

Supplementary Materials

# Antioxidant and Toxic Activity of *Helichrysum arenarium* (L.) Moench and *Helichrysum italicum* (Roth) G. Don Essential Oils and Extracts

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**Table S1.** Principal chemical composition (compounds  $\geq 5.0\%$ ) of *Helichrysum italicum* (Roth) G. Don essential oils investigated in various countries for last ten years \*.

Plant Material	Main Constituents, %
20 native <i>H. italicum</i> ssp. <i>italicum</i> genotypes collected from different locations in Italy and Corsica (France) and grown under similar condition [5]	Several chemotypes were described. Neryl acetate ( $\leq 32.0$ ), $\beta$ - or $\alpha$ -selinene ( $\leq 38.0$ and $\leq 26.5\%$ , respectively), $\beta$ -bisabolene (32.3), $\gamma$ -curcumene ( $\leq 27.7$ ), nerol (18.8), $\beta$ -caryophyllene ( $\leq 18.6$ ), caryophyllene oxide (14.9), carvacrol ( $\leq 14.8$ ), $\beta$ - or $\gamma$ -eudesmol (13.7 and $\leq 20.4$ , respectively), <i>ar</i> -curcumene ( $\leq 8.3$ ), neryl propanoate (6.8); 4,6, 9-trimethyldec-8-en-3, 5-dione ( $\leq 11.0$ ); 5, 7, 10-trimethylundec-9-en-4,6-dione ( $\leq 6.9$ )
ssp. <i>microphyllum</i> from Sardinia, under different weather conditions, from their vegetative period to post-blooming time [6]	Neryl acetate ( $\leq 35.6$ ), eudesmen-5-en-11-ol ( $\leq 23.5$ ), nerol ( $\leq 14.4$ )
ssp. not indicated, from Elba Island [7,9]	Neryl acetate (25.3), $\alpha$ -pinene (14.5), limonene (12.3), $\gamma$ -curcumene (8.7),
ssp. not indicated, flowers from Konavle region, Croatia [8]	$\gamma$ -Curcumene (12.4), $\beta$ -selinene (9.9), $\alpha$ -selinene (5.9)
ssp. <i>italicum</i> from the National Park of Cilento and Diano Valley, Southern Italy [10]	<i>iso</i> -Italicene epoxide (16.8), $\beta$ -costol (7.5)
ssp. <i>italicum</i> from Corsica, France [11]	Neryl acetate (31.0), $\gamma$ -curcumene (10.7), neryl propionate (5.1)
ssp. not indicated, collected in 7 locations of Elba Island (Tuscany, Italy) [12]	Neryl acetate ( $\leq 45.9$ ), nerol ( $\leq 12.8$ )
Diversity of 50 <i>H. italicum</i> (ssp. not indicated) populations collected in Sardinia [13]	Neryl acetate, nerol, neryl propionate, eudesm-5-en-11-ol, $\gamma$ -cadinene, <i>cis</i> - $\beta$ -guaiene, nerolidol
Flowers (ssp. not indicated) collected in the region Konavle, Southern part of Croatia [14]	CO <sub>2</sub> extracts: octacosane (10.2), canellal (10.0), cembrenol (7.4), santonine (6.6) EO: $\alpha$ -pinene (5.0)
Commercial EO sample of <i>H. italicum</i> (ssp. not indicated) from Herba d.o.o, Belgrade, Serbia [15]	$\gamma$ -Curcumene (22.5), $\alpha$ -pinene (15.9), neryl acetate (7.8), $\beta$ -selinene (6.9)
ssp. not indicated, collected from 4 populations (on Brač Island, Biokovo Mt. near Tjarica and near Makarska) in Dalmatia, Croatia [16]	$\alpha$ - <i>trans</i> -Bergamotene (10.2), $\beta$ -acoradiene (10.1), rosifoliol (8.5), neryl acetate (8.1)
ssp. <i>italicum</i> collected in Montenegro [17]	Neryl acetate (28.2), $\gamma$ -curcumene (18.8), neryl propionate (9.1), <i>ar</i> -curcumene (8.3)

ssp. <i>picardii</i> collected from field-growing plants at Vila Real de Santo António, Algarve, Portugal [18]	$\alpha$ -Pinene (53.5), $\gamma$ -curcumene (27.4)
ssp. <i>italicum</i> , 8 EOs from Sicily, plants harvested in the oriental region of Etna volcano; 2 EOs from southern Corsica [19]	Sicilian EOs: $\beta$ -selinene (14.0), rosifoliol (13.9), $\alpha$ -selinene (11.1), aromadendrene (11.0), ledol (8.6), $\beta$ -caryophyllene (7.6), $\alpha$ -pinene (7.1) Corsican EOs: neryl acetate (46.2), $\alpha$ -pinene (12.5), $\gamma$ -curcumene (11.7)
146 <i>H. italicum</i> ssp. <i>microphyllum</i> genotypes from two contrasting habitats (seaside and mountains) in Sardinia [20]	Neryl acetate ( $\leq$ 83.4), <i>cis</i> - $\beta$ -guaiene (58.2), ( <i>E</i> )-nerolidol ( $\leq$ 55.6), eudesmen-7-(11)-en-4-ol ( $\leq$ 46.1), limonene ( $\leq$ 15.7), neryl propionate ( $\leq$ 14.1), nerol ( $\leq$ 13.6), $\gamma$ -curcumene ( $\leq$ 10.6), 1,8-cineole (8.9), neryl-isobutanoate (7.2), <i>epi</i> -cadinol (6.7), $\delta$ -cadinene (6.3)
var. <i>numidicum</i> Pomel from Bibans, Algeria [21]	Isopropyl tetradecanoate (12.1), $\alpha$ -pinene (12.0), hexadecanoic acid (10.0), ( <i>E</i> )-caryophyllene (9.2), ledol (9.1), palustrol (5.6)
ssp. not indicated from Béjaia, North of Algeria [22]	$\alpha$ -Cedrene (13.6), $\alpha$ -curcumene (11.4), geranyl acetate (10.1), limonene (6.1), nerol (5.0)
ssp. not indicated, commercial EO sample from doTERRA Intl., Pleasant Grove, UT, USA [23]	Neryl acetate (35.5), $\gamma$ -curcumene (13.9), $\alpha$ -pinene (8.9)
ssp. not indicated, collected at continental part of Zadar and Šibenik-Knin County, Central Dalmatia, Croatia [24]	$\alpha$ -Pinene (21.6), $\gamma$ -curcumene (21.6), neryl acetate (7.9), $\beta$ -selinene (6.5), <i>iso</i> -italicene (5.4)
ssp. not indicated, plants (of Corsican orifgin) cultivated in Turia village, municipality of Pavel Banya, Stara Zagora region, South Bulgaria harvested in different phenological stages [25]	Citronellol isobutanoate ( $\leq$ 25.8), neryl acetate ( $\leq$ 20.6), $\alpha$ -pinene ( $\leq$ 19.5), $\alpha$ -copaene ( $\leq$ 12.8), eudesm-5-en-11-ol (8.1), $\beta$ -eudesmene ( $\leq$ 5.6)
ssp. <i>italicum</i> , commercial plant samples from Roing, Ljubuški, Bosnia and Herzegovina [26]	$\alpha$ -Pinene ( $\leq$ 29.4), limonene ( $\leq$ 5.5), neryl acetate ( $\leq$ 7.8), italicene ( $\leq$ 7.1), 2,4,6,9-tetramethyldec-8-en-3,5-dione B (7.1), $\beta$ -selinene ( $\leq$ 5.6), $\alpha$ -curcumene (5.0)
ssp. not indicated, collected near Benkovac, Croatia [27]	Neryl acetate (20.4), $\gamma$ -curcumene (14.1), <i>trans</i> - $\alpha$ -bergamotene (7.0)
ssp. <i>italicum</i> cultivated in Marche region, Central Italy [28]	Neryl acetate (15.7), $\alpha$ -pinene (8.2), 4,6,9-trimethyl-8-decene-3,5-dione (italidione I) (7.3), <i>ar</i> -curcumene (5.4), $\beta$ -selinene (5.4)
ssp. <i>italicum</i> , commercial EO from Donna è Company, Monticellu; EO was produced from wild plants collected in the area of Lumiu, Northern Corsica [29]	Neryl acetate (30.4), $\gamma$ -curcumene (10.1), neryl propionate (7.1), <i>ar</i> -curcumene (5.8)
ssp. <i>italicum</i> collected during five different periods in Herzegovina [30]	$\alpha$ -Pinene (15.7), $\gamma$ -curcumene (12.8), 4,6,9-trimethyldec-8-en-3,5-dione (8.7), neryl acetate (6.9), limonene (6.4), $\beta$ -selinene (5.3)
ssp. <i>italicum</i> , commercial EO from Casa Vecchia Corsa, Sartène, Corsica, France [31]	Neryl acetate (35.0), neryl propionate (5.6) in ester-containing chromatographic fraction
Commercial EO sample from Agro Life Montenegro d.o.o. ssp. not indicated, plants collected in Tuzi near Podgorica, Montenegro [32]	In liquid EO: $\beta$ -eudesmene (21.6), $\beta$ -bisabolene (19.9), $\alpha$ -pinene (16.9), neryl acetate (10.7) In the vapour EO phase: $\alpha$ -pinene (78.8)
two subspecies (ssp. <i>microphyllum</i> (plants origin from Bosnia) and ssp. <i>italicum</i> (French origin)) cultivated in the area of Kazanlak, Bulgaria [33]	ssp. <i>microphyllum</i> : $\alpha$ -pinene (20.8), $\gamma$ -curcumene (16.5), $\beta$ -selinene (5.6) ssp. <i>italicum</i> : neryl acetate (33.9), $\gamma$ -curcumene (8.8), rosifoliol (5.5), geranyl propionate (5.0),

ssp. not indicated, from Botanical Garden of the University of Rome, Italy [34]	Neryl acetate (34.0), $\alpha$ -pinene (28.5), nerol (8.0), neryl phenylacetate (7.1), $\beta$ -caryophyllene (5.7)
ssp. not indicated, flowering parts of <i>H. italicum</i> introduced from Čapljina (Bosnia and Hercegovina) and cultivate in the experimental fields of the Institute of Field and Vegetable Crops (IFVCNS) in Bački Petrovac, Novi Sad, Serbia [35]	$\gamma$ -Curcumene (13.6), $\beta$ -selinene (12.2) $\alpha$ -pinene (11.8), $\beta$ -caryophyllene (6.7), neryl acetate (5.5)

\*The published data presented by chronological order (from 2010 to 2021).

**Table S2.** Summary of previously reported data on chemical composition and bioactivity of essential oils of *H. arenarium* (L.) Moench) plants grown in different countries worldwide \*.

Plant Material	EO Preparation Procedure	Main constituents ( $\geq 5.0$ ), %	Biological Activity, Evaluation Method, Results
Inflorescences of <i>H. arenarium</i> (Helichrysi flos - GYNKI B 145/98 MSZ 1.0.) collected in the region of Caucasus were purchased from Hungarian commercial herb distributor [78]	EO was obtained by HD procedure described in the VII. Hungarian Pharmacopoeia. Yield of EO was 0.09%.	Methyl palmitate (28.5), dodecanoic acid (11.9), decanoic acid (9.8), octanoic acid (6.0)	Not investigated
Hungarian and Polish mercantile samples and a plant sample cultivated in Hungary (Soroksár) [79]	EOs prepared by steam-distillation	Methyl palmitate (8.0-28.5), capric acid (8.5-19.8%), lauric acid (6.5-14.6), $\delta$ -cadinene (5.2-7.9), thymol (7.7), pelargonic acid (5.2-6.9), carvacrol (6.2), caprylic acid (6.0)	Not investigated
Plants were collected from Kladovo, Serbia and Montenegro in the flowering period [56]	EO was obtained from the Faculty of Technology and Metallurgy, University of Belgrade (from S. Šiler-Marinković), other details not specified.	Di-epi- $\alpha$ -cedrene (17.9), $\alpha$ -ylangene (14.0), cyclosativene (11.9), limonene (11.4), $\alpha$ -selinene (8.2)	Antifungal activity by microdilution test using <i>Aspergillus niger</i> , <i>A. flavus</i> , <i>Cladosporium cladosporioides</i> , <i>Penicillium funiculosum</i> and <i>Trichoderma viride</i> . MIC = 10–30 $\mu$ g/mL; MFC = 15–60 $\mu$ g/mL. Antimicrobial activity against the Gram (+) ( <i>Micrococcus luteus</i> , <i>Staphylococcus aureus</i> , <i>Staphylococcus epidermidis</i> ) and Gram (–) bacteria ( <i>Escherichia coli</i> , <i>Pseudomonas tolaasii</i> , <i>Salmonella enteritidis</i> , <i>Salmonella typhimurium</i> ) and yeast <i>Candida albicans</i> by a bioautographic test on TLC plates. EO showed a strong antimicrobial activity: a

			volume of 1 µL had antimicrobial activity against all bacterial and yeast species tested.
<i>H. arenarium</i> plants with yellow and orange [f. <i>aurantiacum</i> (Pers.) Bleck.] flowers collected from the same natural habitats in Svencionys and Ukmerge districts, Eastern Lithuania [80]	HD for 2 h, using a mixture of hexane and diethyl ether as a collecting solvent.	Heneicosane (32.1), β-caryophyllene (13.5–25.6), δ-cadinene (9.7 and 14.4), octadecane (8.9)	Not investigated
Plants collected from natural populations, and cultivated in Vilnius, Lithuania. Aerial parts gathered at flowering stage from the field collection [81]	EO from inflorescences of six different colours: citric, citric-yellow, yellow, yellow-brown, orange, and orange-brown. HD for 3 h, a mixture of pentane and diethyl ether (1:1) used as a solvent.	<i>n</i> -Nonadecane (12.2), trans-caryophyllene (5.3–8.8), δ-cadinene (5.1–8.2), tetradecanoic acid (7.0 and 7.8), caryophyllene oxide (5.7), heneicosane (4.9–5.1)	Not investigated
Aerial parts in Khorasan Razavi Province (the Northeast of Iran) [62]	Essential oil of aerial parts of the <i>H. arenarium</i> L. was extracted by steam distillation using a Clevenger apparatus according to the method of British Pharmacopoeia.	Not detailed	<i>In vitro</i> on the growth of seven microbial species: <i>Bacillus subtilis</i> , <i>Escherichia coli</i> , <i>Staphylococcus aureus</i> , <i>Saccharomyces cerevisiae</i> , <i>Candida albicans</i> , <i>Aspergillus flavus</i> and <i>Aspergillus parasiticus</i> , using a micro-dilution method. Results showed that <i>B. subtilis</i> (MIC = 781.25 and MBC = 6250 µg/ml) was more resistance than the two other bacterial species. Among the two yeasts tested, <i>S. cerevisiae</i> (MIC = 97.65 and MFC = 781.25 µg/ml) was more sensitive than <i>C. albicans</i> (MIC = 195.31 and MFC = 3125 µg/ml) while among the fungal species, growth of <i>A. parasiticus</i> inhibited at lower concentration of the EO rather than <i>A. flavus</i> . In liquid medium (Mullerhinton broth), EO exhibited the same MIC value against both fungal strains (48.82 µg/ml), while in solid medium (Mullerhinton

			agar) different activity was observed against <i>A. flavus</i> and <i>A. parasiticus</i> (MFC = 6250 and 390.625 µg/ml, respectively).
Plants collected in Khorasan Razavi Province, Northeast of Iran [64]	EO from aerial parts extracted by water steam distillation, using a Clevenger apparatus according to the method of British Pharmacopoeia. Other details not specified.	α-Pinene (32), 1,8-cineole (16), α-humulene (15), β-caryophyllene (8)	Antimicrobial activity <i>in vitro</i> on the growth of three food born pathogens: <i>Serratia marcescens</i> , <i>Streptococcus agalactiae</i> and <i>Staphylococcus aureus</i> , using a micro-dilution method. MIC: 406, 812 and 812 µg/mL for <i>S. marcescens</i> , <i>S. agalactiae</i> and <i>S. aureus</i> ; respectively. MBC: 812, 3250 and 3250 µg/mL, respectively.
<i>H. arenarium</i> , native from different geographical areas, but acclimatised and grown in homogeneous conditions in Italy (CREA-Sanremo collection) [83]	EO extraction: leaves and flowers, separately from the three <i>Helichrysum</i> species were subjected to HD by Clevenger-type apparatus (3h). EOs had very low yield (≤0.01%), so they were collected with n-hexane (HPLC grade) and stored at -4 °C in the dark until use. Solid Phase Micro Extraction analyses (SPME): The analyses were performed using Supelco SPME device coated with polydimethylsiloxane (PDMS, 100 µm) for VOC emission of flowers and leaves.	In flower EO: methyl ester of pentadecanoic acid (31.0), β-caryophyllene (27.5), β-pinene (7.4) In leaf Eo: β-caryophyllene (46.0), 1,8-cineole (8.3), limonene (5.5) Volatile emission by SPME analysis: In flowers: β-pinene (55.2), α-pinene (15.8), limonene (5.2), 1,8-cineole (5.1); in leaves, (Z)-3-hexenol acetate (49.6), butanoic acid, (Z)-3-methyl-3-hexenyl ester (11.1), 1,8-cineole (7.4), β-pinene (5.8)	Not investigated
Plants collected at full flowering from 7 locations (Birstonas, Varena Alytus, Ukmerge, and Moletai districts, and Neringa city) in Lithuania, mainly in the coniferous woods [82]	Leaves and inflorescences of various intensities of the yellow colour. HD for 3 h, mixture of pentane and diethyl ether (1:1) used as a collecting solvent. The yield of EOs was ≤0.5%.	β-Caryophyllene (5.8–36.2), octadecane (7.1–22.3), heneicosane (7.9–20.0) δ-cadinene (9.0), 1,8-cineole (8.9), γ-cadinene (5.8)	Not investigated
The aerial parts were hand-harvested during flowering from Alax in Inner Mongolia, China [74]	By HD concatenated with a liquid-liquid extraction. Traditional Clevenger apparatus. was modified: two separation columns were	β-Spatulenol (19.9–24.0), ledol (6.2–10.0), bicyclogermacrene (5.7), aromadendrene (5.2)	Not investigated

added. The first one is the conventional oil-water separation column, and the second one is the extraction column for EO components from hydrosol using organic solvents. The theoretical yield of the total EO was 5.10 mg/g.

The aerial parts were collected in full flowering stage from Yozgat Akdagmadeni district (at altitude of 1760 m), Turkey [84]	HD for 2 h in a laboratory glass apparatus of the British Pharmacopoeia. The yield of EO was 0.07%.	Oleic acid (30.3), ethyl hexadecanoate (20.2), linoleic acid (18.9)	Not investigated
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\*The published data presented by chronological order (2000–2020); HD-hydrodistillation procedure.

**Table S3.** Chemical composition of essential oils obtained from *H. arenarium* inflorescences and leaves and *H. italicum* inflorescences.

Compound <sup>a</sup>	<sup>b</sup> <i>RIn</i> <sub>Lit</sub>	<sup>c</sup> <i>RIn</i> <sub>exp</sub>	<sup>d</sup> <i>RIp</i> <sub>exp</sub>	<i>H. aren.</i> Flowers	<i>H. aren.</i> Leaves	<i>H. ital.</i> Flowers
2-Heptanone	892	891		0.6 ± 0.25	0.1 ± 0	
<i>n</i> -Nonane	900	900		0.4 ± 0.10	0.3 ± 0.15	
Heptanal	902	903		0.3 ± 0.10	1.1 ± 0.40	
α-Pinene *	932	938	1035	0.3 ± 0.25	4.2 ± 1.15	6.5 ± 1.50
Camphene	954	955	1070		0.4 ± 0.30	
2-Z-Heptenal		959	1319		0.7 ± 0.20	
Benzaldehyde	960	964		1.6 ± 0.56		
β-Pinene*	979	978	1113	0.1 ± 0.21	0.8 ± 0.25	
1-Octen-3-ol	979	980			0.8 ± 0.10	
1-Octen-3-one		982	1285	0.1 ± 0.10		
2-Pentilfuran		996	1237	1.7 ± 0.55	2.3 ± 0.30	
trans-2-(2-Pentenyl)furan		1003	1284	0.4 ± 0.20	1.2 ± 0.25	
<i>n</i> -Octanal	999	1004		0.3 ± 0.30	0.9 ± 0.35	
Limonene	1029	1030	1196	tr.	3.0 ± 0.50	1.6 ± 0.60
1,8-Cineole*	1031	1032	1218		3.9 ± 0.60	
γ-Terpinene	1060	1060	1255	tr.	0.7 ± 0.70	
Linalool*	1097	1098	1550			2.1 ± 0.45
α-Campholenal	1126	1125			0.1 ± 0.06	
Camphor*	1146	1145	1518			0.5 ± 0.20
<i>n</i> -Nonanal	1101	1103	1398	0.7 ± 0.20	10.4 ± 1.50	
2 <i>E</i> -Nonen-1-al	1161	1160		0.5 ± 0.50	0.9 ± 0.25	
Isoborneol	1162	1164	1660	0.1 ± 0.10		0.4 ± 0.20
Terpinen-4-ol	1177	1181	1604		0.4 ± 0.25	
Octanoic (Caprylic) acid		1185	2070?	0.7 ± 0.65	0.2 ± 0.2	
α-Terpineol	1189	1188	1698	0.6 ± 0.45	1.3 ± 0.35	0.8 ± 0.40
<i>n</i> -Dodecane	1200	1200		0.2 ± 0.20		
Decanal	1202	1202	1503	0.9 ± 0.20	2.1 ± 2.0	
Geraniol	1253	1255	1845			1.3 ± 0.3
2 <i>E</i> -Decenal	1264	1264			0.4 ± 0.40	
Nonanoic (Pelargonic) acid	1271	1269		0.8 ± 0.70	1.2 ± 0.25	

<i>E</i> -Anethole	1285	1285				1.1 ± 0.35
2-Undecanone	1294	1292		0.2 ± 0.15		
Undecanal	1307	1305	1602	0.4 ± 0.15	0.4 ± 0.35	
2 <i>E</i> ,2 <i>E</i> -decadienal	1317	1315			1.5 ± 1.45	
Eugenol	1359	1363	2179	0.8 ± 0.25		
α-Ylangene	1375	1372	1491	0.2 ± 0.15	0.4 ± 0.15	
α-Copaene	1377	1376	1497	1.4 ± 0.60	2.1 ± 0.45	
<i>n</i> -Decanoic (Capric) acid		1382	2275	1.2 ± 0.55	1.1 ± 0.15	
<i>E</i> -β-Demascenone	1385	1388		0.5 ± 0.00		
<i>n</i> -Tetradecane	1400	1400		1.5 ± 0.25		
<i>iso</i> -Italicene	1402	1405				0.7 ± 0.15
Italicene	1406	1406				2.2 ± 0.75
<i>n</i> -Dodecanal	1409	1408		0.5 ± 0.35		
<i>cis</i> -α-Bergamotene	1413	1412				0.2 ± 0.06
<i>E</i> -Caryophyllene*	1419	1418	1608	5.4 ± 0.55	6.5 ± 0.55	3.3 ± 0.30
<i>trans</i> -α-Bergamotene	1435	1439				0.7 ± 0.06
Aromadendrene	1441	1440	1615	0.2 ± 0.1	0.2 ± 0.15	0.8 ± 0.30
α-Humulene*	1455	1455	1680	0.9 ± 0.2	0.4 ± 0.15	
<i>Z</i> -β-Farnesene			1645		1.8 ± 0.75	
α-Acoradiene	1466	1468	1670			0.5 ± 0.0
γ-Muurolene	1480	1478	1690	0.4 ± 0.31	1.1 ± 0.10	
γ-Curcumene	1483	1486				21.5 ± 2.50
<i>E</i> -β-Ionone	1489	1501	1955		1.7 ± 0.75	
β-Selinene	1490	1490	1732			13.6 ± 1.65
α-Selinene	1498	1499				8.1 ± 0.55
<i>n</i> -Pentadecane	1500	1500		0.4 ± 0.15		
α-Muurolene	1500	1502		0.4 ± 0.35	0.6 ± 0.55	
γ-Cadinene	1514	1515	1764	0.6 ± 0.35	0.9 ± 0.90	0.8 ± 0.20
δ-Cadinene	1523	1520	1760	1.6 ± 0.35		1.3 ± 0.65
Italicene ether	1538	1533				0.6 ± 0.06
α-Cadinene	1539	1536	1775		0.3 ± 0.25	
α-Calacorene	1546	1545	1934	0.2 ± 0.15		
β-Calacorene	1566	1568			0.3 ± 0.30	
<i>E</i> -Nerolidol	1563	1567	2045		0.2 ± 0.20	
Caryophyllenyl alcohol	1572	1572			2.3 ± 0.35	
Dodecanoic (Lauric) acid		1577	2520	6.1 ± 1.35	2.0 ± 1.55	3.0 ± 0.15
Spathulenol	1578	1580				1.1 ± 0.10
β-Caryophyllene oxide*	1583	1584	2000	0.6 ± 0.35	1.2 ± 0.35	1.0 ± 0.15
Guaiol	1601	1602				1.1 ± 0.45
Humulene epoxide II	1608	1608	2053		0.2 ± 0.20	
1,10-di- <i>epi</i> -Cubenol	1619	1620			0.6 ± 0.60	
14-hydroxy-9- <i>epi</i> - <i>E</i> -Caryophyllene	1670	1670		0.4 ± 0.2	0.3 ± 0.30	
β-Eudesmol	1649	1650	2237			8.3 ± 0.35
α-Cadinol	1654	1655	2249	0.7 ± 0.3	0.4 ± 0.35	
Selin-11-en-4-α-ol	1660	1660				0.7 ± 0.01
Tridecanoic (Tridecylic) acid		1671	2605			
<i>n</i> -Tetradecanol	1673	1672	1917	2.8 ± 0.35		
Eudesm-7-(11)-en-4-ol	1700	1703				4.4 ± 0.40
<i>n</i> -Tetradecanoic (Myristic) acid		1741	2713	14.9 ± 1.05	8.7 ± 1.35	0.5 ± 0
Phytone (Hexahydrofarnesyl acetone)		1838	2113	4.4 ± 0.55	1.4 ± 0.85	
<i>n</i> -Pentadecanoic (Pentadecylic) acid		1855	2820	2.1 ± 1.20	1.9 ± 0.95	

<i>n</i> -Hexadecanoic (Palmitic) acid		1945	2911	$23.8 \pm 1.13$	$18.8 \pm 0.70$	$0.2 \pm 0.06$
Phytol	1943	1948		$0.4 \pm 0.10$	$0.2 \pm 0.15$	
<i>n</i> -Eicosane	2000	2000		$0.3 \pm 0.15$	$0.4 \pm 0.10$	
Methyl linolenate		2075	2590	$5.3 \pm 0.75$	$2.7 \pm 1.65$	
<i>n</i> -Heneicosane	2100	2100		$2.1 \pm 0.25$	$0.9 \pm 0.25$	$0.2 \pm 0.06$
<i>n</i> -Docosane	2200	2200		$3.8 \pm 0.35$	$0.3 \pm 0.10$	$0.3 \pm 0.06$
<i>n</i> -Tricosane	2300	2300		$0.3 \pm 0.12$		
Average Total				$96.4 \pm 1.52$	$99.1 \pm 0.44$	$89.4 \pm 0.15$

<sup>a</sup> Constituents are listed in order of their elution from a non-polar DB-5 (which is identical to a Rxi-5MS) column, compounds are identified by their mass spectra and retention indices on both (polar HP-FFAP and nonpolar Rxi-5MS) columns; \* Additional identification with reference compound; tr.-traces (<0.05%). <sup>b</sup>  $RIn_{Lit}$ : Kovat's indices for the nonpolar column DB-5 taken from the literature [85]. <sup>c</sup>  $RIn_{exp}$ : Retention indices determined experimentally on the nonpolar column Rxi-5MS (which is identical to DB-5). <sup>d</sup>  $RIp_{exp}$ : Retention indices determined experimentally on the polar column HP-FFAP.



**Figure S1.** Geographical indication of *H. arenarium* (L.) Moench sampling site in Eastern Lithuania (Utena district).

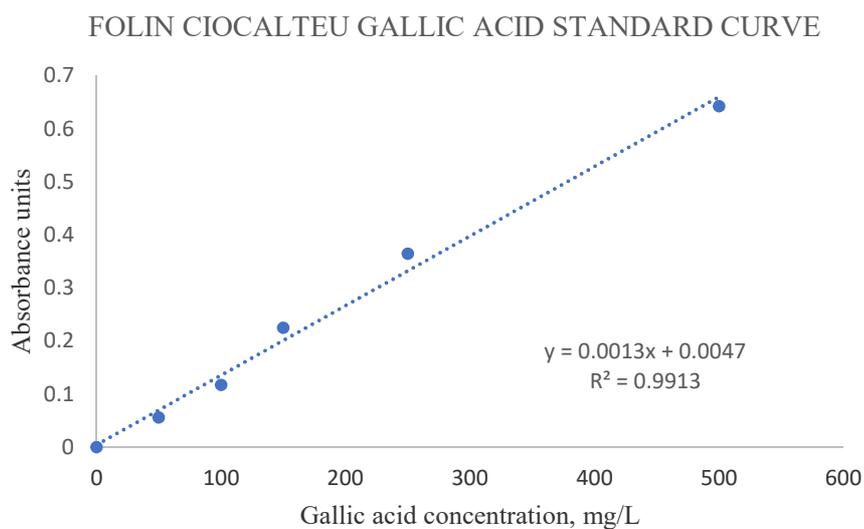


Figure S2. Gallic acid standard calibration curve.

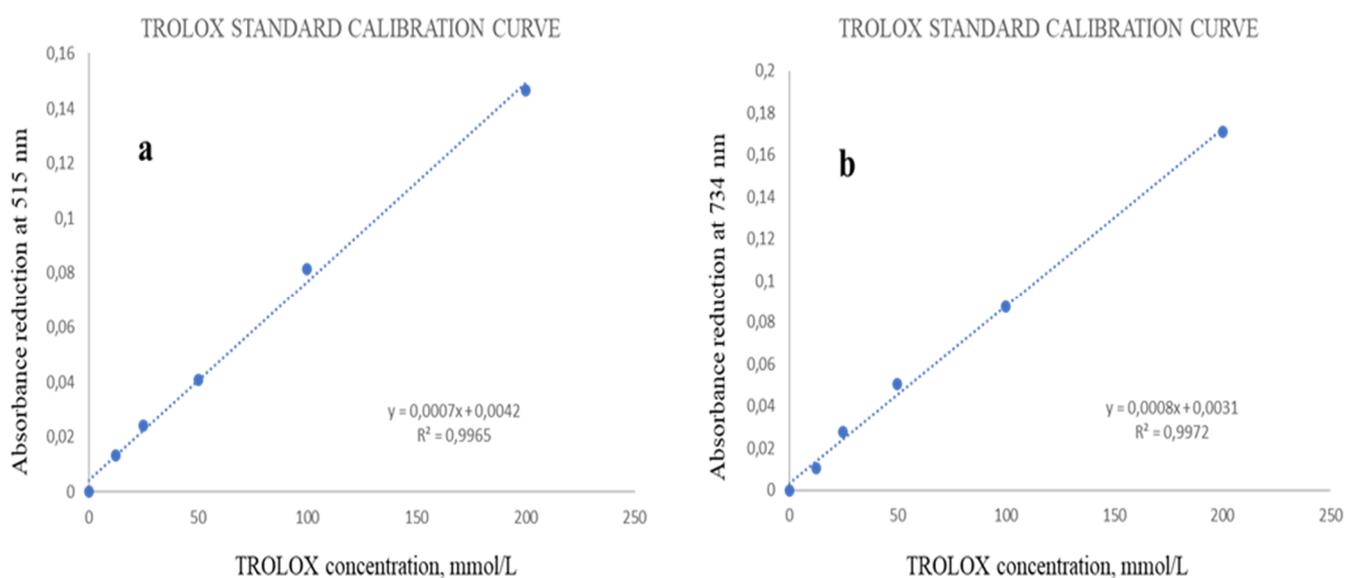
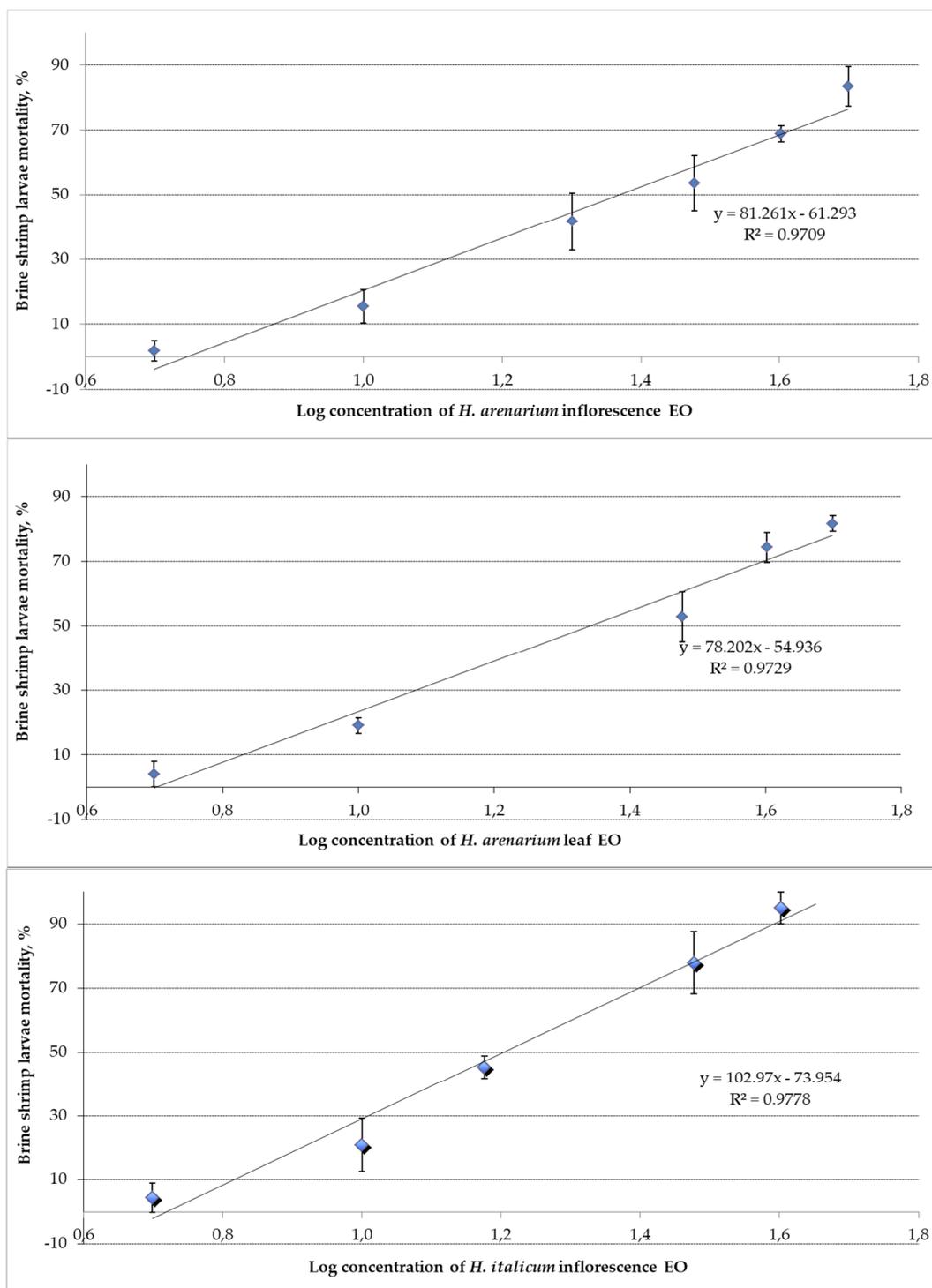


Figure S3. TROLOX standard calibration curves: (a) DPPH<sup>•</sup> assay; (b) ABTS<sup>•+</sup> assay.



**Figure S4.** Dependence of brine shrimp (*Artemia salina*) larvae lethality (%) on Log of essential oil concentration in saline water.