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Analysis of Essential Oil from Pericarps of *Chimonanthus praecox*

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In this work, the essential oil from pericarps of *Chimonanthus praecox* was extracted by hydrodistillation and analyzed by gas chromatography-mass spectrometry (GC-MS). The essential oil in pericarps of both the wild and cultivated wintersweets were mainly characterized by sesquiterpenoids with more than 60 % and, shared 19 components, which indicated the close genetic relationship between the wild and cultivated wintersweets. At the same time, the outstanding differences of both yields and chemical components of the essential oils between the wild wintersweets from different populations, as well as between the wild and cultivated wintersweets, were observed. Pericarps of the tested wild wintersweets possessed higher essential oil contents and more abundant chemical components than those of the cultivated one, of which, the wild wintersweet at Qinghuazhen was more excellent than that at Taipingzhen, suggesting that partial genetic materials controlling some particular natural products might be varied and lost in the processes of evolution and domestication from wild to cultivated wintersweet and there are, to a certain extent, genetic diversity in wild wintersweets. These results revealed that the oil vacuole tissues of wintersweet flowers might be further developed to the fruits, even kept within the dried pericarps that are developed from the ovary walls, which implied that there are probably abundant oil vacuole cells in the pistil of wintersweet.

Key Words: *Chimonanthus praecox*, Pericarp, Essential oil, Hydrodistillation, Gas chromatography-mass spectrometry.

INTRODUCTION

Chimonanthus praecox (L.) Link, belonging to the Calycanthaceae family, is a deciduous shrub native to China, which has survived from the tertiary relic period[1]. It has been called by various synonyms such as Gol-e Yakh in Iran, Roubai in Japan, Lamei in China and wintersweet in England[2]. It is a famous traditional fragrant flower plant with high ornamental value in China. The flowers, leaves, fruits and roots of wintersweet are also a Chinese folk herbal medicine for the treatment of colds, rheumatoid pains, coughs, asthma and other disorders[2-6].

In recent years, developing the essential oil production of wintersweet has been more and more attended due to the excellent aroma[1]. Some researchers have investigated the essential oils in wintersweet flowers[2,7]. However, less information has been reported on essential oil in pericarps of both wild and cultivated wintersweets. The information on essential oil and its chemical component in wintersweet pericarps is important to better understanding the pericarp aromatic and medical properties and to utilizing it in the fragrant and medicinal industries in addition to probing the developing relationship of oil vacuoles between the pericarps and the flowers, because the fruits are further developed from the flowers. The objectives of this study are to extract the essential oil in pericarps from both wild and cultivated wintersweets with steam distillation and analyze the chemical components with GC-MS and to compare the characteristics of essential oil between them for more effectively researching and developing the special products of wintersweet plants.

EXPERIMENTAL

The wild wintersweet (*Chimonanthus praecox*) plants and flowers are with red or purple speckles on the inside perianths, were randomly selected at the Taipingzhen and Qinghuazhen of Wanyuan, Sichuan, China, respectively. And, the ornamental cultivar Suxinlamei with perfect yellow flowers grown at the Taipingzhen was used for the control. The mature and dried fruits were picked from the plants. The pericarps were separated from the seeds by hands and then pulverized for 0.85 mm using the vegetation disintegrator FW100.

Essential oil extraction: 50 g pericarpic powders were weighed and placed in a clevenger apparatus, then distilled water for 450 mL was infused. The essential oil was extracted by hydrodistillation for 2.5 h as the method in Pharmacopoeia of the People's Republic of China (2005). The essential oil was collected in EP (eppendorf) tubes using micropipettor and dried by anhydrous sodium sulphate and stored in ultra low temperature freezer at -80 ºC until chemically analyzed.

GC-MS analysis: To detect the chemical components of essential oil, GC-MS (gas chromatography-mass spectrometry) analysis was performed on an Agilent 5973N MS system coupled with an HP 6890 gas chromatograph, equipped with fused silica capillary column coated with 5 % phenyl methyl siloxane methyl silicone (HP-5MS), 30 m × 0.25 mm, film thickness 0.25 μm. Analytical conditions were as follows: the carrier gas was helium at 1 mL/min and injection volume of each sample was 1 μL and split ratio was 1:50. The injection port and the detector temperatures were 250 and 280 ºC, respectively. Oven temperature was programmed as follows: the column temperature was programmed from 60 to 130 ºC at a rate of 10 ºC/min, isothermal at 130 ºC for 16 min, then increased to 250 ºC at a rate of 10 ºC/min and subsequently held isothermal for 16 min. Ionization of the sample components was performed in the EI mode (70 eV).

Identification of components: The GC-MS data were processed using the Agilent MSD Productivity ChemStation Software. The individual components were identified by comparing their mass spectra and retention indices with those reported in the literatures or stored in the NIST02 and NIST05 versions of mass spectral database (National Institute of Standards and Technology, Washington, DC, USA), respectively. Retention indices were calculated using retention times of *n*-alkanes that were injected under the same chromatographic conditions as the essential oil analysis.

RESULTS AND DISCUSSION

Yield of the essential oils: Pale yellow essential oil was obtained from both the cultivated and wild wintersweet pericarps by hydrodistillation. The obvious differences of essential oil yield were observed between the different wild wintersweet accessions, and between wild and cultivated plants. The wild wintersweet pericarps at Qinghuazhen produced higher essential oil yield for 0.23 % than that at Taipingzhen for 0.17 %, both of which yielded more essential oil than that of the cultivar Suxinlamei, only for 0.12 % (Table-1).

Chemical components of the essential oils: The differences of chemical components of the essential oils existed between the wild wintersweets of the two analyzed areas, as well as between the wild and cultivated materials (Table-2). Sixty-six and fifty-one peaks were obtained in the total ion chromatogram of chemical components of the essential oils in wild wintersweet pericarps at Qinghuazhen and Taipingzhen, respectively, while 48 peaks of the essential oil in the cultivar Suxinlamei (Fig. 1). A total of 37 chemical components were identified, accounting for 74.05 % of the total volatiles of the wild plant at Qinghuazhen, more than and covered all the 29 components accounting for 67.3 % of that at Taipingzhen, both of which were more than that of cultivar Suxinlamei with 21 components accounting for 73.98 %. Of which, 19 were shared by the wild and cultivated wintersweets (Table-2).

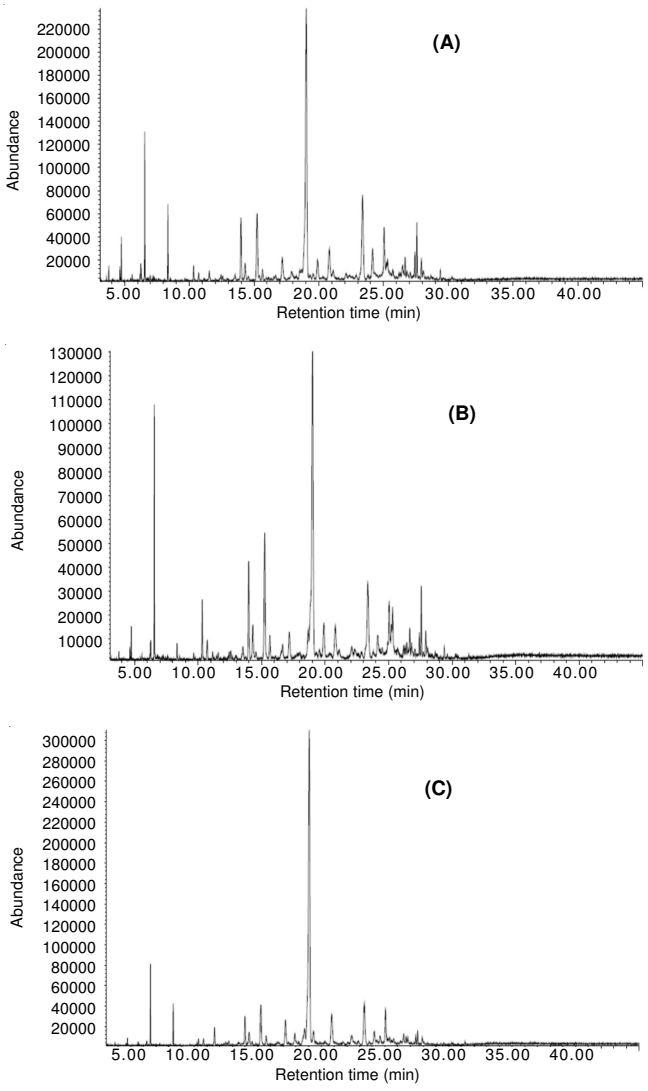

Fig. 1. Total ion chromatogram of chemical components of essential oil in pericarps of the analyzed wintersweets ((A) Wild wintersweet at Qinghuazhen, (B) Suxinlamei, (C) wild wintersweet at Taipingzhen)

TABLE-1
YIELD AND TYPES OF CHEMICAL COMPONENTS OF ESSENTIAL OIL IN PERICARPS OF THE ANALYZED WINTERSWEETS (%)

Essential oil		Suxinlamei	Wild wintersweet	
			Taipingzhen	Qinghuazhen
Yield (%)		0.12	0.17	0.23
Types of chemical components	Monoterpenoids	4.666	3.386	8.213
	Sesquiterpenoids	67.528	60.474	60.468
	Aromatics	Not detected	0.034	0.094
	Aliphatics	1.787	2.991	4.273

TABLE-2
CHEMICAL COMPONENTS AND THEIR CONTENT OF ESSENTIAL OIL IN PERICARPS FROM THE ANALYZED WINTERSWEETS

No.	Retention time (min)	Component	RI	Formula	m.w.	Suxinlamei[a]	Wild wintersweet[a]	
							Qinghuazhen	Taiping zhen
1	3.594	Pinene	930.1	$C_{10}H_{16}$	136	–[b]	0.063	–
2	3.771	Camphene	944.4	$C_{10}H_{16}$	136	0.071	0.256	0.039
3	4.091	Sabinene	970.2	$C_{10}H_{16}$	136	–	0.03	–
4	4.64	Dolcymene	1014.6	$C_{10}H_{14}$	134	0.07	0.27	0.049
5	4.697	Limonene	1019.2	$C_{10}H_{18}O$	154	–	0.049	–
6	4.743	Eucalyptol	1023.1	$C_{10}H_{16}$	136	0.202	0.802	0.118
7	5.577	Linalool	1090.5	$C_{10}H_{18}O$	154	0.128	0.143	0.051
8	6.194	L-pinocarveol	1136.2	$C_{10}H_{16}O$	152	–	0.179	–
9	6.246	Berbenol	1140	$C_{10}H_{16}O$	152	0.126	0.378	0.18
10	6.28	Camphor	1142.4	$C_{10}H_{16}O$	152	0.126	0.261	0.139
11	6.554	Borneol	1162.1	$C_{10}H_{18}O$	154	2.379	3.266	1.991
12	6.783	Benzenemethanol, α,α,4-trimethyl-	1178.6	$C_{10}H_{14}O$	150	–	0.094	0.034
13	6.834	Cryptone	1182.6	$C_9H_{14}O$	138	–	0.101	–
14	6.966	Myrtenol	1193.1	$C_{10}H_{16}O$	152	–	0.204	–
15	7.16	Berbenone	1205	$C_{10}H_{14}O$	150	–	0.09	0.051
16	7.246	trans-Carveol	1210.2	$C_{10}H_{16}O$	152	–	0.098	0.049
17	7.595	D-Carvone	1230.6	$C_{10}H_{14}O$	150	–	0.038	–
18	8.343	Borneol acetate	1275.8	$C_{12}H_{20}O_2$	196	1.564	2.01	0.663
19	8.538	Thymol	1287.4	$C_{10}H_{14}O$	150	–	0.076	0.056
20	10.218	Ylangene	1358.9	$C_{15}H_{24}$	204	0.148	–	–
21	10.332	α-Cubebene	1363.4	$C_{15}H_{24}$	204	–	0.507	0.525
22	10.727	β-Elemene	1379.2	$C_{15}H_{24}$	204	–	0.385	0.326
23	11.538	Santalene	1407.1	$C_{15}H_{24}$	204	–	0.571	0.6
24	12.447	Chamigrene	1429.9	$C_{15}H_{24}$	204	–	0.265	0.262
25	12.573	α-Gurjunene	1433.2	$C_{15}H_{24}$	204	–	0.237	0.203
26	12.727	Humulene	1436.9	$C_{15}H_{24}$	204	0.309	–	–
27	13.984	Eudesma-4(14),11-diene	1468.2	$C_{15}H_{24}$	204	2.216	3.71	2.962
28	14.31	Germacrene D	1476.9	$C_{15}H_{24}$	204	1.293	1.279	1.263
29	14.562	α-Muurolene	1482.9	$C_{15}H_{24}$	204	0.29	0.325	0.316
30	15.236	γ-Cadinene	1499.5	$C_{15}H_{24}$	204	4.744	5.619	5.712
31	17.888	Nerolidol	1540	$C_{15}H_{26}O$	222	1.254	1.124	0.834
32	19.031	Caryophyllene oxide	1557.3	$C_{15}H_{24}O$	220	45.71	28.517	30.826
33	19.905	Carotol	1570.4	$C_{15}H_{26}O$	222	–	1.757	1.27
34	20.82	1,5,5,8-Tetramethyl-12-oxabicyclo[9.1.0] dodeca-3,7-diene #	1584.6	$C_{15}H_{24}O$	220	4.286	3.475	3.546
35	23.38	T-Cadinol	1630.2	$C_{15}H_{26}O$	222	6.035	9.265	8.392
36	24.152	Viridiflorol	1645.1	$C_{15}H_{26}O$	222	1.787	3.03	2.991
37	27.421	Nootkatone	1721.9	$C_{15}H_{22}O$	218	0.504	1.175	1.304
38	27.57	Aristolone	1728.6	$C_{15}H_{22}O$	218	0.739	2.257	2.133
39	27.924	7-Methyl-4-(1-methylethylidene)bicyclo [5.3.1]undec-1-en-8-ol #	1744.5	$C_{15}H_{24}O$	220	–	1.142	–

Retention index (RI) according to C8-C40 *n*-alkanes on the HP-5MS column in parentheses. [a]Relative content (%). [b]Not detected.

The similar preponderant types but the different number and relative content of the components among the analyzed materials were observed (Table-2). The wild wintersweet at Qinghuazhen was with the main components accounting for 61.15 % including caryophyllene oxide (28.52 %), T-cadinol (9.27 %), γ-cadinene (5.62 %), eudesma-4(14), 11-diene (3.71%), 1,5,5,8-tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene# (3.48 %), borneol (3.27 %), viridiflorol (3.03 %), aristolone (2.26 %), borneol acetate (2.01 %). The wild plant at Taipingzhen had the main components for 56.56 % consisting of caryophyllene oxide (30.83 %), T-cadinol (8.38 %), γ-cadinene (5.71 %), 1,5,5,8-tetramethyl-12-oxabicyclo[9.1.0] dodeca-3,7-diene# (3.55 %), viridiflorol (2.99 %), eudesma-4(14),11-diene (2.96 %), aristolone (2.13 %). And, the cultivar Suxinlamei was with these for 65.37 % comprising caryophyllene oxide (45.71 %), T-cadinol (6.04 %), γ-cadinene (4.74 %),1,5,5,8-tetramethyl-12-oxabicyclo[9.1.0]dodeca-3,7-diene# (4.29 %), borneol (2.38 %), eudesma-4(14),11-diene (2.22 %).

The maximum component, caryophyllene oxide of the essential oil, has high value in medicinal properties. It has antitumor activities[8], antispasmodic[9] and antimalarial effects[10] and is able to resist some stubborn bacterials like *Staphyloccocus aureus*[11]. Likewise, T-cadinol, the second preponderant component of the essential oil, has many healing properties like relaxing smooth muscle, treating diarrhea, inhibiting duodenal juices accumulation caused by cholera toxin in mouse[12], having calcium antagonistic effect[13] and bactericidal effect[14] and other bioactivities like killing mite worm[15]. Apparently, the essential oil of wintersweet pericarps possesses high medicinal value and therefore, is worthwile for further research and development.

Types of chemical components of the essential oils: The major chemical components of essential oils in pericarps from both the two wild and cultivated wintersweet were terpenes, which were primarily characterized by sesquiterpenoids, accounting for 60.474, 60.468 and 67.528 %, respectively. It might be associated with the close genetic relationship between them because of the cultivar Suxinlamei with perfect yellow flowers being the mutant from wild wintersweet with purple or red speckles' flowers. These results revealed that the oil vacuole tissues of wintersweet flowers might be further developed to the fruits, even kept within the dried pericarps that are developed from the ovary walls, which implied that there are probably abundant oil vacuole cells in the pistil of wintersweet.

However, some obvious differences existed in between the wild and cultivated wintersweets as well as the wild wintersweets (Table-1). The aromatics existing in the wild wintersweets,were not observed in the cultivar Suxinlamei. And, there were more abundant aliphatics in the wild wintersweets (2.991 and 4.273 %) than in the cultivar Suxinlamei (1.787 %). In addtion, the monoterpenoids, aliphatics and aromatics in the wild winteersweet at Qinghuazhen were 2.4, 1.4 and 2.8 times more than those at Taipingzhen, respectively.

The wild wintersweet pericarps from the two different areas produced higher yield and contained more abundant chemical components of essential oil than the cultivated Suxinlamei ones, though the wild wintersweet lacked of the two components Ylangene and Humulene of cultivated wintersweet. These results suggested that partial genetic materials controlling some particular natural products might be varied and lost in the processes of evolution and domestication from wild to cultivated wintersweet. It is possible to breed more excellent wintersweet cultivars with both higher yield and more abundant chemical components of essential oil in pericarps by the genetic recombination between wild and cultivated wintersweets, which might enjoy the ornamental in winter as well as provide the raw and processed materials for essential oil and medicinal industries in summer.

The yield and chemical components of essential oil between the tested wild wintersweet pericarps were outstandingly different. The wild wintersweet pericarps contained higher yield and more abundant chemical components at Qinghuazhen than that at Taipingzhen, which might be associated with both the genetic and environmental factors. So, it is possible to explore better genetic resources in different wild wintersweet populations.

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