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Review

The essential soils on the anthemideae

Jaime A. Teixeira da Silva

Faculty of Agriculture, Kagawa University, Miki-cho, Ikenobe, 2393, Kagawa-ken, 761-0795, Japan. E-mail: jaimetex@yahoo.com; Telfax: +81 (0)87 898 8909.

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Numerous members of the Anthemideae are important cut-flower and ornamental crops, as well as medicinal and aromatic plants, many of which produce essential oils used in folk and modern medicine, the cosmetic and pharmaceutical industries. These oils and compounds contained within them are used in the pharmaceutical, flavour and fragrance industries. Moreover, as people search for alternative and herbal forms of medicine and relaxation (such as aromatherapy), and provided that there are no suitable synthetic substitutes for many of the compounds or difficulty in profiling and mimicking complex compound mixtures in the volatile oils, the original plant extracts will continue to be used long into the future. This review highlights the importance of secondary metabolites and essential oils from principal members of this tribe, their global social, medicinal and economic relevance and potential.

Key words: Apoptosis, artemisinin, chamomile, essential oil, feverfew, pyrethrin, tansy.

THE ANTHEMIDAE

Chrysanthemum (Compositae or Asteraceae family, subfamily Asteroideae, order Asterales, subclass Asteridae, tribe Anthemideae), sometimes collectively termed the *Achillea*-complex or the *Chrysanthemum*-complex (tribes Astereae-Anthemideae) consists of 12 subtribes, 108 genera and at least another 1741 species (Khallouki et al., 2000). Anthemideae is one of the most well investigated tribes of the Asteraceae, which together with members of Astereae, Cynareae and Heliantheae tribes share compounds such as acetylenes and related compounds such as alkamines, sulphur compounds, isocoumarins and lactones. Essential oils, secondary metabolites and medicinally important compounds with or without bioactivity, have been isolated from *Achillea*, *Anthemis*, *Artemisia*, *Balsamita*, *Chrysanthemum*, *Matricaria*, *Santolina* and *Tanacetum*.

Numerous chrysanthemum plants, apart from their ornamental value, are highly aromatic due to the many volatile components of their essential oils (Table 1), many of which are used in the flavour and fragrance industries, others in alcoholic beverages such as nojigiku alcohol from *Chrysanthemum japonense*. In some countries, such as Japan, both edible (garland, ryouri, shun or shokuyo giku, or chopsuey green, *C. coronarium*) and garnish (tsuma giku, *C. morifolium*) chrysanthemums are popular. The petals of *D. grandiflora* 'Enmeiraku' (or Mottenohoka) containing antioxidant properties and are a popular food in Yamagata, Japan.

SECONDARY METABOLITES AND ESSENTIAL OILS IN MEDICINE AND INDUSTRY

Secondary metabolism in a plant not only plays a role for its survival by producing attractants for pollinators, and a chemical defence against predators and diseases. Often high light or UV leads to the production of anthocyanins, flavones, sinapyl esters, isoflavonoids and psoralens; wounding to coumestrol, coumarin, psoralen, chlorogenic acid, ferulate ester, wall bound phenolic acid, lignin and suberin production; pathogen attack to pterocarpan, isoflavan, prenylated isoflavonoid, stilbene, coumarin, furanocoumarin, 3-deoxyanthocyanidin, flavonol and aurone production; low temperature to anthocyanin production; low nitrogen, phosphate or iron in the soil results in flavonoid/ isoflavonoid, anthocyanin and phenolic acid production, respectively (Heath, 2002; Pichersky and Gershenzon, 2002).

Many secondary metabolites are also an important trait for our food (taste, colour, scent), while others yet such as alkaloids, anthocyanins, flavonoids, quinines, lignans,

S. chamaecyparissus	Cultivated, insular, peninsular populations; artemisia ketones (28-36), T-cadinol (5-24)	Camphor (9-25)	62-94	n.s.	Geotypes	Perez and Velasco 1992
Art. judaica	Artemisia ketone 0-41, camphor tr-20, Artemisia alcohol 0-31, (E)-ethyl cinnamate 4-9	Davanone 0.2-0.9	62	0.5	Sinai	Ravid et al. 1992
Art. salsoloides	Camphor 42, 1,8-cineole 17, camphene 5, terpinen-4-ol 5, β-thujone 3	cis-Chrysanthenol 0.45	109	0.5	Himalayas	Weverstahl et al. 1992a
Art. moorcroftiana	α-Thujone 13, artemisia ketone 10, β-pinene 8, 1,8-cineole 6, camphor 5, β-thujone 4	Vulgarone B 3	n.s.	n.s.	Green odour	Weverstahl et al. 1992b
A. biebersteinii	(Aerial part) 1,8-cineole 46, camphor 18, α-terpineol 8, borneol 3, sabinene 3	Thymol 0.1	47	0.8	Turkey	Chialva et al. 1993
An. nobilis	(Flower bud, aerial part, cell suspension) α-farnesene 0-91, 3-OH 2-butanone 0-72	Nerolidol 0-1.2	30	0.08-0.3	Crown galls	Fauconnier et al. 1993
T. longifolium	Aerial part: trans-sabinyl acetate 43, trans-sabinol 13; root: terpinen-4-ol 26, sabinene 23	Neothujyl alcohol 0.6	49	0.3:0.1	Blue/yellow	Kaul et al. 1993
Art. absinthium	Normal root: α-fenchene, β-myrcene 6; hairy root: neryl isovalerate 47, neryl butyrate 6	β-pinene 1	6	1.4:0.7	Transgenic	Kennedy et al. 1993
Art. petrosa	Wild plants: 1,8-Cineole, β-pinene, borneol	n.s.	41	n.s.	Greece	Souleles 1993
†	(Z)-3-hexen-1-ol acetate 19-55, (Z),(E)-α-farnesene/β-myrcene 3-14 (53% monoterpenes)	3-36% unidentified	43	n.s.	Headspace	Storer et al. 1993
S. sieberi	Camphor 44, 1,8-cineole 19, camphene 5, terpinen-4-ol 3, α-terpineol 2	Bisabolene derivatives tr	>37	n.s.	Flowering	Weverstahl et al. 1993
Art. annua	China: artemisia ketone 64, Artemisia alcohol 8; Vietnam: camphor 22, germacrene-D 18	Artemisinin 0.2-1	n.s.	n.s.	Seeds	Woerdenbag et al. 1993
Art. annua	Artemisia ketone 59, camphor 16, 1,8-cineole 10, germacrene-D 2, pinacarvone 2	Artemisia alcohol 0.15	35	n.s.	Geotypes	Ahmad and Misra 1994
Art. argentea	(Aerial part) α-phellandrene 25-27, isopinocamphone 7-12, β-eudesmol 8	Chamazulene tr	50	0.2-0.25	Madeira	Figueiredo et al. 1994
Art. apiaceae	Primarily camphene, camphor borneol, caryophyllene	n.s.	34	0.23-0.37	Korean	Kim and Jang 1994
A. spp. (10)	Primarily β-pinene, 1,8-cineole, camphor (mono-); germacrene D (sesqui-)	Bisabolene oxide tr	n.s.	n.s.	Geotypes	Maffei et al. 1994
Art. afra	(Vegetative) cis-2,7-dimethyl-4-octene-2,7-diol 19, 1,8-cineole 18, tricosane 14	α-Thujone 1.66	21	0.3-1.4	GC-MS	Moody et al. 1994
C. recutita	19 cuticular waxes; bisabolol oxide A 50, bisabolol oxide B 17, cis-dicycloether 10	Matricine 4	53	1.2	SCFE	Reverchon and Senatore 1994
S. neapolitana	γ-Muurolene 32, α-pinene 16, borneol 9	Ylangene 0.1	41	0.3	Italy	Senatore and de Feo 1994
A. asplenifolia	(Parent plant:in vitro) β-pinene (47:46), 1,8-cineole (10:10), β-caryophyllene (13:14)	Camphor <0.1	13	n.s.	GC-MS	Wawrosch et al. 1994
An. nobilis	(E,E)-α-farnesene 21-57, germacrene D 10-28 (growth stage and light-dependent)	Angelic acid tr.-5	11	n.s.	GC-MS	Asai et al. 1995
A. millefolium	Germacrene-D (11-42), eugenol (9-36), p-cymene (0.1-41)	~55% unidentified	19	0.001	GC-MS	Figueiredo et al. 1995
A. biebersteinii	(Aerial parts) piperitone 4-50, 1,8-cineole 11-30, camphor 9-17, α-terpineol+borneol 1-5	Grandisol 0.4	31	0.55-0.85	Turkey	Küsmenoglu et al. 1995
Art. umbelliformis	α-Thujone 68, β-thujone 18, 1,8-cineole 2, sabinyl acetate 2	Artemisia ketone tr	62	n.s.	Italy	Mucciarelli et al. 1995
Art. campestris I	Caryophyllene oxide 18, unknown alcohol 11, α-pinene 15, β-pinene 10, limonene 5	Artemisia alcohol tr				Ibid.
Art. genipi	α-Thujone 80, β-thujone 10	Spathulenol 0.8				Ibid.
Art. petrosa	α-Thujone 70, β-thujone 17, spathulenol 4	Ascaridole 0.2				Ibid.
Art. vallesiaca	Camphor 41, borneol 28, 1,8-cineole 15, camphene 7	γ-Selinene tr				Ibid.
Art. absinthium	cis-Epoxy-ocimene 25, trans-chrysanthenyl acetate 22, camphor 17, spathulenol 8	Bisabolol oxide 5				Ibid.
Art. chamaemelifolia	Unknown alcohol 27, trans-nerolidol 23, carvacrol 16, 1,8-cineole 15, spathulenol 3	α-Bisabolol 3				Ibid.
Art. glacialis	Camphor 32, 1,8-cineole 15, spathulenol 9, caryophyllene oxide 6, camphene 6	Yomogi alcohol 0.2				Ibid.
Art. campestris II	1,8-cineole 19, spathulenol 18, α-pinene 17, epi-cubenol 14, β-pinene 11	trans-Nerolidol 0.2				Ibid.
Art. alba	Camphor 39, cuminaldehyde 14, isopinocamphone 10, camphene 4, bornyl acetate 4	Myrtenol 0.3				Ibid.
Art. abrotanum	1,8-Cineole 34, bisabolol xide 18, ascaridole 16, trans-nerolidol 4, p-cymene 8	Terpinen-4-ol 2.2				Ibid.
Art. annua	1,8-Cineole 23, α-pinene 20, pinocarveol 6, carvacrol 4	Nojigiku alcohol 0.1				Ibid.
Art. verlotiorum	Caryophyllene oxide 21, borneol 18, camphor 11, 1,8-cineole 11, spathulenol 8	Cuminaldehyde 3.4				Ibid.
Art. vulgaris	Camphor 48, camphene 9, verbenone 9, trans-verbenol 7, β-caryophyllene 4	α-Copaene 1.1				Ibid.

C. coronarium	Camphor 29, α-pinene 15, β-pinene 10, lyratyl acetate 10	Chamazulene 5	15	n.s.	Antifungal 12	Alvarez-Castellanos et al. 2001
T. armenum	(Leaf:herb) 1,8-cineole (31:11), camphor (9:27), α-pinene (4-0.5)	Sabinaketone tr	64:77	0.62-0.67	GC, GC-MS	Başer et al. 2001
T. balsamita	Carvone 52, α-thujone 12, germacrene-D 3	Trans-dihydrocarvone	74	0.38		Ibid.
T. chiliophyllum	Camphor 17, cis-chrysanthenol acetate 16, α-thujone 13, linalool 4, hotrienol 3	Heneicosane	50	0.40		Ibid.
T. haradjani	Camphor 16, 1,8-cineole 10, terpinen-4-ol 7, α-terpineol 4,	T-cadinol	102	0.55		Ibid.
M. recutita	EN-IN-dicycloether 20, apigenine-7-glycoside 20	Chamazulene 10	n.s.	n.s.	Postharvest	Böttcher et al. 2001
S. canescens	Artemisia ketone 61, camphor 20, α-pinene 2	SDA tr.	28	0.13-0.79	Tissue culture	Casado et al. 2001a,b
S. insularis	(CO_2:hydrodistilled) β-myrcene 15:17, β-phellandrene 9:8, spathulenol 5:5	Khusimone 0.3	44:61	n.s.	Supercritical	Cherchi et al. 2001
Art. absinthium	$C_{10}H_{16}O$ 54, β-thujone 12, β-sabinene 2	28% unidentified	6	n.s.	Acaricidal	Chiasson et al. 2001
T. vulgare	β-Thujone 88-92, camphor 1	6-10% unidentified	5	n.s.	Acaricidal	Ibid.
A. millefolium	(July:Sept) β-caryophyllene (39:26), germacrene-D (8:9), p-cymene (7:12)	Camphor (4:7)	33:35	n.s.	Headspace	Cornu et al. 2001
Arg. adauctum3x ssp	(adauctum: caryophyllene oxide 12, borneol 5; gracile: santolina triene 20, chamazulene 8; erythrocarpon: geranyl isovalerate 45, geranyl caproate 9	(Z)-β-santalol 0-1.3	138	0.2-0.4	GC, GC-MS	Couladis et al. 2001 Ibid.
B. suaveolens	(Essential oil:aromatic water) carvone (44:75), α-thujone (16:6), 1,8-cineole (3:4)	Selin-11-en-4-α-ol	80:27	n.s.	GC, GC-MS	Gallori et al. 2001
T. argyrophyllum	(Leaf:flower) α-thujone (52:63), 1,8-cineole (11:4), β-thujone (5:4), camphor (3:2)	Yomogi alcohol (0.6:1)	68:49	1.03:0.96	GC, GC-MS	Gören et al. 2001
T. argenteum	Caryophyllene oxide 13, α-thujone 12, β-caryophyllene 5, caryophylladienol II 4	Spathulenol 1.2	48	0.04		Ibid.
T. praeteritum	(Subsp. praeteritum:massicyticum) α-thujone (0:51), 1,8-cineole (12:4), β-thujone (0:10)	Cinnamaldehyde (0.1:0)	46:70	1.09:0.92		Ibid.
Arg. adauctum	β-Pinene 27, santolinatriene 23	n.s.	60	n.s.	GC-MS	Palá et al. 2001b
C. recutita	(Flower) cis- and trans-dicycloether 30, α-bisabolol/chamazulene 21, β-farnesene 9	γ-Cadinene 1-2	8	n.s.	Supercritical	Povh et al. 2001
Art. marschaliana	(Aerial part) α-pinene 25, germacrene-D 24, bicyclogermacrene 15, spathulenol 10	Longifulene 0.4	20	0.2	Iranian plants	Ahmadi et al. 2002
Art. judaica	(Aerial part) piperitone 45, trans-ethyl cinnamate 21, ethyl-3-phenyl propionate 11	2,6-Dimethyl phenol 1.4	25	1.4	Antioxidative	El-Massry et al. 2002
T. santolinoides	(Aerial part) thymol 18, trans-thujone 18, trans:cis-chrysanthenyl acetate 13: 9	1,8-Cineole 5	n.s.	n.s.	Antimicrobial	El Shazly et al. 2002
Art. campestris	(Aerial part) γ-terpinene, capillene, 1-phenyl-2,4-pentadyene, spathulenol	Methyleugenol tr	51	n.s.	Phenology	Juteau et al. 2002a,b
Art. annua	Camphor 44, germacrene-D 16, trans-pinocarveole 11, β-selinene 9, β-caryophyllene 9	Artemisia ketone 3	26	0.5	GC-MS	Ibid.
Art. afra	(Aerial part) Thujanone 53, camphor 16, 1,8-cineole 14, camphene 3	β-Thujene 0.2	15	n.s.	Antimicrobial	Muyima et al. 2002
Art. sieberi	Camphor 49, 1,8-cineole 11, bornyl acetate 6, trans-verbenol 3, lavandulol 3	Chrysanthemyl acetate 0.7	40	1.02	GC-MS	Sefidkon et al. 2002
Art. santolina	Neryl acetate 13, bornyl acetate 11, trans-verbenol 10, lavandulol 9, linalool 7	Globulol 1.5	39	0.83		Ibid.
Art. aucheri	Verbenone 22, camphor 21, 1,8-cineole 8, trans-verbenol 8, piperitone 3	Spathulenol 0.9	26	0.84		Ibid.
A. setacea	1,8-Cineole 19, sabinene 11, camphor 5, α-pinene 4, bisabolone oxide 4, terpinen-4-ol 3	trans-Pinocarveol 1	27	n.s.	Leaves and flowers	Ünlü et al. 2002
A. teretifolia	1,8-Cineole 20, borneol 12, camphor 11, thujone 5, sabinene 5, trans-piperitol 3	Thymol 0.7	28	n.s.		Ibid.
A. santolina	1,8-Cineole 18, camphor 18, 4-terpineol 7, p-cymene 4, trans-sabinene hydrate 3	Santolina triene 1.3	45	0.18	GC, GC-MS	Bader et al. 2003
A. biebersteinii	cis-Ascaridole 36, p-cymene 32, carvenone oxide 6, camphor 5, carvacrol 1	Terpinolene tr	34	0.20		Ibid.
Arg. pedemontana	Chemotype 1: Camphor 49, 1,8-cineole 13	Chemotype 2: davanone 28	56	n.s.	GC-MS	Perez et al. 2003

A. Achillea, *Arg. Argyranthemum*, *Art. Artemisia*; *B. Balsamita*, *C. Chrysanthemum*, *M. Matricaria*, *S. Santolina*, *T. Tanacetum*. TNC = total number of identified compounds; [1]= 1 x var. *sinense* Makino, 1 x var. *sinense* Makino forma *esculentum* Makino (Ruouri giku). **†** = *D. zawadskii* x *D.*X *grandiflora* . HPLC high performance liquid chromatography; ^{1}H NMR nuclear magnetic resonance, GC-MS gas chromatography-mass spectroscopy, LC liquid chromatography, TLC thin layer chromatography. PP = plant part; n.s. not specified; tr = trace amounts.

morifolium, which also has antiallergic, antibacterial, antifungal, antiviral, antispirochetal, anti-inflammatory, anticarcinogenic (caused by triterpene triols and diols) or tumor-inhibition, lens aldose reductase inhibition and antioxidant activities (Murayama et al., 2002).

Santolina **spp**

S. chamaecyparissus is a shrub with yellow inflorescences widely used in Mediterranean folk medincine. The flowers are used for their analgesis, anti-inflamatory, antiseptic, antispasmodic, bactericidal, fungicidal, digestive and vulnerary properties, and is used in phytotherapy for different kinds of dermatitis. Several products (acetylenes, essential oils, flavonoids, sesquiterpenes) obtained from *Santolina* spp. have been investigated for their biological activities, both from *in planta* and callus cultures, albeit with lower yields in the latter. Coumarins from *S. oblongifolia* have anti-inflammatory properties attributed to the action of apigenin, luteolin, quercetin, herniarin, scopoletin, scopolin and aesculetin (Silvan et al., 1998). *S. insularis* essential oil obtained *in toto* had strong antiviral activity on Herpes simplex virus types 1 and 2 (de Logu et al., 2000). *S. insularis* oils derived by supercritical CO_2 extraction are superior to those derived from hydrodistillation or liquid CO_2 extraction. The essential oils of *S. rosmarinifolia* show great differences at the subspecies level and with a change in season (Palá et al., 2001a), where in ssp. *rosmarinifolia* and *canescens*, the major components are monoterpenes, while in ssp. *pectinata* and *semidentata* they are sesquiterpenes (Palá et al., 1999).

Tanacetum **spp**

Tanacetum species, totaling over 200 and distributed over Europe and West Asia and growing up to altitudes of 2,000 meters contains several strongly scented annual and perennial species. It had been used as an insecticide as well as an insect repellant by native Americans after its introduction into North America in the 18th century. Interest is increasing in species of *Tanacetum* due to its essential oils (stomachic, cordial and used as a food preservative), bitter substances and the presence of sesquiterpene lactones, which exhibit biological activities like cytotoxicity, growth regulating, antimicrobial effects and allergic contact dermatitis. Antibacterial activity by sivasinolide, a sesquiterpene lactone from *T. densum* is effective against *Bacillus subtilis* and *Klebsiella pneumoniae* (Gören et al., 1992).

Tanacetum products are widely distributed in health food shops, while tansy extracts can be effectively used as antioxidants in rapeseed oil (Bandonienė et al., 2000). Epileptogenic properties (i.e. powerful convulsants) of tansy have been known for a long time since it has highly reactive monoterpene ketones such as camphor, pinocamphone, thujone, 1,8-cineole, pulegone, sabinylacetate and fenchone (Burkhardt et al., 1999). *T. corymbosum* essential oil exhibit anticoagulant properties and antifibrinolytic activity (Thomas, 1989b).

Several terpenoids were identified in tansy oils: *α*-pinene (the major component of turpentine, an oleoresin), *α*-terpinene, *γ*-terpinene, *α*-cubenene, dihydrocarvone, artemisia ketone, chrysanthenyl acetate, borneol, *α*- and *β*-thujone, chrysanthenone, camphor and carvone. Even though the active principles behind the mosquito repellant activity of *Tanacetum* oils could not be identified, camphor and bornyl acetate were the most active repellants against potato beetles (Schearer, 1984), while other *Tanacetum* (var. *argyrophyllum* or *praeteritum*) had antibacterial and antifungal activity, probably attributed to the action of the sesquiterpene lactones (Gören et al., 1996b) or in the case of var. *argenteum*, cytotoxic and antifeedant activities (Gören et al., 1996a).

Tanacetum contains similar compounds to *C. coronarium*, although quantities of *α*- and *β*-pinene are lower; *T. parthenium* (feverfew) has a high camphor (44%) and trans-chrysanthemyl acetate (23%) content, while *T. vulgare* (chemotype-dependent) contains lyratyl acetate, thujone and germacrene. One *T. vulgare* contained 95% davanone (Appendino et al., 1984). Other *Tanacetum* extracts, chamazulene and dihydrochamazulenes, of blue colour are used in the perfumery (cosmetic) and pharmaceutical industries. Natural camphor exists as both (–)-form and (+)-form, the former less common than the latter. (*1R*)(+)-Camphor is crystallized from the essential oil by distilling the wood of *Cinnamomum camphora*, but the (*1S*)(–) form has been found in *Tanacetum* and *Artemisia* . (–)-Camphor is the main constituent of *T. balsamita* (=*Balsamita major*), having a characteristic minty smell, also of spearmint.

Future perspectives

This review summarizes and characterizes the importance of essential oils found from a wide range of Anthemidae genera. A number of compounds in these oils (and the oils themselves) have medicinal, (ethno) pharmacological properties and are used in the cosmetic, flavour and fragrance industries. Moreover, some compounds of commercial interest constitute high percentages of the essential oil, and their purification may lead to a success in molecular farming using bioreactor systems.

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