Aromatic Plants of French Polynesia. I. Constituents of the Essential Oils of Rhizomes of Three Zingiberaceae: *Zingiber zerumbet* Smith, *Hedychium coronarium* Koenig and *Etlingera cevuga* Smith

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ABSTRACT: The essential oils from the rhizomes of *Zingiber zerumbet*, *Hedychium coronarium* and *Etlingera cevuga* from Tahiti Island were studied by capillary GC and GC/MS. The oil of *Z. zerumbet* was found to be rich in oxygenated derivatives of α-humulene, in particular zerumbone (65.3%), while *H. coronium* oil contained β-pinene (24.8%) and 1,8-cineole (40.2%) as major constituents. The oil of *E. cevuga* possessed a characteristic chemical composition where methyl eugenol (47.4%) and (Z)- and (E)-methyl isoeugenol (18.8%) together accounted for more than 60% of the oil.

KEY WORD INDEX: Essential oil, *Zingiber zerumbet*, *Hedychium coronarium*, *Etlingera cevuga*, Zingiberaceae, rhizome oils, essential oil composition, 1,8-cineole, zerumbone, methyl eugenol.

INTRODUCTION: While investigating the chemical composition of aromatic medicinal plants of Tahiti (French Polynesia), we first analyzed endemic or pantropical species of the Zingiberaceae family: *Hedychium coronarium* Koenig, which is native to Indo-China and

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recently introduced in Tahiti, *Zingiber zerumbet* Smith and *Etlingera cevuga* Smith, both are wild growing species.

H. coronarium may be found growing wild as a luxuriant green herb which is a meter or more high; smooth, oblong leaves which measure 11-60 x 3-11 cm without stems but with a large ligule at the tip of leaf steath, and alternate along the stem in two rows. In season, at the tip of the leafy stem appears a spike containing many heavily perfumed pure white flowers two or three to each bract, the single stigma-tipped stamen protudes belong the three long, narrow petals, a pair of petal-like staminodes and a broad lip bearing a pale, greenish, heart-shaped spot.

Z. zerumbet is a 0.6-2 m high erect herbaceous, perennial plant; its rhizomes are large, tuberous and pale yellow within; the leaves are distichous, ovate-lanceolate, acuminate, 15-40 cm long and 4-9 cm wide, short-petioled and hairy beneath; each leaf has a prominent ligule which is erect, elongate, obtuse and about 1.5-3.5 cm long; the flowering scape is produced directly from the rhizomes; the peduncle is covered with 4-8 pubescent sheaths which are slightly two-lobed at the apex, reddish at the base; the inflorescence is a spike, ovoid to oblong with an obtuse apex; the bracts are numerous, obovate, imbricate, with mucronate apices, persistent, with a membranaceous margin, greenish and pink-edged when young but red after flowering, and hold a mucilaginous substance; the sessile flower is cream-colored, irregular; the fruit is a capsule, white, glabrous; the seeds are numerous, ellipsoidal, black and covered with white aril.

E. cevuga Smith (syn. *Amomum cevuga*; *Geanthus cevuga* Seeman) is a herbaceous, perennial and rhizomatic plant 2.5-4 m high; shoots foliaceous and floriferous; leaf blades oblong-lanceolate-caudate-acuminate, about 35 cm long, 6 cm wide, glabrous or pubescent along the margin; petiole about 1 cm long; ligule 4-8 mm long; outer bracts numerous, imbricate, red the upper ones, subacute, the lower ones shorter, entire or amarginate; floriferous bracts tubular, calyx-like, bi-lobed, pilose to the outer surface; flowers shortly pedicellate; calyx tubular, with three triangular acute teeth; corolla white to light-pink, the tube as long as the calyx, labellum obovate, emarginate, light pink; ovulary and stigma villose; filament very short; anther 5-7 mm long, villose.

Rhizomes of *Z. zerumbet* such as "Rea moeruru" (Tahitian), "Eka pui" (Marquesian), "Ava pui" (Samoan), are used by traditional healers in many Polynesian medicinal preparations for the treatment of different diseases, for example, carbuncles, diarrhea, otalgia, gonorrhea (1). *H. coronarium*, introduced in Tahiti by Chinese people quite recently, was used by them as medicine for a variety of ailments. The Polynesians love this plant because of the exquisite fragrance of its flowers which they use to make a "hei" (necklace). Leaves of *E. cevuga*, "Opuhi:" tahitian; "Eka pua vao:" marquesian, are used as traditional yellow dyeing material for dyeing vegetal fibers of "More" or "Tapa" (traditional dresses).

Some studies have been reported on volatile components of rhizomes of *H. coronarium* (2-4). The essential oil of the Indian species contains 41% 1,8-cineole, 26% monoterpenic hydrocarbons and 18% β-caryophyllene (2); in the Egyptian species, the presence of methyl eugenol, methyl salicylate and methyl anthranilate (4) is noted. The main components of the *Z. zerumbet* oil have been identified through many studies (5-10): its components are principally oxygenated derivatives of humulene such as zerumbone, which is the major component with a proportion varying between 35% to 59%. A terpinen-4-ol variety has been observed in the Philippines (9). To our knowledge no study concerning the chemical composition of *E. cevuga* has been performed to this date.

EXPERIMENTAL: Rhizomes of *E. cevuga* and *H. coronarium* were collected in June 1991 from the districts of Tiarei and Tautira in Tahiti Island; rhizomes of *Z. zerumbet* were collected in March 1992 from Tiarei. Freshly collected specimen were sliced and steam distilled in a Clevenger-type apparatus (for 6-8 h). The essential oil obtained was dried, after decantation, over anhydrous sodium sulfate.

GC analysis was performed on a Varian 3400 equipped with two fused silica capillary columns (30 m long, 0.32 mm i.d. coated with DB-5 and 30 m long, 0.32 mm i.d., coated with DB-Wax: J&W Scientific, California), programmed from 80-200°C at 2°C/min with a final hold time of 30 min.

GC/MS analysis was performed on a Hewlett-Packard capillary GC-quadrupole MS system (Model 5970) fitted with a 25 m x 0.23 mm i.d. fused silica column, coated with DB-1, programmed from 50-200°C at 5°C/min. Authentic reference compounds, as well as published mass spectra (11,12) and retention indices (13,14) were used as basis for the identification of compounds.

RESULTS AND DISCUSSION: The hydrodistillation of rhizomes of *H. coronarium* gave a pale yellowish mobile oil in about 0.07% (v/w) yield. Rhizomes of *Z. zerumbet* gave a viscous oil in about 0.37% (v/w) yield, while the pale yellow oil of *E. cevuga* was produced in about 0.04% (v/w) yield.

The results of the oil analyses are reported in Table I. The area percentage composition was obtained by peak area normalization without taking into consideration relative response factors.

The qualitative composition of *Z. zerumbet* oil is in accordance with the previously published literature (5,9,10). The high content of zerumbone (>65%) is somewhat similar to that obtained from the Fijian species (10). When the species was cultivated in India (5) or in the Philippines (9), it was characterized by markedly lower amounts of that "α, β-unsaturated ketone" (36-38%) and a correlative higher proportion of α-humulene (17-27%) as compared to 3.5% in our oil.

Concerning the *H. coronarium* specimen, bearing in mind that the genus is native to Indonesia, the chemical composition of its oil is as expected considering its origin (2). The major component was 1,8-cineole (40%), while significant amounts of α-pinene (8%) and β-pinene (25%) were also characterized. The same major compound has been identified in several oil samples from *H. spicatum* var. *acuminatum* (2,15), which contain distinctly higher amounts of linalool. A recent publication relates that linalool accounts for more than 80% of the oil obtained from another species, *H. aurantiacum* (16).

The oil of *E. cevuga* differs notably from the two other oils. The predominance of eugenol derivatives such as: methyl eugenol (47%) and (E)-methyl isoeugenol (18%) is the main difference. Other components, α- and β-pinene (17%), (E)-nerolidol (3%), linalool (3%) and eugenol (2%) are not very significant. The rhizome oil composition is qualitatively similar to that found in stems or leaves of the plant, but with lower amounts of methyl eugenol (17).

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Table I. Percentage composition of essential oils from rhizomes of three Zingiberaceae: *Etlingera cevuga*, *Hedychium coronarium* **and** *Zingiber zerumbet*

Constituent	KI	*Z. zerumbet*	*H. coronarium*	*E. cevuga*	Identification method
2-heptanol	894	-	-	0.5	K,C,MS
α-thujene	928	0.1	0.5	0.1	K,C,MS
α-pinene	937	1.1	7.8	3.9	K,C,MS
camphene	953	4.1	1.0	0.2	K,C,MS
sabinene	976	0.1	1.1	-	K,C,MS
β-pinene	982	0.3	24.8	3.0	K,C,MS
myrcene	990	-	1.6	0.8	K,C,MS
α-phellandrene	1009	0.1	2.5	2.3	K,C,MS
α-terpinene	1019	-	0.8	0.2	K,C,MS
p-cymene	1025	0.4	0.6	0.7	K,C,MS
limonene	1030	0.5	1.0	1.8	K,C,MS
1,8-cineole	1035	1.3	40.2	1.1	K,C,MS
(Z)-β-ocimene	1040	-	-	0.3	K,MS
(E)-β-ocimene	1044	-	0.1	0.6	K,MS
γ-terpinene	1058	-	1.8	0.1	K,C,MS
terpinolene	1091	-	0.7	0.2	K,C,MS
linalool	1099	0.4	0.9	3.1	K,C,MS
camphor	1139	2.2	0.2	0.1	K,MS
borneol	1167	2.3	1.5	0.1	K,C,MS
terpinen-4-ol	1178	0.2	3.0	0.3	K,C,MS
α-terpineol	1192	0.4	5.4	0.7	K,C,MS
2-decanol	1198	-	-	0.3	K,MS
methyl chavicol	1208	-	-	0.3	K,C,MS
n-decanol	1261	-	-	0.6	K,MS
bornyl acetate	1281	0.6	0.2	0.1	K,C,MS
2-undecanone	1299	-	-	0.1	K,MS
α-terpinyl acetate	1349	-	0.5	-	K,C,MS
eugenol	1360	-	-	2.6	K,C,MS
geranyl acetate	1377	-	-	0.4	K,C,MS
methyl eugenol	1415	-	-	47.4	K,C,MS
β-caryophyllene	1422	0.3	0.3	0.1	K,C,MS
α-humulene	1448	0.5	0.5	-	K,C,MS
(Z)-methyl isoeugenol	1456	-	-	0.6	K,MS
dodecanol	1472	-	-	0.5	K,MS
(E,E)-α-farnesene	1485	0.1	0.1	0.1	K,MS
(E)-methyl isoeugenol	1504	-	-	18.2	K,MS
β-bisabolene	1508	0.1	0.2	-	K,C,MS
δ-cadinene	1520	-	0.2	0.2	K,MS
eugenyl acetate	1527	-	-	0.7	K,C,MS
(E)-nerolidol	1560	0.1	0.1	3.0	K,C,MS
spathulenol	1577	-	-	0.2	K,MS
β-caryophyllene oxide	1585	1.2	0.2	0.2	K,MS
humulene epoxide-I	1591	2.9	-	-	MS
humulene epoxide-II	1597	4.4	0.3	0.1	MS

Table I. (cont.)

Constituent	KI	Z. zerumbet	H. coronarium	E. cevuga	Identification method
humulene epoxide-III	1619	2.4	-	-	MS
unknown	1630	-	0.3	0.2	
T-cadinol	1637	0.5	0.1	0.2	K,MS
β-eudesmol	1648	0.7	-	-	K,MS
humulenol-I	1655	0.2	-	-	MS
humulenol-II	1664	0.9	-	-	MS
zerumbone	1731	65.3	-	-	MS
unknown	1799	-	0.5	0.9	
humulene dioxide ?	1985	0.3	-	-	MS

KI = Kovats Index on DB-5 capillary column
K = Comparison of retention data with published data
C = Co-chromatography with authentic samples
MS = Identification based on mass spectral data

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