The Essential Oil from Rosmarinus officinalis L.

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The essential oil produced from the leaves of *Rosmarinus officinalis L.* is an important essential oil of Spain. The Rosmarinus officinalis L. plant grows wild and has been cultivated in different parts of the country. The following sections treat history, botany, essential oil production, physicochemical properties, chemical composition, olfactive properties and applications, and production and trade of the essential oil of Rosemary. The preparation of terpeneless essential oil of Rosemary will be mentioned.

To obtain a general impression about Rosemary oil one should consult the existing books and reviews.[1–11] The publications cited in these books and reviews will be mentioned, as will other publications and this author's investigations and ideas.

History

In Greek and Roman times the use of plants of Rosemary was important in religious ceremonies and public festivities. The plant was regarded as a symbol of fidelity.[12a] Rosemary plants are still used today in southern parts of Spain during religious processions, when the plants are strewn on the streets.

The name of rosemary originated from the Latin words *rŏs* (rŏris) = dew and *mărīnus* = from the sea, in other words, dew of the sea. Other authors[12b] suggested that the name rosemary could also stem from the Greek words *rhops* = brush and *myrinos* = aromatic.[2] The latter explanation is improbable because the old Greek name for rosemary is *libanotis,* which means a kind of incense. The Romans also called rosemary the flower of the sea, and Linneaus indicated that sailors on the Mediterranean sea could smell the odour of rosemary even before they could see the land.[2,12a]

The steam distillation of rosemary plants has been known throughout history. In the early middle ages a Hungarian queen used the alcoholic waters of rosemary plants to bathe and for body care; this lady reached a great age and therefore an alcoholic (spirit of wine) distillate of Rosemary got the name of Hungarian Water. Even in the 13th century this material probably was the first popular perfume.[13,14] According to

Sample of Rosemary Oil, available from Bordas/EL Scott, can be found on page 66.

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Table I. Subspecies and Varieties of *Rosemarinus officinalis L.*

Turrill(25)	Falchi Delitala and Soccolini(26)
1. Rosmarinus officinalis L.	Rosmarinus officinalis L.
var. genuina Turril	var. genuina, forma typica Turrill
forma erectus Pasq.	var. genuina, forma erectus Pasq.
forma humilis Ten	var. genuina, forma humilis Ten
forma albiflorus Beg.	var. genuina, forma albiflorus Beg.
var. rigidus Car et Snt.Lag	var. rigidus Car et Lag.
var. augustifolius Guss	var. augustifolius Gass
var. latifolius Beg.	var. latifolius Beg.
var. pubescens Pamp.	var. pubescens Pamp.
	var. pubescens forma typica Pamp.
	var. pubescens forma roseus Pamp.
2. Rosmarinus laxiflorus de Noé	
3. Rosmarinus lavandulaceus de Noé	
4. Rosmarinus tournefortii de Noé (arab Khlil)	

Villanovus, rosemary oil and turpentine oil were distilled for medical purposes during the middle ages.[13] Later on Lullus,[15] one of Villanovus' students, described the steam distillation of rosemary oil in more detail. The steam distillation of the essential oil of rosemary had been described in the middle ages in several medical books. Rosemary oil, obtained by empyreumatic distillation of the plants, has been described by Lindner.[16]

Investigation of rosemary oil started in the 18th century; however, the presence of camphor (so-called rosemary-camphor) in rosemary oil had already been discovered in 1685.[16] More detailed analysis of rosemary oil continued in the second half of the 19th century.

During that time, the following proudcts were detected in the oil:

d- and l-camphor[17,18]
α-pinene and camphene[19]
borneol and acetate[20]
1,8-cineole[21]
a sesquiterpene (probably caryophyllene)[22]

Thus in fact before the beginning of the 20th century the most important constituents of rosemary oil had been identified.

Botany

The rosemary plant *(Rosmarinus officinalis L.)* belongs to the most important family of the Labiateae (Lamiaceae), which comprises up to 200 genera and about 3,500 species. The species *Rosmarinus officinalis L.* is a dense evergreen shrub with a characteristic aromatic smell, and lavender-like leaves. It is a much branched, densely bushy shrub, up to 1.5 m high (mostly 50 cm) which remains evergreen throughout the year. The leaves are 2 to 4 cm long, by 1 to 3 mm thick, almost cylindrical, folded inwards along the margin; they are leathery, green and rough above, white-hairy below. The flowers are situated in little clusters towards the ends of the branches.

The calyx is mealy, two-lipped, the upper with a single broad oval lobe, the lower with two-pointed triangular lobes; the corolla is two-lipped with the two violet stamens and the long curved style projecting from it. The habitat is all the Mediterranean countries, especially in the maquis and dry hills near the coast. The plant blooms throughout the year; however, the flowering is most abundant in Spring.[12a,12b,23,24]

According to several publications the rosemary plant should be only one species in the genus *Rosmarinus;* however, several subspecies and varieties exist.

Turrill[25] and Falchi Delitala and Soccolini[26] gave a review of different (sub) species and varieties, which are depicted in Table I.

Lopez Gonzalez[12b] found in Spain, apart from *Rosmarinus officinalis L.*, which grows there abundantly, another species, namely *Rosmarinus eriocalyx Jordan & Fourr.*, which also grows in North Africa. Moreover from the latter comes a subspecies or variety which is named *Rosmarinus tomentus Huber-Morath & Maire.*

Thus the genus Rosmarinus with its species, subspecies and varieties is rather complicated, and it is doubtful that within this genus only one species should exist.

Guenther[1] wrote, according to a private communication of R. Bordas,[27] that, so far as Spanish rosemary oil is concerned, botanists at the University of Seville do not doubt that the rosemary growing in Spain is of the same variety (probably meant species) everywhere, and they attribute the difference in the physicochemical properties of the Spanish oils to local conditions. The regions of Murcia and Almeria (de Levante quality) possess a warmer climate than Granada and Jaén (Granadino quality) especially during winter. At least until 1970 the difference between these two qualities was noted.

The botany of the varieties and subspecies of *Rosmarinus officinalis L.* growing in Yugoslavia (Dalmatia) was described by Gvozdanovic[28] and Tucakov.[29]

Paulet and Vacquier reported on the occurrence of two types of rosemary in France (Langedoc—La Garrigue), one with blue-violet flowers and dark green leaves, the other with white flowers and green-yellow leaves.[30] Most of the plants growing wild in that region were hybrids of the two types. Graafland declared the two types morphologically identical;[31] yet the odour of the oils obtained from the two types differed markedly.[1] Granger et al. mentioned that with *Rosmarinus officinalis L.* there exists a great probability for the differentiation of the well-defined biogenetic types.[32] Three principal types should exist with respect to the structure of carbon-skeleton of the main constituents of the essential oils, e.g., the eucalyptol type (Italy, Morocco, Tunisia), the camphor-borneol type (Spain), the α-pinene-verbenone type (France/Corsica).

Apart from the existence of subspecies and varieties of *Rosmarinus officinalis L.* it is probable that within the species there are infraspecific differences, where morphologically identical plants possess different compositions of essential oils.

Essential Oil Production

Oil of rosemary is produced in the following countries: Algeria, France, Greece, Italy, Morocco, Portugal, Russia, Spain, Tunisia and Yugoslavia.

Although the isolation method of the essential oil of rosemary is rather simple, namely, the steam distillation of the twigs with leaves or leaves only, several interesting aspects can be discussed with respect to yield, physicochemical properties and chemical composition.

Some of these aspects are: the time of harvesting, the condition of the twigs and/or leaves, the apparatus, and the technical performance of the steam distillation. Mostly the leaves with the twigs are steam distilled, at least in field distillation plants. One might suppose that the leaves with the twigs should give a higher camphor content in the final oil than the leaves without twigs, however, this has not been proven up to now.

Table II. Yields of Rosemary Oil with Different Plant Material

Plant Material (reference)	% Yield	Origin
Dried leaves (34)	2	France
Flowers (34)	1.4	France
Fresh plants (35)	0.6–0.75	Spain
Fresh plants (3)	0.6–1.0	Spain
Cultivated fresh plants (33)	0.53–0.58	Spain
Wild plants (33)	0.57	Spain
Fresh leaves (36)	1.27–1.67	Yugoslavia
Dried leaves (36)	1.44–2.57	Yugoslavia
Young leaves (37)	1.5	Yugoslavia
Old leaves (37)	0.5	Yugoslavia

Table III. Steam Distillation of Rosemary Oil

Distillation Time (reference)	Yield	Origin
1.5 hours (38)	0.38–0.5	France
3–4 hours (35)	0.6–0.75	Spain
2–3 hours (3)	0.6–1.0	Spain
1.5 hours (39)	82% of the oil	Yugoslavia
0.5 hours (39)	15% of the oil	Yugoslavia

It is very probable that the camphor content is dependent on the age of the plants; old plants have lignified twigs with a higher camphor content than young plants,[33] Rasmussen et. al.[37] found that young leaves contained more (1.5%) oil than old ones (0.5%). Moreover they found that in old leaves the tops of the leaves contain about 10% more camphor than the lowest part of the leaves; the monoterpene content was just the opposite.

An excellent rosemary oil can be obtained from the leaves only.

In most countries, freshly harvested plants (twigs with leaves) are steam distilled; in Yugoslavia, however, dried plants (mostly leaves only) are used.

Yields of the oils with different plant material are shown in Table II.

Three types of apparatus have been used for the steam distillation of rosemary plants. These apparatus have been described in detail by Guenther,[1] Gildemeister and Hoffmann[5] and Gil

Table IV. Physicochemical Properties of Rosemary Oils from Different Origin

Origin (reference)	Specific Gravity	Optical Rotation	Refractive Index	Solubility L80° EtOH	Ester No. % as Bornylacetate	Alcohol No. % as Borneol	Acid Number	% Cineole/ Camphor
Spain								
Lorca (1)	0.898	-0°32'	-	5.5-6	2.6	10.4	-	-
Cieza (1)	0.896	-2°14'	-	6-7	2.8	10.6	-	-
Calaspara (1)	0.896	-2°27'	-	10	2.8	10.9	-	-
Jaen (1)	0.898	+4°53'	-	6	2.8	14.8	-	-
Granada (1)	0.898	+4°47'	-	10	2.9	15.1	-	-
general (5)	0.893-0.910	-0°58' to +11°30'	1.4682-1.4712	4.5-5 turbid 10	1.0-3.9(5.4)	8.7-15.6	-	-
general (41)	0.898-0.920	-6° to +11°30'	1.468-1.473	1-8	0.8-7	8.5-14.3	-	-
general (33)	0.897-0.926	+7.5° to +9.5°	1.4660-1.4728	-	1-2.2	4.65-16.03	0.88-2.8	-
wild (33)	0.902	-1.4°	1.4696	6.7	3.1	11.9	-	19.8/20
Cultivated (different zones) (33)	0.892-0.895	-0.3° to -7.5°	1.4708-1.4729	7.9-8.1	3.1-3.8	8.5-12.5	-	13.5/5
Portugal (42)	0.8692	+6°32'	1.4719	10	2	7.15	0.14	-
general (50)	0.865-0.885	+7° to +22°	1.470-1.474	8-15	3	-	1	-
France								
general (5)	0.900-0.920	up to +13°10'	1.467-1.472	1-8	1.0-4.9	8.0-11.3		
Matelettes (1)	0.9135	+15°24'	-	soluble in 75°	-	-	-	-
Couvent de Mourg. (1)	0.912	+12°9'	-	1	-	-	-	-
La Paillade (1)	0.909	+5°15'	-	1.8	-	-	-	-
cultivated (5)	0.926	+9°6'	-	0.7	-	-	-	-
Tunisia (43)	0.9165	+2o10'	1.4701	1.1-10	2.9	11.0	-	41.4/0
(43)	0.9164	+2°20'	1.4693	3-10	2.0	11.4	-	40.8/0
(yearly/monthly) (1)	0.907-0.919	+2°30' to +5°12	1.467-1.471	1-4	-	-	-	-
February (44)	0.9156	+3°4'	1.4683	1.5	0.65	12.4	0.3	-
July (44)	0.9136	+2°48'	1.4681	1	0.65	11.65	0.3	-
Yugoslavia (5) (Dalmatia)	0.894-0.913	+0°43 to +5°53'	1.466-1.468	1-8	1.8-7	8.4-14.3	0.7	
(1)	0.899-0.904	+2°6 to +2°38'	-	2	2.3-4.2	10.5-11.8	-	
(36)	0.901-0.915	1°20 to +13°22'	1.464-1.469	0.7-9	1.7-6.9	-	0.24-1.24	
Italy								
Grosseto (general) (45)	0.894-0.915	-2°0' to +13°0'	1.465-1.470	10	2-7	-	-	
Sicily (leaves) (46)	0.9208	+5.60	1.4659	1.25	3.7	6.44	0.6	
Sicily (twigs and leaves) (46)	0.91798	+8.98	1.4660	6.6	5.25	8.64	1.0	
Lucanien, tyrrhen. (47)	0.9050	+4°18'	1.4708	3	3.92	16.63	-	36/8.25
Lucanien, ionish (47)	0.8988	+2°18'	1.4698	10	7.18	7.66	-	38/6.95
Sardin (48)	0.9118	+21.5°	1.471	-	17.1	18.8	-	9.2/9.38
				90°				
Russia								
Crimea (49)	0.8954	-9°20'	1.4727	0.2	ca 3	ca 12	0.72	
Caucasus (49)	0.9364	+3°3'	1.4735	0.1	ca 7	ca 25	7.76	
Morocco								
I (1)	0.905	+0°14'	1.4690	1.5	1.6	10.3	-	
II (1)	0.915	+1°16'	1.4660	1	2.0	10.1	-	

Montero.[3]

The old-fashioned method is the field steam distillation of the plants in direct contact with the water which is distilled. The water is heated with the extracted and dried plants. Water for distillation and cooling is obtained from small rivers on the spot. Although plant material, water, energy and labour (families) is cheap, the yields of this method are rather low, below 0.5%.

A better method is the field steam distillation in stills with a double bottom, where the plants are on a type of grill and not in direct contact with water. These apparatus are still producing oil in Spain. This method affords the oil in a yield of 0.5-1.0%.

A more modern method uses steam, directly or indirectly produced, of 2 to 3 atmosphere pressure. This method gives oils in yields of 1.0-1.5% with freshly harvested plants and of 1.5-2.5% with dried leaves.

The technical performance of the steam distillation is important for the yield and the olfactive quality of the oil. Distillation times varying from 1.5 to 4 hours have been mentioned in the literature. It is probable that a longer distillation time causes a higher content of sesquiterpenoids in the final oil with a less appreciated olfactive quality (see Table III).[33,37]

There exists, of course, a relation between the dimension of the distillation apparatus, the amount of plant material, the weight of the distilled water, the distillation time and the yield of the essential oil. All these figures seldom have been mentioned in one and the same publication.

In an average field-still (with grill) of about 1 M^3 (1,000 l) one charges 300 kg of freshly harvested plant material and 30 kg of water, the distillation time is about 2 hours (total time: charging, heating up, cooling down and discharging is about 4 hours), yielding 1.5-2.0 kg of oil (0.5 to 0.7%).

Physicochemical Properties

The physicochemical properties, such as specific gravity, optical rotation, refractive index and solubility in 80% ethanol of rosemary oils from different origin are shown in Table IV.

One may question what can be learned from the physicochemical properties. Formerly, perhaps until 1960, these properties were often used to verify whether an oil was really natural, reconstituted or even compounded.

Nowadays it is possible to reconstitute or compound an oil with the desired physicochemical properties. With modern techniques, however, it is easy to observe whether a certain oil is compounded[40] (see **Olfactive Properties:** reconstitution).

Generally, the following remarks can be made with respect to the physicochemical properties. The specific gravity is lower for an oil with more monoterpenes. The optical rotation, which may vary between $-8°$ and $+25°$, mostly depends on the rotation of α-pinene, camphene and camphor; a higher 1,8-cineole content decreases the optical rotation. The refractive index is roughly a function of the total monoterpene content and their oxygen derivatives; a higher monoterpene content gives a higher refractive index; with the content of the oxygen derivatives the opposite holds true. The solubility in 80% ethanol depends principally on the total amount of hydrocarbons (aliphatics, mono- and sesquiterpenes), the higher this amount, the worse is the solubility.[33]

The chemical properties, like ester, alcohol and acid numbers are depicted in Table IV.

The acid number generally is below 3 (less than 3 mg KOH—about 0.05 maeq.—per g oil). This means for a monoterpene acid (MW ca 170) approximately 1% free acid. However, so far nobody has investigated the identities of the free acids in rosemary oil (see **Chemical Composition**).

The alcohol number may vary a lot, e.g., from 5 to 50; this corresponds with 2%-20% monoterpene alcohols, often calculated as borneol. Although borneol is the most important monoterpene alcohol in rosemary oil, other representatives of this group also play a role.

The ester number mostly corresponds with the amount of monoterpenyl acetates, i.e., bornyl acetate, for which concentrations of 1%-10% (exceptionally 18%) have been observed.

Chemical Composition

The chemical composition of an average oil of rosemary looks rather simple, but may be complicated. If, in the analysis, one goes down to 1 promille, one can easily detect 50 constituents; down to 1 part per million (ppm) about 100 components can be detected, and on a 1 part per billion level (ppb), circa 200 constituents can be found.

The chemical composition of oils of rosemary can differ a lot. These variations may be due to:[32,33,47,51]

—(sub) species of Rosmarinus
—infraspecific hybridization
—climatological environments
—parts and age of the plants
—time of harvesting
—isolation methods

In Table V the approximate number of components and percentages of the different groups of compounds are shown. The different groups of compounds will be discussed in more detail.

Aliphatic Compounds

The very volatile part of rosemary oil contains lower aliphatic compounds, from which 2- and 3-methyl-butanal are the most important ones; their content, however, is normally below 0.1%.

The most important aliphatic compounds in rosemary oil are octanone-3, 3-octanol and 1-octen-3-ol; the content of these compounds may be up to 0.5%.[40,52,53]

Table V.

Type of Compound	Approximate No. of Components	Approximate Percentage
Aliphatic compounds	10-20	0.5-1
Monoterpenes	20-25	40-55
1,8-cineole	1	20-50
Monoterpene alcohols	5-10	5-10
Monoterpene acetates	2-5	2-12
Monoterpene carbonyls	5-10	5-20
Sesquiterpenes	15-20	2-6
Sesquiterpene oxygen derivatives	5-10	1-2
Benzenoid compounds	5-10	0.5-1
Trace components	5-10	0.5

Table VI. Quantitative Analyses of Spanish Rosemary Oils

	Barranco et al.(33)	Dragoco(55)	Schwenker & Kloehn(56)	Rhyu(79)	Formacek & Kubeczka(82)
Monoterpenes	46.1(a)	47.3	47.2	47.1	44.4
tricyclene	-	-	-	-	0.4(d)
alpha-thujene	0.6	-	-	-	} 21.6(e)
alpha-pinene	24.5	26.0	22.7	24.5	
camphene	10.0	9.0	8.3	9.9	7.7
beta-pinene	} 7.2	0.5	5.8	0.7	3.7
sabinene		-	-	+	0.1
delta-3-carene	-	-	-	-	0.1
myrcene	1.1	5.0	3.0	5.1	2.1
cis- and trans-ocimene	-	1.3	-	-	-
alpha-phellandrene	0.3	-	-	0.1	0.2
alpha-terpinene	+	-	-	0.5	0.2
limonene	+(a)	3.0	3.5	4.3	5.3
beta-phellandrene	-	-	-	-	+(f)
gamma-terpinene	0.7	-	-	0.2	0.4
para-cymene	1.1	2.0	3.9	2.7	2.5
terpinolene	0.6	0.5	-	0.1	0.5
1,8-Cineole	19.8(a)	21.0	30.1	19.5	22.8(f)
Monoterpene Alcohols	5.5	2.2	6.6	16.8	4.6
borneol	3.1	1.2	2.5	-	1.4
alpha-terpineol	1.1	-	1.2	1.8	0.9
delta-terpineol	-	-	-	-	-
terpinen-4-ol	0.8	-	2.1(b)	0.9	1.4
linalool	0.5	1.0	0.8	14.1(x)	0.9
Bornyl acetate	3.6	3.3	+(c)	-	0.5
Camphor	20.0	13.0	13.2(c)	0.1(x)	21.6
Verbenone	1.2	1.5	0.6	-	-
Thujone	-	-	-	0.1	-
Sesquiterpenes	1.8	0.1	1.3	0.9	-
caryophyllene	1.8	-	1.3	0.9	-
alpha-humulene	-	-	+(b)	-	-
alpha-bisabolene	-	0.1	-	-	-

(a) limonene + 1,8-cineole
(b) terpinen-4-ol + alpha-phellandrene
(c) bornyl acetate + camphor
(d) average of 2 analyses
(e) alpha-thujene + alpha-pinene
(f) beta-phellandrene + 1,8-cineole
(x) possibly linalool and camphor confused

Monoterpenes

So far about twenty-two monoterpenes have been identified in rosemary oil, from which α-pinene and camphene are dominant. The monoterpenes may be important precursors for olfactive interesting compounds (see **Biogenesis** and **Chemical Formation**).

1,8-Cineole

The content of 1,8-cineole in rosemary oil can vary from circa 20% to about 50%. In Spanish, Yugoslavian and some Italian oils, this content is rather low, whereas in French, Tunisian and Greek oils it is high (see Tables VI-VIII).

Monoterpene Alcohols

Borneol, which can vary in content from a few percent up to 20%, is the dominant of the monoterpene alcohols. Other monoterpene alcohols, like linalool and terpineols, may also play a role in the olfactive quality of the oil.

Monoterpene Acetates

Bornyl acetate is the most important monoter-

Table VII. Quantitative Analyses of French Rosemary Oils

	Lawrence(40)	Schwenker & Kloehn(56)	Lamparsky(g) & Schenk(59)	Rhyu(79)	Granger et al.(32)
Monoterpenes	24.7	27.8	29.8	35.3	32.5
tricyclene	-	-	+	-	-
alpha-thujene	+	-	+	-	-
alpha-pinene	12.5	10.4	11.4	17.2	12.5
camphene	4.0	4.2	4.5	6.8	8.0
beta-pinene	1.3	7.6	7.8	0.6	4.0
sabinene	-	-	+	+	-
delta-3-carene	} 1.3(a)	-	+	-	-
myrcene		1.4	1.2	2.6	4.0
cis- and trans-ocimene	-	-	+	-	-
alpha-phellandrene	-	-	+	0.1	-
alpha-terpinene	0.4	-	-	0.7	-
limonene	3.0	2.1	2.9	3.4	2.0
beta-phellandrene	-	-	-	-	-
gamma-terpinene	0.4	-	+	1.1	-
para-cymene	1.8	2.1	2.0	2.7	2.0
terpinolene	-	-	+	0.1	-
alpha-p.dimethylstyrene	-	-	+	-	-
1,8-Cineole	47.0	49.2	41.0	20.5	18.5
Monoterpene Alcohols	6.2	7.2	5.8	21.5(i)	6.3
borneol	4.0(b)	3.1	} 5.8(h)	-	6.3
alpha-terpineol	+(b)	1.6		3.0	-
delta-terpineol	0.9	1.6(e)	+	-	-
terpinen-4-ol	1.3	-	-	1.2	-
linalool	+(c)	0.9	-	17.3(i)	-
Bornyl acetate	0.9	+(f)	1.3	-	-
Camphor	10.7	13.2	10.4	0.1(i)	25.0
Verbenone	+	+	-	-	5.5
Carvone	0.4(d)	-	-	-	-
Thujone	-	-	-	0.3	-
Sesquiterpenes	8.8	2.9	5.2	1.8	5.0
caryophyllene	4.9	2.9	4.8	1.7	4.0
alpha-humulene	+(b)	+(e)	+	-	1.0
copaene	0.6	-	0.15	0.1	-
gamma-muurolene	3.1	-	0.15	-	-
beta-bisabolene	+(d)	-	0.04	-	-
others	+	-	0.06	-	-

(a) delta-3-carene + myrcene
(b) borneol + alpha-terpineol
(c) linalool + camphor
(d) carvone + beta-bisabolene
(e) delta-terpineol + alpha-humulene
(f) bornyl acetate + camphor
(g) possibly French oil
(h) borneol + alpha-terpineol
(i) linalool + camphor

pene actetate. Small amounts of other acetates, like isobornyl and linalyl, have also been detected.

It has been suggested that bornyl valerate is also present in Spanish rosemary oil;[3] however, this has not been proven in more recent analysis.[40,52,53]

Monoterpene Carbonyls

The monoterpene carbonyls, i.e. ketones, are very important for the olfactive properties of rosemary oil (see **Olfactive Properties**). In general oil of rosemary is a typical monoterpene carbonyl-containing oil. Camphor is the most dominant compound which has been detected in

Table VIII. Quantitative Analyses of Rosemary Oils from Different Origin

	Italy Falchi Delitala et al.(26)	Yugoslavia Grzunov et al.(39)	Yugoslavia Koedam(39)	Greece Skrubis(57)	Egypt Karawya(68)
Monoterpenes	53.6	37.6	43.6	14.7	60.3
tricyclene	-	-	+	-	-
alpha-thujene	-	-	+	-	-
alpha-pinene	32.3	22.4	26.5	10.3	13.7
camphene	8.9	4.7	8.2	4.0	19.2
beta-pinene	3.8	3.7	2.0	-	4.0
sabinene	-	-	+	-	6.4
delta-3-carene	-	-	-	-	-
myrcene	4.7	3.4	1.3	-	10.2
cis- and trans-ocimene	-	-	-	-	-
alpha-phellandrene	-	-	0.1	-	-
alpha-terpinene	-	-	0.3	-	-
limonene	3.9	2.8	2.2	0.4	-
beta-phellandrene	-	-	-	-	4.5
gamma-terpinene	-	0.7	0.6	-	-
para-cymene	+(a)	1.9	1.8	-	2.3
terpinolene	-	-	0.3	-	-
1,8-Cineole	17.0	39.2	28.3	51.2	19.2
Monoterpene Alcohols	10.5	12.8	9.0	5.1	6.5
borneol	4.4	12.8(b)	4.5	4.7	5.4
alpha-terpineol	-	-	2.3	-	-
delta-terpineol	-	-	2.1	-	-
terpinen-4-ol	5.2	-	0.1	-	-
linalool	0.9	+(c)	-	0.4	1.0
geraniol	-	-	-	-	0.1
Bornyl acetate	11.8	7.3(d)	2.4	-	2.0
Linalyl acetate	-	0.2	-	-	1.4
Fenchyl acetate	-	0.1 -	-	-	
Camphor	5.4	12.3(c)	11.6	5.5	3.2
Verbenone	-	-	-	-	-
Sesquiterpenes					
caryophyllene	-	-	3.4	-	-
alpha-humulene	-	-	0.5	-	-

(a) para-cymene and 1,8-cineole
(b) borneol and other alcohols
(c) linalool and camphor
(d) borneol and other esters

amounts from 5% to 25%. Moreover, verbenone, identified in 1% to 2% is an olfactive characteristic compound.

Monoterpene Acids

Up to now, no monoterpene acids have been found in rosemary oil. However, with respect to the acid number (see **Physicochemical Properties**) up to 1% monoterpene acids can be present. It is probable that myrtenic and campheonic acid are present in the oil; these compounds can be derived from α-pinene and camphene respectively.

Sesquiterpenes

Up to thirty sesquiterpenoids, mainly sesquiterpenes, have been detected in rosemary oil up to now.[8,40,59] Caryophyllene and humulene are the most important sesquiterpenes in the oil. All sesquiterpenes may be precursors for important oxygenated products (see below).

Sesquiterpene Oxygen Derivatives

So far, a few sesquiterpene oxygen derivatives, namely caryophyllene and humulene epoxides, have been detected in rosemary oil. It is probable, however, that more olfactive interesting sesquiterpenoids, like alcohols and carbonyls are present in the oil.[8]

Benzenoids

Substituted benzenoids, like anethol, methyl eugenol, safrol, carvacrol and thymol, have been observed as trace components in rosemary oil.[40,53,59] It is surprising that no aryl ketones derived from the unsaturated substituted benzenoids have been found in rosemary oil.

Trace Components

Several groups of trace components are of interest for the olfactive quality of rosemary oil. These groups may be lower aliphatic compounds, monoterpenyl acids, sesquiterpenoid alcohols and carbonyls, aryl ketones and nitrogen derivatives.

One can isolate several nitrogen-containing compounds from rosemary oil below the part per million level, i.e., substituted pyridins, pyrazins and thiazoles. The total concentration of these constituents is mostly below 1 ppm. However, because these compounds can have very low threshold values, they may play a role in the overall olfactive properties of the oil.

Analysis of Rosemary Oil

A quantitative analysis of rosemary oil from Spanish origin is given together with other analyses of oil from different origin (see Table VI-VIII). The chemical composition, qualitatively, of rosemary oil according to literature investigations follows.

A survey of the literature reveals that the oil of rosemary has been the subject of considerable study. A summary of the reference works can be seen below.

Monoterpenes (22)

alpha-pinene 19, 20, 26, 32, 33, 39, 40, 51-59, 67-82
camphene 19, 26, 33, 39, 40, 51-59, 67-82
myrcene 26, 33, 39, 40, 51-56, 59, 67-82
beta-pinene 26, 33, 39, 40, 51-56, 59, 67-82
alpha-phellandrene 33, 52-54, 59, 75, 77, 79, 82
alpha-terpinene 33, 40, 51-54, 59, 75-82
para-cymene 26, 33, 39, 40, 51-56, 59, 75-82
limonene 21, 26, 51-59, 67-82
gamma-terpinene 33, 39, 51-54, 75, 77, 78, 79, 81, 82
terpinolene 33, 51-55, 59, 75, 77, 81, 82
cis/trans-ocimene 40, 55, 59
delta-3-carene 40, 52, 53, 54, 59, 75, 77, 81, 82
delta-4-carene 52, 53, 75, 77
sabinene 40, 52, 53, 59, 75, 77, 82
alpha-thujene 33, 40, 52, 53, 59, 75, 77, 82
alpha-para-dimethylstyrene 40, 59
bornylene 52, 53, 75, 77, 82
alpha-fenchene 52, 53, 59, 75, 77
beta-fenchene 52, 53, 75, 77
tricyclene 52, 53, 59, 75, 77, 82
santene 52, 53, 75, 77
beta-phellandrene 52, 53, 54, 68, 75, 77, 82

Sesquiterpenes (23)

caryophyllene 22, 33, 40, 51, 52, 53, 56, 59, 75, 77, 79, 80, 81
isocaryophyllene 59
alpha-humulene 40, 52, 53, 75, 77
cis-alpha-bisabolene 40, 55
alpha-copaene 40, 59, 79
gamma-muurolene 40, 59
beta-bisabolene 40, 59
ledene 40
gamma-cadinene 40, 59
delta-cadinene 40, 59
alpha-selinene 40
alpha-cubebene 40, 59
calacorene 40, 59
alpha-corocalene 40
cadalene 40, 59
ar-curcumene 40, 59
alpha-ylangene 59
alpha-muurolene 59
trans-beta-farnesene 59
sesquiphellandrene 59
cadina-1(6)-4-diene 59
beta-maaliene 59
calamene 40, 59

Terpenoid Ether/Oxides (3)

1,8-cineole 21, 26, 33, 39, 40, 51-59, 67-82
caryophyllene epoxide 40
alpha-humulene epoxides (2 isomers) 40

Monoterpene Alcohols

borneol 19, 20, 26, 33, 40, 51-59, 62-82
isoborneol 52, 53, 75, 77, 78, 82
alpha-terpineol 26, 33, 51, 52, 53, 59, 75, 77, 79-82
cis-beta-terpineol 59
delta-terpineol 40, 52, 53, 59, 75, 77
terpinen-4-ol 26, 33, 40, 51-53, 59, 75-82
linalool 26, 33, 39, 40, 51-53, 57, 59, 68, 69, 75, 76, 81, 82
geraniol 68, 80
para-cymen-8-ol 40, 59
alpha-fenchol 40, 52, 53, 75, 77
trans-sabinenehydrate 59
cis-sabinenehydrate 59
trans-p-menth-2-en-1-ol 59
cis-p-menth-2-en-1-ol 59
p-menth-1-en-4-ol 59
p-menth-1(7)-en-4-ol 59
alpha-phellandren-8-ol 59
beta-phellandren-8-ol 59
sabinol 59
cis-piperitol 59

Monoterpenoid Esters (6)

bornyl acetate 19, 20, 26, 33, 51-56, 64-82
bornyl valerate 3
isobornyl acetate 39, 78
linalyl acetate 39, 57, 68
alpha-terpinyl acetate 39
alpha-fenchyl acetate 39

Monoterpene Ketones (7)

camphor 17, 18, 26, 32, 33, 39, 40, 51-82
alpha-fenchone 52, 53, 75, 77
verbenone 32, 33, 40, 51-53, 73, 75, 77
verbanone 55
carvone 40
alpha-thujone 52, 79
beta-thujone 52, 79

Aliphatic Compounds (6)

3-hexanone 40
octanol 78
3-octanol 40
1-octen-3-ol 40
3-octanone 52, 53, 75, 77

Table IX.

Origin No. of Analyses	Alpha-Pinene	Verbenone	Camphor	Borneol	Eucalyptol
France (17)	12.5	5.5	25	6.3	18.5
Italy (3)	10	trace	11	6.3	44
Morocco (1)	12	trace	15	5	40
Tunisia (3)	11	2	10	7.3	48
Spain (6)	21	3	17.5	3	23
Portugal (1)	12	trace	9	trace	14
Algeria (1)	26	22	8	4	trace
Corsica (3)	24	27	3	2	6
Yugoslavia (1)	22	trace	13	7	32
Greece (1)	23	trace	7	trace	28

Benzenoids (13)

phenol 59
o-cresol 59
m-cresol 59
p-cresol 59
p-vinylphenol 59
carvacrol 40, 59, 79
thymol 40, 59, 79
eugenol 59
methyl eugenol 40, 59
trans-anethol 40
chavicol 59
methyl chavicol 59
safrol 52, 53, 75, 77

Five samples of freshly distilled Spanish rosemary oil were examined using modern analytical techniques. The oils contained up to sixty constituents, GC detectable, from which forty-five were identified, comprising 99% of the oils. A summary of the results obtained from this study can be seen below.

Aliphatic Compounds (less than 1%)

C_5-aldehydes
3-octanone
1-octen-3-ol

Monoterpenes (45-50%)

alpha-pinene (20-25%)
camphene (8-10%)
beta-pinene (3-7%)
myrcene (3-5%)
limonene (3-5%)
others (3-5%):
- alpha-thujene
- sabinene
- alpha-phellandrene
- cis/trans-ocimene
- alpha-terpinene
- gamma-terpinene
- terpinolene
- para-cymene
- alpha-para-dimethylstyrene

Sesquiterpenes: (2-4%)

caryophyllene (2-3%)
alpha-humulene (less than 1%)
alpha-bisabolene (less than 0.5%)
others (less than 0.5%):
- copaene
- gamma-muurolene
- delta-cadinene
- caryophyllene epoxide

1,8-Cineole(15-20%)

Monoterpenoid Alcohols (4-6%)

borneol (1-3%)
isoborneol (less than 0.1%)
linalool (1-2%)
nerol (less than 0.1%)
geraniol (less than 0.1%)
alpha-terpineol (1-2%)
delta-terpineol (0.5-1%)
terpinen-4-ol (1-2%)

Monoterpenoid Esters (1-3%)

bornyl acetate (1-2%)
isobornyl acetate (less than 0.1%)
linalyl acetate (less than 1%)

Benzenoids (less than 1%)

eugenol
methyl-eugenol
carvacrol
thymol

Monoterpenoid Carbonyls (18-23%)

camphor (15-20%)
verbenone (2-6%)
others (1-2%):
- fenchone
- alpha- and beta thujone
- carvone
- isopinocamphone

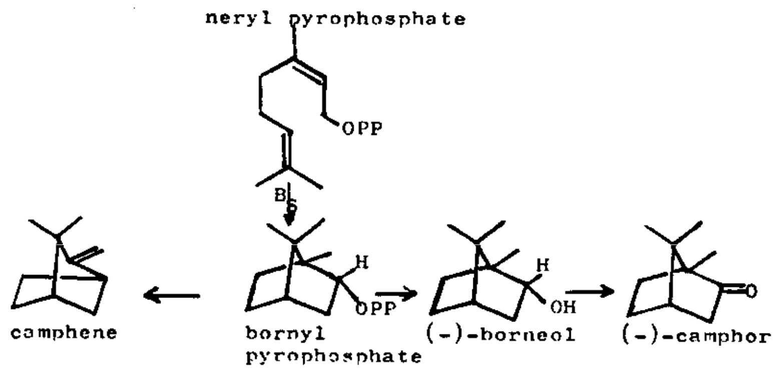

Figure 1

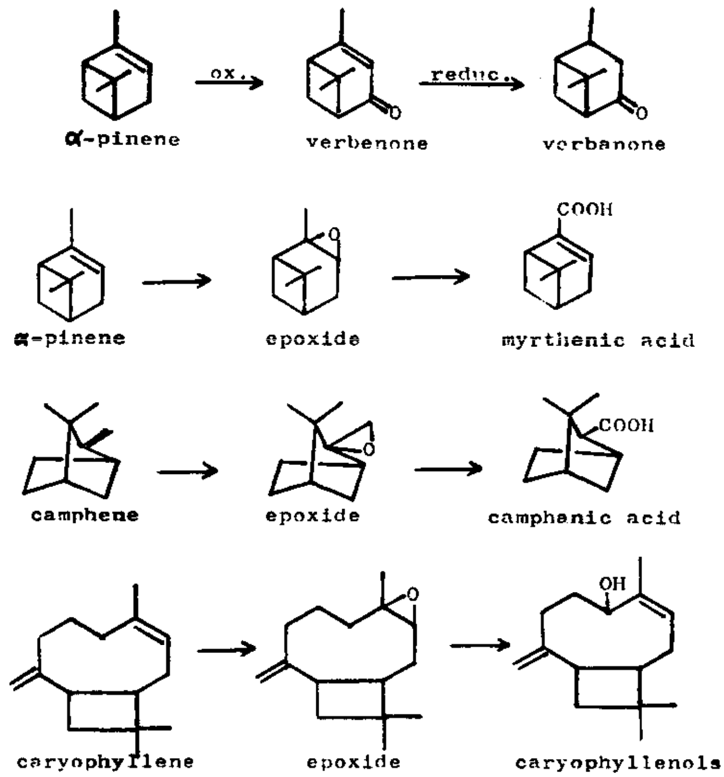

Figure 2

Granger et al. mentioned three principal types of rosemary oils, namely, eucalyptol type (Italy, Morocco, Tunisia), camphor/borneol type (Spain) and alpha-pinene/verbenone type (Algeria, Corsica).[32] The results of their analyses are shown in Table IX.

From the above data may be concluded that at least two types of rosemary oils exist, e.g., one with a high cineole-content (from France, Greece, Tunisia and parts of Italy), and one with a low cineole-content (from Spain, Yugoslavia and other parts of Italy).

Biogenesis and Chemical Formation

One or two biosynthetic pathways have to take place for the formation-sequence of α-pinene/camphene—cineole, borneol (and acetate)—camphor/verbenone in rosemary plants.

Lawrence has postulated a biosynthetic scheme for the formation of camphene, borneol and camphor from nerylpyrophosphate via bornylpyrophosphate (see figure 1).[83]

Genetic Interpretation

B_S (bornyl pyrophosphate synthetase) is a substrate specific synthetase capable of forming bornyl pyrophosphate from neryl pyrophosphate.

Because the essential oil is easily released from the hairy oil glands on the rosemary leaves, the oil is practically in direct contact with the open air (oxygen, sunshine and temperatures up to 40°C). Therefore, it is probable that normal chemical reactions, for example, photochemical oxidations of mono- and sesquiterpenes, occur on plants. Under these circumstances terpenes may be converted via epoxides to carbonyls (aldehydes and ketones) and acids. Some of these reactions are depicted in figure 2.

The leaves of rosemary plants contain, apart from the essential oil, ursolic acid (2%-4%), flavong-lycosides, and tannin (rosemary acid) (see figure 3). It is possible that some of these compounds degrade to volatile products which may occur in the oil.[84]

Olfactive Properties and Application

According to Guenther,[1] in Dalmatia (Yugoslavia) only the leaves of the rosemary shrub are distilled, and that is probably the cause for the finer odour of the Dalmatian oils, as compared with the Spanish oils which are distilled from the aboveground parts, including the woody twigs.

The odour of two oils from France were also much finer than that of Spanish oils.

The total borneol content of Tunisian oils was somewhat higher than that of Spanish, Dalmatian and French oils. The odour of Tunisian oil was very fine.

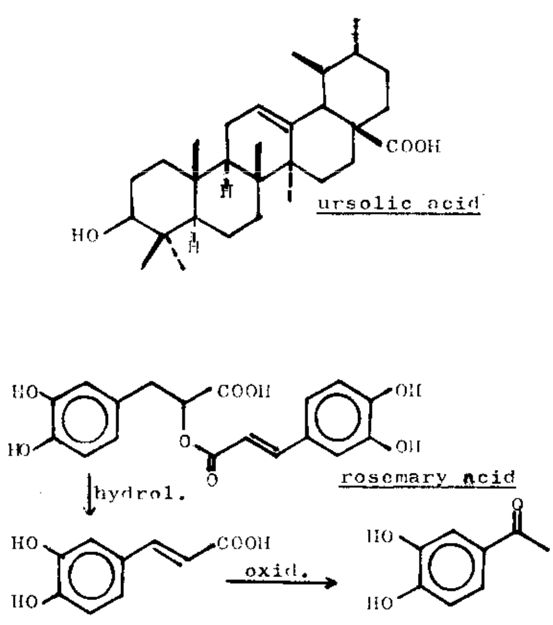

Figure 3

Guenther also mentioned that the oil of rosemary serves in certain medicinal preparations and as an ingredient in rubefacient linaments.

The bulk of rosemary oil, however, is used for the scenting of soaps and technical products, and for the denaturing of alcohol. The oil is useful also in room sprays and inhalants. Because of their harsher note and usually lower price, the Spanish and Moroccan oils lend themselves more advantageously for use in soaps and technical preparations, whereas the usually higher priced Dalmatian oil, with its finer, more delicate odour is preferred for the flavouring of all kinds of food products, such as meats, sausages, soups, and table sauces. The Dalmatian oil thus replaces the dried leaves as condiment. Selected lots of Tunisian and Spanish oils may be used for the same purpose.

Gildemeister and Hoffman published that oil of rosemary is used in medicine as well as in perfumery.[5] In the latter case the oil is especially useful for perfuming of soap and insecticides and, moreover, for the preparation of less expensive perfume compositions. Rosemary oil has an additional benefit: it does not discolour soap.

Garcia Araez described the essential oil of rosemary as a colourless or slightly green-yellow liquid with a camphoraceous odour, penetrating with a sharp and aggressive topnote.[2] He classified the oil between the pastoral essential oils in the second group for its tenacity, very useful for giving body to eau de colognes and toilet waters.

Gil Montero[3] and Poucher[4] mentioned the use and application of rosemary oil in more detail and gave some formulations for perfume compositions.

Lawrence published that the oil of rosemary obtained from the dried leaves of *Rosmarinus officinalis L.* is of great importance to the fragrance industry because of its fresh herbaceous topnotes and its clean, tenacious woody-balsamic dry out.[40] The same author distilled an oil in the laboratory from dried rosemary leaves obtained in the spice trade. As a result the oil obtained by steam distilling these leaves did not necessarily represent a high quality of oil.

Summarizing the olfactive properties it seems worthwhile to give a qualitative odour description of normal oil of rosemary from Spanish origin (see Table X).

Reconstitution of Rosemary Oil

A lot of work has been carried out to reconstitute or compound an essential oil of rosemary from a particular origin by blending other essential oils and/or terpenes (mainly α-pinene), 1,8-cineole, borneol and acetate, camphor and verbenone.[1,5,40] It seems easy to reconstitute the original oil in a chemical way; however, to reproduce the olfactive properties of the natural product is extremely difficult and practically impossible. Moreover, the best reproduction of the oil is often more costly than the natural product.

Several reasons for the failure of the reproduction can be named.

- —analysis is incomplete (some olfactive character-impact components are missing)
- —omission of grace components like volatile aliphatics, nitrogen derivatives and acidic constituents
- —presence of thermolabile components, difficult to analyse, as precursors for olfactive interesting compounds, like mono- and sesquiterpene epoxides
- —used raw materials are contaminated or optically impure
- —the influence of (unknown) sesquiterpenoids on the overall odour character
- —influence of substituted benzenoids and sesquiterpenoids on the tenacity (longlastingness) of the complete oil.

Terpeneless Rosemary Oil

Rosemary plants are cultivated and the young twigs with leaves are harvested, from which the leaves only are steam distilled in stainless steel apparatus. The obtained oil is chromatographed on specially prepared columns at low temperature to remove the hydrocarbons (mono- and sesquiterpenes). The final oil has a very delicate odour, with a fresh herbal, floral top and middle note, and a hay-like, balsamic-woody dry out.

This oil performs excellently in alcoholic and cosmetic perfumery. Moreover, it rounds off and blends every perfume compound for detergents and soaps, which it does not discolour, and has a good tenacity.

Table X. Qualitative Odour Description of Rosemary Oil of Spanish Origin

Odour Quality	Possible Cause
Topnote	
fresh, ethereal	lower aliphatics
harsh	monoterpenes
fruity	higher aliphatics (ketone)
Middle Note	
green	trace components
fresh, minty, eucalyptol	1,8-cineole
coniferous	borneol (and acetate)
camphoraceous	camphor
herbal	verbenone
Bottom Note	
sweet aromatic	subst. benzenoids
hay-like	idem and other carbonyls
Dry Out	
woody-balsamic	sesquiterpenoids

Production and Trade

The production of rosemary oil in 1933 in different countries was published[85] as follows: Spain 70,000-100,000 kg, Dalmatia (Yugoslavia) 10,000-15,000 kg, France 5,000-10,000 kg, and Italy 1,500 kg.

Acording to Schimmel[86] and Fatter[87] the production of rosemary oil in Tunisia decreased from 45,000 kg in 1937 to 2,000 kg in 1943, but increased again to about 20,000 kg in 1948.

Girard in 1947 published the status of the question of concretes and absolutes, including rosemary oil.[88]

Trabaud discussed the world production of rosemary oil and its characteristics.[82]

In Spain, before the civil war (1931-1935), the production of rosemary oil was estimated at 60,000 to 150,000 kg annual. During this war the production sank to about 10,000, however, during the second world war the production increased again to circa 175,000 kg,[1,90] Guenther mentioned that the average annual production in Spain during the forties was about 80,000 kg.[91]

In 1970, the production of rosemary oil in Spain was about 150,000 kg, namely circa 59,000 kg Granadino quality and circa 91,000 kg for Levante quality.[92]

According to more recent publications,[93] the United States imported between 90-100 tons of rosemary oil annually from 1973 to 1980; moreover it imported circa 250 metric tons annually of rosemary plant material.

The annual consumption in the period 1980-1984 for the whole world can be estimated at 400 to 500 tons; in Western Europe 150-200, in the U.S.A. 100-150 and in the rest of the world 100-150 tons are used. The production in Spain is about 200 tons annually.

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