

Isolation of Volatile Compounds with Repellent Properties against *Aedes albopictus* (Diptera: Culicidae) using CPC technology

**Anastasia Liakakou^{1#}, Apostolis Angelis^{1#}, Dimitrios P. Papachristos², Nikolas Fokialakis¹,
Antonios Michaelakis², Leandros A. Skaltsounis^{1*}**

¹Laboratory of Pharmacognosy and Natural Products Chemistry, School of Pharmacy, University of Athens, Panepistimioupoli, Zografou, 15771 Athens

²Benaki Phytopathological Institute, Scientific Directorate of Entomology and Agricultural Zoology, 14561, Kifissia;

[#]Equal contribution

^{*}Corresponding authors: Leandros A. Skaltsounis , Email; skaltsounis@pharm.uoa.gr

Contents

| | |
|--------------------|---|
| Table S1. | Species, plant parts, weight of extraction material, volume and yield of recovered EOs. |
| Table S2: | Identified compounds of PNI, PHE, PPI, JTU and JOX essential oils |
| Figure S1a: | TIC Chromatogram of PNI |
| Figure S1b: | TIC Chromatogram of PHE |
| Figure S1c: | TIC Chromatogram PPI |
| Figure S1d: | TIC Chromatogram of JTU |
| Figure S1e: | TIC Chromatogram of JOX |
| Figure S2a: | GC-MS chromatograms of upper (A) and lower phases (B) of JOX treated with the biphasic system Hept./ACN/ BuOH 1.6 / 1.6 / 0.2 (v/v/v) |
| Figure S2b: | GC-MS chromatograms of upper (A) and lower phases (B) of PPI treated with the biphasic system Hept./ACN/ BuOH 1.6 / 1.6 / 0.2 (v/v/v) |
| Figure S3: | TLC chromatograms of CPC fractions from analyses of JOX and PPI essential oils |

Table S1. Information regarding the species, plant parts, weight of extraction material, volume and yield (%v/w of tested materials) of recovered EOs.

| Species | Plant Part | Weight of row material (gr) | Volume of EO (ml) | % v/w |
|---|------------|-----------------------------|-------------------|-------|
| <i>P. nigra</i> subsp. <i>nigra</i> | wood | 141 | 0.2 | 0.07 |
| <i>P. heldreichii</i> | wood | 88 | 0.6 | 0.57 |
| <i>P. pinea</i> | needles | 14603 | 4 | 0.03 |
| <i>J. turbinata</i> | fruits | 330 | 6.5 | 1.96 |
| <i>J. oxycedrus</i> subsp. <i>deltoides</i> | fruits | 251 | 6.7 | 2.66 |

Table S2: Constituents listed in order of elution from a HP-5 MS column. (PNI: *P. nigra* subsp. *nigra*, PHE: *P. heldreichii*, PPI: *P. pinea*, JTU: *J. turbinata*, JOX: *J. oxycedrus* subsp. *deltoides*).

| aConstituents | KI | PNI | PHE | PPI | JTU | JOX |
|-----------------------------------|------|-------|-------|-------|-------|-------|
| Tricyclene | 921 | - | - | - | 0.48 | - |
| α -Pinene | 930 | 32.82 | 1.49 | 9.50 | 81.72 | 15.65 |
| Camphene | 944 | 0.71 | - | - | 0.85 | 0.12 |
| Sabinene | 966 | 0.12 | - | 0.59 | 0.52 | 0.21 |
| β -Pinene | 970 | 0.31 | - | 3.77 | 2.77 | 1.05 |
| Myrcene | 981 | 0.07 | - | 2.46 | 3.65 | 43.53 |
| α -Phelladrene | 993 | - | - | 0.56 | - | - |
| o-Cymene | 1019 | 0.38 | - | - | - | - |
| Limonene | 1021 | 1.70 | 3.48 | 45.05 | 1.70 | 4.00 |
| β -Phelladrene | 1024 | - | - | 12.79 | 0.16 | - |
| δ -3-Carene | 1027 | - | - | - | 0.21 | - |
| (E) - β - Ocimene | 1041 | - | - | 1.45 | - | - |
| Terpinolene | 1079 | - | - | 0.70 | - | 0.87 |
| Nonanal | 1092 | 0.50 | - | - | - | - |
| α -Fenchol | 1108 | 1.02 | - | - | - | - |
| α -Campholenal | 1120 | 2.26 | - | - | 0.37 | - |
| <i>cis</i> - β -Terpineol | 1136 | - | 47.41 | - | - | - |
| Camphen hydrate | 1140 | 0.41 | - | - | - | - |
| <i>trans</i> - β -Terpineol | 1151 | - | 6.56 | - | - | - |
| Pinocarvone | 1152 | 1.12 | - | - | - | - |
| p-Mentha1,5diene-8ol | 1162 | 0.89 | - | - | - | - |
| Terpinene4ol | 1165 | 0.86 | - | - | 0.21 | 0.23 |
| α -Terpineol | 1177 | - | 9.19 | - | 0.58 | - |
| Verbenone | 1199 | 0.08 | - | - | 0.55 | - |
| Carveol <i>trans</i> | 1211 | - | - | - | - | - |

| | | | | | | |
|--------------------------------------|------|------|-------|------|------|-------|
| Carvone | 1237 | 0.18 | - | - | - | - |
| Bornyl acetate | 1276 | - | - | - | - | 0.24 |
| Azulene | 1289 | 0.18 | - | - | - | - |
| δ -Elemene | 1327 | - | - | 0.51 | - | - |
| α -Terpinyl acetate | 1339 | - | - | - | 0.27 | - |
| α -Cubebene | 1340 | 0.07 | - | - | - | 0.22 |
| Citronellyl acetate | 1342 | - | - | - | - | 0.20 |
| α -Longipinene | 1343 | - | 0.77 | - | - | - |
| Longicyclene | 1361 | - | 0.18 | - | - | - |
| α -Ylangene | 1362 | 0.53 | - | - | - | - |
| α -Copaene | 1364 | 1.06 | - | - | - | 0.43 |
| Geranyl acetate | 1370 | - | - | - | - | 0.46 |
| β -Bourbonene | 1378 | 1.14 | - | - | - | |
| β -Elemene | 1379 | - | - | - | 0.15 | 0.12 |
| Longifolene | 1399 | - | 11.88 | 1.12 | - | - |
| β -Caryophyllene | 1410 | 0.28 | - | 6.40 | 0.99 | 1.76 |
| β -Copaene | 1423 | 0.59 | - | - | - | 0.10 |
| α -Guaiene | 1430 | - | - | 0.30 | - | |
| Z- β -Farnesene | 1434 | - | - | - | - | 0.36 |
| α -Humulene | 1442 | - | - | 1.12 | 0.53 | 1.58 |
| γ -Muurolene | 1468 | 4.93 | - | - | - | 0.32 |
| α -Amorphene | 1474 | 0.35 | - | - | - | |
| Germacrene D | 1475 | 0.61 | - | 3.29 | - | 23.77 |
| β -Selinene | 1487 | - | 0.77 | - | - | - |
| δ -Selinene | 1490 | - | - | 3.29 | - | - |
| <i>trans</i> -Muurola 4 (14) 5 diene | 1492 | - | - | - | 2.40 | - |
| γ -Amorphene | 1489 | 1.06 | - | - | - | - |
| α -Selinene | 1490 | - | 0.21 | - | - | - |
| α -Muurolene | 1499 | 1.11 | - | - | 0.08 | 0.46 |
| δ -Amorphene | 1505 | 0.17 | - | - | - | - |
| γ -Cadinene | 1505 | 2.54 | - | - | - | 0.26 |
| δ -Cadinene | 1514 | 5.23 | - | 0.73 | - | 2.74 |
| <i>trans</i> -Calamenene | 1518 | 1.04 | - | - | - | - |
| α -Cadinene | 1530 | 0.31 | - | - | - | - |
| α -Calacorene | 1539 | 0.66 | - | - | - | - |
| β -Calacorene | 1557 | 0.28 | - | - | - | - |
| Guaiol | 1593 | - | - | 4.25 | - | - |
| 1-epi Cubenol | 1612 | 0.56 | - | - | - | - |
| epi- α -Murrolol | 1632 | 0.68 | - | - | - | - |
| α -Cadinol | 1644 | 1.77 | - | - | - | - |
| Cadalene | 1669 | 0.81 | - | - | - | - |

| | | | | | | |
|---------------------------|------|-------|------|-------|-------|-------|
| Cembrene | 1929 | - | 6.26 | - | - | - |
| (3E)-Cembrene A | 1943 | - | 0.59 | - | - | - |
| Thunbergol | 2057 | | 9.61 | | | |
| Sandaracopimarinal | 2172 | 18.10 | - | - | - | - |
| Sandaracopimarinol | 2268 | 3.45 | - | - | - | - |
| Abietal | 2300 | 1.17 | - | - | - | - |
| Total identified (%) | | 92.11 | 98.4 | 97.88 | 98.19 | 98.68 |
| Total number of compounds | | 43 | 13 | 18 | 19 | 23 |

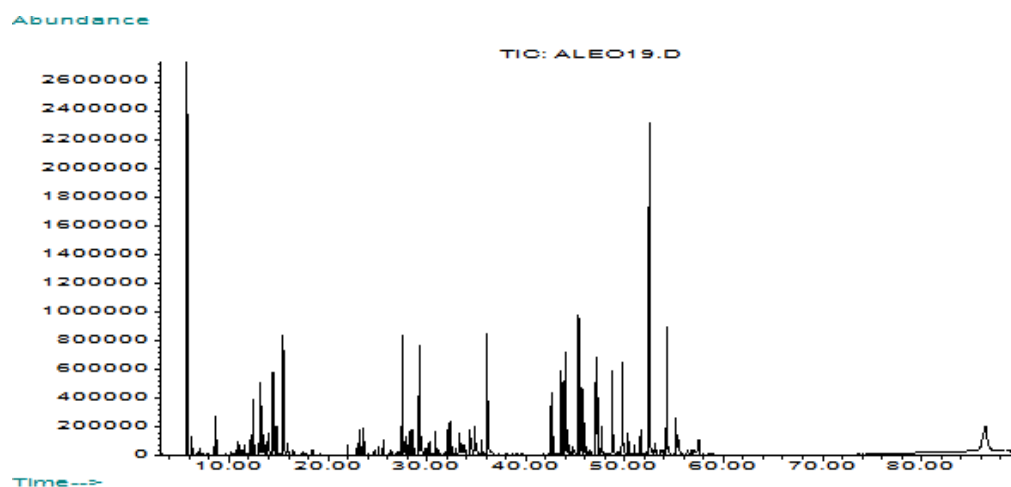


Figure S1a: TIC Chromatogram of *Pinus nigra* subsp. *nigra* (wood)

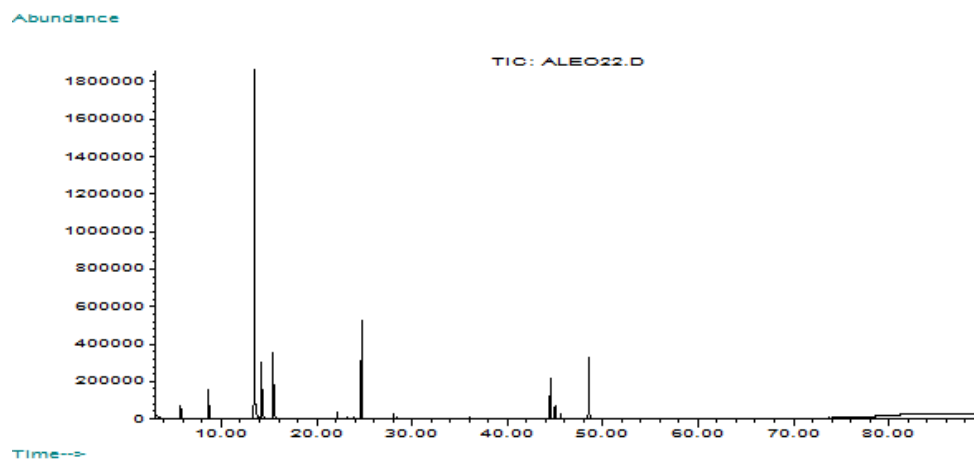


Figure S1b: TIC Chromatogram of *Pinus heldreichii* (wood)

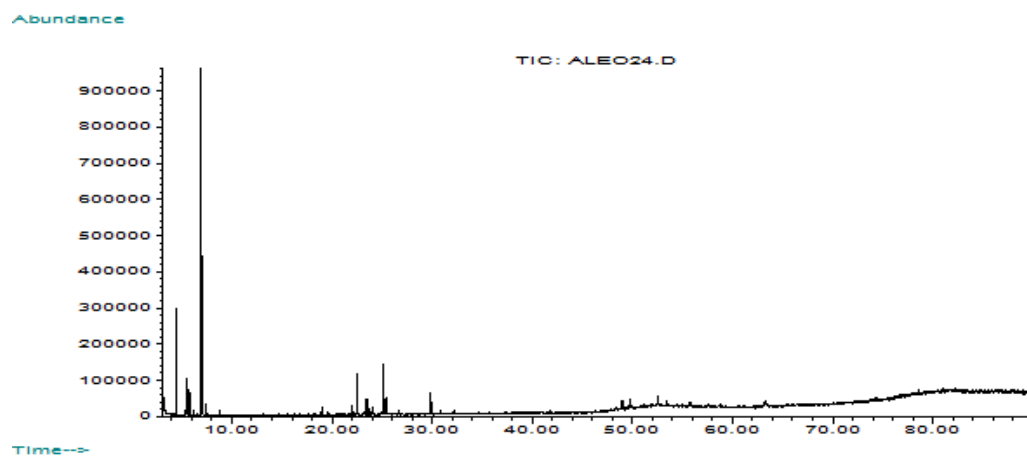


Figure S1c: TIC Chromatogram *Pinus pinea* (needles)

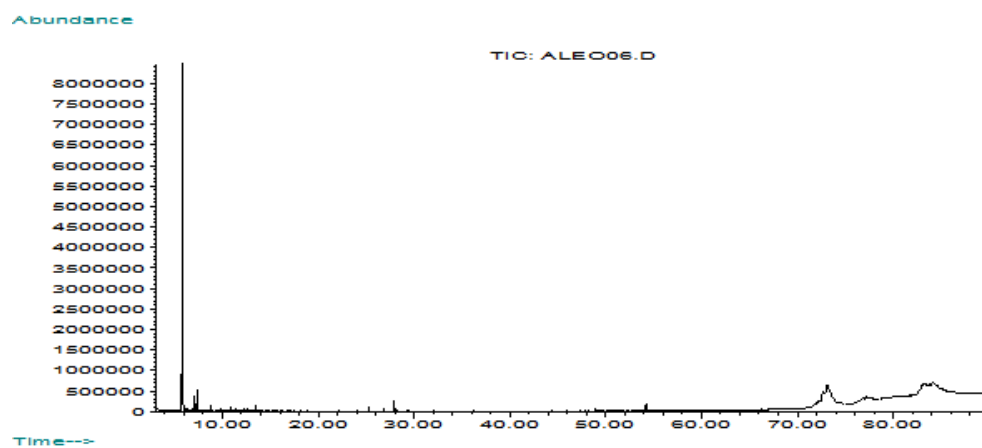


Figure S1d: TIC Chromatogram of *Juniperus turbinata* (fruits)

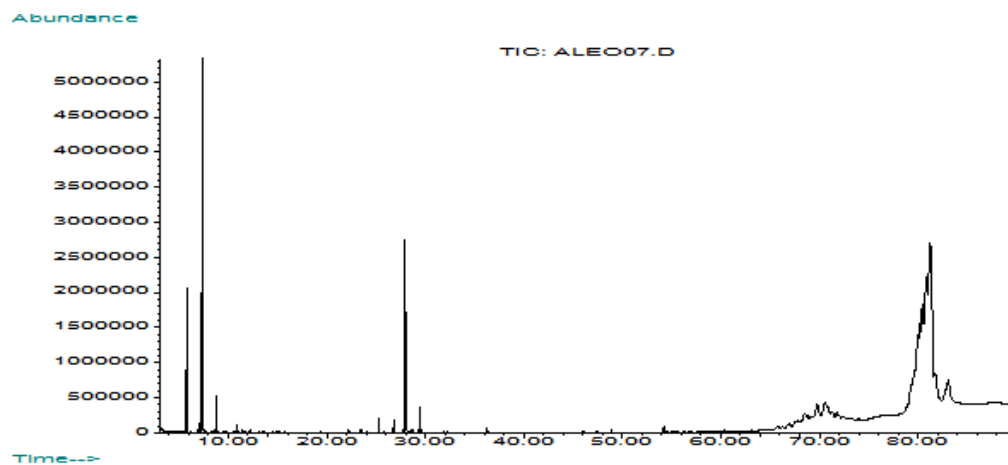


Figure S1e: TIC Chromatogram of *Juniperus oxycedrus* subsp. *deltooides* (fruits)

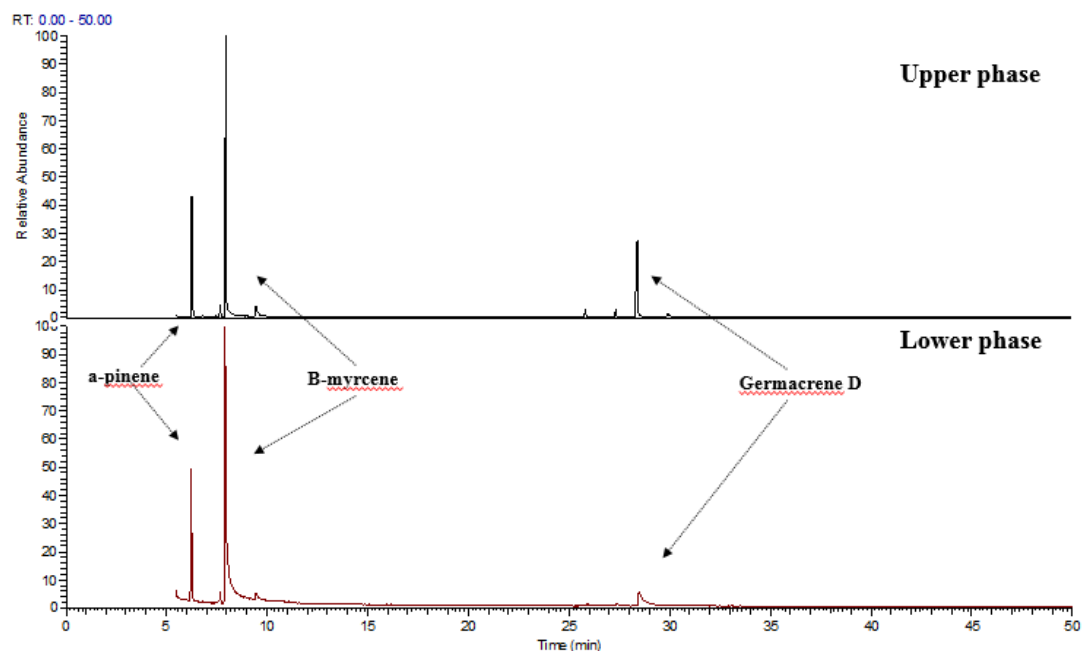


Figure S2a: Upper (A) and lower phase (B) GC-MS chromatograms obtained from *J. oxycedrus* subsp. *deltooides* essential oil with Heptane / Acetonitrile / Butanol biphasic system 1.6 / 1.6 / 0.2 v

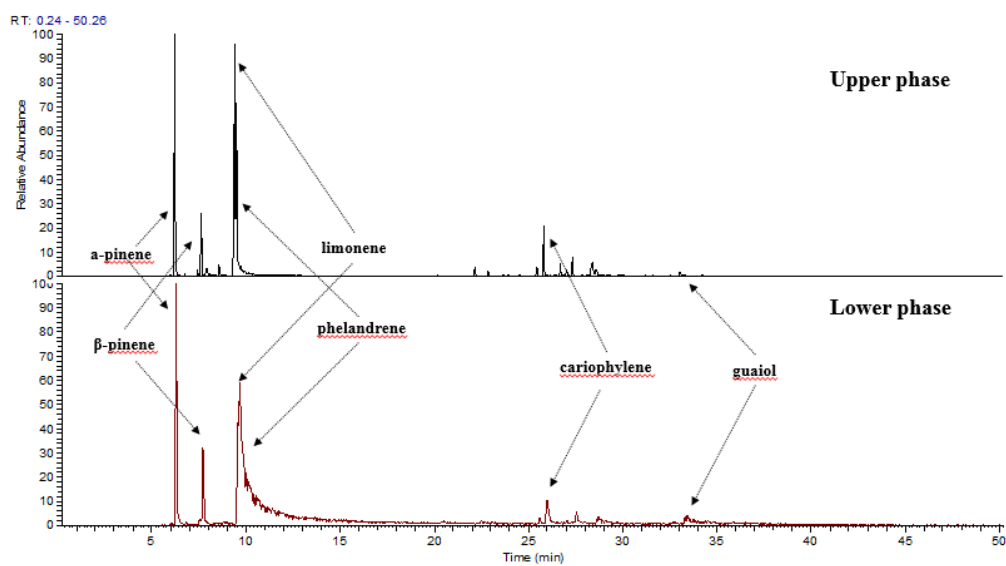


Figure S2b: Upper (A) and lower phase (B) GC-MS chromatograms obtained from *P. pinea* essential oil with Heptane / Acetonitrile / Butanol biphasic system 1.6 / 1.6 / 0.2 v

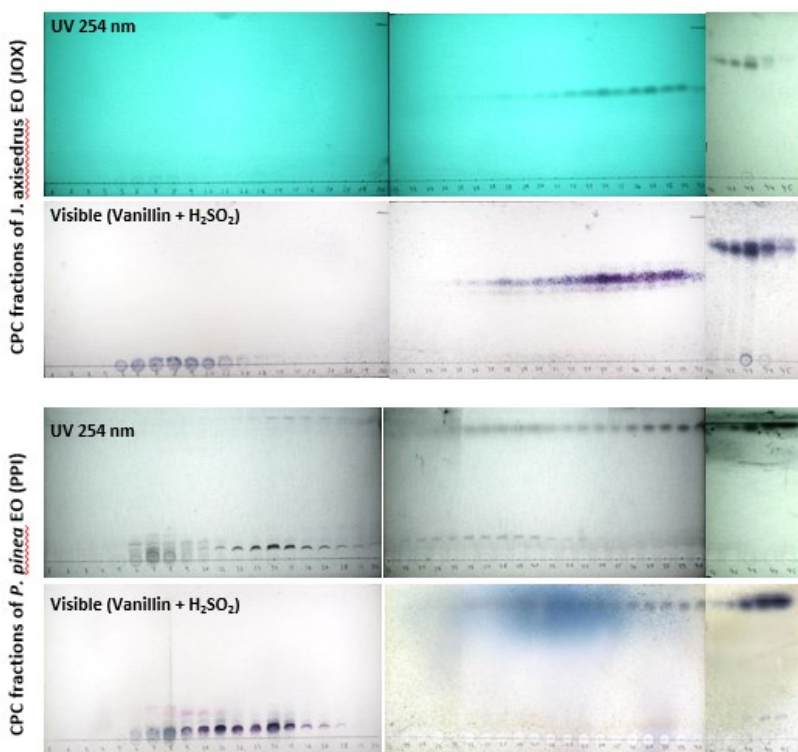


Figure S3: TLC chromatograms of fractions from CPC analyses of the essential oils of *J. oxycedrus* subsp. *deltoides* (upper) and *P. pinea* (lower) observation at UV-254 nm and visible after spraying with vanillin sulfate solution and heating