Botanical Adulterants Prevention Program BOTANICAL ADULTERANTS PREVENTION OULLETIN Adulteration of Oregano Herb and Essential Oil

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Oregano Origanum vulgare Photo ©2019 Steven Foster

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Goal: The goal of this bulletin is to provide timely information and/or updates on issues of intentional and accidental adulteration and mislabeling of oregano herb and essential oil (EO).* It provides information on issues of adulteration and mislabeling of oregano (Origanum vulgare subsp. hirtum, O. onites) herb used as a spice and herbal remedy, in particular with winter savory (Satureja montana, Lamiaceae) herb, sweet marjoram (Origanum majorana, Lamiaceae) herb, Cistus spp. (Cistaceae) leaf, olive (Olea europaea, Oleaceae) leaf, thyme (Thymus spp., Lamiaceae) herb, summer savory (Satureja hortensis, Lamiaceae) herb, strawberry (Fragaria spp., Rosaceae) leaf, sumac (Rhus spp., Anacardiaceae) leaf, hazelnut (Corylus avellana, Betulaceae) leaf, myrtle (Myrtus communis, Myrtaceae) leaf, and colored wheat (Triticum aestivum, Poaceae) bran. Additional species in the family Lamiaceae from the genera Origanum, Thymus, Poliomintha, Plectranthus, Monarda, and Hyptis or in the family Verbenaceae from the genera Lippia, Aloysia, and Lantana are known to have "oregano" as part of their common name, and may thus be mistaken for oregano by purchasing personnel who lack proper training.

The bulletin also addresses adulteration of oregano (*Origanum vulgare* subsp. *hirtum*) EO, in particular with *Origanum* and *Thymus* species, Spanish thyme blend, and synthetic carvacrol and thymol. This bulletin may serve as

guidance for quality control personnel, the international herbal products industry and the extended natural products community in general. It is also intended to present a summary of the scientific data and methods on the occurrence of species substitution, adulteration, the market situation, and economic and safety consequences for the consumer and the industry.

1. General Information

1.1 Common name: Oregano^{1,2†}

1.2 Other common names²⁻⁵

Greek oregano:

English: Wild marjoram *Danish:* Almindelig merian, merianurt *Dutch:* Oregano, wilde marjolein *French:* Origan, origan commun *German:* Dostenkraut, gemeiner Dost, Oregano, wilder Majoran *Greek:* Ρίγανη (rigani) *Italian:* Maggiorana selvática, origano, origano comune, *Norwegian:* Bergmynte, oregano *Portuguese:* Orégano, orégão *Spanish:* Orégano

* Some authors differentiate the volatile oil obtained by steam distillation from essential oil. For the purpose of this document, no such distinction has been made.



[†] The common name "oregano" is used for both, *O. vulgare* subsp. *hirtum* and *O. onites*. However, *Origanum vulgare* subsp. *hirtum* is also known as *Greek oregano*, while *O. onites* is commonly referred to as Turkish oregano (see section 1.6).

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Swedish: Kungsmynta, oregano *Turkish*: Kekik, Keklik otu

Turkish oregano:

English: Cretan oregano, Oikea oregano, pot marjoram *Danish:* Fransk merian, Tyrkisk merian, pottemerian *Dutch:* Potmarjolein, Kretenzische oregano, Turkse oregano *French:* Origan de Chypre, origan crêtois, origan turc

German: Französischer Majoran, Kretischer Dost, Kretischer Oregano, Türkischer Oregano *Greek*: Τούρκικη ρίγανη (Tourkiki rigani) *Italian*: Origano siciliano, origano cretico *Norwegian*: Fransk merian *Portuguese*: Orégano turco *Spanish*: Orégano cretense, orégano italiano, orégano, orégano turco *Swedish*: Smyrnamejram, spansk timjan, spansk dosta,

fransk melorian, krukmejram

Turkish: Bilyali kekik, Izmir kekiği

1.3 Accepted Latin binomials:

Origanum vulgare L. subsp. hirtum (Link) letswaart^{1,6} and Origanum onites L.⁷

1.4 Synonyms:

Origanum vulgare L. subsp. hirtum: Origanum heracleoticum auct. non L.,¹ Origanum hirtum Link,⁸ Origanum megastachyum Link,⁹ Origanum neglectum Vogel,⁹ Origanum smyrnaeum Sm.,⁹ Origanum vulgare subsp. hirtum (Link) A. Terracc.¹⁰

Origanum onites: Majorana cretica Mill., Majorana onites (L.) Benth., Origanum orega Vogel, Origanum pallidum Desf., Origanum smyrnaeum L.⁹

1.5 Botanical family: Lamiaceae

1.6 Vernacular name use:

There is much confusion regarding the vernacular name oregano. It is a name given to dozens of plant species from the families Asteraceae, Fabaceae, Lamiaceae, and Verbenaceae.¹¹⁻¹⁴ The main genera are Origanum, Lippia, Thymus, Plectranthus, Lantana, Aloysia, Monarda, and Hyptis. Their commonality is that their leaves or flowering tops have the flavor recognized as being oregano provided by the mix of carvacrol and thymol and other volatile compounds.^{12,14,15} Two main genera are available commercially in North America: Origanum and Lippia. Lippia graveolens (Mexican oregano) is most frequently used in the United States, Mexico, and Japan.¹¹ Mexican oregano may also include species from the genera Lantana or Hyptis. Spanish oregano is not derived from an Origanum or Lippia species, but from Thymbra capitata (syn. Thymus capitatus).^{12,13,15} Plectranthus amboinicus (syn. Coleus amboinicus, Lamiaceae) is known as Cuban oregano or Indian mint.^{14,16} Limitations with regard to the species permissible for use as a dietary supplement/herbal medicine or spice ingredient are indicated below.

1.7 Interchangeable use of species:

Dietary supplement and herbal medicine: According to the American Herbal Products Associations Herbs of Commerce, 2nd edition,¹ products made of Origani herba consist of flowers and leaves of O. vulgare subsp. hirtum, while the European Pharmacopoeia (Ph. Eur.) allows two species, O. vulgare subsp. hirtum and O. onites, to be sold as oregano.¹⁷ Dietary supplement ingredients derived from Lippia graveolens have to be listed as "Mexican oregano" in the United States.¹

Spice: The European Spice Association (ESA) and the American Spice Trade Association list two species that can be sold as oregano, O. vulgare subsp. hirtum and O. onites, while the International Organization for Standardization (ISO) has a broader definition for oregano which includes all species of the genus Origanum with the exception of O. majorana.18 Spices derived from Lippia species (Verbenaceae), particularly L. graveolens, have to be labeled "Mexican oregano." The draft standard developed by the Joint FAO/WHO Food Standards Codex Committee on Spices and Culinary Herbs (CCSCH) includes a number of species that can be marketed as dried oregano, including O. vulgare subsp. hirtum, O. onites, O syriacum, O. vulgare x majoricum, O. vulgare subsp. virens, O. vulgare subsp. viride, O. vulgare subsp. vulgare, Satureja montana, Lippia graveolens, L. berlandieri, L. palmeri, and Poliomintha longiflora (Lamiaceae). Origanum vulgare subsp. hirtum is offered under the trade name "Greek oregano," while O. onites is traded as "Turkish oregano," "Cretan oregano," or "Oikea oregano." Species of the Lippia and Poliomintha genus are traded as "Mexican oregano."19 Additional regional differences in what plant species is used as oregano have been reported.20

Essential oil (EO): Oregano oil is generally known as the essential oil obtained by steam distillation of the flowers and leaves of *Origanum vulgare* L. subsp. *hirtum*. The International Nomenclature of Cosmetic Ingredients (INCI) name for oregano oil is "*Origanum vulgare* leaf oil," "*Origanum vulgare* flower/leaf/stem extract," or "*Origanum vulgare* leaf extract." It is not known to what extent oils from other *Origanum* species can be/are sold as oregano oil, although the essential oils obtained from *O. compactum, O. onites*, and *Thymbra capitata* are commonly labeled as oregano oils.^{21,22} According to the International Trade Centre, oregano oil originating in Turkey may be derived from a range of species such as *Origanum, Satureja, Thymbra*, and *Thymus.*²³

1.8 Distribution:

Origanum vulgare is originally from the Mediterranean region, but is more widely distributed across Europe (including the British Isles), as well as south and central Asia, and, in North America in the lower 48 states and Canada.^{24,25}

Origanum vulgare is represented by six different subspecies (with a circumscribed regional distribution): O. vulgare subsp. glandulosum (Algeria, Tunisia); O. vulgare subsp. gracile (Central Asia, Iran, India, Turkey, Afghanistan, Pakistan); O. vulgare subsp. hirtum (Balkans, Greece, Cyprus, Turkey); O. vulgare subsp. virens (Morocco, Canary Islands, Azores, Balearic Islands, Spain, Portugal, Madeira); O. vulgare subsp. viridulum (widerspread across southern Europe to Iraq, Iran, Afghanistan, Pakistan); and O. vulgare subsp. vulgare (widespread across Europe and Asia, naturalized in North America and Venezuela).^{8,24,26,27} Origanum onites has a limited geographic range, occurring mainly in Southern Greece, the Greek islands of the Aegean sea, in some areas of western and southern Turkey and the island of Sicily.^{8,24,26-28}

1.9 Plant part and form:

Origanum vulgare is a perennial herb, with green fresh leaves, 3-28 mm long and 2.5-19 mm wide, petiolated or sessile. Laminae tend to be ovate or ovate-elliptic, with serrated or not serrated margins; acute or obtuse apex. Flowers are rare, found as broken parts of the corymbs. Bracts are greenish-yellow and imbricate. The calyx is corolla-like and inconspicuous. The corolla is white, on top of inflorescences, slightly conspicuous or inconspicuous.¹⁷ Dried leaves and flowers separated from the stems (stems contain lower amounts of essential oil) of oregano are used to produce oregano herb and oil.

The EO obtained by steam distillation from *O. vulgare* subsp. *hirtum* has been most frequently used due to its high EO content and a well-studied composition.^{27,29-31} *O. vulgare* subsp. *hirtum* has two main chemotypes, thymol-type and carvacrol-type, with intermediate chemotypes containing various amounts of carvacrol and thymol and a high content of the two precursors, *p*-cymene and γ -terpinene.^{27,32} According to ISO, oregano oil is defined as a yellow to dark brownish-red colored oil with a characteristic organoleptic profile (having aromatic, phenolic, and slightly spicy base qualities). The essential oil composition is 60-80% carvacrol, 4-10% *p*-cymene, 3-9% γ -terpinene, and small amounts of thymol (0.5-5%).⁶

1.10 General use(s):

Medicinal uses for oregano date back to the ancient Greek and Roman empires, where leaves were used to treat different conditions, such as skin sores, aching muscles, and as an antiseptic.³³ *Origanum vulgare* is employed as a stimulant, carminative, diaphoretic, and nerve tonic. It is also used in the treatment of respiratory ailments (asthma, coughs), headaches, toothaches, rheumatism, as an antibiotic, expectorant in bronchitis, antispasmodic, appetite stimulant, diuretic, diaphoretic, and as a mild sedative.^{2,12,33-35} Reported effects include: vasodilation, reduction of blood clotting, as well as sugar-lowering properties.³⁶ Oregano oil, when applied topically, works as an emollient, antipruritic (in spider and insect bites), and is used to treat frostbite and as a protectant against infections in cracks, bruises,

and insect bites.^{33,35} Oregano herb and oil have been used in European phytomedicine and in the United States as an analgesic in lozenges, and as an antiseptic in oral health. Carvacrol, the main active compound in oregano oil, is believed to be responsible for most of oregano oil's purported health benefits. A number of in vitro and in vivo studies support antibiotic, antifungal, anti-inflammatory, and immune-stimulant properties, as well as carvacrol's effects on digestive and cardiovascular health.¹⁵ Carvacrol is also used in the United States as a food additive, with demonstrated antiseptic and antioxidant properties in food preservation.¹⁵

2. Market

2.1. Importance in the trade:

2.1.1 Oregano oil

The use of oregano oil as an ingredient in commercial products is quite extensive. According to KHC Başer, PhD, Turkey dominates the global supply of oregano oil, providing around 70% of global needs. Annual Turkish oregano oil production in 2014 was estimated to be in the range of 15 to 20 metric tons. The oil is derived from a number of species from the genera *Origanum*, *Thymus*, *Thymbra*, and *Satureja*.^{15,23} Most oil production is from wild harvested material. Main production sites are in the Aegean and Mediterranean regions, with more limited production in the Marmara region. The optimum harvest time for oil production is just as the flower buds form. In 2018, Turkey exported approximately 66 metric tons of oregano oil for a return of US \$5.3 million. (KHC Baser email to S. Gafner, May 22, 2019)

2.1.2 Oregano herb

Historically, Mediterranean oregano herb was produced in Turkey, Greece, Albania, Spain, Italy, Israel, Morocco, and Egypt.^{11,37,38} Currently, Chile and Peru are also among the main producing countries.³⁹

A report from 2002 estimated that over 10,000 tons of dried oregano were harvested from wild sources in Turkey annually; about a tenth of that production was believed to be used domestically as condiment or herbal tea, and the remaining 9,000 tons used for EO production or exported as herb. *Origanum onites* is cultivated in the Aegean part of Turkey in fields totaling over 6,300 ha in the provinces of Denizli (4000 ha), Izmir (2000 ha), and Isparta (300 ha). Organic farming of *O. onites* is also practiced in Western provinces of Turkey.³⁷

In 2011, approximately 12,000 metric tons of oregano (*O. vulgare, O. onites, L. graveolens*) were traded globally with Turkey (8,182 tons), Peru (3,324 tons), and Mexico (215 tons) as the main producers, although numbers from the Food and Agriculture Organization of the United Nations (FAO) suggest that production in Mexico is much higher (see below).^{40,41}

More recent figures from a 2016 estimate that the world

trade volume of oregano is about 12,000-13,000 tons/year, although data from the Turkish Statistical Institute (TURK-STAT) indicate a production of 14,724 tons oregano in 2016 in Turkey alone.⁴² Turkey exported approximately 9-12 thousand tons of oregano in the years 2011-2016, with approximately 80% cultivated oregano and the rest wildcrafted.²⁹

Turkey is the major supplier of Mediterranean oregano herb consisting of over 90% of oregano exports.³⁷ *Origanum onites* (Turkish oregano) tops the list of commercial *Origanum* species of Turkey. It is obtained both from wild and cultivated plants. The other *Origanum* species collected from the wild for commercial use are the following: *O. majorana* (sweet marjoram), *O. vulgare* subsp. *hirtum* (Greek oregano), *O. minutiflorum* (Spartan oregano), *O. syriacum* var. *bevanii* (Israeli oregano), and *O. acutidens*, another Turkish endemic species.³⁷ Herb production reached 17,666 tons in 2018 with a value of US \$57 million; unit export value was US \$3.2/kg. (KHC Baser email to S. Gafner, May 22, 2019)

SADER, the Mexican government's Secretariat of Agriculture and Rural Development, reports that Mexico was the second largest producer of oregano (although not Mediterranean oregano) in 2013 and that oregano production was the highest in the central and northern Mexican states of from San Luis Potosi north to Baja California,⁴³ with Chihuahua producing 1,000 tons in the year 2015.⁴⁴ The FAO stated that Mexico produced between 5,000-10,000 tons of Mexican oregano per year in between 2014-2018.⁴¹ The Mexican oregano (*L. graveolens*) is used domestically and also exported to the international market, representing approximately 35–40% of oregano in the world. Consumption of Mexican oregano is restricted mainly to two countries outside of Mexico – the United States and Japan – which account for 90% of the total exports.^{44,45}

2.1.3 Oregano dietary supplements

Oregano dietary supplements have seen a consistent growth in sales from 2015-2018 in the United States, with

most of the sales in the natural channel (co-ops, associations, independent retailers and large regional chains), where it has been among the top 12 best-selling herbs.

2.2 Market dynamics:

In 2011, high quality dried oregano (min. 2.5% EO content) of Turkish origin was sold at wholesale prices of US \$2.58/kg while material of lower quality was available at US \$1.58/kg. In the same year, oregano (*Origanum vulgare*) from Peru was priced at US \$3.20/kg and Mexican oregano (*Lippia* spp.) originating in Mexico at US \$5.51/kg.⁴⁰ Prices of dried Turkish oregano in 2016 were US \$4.30/kg in the US and US \$4.56 in Europe,⁵⁰ while Mexican oregano was sold by wildcrafters at a fraction of a dollar (8-10 pesos/kg) and at US \$5.00/kg (100 pesos) by suppliers.⁵¹

An informal investigation of the retail prices for "oregano" essential oil from Chinese distributors (depending on amount purchased and if whole herb or leaf) on Alibaba. com showed products sold for as low as US \$1/kg, although starting prices were more often in the US \$10-25/kg range, which is well below the normal price range for authentic oregano oil. Based on information from the International Trade Centre, the price of pure oregano oil from Mediterranean source was in the range of 70-120 \in /kg in 2014,^{23,50} with corresponding prices in US \$ fluctuating between 80-150/kg.

3. Adulteration

3.1 Known adulterants:

3.1.1 Known adulterants of oregano herb

A survey of the European market from 2001 to 2007 revealed products frequently labeled and sold as "true" Mediterranean oregano contained: (1) plants lacking a clearly detectable EO profile, added as bulk extraneous material including berries (*Rubus* spp., Rosaceae), hoary rockrose (*Cistus × incanus*, Rosaceae), Sicilian sumac (*Rhus coriaria*, Anacardiaceae) and, (2) essential oil-bearing plants with oregano-like flavor, mostly belonging to the Lamiaceae

Channel	2015		2016		2017		2018	
	Rank	Sales [US\$]						
Natural ^a	10	7,755,387	12	7,510,751	10	8,693,675	10	9,925,727
Mainstream Multi-Outlet ^b	59	589,106	66	754,693	61	921,834	n/a	n/a

Table 1: Sales data for oregano dietary supplements in the herbal category in the United States from 2015-2018.

^a According to SPINS (SPINS does not track sales from Whole Foods Market.)

^b Data from 2015–2017 according to SPINS/IRI. This channel includes the food, drug, and mass-market sector, military commissaries, and select buyer's clubs and so-called dollar stores. SPINS/IRI data does not include discount department store sales, e.g., possible sales at Walmart and club stores are excluded, products sold through the internet or health care practitioners, and sales through independent stores who do not use check out scanners.

n/a: not available

Sources: Smith et al.,⁴⁶⁻⁴⁹ T. Smith (American Botanical Council) e-mail to S. Gafner, February 6, 2018, and K. Kawa (SPINS) e-mail to S. Gafner, July 11, 2016.

family (*S. montana* and *O. majorana*).^{52,53} Other reports indicate adulteration of oregano leaves may occur with anise (*Pimpinella anisum*, Apiaceae) seeds, myrtle (*Myrtus* spp., Myrtaceae) leaves, European hazelnut (*Corylus avellana*, Betulaceae) leaves, olive (*Olea europaea*, Oleaceae) leaves, and artificially colored wheat (*Triticum aestivum*, Poaceae) bran.^{18,37,52,54-56}

Given that the definition of the oregano plant encompasses several species of plants with similar properties, commercial decisions to purchase one or other oregano species has been largely based on availability of raw material, cost, and regional acceptability. There is a clear morphological and organoleptic distinction which can be made between Mediterranean and Mexican oregano for identity purposes using microscopic, macroscopic, and organoleptic methods. However, North American and European market trends are predominantly driven by regional preferences and market acceptability, represented in the U.S. by the American Spice Trade Association (ASTA) and in Europe by the European Spice Association (ESA) with their own definitions and cleanliness specifications (see section 1.6).¹⁸

3.1.2 Known Adulterants of Oregano Essential Oil

Adulteration, such as the use of lower-cost synthetic or natural material added to natural EOs, is a significant problem in the EO industry.⁵⁷ Adulteration of oregano oil occurs with synthetic thymol, carvacrol (or thymol-carvacrol mixtures), or limonene added from different sources.58,59 Carvacrol can be produced in a synthetic manner using a number of starting materials, e.g., by alkylation of o-cresol.^{60,61} "Thyme oil white" from Spain is a frequent adulterant used to dilute pure EOs. This is a blend made out of several natural fractions of Spanish thyme (Thymus zygis) oil, plus other oregano species (besides O. vulgare) with added natural p-cymene. (E. Schmidt email, November 29, 2017) Spanish thyme oil contains around 40-50% thymol and 4% carvacrol.58 Adulteration with EO from Thymus spp., or synthetic thymol has also been found during routine quality control assessments in industry laboratories (C. Beaumont, [doTerra] oral communication to S. Gafner, February 7, 2019). Synthetic thymol can be obtained in a similar manner as synthetic carvacrol, e.g. using *m*-cresol as a starting material which is subjected to alkylation with propylene.62

3.2 Sources of information supporting confirmation of adulteration:

3.2.1. Oregano spice

In its guidance document, ASTA reports that oregano adulteration is a common practice. The organization reported the sale of oregano with extraneous matter from different parts of other plants of similar appearance and determined that this is typically added at the cutting/grinding/blending stage.⁶³ Oregano is frequently adulterated with *Cistus* spp., sumac, and myrtle. The *Cistus* genus has



20 species – perennial shrubs found on dry or rocky soils throughout the Mediterranean region, from Morocco and Portugal to the Middle East. *Cistus* leaves have a dark green color that, when added to oregano, makes the adulterated spice visually more appealing than pure oregano.⁶³ Other herbs which have been found in oregano include winter savory and sweet marjoram. Seven commercial Mediterranean oregano samples also exhibited different contamination levels of *O. majorana* (ranging between 0% and 95%) and *S. montana* (ranging between 0% and 9.7%). These samples were obtained from wholesale traders during a survey carried out between 2001 and 2009.⁵²

In a more recent report from the United Kingdom, 53 commercially available oregano samples were purchased at various retailers, including convenience shops, supermarkets, and market places in the UK and Ireland.¹⁸ In addition, 25 samples were also purchased from online retailers from EU and non-EU countries. Samples were tested for identity using spectrometric methods (FTIR and LC-HRMS). Approximately 24% of the total 78 oregano samples tested were adulterated and the amount of adulterant ranged from 30% to over 70%; two samples had no oregano present.

Similar figures were found for oregano purchased from retail stores and on the Internet. The most common adulterants found in the samples were olive leaves and myrtle leaves. No difference was found between samples purchased in supermarkets and those online.¹⁸

CHOICE, a consumer advocacy group in Australia, had 12 commercial oregano products tested, representing more than 80% of the value of Australia's oregano sales in the herb and spice product category. Samples were analyzed for authenticity by Fourier transform infrared (FTIR) spectroscopy and subsequent statistical testing. The results gave evidence that seven of these products contained less than 50% oregano and were adulterated with olive leaves or sumac leaves.⁶⁴⁻⁶⁶ The consumer advocate group referred the issue to the Australian Competition and Consumer Commission.^{67,68} Based on the complaint, ACCC initiated its own investigation into a number of oregano brands, and found that one brand manufactured in Australia and three other products sold as oregano leaves were mislabeled since they contained substantial amounts of undeclared olive leaves.67,68

Finally, in a presentation on the quality control of spices, Larry Lichter, Vice-President of Global Quality and Food Safety at McCormick & Co., Inc., reported that oregano adulteration is common in non-qualified supply chains (i.e., the "spot market", also known as "cash market", where commodities are immediately delivered after the sale), with sumac, olive, strawberry, myrtle, hazelnut, and cistus leaves being the principal adulterants.⁶⁹

3.2.2 Oregano oil

In 2014, an alert on the EU RASFF (Rapid Alert System for Food and Feed) portal was placed on a nutritional supplement distributed in the Czech Republic, Germany, Finland, and Slovakia. The oregano oil product in capsules packed in Finland was found to be adulterated with litsea (*Litsea cubeba*, Lauraceae) oil, a widely used EO in the perfumery and cosmetic industry, but considered to be a potential allergen due to its citral content.⁷⁰

Essential oil expert Erich Schmidt, PhD in his chapter on the adulteration of EOs in the *Handbook of Essential Oils*, 2nd edition, reports that: "Origanum oil is adulterated with synthetic thymol and carvacrol, or with limonene from different sources." Adulteration with carvacrol is also mentioned in Hager's Handbuch der Pharmazeutischen Praxis.¹³

3.3 Accidental or intentional adulteration:

Both intentional and accidental substitution seems to occur, based on anecdotal and scientific evidence.^{18,53} The scenario is complicated by the large heterogeneity of the *Origanum* genus and by the denomination of different botanical genera under a single generic name. According to ASTA, oregano herb adulteration is economically motivated and intentional.⁶³ Oregano is frequently adulterated with *Cistus spp.*, to intentionally give a dark green color

when added to oregano. This makes the adulterated spice more visually appealing than pure oregano. The addition of undeclared plant materials, such as myrtle, olive, sumac, and hazelnut leaves, may be accidental, but in most cases these ingredients are added for financial gain. Adulteration with savory, thyme, or marjoram frequently reported in the literature is likely accidental.

The adulteration of oregano EO appears to be economically motivated. The so-called "thyme oil white" (see section 3.1.2) is available at a lower cost than oregano oil, and therefore admixture of this oil with oregano oil can increase profit for the seller.^{58,71}

3.4. Frequency of occurrence:

The frequency of adulteration of oregano herb appears to be high. Marieschi et al. found that 61 of the 84 samples (72.6%) obtained from international wholesale traders exporting to the European Union contained more than the permissible 2% of foreign matter, including up to 77% winter savory and up to 95% marjoram. Almost all the samples contained stem parts above the permissible 2% content (stem contents between 1.5-12% were detected).⁵³

Investigations into the composition of commercial oregano spice samples in Argentina found that all samples contained 5% or more of foreign matter. Using macroscopic and microscopic features, colored wheat bran was detected in four out of six (67%),⁵⁶ and 10 out of 36 (28%) samples,⁵⁵ respectively.

Between 30% to over 70% of oregano samples in the UK and Ireland tested were found to include other non-oregano materials, with a follow-up investigation of 54 oregano samples suspected to be adulterated finding that 85% of the products contained 20% or more undeclared bulking agents (cistus, myrtle, olive or sumac leaves).^{18,72} An investigation in Denmark found that four out of 10 oregano herb samples contained between 30-50% bulking agents such as cistus and olive leaves.⁷³ Over 50%, or seven out of 12 of the popular brands tested in Australia, also contained substantial amounts of other plant materials.⁶⁴

The frequency of adulteration of oregano oil is not known as the information is scant, but suggestive of widespread adulteration. Erich Schmidt communicated that all of the 38 commercial samples of oregano oil analyzed in his laboratory were adulterated with Spanish white thyme oil and *p*-cymene. (E. Schmidt, email November 29, 2017).

3.5 Possible safety/therapeutic issues:

According to ASTA, oregano herb is commonly adulterated with non-toxic, less expensive leaves, such as sumac, cistus, or myrtle. This economically motivated adulteration is done with material that does not present a significant public health risk.⁶³

As mentioned above, one of the adulterating materials of oregano oil was found to be *Litsea cubeba* oil.⁷⁰ This is an EO from a tree widely found in China, Indonesia, and other parts of Southeast Asia that has uses in the fragrance

industry. The content of citral in the oil is typically about 70% and is known to be a skin irritant and skin sensitizer.⁷⁴ Cosmetic regulations in the EU require citral, when present above 0.01% in rinse-off products (soaps, shampoos), or above 0.001% in leave-on products (creams, deodorants)), to be declared as an allergenic compound.⁷⁵

3.6 Analytical methods to detect adulteration:

Macroscopic and microscopic methods of analysis for the authentication of oregano herb have recently been reported.^{17,53,54,76} Macroscopic identification criteria can be found, e.g., in the monograph of the European Pharmacopoeia,¹⁷ or the publication by Varela et al.⁵⁵ Bononi et al. published microscopy images of oregano, cistus, hazelnut, marjoram, olive, sumac, and savory leaves.⁵⁴ The European Pharmacopoeia also details a thin-layer chromatography (TLC) test which can be used for oregano herb and oregano EO.17 The extracted oregano herb sample or the corresponding EO are run on a TLC silica gel plate R with methylene chloride as the eluent and anisaldehyde as the reagent. Thymol is characterized visually as a pink zone and carvacrol as a pale violet zone on the TLC plate. A gas chromatography (GC) procedure for oregano EO has also been reported.¹⁷ In addition, ISO provides a method to determine the identity of oregano oil in accordance with ISO standard 11024 (all parts).6

Gas chromatography mass spectrometry (GC-MS) and high-performance liquid chromatography mass spectrometry (HPLC-MS) methods to distinguish oregano leaf from olive leaf, one of the most common oregano herb adulterants, have been published by Bononi et al.^{77,78} These methods take advantage of the presence of phenolic compounds,

e.g., oleuropein in olive leaves, which are absent in oregano. Black et al. developed and fully validated a two-tier approach utilizing FTIR with multivariate statistical analysis and ultra high-performance liquid chromatography high resolution mass spectrometry (UHPLC-HRMS) to screen for and confirm oregano adulteration by other plant materials, such as cistus, hazelnut, myrtle, olive, and sumac.¹⁸ The same FTIR method was used by Wielogorska et al., and compared to a UHPLC-MS/ MS method using ions specific for hazelnut, myrtle, olive, and sumac to detect adulteration. The detection limit (LOD) for FT-IR was established to be 10%; LODs for the UHPLC-MS/MS method were below 2%, except for cistus leaves, for which no suitable marker ion was found.72

An unusual approach to detect oregano herb adulteration was published by Drabova et al.⁶⁹ These authors developed a statistical model to predict oregano adulteration based on the GC-MS/MS analysis of 183 pesticides and HPLC-MS/ MS of 335 pesticides in 42 authentic and 34 adulterated oregano samples. Using multivariate statistics, a model was constructed with a good prediction ability according to the authors. Pyriproxyfen, cyfluthrin, and cyhalothrin were identified as the most important markers of possible adulteration.⁷⁹

In response to the CHOICE consumer group food alert in the UK and Australia,⁶⁴ Reading Scientific Services, Ltd., used microscopy and subsequent analyses of the constituents by GC-MS. In addition, oregano samples were analyzed with Next Generation Sequencing (NGS) for rapid determination of the DNA sequences of all ingredients in a sample.⁸⁰

Other rapid methods for detection of oregano herb adulterants in crude material are based on developing and successfully implementing test for molecular markers. These methods include the comparison of random amplified polymorphic DNA (RAPD) markers of oregano and *Rubus* spp., *Cistus incanus*, and *Rhus coriaria* added to oregano as bulk extraneous material. Out of 20 random primers tested, 13 produced sequences that allowed discrimination between *Origanum* spp. and *Rubus caesius*, *R. coriaria*, and *C. incanus*, with a limit of detection of 1%.⁵³

Sequence characterized amplified region (SCAR) markers for *Satureja montana*, *O. majorana*, and olive leaf adulterants were also developed from RAPD markers.^{52,81,82} The SCAR primers gave rise to specific sequences and subsequent characterization of bands after gel electrophoresis allowed the detection of oregano adulterating plants at a concentration of 0.5%.^{52,81}



Volatile adulterants in oregano oil lack the genetic information from the plant and thus cannot be identified with DNA-based methods. The usefulness of GC fingerprints to authenticate oregano EOs is hampered by the high intraspecific variability in the EO composition.^{83,84} To detect adulteration of oregano oil with synthetic carvacrol, a GC-stable isotope ratio (SIR)-MS method is used to measure the stable isotopes. Differences in the ²H/¹H ratio (δ^2 H) between natural carvacrol (δ^2 H: -272 to -264), and synthetic carvacrol (δ^2 H: -190 to -225) can be used to determine the presence of undeclared synthetic carvacrol. (E. Schmidt email, November 30, 2017)

4. Conclusions

Adulteration of oregano herb is prevalent, with a number of foreign materials including *Origanum* spp., *Cistus* spp., *Thymus* spp., *Satureja* spp., sumac, olive, and myrtle leaves frequently detected. The addition of undeclared colored wheat bran in commercial oregano spice products has been reported from Argentina. Confusion and variations in the definition of oregano, and the number of species commercially bearing the same name may result in accidental adulteration by less thoroughly trained wildcrafters. Regional distinctions and guidelines are established by spice trade associations, limiting impurities of extraneous materials to 1-2%.

The prevalence of oregano oil adulteration is just beginning to be understood, and not much information has been confirmed by laboratory tests and reports in the scientific literature. It appears that more affordable EO components and byproducts from EO processing from the fragrance and flavor industries are being mixed with oils derived from Origanum, Thymus, and species from other genera. Although ISO has established a standard for only Mediterranean oregano oil based on the distinct flavor and aroma provided by high carvacrol and low thymol content, the rationale for this decision does not seem to take into consideration the complexity of the oregano herb definition including more than two dozen different plants. Moreover, oregano oil health benefits are very likely shared by several plant species with the common name "oregano" and not exclusively to Mediterranean oregano.²³ The potential lower cost of raw materials may increase the opportunity for adulteration of Mediterranean oregano oil with essential oils, or oil fractions from a large number of other Origanum species and other plants with a similar fragrance profile.

The adulteration of oregano herb with low-cost materials from other plants is not considered a health issue. However, some concerns have been raised with certain adulterants of oregano oil: Allergic reactions may occur in people sensitive to citral when oregano oil contains sufficient amounts of undeclared essential oils with a high concentration of citral.

The ability to sell adulterated oregano at a fraction of the price of high-quality authentic material leads to a substantial market disadvantage for reputable oregano suppliers and manufacturers. It also increases the pressure on these companies to provide ingredients/products at lower cost to stay competitive. Appropriate quality control procedures need to be in place to ensure the authenticity of oregano-based ingredients.

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REVISION SUMMARY

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