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SALICYLALDEHYDE RICH LEAF ESSENTIAL OIL COMPOSITION OF *FILIPUNDULA VESTITA* FROM WESTERN HIMALAYA OF UTTRAKHAND, INDIA

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Abstract. Salicylaldehyde has many applications as an intermediate in chemical industries. Genus *Filipendula* is a potential source of salicycldehyde. Essential oils are prescribed for a variety of health problems by traditional systems of medicine, all over the world. In present study, leaf essential oil composition of *Filipendula vestita* (Wall. Ex G. Don) Maxim. (Family: Rosaceae) from Uttrakhand, India was analyzed using gas chromatography (GC-FID) and gas chromatography-mass spectrometry (GC-MS) methods. The volatile oil was dominated by salicylaldehyde 51.5 %, methyl salicylate 24.5 %, salicylic acid butyl ester 5.70 %, carvone 4.30 %, santene 3.50 %, as major constituents.

Keywords: Filipendula vestita, Rosaceae, essential oil, salicylaldehyde, methyl salicylate.

Classification numbers: 1.4.6.

1. INTRODUCTION

Filipendula genus represented by about twelve species of perennial, herbaceous flowering plants, native to the temperate regions of the Northern Hemisphere. Well-known species include Meadowsweet (*F. ulmaria*) and Dropwort (*F. vulgaris*), both native to Europe, and Queen-of-the-forest (*F. occidentalis*) and Queen-of-the-prairie (*F. rubra*), native to North America, *Filipendula* species are used as food plants by the larvae of some Lepidopteran species, including Emperor moth, gray pug, Grizzled skipper, among others. *F. vestita* (Wallich ex G. Don) Maxim. is an erect leafy perennial, distributed up to 2100-3300 m also reported in Western Himalayan region of Uttrakhand [1, 2].

Previous studies about *Filipendula* species showed that extracts of *F. glaberrima* have been used in traditional medicines of Europe and other countries as anti-inflammatory, analgesic, anti-rheumatic, diuretic, astringent, and diaphoretic agents. Also monotropitin, (+)-catechin, and daucosterol isolated from the flower and fruit oils of *F. glaberrima* [3 - 5]. GC/MS analysis of leaf essential oil of *F. glaberrima* revealed β-farnesol (2.96 %), l-α-terpineol (2.43 %), benzenemethanol (2.87 %), (*Z*)-3-hexen-1-ol (5.23 %), and 2,6-bis (1,1-dimethylethyl)-4-methylphenol (1.91 %) as major constituents. The essential oil from *F. glaberrima* showed a significant toxic effect against early fourth stage larvae of *Aedes aegypti* L, an approach to

reduce the population of mosquitoes would be to target the larvae in South Korea also. Methanolic extracts from *F. glaberrima* (whole parts) showed cosmetic active against matrix metalloproteinase-1. However, little information is available on such biological activity of *F. glaberrima*. Although minimum number of biological activity has been discovered from *F. glaberrima*, its biological activity has not been fully characterized [3-7].

F. ulmaria and *F. hexapetala* other two previously studied species of this genus, which are commonly occur in Poland have been used in folk medicine and phytotherapy for a long time. Due to its anti–inflammatory, anti-pyretic and anti-rheumatic properties, *F. ulmaria* (meadowsweet, queen of the meadow flowers) is mainly used in therapy. The aerial parts of *F. hexapetala* contain *n*-tricosane (17.9 %), the oil was characterized by a high content of salicylic acid derivatives, salicylaldehyde (13.7 %), benzyl salicylate (6.8 %), and methyl salicylate (6.7 %). Besides, *n*-nonanal (11.9 %), 2-heptadecanone (6.2 %), and linalool (5.2 %) were present in significant amounts. The high content of salicylaldehyde (36.0 %) and methyl salicylate (19.0 %) was also found in the essential oil from aerial parts of *Filipendula ulmaria*. Recent report showed that the extract from roots of *F. hexapetala* has interferon like activity. Water-methanol extract from *F. ulmaria* contains a variety of phenolic compounds, such as caffeic, *p*-coumaric and vanillic acid, myricetin, etc, which demonstrate antibacterial activity. The efficacy of *F. ulmaria* extract against selected foodborne psychrotrophic bacteria was also tested using solid laboratory media and low incubation temperatures for better simulation of food preservation conditions. Higher concentrations of the extract, compared to minimum inhibitory concentration determined in the broth, were needed for satisfactory inhibition of spoilage bacteria. Potential use of *F. ulmaria* extract as natural food preservative was also examined against natural spoilage flora and inoculated pathogenic bacteria on fish flesh and fish roe product (tarama salad) [8-13].

2. MATERIAL AND METHODS

2.1. Plant collection, identification and isolation of essential oils

The fresh leaves of *F. vestita* (Wallich ex G. Don) Maxim. were collected from Milam glacier (latitude 30.48° N, longitude 80.10° E and an altitude of 3400 m) in Uttrakhand, India. The plant material was authenticated from Botanical Survey of India, Dehradun. The voucher specimen (Chem./DST/02) has been deposited in the Phytochemistry laboratory of the Chemistry Department, Kumaun University, Nainital. Fresh leaves (1 kg) were subjected to steam distillation using a copper electric still, fitted with spiral glass condensers for three hours. The distillates were saturated with NaCl and extracted with *n*-hexane and dichloromethane. The organic phase was dried over anhydrous sodium sulfate and the solvents were distilled off in a rotary vacuum evaporator at 30 ^{o}C and the percentage oil content was calculated on the basis of fresh weight of plant materials. The crude oil was kept in a cool and dark place until further analyses.

2.2. GC and GC-MS analysis

The oils were analyzed by using a Nucon 5765 gas chromatograph (Rtx-5 column, 30 m × 0.32 mm, FID), split ratio 1: 48, N_2 flow of 4 kg/cm^2 and on Thermo Quest Trace GC 2000 interfaced with MAT Polaris Q Ion Trap Mass spectrometer fitted with a Rtx-5 (Restek Corp.) fused silica capillary column (30 m × 0.25 mm; 0.25 µm film coating). The column temperature was programmed 60 – 210 ^{o}C at 3 ^{o}C/min using He as carrier gas at 1.0 mL/min. The injector

temperature was 210 ^{o}C, injection size 0.1µL prepared in hexane, split ratio 1:40. MS were taken at 70 eV with a mass range of 40 - 450 amu.

2.3. Identification of the components

Identification of constituents were done on the basis of Retention Index (RI, determined with reference to homologous series of n-alkanes (C_9-C_{24}, Polyscience Corp., Niles IL) under identical experimental condition), co injection with standards (Sigma and known essential oil constituents (standard isolates), MS Library search (NIST and WILEY), by comparing with the MS literature data [14]. The relative amounts of individual components were calculated based on GC peak area (FID response) without using correction factor.

3. RESULT AND DISCUSSION

The essential oils composition of leaves of *F. vestita* were analyzed and compared by using capillary GC and GC-MS. Essential oil yield was 0.30 % (v/w). The GC and GC/MS analysis led to the identification of nine constituents forming 98.3 % of the total oil compositions. The identified constituents with their relative content and class composition are given in Table 1.

Table 1. Leaf essential oil composition of *Filipendula vestita* from Uttrakhand, India.

Sr. No.	RI[a] Exp.	RI[b] lit.	Compound	Content	Identification*
1.	882	880	Santene	3.5	RI,MS
2.	1020	1024	Limonene	0.7	RI,MS
3.	1043	1045	Salicylaldehyde	51.5	RI,MS
4.	1095	1097	Linalool	3.60	RI,MS
5.	1190	1192	Methyl salicylate	24.5	RI,MS
6.	1240	1243	Carvone	4.3	RI,MS
7.	1469	1471	Salicylic acid butyl ester	5.7	RI,MS
8.	1673	1675	α-asarone	2.0	RI,MS
9.	1862	1865	Benzyl salisylate	2.5	RI,MS
Class composition					
Monoterpene hydrocarbons				4.2	
Oxygenated monoterpenes				89.6	
Sesquiterpene hydrocarbons				-----	
Oxygenated sesquiterpenes				4.5	
Total Identified %				**98.3**	

*Mode of identification: Retention Index (LRI, Based on homologous series of n-alkanes; C_8-C_{24}), coinjection with Standards/Peak enrichment with known oil constituents, MS (GC-MS), (< 0.1 %); (-) = not detected; [a]RI: Retention index on Rtx-5 column (30 m × 0.25 mm; 0.25 µm film coating); [b]RI: Literature value 14].

The essential oil composition of the leaves of *F. vestita* was mainly constituted by salicylaldehyde 51.5 %, methyl salicylate 24.5 %, salicylic acid butyl ester 5.70 %, carvone 4.30 %, linalool 3.60 %, santene 3.50 %, benzyl salisylate 2.5 %, α-asarone 2.0 % and limonene 0.7 %. As we have seen the essential oil of *F. vestita* showed high percentage of salicylaldehyde

and methyl salicylate which is common in other species of this genus also. Previous studies showed the ratio of these two compounds has very good bioactivities, like the leaf essential oil of *F. vulgaris*, consisting mainly of salicylaldehyde (68.6 %), was screened for its antimicrobial activity by the disk diffusion and micro-dilution broth assays. The essential oil remarkably inhibited the growth of all of the tested bacteria and fungi. It seems that the antimicrobial nature of *F. vulgaris* essential oil can be attributed to the synergistic interactions of the compounds constituting the oil rather than to the presence of a single inhibitory agent. A synergy in salicylaldehyde/linalool mixtures was observed with a maximum interaction situated in the range between 60:40 and 80:20 (mol ratio). At this concentration range (at a dose of 1.7 μg/disk) no microbial growth was observed while the respective pure compounds, at the corresponding quantities, are shown to be dramatically less active. The MIC value for the 60:40 mixtures was determined to be less that 0.009 mg/ml. In addition, an antagonistic relationship between salicylaldehyde and methyl salicylate was established. The maximum (negative) interaction was shown to correspond approximately to the mixture at the 40:60 (methyl salicylate/salicylaldehyde) mol. ratios resulting in the complete loss of activity at the investigated dose [15-18]. Previously I have reported the root essential oil composition of *F. vestita* dominated by methyl salicylate (56.0 %), salicylaldehyde (15.60 %), santene (9.40 %), and limonene (6.30 %) [19] as the major marker constituents, but the leaf essential oil is reporting presently is dominated as salicylaldehyde (51.50 %).

4. CONCLUSIONS

Genus *Filipendula* is a potential source of salicycldehyde. Essential oils are prescribed for a variety of health problems by traditional systems of medicine, all over the world. Thus, essential oil of *Filipendula vestita* growing in higher altitudes of Himalayan region could be used as potential natural source of salicylaldehyde, methyl salicylate and related esters, which may be used as a raw material for herbal industries.

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