

CORSICA VS BALKAN IMMORTELE ESSENTIAL OIL – COMPARISON OF CHEMICAL PROFILES

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SUMMARY

Helichrysum italicum (Roth) G. Don., (Asteraceae) commonly known as immortelle, everlasting or curry plant is a typical Mediterranean plant, which grows spontaneously and as cultivated in the region of West Mediterranean countries (Portugal, Italy, and France), and region of Balkan-Mediterranean countries (Croatia, Bosnia and Herzegovina, and Serbia). This geographical division caused a difference in the quality of immortelle essential oil, which is present on the market under the INCI name: *Helichrysum italicum* flower oil (CAS number: 90045-56-0, EINECS number 289-918-2). Although this oil is in great demand on the international market, there is still no official standard that determines its quality. However, there is a valuation into two types of oil – Corsica vs Balkan immortelle essential oil, determined by quantity of main compounds: neryl acetate and α -pinene. The aim of this research was to collect publicly available GC-MS profiles of commercially offered immortelle essential oils (provided by manufacturers or companies engaged in the trade of essential oils), in order to estimate the market situation. The results showed that the offer on the market of immortelle essential oil indicates the existence of two types of oil: Corsica (with 29.8-44.0% neryl acetate, and 0.6-10.2% α -pinene) and Balkan (with 1.9-28.3% neryl acetate and 12.8-29.4% α -pinene), which overlaps with the statements of traders and scientists.

KEYWORDS: essential oil price, medicinal plants cultivation

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ABBREVIATIONS: CAS Chemical Abstracts Service (gives a unique numeric identifier for the substances described in the scientific literature); EINECS European Inventory of Existing Chemical Substances (provides a register of chemical substances commercially available within the European Union); GC-MS Gas Chromatography-Mass

Spectrometry; INCI International Nomenclature of Cosmetic Ingredients (ensures that the list of ingredients on the labels of cosmetic products is the same in all countries); ISO The International Organization for Standardization.

INTRODUCTION

Helichrysum italicum (Roth) G. Don., commonly known as immortelle, everlasting or curry plant is a typical Mediterranean plant growing in arid areas of Southern Europe on alkaline, dry, sandy, and poor soil (Kramberger et al., 2022; Primitivo et al., 2022). It is perennial shrub with woody base and several upright tomentose stems 30-50 cm high (Manitto & Monti, 1972). The leaves are elongated, linear, with rounded top, hairy, green on the upper side, and on the underside silvery, due to tomentose and glandular hairs (Glumac et al., 2023). The inflorescence is typical for Asteraceae family; flower heads, or capitula, is with numerous bright yellow flowers having a strong curry-like aroma (Primitivo et al., 2022), Picture 1.

Immortelle essential oil is available on the market under the INCI name: *Helichrysum italicum* flower oil (CAS number: 90045-56-0, EINECS number 289-918-2). The demand for immortelle essential oil has recorded a constant growth during the last two decades, especially in the cosmetics and pharmaceutical industries as fragrance compound, or in preparations for the protection of sensitive and irritated skin (Andreani et al., 2019; Tadić et al., 2021; Kunc et al., 2022). The increasing interest for this essential oil has led to a significant increase in prices on the world market, up to 2,000 € per



Picture 1. *Helichrysum italicum* plant and dried flowers

kg (Genčić et al., 2021). The price of immortelle essential oil is high, mainly due to the low content of essential oil in the flowers (0.15–0.38%), as well as to limited growing area (Erbaş et al., 2023; Nebrigić et al., 2023), similar as in case of damask rose (Aćimović et al., 2023).

Through the ages, in the countries where immortelle grows spontaneously, it was collected from nature and mainly used in traditional medicine: internally as infusion for treating digestive and respiratory problems, and externally in form of ointments for wound healing, as well as spice to pork meat and stuffing (Genčić et al., 2021; Kenig et al., 2021; Kramberger et al., 2021; Andjić et al., 2022; Erbaş et al., 2023). However, in the last two decades the growing interest and the high price of essential oil has triggered the need for commercial cultivation of immortelle in the fields (Hladnik et al., 2023). Since then, the production area under immortelle grew rapidly, and the market quickly became saturated and the price began to fall, which further implied the plowing and destroying of immortelle plantations after 2020 in Serbia, even though this plant could be suitable crop, with two harvests per year (Aćimović et al., 2021; 2022a).

Today, immortelle is commercially cultivated in West Mediterranean countries (Italy and France), as well as in Balkan countries (Croatia, Bosnia and Herzegovina, and Serbia), however there are attempts to expand to Bulgaria, Hungary, Turkey, and Algeria (Bouchaala et al., 2016; Djihane et al., 2017; Tzanova et al., 2018; Balázs et al., 2022; Zheljaskov et al., 2022; Erbaş et al., 2023). However, the quality of the essential oil has the greatest importance in the formation of the price, especially the italdione content. Although there is no ISO standard for

immortelle essential oil, the market recognizes two types of oil: Corsica (with more than 30% neryl acetate, and less than 10% α -pinene) and Balkan (with less than 30% neryl acetate and more than 10% α -pinene).

The aim of this research was to collect publicly available GC-MS profiles of commercially offered immortelle essential oils (provided by manufacturers or companies engaged in the trade of essential oils) in order to estimate the market situation.

MATERIAL AND METHODS

GC-MS profiles or certificates of commercially offered immortelle essential oils (provided by manufacturers or essential oil trading companies) were collected and analyzed to provide information on trends and demand in the international market for immortelle essential oil.

RESULTS AND DISCUSSION

By searching publicity available GC-MS profiles of commercially offered immortelle essential oils (provided by manufacturers or companies engaged in the trade of essential oils), 60 certificates were collected. A half of them belongs to Corsica type (Supplementary file Table 1), and another half belongs to Balkan type (Supplementary file Table 2).

The dominant compound of Corsica type immortelle essential oil according to this data is neryl acetate (29.8–44.0%), followed by γ -curcumene (4.7–17.4%), ar-curcumene (2.0–6.8%), italdiones (1.1–15.8%), β -caryophyllene ($\leq 9.1\%$), β -selinene ($\leq 5.1\%$), α -selinene ($\leq 3.1\%$), α -copaene ($\leq 2.1\%$), neryl propionate (0.8–6.3%), α -pinene (0.6–10.2%), italicene (0.5–

5.5%), nerol (0.4-3.8%), linalool (0.3-4.2%), iso-italicene (0.2-3.0%), and limonene (0.1-7.3%).

In the Balkan type the dominant compounds is α -pinene (12.8-29.4%), followed by γ -curcumene (6.8-29.4%), β -selinene (3.0-10.4%), β -caryophyllene (2.6-9.1%), italicene (2.6-5.0%), ar-curcumene (2.1-13.9%), neryl acetate (1.9-28.3%), α -selinene (1.8-5.3%), italdiones (1.5-10.6%), α -copaene (1.1-4.0%) limonene (0.4-4.6%), linalool (0.2-6.0%), nerol (0.2-2.5%), iso-italicene (0.1-3.4%), and neryl propionate (\leq 3.2%).

All these oils listed in Supplementary file Tables 1 and 2 are commercial, obtained by steam distillation process, while in scientific papers primarily hydro-distillation in Clevenger apparatus is used for essential oil isolation. It is known that these two techniques give differences in essential oil composition, what is previously reported for this plant species (Jažo et al., 2022), as well as for many others such as clary sage (Aćimović et al., 2022b), rosemary (Jaimand et al., 2018), agarwood (Nasardin et al., 2018), and laurel (Řebíčková et al., 2020).

There is a significant number of scientific papers dealing with the chemical composition of immortelle essential oil. It is mainly focused on regions; Balkan: Serbia (Aćimović et al., 2021; Stanojević et al., 2022), Bosnia and Herzegovina (Talić et al., 2019; 2021; Glumac et al., 2023; Nebrigić et al., 2023), Croatia (Ćavar Zeljković et al., 2015; Malenica Staver et al., 2018; Dzamic et al., 2019; Jažo et al., 2022; Kunc et al., 2022), Montenegro (Kladar et al., 2015; Šćepanović et al., 2019; Oliva et al., 2020), Bulgaria (Tzanova et al., 2018; Zheljaskov et al., 2022), and West Mediterranean: Portugal (Costa et al., 2015; Ferraz et al., 2022), Italy (Leonardi et al., 2013; Fraternali et al., 2019; Gismondi et al., 2020), and France (Mouahid et al., 2017; Lamaire et al., 2023).

Systematic review of scientific literature dealing with 104 immortelle accessions in total, showed the presence of ten potential chemotypes of essential oil: high neryl acetate (with 50.5-83.4%), moderate neryl acetate (with 19.5-48.0%), neryl acetate + ar-curcumene (3.9-20.3% and 0.8-14.5%, respectively), ar-curcumene + γ -curcumene (17.9-28.6% and 12.0-22.0%, respectively), γ -curcumene (13.6-27.7%), high α -pinene (25.2-53.5%), moderate α -pinene (5.6-20.0%), juniper camphor (25.3-45.1%), β -selinene (11.6-38.0%), and italdiones chemotype (Aćimović et al., 2021). According to this classification, the high neryl acetate chemotype and partially moderate neryl acetate belong to Corsica type, while neryl acetate + ar-curcumene, ar-curcumene + γ -curcumene, γ -curcumene, high α -pinene and moderate α -

pinene belong to Balkan type. From the other side, juniper camphor, β -selinene and italdiones chemotype are occurring probably as genetic variations (Morone-Fortunato et al., 2010; Galbany-Casals et al., 2011), inter-specific hybridization (Taban et al., 2015), or adaptation to environment (Melito et al., 2016; Andres-Sanchez et al., 2019).

Taking in account that the activity of immortelle essential oil depends on percentage of compounds (Genčić et al., 2021), it is important to introduce the official categorization of immortelle essential oils through standard, because different types of immortelle essential oils have different potential applications. For example, neryl acetate is important flavor compound widely used in perfumes and cosmetics (Sun et al., 2022), while for α -pinene were reported anti-inflammatory and hypoglycemic activities (Özbek & Yılmaz, 2017), as well as neuroprotective (Khoshnazar et al., 2020) and anticancer effects (Aydin et al., 2013).

Therefore, the type of immortelle oil rich in α -pinene (Balkan) is more suitable for application in pharmaceutical industry, while the type rich in neryl acetate (Corsica) is highly valuable in cosmetic and perfumery.

CONCLUSIONS

The offer of immortelle essential oil on the market indicates the existence of two types of oil: Corsica (with 29.8-44.0% neryl acetate, and 0.6-10.2% α -pinene) and Balkan (with 1.9-28.3% neryl acetate and 12.8-29.4% α -pinene), which overlaps with the statements of traders and scientists.

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SAŽETAK

'KORZIKA' VS 'BALKAN' TIP ETARSKOG ULJA SMILJA - POREĐENJE HEMIJSKIH PROFILA

MILICA AĆIMOVIĆ

Helichrisum italicum (Roth) G. Don., (Asteraceae), poznata kao smilje, tipična je mediteranska biljka, koja spontano raste i uzgaja se u regionu zapadnomediteranskih (Portugal, Italija, Francuska) i balkansko-mediteranskih zemalja (Hrvatska, Bosna i Hercegovina, Srbija). Ova geografska podela uslovlila je razliku u kvalitetu etarskog ulja smilja, koje je na tržištu prisutno pod INCI nazivom: cvetno ulje *Helichrisum italicum* (CAS broj: 90045-56-0, EINECS broj 289-918-2). Iako je ovo ulje veoma traženo na međunarodnom tržištu, još uvek ne postoji zvanični standard koji određuje njegov kvalitet. Međutim, postoji podela na dve vrste etarskog ulja smilja- 'Korzika' i 'Balkan', na osnovu količine jedinjenja: neril acetata i α -pinena. Cilj ovog istraživanja bio je prikupljanje javno dostupnih GC-MS profila komercijalnih eteričnih ulja smilja (koju obezbeđuju proizvođači ili kompanije koje se bave prometom eteričnih ulja), kako bi se procenila situacija na tržištu. Rezultati su pokazali da ponuda na tržištu eteričnog ulja smilja ukazuje na postojanje dve vrste ulja: 'Korzika' (sa 29,8-44,0% neril-acetata i 0,6-10,2% α -pinena) i 'Balkan' (sa 1,9-28,3% neril-acetata i 12,8-29,4% α -pinena), što odgovara izjavama trgovaca i istraživača.

KLJUČNE REČI: cena eteričnog ulja, gajenje lekovitog bilja

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