COMPOSITION AND VARIABILITY OF THE ESSENTIAL OIL OF *Houttuynia* OF CHINA

Li Chen,[1] Wei Wu,[2] Chunyan Huang,[2] Yuxia Yang,[2] and You-Liang Zheng[3*]

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Houttuynia Thunb. is one of the important genera in Saururaceae. According to Florae Reipublicae Popularis Sinicae and Flora Sichuanica, *Houttuynia cordata* Thunb. (Yuxingcao in Chinese) is the only species in the genus *Houttuynia* [1, 2]. It is mainly used in Chinese medicine to reduce fever to ease malnutrition, and to clear the body of toxins; it is also used to treat lung carbuncles and has anti-bacterial properties as well. Moreover, it is a delicious wild vegetable in China. Now it has been identified as one of the most potential medical and edible plant genetic resources by the Chinese State Health Department. Recently, Zhu et al. classified a new species *H. emeiensis* Z. Y. Zhu et S. L. Zhang in the genus *Houttuynia* collected at Emei mountain in Sichuan province, China. *H. emeiensis* is locally called white Yu Xingcao, also used as a Chinese medical herb and vegetable [3].

The extract or essential oil of *H. cordata* contains as its main compounds antibacterial, anti-virus, and anti-inflammatory compounds. *H. cordata* essential oil is produced by hydrodistillation of the whole plant of *H. cordata*. Many studies have shown that the different populations of the same species or genus can be divided into different chemotypes based on their essential oil compositions [4–8]. Some chemotypes were found to have different antibacterial and anti-virus activity [9, 10]. The essential oil components of different *H. cordata* populations from different provinces are found to bedifferent [11–15]. Some reports have shown that the chemical compositions of the polyploid are different from the haploid plant [16–18]. Our previous study has also tentatively shown that the essential oil components of populations with a different chromosome number in *Houttuynia* are distinguished using thin layer chromatography [19]. But the essential oil components of these populations with different chromosome numbers are not yet available in detail.

According to Chinese Pharmacopoeia 2005 [20], the whole plant of *H. cordata*, including its aerial stems, leaves, and underground stems, is itself a kind of Chinese medicine. However, little is known about the difference in the essential oil compositions of *Houttuynia* with respect to its fresh aerial parts (including aerial stems and leaves) and underground parts (actually underground stems).

The objectives of the presented research are (i) to study the interspecific and intraspecific chemical polymorphism of *Houttuynia* and to identify the chemotypes of *Houttuynia* in Sichuan and Chongqing province in China (Table 1); (ii) to throw light on the relationship between chemical compositions and cytotypes; (iii) to compare the essential oil compositions of fresh aerial parts of *Houttuynia* with its underground parts.

In total, 21 components could be identified from the essential oils (EO) of the aerial parts of *Houttuynia* of all plant terra in two years, accounting for from 87.29% to 95.83% of the whole volatiles of the aerial parts, which are listed in Table 2. A high level of chemical polymorphism in the EO of the aerial parts of *Houttuynia* was observed in this investigation. Among 21 components, only 14 were found in all the accessions. The other components only occurred in one or more accessions. No significant chemical component differences were detected between the *H. cordata* and *H. emeiensis* accessions.

1) Information & Engineering Technology College, Sichuan Agricultural University, Ya′an, 625014, Sichuan Province, China; 2) Agronomy College, Sichuan Agricultural University, Ya′an, 625014, Sichuan Province, China; 3) Triticeae Research Institute, Sichuan Agricultural University, Ya′an, 625014 Sichuan Province, China, tel: 86 835 2883153, fax: 86 0835 2882620, e-mail: ylzheng@sicau.edu.cn, chenli167541@yahoo.com.cn. Published in Khimiya Prirodnykh Soedinenii, No. 6, pp. 629-632, November-December, 2008. Original article submitted April 4, 2007.

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TABLE 1. The Collection Localities, Chromosome Numbers, and Chemotypes of 18 *Houttuynia* Accessions

Accession Voucher No.	Localities	Chromosome number (2n)	Chemotype
w01-86*	Emei Mountain, Sichuan	36	D
w01-1*	Jioufeng, Leshan, Sichuan	36	D
w01-5	Qingxichang, Qianwei Sichuan	36	D
w01-48	Neijiang, Sichuan	36	D
w01-39	Hanlin, Pengshui, Chongqing	54	D
w01-43	Ma’erkang, Sichuan	54	D
w01-81	Taihusi, Yingjing, Sichuan	72	D
w01-16	Yibin, Sichuan	80	M
w01-80	Huaishu, Mianyang, Sichuan	80	M
w01-6	Jiujing, Qianwei, Sichuan	81	M
w01-71	Mozi, Bazhong, Sichuan	81	M
w01-37	Zongdu, Youyang, Chongqing	82	M
w01-96	Zhongfengsi, Emei, Sichuan	82	M
w01-34	Junling, Xiushan, Chongqing	86	M
w01-98	Helong, Ya’an, Sichuan	86	M
w01-99	Ziyang, Sichuan	88	M
w01-94	Wangyu, Ya’an, Sichuan	90	M
w01-100	Qionglai, Sichuan	90	M

*Represents *H. emeiensis*; D and M represent decanal and myrcene chemotypes, respectively.

On the basis of the GC/MS data of the aerial part essential oil, two chemotypes could be described within the investigated *Houttuynia* accessions by using cluster analysis and principal component analysis (Table 2). Chemotype decanal (chemotype D) was dominated by decanal with high concentrations over 25%, e.g., w01-86 (61.81%) and w01-43 (59.81%). All the accessions classified into chemotype D have chromosome number less than 80. The w01-16, w01-80, w01-71, w01-6, w01-37, w01-96, w01-92, w01-100, w01-98, w01-34, w01-94, and w01-99 with chromosome number over 80 were grouped into chemotype myrcene (chemotype M). Compared with chemotype D, a higher concentration of myrcene (average 18.48%) could be detected in chemotype M. Additionally, chemotype D showed less complex EO matrixes than chemotype M.

All the compounds of chemotypes D and M are presented in Table 3. Chemotype D showed lower amounts of monoterpenes and sequiterpenes, on average 5.72% and 0.16%, varying from 1.12 to17.65% and 0 to 0.38%, respectively. The other nonterpene straight-chain components made up 76.94% to 94.71% of the whole EO components. Of them, the straight-chain aldehydes represented the main chemical structures in chemotype D. In general, chemotype M showed higher amounts of monoterpenes (average 36.16%) and sequiterpenes (1.38%), while the concentration of other straight-chain components was 52.92%, ranging from 37.88% to 64.89%. Furthermore, chemotypes D and M had two subtypes, respectively. In chemotype D, the subtype including only accession w01-48 has a higher concentration of dodecanal and a lower concentration of decanal than the other chemotype D accessions. In chemotype M, the subtype including w01-80, w01-71, w01-6, and w01-37 has a lower concentration of decanal and dodecanal than the other chemotype M accessions.

In total, 16 compounds were characterized from the essential oils (EO) of the underground parts of *Houttuynia* of all plant terra in two years. Of them, all compounds were found in all accessions, accounting to 94.97% of the whole volatiles of the underground parts. No obvious chemical component difference was detected between the underground parts of *H. cordata* and *H. emeiensis* accessions.

On the basis of the GC-MS data of the EO of the underground parts (Table 4), no typical chemotype could be described within the investigated *Houttuynia* accessions with a total of 16 compounds (Table 4). Table 4 showed that w01-86, w01-1, and w01-48 had higher amounts of 2-ethyl-1-decene and *H. emeiensis* separated from *H. cordata* accessions. However, there was no significant chemical difference between the EO of the underground parts of the different *Houttuynia* accessions.

The Euclidean distance values among all the accessions based on the EO components of the underground parts of *Houttuynia* were far less than that based on the EO of the aerial parts. Even *H. emeiensis* seemed separated from *H. cordata* accessions, the Euclidean distance between it and the other accessions in this study being just 0.17, which indicated a closer relationship among all accessions based on the EO data of the underground parts.

TABLE 2. Essential Oil Composition of the Aerial Part of 18 *Houttuynia* Accessions, %

Compound	w01-86	w01-1	w01-5	w01-48	w01-39	w01-43	w01-81	w01-16	w01-80
α-Pinene	0.23	0.42	1.00	0.25	0.08	0.20	1.25	2.95	2.53
Camphene	0.08	0.09	0.09	0.08	0.00	0.03	0.28	0.07	0.58
β-Phellandrene	0.00	0.00	2.46	0.00	0.16	0.00	2.06	4.19	8.67
β-Pinene	0.41	0.67	0.99	0.32	0.16	0.24	1.04	3.35	1.96
Myrcene	0.75	1.68	5.19	2.35	0.50	0.64	10.27	12.19	28.17
Limonene	0.07	0.13	0.12	0.09	0.00	0.02	0.33	0.15	0.76
E-Ocimene	0.00	0.00	0.69	0.00	0.00	0.00	0.65	1.49	1.17
Nonanal	3.36	2.51	0.97	1.75	0.92	1.89	0.50	0.24	0.09
1-Nonanol	0.62	0.86	0.31	0.83	0.75	0.42	0.17	0.15	0.07
4-Terpineol	0.00	0.00	1.12	0.00	0.17	0.00	1.14	1.53	3.18
Decanal	61.81	54.81	49.38	29.88	52.95	59.81	46.36	28.51	13.83
1-Decanal	8.97	15.39	7.42	13.85	18.86	10.90	6.35	3.19	3.02
Bornyl acetate	0.16	0.21	0.15	0.17	0.05	0.18	0.62	0.16	1.21
2-Undecanone	0.23	0.53	4.39	0.07	0.11	0.16	2.94	14.97	7.33
Undecanal	4.99	5.01	4.32	7.10	5.03	6.29	3.14	2.55	1.77
Dodecanal	9.10	9.65	11.57	25.88	14.75	13.35	13.54	5.00	3.30
Caryophyllene	0.00	0.28	0.17	0.27	0.00	0.00	0.38	1.55	2.85
1-Dodecanol	1.06	2.18	0.90	6.31	1.23	1.31	1.82	0.14	0.68
Decanoyl acetaldehyde	0.00	0.00	0.86	0.00	0.00	0.00	0.24	5.52	1.41
2-Tridecanone	0.20	0.45	0.34	0.08	0.07	0.05	0.33	0.34	1.55
2-Ethyl-1-decene	0.17	0.33	1.79	0.09	0.04	0.04	1.56	3.85	4.82

Compound	w01-6	w01-71	w01-37	w01-96	w01-34	w01-98	w01-99	w01-94	w01-100
α-Pinene	1.52	2.21	2.15	1.45	2.63	1.71	2.52	1.60	3.27
Camphene	0.22	0.64	0.28	0.35	0.36	0.26	0.50	0.22	0.12
β-Phellandrene	4.38	4.96	3.85	5.56	3.33	4.26	7.72	6.20	11.95
β-Pinene	1.75	1.51	2.03	1.31	2.35	1.37	2.25	1.47	3.12
Myrcene	9.98	27.39	16.67	11.59	19.76	22.19	21.54	13.43	20.33
Limonene	0.41	0.65	0.58	0.53	0.63	0.53	0.75	0.48	0.17
E-Ocimene	2.15	1.38	3.14	2.56	4.73	2.19	3.99	4.01	5.17
Nonanal	0.31	0.12	0.04	0.27	0.11	0.03	0.06	0.24	0.05
1-Nonanol	0.25	0.12	0.05	0.11	0.05	0.03	0.03	0.03	0.07
4-Terpineol	2.28	1.54	1.91	3.91	0.99	3.63	4.19	2.35	7.27
Decanal	10.73	12.21	8.20	14.38	21.72	19.57	19.96	27.02	16.22
1-Decanal	3.37	3.36	1.46	1.35	3.03	1.90	0.80	3.43	1.14
Bornyl acetate	0.46	1.34	0.93	0.64	1.04	0.81	0.98	0.47	0.42
2-Undecanone	13.91	15.90	20.18	1.59	7.22	3.42	5.33	4.87	6.55
Undecanal	0.44	1.00	0.47	7.36	3.34	0.29	0.15	3.19	0.24
Dodecanal	0.55	0.54	2.52	27.83	13.99	20.28	14.27	19.86	10.89
Caryophyllene	1.70	1.76	1.45	1.07	1.97	1.30	0.54	0.47	0.49
1-Dodecanol	0.00	0.00	0.05	2.65	1.04	1.20	0.30	1.28	0.85
Decanoyl acetaldehyde	14.51	5.85	8.72	0.53	0.69	1.87	1.16	1.51	1.37
2-Tridecanone	1.30	1.13	1.27	1.17	1.30	0.77	0.78	0.58	0.39
2-Ethyl-1-decene	19.54	9.05	12.00	1.08	1.07	2.67	1.26	1.48	1.37

Twelve compounds were characterized both in the EO of the aerial and underground parts of *Houttuynia* accessions, including α-pinene, camphene, β-phellandrene, β-pinene, myrcene, limonene, 4-terpineol, bornyl acetate, 2-undecanone, caryophyllene, decanoyl acetaldehyde, and 2-tridecanone. Nine compounds were found in the EO of the aerial parts, and only four were found in the EO of the underground parts.

TABLE 3. Means Percentage Content of Main EO Constituents in the Aerial Parts of Various *Houttuynia* Chemotypes

Compounds	Chemotypes	
	D	M
Monoterpene hydrocarbons	5.15 (0.90-1.88)	32.41 (20.41-44.14)
Alcohol	0.35 (0.00-1.14)	2.98 (0.99-7.27)
Esters	0.22 (0.05-0.62)	0.77 (0.16-1.34)
Total	5.72 (1.12-17.65)	36.16 (23.15-51.83)
Sesquiterpene hydrocarbons	0.16 (0.00-0.38)	1.38 (0.47-2.85)
Straight chain hydrocarbons	0.58 (0.04-1.79)	5.29 (1.07-19.54)
Alcohol	14.36 (8.33-20.99)	3.20 (1.12-4.75)
Aldehyde	71.52 (63.54-81.34)	30.34 (11.23-50.31)
Ketones	1.42 (0.16-4.73)	10.17 (2.76-21.44)
Total	88.03 (76.94-94.71)	52.92 (37.88-64.89)
Sum total	93.91 (89.37-95.83)	90.45 (87.28-94.20)

TABLE 4. Essential Oil Composition of the Underground Parts of 18 *Houttuynia* Accessions, %

Compound	w01-86	w01-1	w01-5	w01-48	w01-39	w01-43	w01-81	w01-16	w01-80
α-Pinene	8.86	9.49	9.24	13.55	10.98	16.14	11.26	10.79	6.44
Camphene	1.10	1.15	1.34	1.45	0.61	2.28	1.17	1.16	4.31
β-Phellandrene	1.36	1.40	16.40	1.99	12.51	1.84	10.96	7.08	15.42
β-Pinene	16.89	18.84	15.22	25.78	22.65	27.50	21.73	20.04	15.37
Myrcene	6.28	6.55	8.51	7.96	4.79	6.81	8.19	10.94	7.90
Limonene	2.31	2.87	5.81	3.76	2.42	4.18	4.44	5.55	4.52
4-Terpineol	0.50	0.67	2.21	0.85	2.59	0.43	1.11	1.61	1.14
α-Terpineol	0.37	0.59	0.31	0.88	0.35	0.41	1.76	0.34	1.11
Bornyl acetate	1.58	1.96	2.31	1.58	0.31	3.24	2.15	1.94	11.14
2-Undecanone	20.28	24.15	18.86	13.76	21.61	20.08	19.41	25.69	11.75
2-Dodecanone	1.31	1.93	0.83	2.33	2.16	1.48	0.88	0.76	1.03
Caryophyllene	1.44	1.96	0.67	1.61	0.61	0.84	0.58	0.96	3.20
Decanoyl acetaldehyde	5.52	3.15	3.53	2.55	2.48	2.71	1.72	4.20	0.56
2-Tridecanone	2.90	3.67	1.09	2.38	1.77	1.95	1.16	1.13	7.57
2-Ethyl-1-decene	22.99	14.45	7.01	12.38	9.83	7.82	7.63	5.46	0.09
Isoelemicin	0.96	1.43	0.20	1.00	0.39	0.32	0.52	0.21	0.00

Compound	w01-6	w01-71	w01-37	w01-96	w01-34	w01-98	w01-99	w01-94	w01-100
α-Pinene	8.62	9.72	11.61	6.93	12.88	12.62	11.71	10.05	9.59
Camphene	1.07	1.72	1.38	1.05	1.44	1.43	1.27	1.28	1.25
β-Phellandrene	5.70	7.84	8.14	11.41	7.35	9.03	8.74	17.21	13.97
β-Pinene	15.95	16.33	22.41	12.88	24.86	23.45	22.11	17.90	17.54
Myrcene	10.53	12.87	7.74	8.11	8.28	8.70	7.87	8.66	7.02
Limonene	4.63	4.46	7.02	4.51	7.07	5.89	5.81	6.04	5.76
4-Terpineol	1.52	2.12	1.95	3.15	1.45	2.28	1.94	3.15	2.62
α-Terpineol	0.39	0.36	0.46	0.54	0.39	0.50	0.31	0.31	0.36
Bornyl acetate	2.18	3.80	3.01	1.57	2.67	2.35	1.89	1.58	1.91
2-Undecanone	25.99	20.93	22.00	21.07	20.78	19.28	21.30	20.66	22.60
2-Dodecanone	1.01	0.56	0.71	1.49	0.83	0.32	0.27	0.55	0.33
Caryophyllene	1.28	0.92	0.56	0.92	0.51	0.68	0.55	0.48	0.41
Decanoyl acetaldehyde	5.95	3.35	2.55	6.47	3.17	3.35	3.14	2.21	3.67
2-Tridecanone	1.25	0.62	0.51	0.99	0.75	0.73	0.57	0.46	0.66
2-Ethyl-1-decene	7.44	8.29	5.77	9.50	3.93	4.93	5.90	3.54	6.60
Isoelemicin	0.48	0.76	0.23	0.12	0.07	0.24	0.10	0.07	0.17

TABLE 5. Variation Ranges, Mean, Standard Deviations (std.), and Coefficient of Variations (CV) of the Content of Main EO Constituents in the Aerial and Underground Parts of *Houttuynia*, %

Compound	Aerial parts			Underground parts		
	Variation range	Mean (std)	CV, %	Variation range	Mean (std)	CV, %
Monoterpene hydrocarbons	0.90-44.14	21.81±15.46	70.91	36.80-61.88	53.75±7.13	13.26
Alcohol	0.00-7.24	1.96±1.92	98.19	0.84-3.69	2.28±0.80	35.10
Esters	0.05-1.34	0.56±0.41	73.60	0.31-11.14	2.62±2.26	86.24
Total	1.12-51.83	24.32±17.32	71.20	39.24-67.35	58.65±7.95	13.55
Sesquiterpene hydrocarbons	0.00-2.85	0.90±0.83	92.01	0.41-3.20	1.01±0.70	68.89
Straight chain hydrocarbons	0.04-19.54	3.46±5.16	149.13	0.09-22.99	7.98±4.96	62.24
Alcohol	1.12-20.99	7.54±6.57	87.17	-	-	-
Aldehyde	11.23-81.34	46.35±23.85	51.48	0.56-6.47	3.35±1.46	43.66
Ketones	0.16-21.44	6.77±6.50	96.01	18.47-29.74	23.29±2.95	12.67
Total	37.88-94.71	66.57±19.51	29.30	21.00-53.00	34.61±7.44	21.49
Aromaticity total	-	-	-	0.00-1.43	0.40±0.39	97.08
Sum total	87.28-95.83	91.80±2.75	2.99	90.71-98.03	94.67±1.87	1.98

The difference between the chemical components of the EO in the aerial and underground parts of *Houttuynia* was distinct. By comparing the components of the whole volatiles in the aerial and underground parts (Table 5), it can be seen that the EO of the aerial parts of *Houttuynia* had lower amounts of monoterpenes and sequiterpenes, on average 24.32% and 0.90%, in the range 1.12% to 51.83% and 0.00% to 2.85%. It had higher amounts of non-terpene straight-chain components (average 66.57%), especially higher straight-chain aldehydes, which was on the average 46.35%, ranging from11.23% to 81.34%. In contrast, the EO of the underground parts of *Houttuynia* had higher amounts of monoterpenes (average 58.65%), in the range 39.24% to 67.35 %. Of them, monoterpene hydrocarbons occurred at the highest percentage, with mean 53.75%, ranging from 36.80% to 61.88%. Non-terpene straight-chain components in the EO of the underground parts were present in lower amounts (average 35.01%), in the range 21.00% to 53.96%. No straight- chain acids and alcohols were detected, but the percentage of straight-chain ketones (23.29% on average) was higher. The principal compounds in the EO of the aerial parts were myrcene, decanal and dodecanal, and in the EO of the underground parts they were α-pinene, β-pinene, and 2-undecanone.

Almost all the coefficients of variations (CV) of the content of main constituents in the aerial part of *Houttuynia* were much higher than that in underground parts (Table 5), which indicated that the chemical polymorphism of the aerial parts of different *Houttuynia* accessions was much higher, while that of the underground parts was relatively lower.

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