A MINI REVIEW ON THE PHYTOCHEMICAL COMPOSITION AND BIOLOGICAL ACTIVITIES OF AGASTACHE RUGOSA

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Abstract: *Agastache rugosa (Fisch. & C.A.Mey.) Kuntze, a medicinal and ornamental plant in the Lamiaceae family, is mostly found in East Asian countries such as Vietnam, Korea, China, and Japan. All parts of this plant are used as traditional medicines to treat abdominal pain, congestion, chills, diarrhea, nausea, and vomiting, and dispel dampness. Phytochemical studies of this plant revealed that it is a source of specialized metabolites including flavonoids, phenylpropanoids, lignans, and terpenoids, which have useful pharmacological activities such as antioxidant, anti-inflammatory, anti-allergic, anti-microbial, anti-depression, anti-cancer, anti-viral, anti-asthmatic, and cardiovascular activities. Among them, acacetin (1), tilianin (2), and rosmarinic acid (19) are the main active compounds of A. rugosa. However, most of the phytochemical and pharmacological studies belong to A. rugosa species originating from Korea, China, and Japan. To date, there has only been one analysis report on the constituents of the leaf and flower oils of A. rugosa in Vietnam. This review briefly summarizes the chemical constituents and biological properties of A. rugosa that have been recently reported.*

Keywords: *Lamiaceae; Biological actions; Phytoconstituents; Agastache rugosa; Review.*

1. Introduction

Agastache is a small genus of the Lamiaceae family, comprising 22 species of perennial aromatic medicinal herbs (Zielińska & Matkowski, 2014). Of them, *A. rugosa* (Fisch. & C.A.Mey.) Kuntze is mostly found in East Asian countries such as Vietnam, Korea, China, and Japan (Li et al., 2013). *A. rugosa* has been used as a wild vegetable and herbal drug for the treatment of anorexia, vomiting, and other intestinal disorders (Li et al., 2013). This herb is known under many different names, such as Korean mint, purple giant hyssop, Indiana mint, wrinkled giant hyssop, "Tho Hoac Huong" (in Vietnam), “Huo Xiang” (in China), and "Kakko" (in Japan) (Itokawa et al., 1981; Loi, 1988). All parts of this plant are used as traditional pharmaceuticals to treat different disorders in various civilizations’ medical systems (Zielińska & Matkowski, 2014). In Korea, this mint-fragranced plant has long been used for the treatment of abdominal pain, congestion, chills, and diarrhea. In addition, it is also used as a wild vegetable, a spice a spice for fish-based foods (Hong et al., 2020). In Chinese traditional medicine, this plant has been used to treat nausea, and vomiting, and dispel dampness (Cao et al., 2017).

2. Research overview

Previous biological studies showed that extracts of *A. rugosa* have antioxidant, anti-HIV, antiatherogenic, antifungal, hypolipidemic, carminative, and antipyretic properties (Gong et al., 2012; Seo et al., 2019; Tuan et al., 2012). Especially, the essential oil of *A. rugosa* possesses various pharmacological properties such as antibacterial, antifungal, antioxidant, anticancer, antiviral, nematicidal, insecticidal, wrinkle improver, stress reliever, and Alzheimer's disease alleviator (Hong et al., 2020). Phytochemical studies of this plant revealed that it is a source of specialized metabolites including flavonoids, phenylpropanoids, lignans, and terpenoids, which have useful pharmacological activities such as

antioxidant, anti-inflammatory, anti-allergic, anti-microbial, anti-depression, anti-cancer, anti-viral, anti-asthmatic, and cardiovascular activities (Lam et al., 2020; Seo et al., 2019). However, most of the above studies belong to *A. rugosa* species originating from Korea, China, and Japan. There is almost no phytochemical research on non-volatile compounds and their biological activities from *A. rugosa* originating in Vietnam. To date, there has only been one analysis report on the constituents of the leaf and flower oils of *A. rugosa* from Vietnam (Dung et al., 1996). This review briefly summarizes the chemical constituents and pharmacological properties of *A. rugosa* that have been recently reported.

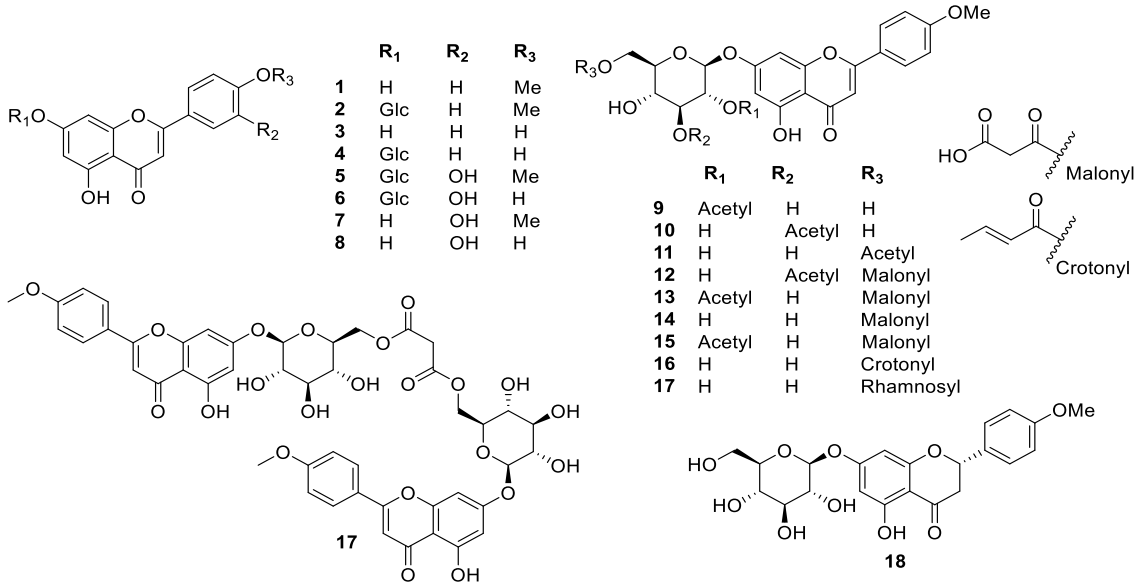
3. Materials and methods

The available information on the chemical constituents and biological properties of *A. rugosa* was systematically collected via piles of resources including classic books about Vietnamese herbal medicine, and scientific databases including Pubmed (https://pubmed.ncbi.nlm.nih.gov/), Google Scholar (https://scholar.google.com/), ACS, Web of Science, and Science Direct. Main keywords used for the reference search included: *Agastache rugosa, Agastache* genus, Lamiaceae, "Thổ hoắc hương", *Agastache* + phytochemistry, *Agastache* + pharmacological property. The 2D structures of compounds were constructed using ChemBioDraw Ultra 21.0.0.28 (Cambridgesoft, USA).

4. Results

4.1. Phytochemical constituents of A. rugosa

Figure 1. Chemical structure of 18 flavonoids 1-18 from *A. rugosa*



4.1.1. Flavonoids

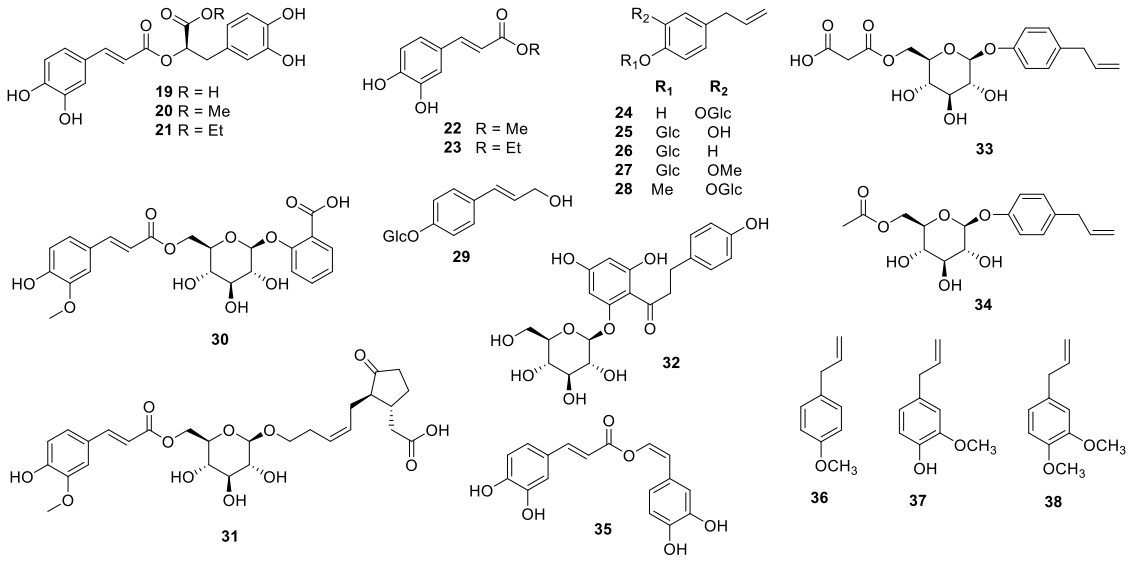
Flavonoid is the main constituent of *A. rugosa*. To date, 18 flavonoids have been isolated and structurally characterized from *A. rugosa* including acacetin (1) (An et al., 2018; Itokawa et al., 1981; Park et al., 2016; Seo et al., 2019), tilianin (2, acacetin-7-*O*-*β*-D-glucopyranoside) (An et al., 2018; Itokawa et al., 1981; Park et al., 2016; Seo et al., 2019), apigenin (3) (Park et al., 2016; Seo et al., 2019), apigetrin (4) (Park et al., 2016; Seo et al., 2019), diosmetin-7-*O*-*β*-D-glucopyranoside (5) (Seo et al., 2019), luteoloside (6) (Seo et al., 2019), diosmetin (7) (Seo et al., 2019), luteolin (8) (Seo et al., 2019), isoagastachoside (9) (Seo et al., 2019), acacetin-7-*O*-(3″-*O*-acetyl)-*β*-*D*-glucopyranoside (10) (Park et al., 2016; Seo et al., 2019), agastachoside (11) (Itokawa et al., 1981), acacetin-7-*O*-(3″-*O*-acetyl-6″-*O*-malonyl)-β-D-glucopyranoside (12) (Seo et al., 2019), acacetin-7-*O*-(2″-O-acetyl-6″-*O*-malonyl)-*β*-*D*-glucopyranoside (13) (An et al., 2018; Seo et al., 2019), acacetin-7-*O*-(6″-*O*-malonyl)-*β*-*D*-glucopyranoside (14) (Seo et al., 2019), acacetin 7-*O*-*β*-(6"-*O*-(*E*)-

crotonylglucopyranoside) (15) (Park et al., 2016), linarin (16, acacetin-7-*O*-rutinoside) (Itokawa et al., 1981), agastachin (17) (Itokawa et al., 1981), and (2*S*)-poncirenin (18) (Seo et al., 2019). Their structures are shown in Figure 1. Among them, tilianin (2) was a major constituent (1.96% of *A. rugosa* methanol extract) (Hong et al., 2001).

4.1.2. Phenylpropanoids

Together with flavonoids, phenylpropanoids are also the main constituent of *A. rugosa.* Phenylpropanoid compounds that have been isolated from *A. rugosa* include: rosmarinic acid (19) (An et al., 2018; Seo et al., 2019), methyl rosmarinate (20) (Seo et al., 2019), ethyl rosmarinate (21) (Seo et al., 2019), methyl caffeate (22) (Seo et al., 2019), ethyl caffeate (23) (Seo et al., 2019), 1-hydroxy-2-*O*-*β*-D-glucopyranosyl-4-allylbenzene (24) (Park et al., 2016; Seo et al., 2019), 1-*O*-*β*-D-glucopyranosyl-2-hydroxy-4-allylbenzene (25) (Park et al., 2016; Seo et al., 2019), chavicol-*β*-D-glucopyranoside (26) (Park et al., 2016; Seo et al., 2019), citrusin C (27) (Park et al., 2016; Seo et al., 2019), 3-hydroxyestragole-*O*-*β*-glucopyranoside (28) (Park et al., 2016), (*E*)-4-hydroxycinnamyl alcohol-4-*β*-glucopyranoside (16-29) (Park et al., 2016), (3*R*,7*R*)-tuberonic acid-12-*O*-[6′-*O*-(*E*)-feruloyl]-*β*-*D*-glucopyranoside (30) (Seo et al., 2019), salicylic acid-2-*O*-[6′-*O*-(*E*)-feruloyl]-*β*-D-glucopyranoside (31), phlorizin (32) (Seo et al., 2019), chavicol-1-*O*-(6′-*O*-methylmalonyl)-*β*-D-glucopyranoside (33) (Seo et al., 2019), chavicol-1-*O*-(6′-*O*-acetyl)-*β*-D-glucopyranoside (34) (Seo et al., 2019), nepetoidin B (35) (Seo et al., 2019), estragole (36) (Li et al., 2013), eugenol (37) (Li et al., 2013), and methyleugenol (38) (Li et al., 2013). Their structures are shown in Figure 2. Among these, the most typical compound is rosmarinic acid (RA, 19). The highest amount of RA was detected in flowers, where its content was 48.43 µg/g d.w., as well as in roots (30.97 µg/g) and leaves (22.14 µg/g). The lowest content of RA was reported in stems (9.14 µg/g) (Zielińska & Matkowski, 2014). While, estragole (36, 8.55%), eugenol (37, 7.54%), and methyleugenol (38, 50.51%) were determined as the principal compounds of *A. rugosa* essential oil (Li et al., 2013).

Figure 2. Chemical structures of phenylpropanoids 19-38 from *A. rugosa*

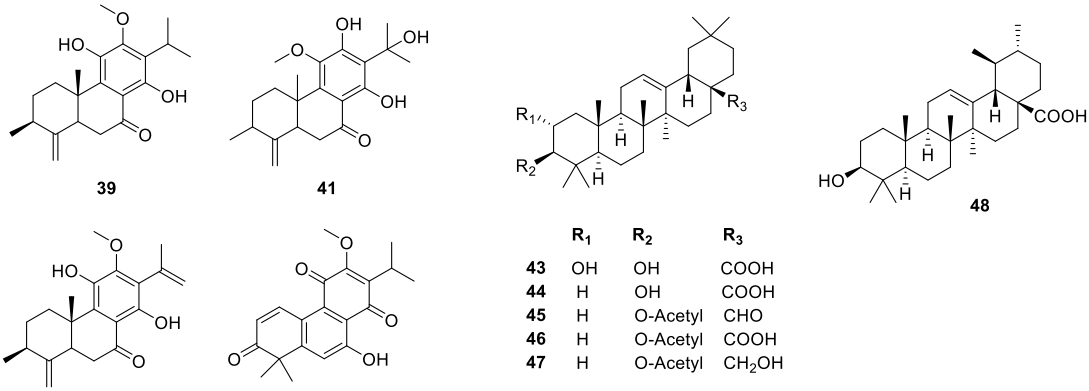


4.1.3. Terpenoids

Through GC-MS analysis (Li et al., 2013), many monoterpenoids and sesquiterpenoids were discovered in the *A. rugosa* essential oil. Typical compounds with high concentrations can be mentioned as follows: thymol, pulegone, limonene, thymoquinone, and caryophyllene (2.38%) (Li et al., 2013). There are four diterpenes isolated from the root of Korea *A. rugosa*, including agastanol (39) (Lee et al., 1994) dehydroagastol (40) (Lee et al., 1994; Zou & Cong, 1991), 19(4→3)-abeo-12,14,15-trihydroxy-11-methoxy-abiet-4(18),8,11,13-

tetraen-7-one (41) (Han et al., 1987b), and agastaquinone (42) (Lee et al., 1995; Min et al., 1999). In addition, six triterpenes: maslinic acid (43) (Zou & Cong, 1991), oleanolic acid (44) (Zou & Cong, 1991), 3-*O*-acetyloleanolic aldehyde (45) (Han et al., 1987a; Zou & Cong, 1991), 3-*O*-acetyloleanolic acid (46) (Han & Byon, 1988) erythrodiol-3-*O*-acetate (47) (Han et al., 1987a), and ursolic acid (48) (Cao et al., 2017) were also isolated from *A. rugosa*. Their structures are shown in Figure 3.

Figure 3. Chemical structure of terpenoids 39-48 from *A. rugosa*



4.1.4. Lignans, megastigmanes, steroids, and other compounds

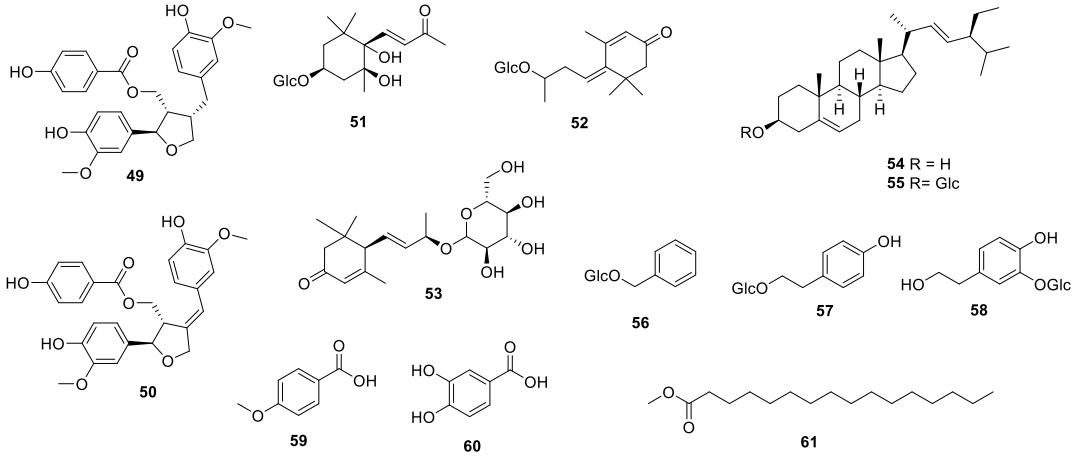
Up to now, there are only 2 lignans isolated and structurally elucidated from *A. rugosa* including: (8S,7′R,8′S)-4-hydroxybenzoic acid 4-(4-hydroxy-3-methoxybenzyl)-2-(4-hydroxy-3-methoxyphenyl) tetrahydrofuran-3-ylmethyl ester (agastinol, 49) and (7′*R*,8′*S*)-4-hydroxybenzoic acid 4-(hydroxy-3-methoxybenzylidene)-2-(4-hydroxy-3-methoxyphenyl)-tetrahydrofuran-3-yl methyl ester (agastenol, 50) (Lee et al., 2002). In addition, other phytochemical investigations of the aerial parts or leaves of *A. rugosa* revealed the presence of megastigmanes [5*β*,6*α*-dihydroxy-3*β*-(*β*-glucopyranosyloxy)-7-megastigmen-9-one (51), (*E*)-4-[3′-(*β*-glucopyranosyloxy)butylidene] -3,5,5-trimethyl-2-cyclohexen-1-one (52), and (6*R*,9*R*)-3-oxo-*α*-ionol-9-O-*β*-glucopyranoside (53) (Park et al., 2016; Seo et al., 2019), steroids [*β*-sitosterol (54) (Cao et al., 2017) and daucosterol (55)] (Zou & Cong, 1991), and other compounds [benzyl *β*-glucopyranoside (56) (Park et al., 2016), salidroside (57) (Park et al., 2016), cimidahurinine (58) (Park et al., 2016), anisic acid (59) (Seo et al., 2019), protocatechuic acid (60) (Cao et al., 2017), and methyl hexadecanoate (61) (Cao et al., 2017)] (Figure 4).

4.2. Biological properties of A. rugosa

The essential oil of *A. rugosa* possesses various pharmacological properties such as antibacterial, antifungal, antioxidant, anticancer, antiviral, nematicidal, insecticidal, wrinkle improver, stress reliever, and Alzheimer's disease alleviator (Hong et al., 2020). The phytotoxic and antimicrobial activities of the *A. rugosa* essential oils could result from one of its main constituents, estragole (36). Estragole isolated from *A. rugosa* was more efficient against human pathogenic fungi as a pure compound than as a crude essential oil (Zielińska & Matkowski, 2014). Previous reports indicated that acacetin (1), tilianin (2), and rosmarinic acid (19) are the main active compounds of *A. rugosa* (Lam et al., 2020; Tuan et al., 2012; Zielińska & Matkowski, 2014). They are well-known for their pharmacological activities such as antioxidant, anti-inflammatory, anti-allergic, anti-microbial, anti-depression, anti-cancer, anti-viral, anti-asthmatic, and cardiovascular activities (Lam et al., 2020; Seo et al., 2019; Tuan et al., 2012; Zielińska & Matkowski, 2014). The high content of these active compounds contributes to the pharma- cologically useful properties of this species. The concentrations of acacetin (1), tilianin (2), and rosmarinic acid (19) in *A. rugosa* were the highest in the flowers (Tuan et al., 2012). *A. rugosa* diterpenes: agastanol (39) and dehydroagastol (40) showed cytotoxic activities against human cancer cell lines. While agastanol (39) and agastaquinone (42) exhibited significant inhibitory effects against

human immunodeficiency virus type 1 (HIV-1) protease activity with IC_{50} values of 360 and 87 μM, respectively. Most recently, the remarkable anti-inflammatory properties of *A. rugosa* flavonoids (1, 2, 6, 7, 9, and 12) and phenyl propanoids (31, 33, and 34) have also been documented through their inhibitory effects against the production of prostaglandin E_2 (PGE_2) in LPS-induced RAW 264.7 macrophages (Seo et al., 2019).

Figure 4. Chemical structure of compounds 49-61 from *A. rugosa*



5. Discussions

Of the three most important medicinal species in the genus *Agastache*, *A. rugosa* is the main object in most of the published bioactivity data. *A. rugosa* is the only species native to East Asia, and it is an important herbal drug in Chinese, Korean and Japanese traditional medicine. As such, it has been frequently studied for various pharmacological activities in both in vitro and animal models. Pharmacological results have validated the use of *A. rugosa* in traditional medicine. As literature demonstrated, flavonoids and rosmarinic acid derivatives are the main constituents and responsible for most of the biological activities shown by this plant. However, the detailed active compounds and the underlying mechanisms remain a work in progress. In addition, more attention should be paid to the phytochemical investigation of *A. rugosa* species originating from Vietnam.

6. Conclusions

All the above-mentioned findings suggest the importance of *A. rugosa* for East Asian traditional medicine, which can be expected to extend to other regions, similar to the already more popular herbs from the Lamiaceae family. All parts of this plant are used as traditional medicines to treat abdominal pain, congestion, chills, diarrhea, nausea, and vomiting, and dispel dampness. To date, sixty-one compounds have been isolated from *A. rugosa* via chromatography methods. Their structures were classified into some main groups of flavonoids, phenylpropanoids, lignans, and terpenoids, which have useful pharmacological activities such as antioxidant, anti-inflammatory, anti-allergic, anti-microbial, anti-depression, anti-cancer, anti-viral, anti-asthmatic, and cardiovascular activities. Among them, acacetin (1), tilianin (2), and rosmarinic acid (19) are the main active compounds of *A. rugosa*. However, most of the phytochemical and pharmacological studies belong to *A. rugosa* species originating from Korea, China, and Japan. To date, there has only been one analysis report on the constituents of the leaf and flower oils of *A. rugosa* in Vietnam. Therefore, future studies on the chemical composition and biological activity of *A. rugosa* originating from Vietnam are very necessary and need to be focused on.

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TỔNG QUAN CÁC NGHIÊN CỨU VỀ THÀNH PHẦN HÓA HỌC VÀ HOẠT TÍNH SINH HỌC CỦA LOÀI THỔ HOẮC HƯƠNG *AGASTACHE RUGOSA* (FISCH. & C.A.MEY.) KUNTZE

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Tóm tắt: *Agastache rugosa (Fisch. & C.A.Mey.) Kuntze là một trong các loài hoắc hương được tìm thấy ở miền Bắc Việt Nam, thuộc họ Hoa môi (Lamiaceae) và còn có tên gọi khác là thổ hoắc hương. A. rugosa là cây thân cỏ có mùi thơm, phân bố phổ biến ở các quốc gia Đông Á như Trung Quốc, Hàn Quốc, Nhật Bản và Việt Nam. Loài dược liệu này từ lâu được sử dụng trong Y học cổ truyền ở các quốc gia này để điều trị các chứng đau bụng, nghẹt mũi, ớn lạnh, tiêu chảy và buồn nôn. Các nghiên cứu hóa thực vật của loại cây này cho thấy sự có mặt của các chất chuyển hóa chuyên biệt bao gồm flavonoid, phenylpropanoid, lignan và terpenoid, với các hoạt tính sinh học hữu ích như chống oxy hóa, chống viêm, chống dị ứng, kháng khuẩn, chống trầm cảm, chống ung thư, kháng virus, cũng như tác dụng trong điều trị bệnh hen xuyễn và điều hòa tim mạch. Tuy nhiên, hầu hết các nghiên cứu trên đều thuộc về các loài A. rugosa có nguồn gốc từ Hàn Quốc, Nhật Bản và Trung Quốc. Ở Việt Nam, hiện nay chỉ có duy nhất một nghiên cứu về thành phần hóa học của tinh dầu được chiết xuất từ lá và hoa của loài A. rugosa. Bài báo này tổng kết lại tất cả các nghiên cứu về thành phần hóa học và hoạt tính sinh học của loài A. rugosa đã được công bố cho đến nay.*

Từ khóa: *Họ Lamiaceae; Hoạt tính sinh học; Thành phần thực vật; Thổ hoắc hương; Tổng quan.*