Specific characteristics of essential oils of four Artemisia species from the Mongolian Trans-Altai Gobi

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Abstract: The essential oil compositions of four Artemisia species in Mongolian Trans-Altai Gobi were studied by gas chromatography (GC) and gas chromatography-mass spectrometry (GC-MS). The oil from A.macrocephala Jacq and A.dracunculus Ledeb. were characterized by the presence of monoterpenes hydrocarbons and oxygenated monoterpenoids predominately. E-nerolidol (26.95%), methylheugenol (23.29%) and sabinene (13.21%) were found as main components in the essential oils of A.dracunculus. A.macropcephala was characterized by the presence of chamazulene (13.8%), cineol (11.7%), myrcene (9.0%), germacrene-D (7.1%). A.anethifolia Web was characterized by the presence of fragrant compounds as camphor (26.05%), α-thujone (10.1%), borneol (5.1%). Davanone and davanone derivatives were also detected in the sample in amount of 7.7% in total. A.scoparia Waldst differed by domination of monoterpen hydrocarbons (78.9%) with (Z)-β-ocimene (29.24%), α-pinene (15.19%), limonene (10.27%) and myrcene (9.61%).

Keywords: Wormwood, essential oil composition, chamazulene, 1,8-cineol, camphor,

INTRODUCTION

The genus of Artemisia (family Asteraceae) which contains many useful aromatic and medicinal plants, comprises of about 400 species found in the northern hemisphere [1]. The genus Artemisia presents 103 species that are found wild all over the Mongolia [2, 3]. Artemisia popularly known as “sagebrush” or “wormwood” is a source of valuable drugs and essential oils. Because of medicinal importance and intricate chemical composition of several varieties and chemotypes, Artemisia continues to be subject of wide interest for chemists and taxonomists. The genus Artemisia produces a great number of terpenoid compounds in glandular trichomes which have been found to be biologically active [4]. Currently, the pharmaceutical [5-8], food science [9, 10], fragrance [9, 11] and in perfumery [12], cosmetical industries [13] are intensively studying the terpenoids of Artemisia and their sesquiterpenoid lactones [14-16]. The Artemisia species are rich in volatile oils that exhibit a wide spectrum of biological activity as anti-inflammatory, antibacterial, antifungal, anti-oxidant, antiviral, allelopathic etc [17, 18]. A.macropcephala, A.scoparia are used as an antiseptic, anti-inflammatory, antihemimtic, tonic as well as for the treatment of stomachache and toothache in Mongolian traditional medicine [4, 5, 8].

A literature survey revealed only a few reports on essential oil composition of A.macrocephala, A.scoparia and many researches for A.dracunculus is known Tarragon [19-22].

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Tarragon possesses insecticide and radical-scavenging activities. Antifungal, antitumor and DNA-damaging effects were also reported for the essential oil of A.dracunculus [23]. Worthy of mention is the use of the aromatic leaves of A.dracunculus in perfumery and range of food applications which include soups, causes, salad dressing and in the Tarragon vinegar [9, 10, 23, 24].

Previously, reported constituents of A.macropcephala were flavonoids, alkaloids, saponins and terpenes including α-pinine, β-pinine, limonene, p-cymene, borneol and 1,8-cineole in the essential oil of the plant. The oil also contains camphor propionic acid, acetic acid, enanthic acid and isovaleric acid. [21, 22, 25]. Mohammad Shoaib et al., examined that the essential oil of A.macropcephala possesses acetylcholinesterase and butyrylcholinesterase inhibitory potential and revealed from the study beneficial applications of the oil in treatment of various neurodegenerative disorders including Alzheimer’s disease, Parkinson’s disease, ataxia and all other forms of dementia [25]. A.scoparia has medicinal properties like anti-cholesterolemic, antipyretic, anti-septic, antibacterial, diuretic, cholagogue, vasodilator [19]. The seeds and young flowering stem of the plant, yield an essential oil that also finds extensive use in medicine. Essential oil of this plant has strong antioxidant and insecticidal activity against stored-product insects [6, 20, 21].

In 1976, Shatar et al., examined the chemical composition of A.scoparia oil produced from plants grown in Mongolian Gobi. The compounds identified in the GC were as follows: α-pinene (15.0%), camphene (12.0%), β-pinene (1.2%), sabinene
(6.0%), 3-ð-carene (2.4%), myrcene (2.5%), terpinene (4.3%), ß-phellandrene (3.5%), ß-phellandrene (3.0%), p-cymene (5.0%), longicyclene (2.5%), longifolene (0.3%), ß-bisabolene (0.8%), ß-santalene (0.2%), ß-himachalene (2.5%), ß-bisabolene (0.4%), ð-cadinene (1.8%) and curcumene (0.2%) [21]. We reported the chemical composition of the essential oil of Mongolian twenty two Artemisia species previously [24, 26-29]. The essential oil composition of Artemisia anethifolia has not been investigated so far. As a part of our studies on Artemisia species in Mongolian Trans-Altai Gobi, we have now investigated the essential oil composition of A.dracunculus, A.macrocephala and A.scoparia.

EXPERIMENTAL

Plant material: Aerial parts of A.scoparia, A.anethifolia, A.macrocephala and A.dracunculus were collected from wild growing plants in Mongolian Trans-Altai Gobi at full flowering and fruiting stage in July-September 2011-2013. The exact dates of each harvesting are showed in Table 1.

A voucher specimen has been deposited in the Herbarium fund of the Institute of Botany of the Mongolian Academy of Sciences, Ulaanbaatar, Mongolia.

Isolation of oil: Air-dried aerial parts (70-80 g) were subjected to a hydro distillation in Clevenger type apparatus for 3 h. The samples were yielded 0.43% (w/w) of A.scoparia oil, 0.75% of A.anethifolia oil, 0.23% of A.macrocephala oil and 0.56% of A.dracunculus oil. The oils were dried over anhydrous calcium chloride and stored in sealed vials at 4°C before analysis.

Gas chromatography (GC) and gas chromatography and mass spectrometry (GC/MS): GC analysis was carried out on Hewlett Packard HP 5890II Gas Chromatograph fitted with an fused silica DB-Wax column (30m×0.25mm×0.25µm); carrier gas nitrogen, linear velocity 38 ml/min, split ratio 30:1. The injector and detector temperature was 250°C, column temperature was programmed from 80 to 200°C at a rate 2°C/min. 0.5 µl solutions of essential oil samples in dichloromethane (1%) were subjected to the injector. Quantitative data were obtained from an electronic integration of the flame ionization detector (FID) peak area. GC-MS analysis was performed on HP 5971A instrument with MS detector 5890II of the same company which was operated in EI mode (70eV). GC-MS fitted with a Supelcowax 10 column (60m×0.25mm×0.25µm); carrier gas helium, linear velocity 10 ml/min, split ratio 30:1. The injector and detector temperature was 250°C and 280°C; column temperature was programmed from 80 to 120°C at a rate 3°C/min. All GC condition and capillary column used were as described above but a carrier gas was helium.

Identification of components: The separated components were identified by matching with mass-spectral library data and by comparison of Kovat's indices with those of authentic components and with published data [30-34].

RESULTS AND DISCUSSION

The essential oils were isolated from the aerial parts of four Artemisia species at full flowering and fruiting stage were obtained in 3 replications and taken means of the oil yields as shown in Table 1. Additionally the place and time of collection of the plant materials are given in Table 1.

<table>
<thead>
<tr>
<th>Plant name</th>
<th>Date of collection</th>
<th>Vegetation period</th>
<th>Oil yield, %</th>
</tr>
</thead>
<tbody>
<tr>
<td>A.scoparia</td>
<td>Waldst August 2012</td>
<td>In full bloom</td>
<td>0.43</td>
</tr>
<tr>
<td>A.anethifolia</td>
<td>Web August 2012</td>
<td>In full bloom</td>
<td>0.75</td>
</tr>
<tr>
<td>A.macrocephala</td>
<td>Jacq September 2013</td>
<td>Ripe fruits</td>
<td>0.23</td>
</tr>
<tr>
<td>A.dracunculus</td>
<td>L July 2011</td>
<td>In full bloom</td>
<td>0.56</td>
</tr>
</tbody>
</table>

The essential oils were obtained from aerial parts of wild growing A.dracunculus, A.anethifolia, A.macrocephala and A.scoparia in Mongolian Trans-Altai Gobi. GC and GC-MS analyses led to detection of 88 constituents accounting 88.30-94.03% of the oils (Table 2). The compounds were identified on the basis of their mass spectral characteristics and retention indices on non polar HP-5MS column.

A.dracunculus essential oil contained 44 compound, accounting 88.30% of the oil. As it seen in Table 2, the oil was rich oxygenated terpenoids: Monoterpenoids 33.66% and sesquiterpenoids 28.66%. Methylchavicol (23.29%), E-nerolidol (26.95%), sabinen (13.21%) were dominating in the investigated sample. The different observers revealed from previous studies which are noted to be principle chemotype of methylchavicol (35.7-60.46%), sabinen (0.11-46.78%), ocimene (0.3-13.54%), ß-terpinene (2.43-17.01%) for essential oil of A.dracunculus from South Siberia, USA, Russia and Italia [10, 29] while the investigated sample has specific chemotype that including considerable amounts of methylchavicol and aromatic alcohols. It can be shown that the A.dracunculus from Mongolian Trans-Altaí Gobi can be used for raw material in the medicines, perfumes, cosmetic and food producing.

The oil of A.anethifolia contained 47 compounds, accounting 88.17% of the oil (Table 2). The sample differed from the described by domination of monoterpenoids (56.94%), among which are camphor (26.05%), ß-cadinene (14.35%). In addition, borneol (5.11%), hexadien-2,4-phenol (4.51%), caryophyllene oxide (2.53%), germacrene-D (2.53%), caryophyllene (2.07%), squalenol (2.17%) were detected as prominent monoterpenoids. A literature survey showed any reports concerning essential oil composition of A.anethifolia. The oil differed from the described above by the presence of davanone type compounds (7.73%) davone-1, davone-2, davone-3, davone-4 and davanyl ether. Literature survey showed that they accompanied davana type compounds in essential oils and could be described as...
Table 1. Essential oil yields of four Artemisia species, %

<table>
<thead>
<tr>
<th>Compounds</th>
<th>A. dracunculus</th>
<th>A. anethifolia</th>
<th>A. macrocephala</th>
<th>A. scoparia</th>
</tr>
</thead>
<tbody>
<tr>
<td>α-thujene</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.14</td>
</tr>
<tr>
<td>α-pinene</td>
<td>-</td>
<td>0.61</td>
<td>1.4</td>
<td>15.19</td>
</tr>
<tr>
<td>camphene</td>
<td>-</td>
<td>2.71</td>
<td>1.2</td>
<td>0.18</td>
</tr>
<tr>
<td>sabinene</td>
<td>13.21</td>
<td>0.24</td>
<td>0.6</td>
<td>3.85</td>
</tr>
<tr>
<td>β-pinene</td>
<td>1.10</td>
<td>0.31</td>
<td>0.4</td>
<td>5.55</td>
</tr>
<tr>
<td>myrcene</td>
<td>0.88</td>
<td>-</td>
<td>8.0</td>
<td>9.61</td>
</tr>
<tr>
<td>α-phellandrene</td>
<td>-</td>
<td>-</td>
<td>3.6</td>
<td>-</td>
</tr>
<tr>
<td>α-terpinene</td>
<td>0.26</td>
<td>-</td>
<td>0.4</td>
<td>0.26</td>
</tr>
<tr>
<td>p-cymol</td>
<td>0.58</td>
<td>0.19</td>
<td>1.4</td>
<td>-</td>
</tr>
<tr>
<td>limonene</td>
<td>1.60</td>
<td>-</td>
<td>1.2</td>
<td>10.27</td>
</tr>
<tr>
<td>(E)-β-ocimene</td>
<td>0.80</td>
<td>-</td>
<td>-</td>
<td>1.02</td>
</tr>
<tr>
<td>(Z)-β-ocimene</td>
<td>1.24</td>
<td>-</td>
<td>-</td>
<td>29.47</td>
</tr>
<tr>
<td>γ-terpinene</td>
<td>0.71</td>
<td>-</td>
<td>0.7</td>
<td>-</td>
</tr>
<tr>
<td>terpinolene</td>
<td>0.45</td>
<td>-</td>
<td>0.5</td>
<td>-</td>
</tr>
</tbody>
</table>

Monoterpenic hydrocarbons 22.55 4.06 20.4 78.94

1.8-cineol 1.10 1.21 11.7 2.61
lavandulacton - 0.33 - -
artemisia keton - 0.75 - -
(E)-sabinenehydrate 0.20 0.25 - -
(Z)-sabinenehydrate 0.33 0.27 1.6 0.09
linalool 0.31 0.21 0.2 -
α-thujone 0.23 9.95 - -
β-thujone - 4.40 - 0.07
(E)-p-ment-2-en-1-ol - 0.93 - -
chrysanthenon - 0.24 - -

pinocarveol - - 0.79 -
camphor - 26.05 1.4 -
(E)-chrysanthenol - 0.47 0.2 -
borneol - 5.11 0.9 0.45
terpin-4-ol 4.38 0.76 0.5 0.19
isogeraniol - - 1.2 -

α-terpineol 2.68 0.38 1.9 0.08
germaylacetate - - - 0.36
methylchavicol 0.46 - - -
myrtenol - 1.06 - -

(Z)-piperitol - 1.25 - -
(Z)-carveol - 0.14 - -
bomyliformat - 0.19 - -
carvon - 0.20 - -
methylysalicate - - - -

bornylacetate 0.21 1.03 - -
(Z)-piperitolacetate 0.59 - - -
methyleugenol 23.39 - - -

Monoterpenoids 33.66 56.94 20.6 1.26

α-santalene - - - 1.12
copaene - - 0.5 - -
β-bourbonene - - - 0.5 - -
β-elemene - - - 0.7 - -
caryophyllene 1.00 2.07 2.0 1.60
humulene - - 0.2 0.12
β-farnesene 0.29 - 2.0 0.45
selina-4,11-dien - - 1.3 -

Germacrene-D 0.29 2.53 7.1 -

β-selinene - - 4.0 -
γ-muurolene 0.19 - - -
bicyclogermacrene 0.86 - 1.0 2.27
(E,E)-α-farnesene - - - 0.44
γ-cadinene - - - 0.33
δ-cadinene 0.40 0.49 - -
selin-3,7(11)-dien - - 2.6 0.90
Chamazulen - - 13.8 -

Sesquiterpene hydrocarbons 3.43 6.89 35.2 11.75
biogenetically connected with them or their degradation products [1, 10, 17].

Further, 32 compounds were identified in the A. scoparia essential oil, representing 94.03% of the studied oil. (Z)-β-ocimene (29.47%), α-pinene (15.19%) and limonene (10.27%) were found to be the main terpenoids in analyzed sample. Myrcene (9.51%), β-pinene (5.55%), sabinene (3.85%) were detected in the relatively low concentrations in the investigated sample. Spathulenol, nerolidol, caryophyllene oxide, α-cadinol and epi-α-cadinol were the only five sesquiterpenoids detected in amounts of 2.08% in total. It should be noted a high percentage of β-ocimene (29.47%) in the oil. This compound has not been detected previously in a plant originated from Khangai region, Mongolia. Instead, α-pinene, camphene are reported as principle components in the plant [21]. Comparison of the obtained results with those published so far showed similar essential oil profile which was characterized by high percentage of monoterpene hydrocarbons and aromatic compounds. The observed differences were significant and could be due to the effect of ecological conditions or stage of the plant development [29].

Finally, in A. macrocephalla oil were detected 44 compounds, representing 92.4% of the oil. The obtained results have considerably enlarged data for A. macrocephalla essential oil. So far, monoterpenoids were known as main components of the oil with borneol (10.8%), cineol (10.7%), isoborneol (8.3%), camphor (6.3%) and chamazulene without any quantitative data [21], camphene, limonene, p-cymol, α-thujone [7], caryophyllene, β-selinene [1], 1.8-cineole, γ-terpinene, sabinene, limonene [29]. These components were presented in the investigated sample, but the oil was characterized by high contents of chamazulene (13.8%), 1.8-cineole (11.7%). Other prominent compounds were myrcene (9.0%) and germacrene (7.1%). A. macrocephala in Mongolian Trans-Altaï Gobi could be more anti-inflammatory activity than others which are growing in different areas of Mongolia because of chamazulene content according to the studies by researchers at Siberian State Medical University showed high anti-inflammatory activities for azulene (derivative product is chamazulene) containing essential oils of wormwood [35].

CONCLUSION
According to the results, specific chemotypes in the essential oils from A. scoparia, A. dracunculus, A. macrocephala in Mongolian Trans-Altaï Gobi are revealed comparing with previous researches of the three Artemisia species are wild growing in different regions of the Mongolia. Furthermore chemical compositions of the essential oil from A. anethifolia in the Gobi were investigated firstly [1, 3, 7, 10], which are dominated by monoterpenoids. Determining components as main from the Artemisia species are methyleugenol in A. dracunculus, aromatic alcohols and ketones in A. macrocephala, chamazulene in A. macrocephala possess high bioactivity properties and the essential oils could be effective supplements for medicine, food, perfumer industries.

REFERENCES

<table>
<thead>
<tr>
<th>Compound</th>
<th>A. scoparia</th>
<th>A. dracunculus</th>
<th>A. macrocephala</th>
</tr>
</thead>
<tbody>
<tr>
<td>lavandulisolobutanoate</td>
<td>-</td>
<td>-</td>
<td>1.0</td>
</tr>
<tr>
<td>lavanduly-3-methylbutanoate</td>
<td>-</td>
<td>-</td>
<td>2.2</td>
</tr>
<tr>
<td>E-nerolidol</td>
<td>26.95</td>
<td>-</td>
<td>0.3</td>
</tr>
<tr>
<td>hexadien-2,4,1-phenyl-4-neryl-2-methylbutanoate</td>
<td>-</td>
<td>4.51</td>
<td>-</td>
</tr>
<tr>
<td>E,E-α-fernezelin</td>
<td>-</td>
<td>0.45</td>
<td>-</td>
</tr>
<tr>
<td>davanyl ether</td>
<td>-</td>
<td>2.21</td>
<td>-</td>
</tr>
<tr>
<td>neryl-3-methylbutanoate</td>
<td>-</td>
<td>-</td>
<td>2.2</td>
</tr>
<tr>
<td>geranyl-2-methylbutanoate</td>
<td>-</td>
<td>-</td>
<td>1.4</td>
</tr>
<tr>
<td>copaborneol</td>
<td>-</td>
<td>-</td>
<td>2.2</td>
</tr>
<tr>
<td>geranyl-3-methylbutanoate</td>
<td>-</td>
<td>-</td>
<td>0.4</td>
</tr>
<tr>
<td>davanon-1</td>
<td>-</td>
<td>1.47</td>
<td>-</td>
</tr>
<tr>
<td>davanon-2</td>
<td>-</td>
<td>2.19</td>
<td>-</td>
</tr>
<tr>
<td>davanon-3</td>
<td>-</td>
<td>1.10</td>
<td>-</td>
</tr>
<tr>
<td>spathulenol</td>
<td>1.39</td>
<td>-</td>
<td>0.9</td>
</tr>
<tr>
<td>caryophyllene oxide</td>
<td>0.32</td>
<td>2.17</td>
<td>0.4</td>
</tr>
<tr>
<td>epi-α-cadinol</td>
<td>-</td>
<td>2.53</td>
<td>-</td>
</tr>
<tr>
<td>davanon-4</td>
<td>-</td>
<td>0.76</td>
<td>-</td>
</tr>
<tr>
<td>β-bisabolol oxide</td>
<td>-</td>
<td>0.81</td>
<td>-</td>
</tr>
<tr>
<td>γ-eudesmol</td>
<td>-</td>
<td>-</td>
<td>0.3</td>
</tr>
<tr>
<td>bisabolol</td>
<td>-</td>
<td>2.08</td>
<td>-</td>
</tr>
<tr>
<td>T-cadinol</td>
<td>-</td>
<td>-</td>
<td>0.4</td>
</tr>
<tr>
<td>caryophyll-4-en-13-ol</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>Total identified compounds</td>
<td>88.30</td>
<td>88.17</td>
<td>92.4</td>
</tr>
<tr>
<td>Sesquiterpenoids</td>
<td>28.66</td>
<td>20.28</td>
<td>16.2</td>
</tr>
<tr>
<td>Total identified compounds</td>
<td>88.30</td>
<td>88.17</td>
<td>92.4</td>
</tr>
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</table>
Mongolia. Valang, Moscow, 36 (in Russian).