Abstract: Endophytic fungi are an important group of microorganisms and one of the least studied. They enhance their host’s resistance against abiotic stress, disease, insects, pathogens and mammalian herbivores by producing secondary metabolites with a wide spectrum of biological activity. Therefore, they could be an alternative source of secondary metabolites for applications in medicine, pharmacy and agriculture. In this review, we analyzed patents related to the production of secondary metabolites and biotransformation processes through endophytic fungi and their fields of application. We examined 245 patents (224 related to secondary metabolite production and 21 for biotransformation). The most patented fungi in the development of these applications belong to the Aspergillus, Fusarium, Trichoderma, Penicillium, and Phomopsis genera and cover uses in the biomedicine, agriculture, food, and biotechnology industries.

Keywords: endophytic fungi; patents; secondary metabolites; biotransformation; biological activity

1. Introduction

The term endophyte refers to any organism (bacteria or fungi) that lives in the internal tissues of a host. This endophyte–host association is complex: it is normally driven without causing harm or apparent disease symptoms and provides benefits in survival, fitness, biodiversity, and ecosystem function for both parties by enhancing the response to environmental stress and producing the same or similar compounds that originate in the host [1–3]. In particular, fungal endophytes have been the focus of many studies due to their prospective promise in the production of secondary metabolites with pharmacological, agricultural, industrial, or biotechnological applications [4–6].

Endophytic fungi were discovered over a century ago; however, it was not until about three decades ago, with the discovery of the taxol-producing endophytic fungus Taxomyces andreanae, that they gained remarkable relevance due to the abovementioned production of active secondary metabolites [7–10].

As was the case with taxol, the process for the isolation and purification of metabolites in adequate yields remains a major concern; low yields due to the exploitation of the host for the extraction process of metabolites are also associated with environmental impacts, and new strategies such as involving the use of endophytic microorganisms instead of the host themselves have offered new niches that should be meticulously investigated and used as a base for sustainable research and development [11,12].
in the process of finding new drug candidates or precursors for the synthesis of new molecules. We also cover the production of secondary metabolites in biotransformation processes by using endophytic fungi.

2. Materials and Methods

This review was conducted mainly through searches of the databases Scifinder® and Google Patents. Our search was made under the subjects “endophytic fungi” and “patents” covering the period from 2001 to 2019. 4670 references were found. After removing duplicates, we selected those related to the production of secondary metabolites and biotransformation. Resulting in 245 documents from which 224 were related to any kind of secondary metabolite derived from endophytic fungi and 21 detail biotransformation processes of metabolites through endophytic fungi. The patents covered in this study are described in Tables 1 and 2.

3. Results

The description and analysis of patents was divided into two sections: those that are connected to the production of secondary metabolites and those associated with biotransformation processes. Likewise, two tables were constructed in which the main generalities of each patent are summarized.

3.1. Production of Secondary Metabolites

Early patents consisted mainly of registering the endophytic strains capable of producing specific compounds or those that represented a novel source of active metabolites (chanoclavine in EP1142986A2; resveratrol in CN1948459A; gallic acid in CN101280279A; taxol in CN101486974A) and very few applications. However, over time, patents were developed to include the registration of methods and procedures to produce and recover the compounds of interest (with a known biomedical application) or to optimize or increase their production (podophyllotoxin in US20040248265A1; taxol in CN1624103A; camptothecin in US20060134762A1; huperzine A in CN101275116A). In the last ten years, patents have been focused on using novel or enhanced fermentation processes to obtain high yields of products and provide possible applications for the metabolites (alpha-pyrone in CN110563740A; epimedins A–C in CN10511876A; differanisole A in CN109971655A; 5, 8-ergosterol epoxide in CN109971651A). The distribution of the patents in relation with the principal areas of application are illustrated in Figure 1. The production of taxol and huperzine A were considered as other application outside of their anticancer and anti-Alzheimer property respectively, due to the number of patents and economic importance.

The principal applications consist on providing metabolites that are precursors of bioactive molecules (baccatin III and cephalomannine in CN103194502A) and those that can be use as anticancer, antitumor, antineoplastic or immunosuppressive agents (anthraquinone compounds in CN102586355A; cerrenin D in CN109456191A; alterporriol P in CN102633616A; dalesconol A and B in CN104031948A; quinazolone alkaid compound in CN103570744A); in pesticides, insecticidal, algcal control (diterpene alkaid-like compounds in CN102190699A); as antibacterial, antibiotic, antimicrobial, bacteriostatic (beauvericin in CN101240249A; diterpene alkaid compound in CN102190612A); as antifungal and antymycotics (Trichoderma acid in CN103083290A); in neurodegenerative diseases and neuroprotective agent (huperzine A in CN102191294A); as agents in pharmacy, food, cosmetics, agriculture and health care products (pseutorin A in CN104774774A; alterlactone in CN110093838A); antioxidant (flavipin in CN103087923A); anti-inflammatory and anti-rrheumatic (1,4-napthoquinones in CN109293494A); in cardiovascular diseases (breviscapine in CN1421522A); anti-diabetes (2 isabolen sesiquiterpenes in CN109096056A); anti-tuberculosis (enniatin compounds in CN101469939A); antiviral (alterporriol Q and R in CN102643186A); as pigments; hepatoprotective agents (pyrrole-type compounds in CN103667073A); in biofuels. Table 1 displays the patents, endophytic fungi, host organism, secondary metabolites, and disclosed applications. The structures of the compounds listed in Tables 1 and 2 are shown in Figure S1 (see the Supplementary Information).
The principal endophytic fungi reported in this section of patents belong to the genera *Aspergillus*, *Fusarium*, *Trichoderma*, *Penicillium*, and *Phomopsis* with 31, 24, 18, 16, and 8 patents, respectively, and compounds such as taxol or paclitaxel, huperzine A, camptothecin, podophyllotoxin, and resveratrol. Methods for enhancing their production represented most of the registered applications. Furthermore, the diversity of compound structures demonstrates the capability of fungi to synthesize simple or very complex molecules.

Mostly, *Aspergillus* endophytes from plants of the genera *Taxus* and *Torreya* are described as having applications related to obtaining the highest yield of paclitaxel or its precursors, like baccatin III and cephalomannine, due to their anticancer activity. Endophytes from *Huperzia serrata* have been linked to the production of huperzine A and its analogs due to their anti-senile dementia and anti-neurodegenerative applications. Plant endophytes such as *Nothapodytes nimmoniana* and *Camptotheca acuminata* have been linked to the production of the antineoplastic agent camptothecin and some analogs. Production of the lignan-type compound podophyllotoxin has been described for several endophytes. This compound has high biomedical potential as an anticancer, antiviral, and antibacterial agent, among others, and is the precursor of the anticancer drugs etoposide and teniposide. The stilbenoid compound found on grape skin, resveratrol, could have promising therapeutic actions against obesity, type II diabetes mellitus, metabolic syndrome, cancer, autism, dementia, and Alzheimer’s disease [13]. Therefore, a number of patents involving endophytes of the genera *Cladosporium*, *Fusarium*, *Alternaria*, and *Penicillium* for its production were registered. The demand for natural resveratrol has gained traction in various end-use industries.
Table 1. Endophytic fungi and their methods of production of natural products.

| Patent No. | Endophyte | Host | Patent Details | Ref. |
|------------|-----------|------|----------------|------|
| EP1342986A2 | Neotyphodium sp. | Not disclosed | Channoclavine (1)-production. | [14] |
| US6329193B1 | Cladosporium macrocarpon | Taxus spp. | Production of taxol. | [15] |
| CN1421522A | Alternaria sp. | Erigeron sp. | Production of brevescapine B (2) and other flavonoids for the treatment of cardiovascular diseases and for preparing antitumor medicine. | [16] |
| US6638742B1 | Alternaria sp. | Alnus rubra, Corylus sp., Cytisus scoparius, Ginkgo sp. | Methods for obtaining and recovering taxanes, including paclitaxel (3), from novel sources. | [17] |
| US6137388B1 | Cryptosporiopsis cf. quercina | Tripterygium wilfordii | Isolation of cryptocandin possessing antifungal activity. | [18] |
| US20040185031A1 | Muscodor vitigenus | Paullinia paullinoides | Novel fungi that produces naphthaleine and applications. | [19] |
| US20040206997A1 | Muscodor albus | Cinnamomum tree | Novel fungi and production of organic volatile antibiotics effective in the treatment of human and animal waste. | [20] |
| US20040249365A1 | Phialocephala fortinii | Podophyllum sp. | Identification of podophyllotoxin-producing fungi and methods for recovering podophyllotoxin (4) from such fungi. | [21] |
| WO2004106487A1 | Neotyphodium lolii | Pooidae grass | Preparation of the sesquiterpenoids trichotec-9-en-4-ol, 12, 13, epoxy-, and 4-epoxy-7,12,13-trihydroxy-5a,8α-epoxytetraol (17), and succinic acid (18) as antimicrobial active ingredients. | [40] |
| CN1624103A | Mix of Taxus endophytes | Taxus chinensis | Increase the production of taxol and taxol precursors. | [41] |
| US6911338B2 | Muscodor sp. | Cinnamomum zeylanicum, Grevillea pteridifolia | Preparing of compound 2,3-diamino-6-hydroxy-benzoic acid-2-ethyl-hexyl ester (10), including method, and its application in pharmacy. | [30] |
| CN1850765A | Halosorrellina sp. | mangrove | Preparation of sesquiterpenoids trichotec-9-en-4-ol, 12, 13, epoxy-, and 4β-acebacil (11) as pesticides. | [31] |
| US2006034762A1 | Fungal strain MTCC 5124 | Mappia sp. | Preparation of the sesquiterpenoids trichotec-9-en-4-ol, 12, 13, epoxy-, and 4β-acebacil (11) as pesticides. | [32] |
| US707985B2 | Muscodor albus | Cinnamomum zeylanicum | Novel fungi and production of organic volatile antibiotics effective in the treatment of human and animal waste products. | [27] |
| CN1896232A | Fusarium sp. | Ginkgo biloba | Production of plasmin. | [28] |
| CN1894859A | Cladosporium sp. | Parthenocissus tricuspidatae | Production of resveratrol (9). | [29] |
| CN1951907A | Aspergillus niger | Euphorbia sp. | Preparation of compound 2,3-diamino-6-hydroxy-benzoic acid-2-ethyl-hexyl ester (10), including method, and its application in pharmacy. | [30] |
| CN101037656A | Trichoderma harzianum | Bux cornuta | Preparation of the sesquiterpenoids trichotec-9-en-4-ol, 12, 13, epoxy-, and 4β-acebacil (11) as pesticides. | [31] |
| CN101041840A | Trichoderma harzianum | Bux cornuta | Preparation of the sesquiterpenoids trichotec-9-en-4-ol, 12, 13, epoxy-, and 4β-acebacil (11) as pesticides. | [32] |
| US7192939B2 | Pestalotiosis microspora | Terminalia morbenesis | Novel fungi strains capable of producing novel antioxidant and antiviral agents. | [33] |
| CN101195804A | Acremonium endophytes | Huperzia serrata | Production of huperzine A (12) analogous through strain liquid fermentation of the endophytic fungi. | [34] |
| CN101239451A | Aspergillus clavatothecum | mangrove | Preparation of bipherol compound (13) including preparation method and application. | [35] |
| CN101275036A | Mix of endophytes | Huperzia serrata | Preparation of huperzine A. | [36] |
| CN101240249A | Fusarium sp. | Dioscorea zingiberensis | Production of beauvericin (14) description of its antibacterial activity. | [37] |
| CN101280279A | Phomopsis sp. | Acer ginnala | Production of gallic acid (15). | [38] |
| US7341862B2 | Muscodor albus | Cinnamomum zeylanicum | Novel fungi and production of organic volatile antibiotics effective in the treatment of human and animal waste products. | [39] |
| CN10142971A | Fusarium sp. | Paris polyphylla var. yunnanensis | Production of 5α, 8α-ergosterol peroxide-6, 22-diene-3β-ol (16), ergosterol-8(9), 22-diene-3β, 5α, 6β, 7α-tetraol (17), and succinic acid (18) as antimicrobial active ingredients. | [40] |
| CN101468977A | Phomopsis sp. | Asadricha indica | Novel pseudo-phomallactone (19) antibacterial compound from fermentation products of an endophytic fungus strain. | [41] |
| CN101468996A | Phomopsis sp. | Asadricha indica | Source of ten-membered lactone 7α-acetoxy-multipolide A (20) and its applications. | [42] |
| CN101481379A | Cladosporium globoseum | Ginkgo biloba | Obtaining chaetomugilin D (21) from an acetic acid ethyl ester extract of fermentation liquor. | [43] |
| CN101486974A | Aspergillus niger | Taxus cuspidata | Production of taxol from endophytic fungus. | [44] |
| CN101503658A | Not disclosed | Locoweed | Separation of an endophytic fungus producing swainsonine (22). | [45] |
Table 1. Cont.

| Patent No. | Endophyte | Host ¹ | Patent Details | Ref. |
|------------|-----------|--------|----------------|-----|
| CN101525611A | Fusarium sp. | Chrysanthemum sp. | Plasmin preparation. | [46] |
| CN101586082A | Aspergillus candidus | Taxus x media | Production of taxol. A method for preparing taxol is also given. | [47] |
| US20090142816A | Gloeocadium sp. | Eucryphia cordifolia | Production of volatile compounds and hydrocarbons to generate biofuels. | [48] |
| CN101619291A | Chaetomium cupreum | Macaranga cordata | Preparatin of 3,6'-tetrahydroxy-4,4'-dimethyl-2-hexa-2,5-diene-2,5,5'-tetraone (23) with antitumor properties. | [49] |
| CN101669939A | Not disclosed | Mangrove | Ennatin compound (24) that aids in the preparation of anti-tubercle drugs. | [50] |
| CN10170120A | Fusarium proliferatum | Mangrove | Improving the output of anticancer anthraquinone compound (25) by utilizing different vaccination methods. | [51] |
| CN101875905A | Shiraia bambusicola | Phyllostachys edulis seed | High-yield hypocrellin-producing strain that carries out hypocrellin (26) production by fermentation. | [52] |
| CN101914452A | Penicillium chrysogenum | Not disclosed | Huperzine A-producing strain. | [53] |
| KR201004252A | Scolosbasidium tohaeyschae | Soybean | Gibberellin (27) production using soybean endophyte. | [54] |
| WO2010062199A | Aspergillus sp. | Garcinia scorchtini | Cyclic peptides with utility in anticancer treatments. | [55] |
| CN101942393A | Shiraia sp. | Taxus serrata | Production of huperzine A. | [56] |
| CN102080110A | Not disclosed | Neothamnopsis minnimana | Technical process for synthesizing a camptothecin sugar derivative. | [57] |
| CN102187870A | Colletotrichum gloeosporioides | Huperzia serrata | High-producing strain and method for huperzine A production. | [61] |
| CN102168017A | Aspergillus oryzae | Red algae | Use of diterpenoidal alkaloid (31) secondary metabolites as pesticides. | [62] |
| CN102190612A | Aspergillus oryzae | Red algae | Preparation of diterpenoidal alkaloid (32) with bacteriostatic activity that can be used for preparing antimicrobial agents. | [63] |
| CN102190614A | Aspergillus oryzae | Red algae | Use of diterpenoidal alkaloid (33) as an insecticide agent. | [64] |
| CN102190608A | Aspergillus oryzae | Marine algae | Preparation and application of alga endophytic fungi diterpenoidal alkaloid compound (34). | [65] |
| CN102191294A | Azcomonium endophyllum | Huperzia serrata | Production of huperzine A as an anti- senile dementia pharmaceutical ingredient. | [66] |
| CN10220247A | Verticillium dahlia | Radix glycyrrhizae | Preparation of diterpene alkaloid (35) for use as an insecticide. | [67] |
| IN201004117A | Phomopsis wenchengensis | Not disclosed | Production of glycyrrhetic acid (36). | [68] |
| JP2011051953A | Colletotrichum gloeosporioides | Heterosiphonia sp. | Manufacture of neohexa-hydro-curcumin (37). | [70] |
| WO201114634A | Hypoxylon sp./Nodulisporium sp./Dalmania sp./Nasudora sp. | Persia indica | Production of volatile organic compounds from these fungi. | [71] |
| CA2766412A | Fungal endophyte of Pinus strobus | Pinus strobus | Antifungal metabolites (38–44). | [72] |
| CN102525154A | Penicillium steckii | Trysterigmatium wilfordii | Production of triptolide (45). | [73] |
| CN10241783A | Phomopsis sp. | Camptotheca acuminata | Production and method for preparation of camptothecin. | [74] |
| CN10246643A | Trichoderma atroviride | Cephalotaxus fortunei | New compound (46) in secondary metabolites of C. fortunei endophytic fungi and its preparation method and application thereof. | [75] |
| CN102595917A | Fusarium sp. | Podophyllum hexadrum | Preparation of podophyllotoxin. | [76] |
| CN102596355A | Fusarium proliferatum | Mangrove | Method for producing anticancer anthraquinone compounds. | [77] |
| CN102628018A | Aspergillus niger | Sclerotium chinense | Improved production of the main components schisandrol A (47), schisandrin A (48), deoxyschizandrin (49), schisandrin B (50) from S. chinensis through fermentation. | [78] |
| CN102633616A | Alternaria sp. | Sarcopteryx sp. | Preparation of the anthraquinone dimer allopodol PI (51) as an antineoplastic agent. | [79] |
| CN102643167A | Alternaria versicolor | Marine algae | Fermentation preparation and application as an antibacterial and insecticidal agent of albicans-11,14-diol (52). | [80] |
| CN102643186A | Alternaria sp. | Sarcopteryx sp. | Preparation of the anthraquinone dimers allopodol Q (53) and allopodol R (54) for antiviral drugs. | [81] |
Table 1. Cont.

| Patent No. | Endophyte | Host | Patent Details | Ref. |
|------------|-----------|------|----------------|------|
| CN102643755A | Penicillium chrysogenum | Glycyrrhiza glabra | Endophytic fungus that improves the content of glycyrrhetinic acid by fermenting licorice. | [82] |
| CN102653720A | Colletotrichum gloeosporioides | Huperzia serrata | Endophytic fungus capable of generating huperzine A. | [83] |
| CN102660466A | Aspergillus penicillioides | Schizandra chinensis | Improves the content of the active ingredients of S. chinensis: schizandrin, schisantherin, deoxyschizandrin, and schisandrin B. | [84] |
| CN102660467A | Fusarium oxysporum | Glycyrrhiza glabra | Fungal strain that produces glycryrrhetic acid. | [85] |
| CN102676992A | Trichoderma atroviride | Salvia miltiorrhiza | Endophytic fungus that aids in the production of tanshinone I (55) and tanshinone IIa (56). | [86] |
| CN102701935A | Trichoderma longibrachiatum | Seaweed | Preparation of tetrinuclear diterpenoid (57) with pesticidal and bacteriostatic activity. | [87] |
| CN102703327A | Cladosporium sp. | Aconitum leucomeatum | Fungal strain capable of synthesizing aconitine (58) for the preparation of antitumor, anti-inflammatory, and antihypertensive drugs. | [88] |
| CN102719362A | Alternaria sp. | Merlot grapes | Fungal strain capable of producing a large amount of resveratrol in the fermentation process. | [89] |
| CN10272427A | Fusarium proliferatum | Ceythropia glabra | Separation method for swainsonine-producing endophytic fungus. | [90] |
| CN10272428A | Fusarium oxysporum | Cajanus cajan | Endophytic fungal strain with a high yield of cajaniin C24 acid (59). | [91] |
| CN10276077A | Acrocomium sp. | Sophora alopecuroides | Synthesis of matrine (60). | [92] |
| WO2012020364A1 | Fungal strain MTCC 5544 | Pongamia pinnata | Preparation of 2',4'-dihydroxy-6'-methoxyl-3',5'-dimethylhydroquinone (61). | [93] |
| CN103073527A | Phomopsis sp. | Illgera odorata | Dipetide derivative (62) for the treatment of cancer. | [94] |
| CN103074236A | Trichoderma atroviride | Camptotheca acuminata | Preparation of libertellone G (63) as a novel medicine for treating Alzheimer’s disease. | [95] |
| CN103083290A | Trichoderma sp. | Not disclosed | Production and application of camptothecin. | [96] |
| CN103085574A | Chaosporium globosum | Cinquefoil biloba | Trichoderma acid (64) is involved in the preparation of antifungal agents. | [97] |
| CN103087923A | Colletotrichum sp. | Not disclosed | The endophytic fungus and metabolite flavipin (65) acts as an antioxidant. | [98] |
| CN103103134A | Colletotrichum sp. | Not disclosed | Production of huperzine A. | [99] |
| CN103134950A | Nodulisporium syzygiforme | Taxus sp. | Separation and purification of taxol by biological fermentation as well as precursors such as baccatin III (66) and cephalomannine (67). | [100] |
| CN103288807A | Not disclosed | Trepertigium wifidii | Separation of alkaloids (68–70) with pharmaceutical application. | [101] |
| CN103360531A | Xylaria sp. | Asatrafurin indica | Obtaining three isopimarane diterpenoid compounds (71–73) with antifungal activity and potential applications in new agricultural or medical antifungal medications. | [102] |
| CN103436451A | Colletotrichum sp. | Cyclopseudospora | Production of haematochrome, including its production via a fermentation method. | [103] |
| IN2011DE03381A | Diaporthe sp. | Pandanus amaryllifolius | Antitubercular diaportheone B analogs (74–75) and their synthesis. | [104] |
| WO2012020364A1 | Fungal strain MTCC 5544 | Myroxylon balsamum, Taxodium distichum | System and method for producing volatile organic compounds. | [105] |
| US2013013713A1 | Colletotrichum sp. | Persica indica | Production of antifungal and immunosuppressive compounds. | [106] |
| US20130224315A | Colletotrichum sp. | Pterospermum sp. | Production of volatile organic compounds and methods of use. | [107] |
| US20130252289A1 | Muscodor strobili | Not disclosed | Production of volatile organic compounds from microorganisms. | [108] |
| US20130252289A1 | Several fungi such as Nodulisporium sp., Hypoxylon sp., Sclerotinia sp., Daldinia sp., Xylaria sp. | Thelypteris angustifolia, Persica indica, Citrus aurantiifolia, Myrobalan balsamum, Taxodium distichum | Production of volatile organic compounds from microorganisms. | [109] |
| US20130302480A1 | Muscodor crispans | Ananas sp., Ananas annosamoides | Production of compounds with wide range of applications in agriculture, industrial, building, pharmaceutical and/or personal care products. | [110] |
| WO2013161834A1 | Fusarium solani | Taxis celebica | Cost-effective process for commercial production of paclitaxel. | [111] |
| CN10357044A | Scopulariopsis sp. | Carioa sp. | Preparation method for the quinazoline alkaloid compound (76) and its application as a tumor cell growth inhibitor. | [112] |
| CN1036203726A | Fungal strain L1 CGMCC No. 4558 | Polygophum cuspidatum | Extraction of resveratrol from fermented liquor. | [113] |
| CN103642864A | Shiria bhamciculara | Huperzia serrata | Preparation of hypocrocin compounds. | [114] |
| CN10366070A | Trichoderma sp. | Huperzia serrata | Preparation and application of huperzine A. | [115] |
| CN10366070A | Colletotrichum sp. | Huperzia serrata | Preparation of 4α,13α-epoxy-huperzine A (77). | [116] |
| CN103670073A | Penicillium laticzella | Huperzia serrata | Preparation of pyrrole type (78) liver-protecting medicines. | [117] |
Table 1. Cont.

| Patent No. | Endophyte | Host | Patent Details | Ref. |
|------------|-----------|------|----------------|-----|
| CN103620332A | Pycnoporus sanguineus | Huperzia serrata | Production of huperzine A. | [118] |
| CN103911293A | Betrysophila dorthea | Taxus chinensis | Strain with a high paclitaxel yield and method for producing paclitaxel. | [119] |
| CN103966109A | Aspergillus fumigatus | Schizandra chinensis fruit | Endophytic fungus that is capable of producing protocatechuic aldehyde (79). | [120] |
| CN104031948A | Daedonia eschscholtzii | Ginkgo biloba | Preparation of a spiro-dinaphthalene compound (83). | [124] |
| CN104059044A | Trichoderma sp. | Mangrove | Preparation of a xanthine derivative (82) as a microbial pesticide and fungicide. | [122] |
| CN104073529A | Not disclosed | Taxus x media seed | Preparation of taxol. | [123] |
| CN104086522A | Lasiodiplodia | Camptotheca acuminata | Preparation and dyeing of red pigment haematocrome. | [125] |
| CN104096991A | Not disclosed | Ginkgo biloba | Preparation of taxol. | [123] |
| US20140082271A | Not disclosed | Lobatis fraseri or Nothofagus cunninghamii | Isolation of antibiotic compound. | [126] |
| CN10425678A | Cladosporium cladosporioides | Fumigatus sp. | Production of forsythoside A (84), forsythoside B (85), and forsythin (86) and their applications. | [127] |
| CN104357525A | Acremonium dichromosporum | Clyropyricia sp. | Production of glycyrrhetinic acid by using microbial fermentation. | [128] |
| CN104450558A | Not disclosed | Gardenia jasminoides | Method for isolation and screening of endophytic fungi and for large-scale preparation of high-purity genipin (87). | [129] |
| CN104450331A | Fusarium tricinctum | Fritillaria cirrhosa | Obtains pemminine (88) and peimimisine (89) alkaloids. | [130] |
| CN104593443A | Botrysophila rhodina | Aquilaria sinensis | Preparation of aglawood chromone (90–94) components. | [131] |
| CN104726345A | Mixture of fungi including | Taxus spp. | High production of baccatin III. | [132] |
| CN10476728A | Not disclosed | Gastrodia elata | Preparation of gastrodin (95). | [133] |
| CN104774771A | Aspergillus fumigatus | Glycyrrhiza sp. | Production of pseutorin A (96) as a food preservative. | [134] |
| CN104789613A | Alternaria sp. | Spiraea salicifolia | Extraction and separation of bacteriostatic component (97) from fermentation broth. | [135] |
| CN104805017A | Fusarium solani | Penicillia sp. | Generation and application of β-glucosidase. | [136] |
| CN104877910A | Alternaria sp. | Nothapodytes pittosporoides | Preparation of breveldin (98). The compound has antifungal and insecticide activity and is an ideal veterinary and agriculture candidate drug. | [137] |
| CN105009173A | Mortierella sp. | Huperzia serrata | Preparation of taxol. | [123] |
| CN105009174A | Fusarium sp. | Paonia sp. | Production of paenol (99). | [139] |
| CN105009175A | Talaromyces sp. | Paonia sp. | Production of paenol. | [140] |
| CN105009176A | Fusarium sp. | Paonia sp. | Production of paenol. | [141] |
| CN105200091A | Geomyces sp. | Nerium indicum | Preparation and application of ethyl vincamine (100). | [142] |
| US20150073048A | Aspergillus fumigatus | Ananas ananassoides | Preparation of antimicrobial composition and methods of use. | [143] |
| WO20150928869A | Trichoderma longibrachiatum | Bousselia serrata | Preparation of brachatin D (101). | [144] |
| CN105238697A | Chaetomium sp. | Paonia sp. | Production of baccatin with endophytic fungus from peony. | [145] |
| CN105238700A | Epicoccum nigrum | Wild soybean | High yielding oleonic acid endophyte. | [146] |
| CN105274005A | Aspergillus fumigatus | Taxus x media | Taxol production. | [147] |
| CN105316238A | Trichoderma sp. | Taxus chinensis | Method for culturing and screening taxol-producing fungus. | [148] |
| CN105349431A | Phoma glomerata | Salvia miltiorrhiza | Generation and application of salvianolic acid C (102). | [149] |
| CN105488024A | Fusarium mautre | Taxus x media/Vilateria jatamansi | Increases the yield of paclitaxel in an endophytic fungus fermentation product. | [150] |
| CN105507896A | Phoma glomerata | Salvia miltiorrhiza | Generation of ergosterol (103). | [151] |
| CN10550821A | Aspergillus sp. | Not disclosed | Preparation of taxol-containing culture. | [152] |
| CN10562040A | Mucor racemosus | Huperzia serrata | Application of a fungal strain with highly efficient expression of huperzine A. | [153] |
| CN10563861A | Chaetomium globosum | Cajanus cajan | Application of a fungal strain with a high yield of flavin. | [154] |
| CN105925546A | Phomopsis liquidambari | Magnoflorus | Preparation method for cytochalasin H (104). | [155] |
| CN106010990A | Phoma glomerata | Acrida cinerea | Strain capable of producing of perlolyrine (105) and a method for preparation. | [156] |
| CN106047715A | Trichoderma sp. | Nothapodytes pittosporoides | Extraction of camptothecin. | [157] |
| WO2016034751A | Aspergillus fumigatus | Schisandra chinensis | Production of taxol. | [123] |
| Patent No.       | Endophyte                  | Host | Patent Details                                                                 |
|------------------|---------------------------|------|--------------------------------------------------------------------------------|
| CN106433416A     | Penicillium citrinum      |     | Preparation of penicillins (107–113) as antibiotic drugs.                        |
| CN106343361A     | Aspergillus niger         |      | Preparation of indane derivatives (114–115).                                   |
| CN106497803A     | Fusarium verticillioides  |      | Fungal strain with hyperzine A-producing function and its use in the biosynthesis |
|                  |                           |      | of medicine for treating Alzheimer's disease and vascular dementia.             |
| CN10668984A      | Fusarium oxysporum        |      | Production of hyperzine A and its application in the treatment of dementia.     |
| CN10658894A      | Neocrypta sp.             |      | Preparation of compound (116) derived from Tibetian medicine endophytic fungi. |
| CN106662647A     | Not disclosed             |      | Fermentation extraction of azadirachtin (117).                                |
| CN106701994A     | Neoclynostrum sp.         |      | Production of pyrocide A (118) and pyrocide B (119).                           |
| CN106946955A     | Pezicula sp.              |      | Production of mycosubaric compounds (120–124) that aid in the preparation of   |
|                  |                           |      | drugs for preventing and controlling plant fungal disease.                     |
| CN10696722A      | Aspergillus flavus        |      | Paclitaxel production.                                                         |
| CN10696762A      | Aspergillus niger         |      | Production of taxane compound baccatin III.                                   |
| CN106783856A     | Nigrospora sphaerica      |      | Production of large amounts of bostrycin (125).                               |
| CN107054145A     | Pestalotiopsis visnate    |      | In vitro production of nucleosides, preferably, adenosine, guanylyl, uridine,  |
|                  |                           |      | and inosine.                                                                    |
| CN107098118A     | Aspergillus aculeatus     |      | Efficient taxol producing endophytic fungi.                                    |
| CN107138972A     | Epicoccum nigrum          |      | Endophytic fungus capable of generating pectin through liquid fermentation.    |
| CN107129936A     | Penicillium sp.           |      | Production of paclitaxel.                                                      |
| CN107254504A     | Fusarium sp.              |      | Increasing the scutellarin (126) content with microbial agents.               |
| CN107354182A     | Purpureocillium llacatum  |      | Preparation of (±)-4-benzyl-2-oxazolidinone (127) by fermentation.           |
| WO2017049353A    | Daldinia sp.              |      | Production of volatile organic compounds as insecticidal and antifungal agents.|
| WO2017066233A    | Stemphyllum solani        |      | To obtain compounds (128–129) for use as biocides.                            |
| CN107066817A     | Ascomycota sp.            |      | Production of ascomylactam compounds (130–131).                               |
| CN107072345A     | Fusarium sp.              |      | Endophytic fungi and application in the steroids saponin diosgenin (132) and   |
|                  |                           |      | ruscogenin (133).                                                             |
| CN107072346A     | Penicillium oxalicum      |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN107072347A     | Cladosporium sp.          |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN107072348A     | Penicillium sp.           |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN1070729716A    | Penicillium sp.           |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN1070729717A    | Schizopyllum sp.          |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN1070797918A    | Aspergillus sp.           |      | Endophytic fungi and application in the steroid saponin diosgenin and ruscogenin.|
| CN107095579A     | Aspergillus niger         |      | Application in the preparation of the steroid saponin diosgenin and ruscogenin.|
| CN108264473A     | Penicillium decumbens     |      | Preparation of 8α, 15α-epoxy-huperzine A, which has a curative neuroprotective effect.|
| CN108277164A     | Diaporthe sp.             |      | Preparation of the steroid saponin.                                           |
| CN108388831A     | Aspergillus tabungensis    |      | Indene derivative (136) that aids in the preparation of an anti-inflammatory drug.|
| CN108667989A     | Trichoderma asperellum    |      | Production of furance (137) derivative with good antibacterial activity.        |
| CN10858636A      | Aspergillus tabungensis    |      | Preparation of diketopiperazine compound (138), which has antibacterial application.|
| CN10866497A      | Dalania aschdoltii        |      | Extraction method and application of a bicoumarin derivative (139).            |
| CN108723867A     | Phoma sp.                 |      | Preparation and application of polyketides (140–141).                          |
| CN108913731A     | Pestalotiopsis sp.        |      | Preparation of antibacterial compounds (142–143).                              |
| CN10902445A      | Fusarium proliferatum     |      | Preparation and application of pestalotistone M (144) which has                |
|                  |                           |      | immunosuppressive activity.                                                    |
| CN10906056A      | Aspergillus flavus        |      | Preparation of bisabolane sesquiterpene compounds (154–155) as anti-type II diabetes mellitus drugs. |
| IN201641023516A  | Phomopsis sp.             |      | Method of producing colchicine (156) from an endophyte using epigenetic modifiers.          |
| IN201721003410A  | Phoma sp.                 |      | Isolation, fermentation, purification, and characterization of the antibacterial compound 2'-hydroxyoxygenstein (157). |
Table 1. Cont.

| Patent No. | Endophyte | Host | Patent Details | Ref. |
|------------|-----------|------|----------------|-----|
| CN109776561A | Cytopora rhizophorae | Morinda officinalis | Preparation of cytozolin B (200) and C (201) that are applied in the preparation of antitumor drugs. | [216] |
| CN109810906A | Bionectria pityrodes | Tammarix sp. | Preparation of phenolic acid compound (202) through fermentation. | [219] |
| CN109968668A | Penicillium notatum | Gastrodia elata | Fermentation and production of penicilllic acid (182). | [215] |
| CN110053428A | Trichoderma asperellum | Seaweed | Preparation of 5,8-peroxyde of ergosterol. | [218] |
| CN110053053A | Trichoderma asperellum | Seaweed | Production of dihydroxyquinone C (206) that is used as an antineoplastic drug for treating human pulmonary squamous carcinoma and breast carcinoma. | [219] |
| CN110093363A | Alterania sp. | Polysomyx corymbosa | Preparation of compound alterlactone (207) that is used as a disinfectant in agriculture. | [220] |
| CN110218020A | Pseudopithomyces sp. | Sonnentidae ceauslaris | Preparation of desippeptide compound (208). | [221] |
| CN11029127A | fungal strain TGM112 | Mangrove | Preparation of butyrolactone compounds (209–211). | [222] |
| CN110257255A | Dalinia escholzitzi | Mangrove | Preparation of chrome derivatives (212–216). | [223] |
| CN110257260A | Boeremia exigua | Atractylodes macrocephala | Preparation of the Atractylodes lactones I (217) and II (218). | [224] |
| CN110272928A | Colletotrichum boninense | Hupera serrata | New microbe resource for the production of hupizine A industrial fermentation. | [225] |
| CN110285728A | Dalinia escholzitzi | Mangrove | Preparation of tetraedrine derivatives (219–223). | [226] |
| CN1102915116A | Aspergillus sp. | Tammarix sp. | Production of a variety of fatty acids and their application. | [227] |
| CN110302215A | Penicillium sp. | Taxus x media | Fungal crude extract, it's applications, e.g., as being a source of paclitaxel analog. | [228] |
| CN110438015A | Aspergillus miamrii | Citron orange fruit | Fungal strain its fermentation to produce hesperidinase. | [229] |
| CN110438018A | Acremonium pilosum | Malania sp. | Preparation of fusidic acid (224). | [230] |
| CN110518876A | Aspergillus fumigatus | Koken Epimedium | The culture method of this fungal strain and its metabolites epimers A–C (225–227). | [231] |
| CN110537040A | Fusarium oxysporum | Edgeworthia chrysanth/| Methods for preparation and application of alpha-pyrone (228). | [232] |
| CP0271200132A | Aspergillus japonicus | Achyranthes aspera | Production of the novel antibacterial compound fraxidin (229). | [233] |

1 Some patents just provided a common name for the host organism.
3.2. Biotransformation by Endophytic Fungi

Biotechnological processes enable the production of useful molecules with a decrease in the generation of pollutants, reducing the use of solvents and reagents, minimizing the consumption of energy, and providing a way to obtain active compounds with greater specificity and efficiency. The use of endophytic fungi in biotechnological processes, such as biotransformation, is in its early stages of development and has some limitations [238]. However, there have been some reports of fungi that have been used in biotransformation [239–242].

Table 2 lists a group of patents that illustrate the efforts toward using endophytic fungi to obtain molecules of biological importance such as the ginsenosides [243] and glycyrrhetinic acid monoglucuronide [244].

Fungi from the genera Absidia, Zygorhynchus, Xylaria, and Fusarium have been patented to obtain ginsenoside Rd by the transformation of ginsenoside Rb1. Fungi from the genera Microsphaeropsis, Aspergillus, and Chaetomium have been patented for the biotransformation of glycyrrhizinic acid into glycyrrhetinic acid monoglucuronide.
Table 2. Endophytic fungi applied for biotransformation.

| Patent No. | Endophyte | Host          | Patent Details                                                                                                                                                                                                 | Ref. |
|------------|-----------|---------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|------|
| CN102080348A | *Absidia glauca* | *Panax gingsen* | Conversion of ginsenoside Rb1 (230) to prepare ginsenoside Rd (231).                                                                                                                                     | [245]|
| CN102080349A | *Zygophyllum moelleri* | *Panax gingsen* | Preparation of ginsenoside Rd from ginsenoside Rb1.                                                                                                                                                       | [246]|
| CN102154123A | *Fusarium sp.* | *Dioscorea nipponica* | Biotransformation conversion conditions of diosgenin saponins.                                                                                                                                             | [247]|
| CN102199548A | *Penicillium exiguum* | *Polygonum cuspidatum* | Microbial transformation of resveratrol from polydatin (232).                                                                                                                                             | [248]|
| CN10221486A | *Penicillium exiguum* | *Polygonum cuspidatum* | Conversion of polydatin into resveratrol.                                                                                                                                                                  | [249]|
| CN102363350A | *Penicillium sp.* | *Not disclosed* | Biotransformation of raisin extract. Preparation and application in flavoring.                                                                                                                             | [250]|
| CN10275443A | *Penicillium purpurogenum* | *Dipsacus sp.* or *Sabina vulgaris* | Separation and purification method for bioconversion of podophyllotoxin into sulfur-substituted derivatives.                                                                                             | [251]|
| CN103695476A | *fungal strain L1 CCMCC No. 4558* | *Not disclosed* | Conversion of polydatin to resveratrol.                                                                                                                                                                   | [252]|
| CN103981104A | *Microphallus arundinis* | *wild rice* | Biotransformation of glycyrrhizic acid (233) into liquiritin (234).                                                                                                                                       | [253]|
| CN103992953A | *Aspergillus flavus* | *wild rice* | Transform glycyrrhizic acid into glycyrrhetinic acid monoglucuronide.                                                                                                                                       | [254]|
| CN106591142A | *Xylariales sp.* | *Not disclosed* | Conversion of Panax notoginseng saponin to prepare vina-ginsenoside R13 (236), notoginsenoside J (237) and American saponin ginseng L16 (238).       | [255]|
| CN106893677A | *Fusarium sp.* | *Herba Andrographis* | Conversion of glycyrrhizic acid into glycyrrhetinic acid monoglucuronide.                                                                                                                                    | [256]|
| CN109536561A | *Fusarium oxysporum* | *Gentiana sp.* | Transformation of andrographolide diterpenoids (239–242).                                                                                                                                             | [257]|
| CN109731270A | *Fusarium proliferatum* | *Cajanus cajan* | Conversion of gentiopicroside (243) into two separate compounds with hepatoprotective activity.                                                                                                           | [258]|
| CN108705553A | *Plectosphaerella cucumerina* | *Huperzia serrata* | Efficient conversion of androstenedione to testolactone and androstone diene diketone.                                                                                                                    | [259]|
| CN10936561A | *Fusarium oxysporum* | *Not disclosed* | Conversion of ginsenoside Rb1 into ginsenoside Rd and its application.                                                                                                                                      | [260]|
| CN110527632A | *Phomopsis sp.* | *Not disclosed* | Bioconversion of betulinic acid (245).                                                                                                                                                                   | [261]|
| CN110423469A | *Lasiodiplodia pseudotheobromae* | *Illicium verum* | trans-trans-Anethole (246) conversion to generate different vanillic acids (247).                                                                                                                        | [262]|
| WO2019024295A1 | *Oxotropia truncata* | *Curcuma sp.* | Microbial bioconversion of curcuminoinds to caletine A (248).                                                                                                                                            | [263]|
| WO2019070219A2 | *Alternaria euryceras* | *Astragalus condensatus, A. angustifolius* | Production of a telomerase activator, biotransformation with endophytic fungi to obtain new/molecular molecules from the saponins from natural sources and method for discovery molecules that increase telomerase enzyme activation. | [264]|

1 Some patents just provided a common name for the host organism.
4. Discussion

The study of endophytic fungi as a source of bioactive secondary metabolites has its first beginnings in 1993 with the discovery of taxol [4], until then, the primary sources of active natural molecules were isolated mainly from plants [266]. About two decades ago, the study of endophytic fungi as producers of active molecules has been emphasized due to obtaining compounds originally produced by plants or due to the production of novel secondary metabolites [11,267]. Thus, fungi from genus Aspergillus, Fusarium, Penicillium and Pestalotiopsis has been recognized as producers of anticancer compounds and having pharmaceutical potential [12,268]. It is estimated that only around 1% of the microorganisms have been cultivated, and within this groups, endophytic fungi corresponded to the least studied [269].

Through this review, we have demonstrated the wide number of endophytic fungi involved in the development of methods and techniques for the application of isolation and fermentation to obtain secondary metabolites with high potential and applications in biomedicine, agriculture, and biotechnology processes. Figure 2 shows the number of patents registered for secondary metabolites and biotransformation processes through endophytic fungi for the period from 2001 to 2019. We found 224 patents related to secondary metabolites and 21 patents related to biotransformation. Aspergillus, Fusarium, Trichoderma, Penicillium, and Phomopsis were the most representative genera for secondary metabolites.

![Figure 2. Number of registered patents from 2001 to 2019 linked to endophytic secondary metabolites and biotransformation processes through endophytic fungi.](image)

Fusarium and Penicillium were the most commonly registered endophytic fungi genera among the 21 patents reviewed for biotransformation processes. Figure 3 shows the number of patented genera. The most notable applications patented were antimicrobial, antibacterial, anticancer, and those related to neurodegenerative diseases. For biotransformation processes, the conversion of ginsenosides and glycyrrhizinic acid were the most patentable applications due to their importance and potential in the pharmaceutical and food industries.

Tables 1 and 2 showed that the majority of the endophytic fungi were derived from plants, but we could also find patents where the host was soft corals or insects.
Figure 3. Number of patents reported for various endophytic fungi by genera.

The global market for compounds like taxol is expected to reach USD $99 million by 2021 [270], and for resveratrol, the projected growth from 2018 to 2028 in revenue terms is 8.1% from USD $97.7 million [271]. Under the objectives of the 1992 Convention on Biological Diversity for the sustainable use of its components and the Nagoya Protocol on Access to Genetic Resources and the Fair and Equitable sharing of benefits derived from the use of genetic resources [272], endophytic fungi and their derived compounds could open a new set of industries and economics in development countries with high biodiversity for the low-cost yield of high-profit molecules that can be applied in the fields discussed in this review.

**Supplementary Materials:** The following are available online at http://www.mdpi.com/2309-608X/6/2/58/s1, Figure S1: Structures of the secondary metabolites listed in Tables 1 and 2.

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