Aromatic Components of the Essential Oils of Four Chinese Medicinal Plants (*Asarum petelotii, Elsholtzia souliei, Eupatorium adenophorum, Micromeria biflora*) in Yunnan

Jingkai Ding, Yu Xuejian, Wu Yu, Zhihui Ding, Zonglian Chen
Kunming Institute of Botany, Academic Sinica, Kunming, China

Nanao Hayashi and Hisasi Komae
Faculty of Integrated Arts and Sciences, Hiroshima University, Kagamiyama 1-7-1, Higashi-Hiroshima 724, Japan

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The volatile components of the essential oils of four Chinese medicinal Plants (*Micromeria biflora, Elsholtzia souliei, Eupatorium adenophorum, Asarum petelotii*) were investigated by GC and GC-MS. Large amounts of terpenes (geranial, naginataketone, *p*-cymene, β-himachalene), or phenol ethers (apiole, elemicin, 1,2,3,4-tetramethoxy-5-allyl-benzene) were identified in the oils.

Introduction

Yunnan in the southern part of China is the biggest treasure-house of plant resources (Zhang *et al.*, 1988) with 15,000 higher plants which are precious, rare or nearly extinct. Most of the plants are resource plants providing food, medicine, and industrial material. Among the resource plants, those most worth mentioning are the medicinal plants. According to statistics, there are over 2000 wild medicinal plant in Yunnan, from which many new medicines have been produced in recent years. In the present study, the essential oils of four medicinal plants (*Micromeria biflora, Elsholtzia souliei, Eupatorium adenophorum, Asarum petelotii*) in Yunnan were analyzed by means of GC and GC-MS. These plants have a characteristic odor and contain a considerable amount of essential oil. The identified components constituted 83.83–98.64% of the oils. These essential oils were found to be rich in monoterpenes (72.14–87.88%) or phenol ether (72.13–79.37%). We have found that these oils contain 11–55 compounds as volatile compounds.

Reprint requests to Prof. N. Hayashi.
Telefax: 0824-24-0758.

Experimental

Isolation and identification of the essential oils

The four medicinal plants were collected in Yunnan province (in the southern part of China) in August and identified by one of the authors (Z. Chen). The voucher specimens are available for inspection at Herbarium of the Kunming Institute of Botany, Academy of Sciences of China. The essential oils of the four plants (*Asarum petelotii* O.C. Sclmuilt, *Elsholtzia souliei* Lévl., *Eupatorium adenophorum* Spreng., *Micromeria biflora* (Buch. Ham. *ex* D. Don) Benth.) were obtained by water distillation. The yields of water-distilled oils were 0.20–0.99% of the fresh samples (49–181 g). *Asarum petelotii* collected at Ping-bian in Yunnan; leaves 119 g, oil 0.23 g (yield 0.20%); roots 153 g, oil 0.59 g (yield 0.39%). *Elsholtzia souliei* collected at Anning in Yunnan; leaves 49 g, oil 0.48 g (yield 0.99%). *Eupatorium adenophorum* collected in Kunming; leaves 212 g, oil 0.74 g (yield 0.35%). *Micromeria biflora* collected at Golden Palace in Kunming; whole plants 181 g, oil 0.55 g (yield 0.30%).

The individual components of the essential oils were identified by means of GC-MS. The data were obtained with a Finnigan 4510 GC-MS instrument using SE-54 silica capillary column

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(30 m×0.25 mm) under the following conditions: column T; 80–200 °C at a rate of 3 °C per min, ion source T; 140 °C, ionization voltage; 25 or 70 eV. The identification of each components in the oils was made by comparing the mass spectra with a collection of literature spectra, or with those of authentic samples, and these was also checked against the retention times on a gas chromatogram.

Results and Discussion

Analytical data

The analytical results of the essential oils of the four medicinal plants examined in this work were as follows:

Asarum petelotii O.C. Sclmuilt (Aristolochiaceae)

Leaves: leaf oil 0.20% yield, α-thujene (0.01%), α-pinene (3.11%), camphene (1.92%), sabinene (0.09%), β-pinene (1.94%), α-phellandrene (0.03%), limonene (0.07%), *cis*-β-ocimene (0.61%), 1,8-cineole (0.11%), *trans*-β-ocimene (0.58%), δ-3-carene (0.01%), terpinolene (0.01%), linalool (0.02%), borneol (2.54%), terpinen-4-ol (0.16%), α-terpineol (0.03%), *iso*-amylcaprylate (0.01%), 2,4,6-tetramethylacetophenone (0.02%), bornyl formate (0.02%), *iso*-bornyl acetate (0.02%), bornyl acetate (0.17%), safrole (0.03%), δ-elemene (0.03%), terpinyl acetate (0.09%), α-copaene (0.01%), α-bergamotene (0.02%), β-elemene (0.06%), α-cedrene (0.03%), δ-selinene (0.05%), β-caryophyllene (0.04%), β-bergamotene (0.26%), α-guaiene (0.20%), α-humulene (0.04%), α-patchoulene (0.06%), cuparene (0.22%), γ-elemene (0.21%), δ-guaiene (0.33%), myristicin (0.77%), elemicin (26.44%, structure I in Fig. 1), 1,2,3,4-tetramethoxy-5-allylbenzene (21.49%), *iso*-elemicin (0.05%), *iso*-apiole (1.88%), *trans*-asarone (0.11%), apiole (28.60%), structure II), unknown (7.50%).

Roots: root oil 0.39% yield, α-pinene (0.05%), camphene (0.02%), sabinene (0.01%), β-pinene (0.19%), α-phellandrene (0.01%), limonene (0.01%), *cis*-β-ocimene (0.13%), 1,8-cineole (0.06%), *trans*-β-ocimene (0.15%), linalool (0.01%), *iso*-butylcaproate (0.01%), borneol (0.03%), *iso*-amylcaprylate (0.03%), bornyl acetate (0.05%), safrole (0.02%), δ-elemene (0.06%),

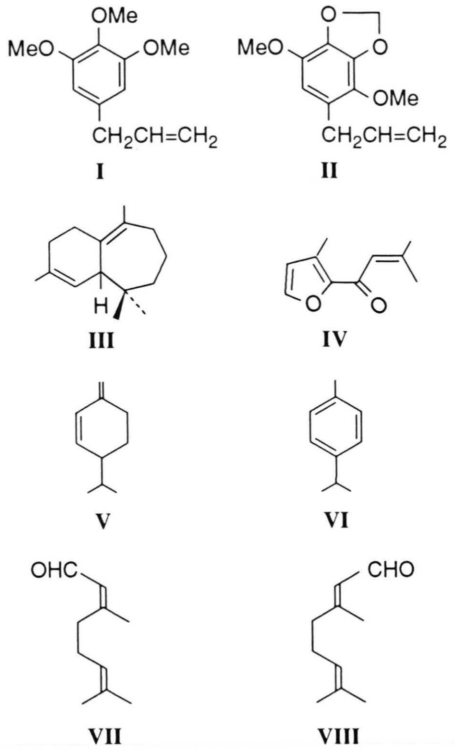

Fig. 1. Structure of main compounds of the essential oils of four medicinal plants.

terpinyl acetate (0.12%), α-copaene (0.04%), α-bergamotene (0.43%), β-elemene (0.63%), α-cedrene (0.04%), δ-selinene (0.11%), β-caryophyllene (0.16%), β-bergamotene (2.51%), *cis*-β-farnesene (1.34%), *trans*-β-farnesene (2.11%), β-himachalene (20.72%, structure III), myristicin (3.90%), elemicin (0.27%), nerolidol (2.34%), palustrol (0.47%), 1,2,3,4-tetramethoxy-5-allylbenzene (15.00%), *iso*-apiole (2.52%), apiole (30.42%), farnesol (0.11%), ethyl palmitate (0.15%), unknown (16.17%).

Elsholtzia souliei Lévl. (Labiatae)

Leaves: leaf oil 0.99% yield, 2-hexen-1-ol (0.08%), 3-methyl-2-butenoic acid (0.07%), pentanoic acid (0.02%), 2-(2-butenyl)-cyclopent-2-en-1-one (21.46%), linalool (1.74%), elsholtzione (4.24%), naginata ketone (66.13%, IV), geranyl acetate (0.03%), bourbonene (0.11%), β-caryophyllene (2.67%), β-cubebene (0.01%), α-humulene (0.31%), γ-muurolene (0.10%), β-berga-

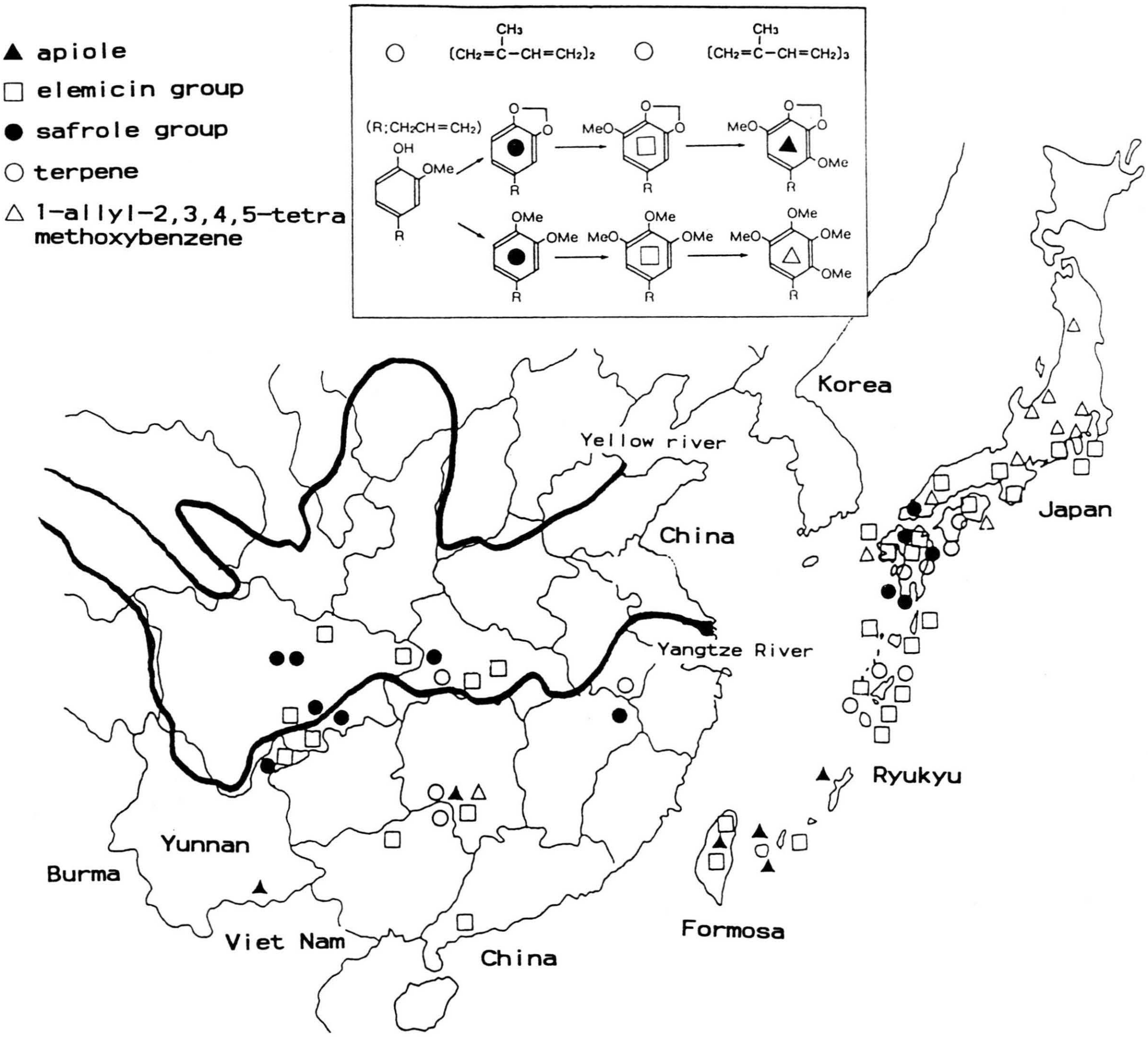

Fig. 2. Geographical distribution of *Asarum* species with phenol ethers and terpenes.

motene (0.30%), γ-elemene (1.37%), unknown (1.36%).

Eupatorium adenophorum Spreng. (Compositae)

Leaves: leaf oil 0.35% yield, α-pinene (1.37%), camphene (17.65%), β-pinene (1.42%), β-phellandrene (20.51%, V), *p*-cymene (30.89%, VI), limonene (2.80%), geraniol (3.44%), bornyl acetate (1.96%), linalyl acetate (0.68%), β-caryophyllene (1.52%), β-bisabolene (6.00%), calarene (2.76%), γ-elemene (0.63%), β-farnesene (6.78%), unknown (1.64%).

Micromeria biflora (Buch. Ham. *ex* D. Don) Benth. (Labiatae)

Whole plant: essential oil 0.30% yield, 6-methyl-5-hept-2-one (0.08%), *cis*-linalool oxide (0.08%), *trans*-linalool oxide (0.07%), linalool (0.27%), neral (37.09%, VII), geranial (50.52%, VIII), nonyl acetate (0.04%), citronellyl acetate (0.12%), β-caryophyllene (0.22%), α-humulene (0.04%), γ-muurolene (0.44%), unknown (11.03%).

In their phytochemical study, Yang *et al.* (1984, 1986) reported the analysis of the volatile components of crude drugs (dry material) of 31 Chinese species in *Asarum*. In the present work, the oil of *A. petelotii* contained phenol ethers in higher amounts than terpenes. The essential oils contained a unique phenol ether, apiole in the leaves (28.6%) and roots (30.4%) together with considerable amounts of elemicin (26.44%) in the leaves and β-himachalene (20.72%) in the roots.

Phytochemically, the presence of apiole in the essential oils of *A. petelotii* is significant in *Asarum*. The components of this species are very similar to that of the *Asarum* (or *Heterotropa*) species (Fig. 2) distributed in Formosa, in China, and in Ryukyu Islands in Japan (Lawrence *et al.,* 1988). The plant can be used to make medicine. *Elsholtzia souliei* contained a high level of naginata ketone (66.13%) as a main component together with small amounts of elsholtzione (4.24%). The plant can be used to make medicine. Fujita *et al.* (1965) reported that the Japanese *Elsholtzia ciliata* Hyl. (obtained from Osaka) contained naginata ketone (58.3–66.1%) as a main component together with elsholtzione (2.0–5.4%). The results showed that the above two species are related phytochemically. On the other hand, *Elsholtzia oldhami* Hemsl. in Formosa contains elsholtzione (80%) as a main component together with a considerable amount of naginata ketone (15%) (Fujita *et al.,* 1964). The leaf oil of *Eupatorium adenophorum* contained a high level of *p*-cymene (30.89%) and β-phellandrene (20.5%) as main components. The whole plant is poisonous for a domestic animal and can be used for an insecticide. The essential oil of the whole plants of *Micromeria biflora* have the strong odor of citral and contained a high quantity of geranial (50.52%) and neral (37.09%).

The whole plant is effective in reducing cold and stomach trouble.

Acknowledgements

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Fujita J. (1958, 1959), The essential oils of the genus of *Elsholtzia*. J. Chem. Soc. Japan **79,** 1067; *ibid.***80,** 1495.

Fujita J. (1964), The essential oil of *Elsholtzia souliei*. The 8th symposium paper of TEAC (Japan), pp. 148–154.

Lawrence B. M., Mookherjee B. D. and Willis B. J. (1988), Development in Food Science **18,** Flavors and Fragrances, pp. 259–282.

Yang C., Zhang J., Pang J., Xu Z., Zhu Q. and Wang G. (1984, 1986), GC-MS analysis of the essential oil of *Asarum*. Zhongyao Tongbao (Bull. Chinese Mat. Med.), 9, 27, 175.

Zhang Qitai, Feng Zhizhou and Yang Zenghong (1988), Rare Flowers and Unusual Trees. The China Esperanto Press, Beijing, pp. 24–27.