Chapter 44

Curry Plant (*Helichrysum* sp.) Oils

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INTRODUCTION

Madagascar's flora is one of the richest in the world and has many medicinal and aromatic plants. The essential oils that are extracted are products with high added value that could contribute to economic island development. Among these aromatic and medicinal plants, we will focus on the genus *Helichrysum*. Varietal differences occur worldwide, with more than 600 species (Bigovic et al., 2010).

The genus *Helichrysum* (from the Greek *helios*, meaning sun, and *chrysos*, meaning gold) belongs to the Asteraceae family which incudes many species rich in essential oils and aromatic compounds. The Asteraceae family, also known as the Compositae family, includes 1600–1700 genera and 24,000–30,000 species (Funk et al., 2005). All species are well distinguished by the arrangement of the florets and the fruit. Plants belonging to this family may appear on different forms, as it includes herbs, succulents, lianas, epiphytes, trees, or shrubs. They are located everywhere, except Antarctica (Funk et al., 2005).

The uses of several members of the Asteraceae family are in medicine, as ornaments, and for trade. Among species we can cite those of commercial impact as the food crops *Lactuca sativa* (lettuce), *Cichorium intybus* (chicory), *Cynarascolymus* (globe artichoke), *Smallanthus sonchifolius* (yacon), and *Helianthus tuberosus* (Jerusalem artichoke).

Seeds of some species are used for the production of cooking oil such as the seeds of *Helianthus annuus* (sunflower) and *Carthamus tinctorius* (safflower) (Gao et al., 2010). Figure 1 illustrates some flowering plants which belong to the Asteraceae family.

Several researchers have focused on the chemical composition of the essential oil of some species belonging to this family. These volatile compounds may be applied in edible, medicinal, and herbal plants which make them safe when used in food products. Essential oils and their constituents have been largely employed as flavoring agents in foods and much of them exhibited a large antimicrobial activity (Alzoreky and Nakahara, 2002).

Raala et al. (2011) studied the content and composition of the essential oils of five Asteraceae species from Estonia, namely *Chamomilla recutita*, *Chamomilla suaveolens*, *Matricaria perforate*, *Anthemistinctoria*, and *Leucanthemum vulgare*. The oil yields ranged from trace amounts up to 0.2%. The chemical composition was established using gas chromatography (GC)–flame ionization detector and GC–mass spectrometry (MS) methods. The identified compounds varied significantly from one species to another. The main constituents of the essential oils of the studied Asteraceae species were as follows: *C. recutita*: bisabolol oxide A (39.4%), bisabolone oxide A (13.9%), (Z)-En-yne-dicycloether (11.5%), bisabolol oxide B (9.9%), α-bisabolol (5.6%), and chamazulene (4.7%); *C. suaveolens*: (Z)-En-yne-dicycloether (37.2%), geranylisovalerate (22.9%), (E)-β-farnesene (15.6%); *Anthemistinctoria*: α-eudesmol (10.2%), γ-cadinol (8.7%), γ-cadinene (4.0%); *M. perforata*: (Z,Z)-matricaria ester (77.9%), (E)-β-farnesene (3.5%), matricaria ester isomer (3.5%), and matricaria lactone (3.0%); and *L. vulgare*: (E)-β-farnesene (7.3%), hexadecahydrocyclobuta[1,2:3,4]dicyclooctene (5.3%), decanoic acid (4.9%), and γ-eudesmol (4.5%).

BOTANICAL ASPECTS

The genus *Helichrysum* (Asteraceae family) is abundantly prevalent throughout the world (Europe, Africa, Australia, North America) and is represented by more than 600 species. One hundred and fifteen of them are located in Madagascar; most are endemic, and some are used in folk medicine. Essential oils and extracts are obtained from the whole plant or from several parts of the plant.

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FIGURE 1 Flowering plants (Asteraceae family) (Funk et al., 2005).

This chapter will study of essential oils of seven Malagasy species, namely *Helichrysum bracteiferum* (DC) H. Humbert, *Helichrysum gymnocephalum* (DC), H. Humbert, *Helichrysum selaginifolium* Vig. Humboldt, *Helichrysum hypnoides* (DC) Vig. Humboldt, *Helichrysum cordifolium* (DC), and *Helichrysum faradifani* Sc Ell. We will start with a botanical description of each one, then focus on the chemical composition of their essential oils. The botanical description and embranchment is summarized in Table 1. It is important to note that both species *H. bracteiferum* and *H. gymnocephalum* are often confused because of their great morphological similarity. They both share the same common name Rambiazina, but they can be differentiated according to the size of their leaves. The first, with small leaves is classified as the male and called *lahy*, while the second, with broader leaves, is classified as the female and called *vavy* (Rasoanaivo and De La Gorce, 1998).

TABLE 1 Botanical Description of *Helichrysum gymnocephalum*

The Reign	Plant
Class	Dicotyledons
Sub-class	Asteridae
Order	Asterales
Family	Asteraceae
Genus	*Helichrysum*
Species	*gymnocephalum*

***Helichrysum bracteiferum* (DC) H. Humbert**, vernacular name is Rambiazina, is a shrub of 1–3 m high, with ultimate branches covered with dense brown tawny tomentum formed with small fine hairs. Leaves are sessile, attenuate at the base, and small (20–30 × 4–5 mm). Flowers are among two or three, all contained in flower heads, and are all homogamous. The fruits, called Achenes, are papillose (Humbert, 1962). This species is found in several places of Madagascar, particularly the siliceous rock in the middle of the ericoid vegetation peaks in the massif of Tsaratanana (North), the massif of Andrangovalo (East Lake Alaotra), or the massif of Ankaratra (south of Antananarivo).

***Helichrysum selaginifolium* Vig. and Humboldt** is a branching shrub ericoid,1–1.5 m in height, or it is a suffrutescent plant of 10–60 cm in places subject to periodic fires. It has very slender twigs (ultimate branches of 1–1.5 mm in diameter), prepared, and tomentose. The leaves are narrowly deltoid-acuminate (long May to September, mm wide and 0.5 mm at the base). Heterogamous flower heads are very small and grouped in globular clusters of 10–20, many of which are female. Flowering lasts from August to October. The achenes are generally papillose.

***Helichrysum hypnoides* (DC) R. Vig. and H. Humboldt** is an extremely branching ericoid shrub, 10–80 cm tall, with thin but highly lignified stems (brittle) and covered with a fine tomentum greyish white cobweb blanc. The leaves are sessile, lanceolates, and very small (3–5 mm long and 0.5–1.5 mm wide at the base). Heterogamous flower heads are all or most solitary, or two or three on top of twigs or partially close in small terminal clusters of three to 15 and are arranged in heads (several are female). The petals are red or brownish in color, especially at the top. Flowering lasts from March to October. The achenes are papillose.

***Helichrysum faradifani* Sc. Ell.**, vernacular name *ahibalala* (grasshoppers's grass), is a shrub measuring up to 60 cm in height and is abundant on the whole island of Madagascar. It is particularly present in rocky or sandy places. This species is used in traditional medicine. Flowering lasts all year.

***Helichrysum cordifolium* DC**, common name Fotsiavadika Tsimanandra, is a very rower suffrutescent plant (0.5–1.2 m high and sometimes up to 2–3 m, supported on the bushes). It has juvenile branches covered with loose tomentum. There are about 15 flowers, one to five of which are female. Flowering lasts all year following localities. It is found in forest edges, in ravines, etc. It is located in the center of the island and there are also a few specimens in the Comoros (probably introduced as a medicinal plant).

We will next focus on the botanical description of four species growing in Greece, namely *Helichrysum orientale, Helichrysum heldreichii, Helichrysum italicum* subsp. Microphyllum, and *Helichrysum doeryeri.*

- ***Helichrysum orientale* (L.)** is a non aromatic species with flat leaves; 20–60 mm at base and 7–10 mm in diameter.
- ***Helichrysum heldreichii* Boiss** is rare and endemic to West Crete. This species grows on vertical cliffs. The leaves are linear and long (more than 30 mm).
- ***Helichrysum italicum* (Roth) G. Don fil** is a Mediterranean species. It is a shrub with yellow flowers, with a height of 50–70 cm. This species grows on dry cliffs and sandy soil. *H. italicum* consists of two subspecies: subsp. *microphyllum* (Willd.) Nyman (Sardinia, south of Corsica), and subsp. *Italicum* (Tuscany, Corsica). Some phenotypic differences occur between the subspecies, for example, the height, ramification type, and size of the leaves (Bianchini et al., 2003).
- ***Helichrysum italicum* (Roth) G. Don in Loudon ssp. *microphyllum* (Willd.) Nyman** is an aromatic shrub. The leaves are very small (10 mm in length) with revolute margins. This species grows on cliffs and in rocky places of the islands in the Mediterranean region (Roussis et al., 2000). This species is also located in Tuscan Archipelago Islands, in Italy.
- ***Helichrysum doeryeri* Rech. Fil** is rare and endemic to east Crete. This species grows on mountain cliffs. It is a perennial plant characterized by very short stems and lower leaves. The capitulars are usually large with white outer bracts and form small bundles. Roussis et al. (2000) published the first analysis of essential oils in the Greek *Helichrysum* species which is described in the section related to the chemical composition of *Helichrysum* essential oils (see Usage and Applications).

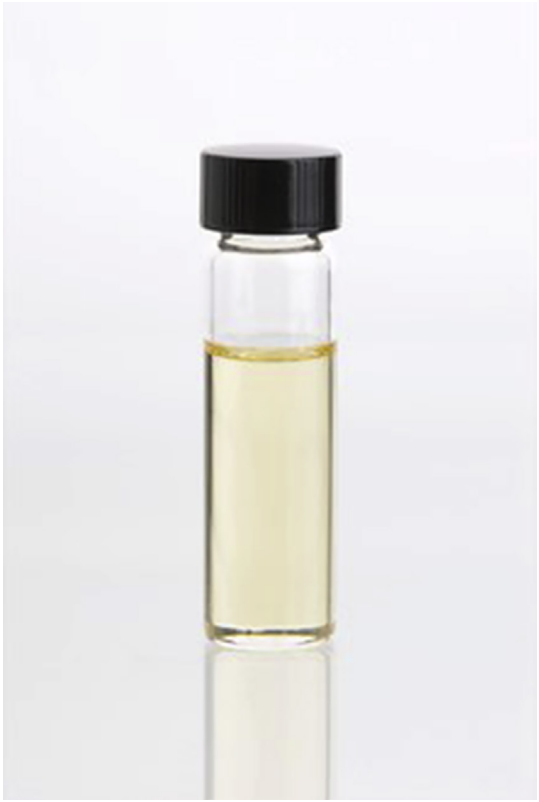

FIGURE 2 *Helichrysum italicum* essential oil in a clear glass vial.

In addition to Madagascar, Greece, and Italy, other *Helichrysum* species have been found, such as ***Helichrysum cymosum* (L.)**, an aromatic perennial herb with yellow flowers and characteristic odors, widely distributed in southern tropical Africa. This species is used in traditional medicine to treat respiratory ailments and wound infections (Van Vurrenet al., 2006).

Libya is rich in ***Helichrysum stoechas*** species, but there is not sufficient botanical information concerning this species.

In Iran, 19 species of the genus *Helichrysum* were found, eight of them were endemic ***Helichrysum oligocephalum DC***, spread over the entire country, and were most common in the west of Iran. As far as we know, there is only one report describing the phytochemical composition of the essential oil of *H. oligocephalum* (Ebrahim Sajjadi et al., 2009).

Usage and Applications

Several studies have reported that there is great phenotypic variability in different species of the genus *Helichrysum*. There are also the problems of intra- and interspecific variability, all leading to significant variation in chemical composition. Some studies have analyzed the chemical composition of various *Helichrysum* species located in several places in the world (Madagascar; Turkey; Italy, Libya, Greece, Iran). The obtained essential oils varied significantly.

This variability may be attributed to the climatic conditions, harvested time, location, and geographic conditions. Afoulous et al. (2011) used *gymnocephalum* leaves harvested in July 2008; however, this plant was harvested in March 1997 by Möllenbeck et al. (1997) and during November–December 1994 by Cavalli et al. (2001). Therefore, it is important to note that despite the significant quantitative changes in *H. gymnocephalum* essential oil composition, 1,8-cineole remains the major component of *Helichrysum* essential oil. Roussis et al. (2000) showed that variability in the chemical constituents of the essential oil of *Helichrysum* species was detected according to the period of anthesis (before and after anthesis). Figure 2 shows a picture of a vial of *H. italicum* essential oil.

1,8-cineol has known antimicrobial properties (Pattnaik et al., 1997) and may possibly contribute to the antimicrobial effect of the *H. cymosum* oil. Table 2 shows the major components found in the different tested essential oils.

As we mentioned above, essential oils obtained from aerial parts of various *Helichrysum* species present a mixture of volatile compounds. Their chemical profiles vary with seasons, geographical conditions, plant parts used, the harvesting time, and even method of isolation. These secondary metabolites are known by their biological activities. In fact, many studies focused on the ability of essential oils to inhibit the development of food spoilage organisms, especially food-borne

TABLE 2 Major Compounds of *Helichrysum* Essential Oil

Helichrysum Species and Localization	Major Compounds of *Helichrysum* Essential Oil	References
Helichrysum gymnocephalum (Madagascar)	1,8-cineole (17.2–14.6%), borneol (16.2–10.4%), (E)-caryophyllene (12.7–9.9%), β-pinene (8.2–6.4%), and eugenol (2.2–13.5%)	De Medici et al. (1992)
	1, 8-cineole (20.4–14.1%), 3-ethyl-2,5-dimethylhexan-1,3-diene (17.2–2, 8%), α-farnesene (12.7–1.8%), β-pinene (3.1–10, 6%), (E)-caryophyllene (1.2–9.5%) and α-humulene (0.6–13.2%)	Theron et al. (1994)
	1, 8-cineole (66.7%)	Möllenbeck et al. (1997)
	1, 8-cineole (59.7%)	Cavalli et al. (2001)
	1, 8-cineole (47.4%), bicyclosesquiphellandrene[a] (5.6%), γ-curcumene[a] (5.6%), α-amorphene[a] (5.1%), and bicyclogermacrene[a] (5%)	Afoulous et al. (2011)
Helichrysum cymosum (L.) Less. (Tanzania)	*Trans*-caryophyllene (27.02%), caryophyllene oxide (7.65%), *p*-cymene (7.55%), Δ-3-carene (6.84%), and α-fenchene (6.25%)	Bougatsos et al. (2004)
Helichrysum fulgidum (L.) (Tanzania)	Caryophyllene oxide (12.45%), β-pinene (8.72%), spathulenol (7.88%), t-muurolol (7.31%), β-bourbonene (7.11%) and camphor (5.35%)	Bougatsos et al. (2004)
Helichrysum selaginifolium (Madagascar)	β-pinene (38.2%), α-pinene (16.3%), caryophyllene (7.5%), 1,8-cineole (7.1%)	Cavalli et al. (2001)
Helichrysum bracteiferum (Madagascar)	1,8-Cineole (27.3%), β-pinene (11.9%), α-pinene (5.9%), α-humulene (10.1%), and (E)-caryophyllene (7.1%)	Cavalli et al. (2001)
Helichrysum cordifolium (Madagascar)	(E)-caryophyllene (55.6%)	Cavalli et al. (2001)
Helichrysum faradifani (Madagascar)	(E)-Caryophyllene (34%) and linalool (16.1%)	Cavalli et al. (2001)
Helichrysum hypnoides (Madagascar)	(E)-Caryophyllene (35%) and 1,8-cineole (13.4%)	Cavalli et al. (2001)
Helichrysum orientale (Greece)	Nonacosane (11.1%)	Roussis et al. (2000)
Helichrysum heldreichii (Greece)	(E)-caryophyllene (38.5%) and caryophyllene derivatives (a total of 21.5%)	Roussis et al. (2000)
Helichrysum italicum subsp. *microphyllum* (Greece)	β-selinene (17.2%) and γ-curcumene (13.7%)	Roussis et al. (2000)
Helichrysum doeryeri (Greece)	Four eudesmol isomers (31.4%)	Roussis et al. (2000)
Helichrysum cymosum (southern tropical Africa)	α-pinene (12.4%) and 1,8-cineole (20.4%)	Van Vurren et al. (2006)
Helichrysum cymosum (Tanzania)	*Trans*-caryophyllene (27.02%), caryophyllene oxide (7.65%), *p*-cymene (7.55%), *Δ*-3-carene (6.84%), and *α*-fenchene (6.25%).	Bougatsos et al. (2004)
Helichrysum stoechas (Libiya)	α-pinene (59%), limonene (16,7%), and α-bisabolol (9.6%)	Sobhy and El feky (2007)
Helichrysum oligocephalum (Iran)	Thymol (14.4%)	Ebrahim Sajjadi et al. (2009)
Helichrysum graveolens (Bieb.) (Turkey)	α-cubebene (10.5%), β-caryophyllene (9.4%), caryophyllene oxide (8.2%) and azulene-octahydro (7.5%)	Bagci et al. (2013)

[a]*Newly identified compounds.*

TABLE 3 The Application Fields of the Major Components of *Helichrysum* Essential Oils

Major Components	Application	References
1,8-cineole	Antimicrobial properties against *Staphylococcus aureus*, *Bacillus cereus*, *Micrococcus luteus*, *Enterococcus faecalis* and aroma characteristics, such as fresh, herb, spice	Ayala-zavala et al. (2009)
	Antibacterial properties in vitro or in food models	Burt (2004)
	Antioxidant activities	Wang et al. (2008)
α-pinene	Antibacterial properties in vitro or in food models	Burt (2004)
	Antimicrobial agent against *Sarcina* spp.	Holley and Patel (2005)
	Antioxidant activity	Wang et al. (2008) and Dai et al. (2013)
	Antibacterial, antifungal, antiinflammatory, insecticidal, and antioxidant properties, also traditionally used as a flavoring agent and antimicrobial material in food	Tantaoui-Elaraki et al. (1993)
β-pinene	Antibacterial properties in vitro or in food models	Burt (2004)
	Antioxidant activities	Wang et al. (2008)
(E)-Caryophyllene	Antimicrobial and antioxidant activities	Mimika-Dukic et al. (2004)
Trans-caryophyllene	Antioxidant activity	Legault and Pichette (2007)
	Antibacterial activity	Rahman et al. (2008)
thymol	Antibacterial properties in vitro or in food models with an important minimal inhibitory concentration against pathogens such as *Escherichia coli*, *Salmonella typhimurium*, *S. aureus*, *Listeria monocytogenes*, and *B. cereus*	Burt (2004)
	Antimicrobial properties against *B. cereus*, *Clostridium botulinum*, *E. faecalis*, *E. coli*, *S. aureus*, *L. monocytogenes*, *Aspergillus flavus*, *Aspergillus niger*, *Penicillium corylophilum*, *K. pneumoniae*, *Pseudomonas aeruginosa*, *S. aureus*, *Salmonella* sp., and aroma characteristics such as spice, citrus, and wood	Ayala-zavala et al. (2009)

pathogens (Burt, 2004). Nowadays, there is a great interest in natural products to provide solutions for food protection against microorganisms, agriculture products, preservation of raw materials, and crop products against any chemicals substances that are toxic and damage health (Ayala-Zavala et al., 2009).

To have great idea about some biological activities, we developed Table 3 which shows an overview of the fields of application of the major compounds found in *Helichrysum* essential oils.

Usage and Applications in Food Science

Species belonging to the genus *Helichrysum* have various preservative capacities, mainly antimicrobial and antiseptic effects on food products. Particularly, leaves of *H. gymnocephalum* have been used as tea (Boiteau and Allorge-Boiteau, 1993). Plants that belong to the genus *Helichrysum* are often used for food, and the leaves are cooked and eaten in Africa (Mathekga et al., 2000). Figure 3 shows a photo of *H. gymnocephalum*.

The species *H. italicum* is known as "immortal of Italy". This plant is very aromatic, and is used in food because of its flavour, close to curry. However, this plant is not used in curry, which is an Indian blend of spices, but its scent is very similar. Its leaves pleasantly perfumed for all kinds of food preparation. In fact, its aroma of curry marries well with south Asian culinary habitudes, and Middle Eastern grilled meats, fresh cheeses, salads.

Helichrysum essential oil does not tolerate long cooking. When used with potatoes, it gives a taste of the Maquis to the preparation. The essential oil aroma of several *Helichrysum* species goes well with garlic dressing. Also, fresh or dried

FIGURE 3 Photo of *Helichrysum gymnocephalum*.

leaves, as well as essential oil, can flavor many savory dishes such as rice, vegetables, poultry, meat, and fish and it goes well with marinades for the barbecue.

Mancini et al. (2011) used GC and GC–MS to study the chemical composition of the essential oil of *H. italicum* (Roth) Don subsp. *italicum* collected in the National Park of Cilento and Diano Valley, Southern Italy. Forty four compounds of 45 constituents were identified in the oil, and the oxygenated sesquiterpenes were preponderant. The essential oil was evaluated for its possible in vitro phytotoxic activity against germination and early radical elongation of radish and garden cress. The essential oil contributed to the significant inhibition of the radical elongation of radish when applied at the highest doses. This example elucidated the use of *H. italicum* (Roth) Don subsp. *Italicum* essential oil in agriculture.

Bougatos et al. (2004) investigated the chemical composition of two essential oils obtained from the aerial parts of *H. cymosum* and *Helichrysum fulgidum* from Tanzania using GC and GC–MS. Sixty-five compounds, representing 92.4% and 88.2%, respectively, of the two oils, were identified. *Trans*-caryophyllene, caryophyllene oxide, β-pinene, *p*-cymene, spathulenol, and β-bourbonene were found to be the main components. Moreover, the oils were tested against two Gram-positive bacteria, *Staphylococcus aureus* (ATCC 25923) and *Staphylococcus epidermidis* (ATCC 12228), four Gram-negative bacteria *Escherichia coli* (ATCC 25922), *Enterobacter cloacae* (ATCC 13047), *Klebsiella pneumoniae* (ATCC13883), and *Pseudomonas aeruginosa* (ATCC 227,853). Essential oils were also tested against the pathogenic fungi *Candida albicans* (ATCC 10231), *Candida tropicalis* (ATCC 13801), and *Candida glabrata* (ATCC 28838). Results showed that the oil of *H. fulgidum* exhibited significant antimicrobial activity, while the oil of *H. cymosum* did not exhibit any antimicrobial activity.

Afoulous et al. (2011) showed that the essential oil of *H. gymnocephalum* had poor antioxidant activity against two tests: 2,2-diphenyl-1-picrylhydrazyl (IC_{50} value >1000 mg/L) and 2,2'-azino-bis(3-ethylbenzothiazoline-6-sulphonic acid) ($IC_{50} = 1487.67 \pm 47.70$ mg/L).

The antimicrobial activity of the essential oils of four *Helichrysum* species growing in Greece (*H. orientale, H. heldreichii, H. italicum* subsp. *Microphyllum*, and *H. doeryeri*) were investigated. *H. italicum* subsp. *microphyllum* exhibited the strongest antibacterial activity against *S. aureus*, *S. epidermidis*, *P. aeruginosa*, *Es.coli*, and *En.cloacae*. While only the oil of *H. oriental* significantly inhibited *Klebsiella pneumonia* (Roussis et al., 2000).

Van Vurren et al. (2006) studied the antimicrobial and toxicity activities of the essential oil of *H.cymosum.* The antimicrobial activity of the essential oil was determined using the *p*-iodonitrotetrazolium chloride (INT) microplate method, where the essential oil was tested against 10 pathogens. The result showed that the minimum inhibitory concentration ranged from 1 to 8 mg/mL. However, Bougatsos et al. (2004) reported that essential oil of *H. cymosum* was inactive against all pathogens tested (six bacteria and three pathogenic fungi).

Sobhy and EL Feky (2007) reported the antimicrobial activity of essential oil of *H. cymosum* against bacteria and fungi such as *S. aureus*, *S. epidermis*, *Es. coli*, and *C. albicans*.

SUMMARY POINTS

- *Helichrysum* essential oil is obtained from the different species of the genus *Helichrysum* which belongs to the Asteraceae family.
- The principal fields of use of *Helichrysum* essential oil are pharmacology, medicine, and the food industry.
- The chemical composition of *Helichrysum* essential oil varies according its geographical provenance, the period of essential oil isolation, and the parts of the plant used, harvested time, as well as the method of extraction.
- The main components of *Helichrysum* essential oil are 1,8-cineole, borneol, (E)-caryophyllene, β-pinene, eugenol, 3-ethyl-2,5-diméthylhexan-1,3-diene, α-farnesene, α-humulene, bicyclosesquiphellandrene, γ-curcumene, bicyclogermacrene, α-amorphene, *trans*-caryophyllene, caryophyllene oxide, *p*-cymene, spathulenol, *β*-bourbonene, camphor, α-pinene, linalool, and thymol.
- Biological activities such as antioxidant, antifungal, and antibacterial activity contribute to the application of *Helichrysum* essential oil in the food industry and agriculture.
- The antimicrobial activity of *Helichrysum* essential oil has been attributed to 1,8-cineol for both fungi and bacteria.
- *Helichrysum* has many food uses, such as in poultry and salads etc.

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