Original article (Orijinal araştırmalar)

Chemical composition and insecticidal potential of different Origanum spp. (Lamiaceae) essential oils against four stored product pests

This study was conducted to determine the contact and fumigant toxicity of plant essential oils extracted from four Origanum spp. against four stored product pests, Rhyzopertha dominica (F., 1792) (Coleoptera: Bostrichidae), Tribolium confusum Jacq. Du Val, 1863 (Coleoptera: Tenebrionidae), Sitophilus granarius (L., 1875) and Sitophilus oryzae (L., 1763) (Coleoptera: Curculionidae). Chemical composition of the essential oils was determined using GC-MS. The essential oils extracted from Origanum onites L. and Origanum vulgare L. var. hirtum caused 100% mortality of R. dominica 24 h after application was 0.046 µl/insect. Single concentration fumigant study indicated that O. onites and O. vulgare var. hirtum essential oils cause high mortality (91 and 70%, respectively) of R. dominica within 24 h. Essential oils of O. vulgare showed the highest activity against R. dominica with LC50 and LC90 values of 0.0052 and 0.0144 µl/ml, respectively. The main components of O. onites essential oil were thymol (22.9%), γ-terpinene (13.0%), p-cymene (12.9%) and carvacrol (7.2%). Similarly, the essential oils of O. vulgare var. hirtum were composed of carvacrol (32.5%), thymol (16.1%), p-cymene (12.2%) and γ-terpinene (7.9%). Likewise, the essential oil of O. vulgare var. verticium had carvacrol (35.0%), p-cymene (11.6%), γ-terpinene (10.3%) and thymol (9.1%). Nonetheless, O. vulgare x O. onites essential oil had carvacrol (15.2%), cis-sabinene hydrate (14.6%), terpinen-4-ol (14.6%) and γ-terpinene (8.7%).

Keywords: Contact activity, essential oils, fumigant activity, GC-MS, Lamiaceae

Abstract

Bu çalışmanın amacı dört Origanum türünden elde edilen uçucu yağların kontakt ve fumigant etkinliklerini Rhyzopertha dominica (F., 1792) (Coleoptera: Bostrichidae), Tribolium confusum Jacq. Du Val, 1863 (Coleoptera: Tenebrionidae), Sitophilus granarius (L., 1875) ve Sitophilus oryzae (L., 1763) (Coleoptera: Curculionidae) erginlerine karşı belirlemektir. Uçucu yağların kimyasal kompozisyonu GC-MS cihazı kullanılarak belirlenmiştir. Denemeler laboratuvar koşullarında 2019 yılında Ziraat Mucadele Merkez Araştırma Enstitüsü'nde yürütülmüştür. Rhyzopertha dominica ve T. confusum erginlerinde Origanum onites L. ve Origanum vulgare L. var. hirtum uçucu yağları %100 ölüme neden olmuştur. Yirmi dört saat sonunda O. vulgare var. verticium bitki uçucu yağının R. dominica için LD50 değeri 0.046 µl/böcek olarak hesaplanmıştır. Tek konsantrasyon fumigant etki denemeleri sonucunda O. onites ve O. vulgare hirtum uçucu yağları 24 saat sonunda R. dominica'ya karşı yüksek aktivite (sirasıyla %91 ve %70) göstermiştir. Fumigant konsantrasyon etki denemeleri sonucunda bitki uçucu yağlarından O. vulgare uçucu yağı R. dominica için en yüksek etkinliğini göstermiş ve 24 saat sonunda LC50 ve LC90 değerleri sırasıyla 0.0052 µl/ml ve 0.0144 µl/ml olarak hesaplanmıştır. Origanum onites’in ana bileşenleri, thymol %22.9; γ-terpinene %13.0; p-cymene %12.9; carvacrol %7.2, O. vulgare var. hitistim ana bileşenleri carvacrol %32.5; thymol %16.1; p-cymene %12.2; γ-terpinene %7.9, O. vulgare var. verticium ana bileşenleri carvacrol %35.0; p-cymene %11.6; γ-terpinene %10.3; thymol %9.1, O. vulgare x O. onites’in ana bileşenleri carvacrol %15.2; cis-sabinene hydrate %14.6; terpinen-4-ol %14.6 ve γ-terpinene %8.7 olarak belirlenmiştir.

Anahtar sözcükler: Kontakt etkinlik, uçucu yağ, fumigant etkinlik, GC-MS, Lamiaceae
Introduction

Cereals can be infested by many pests after harvest if not kept under appropriate storage conditions. Qualitative and quantitative losses occur in the stored products due to these pests. Different cultural, physicomachinery and chemical control methods are used to reduce the damage caused by stored product pests. Chemical control is the most widely and extensively used method to manage these pests globally. The most commonly used synthetic chemicals to control these pests are methyl bromide and aluminum phosphide (Bond, 1984; Taylor, 1994; Mutungi et al., 2014). The use of these chemicals is being prohibited in the scope of Montreal protocol due to their toxicity against warm-blooded organisms and damage to ozone layer but phosphide is the major fumigant in current use.

Plants employ various defense mechanisms to protect themselves from enemies. Various secondary metabolites synthesized within the plant cells occupy an important place among these mechanisms. These compounds having insecticidal and behavioral activities against various pests (Güncan & Durmuşoğlu, 2004) can be classified as alkaloids, glycosides, phenols, terpenoids, tannins and saponins (Shanker & Solanki, 2000). The plant essential oils contain terpenic or non-terpenic volatile compounds that are hydrocarbons and their derivatives (Başer, 2009).

Origanum (Lamiaceae) includes important medicinal aromatic plants and many studies have been conducted on their biological activities. Different activities of Origanum spp. such as antioxidant (Dutra et al., 2019), cytotoxic (Coccimiglio et al., 2016), antimicrobial (Lesjak et al., 2016; Reyes-Jurado et al., 2019), anti-acetylcholinesterase (Abou-Taleb et al., 2016; Hajlaoui et al., 2016; López et al., 2018), antibacterial (da Cunha et al., 2018; Wijesundara & Rupasinghe, 2018), repellent (Govindarajan et al., 2016; La Pergola et al., 2017; Giatropoulos et al., 2018), antifungal (Vinciguerra et al., 2018), allelopathic (Boukaew et al., 2017), phytotoxic (Ibáñez & Blázquez, 2018; Gru'ová et al., 2019) insecticidal (Kim et al., 2016; Szczepanik et al., 2018; Benelli et al., 2019) have been determined in a number of studies.

The studies conducted to the determine the essential oil composition of Origanum spp. have reported that the main are carvacrol (Martucci et al., 2015; Lesjak et al., 2016), thymol (Mechergui et al., 2016), γ-terpinene (Hajlaoui et al., 2016; Lesjak et al., 2016), p-cymene (Martucci et al., 2015; Hajlaoui et al., 2016; Mechergui et al., 2016), terpinen-4-ol (Hajlaoui et al., 2016), linalool (Aligiannis et al., 2001), sabinene (Hajlaoui et al., 2016), α-terpinene (Hajlaoui et al., 2016), cis-sabinene hydrate (Hajlaoui et al., 2016), terpinene, α-pinene (Martucci et al., 2015) and 4-terpineol (Couto et al., 2015).

Coleoptera is the largest insect order and includes the most common and important stored product pests. The pests belonging to this order live in a wide variety of habitats. Stored product pests have different behavior patterns; thus, some of them are regarded as primary pests, while others are defined as secondary pests. The Curculionidae family includes some of the stored product pests. Sitophilus spp. belong to this family and considered as primary pests. The Tenebrionidae family comprises of >10,000 species, of which 100 are stored product pests, and are regarded as secondary pests. Pests belonging to Tribolium are in this family.

The contact and fumigant toxicity of essential oils extracted from four Origanum spp., Origanum onites L., Origanum vulgare L. var hirtum, Origanum vulgare L. var verticium and O. vulgare x O. onites (Lamiaceae) were determined against four important stored product pests, Rhizopertha dominica (F., 1792) (Coleoptera: Bostrichidae), Tribolium confusum Jacquelin Du Val, 1863 (Coleoptera: Tenebrionidae), Sitophilus granarius (L., 1875) and Sitophilus oryzae (L., 1763) (Coleoptera: Curculionidae). In addition, the essential oil components of these species were determined by GC-MS. Many studies have been conducted on the effect of Origanum spp. on storage pests, but the insecticidal activity of the essential oil of O. vulgare x O. onites was studied for the first time. The results of the study will help to devise alternative and environmentally safe management strategies for control of stored product pests.
Materials and Methods

Plant material

Shoots of *O. onites*, *O. vulgare* var. *hirtum*, *O. vulgare* var. *verticium* and *O. vulgare* x *O. onites* were collected during the flowering period July 2018 from the production area of Field Crops Central Research Institute, Ankara, Turkey. All vegetative parts of the plant were used in the production of essential oils. The species were identified by PhD Reyhan Bağdat Bahtiyarca. The herbaria of these species were prepared and deposited at the Directorate of Plant Protection Central Research Institute, Ankara, Turkey.

Extraction of essential oils

The aerial parts (100 g each) of the air-dried plant samples of all the species were separately hydro-distilled for 4 h using a Clavenger apparatus. Oils yields were 2.2, 4.6, 2.8 and 3.1% for *O. onites*, *O. vulgare* var. *hirtum*, *O. vulgare* var. *verticium*, and *O. vulgare* x *O. onites*, respectively. The extracted oils were stored at −20°C until analyzed.

Analysis of essential oils

The GC-MS analysis was performed with an Agilent 5975C InertXL EI/CI MSD system. In the preparation of essential oil samples for analysis GC-MS 20 ml of essential oil and 180 ml of hexane was added to vials. The GC-MS analysis was conducted using an Innowax FSC column (60 m x 0.25 mm) containing helium carrier gas (1 ml/min) with temperature program. The oven temperature was kept at 60°C for 10 min and then raised to 220°C at 4°C/min. The oven was kept at this temperature for 10 min and then temperature was raised to 240°C at 1°C/min. Mass spectra were recorded in the 70 eV mass range/load ratio of 35-450. GC/FID analysis was performed simultaneously in the same column where GC-MS analysis was conducted with same gas, gas flow and temperature used in GC-MS analysis. RRI (relative retention index) values of the essential oil components were compared with those previously reported in the literature (Başer et al., 1998, 2000, 2001, 2002a, b, 2009; Kirimer et al., 2000; Demirci et al., 2003, 2004, 2006; Jiang & Kubota, 2004; Lourens et al., 2004; Kürkçüoğlu et al., 2006; Tabanca et al., 2006; Özkan et al., 2008; Bardakci et al., 2012; Maggio et al., 2012; Polatoğlu et al., 2012a, b, c, 2013, 2017).

Insect rearing

The insect cultures were obtained from the stock cultures of the Plant Protection Central Research Institute, Ankara, Turkey. A mixture of ground soft bread wheat and dry yeast [*Saccharomyces cerevisiae* Meyen ex E. C. Hansen, 1883 (Saccharomycetales: Saccharomycetaceae)] was used to rear *T. confusum* and *R. dominica*. The wheat was crushed to coarse size in feed crushing machine and kept in freezer at −18°C for 72 h to eliminate possible contamination by insect and mites. Dry yeast was ground in a grinding mill, sieved through 100 mesh sieves and added to wheat at 5% w/w. Whole wheat grains were used for rearing *S. granarius* and *S. oryzae*. In order to obtain the adults of desired age, adult emergence was recorded daily about 3 weeks after the eggs were taken into jars. The adults emerging between 7 and 28 d after first emergence were used in the study.

Contact toxicity assay

In single-dose contact activity assays, essential oils were prepared with acetone at a concentration of 0.15% v/v and applied to the ventral of each insect abdomen (1 μl/insect) with micro applicator (Hamilton, Bonaduz, GR, Switzerland). The same amount of acetone was applied to the insects in control treatment of the study. Twenty adult individuals were used in each replication, which were transferred to Petri dishes (6 cm diameter) containing food, and mortality was recorded after 24 and 48 h. The insects unable to move synchronously upon touching with a sable brush were considered as to be dead. The Petri dishes were kept in an incubator at 25±2°C and 65% RH (Polatoğlu et al., 2013). The experiment was laid out according
in a completely randomized design with five replicates. The plant essential oils showing 70% or higher mortality were included in the dose-response assays. The essential oils of all Origanum spp. were applied against R. dominica, T. confusum, S. granarius and S. oryzae at different doses ranging from 0.025 to 0.2% v/v and LD$_{50}$ and LD$_{90}$ values were calculated.

**Fumigant toxicity assay**

Glass tubes (10 ml) with airtight caps were used in single concentration fumigant activity assays. Five adult individuals were released in each tube. Discs of 10 mm diameter were cut from Whatman No1 filter paper and attached to the caps of the glass tubes with a needle. Concentrations of essential oils 0.1% v/v were prepared with acetone and 10 µl was applied to each filter paper disc with a micropipette. The same amount of acetone was applied to the insects in a control treatment. The tubes were kept under fumehood for 5 min to allow the acetone to evaporate. The silicon septic caps of the tubes were then closed with a motor creeper. The tubes were incubated in a temperature controlled climatic chamber at 25±2°C and dying insects were recorded after 24 and 48 h of exposures (Polatoglu et al., 2013). The experiment was laid out in a completely randomized design with 18 replicates. The plant essential oils showing 70% or higher mortality were included in dose-response assays. The essential oils of O. onites, O. vulgare var. hirtum and O. vulgare var verticium were applied against R. dominica and S. oryzae at different doses ranging from 0.025 to 0.2% v/v and LC$_{50}$ and LC$_{90}$ values were calculated.

**Statistical analysis**

The mortality data recorded in single-dose assays were converted to percent mortality and then transformed by arcsine transformation. One-way analysis of variance was used to test the significance, and treatment means were separated by Tukey’s multiple comparison test. The statistical analyses were carried out on MINITAB (Release 16) computer program. The data recorded from dose-response assays were analyzed by Polo-PC probit package program and LC/LD$_{50}$ and LC/LD$_{90}$ values and confidence intervals were computed. Principle component analysis (PCA) was performed with GenStat statistical software.

**Results and Discussion**

**Composition of essential oils**

A total of 54 compounds were identified from the essential oil of O. onites, which represented 99.1% of the essential oil. Similarly, 50 compounds were recognized from O. vulgare var. hirtum essential oil, which constituted 97.9% of the oil. The GC-MS analysis identified 43 compounds in the essential oil of O. vulgare var. verticium, and the identified compounds represented 98.7% of the total essential oil. Likewise, 57 essential oil components of O. vulgare x O. onites were identified and represented 97.0% of the oil (Table 1).

The major components of O. onites essential oil were thymol (22.9%), γ-terpinene (13.0%), p-cymene (12.9%) and carvacrol (7.2%). Similarly, the main components of O. vulgare var. hirtum essential oil were carvacrol (32.5%), thymol (16.1%), p-cymene (12.2%) and γ-terpinene (7.9%). Likewise, the major components identified from the essential oil of O. vulgare var. verticium were carvacrol (35.0%), p-cymene (11.6%), γ-terpinene (10.3%) and thymol (9.1%). Nonetheless, the major essential oil components of O. vulgare x O. onites were carvacrol (15.2%), cis-sabinene hydrate (14.6%), terpinen-4-ol (14.6%) and γ-terpinene (8.7%). PCA divided the species in two groups based on their essential oil components. The PCA indicated that O. vulgare var. hirtum and O. vulgare var. verticium had similar essential oils, but O. onites and O. vulgare x O. onites are different (Figure 1).
Table 1. Essential oil composition of *Oregano* (Ao), *O. vulgare* var. *verticium* (Ovv), *O. vulgare* var. *hirtum* (Ovh) and *O. vulgare* × *Oregano onites* (Ovo) (All components were identified by mass spectrometry database matches and comparison of relative retention index from the literature)

| Compound                  | RRI   | RRI L. | Ao (%) | Ovv (%) | Ovh (%) | Ovo (%) |
|---------------------------|-------|--------|--------|---------|---------|---------|
| α-Pinene                  | 1024  | 1026   | 0.80   | 1.75    | 1.52    | 0.89    |
| α-thujene                 | 1028  | 1028   | 2.16   | 0.46    | 2.54    | 1.38    |
| Camphene                  | 1070  | 1069   | 0.25   | 0.32    | 0.28    | 0.06    |
| Hexanal                   | 1090  | 1087   |        |         | -       | -       |
| β-Pinene                  | 1115  | 1114   | 0.19   | 0.19    | 0.29    | 0.38    |
| Sabinene                  | 1129  | 1126   |        |         | -       | -       |
| 5-3-carene                | 1156  | 1159   | 0.15   | 0.20    | 0.20    | 0.03    |
| Myrcene                   | 1171  | 1168   | 2.83   | 3.18    | 3.06    | 1.69    |
| p-Mentha-1(7).8-diene     | 1177  | 1183   |        |         | -       | -       |
| α-Terpine B               | 1187  | 1183   | 4.95   | 2.27    | 2.21    | 6.12    |
| Dehydro 1.8-cineole       | 1197  | 1194   |        |         | -       | -       |
| Limonene                  | 1206  | 1202   | 0.64   | 0.51    | 0.50    | 0.70    |
| 1.8-Cineole (=Eucalyptol) | 1214  | 1212   |        | 0.06    | -       | 0.06    |
| β-Phellandrene            | 1216  | 1218   | 0.39   | 0.38    | 0.42    | 0.98    |
| (E)-2-Hexanal             | 1229  | 1232   | 0.18   | 0.07    |         | 0.14    |
| β-2-ocimene               | 1244  | 1246   | 0.12   |         | 0.07    | 0.57    |
| γ-Terpine B               | 1257  | 1251   | 13.00  | 7.93    | 10.33   | 8.69    |
| β-E-ocimene               | 1261  | 1265   | 0.10   | 0.08    | 0.08    | 0.10    |
| 5-Methyl-3-heptanone      | 1263  | 1265   |        | 0.29    | 0.38    | -       |
| p-cymene                  | 1281  | 1277   | 12.94  | 12.17   | 11.62   | 3.54    |
| α-Terpinolene             | 1292  | 1290   | 0.35   | 0.23    | 0.11    | 2.24    |
| 1-Octenyl acetate         | 1387  | 1386   |        | 0.07    | -       | 0.16    |
| 3-Octanol                 | 1397  | 1393   | 0.25   | 0.09    | 0.14    | -       |
| α. p-Dimethylstyrene      | 1451  | 1452   |        | 0.09    | -       | -       |
| 1-Octen-3-ol              | 1456  | 1457   | 2.01   | 0.75    | 1.23    | 0.16    |
| trans-Sabinene hydrate    | 1473  | 1469   | 2.12   | 0.43    | 0.71    | 4.40    |
| α-Campholene aldehyde     | 1505  | 1500   | 0.14   |         | -       | -       |
| Linalool                  | 1555  | 1552   | 0.68   | 1.24    | 0.09    | 1.95    |
| cis-Sabinene hydrate      | 1557  | 1554   | 1.34   | 0.36    | 0.40    | 14.58   |
| Linalyl acetate           | 1568  | 1565   | 0.85   | 0.34    | -       | 0.28    |
| trans-p-Menth-2-en-1-ol    | 1575  | 1570   | 0.30   | 0.10    | -       | 2.24    |
| Bornyl acetate            | 1596  | 1593   |        | 0.08    | -       | 0.12    |
| trans-β-bergamotene       | 1598  | 1594   | 0.11   |         | -       | -       |
| β-Caryophyllene           | 1616  | 1609   | 6.82   | 5.77    | 8.71    | -       |
| Carvacrol methyl ether    | 1619  | 1614   |        | 0.36    | 0.17    | -       |
| Terpentine-4-ol           | 1620  | 1611   |        | -       | -       | 14.57   |
| Aromadendrene             | 1625  | 1628   | 0.21   | -       | 0.06    | -       |
| cis-Dihydrocarvone         | 1627  | 1624   | 0.18   | 0.06    | 0.41    | -       |
| p-Menth-3-en-1-ol (=Terpinen-1-ol) | 1639  | 1638   | 0.13   | 0.07    | -       | -       |
| Terpinen-1-ol             | 1640  | 1628   |        | -       | -       | 1.37    |
| trans-Dihydrocarvone       | 1647  | 1645   |        | -       | 0.03    | -       |
| cis-Isodihydrocarvone      | 1649  | 1645   |        | -       | -       | 0.48    |
| trans-Pinoacarveol         | 1672  | 1667   | 0.08   | -       | -       | 0.06    |
| α-Humulene (=α-Caryophyllene) | 1690  | 1685   | 0.19   | 0.35    | 0.83    | 0.16    |
| trans-Piperitol            | 1693  | 1688   |        | -       | -       | 0.59    |
| γ-Murolene                | 1706  | 1702   | 0.14   | 0.08    | -       | -       |
| α-Terpineol               | 1710  | 1706   | 0.67   | 0.62    | 0.25    | 4.37    |
| Borneol                   | 1717  | 1717   | 1.57   | 0.95    | 0.65    | 0.16    |
| Germacrene D              | 1730  | 1726   |        | -       | -       | 0.05    |
| β-Bisabolene              | 1742  | 1741   | 5.58   | 0.71    | 1.49    | 0.31    |
| Bicyclergermacrene        | 1756  | 1755   |        |         | -       | 0.56    |
Table 1. Continued

| Compound                        | RRI      | RRI L. | Ao (%) | Ovv (%) | Ovh (%) | Ovo (%) |
|---------------------------------|----------|--------|--------|---------|---------|---------|
| cis-Piperitol                   | 1759     | 1756   | 0.16   | -       | -       | 0.89    |
| Carvone                         | 1760     | 1755   | -      | 0.11    | -       | -       |
| Geranyl acetate                 | 1769     | 1765   | 0.22   | 0.12    | -       | 0.06    |
| γ-Cadinene                      | 1781     | 1774   | 0.23   | 0.09    | 0.08    | -       |
| β-Sesquiphellandrene            | 1787     | 1783   | 0.22   | -       | -       | -       |
| trans-Carveol                   | 1850     | 1845   | -      | -       | -       | 0.10    |
| Geraniol                        | 1856     | 1852   | 0.21   | 0.11    | -       | 0.08    |
| p-Cymen-8-ol                    | 1865     | 1860   | 0.09   | 0.08    | 0.08    | 0.06    |
| Thymyl acetate                  | 1870     | 1868   | 0.34   | -       | -       | -       |
| Carvacryl acetate               | 1894     | 1890   | -      | 0.32    | 0.48    | 0.16    |
| Piperitenone oxide              | 1987     | 1983   | 0.13   | -       | 0.17    | -       |
| Isocaryophylene oxide           | 2007     | 2001   | 0.13   | 0.41    | 0.52    | -       |
| Caryophyllene oxide             | 2022     | 2007   | 1.15   | 4.02    | 3.56    | 0.07    |
| (E)-Nerolidol                   | 2052     | 2045   | 0.13   | -       | -       | -       |
| Humulene epoxide-III            | 2080     | 2081   | -      | 0.26    | 0.25    | -       |
| Elemol                          | 2101     | 2096   | 0.31   | -       | -       | -       |
| Globulol                        | 2102     | 2098   | -      | -       | -       | 0.07    |
| Cumin alcohol                   | 2124     | 2113   | -      | -       | 0.07    | -       |
| Spathulenol                     | 2154     | 2142   | 0.58   | 0.15    | 0.31    | 0.71    |
| Isothymol                       | 2185     | 2180   | 0.12   | 0.39    | 0.25    | -       |
| Eugenol                         | 2194     | 2187   | 0.08   | -       | -       | -       |
| Thymol                          | 2203     | 2198   | 22.94  | 16.05   | 9.13    | 0.46    |
| 4-Isopropyl-2-methylphenol      | 2223     | 2219   | 0.12   | 0.43    | 0.32    | -       |
| Carvacrol                       | 2236     | 2239   | 7.22   | 32.49   | 35.00   | 15.23   |
| α-Eudesmol                      | 2252     | 2242   | 0.54   | -       | -       | -       |
| α-Cadinol                       | 2260     | 2255   | -      | -       | -       | 0.04    |
| β-Eudesmol                      | 2262     | 2250   | 0.50   | -       | -       | -       |
| Caryophylla-2(12),6(13)-dien-5β-ol caryophylladienol-I | 2325 | 2317 | - | - | - | 0.06 |
| 14-Hydroxy-β-caryophyline       | 2362     | 2357   | 1.23   | 0.07    | -       | -       |
| Manoyl oxide                    | 2384     | 2375   | 0.35   | -       | -       | -       |
| Caryophylla-2(12),6-dien-5β-ol (=caryophyllenol-II) | 2404 | 2392 | - | - | - | 0.04 |
| Aromadendren oxide              | 2406     | 2399   | -      | -       | -       | 0.11    |
| Pseudo phytol                   | 2550     | 2551   | -      | -       | -       | 0.15    |
| Monoterpene hydrocarbons        | 38.87    | 29.76  | 33.23  | 31.44   | -       | -       |
| Oxygenated monoterpenes         | 47.35    | 56.51  | 50.65  | 63.98   | -       | -       |
| Sesquiterpene hydrocarbons      | 7.78     | 6.35   | 9.83   | 0.16    | -       | -       |
| Oxygenated sesquiterpene        | 4.57     | 4.91   | 4.64   | 1.10    | -       | -       |
| Oxygenated diterpenes           | 0.35     | -      | -      | 0.15    | -       | -       |
| Others                          | 0.18     | 0.36   | 0.38   | 0.15    | -       | -       |
| Total                           | 99.10    | 97.89  | 98.73  | 96.98   | -       | -       |

RRI, relative retention index; RRI L., RRI of the compound at same GC column and similar GC condition.
The chemical composition of the essential oils obtained from *O. onites*, *O. vulgare* var. *hirtum* and *O. vulgare* var. *verticium* was in line with the findings of previous studies (Aligiannis et al., 2001; Hajlaoui et al., 2016; Mechergui et al., 2016). However, the percentage of different compounds in total oil varied. In a previous study, *Tanacetum chiliophyllum* (Fisch. & C. A. Mey.) Sch.Bip. var. *chiliophyllum* (Asteraceae) was collected from the same region at different times. The essential oil components of the species as well as biological activities varied with respect to collection time (Polatoğlu et al., 2012c). On the other hand, in a previous study it was reported that the essential oil of *O. vulgare*, contains pulegone, menthone, cis-isopulegone, piperitone and β-myrcene (Abdelgaleil et al., 2016). The current study used subspecies of *O. vulgare*, i.e., *O. vulgare* var. *hirtum* and *O. vulgare* var. *verticium* and the main components of essential oils were carvacrol, thymol, p-cymene and γ-terpinene. Numerous studies have suggested that plant essential oils and their main components have considerable potential to be used in the management of different pests (Isman, 2000; Koul et al., 2008; Lopez et al., 2008; Tripathi et al., 2009).

**Contact toxicity of essential oils**

Single-dose assay indicated that essential oils of all *Origanum* spp. exhibited >70% contact activity against *R. dominica* after 24 h (F=289; df=4,24; P < 0.001). The essential oils of *O. onites* and *O. vulgare* var. *hirtum* caused 100% mortality of *R. dominica*. Similarly, the essential oils of *O. onites* and *O. vulgare* var. *hirtum* caused 100% mortality in *T. confusum*, whereas 18.3% and 7.7% mortality were recorded with essential oils of *O. vulgare* var. *verticium* and *O. vulgare* x *O. onites*, respectively (F=58.3; df=4,24; P < 0.001). *S. oryzae* showed high sensitivity to applied essential oils as >90% mortality was recorded with the essential oils of all species except *O. vulgare* x *O. onites* (F=150; df=4,24; P < 0.001). The essential oils included in the study indicated high contact activity against *S. granarius* as >99.2% mortality was recorded with all essential oils after 24 h except *O. vulgare* x *O. onites* which caused 21.5% mortality (F=537; df=4,24; P < 0.001). The activity of plant essential oils was linearly increased with time after 48 h (Table 2).
The results of the current study.

Response (Negehban et al., 2018) showed varying biochemical differences of different pest species. Previous studies have shown that insects of the same genus or different species to which the same plant essential oil or extract were applied showed varying biological activities under laboratory conditions against storage pests (Ertürk et al., 2017; Shahriari et al., 2017). The highest contact activity against Sitophilus oryzae var. hirtum and O. onites with LD50 values of 0.068 and 0.070 µl/insect, respectively. The highest contact activity against T. confusum was recorded for the essential oils of O. vulgare var. verticium and O. vulgare var. hirtum with LD50 values of 0.095 and 0.103 µl/insect, respectively. The lowest LD50 value of 0.061 µl/insect against S. oryzae was recorded for the essential oil of O. vulgare var. verticium. The highest activity against S. granarius after 24 h was determined for the essential oil of O. vulgare var. verticium with LD50 value of 0.066 µl/insect. Keeping in view the LD50 values, the highest activity against S. granarius was recorded with the essential oil of O. vulgare var. hirtum having LD50 value of 0.092 µl/insect (Table 3).

The biological activity of Origanum spp. has been tested by different researchers against various storage pests in earlier studies (Kim et al., 2010; Qari & Abdel-Fattah, 2017; Benelli et al., 2019). Furthermore, some of the main components of the essential oils of this genus have been tested for their biological activities under laboratory conditions against storage pests (Ertürk et al., 2017; Shahriri et al., 2018). The highest contact activity against R. dominica was recorded with the essential oil of O. vulgare var. verticium after 24 h (LD50 0.046 µl/insect) in the current study. Two of the main constituents of the essential oil of O. vulgare var. verticium were carvacrol and thymol. The main components of essential of Satureja spp. are carvacrol and thymol, displayed contact activity against Tribolium castaneum (Herbst, 1797) (Coleoptera: Tenebrionidae) with LD50 value of 20.1-40.6 µg/adult (Taban et al., 2017). The four storage pests included in the study exhibited varying response to the essential oil of the same species. This might be explained with the chemical composition of the essential oil, as well as the physiological and biochemical differences of different pest species. Previous studies have shown that insects of the same genus or different species to which the same plant essential oil or extract were applied showed varying response (Negehban et al., 2007; Guo et al., 2017; Liang et al., 2017). These results are consistent with the results of the current study.

Table 2. Single-dose (0.15% v/v) contact activities of different Origanum spp. essential oils against test insect species

|        | Control | Ao | Ovh | Ovv | Ovo |
|--------|---------|----|-----|-----|-----|
| 24 ETH |         |    |     |     |     |
| Rd     | 0.2±0.45 c1 | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 99.8±0.45 a | 75.7±0.76 b |
| Tc     | 0.2±0.45 c  | 99.6±0.92 a  | 99.2±0.68 a  | 92.8±1.08 b  | 48.0±0.38 c |
| So     | 1.7±1.37 d  | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 99.2±0.68 a | 21.5±0.43 b |
| Sg     | 0.0±0.00 c  | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 25.7±0.34 b |
| 48 ETH |         |    |     |     |     |
| Rd     | 0.2±0.45 c  | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 99.8±0.45 a | 82.4±0.35 b |
| Tc     | 0.2±0.45 c  | 99.6±0.92 a  | 99.8±0.45 a  | 19.9±5.83 b  | 8.4±1.56 bc |
| So     | 1.7±1.37 c  | 100.0±0.00 a | 100.0±0.00 a | 92.8±1.05 a  | 73.3±0.38 b |
| Sg     | 0.0±0.00 c  | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 100.0±0.00 a | 25.7±0.34 b |

1 Values followed by the same letter within a row are not statistically different (ANOVA P < 0.05, Tukey test). Ao, Origanum onites; Ovh, O. vulgare var. hirtum; Ovv, O. vulgare var. verticium; Ovo, O. vulgare x O. onites; ETH, exposure time (h); Rd, Rhyzopertha dominica; Tc, Tribolium confusum; So, Sitophilus oryzae; and Sg, S. granarius.
Table 3. The results of dose-response assays used to determine the contact activity of different essential oils against test insect species

| Essential oil | Insect | ETH | Slope±SE | LD_{50} (µl/insect) | LD_{95} (µl/insect) |
|---------------|--------|-----|----------|---------------------|---------------------|
| Ao            | Rd     | 24  | 5.97±0.55| 0.070 (0.065-0.075) | 0.115 (0.105-0.129) |
|               | 48     |     | 5.51±0.53| 0.067 (0.062-0.073) | 0.115 (0.104-0.131) |
|               | Tc     | 24  | 4.33±0.43| 0.083 (0.073-0.092) | 0.165 (0.143-0.202) |
|               | 48     |     | 4.15±0.42| 0.080 (0.070-0.090) | 0.164 (0.141-0.204) |
|               | So     | 24  | 5.87±0.63| 0.075 (0.067-0.082) | 0.124 (0.115-0.138) |
|               | 48     |     | 5.26±0.48| 0.054 (0.047-0.060) | 0.094 (0.085-0.108) |
|               | Sg     | 24  | 9.37±0.72| 0.075 (0.072-0.079) | 0.103 (0.098-0.110) |
|               | 48     |     | 9.44±0.83| 0.072 (0.069-0.075) | 0.099 (0.093-0.106) |
| Ovh           | Rd     | 24  | 4.40±0.47| 0.068 (0.058-0.076) | 0.132 (0.113-0.171) |
|               | 48     |     | 4.26±0.46| 0.065 (0.054-0.074) | 0.130 (0.110-0.172) |
|               | Tc     | 24  | 9.54±0.97| 0.103 (0.099-0.108) | 0.141 (0.133-0.152) |
|               | 48     |     | 9.15±0.99| 0.102 (0.097-0.106) | 0.140 (0.103-0.152) |
|               | So     | 24  | 7.64±0.70| 0.069 (0.065-0.073) | 0.102 (0.096-0.111) |
|               | 48     |     | 6.91±0.64| 0.065 (0.060-0.069) | 0.100 (0.093-0.108) |
|               | Sg     | 24  | 10.19±0.88| 0.068 (0.065-0.071) | 0.092 (0.087-0.098) |
|               | 48     |     | 10.51±0.92| 0.067 (0.064-0.070) | 0.089 (0.084-0.095) |
| Ovv           | Rd     | 24  | 3.78±0.53| 0.046 (0.028-0.057) | 0.100 (0.081-0.153) |
|               | 48     |     | 3.32±0.54| 0.038 (0.017-0.051) | 0.093 (0.074-0.150) |
|               | Tc     | 24  | 9.55±0.83| 0.095 (0.091-0.099) | 0.130 (0.123-0.139) |
|               | 48     |     | 9.93±0.86| 0.093 (0.089-0.097) | 0.125 (0.119-0.133) |
|               | So     | 24  | 3.15±0.46| 0.061 (0.050-0.070) | 0.156 (0.131-0.207) |
|               | 48     |     | 3.33±0.70| 0.032 (0.018-0.041) | 0.077 (0.065-0.096) |
|               | Sg     | 24  | 7.19±0.64| 0.066 (0.062-0.071) | 0.100 (0.092-0.112) |
|               | 48     |     | 8.25±0.74| 0.065 (0.061-0.068) | 0.092 (0.086-0.101) |
| Ovo           | Rd     | 24  | 3.46±0.43| 0.100 (0.090-0.111) | 0.234 (0.192-0.317) |
|               | 48     |     | 3.75±0.42| 0.091 (0.083-0.101) | 0.200 (0.170-0.256) |

Ao, Origanum onites; Ovh, O. vulgare var. hirtum; Ovv, O. vulgare var. verticium; Ovo, O. vulgare x O. onites; ETH, exposure time (h); Rd, Rhynzopethra dominica; Tc, Tribolium confusum; So, Sitophilus oryzae; and Sg, S. granarius.

**Fumigant toxicity of essential oils**

Single-dose (0.1 v/v) fumigant assays exhibited a varying degree of fumigant activity according to insect species and exposure time (Table 4). The plant essential oils of O. onites and O. vulgare var. hirtum showed 90.95% and 70.42% activity against R. dominica after 24 h (F=55.0; df=4,89; P < 0.001). The other essential oils did not exhibit a significant activity against this pest. Among different essential oils tested, only O. onites essential oil gave 52.7% mortality of T. confusum, which was statistically different from the control treatment (F=48.8; df=4,89; P < 0.001). When essential oils were evaluated for fumigant activity
against *S. oryzae*, essential oil of *O. vulgare* var. *verticium* gave 75.5% mortality after 24 h, followed by *O. onites* essential oil which caused 70.3% mortality (*F*=30.9; *df*=4,89; *P* < 0.001). None of the tested essential oils had significant toxicity to *S. granarius*.

Table 4. Single-dose fumigant activities of different essential oils against test insect species

| Insect  | Control | Ao | Ovh | Ovv | Ovo |
|---------|---------|----|-----|-----|-----|
| 24 ETH  |         |    |     |     |     |
| *Rd*    | 0.0±0.00 b | 91.0±2.08 a | 70.4±2.90 b | 44.0±0.93 c | 26.7±1.90 c |
| *Tc*    | 0.0±0.00 b | 52.7±2.05 a | 0.8±1.03 b | 1.1±0.91 b | 0.4±0.84 b |
| *So*    | 1.7±1.06 c | 70.3±2.50 a | 64.3±1.87 a | 75.5±2.09 a | 32.1±2.31b |
| *Sg*    | 0.0±0.00 b | 0.07±0.28 b | 16.4±2.82 a | 3.6±1.88 b | 0.0±0.00 b |
| 48 ETH  |         |    |     |     |     |
| *Rd*    | 0.0±0.00 c | 91.1±2.87 b | 99.9±0.61 a | 99.7±0.53 a | 78.9±2.64 b |
| *Tc*    | 0.0±0.00 c | 84.7±3.22 a | 17.5±1.75 b | 3.7±1.48 c | 0.1±0.28 c |
| *So*    | 1.7±1.06 d | 99.4±0.74 a | 87.2±2.86 b | 92.9±1.48 ab | 38.0±1.48 c |
| *Sg*    | 0.0±0.00 c | 23.7±1.60 b | 63.4±2.06 a | 26.5±2.66 b | 1.0±1.31 c |

1 Values followed by the same letter within a row are not statistically different (ANOVA P < 0.05, Tukey test). *Ao*, *Origanum onites*; *Ovh*, *O. vulgare* var. *hirtum*; *Ovv*, *O. vulgare* var. *verticium*; *Ovo*, *O. vulgare* × *O. onites*; ETH, exposure time (h); Rd, *Rhyzopertha dominica*; Tc, *Tribolium confusum*; So, *Sitophilus oryzae*; and Sg, *S. granarius*.

In dose-response assays, essential oil of *O. onites* showed the highest activity against *R. dominica* and LC50 and LC90 values after 24 h were 0.0052 and 0.0144 µl/ml air, respectively (Table 5). These values were 0.0047 and 0.0124 µl/ml air, respectively after 48 h. The essential oil of *O. onites* showed a significant fumigant activity against *S. oryzae* after 24 h with LC50 and LC90 values of 0.0135 and 0.0653 µl/ml air, respectively. These LC50 and LC90 values after 48 h were 0.0101 and 0.0512 µl/ml air, respectively. The LC50 and LC90 values of *O. vulgare* var. *hirtum* against *R. dominica* after 24 h were 0.0080 and 0.0144 µl/ml air, respectively. The essential oil of *O. vulgare* var. *verticium* was evaluated for fumigant activity only against *S. oryzae*, and LC50 and LC90 values at the end of 24 h were 0.0104 and 0.0262 µl/ml air, respectively.

Table 5. The results of dose-response assays used to determine the fumigant activity of different essential oils against test insect species

| Essential oil | Insect  | ETH | Slope±SE | LC50 (µl/ml) (95% fiducial Limit) | LC90 (µl/ml) (95% fiducial Limit) |
|---------------|---------|-----|----------|----------------------------------|----------------------------------|
| *Ao*          | *Rd*    | 24  | 2.91±0.27| 0.0052 (0.0046-0.0058)            | 0.0144 (0.0122-0.0180)           |
|               | 48      |     | 3.03±0.28| 0.0047 (0.0041-0.0052)            | 0.0124 (0.0107-0.0151)           |
|               | *So*    | 24  | 1.87±0.38| 0.0135 (0.0111-0.0200)            | 0.0653 (0.0345-0.0745)           |
|               | 48      |     | 1.81±0.36| 0.0101 (0.0080-0.0136)            | 0.0512 (0.0272-0.0654)           |
| *Ovh*         | *Rd*    | 24  | 5.01±0.58| 0.0080 (0.0070-0.0087)            | 0.0144 (0.0132-0.0164)           |
|               | 48      |     | 5.37±0.76| 0.0065 (0.0051-0.0074)            | 0.0112 (0.0103-0.0127)           |
| *Ovv*         | *So*    | 24  | 3.19±0.40| 0.0104 (0.0092-0.0119)            | 0.0262 (0.0201-0.0435)           |
|               | 48      |     | 2.76±0.38| 0.0087 (0.0074-0.0099)            | 0.0252 (0.0190-0.0446)           |

1 Values followed by the same letter within a row are not statistically different (ANOVA P < 0.05, Tukey test). *Ao*, *Origanum onites*; *Ovh*, *O. vulgare* var. *hirtum*; *Ovv*, *O. vulgare* var. *verticium*; *Ovo*, *O. vulgare* × *O. onites*; ETH, exposure time (h); Rd, *Rhyzopertha dominica*; Tc, *Tribolium confusum*; So, *Sitophilus oryzae*; and Sg, *S. granarius*. 
Origanum spp. used in the study showed significant fumigant activity against S. oryzae and R. dominica. Several earlier studies determined the fumigant of plant essential oils against S. oryzae (Kim et al., 2003; Kim & Park 2008; Cardiet et al., 2012) and R. dominica (Shaaya et al., 1991; Lee et al., 2004). The current study indicated that the essential oil of O. onites var. hirtum had the strongest fumigant activity against R. dominica, while O. vulgare had the strongest fumigant activity against S. oryzae. Lee et al., (2001) indicated that essential oil of eucalyptus exhibited fumigant activity with LD50 of 28.9 µl/ml air against tested insect species. Previously, several studies have determined the insecticidal activity of essential oils of the Lamiaceae family against storage pests (Chu et al., 2011; Conti et al., 2011; Kim et al., 2016). The main components of the essential oils exhibiting the highest fumigant activity in the current study are thymol and carvacrol. Previous studies with these two essential oil components or essential oils containing high percentage of these components have found a high fumigant activity against different storage pests (Erler, 2005; Kim & Park, 2008).

In this study, insecticidal effects of essential oils obtained from Origanum spp. against four important stored product pests that cause significant damage in warehouses were tested under laboratory conditions and main components of plant essential oils were determined. As a result of the study, it was determined that these plant essential oils have both contact and fumigant activity. It was also concluded that the activity varies depending on the chemical composition of plant essential oils and the insect species applied. This study is a basic study and its future applicability will be demonstrated with the studies to be done.

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References

Abdelgaleil, S. A., M. I. Mohamed, M. S. Shawir & H. K. Abou-Taleb, 2016. Chemical composition: insecticidal and biochemical effects of essential oils of different plant species from Northern Egypt on the rice weevil. Sitophilus oryzae L. Journal of Pest Science, 89 (1): 219-229.

Abou-Taleb, H. K., M. I. Mohamed, M. S. Shawir & S. A. Abdelgaleil, 2016. Insecticidal properties of essential oils against Tribolium castaneum (Herbst) and their inhibitory effects on acetylcholinesterase and adenosine triphosphatases. Natural Product Research, 30 (6): 710-714.

Alijani, N., E. Kalpoutzakis, S. Mitaku & I. B. Chinou, 2001. Composition and antimicrobial activity of the essential oils of two Origanum species. Journal of Agricultural and Food Chemistry, 49 (9): 4168-4170.

Bardakci, H., B. Demirci, E. Yesilada, H. Kirmizibekmez & K. H. C. Başer, 2012. Chemical composition of the essential oil of the subterranean parts of Valeriana alliariifolia. Records of Natural Products, 6 (1): 89-92.

Başer, K. H. C., 2009. Uççu yağlar ve aromaterapi. Fitomed, 7: 8-25.

Başer, K. H. C., B. Demirci, F. Demirci, S. Koçak, Ç. Akıncı, H. Malyer & G. Güleryüzl, 2002a. Composition and antimicrobial activity of the essential oil of Achillea multifida. Planta Medica, 68 (10): 941-943.

Başer, K. H. C., B. Demirci, N. E. Kirimer, F. Satîl & G. Tumen, 2002b. The essential oils of Thymus migricus and T. fedtschenkoi var. handelii from Turkey. Flavour and Fragrance Journal, 17 (1): 41-45.

Başer, K. H. C., B. Demirci, M. Kurkuçoğlu, F. Satin & G. Tumen, 2009. Comparative morphological and phytochemical characterization of Salvia cadmica and S. smyrnaea. Pakistan Journal of Botany, 41 (4): 1545-1555.

Başer, K. H. C., B. Demirci, N. Tabanca, T. Özkek & N. Gören, 2001. Composition of the essential oils of Tanacetum armenum (DC.) Schultz Bip., T. balsamita L., T. chilophyllum (Fisch. & Mey.) Schultz Bip. var. chilophyllum and T. haradjani (Rech. fil.) Grierson and the enantiomeric distribution of camphor and carvone. Flavour and Fragrance Journal, 16 (3): 195-200.
Basar, K. H. C., M. Kurcoglu & Z. Aytac, 1998. Composition of the essential oil of *Salvia euphratica* Montbret ex *Bentham var. euphratica* from Turkey. Flavour and Fragrance Journal, 13 (1): 63-64.

Basar, K. H. C., T. Ozek, B. Demirci & H. Duman, 2000. Composition of the essential oil of *Glaucesciadium cordifolium* (Boiss.) Burtt et Davis from Turkey. Flavour and Fragrance Journal, 15 (1): 45-46.

Benelli, G., R. Pavela, R. Petrelli, L. Cappellacci, F. Bartolucci, A. Canale & F. Maggi, 2019. *Origanum syriacum* subsp. *syriacum*: From an ingredient of Lebanese ‘manoushe’ to a source of effective and eco-friendly botanical insecticides. Industrial Crops and Products, 134: 26-32.

Bond, E. J., 1984. Manual of Fumigation for Insect Control. FAO Plant Production and Protection Paper No: 54, 432 pp.

Boukaew, S., P. Prasertsan & S. Sattayasamitsathit, 2017. Evaluation of antifungal activity of essential oils against aflatoxigenic *Aspergillus flavus* and their allelopathic activity from fumigation to protect maize seeds during storage. Industrial Crops and Products, 97: 558-566.

Cardiet, G., B. Fuzeau, C. Barreau & F. Fleurat-Lessard, 2012. Contact and fumigant toxicity of some essential oil constituents against a grain insect pest *Sitophilus oryzae* and two fungi. *Aspergillus westerdijkiae* and *Fusarium graminearum*. Journal of Pest Science, 85 (3): 351-358.

Chu, S. S., S. L. Liu, Q. Z. Liu, Z. L. Liu & S. S. Du, 2011. Composition and toxicity of Chinese *Dracocephalum moldavica* (Labiatae) essential oil against two grain storage insects. Journal of Medicinal Plants Research, 5 (18): 4621-4626.

Coccimiglio, J., M. Alipour, Z. H. Jiang, C. Gottardo & Z. Suntres, 2016. Antioxidant, antibacterial, and cytotoxic activities of the ethanolic *Origanum vulgare* extract and its major constituents. Oxidative Medicine and Cellular Longevity, 2016: 1-8.

Conti, B., A. Canale, P. L. Cioni, G. Flamini & A. Rifici, 2011. *Hyptis suaveolens* and *Hyptis spicigera* (Lamiaceae) essential oils: qualitative analysis. contact toxicity and repellent activity against *Sitophilus granarius* (L.) (Coleoptera: Dryophthoridae). Journal of Pest Science, 84 (2): 219-228.

Couto, C. S., N. R. Raposo, S. Rozental, L. P. Borba-Santos, L. M. Bezerra, P. A. de Almeida & M. A. Brandao, 2015. Chemical composition and antifungal properties of essential oil of *Origanum vulgare* Linnaeus (Lamiaceae) against *Sporothrix schenckii* and *Sporothrix brasiliensis*. Tropical Journal of Pharmaceutical Research, 14 (7): 1207-1212.

da Cunha, J. A., C. de Avila Scheeren, V. P. Fausto, L. D. W. de Melo, B. Henneman, C. P. Frizzo, R. de Almeida Vaucher, A. C. de Vargas & B. Baldisserotto, 2018. The antibacterial and physiological effects of pure and nanoencapsulated *Origanum majorana* essential oil on fish infected with *Aeromonas hydrophila*. Microbial Pathogenesis, 124: 116-121.

Demirci, B., K. H. C. Basar & M. Y. Dadandi, 2006. Composition of the essential oils of *Phlomis rigidia* Labill. and *P. samia* L. Journal of Essential Oil Research, 18 (3): 328-331.

Demirci, B., K. H. C. Basar, B. Yildiz & Z. Bahcecioğlu, 2003. Composition of the essential oils of six endemic *Salvia* spp. from Turkey. Flavour and Fragrance Journal, 18 (2): 116-121.

Demirci, F., D. H. Paper, G. Franz & K. H. C. Basar, 2004. Investigation of the *Origanum onites* L. essential oil using the Chorioallantoic Membrane (CAM) assay. Journal of Agricultural Food Chemistry, 52 (2): 251-254.

Dutra, T. V., J. C. Castro, J. L. Menezes, T. R. Ramos, I. N. do Prado, M. M. Junior, J. M. G. Mikcha & B. A. de Abreu Filho, 2019. Bioactivity of oregano (*Origanum vulgare*) essential oil against *Alicyclobacillus* spp. Industrial Crops and Products, 129: 345-349.

Erlér, F., 2005. Fumigant activity of six monoterpenoids from aromatic plants in Turkey against the two stored-product pests confused flour beetle, *Tribolium confusum* and Mediterranean flour moth, *Ephestia kuehniella*. Journal of Plant Diseases and Protection, 112 (6): 602-611.

Ertürk, S., A. Yilmaz, T. Akdeniz Firat & M. Alkan, 2017. Fumigant effect of trans-anethole and carbon dioxide mixture against to *Rhysopertha dominica*, *Tribolium castaneum* and *Sitophilus oryzae*. Plant Protection Bulletin, 57 (3): 391-400.

Giatropoulos, A., A. Kimbaris, A. Michaelakis, D. P. Papachristos, M. G. Polissiou & N. Emmanuelou, 2018. Chemical composition and assessment of larvicidal and repellent capacity of 14 Lamiaceae essential oils against *Aedes albopictus*. Parasitology Research, 117 (6): 1953-1964.
Govindarajan, M., S. Kadaikunnan, N. S. Alharbi & G. Benelli, 2016. Acute toxicity and repellent activity of the *Origanum scabrum* Boiss. & Heldr. (Lamiaceae) essential oil against four mosquito vectors of public health importance and its biosafety on non-target aquatic organisms. Environmental Science and Pollution Research, 23 (22): 23228-23238.

Gružová, D., M. Pšuchová, J. Fejér, L. De Martino, L. Caputo, V. Sedláček & V. De Feo, 2019. Influence of six essential oils on invasive *Solidago canadensis* L. seed germination. Natural Product Research, 33: 1-3.

Guo, S. S., W. J. Zhang, C. X. You, J. Y. Liang, K. Yang, Z. F. Geng & C. F. Wang, 2017. Chemical composition of essential oil extracted from *Laggera pterodonta* and its bioactivities against two stored product insects. Journal of Food Processing and Preservation, 41 (2): 1-9.

Güncan, A. & E. Durmuşoğlu, 2004. Bitkisel kökenli doğal insektisitler üzerine bir değerlendirme. Hasad Dergisi, 233: 26-32.

Hajlaoui, H., H. Mighri, M. Aouni, N. Gharsallah & A. K. Ksibi, 2016. Essential oils and their components against adult rice weevil (Sitophilus oryzae L.). Journal of Agricultural and Food Chemistry, 52 (13): 4197-4203.

Ibáñez, M. & M. Blázquez, 2018. Phytotoxicity of essential oils on selected weeds: Potential hazard on food crops. Plants, 7 (4): 1-15.

Isman, M. B., 2000. Plant essential oils for pest and disease management. Crop Protection, 19 (8-10): 603-608.

Jiang, L. & K. Kubota, 2004. Differences in the volatile components and their odor characteristics of green and ripe fruits and dried pericarp of Japanese pepper (*Xanthoxylum piperitum* DC.). Journal of Agricultural and Food Chemistry, 52 (13): 4197-4203.

Kim, J. & I. K. Park, 2008. Fumigant toxicity of Korean medicinal plant essential oils and components from *Asiasarum sieboldi* root against *Sitophilus oryzae* L. Flavour and Fragrance Journal, 23 (2): 79-83.

Kim, S. W., H. R. Lee, M. J. Jang, C. S. Jung & I. K. Park, 2016. Fumigant toxicity of Lamiaceae plant essential oils and blends of their constituents against adult rice weevil *Sitophilus oryzae*. Molecules, 21 (3): 361.

Kim, S. I., J. Y. Roh, D. H. Kim, H. S. Lee & Y. J. Ahn, 2003. Insecticidal activities of aromatic plant extracts and essential oils against *Sitophilus oryzae* and *Callosobruchus chinensis*. Journal of Stored Products Research, 39 (3): 293-303.

Kim, S. I., J. S. Yoon, J. W. Jung, K. B. Hong, Y. J. Ahn & H. W. Kwon, 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.

Kırımer, N., N. Tabanca, T. Öztek, G. Tümen & K. H. C. Başer, 2000. Essential oils of annual *Sideritis* species growing in Turkey. Pharmaceutical Biology, 38 (2): 106-111.

Koul, O., S. Walia & G. S. Dhalwali, 2008. Essential oils as green pesticides: potential and constraints. Biopesticides International, 4 (1): 63-84.

Kürkçüoğlu, M., K. H. C. Başer, G. İşcan, H. Malyer & G. Kaynak, 2006. Composition and anticalendrical activity of the essential oil of *Chaerophyllum byzantinum* Boiss. Flavour and Fragrance Journal, 21 (1): 115-117.

La Pergola, A., C. Restuccia, E. Napoli, S. Bella, S. Brighina, A. Russo & P. Suma, 2017. Commercial and wild Sicilian *Origanum vulgare* essential oils: chemical composition, antimicrobial activity and repellent effects. Journal of Essential Oil Research, 29 (6): 451-460.

Lee, B. H., P. C. Annis & W. S. Choi, 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.

Lee, B. H., W. S. Choi, S. E. Lee & B. S. Park, 2001. Fumigant toxicity of essential oils and their constituent compounds towards the rice weevil. *Sitophilus oryzae* (L.). Crop Protection, 20 (4): 317-320.

Lesjak, M., N. Simin, D. Orcic, M. Franciskovic, P. Knezevic, I. Beara, V. Aleksic, E. Svircev, K. Buzas & N. Mimica-Dukic, 2016. Binary and tertiary mixtures of *Satureja hortensis* and *Origanum vulgare* essential oils as potent antimicrobial agents against *Helicobacter pylori*. Phytotherapy Research, 30 (3): 476-484.

Liang, J. Y., W. T. Wang, Y. F. Zheng, D. Zhang, J. L. Wang, S. S. Guo & J. Zhang, 2017. Bioactivities and chemical constituents of essential oil extracted from *Artemisia anethoides* against two stored product insects. Journal of Oleo Science, 66 (1): 71-76.
López, V., M. Cascella, G. Benelli, F. Maggi & C. Gómez-Rincón, 2018. Green drugs in the fight against Anisakis simplex-larvicidal activity and acetylcholinesterase inhibition of Origanum compactum essential oil. Parasitology Research, 117 (3): 861-867.

López, M., M. Jordán & M. Pascual-Villalobos, 2008. Toxic compounds in essential oils of coriander, caraway and basil active against stored rice pest. Journal of Stored Products Research, 44: 273-278.

Lourens, A. C. U., D. Reddy, K. H. C. Başer, A. M. Viljoen & S. F. Van Vuuren, 2004. In vitro biological activity and essential oil composition of four indigenous South African Helichrysum species. Journal of Ethnopharmacology, 95 (2): 253-258.

Maggio, A., S. Rosselli, M. Bruno, V. Spadaro, F. M. Raimondo & F. Senatore, 2012. Chemical composition of essential oil from Italian populations of Artemisia alba Turra (Asteraceae). Molecules, 17 (9): 10232-10241.

Martucci, J. F., L. B. Gende, L. M. Neira & R. A. Ruseckaitė, 2015. Oregano and lavender essential oils as antioxidant and antimicrobial additives of biogenic gelatin films. Industrial Crops and Products, 71: 205-213.

Mechergui, K., W. Jaouadi, J. P. Coelh & M. L. Khouja, 2016. Effect of harvest year on production. chemical composition and antioxidant activities of essential oil of oregano (Origanum vulgare subsp glandulosum (Desf.) Ietwaart) growing in North Africa. Industrial Crops and Products, 90: 32-37.

Mutungi, C., H. Affognon, A. Njoroge, D. Baributsa & L. Murdock, 2014. Storage of mung bean (Vigna radiata [L.] Wilczek) and pigeonpea grains (Cajanus cajan [L.] Millsp) in hermetic triple-layer bags stops losses caused by Callosobruchus maculatus (F.) (Coleoptera: Bruchidae). Journal of Stored Product Research, 58: 39-47.

Negehan, M., S. Moharramipour & F. Sefidkon, 2007. Fumigant toxicity of essential oil from Artemisia sieberi Besser against three stored-product insects. Journal of Stored Products Research, 43 (2): 123-128.

Özkan, A. M. G., B. Demirci, F. Demirci & K. H. C. Başer, 2008. Composition and antimicrobial activity of essential oil of Ferulago longistylis Boiss. fruits. Journal of Essential Oil Research, 20 (6): 569-573.

Polatoğlu, K., B. Demirci, F. Demirci, N. Güren & K. H. C. Başer, 2012c. Biological activity and essential oil composition of two new Tanacetum chiliophyllum (Fisch. & Mey.) Schultz Bip. var. chiliophyllum chemotypes from Turkey. Industrial Crops and Products, 39: 97-105.

Polatoğlu, K., F. Demirci, B. Demirci, N. Güren & K. H. C. Başer, 2012a. Essential Oil Composition and Antimicrobial Activities of Tanacetum chiliophyllum Fisch Mey Schultz Bip var monocephalum Grierson from Turkey. Record of Natural Products, 6 (4):184-188.

Polatoğlu, K., Ö. C. Karakoç & N. Gören, 2013. Phytotoxic, DPPH scavenging, insecticidal activities and essential oil composition of Achillea vermicularis. A. teretifolia and proposed chemotypes of A. biebersteinii (Asteraceae). Industrial Crops and Products, 51: 35-45.

Polatoğlu, K., A. Sen, A. Kandemir & N. Gören, 2012b. Essential oil composition and DPPH scavenging activity of endemic Tanacetum mucroniferum Hub.-Mor. & Grierson from Turkey. Journal of Essential Oil Bearing Plants, 15 (1): 66-74.

Polatoğlu, K., H. Servi, Ö. Özçinar, A. Nalbantsoy & S. Gücel, 2017. Essential Oil Composition of Endemic Arabis purpurea Sm. & Arabis cyperia Holmboe (Brassicaceae) from Cyprus. Journal of Oleo Science, 66 (1): 65-70.

Qari, S. H. & N. A. Abdel-Fattah, 2017. Genotoxic studies of selected plant oil extracts on Rhyzopertha dominica (Coleoptera: Bostrichidae). Journal of Taibah University for Science, 11 (3): 478-486.

Reyes-Jurado, F., T. Cervantes-Rincón, H. Bach, A. López-Malo & E. Palou, 2019. Antimicrobial activity of Mexican oregano (Lippia berlandieri). thyme (Thymus vulgaris) and mustard (Brassica nigra) essential oils in gaseous phase. Industrial Crops and Products, 131: 90-95.

Shaaya, E., U. Ravid, N. Paster, B. Juven, U. Zisman & V. Pissarev, 1991. Fumigant toxicity of essential oils against four major stored-product insects. Journal of Chemical Ecology, 17 (3): 499-504.

Shahriari, M., A. Zibaee, N. Sehebzadeh & L. Shamakhi, 2018. Effects of α-pinene. trans-anethole. and thymol as the essential oil constituents on antioxidant system and acetylcholine esterase of Ephestia kuehniella Zeller (Lepidoptera: Pyralidae). Pesticide Biochemistry and Physiology, 150: 40-47.

Shanker, C. & K. R. Solanki, 2000. Botanical insecticides: A historical perspective. India Asian Agrihistory, 4 (2): 21-30.
Szczepanik, M., M. Walczak, B. Zawitowska, M. Michalska-Sionkowska, A. Szumny, C. Wawrzeńczyk & M. S. Brzezinska, 2018. Chemical composition, antimicrobial activity and insecticidal activity against the lesser mealworm *Alphitobius diaperinus* (Panzera) (Coleoptera: Tenebrionidae) of *Origanum vulgare* L. ssp. *hirtum* (Link) and *Artemisia dracunculus* L. essential oils. Journal of the Science of Food and Agriculture, 98 (2): 767-774.

Taban, A., M. J. Saharkhiz & M. Hooshmandi, 2017. Insecticidal and repellent activity of three *Salvia* species against adult red flour beetles. *Tribolium castaneum* (Coleoptera: Tenebrionidae). Acta Ecologica Sinica, 37 (3): 201-206.

Tabanca, N., B. Demirci, T. Ozek, N. Kirimer, K. H. C. Başer, E. Bedir, I. A. Khan & D.E. Wedge, 2006. Gas chromatographic-mass spectrometric analysis of essential oils from *Pimpinella* species gathered from Central and Northern Turkey. Journal of Chromatography, 1117 (2): 194-205.

Taylor, R. W. D., 1994. Methyl bromide- Is there any future for this noteworthy fumigant? Journal of Stored Products Research, 30: 253-260.

Tripathi, A., S. Upadhyay, M. Bhuiyan & P. Bhattacharya, 2009. A review on prospects of essential oils as biopesticide in insect-pest management. Journal of Pharmacogn and Phytotherapy, 1: 52-63.

Vinciguerra, V., F. Rojas, V. Tedesco, G. Giusiano & L. Angiolella, 2018. Chemical characterization and antifungal activity of *Origanum vulgare*, *Thymus vulgaris* essential oils and carvacrol against *Malassezia furfur*. Natural Product Research, 4: 1-5.

Wijesundara, N. M. & H. V. Rupasinghe, 2018. Essential oils from *Origanum vulgare* and *Salvia officinalis* exhibit antibacterial and anti-biofilm activities against *Streptococcus pyogenes*. Microbial Pathogenesis, 117: 118-127.