Phytochemicals and Their Biological Activities of Plants in *Tagetes* L.

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Abstract: *Tagetes* L., the genus in the family Asteraceae, consists of about 30 species spread in South and Middle America as well as Mexico. More than one hundred secondary metabolites have been obtained in phytochemical investigation on the species, some of which have potent biological activities. The advances in phytochemical studies and biological activities of the plants in *Tagetes* L. from 1925 to 2011 are summarized in this paper.

Key words: Asteraceae; biological activities; secondary metabolite; *Tagetes* L.
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Introduction

The plants in *Tagetes* L., popularly known as marigold, is originally used as an ornamental plant (Wang, Xu, and Zhao, 2002). There are about 30 species in *Tagetes* L. in South and Middle America as well as Mexico. Both *T. erecta* L. and *T. patula* L. are mainly distributed in China. *T. erecta* has been widely cultured in China and *T. patula* is mainly cultured in Guizhou, Yunnan, Guangdong, Hainan, and Guangxi Provinces of China. The species of *Tagetes* L. have not been admitted in *Chinese Pharmacopoeia 2010*, however, their chemical compounds and bioactivities have been intensively studied in recent years. The phytochemical studies of the plants in *Tagetes* L. could be traced back to 1920s. Till now, about 126 secondary metabolites with various carbon skeletons, phenolic derivatives, phenylpropanoids, thiophene derivatives, benzofunan derivatives, triterpenoids, steroids, alkaloids, flavonoids, carotenoids, and others have been obtained from the species of *Tagetes* L. Some of them showed potent activities as leading compounds of the new drugs.

Some species in *Tagetes* L. have been widely used as folk medicine for calming the liver, dominating heat, expelling wind, and reducing phlegm (Lin, 2009), and also have been used as natural insecticides and fungicides (Vasudevan, Kashyap, and Sharma, 1997). For example, the extract from the roots of *T. erecta* was lethal or inhibitory to the hatching of *Meloidogyne javanica* (Treub) Chitw. and *M. arenaria* (Neal) Chitwood (Vasudevan, Kashyap, and Sharma, 1997). The extracts from some species in *Tagetes* L. also exhibited antifungicidal activity (Vasudevan, Kashyap, and Sharma, 1997). Various oils in plants of *Tagetes* L. were found to inhibit Gram-positive bacteria and fungi (Vasudevan, Kashyap, and Sharma, 1997). In addition, flower pigments in the plants of *Tagetes* L., as natural colorant, have been widely used in food and drink (Vasudevan, Kashyap, and Sharma, 1997). Other applications, such as the anti-oxidative and anti-mutagenic activities, have also emerged, which gradually captured the interest of scientists.

The lack of updated reviews on this topic urged us to speculate the topic from phytochemical and biological views.

Chemical constituents

Phenolic derivatives

Four phenolic derivatives (**1** — **4**) have been isolated from the species of *Tagetes* L. Among them, the positions C-1 and C-4 are substituted by -COOH and -OH, respectively, while no substitution occurs at the position C-2 or C-6. The positions C-3 and C-5 are often substituted by -OMe or -OH.

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Phenylpropanoids

Six phenylpropanoids (**5**—**10**) isolated from plants in *Tagetes* L. have been reported. Compounds **7** and **8** are a pair of isomers with differences at the configuration of the positions C-1, C-3, C-4, and C-5. Compounds **9** and **10** are two classical coumarin derivatives.

Thiophene derivatives

Sixteen thiophene derivatives (**11** — **26**) in the species of *Tagetes* L. have been identified. α-Terthienyl (**11**) was discovered from the flowers of the lemon variety of *T. erecta* by Zechmeister and Sease in 1947 (Vasudevan, Kashyap, and Sharma, 1997). Compound **13** was identified in 1959. Then, in 1962, compounds **17**, **21**, **22**, and **25** were isolated from the ether extracts of *T. erecta* and *T. patula* (Wang, Xu, and Zhao, 2002). Compounds **14** and **15** were reported in 1964 by Atkinson *et al* (Wang, Xu, and Zhao, 2002). The others were reported subsequently during the years 1966—2011. α-Terthienyl is representative in this class of compounds. It is found to distribute in five species, *T. erecta* Linn., *T. patula* Linn., *T. tenuifolia* Cav., *T. microglossa* Benth., and *T. jalisciencis*.

Among these compounds, **13**—**26** are bithiophene derivatives, of which substituted groups are introduced to the positions C-5 and C-5′. Alkyne rarely appeared as a substituted group. However, from this genus, nine bithiophenes (**14** — **17**, **21** — **24**, and **26**) were substituted with alkynes. The biosynthesis study of 5-(3,4-diacetoxy-1-butynyl)-2,2′-bithiophene (**16**) showed that the compound **16** was converted to 5-(3,4-dihydroxy-1-butynyl)-2,2′-bithiophene by a highly specific 5-(3,4-diacetoxy-1-butynyl)-2,2′-bithiophene: 4-acetate esterase, and 5-(3-hydroxy-4-acetoxy-1-butynyl)-2,2′-bithiophene was the intermediate product in the process (Pensl and Sűtfeld, 1985). Compound **22** was yielded by biosynthesis in root cultures which was grown with [U-$^{13}C_6$] glucose or [1-^{13}C] glucose of *T. patula*, and the data suggested that acetyl-CoA or a closely related compound (e.g. malonyl-CoA) might be as building blocks and their orientations in the bithiophene (Margl *et al*, 2001).

Benzofunan derivatives

Six benzofunan derivatives (**27** — **32**) were identified in the species of *Tagetes* L. Their names, structures, and resources were listed in Table 1. The structures of the compounds were determined by spectral analysis and compared with published data. The molecular structure of isoeuparin (**27**) was characterized by single crystal X-ray diffraction. What is interesting is that the identified benzofunan compounds only exist in *T. patula*. It may provide evidence to the plant taxonomy in the species of *Tagetes* L.

Terpenoids and steroids

Terpenoids (**33**—**44**) were found and isolated from *T. erecta.* Auto-oxidation often occurred among compounds **37** — **42** (Faizi and Naz, 2004). Auto-oxidation converted compound **37** into compounds **38** and **40**, and transformed compound **38** into compound **42** (Faizi and Naz, 2004). Compound **44**, the only sesquiterpenoid, was also isolated from *T. erecta.* However, five steroids, compounds **45**—**49**, were also isolated from *T. erecta* and *T. patula*.

Alkaloids

To date, two alkaloids (**50**—**51)** are found from this genus. Jafrine (**50**) is an inherently unstable and structurally novel tetrahydro-β-carboline alkaloid (Faizi and Naz, 2002). Auto-oxidation made jafrine transform into 2-acetyl tryptamine derivatives (Faizi and Naz, 2002). Liu, Su, and Wang (2007) reported that alkaloids with different polarity indeed existed in the roots of *T. erecta*. However, the chemical components have not been investigated, and further investigation is called for.

Flavonoids

Flavonoids are the main components within the genus *Tagetes* L., and may have the meaning of chemosystematic interpretations in some extent. A total of forty-nine flavonoids (**52**—**100**) have been identified from the genus *Tagetes* L. Flavonoids exist within this genus in the free or glycoside form. This class within the genus could be divided into two subgroups, flavone derivatives and flavonol derivatives.

Flavone derivatives Only three flavone derivatives have been isolated from the genus. They are luteolin (**52**) from *T. multiflora* Kunth and *T. rupestris* Cabrera, luteolin 7-*O*-glucoside (**53**) from *T. multiflora*, and chrysocriol-7-*O*-(6-*O*-α-*L*-arabinofuranosyl)-β-*D*-glucopyranoside (**54**) from *T. patula.*

Flavonol derivatives Except for compounds **52**—**54**, the flavonoids isolated from the genus belong to the subgroup of flavonol derivatives. All of them bore oxygen functions at the positions C-5, 7, and 4′.

However, C-2′, 6′, and 8 have never been functionalized. When it comes to their glycosides, the glycose was often attached to positions C-3 and C-7. For instance, quercetagetin (**55**), isolated from six species, is the representative with six hydroxyls at positions C-3, 5, 6, 7, 3′, and 4′, respectively. Its monosubstitued methyl derivatives (**56** — **57**), disubstitued methyl derivatives (**58** — **63**), and trisubstitued methyl derivatives (**64**—**66**), as well as its glycosides with the glucose at the position C-3 or 6 or 7, are also found in the genus. Moreover, other glycosides, substituted at position C-7 of quercetagetin, such as protocatechuic acid glycoside (**70**), coumaric acid glycoside (**71**), and caffeic acid glycoside (**72**) have also been isolated from the genus. The only diglucoside is quercetin-3-diglucoside (**96**), which was isolated from *T. coronopifolia* Willd. Quercetin (**89**) could be changed into quercetagetin (**55**) by a-ring specific hydroxylation in position C-6, which is catalyzed by a cytochrome P450 dependent mono-oxygenase (Halbwirth, Forkmann, and Stich, 2004).

Carotenoids

Carotenoids are the important components from the petals extracts in the species of *Tagetes* L. Carotenoids within this genus mainly composed of all-*trans*-lutein (**101**), β-carotene (**109**), zeaxanthin (**110**), and lutein esters. Lutein, bearing one hydroxyl group at each ionone ring, could be esterified with the saturated fatty acids, resulting in mono- and diacylated derivatives, such as lutein dipalmitate diesters (**102**), lutein myristate palmitate diesters (**103**), and lutein violaxanthin monoesters (**107**).

Others

In addition to the types mentioned above, other compounds have also been isolated from plants in *Tagetes* L. (**111** — **126**). The compound names and sources listed in Table 1 and the structures of 126 compounds are shown in Fig. 1.

Table 1 Compounds in plants of *Tagetes* L.

No.	Compounds	Plant resources	References
Phenolic derivatives			
1	syringic acid	*T. erecta*	(Huang, Zhou, and Wang, 2006)
2	3,4-dihydroxybenzonicacid	*T. erecta*	(Huang, Zhou, and Wang, 2006)
3	gallic acid	*T. erecta*	(Huang, Zhou, and Wang, 2007)
4	3,4-dihydroxy-5-methoxy-benzoicacid	*T. erecta*	(Huang, Zhou, and Wang, 2007)
Phenylpropanoids			
5	caffeic acid-*O*-glucoside	*T. maxima*	(Irene *et al*, 2004)
6	rosmarinic acid	*T. maxima*	(Irene *et al*, 2004)
7	neochlorogenic acid	*T. maxima*	(Irene *et al*, 2004)
8	chlorogenic acid	*T. maxima*	(Irene *et al*, 2004)
9	7-methoxy coumarin	*T. lucida*	(Oranday *et al*, 2008)
10	scopoletine	*T. lucida*	(Oranday *et al*, 2008)
Thiophene derivatives			
11	α-terthieny1	*T. erecta*	(Vasudevan, Kashyap, and Sharma, 1997; Xu, Wang, and Shi, 2011)
		T. patula	(Vasudevan, Kashyap, and Sharma, 1997)
		T. tenuifolia	(Vasudevan, Kashyap, and Sharma, 1997)
		T. microglossa	(Vasudevan, Kashyap, and Sharma, 1997)
		T. jalisciencis	(Vasudevan, Kashyap, and Sharma, 1997)
12	5-methyl-2,2′-5′,2″-terthiphene	*T. minuta*	(Wang *et al*, 2002)
13	5-butyl-2,2′-bithienyl	*T. erecta*	(Vasudevan, Kashyap, and Sharma, 1997)
14	5-(but-l-chloro-2-ol-3-ynyl)-2,2′-bithienyl	*T. minuta*	(Wang, Xu, and Zhao, 2002)
15	5-(but-l-ol-3-ynyl)-2,2′-bithienyl	*T. minuta*	(Wang, Xu, and Zhao, 2002)
16	5-(3,4-diacetoxy-1-butynyl)-2,2′-bithiophene	*T. patula*	(Menelaou *et al*, 1991)
17	5-(4-acetoxy-l-butynyl)-2,2′-bithiophene	*T. patula*	(Menelaou *et al*, 1991)

(To be continued)

(Continued Table 1)

No.	Compounds	Plant resources	References
18	5,5′-dimethyl-2,2′-bithienyl	*T. erecta*	(Wang, Xu, and Zhao, 2002)
19	5-vinyl-2,2′-bithiophene	*T. erecta*	(Wang, Xu, and Zhao, 2002)
20	5-methyl-5′-vinyl-2,2′-bithiophene	*T. minuta*	(Wang, Xu, and Zhao, 2002)
21	5′-hydroxymethyl-5-(3-butene-1-ynyl)-2,2′-bithiophene	*T. patula*	(Bano *et al*, 2002)
22	5-(3-butene-1-ynyl)-2,2′-bithiophene	*T. patula*	(Menelaou *et al*, 1991; Parodi, Fischer, and Flores, 1988)
		T. erecta	(Wang, Xu, and Zhao, 2002)
		T. minuta	(Wang, Xu, and Zhao, 2002)
23	5′-methyl-5-(3-buten- 1-ynyl)-2,2′-bithiophene	*T. patula*	(Vasudevan, Kashyap, and Sharma, 1997)
24	5′-methyl-5-[4-(3-methyl-1-oxobutoxy)-1-butynyl]-2,2′-bithiophene	*T. patula*	(Bano *et al*, 2002)
25	*cis*-5-(1-acetoxy-but-3-enyl)-2,2′-bithienyl	*T. patula*	(Atkinson, Curtis, and Phillips, 1964)
		T. erecta	(Atkinson, Curtis, and Phillips, 1964)
26	1-[5′-(1-propyn-1-yl)-[2,2′-bithiophen]-5-yl]-ethanone	*T. erecta*	(Xu, Wang, and Shi, 2011)
Benzofunan derivatives			
27	isoeuparin	*T. patula*	(Parodi, Fischer, and Flores, 1988)
28	dehydrotremetone	*T. patula*	(Menelaou *et al*, 1991)
29	14-isobutyryloxyeuparin	*T. patula*	(Menelaou *et al*, 1991)
30	2,3-dihydro-14-isobutyryloxyeuparin	*T. patula*	(Menelaou *et al*, 1991)
31	hydroxytremetone	*T. patula*	(Sűtfeld, Balza, and Towers, 1985; Tang, Wat, and Towers, 1987)
32	euparin	*T. patula*	(Sűtfeld, Balza, and Towers, 1985; Tang, Wat, and Towers, 1987)
Terpenoids			
33	β-amyrin	*T. erecta*	(Huang, Zhou, and Wang, 2006)
34	lupeol	*T. patula*	(Bano *et al*, 2002)
35	erythrodiol	*T. erecta*	(Xu, Wang, and Shi, 2011)
36	erythrodiol 3-palmitate	*T. erecta*	(Faizi and Naz, 2004)
37	3-*O*-[(9*Z*)-hexadec-9-enoyl]erythrodiol	*T. erecta*	(Faizi and Naz, 2004)
38	3-*O*-[(9*Z*)-hexadec-9-enoyl]-β-amyrin	*T. erecta*	(Faizi and Naz, 2004)
39	11α,12α:13β,28-diepoxyoleanan-3β-yl (9*Z*)-hexadec-9-enoate	*T. erecta*	(Faizi and Naz, 2004)
40	13β,28-epoxyolean-11-en-3β-yl (9*Z*)-hexadec-9-enoate	*T. erecta*	(Faizi and Naz, 2004)
41	28-hydroxy-11-oxoolean-12-en-3β-yl (9*Z*)-hexadec-9-enoate	*T. erecta*	(Faizi and Naz, 2004)
42	11-oxoolean-12-en-3β-yl (9*Z*)-hexadec-9-enoate	*T. erecta*	(Faizi and Naz, 2004)
43	dammarenediol 3-*O*-*n*-palmitate	*T. erecta*	(Huang, Zhou, and Wang, 2007)
44	oplodiol	*T. erecta*	(Xu, Wang, and Shi, 2011)
Steroids			
45	β-sitosterol	*T. erecta*	(Huang, Zhou, and Wang, 2006; Xu, Wang, and Shi, 2011)
46	β-daucosterol	*T. erecta*	(Xu, Wang, and Shi, 2011)

(To be continued)

(Continued Table 1)

No.	Compounds	Plant resources	References
47	7β-hydroxysitosterol	*T. erecta*	(Xu, Wang, and Shi, 2011)
48	stigmasterol	*T. erecta*	(Huang, Zhou, and Wang, 2006)
49	cholesterol	*T. patula*	(Bano *et al*, 2002)
Alkaloids			
50	jafrine	*T. patula*	(Faizi and Naz, 2002)
51	6-ethoxy-2,4-dimethylquinoline	*T. erecta*	(Xu, Wang, and Shi, 2011)
Flavonoids			
52	luteolin	*T. multiflora*	(Abdala, 2003)
		T. rupestris	(Vasudevan, Kashyap, and Sharma, 1997)
53	luteolin-7-*O*-glucoside	*T. multiflora*	(Abdala, 2003)
54	chrysocriol-7-*O*-(6-*O*-α-*L*-arabinofuranosyl)-β-*D*-glucopyranoside	*T. patula*	(Wang, Xu, and Zhao, 2002)
55	quercetagetin	*T. erecta*	(Huang, Zhou, and Wang, 2006; Vasudevan, Kashyap, and Sharma, 1997; Xu, Wang, and Shi, 2011)
		T. patula	(Vasudevan, Kashyap, and Sharma, 1997)
		T. multiflora	(Abdala, 2003)
		T. tenuifolia	(Abdala, 2001)
		T. stenophylla	(Abdala, 2000)
		T. rupestris	(Vasudevan, Kashyap, and Sharma, 1997)
56	quercetagetin-7-methyl ether	*T. erecta*	(Huang, Zhou, and Wang, 2006; Xu, Wang, and Shi, 2011)
57	quercetagetin-5-methyl ether	*T. erecta*	(Huang, Zhou, and Wang, 2007)
		T. patula	(Bhardwaj *et al*, 1980)
58	quercetagetin-5,7-dimethl ether	*T. erecta*	(Huang, Zhou, and Wang, 2007)
59	quercetagetin-3,6-dimethylether	*T. maxima*	(Irene *et al*, 2004)
60	quercetagetin-4′,6-dimethylether	*T. maxima*	(Irene *et al*, 2004)
61	quercetagetin-4′,6-dimethyl-*O*-hexoside	*T. maxima*	(Irene *et al*, 2004)
62	quercetagetin-3′,6-dimethylether	*T. maxima*	(Irene *et al*, 2004)
63	quercetagetin-3′,6-dimethyl-*O*-hexoside	*T. maxima*	(Irene *et al*, 2004)
64	quercetagetin-3,6,3′-trimethylether	*T. maxima*	(Irene *et al*, 2004)
65	quercetagetin-3,6,4′-trimethylether	*T. maxima*	(Irene *et al*, 2004)
66	quercetagetin-3,6,4′-trimethyl-7-*O*-glucoside	*T. maxima*	(Irene *et al*, 2004)
67	quercetagetin 3-*O*-glucoside	*T. erecta*	(Huang, Zhou, and Wang, 2007; Wang, Xu, and Zhao, 2002; Yang *et al*, 2003)
		T. gracilis	(Abdala, 2003)
68	quercetagetin-7-*O*-glucoside	*T. erecta*	(Irene et al, 2004; Xu, Wang, and Shi, 2011)
		T. patula	(Vasudevan, Kashyap, and Sharma, 1997)
		T. gracilis	(Abdala, 2003)
		T. coronopifolia	(Abdala, 2003; 1997)
		T. maxima	(Parejo *et al*, 2005)
69	quercetagetin 6-*O*-β-*D*-glucopyranoside	*T. mandonii*	(Dagostino *et al*, 1997)
70	quercetagetin-*O*-procatechuoylhexoside	*T. maxima*	(Irene *et al*, 2004)
71	quercetagetin-7-*O*-(6-*O*-coumaroyl-β-*D*-glucopyranoside)	*T. maxima*	(Irene *et al*, 2004; Parejo *et al*, 2005)

(To be continued)

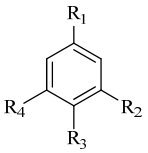
(Continued Table 1)

No.	Compounds	Plant resources	References
72	quercetagetin-7-*O*-(6-*O*-caffeoyl-β-*D*-glucopyranoside)	*T. maxima*	(Irene *et al*, 2004; Parejo *et al*, 2005)
73	quercetagetin-7-*O*-(6-*O*-galloyl-β-*D*-glucopyranoside)	*T. maxima*	(Irene *et al*, 2004; Parejo *et al*, 2005)
74	kaempferol	*T. patula*	(Wang, Xu, and Zhao, 2002)
		T. erecta	(Wang, Xu, and Zhao, 2002; Xu,Wang, and Shi, 2011)
		T. maxima	(Irene *et al*, 2004)
		T. tenuifolia	(Abdala, 2001)
75	kaempferitrin	*T. patula*	(Vasudevan, Kashyap, and Sharma, 1997)
		T. erecta	(Vasudevan, Kashyap, and Sharma, 1997)
		T. tenuifolia	(Abdala, 2001)
		T. rupestris	(Vasudevan, Kashyap, and Sharma, 1997)
76	kaempferol-*O*-caffeoylhexoside	*T. maxima*	(Irene *et al*, 2004)
77	6-hydroxykaempferol-7-*O*-(6-*O*-caffeoyl-β-*D*-glucopyranoside)	*T. maxima*	(Irene *et al*, 2004)
78	6-hydroxykaempferol-*O*-galloylhexoside	*T. maxima*	(Irene *et al*, 2004)
79	6-hydroxykaempferol-*O*-hexoside	*T. maxima*	(Irene *et al*, 2004)
80	myricetin	*T. patula*	(Vasudevan, Kashyap, and Sharma, 1997; Wang, Xu, and Zhao, 2002)
81	myricetin 7-*O*-glucoside	*T. coronopifolia*	(Vasudevan, Kashyap, and Sharma, 1997)
82	myricetin 3-*O*-glucoside	*T. coronopifolia*	(Abdala, 2003; Vasudevan, Kashyap, and Sharma, 1997)
83	patuletin	*T. maxima*	(Guinot *et al*, 2008)
		T. patula	(Vasudevan, Kashyap, and Sharma, 1997)
		T. stenophylla	(Abdala, 2000)
84	patulitrin	*T. maxima*	(Guinot *et al*, 2008)
		T. patula	(Vasudevan, Kashyap, and Sharma, 1997)
		T. stenophylla	(Abdala, 2000)
85	patuletin-*O*-pentoside	*T. maxima*	(Irene *et al*, 2004)
86	patuletin-*O*-caffeoylhexoside	*T. maxima*	(Irene *et al*, 2004)
87	patuletin-*O*-galloylhexoside	*T. maxima*	(Irene *et al*, 2004)
88	patuletin-*O*-hexoside	*T. maxima*	(Irene *et al*, 2004)
89	quercetin	*T. erecta*	(Irene *et al*, 2004)
		T. tenuifolia	(Abdala, 2001)
		T. rupestris	(Vasudevan, Kashyap, and Sharma, 1997)
90	quercetin-3-*O*-galactoside	*T. maxima*	(Irene *et al*, 2004)
91	quercetin-3-*O*-pentoside	*T. maxima*	(Irene *et al*, 2004)
92	quercetin-3-*O*-glucoside	*T. maxima*	(Irene *et al*, 2004)
		T. tenuifolia	(Abdala, 2001)
		T. coronopifolia	(Abdala, 2003; 1997)
93	quercetin 3-*O*-rhamnoside	*T. tenuifolia*	(Abdala, 2001)
94	quercetin-7-*O*-glucoside	*T. gracilis*	(Abdala, 2003)
		T. multiflora	(Abdala, 2003)
95	quercetin-5-*O*-glucoside	*T. multiflora*	(Abdala, 2003)
96	quercetin 3-diglucoside	*T. coronopifolia*	(Abdala, 2003; 1997)

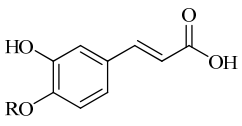
(To be continued)

(Continued Table 1)

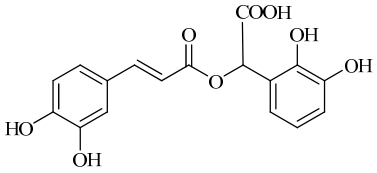
No.	Compounds	Plant resources	References
97	isorhamnetin	*T. maxima*	(Irene *et al*, 2004)
		T. stenophylla	(Abdala, 2000)
98	isorhamnetin-7-*O*-galactoside	*T. patula*	(Tripathi, Paliwal, and Singh, 1991)
99	isorhamnetin-3-*O*-galactoside	*T. stenophylla*	(Abdala, 2000)
		T. maxima	(Irene *et al*, 2004)
100	isorhamnetin-7-*O*-glucoside	*T. stenophylla*	(Abdala, 2000)
Carotenoids			
101	lutein	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
		T. patula	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
102	lutein dipalmitate diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
103	lutein myristate palmitate diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
104	lutein lauristate myristate diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
105	lutein dimyristate diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
106	lutein palmitate stearistate diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
107	lutein violaxanthin monoesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
108	lutein neoxanthin violaxanthin diesters	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
109	β-carotene	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
110	zeaxanthin	*T. erecta*	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
		T. patula	(Breithaupt, Wirt, and Bamedi, 2001; Scalia and Francis, 1989)
Others			
111	γ,η-dimethyl-$\triangle^{\alpha}$-octen-2-one	*T. glandulifera*	(Jones and Smith, 1925)
112	tagetone	*T. glandulifera*	(Jones and Smith, 1925)
113	β-farnesene	*T. patula*	(Menelaou *et al*, 1991)
114	16*Z*,19*Z*-pentacosadienoic acid	*T. erecta*	(Huang, Zhou, and Wang, 2007)
115	monolinoleoyl glycerol	*T. erecta*	(Huang, Zhou, and Wang, 2007)
116	uracil	*T. erecta*	(Yang *et al*, 2003)
117	mannitol	*T. erecta*	(Yang *et al*, 2003)
118	9*Z*,12*Z*,15*Z*-octadectrien-1-ol	*T. erecta*	(Huang, 2007)
119	*n*-hexadecane	*T. erecta*	(Huang, 2007)
120	*n*-tetratriacontane	*T. erecta*	(Huang, 2007)
121	3-α-galactosyl disyringic acid	*T. erecta*	(Huang, 2007)
122	3-β-galactosyl disyringic acid	*T. erecta*	(Huang, 2007)
123	vitamin E	*T. erecta*	(Huang, Zhou, and Wang, 2007)
124	(3*S*,6*R*,7*E*)-hydroxy-4,7-megastigmadien-9-one	*T. erecta*	(Xu, Wang, and Shi, 2011)
125	palmitin	*T. erecta*	(Xu, Wang, and Shi, 2011)
126	ethylene glycol linoleate	*T. erecta*	(Xu, Wang, and Shi, 2011)



	R_1	R_2	R_3	R_4
1	COOH	OMe	OH	OMe
2	COOH	H	OH	OH
3	COOH	OH	OH	OH
4	COOH	OMe	OH	OH



5 R=Glc



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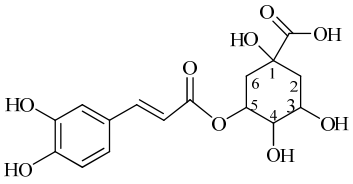
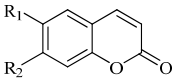
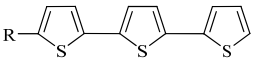


7

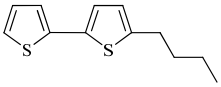
8 1*R*, 3*R*, 4*S*, 5*R*



9 R_1=H R_2=OMe
10 R_1=OMe R_2=OH



11 R=H
12 R=Me



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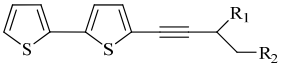
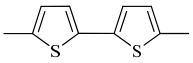


14 R_1=OH R_2=Cl
15 R_1=H R_2=OH
16 R_1=R_2=OAc
17 R_1=H R_2=OAc



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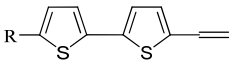
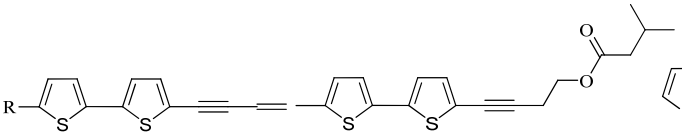
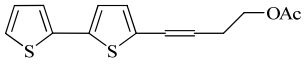


19 R=H
20 R=Me



21 R=CH_2OH
22 R=H
23 R=Me

24



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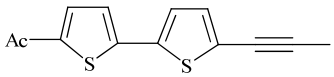

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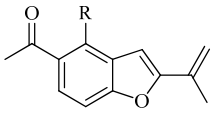
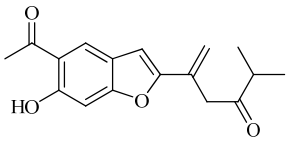
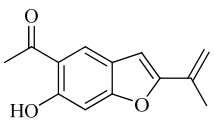


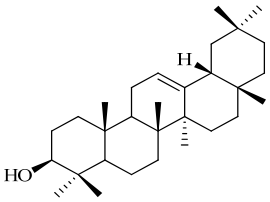
27 R=OH
28 R=H



29 2,3-dihydro
30



31 2,3-dihydro
32



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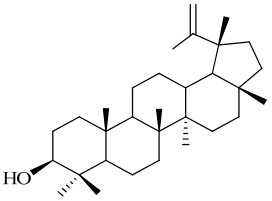

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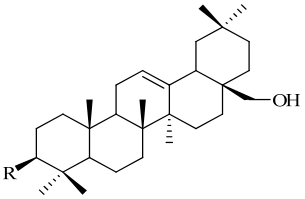
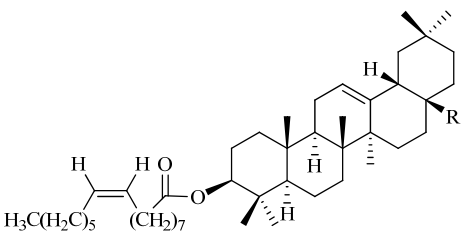
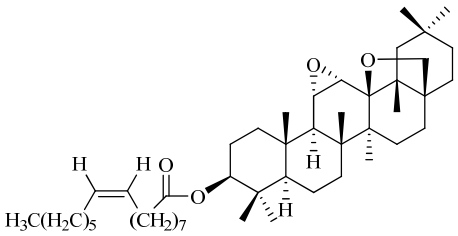


35 R=OH
36 R=$OCO(CH_2)_{14}CH_3$



37 R=CH_2OH
38 R=Me



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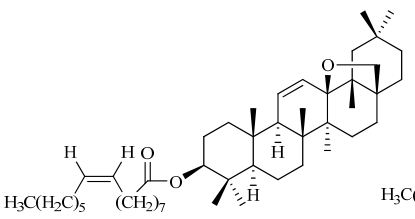
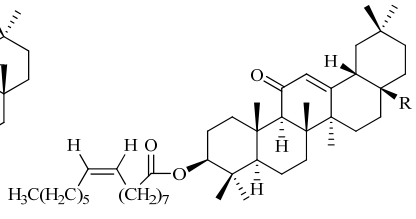
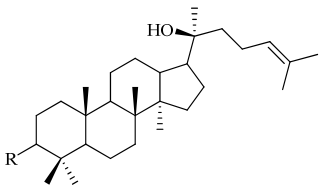


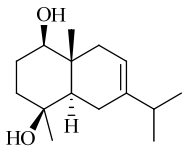
40



41 R=CH_2OH
42 R=Me



43 R=$OCO(CH_2)_{14}CH_3$



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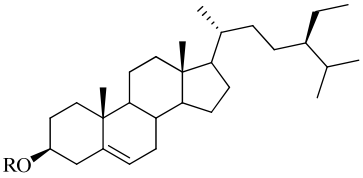
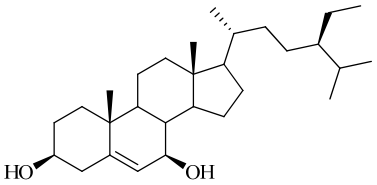


45 R=H
46 R=Glc



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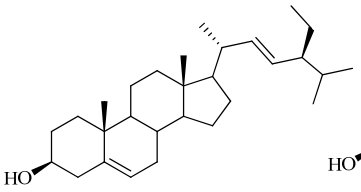

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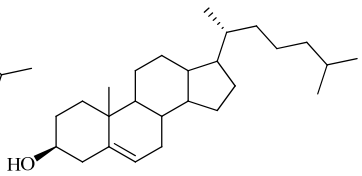

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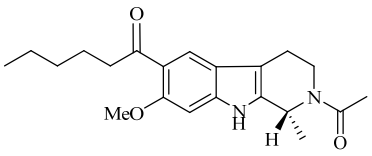

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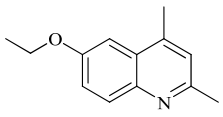

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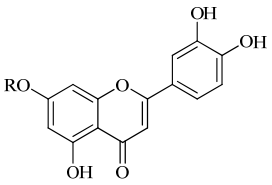
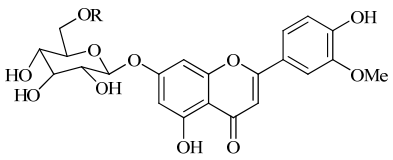
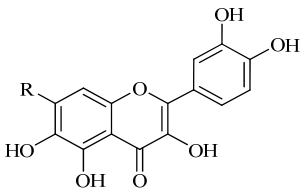


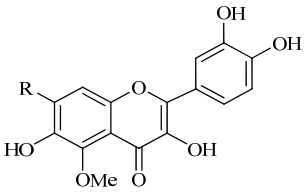
52 R=H
53 R=Glc



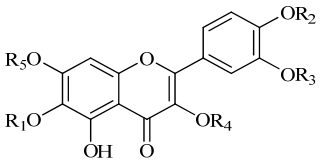
54 R=α-*L*-arabinofuranose



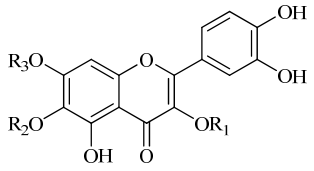
55 R=OH
56 R=OMe



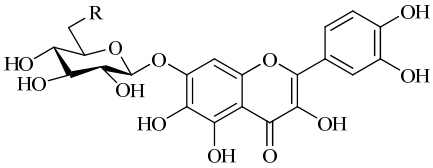
57 R=OH
58 R=OMe



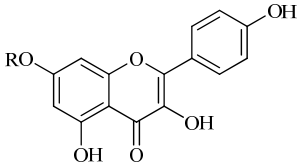
59 R_1=R_4=Me R_2=R_3=R_5=H
60 R_1=R_2=Me R_3=R_4=R_5=H
61 R_1=R_2=Me R_3=R_4=H R_5=hexose
62 R_1=R_3=Me R_2=R_4=R_5=H
63 R_1=R_3=Me R_2=R_4=H R_5=hexose
64 R_1=R_3=R_4=Me R_2=R_5=H
65 R_1=R_2=R_4=Me R_3=R_5=H
66 R_1=R_2=R_4=Me R_3=H R_5=Glc



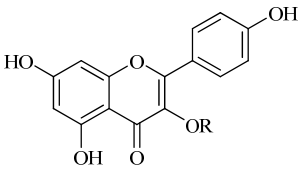
67 R_1=Glc R_2=R_3=H
68 R_1=R_2=H R_3=Glc
69 R_1=R_3=H R_2=Glc



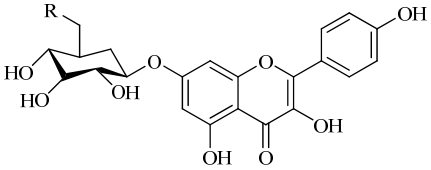
70 R=protocatechuic acid
71 R=coumaric acid
72 R=caffeic acid
73 R=galic acid



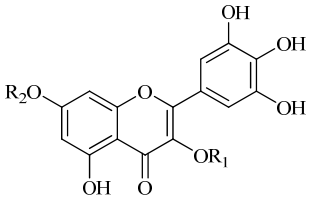
74 R=H
75 R=Glc



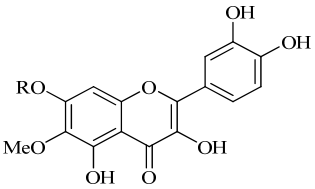
76 R=caffcoylhexose



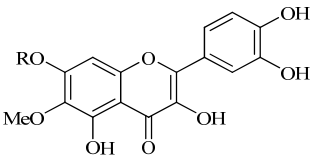
77 R=caffeic acid
78 R=galic acid
79 R=H



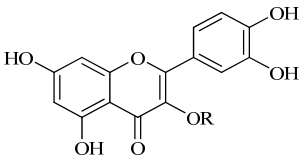
80 R_1=R_2=H
81 R_1=H R_2=Glc
82 R_1=Glc R_2=H



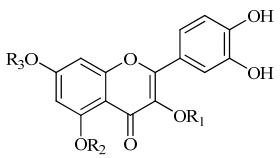
83 R=H
84 R=Glc



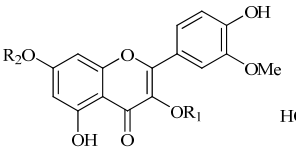
85 R=pentose
86 R=caffeoylhexose
87 R=galloylhexose
88 R=hexose



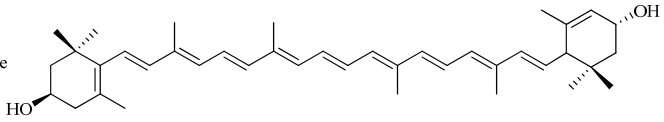
89 R=H
90 R=galactose
91 R=pentose
92 R=Glc
93 R= rhamnose



94 R_1=R_2=H R_3=Glc
95 R_1=R_3=H R_2=Glc
96 R_1=Glc-Glc R_2=R_3=H



97 R_1=R_2=H
98 R_1=H R_2=galactose
99 R_1=galactose R_2=H
100 R_1=H R_2=Glc



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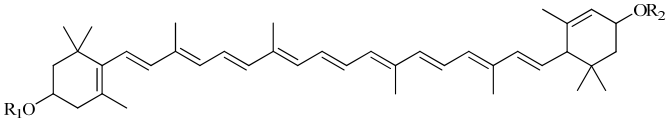
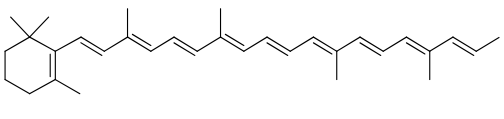


102 R_1=$C_{15}H_{31}CO$ R_2=$C_{15}H_{31}CO$
103 R_1=$C_{13}H_{25}CO$ R_2=$C_{15}H_{31}CO$
104 R_1=$C_{11}H_{23}CO$ R_2=$C_{13}H_{25}CO$
105 R_1=$C_{13}H_{25}CO$ R_2=$C_{13}H_{25}CO$
106 R_1=$C_{15}H_{31}CO$ R_2=$C_{17}H_{33}CO$
107 R_1=H R_2=violaxanthin
108 R_1=neoxanthin R_2=violaxanthin



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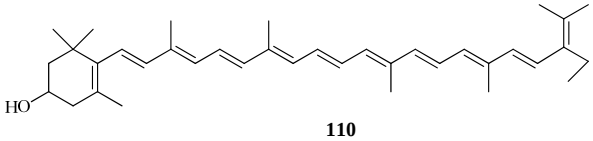

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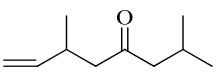

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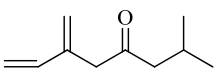

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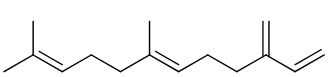

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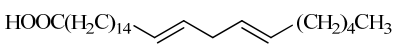

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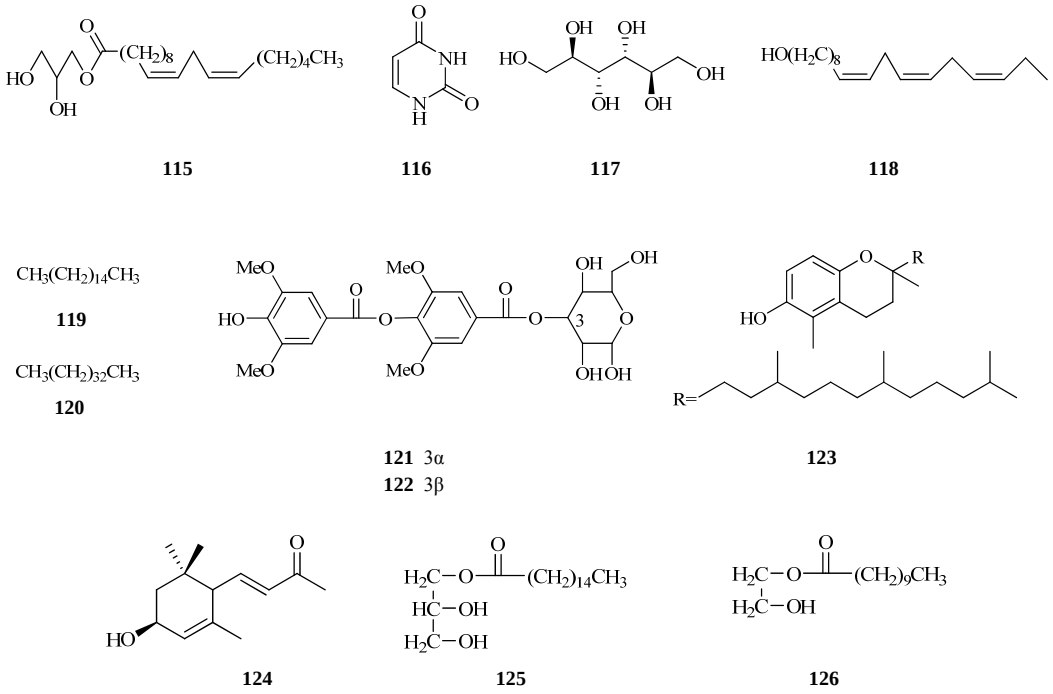

Fig. 1 Structures of compounds in plants of *Tagetes* L.

Biological Activities

Agricultural application

The nematocidal, fungicidal, and insecticidal activities of the species of *Tagetes* L. and the original activity found by different groups have been reviewed by Vasudevan, Kashyap, and Sharma (1997). Many relative studies have been reported in recent years.

Nematocidal activity The activity of the plants in *Tagetes* L. is mainly origined from thiophene derivatives. α-Terthienyl is the most effective compound compared with the other thiophene derivatives and the blank control (Vasudevan, Kashyap, and Sharma, 1997). The test of cold aqueous extracts from African marigold (*T. erecta*) for controlling tomato root knot nematode diseases showed that the whole plant extracts of *T. erecta* were more efficacious than stem extracts, and both of them were more effective than root extracts. Plant height, leaf number, and fruit yield were all better than the blank control. It is possible that *T. erecta* will be an alternative crop for intercropping (Natarajan *et al*, 2006), and it may be used as a kind of effective natural insecticide in the future.

Antifungicidal activity

The methanol extract of *T. patula* flowers was found to possess antimicrobial activity against a number of bacteria in the preliminary assay, the bioassay-guided fractionation of which led to the isolation of patuletin (**83**) as the active antibacterial principle with a minimum inhibitory concentration (MIC) value of 12.5 g/disk against *Corynebacterium* spp., *Staphylococcus* spp., *Streptococcus* spp., and *Micrococcus luteus*. Its glucoside, patulitrin (**84**), was found to be weakly activated. The antimicrobial activity of the extract from *T. patula* was reported for the first time (Faizi *et al*, 2008). The methanol extracts obtained from ten cultivars of *T. patula* were also assayed on two phytopathogenic fungi, *Botrytis cinerea* Pers. and *Fusarium moniliforme* Sheldon. The extract showed a high dose-dependent inhibition on *B. cinerea*, with remarkable difference between light and dark treatment (Mares *et al*, 2002). The scanning electron microscopy (SEM) and transmission electron microscopy (TEM) observations of *Pythium ultimum* revealed that *T. patula* extract induced alterations on cell fungal membranes with a photoactivation mechanism (Mares *et al*, 2004). The antifungicidal activity of *T. patula* was also reported by Chinese workers (Chen, Wang, and Zhang, 2004; Wei, 2005; Wang and Guo, 2004). These results gave us a clue that the compounds existing in *T. patula* especially the flavonoids, might be developed as fungicide in the future, and they might be effective on more fungi.

The essential oils of *T. minuta* obtained by hydrodistillation were found to have antibacterial activity, especially against Gram-positive bacteria. The MICs for the oil from UK greenhouse-grown plants were 6.25－25 g/mL for Gram-positive bacteria and 25－50 g/mL for Gram-negative bacteria, with the lowest MIC of 6.25 g/mL against *Streptococcus faecalis*. Oil obtained from plants from South Africa had MICs of 50 － 100 g/mL against Gram-positive and Gram-negative bacteria (Senatore *et al*, 2004). The plants from different regions have significantly different effects towards the same bacteria. It may provide us information about the plant cultivation. Using a serial microdilution assay, it was reported that the aqueous and organic extracts of *T. minuta* had the antifungal activity against isolates of four agriculturally important fungi (Thembo *et al*, 2010). All extracts except for the water extracts showed growth inhibitory activity against most isolates of the *Fusarium* spp. No inhibition of *Aspergillus* spp. tested was observed, but conidium formation was stimulated on plates treated with the plant extracts when visually compared to the growth controls. The results indicated that chemical constituents from these species may be developed as potential agrochemical fungicides.

Five successive extracts from the roots of *T. erecta* were evaluated for *in vitro* antimicrobial activity against seven microbial strains (Gupta and Vasudeva, 2010). All extracts exhibited significant antimicrobial activity against three Gram-positive and two Gram-negative bacteria and two fungal strains with MIC values ranging 12.5 － 100 μg/mL. The observation provides support for the ethnobotanical use of the plant.

Insecticidal activity

Essential oils extracted from genotype 1 of *T. minuta* (TM-1), genotype 2 of *T. patula* (TP-2), and genotype 13 of *T. erecta* (TE-13) were the most toxic against three stored-product beetle species, *Callosobruchus maculatus* (Fabricius), *Sitophilus oryzae* (Linnaeus), and *Tribolium castaneum* (Herbst). These three genotypes were then evaluated for adult toxicity, oviposition deterrence, and ovicidal and population reduction activities. Essential oils of genotypes TM-1 and TP-2 induced 100% adult mortality for all the three insect species at doses of 5% and 500 μg/insect in fumigant and contact toxicity bioassays, respectively (Krishna *et al*, 2005). Two bioassays were carried out to evaluate the effect of the essential oils of *T. patula* on the behavior and insecticide activity on adults of *S. zeamais.* The results showed that this essential oil (10 μL) was efficient to control adults of *S. zeamais* (Restello, Menegatt, and Mossil, 2009).

The crude fractions in *n*-hexane and ether of the seeds of *T. minuta* (Shahzadi *et al*, 2010) growing in north of Pakistan were applied to three species of common grain pests, *Tribolium castaneum*, *Rhyzopertha dominica*, and *Callosobruchus analis* F. Insecticidal activity (similar to 70%) was observed for both *n*-hexane and ether fractions against common grain pests especially for *R. dominica*.

The insecticidal activity of the methanol and ethyl acetate extracts of *T. minuta* L. (Ireri *et al*, 2010) were investigated against *Phlebotomus duboscqi* Neveu-Lemaire at the doses of 2.5, 5, and 10 mg/mL. The extracts had the significant mortality rate ($P < 0.05$) in both male and female bioassays but were not significantly different between sexes. The different concentration used showed significantly different mortality rates and 10 mg/mL was the most effective concentration. These extracts were found to be insecticidal to adult sandflies.

Herbicidal activity

The study on the potential herbicidal activity of *T. minuta* leaf powder (at 0.1, 0.2, and 0.4 kg/m^2) towards two invasive weeds *Echinochloa crus-galli* and *Cyperus rotundus* of rice fields noted that *T. minuta* leaf powder applied to rice field soil significantly reduced emergence and growth of both the weed species in pots under greenhouse and in rice field plots. At 0.1 and 0.2 kg/m^2 doses of application, yield of rice increased significantly and the effect was similar to that observed with the herbicidal application under field conditions. It could be concluded that *T. minuta* possesses weed-suppressing ability and could be used as a natural herbicide (Batish *et al*, 2007).

Antimalarial activity

Five successive extracts from the roots of *T. erecta* (Gupta and Vasudeva, 2010) were evaluated for *in vitro* antiplasmodial activity against chloroquine sensitive and resistant strains of *Plasmodium falciparum* by the schizont maturation inhibition assay. The ethyl acetate fraction exhibited significant antiplasmodial efficacy with the half maximal inhibitory concentration (IC_{50}) of

0.02 and 0.07 mg/mL against the chloroquine sensitive and resistant strains of *Plasmodium falciparum*, respectively.

The antimalarial activity of the solvent extraction from the seeds of *T. erecta* (Shahzadi *et al*, 2010) growing in north of Pakistan was reported. The results indicated that the *n*-hexane fraction showed significantly better results than ether-fraction for antimalarial activity.

Anti-oxidative activity

The flavonoids and carotenoids isolated from the species of *Tagetes* L. led to the anti-oxidative activity. Parejo *et al* (2005) reported the anti-oxidative activity of acylated quercetagetin glycosides from *T. maxima*. The anti-oxidative activity of the acylated quercetagetin glycosides by checking the scavenging activity against three radicals, 2,2-diphenyl-1-picrylhydrazyl free radical (DPPH·), hydroxyl (·OH), and superoxide showed that all the quercetagetin-type flavonoids as well as the 6-hydroxykaempferol derivatives exhibited higher radical scavenging activity in all the tests, in comparison with that of the standards used. Quercetagetin-7-*O*-(6-*O*-caffeoyl-β-*D*-glucopyranoside) (**72**) was the most active, as its activity to DPPH· was about 2.73 ± 0.04, to ·OH was about 1.10 ± 0.08, and to superoxide was about 89.28 ± 0.61. These results exhibited that compound **72** had higher anti-oxidative activity than the referenced compounds (Parejo *et al*, 2005). The anti-oxidative activity of lutein (**101**) was examined by using the photochemiluminescence (PCL) assay and the β-carotene-linoleic acid model system ([β]-CLAMS). Lutein showed greater anti-oxidative activity than the other two common carotenoids, β-carotene (**109**) and lycopene (Li *et al*, 2007; Li, 2001). The anti-oxidative activity mechanism of β-carotene was reported. Using the electron-spin resonance (ESR) spectroscopy coupled to the spin-trapping technique, they observed the first direct evidence that carotenoids quenched peroxyl radicals (Iannone *et al*, 1998). Li *et al* (2007) also reported the anti-oxidative activity of phenols, flavonoids, and lutein esters by the radical-scavenging test. The essential oil from flowers of *T. erecta* was evaluated for anti-oxidative activity *in vitro* using DPPH·, the thiocyanate, β-carotene bleaching, free radical scavenging activity, and oxidation of deoxyribose assay (Martha *et al*, 2006).

Antimutagenic activity

The highest amount of lutein was found in natural plants of *Tagetes* L. High dietary intake of lutein has been associated with risk reduction of many chronic diseases, including age-related macular degeneration (AMD), cancer, and cardiovascular diseases. Lutein in food is generally regarded as safety (Wang *et al*, 2006). The mutagenicity and antimutagenicity of lutein at 334, 668, and 1335 μg/plate were examined using the standard Ames test in the presence and absence of S9 mix. Lutein was not only found to be non mutagenic at all doses, but it also showed an antimutagenic effect in a dose-dependent manner. Similar results were found in a chromosome aberration test using the Chinese hamster ovary cells for the evaluation of clastogenicity and anti-clastogenicity of lutein at doses of 66.8, 133.5, and 267.0 mg/L (Wang *et al*, 2006). The effect of xanthophylls extracted from *T. erecta* on the AFB_1 mutagenicity was observed by the salmonella plate incorporation test, using strain YG1024. The effect of lutein on the DNA-repair system in YG1024 was investigated by a pre-incubation test. Pure lutein and xanthophylls inhibited the mutagenicity of AFB_1 in a dose-dependent manner. The percentages of the inhibition on the AFB_1 mutagenicity were 37% and 76% for lutein, xanthophyl plus at the dose of 2 μg/plate, respectively. Lutein had a modest effect on the DNA-repair system of YG1024. The results showed the possible inhibitory mechanism of lutein against the AFB_1 mutagenicity (Gonzalez de Mejia, Ramos-Gomez, and Loarca-Pina, 1997).

Other activities

T. erecta could be used as a wound healing agent in buffalo calves and could improve the anti-oxidative action in *D*-galactose induced aged rats (Kumar *et al*, 2006; Pei, Hui, and Dong, 2007). Quercetagetin and patuletin inhibited rat lens aldose reductase (Li *et al*, 1991). *T. minuta* essential oil showed anxiogenic-like effects on T-maze and tonic immobility behavior in domestic chicks (Marin *et al*, 1998). Solvent extracts of *T. erectus* have antinociceptive and anti-inflammatory effects (Shinde *et al*, 2009).

Conclusion

For the chemical researches, it could be concluded that the chemical constituents within the species of *Tagetes* L. exhibited a variety of skeletal arrangements

and some showed significant bioactivities. Some new structures have been reported recently, but there is no report on them. From the bioacvities research, we found that the variety kinds of compounds existed in the genus *Tagetes* L. might have different bioactivies, and some activities might be due to one kind of compounds at a range of doses level. According to the above reports, flavonoids derivatives are rich in *Tagetes* L. Quercetagetin, distributed in six species, as the characteristic of flavonoids, exhibits better anti-oxidative activity. Lutein, mainly distributed in the petals of *T. erecta* and *T. patula* in the form of lutein esters, has high anti-oxidative and antimutagenic activities, additionally, it has high colouring activity. Thiophene derivatives showed better nematocidal activity. No matter what the bioactivities of the medical or the agricultural application, the study on the bioactivity and the relative mechanism of the single compound are limited, it is worthy to research on this aspect further.

Both *T. patula* and *T. erecta* have been intensively studied on their phytochemicals and bioactivities. The organic solvent extracts of the two plants might be developed as agrichemical agents, large work should be done to work it out. The structure-activity relationships of isolated compounds from the species of *Tagetes* L. and their bioactivities have not been observed. In order to search for more potential bioactive species of *Tagetes* L. and components, investigation on lots of other species should be further carried out in phytochemicals and biological activity.

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