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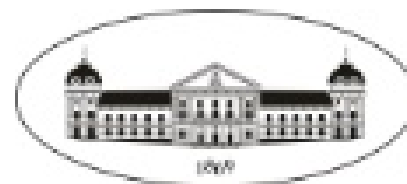
Exudate compounds of *Origanum* species

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Origanum species are valuable medicinal and culinary herbs, more that their biocidal properties are very important for organic farming. The first substances involved in allelopathic interactions in nature are the exudate (surface) compounds. In the present study, acetone exudates of ten samples of *Origanum* species were comparatively analyzed by GC/MS and TLC. Plant material of *Origanum dictamnus* L., *Origanum vulgare* L. and *Origanum vulgare* subsp. *hirtum* as the latter taxa was represented by 8 patterns with different origin were studied. Flavonoid aglycones, terpenes, fatty acids and alcohols, alkanes, triterpene acids and phenolic derivatives were identified. Methylated derivatives of flavones and non-methylated flavanones (naringenin and eriodictyol) were identified as the most common flavonoid aglycones. The most complex flavonoid profile was detected for *O. vulgare* ssp *hirtum* samples. A few differences in the flavonoid profiles of *O. vulgare* ssp *hirtum* from different origin of were found. Carvacrol was determined as main component of *O. vulgare* subsp. *hirtum* samples, whereas in *O. vulgaris* exudate long-chain fatty alcohol – hexacosanol was found as abundant compound. The data obtained complement the knowledge of the distribution and role of exudate compounds.

Keywords: *Origanum vulgare* subsp. *hirtum*; *Origanum dictamnus*; Greek oregano; flavonoid aglycones, carvacrol; long-chain fatty alcohols

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Results and Discussion

Ten exudates of *Origanum* samples listed at Table 1 were comparatively analyzed for determination of their main constituents.

Table 2. Identified flavonoid aglycones in the studied samples by TLC. Od *O. dictamnus*; Ov *O. vulgare*; Oh1- Oh8 *O. vulgare* subsp. *hirtum* (details Table 1) Me – methyl ether

Compounds	Od	Ov	Oh1	Oh2	Oh3	Oh4	Oh5	Oh6	Oh7	Oh8
Apigenin	•	•	•	•	•	•	•	•	•	•
Scutellarein 6,7-diMe	•			•			•		•	
Scutellarein 6,7,4'-triMe					•					
Scutellarein 6,7,8-triMe (Xantomicrol)	•						•	•		•
Luteolin	0	•	•	•	•	•	•	•	•	•
Naringenin			•	•	•	•	•	•	•	•
Eriodictyol		0	•	•	•	•	•	•	•	•

Table 3. Main identified compounds in the studied samples by GC/MS

Compounds	Od	Ov	Oh1	Oh2	Oh3	Oh4	Oh5	Oh6	Oh7	Oh8
Carvacrol	13.6	0.9	49.2	14.7	30.8	5.1	29.1	31.6	39.1	14.5
Caryophyllene	1.4	0.8	3.3		0.8			1.3		
Hydroquinone derivative	3.4		19.6	0.5	0.4	0.4	5.6	3.2	7.6	1.6
Caryophyllene oxide	2.7	0.4	0.4		0.8				0.3	0.1
Hexadecanoic acid		1.1		0.8	0.4	0.3	0.6	0.1		0.1
Hexacosanol	13.4	61.1	2.3	36.5	31.2	46.5	8.9	12.6	5.7	33.4
Triterpene	2.1	0.5	4.6	1.1	0.3	2.9	2.2	1.2	1.1	0.6
Ursolic acid	8.2	6.9	10.4		0.2	0.8	1.3	0.2		

Conclusions

In the present study metabolite profiles of exudates of ten samples on three taxa were determined. Monoterpene phenol (carvacrol), long-chain primary fatty alcohol (hexacosanol), ursolic acid, methylated flavones and non-methylated flavanones were determined as main bioactive compounds. These are substances with proved strong biocidal activity that suggests their protective role for plants.

The quantities are expressed in relative percentages (area %)

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Supplementary Materials

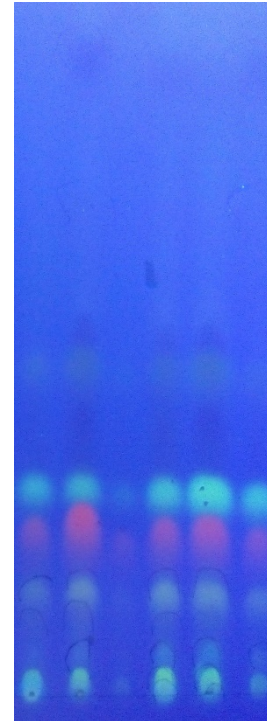
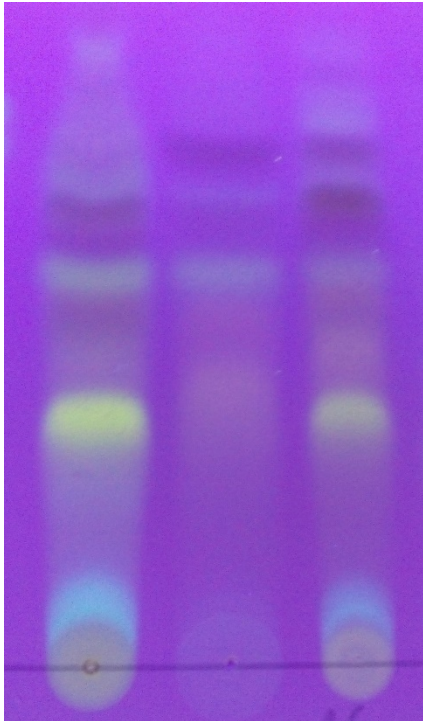
Table 1. Description of studied plant material

No	Taxon	Description of origin
Od	<i>O. dictamnus</i>	Plant collection Kazanlak, source material (seeds) purchased from seed plot https://zelena-prolet.com/
Ov	<i>O. vulgare</i>	Natural population, Trigrad, Bulgaria
Oh1	<i>O. vulgare</i> <i>subsp. hirtum</i>	Natural population, at the Struma valley Bulgaria
Oh2	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection IBER, source material (seeds) from natural population http://www.iber.bas.bg/sites/default/files/projects/plantscollection
Oh3	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, source material (seeds) purchased from Germany company https://www.pharmasaat.de
Oh4	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, source material from natural population, northern Greek
Oh5	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, Hebros variety
Oh6	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, candidate variety
Oh7	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, hybrid 1, seed progeny of <i>O. vulgare</i> subsp. <i>hirtum</i> obtained by free pollination of <i>O. vulgare</i> subsp. <i>hirtum</i> and <i>O. vulgare</i>
Oh8	<i>O. vulgare</i> <i>subsp. hirtum</i>	Plant collection Kazanlak, hybrid 2, seed progeny of <i>O. vulgare</i> subsp. <i>hirtum</i> obtained by free pollination of <i>O. vulgare</i> subsp. <i>hirtum</i> and <i>O. vulgare</i>

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