide	0.2	858	867

3.2. *Red Flour Beetle Experiment*

Fumigant Toxicity of Tested Essential Oils

Figure 1 shows the statistical significance of the main effects (oil source, oil concentration, and time of exposure). Oil of *S. terebinthifolius* unripe fruits showed the highest mortality of *T. castaneum* (Figure 1A). With increased oil concentration and exposure time, mortality increased significantly (Figure 1B,C). Additionally, the interaction between two factors (Figure 1D–F) showed significant effects on the mortality percentage of *T. castaneum.*

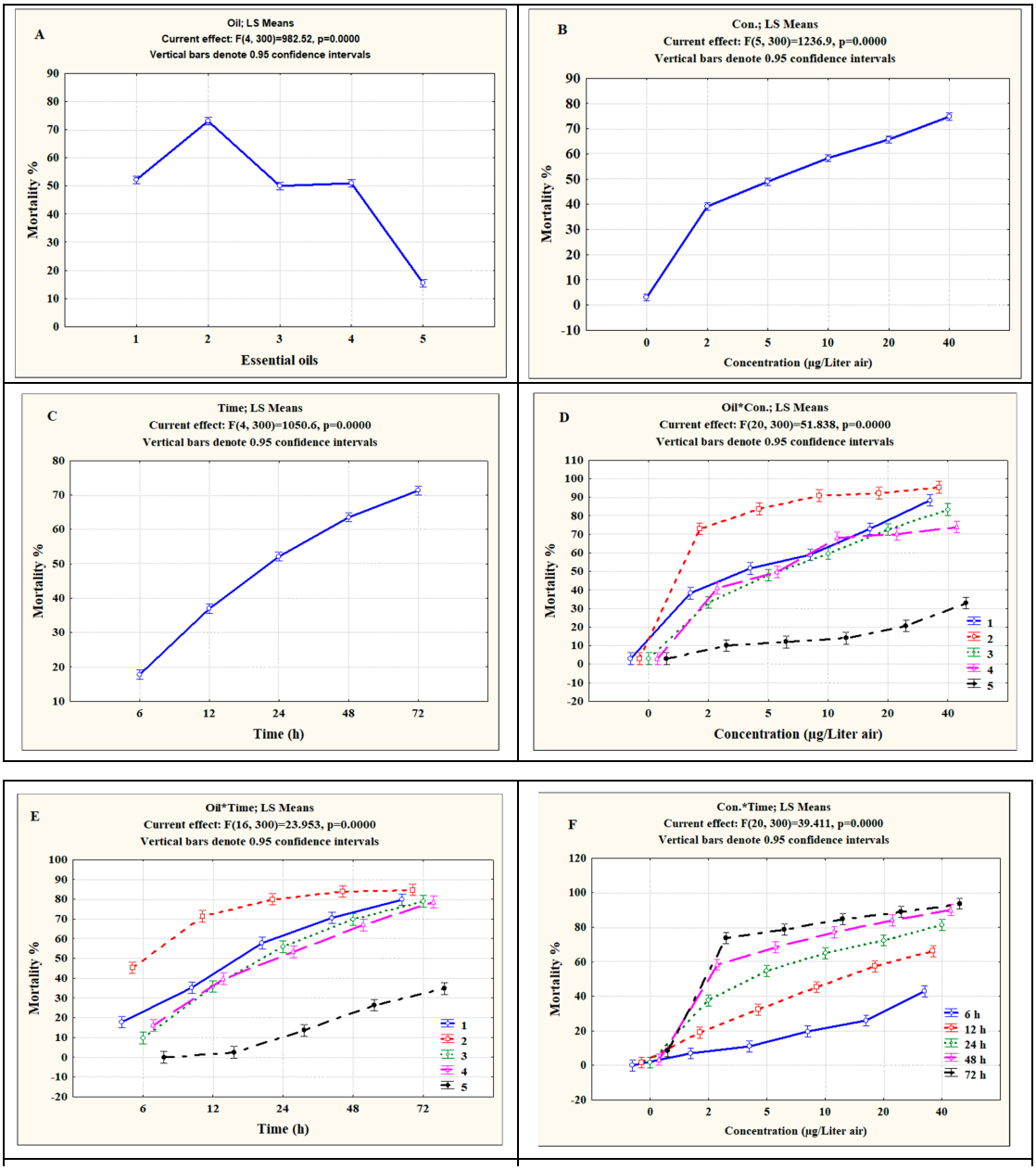

Figure 1. Effects among parameters of tested EOs on *T. castaneum* mortality: concentrations (0, 10, 25, 50, and 100 µg/L air) and exposure time (6, 12, 24, and 72 h). (**A**–**C**) Results of *T. castaneum* mortality when only one parameter effect was studied separately. (**D**–**F**) Results when effects between two parameters on *T. castaneum* mortality were studied together: (**D**) EO and concentration, (**E**) EO and exposure time, and (**F**) concentration and exposure time. 1: *S. terebinthifolius* ripe fruit oil. 2: *S. terebinthifolius* unripe fruit oil. 3: *S. terebinthifolius* leaf oil. 4: *P. guajava* leaf oil. 5: *O. majorana* air-dried aerial parts EO.

The mortality values were 31.66% and 75% after 24 h of exposure to EO of *O. majorana* leaves and *P. guajava* leaves, respectively, and were 56.66% and 93.33% after 48 h. The mortality values were 68.33% and 100% after 72 h with EO *O. majorana* leaves and *P. guajava* leaves at 40 µg/L air, respectively (Table 4). The great effect of EO of *S. terebinthifolius* unripe fruits on adult *T. castaneum* after 6 h of exposure at 40 µg/L air was shown by adult mortality of 76.66%. After 6 h at 40 µg/L air with EO of *S. terebinthifolius* leaves, *O. majorana* leaves, and *P. guajava* leaves, the adult mortality was under 50%, while it was 61.66% and 76.66% with EO of *S. terebinthifolius* ripe and unripe fruits, respectively.

Table 4. Mortality (%) of *Tribolium castaneum* as affected by different concentrations of five essential oils with different time periods using a fumigant application.

Source of Essential Oil	Concentration (µg/L Air)	Time (h)				
		6	12	24	48	72
S. terebinthifolius ripe fruits	0	0	1.6 ± 2.8	1.66 ± 2.88	3.3 ± 2.8	8.3 ± 2.8
	2	0	16.6 ± 14.4	33.33 ± 14.43	58.3 ± 7.6	83.3 ± 7.6
	5	6.6 ± 5.7	30 ± 5	58.33 ± 7.63	76.6 ± 10.4	86.6 ± 5.7
	10	6.6 ± 5.7	33.3 ± 2.8	70.00 ± 0.00	85 ± 5	100
	20	31.6 ± 2.8	50 ± 5	83.3 ± 7.6	100	100
	40	61.6 ± 12.5	80 ± 10	100	100	100
S. terebinthifolius unripe fruits	0	0	1.6 ± 2.8	1.6 ± 2.8	3.3 ± 2.8	8.33 ± 2.88
	2	35 ± 5	51.6 ± 7.6	78.3 ± 7.6	100	100
	5	43.3 ± 11.5	75 ± 5	100	100	100
	10	55 ± 18	100	100	100	100
	20	61.6 ± 12.5	100	100	100	100
	40	76.6 ± 5.7	100	100	100	100
S. terebinthifolius leaves	0	0	1.6 ± 2.8	1.6 ± 2.8	3.3 ± 2.8	8.3 ± 2.8
	2	0	8.3 ± 2.8	28.3 ± 5.7	55 ± 5	75 ± 10
	5	0	26.6 ± 15.2	50 ± 5	73.3 ± 2.8	90 ± 5
	10	8.3 ± 7.6	33.3 ± 7.6	70 ± 8.6	86.6 ± 5.7	100
	20	10	68.3 ± 17.5	85 ± 10	100	100
	40	40 ± 10	76.6 ± 5.7	100	100	100
O. majorana leaves	0	0	1.66 ± 2.8	1.6 ± 2.8	3.3 ± 2.8	8.3 ± 2.8
	2	0	0	6.6 ± 2.8	16.6 ± 2.8	26.6 ± 2.8
	5	0	0	10	21.6 ± 2.8	28.3 ± 2.8
	10	0	0	11.6 ± 5.7	26.6 ± 2.8	31.6 ± 5.7
	20	0	5 ± 5	20 ± 5	33.3 ± 5.7	45 ± 5
	40	0	8.3 ± 2.8	31.6 ± 7.6	56.6 ± 15.2	68.3 ± 12.5
P. guajava leaves	0	0	1.6 ± 2.8	1.6 ± 2.8	3.3 ± 2.8	8.3 ± 2.8
	2	0	18.3 ± 7.6	41.6 ± 7.6	61.6 ± 5.7	83.3 ± 12.5
	5	5 ± 5	30 ± 5	55 ± 5	70 ± 5	88.3 ± 12.5
	10	28.3 ± 7.6	60 ± 10	73.3 ± 7.6	86.6 ± 5.7	91.6 ± 14.4
	20	26.6 ± 2.8	63.3 ± 12.5	73.3 ± 7.6	86.6 ± 5.7	100
	40	36.6 ± 5.7	65 ± 13.2	75 ± 13.2	93.3 ± 11.5	100
$LSD^*_{0.05}$ = 10.077						

* LSD: Least Significant Difference

After 72 h at 5 µg/L air with EOs of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves, *O. majorana* leaves, and *P. guajava* leaves, adult mortality was 86.66%, 100%, 90%, 28.33%, and 88.33%, respectively, while the control was recorded as a standard reference. After 12 and 24 h of exposure with acetone as the control, mortality was 1.66%, but was 3.33% after 48 h and 8.33% after 72 h (Table 4).

The fumigant experiment applied to adult *T. castaneum* with different times and concentrations showed that adult mortality increased gradually with increased concentrations from 2 to 40 µg/L air and time from 6 h to 72 h of exposure.

Figure 2 illustrates the effects of tested EOs on adult *T. castaneum*, with dead insects (shown in black) due to accumulation of CO_2 in the tracheas of insects treated with the fumigation method, when compared to normal *T. castaneum* (in brown).

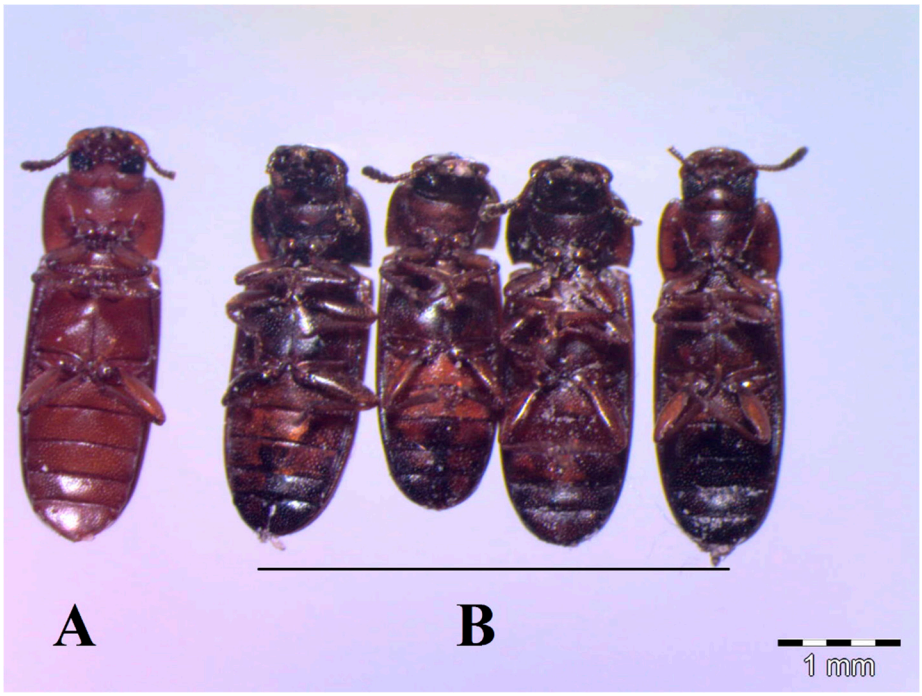

Figure 2. (**A**) Adult *T. castaneum* from the control group. (**B**) Adult mortality of *T. castaneum* after treatment with *S. terebinthifolius* unripe fruit EO at higher concentrations using the fumigant method with different time periods.

Table 3 presents the mortality percentages of *T. castaneum* as affected by the three factors, oil source, oil concentration, and time period, with fumigant application. After 48 h of treatment, the mortality ranged from 58.3% to 100% with EO of *S. terebinthifolius* ripe fruits, was 100% with EO of *S. terebinthifolius* unripe fruits, ranged from 55% to 100% with EO of *S. terebinthifolius* leaves, and ranged from 16.6% to 56.6% with EO of *O. majorana* leaves, and 61.6% to 93.3% with EO of *P. guajava* leaves. By comparison, mortality was 8.3% with the control (Table 2).

The lethal concentration causing 50% mortality (LC_{50}) of *T. castaneum* was calculated for tested EOs at different time periods (6, 12, 24, 48, and 72 h). After 6 h of treatment, the LC_{50} was 33.3, 6.8, >40, >40, and >40 µg/L air for EOs of *S. terebinthifolius* ripe fruits, *S. terebinthifolius* unripe fruits, *S. terebinthifolius* leaves, *O. majorana* leaves, and *P. guajava* leaves, respectively. After 24 h of treatment, the LC_{50} was 4.2, <2, 5.1, >40, and 6.1 µg/L air for EOs of *S. terebinthifolius* ripe fruits, *S. terebinthifolius* leaves, *O. majorana* leaves, and *P. guajava* leaves, respectively. After 24 h of treatment, the LC_{50} of *S. terebinthifolius* unripe fruit oil was under 2 µg/L air, which means that the EO of *S. terebinthifolius* ripe fruits had a stronger effect on *T. castaneum* adults than other tested EOs using the fumigation method (Table 5).

Table 5. Probit regression line parameters of *T. castaneum* for five essential oils at five interval concentrations.

Tested Essential Oil	Period (h)	LC_{50} (µg/L Air)	95% Confidence Limits		Slope ± SE*	Chi^2
			Lower	Upper		
S. terebinthifolius ripe fruits	6	33.3	20.3	54.6	2.1 ± 0.1	0.59
	12	15.5	8	30.2	1.2 ± 0.1	0.92
	24	4.2	2.2	8.1	1.3 ± 0.1	0.98
	48	<2				
	72	<2				
S. terebinthifolius unripe fruits	6	6.8	2.5	18.4	0.8 ± 0.2	0.99
	12	2	1.1	3.8	1.5 ± 0.1	NA
	24	<2				
	48	<2				
	72	<2				
S. terebinthifolius leaves	6	>40				
	12	14.5	8.5	24.5	1.7 ± 0.1	0.95
	24	5.1	2.8	8.8	1.5 ± 0.1	0.99
	48	<2				
	72	<2				
O. majorana leaves	6	>40				
	12	>40				
	24	>40				
	48	>40				
	72	37.9	13.9	103.1	0.8 ± 0.2	0.85
P. guajava leaves	6	>40				
	12	9.5	4.5	16.4	1.4 ± 0.3	0.93
	24	6.1	1.8	19.7	0.6 ± 0.2	0.97
	48	<2				
	72	<2				

* SE: Standard error

3.3. Insecticidal Activity of Essential Oil on C. Pipiens

3.3.1. Immature Stages

Table 6 shows the significant effects of oil concentrations and oil sources and their interaction with mortality and longevity of *C. pipiens* at different stages (larval, pupal, and adult). All treatments showed highly significant effects on mortality and longevity except the interaction between the EO source and the EO concentration for longevity at the larval stage.

Table 6. Analysis of variance for the effect of main treatments and their combinations on *C. pipiens*.

S.O.V.*	DF	Sum of Squares	Mean Square	F-test Value	Pr > F
Larval stage			Mortality after 24 h (%)		
Oil concentration (A)	4	6235.73	1558.93	186.18	<0.0001
Oil source (B)	4	1504	376	44.90	<0.0001
A × B	16	988.26	61.76	7.38	<0.0001
Error	50	418.66	8.37		
			Mortality after 48 h (%)		
A	4	7212.58	1803.146	193.19	<0.0001
B	4	1546.98	386.74	41.44	<0.0001
A × B	16	959.14	59.94	6.42	<0.0001
Error	50	466.66	9.33		
			Total mortality (%)		
A	4	22,063.14	5515.78	415.35	<0.0001
B	4	7092.48	1773.12	133.52	<0.0001
A × B	16	2720.85	170.053	12.81	<0.0001
Error	50	664	13.28		
			Longevity (days)		
A	4	992.89	48.22	30.94	<0.0001
B	4	167.718	41.929	5.23	0.0013
A × B	16	78.8658	4.9291	0.61	0.8571
Error	50	401.153	8.023		
Pupal stage			Mortality (%)		
A	4	26,178.66	6544.66	475.63	<0.0001
B	4	7129.06	1782.26	129.53	<0.0001
A × B	16	2540.26	158.76	11.54	<0.0001
Error	50	688.	13.76		
			Longevity (h)		
A	4	2292.65	573.16	17.31	<0.0001
B	4	2734.14	683.53	20.65	<0.0001
A × B	16	1040.83	65.05	1.96	0.0355
Error	50	1655.27	33.11		
Adult stage			Mortality (%)		
A	4	47,423.78	11,855.94	835.71	<0.0001
B	4	9540.05	2385.01	168.12	<0.0001
A × B	16	2717.54	169.84	11.97	<0.0001
Error	50	709.33	14.18		
			Longevity (days)		
A	4	6651.02	1662.75	94.69	<0.0001
B	4	1961.75	490.43	27.93	<0.0001
A × B	16	767.807	47.98	2.73	0.0034
Error	50	878.04	17.56		

* SOV: source of variance. DF: degree of freedom.

The EOs were tested for their toxicity against the second instar larvae of *C. pipiens*. The five EOs showed pronounced insecticidal activity on immature stages (larva and pupa). After 24 h of treatment with EO of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves, *O. majorana* leaves, and *P. guajava* leaves, the larval mortality was 15.3%, 34.6%, 30.6%, 36.6%, and 16.6% at 100 mg/L, respectively (Table 7). The larval mortality recorded after 48 h of treatment with the tested EOs was 17.3%, 36.6%, 32.6%, 38.6%, and 18.6% at 100 mg/L, respectively.

The total larval mortality was recorded during the larval stage for each concentration to examine the larvicidal activity of the tested EOs against *C. pipiens*. Table 7 shows that total larval mortality ranged from 26% to 33.3% with EO of *S. terebinthifolius* ripe fruits, 40.6% to 68% with EO of *S. terebinthifolius* unripe fruits, 30% to 50% with EO of *S. terebinthifolius* leaves, 42.6% to 78% with EO of *O. majorana* leaves, and 24% to 36.6% with EO of *P. guajava* leaves at 10 to 100 mg/L, and was 3.3% as a control. Mortality increased with growing concentration and time of exposure.

The present data confirm that the EOs of *O. majorana* leaves and *S. terebinthifolius* unripe fruits and leaves were more effective as larvicide than EOs of *S. terebinthifolius* ripe fruits and *P. guajava* leaves on *C. pipiens* at a higher concentration (100 mg/L).

Figures 3 and 4 show the destroyed digestive system (rupture) in larvae of *C. pipiens*, which results in increased larval mortality within a short time (24–48 h) with treatment by EO of *S. terebinthifolius* unripe fruits, while EO of *O. majorana* leaves led to a 78% mortality at 100 mg/L.

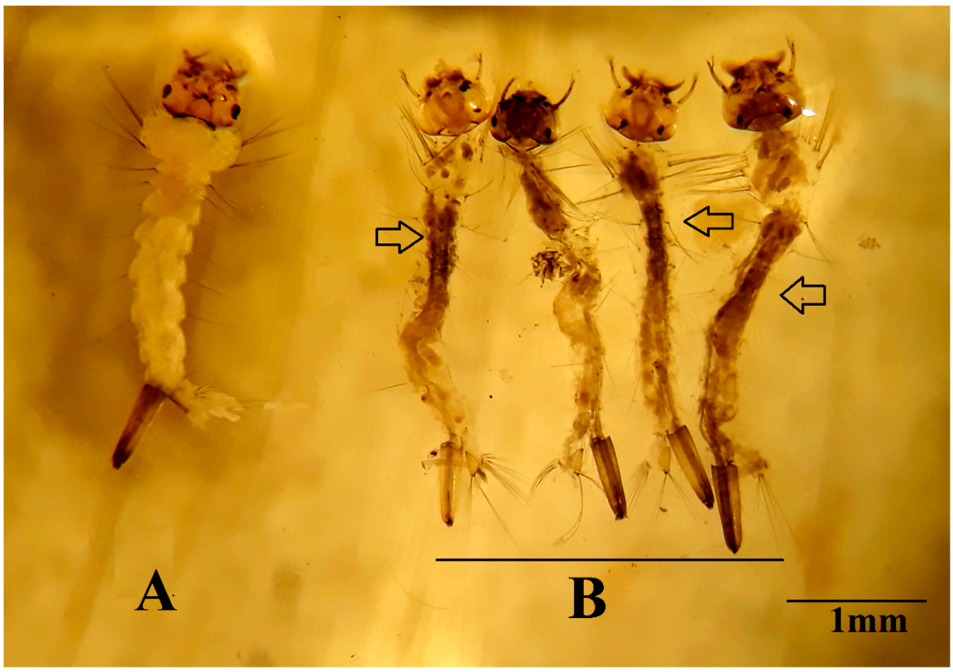

Figure 3. (**A**) Normal *C. pipiens* larva from the control. (**B**) Abnormal larvae produced after treatment with oil of *S. terebinthifolius* unripe fruits at 100 mg/L showing a destroyed digestive system, especially midgut (arrows).

Table 7. Insecticidal effect of tested essential oils on biological activity of *C. pipiens*.

Tested Essential Oil	Concentration (mg/L)	Larval Stage				Pupal Stage		Adult Stage	
		Mortality after 24 h (%)	Mortality after 48 h (%)	Total Mortality (%)	Longevity (days)	Mortality (%)	Longevity (h)	Mortality (%)	Longevity (days)
S. terebinthifolius ripe fruits	0	0.6 ± 1.1	0.6 ± 1.1	3.3 ± 1.1	8.3 ± 0.6	4 ± 2	32.2 ± 6.7	5.3 ± 1.1	44.3 ± 6.1
	10	10 ± 2	10 ± 2	26 ± 2	11.2 ± 0.8	26 ± 2	36.7 ± 10.5	36 ± 2	44.6 ± 2.1
	25	10.6 ± 3	11.3 ± 2.3	26.6 ± 3	14.4 ± 2.9	26.6 ± 3	46.1 ± 12.4	38 ± 5.2	40.6 ± 3
	50	12 ± 2	13.3 ± 2.3	30.6 ± 3	18.5 ± 3.4	32.6 ± 3	57 ± 4.4	49.3 ± 3	32 ± 6.4
	100	15.3 ± 1.1	17.3 ± 1.1	33.3 ± 3	20.6 ± 2.1	40 ± 4	63.4 ± 1.6	58 ± 4	27.3 ± 3.2
S. terebinthifolius unripe fruits	0	0.6 ± 1.1	0.6 ± 1.1	3.3 ± 1.1	8.3 ± 0.6	4 ± 2	32.2 ± 6.7	5.3 ± 1.1	44.3 ± 6.1
	10	13.3 ± 4.1	14 ± 4	40.6 ± 9.4	9.4 ± 1.8	42.6 ± 9.4	25.1 ± 6.5	58.6 ± 9.4	33.7 ± 4.7
	25	18 ± 2	18.6 ± 1.1	48 ± 2	10.5 ± 2.4	45.3 ± 5.7	29.8 ± 4.7	63.3 ± 5.7	22.6 ± 4
	50	28 ± 2	29.3 ± 2.3	60 ± 2	13.7 ± 3.2	64 ± 2	34.4 ± 2.5	84 ± 2	18.1 ± 1.6
	100	34.6 ± 5.7	36.6 ± 5.7	68 ± 3.4	16.3 ± 0.9	72 ± 3.4	37.1 ± 6.3	94 ± 6	15.4 ± 4.4
S. terebinthifolius leaves	0	0.6 ± 1.1	0.6 ± 1.1	3.3 ± 1.1	8.3 ± 0.6	4 ± 2	32.2 ± 6.7	5.3 ± 1.1	44.3 ± 6.1
	10	9.3 ± 1.1	10 ± 2	30 ± 2	8.5 ± 5.1	30 ± 2	33.3 ± 3.9	46 ± 2	34.4 ± 7.3
	25	15.3 ± 3	16 ± 2	36 ± 2	8.9 ± 4.4	38 ± 2	34.5 ± 0.7	54 ± 2	23.6 ± 3.1
	50	27.3 ± 4.1	28.6 ± 5	48.6 ± 5	14.7 ± 5.5	52.6 ± 5	38.5 ± 4.9	70.6 ± 5	18.8 ± 2.2
	100	30.6 ± 5	32.6 ± 5	50 ± 2	18.2 ± 2.4	58 ± 2	43.1 ± 6.4	78 ± 2	20.5 ± 3.2
O. majorana leaves	0	0.6 ± 1.1	0.6 ± 1.1	3.3 ± 1.1	8.3 ± 0.6	4 ± 2	32.2 ± 6.7	5.3 ± 1.1	44.3 ± 6.1
	10	12 ± 5.2	12.6 ± 6.4	42.6 ± 6.4	8.1 ± 2.7	44.6 ± 6.4	22.1 ± 4.1	62.6 ± 6.4	30.4 ± 2
	25	19.3 ± 1.1	20 ± 2	50 ± 2	9.3 ± 2.4	52 ± 2	28.8 ± 3	70 ± 2	19.4 ± 1.2
	50	30.6 ± 3	32 ± 4	62 ± 4	12.3 ± 4.9	66 ± 4	30.7 ± 5.1	86 ± 4	14.8 ± 1.4
	100	36.6 ± 4.1	38.6 ± 4.1	78 ± 2	14.8 ± 2.8	82 ± 2	34.7 ± 3.7	100	11.8 ± 2.4
Psidium guajava leaves	0	0.6 ± 1.1	0.6 ± 1.1	3.3 ± 1.1	8.3 ± 0.6	4 ± 2	32.2 ± 6.7	5.3 ± 1.1	44.3 ± 6.1
	10	8 ± 2	8 ± 2	24 ± 2	9.5 ± 1.5	24 ± 2	31.6 ± 3.4	34.6 ± 2.3	45 ± 3
	25	9.3 ± 1.1	10	26	13.7 ± 3.1	27.3 ± 2.3	35.2 ± 3.6	37.3 ± 2.3	38.6 ± 1.1
	50	12.6 ± 3	14 ± 3.4	30 ± 3.4	17.4 ± 1.9	33.3 ± 3	41.7 ± 3.4	51.3 ± 3	21.7 ± 4.3
	100	16.6 ± 1.1	18.6 ± 1.1	36.6 ± 8.1	19.9 ± 2.6	42.6 ± 5	46.4 ± 3.6	63.3 ± 3	24.9 ± 0.8
P-value		0.0002	<0.0001	<0.0001	0.8571	<0.0001	0.0355	<0.0001	0.0034

Figure 4. Abnormal larvae produced after treatment with *O. majorana* leaf oil at 100 mg/L showing a destroyed digestive system leading to a transparent midgut.

The effects of the tested EOs on immature stages were recorded as mortality percentages. As shown in Table 3, the mortality percentages increased gradually with increased oil concentration (from 10 to 100 mg/L). Pupal mortality ranged from 26% to 40% with EO of *S. terebinthifolius* ripe fruits, 42.6% to 72% with EO of *S. terebinthifolius* unripe fruits, 30% to 58% with EO of *S. terebinthifolius* leaves, 44.6% to 82% with EO of *O. majorana* leaves, and 24% to 42.6% with EO of *P. guajava* leaves at 10 to 100 mg/L. The tested EO of *S. terebinthifolius* ripe fruits, *S. terebinthifolius* unripe fruits, *S. terebinthifolius* leaves, *O. majorana* leaves, and *P. guajava* leaves affected larval and pupal longevity of *C. pipiens*. Larval longevity at 100 mg/L was 20.6, 16.3, 18.2, 14.8, and 19.9 days, respectively, while it was 8.3 days in the control (Table 7).

On the other hand, pupal longevity was affected by treatment with 100 mg/L of EO of *S. terebinthifolius* ripe or unripe fruits and leaves, *O. majorana* leaves, and *P. guajava* leaves, with values at 63.4, 37.1, 43.1, and 46.4 h, respectively, while it was 32.2 h in the control (Table 4).

3.3.2. Adult Stage

Adult mortality ranged from 36% to 58% with EO of *S. terebinthifolius* ripe fruits, 58.6% to 94% with EO of *S. terebinthifolius* unripe fruits, 46% to 78% with EO of *S. terebinthifolius* leaves, 62.6% to 100% with EO of *O. majorana* leaves, and 34.6% to 63.3% with EO of *P. guajava* leaves at 100 mg/L, and was 5.3% in the control. Mortality increased with a growing concentration and time of exposure (Table 7).

Adult longevity reached 27.3, 15.4, 20.5, 11.8, and 24.9 days with 100 mg/L of EO of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves, *O. majorana* leaves, and *P. guajava* leaves, respectively, and was 44.3 days with the control. EO from *O. majorana* leaves and *S. terebinthifolius* unripe fruits strongly reduced adult longevity by approximately 65% to 73% when compared with the control, which means that both EOs had insecticidal activity on the adult stage, which is an important vector for severe and highly infectious diseases in humans.

3.4. Lethal Concentrations of LC_{50}

The results were obtained using probit regression line parameters of *C. pipiens* with five essential oils at five interval concentrations, and the lethal concentration causing 50% mortality (LC_{50}) was calculated for the tested EOs on larval and adult stages at different time periods (after 6, 12, 24, 48, and 72 h) to examine the larvicidal and insecticidal activity.

The LC_{50} values of total larval mortality were >100, 31.2, >100, 24.1, and >100 mg/L for EOs of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves, *O. majorana* leaves, and *P. guajava* leaves (Table 8), respectively. This means that the oils of *O. majorana* leaves and *S. terebinthifolius* unripe fruits had stronger larvicidal activity against *C. pipiens* larvae than the other tested EOs applied by the dipping method.

Table 8. Probit regression line parameters of *Culex pipiens* for five essential oils at five interval concentrations.

Oil Source	Insect Mortality	LC_{50} (mg/L)	95% Confidence Limits		Slope ± SE	Chi^2	R^2
			Lower	Upper			
S. terebinthifolius ripe fruits	TL*	>100	–	–	–	–	–
	Ad*	>50	–	–	–	–	–
S. terebinthifolius unripe fruits	TL	31.2	8.8	109.6	0.721 ± 0.2	0.9	0.9
	Ad	10.9	4.7	24.9	1.185 ± 0.2	0.8	0.9
S. terebinthifolius leaf	TL	>100	–	–	–	–	–
	Ad	20.1	6.9	57.3	0.872 ± 0.2	0.9	0.9
O. majorana leaf	TL	24.1	8.9	64.7	0.925 ± 0.2	0.9	0.9
	Ad	9.7	4.6	20.1	1.414 ± 0.1	0.8	0.9
P. guajava leaf	TL	>100	–	–	–	–	–
	Ad	>50	–	–	–	–	–

* TL, total larval. Ad, adult.

In addition, Table 8 shows that the LC_{50} of adults was >50, 10.9, 20.1, 9.7, and >50 mg/L for EOs of *S. terebinthifolius* ripe fruits, unripe fruits, and leaves, *O. majorana* leaves, and *P. guajava* leaves, respectively. Therefore, the essential oils of *O. majorana* leaves and *S. terebinthifolius* unripe fruits had strong insecticidal activity against *C. pipiens.*

4. Discussion

4.1. Chemical Constituents of the Essential Oils

Several compounds have been identified in the studied plant materials. α-Pinene was identified with a high percentage in EO from unripe fruits of *S. terebinthifolius*, which agreed with Ennigrou et al. [21], who reported that α-pinene was found in amounts of 26.3% (immature fruits) and 13.9% (mature fruits). α-Cadinol, elemol, germacrene-D, and Δ-3-carene are the most common compounds identified in the EO of leaves and fruits of *S. terebinthifolius* [53]. Δ-3-carene (25.9%) was the most abundant compound in EO of *S. terebinthifolius* ripe fruits. Previously it was reported that the main chemical compounds of EO from *S. terebinthifolius* ripe fruits from Brazil were myrcene, limonene, and germacrene-D [54], while, in another report, Δ-3-carene, and α-pinene dominated in fruit EO [23].

Δ-3-Carene, limonene, α-phellandrene, and α-pinene were reported as the major components of the EO of *S. terebinthifolius* fruits grown in Brazil [55]. Limonene, α-phellandrene, α-pinene, and germacrene-D were identified as the main compound of fruit essential oils of *S. terebinthifolius* from Reunion Island [56]. *S. terebinthifolius* fruit EOs in Germany showed α-apinene, α-phellandrene, β-phellandrene, and limonene [57], α-phellandrene, γ-cadinene, β-phellandrene, *p*-cymene, and

α-pinene from Sfax (Southern Tunisia) [58], and from *S. terebinthifolius* ripe fruits in Egypt were α-pinene, α-phellandrene, limonene, α-terpineol, α-cadinol, β-pinene, elixene, α-pinene, and germacrene D [8,59,60].

4-Terpinene, γ-terpinene, α-terpinene, and sabinene were the main compounds of EO from *O. majorana* [27]. Another study showed that the major chemical components of *O. vulgare* EO were carvacrol and terpinen-4-ol [61]. The insecticidal activity of *Origanum* against larvae of *C. pipiens* was found [62], where thymol (LC_{50} = 36 mg/L) and carvacrol (LC_{50} = 37.6 mg/L) were responsible for this activity.

α-Pinene, (*E*)-caryophyllene, (*E*)-nerolidol, and cedran-8-ol were the main compounds in *P. guajava* leaf EO, which agreed with in one study [63], while another study identified α-pinene and 1,8-cineole as the major components [64]. (*E*)-nerolidol was found in 18.5% and 17% amounts in EOs from young and mature leaves of *P. guajava* varieties, while β-caryophyllene was identified as a major constituent in EO from five Brazilian guava cultivars [65].

Another study showed that (*E*)-caryophyllene, caryophyllene oxide, and α-humulene were the main compounds in the essential oil of *P. guajava* leaves collected from Espírito Santo, Brazil [24]. The compounds *iso*-caryophyllene, veridiflorene, farnesene, dl-limonene, Δ-cadinene, α-copaene, and α-humulene were found to be abundant in the EO of plants collected from the Alsharqia region, Sultanate of Oman [66], while α-terpinyl acetate, trans-caryophyllene, nerolidol, α-cadinol, α-copaene, α-humulene, and aryphyllene oxide were found in plants collected from Northeast India [67].

4.2. Fumigant Toxicity on T. Castaneum

The LC_{50} ranged from <2 to 33.3 µg/L air for EO of *S. terebinthifolius* ripe fruits, <2 to 6.8 µg/L air for EO of *S. terebinthifolius* unripe fruits, <2 to 65.1 µg/L air for EO of *S. terebinthifolius* leaves, 37.9 to >40 µg/L air for EO of *O. majorana* leaves, and <2 to 60.2 µg/L air for EO of *P. guajava* leaves, which means that the EO of *S. terebinthifolius* unripe fruits had a stronger effect on *T. castaneum* adults than the other tested EOs using the fumigation method. Our results agree with those of Abdelgaleil et al. [68] who reported that the EO of *O. vulgare* (LC_{50} = 1.6 µg/L air) was the most potent toxicant against *S. oryzae* adults. At the same time, EO of *S. terebinthifolius* possessed strong fumigant toxicity (LC_{50} <30 mg/L air).

Savory and marjoram EOs had 72.5% and 67.5% mortality, respectively, on *T. castaneum* adults when exposed to 150 µL/L air for 24 h [69]. The insecticidal activity of oil of *Origanum* leaves in a vapor-phase toxicity bioassay against *T. castaneum* adults reached LC_{50} = 73.7 µL/L air [70], while the EOs obtained from leaves and flowers showed insecticidal activity against *T. castaneum* adults [71]. Thymol and other compounds of *O. majorana* EO, showed insecticidal activity against *S. oryzae* and *R. dominica* adults [72].

The *P. guajava* treatments caused significantly higher mortality at 21 days of exposure when compared to the control. None of the treatments of *P. guajava* achieved 100% mortality throughout the experimental period. Since mortality was found to be directly proportional to exposure time and concentration, increased mortality might be attained by increasing either or both [73].

For the mode of toxic action, some monoterpenes had an inhibitory effect on acetylcholinesterase activity [74,75], bound with octopamine receptors [76] and GABA-gated chloride ion channels [77].

4.3. Mosquitocide Activity of Tested Essential Oils

In this study, five EOs belonging to several classes was examined to compare their relative toxicity against *C. pipiens* larvae. The EOs of *O. majorana* leaves and *S. terebinthifolius* leaves and unripe fruits showed larvicidal toxicity. The tested EOs had LC_{50} values for the larval and adult stages under 100 mg/L (9.7–90.9 mg/L), except for the EOs of *S. terebinthifolius* ripe fruits and leaves as well as *P. guajava* leaves, which had LC_{50} of total larval mortality of 18,475.3, 115.6, and 1719.1 mg/L, respectively. Therefore, the EOs of *O. majorana* leaves, *S. terebinthifolius* leaves, and unripe fruits EOs have potential as effective mosquitocides. In addition, the bioactivity of most monoterpenes against *C. pipiens* was evaluated in the present experiment. The leaves of the *Origanum* herb are rich in EO, which confers

its characteristic and fragrance. The larval toxicity of some plant extracts, EOs, and phytochemicals against *C. pipiens* has been reported [78–81].

With the present results, total larval mortality at 10 to 100 mg/L ranged from 40.6% to 68% with EO of *S. terebinthifolius* unripe fruits, while it was 42.6% to 78% with EO of *O. majorana* leaves. Mortality increased with a growing concentration and time of exposure. The present data confirms that the EOs of *O. majorana* leaves as well as *S. terebinthifolius* unripe fruits and leaves had a more larvicidal effect than EOs of *S. terebinthifolius* ripe fruits and *P. guajava* leaves on *C. pipiens* at the higher concentration (100 mg/L).

The majority compounds 4-terpinene, γ-terpinene, α-terpinene, and sabinene of *O. majorana* EO showed larvicidal activity against *C. pipiens* with LC_{50} and LC_{90} values of 258.7 mg/L and 580.4 mg/L, respectively [27].

EO from *O. vulgare*, with the main compounds of carvacrol and terpinen-4-ol, had a significant toxic effect against early third-stage larvae of *Anopheles stephensi* and *An. subpictus*, *C. quinquefasciatus*, and *C. tritaeniorhynchus*, which had LC_{50} values of 67, 74.1, 80.3, and 84.9 µg/mL, respectively [60].

The tested EOs of *S. terebinthifolius* ripe/unripe fruits and leaves, *O. majorana* leaves, and *P. guajava* leaves affected larval and pupal longevity of *C. pipiens* due to prolonged larval longevity. Larval longevity at 100 mg/L was 20.6, 16.3, 18.2, 14.8, and 19.9 days, respectively. Similar to Abd El Meguid et al. [82], the toxicological activity of four plant oils including *O. majorana* had prominent mosquitocidal activity against *A. caspius* and *C. pipiens*, along with toxic effects against larvae and pupae.

The most abundant identified compound of EOs of *S. terebinthifolia* fruits and seeds was Δ-3-carene and the least abundant identified compound was γ-elemene. The EOs were observed to have mosquitocidal activity against *An. gambiae*, *An. Arabiensis*, and *C. quinquefasciatus*. The mortality of *C. quinquefasciatus* ranged from 0.5% to 96.7%, and of *An. gambiae* from 13.7% to 97.9% [23].

From the present results, the adult mortality ranged from 36% to 58% with EO of *S. terebinthifolius* ripe fruits, 58.6% to 94% with EO of *S. terebinthifolius* unripe fruits, 46% to 78% with EO of *S. terebinthifolius* leaves, 62.6% to 100% with EO of *O. majorana* leaves, and 34.6% to 63.3% with EO of *P. guajava* leaves at 100 mg/L. The LC_{50} of adult *C. pipiens* was 65.8 mg/L for EO of *P. guajava* leaves. Our results align with Sowmyashree et al. [83], who reported LC_{50} and LC_{90} values of EO of *P. guajava* at 24 h of 40.2 ppm, 56.4 ppm, 38 ppm, and 51.5 ppm.

From the previously identified chemical components in the tested EOs, it can be considered that they have insecticidal properties against immature stages of *C. pipiens* and the adult stage of *T. castaneum.*

5. Conclusions

The present data confirm that the essential oils of *O. majorana* leaves and *S. terebinthifolius* unripe fruits and leaves have more larvicidal effect than those of *S. terebinthifolius* ripe fruits and *P. guajava* leaves on *C. pipiens* at a higher concentration (100 mg/L) when applied by the dipping method. Additionally, EOs of *S. terebinthifolius* unripe and ripe fruits, *P. guajava* leaves, and *S. terebinthifolius* leaves have more adulticidal effect than *O. majorana* leaf oil against *T. castaneum* when applied by the fumigant method.

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