Chemical Constituents and Biological Activities of Plants from the genus *Ehretia* Linn.

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Abstract: The plants of the genus *Ehretia* Linn. composed of about 50 species mainly distributed in tropical Asia and Africa. They have been used as folk medicines or traditional tea to treat various ailments in China for a long time. This contribution reviews the chemical constituents isolated from the plants of the genus *Ehretia* Linn. and related biological activities of these species in the past few decades. The compounds in the genus mainly belong to the classes of phenolic acids, flavonoids, benzoquinones, cyanogenetic glycosides, and fatty acids. The main biological activities include antioxidant, anti-inflammatory, antisnake venom, and anti-allergic activities.

Key words: biological activities; chemical constituents; *Ehretia* Linn.
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Introduction

The genus *Ehretia* Linn. (Boraginaceae *s.l.*, subfamily Ehretiaceae) contains about 50 species mainly distributed in tropical Asia and Africa (Gurke, 1893). All species of the genus are arbors or shrubs. The leaves, barks, roots, branches, fruits, and heartwoods are used as the herbal medicines separately (Wang, 1979). In China, the genus *Ehretia* Linn. is represented by 12 species and 1 variety. Six of them including *E. corylifolia* C. H. Wright, *E. longiflora* Champ., *E. macrophylla* Wall. Ex Robx., *E. macrophylla* var. *glabrescens* Y. L. Liu, *E. thyrsiflora* Nakai., and *E. tsangii* Johnst. are used as traditional Chinese herbal medicines because of their extra-ordinary antipyretic detoxicate activities (Zeng and Zeng, 1994). *E. thyrsiflora* has ever locally been used as one source of the bitter tea called *Kudingcha* (He, Liu, and Yang, 1992) in Southern China. *Ehretia* Linn. species were used as ethnopharmaceuticals in folklore for the treatment of various ailments such as inflammation, cough, itches, swellings, pain, diarrhea, dysentery, fever, cachexia, and syphilis (Iqbal *et al*, 2005). These properties were thought to be benefited from the antioxidant activity involving various mechanisms such as free radical scavenging, electron or hydrogen atom donation, and metal cation chelating related to the main class compounds such as polyphenols (Balasundram, Sundram, and Samman, 2006) and benzoquinones. We summarized the research progress on phytochemicals and biological activities of the genus *Ehretia* Linn. over the past decades in this review to provide scientific evidence for better utilization as herbal medicines of the genus.

Chemical constituents

During the past decades, phenolic acids, flavonoids, benzoquinones, cyanogenetic glycosides, fatty acids, and others were isolated from the genus *Ehretia* Linn. Main compounds and the corresponding plant sources are collected in Table 1.

Phenolic acids Phenolic acids are one important type of components in this genus. Twenty phenolic acids (**1**–**20**) were isolated from the genus. Most of them are biosynthesized from caffeic acid to various derivates form, such as monomer, oligomer, and its glycoside. The structures are demonstrated in Fig. 1.

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Table 1 Chemical constituents (1-68) from the plants of *Ehretia* Linn.

No.	Compounds	Sources	Reference
	Phenolic acids		
1	Methyl 2-*O*-feruloyl-1a-*O*-vanillactate	*E. obtusifolia*	Iqbal *et al*, 2005
2	Caffeic anhydride	*E. obtusifolia*	Iqbal *et al*, 2005
3	Rosmarinic acid	*E. obtusifolia*	Iqbal *et al*, 2005
		E. thyrsiflora	Li *et al*, 2009a
4	Methyl rosmarinate	*E. obtusifolia*	Iqbal *et al*, 2005
		E. thyrsiflora	Li *et al*, 2008
5	Icariside E5	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
		E. thyrsiflora	Li *et al*, 2009a
6	Ehletianol D	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
7	1-(4-hydroxy-3-methoxyphenyl)-2-{2-methoxy-4-[1-(*E*)propen-3-ol] -phenoxy}-propane-3-diol *(erythro)*	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
8	1-(4-hydroxy-3-methoxyphenyl)-2-{2-methoxy-4-[1-(*E*)propen-3-ol] -phenoxy}-propane-l, 3-diol *(threo)*	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
9	Buddlenol B	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
10	Ehletianol C	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
11	Trans-4-hydroxycyclohexyl-2-*O*-*p*-coumaroyl-β-*D*-glucopyranoside	*E. obtusifolia*	Iqbal *et al*, 2005
12	Ehletianol A	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
13	Ehletianol B	*E. ovalifolia*	Yoshikawa, Kageyama, and Arihara, 1995
14	*Trans*-ferulic acid	*E. thyrsiflora*	Li *et al*, 2009a
15	Caffeic acid	*E. thyrsiflora*	Li *et al*, 2008
16	*p*-hydroxybenzoic acid	*E. thyrsiflora*	Li *et al*, 2008
17	Lithospermic acid B	*E. thyrsiflora*	Li *et al*, 2009a
18	Danshensu	*E. thyrsiflora*	Li *et al*, 2009a
19	Cinnamic acid	*E. thyrsiflora*	Li *et al*, 2009a
20	(*E*)-ethyl caffeate	*E. thyrsiflora*	Li, 2009
	Flavonoids		
21	Ovalifolin	*E. ovalifolia*	Khattab, Grace, and El-Khrisy, 2001
22	Apigenin	*E. ovalifolia*	Khattab, Grace, and El-Khrisy, 2001
23	Luteolin	*E. ovalifolia*	Khattab, Grace, and El-Khrisy, 2001
24	Quercetin-3-*O*-α-*D*-arabinoside	*E. thyrsiflora*	Li *et al*, 2008
25	Kaempferol-3-*O*-α-*D*-arabinoside	*E. thyrsiflora*	Li *et al*, 2008
26	Quercetin-3-*O*-β-*D*-glucopyranoside	*E. thyrsiflora*	Li *et al*, 2009a
27	Hyperoside	*E. thyrsiflora*	Li *et al*, 2009a
28	Kampferol-3-*O*-β-*D*-galactopyranoside	*E. thyrsiflora*	Li *et al*, 2009a
29	Kampferol-3-*O*-β-*D*-glucopyranoside	*E. thyrsiflora*	Li *et al*, 2009a
30	Kaempferol-3-*O*-arabinosylgalactoside	*E. thyrsiflora*	Li *et al*, 2009a
31	Quercetin-3-*O*-arabinosylgalactoside	*E. thyrsiflora*	Li *et al*, 2009a
32	Quercetin	*E. thyrsiflora*	Li *et al*, 2008
33	Kampferol	*E. thyrsiflora*	Li *et al*, 2008
	Benzoquinones		
34	1,4-naphthoquinone lewisone	*E. laevis*	Thapliyal and Yadav, 2003
35	Ehretianone	*E. buxifolia*	Selvanayagam *et al*, 1996
36	Allomicrophyllone	*E. microphylla*	Yamamura *et al*, 1995
37	Microphyllone	*E. microphylla*	Yamamura *et al*, 1995; Agarwal *et al*, 1980
38	Dehydromicrophyllone	*E. microphylla*	Yamamura *et al*, 1995
39	Hydroxymicrophyllone	*E. microphylla*	Yamamura *et al*, 1995
40	Cyclomicrophyllone	*E. microphylla*	Yamamura *et al*, 1995
	Cyanogenetic glycosides		
41	Simmondsin	*E. philippinensis*	Simpol *et al*, 1994
42	Ehretioside A_1	*E. philippinensis*	Simpol *et al*, 1994
43	Ehretioside A_2	*E. philippinensis*	Simpol *et al*, 1994
44	Ehretioside A_3	*E. philippinensis*	Simpol *et al*, 1994
45	Ehretioside B	*E. philippinensis*	Simpol *et al*, 1994
46	Polyalcohol	*E. philippinensis*	Simpol *et al*, 1994
	Fatty acids and related derivatives		
47	Araneosol	*E. ovalifolia*	Khattab, Grace, and El-Khrisy, 2001
48	(10*E*, 12*Z*, 15*Z*)-9-hydroxy-10, 12, 15-octadecatrienoic acid methyl ester	*E. dicksonii*	Dong, Oda, and Hirota, 2000
49	(9*Z*, 11*E*)-13-hydroxy-9, 11-octadecadienoic acid	*E. dicksonii*	Dong, Oda, and Hirota, 2000
50	(9*Z*, 11*E*)-13-oxo-9, 11-octadecadienoic acid	*E. dicksonii*	Dong, Oda, and Hirota, 2000
51	2-methoxyl benzoic acid octyl ester	*E. thyrsiflora*	Li, 2009
52	Di (octadecyl) phthalate	*E. thyrsiflora*	Li, 2009
53	Tetradecenoic acid, 2, 3-dihydroxypropyl ester	*E. thyrsiflora*	Li, 2009
	Alkaloids		
54	Ehretinine	*E. aspera*	Suri *et al*, 1980
55	Allantoin	*E. thyrsiflora*	Li, 2009
	Others		
56	Bauerenol	*E. laevis*	Dan and Dan, 1982
57	Bauerenol acetate	*E. laevis*	Dan and Dan, 1982
58	α-amyrin	*E. laevis*	Dan and Dan, 1982
59	Betulin	*E. laevis*	Dan and Dan, 1982
60	Lupeol	*E. laevis*	Dan and Dan, 1982
61	Betulinic acid	*E. laevis*	Dan and Dan, 1982
62	Stigmasterol	*E. buxifolia*	Selvanayagam *et al*, 1996
63	Stigmastanol	*E. buxifolia*	Selvanayagam *et al*, 1996
64	Campesterol	*E. buxifolia*	Selvanayagam *et al*, 1996
65	α-spinasterol	*E. buxifolia*	Selvanayagam *et al*, 1996
66	Cholesterol	*E. buxifolia*	Selvanayagam *et al*, 1996
67	β-sitosterol	*E. laevis*	Dan and Dan, 1982
68	Daucosterol	*E. thyrsiflora*	Li, 2009

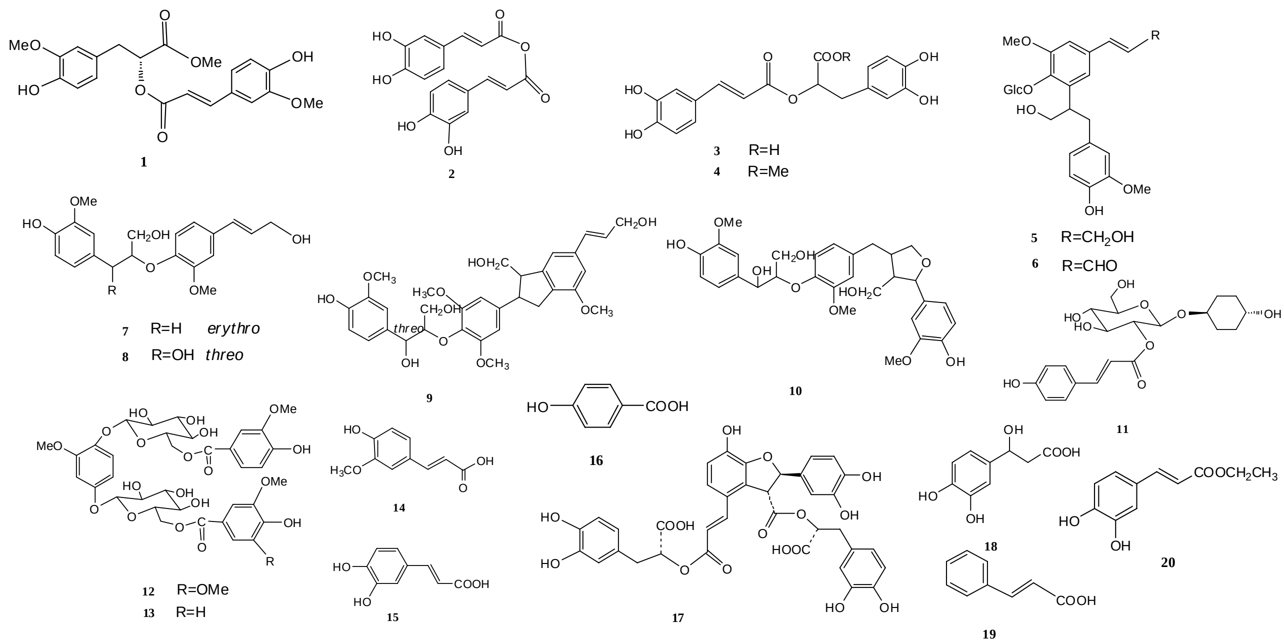

Fig. 1 Chemical structures of phenolic acids isolated from the plants of *Ehretia* Linn.

Flavonoids Thirteen flavonoids (**21**–**33**) have been isolated from this genus so far (Fig. 2).

Benzoquinones Seven benzoquinones (**34**–**40**) were isolated from the genus *Ehretia* Linn. This class of compounds is one of the most characteristics in the family Boraginaceae. Most of them were biosynthesized from the compound 4-hydroxybenzoic acid (4HB) and monoterpenes. They were all related to the bioactivities of hematischesis, antibacterial, and antivirus (Fig. 3).

Cyanogenetic glycosides Six compounds (**41**–**46**) were isolated from *E. philippinensis* (Simpol *et al*, 1994) (Fig. 4).

Fatty acids and related derivatives Eight compounds (**47**–**53**) were isolated from *E. ovalifolia* (Khattab, Grace, and El-Khrisy, 2001), *E. dicksonii* (Dong, Oda, and Hirota, 2000), and *E. thyrsiflora* (Li, 2009) (Fig. 5).

Alkaloids Pyrrolizidines which are widespread distributed in the family Boraginaceae, especially in

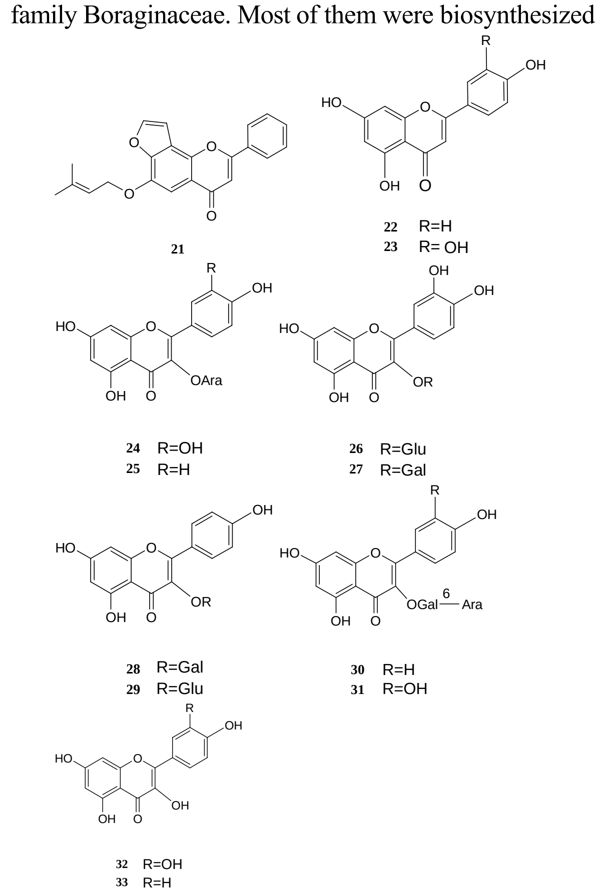

Fig. 2 Chemical structures of flavonoids isolated from the plants of *Ehretia* Linn.

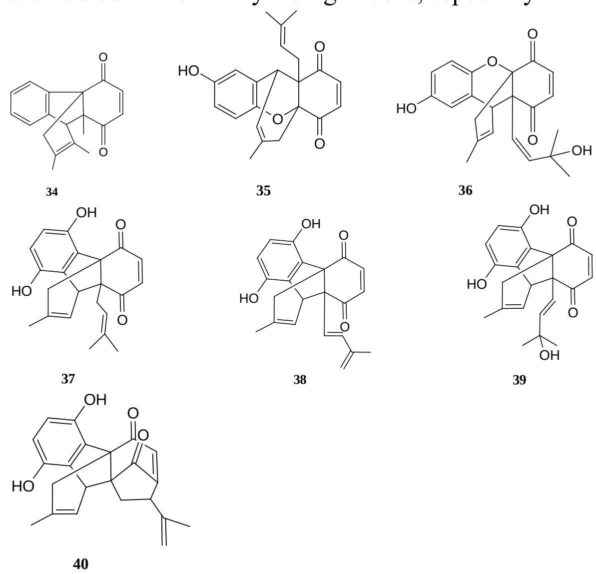

Fig. 3 Chemical structures of benzoquinones isolated from the plants of *Ehretia* Linn.

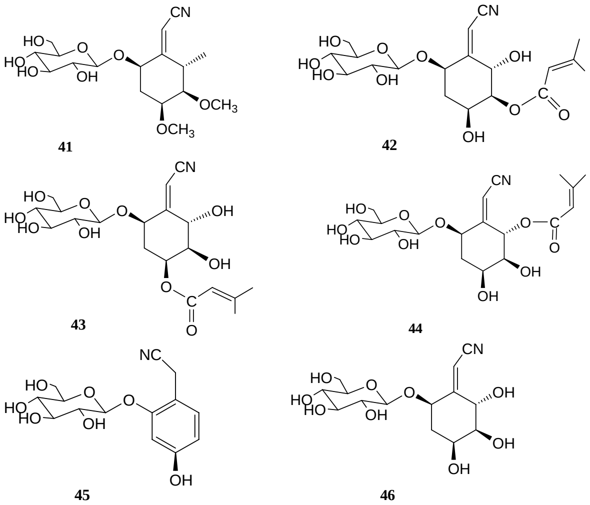

Fig. 4 Chemical structures of cyanogenetic glycosides isolated from *Ehretia* Linn.

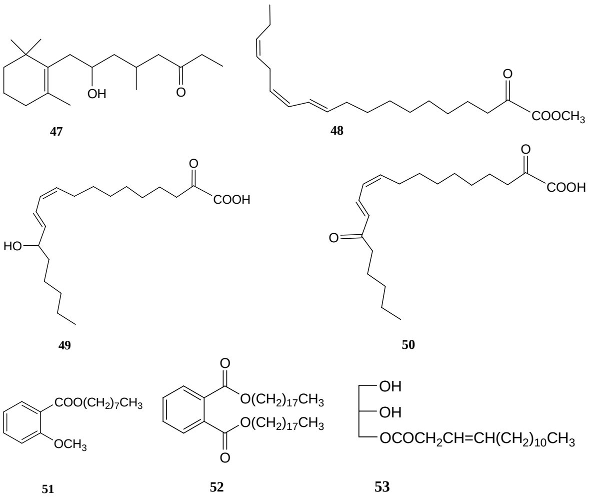

Fig. 5 Chemical structures of fatty acids and related derivatives isolated from plants of *Ehretia* Linn.

the subfamily of Boraginoideae and Heliotropioideae are relatively few reported in the subfamily of Ehretiaceae. In the genus *Ehretia* Linn., compound **54** belonging to retronecine-type pyrrolizidine was isolated from *E. aspera* (Suri *et al*, 1980). Oxidative product of uric acid by purine catabolism, allantoin (**55**) was also reported from *E. thyrsiflora* (Li, 2009) (Fig. 6).

Others Six triterpenoids such as bauerenol (**56**), bauerenol acetate (**57**), α-amyrin (**58**), betulin (**59**), lupeol (**60**), and betulinic acid (**61**) were isolated from *E. laevis* (Dan and Dan, 1982). Seven sterols

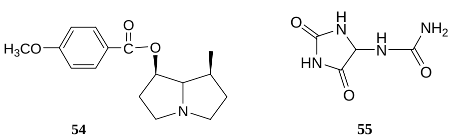

Fig. 6 Chemical structures of compounds 54–55 isolated from plants of *Ehretia* Linn.

including stigmasterol (**62**), stigmastanol (**63**), campesterol (**64**), α-spinasterol (**65**), cholesterol (**66**), β-sitosterol (**67**), and daucosterol (**68**) were isolated from *E. buxifoli* (Selvanayagam *et al*, 1996), *E. laevis* (Dan and Dan, 1982), and *E. thyrsiflora* (Li, 2009) (Fig.7).

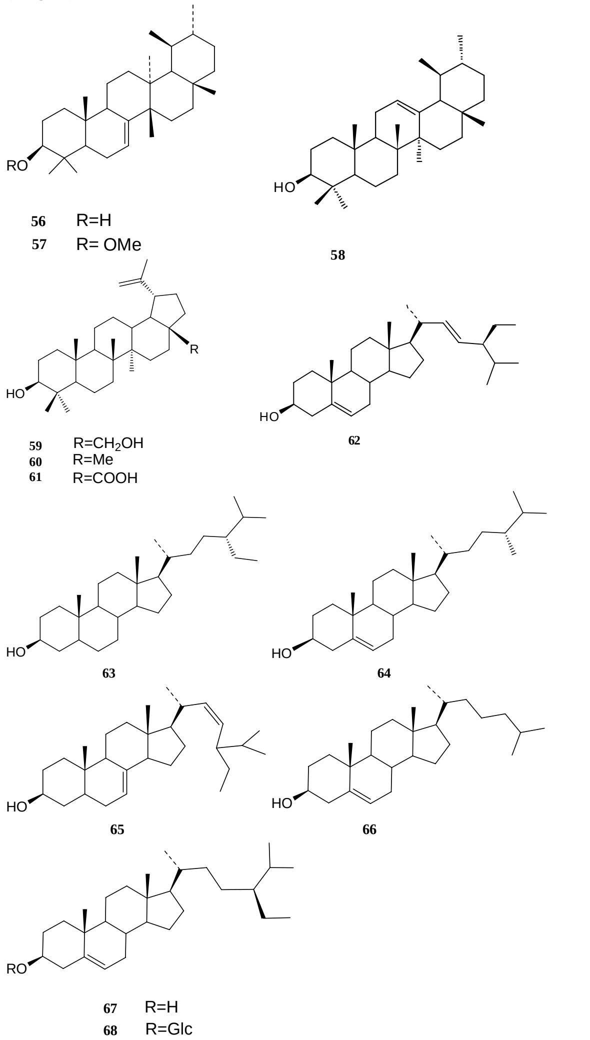

Fig. 7 Chemical structures of compounds 56–68 isolated from plants of *Ehretia* Linn.

Biological activities

Antioxidant effect The antioxidant activities of crude extract from the leaves of *E. thyrsiflora* and its four fractions of dichloromethane, ethyl acetate, *n*-butanol, and water were tested by using different *in vitro* assays of the scavenging effects on DPPH, reducing power and Fe^{2+}-chelating activity. Ethyl acetate fraction containing abundant polyphenols showed comparatively strong antioxidant activities in these assays (Li *et al*, 2009b).

Anti-inflammatory effects Methyl 2-*O*-feruloyl-1a-*O*-vanillactate (**1**), caffeic anhydride (**2**), rosmarinic acid (**3**), methyl rosmarinate (**4**), and *trans*-4- hydroxycyclohexyl-2-*O*-*p*-coumaroyl-β-*D*-glucopyranoside (**11**) isolated from the ethyl acetate fraction of *E. obtusifolia* inhibited lipoxygenase in a concentration dependent manner with K_i values ranging 0.85–57.6 μmol/L (Iqbal *et al*, 2005). Pharmacologically experimental evidences provided scientific explanation for the folkloric use of herbal medicines in the genus *Ehretia* Linn. in the treatment of some ailments associated with lipoxygenase and inflammatory.

(10*E*,12*Z*,15*Z*)-9-hydroxy-10,12,15-octadecatrienoic acid methyl ester (**48**) isolated from *E. dicksonii* suppressed TPA-induced inflammation on mouse ears at a dose of 500 μg/mL. The inhibitory effect (IE_{50}) was 43%. (9*Z*,11*E*)-13-hydroxy-9,11-octadecadienoic acid (**49**) and (9*Z*,11*E*)-13-oxo-9,11-octadecadienoic acid (**50**) showed potent activity at a dose of 500 μg/mL (IE_{50} values were 63% and 79%, respectively). Compounds **48**–**50** also showed inhibitory activity toward soybean lipoxygenase at a concentration of 10 μg/mL (Dong, Oda, and Hirota, 2000).

Antisnake venom effects The antisnake venom activity was determined using the LD_{50} of the venom in mice by the subcutaneous route. The methanol extract of the root bark of *E. buxifolia* showed strong neutralization, prophylactic, and curative activity. In the neutralization studies, ehretianone (**35**) showed significant protection at a dosage level of 3.75 mg/kg in mice when challenged with the LD_{50} of the venom. In the prophylactic treatment, compound **35** was administered 30 min before venom injection and the mortality was reduced by 35% compared with control. In the curative study, the same dosage of compound **35** gave significant protection up to 5 min after venom injection (Selvanayagam *et al*, 1996).

Anti-allergic effects The ethyl acetate fraction of *E. microphylla* showed inhibitory activity on exocytosis in antigen-stimulated rat basophils. The bioassay-guided separation afforded five benzoquinones. Microphyllone (**37**) and allomicrophyllone (**36**) showed strong activities (IC_{50} values were 33 and 36 μmol/L, respectively). Dehydromicrophyllone (**38**) and cyclomicrophyllone (**40**) had weaker activity (IC_{50} values were 62 and 106 μmol/L, respectively). Hydroxymicrophyllone (**39**) was inactive (Yamamura *et al*, 1995).

The *n*-butanol and ethyl acetate extracts of *E. philippinensis* were found to have antihistamine release activity against compound 48/80 (a polymer prepared by heating 4-methoxy-*N*-methylphenylamine with formaldehyde in acid solution and a very potent histamine liberator). Rosmarinic acid (**3**) (IC_{50} was 18 μmol/L) and methyl rosmarinate (**4**) (IE_{50} was 66% at 107 μmol/L) isolated from *E. philippinensis* had obviously antihistamine release activity against compound 48/80. But they didn't show any activity against compound concanavalin A. The bioassay-guided separation of the active fraction afforded 6 cyanogenetic glycosides (**41**–**46**), but didn't show the same antihistamine release activity like the fraction (Simpol *et al*, 1994).

Twenty-three medicinal plants from Philippine were tested for their antihistamine release properties, and *E. microphylla* was found to have strong activity. From methanol extract of the leaves of *E. microphylla*, rosmarinic acid (**3**) was isolated as one of the active inhibitory constituents (Rimando *et al*, 1987).

Conclusion

There are 50 species of the genus *Ehretia* Linn. in the world, in which 12 species and 1 variety are in China. Six of them were used as traditional Chinese herbal medicines for heat-clearing and detoxifying in the body or treatment of the inflammatory. *E. thyrsiflora* has ever been locally used as a substitution of the traditional Chinese herbal tea *kudingcha* in Southwest of China because of the similar plant morphology and ethnopharmacology. In literatures, retronecine-type pyrrolizidine alkaloids in the family Boraginaceae are hepatotoxic. The poisoning can lead to veno occlusive diseases with symptoms like arterial hypertension, right ventricular hypertrophy, abdominal pain, ascites, hepatomegaly and elevated serum transaminase level (Bah, Bye, and Pereda-Miranda, 1994; Ng, Cheng, and Xu, 2009). Although this type of alkaloids was not reported in *E. thyrsiflora,* compound **54** isolated from *E. aspera* suggested that the utilization of herbal medicines in this genus should be cautious. Abundant polyphenols in this genus are thought to be associated with the antioxidative and anti-inflammatory activities. Benzoquinones with particular distribution and important bioactivities of hematischesis, anti-bacterial, and anti-viral could be used as marker compounds. The chemical constituents existed in the genus *Ehretia* Linn. attract more scientists to pay close attention for the potent and prospect activity of these species just like other species from the family Boraginaceae.

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