

Essential oils of *Satureja boliviana* and *S. parvifolia* growing in the region of Jujuy, Argentina

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ABSTRACT: Essential oils of aerial parts of *Satureja parvifolia* and *S. boliviana* of Argentina were analysed by GC–MS and GC–FID. Fifty-six components were identified. The main compound of *S. parvifolia* essential oil is piperitenone oxide. The most abundant constituents identified in *S. boliviana* essential oil were γ -terpinene, β -caryophyllene, germacrene-D, bicyclogermacrene, 1,8-cineol and linalool. Copyright   2000 John Wiley & Sons, Ltd.

KEY WORDS: *Satureja parvifolia* (Phil.) Epl.; *Satureja boliviana* Briq.; Lamiaceae; essential oil composition; piperitenone oxide; γ -terpinene; β -caryophyllene; bicyclogermacrene; germacrene-D; linalool

Introduction

Satureja boliviana Briq. and *S. parvifolia* (Phil.) Epl. are medicinal plants growing in the Andean countries (Peru, Bolivia and Argentina). They are known as 'mu a' or 'khoa' by Kechuas Indians and 'poleo' by Spanish people.¹

Both species are traditionally used in medicine and in cooking. An infusion of the leaves of *S. boliviana* is used to relieve rheumatism pains, while the stems are also employed to relieve migraines or as a stomachic, sudorific or insecticide.² The branches are used as fuel. The aerial parts are widely used: an infusion is employed as a digestive or antispasmodic or in the treatment of colds; a decoction is used as a vermifuge and the essential oil is sprayed on potatoes to protect them from pests.^{3–5}

S. parvifolia is traditionally used in Argentina against digestive disorders, colds and female sterility. This plant is also known as an aphrodisiac and emmenagogue.^{5,6} It is employed to relieve a disease of the region called *puna*, due to altitude⁶ (personal communication).

In the present work we examined the chemical composition of two essential oil samples from Argentinian *S. boliviana* and *S. parvifolia* collected in the Quebrada and Valley of Jujuy in the North West of Argentina. Analysis were performed simultaneously in Argentina (GC–FID, GC–MS) and in France (GC–MS). Pre-

vious investigations of essential oil of *S. boliviana* and *S. parvifolia* indicated that the chemical composition varied with origin.^{4,7–16}

Experimental

Plant Material

Aerial parts of *S. boliviana* were collected by Ana Molina at the flowering stage in Argentina near San Salvador de Jujuy in the Valley area (altitude above 1000 m). Aerial parts of *S. parvifolia* were collected at the full flowering period by Carmen Ines Viturro in Quebrada of Jujuy (altitude above 3300 m). The samples were identified by O. Ahumada (Professor at National University of Jujuy). Voucher specimens (Ahumada 6640 and Ahumada 9159, respectively) have been deposited at the Herbarium of the Faculty of Agrarian Sciences, National University of Jujuy, Argentina (JUA).

Oil Isolation

Fresh aerial parts of the plants were hydrodistilled for 3 h using a Clevenger-type apparatus. The oils obtained were dried over anhydrous sulphate. Essential oil yields were 0.27% in *S. boliviana* and 0.57% in *S. parvifolia* (v/w of fresh sample). Samples have been stored after reception.

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Essential Oil Analysis

Both samples were investigated by GC-MS and GC using fused silica columns of equivalent stationary phases. These phases were 5% diphenyl, 95% dimethylpolysiloxane (SPB-5, HP-5MS, HP-5, DB-5), 5% phenyl equivalent, 95% polysilphenylenesiloxane (BPX5). Complementary analyses were performed on an apolar DB-1 column to confirm the identification of the compounds.

GC-MS Analysis

In France, *S. boliviana* and *S. parvifolia* essential oils were analysed in a ATI UNICAM 610 instrument linked to a mass spectrometer, ATI UNICAM 120, under the following conditions: column BPX5 (25 m × 0.22 mm, film thickness 1 µm) programmed at 60–250°C (10 min) at 3°C/min; carrier gas, helium (15 psi); injector temperature, 240°C; detector temperature, 250°C; mass spectra, electronic impact. The samples were diluted in chloroform (1 : 10) and 0.3–0.5 µl were injected.

In Argentina, *S. boliviana* and *S. parvifolia* fresh essential oils were analysed by GC-MS using two different apparatuses with a column of equivalent stationary phase. The *S. boliviana* sample was analysed in a GC HP 6890 apparatus linked to a MS HP 5972 spectrometer with a HP-5MS column (30 m × 0.25 mm, film thickness 0.25 µm); carrier gas, helium (15 psi); injector temperature, 240°C (splitless mode); detector temperature, 300°C; column temperature, 60–280°C at 4°C/min; the sample was diluted with acetone (4 µl/ml).

The *S. parvifolia* sample was analysed by GC-MS Shimadzu QP-5000, using a SPB-5 column (30 m × 0.25 µm, film thickness 0.25 µm) under the following conditions: oven temperature, 60°C (0 min)–280°C (10 min) at 4°C/min, injector 240°C, split ratio 1 : 25; carrier gas, helium; interface temperature, 280°C; detector temperature, 300°C.

GC-FID Analysis

Both samples were investigated in Argentina by GC-FID in a KNK 3000 G (KONIK) equipped with a flame ionization detector (FID), using a HP-5 column (30 m × 0.25 mm, film thickness 0.25 µm). GC Analytical conditions were as follows: injector temperature, 250°C; split ratio 1 : 50; column temperature: 60–280°C at 6°C/min; carrier gas hydrogen, flow rate 1 ml/min, detector temperature 300°C. Complementary analysis of *S. parvifolia* sample was performed in the KNK 3000 G using a DB-1 column (30 m × 0.25 mm, film thickness 0.25 µm) under the following conditions: 60–280°C at 4°C/min, carrier gas helium, injector temperature 240°C, detector temperature 300°C.

Components were identified by comparison of their Kováts indices^{17,18} and their mass spectra with those of several databases.^{17,19} Kováts indices were calculated on the basis of a linear regression programme relating the indices of *n*-alkanes to the number of scans of the spectrometer obtained by analysis of an alkane mixture in the same conditions. Kováts indices given by the databases are specific for DB-5 and OV-101 columns, whose stationary phase are similar to those used here.^{17,18}

Component concentrations were calculated from GC-FID areas.

Results and Discussion

The identified compounds of *S. boliviana* and *S. parvifolia* essential oils are listed in Table 1 in order of their elution from the DB-5 column.

Fifty-six volatile components, representing 99.6% of the total oil were identified in the *S. boliviana* sample. The main components and their percentages of *S. boliviana* essential oils from different origins are listed in Table 2. At first, chemical composition of this essential oil differs with those of Bolivian and Peruvian species whose main components are menthone, isomenthone and pulegone. None of these compounds is identified in the studied Argentinian sample. In addition, this essential oil contains nearly equal amounts of monoterpenes (50.1%) and sesquiterpenes (49.3%). The main compounds are γ -terpinene (15.4%), β -caryophyllene (10.2%), germacrene-D (8.9%) and bicyclogermacrene (8.3%). Monoterpene hydrocarbons represent 27.7%, oxygenated monoterpenes 22.4%, sesquiterpene hydrocarbons 38.4% and oxygenated sesquiterpenes 10.9%. Consequently, the chemical composition of this sample is different from the Tucuman Argentinian one studied by Tomasini *et al.* (1973).⁹ The latter was richer in monoterpenes, and the main components were monoterpene hydrocarbons (camphene, 7.9% and *p*-cymene, 27.5%) and esters (bornyl acetate, 9.8%, neryl acetate, 18.6% and geranyl acetate, 11.8%). The chemical composition differences of these essential oils seem to indicate chemotype existence and show the high influence of environmental factors such as geographic location and light intensity.

Fifty-six compounds, representing 97.5% of the essential oil, were identified in *S. parvifolia*. This sample is richer in monoterpene compounds (93.9%), essentially in oxygenated monoterpenes (89.7%). It contains only 3% of sesquiterpene compounds (oxygenated, 1.9%; hydrocarbons, 2.1%). These proportions are in accordance with those previously calculated in essential oils of the same species, as mentioned in Table 3.^{14–16} The main compounds were piperitenone oxide (69.8%), piperitenone (5.6%) and pulegone (4.4%). Argentinian

Table 1. Comparative percentage composition of *S. boliviana* and *S. parvifolia* collected in the region of Jujuy, Argentina

| Compounds | <i>S. boliviana</i> (%) | <i>S. parvifolia</i> (%) |
|---|-------------------------|--------------------------|
| Tricyclene | 0.7 | |
| α -Thujene | 0.1 | 0.1 |
| α -Pinene | 0.6 | 0.3 |
| Camphene | 1.4 | t |
| Sabinene | 0.8 | 0.4 |
| β -Pinene | 0.4 | 0.5 |
| Myrcene | 0.2 | 0.9 |
| 1-Octen-3-ol | 0.6 | |
| 3-Ocanol | t | t |
| <i>o</i> -Cymene | 1.4 | |
| <i>p</i> -Cymene | 2.8 | 0.1 |
| Limonene | 0.6 | 0.9 |
| 1,8-Cineole | 7.4 | 1.2 |
| <i>cis</i> - β -Ocimene | | 0.1 |
| <i>trans</i> - β -Ocimene | 3.4 | 0.4 |
| γ -Terpinene | 15.4 | |
| <i>cis</i> -Sabinene hydrate | | t |
| <i>cis</i> -Linalool oxide | | t |
| <i>m</i> -Cymenene | | t |
| Linalool | 4.8 | 1.0 |
| <i>trans</i> -Pinan-2-ol | 3.9 | |
| <i>trans</i> -Thujone | | 0.1 |
| Dihydrolinalool | | 0.1 |
| <i>trans</i> -Verbenol | | 0.1 |
| Isopulegol | | t |
| Isoborneol | t | t |
| <i>trans</i> - α -Dihydroterpineol | | t |
| Borneol | 1.0 | |
| Menthol | | t |
| Terpin-4-ol | 0.6 | t |
| <i>p</i> -Cymen-8-ol | | 0.6 |
| α -Terpineol | | 0.4 |
| Myrtenal + myrtenol | | 0.6 |
| Verbenone | | t |
| Cuminaldehyde | | 0.6 |
| <i>trans</i> -Carveol | | t |
| Citronellol | | 0.6 |
| Pulegone | | 4.4 |
| Carvone | | 0.6 |
| Piperitone | | 0.8 |
| Linalyl acetate | 0.4 | |
| Geraniol | | 0.3 |
| <i>trans</i> -Myrtanol | | 0.8 |
| Citronellyl formiate | | 1.4 |
| Methyl nerolate | | 0.6 |
| Bornyl acetate | 2.2 | |
| Thymol | | t |
| Pinocarvyl acetate | 0.3 | |
| 6-Hydroxy-carvotanacetone | | 0.1 |
| <i>cis</i> -Pinocarvyl acetate + isoamyl benzyl ether | | t |
| <i>trans</i> -Carvyl acetate | | t |
| δ -Elemene | 0.9 | |
| Piperitenone | | 5.6 |
| Piperitenone oxide | | 69.8 |
| Neryl acetate | 0.1 | |
| α -Copaene | 0.2 | |
| β -Bourbonene | 0.1 | |
| β -Elemene | 0.3 | |
| α -Gurjunene | 0.1 | |
| <i>trans</i> -Caryophyllene | 10.2 | 0.9 |
| β -Gurjunene | 0.1 | |
| Aromadendrene | 0.2 | |
| <i>cis</i> - β -Farnesene | | 0.1 |
| α -Humulene | 0.9 | |
| <i>cis</i> -Muurola-4(14),5-diene | 0.1 | |
| allo-Aromadendrene | 0.2 | |
| γ -Muurolene | 0.5 | t |
| Germacrene D | 8.9 | |
| β -Selinene | | t |

Table 1. Continued

| Compounds | <i>S. boliviana</i> (%) | <i>S. parvifolia</i> (%) |
|--|-------------------------|--------------------------|
| Bicyclogermacrene | 8.3 | 1.0 |
| β -Dihydroagarofuran | 1.7 | |
| α -Muurolene | 1.1 | |
| γ -Cadinene | 0.7 | 0.1 |
| (<i>Z</i>)-Calamenene | 1.7 | |
| δ -Cadinene | 2.8 | |
| Germacrene-D-4-ol | 0.3 | |
| Spathulenol | 2.1 | 1.7 |
| Caryophyllene oxide | 1.8 | 0.2 |
| Viridiflorol | 0.6 | |
| <i>cis</i> -Dihydro-occidentalol | 0.1 | |
| <i>cis</i> -Isolongifolanone | 0.1 | |
| β -Oplopenone + humulene epoxide II | 0.4 | |
| β -Acorenol | | t |
| <i>epi</i> - α -Cadinol | 1.4 | |
| Cubenol | 0.3 | |
| α -Cadinol | 1.4 | t |
| Oplopanone | 0.1 | |
| <i>cis</i> -Dihydro-occidentalol acetate | 0.1 | |
| <i>trans</i> -Dihydro occidentalol acetate | 0.1 | |

Compounds are listed in order of elution on a DB-5 column. t = trace (<0.05%).

Table 2. Comparative composition of the essential oils of *S. boliviana* from different regions

| Origin | Peru ⁴ (aerial parts, %) | Peru ⁷ (aerial parts, %) | Peru ⁸ (aerial parts, %) | Peru ⁸ (aerial parts, %) | Argentina (Tucuman) ⁹ (leaves, %) | Argentina (Jujuy) (aerial parts, %) |
|---------------------------------|--|--|--|--|--|---|
| α -Pinene | 0.3 | 0.5 | 0.3 | 0.7 | 3.0 | 0.1 |
| Camphene | | 0.1 | t | 0.2 | 7.9 | 1.4 |
| <i>p</i> -Cymene | 3.4 | 1.8 | 4.3 | 5.4 | 27.5 | 2.8 |
| 1,8-Cineole | 4 | 4.2 | 4.2 | 9.8 | 1.8 | 7.4 |
| <i>trans</i> - β -Ocimene | 0.1 | | | t | | 3.4 |
| γ -Terpinene | 0.8 | | 2.3 | 0.4 | | 15.4 |
| Linalool | 3.1 | 1.8 | 3.0 | 1.4 | 12.5 | 4.8 |
| Menthone | 24.2 | 54.1 | 12.6 | 0.7 | | |
| Isomenthone | 29.7 | 15.1 | 29.0 | 27.0 | | |
| Isopulegone | | 5.5 | 2.3 | 0.6 | | |
| <i>cis</i> -Dihydrocarvone | 6.3 | | | | | |
| Pulegone | 10.7 | 2.2 | 12.6 | 20 | | |
| Piperitone | 2.1 | 1.7 | 1.3 | 0 | | |
| Linalyl acetate | 0.1 | | t | | 3.8 | |
| Bornyl acetate | | | | 0.8 | 9.8 | 2.2 |
| Thymol | 4.5 | 0.2 | 6.8 | 14.7 | + | |
| Carvacrol | 0.1 | 6.8 | 0.4 | 0.7 | + | |
| Neryl acetate | | | | | 18.6 | 0.1 |
| Geranyl acetate | | | 0.1 | | 11.8 | |
| β -Caryophyllene | 1.6 | | 1.8 | 0.7 | | 10.2 |
| γ -Elemene | 1.3 | | | | | |
| Germacrene D | | | 0.2 | 0.2 | | 8.9 |
| Bicyclogermacrene | | | 3.0 | | | 8.3 |
| δ -Cadinene | | | 0.6 | 0.1 | | 2.8 |
| Spathulenol | 1.3 | | 0.3 | 2.3 | | 2.1 |
| Caryophyllene oxide | t | | 1.0 | 0.2 | | 1.8 |
| β -Bourbanol | | | 2.4 | | | |
| Monoterpenes | 87.9 | 88.7 | | | | 50.1 |
| Monoterpene hydrocarbons | 6.4 | 3.7 | 11.0 | 10.9 | | 27.7 |
| Oxygenated monoterpenes | 81.5 | 85.0 | 78.5 | 77.6 | | 22.4 |
| Sesquiterpenes | | | | | | 49.3 |
| Sesquiterpene hydrocarbons | 4.8 | 1.9 | 1.3 | 2.3 | | 38.4 |
| Oxygenated sesquiterpenes | 1.0 | 1.7? | 3.1 | 3.1 | | 10.9 |

Table 3. Comparative composition of the essential oils of Argentinian *S. parvifolia* from different regions

| Origin | Tucuman ¹⁴ (leaves, %) | Tucuman ¹⁵ (leaves, %) | Córdoba ¹⁶ (leaves, %) | Jujuy (aerial parts, %) |
|----------------------------|--------------------------------------|--------------------------------------|--------------------------------------|----------------------------|
| α -Pinene | 2.3 | 0.3 | 0.4 | 0.3 |
| β -Pinene | 1.2 | 0.2 | 0.4 | 0.5 |
| Myrcene | 1.0 | 0.2 | 0.5 | 0.9 |
| <i>p</i> -Cymene | | 14.0 | | 2.8 |
| Limonene | 4.4 | 0.7 | 5.1 | 0.9 |
| 1,8-Cineole | 2.5 | t | 1.1 | 1.2 |
| γ -Terpinene | | 11.3 | | 15.4 |
| Menthone | | 0.1 | 6.1 | |
| Isopulegone | | 0.5 | | |
| Menthol | | | 20.2 | t |
| Pulegone | | 0.9 | 3.6 | 4.4 |
| Piperitone | 12.8 | | 1.9 | 0.8 |
| Thymol | | 8.9 | | |
| Carvacrol | | 34.0 | | |
| Carvacryl acetate | | 14.7 | | |
| Pipertenone oxide | 41.5 | | 15.0 | 69.8 |
| Piperitonone | | | | 5.6 |
| Piperitone oxide | 19.3 | | 30.1 | |
| β -Caryophyllene | 9.1 | | | 0.9 |
| Bicyclogermacrene | | | | 1.0 |
| Monoterpenes | | | | |
| Monoterpene hydrocarbons | | 31.4 | 6.0 | 3.7 |
| Oxygenated monoterpenes | | 63.8 | 88.1 | 89.2 |
| Sesquiterpenes | | | | |
| Sesquiterpene hydrocarbons | | 0.1 | | 2.1 |
| Oxygenated sesquiterpenes | | 0.7 | 3.0 | 1.9 |

species have been investigated previously. The chemical composition of this sample differs with those of Tucuman samples studied respectively by de Iglesias¹⁴ and Muschietti.¹⁵ The main components of the first sample obtained from leaves were piperitone (12.8%), piperitone oxide (41.5%) and piperitenone oxide (19.3%), while those found in second sample obtained from leaves were carvacrol (34.0%), carvacryl acetate (14.7%), *p*-cymene (14.0%) and γ -terpinene (11.3%). In addition, the chemical composition of the sample analysed differs also from those of the leaf essential oil of the same species from area of Cordoba: the main components of the Cordoba sample were piperitone oxide (30.1%), menthol (20.2%) and piperitenone oxide (15.0%).¹⁶ The difference in oil composition of these *Satureja* oils is probably due to the existence of different chemotypes or to the influence of soil, altitude and weather conditions. These influences are widely known in the chemical composition of essential oils from plants of the Lamiaceae.^{20–22}

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