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Review

Research Progress of Benzothiazole and Benzoxazole Derivatives in the Discovery of Agricultural Chemicals

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Abstract: Benzoxazole and benzothiazole have a broad spectrum of agricultural biological activities, such as antibacterial, antiviral, and herbicidal activities, which are important fused heterocyclic scaffold structures in agrochemical discovery. In recent years, great progress has been made in the research of benzoxazoles and benzothiazoles, especially in the development of herbicides and insecticides. With the widespread use of benzoxazoles and benzothiazoles, there may be more new products containing benzoxazoles and benzothiazoles in the future. We systematically reviewed the application of benzoxazoles and benzothiazoles in discovering new agrochemicals in the past two decades and summarized the antibacterial, fungicidal, antiviral, herbicidal, and insecticidal activities of the active compounds. We also discussed the structural–activity relationship and mechanism of the active compounds. This work aims to provide inspiration and ideas for the discovery of new agrochemicals based on benzoxazole and benzothiazole.

Keywords: benzoxazole; benzothiazole; agrochemical; SAR; mechanism

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1. Introduction

In global agricultural production, plant diseases, insects, and weed damage are the main causes of crop yield loss [1,2]. Fungi [3,4], bacteria [5–7], plant viruses [8,9], pests [10,11], weeds [12], nematodes [13–16], and mites [17] cause huge economic losses to the world's agriculture every year. At present, the use of agrochemicals is still one of the most effective means to control plant diseases, insects, and grass damage, especially in the management of pest resistance and resistant weeds [18,19]. More importantly, when pests (such as armyworms [20], locusts [21], and walkers [22]) break out in large areas, the use of highly efficient chemical pesticides is the most effective strategy for rapid pest control [23]. However, long-term use of traditional agrochemicals will not only pollute the environment but also increase the resistance of pathogens [24], resulting in more difficult management of plant diseases, insects, and weeds [7,25,26]. Therefore, the development of new agrochemicals with unique action mechanisms to replace traditional pesticides is an urgent problem to be solved in the management of plant diseases, pests, and grass diseases.

Benzoxazole is a combination of a benzene ring and an oxazole ring; benzothiazole is the bioisostere of benzoxazole. They are widely used in drug research and development as the core scaffold structure [27–32] and play an important role in drug discovery. Twenty years ago, the research on benzothiazole and benzoxazole was widely focused on

the field of medicine [33–36]; on the contrary, there was little research in the field of agrochemicals. However, 10 years ago, there was a large amount of research on benzothiazole and benzoxazole in new agrochemicals. In terms of commercial agrochemicals, benzoxazole and benzothiazole agrochemicals play an important role. For example, the herbicides metamifop (Figure 1) and fenoxaprop-p-ethyl are acetyl-coenzyme A carboxylase inhibitors, which inhibit the growth of grasses mainly by inhibiting the synthesis of plant fatty acids, eventually leading to the death of plants [37–40]. Mefenacet, a systemic herbicide, is an inhibitor of cell generation and division, which can prevent cell division and elongation in weed meristem and has a good control effect on barnyard grass [41]. The fungicide benthiavalicarbisopropyl has an inhibitory effect on the sporangia formation and germination of Phytophthora at low mass concentrations. The mechanism of action is still unclear, but it does not affect the oxidation and synthesis of nucleic acid and protein [42,43]. The antiviral agent Dufulin has been widely used against tomato virus disease, cucumber virus disease, tobacco virus disease, and southern rice black-streaked dwarf virus disease [44–46]. Oxazosulfyl, the first benzoxazole insecticide with a broad spectrum of insecticidal activity, is currently mainly used to control rice pests, but its mechanism of action is still unclear [47,48].

Benzoxazole and benzothiazole have stable structures and are easily modified, which play an important role in the discovery of new agrochemicals. Research on the discovery of new agrochemicals based on benzoxazole and benzothiazole scaffolds may be strengthened in the future. There is no comprehensive review of benzoxazole and benzothiazole derivatives in the discovery of novel agrochemicals. Herein, we summarize the benzoxazole and benzothiazole derivatives in the application of new types of agricultural chemicals, perform analysis of the benzoxazole and benzothiazole compounds in terms of antibacterial, antifungal, antiviral, weeding, and insecticidal activity, and discuss the structure–activity relationship (SAR) and mechanism of action. It is hoped that this review provides new clues and inspiration for the discovery of new benzoxazole and benzothiazole agrochemicals.

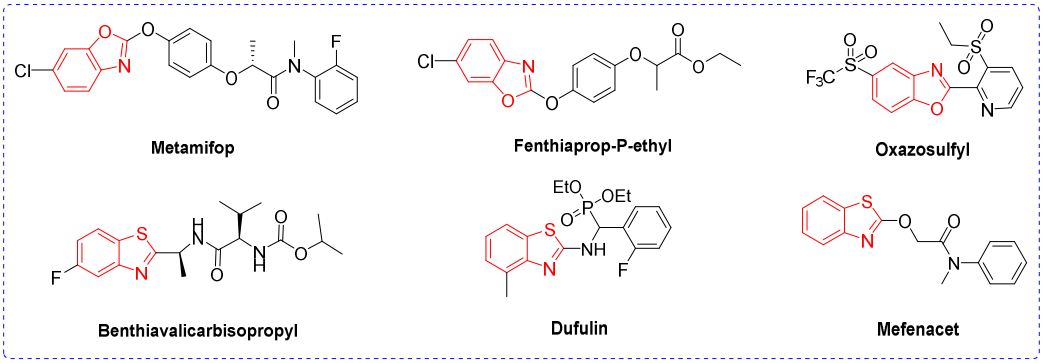

Figure 1. Chemical structure of some pesticides containing benzoxazole or benzothiazole scaffolds.

2. Antibacterial Activity

Diseases caused by plant bacteria have seriously restricted the safe production of crops and caused huge output and economic losses to world agriculture every year [49,50]. However, sustained and effective management of these plant bacterial diseases is extremely difficult and often requires integrated management strategies [51–53]. The long-term use of chemical antimicrobials has led to the evolution of resistance in bacteria [54]. This puts forward higher requirements for the development of antimicrobial agents and the management of plant bacterial diseases.

Some benzoxazole derivatives or benzothiazole derivatives have good antibacterial activity (Figure 2). For example, the EC_{50} values of compound **1** against *Xanthomonas oryzae pv.oryzicola (Xoc)* and *Xanthomonas citri* subsp. *Citri (Xac)* were 47.6 mg/L (Table 1) and 36.8 mg/L, respectively [55]. In addition, compound **1** showed good antibacterial activity

by up-regulating the expression of Succinate dehydrogenase (SDH) during oxidative phosphorylation, thereby inhibiting bacterial reproduction. At a concentration of 100 mg/L, the inhibition rate of compound **2** against *Xanthomonas oryzae pv.oryzae (Xoo)* was 52.4%. Based on compound **2**, the methoxy group was replaced with the nitro group, and the methyl group at position-2 of the benzene ring was replaced with the trifluoromethyl group at position-4 of the benzene ring. The inhibition rate of compound **3** on *Ralstonia solanacearum* (*Rs*) was 71.6% [56]. In addition, the introduction of the pyridine e group increased the broad spectrum of antibacterial compounds. For example, the antibacterial activities of compound **4** against *Xoo*, *Xac*, and *Rs* were 52.40%, 50.97%, and 36.49%, respectively. If the pyridyl group was replaced by the electron-withdrawing group, the antibacterial activity of the compound was enhanced. For example, the EC_{50} value of compound **5** against *Xoo* was 38.97 mg/L, while the EC_{50} value of compound **6** against *Xac* was 13.42 mg/L [57]. The EC_{50} value of compound **7** against *Xoo* was 11.4 mg/L. In addition, compound **7** can not only change cell morphology, but also reduce the pathogenicity of *Xoo* to rice by inhibiting the formation of cell biofilms, thereby affecting cell division [58]. The EC_{50} values of compounds **8** and **9** against *Xoo* were 76.1 and 86.1 mg/L. However, the antibacterial activity of compound **10** (EC_{50} = 20.0 mg/L) was significantly increased when a fluorine atom was introduced into the para position of the benzene ring. In addition, the introduction of para-methyl or ortho-chlorine atoms made the compounds exhibit good antibacterial activity against *Xac*. For example, compounds **11** and **12** had EC_{50} values of 35.7 and 28.5 mg/L for *Xac*. Interestingly, compound **11** can cause fold and damage to cell surface morphology, and the higher the concentration of the compound, the greater the degree of damage on the cell surface [59].

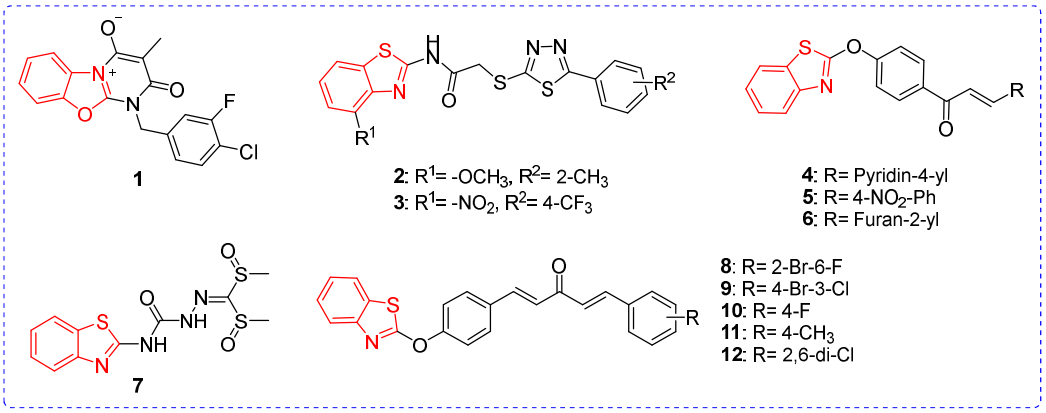

Figure 2. Chemical structure of benzoxazole and benzothiazole antibacterial active compounds **1**–**12**.

Table 1. Benzoxazole or benzothiazole antibacterial derivatives with antifungal activity.

Compound	Bacteria	Concentration	Antibacterial Activity	SAR/Physiology and Biochemistry
1	*Xoc*		47.6 [a]	The expression of SDH during oxidative phosphorylation is up-regulated.
	Xac		36.8 [a]	
2	*Xoo*	100 mg/L	52.4%	
3	*Rs*	100 mg/L	71.6%	The introduction of the nitro group and trifluoromethyl group plays a key role.
4	*Xoo*	100 mg/L	52.40%	The introduction of electron-withdrawing groups enhances the antibacterial activity of the compounds.
	Xac		50.97%	
	Rs		36.49%,	
7	*Xoo*		11.4 [a]	Cell morphology is altered and biofilm formation is inhibited.

10	*Xoo*	20.0 [a]	The introduction of the fluorine atom plays a key role.
11	*Xac*	35.7 [a]	The cell surface morphology is folded and damaged.
12	*Xac*	28.5 [a]	

[a] median effective concentration (EC_{50}, mg/L).

3. Antifungal Activity

There are a wide variety of fungal diseases in plants, and their distribution is widespread [60,61]. Fungal diseases not only affect the yield and quality of crops, but also some fungi can secrete toxins and metabolites that are harmful to humans when they infect crops [62,63]. At present, the use of chemical agents is still one of the main methods of fungal disease activity management. In recent years, the research on benzoxazole and benzothiazole fungicidal compounds has made great progress.

Some benzoxazoles or benzothiazoles have shown excellent fungicidal activity. For example, compound **13** (Figure 3) had an EC_{50} value of 0.3 mg/L (Table 2) for *Alternaria brassicae*, which was superior to the commercial agent carbendazim (EC_{50} = 47.0 mg/L) [64]. At a concentration of 90 mg/L, the protective effect and treatment activities of compounds **14** and **15** against *Botrytis cinerea (B. cinerea)* were greater than 88% [65]. The EC_{50} value of compound **16** for *B. cinerea* was 2.40 mg/L, and the introduction of fluorine or chlorine atoms to the phenyl was conducive to the improvement of fungicidal activity of the compound. For example, compounds **17** and **18** for *B. cinerea* had EC_{50} values of 1.81 and 1.69 mg/L. In addition, compound **16** may show fungicidal activity by binding to the active site of the sec14p target of fungi [66].

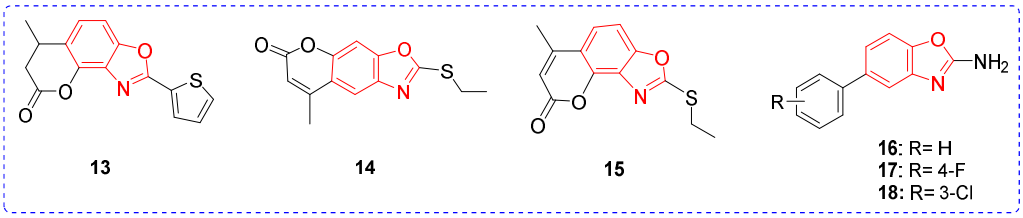

Figure 3. Chemical structure of benzoxazole antifungal active compounds **13**–**18**.

Table 2. Benzoxazole derivatives with antifungal activity.

Compound	Fungus	Concentration	Antifungal Activity	SAR/Molecular Docking
13	*Alternaria brassicae*		0.3 [a]	
14	*Botrytis cinerea*	90 mg/L	>88%	
16	*Botrytis cinerea*		2.40 [a]	Compound 16 may show fungicidal activity by binding to the active site of the sec14p target of fungi
18	*Botrytis cinerea*		1.69 [a]	The introduction of electron-absorbing groups is beneficial for antifungal activity.

[a] median effective concentration (EC_{50}, mg/L).

The IC_{50} value of compound **19** (Figure 4) for *B. cinerea* was 1.4 μM (Table 3), and the addition of methylene between benzothiazole and aryl increased the fungicidal activity of the compound [67]. At a concentration of 50 mg/L, the inhibitory rates of compound **20** against *Rhizoctonia solani (R. solani)*, *B. cinerea*, *Dothiorella gregaria (D. gregaria)*, and *Colletotrichum gossypii (C. gossypii)* were 92%, 97%, 89%, and 78%. Moreover, the introduction of chlorine atoms and trifluoromethyl compounds was not beneficial to the fungicidal activity of the compounds. For example, the inhibitory rates of compound **21** against *R. solani*, *B. cinerea*, *D. gregaria*, and *C. gossypii* were 40%, 67%, 35%, and 37% [68]. The EC_{90}

values of compound **22** on *Sphaerotheca fuliginea (S. fuliginea)* and *Pseudoperoniospora cubensis (P. cubensis)* were 6.17 and 46.32 mg/L, respectively [69]. The inhibition rates of compound **23** on *S. fuliginea* and *P. cubensis* were 67% [70] because the introduction of large steric groups reduced the fungicidal activity of the compound. Compounds **24**, **26**, and **28** showed inhibition rates of 69%, 55%, and 65% against *Phytophthora infestans (P. infestans)* at concentrations of 100 ppm. The fungicidal activities of compounds **24**, **26,** and **28** were reduced when chlorine atoms on the position-2 of the benzene ring were replaced by position-4 fluorine atoms of the benzene ring. For example, compounds **25**, **27**, and **29** have inhibition rates against *P. infestans* of 58%, 53%, and 58% [71].

The position-2 of benzothiazoles replaced by thioether is a good fungicidal scaffold structure, which has the value of further optimization and derivation. Currently, the framework is mainly combined with benzene, furanone, and thiadiazole. In the future, it may be considered to introduce thiazole, oxazole, and pyridine on sulfur atoms to optimize the structure.

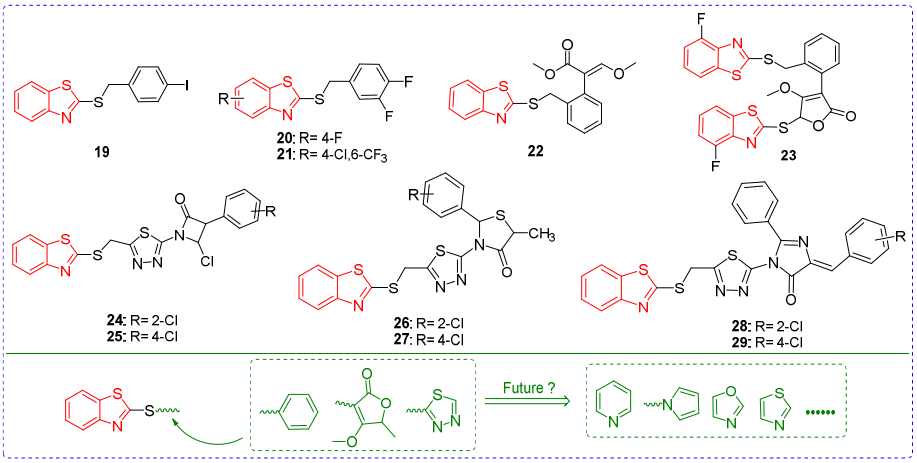

Figure 4. Chemical structures and modified fragments analysis of benzothiazole antifungal active compounds **19**–**29**.

Table 3. Benzothiazole derivatives with antifungal activity.

Compound	Fungus	Concentration	Antifungal Activity	SAR
19	*Botrytis cinerea*		1.4 [a]	The addition of methylene between benzothiazole and aryl increased the fungicidal activity of the compound
20	*Rhizoctonia solani*, *Botrytis cinereal*, *Dothiorella gregaria*, *Colletotrichum gossypii*	50 mg/L	92% 97% 89% 78%	
22	*Sphaerotheca fuliginea*, *Pseudoperoniospora cubensis*		6.17 [b] 46.32 [b]	
23	*Sphaerotheca fuliginea*, *Pseudoperoniospora cubensis*	50 mg/L	67% 67%	The introduction of large steric groups reduces the fungicidal activity of the compound.
25 **27** **29**	*Phytophthora infestans*	100 ppm	58% 53% 58%	The fungicidal activity improves when chlorine atoms on position-2 of the benzene ring are replaced by position-4 fluorine atoms.

[a] half maximal inhibitory concentration (IC_{50}, μmol/L). [b] concentration for 90% of maximal effect (EC_{90}, mg/L).

Amide bonds can form hydrogen bonds with target proteins, and compounds obtained by an organic combination of benzothiazole and amide often show good fungicidal activity [72]. At the concentration of 1000 mg/L, compound **30** (Figure 5) showed an inhibition rate of 88.9% (Table 4) against *B. cinereal*—the 4-nitrophenyl group was beneficial to improve the fungicidal activity of the compound. Interestingly, compound **30** showed better fungicidal activity in vivo than in vitro, suggesting that compound **30** may enhance plant disease resistance [73]. At a concentration of 50 mg/L, the inhibition rates of compound **31** on *B. cinerea* and *Gibberella zeae (G. zeae)* were 80% and 75%, respectively, suggesting that the introduction of permethric acid had no significant contribution to the fungicidal activity of the compound [74]. The EC_{50} values of Compound **32** against *Ustilago tritici*, *Puccinia striiformis*, *Puccinia triticina*, *Blumeria graminis*, *Dickeya oryzae*, and *Ustilag ohordei*are were all less than 0.8 mmol/L [75]. The inhibition rates of compounds **33** and **34** against *Helminthosporium maydis* were 78.6% and 80.6%. The fungicidal activity of the compound was not significantly improved by the introduction of electron-donating or electron-absorbing groups at position-6 of the benzothiazole ring. This suggests that the fungicidal activity of the compound in this structure is independent of the electron density at position-6 of the benzothiazole ring. In the future, spatial effects, hydrogen bonding, and water transport may be considered [76]. When thiazoles in the structure of compounds **33** and **34** were replaced with oxazoles, the fungicidal activity and broad spectrum of the compounds increased. For example, compound **35** had inhibition rates of 93.8%, 94.1%, 93.4%, 94.6%, and 94.5% against *R. solani*, *B. cinereal*, *G. zeae*, *Helminthosporium maydis*, and *Sclerotinia sclerotiorum (S. sclerotiorum)* [77]. Compound **36** showed a certain inhibitory effect on *Fusarium oxysporum (F. oxysporum)* (MIC 12.5 mg/mL) [78].

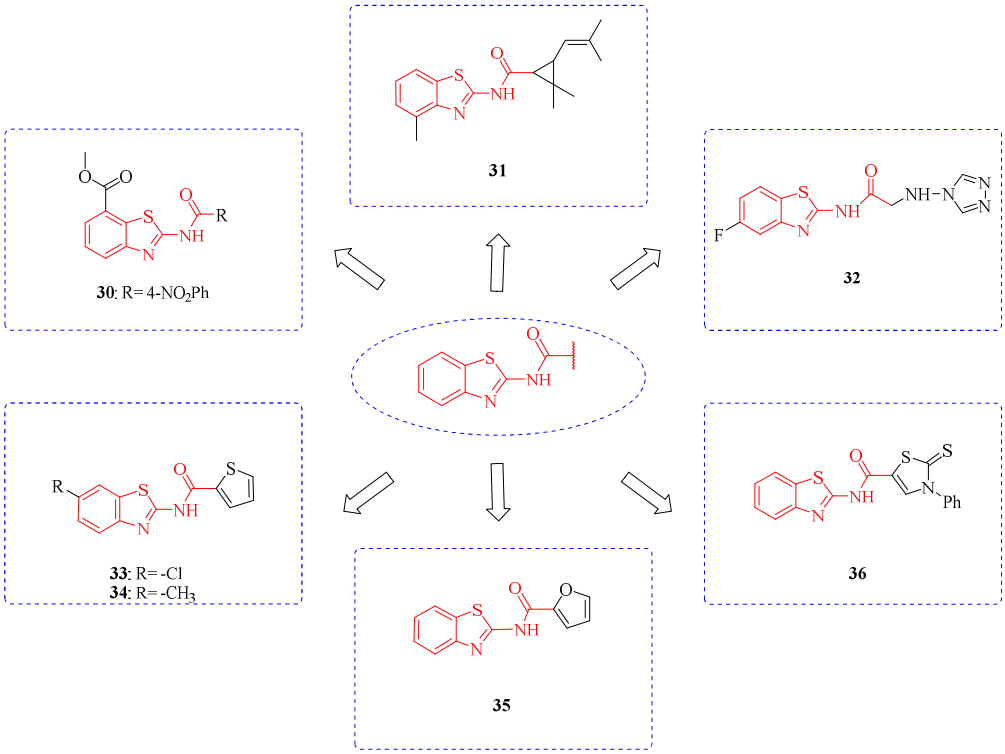

Figure 5. Chemical structures of benzothiazole fungicidal active compounds **30**–**36**.

Table 4. Benzothiazole derivatives with antifungal activity.

Compound	Fungus	Concentration	Antifungal Activity	SAR
30	*Botrytis cinereal*	1000 mg/L	88.9%	The introduction of the nitrophenyl group increases antifungal activity.
31	*Botrytis cinereal*, *Gibberella zeae*	50 mg/L	80% 75%	The introduction of permethric acid had no significant contribution to the fungicidal activity of the compound
32	*Ustilago tritici*, *Puccinia striiformis*, *Puccinia triticina*, *Blumeria graminis*, *Dickeya oryzae* *Ustilag ohordei*are		<0.8 [a]	
35	*Rhizoctonia solani*, *Botrytis cinereal*, *Gibberella zeae*, *Helminthosporium maydi*s, *Sclerotinia sclerotiorum*	50 mg/L	93.8%, 94.1%, 93.4%, 94.6%, 94.5%	The introduction of oxazoles plays a key role
36	*Fusarium oxysporum*		12.5 [b]	

[a] median effective concentration (EC_{50}, mmol/L). [b] minimum inhibitory concentration (MIC, mg/mL).

At the concentration of 100 mg/L, compound **37** (Figure 6) had inhibition rates of 38% (Table 5) to *Alternaria alternata* and 39% to *Aspergillus niger*, respectively. In addition, compound **37** may show fungicidal activity by inhibiting spore germination [79]. Under the condition of concentration of 250 mg/L, compound **38** *G. zeae* inhibition rate was 53.5% [80]. At the concentration of 100 mg/L, the inhibition rate of compound **39** against *Sclerotinia sclerotiorum* was 87.5%. However, the substitution of the alkyl group with the aromatic ring is not conducive to the fungicidal activity of the compound, for example, compound **40** showed 43.8% inhibition of *S. sclerotiorum* [81]. Under the condition of 50 mg/L, the inhibition rate of compound **41** to *R. solani* was 70.43% [82]. The inhibition rate of compound **42** against *F. oxysporum* was 60.53% [83]. At the concentration of 10 mg/L, the average inhibitory zone diameter of compound **43** against *Aspergillus oryzae (A. oryzae)* was 0.81 mm. However, the replacement of chlorine atoms with nitro groups had no significant effect on the fungicidal activity of compounds; for example, the average diameter of the inhibition zone of compound **44** against *A. oryzae* was 0.81 mm [84]. At the concentration of 50 mg/L, the inhibitory activities of compounds **45** and **46** against *Rape sclerotinia rot* were 80.08% and 81.61%, respectively [85]. The ED_{50} values of compounds **47** and **48** for *R. solani* are 0.96 μM and 1.48 μM, respectively, which may be due to amines having stronger alkalinity than imines. In addition, compound **48** binds to the CYP51 site of fungi, hindering the synthesis of fungal cell membranes and, thus, inhibiting the normal growth of fungi [86].

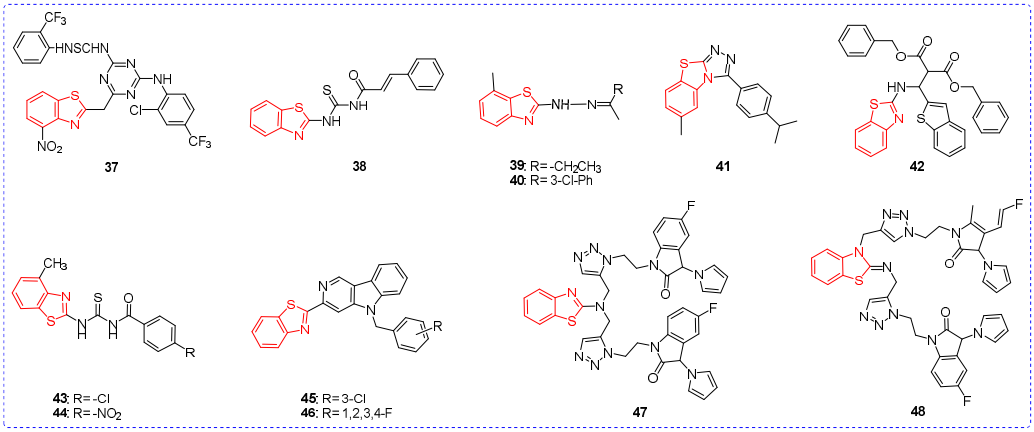

Figure 6. Chemical structures of benzothiazole fungicidal active compounds **37**–**48.**

Table 5. Benzothiazole derivatives with antifungal activity.

Compound	Fungus	Concentration	Antifungal Activity
37	*Alternaria alternate*, *Aspergillus niger*	100mg/L	38% 39%
38	*Gibberella zeae*	250 mg/L	53.5%
39	*Sclerotinia sclerotiorum*	100mg/L	87.5%
41	*Rhizoctonia solani*,	50 mg/L	70.43%
42	*Fusarium oxysporum*	50 mg/L	60.53%
43	*Aspergillus oryzae*	10 mg/L	0.81 [a]
46	*Rape sclerotinia rot*	50 mg/L	81.61%
47	*Rhizoctonia solani*,		0.96 [b]

[a] The inhibitory zone diameter(mm). [b] median effective concentration (EC_{50}, μM).

4. Antiviral Activity

Effective management of plant viral diseases has been one of the hotspots in the field of plant protection [87–89]. Plants do not have a complete immune metabolism system, and, once the virus invades the plant, it will reproduce indefinitely in the plant until the plant dies [90,91]. Therefore, plant viral diseases are more difficult to manage than bacterial diseases, fungal diseases, pests, and weeds [92–94]. Many studies have been conducted on benzothiazoles against plant virus diseases; some have good antiviral activities. For example, at the concentration of 500 mg/L, the treatment activities of compounds **49** and **50** (Figure 7) against tobacco mosaic virus (TMV) were 52.23% and 54.41% (Table 6), respectively [95]. The electron-donating group in the benzothiazole ring may be an important factor for the antiviral activity of compounds **49** and **50**. The protective activity of compound **51** against TMV was 39.27%. In addition, the introduction of chlorine atoms increased the antiviral activity of the compound; for example, the protective activities of compounds **52** and **53** against TMV were 55.96% and 54.21% [96]. The inhibition rate of compound **54** against TMV was 28.2%, while its racemic activity against TMV was 35.4% [97]. Compounds **55** and **56** had treatment activities against TMV of 37.9% and 35.8%. When the alkyl part of the amino phosphonate of these compounds was ethyl, the compounds showed better antiviral activity. For example, the treatment activity of compound **57** against TMV was 48.1% [98]. The treatment activity of compound **58** against TMV was 48.2%. Replacing the fluorine atom of compound **59** with a methoxy group had no significant effect on the antiviral activity of the compound. For example, the treatment activity of compound **59** against TMV was 47.2% [99]. The treatment, protection, and passivation of compound **60** against TMV were 33.2%, 65.1%, and 45.7%, while, for compound **61** against TMV, they were 74.3%, 78.7%, and 94.3%. Molecular docking found that benzo-

thiazole rings are important for the antiviral activity of these compounds, and the hydrazone's structure can affect the compounds' antiviral activity [100]. The combination of benzothiazoles with diesters or amino phosphonate had good antiviral activity, which showed the advantage of the skeleton structure in antiviral activity. Currently, benzothiazole, thiazole, benzothiophene, and benzofuran structures are mainly introduced into benzothiazole scaffolds. In the future, the introduction of thiazole, oxazole, and morpholine rings may be considered to find molecules with higher antiviral activity.

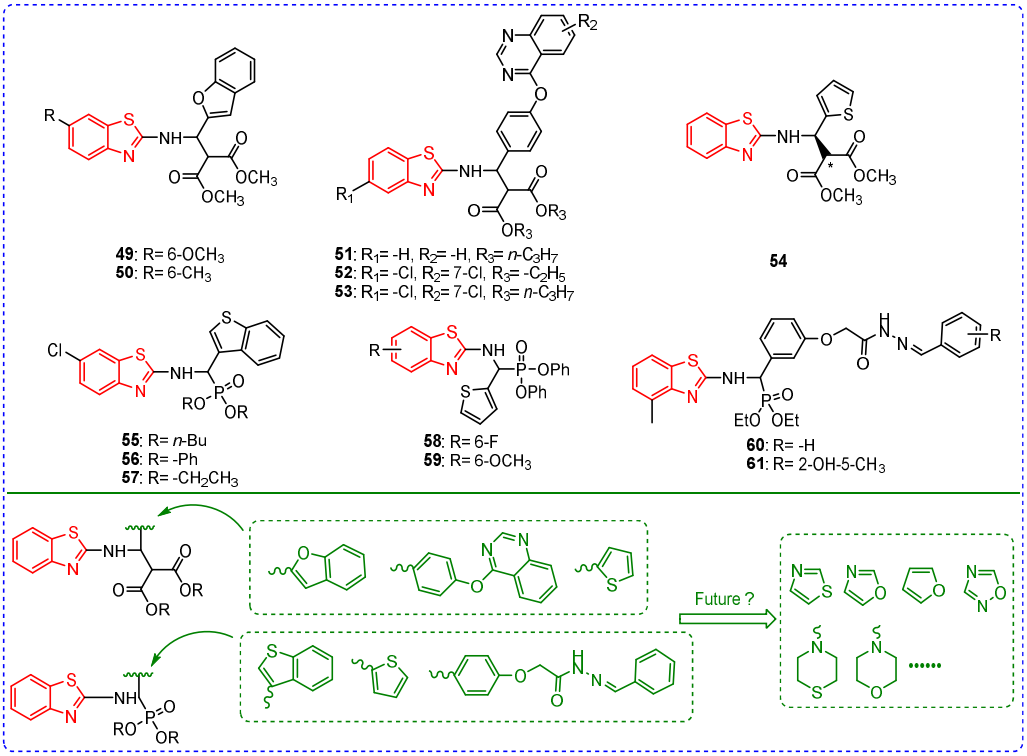

Figure 7. Chemical structures and modified fragments analysis of benzothiazole antiviral active compounds **49**–**61**.

Table 6. Benzothiazole derivatives with antiviral activity.

Compound	Virus	Concentration	Antiviral Activity	SAR
50	TMV	500 mg/L	54.41% [a]	The electron-donating group in the benzothiazole ring may play a key role
52	TMV	500 mg/L	55.96% [b]	The introduction of the chlorine atom plays a key role
54	TMV	500 mg/L	28.2%	
57	TMV	500 mg/L	48.1% [a]	
58	TMV	500 mg/L	48.2% [a]	
61	TMV	500 mg/L	74.3% [a] 78.7% [b] 94.3% [c]	The hydrazone's structure can affect the compounds' antiviral activity.

[a] treatment activity, [b] protective activity, [c] passivation activity.

At a concentration of 500 mg/L, the treatment activity of compound **62** (Figure 8) against TMV was 52.9% (Table 7), and the replacement of straight-chain alkanes with branched-chain alkanes resulted in a decrease in the antiviral activity of the compound; for example, compound **63** had a treatment activity against TMV of 46.6% [101]. The substitution of alkyl of compound **64** (30.9%) with benzene ring was beneficial to the improvement of the anti-TMV activity of compound **64** (30.9%). For example, compounds **65**, **66**, and **67** had anti-TMV activities of 32.1%, 38.1%, and 44.0%, respectively, at a concentration

of 0.05% [102]. Under the condition of concentration of 50 mg/L, the inhibition rate of compound **68** against Cucumber mosaic virus (CMV) was 46.3%, while the growth of the alkyl chain had little effect on the antiviral activity of the compound; for example, the inhibition rate of compound **69** against CMV was 45.1% [103]. At the concentration of 500 mg/L, the inhibition rate of compound **70** on TMV was 44.5%, while the substitution position of the methyl group in the benzothiazole ring had no significant effect on the antiviral activity of the compound. For example, the inhibition rate of compound **71** on TMV was 45.1% [104]. The treatment activity of compound **72** against TMV was 39.3%. When the oxazole ring was replaced by a thiazole ring, the antiviral activity of the compound increased. For example, the treatment activity of compound **73** against TMV was 52% [105]. The protective and passivation activities of compound **74** against TMV were 78.3% and 79.5%, and the protective and passivation activities of compound **75** against TMV were 83.3%. The replacement of chlorine atoms with nitro atoms did not significantly change the antiviral activity of the compound [55].

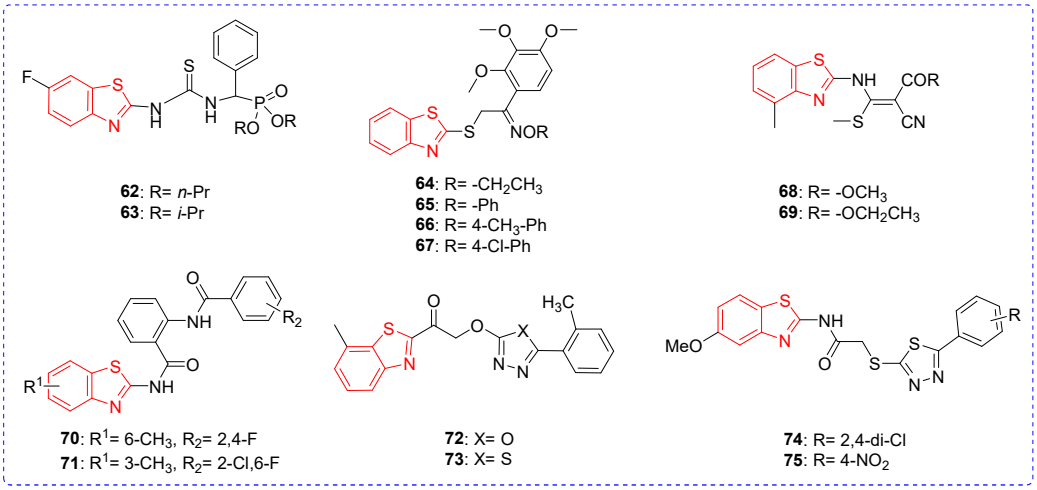

Figure 8. Chemical structures analysis of benzothiazole antiviral active compounds **62**–**75.**

Table 7. Benzothiazole derivatives with antiviral activity.

Compound	Virus	Concentration	Antiviral Activity	SAR
62	TMV	500 mg/L	52.9% [a]	Straight-chain alkane is beneficial to the antiviral activity of the compound
67	TMV	500 mg/L	44.0%	The introduction of the benzene ring is beneficial to the improvement of the anti-TMV activity of the compound
68	CMV	50 mg/L	46.3%	The growth of the alkyl chain had little effect on the antiviral activity of the compound
71	TMV	500 mg/L	45.1%	
73	TMV	500 mg/L	52%	The introduction of the thiazole ring is beneficial to the antiviral activity of the compound
75	TMV	500 mg/L	83.3% [b,c]	

[a] treatment activity, [b] protective activity, [c] passivation activity.

5. Herbicidal Activity

Weeds compete with crops for nutrients, sunlight, and water, harming the normal growth and yield of crops. Furthermore, some weeds contain toxins in their seeds or pollen that can harm human health [106,107]. The use of chemical herbicides is the most effective and cost-effective way to manage weeds [108,109]. Currently, 263 species of weeds worldwide have shown resistance to 23 herbicides [110,111]. Therefore, the discovery of new herbicides is an urgent need for weed management [112,113].

Although the herbicidal activities of benzoxazole and benzothiazole derivatives have been less reported, some compounds have shown excellent herbicidal activities. For example, compounds **76** and **77** (Figure 9) both achieved 90% (Table 8) herbicidal activity against the monocotyledon weeds *Digitaria sanguinalis* and *Setaria viridis* at a concentration of 75 g/ha [114]. In addition, compounds **76** and **77** showed good safety on the stems and leaves of rice. At a concentration of 100 μg/L, compound **78** had 93% and 85% herbicidal activities against the roots and stems of *Chenopodium album (C. album)*, respectively. In addition, compound **78** may show herbicidal activity by inhibiting the growth of the taproot and stem of the *C. album* [68]. Under the condition of 37.5 g/hm^2, compound **79** showed 100% inhibition rate against *Setaria viridis*, *Ditaria sanguinalis*, and *Abutilon theophrasti*. The introduction of the alkoxy group was beneficial to increase the herbicidal activity of the compound [115]. The inhibition rate of compound **80** to *Amaranthus retroflexus (A. retroflexus)* was 100% at 1400 g/ha, and the introduction of the nitro group improved the herbicidal activity of the compound [116]. Compounds **81**, **82**, and **83** showed 99% herbicidal activities against *A. retroflexus* at a concentration of 10 mg/L, and the introduction of fluorine may have increased the herbicidal activity of the compounds [117]. Under the condition of 37.5 g/hm^2, the inhibition rate of compound **84** against *Abutilon theophrasti*, *Cyperus iria*, *Rumex acetasa*, and *Eclipta prostrate* was greater than 80%, which has the prospect of further development [118].

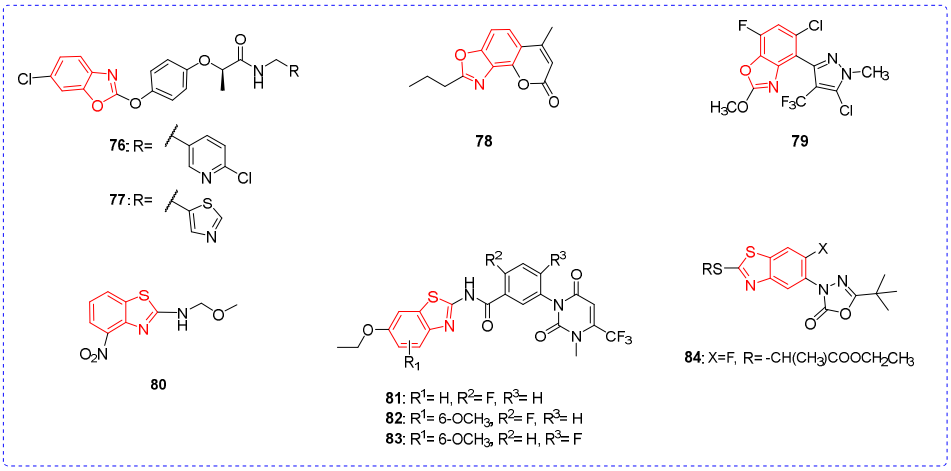

Figure 9. Chemical structures analysis of benzoxazole and benzothiazole herbicidal active compounds **76**–**84**.

Table 8. Benzoxazole and benzothiazole derivatives with herbicidal activity.

Compound	Weeds	Concentration	Herbicidal Activity	SAR/Physiology and Biochemistry
76	*Digitaria sanguinalis*, *Setaria viridis*	75 g/ha	90%	The compound shows good safety on the stems and leaves of rice
78	the roots of *Chenopodium album*, the stems of *Chenopodium album*	100 μg/L	93% 85%	The compound inhibits the growth of the taproot and stem of the *Chenopodium album*
79	*Setaria viridis*, *Ditaria sanguinalis*, *Amaranthus retroflexus*	37.5 g/hm^2	100%	The introduction of the alkoxy group was beneficial to increase the herbicidal activity.
81	*Amaranthus retroflexus*	10 mg/L	99%	The introduction of fluorine may have increased the herbicidal activity of the compounds
84	*Abutilon theophrasti*, *Cyperus iria*, *Rumex acetasa*, *Eclipta prostrate*	37.5 g/hm^2	>80%	

6. Insecticidal Activity

The wide variety of pests is an important factor in crop yield reduction and some pests are characterized by the outbreak, such as *Pyrausta nubilalis* [119], *Helicoverpa armigera* [120], *Oriental armyworm* [121,122], and *Locust* [123–125]. Traditional insecticides have played an irreplaceable role in pest control, and the long-term use of traditional insecticides not only leads to the rapid increase in pest resistance but also pollutes the environment and threatens human health [126–128]. The discovery of insecticides has always been a hot topic in pesticide research [129]. However, there are relatively few reports on the insecticidal activity of benzoxazole and benzothiazole, which may be strengthened in the future. The Maximum Likelihood Programmer (MLP) calculation showed that the combination of benzothiazole and pyridine could increase the antifeedant activity of the compounds. For example, LC_{50} of compounds **85**–**88** (Figure 10) against *Spodoptera litura* were 0.38, 0.24, 0.10, and 0.07, respectively [130,131]. The insecticidal activity of compounds **86**, **87**, and **88** was significantly higher than that of compound **85**, which may be due to the different electronegativity of groups introduced at position-6 of benzothiazole. Perhaps this is a hint that we can try to introduce strong electron-absorbing groups such as nitro and trifluoromethyl to benzothiazole in the future to find new insecticides.

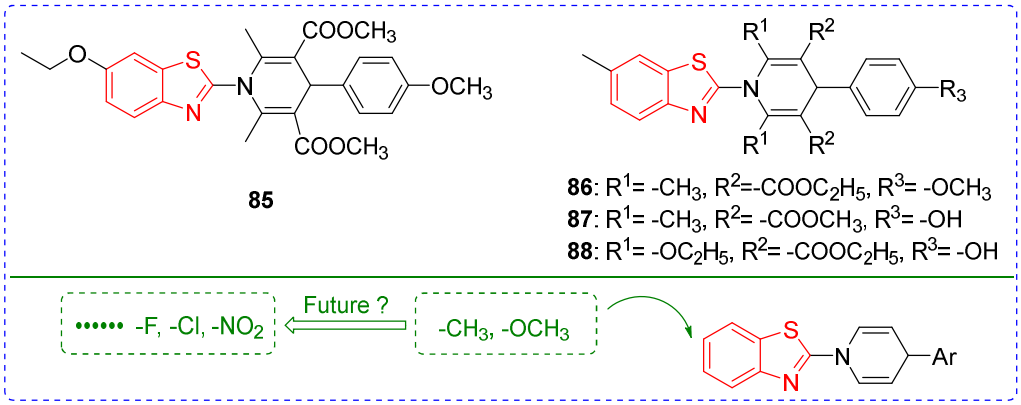

Figure 10. Chemical structures and modified fragments analysis of benzothiazole insecticidal active compounds **85**–**88**.

At a concentration of 1 mg/L, the insecticidal activity of compound **89** (Figure 11) against *Spodoptera exigua* was 100% (Table 9); perhaps the strong electron-absorbing group trifluoromethyl played an important role in the insecticidal activity of compound **89** [132]. The insecticidal activity of compound **90** against *Mythimna separata Walker* was 62.1%, which was better than that of the lead compound magnolol [133]. Under the concentration of 5 g/L, the mean killing time of compound **91** to cockroaches was 147 min, which was better than that of commercial Parathion (280 min) [134]. The LC_{50} of compound **92** for *Tetranychus urticae* was 0.07 mg/L [135]. The insecticidal activity of compound **93** against *Aphis* was 54% at a concentration of 200 mg/mL [136]. The ED_{50} value of compound **94** for *Achaea janata (A. janata)* was 19.3 μg/cm^2. The insecticidal activity of the compounds was significantly improved when fluorine atoms on the benzene ring were replaced with methoxide. For example, compounds **95** and **96** had ED_{50} values of 7.0 and 5.2 μg/cm^2 for *A. janata*, respectively. Meanwhile, the insecticidal activities of compounds **95** and **96** against *Spodoptera litura* were greater than 95% at a concentration of 0.2 μg/insect [137]. The LC_{50} value of compound **97** against *Bollworm* was 4.90 mg/L [138]. The insecticidal activity of compound **98** against the *Diamondback moth* was 88% at a concentration of 1 mg/L. In addition, at high concentrations, compound **99** showed good insecticidal activity by activating the release of calcium ions from the central neurons of insects [139].

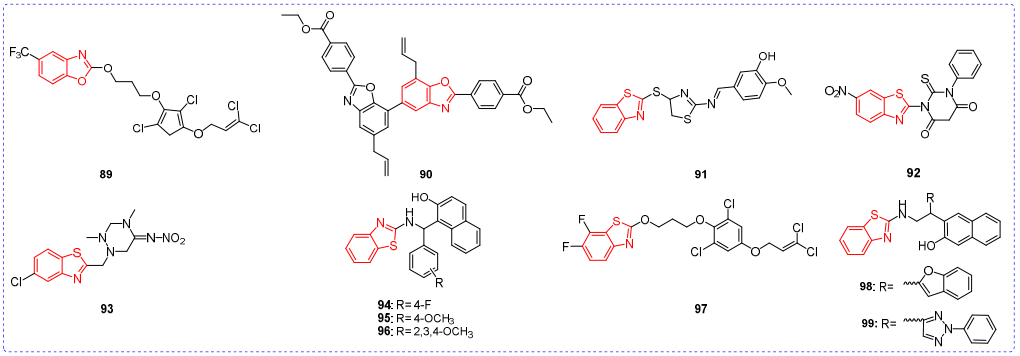

Figure 11. Chemical structures of benzothiazole insecticidal active compounds **89**–**99**.

Table 9. Benzoxazole and benzothiazole derivatives with insecticidal activity.

Compound	Pests	Concentration	Insecticidal Activity	SAR
88	*Spodoptera litura*		0.07 [a]	The introduction of the ethoxy group may play a key role
89	*Spodoptera exigua*	1 mg/L	100%	The strong electron-absorbing group may play a key role
90	*Mythimna separata Walker*	1 mg/L	62.1%	
91	cockroaches	5 g/L	147 [b]	
92	*Tetranychus urticae*		0.07 [c]	
93	*Aphis*	200 mg/mL	54%	
96	*Achaea janata*		5.2 [d]	Fluorine atom on the benzene ring improves the insecticidal activity of the compound
97	*Bollworm*		4.90 [c]	
98	*Diamondback moth*	1 mg/L	88%	

[a] the calculation of LC_{50}/ LD_{50} using the Maximum Likelihood Programmer (MLP). [b] the mean killing time (min). [c] lethal concentration 50 (LC_{50}, mg/L). [d] a median effective concentration (EC^{50}, μg/cm^2).

7. Conclusions

Benzothiazoles and benzoxazoles not only have a bicyclic structure, but also have seven modifiable sites, illustrating the important value of benzothiazoles and benzoxazoles in the discovery of pesticides. It is worthy to carry out more exploration and research based on benzothiazoles or benzoxazoles. In recent years, benzoxazole and benzothiazole derivatives have been increasingly studied as fungicides, antimicrobials, herbicides, antiviral agents, and insecticides. However, the research on the mechanism of action and the discovery of new targets of benzoxazole and benzothiazole derivatives compounds is still weak and needs to be further strengthened in the future, which is a key factor restricting the discovery of new green pesticides. We systematically reviewed the application of benzoxazole and benzothiazole derivatives compounds in the discovery of new agrochemicals, summarized the antibacterial, fungicidal, and antiviral agents, as well as herbicidal and insecticidal activities, of the compounds, and discussed the structural–activity relationship and mechanism of action of the active compounds, aiming to provide new clues and inspiration for the discovery of new pesticides.

Author Contributions: J.C. conceived and designed the title; the data were analyzed and interpreted by Y.Z. (Yue Zou), Y.Z. (Yong Zhang), X.L., H.S., Q.C. and S.W.; Y.Z. (Yue Zou) wrote the paper; J.C. and C.Y. critically revised the paper with regard to important intellectual content. All authors have read and agreed to the published version of the manuscript.

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Abbreviations

SAR	Structural–activity relationship
Xoc	*Xanthomonas oryzae* pv.*oryzicola*
Xac	*Xanthomonas citri* subsp. *Citri*
Xoo	*Xanthomonas oryzae* pv.*oryzae*
SDH	Succinate dehydrogenase
Rs	*Ralstonia solanacearum*
B. cinerea	*Botrytis cinereal*
R. solani	*Rhizoctonia solani*
D. gregaria	*Dothiorella gregaria*
C. gossypii	*Colletotrichum gossypii*
S. fuliginea	*Sphaerotheca fuliginea*
P. cubensis	*Pseudoperoniospora cubensis*
P. infestans	*Phytophthora infestans*
G. zeae	*Gibberella zeae*
S. sclerotiorum	*Sclerotonia sclerotiorum*
F. oxysporum	*Fusarium oxysporum*
A. oryzae	*Aspergillus oryzae*
TMV	tobacco mosaic virus
CMV	Cucumber mosaic virus
C. album	*Chenopodium album*
A. retroflexus	*Amaranthus retroflexus*
A. janata	*Achaea janata*
MLP	Maximum Likelihood Programmer

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