

COMPOSITION, CHEMOTYPES AND SUB-CHEMOTYPES OF ESSENTIAL OILS FROM *CORIANDRUM VULGARE* AND *CARUM CARVI* FRUITS CULTIVATED IN DIFFERENT COUNTRIES

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Coriandrum sativum L. and *Carum carvi* L. of the Apiaceae family are among the most cultivated plants, as they have been used for a long time as spices and essential oil (EO) bearing plants.

The aim. The aim from of this work is to examine the composition of EOs from commercial samples of *C. sativum* originating from 6 countries and *C. carvi* EOs from 2 countries, to establish the variability of the content of their components and to identify possible chemotypes of this species.

Materials and methods. The EOs were hydrodistilled from the dried fruits of *C. carvi* and *C. sativum*, and their chemical composition was determined using GC/MS. Samples were obtained from retail pharmacies in 6 different countries.

Research results. In the samples of coriander EOs, 50 compounds were detected. The dominant group of compounds is acyclic monoterpenoids, ranging from 67.3% (Turkey) to 84.2% (Czech Republic). The dominant component is linalool (61.6–77.9%). According to the content of the dominant major and minor components, it has been established for the first time that the studied samples of linalool-chemotypes can be divided into several sub-chemotypes. It has been noted for the first time that phenolic monoterpenoids were found in samples from subtropical and tropical countries. There is a strong negative correlation between the content of linalool and α -pinene (–0.891); linalool and γ -terpinene (–0.895). In the samples of *C. carvi* EOs, 28 compounds were detected. Both studied samples of caraway fruits contain the maximum amount of carvone (54.6–66.8%), followed by the content of limonene (19.9–30.1%). The EO of the caraway studied samples consists almost entirely of monocyclic monoterpenoids.

Conclusions. The results of our study of coriander fruits essential oil from six countries allowed us to establish its linalool chemotype, which is divided into five subtypes depending on the secondary compounds, that is novelty for research on possible chemotypes of coriander fruit essential oil. The studied samples of coriander fruits EO do not fully comply with the requirements of the ISO 3516:1997 standard; the content of linalool (Turkey) is slightly below the lower limit in accordance with the requirements. The studied samples of caraway EO slightly exceed the limits of the content of the dominant component carvone (Georgia) and contain significantly less limonene (India) in accordance with the requirements of the ISO 8896-2016 standard

Keywords: *Coriandrum sativum*, *Carum carvi*, essential oil, component composition, chemotypes, sub-chemotypes

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1. Introduction

Coriandrum sativum L. and *Carum carvi* L., belonging to the Apiaceae family, are well-known essential oil (EO) plants widely used in both medicine and culinary arts. Coriander, also known as cilantro or Chinese parsley, has been cultivated and used as a culinary herb for centuries. Its distinct flavour and aroma make it a popular ingredient in various cuisines around the world [1]. Over the years, coriander production has witnessed significant changes and trends. Coriander production is not evenly distributed across the globe. Some countries, such as India, China, Mexico, and Ukraine, have succeeded in growing coriander, securing their status as leading producers in the global market. These countries are consistently increasing their coriander pro-

duction in response to the growing global demand. The availability of suitable land, a favourable climate, and skilled farmers contribute to their success in cultivating coriander [2].

Linalool, the main component of coriander EO, has antioxidant, antitumor, neuroprotective, anxiolytic, anticonvulsant, migraine-relieving, analgesic, hypoglycemic, and hypolipidemic effects, as well as lowering blood pressure. *C. sativum* (Coriander) has been documented as a traditional treatment for cholesterol and diabetes patients [3].

C. carvi, or Caraway, also known as Meridian fennel and Persian cumin. Approximately 25 species of *Carum* are recognized, and only *C. carvi* has economic significance, as it is used and grown in several regions.

Caraway (*C. carvi*) is often confused with black caraway (*Carum bulbocastanum* Koch, *Bunicum persicum* Boiss) and Nigella (*Nigella sativa* L.) due to shared common names, but they are botanically different from each other. Caraway fruits are often used in bread, rolls, and baked goods, contributing to their demand in the bakery sector. Their inclusion adds flavour, texture, and visual appeal to baked products. Nowadays, it is cultivated in countries such as Jamaica, India, Canada, the United States and Australia. In India, this spice is known as Kashmiri jeera. Caraway Fruits Market size achieved USD 1.92 billion in 2023 and will grow at 6.8% CAGR (Compound Annual Growth Rate) from 2024 to 2032 [4].

It is known that the chemical composition of EO undergoes changes during ontogenesis, which affects the aroma of the plant, and thus, the coriander fruit aroma is completely different from the aroma of the herb [5–7].

The study of EOs extracted by hydrodistillation from the fruits and vegetative aerial parts of 11 samples of *C. sativum* grown in several regions of mainland Portugal. The EOs were analyzed using gas chromatography and gas chromatography-mass spectrometry. The analysis of coriander EOs revealed two main clusters, the first of which contains EOs from the fruits, where linalool (60–73%), γ -terpinene, and α -pinene are predominant, while the second contains EOs from the vegetative parts, where the main compounds are 2-*trans*-decenal (37–63%) and *n*-decanal (13–30%) [8].

The European Pharmacopoeia 11 regulates the content of EO in the fruits of *C. sativum* to be no less than 3 ml/kg, and the range of linalool content in the EO is 65–78% [9]. The quality of the coriander EO is regulated by the standard ISO 3516:1997 «Oil of coriander fruits (*Coriandrum sativum* L.)» [10]. According to this standard, the chromatographic profile of the EO is characterized by the presence of linalool (65–78%), camphor (4–6%), α -pinene (3–7%), γ -terpinene (2–7%), limonene (2–5%), geranyl acetate (1–3.5%), geraniol (0.5–3%), myrcene (0.5–1.5%), and α -terpineol (0.5–1.5%).

Coriander fruit EO is included among the major EOs in the world [11]. The major constituents of coriander fruit EOs are linalool (19.8–91.77%), α -pinene (0.09–10.7%), γ -terpinene (0.2–9%), geranyl acetate (1.44–8.59%), camphor (2.01–5.67%) terpinolene (0.14–5.85%) and geraniol (1.84–2.6%) [12–14]. Data analysis shows the content of linalool in EO from coriander fruits at levels of 68.9–83.7% in Argentina, 70.5% in Korea, 75.30% and 67.7% in India, 73.11% in Algeria, 65 to 70% and 69.60% in Pakistan. A lower level of linalool is observed in Cuba at 54.57%; in India at 52.26%, in Canada at 63.9–66.2%, in Bangladesh at 37.65%, in Brazil at 58.22%, in South Korea at 53.79%, and in Pakistan at 55.59% [15]. Coriander mainly belongs to the linalool chemotype, but it has several differences in the content of minor substances [16].

C. carvi fruits contain 1–9% EOs consisting of more than 30 compounds. The EO

compounds were included carvone, limonene, γ -terpinene, and as minor components α -pinene, camphene, β -pinene, β -myrcene, β -ocimene, *p*-cymene, terpinolene, limonene oxide, camphor, linalool, linalyl acetate, terpinene-4-ol, β -caryophyllene, dihydrocarvone, α -terpineol, germacrene-D, β -selinene, α -farnesene, citronellol, δ -cadinene, γ -cadinene, cuminaldehyde, nerol, *trans*-carveol, nonadecane, spathulenol, eugenol, thymol and carvacrol [17–23].

The fruits contain other compounds including acetaldehyde, cumuninic aldehyde, furfural, dihydrocarveol, thujone, anethole, linalool, carvenone, sabinene, perillyl alcohol, and phellandrene, as well as glycosides and flavonoids [24].

The quality of the caraway EO is regulated by the standard ISO 8896:2016 “Essential oil of caraway (*Carum carvi* L.)” [25]. According to this standard, the chromatographic profile of the EO is characterized by the presence of carvone (50.0–63.0%), limonene (33.0–45.0%), *cis*-dihydrocarvone (0.1–1.5%), myrcene (0.2–0.7%), *cis*-carveol (0.2–0.5%), and *trans*-carveol (traces – 0.5%).

In our previously published works, the chemical composition of the EO of commercial samples of coriander fruits and caraway fruits from various countries of origin was established [26]. It has been established that the major constituent in the coriander fruits’ EOs was linalool (58.0–80.3%). The other characteristic compounds present in the EOs were γ -terpinene (0.3–11.2%), α -pinene (0.2–10.9%), *p*-cymene (0.1–8.1%), camphor (3.0–5.1%) and geranyl acetate (0.2–5.4%) [27]. The main components of the EOs in caraway fruits were carvone (44.5–95.9%) and limonene (1.5–51.3%) [26]. This work is a continuation of previously initiated research.

The purpose of this work is to examine the composition of EO samples of *C. sativum* originated from 6 countries that have not been previously studied, namely Austria, India, Georgia, Germany, Czech Republic and Turkey and *C. carvi* EO from 2 countries, namely Georgia and India, to establish the variability of the content of their components and to identify possible chemotypes of this species.

2. Planning (methodology) of research

The study protocol describing the different stages of the present research work is presented in the following flow chart (Fig. 1).

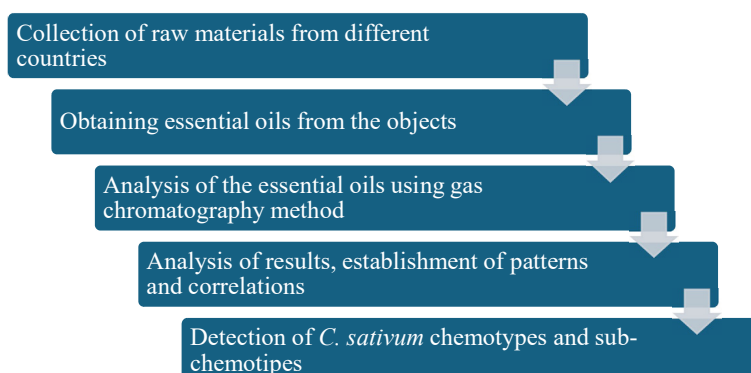


Fig. 1. Study protocol

3. Materials and methods

Materials. Commercial coriander fruits (6 samples) and caraway fruits (2 samples) were used. The commercial plant material was obtained from retail pharmacies or health (herb) shops in different countries, such as Austria, India (cultivated in India, packed in England), Georgia, Germany, the Czech Republic and Turkey. The commercial samples were kept in their original packages.

Hydrodistillation of EO. The EOs were hydrodistilled from the dried fruits of *C. carvi* and *C. sativum* using the method described in the European Pharmacopoeia [9, 26]. The air-dried plant fruits of (10 g of *C. carvi* and 30 g of *C. sativum*) with 200 mL of purified water were hydrodistilled in a 1000 mL round-bottom flask for 1.5 and 2 hours, respectively (2–3 mL/min). Hexane (0.5 mL) was added to a graduated tube to remove the distilled EO.

GC-MS analysis of EO. GC analysis used a Chrom 5 chromatograph with FID on two fused silica capillary columns with bonded stationary phases SPB-5 (30 m × 0.25 mm, Supelco) and SW-10 (30 m × 0.25 mm,

Supelco). The film thickness of both stationary phases was 0.25 µm. Carrier gas was He, and the flow rate of 30–35 cm/s was applied. Split ratio 1 : 150. The temperature program was 50–250°C at 2 °C/min, and the injector temperature was 200 °C. A Hewlett-Packard Model 3390A integrator was used for data processing.

The EO components were identified by comparing their retention indices (RI) on two columns with the RI values of reference standards, our RI data bank and the literature data [27–29]. GC/MS confirmed the results obtained. The percentage composition of the EO was calculated in peak areas (nonpolar column) using the normalization method without correction factors. The relative standard deviation of percentages of EO components of three repeated GC analyses of a single EO sample didn't exceed 5% [29–31].

4. Results

The identified compounds in the fruits EOs of the 6 coriander and 2 caraway samples from different countries are presented in Table 1 and Table 2, respectively.

Table 1

Composition of the essential oil of *Coriandrum sativum* L. fruits

Compound	RI		Content, % in Coriander fruits						Mean, %, <i>n</i> = 6	Variation coefficient
	SPB-5	SW-10	Austria	India*	Georgia	Germany	Czech Republic	Turkey		
1	2	3	4	5	6	7	8	9	10	11
α-Thujene	923	1024	0.1	tr	0.3	nd	nd	0.1	0.1	1.22
α-Pinene	928	1022	7.1	7.2	6.3	1.2	nd	10.1	5.32	0.73
Camphene	942	1065	0.9	tr	1.0	0.2	nd	0.6	0.45	0.99
Sabinene	968	1110	0.4	0.1	0.4	0.1	nd	0.5	0.25	0.83
β-Pinene	970	1102	0.6	0.8	0.4	0.1	nd	0.9	0.47	0.79
Myrcene	989	1162	1.0	0.4	1.0	0.3	tr	1.2	0.65	0.74
α-Phellandrene	1002	1164	nd	0.1	tr	nd	nd	0.4	0.08	1.92
3-Carene	1007	1135	nd	nd	nd	nd	nd	0.6	0.10	2.45
α-Terpinene	1012	1175	0.1	tr	0.1	0.1	nd	0.1	0.07	0.77
<i>p</i> -Cymene	1020	1265	2.1	tr	0.3	1.4	0.2	2.8	1.13	1.02
Limonene	1024	1200	2.3	0.2	2.9	0.9	0.1	1.8	1.37	0.84
1,8-Cineole	1027	1209	0.1	0.2	2.3	0.1	0.1	0.3	0.52	1.70
(Z)-β-Ocimene	1036	1235	nd	nd	nd	nd	nd	0.1	0.02	2.45
γ-Terpinene	1056	1243	7.0	0.5	5.3	1.9	0.6	6.2	3.58	0.82
(Z)-Linalool oxide	1066	1425	0.1	tr	0.1	0.1	0.1	nd	0.07	0.77
(E)-Linalool oxide	1079	1455	0.5	0.7	0.4	0.2	0.1	nd	0.32	0.83
Terpinolene	1086	1282	tr	nd	tr	0.1	nd	0.3	0.07	1.82
Linalool	1100	1545	65.6	69.1	67.5	74.5	77.9	61.6	69.37	0.09
Camphor	1138	1502	3.4	0.3	4.4	3.1	3.5	1.2	2.65	0.59
Citronellal	1148	1480	tr	nd	nd	0.1	0.1	0.1	0.05	1.10
Camphene hydrate	1150	1611	nd	0.1	tr	nd	nd	nd	0.02	2.45
Isoborneol	1160		0.1	nd	nd	nd	nd	nd	0.02	2.45
Borneol	1161	1702	tr	0.1	0.1	1.7	nd	nd	0.32	2.15
Terpinen-4-ol	1173	1600	0.2	tr	0.2	1.2	0.3	0.1	0.33	1.31
α-Terpineol	1187	1700	0.2	0.1	0.3	0.4	0.4	0.2	0.27	0.45
<i>p</i> -Cymen-8-ol	1194	1860	nd	nd	nd	nd	nd	0.1	0.02	2.45
Myrtenol decanal	1200		0.1	nd	nd	0.5	0.3	nd	0.15	1.38
Neral	1232	1663	nd	nd	nd	nd	0.2	0.2	0.07	1.55
β-Citronellol	1238	1767	nd	1.3	nd	nd	nd	nd	0.22	2.45
Carvone	1238	1725	1.3	nd	0.2	0.4	1.6	0.9	0.73	0.87
Geraniol	1256	1854	1.9	0.4	2.1	3.6	2.7	nd	1.78	0.77
Linalyl acetate	1258	1550	nd	0.3	nd	nd	nd	nd	0.05	2.45
Geranial	1266	1735	tr	nd	0.1	0.1	nd	nd	0.03	1.55

Continuation of Table 1

1	2	3	4	5	6	7	8	9	10	11
Perill aldehyde	1269	1786	nd	0.8	nd	nd	nd	nd	0.13	2.45
Bornyl acetate	1281	1531	0.4	nd	nd	nd	0.6	nd	0.17	1.59
(E)-Anetole	1281	1850	nd	1.7	0.1	nd	nd	1.7	0.58	1.48
Thymol	1293	2192	nd	0.2	tr	nd	nd	0.3	0.08	1.59
Carvacrol	1306	2215	nd	0.2	0.1	nd	nd	1.3	0.27	1.92
α -Terpinyl acetate	1353	1690	nd	nd	nd	nd	nd	0.1	0.02	2.45
Neryl acetate	1360	1721	nd	0.3	nd	nd	nd	nd	0.05	2.45
Geranyl acetate	1379	1756	3.2	10.8	2.3	1.9	3.2	4.1	4.25	0.78
Isocaryophyllene	1406	1588	nd	nd	nd	0.1	0.1	nd	0.03	1.55
(E)- β -Caryophyllene	1411	1582	0.1	nd	tr	nd	nd	0.2	0.05	1.67
β -Farnesene	1462	1663	nd	0.5	nd	nd	nd	nd	0.08	2.45
Geranyl acetone	1463	1853	nd	nd	nd	nd	nd	0.4	0.07	2.45
α -Farnesol	1468	2275	nd	0.3	nd	nd	nd	nd	0.05	2.45
(E)- α -Farnesene	1500	1742	nd	nd	nd	nd	nd	0.3	0.05	2.45
α -Cadinol	1652	2221	nd	nd	nd	nd	nd	0.1	0.02	2.45
Myristic acid	1775	2700	nd	0.4	nd	0.1	3.3	nd	0.63	2.08
Palmitic acid	1970	2020	nd	0.3	nd	1.1	0.4	nd	0.30	1.43
Component groups										
Monoterpenoids acyclic			72.3	83.3	73.5	80.8	84.2	67.3	–	–
Monoterpenoids monocyclic			13.3	1.9	11.6	6.5	3.4	13.3	–	–
Monoterpenoids bicyclic			13.1	8.6	13.9	6.9	0	13.9	–	–
Sesquiterpenoids acyclic			0	0.8	0	0	0	0.3	–	–
Sesquiterpenoids bicyclic			0.1	0.8	0	0.1	0.1	0.1	–	–
Monoterpenoids phenolic			0	2.1	0.2	0	0	3.3	–	–
Norsesquiterpenoids			0	0	0	0	0	0.4	–	–
Fatty acids			0	0.7	0	1.2	3.7	0	–	–
Total			98.8	98.2	99.2	95.5	91.4	98.6	–	–

Note: tr – traces (<0.05%); nd – not detected; * – cultivated in India, packed in England.

Table 2

Composition of the essential oil of *Carum carvi* L. fruits

Compound	RI		Content, % in Carvi fruits		Concentration range, % [27]
	SPB-5	SW-10	Georgia	India*	
1	2	3	4	5	6
α -Pinene	930	1020	tr	0.3	0–0.3
Sabinene	968	1112	tr	tr	0–0.3
1-Okten-3-ol	973	1450	nd	6.2	0–6.2
Myrcene	989	1164	0.3	0.1	0–0.4
α -Terpinene	1012	1177	nd	0.2	0–0.2
p-Cymene	1019	1262	nd	0.4	0–0.4
Limonene	1022	1200	30.1	19.9	1.5–51.3
1,8-Cineole	1025	1209	nd	0.4	0–0.4
(E)- β -Ocimene	1047	1255	tr	nd	0–0.1
γ -Terpinene	1053	1243	0.1	0.2	0–0.1
Terpinolene	1084	1272	nd	0.2	0–0.2
Linalool	1100	1545	0.2	0.1	0–0.2
(Z)-Limonene oxide	1128	1445	0.1	0.5	0–0.5
(E)-Limonene oxide	1132	1460	0.2	nd	tr–0.3
Pinocarvone	1162	1540	nd	0.7	0–0.7
(Z)-Dihydrocarvone	1187	1600–1610	tr	0.4	0–0.5
(E)-Dihydrocarvone	1194	1600	0.3	0.4	0–0.4
(E)-Carveol	1200	1804	0.4	0.3	0–0.4
(Z)-Carveol	1230	1835	0.1	nd	0–0.2
Carvone	1235	1730	66.8	54.6	44.5–95.9
Perill aldehyde	1272	1785	0.3	0.2	0.1–0.4
(E)-Anetole	1281	1824	0.1	0.4	0–2.2
Carvone oxide	1288	1741	nd	5.4	0–5.4
Dihydrocarvenyl acetate I	1300	1583	nd	0.4	0–0.4

Continuation of Table 2

1	2	3	4	5	6
Dihydrocarvenyl acetate II	1312	1589	nd	0.5	0–0.5
(E)-Carvenyl acetate	1371	1756	nd	0.9	0–0.9
β -Bourbonen	1384	1500	nd	1.4	0–1.4
(E)- β -Caryophyllene	1410	1581	0.1	0.3	0–0.3
Component groups					
Monoterpenoids acyclic			0.5	0.2	–
Monoterpenoids monocyclic			98.4	84.9	–
Monoterpenoids bicyclic			0.0	1.0	–
Sesquiterpenoids bicyclic			0.1	0.3	–
Sesquiterpenoids tricyclic			0.0	1.4	–
Monoterpenoids phenolic			0.1	0.4	–
Aliphatic alcohols			0	6.2	–
Total			99.1	94.4	–

Note: tr – traces (<0.05%); nd – not detected; * – cultivated in India, packed in England.

5. Discussion

In the samples of coriander EO studied, 50 compounds were detected. High coefficients of variation for the majority of compounds (> 1), which indicate that the content of these compounds varies significantly from sample to sample, were observed for 34 out of 50 identified substances. Low variation coefficients (0.56–0.75) could be seen for α -pinene, myrcene, and camphor. Trace amounts (< 0.05%) were detected in one sample of α -thujene, camphene, myrcene, α -phellandrene, α -terpinene, *p*-cymene, (Z)-linalool oxide, citronellal, camphene hydrate, borneol, terpinen-4-ol, geranial, and (E)- β -caryophyllene, in two samples of terpinolene.

In all the examined samples of coriander fruits, the dominant component is linalool (61.6–77.9%). The smallest value of the coefficient of variation for linalool (0.09) indicates the stability of its content in coriander EO of different origins, which is also confirmed by previous studies [30].

Taking into account the present results and the results of our previous studies, we found a strong positive correlation (more than 70%) between the content of α -pinene and sabinene (0.702); α -pinene and β -pinene (0.868); α -pinene and γ -terpinene (0.712). There is a strong negative correlation between the content of linalool and α -pinene (–0.891); linalool and γ -terpinene (–0.895). The content of linalool and β -pinene correlates at a fairly high level of (–0.673).

All coriander EOs produced in different countries belong to the linalool chemotype: the linalool content exceeds 50%. However, this coriander chemotype has several differences in the content of minor substances, which led to the identification of five coriander sub-chemotypes.

According to the content of dominant major and minor components, the studied linalool chemotype samples can be divided into several sub-chemotypes (sub-Ct), that is novelty for coriander fruit essential oil:

- sub-Ct-1 – linalool > camphor, samples from Germany and Czech Republic;
- sub-Ct-2 – linalool > α -pinene \approx γ -terpinene > camphor, sample from Austria;

– sub-Ct-3 – linalool > α -pinene > γ -terpinene > geranyl acetate > sum monoterpenoids phenolic, sample from Turkey;

– sub-Ct-4 – linalool > geranyl acetate > α -pinene, sample from India;

– sub-Ct-5 – linalool > α -pinene > γ -terpinene > camphor, sample from Georgia.

In all studied samples, the dominant group of compounds is acyclic monoterpenoids (Table 1, Fig. 2). Their content ranges from 67.3% (Turkey) to 84.2% (Czech Republic). It should be noted that the sample from the Czech Republic differs from others in the absence of bicyclic monoterpenoids. Acyclic sesquiterpenoids are found only in samples from India and Turkey. It should be noted that phenolic monoterpenes are found in samples from subtropical and tropical countries such as India, Georgia, and Turkey, which has been established for the first time. The norsesquiterpenoid geranylacetone was found only in the EO of a sample from Turkey.

For the first time, the composition of coriander fruit essential oil from various countries such as Austria, India (grown in India, packaged in England), Georgia, Germany, the Czech Republic, and Turkey was analyzed for compliance with the requirements of ISO 3516:1997: Essential oil of coriander fruit (*Coriandrum sativum* L.). It was found that the tested samples of coriander fruit essential oil did not fully comply with the requirements of the standard. For instance, in the sample from Turkey, the content of linalool (61.6%) is below the lower limit set by ISO (65–78% according to the standard's requirements). The content of camphor meets the ISO requirements (4–6%) only in the sample from Georgia (4.4%). The regulation of α -pinene content (3–7%) is met only by the sample from Georgia (6.3%), while in the samples from Austria and India, its content is slightly above the upper limit (7.1% and 7.2%, respectively). In the sample from Turkey, the content is significantly above the upper limit (10.1%), in the sample from Germany it is much lower than the lower limit (1.2%), and in the sample from the Czech Republic, it is completely absent. The content regulation of γ -terpinene (2–7%) does not comply with the samples from India (0.5%) and the Czech Republic.

lic (0.6%). The content of limonene (2–5%) meets the requirements only in the sample from Georgia (2.3%). The content of geranyl acetate (1–3.5%) significantly exceeds the upper limit in the samples from Turkey (4.1%) and India (10.8%). The content of geraniol (0.5–3%) is slightly below the lower limit of requirements in the sample from India (0.4%) and somewhat above the upper limit in the sample from Germany (3.6%), while in the sample from Turkey it is completely absent. The content of myrcene (0.5–1.5%) meets the requirements only in the EOs from Austria (1.0%), Georgia (1.0%), and Turkey (1.2%). The content of α -terpineol (0.5–1.5%) does not comply with ISO 3516:1997 in any of the studied samples.

The results we obtained regarding the content of linalool in the coriander EO sample from Turkey (61.6%) are significantly lower than those indicated in the published work of Turkish researchers (89.44–91.77%); the content of α -pinene and geranyl acetate in our study is significantly higher, at 10.1% and 4.1% respectively, while according to the literature data, they are 3.06% and 2.77% [32].

Similarly, in the sample from India, the content of linalool in our study is 69.1%, whereas according to other researchers it is 73.2–85.3%. Conversely, based on our data, the content of α -pinene is 7.2% compared to 2.81% according to the literature, and the content of geranyl acetate in our data is 10.8%, while it is absent in a previously published work [33].

In the samples of caraway EO studied, 28 compounds were detected.

Both studied samples of caraway fruits contain the maximum amount of carvone, followed by the content of limonene. In the sample from India, it is also important to note the significant content of carvone oxide (5.4%).

Trace amounts (< 0.05%) were detected in one sample of α -pinene, (*E*)- β -ocimene, and (*Z*)-dihydrocarvone, in two samples of sabinene.

The overall quality of fruits is considered to correlate with the content of EO and its carvone/limonene (C/L) ratio: the higher the ratio, the better the quality [34]. As can be seen from our results, the C/L ratio was 2.22 for the sample from Georgia and 2.74 for the sample from India. Therefore, it can be concluded that the *C. carvi* tested in our study belongs to the carvone chemotype, which is consistent with the data from other researchers [35].

The EO of the studied samples is almost entirely composed of monocyclic monoterpenoids. Their content in the sample from Georgia is 98.4%, while in the sample from India it is 84.9%.

For the first time, the composition of the essential oil from caraway fruit from Georgia and India (cultivated in India, packed in England) for compliance with the requirements of ISO 8896:2016(E) “Essential oil of caraway (*Carum carvi* L.)”. It was found that the tested samples of caraway fruit essential oil did not fully comply with the requirements of the standard. The content of carvone (50.0–63.0% according to the standard requirements) in the sample from Georgia is 66.8%, which exceeds the upper limit of the standard. The content of limonene (33.0–45.0%) in both studied samples is below the standard requirements. The content of (*Z*)-dihydrocarvone (0.1–1.5%) and (*E*)-carveol (*cis*-carveol) (0.2–0.5%) complies with the regulations. The content of myrcene (0.2–0.7%) is lower than the norm in the EO sample from India (0.1%). The content of (*Z*)-carveol (*trans*-carveol) (traces – 0.5%) was not detected in the sample from India. The phenolic terpenoids in both samples are represented by anethole.

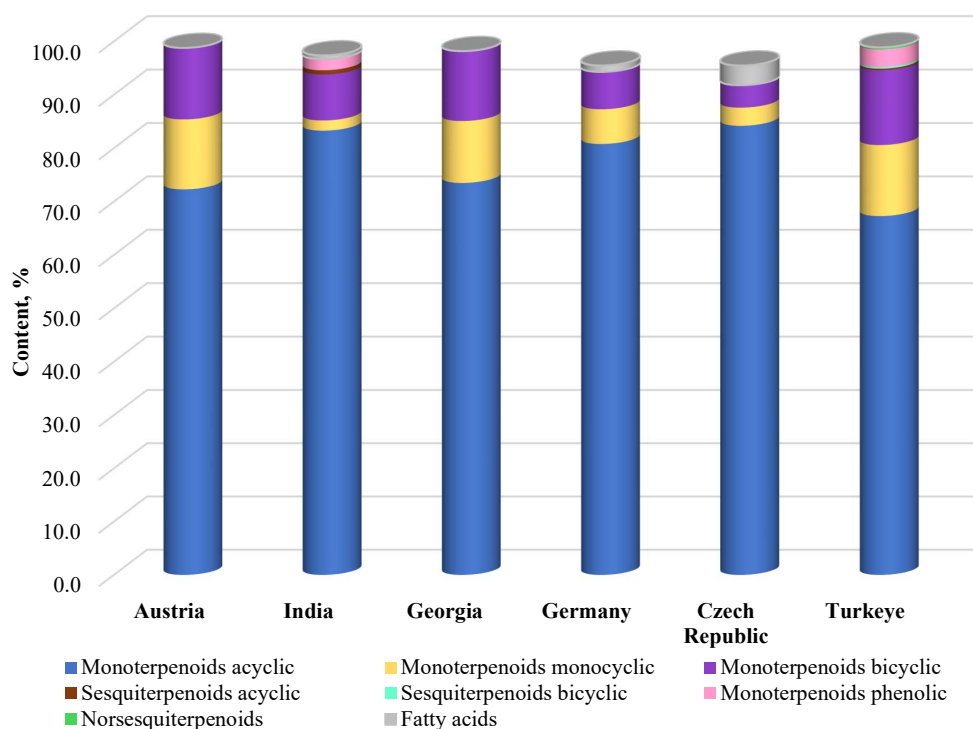


Fig. 2. The content (%) groups of substances in *C. sativum* essential oils

Experimental studies have revealed antibacterial, antiulcer, antioxidant, hepatoprotective, anticonvulsant, analgesic, diuretic, hepatoprotective, antihyperlipidemic and antitumor activity of terpenoids contained in caraway EO [36].

Practical relevance. The established chemotypes and sub-chemotypes allow for a more precise classification and sorting of the raw materials and essential oils of coriander (*Coriandrum sativum* L.) and caraway (*Carum carvi* L.), predicting the pharmacological activity of its medicinal products and ensuring the possibility of targeted search for the necessary raw materials according to the specified parameters. The correlation of substance content allows for the prediction of target molecules in the essential oils of the studied species.

Study limitations. The samples of coriander and cumin fruits used were mainly from European countries. It would be advisable to expand the geography of the raw material origin.

The prospects for further research. The prospects for further research include expanding the dataset of new chemotypes across various countries worldwide and creating conditions for the targeted search for biologically active substances and fruit materials.

6. Conclusion

The results of our study of coriander fruit essential oil from six countries allowed us to establish its linalool chemotype, which is divided into five subtypes depending on the secondary compounds, that is novelty for research on possible chemotypes of coriander fruit essential oil. Acyclic sesquiterpenoids are found only in samples from India and Turkey. It has been noted for the first time that phenolic monoterpenoids were found in samples from subtropical and tropical countries, such as India, Georgia and Turkey. The nor-sesquiterpenoid geranylacetone was found only in the EO of a sample from Turkey. There is a strong negative correlation between the content of linalool and α -pinene (-0.891); linalool and γ -terpinene (-0.895). The studied samples of coriander fruits EO do not fully comply with the requirements of the ISO 3516:1997 standard; the content of linalool (Turkey) is slightly below the lower limit in accordance with the requirements.

The EO of the caraway studied samples consists almost entirely of monocyclic monoterpenoids. The studied samples of caraway EO slightly exceed the limits of the content of the dominant component carvone (Georgia) and contain significantly less limonene (India) in accordance with the requirements of the ISO 8896-2016 standard.

Conflicts of interest

The authors declare that they have no conflict of interest concerning this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Data availability

The datasets used and/or analyzed during the current study are available from the author and/or corresponding author upon reasonable request.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies when creating the current work.

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Authors' contributions

Ain Raal: conception and design, acquisition and data, analysis and interpretation of data, drafting of the manuscript, critical revision of the manuscript for important intellectual content, obtaining funding, administrative, technical or material support, supervision; **Tetiana Ilina:** conception and design, acquisition and data, analysis and interpretation of data, drafting of the manuscript, critical revision of the manuscript for important intellectual content; **Alla Kovalyova:** conception and design, analysis and interpretation of data, drafting of the manuscript, critical revision of the manuscript for important intellectual content; **Anne Orav:** conception and design, acquisition and data, analysis and interpretation of data; **Yuliia Avidzba:** acquisition and data, analysis and interpretation of data, statistical analysis; **Oleksandr Panasenکو:** analysis and interpretation of data, critical revision of the manuscript for important intellectual content, statistical analysis; **Oleh Koshovyi:** conception and design, acquisition and data, analysis and interpretation of data, drafting of the manuscript, critical revision of the manuscript for important intellectual content, statistical analysis.

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