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Orthoquinone, Cyclic, and Acyclic α-Diketone Natural Products: From Food Applications to Pharmacotherapy

Hidayat Hussain[1] | Daijie Wang[1] | Satyajit D. Sarker[2] | Lutfun Nahar[3] | Nilufar Z. Mamadalieva[4,5] | Jianbo Xiao[6]

[1]International Joint Laboratory of Medicinal Food Development and Health Products Creation, Biological Engineering Technology Innovation Center of Shandong Province, Heze Branch of Qilu University of Technology (Shandong Academy of Sciences), Heze, China | [2]Centre for Natural Products Discovery, School of Pharmacy and Biomolecular Sciences, Liverpool John Moores University, Liverpool, UK | [3]Laboratory of Growth Regulators, Palacký University and Institute of Experimental Botany, The Czech Academy of Sciences, Olomouc, Czech Republic | [4]Institute of the Chemistry of Plant Substances of the Academy Sciences of Uzbekistan, Tashkent, Uzbekistan | [5]Department of Pharmacy and Chemistry, Faculty of Dentistry and Pharmacy, Alfraganus University, Tashkent, Uzbekistan | [6]Nutrition and Bromatology Group, Department of Analytical Chemistry and Food Science, Faculty of Science, Instituto de Agroecoloxía e Alimentación (IAA), Universidade de Vigo, Campus Auga, Ourense, Spain

Correspondence: Hidayat Hussain (hussainchem3@gmail.com) | Jianbo Xiao (jianboxiao@uvigo.es)

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ABSTRACT

The structural diversity of natural products is vast and fascinating, and they have been recognized as an enormously diverse source of new lead compounds. Many bioactive secondary metabolites have a 1,2-carbonyl pharmacophore in which the orthoquinone moieties are normally associated with interacting with and molecularly recognizing the cellular target. For therapeutic effects, especially antimicrobial effects, the presence of orthoquinone entities is crucial and in some cases necessary. This review will describe orthoquinone natural products and their use in the food packaging industry along with their other therapeutic properties. Natural products are considered one of the most important sources of approved drugs with diverse mechanisms of action. The orthoquinone moiety enhances bioavailability, biological potential, and stability in many pharmacologically active natural products. The vast majority of the orthoquinone metabolites were found to have antimicrobial, antioxidant, antiviral, cytotoxic, and anti-inflammatory effects. In particular, significant antimicrobial potential towards various microorganisms, especially foodborne pathogens and plant pathogens, is demonstrated by numerous orthoquinones presented in this review. Their antimicrobial effects suggest that they could be used as natural food preserving agents to eliminate or control growth of pathogens and spoilage organisms.

1 | Introduction

The orthoquinone (1,2-dicarbonyl) motif is an important life-related structure that is found in natural products and modern pharmaceuticals. Tanshinone IIA is a transcription factor inhibitor and this molecule and related tanshinones possess numerous biological activities including antimicrobial, antioxidant, anticancer, antidiabetic, antimalarial, anti-Alzheimer, neuroprotective, cardioprotective, antiadipogenic, and anti-inflammatory effects. Tanshinones have been shown to be an attractive therapeutic strategy in clinical trials for pulmonary hypertension, cardiovascular disease, lung disease, polycystic ovarian syndrome, acute myocardial infarction, and childhood acute promyelocytic leukemia (M. Wang et al. 2019; Subedi and Gaire 2021). Majority of tanshinones were reported from *Salvia* (Lamiaceae) and this group of plants is one of the most important traditional Chinese herbs with a long history of use in both medicine and healthy food. In the *Salvia* genus, *S. miltiorrhiza* is also a healthy plant that can be used as tea (Shi et al. 2019).

Natural orthoquinone compounds called licoagrodione (W. Li et al. 1998) and mansonone C (Bettòlo et al. 1965) showed significant antimicrobial activity, whereas sophoradione was cytotoxic to KB tumor cells. Scleroderolide and dunnione showed strong antimicrobial and anticancer effects (C.-C. Shen et al. 2006). The ability of the dicarbonyl moiety to attach to proteins in the body, thereby enhancing bioavailability, has led to the development of many known dicarbonyl-containing molecules that are in clinical use. Examples include the anticancer drugs indibulin and biricodar, the anti-HCV drug boceprevir and the dermatological drug fluocortin butyl, a synthetic corticosteroid with a wide range of topical to systemic activity (M. Wang et al. 2019) (Figure 1). In addition, organic synthesis and materials science often use dicarbonyl-containing compounds as valuable synthetic intermediates and precursors. For example, aromatic-substituted quinoxalines have been synthesized from dicarbonyl-containing compounds. These are widely used as photoinitiators and fluorescence-based sensors (Ashton et al. 2015; L. Wang et al. 2013; C. T. Chen et al. 2006; Sessler et al. 2006).

The food industry can benefit from the use of naturally sourced antioxidants, which are valuable bioactive compounds with proven potential. As well as being used in functional foods, it is becoming more popular as an alternative to synthetic antioxidants. This is because it can enhance stability and prevent oxidative degradation during processing and storage. The circular economy is being given consideration in terms of the use of natural antioxidants from agricultural food byproducts and underutilized plant materials (Lourenço et al. 2019). This review article covers various orthoquinones that possess significant antioxidant potential. For example, the majority of tanshinones and mansonones showed promising antioxidant activity, which is an important finding in the field. Additionally, pyrroloquinoline quinone (PQQ) is the most active orthoquinone, demonstrating potent antioxidant effects. Orthoquinones have a strong antioxidant capacity, which makes them attractive for use in the preservation of food, including meat, edible oils, and plant foods.

Orthoquinones have significant antimicrobial properties, which make them effective against a variety of organisms. In consideration of their antimicrobial properties, these natural products find application in the packaging of foodstuffs, thereby contributing to an augmentation in the shelf life of a wide variety of edibles. Plant diseases are responsible for the failure of an estimated 10%–15% of the world's major crops each year, resulting in direct economic losses of several hundred billion dollars. In particular, 70%–80% of these diseases are caused by pathogenic fungi, which negatively impact crop growth and yield (Peng et al. 2021; Sar et al. 2023). This review reports on many orthoquinones that have significant antifungal properties toward various fungi. Notably, several orthoquinone natural products exhibited potent antifungal activity against plant pathogen fungi.

This review will provide comprehensive and critical coverage of approximately 270 new orthoquinone natural products. The format focuses on the discovery of new orthoquinone natural products from natural sources along with structural diversity and biological effects. Although many biological effects have been reported for numerous orthoquinone natural products. However, we focus on highlighting the antimicrobial and antioxidant properties of orthoquinone metabolites and their role in the food industry. The significant antimicrobial and antioxidant potential of these orthoquinone metabolites is an indication that these fungal metabolites may have potential as natural preservatives in the food industry. In addition, the antimicrobial properties of orthoquinones may also be useful in improving the shelf life of food products.

2 | Diterpene

2.1 | Tanshinones

Tanshinone IIA (**1**) (Figure 2), the first member in the tanshinones family, is iolated from *S. miltiorrhiza* (Danshen). Danshen is listed in the Chinese Pharmacopeia and has various traditional and pharmacological uses ascribed to it (Table 1) (Gao et al. 2012; J. Y. Han et al. 2008; S. H. Han et al. 2008; X. H. Wang et al. 2007; Yagi et al. 1989; Adams et al. 2006; Cheng 2006; L. Zhou et al. 2005; Takahashi et al. 2002). *S. miltiorrhiza* is employed as a health-promotional food and medicinal plant. Literature revealed that *S. miltiorrhiza* was employed as a food supplement due to its significant activities on microcirculation (Hügel 2015; X. Wang et al. 2022). Moreover, various studies have illustrated that Tanshinone IIA (Tan IIA) is the main component of *S. miltiorrhiza* that possesses anti-inflammation activity, functions by expanding blood vessels, has antithrombosis activity, and lowers blood pressure (X. Wang et al. 2022; Y. Huang et al. 2020; H. Chen et al. 2018). Additionally, Tan IIA has been reported to treat neuroinflammation (H. X. Liu et al. 2013; M. L. Liu et al. 2013; X. Liu et al. 2013). In another study, Tan IIA showed significant

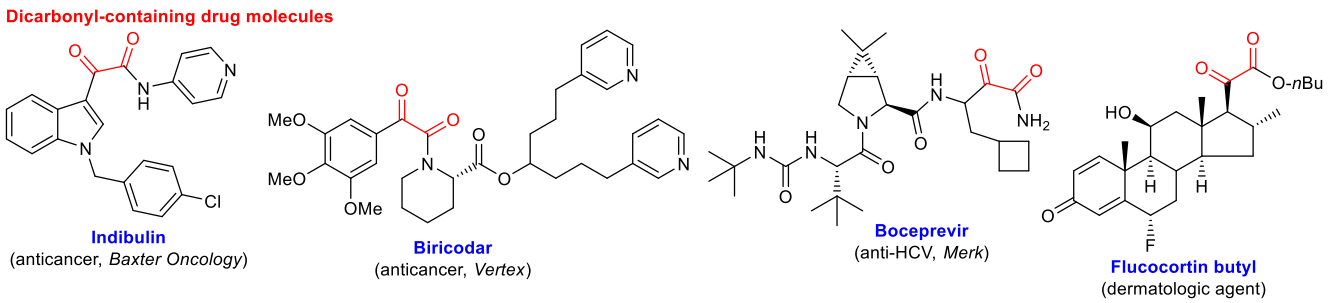

FIGURE 1 | Dicarbonyl comprising drug molecules.

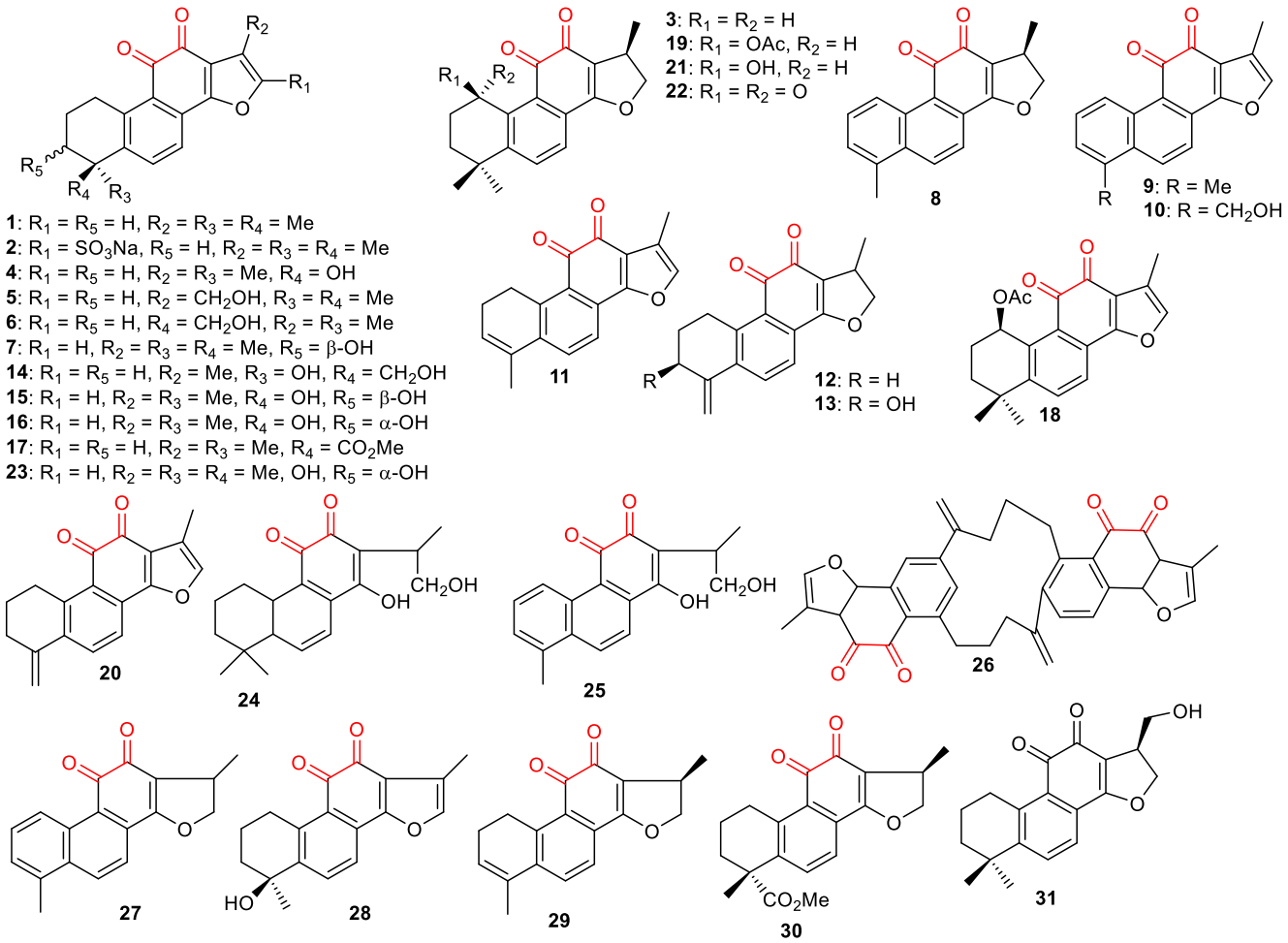

FIGURE 2 | Structures of tanshinones **1–31**.

protective effects which supported the potential of it and *S. miltiorrhiza* as food supplements to treat brain diseases (X. Wang et al. 2022).

Various reviews have been published on the pharmacological applications of Tan IIA (**1**) (R. Guo et al. 2020; Ansari et al. 2021). Numerous experimental and clinical data demonstrated that compound **1** can prevent or slow down the process of cardiovascular diseases, cancer, hepatic fibrosis, ischemic encephalopathy, and neurodegenerative diseases (J. Y. Han et al. 2008; S. H. Han et al. 2008; X. H. Wang et al. 2007; Takahashi et al. 2002). In China, tanshinone IIA (**1**) is employed either alone or as its sodium tanshinone IIA sulfate (**2**) or in the form of tablets (Fufang Danshen Dripping Pill) to treat cardiovascular diseases (Gao et al. 2012). Moreover, Fufang Dripping Pill passed Phase II clinical trials in the US with chronic stable angina pectoris (see http://clinicaltrials.gov/, No. NCT00797953) (Gao et al. 2012).

In addition, pharmacokinetic potentials of compound **1** have been investigated in recent years (Bi et al. 2008; Mao et al. 2007; Qiao et al. 2003). Interestingly, it illustrated antiatherosclerotic effects in various in vivo models viz., mice and rabbits (F. Tang et al. 2007; F. T. Tang et al. 2011; Fang et al. 2007, 2008; X. Li et al. 2010; Du et al. 2005; Gong et al. 2009). Clinically, tanshinone IIA (**1**) has been employed in China in combination theraphy with other lipid-lowering pharmaceutical drugs to treat atherosclerosis along with other cardiovascular issues (Q. Guo 2010). In another study, Y. K. Huang (2010) demonstrated that combination therapy of STS/atorvastatin significantly reduced the level of C-reactive protein and inhibited vascular inflammation in patients with hyperlipidemia, hypertension, and diabetes. Tanshinone IIA (**1**) also illustrated cardioprotective effects (D.-D. Sun et al. 2008; Shan et al. 2009; L. Yang et al. 2009; Y. S. Li et al. 2008; D. X. Zhou et al. 2008; Yan et al. 2008; Z. H. Wang et al. 2006; G. Y. Zhou et al. 2003) and some authors reported that this compound protects against patients having myocardial infarction (G. Xu et al. 2009; W. Xu et al. 2009).

Compounds **1** and **3–17** (Figure 2) were reported to be isolated from *S. miltiorrhiza* and all were evaluated against A549 (lung), SK-MEL-2 (melanoma), SK-OV-3 (ovary), XF498 (CNS), and HCT-15 (colon) cancer cell lines. Interestingly, these compounds possess cytotoxic effects in which the IC_{50} ranged from 0.2 to 8.1 μg/mL (Table 1). Among these compounds, orthoquinones **14–16** were the most potent with IC_{50}: ≤ 1 μg/mL (Ryu et al. 1997). Tanshinone I (**9**) which was reported from *S. miltiorrhiza* had an inhibition factor of IC_{50} = 38 μM). Additionally, (**9**) inhibited $sPLA_2$ and $cPLA_2$ with IC_{50}: 11 and 82 μM, respectively (S. Y. Kim et al. 2002). Cryptotanshinone (**3**) and 15,16-dihydrotanshinone I (**8**) possessed significant antimicrobial effects toward *Bacillus subtilis*, *B. thuringiensis*, *Micrococcus luteus*, *Staphylococcus aureus*, and *Staphylococcus epidermidis*

TABLE 1 | Compounds isolated from *Salvia miltiorrhiza* and their biological effects.

Name	Activity	References
Tanshinone IIA (**1**)	*Anticancer*: A549: IC_{50}: 0.9 μg/mL; SK-OV-3: IC_{50}: 1.3 μg/mL; K-MEL-2: IC_{50}: 0.9 μg/mL; XF498: IC_{50}: 0.4 μg/mL; HCT15: IC_{50}: 0.8 μg/mL; HCT15: IC_{50}: 2.3 μM; BGC-283: IC_{50}: 2.1 μM; A549: IC_{50}: 1.4 μM; and A2780: IC_{50}: 2.8 μM	Ryu et al. (1997), H.-P. Lee et al. (2017), and Lin et al. (2015)
Cryptotanshinone (**3**)	*Anticancer*: A549: IC_{50}: 0.7 μg/mL; SK-OV-3: IC_{50}: 2.5 μg/mL; K-MEL-2: IC_{50}: 1.6 μg/mL; XF498: IC_{50}: 0.5 μg/mL; HCT15: IC_{50}: 1.2 μg/mL; HCT-8: IC_{50}: 3.9 μM; BGC-823: IC_{50}: 8.3 μM; and HCT-8: IC_{50}: 3.9 μM	Ryu et al. (1997), S. A. Kim et al. (2018), and J. Xu et al. (2018)
Tanshinol B (**4**)	*Anticancer*: A549: IC_{50}: 3.4 μg/mL; SK-OV-3: IC_{50}: 3.7 μg/mL; K-MEL-2: IC_{50}: 1.4 μg/mL; XF498: IC_{50}: 1.8 μg/mL; and HCT1 5: IC_{50}: 0.8 μg/mL	Ryu et al. (1997)
Przewaquinone A (**5**)	*Anticancer*: A549: IC_{50}: 2.3 μg/mL; SK-OV-3: IC_{50}: 1.7 μg/mL; K-MEL-2: IC_{50}: 1.9 μg/mL; XF498: IC_{50}: 1.6 μg/mL; and HCT1 5: IC_{50}: 0.8 μg/mL	Ryu et al. (1997)
Tanshinone IIB (**6**)	*Anticancer*: A549: IC_{50}: 5.2 μg/mL; SK-OV-3: IC_{50}: 9.2 μg/mL; K-MEL-2: IC_{50}: 2.9 μg/mL; XF498: IC_{50}: 3.2 μg/mL; and HCT1 5: IC_{50}: 1.7 μg/mL	Ryu et al. (1997)
Hydroxytanshinone IIA (3-βhydroxytanshinone IIA, **7**)	*Anticancer*: A549: IC_{50}: 2.5 μg/mL; SK-OV-3: IC_{50}: 4.1 μg/mL; K-MEL-2: IC_{50}: 7.0 μg/mL; XF498: IC_{50}: 2.7 μg/mL; HCT1 5: IC_{50}: 1.2 μg/mL	Ryu et al. (1997)
15,16-Dihydrotanshinone I (**8**)	*Anticancer*: A549: IC_{50}: 3.0 μg/mL; SK-OV-3: IC_{50}: 1.6 μg/mL; K-MEL-2: IC_{50}: 1.2 μg/mL; XF498: IC_{50}: 0.9 μg/mL; and HCT1 5: IC_{50}: 1.4 μg/mL	Ryu et al. (1997)
Tanshinone I (**9**)	*Anticancer*: A549: IC_{50}: 4.3 μg/mL; SK-OV-3: IC_{50}: 4.1 μg/mL; K-MEL-2: IC_{50}: 2.8 μg/mL; XF498: IC_{50}: 1.6 μg/mL; HCT1 5: IC_{50}: 4.2 μg/mL; and BGC-823: IC_{50}: 3.8 μM	Ryu et al. (1997) and K. M. Kim et al. (2015)
Tanshinol A (**10**)	*Anticancer*: A549: IC_{50}: 1.4 μg/mL; SK-OV-3: IC_{50}: 3.0 μg/mL; K-MEL-2: IC_{50}: 0.7 μg/mL; XF498: IC_{50}: 1.3 μg/mL; and HCT1 5: IC_{50}: 0.8 μg/mL	Ryu et al. (1997)
1,2-Dihydrotanshiquinone I (**11**)	*Anticancer*: A549: IC_{50}: 0.8 μg/mL; SK-OV-3: IC_{50}: 1.5 μg/mL; K-MEL-2: IC_{50}: 3.4 μg/mL; XF498: IC_{50}: 0.6 μg/mL; and HCT1 5: IC_{50}: 0.2 μg/mL	Ryu et al. (1997)
Methylenetanshiquinone (**12**)	*Anticancer*: A549: IC_{50}: 0.4 μg/mL; SK-OV-3: IC_{50}: 0.9 μg/mL; K-MEL-2: IC_{50}: 2.2 μg/mL; XF498: IC_{50}: 0.3 μg/mL; and HCT1 5: IC_{50}: 0.2 μg/mL	Ryu et al. (1997)
3-β-Hydroxymethylene-tanshiquinone (**13**)	*Anticancer*: A549: IC_{50}: 1.5 μg/mL; SK-OV-3: IC_{50}: 3.4 μg/mL; K-MEL-2: IC_{50}: 1.0 μg/mL; XF498: IC_{50}: 1.0 μg/mL; and HCT1 5: IC_{50}: 0.9 μg/mL	Ryu et al. (1997)
Tanshindiol A (**14**)	*Anticancer*: A549: IC_{50}: 0.7 μg/mL; SK-OV-3: IC_{50}: 0.8 μg/mL; K-MEL-2: IC_{50}: 0.2 μg/mL; XF498: IC_{50}: 0.2 μg/mL; HCT1 5: IC_{50}: 0.4 μg/mL	Ryu et al. (1997)
Tanshindiol B (przewaquinone D, **15**)	*Anticancer*: A549: IC_{50}: 0.9 μg/mL; SK-OV-3: IC_{50}: 1.0 μg/mL; K-MEL-2: IC_{50}: 0.4 μg/mL; XF498: IC_{50}: 0.5 μg/mL; and HCT1 5: IC_{50}: 0.7 μg/mL	Ryu et al. (1997)
Tanshindiol C (przewaquinone E, **16**)	*Anticancer*: A549: IC_{50}: 0.7 μg/mL; SK-OV-3: IC_{50}: 0.9 μg/mL; K-MEL-2: IC_{50}: 0.3 μg/mL; XF498: IC_{50}: 0.5 μg/mL; and HCT1 5: IC_{50}: 0.4 μg/mL	Ryu et al. (1997)

(Continues)

TABLE 1 | (Continued)

Name	Activity	References
Methyltanshionate (**17**)	*Anticancer*: A549: IC_{50}: 0.8 μg/mL; SK-OV-3: IC_{50}: 5.0 μg/mL; K-MEL-2: IC_{50}: 6.9 μg/mL; XF498: IC_{50}: 1.5 μg/mL; HCT1 5: IC_{50}: 1.0 μg/mL; HL60: IC_{50}: 16.2 μM; SMMC-7721: IC_{50}: 4.0 μM; MCF7: IC_{50}: 3.4 μM; and SW480: IC_{50}: 6.3 μM	Ryu et al. (1997) and Pan et al. (2012)
Rosmariquinone (miltirone (**32**))	*Anticancer*: A549: IC_{50}: 8.1 μg/mL; SK-OV-3: IC_{50}: 6.6 μg/mL; K-MEL-2: IC_{50}: 3.4 μg/mL; XF498: IC_{50}: 4.7 μg/mL; HCT1 5: IC_{50}: 5.9 μg/mL; and MIAPaCa-2: IC_{50}: 1.9 μM	Ryu et al. (1997)
Dehydromiltirone (**33**)	*Anticancer*: A549: IC_{50}: 4.6 μg/mL; SK-OV-3: IC_{50}: 3.4 μg/mL; K-MEL-2: IC_{50}: 2.3 μg/mL; XF498: IC_{50}: 3.9 μg/mL; and HCT1 5: IC_{50}: 2.8 μg/mL	Ryu et al. (1997)
Multiorthoquinone (**36**)	*Antimicrobial*: *Staphylococcus aureus* (ATCC 6538): MIC: 0.1 μg/mL; *Enterococcus faecalis*: MIC: 2.0 μg/mL; *beta hemolytic Streptococcus*: 0.1 μg/mL; and *Pseudomonas aeruginosa*: 0.5 μg/mL	Ulubelen et al. (1997)
12-Demethylmultiorthoquinone (**37**)	*Antimicrobial*: *Escherichia coli*: 4.6 μg/mL	Ulubelen et al. (1997)

with MICs: of 3.1, 3.1, 6.3, 12.5, and 6.3 μg/mL, respectively (D. S. Lee et al. 1999; N. H. S. Lee et al. 1999).

Rice (*Oryza sativa* L.) is accepted to be one of the most widely used food crops, feeding over 50% of the world's population. Furthermore, rice blast, is the most disasterous disease to destroy rice and is caused by *Magnaporthe oryzae* (J. Liu et al. 2010). Cryptotanshinone (**3**) and tanshinone I (**9**) demonstrated significant antifungal effects toward *M. oryzae* with IC_{50}: 3.4 and 6.1 μg/mL, respectively. On the other hand, 15,16-dihydrotanshinone I (**8**) possessed very promising effects in destroying *M. oryzae* with IC_{50}: 0.91 μg/mL and this compound proved to be three times more potent than the standards, carbendazim, and streptomycin sulfate (IC_{50}: 2.8 μg/mL) (Zhao et al. 2011). In addition, 15,16-dihydrotanshinone I (**8**) displayed good antmicrobial potential toward *Agrobacterium tumefaciens*, *Pseudomonas lachrymans*, *Ralstonia solanacearum*, and *Xanthomonas vesicatoria* with IC_{50}: 4.0, 9.1, 7.7, and 7.5 μg/mL, respectively. Moreover, cryptotanshinone (**3**) illustrated good antimicrobial potential toward *A. tumefaciens*, *Escherichia coli*, *P. lachrymans*, *R. solanacearum*, and *X. vesicatoria* with IC_{50}: 7.2, 7.8, 7.3, 7.1, and 3.6 μg/mL, respectively (Zhao et al. 2011).

Perovskia atriplicifolia Kar. (Lamiacea) is used in salads or employed as a garnish (Pourmortazavi et al. 2003). Phytochemical investigation *P. atriplicifolia* provided (1*R*)-1-acetoxytanshinone IIA (**18**), (1*R*,15*R*)-1-acetoxycryptotanshinone (**19**) and both metabolites illustrated significant butyrylcholinesterase (BChE) inhibition with IC_{50}: 0.84 and 2.7 μg/mL, respectively (Ślusarczyk et al. 2020).

Tanshinone IIA (**1**), cryptotanshinone (**3**), and przewaquinone A (**5**) were reported to possess good antibacterial effects toward *S. aureus*, *B. subtilis* and *Micrococuss flavus* (Mothana et al. 2009). Cryptotanshinone (**5**), tanshinone IIB (**6**), 15,16-dihydrotanshinone I (**8**), tanshinone I (**9**), and methylenetanshinquinone (isolated from *S. miltiorrhiza*, **20**) were found to act as antioxidants (K. Q. Zhang et al. 1990; X. C. Weng and Gordon 1992). Initially, it was proposed that the tanshinones act as antioxidants via lipid radical addition to the *o*-quinone in order to form a stable diradical (X. C. Weng and Gordon 1992). However, it was later reported that tanshinone IIA (**1**) is able to inhibit the lipidperoxidation products association with DNA (Cao et al. 1996). Tanshinone IIA (**1**) is a promising antioxidant toward LDL oxidation and a possible mechanism was proposed involving LDL binding and peroxyl radical scavenging effects (Niu et al. 2000).

Tanshinone IIB (TNB, **6**) exhibited significant potential against *S. aureus* both in in vitro and in vivo models. The TNB (**6**) decreased the expression of Hla significantly at 12 μg/mL concentration. However, it did not affect the transcription of Hla (J. M. Jiang et al. 2022). Additionally, TNB (**6**) potentially decreased the expression of RNAII which increases Hla transcription. It is established that multiple regulators, including MgrA and SaeR, affect Hla transcription. Therefore, it can be proposed that TNB (**6**) act through various pathways. This results in unchanged transcription of Hla. Research has indicated that TNB (**6**) regulates the expression of Hla at post-transcriptional level and decreases its expression. The translation process of Hla in enhanced by RNAIII through its interaction with 5′ UTR of Hla mRNA. The above findings indicate that TNB decreases the expression of RNAIII significantly explaining the inhibition of Hla expression while maintaining the Hla transcription unchanged. Moreover, the Hld expression was enhanced and that of RNAIII was decreased by TNB (**6**) treatment (J. M. Jiang et al. 2022). It is shown that base-pairing between hla mRNA and RNAIII prevents hld gene Shine–Dalgarno sequence. This indicates that an RNAIII molecule which is responsible for Hla translation may not be translated to Hld. This shows that TNB (**6**) blocks interactions between hla mRNA and RNAIII. However, this need to be confirmed in future studies. TNB (**6**) significantly decreased the expression of PSMα1 both at transcription and translation

levels. However, the expression of agrA was not reduced. The psmα expression is under direct control of AgrA which is a transcription factor and part of agr system (J. M. Jiang et al. 2022). The agr system is under positive feedback loop indicating a change in protein expression or mRNA will affect agr system and its own expression. However, present study indicated that TNB (**6**) did not change the expression of agr associated genes such as agrA-D. This shows that TNB can interfere the interactions between psm DNA promoter and AgrA, thereby inhibiting PSMα1 expression. However, additional studies are required that may demonstrate the direct binding between AgrA and TNB (J. M. Jiang et al. 2022).

1β-Hydroxycryptotanshinone (**21**) and 1-oxocryptotanshinone (**22**) were obtained from *Perovskia abrotanoides* and illustrated leishmanicidal effects with IC_{50}: 46.7 and 27.7 μM, respectively. Diterpenes **21** and **22** also demonstrated antiplasmodial effects toward *Plasmodium falciparum* with IC_{50}: 26.9 and 17.6 μM, respectively. Compound **21** exerted cytotoxic effects toward KB-3-1, KB-V1, and lymphocytes with IC_{50}: 5.0, 5.6, and 16.0 μM, respectively (Sairafianpour et al. 2001). 3α-Hydroxytanshinone IIA (**23**) is produced by *Salvia sahendica* and possesses antimicrobial effects toward *Blakeslea trispora* with MIC: 100 μg/mL (Jassbi et al. 2006).

Tanshinones V (**24**) and Vi (**25**) (Figure 2) and neo-przewaquinone A (**26**) were obtained from *S. miltiorrhiza* (Z. Hu and Alfermann 1993; C. Zhang et al. 2013). Neo-przewaquinone A (**26**) illustrated antimicrobial effects toward *Microcystis aeruginosa*, *Scenedesmus obliquus*, and *Chlorella pyrenoidosa* with EC_{50}: 4.6, 10.3, and 14.7 mg/L, respectively (C. Zhang et al. 2013). Przewaquinones B (**27**) and C (**28**) are produced by *Salvia* sp. (Ozarowskia et al. 2017; M. H. Yang et al. 1996). Trijuganone B (**29**) and C (**30**) were isolated from *Salvia trijuga* (X. Lu et al. 1990; X. Z. Lu et al. 1991). 17-Hydroxycryptotanshinone (**31**) was isolated from *Meriandra benghalensis* (Roxb.) Benth. (Lamiaceae) and possesses good antibacterial effects toward *S. aureus*, *B. subtilis* and *M. flavus* (Mothana et al. 2009).

The majority of tanshinones mentioned in this section were isolated from *S. miltiorrhiza* and demonstrated promising antimicrobial and antioxidant effects. *S. miltiorrhiza* is one of the most important traditional Chinese herbs with a long history of use in both medicine and healthy food diets. *S. miltiorrhiza* is also a health giving plant that can be used in its dried form as a tea and has been widely used in the treatment of cardiovascular and cerebrovascular diseases (Shi et al. 2019).

2.2 | Miltirones

Rosmariquinone (miltirone, **32**) is produced by *S. miltiorrhiza* (L. Zhou et al. 2016) and dehydromiltirone (**33**) (Figure 3) was reported from *Salvia lanigera* (Aboul-Ela et al. 2002). Compound **32** possessed antioxidant potential comparable to that of the commonly used phenolics viz., BHT and BHA (Houlihan et al. 1985). On the other hand, *Rosmarinus officinalis* is a special spice available in markets as an antioxidant in the USA and Europe (Nieto et al. 2018). Moreover *R. officinalis* is used as a natural antioxidant, displaying better antioxidant activity than other antioxidants commonly used in food preservation (Etter 2004). In addition, both compounds illustrated cytotoxic effects toward various cancer cells (Table 1).

1-Oxomiltirone (**34**) is isolated obtained from *P. abrotanoides* and illustrated leishmanicidal and antiplasmodial effects (toward *P. falciparum*) with IC_{50}: 17.9 and 13.0 μM, respectively. Compound **34** also exerted cytotoxic effects toward KB-3-1, KB-V1, and lymphocytes with IC_{50}: 46.6, 47.8, and 44.6 μM, respectively (Sairafianpour et al. 2001). 1(R)-Hydroxymiltirone (**35**) is produced by *Salvia argentea* (Michavila et al. 1986). Multiorthoquinone (**36**) and 12-demethylmultiorthoquinone (**37**) were isolated from the *Salvia multicaulis.* It was found that diterpene **36** possessed antimicrobial effects toward *S. aureus*, *Enterococcus faecalis*, *beta hemolytic Streptococcus*, and *Pseudomonas aeruginosa* whereas compound **37** was active against *E. coli* (Ulubelen et al. 1997).

2.3 | Abietane Diterpenoids

Rosmaquinone (**38**), rosmaquinone A (**39**), and rosmaquinone B (**40**) (Figure 3) were isolated from *R. officinalis* L. (Mahmoud et al. 2005). Compounds **38** possessed antimicrobial effects toward *S. aureus*, *S. epidermidis*, *B. subtilis*, and *Bacillus cereus* with MIC: 8–40 μg/mL (Table 2). On the other hand, diterpene **40** was active toward *S. aureus*, *B. subtilis*, and *B. cereus* with MIC: 20–40 μg/mL (Marrero et al. 2009) (Table 2). Sessein (**41**) was reported from *Salvia sessei* and possessed antimicrobial effects toward *E. faecalis*, *E. coli*, and *Staphylococcus haemolyticus* with an MIC: 12.5 μg/mL (Gomez-Rivera et al. 2018).

Hypargenin C (**42**) (Figure 3) is isolated from *Salvia hypargeia* (Dlubelen et al. 1988). On the other hand, salviphlomone (**43**) (Hueso-Rodriguez et al. 1983), 6α-hydroxy-11,12-dioxo-abieta-8,13-diene (**44**), and deoxysalviphlomone (**45**) (Nagy et al. 1999) were isolated from *Salvia phlomoides*, *Salvia recognita*, and *Salvia nutans* (N. Tan et al. 1998). The abietane diterpene, 6α-hydroxy-11,12-dioxo-8,13-abieta-diene (**46**) was obtained from *S. recognita*. Another orthoquinone, 7α,14-dihy-droxy-8,13-abieta-diene-11,12-dione (**46**) was isolated from *Salvia viridis* roots and showed antibacterial effects toward *E. faecalis* with MIC: 50 μM (Rungsimakan and Rowan 2014).

Abietane diterpenoids, known a complanatins A–D (**47–50**) along with xanthopel (**51**) are produced by *Lycopodium complanatum* L. (Lycopodiaceae). Compounds **49** and **50** illustrated cytotoxcity toward MSTO-211H, NCI-H2052, and NCI-H226 with IC_{50} values ranging from 17.3 to 40.4 μM (Y. Weng et al. 2018).

Forskalinone (**51a**) and cleon V (**51b**) were isolated from *Salvia forskahlei* (Ulubelen et al. 1996) and *Plectranthus myrianthus* (Lamiaceae) (Yoshizaki et al. 1979). Forskalinone (**51a**) possessed moderate antimicrobial effects toward *S. epidermidis* and slight activity against *E. faecalis* with MIC of 670 and 168 μg/mL, respectively (Ulubelen et al. 1996). The abietane diterpene sugikurojin F (**52**) was obtained from *Cryptomeria japonica* D. Don (Taxodiaceae) (Yoshikawa et al. 2006). In another study, the 7,20-epoxyabietane diterpenoid, przewalskin G (**53**) was obtained from *Salvia przewalskii* (G. Xu et al. 2009; W. Xu et al. 2009).

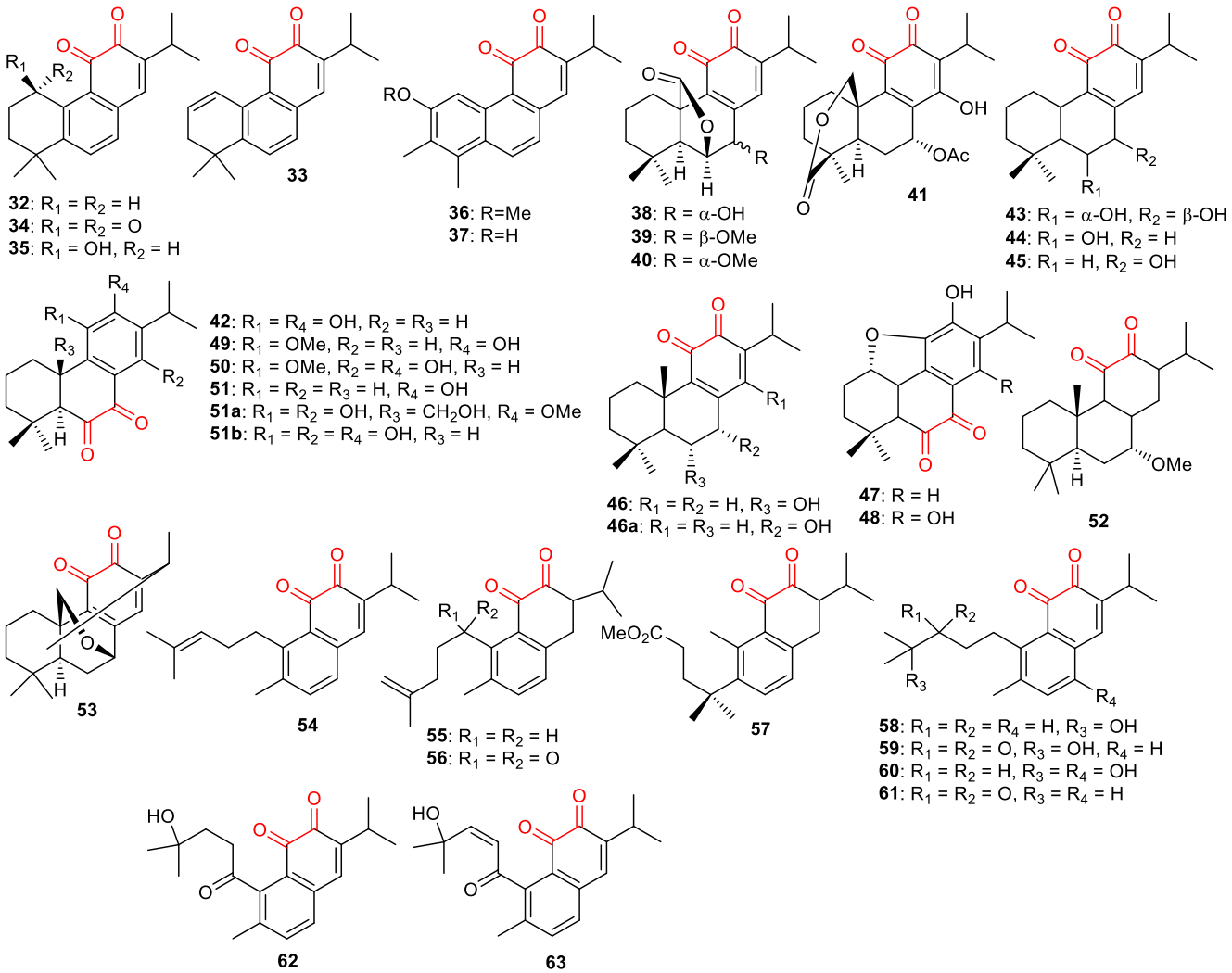

FIGURE 3 | Structures of diterpenes **32–63**.

2.3.1 | *Seco*-Abietane Diterpenoids

Saprorthoquinone (**54**) (Figure 4) was isolated from *Salvia lanigera* and possessed cytotoxic effects toward BC1, LU1, COL2, KB, KB-VI, and P388 cell with ED_{50}: 9.2, 16.4, 3.3, 9.1, and 2.3 μg/mL, respectively (Ulubelen et al. 1999). Aethiopinone (**55**) and 1-oxoaethiopinone (**56**) were reported from *Salvia sclarea* (Kuzma et al. 2007). Diterpene **55** possessed antimicrobial effects toward *S. aureus* (ATCC 29213), *S. epidermidis* (ATCC 12228), *E. faecalis* (ATCC 29212), *S. aureus* (A3), *S. aureus* (1474/01), *S. epidermidis* (ATCC 12228), *S. epidermidis* (RP12), and *S. epidermidis* (6756/99) with MIC: ranging from 9.3 to 75 μg/mL (Kuzma et al. 2007; Walencka et al. 2007) (Table 2). On the other hand, 1-oxoaethiopinone (**56**) demonstrated antimicrobial effecta toward *S. aureus* (ATCC 29213) and *S. epidermidis* (ATCC 12228) with MIC: 150 and 75 μg/mL, respectively (Kuzma et al. 2007). In another study, aethiopinone (**55**) was shown to possess potent antimicrobial effects toward *Bordetella bronchiseptica*, *M. luteus*, *S. aureus*, *S. epidermidis*, and *B. subtilis* (Hernández-Pérez1 et al. 1999).

Salvigerone (**57**) was isolated from *S. lanigera* (I. S. Lee et al. 1998) whereas 4-hydroxysaprorthoquinone (**58**) and 3-keto-4-hydroxysaprorthoquinone (**59**) were obtained from *Salvia prionitis*. Compound **58** illustrated potent inhibition toward topoisomerase with IC_{50}: 0.8μM. On the other hand, compound **59** demonstrated cytotoxic effects toward HL-60 SGC-7901 and MKN-28 with IC_{50}: 4.6, 0.2, and 0.3 μM, respectively (X. Chen et al. 2002). 4,14-Dihydroxysaprorthoquinone (**60**) was isolated from *Salvia eriophora* and showed significant cardiovascular effects (Ulubelen et al. 2002). Salvisyrianone (**61**) was obtained from *Salvia syriaca* (Ulubelen et al. 2000). Prionoids D (**62**) and E (**63**) were obtained from *Salvia prionitis* and diterpene **62** exhibited significant cytotoxic effects toward P-388 cells with IC_{50}: 0.41 μM. On the other hand, prionoid E (**63**) exhibited potent effects toward A-549 cells with IC_{50}: 0.72 μM (J. Chang et al. 2005).

2.3.2 | Nor- and *Abeo*-Abietane Diterpenoids

Saligerone (**64**) (Figure 4), a norditerpenoid missing a C-20 methyl group, was isolated from *S. lanigera* and revealed antimicrobial effects toward *S. aureus*, *P. aeruginosa,* and *Candida albicans* with MIC: 13.0, 13.0, and 52.0 μg/mL, respectively (El-Lakany 2003). *Abeo*-abietane diterpenes **65** and **66** were obtained from *Taxus cuspidata* (Bai et al. 2005). Diterpenes **65** and **66** possess the rearranged 9(10 → 20)-*abeo*-abietane diterpenoid skeleton.

TABLE 2 | Biological effects of abietane diterpenes.

Compound	Activities	References
Rosmaquinone (**38**)	*Antimicrobial*: *Staphylococcus aureus*: MIC: 20 μg/mL; *Staphylococcus epidermidis* (ATCC 12228): MIC: 40 μg/mL; *Bacillus subtilis*: MIC: 8 μg/mL; *Bacillus cereus*: MIC: 10–20 μg/mL; *Bacillus albicans*: MIC: > 40 μg/mL; and *Anticancer*: A549: 28.8 μM; MCF7: 39.5 μM	Marrero et al. (2009)
Rosmaquinone B (**40**)	*Antimicrobial*: *S. aureus*: MIC: 40 μg/mL; *S. epidermidis* (ATCC 12228): MIC: > 40 μg/mL; *B. subtilis*: MIC: 20–40 μg/mL; *B. cereus*: MIC: 20–40 μg/mL; *B. albicans*: MIC: > 40 μg/mL; *Anticancer*: HeLa: 33.8 μM; Hep-2: 42.5 μM: A549: 25.4 μM; and MCF7: 22.1 μM	Marrero et al. (2009)
Sessein (**41**)	*Antimicrobial*: *Enterococcus faecalis:* MIC: 12.5 μg/mL; *Escherichia coli*: 12.5 μg/mL; and *Staphylococcus haemolyticus:* 12.5 μg/mL	Gomez-Rivera et al. (2018)
Aethiopinone (**55**)	*Antimicrobial: S. aureus* (ATCC 29213): MIC: 75 μg/mL; *S. epidermidis* (ATCC 12228): MIC: 37 μg/mL; and *E. faecalis* (ATCC 29212): MIC: 75 μg/mL	Kuzma et al. (2007)
	Antimicrobial: *S. aureus* (ATCC 29213): MIC: 75 μg/mL; *S. aureus* (A3): MIC: 37.5 μg/mL; *S. aureus* (1474/01): MIC: 37.5 μg/mL; *S. epidermidis* (ATCC 12228): MIC: 37.5 μg/mL; *S. epidermidis* (RP12): MIC: 18.7 μg/mL; and *S. epidermidis* (6756/99): MIC: 9.3 μg/mL	Walencka et al. (2007)
1-Oxoaethiopinone (**56**)	*Antimicrobial*: *S. aureus* (ATCC 29213): MIC: 150 μg/mL; *S. epidermidis* (ATCC 12228): MIC: 75 μg/mL; and *E. faecalis* (ATCC 29212): MIC: > 150 μg/mL	Kuzma et al. (2007)

2.4 | Icetexane Diterpenoids

Ortho-quinone-type icetexane diterpenoids, perovskatone **B** (**67**), and przewalskin E (**68**) (Figure 4) were obtained from *P. atriplicifolia*. Compound **67** illustrated moderate anti-HBV effects (Z. Y. Jiang et al. 2015). In another phytochemical investigation, romulogarzone (**69**) was isolated from *Salva ballotaeflora* (Dominguez et al. 1976). Salvatrione (**70**) is a combination of the icetexane diterpene and monoterpene and is isolated from *Salvia bucharica* (Ahmad et al. 2000).

2.5 | Taxane Diterpenes

Wallitaxanes C (**71**) and E (**72**) (Figure 4) were obtained from *Taxus wallichiana* Zucc. (Taxaceae) and it was found that compound **72** featured the rare *abeo*-taxoid skeleton, Diterpenes **71** and **72** demonstrated α-alpha glucosidase inhibition with IC_{50}: 142 and 83.6 μM, respectively. On the other hand, wallitaxane E (**72**) also possessed cytotoxicity toward HeLa Cells with IC_{50}: 4.9 μM (Dang et al. 2017). Taiwantaxin A (**73**) is produced by *Taxus sumatrana* (Miq.) de Laub. and oxygenated functionalities at either C-13 or C-14 are absent in this molecule. Moreover, this diterpene was not active toward PC carcinoma cells (S. S. Wang et al. 2009).

2.6 | Miscellaneous Diterpenoids

Stypoldione (**74**) (Figure 4) is a pentacyclic marine diterpenoid which features an uncommon spiro-*o*-benzoquinonefuran core and was reported as isolated from the alga *Stypopodium zonale* in 1979 (Gerwick et al. 1979; Gerwick and Fenical 1981; Penicooke et al. 2013). Notably, this compound is reported to inhibit a wide range of biochemical and biological processes (Gerwick et al. 1979; Gerwick and Fenical 1981; Penicooke et al. 2013; White and Jacobs 1983; O'Brien et al. 1983; O'Brien et al. 1984; O'Brien et al. 1986, 1989; Jacobs et al. 1985; O'Brien, White, et al. 1984). Stypoldione (**74**) was initially identified for fish toxicity (Gerwick et al. 1979; Gerwick and Fenical 1981) and later this compound was shown to exhibit division of Ehrlich tumor cells, mammalian cells, and P388 leukemia (White and Jacobs 1983; O'Brien et al. 1984, 1986). Elisabatin A (**75**), an amphilectane diterpene, was reported from *Pseudopterogorgia elisabethae* and possessed weak cytotoxicity toward various cancer cells but was not active as an anti-HIV agent. In addition, this molecule revealed 21% inhibition toward *Mycobacterium tuberculosis* (A. D. Rodríguez et al. 1999).

Oryzadione (**76**) (Figure 4), a kaurane diterpene, is produced by the rice plant and possessed antibacterial effects with 40% colony formation of *Xanthomonas campestris* (Kono et al. 2004). The clerodane diterpene named clerodane-6,7-dione (**77**) was obtained from *Teucrium polium* (Marquez and Valverde 1979). The diterpene with an undescribed carbon skeleton named aberrarone (**78**) was obtained from *P. elisabethae*. Aberrarone (**78**) possessed antimalarial effects toward *P. falciparum* with IC_{50}: 10 μg/mL (Rodrıguez et al. 2009). Steglich isolated crinipellin (**79**) from the basidiomycete *Crinippellis stipitaria* (Anke et al. 1986) and it was shown to possess antimicrobial effects toward *Proteus vulgaris*, *Arthrobacter citreus*, *Bacillus brevis*, *B. subtilis*, *Corynebacterium insidiosum*, *Micrococcus roseus*, *Sarcina lutea*, *S. aureus*, and *Saccharomyces cerevisiae* (Kupka et al. 1979).

Mitrephorone A (**80**), an *ent*-trachylobane diterpenoid comprising an oxetane core, was produced by *Mitrephora glabra*. This compound illustrated cytotoxic effects toward KB, MCF-7, H460, and SF-268 cancer cells with IC_{50} values of 8.0, 15.7, 23.3, and 30.9, respectively. In addition, this diterpene orthoquinone possessed moderate antimicrobial effects toward *M. luteus*, *Aspergillus niger*,

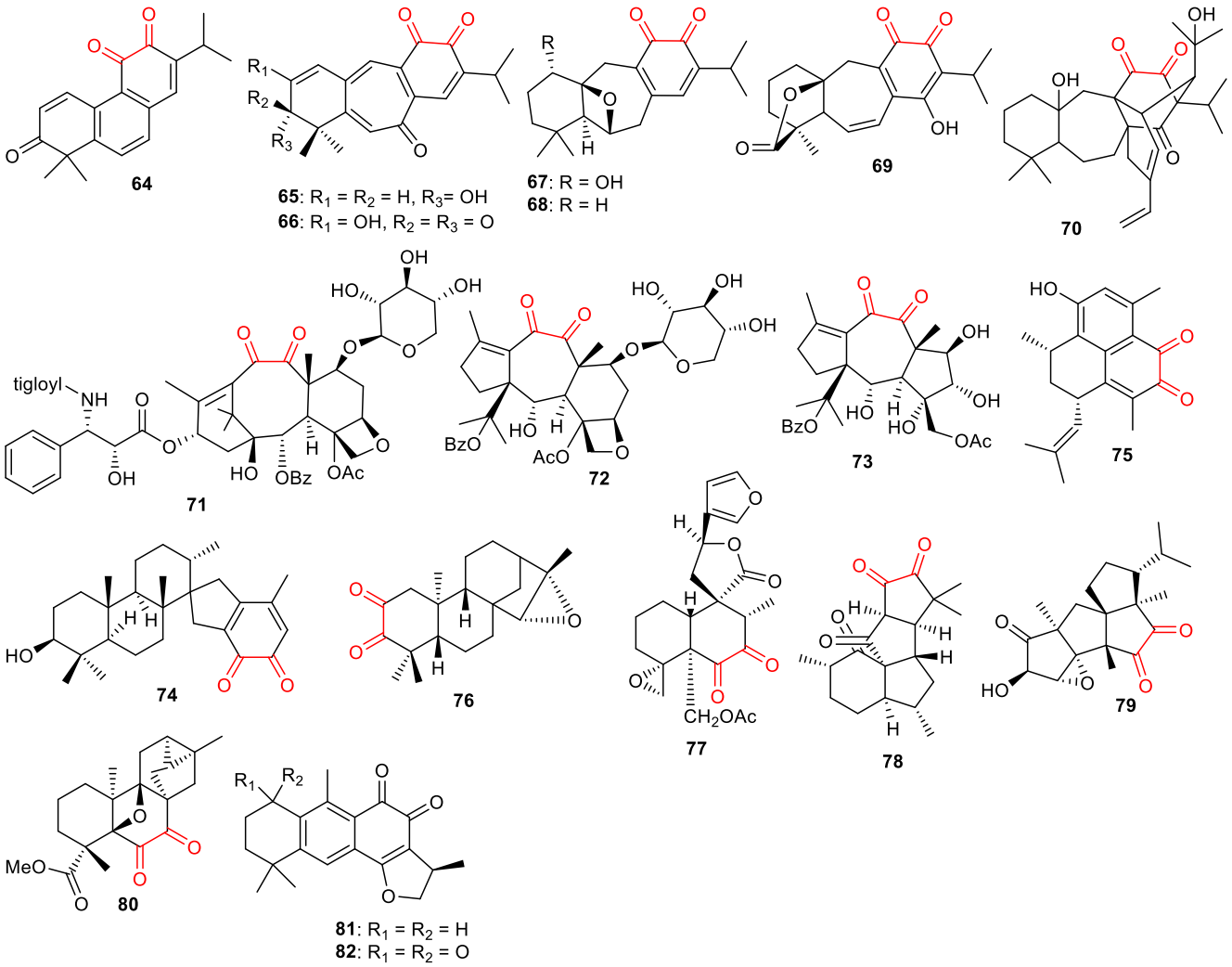

FIGURE 4 | Structures of diterpenoids **64–82**.

S. cerevisiae, and *Mycobacterium smegmatis*. Aegyptinone A (**81**) is produced by *Salvia tebesana* (Eghbaliferiz et al. 2018) whereas (15*R*)-1-oxoaegyptinone A (**82**) was obtained from *P. atriplicifolia* and displayed inhibition toward BChE with an IC_{50} of 15.7 μg/mL (Slusarczyk et al. 2020). Orthoquinone **81** displayed cytotoxic effects toward MCF-7, B16F10, PC-3, and C26 with IC_{50}: values of 8.1, 8.1, 2.1, and 6.9 μM, respectively (Eghbaliferiz et al. 2018).

3 | Unsymmetrical Benzils

2-Hydroxybenzil (**83**) (Figure 5) is the core skeleton of unsymmetrical benzils obtained from the Leguminosae family (Worayuthakarn et al. 2014). Licoagrodione (**84**), is produced by *Glycyrrhiza glabra* L. (Leguminosae) and exhibited antimicrobial effects towards *S. aureus*, *B. subtilis*, *E. coli*, *P. aeruginosa*, *Kliebsiella pneumoniae*, *C. albicans*, *A. niger*, and *Trichophyton mentagrophytes* (W. Li et al. 1998). Scandione (**85**) was obtained from *Derris scandens* (Mahabusarakam et al. 2004) whereas calophione A (**86**) was isolated from *Tephrosia calophylla* (Ganapaty et al. 2009). The later molecule possessed significant cytotoxicity toward RAW and HT-29 cells with IC_{50}: 5.00 and 2.9 μM, respectively (Ganapaty et al. 2009). Furthermore, tenuifodione (**87**) was obtained from *Iris tenuifolia* Pallas (Iridaceae) and saxoguattine (**88**) (Hocquemiller et al. 1984) along with crypto-pleurospermine (**89**), are extensively reported from the Lauraceae, Annonaceae, and Papaveraceae families (Worayuthakarn et al. 2014).

4 | Terphenylquinones

Sarcodon leucopus produced sarcoviolin α (**90**) (Figure 6) and its isomer episarcoviolin α (**91**). A mixture of metabolites **90** and **91** illustrated cytotoxic effects toward SF-268 (CNS), MCF7 (Breast), and NCI-H460 (Lung) cell lines. Moreover, this mixture showed anti-HIV effects with EC_{50}: 5 μg/mL (Calì et al. 2004). Another orthoquinone, hydnellin B (**92**) was reported from *Hydnellum geogerirum* and *Hydnellum suaveolens* (Hashimoto et al. 2006). Singh and coworkers reported on the *p*-terphenyl based *o*-quinone **93** isolated from *Phoma* sp. and was shown to inhibit GMP-dependent protein kinase with IC_{50}: 0.9 μM (C. Zhang et al. 2006). Seven year later Nakazaki et al. (2013) revised structure **93** into **94** based on both synthesis and NMR data.

Spiromentins A–D (**95–98**) were isolated from the fungus *Paxillus atrotomentosus* (Buchanan et al. 1995; J. K. Liu 2006). The

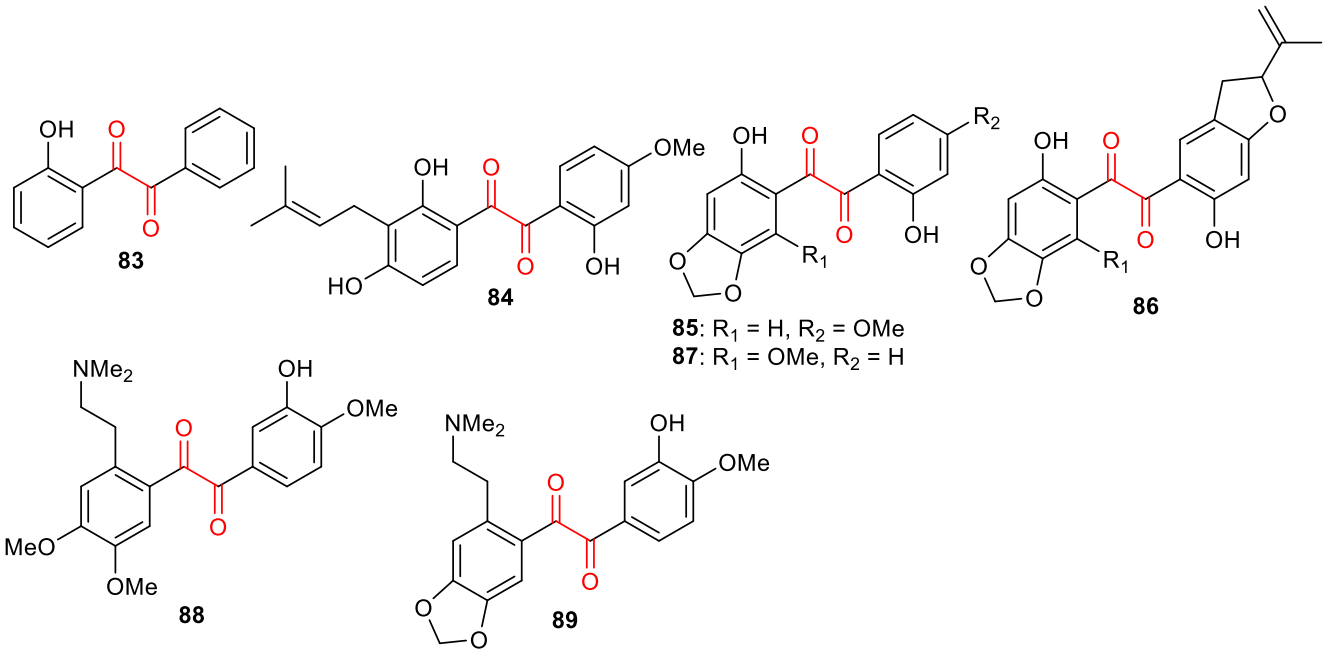

FIGURE 5 | Structures of unsymmetrical benzils **83–89**.

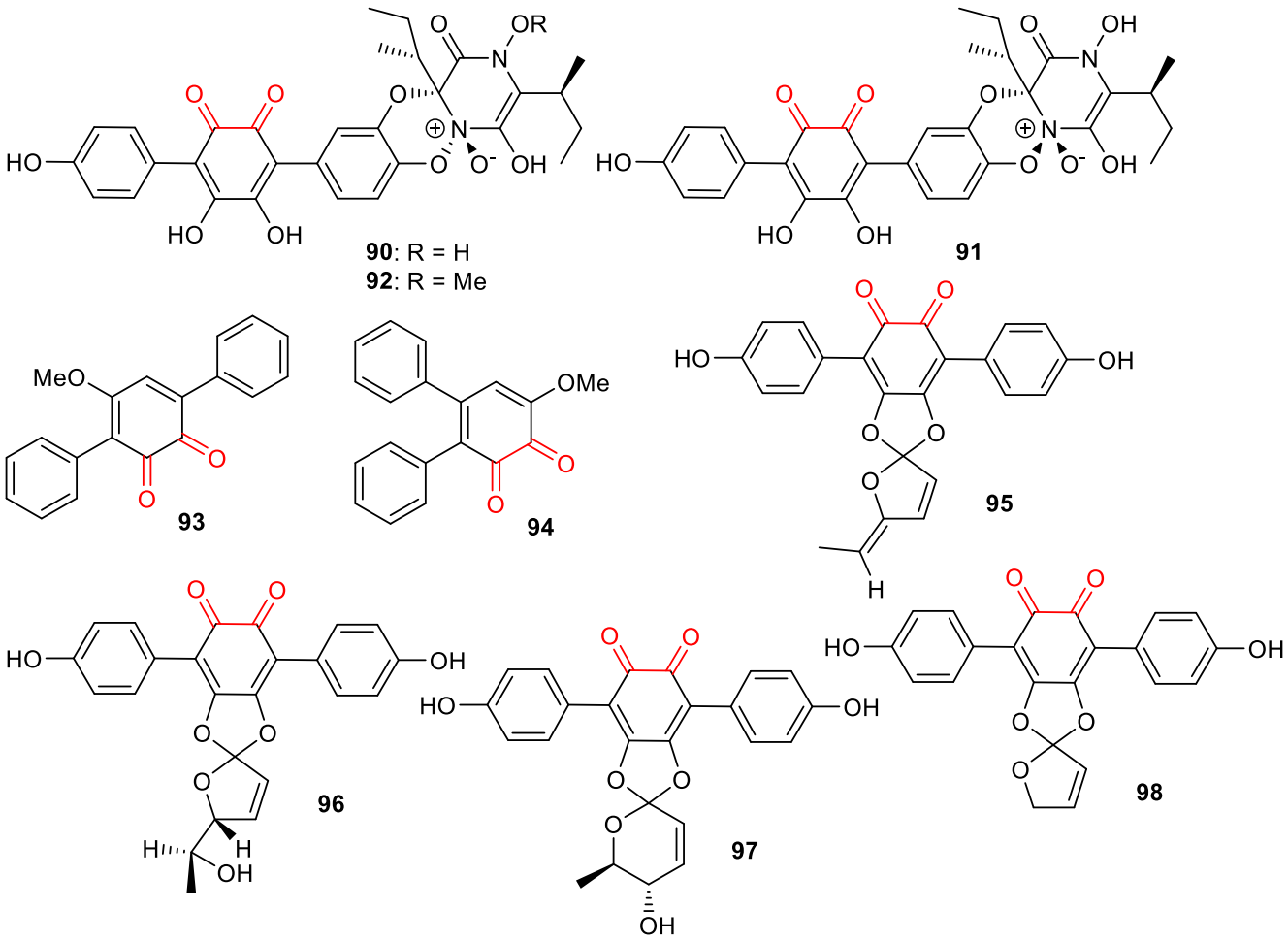

FIGURE 6 | Structures of unsymmetrical benzils **90–98**.

spiromentins C (**97**) and B (**98**) were also reported from *Tapinella atrotomentosa* (Béni et al. 2018). These compounds possess an unusual spiro core in which a 4,5-dihydroxy-1,2-benzoquinone is attached to a lactone acetal skeleton. The spiromentins C (**97**) and B (**98**), illustrated significant antioxidant effects with 16.21 and 11.23 mmol TE/g, respectively, and these compounds were more potent than ascorbic acid (6.97mmol TE/g) (Béni et al. 2018). In addition, spiromentin C (**97**) displayed significant antimicrobial effects against *A. baumannii* and *E. coli* (Béni et al. 2018).

5 | *Ortho*-Naphthoquinones

5.1 | β-Lapachone

β-Lapachone (**99**) (Figure 7) was reported from *Handroanthus impetiginosus* (Mart. Ex DC.) Matto (Bignoniaceae) and has been evaluated for phase II clinical trials to treat pancreatic cancer. Moreover, compound **99** possessed in vivo anticancer, anti-inflammatory, antiobesity, antioxidant, neuroprotective, nephroprotective, and wound healing properties (Gomes et al. 2021).

5.1.1 | Antimicrobial Effects

β-Lapachone (**99**) exhibited significant antimicrobial activity against various strains of *S. aureus*. The MIC vaules were from 8.0 to 32 μg/mL against these strains. β-lapachone in combination with ciprofloxacin als produced additive effect toward LFBM 33 strain. Moreover, β-lapachone produced antimicrobial effect against MRSA strains in combination with fluoroquinolones, beta-lactams, and carbapenems (Macedo et al. 2013). These findings are in line with those reported by Wink and coauthors (Wink et al. 2012) and Y. S. Lee et al. (2010). These authors reported that when naphthoquinones from plants were combined with β lactams (oxacillin and ampicillin) and tested against MRSA strains, synergistic effects were observed. In another research study, it was reported that when β-lapachone (**99**) was combined with isoniazide and tested against *M. smegmatis* and *Mycobacterium fortuitum*, synergistic effect was observed. The effect produced was bactericidal effect which resulted in sterilization of mycobacterial cultures within 96 h (J. L. Silva et al. 2009).

β-Lapachone (**99**) exbibited potentail antibiofilm activity against *S. aureus* and decreased biofilm production (Fernandes et al. 2020). Moreover, β-lapachone produced significant antifungal activity against *Histoplasma capsulatum* and *Coccidioides posadasii* with MIC of 0.12 and 0.25 μg/mL, respectively. The β-lapachone (**99**) effect on cell membrane could be due to its hydrophobic nature, that may affect metabolic and others functions within cells. This fact was shown in current study that showed that β-lapachone alters the permeability of plasma membrane of *H. capsulatum* and *C. posadasii*, exhibiting that its effectiveness is associated with degeneration of the fungal plasma membrane (Brilhante et al. 2016). Moreover, β-lapachone (**99**) inhibits ergosterol synthesis, decreasing the quanity of ergosterol that is extracted from fungal cells post β-lapachone (**99**) treatment. Additionally, β-lapachone also induces oxidative stress within fungal cells as indicated by the formation of reactive oxygen species, such as the hydroxyl radical (OH), superoxide anion radical ($O_2^{\cdot -}$), and hydrogen peroxide (H_2O_2), resulting in apoptotic and cytotoxic effects (Brilhante et al. 2016).

β-lapachone (**99**) exhibited potential antibacterial activity against *S. aureus*, *S. epidermidis*, *P. aeruginosa,* and *S. saprophyticus* with MIC in the range of 0.04–0.20 mM. The effect of β-lapachone on biofilm formation of these four strains was examined in this study. The results showed that except *P. aeruginosa*, β-lapachone (**99**) significantly inhibited biofilm inhibition at MBIC (50%–80%) in these strains (Mir et al. 2023). Among *S. aureus*, *S. epidermidis,* and *S. saprophyticusm*, β-lapachone (**99**) inhibited biofilm formation of *S. epidermidis* and

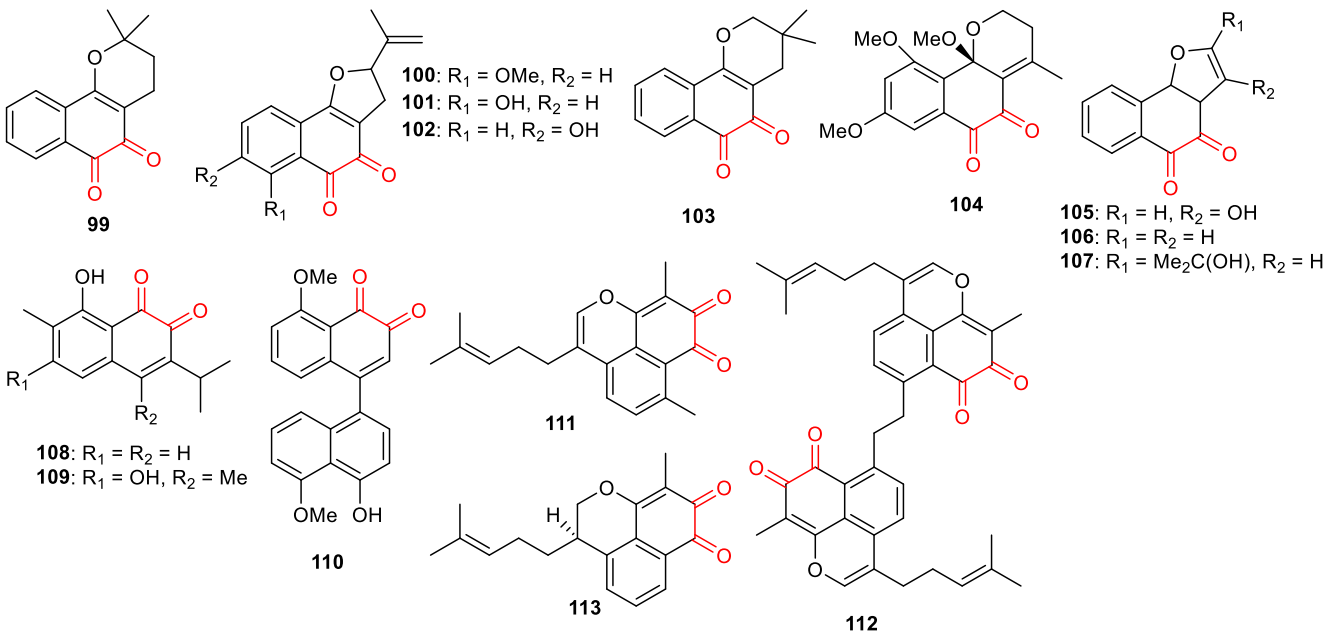

FIGURE 7 | Structure of *ortho*-saphthoquinones **99–113**.

S. aureus very strongly. This β-lapachone-mediated inhibition was 78% for *S. epidermidis* and 75% for *S. aureus* at their respective MBIC. Moreover, β-lapachone (**99**) did not inhibit the growth in these strains at MBIC, indicating that β-lapachone targets factors or processes which are required for biofilm formation of these strains without having impact on biological processes needed for normal growth of bacteria. The β-lapachone (**99**) not only inhibited the biofilm formation, but it also distrupted the mature biofilms of these strains. It was noted that 48-h mature biofilms of *S. epidermidis* were very sensitive to the efefcts of β-lapachone since biofilms were prevented 11.5 times by β-lapachone as compared to control (Mir et al. 2023).

β-Lapachone (**99**) exhibited significant antifungal and antibacterial effects (Table 3). It was noted that all Gram-positive strains but only two Gram-negative strains, including *Brucella melitensis* and *P. aeruginosa,* were sensitive to the effects of β-lapachone treatment. β-lapachone produed very significant effect against *S. aureus* and *Bacillus subthis* among Gram-positive strains whereas only *Brucella* species was sensitive among Gram-negative strains. The Fungal strains were more sensitive as compared to bacterial strains. It was found that β-lapachone was more effective as compared to standard antifungal drug, ketoconazol, particularly against *Candida tropicalis*, *C. albicans*, *T. mentagrophytes,* and *Torulopsis glabrata* mentagrophytes (Table 3).

The liposomes loaded with β-Lapachone (**99**) exhibited significant antimicrobial activity against *Cryptococcus neoformans* and *S. aureus* with MICs in the range of 2.0–8.0 μg/mL (Cavalcanti et al. 2015). Moreover, orthoquinone 102 significantly inhibited *S. epidermidis* and *Staphylococcus haemolyticus* with MIC value of 8.0 μg/mL (Pereira et al. 2006). The β-lapachone (**99**) reduced biofilm formation up-to 92%, *C. albicans* cell wall mannoprotein up-to 28.5% and yeast-to-hyphae transition up-to 92% (Moraes et al. 2018). In other research study, it was shown that β-lapachone (**99**) exhibited significant antifungal activity against *A. niger* and *C. albicans* with 16 mm and 18 zone of inhibition, respectively. The antifungal activity of Flucanzole (standard antifungal drug) was 18 and 19 mm against *A. niger* and *C. albicans*, respectively (Karthikeyan et al. 2016). The β-lapachone (**99**) also produced antibacterial activity against *Streptococcus agalactiae*, *S. dysgalactiae,* and *Streptococcus uberis* with MIC values of 64, 32, and 32 μM, respectively. It is indicated that β-lapachone (**99**) produces its bactericidal activity against *S. uberis* through ROS-dependent mechanism.

TABLE 3 | Antimicrobial effects of β-lapachone (**99**).

Microorganisms	MIC/MFC (μg/mL)
Fungi	
Candida albicans	MIC: 0.03
C. parapsilosis	MIC: 0.03
C. tropicalis	MIC: 0.03
Cryptococcus neoformans	MIC: 0.02
Epidermophyton floccosum	MFC: 4
Microsporum gypseum	MFC: 16
Torulopsis glabrata	MIC: 0.03
Trichophyton interdigitatum	MFC: 4
Trichophyton mentagrophytes	MFC: 8
T. rubrum	MFC: 1
Bacteria	
Bacillus subtilis	MIC: 2
Brucella melitensis	MIC: 0.25
Pseudomonas aeruginosa	MIC: 32
Staphylococcus aureus ATCC 9144	MIC: 2
S. aureus ATCC6538p	MIC: 8
Streptococcus faecalis ATCC 10541	MIC: 8

Abbreviations: MFC: minimal fungicidal; MIC: minimal inhibitory concentrations.

5.2 | Miscellaneous *Ortho*-Naphthoquinones

Lantalucratins A–C (**100–102**) (Figure 7), isopropenylfurano-*ortho*-naphthoquinones, were obtained from *Lantana involucrata* L. (Verbenaceae). Their structures were determined on the basis of NMR and X-ray crystallographic analyses. Compounds **100** and **101** showed cytotoxic activities against various human tumor cell lines, including drug resistant variants, with IC_{50} values of 1.0–4.9 μM (Table 4).

Rhinacanthone (**103**) was obtained from *Rhinacanthus nasutus* (L.) Kurz (Acanthaceae) and illustrated cytotxicty toward HeLa cells with IC_{50}: 1.2 μM along with in vivo effects toward Dalton's lymphoma in mice (Siripong et al. 2009). Additionally rhinacanthone (**103**) was active toward KB, HepG2, and Vero cells with IC_{50}: 9.6, 6.9, and 7.5 μM, respectively (Kongkathip et al. 2003). Colelomycerones A (**104**) is produced by the fungus YMF 1.01029 and possessed good antimicrobial effects toward *Bipolaris maydis*, *Cochliobolus sativus*, *Fusarium verticillioides*, *B. subtilis*, *Brevibacillus laterosporus*, and *S. aureus* (Dong et al. 2009).

Orthoquinones **105–107** (Figure 7) were isolated from *Avicennia alba* L. (Verbenaceae) and their structures determined via extensive spectroscopic techniques (Ito et al. 2000). The Egyptian *S. lanigera* produce a reddish orthoquinone lanigerone (**108**) (I. S. Lee et al. 1998) whereas berryammone A (**109**) was obtained from *Berrya ammonilla* Roxb. (Malvaceae) (Chou et al. 2012). Berryammone A (**109**) possessed potent inhibitory effects on superoxide anion generation by human neutrophils (IC_{50}: 3.2 μM) as well as elastase release by human neutrophils (IC_{50}: 3.7 μM) (Chou et al. 2012). Daldiquinone (**110**), 1,2-naphthoquinone analog was isolated from *Daldinia concentrica* and illustrated antiangiogenesis effects, **110** significantly inhibited HUVEC with IC_{50}: 7.5 μM (Kamauchi et al. 2018).

Biflorin (**111**) (Figure 7) was reported from *Capraria biflora* L. (Scrophulariaceae) in 1954 and demonstrated antimicrobial effects toward *C. albicans*, *Trichosporon margaritipherum* along with other fungi and bacteria. Moreover, this molecule illustrated in vivo (Ehrlich carcinoma, melanoma, and sarcoma 180) and in vitro anticancer effects and one clinical report published a patient with an erythematous perioral lesion (Wisintainer et al. 2014). Furthermore, biflorin (**111**) significantly inhibits B16, MCF-7, HCT-8, HL-60, and CEM cancer cells with IC_{50}: 0.4, 0.4, 0.8, 1.9, and 1.0 μg/mL, respectively (Vasconcellos

TABLE 4 | Biological effects of orthoquinones.

Name	Activity	References
Lantalucratin A (**100**)	*Anticancer*: KB: > 5 μM; KB-VIN: > 5 μM; KB-7d: > 5 μM; KB-CPT: 2.1 μM; A549: 3.3 μM; 1A9: 3.9 μM; 1°9-PTX10: > 5 μM; MCF-7: 1.6 μM; HOS: 4.8 μM; and U87-MG: 4.7 μM	Hayashi et al. (2004)
Lantalucratin B (**101**)	*Anticancer*: KB: 2.3 μM; KB-VIN: 2.4 μM; KB-7d: 2.1 μM; KB-CPT: 1.0 μM; A549: 1.8 μM; 1A9: 1.5 μM; 1°9-PTX10: 2.1 μM; MCF-7: 1.0 μM; HOS: 2.5 μM; and U87-MG: 1.9 μM	Hayashi et al. (2004)
Lantalucratin C (**102**)	*Anticancer*: KB: 6.9 μM; KB-VIN: 6.3 μM; KB-7d: 5.8 μM; KB-CPT: 5.2 μM; A549: 6.0 μM; 1A9: 9.4 μM; 1A9-PTX10: 6.5 μM; MCF-7: 4.9 μM; HOS: 4.7 μM; and U87-MG: 7.3 μM	Hayashi et al. (2004)
Mansonone C (**115**)	*Antimicrobial*: *Phytophthora parasitica*: 84.2%; *Xanthomonas oryzae pv. oryzicola*: ZI: 13.7; and *X. oryzae pv. oryzae*: 12.7	Mongkol and Chavasiri (2016)
Mansonone E (**117**)	*Antimicrobial*: *P. parasitica*: 94.4%; *X. oryzae pv. oryzicola*: ZI: 24.3; *X. oryzae pv. oryzae*: 25.7; *Alternaria porri*: MIC: 250 μg mL^{-1}; *Colletotrichum gloeosporioides*: MIC: 31.2; μg mL^{-1}; *Fusarium oxysporum*: MIC: 500 μg mL^{-1}; and MIC: *P. parasitica*: 31.2 μg mL^{-1}	Mongkol and Chavasiri (2016)
Mansonone F (**118**)	*Antimicrobial*: *Bacillus subtilis* ATCC 6633: MIC (μg/mL): 2; *Bacillus cereus* ATCC 6633: MIC: 4; *Staphylococcus aureus* ATCC 25923: MIC: 1; *S. aureus* ATCC 6538P: MIC: 4; *S. aureus* ATCC 29213: MIC: 4; *S. aureus* TMS 64: MIC: 8; *S. aureus* TMS 33: MIC: 8; *S. aureus* TMS 417: MIC: 4 μg/mL; *S. aureus* Smith: MIC: 4; *Staphylococcus epidermidis*: MIC: 0.5; *Klebsiella pneumoniae*: MIC: 8; *Micrococcus luteus* ATCC 9341: MIC: 1; and *M. luteus* ATCC 10240: MIC: 1	Suh et al. (2006)
Mansonone F (**118**)	*Antimicrobial*: *S. aureus* SG 511: MIC (μg/mL): 1.5; *S. aureus* Giorgio: MIC: 1.5; *S. aureus* 209 P: MIC: 1.5; *S. aureus* 285: MIC: 0.78; *S. aureus* 503: MIC: 0.78; *S. aureus* Smith: MIC: 0.78; *M. luteus* ATCC 9341: MIC: 0.2; and *B. subtilis ATCC 6633*: MIC: 0.78	Shin et al. (2000)
Mansonone G (**119**)	*Antimicrobial*: *P. parasitica*: 81.2%; *X. oryzae pv. oryzicola*: ZI: 17.0; and *X. oryzae pv. oryzae*: 18.3	Mongkol and Chavasiri (2016)
Mansonone H (**120**)	*Antimicrobial*: *P. parasitica*: 33.3%; *X. oryzae pv. oryzicola*: ZI: 18.3; and *X. oryzae pv. oryzae*: 21.7	Mongkol and Chavasiri (2016)
Nocardiones A (**168**)	*Antimicrobial*: *Candida albicans* IFO-1060 (MIC: μg/mL): 6.2; *C. albicans* IFO-1061: 6.2; *C. albicans* ATCC 38247: 6.2; *C. albicans* A-9540: 6.2; *Cryptococcus neoformans*: 1.5; *Saccharomyces cerevisiae* NR5174 S-100: 12.5; *Kluyveromyces fragilis* ATCC 8635: 6.2: *Aspergillus niger*: 6.2; *Aspergillus fumigatus*: 3.1; and *Trichophyton mentagrophytes*: 3.1	Otani et al. (2000)

Abbreviation: ZI: zone of inhibition.

et al. 2005). In another report, Vasconcellos et al. (2010) demonstrated that orthoquinone **111** possessed potent genotoxic, cytotoxic, protective, and antimutagenic effects on *Salmonella tiphymurium*, yeast *S. cerevisiae*, and V79 mammalian cells. Biflorin (**111**) and its dimer bis-biflorin (**112**) was reported to be isolated from *C. biflora* and illustrated significant inhibition toward *Candida auris* polymerase. Based on computational studies, bis-biflorin (**112**) revealed the best interactions with *C. auris* polymerase (da Fonseca et al. 2023).

Orthoquinone **113** was obtained from *Eremophila serrulata* R. Br.(Scrophulariaceae) and screened for antimicrobial effects. It demonstrated significant antimicrobial effects toward *S. aureus* (ATCC 29213) with an MIC: 15.6 μg/mL and a MBC of 125 μg/mL. This molecule further demonstrated antimicrobial effects toward *S. aureus* (ATCC 25923), *Streptococcus pyogenes*, and *Streptococcus pneumoniae* with MBCs and MICs ranging from 7.8 to 125 and 7.8–15.6 μg/mL, respectively (Ndi et al. 2007).

6 | Sesquiterpene Quinones

6.1 | Mansonones

Mansonone A (**114**) (Figure 8) was isolated from *Ulmus americana* L. (Ulmaceae) (Dumas et al. 1983) whereas sesquiterpene quinones mansonones C-I (**115–121**), L (**122**), M (**123**), and O (**124**) were provided by *Thespesia populnea* (L.) Solander ex Correa (Malvaceae), *Ulmus davidiana* Planch, *Mansonia altissima* A. Chev., and *Mansonia gagei* J.R. Drumm. (Malvaceae) (Boonsri et al. 2008; J. P. Kim et al. 1996; Galeffi et al. 1969; C. M. Chen et al. 1990; Tiew et al. 2002). Mansonone E (**117**) possesed good cytotoxic effects toward MCF-7, HeLa, HT-29, and KB cancer lines with IC_{50}: ranging from 0.05 to 0.55 μg/mL. In addition, mansonone D (**116**) demonstrated significant effects toward MCF-7 (IC_{50}: 0.80 μg/mL) and HeLa (IC_{50}: 2.80 μg/mL) (Boonsri et al. 2008). In another study, mansonone **117** was shown to possess significant acetylcholinesterase and

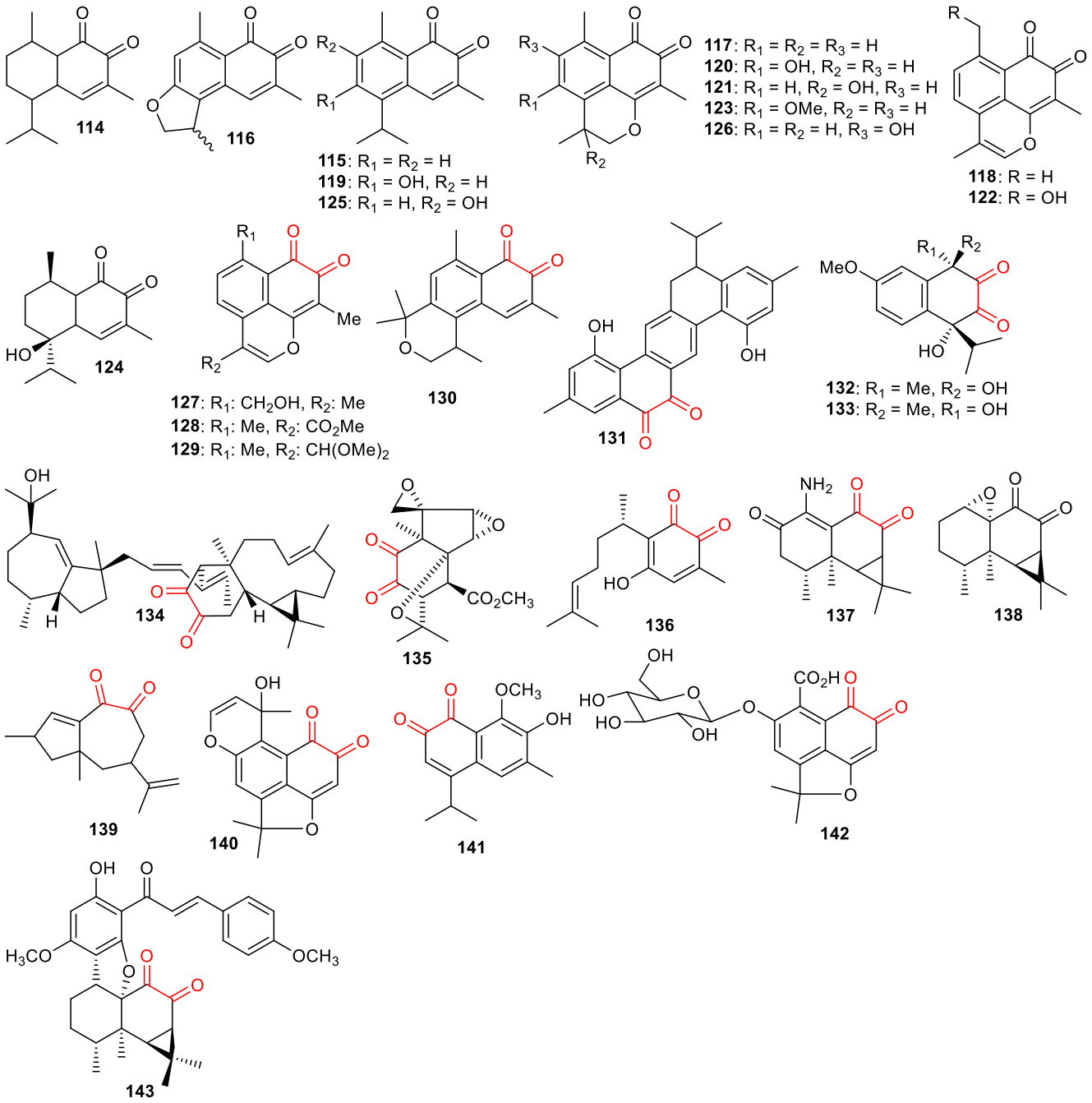

FIGURE 8 | Structures of sesquiterpene orthoquinone **114–143**.

butyrylcholinesterase inhibitions with 81% and 91%, respectively (Changwong et al. 2012). Azanzones A (**125**) and B (**126**) were isolated from *Azanza garckeana* Exell and Hillc. (Malvalceae) (Letcher and Shirley 1992).

6.1.1 | Antimicrobial and Antioxidant Effects

Mansonone **116** showed moderate antibacterial effects toward *B. subtilis* whereas mansonone **117** illustrated weak effects (Boonsri et al. 2008). Mansonones **117** and **118** illustrated significant antioxidant effects with IC_{50}: 0.04 and 0.03 μg/mL, respectively (J. P. Kim et al. 1996). Mansonones **115**, **117**, and **119** possessed potent antimicrobial effects toward *Phytophthora parasitica* with percentage inhibition of 84.2, 94.4, and 81.2, respectively (Mongkol and Chavasiri 2016). Mansonones C (**115**) showed antifungal effects toward *C. albicans* and *Cladosporium cucumerinum* in low concentrations of 0.15 and 0.6 μg/mL, respectively. Similarly, mansonones **117** demonstrated antifungal effects toward *C. albicans* and *C. cucumerinum* in low concentrations of 2.5 and 0.6 μg/mL, respectively (Ioset et al. 2003).

Mansonone E (**117**) possessed potent antifungal effects toward *P. parasitica* with inhibition of 94% whereas mansonones C (**115**) and G (**119**) demonstarted potential antifungal activity toward *P. parasitica* with inhibition of 84% and 81%, respectively (Table 5). Additionally, Mansonone E (**117**) displayed antifungal

TABLE 5 | Sources of orthoquinones **223–244**.

Compound	Source	References	Compound	Source	References
Artabotrine (**223**)	*Artubotrys zeylunicus*	Wijeratna et al. (1995)		*P. wightii*	Boll et al. (1996)
	A. crassifolius	K. K. Tan et al. (2015)		*Stephania cepharantha*	Akasu et al. (1974)
	Artabotrys stenopetalus	Fleischer et al. (1997)	7-Chloro-6-demethylcepharadione B (**229**)	*Houttuyniae cordata*	Gribble (1996)
	A. suaveolens	Barger and Sargent (1939) and Azziz et al. (2013)	1-Desmethoxy-4,5-dioxodehydorasimilobine (**230**)	*Monocyclanthus vignei*	Achenbach et al. (1991)
Aristolodione (**224**)	*Aristolochia chilensis*	Urzua et al. (1987)	1,2-Dimethoxy-4,5-dioxo-6a,7-dehydroaporphine (**231**)	*Piper aborescens*	Duh et al. (1990)
	A. heterophylla	Chan et al. (1999)	4,5-Dioxodehydrocrebanine (**232**)	*Stephania sasakii*	Kumtomc et al. (1980)
	A. kaempferi		Griffithdione (**233**)	*Goniothalamus griffithii*	Y. J. Zhang et al. (1998)
Aristoliukine B (**225**)	*Aristolochia heterophylla*		3-Methoxycepharadione B (**234**)	*Mitrephora maingayi*	D. S. Lee et al. (1999) and N. H. S. Lee et al. (1999)
	A. kaempferi	Wu et al. (2000a, 2000b) and Wu, Leu, et al. (2000)	8-Methoxyoureigidione (**235**)	*Artabotrys zeylanicus*	Wijeratine et al. (1996)
	A. mollissima	Wu, Chan, and Leu (2001) and Wu, Chan, and Lin (2001)	Noraristolodione (**236**)	*Aristolochia indica*	Achari et al. (1982)
Aristoliukine C (**226**)	*Aristolochia kaempferi*	Wu et al. (2000a, 2000b) and Wu, Leu, et al. (2000)		*Fissistigma balansae*	Chia et al. (2000)
Cepharadione A (**227**)	*Aristolochia chilensis*	Urzua et al. (1987)		*F. glaucescens*	Lo et al. (2000)
	A. cucurbitifolia	Wu et al. (2000a, 2000b, 1999) and Wu, Leu, et al. (2000)		*Piper attenuatum*	Desai et al. (1989)
	A. foveolata	Leu et al. (1998)		*P. longum*	Desai et al. (1989)
	A. heterophylla		Norcepharadione A (**237**)	*Annona hayesii*	Rasamizafy et al. (1987)
	A. indica	Che et al. (1984) and Achari et al. (1982)		*Oncodostigma monosperma*	Cave et al. (1986); Abdallah et al. (1989)
	A. kaempferi	Wu et al. (2000a, 2000b) and Wu, Leu, et al. (2000)		*Stephania cepharanta*	Akasu et al. (1975)
	A. kankauensis	Wu et al. (1994)		*Unonopsis pacifica*	Arango et al. (1988)

(Continues)

TABLE 5 | (Continued)

Compound	Source	References	Compound	Source	References
	A. mollissima	Wu, Chan, and Leu (2001) and Wu, Chan, and Lin (2001)	Norcepharadione B (**238**)	*Fissistigma balansae*	Chia et al. (2000)
	A. triangularis			*F. glaucescens*	Lo et al. (2000)
	A. zollingerina	Chiang et al. (1998)		*Goniothalamus tenuifolius*	Y. J. Zhang et al. (1999)
	Piper acutisleginum	Parmar et al. (1998)		*Guatteria ouregou*	Cortes et al. (1986)
	P. argyrophyllum	Singh et al. (1996)		*Houttuynia cordata*	Proebstle and Bauer (1992)
	P. attenuatum	Desai et al. (1989)		*Oxymitra velutina*	Achenbach and Hemrich (1991)
	P. auritum	Hansel et al. (1975)		*Piper attenuatum*	Desai et al. (1989)
	P. betel	Parmar et al. (1998)		*P. boehmerifolium*	Desai et al. (1989)
	P. boehimerifolium	Desai et al. (1989)		*P. hamiltonii*	Desai et al. (1989)
				P. longum	Desai et al. (1989)
	P. bogotense	Pena et al. (2000)	Piperadione (**239**)	*Piper attenuatum*	Desai et al. (1989) and Parmar et al. (1997)
	P. divaricatum	Avella et al. (1994)		*P. hamiltonii*	Desai et al. (1989) and Parmar et al. (1997)
	P. hamiltonii	Desai et al. (1989)	Pontevedrine (**240**)	*Aristolochia elegans*	Wijeratine et al. (1996) and Parmar et al. (1997)
	P. longum	Desai et al. (1989, 1988)		*Glaucium flavum*	Ribas et al. (1971)
	P. manausense	deDiaz et al. (1990)		*Platycapnos spicata*	O. M. Blanco et al. (1993)
	P. methysticum	Jaggy and Achenbach (1992)		*Sarcocapnos enneaphylla*	Tojo et al. (1991)
	P. pedicellosum	Parmar et al. (1998)		*S. saetabensis*	O. Blanco et al. (1991)
	P. thomsoni	Parmar et al. (1998)	Triangularine A (**241**)	*Aristolochia triangularis*	Lewis (2000)
	Stephania cepharantha	Akasu et al. (1974)	1,2,3-Trimethoxy-4,5-dioxo-6a,7-dehydroaporphine (**242**)	*Piper aborescens*	Duh et al. (1990)
	S. sasaki	Kunitomo et al. (1981)		*Pseuduvaria macrophylla*	Mahmood et al. (1986)

(Continues)

TABLE 5 | (Continued)

Compound	Source	References	Compound	Source	References
Cepharadione B (**228**)	*Piper argyrophyllum*	Singh et al. (1996)	Tuberosinone (**243**)	*Aristolochia cinnabar*	Hong et al. (1994) and H. Li et al. (1995)
	P. attenuatum	Desai et al. (1989)		*A. kaempferi*	Wu et al. (2000a, 2000b) and Wu, Leu, et al. (2000)
	P. auritum	Hansel et al. (1975)		*A. tuberosa*	Dayuam et al. (1983)
	P. boehmerifolium	Desai et al. (1989)		*Aristolochin cinabaria*	C. Zhang et al. (1986)
	P. hamiltonii	Desai et al. (1989)	Tuberosinone-N-β-D-glucoside (**244**)	*Aristolochia cinnabarina*	Hong et al. (1994) and H. Li et al. (1995)
	P. longum	Desai et al. (1989, 1988)		*A. tuberosa*	Dayuam et al. (1983)
				Aristolochin cinabaria	C. Zhang et al. (1986)

effects against *Alternaria porri*, *Colletotrichum gloeosporioides*, *Fusarium oxysporum*, and *P. parasitica* with MIC: of 250, 31.2, 500, and 31.2 μg mL^{-1} (Mongkol and Chavasiri 2016). Mansonone E (**117**) also demonstrated significant antibacterial effects toward *Xanthomonas oryzae* pv. *oryzae* and *X. oryzae* pv. *oryzicola* with inhibition zones of 25.7 and 24.3 mm, respectively. The inhibition zone of mansonones C (**115**) and G (**119**) and H (**120**) ranged from 11.3 to 21.7 mm.

Mansonone G (**119**) possessed antibacterial effects toward *B. subtilis*, *E. coli*, and *S. aureus* with MICs of 62.5, 15.6, and 125 μM, respectively (Htoo et al. 2022). In another study, mansonone F (**118**) displayed significant antimicrobial effects against various strains of *S. aureus* along with *Bacillus* sp. and *Micrococcus* sp. (Table 4) (Suh et al. 2006). In addition, mansonone F (**118**) showed significant antimicrobial effects toward various strains of *S. aureus*, *Bacillus* sp. and *Micrococcus* sp. with MIC ranging from 0.2 to 1.5 μg/mL (Table 4) (Shin et al. 2000).

6.2 | Miscellaneous Sesquiterpene Quinones

Sesquiterpene orthoquinones, the davidianones A–C (**127–129**) (Figure 8) were isolated from *U. davidiana*. Notably, compounds **127** and **129** illustrated significant antioxidant effects with IC_{50}: 0.12 and 0.8 μg/mL, respectively, whereas compound **128** possessed moderate antioxidant effects with IC_{50}: 6.9 μg/mL (J. P. Kim et al. 1996). *T. populnea* Solander ex Correa provided the sesquiterpene quinone, populene C (**130**) which was found to be active toward MCF-7, HeLa, HT-29, and KB cancer lines with IC_{50}: ranging from 2.9 to 3.4 μg/mL (Boonsri et al. 2008). Moreover, this molecule also showed antimicrobial effects toward *B. subtilis* with MIC of 4.6 μg/mL (Boonsri et al. 2008). Parviflorene I (**131**), featuring a biscadinane-type skeleton was reported from *Curcuma parviflora* Wall. (Zingeraceae) and illustrated cytotoxic effects toward HeLa cells with $IC_{50:}$ 3.6 μM (Toume et al. 2005).

Tavinin A (**132**) and epi-tavinin A (**133**) (Figure 8) were isolated from *Sterculia tavia* Baill. (Sterculiaceae) and illustrated cytotoxic effects toward ovarian cancer (A2780) with IC_{50}: 5.5 and 6.7 μM, respectively (Dai et al. 2012). Nudibaccatumone (**134**), a trimer featuring two sesquiterpene groups along with phenylpropanoid, is produced by *Piper nudibaccatum* Y. C. Tseng (Piperaceae) (H. X. Liu et al. 2013; M. L. Liu et al. 2013; X. Liu et al. 2013). Coriatone (**135**) was reported from *Coriaria nepalensis* Wall. (Coriariaceae) and this sesquiterpene demonstrated no cytotoxicity toward K562 cells (Y. H. Shen et al. 2004) whereas rigidone (**136**) was reported from the Gorgonian coral *Pseudopterogorgia rigida* (Freyer et al. 1997). Two aristolane sesquiterpenoids, lepidamine (**137**) (J. Tan et al. 2003) and, kanshone C (**138**) (Bagchi et al. 1988) were isolated from *Russula lepida* and *Nardostachys chinensis*, respectively.

Sesquiterpene **139** (Figure 8) is produced by *Syringa pinnatifolia* Hemsl. (Oleaceae) and illustrated significant effects toward *F. oxysporum*, *E. coli,* and *P. vulgaris* but was comparatively less active toward *A. niger*, *Bacillus coagulas*, and *S. aureus*

(Ao et al. 2012). Two cadinane sesquiterpenoids, bombamalone B (**140**), C (**141**), and bombamaloside (**142**) were obtained from *Bombax malabaricum* DC (*B. ceiba* L., Malvaceae) (X. Zhang et al. 2007). Nardoaristolones A (**143**) featuring the aristolane sesquiterpene and chalcone skeleton, was isolated from *N. chinensis* and revealed protective cardiomyocytes effects (H. X. Liu et al. 2013; M. L. Liu et al. 2013; X. Liu et al. 2013).

7 | Phenalenones

Phenalenones are polyketides (heptaketide) bearing fused three-ring frameworks illustrating a wide range of biological effects. (+)-Atrovenetinone (**144**) (Figure 9) was reported from various fungual species viz., *Sirococcus* strain (Ayer and Ma 1992), *Penicillium* sp. (Nakanishi et al. 1995; Shiomi et al. 2005; Narasimhachari and Vining 1963), *Gremmeniella abietina* (Ayer et al. 1986), and *Phoma* sp. (Hussain et al. 2015). Moreover compound **144** is reported to be a myosin chain kinase inhibitor with IC_{50}: 0.86 μM (Nakanishi et al. 1995). Moreover, (−)-atrovenetinone or *ent*-atrove-netinone (**145**), is produced by the fungi *G. abietina* (Ayer et al. 1987, 1986), *Coniothyrium cereale* (Moustafa 2011), and *Penicillium verruculosum* (Nakanishi et al. 1995). The prenylated phenalenone, conioatrovenetinone (**146**) was obtained from the fungus *C. Cereale* (Moustafa 2011). Atrovenetinone (**144**) illustrated good antifungal effects toward *Elymus repens* and possessed moderate antmicrobial effects

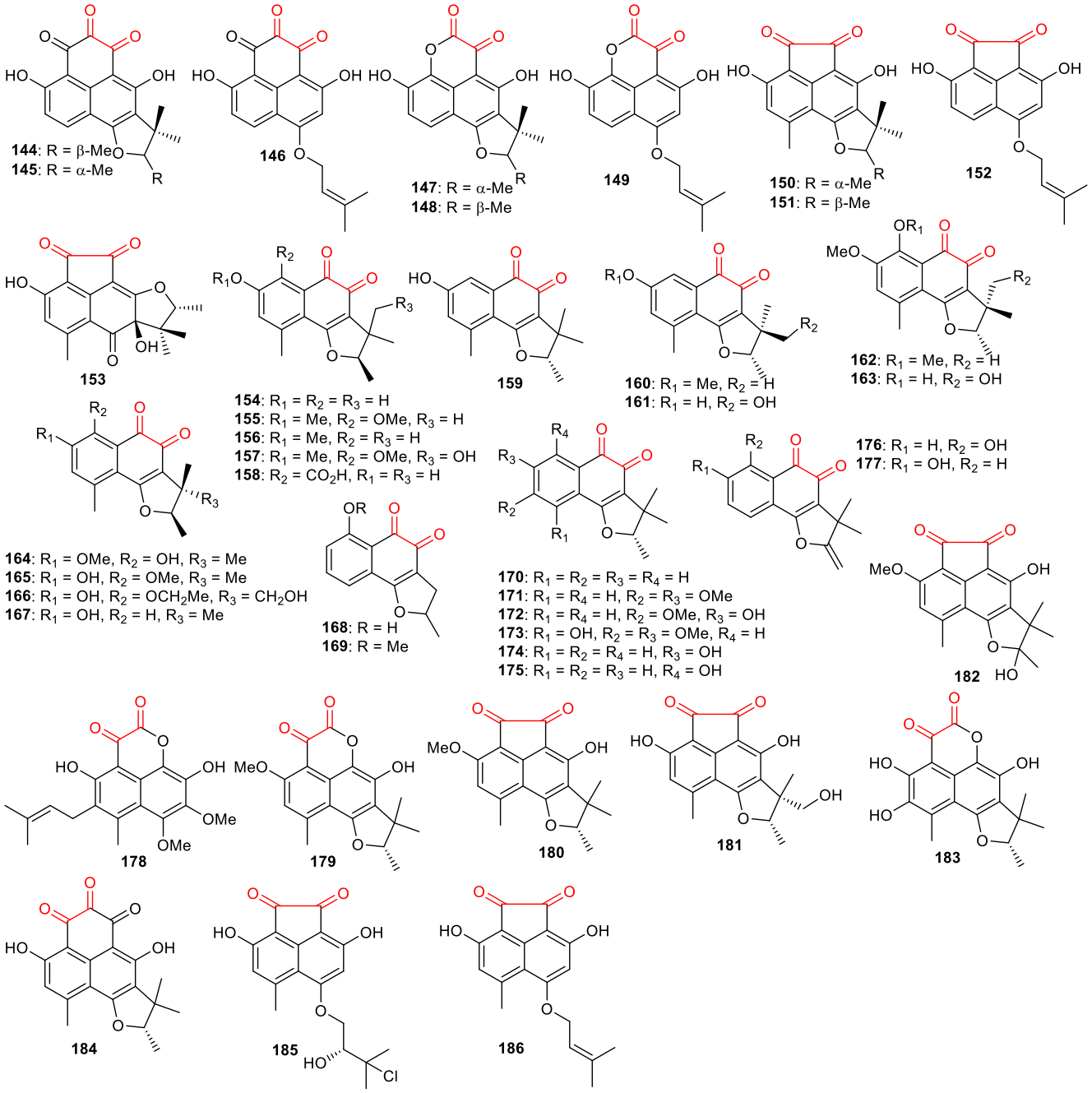

FIGURE 9 | Structures of phenalenones **144–186**.

toward *Mycotypha microspore*, *F. oxysporum,* and *Bacillus megaterium*. On the other hand, this molecule possessed very potent antifungal effects toward *Utricularia violacea* R. Br. (Lentibulariaceae) (Hussain et al. 2015).

(−)-Scleroderolide (**147**) was obtained from *Godronia abietina* (Ayer et al. 1987), *G. cassandrae* (Ayer et al. 1989), *Penicillium* sp., and *C. cereale* (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011). Scleroderolide (**147**) possesses anα-ketolactone (phenylglyoxylate lactone) framework. Moreover, this compound is reported to show antimicrobial effects toward *S. aureus*, *M. luteus*, *B. subtilis*, *Bacteroides fragilis*, *Pyricularia oryzae,* and *Mycobacterium phlei* (Ayer et al. 1989). In another study, scleroderolide (**147**) was shown to possess antimicrobial effects toward *S. aureus* and *E. coli* with MIC of 7 and 9 μg/mL (Q. Li et al. 2018).

A stereoisomer of **147**, (+)-scleroderolide (**148**), was reported from *Penicillium* sp. (Q. Li et al. 2018), whereas coniosclerodérolide (**149**) was isolated from *C. cereale* (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011). Coniosclerodérolide (**149**) illustrated antimicrobial effects similar to metabolite **147** (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011). The acenaphthenequinone analog, (−)-sclerodione (**150**) is produced by *G. abietina* (Ayer et al. 1986) as well as from *C. cereale* (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011) whereas (+)-sclerodione (**151**) was obtained from the fungus *Penicillium* sp. (Q. Li et al. 2018). Compound **151** illustrated moderate antifungal effects toward *E.* repens and possessed very potent antifungal effects s toward *U.* violacea (Hussain et al. 2015). The fungus *C. cereale* produces the heptaketide phenalenone, coniosclerodione (**152**), which has a prenyl group (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011). Peniciphenalenin A (**153**) is produced by *Penicillium* sp. and possesses antiproliferative effects toward glioma cells at a concentration of 100 μM (Q. Li et al. 2018).

(+)-Trypethelone (**154**) (Figure 9), methoxytrypethelone methyl ether (**155**), trypethelone methyl ether (**156**), and 4′-hydroxy-8-methoxytrypethelone methyl ether (**157**) were isolated from *Trypethelium eluteriae* (Mathey et al. 1980). In addition, compounds **154**, **156,** and **157** were also obtained from the fungus *Astrothelium* sp. (L. Y. Sun et al. 2010). Peniciphenalenin D (**158**) is produced by *Penicillium* sp. and possesses antiproliferative effects toward glioma cells at a concentration of 100μM (Q. Li et al. 2018). Additionally, the fungus *C. cereale* produced (−)-trypethelone (**159**) (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011, 2012). Compound **154** showed significant antibacterial effects toward *B. subtilis* with MIC: 5.0 μg/mL. This compound was shown to be active toward *S. aureus col* (MRSA) with an MIC value of 2.5 μg/mL (L. Y. Sun et al. 2010).

Compound **159** possesses antimicrobial effects toward *E. coli*, *S. aureus*, *M. phlei,* and inhibits mouse fibroblast cells with IC_{50}: 7.5 μM (Elsebai, Kehraus, et al. 2011; Elsebai, Natesan, et al. 2011). Phenalenones (−)-2′*S*-trypethelone methyl ether (**160**), (−)-(2′*S*,3′*S*)-4′-hydroxytrypethelone (**161**), (−)-2′*S*-8-methoxytrypethelone methyl ether (**162**), and (−)-(2′*S*,3′*R*)-4′-hydroxy-8-methoxy trypethelone methyl ether (**163**) were isolated from the lichen *Marcelaria cumingii* (Jarupinthusophon et al. 2019). Compounds **160–162** possess cytotoxic effects toward KB and HeLaS3 with IC_{50}: 9.8–53.7 μM. Compound **160** was the most potent toward HeLaS3 (IC_{50}: 5.1 μM) and KB (IC_{50}: 9.8 μM). In addition, compound **160** is cytotoxic toward HCT116 (IC_{50}: 0.3 μM), A549 (IC_{50}: 1.0 μM), and HT29 (IC_{50}: 6.1 μM) (Jarupinthusophon et al. 2019).

8-Hydroxytrypethelone methyl ether (**164**) (Figure 9), trypethelones, 8-methoxytrypethelone (**165**), and 5′-hydroxy-8-ethoxytrypethelone (**166**) along with compounds **157** and **159** were isolated from *Trypethelium eluteriae*. Metabolite **164** illustrated moderate effects with MIC: 25 μg/mL toward *M. phlei*, *M. Szulgai*, *Mycobacterium parafortuitum*, *Mycobacterium flavescens*, *Mycobacterium chitae*, and *Mycobacterium kansasii* whereas compounds **157** and **159** showed antimicrobial effects toward *S. aureus* (MIC: 25 μg/mL) (Srinivasan et al. 2020). Another phenalenone, 5′-hydroxytrypethelone (**167**) along with compounds **154**, **159**, and **164** were also obtained from *T. eluteriae* and tested toward RKO, A549, and HepG2 cell lines. All these compounds possess moderate-to-weak cytotoxic effects toward RKO cells with IC_{50}: 22.6–113.5 μM (Basnet et al. 2019). Nocardiones A (**168**) and B (**169**) were isolated from a strain of *Nocardia* sp. Compound **168** illustrated moderate antifungal effects toward various *Candida* sp. and other fungi and yeasts (Table 3). Moreover metabolite **168** also possess effects toward PTP1B (IC_{50}: 14 μM), Cdc25B (IC_{50}: 17 μM), and FAP-1 (IC_{50}: 89 μM) whereas this compound furthermore induces cell death in U937 myeloid leukemia cells (Otani et al. 2000).

Dunnione (**170**) (Figure 9), a natural product reported from *Streptocarpus dunnii* Mast. (Gesneriaceae), showed various biological effects. Dunnione (**170**) is reported to possess antitumor and antifungal properties as well as NQO1 stimulator (Bian et al. 2015; H. J. Kim et al. 2016). Dunnione (**170**) exhibits nephrotoxicity via regulating Sirt1/Nrf2 and NQO1 axes. Quinones **171–175** are produced by *Sinningia reitzii* L.E. Skog (also Gesneriaceae) and compound **171** illustrated cytoxic effects toward PC-3 and HeLa with IC_{50}: 7.7 and 4.4 μmol/L, respectively (A. S. Silva et al. 2019). *S. reitzii* also produced 8-hydroxydehydrodunnione (**176**) along with 7-hydroxydehydrodunnione (**177**) and compound **176** illustrated potent anti-inflammatory as well as antinociceptive effects (Soares et al. 2017).

Flaviphenalenone C (**178**) (Figure 9) was obtained from the fungus *Aspergillus flavipes* and illustrated α-glucosidase effects with IC_{50}: 78.9 μM. In addition, this molecule possessed cytotoxic effects toward A549 and MCF-7 with IC_{50}: 28.5 and 50.0 μM, respectively (L. H. Zhang et al. 2016). (−)-Bipolarides A (**179**) and B (**180**) are produced by *Lophiostoma bipolare* and compound **180** possesses antibacterial effects toward *B. cereus* with MIC: 25 μg/mL. On the other hand, this latter compound was only weakly active toward MCF-7, KB, and NCI-H187 cancer cells with IC_{50}: 79.4, 96.8, and 56.5 μM, respectively. Interestingly, compound **179** illustrated antimicrobial effects toward *B. cereus* with MIC: 12.5 μg/mL and cytotoxicity toward NCI-H187 with IC_{50}: 60.2 μM (Intaraudom et al. 2015). Peniciphenalenin H (**181**) was obtained from the fungus *Pleosporales* sp. and demonstrated antimicrobial effects toward the *Proteus* species, *B. subtilis*, *Vibrio parahemolyticus*, *MRCNS*, and

MRSA with MIC: 50, 25, 25, 25, and 50 25 μM, respectively (Y. Han et al. 2021). Bipolarol A (**182**) was isolated from the fungus *L. bipolare* and showed weak cytotoxic effects toward MCF-7 with IC_{50}: 110.4 μM (Intaraudom et al. 2015).

(+)-8-Hydroxyscleroderolide (**183**) was isolated from the sponge *Acanthella cavernosa* (Le Goff et al. 2019). Antatrovenetinone (**184**) was obtained from *C. cereale* and this molecule possesses an interesting 1,2,3-triketone system (Elsebai et al. 2016). The fungus *Pleosporales* sp. produced peniciphenalenin G (**185**) and I (**186**). Compound **185** illustrated antimicrobial effects toward *B. cereus*, *P. species*, *M. phlei*, *B. subtilis*, *V. parahemolyticus*, *MRCNS*, and *MRSA* with MIC: 25.0, 50.0, 50.0, 25.0, 50.0, 50, 25.0, and 12.5 μM, respectively (Y. Han et al. 2021).

8 | Alkaloids Containing *o*-Quinone Systems

8.1 | Pyrrole Alkaloids

8.1.1 | Damirones and Related Alkaloids

The alkaloids batzellines A–C (**187–189**) (Figure 10) along with 6-dechlorobatzelline C (**190**), feature the tricyclic pyrrolo[4,3,2-*de*]quinoline core were reported as isolated from the sponge *Batzella* sp. (Sakemi et al. 1989; Dijoux et al. 2005). The chemical structure of batzellines A–C (**187**) was established via X-ray study, whereas those of **188** and **189** were determined by chemical and physical methods. Bewley and their coworkers isolated batzelline D (**191**) from the sponge *Zyzzya fuliginosa* (L. C. Chang et al. 2002). Damirones A (**192**) and B (**193**) were reported from *Damiria* sp. (Stierle and Faulkner 1991) and *Histodermella* sp. (Carney et al. 1993). Later both of the *Histodermella* and *Damiria* sp. were reidentified as *Z. fuliginosa* (Antunes et al. 2005). The structure of damirones A (**192**) has also been confirmed by X-ray diffraction spectroscopy (Utkina et al. 2003). In another investigation of *Z. Fuliginosa*, it provided damirone C (**194**) (Schmidt et al. 1995). The pyrroloquinoline alkaloids, makaluvone (**195**) was reported from *Zyzzya* cf. *marsailis* (Radisky et al. 1993) and later *Zyzzya* cf. *marsailis* was reidentified as *Z. fuliginosa* (Antunes et al. 2005). In addition, makaluvamine O (**196**) was reported to be isolated from the sponge *Smenospongia aurea* (J. F. Hu et al. 2002). Moreover, makaluvamine O (**196**) exerts significant cytotoxic effects toward the HCT-116 HCT-116 cells ($p21^{-/-}$, $p53^{-/-}$, $p21^{+/+}$, and $p53^{+/+}$) with IC_{50s} 94 to 8.6 μM. Notably, this compound was very effective toward $p21^{-/-}$ with IC_{50}: 8.6 μM (Tasdemir et al. 2002). In addition, makaluvamine O (**196**) illustrates moderate antimalarial effects toward *P. falciparum* with IC_{50}: 0.94 μM (J. F. Hu et al. 2002).

In 1993, the Steglich group reported the first fungal pyrroloquinoline alkaloid, haematopodin (**197**) (Figure 10) to be isolated from *Mycena haematopus* (Baumann et al. 1993). In addition to this pyrroloquinoline alkaloid, mycenarubins A (**198**) and B (**199**) were also isolated from *Mycena rosea* (Peters and Spiteller 2007a) whereas the indolo-6,7-quinone alkaoid, sanguinolentaquinone (**200**) was obtained from *Mycena sanguinolenta* (Peters and Spiteller 2007b). Pelianthinarubins A (**200a**) and B (**201**) featured the S-hercynine group were reported from *Mycena pelianthina* and no activity toward *E. coli*, *B. subtilis*, *B. brevis*, and *C. cucumerinum* was observed (Pulte et al. 2016). Quite recently mycenaflavin B (**202**) were isolated from *M. haematopus* (Lohmann et al. 2018).

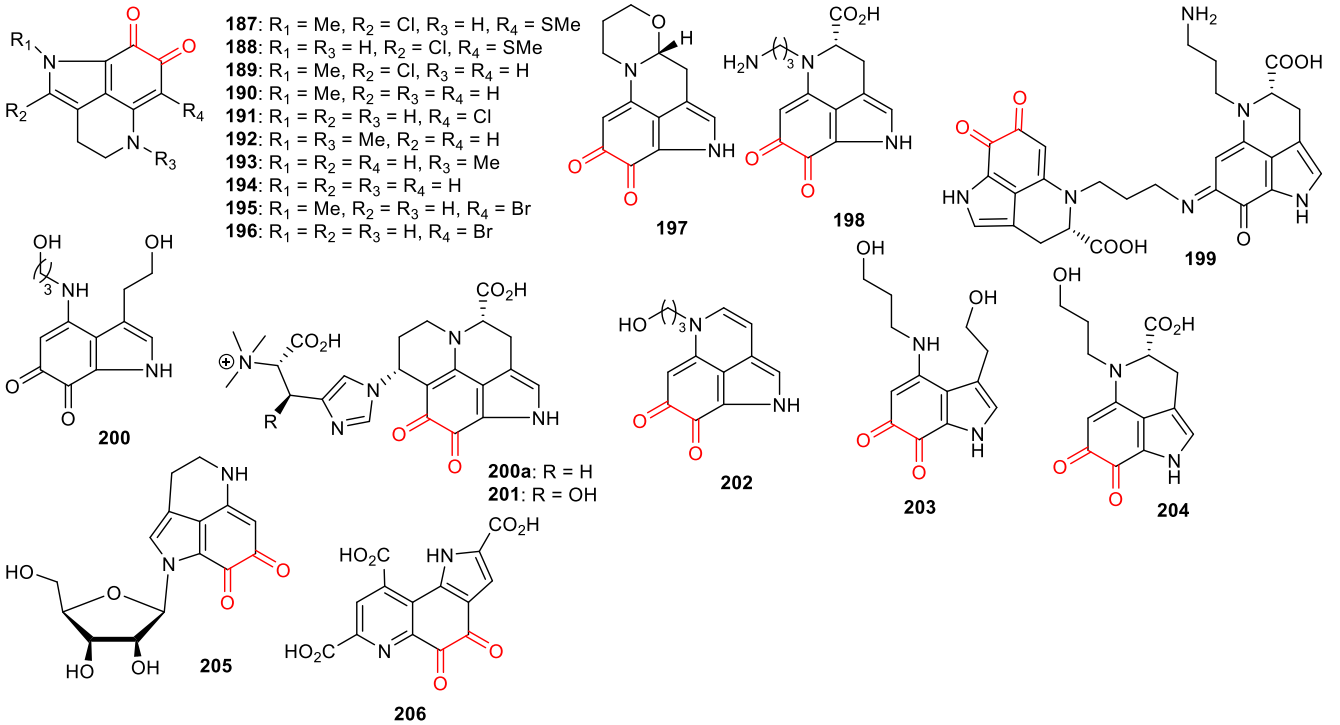

FIGURE 10 | Structures of alkaloids **187-206**.

In 2007, Peters et al. reported on the pyrroloquinoline alkaloid sanguinolentaquinone (**203**) (Figure 10) (Peters and Spiteller - 2007b) they isolated from *M. sanguinolenta* whereas mycenarubin F (**204**) was obtained from *M. haematopus* (Peters et al. 2008). In another study, the pyrroloquinoline *N*-glycoside, *N*-1-β-D-ribofuranosyldamirone C (**205**) was isolated from *Strongylodesma aliwaliensis* (Keyzers et al. 2004).

8.1.2 | Pyrroloquinoline Quinone

Pyrroloquinolinequinone (**206**, PQQ) (Figure 10), also known as methoxatin, was isolated from cultures of methylotropic bacteria (Akagawa et al. 2016). Moreover, PQQ **206** is reported to be present in human milk, daily eaten foods, including vegetables and meats (Kumazawa et al. 1995; Kumazawa et al. 1993; Noji et al. 2007). Numerous dietary supplements have been marketed in the USA comprising $PQQNa_2$ and these products have no side effects (Akagawa et al. 2016).

8.1.2.1 | Growth Promotion. In 1984 (Shimao et al. 1984), it was known that PQQ (**206**) which was isolated from *Pseudomonas* sp. that is poly(vinyl alcohol)-degrading bacteria is required for bacterial growth. These findings were also extended to other bacterial species such as *Acinetobacter* and yeasts (Ameyama et al. 1984). The PQQ (**206**) is also present in *Pseudomonas fluorescens* B16, and when it is applied to *Solanum lycopersicum* L (tomato plant, family: Solanaceae), flower number, height of the plant, fruit weight, and fruit number were increased and when it was applied to *Cucumis sativus* L (cucumber plant.), PQQ significantly increased its growth (Choi et al. 2008).

It is known that mineral phosphate is required environmental element for the growth of plants. It is provided by rhizobacteria from the genera *Agrobacterium*, *Rhizobium*, *Acinetobacter*, *Erwinia*, *Achromobacter*, *Bacillus*, *Micrococcus*, *Pseudomonas*, and *Flavobacterium* (Vyas and Gulati 2009; Bashan et al. 2013). Microorganisms that can solubilize mineral phosphate contain compound 206 which acts a cofactor (Babu-Khan et al. 1995). Their effect in increasing plant growth was reported (H. Rodriguez and Fraga 1999).

8.1.2.2 | Antimicrobial Effects. To control the synthesis of PQQ (**206**) is very important as indicated by bacterial species *Rahnella aquatilis* HX2 (Li et al. 2014), which can solubilize hydroxyapatite and is indicated as a biocontrol (Li et al. 2014; Calvo et al. 2007; Godziszewska et al. 2017) although being pathogenic in nature. When the PQQ synthetic genes pqqB and pqqA were mutated separately, both compound 206 and gluconic acid were not produced. Therefore, its antibacterial activity toward sunflower crown gall disease was decreased as it did not solubilize hydroxyapatite (Li et al. 2014). The *Enterobacter intermedium* 60-2G is a bacterial species that can solubilize phosphate and produces systemic resistance to *Erwinia carotovora*, soft rot plant pathogen, by gluconic acid formation, showing quinone 206 as a cofactor (J. Y. Han et al. 2008; S. H. Han et al. 2008). As predicted, mutation of pqqB or pqqA prevented the biosynthesis of 206, and microorganism could not solubilize mineral phosphate or decrease *E. carotovora* or Magnaporthe grisea KI-409 the rice pathogen (J. Y. Han et al. 2008; S. H. Han et al. 2008). From bioinformatics research data (vide infra), PQQ-producing microorganisms that are identified as pathogenic to humans, plants, or other mammals were identifies (Y.-Q. Shen et al. 2012) as an important and potential lead for drug development and plant protective agents. It is recognized as pathogenic in nature on pineapple plant in Philippines since 1928 (Serrano 1928), it produces its effects on Sudan grass, maize, watermelons, rice, honeydew melons, onion, sugar cane, and tomatoes and results in critical economic losses (Coutinho and Venter 2009). It is also threat to inherent plants, and therefore traditional drug sources and biodiversity conservation (Anderson et al. 2004). The aerobic microorganisms which accumulate hydroxyapatite, their genome sequence indicated homologs of glucose dehydrogenase (mGDH) and a possible PQQ synthetic operon (vide infra) similar to the pqqABCDEF cluster from bacterium, *Klebsiella pneumoniae* (Andreeva et al. 2011).

8.1.2.3 | Antioxidative Effects. The PQQ (**206**) has potential free radical scavenging effect due to chemiluminescence inhibition caused by carrageenin and zymosan, N-formyl-methionyl-leucyl-phenylalanine, in mouse model of peritoneal cells, and of xanthine-xanthine oxidase reaction in brain homogenate of rats (Hamagishi et al. 2008). It has more potency than idebenone, ascorbic acid, and α-tocopherol. The 7- and 9-carboxy and ortho-quinone functional groups were noted as important for the effect. Oxidative stress induced by Rose Bengal was prevented by 1, and oxidative damage to plasmid DNA induced with γ-irradiation was inhibited by 1, through ROS scavenging activity (Misra et al. 2004). As compared to niestriol, the PQQ (4 mg/kg) was more efficient in protecting from γ-irradiation, enhancing hemopoietic recovery of white blood cells, reticulocytes, and bone marrow cells (Xiong et al. 2011). In vitro model, the PQQ decreased oxidative injury due to H_2O_2 in cardiomyocytes of male rat through reducing membrane potential depolarization of mitochondria and decreasing ROS level (Tao et al. 2007). The PQQ (206) enhanced PGC-1α mRNA, its expression of protein and induced protection toward 3-nitropropionic acid and exogenous sodium azide in mouse model of Hepa1−6 cells (Chowanadisai et al. 2010). The PQQ (206) also increased the elevation of mouse fibroblasts by extracellular signal-regulated kinases activation (ERK 1/2) (Kumazawa et al. 2007). Moreover, the PQQ (> 10 nM) increased human epithelial cells (A31) proliferation by tyrosine autophosphorylation of epidermal growth factor receptor (Kimura et al. 2012). The PQQ protected from oxidative stress caused by lipid peroxidation in mitochondria and inactivation of respiratory chain within mitochondria. It was noted that Jurkat cells that are oxidant-sensitive were damaged by 1 (250 μM) due to H_2O_2 generation in RPMI 1640 culture medium (He et al. 2003).

8.2 | Carbozole Alkaloids

Alkaloids comprising an orthoquinone group, viz., carbazoquinocins A–F (**207–212**) (Figure 11) were isolated from *Streptomyces violaceus* and tested for their biological effects. Alkaloid

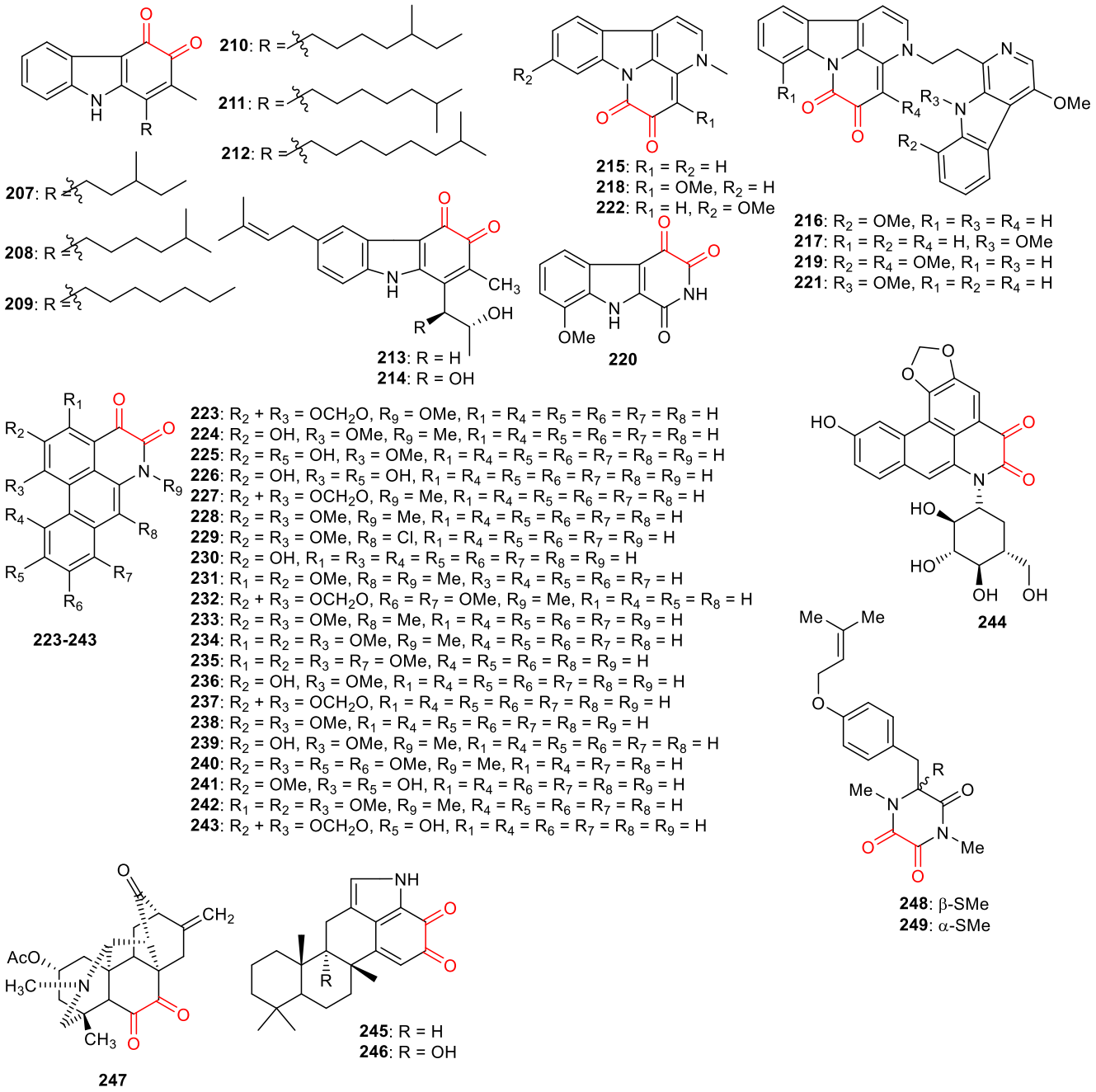

FIGURE 11 | Structures of alkaloids **207–249**.

208 bearing a 5-methylhexyl group was the most potent in antioxidant effects with IC_{50}: 0.12 mM. Moreover, the heptyl group slightly decreases the ativity (compound **209**: IC_{50}: 0.22 mM) whereas a 5-methylpentyl and 7-methyloctyl group significantly decreases antioxidant effects as compound **207** and **212** possessed IC_{50}: 0.41 and 0.42 mM, respectievly. Compounds **210** and **211** possess good antioxidant effects with IC_{50}: 0.37 and 0.33 mM, respectievely (M. Tanaka et al. 1995). Carquinostatins A (**213**) and B (**214**) are produced by *Streptomyces exfoliatus*. Compound **213** completely inhibited the toxicity of L-glutamate in N18-RE-105 cells at a concentration of > 20 nM whereas orthoquinone **214** had an $EC_{50:}$ 72 nM. In addition, compound **213** possessed antioxidant effects and its effects are comparable to that of vitamin E (Shin-Ya et al. 1993, 1997).

8.3 | Carboline-Type Alkaloids

Picrasidine L (**215**), M (**216**), N (**217**), O (**218**), U (**19**), and V (**220**) (Figure 11) were reported from the root bark of *Picrasma quassioides* (D. Don) Benn. (Simaroubaceae) (Ohmoto and Koike 1985a, 1985b; Koike et al. 1990). Amarastelline A (**221**) was reported from *Quassia amara* L. (Simaroubaceae) with fluorescent properties and possessed cytotoxic potentials toward HeLa cancer cells (Taniguchi et al. 2012). Moreover picrasidine L (**215**) illustrated potent PTP1B effects with IC_{50}: 19.8 μM and a kinetic study demonstrated that this compound is a competitive inhibitor with *Ki*: 7 μM (Sasaki et al. 2015). Eurycomine E (**222**) was reported from *P. quassioides* and illustrated cerebral protective activity (Sasaki et al. 2016) as well as potent PTP1B

effects with IC_{50}: 19.1 μM (Sasaki et al. 2015). Furthermore a kinetic study demonstrated that this compound is a noncompetitive inhibitor with *Ki*: 12.9 μM (Sasaki et al. 2015).

8.4 | 4,5-Dioxoaporphines

4,5-Dioxoaporphines account for a small section of the aporphine alkaloids. Artabotrine (**223**) (Figure 11), *N*-methoxylated alkaloid was reported from *Artubotrys zeylunicus* (Wijeratna et al. 1995), *Artabotrys crassifolius* (K. K. Tan et al. 2015), and *Artabotrys suaveolens* (Barger and Sargent 1939; Azziz et al. 2013). Artabotrine (**223**) illustrated moderate cytotoxicity effects toward HL60, RPMI8226, HCT116, and MCF7 with IC_{50}: ranging from 5.5 to 24.5 μM (Barger and Sargent 1939). Compound **223** was also reported to show good inhibitory effects toward P-388 leukemia cells (Wijeratna et al. 1995). Moreover, artabotrine (**223**) illustrated antibacterial effects toward various bacteria viz., *Listeria monocytogenes*, *P. vulgaris*, *Bacillus cereus*, *M. luteus*, *Rhodococcus equi*, *S. agalactiae*, and *S. aureus* and with MIC: ranging from 1.25 to 5 mg/mL (K. K. Tan et al. 2015). Orthoquinones **224–244** and their plant sources are presented in Table 5.

8.5 | Miscellaneous Alkaloids

Two indole-based alkaloids, mycoleptodiscins A (**245**) and B (**246**) (Figure 11), were isolated from the fungus *Mycoleptodiscus* sp. and **246** was shown to possess significant cytotoxic effects toward H460, A2058, H522T1, and PC3 cancer cell lines with IC_{50} ranging from 0.60 to 0.78 μM (Ortega et al. 2013). Mycoleptodiscin B (**246**) showed significant antimicrobial effects toward *B. subtilis* and *S. aureus* with MICs of 0.5 and 1 μg/mL, respectively. On the other hand, **246** was less potent toward methicillin resistant *S. aureus* and *C. albicans* with MIC values of 32 and 64 μg/mL (Dissanayake et al. 2016). Vilmorrianone (**247**), a diterpenoid alkaloid, is produced by *Aconitum vilmorrianurn* (Ding et al. 1991). The fungus *Penicillium crustosum* was isolated from the soil province, Yunnan China and this fungus produced two piperazine-triones viz., **248** and **249** (H. Wang et al. 2015).

9 | Miscellaneous Compounds

Dipuupehedione (**250**) (Figure 12), a dimer of puupehenone, is produced by the sponge *Hyrtios* sp. and is active on KB cells with IC_{50}: 3 μg/mL (Bourguet-Kondracki et al. 1996). Biruloquinone (**251**) was isolated from the fungus *Cladonia macilenta* and showed potent anti-Alzheimer and neuroprotective effects (Heng et al. 2013). This molecule also showed antioxidant activity with EC_{50}: 0.49 mmol/L (Huan and Tian 2015). Theanaphthoquinone (**252**), a catechin derivative is produced by tea leaf of *Camellia sinensis* (L.) Kuntze (Theaceae) (T. Tanaka et al. 2000). Dehydroatalantin (**253**), was reported from *Atalantia monophylla* (L.) DC (Rutaceae) (Dreyer et al. 1976) whereas another limonoid **254** was produced by *Atalantia zeylanica*, which was also named dehydroatalantin which is confusingly the same as the limonoid **253** (Bennett et al. 1994).

Terretonins B (**255**) (Figure 12), a sesterterpenoid, was obtained from *Aspergillus terreus* (C. Li et al. 2005; G. Y. Li et al. 2005). Obionin A (**256**) was initially isolated from the fungus *Leptosphaeria obiones* (Poch and Gloer 1989) and later from the fungus *Microcyclospora malicola* (Surup et al. 2015). Moreover, polyketide **256** illustrated antimicrobial effects toward various bacteria and fungi such as *M. luteus*, *Mycobacterium diernhoferi*, *Nocardioides simplex*, *Nocardia* sp., *S. aureus*, *Aspergillus clavatus*, *Mucor hiemalis*, *Nematospora coryli*, *Rhodotorula glutinis*, *Schizosaccharomyces pombe*, and *Trichosporon oleaginosus* (Surup et al. 2015). Obionin B (**257**) was obtained from an unidentified fungus of the order pleosporales and possesssed cytotoxicity toward MCF-7, H460, SF-268, HT-29, and MDA-MB-435 with IC_{50}: 11.1, 7.6, 13.2, 3.1, and 7.3 μM, respectively (Ayers et al. 2011).

Spiromentins B (**258**) and C (**259**) are produced by the fungus *P. atrotomentosus* (Buchanan et al. 1995) whereas fordianaquinone A (**260**) was reported from *Lysimachia fordiana* Oliv and illustrated weak cytotoxicity toward A549 cells (X. A. Huang et al. 2007). Herbimycin E (**261**) was isolated from *Streptomyces* sp. and this molecule has high binding affinities toward Hsp90α which is employed in the treatment of neurodegenerative diseases (Shaaban et al. 2013).

Laccaridiones A–C (**262–264**) (Figure 12) and leptosphaerodione (**265**) were isolated from *Montagnula* sp. (Berg et al. 2000) and compounds **262** and **263** inhibited trypsin, zinc-protease, papain, thermolysin, collagenase, and with IC_{50}: ranging from 2.5 to 18.8 μg/mL (Almeida et al. 2019). Additionally, compound **264** displayed cytotoxic effects toward A2058 and MCF-7 with IC_{50} of 13.2 and 3.7 μM, respectively (Almeida et al. 2019). In another study, quinones **262** and **263** potently inhibited *C. albicans* adhesion to endothelial and epithelial (Falkensammer et al. 2008). Laccaridione B (**263**) illustrated cytotoxic effects toward L-929, K-562, and HeLa with IC_{50}: 2.4, 1.8, and 13.9 μg/mL, respectively (Berg et al. 2000).

The pulvinic acid dimer, badione A (**266**) was obtained from the fungus *Scleroderma citrinum* (Winner et al. 2004) and was also reported from *Xerocomus badius* (Steffan and Steglich 1984). (+)- and (−)-ecarlottone (**267**) were isolated from *Fissistigma latifolium* and illustrated significant antagonistic effects for proteins Bcl-xL/Bak (*Ki*: 4.8 μM) and Mcl-1/Bid (*Ki*: 2.4 μM). On the other hand, the racemic mixture **267** possesses potent antagonistic effects for protein Mcl-1/Bid (*Ki*: 3.1 μM) while being less effective for protein Bcl-xL/Bak (*Ki*: 17.5 μM) (Geny et al. 2017). Albidin (**268**) was produced by the fungus *Penicillium albidum* (Grove and Hitchcock 1986).

10 | Antimicrobial and Antioxidant Orthoquinones in Food Applications

10.1 | Fresh-Keeping During Food Storage and Processing

Naturally sourced antioxidants are valuable bioactive compounds with proven potential to benefit the food industry. In addition to its use in functional foods, it is increasingly being

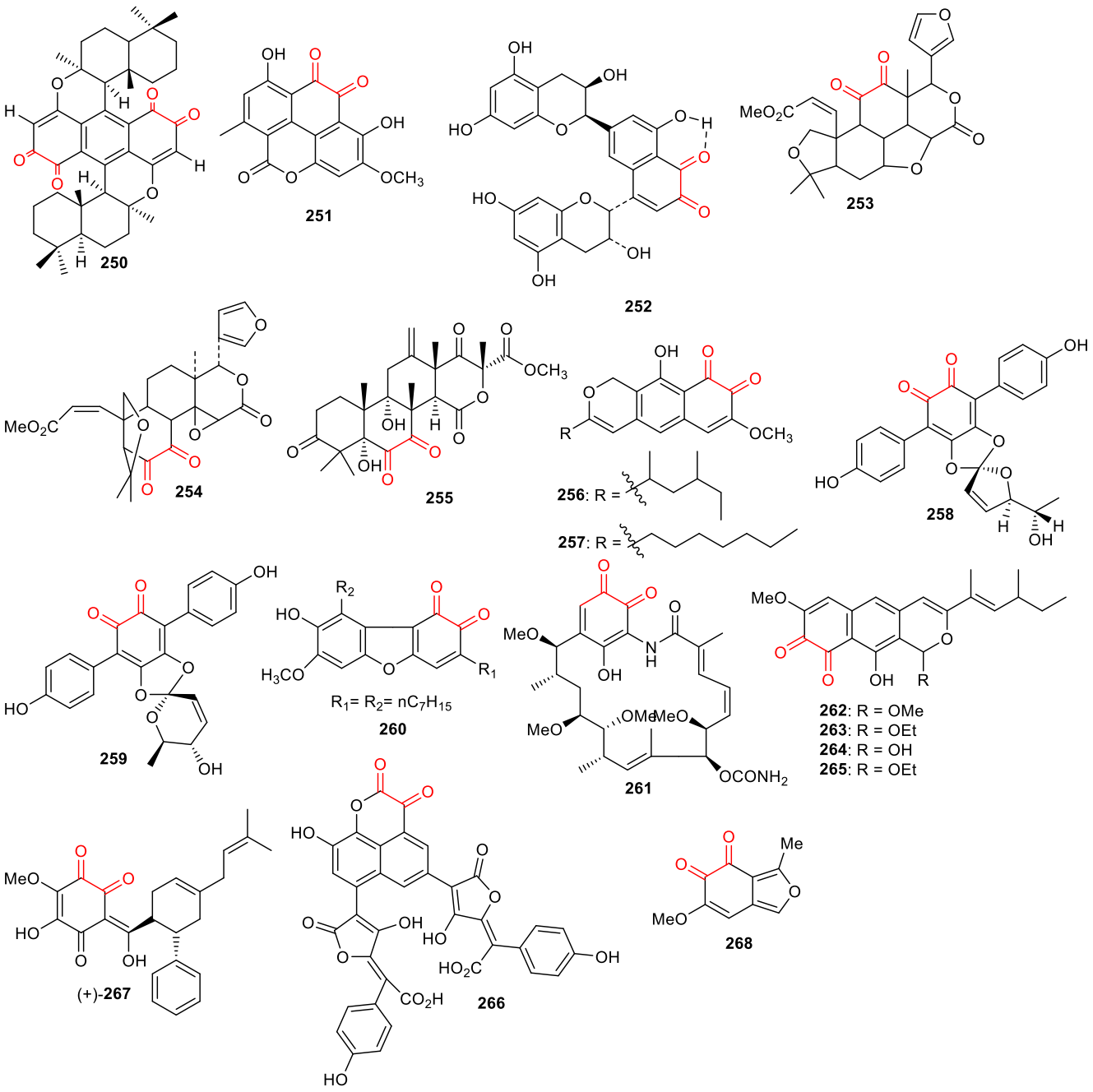

FIGURE 12 | Structures of miscellaneous compounds **250–268**.

used as an alternative to synthetic antioxidants to enhance stability and prevent oxidative degradation during processing and storage. Efforts are being made to use natural antioxidants from agricultural food byproducts and underutilized plant materials as part of a circular economy (Lourenço et al. 2019).

Since food is not consumed immediately after production, it has to be stored and transported. Therefore, suppliers must ensure that it is delivered to consumers in a safe and high-quality manner, while maintaining the same or higher nutritional value than when it was produced. Strategies to improve overall food quality and shelf-life are constantly being sought, often by reducing or inhibiting oxidative damage; the addition of antioxidants is one strategy to retard oxidation of biomolecules. There is great potential for natural antioxidants to replace synthetic antioxidants as they are readily available from natural sources (Lourenço et al. 2019). Various orthoquinones possessed significant antioxidant potentials. For example, the majority of tanshinones and mansonones showed promising antioxidant activity. In addition, pyrroloquinoline quinone (**206**, PQQ) is the most active orthoquinone showing potent antioxidant effects. The strong antioxidant capacity of antioxidant orthoquinones makes them attractive for food preservation, including meat, edible oils, and plant foods.

10.2 | Food Safety

Food is a constant nutritional need for human life. However, unwanted microorganisms can contaminate foods, food

processing facilities, or manufacturing environments, resulting in the risk of human illness. Foodborne pathogens are microbes that can cause illness in humans by virulence mechanisms, sometimes even in low infectious doses. The control of such microorganisms has evolved throughout human history. It has made it possible to extend the shelf-life of food and to produce safer and more

TABLE 6 | Antimicrobial effects of orthoquinones against various organisms.

Bacteria		
Bacillus subtilis	*Corynebacterium insidiosum*	*Streptococcus agalactiae*
Bacillus subtilis ATCC 6633	*Micrococcus roseus*	*Streptococcus pyogenes*
Bacillus thuringiensis	*Sarcina lutea*	*Streptococcus pneumoniae*
Bacillus cereus	*Mycobacterium smegmatis*	*Brevibacillus laterosporus*
Bacillus cereus ATCC 6633	*Mycobacterium diernhoferi*	*Salmonella tiphymurium*
Bacillus brevis	*Mycobacterium phlei*	*Klebsiella pneumoniae*
Bacillus coagulas	*Mycobacterium szulgai*	*Bacteroides fragilis*
Bacillus megaterium	*Mycobacterium flavescens*	*Vibrio parahemolyticus*
Staphylococcus aureus	*Mycobacterium chitae*	*Brevibacillus brevis*
S. aureus (ATCC 29213)	*Mycobacterium kansasii*	*Bordetella bronchiseptica*
S. aureus (A3)	*Mycobacterium parafortuitum*	*Enterococcus faecalis*
S. aureus (1474/01)	*Listeria monocytogenes*	*E. faecalis* (ATCC 29212)
S. aureus ATCC 25923	*Rhodococcus equi*	*Micrococuss flavus*
S. aureus ATCC 6538P	*Nocardia* sp.	*Microcystis aeruginosa*
S. aureus TMS 64	*Ralstonia solanacearum*[a]	*Xanthomonas vesicatoria*[a]
S. aureus TMS 33	*Xanthomonas oryzae*	*Xanthomonas campestris*
S. aureus TMS 417	*R. solanacearum*[a]	*Agrobacterium tumefaciens*
S. aureus Smith	*Pseudomonas aeruginosa*	*Micrococcus luteus*
S. aureus SG 511	*Pseudomonas lachrymans*[a]	*M. luteus* ATCC 9341
S. aureus Giorgio	*Staphylococcus haemolyticus*	*M. luteus* ATCC 10240
S. aureus 209 P	*Escherichia coli*	*Staphylococcus epidermidis*
S. aureus 285	*Proteus vulgaris*	*S. epidermidis* (ATCC 12228)
S. aureus 503	*Kliebsiella pneumoniae*	*S. epidermidis* (RP12)
S. aureus col (MRSA)	*Arthrobacter citreus*	*S. epidermidis* (6756/99)
Fungi		
Candida albicans	*Aspergillus clavatus*[b]	*Aspergillus niger*[b]
C. albicans ATCC 38247	*Aspergillus fumigatus*[b]	*Alternaria porri*
C. albicans IFO-1060	*Fusarium oxysporum*[b]	*Colletotrichum gloeosporioides*[b]
C. albicans IFO-1061	*Fusarium verticillioides*[b]	*Mycotypha microspore*
C. albicans A-9540	*Trichophyton mentagrophytes*	*Pyricularia oryzae*[b]
Candida auris	*Bipolaris maydis*[b]	*Cryptococcus neoformans*
Cladosporium cucumerinum[b]	*Nocardioides simplex*	*Mucor hiemalis*
Nematospora coryli[b]	*Cochliobolus sativus*[b]	*Trichosporon margaritipherum*
Yeast		
Saccharomyces cerevisiae	*Trichosporon oleaginosus*	*Kluyveromyces fragilis*
S. cerevisiae NR5174 S-100	*Schizosaccharomyces pombe*	*Rhodotorula glutinis*
Algae		
Scenedesmus obliquus	*Chlorella pyrenoidosa*	
Oomycete		
Phytophthora parasitica		

[a]Plant pathogenic bacteria.
[b]Plant pathogenic fungi.

nutritious food. Microbial control methods are mainly aimed at inhibiting the growth of unwanted microorganisms. This is done by applying physical or natural antimicrobial strategies (Abdelhamid and El-Dougdoug 2020).

L. monocytogenes, *S. tiphymurium*, *E. coli*, *E. faecalis*, *S. aureus*, *B. subtilis*, *S. cerevisiae*, and *V. parahemolyticus* were the most common foodborne microorganisms (Abdelhamid and El-Dougdoug 2020). Notably, majority of orthoqunone natural products illustrated significant antimicrobial effects toward various foodborne microorganisms (Table 6). These orthoquinones can be effectively used to protect consumers from foodborne pathogens and premature spoilage of food, either alone or in combination with other orthoquinones or other antimicrobial natural products.

10.3 | Food Packaging

Many of the current studies into the incorporation of antimicrobials into active packaging continue to be both exciting and challenging for researchers. The ability of active packaging to carry food additives such as antimicrobials and antioxidants has been thoroughly investigated. The majority of these antimicrobial solutions are incorporated into edible coatings or films in active packaging and are released onto the surface of the food in a controlled manner. Rather than applying the antibacterial directly to the surface of the food, and slowing its penetration into the food, this method can be a better alternative (Batiha et al. 2021). Orthoquinones have significant antimicrobial properties against a variety of organisms (Table 6). Due to their antimicrobial effects, these natural products can be used in the packaging of food products and can help to increase the shelf life of various food products.

10.4 | Applications in Agriculture

Natural antimicrobials have broad agricultural applications, including animal health, plant disease control, and plant growth. Continued research and development in this area may further extend agricultural applications of natural antimicrobials and contribute to sustainable and safe food systems. Plant diseases cause an estimated 10%–15% of the world's major crops to fail each year, resulting in direct economic losses of up to several hundred billion dollars. In particular, 70%–80% of diseases are caused by pathogenic fungi, which have a negative impact on crop growth and yield. Phytopathogens reduce crop yield and quality, causing enormous losses in agricultural production (Peng et al. 2021; Sar et al. 2023). Many of the orthoquinones reported in this review had significant antifungal properties (Table 6) toward various fungi. In particular, several orthoquinone natural products showed significant antifungal effects toward plant pathogen fungi including *C. cucumerinum*, *N. coryli*, *A. clavatus*, *Aspergillus fumigatus*, *A. niger*, *F. oxysporum*, *F. verticillioides*, *B. maydis*, *C. sativus*, *C. gloeosporioides*, and *P. oryzae*. In addition, some orthoquinones were also active toward plant pathogen bacteria such as *R. solanacearum*, *P. lachrymans*, and *X. vesicatoria*.

11 | Conclusion and Future Perspectives

This review describes numerous orthoquinone metabolites along with their antimicrobial, antioxidant, and other promising biological effects. In addition, the review included a large library of orthoquinone secondary metabolites, complemented by extensive biological studies as well as the most comprehensive database of recent literature. The vast majority of orthoquinone natural products illustrated antimicrobial potential toward several microorganisms (including plant pathogens), namely, *Candida* sp., *Fusarium* sp., *Aspergillus* sp., *Cryptococcus* sp., *Botrytis* sp., *Saccharomyces* sp., *Escherichia* sp., *Bacillus* sp., and *Staphylococcus* sp.

Antimicrobial resistance is a major obstacle to modern medicine and to succeeding in treating infectious diseases. In particular, resistance to antimicrobial leads is also significantly affecting food production around the world. This is because agriculture and food production have depended on using chemicals to control pathogenic microbes. As a result, it has been difficult to eradicate or combat pathogens over recent decades, leading to food insecurity and food production issues. The antimicrobial potential of these orthoquinone metabolites is an indication that these fungal metabolites may have potential as natural preservatives in the food industry. In addition, the antimicrobial properties of orthoquinones may also be useful in improving the shelf life of food products.

This review has resulted in a reasonable number of new lead structures. These have the potential to enter and complement the current drug development pipeline. We believe that this detailed review of orthoquinone metabolites will further stimulate the use of orthoquinone pharmacophore generation strategies (chemoselective and chemoenzymatic) as a platform that can serve as a useful medicinal chemistry tool for optimizing secondary metabolites, offering exciting opportunities to improve any future secondary metabolite therapeutic potential.

Author Contributions

Hidayat Hussain: conceptualization, writing – original draft, writing – review and editing, formal analysis, validation. **Daijie Wang:** conceptualization, formal analysis, resources. **Satyajit D. Sarker:** investigation, validation. **Lutfun Nahar:** validation, writing – review and editing. **Nilufar Z. Mamadalieva:** methodology. **Jianbo Xiao:** funding acquisition, writing – review and editing, project administration, resources, conceptualization.

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The authors have nothing to report.

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The authors declare no conflicts of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon request. The data are not publicly available due to privacy or ethical restrictions.

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