Aspergillus and *Penicillium* Species in Biodegradation of Pesticides

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Since filamentous fungi of *Penicillium* and *Aspergillus* genera can colonize very diverse niches, and Ascomycota seems to be the dominant phylum within the microbial group in various contaminated substrates, they possess great potential in the remediation of pesticide-contaminated sites. Different species can remove the pesticides at different rates, and to various extents; however, the fungal ability to resist high concentrations of pesticides is almost unparalleled compared to other microbial groups. Their performance may be further improved by applying indigenous strains isolated from pesticide-contaminated soils and sediments.

biotransformation filamentous fungi organochlorine organophosphorus pesticides

1. Introduction

The current agricultural practice is contingent on the use of various pesticides since there is an urgent need to enhance the crop production to supply the rapidly increasing food-demand [1]. Throughout the globe, approximately 2 million tons of pesticides are produced and utilized annually, and it was estimated that in 2020, the global pesticide usage was approximated to be 3.5 million tons [2]. The application of pesticides is apparently advantageous since it diminishes the crop infestations, and thus, limits the harvest losses and positively affects the crops quality [3]. However, due to their potent biological activity as toxins and owing to their extensive or injudicious application, the heavy soil treatment with pesticides can endanger the wildlife. The pesticides and their toxic degradation products can enter the plant tissues and build up in the food chain or remain in the soil and water environments and negatively affect the soil fertility and water quality [4]. Thus, the pesticides pose a significant risk to the environment and to human health. The alarming increase in the production and usage of pesticides triggered the notion to reduce the impact of pesticides and find sustainable alternative solutions to protect the crops [5].

Although the role of many pesticides in the environmental deterioration has not been adequately resolved, it is indisputable that they have adverse effects on various non-target organisms [6]. Thus, the research on advanced practices protecting wildlife, which highlights a more cautious use of synthetic agrochemicals, careful risk assessment, and licensing, is very much needed [7]. Therefore, the development of ecofriendly technologies focused on reducing the utilization of synthetic pesticides, especially those with high persistence in the environment, has been addressed [8][9]. More importantly, there is still an issue of developing a proper treatment technology for the remediation of pesticide-contaminated soils. This is a complex problem, since the areas with point-source contamination are usually agricultural soils whose properties should be maintained; thus, aggressive technologies should be omitted [10]. Among more recent and ecologically acceptable emerging technologies is

bioremediation, which involves the utilization of indigenous microflora, adapted or genetically engineered microorganisms or their enzymes for the degradation and conversion of pesticides to another form via co-metabolism or mineralization [11][12].

2. Degradation of Organochlorine Pesticides by *Aspergillus* and *Penicillium* Species

The fungal performance in the biodegradation of the most common organochlorine pesticides (endosulfan and lindane) in culture media, as well as the transformants or end products (**Figure 1**) formed by the fungi belonging to *Aspergillus* and *Penicillium* genera are listed in **Table 1**.

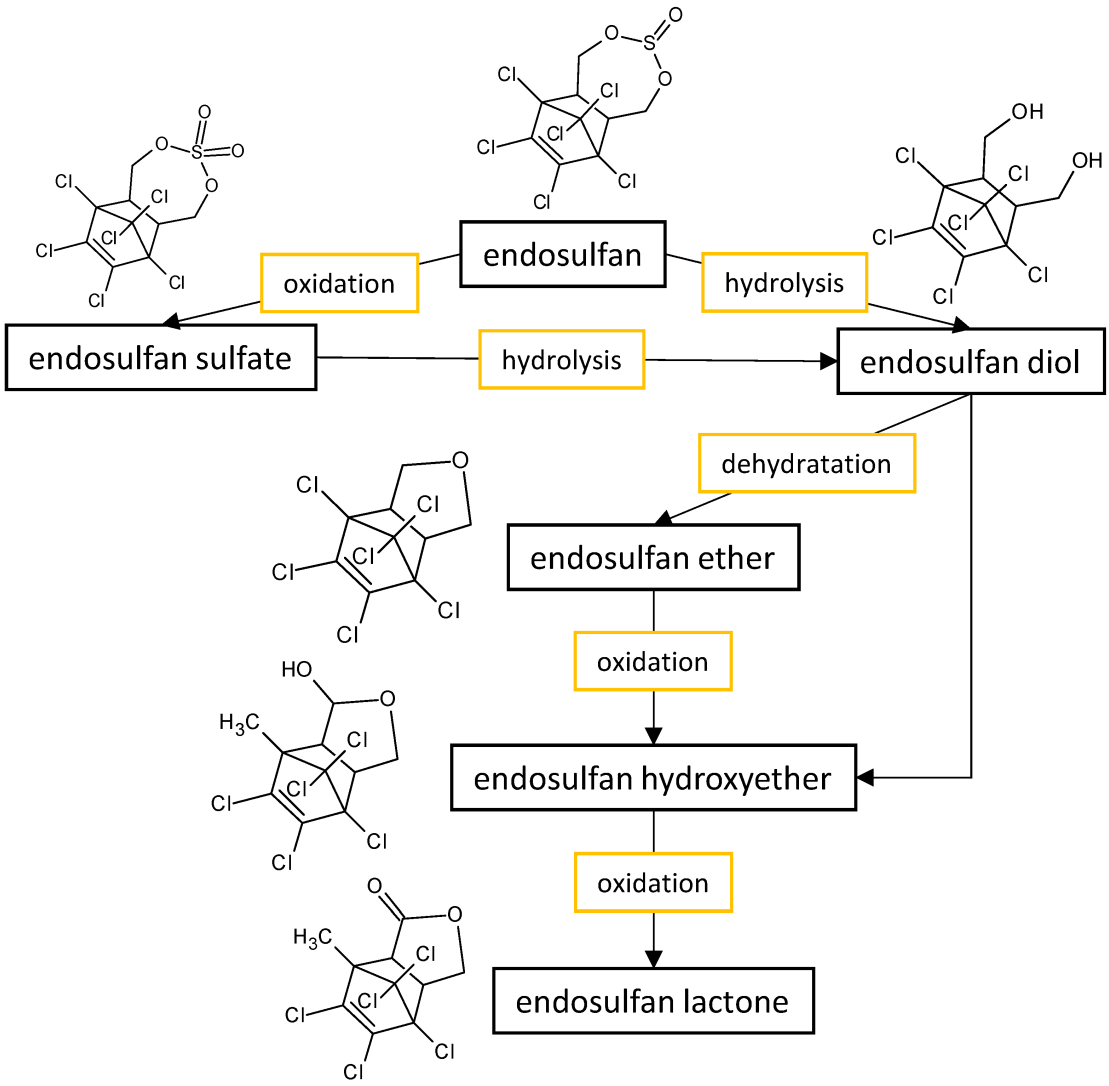

Figure 1. Proposed metabolic pathway for the degradation of endosulfan by *Aspergillus* and *Penicillium* fungal strains.

Table 1. Cultivation conditions and reported performances of filamentous fungi belonging to the genera *Aspergillus* and *Penicillium* in the biodegradation of organochlorine pesticides.

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
Penicillium sp. CHE23	94.8%	modified mineral salt medium with ~57 mg·L^{-1} of **endosulfan** as carbon source incubated for 144 h at 30 °C and 150 rpm	not reported	Romero-Aguilar et al. [13]
Aspergillus niger	complete degradation	Czapek–Dox broth with 350 mg·L^{-1} of β-**endosulfan** source incubated for 120 h at 30 °C and 120 rpm	endosulfan sulfate being most persistent metabolite; after 120 h, complete mineralization was suggested	Bhalerao and Puranik [14]
Aspergillus niger ARIFCC 1053	complete degradation	Czapek–Dox broth with 1000 mg·L^{-1} of technical-grade **endosulfan** incubated for 7 days at 30 °C and 120 rpm	complete mineralization	Tejomyee [15]
Aspergillus niger	98.6%	Czapek–Dox broth with 15.4 µg·g^{-1} of technical-grade **endosulfan** incubated for 15 days at 30 °C and intermittent shaking	not reported	Mukherjee and Gopal [16]
Aspergillus terreus, (*Cladosporium oxysporum*)	91.5%, (89%)	potato dextrose broth with 1.89 µg·g^{-1} of technical-grade **endosulfan** incubated for 15 days at 25 °C	trace concentrations of endosulfan sulfate were detected during incubation; no end products were reported	Mukherjee and Mittal [17]
Aspergillus terricola, *Aspergillus terreus*, (*Chaetosartorya stromatoides*)	~90%	non-sulfur medium enriched with 100 mg·L^{-1} of α or β **endosulfan** isomers incubated for 12 days at 30	major metabolic products being endosulfan diol and endosulfan ether	Hussain et al. [18]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
		°C and 150 rpm (pH 6)		
Aspergillus niger AE	complete degradation (0.1% endosulfan), and 76% (0.5% endosulfan)	Czapek–Dox broth spiked up to 5000 mg·L^{-1} (0.5%) of **endosulfan** incubated for 8 days at 30 °C and 180 rpm (optimum at pH 4)	not reported	Mukhtar et al. [19]
Aspergillus niger, *Aspergillus flavus*, *Penicillium chrysogenum*	77% (AN), 72% (AF), 69% (PC)	potato dextrose broth with 10 mg·L^{-1} of **endosulfan** incubated for 35 days at 29 °C	desulphurized transformants of endosulfan, while chlorine atoms remained imperforated	Ahmad [20]
Aspergillus sydoni	95% (α isomer), 97% (β isomer)	Czapek–Dox broth with 100 mg·L^{-1} of α or β **endosulfan** isomers incubated for 18 days at 30 °C and 150 rpm	major metabolic products being endosulfan sulfate	Goswami et al. [21]
Aspergillus tamarii JAS9, (*Botryosphaerialaricina* JAS6)	kinetic analysis shows that 50% of α and β endosulfan isomers were degraded in 1.7 and 2.2 days by JAS9, respectively (4.2 days for 50% reduction of β endosulfan by JAS6)	M1 medium with 1000 mg·L^{-1} of technical-grade **endosulfan** as carbon source incubated for 10 days at 30 °C and 120 rpm	not reported	Silambarasan and Abraham [22]
fungal consortium (*Botryosphaeria laricina* JAS6, *Aspergillus tamarii* JAS9 and *Lasiodiplodia* sp. JAS12)	complete degradation (a 50% degradation calculated on the 3rd day of incubation)	M1 medium with 1000 mg·L^{-1} of technical-grade **endosulfan** as carbon source incubated for 120 h at 30 °C and 120 rpm	complete mineralization	Abraham and Silambarasan [23]
Penicillium camemberti	70%	acetate-free basal medium supplemented with	not reported	Taşeli [24]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
		1 mM **lindane** incubated for 120 h at 25 °C and 80 rpm (pH 5)		
Aspergillus fumigatus	complete degradation	**lindane** initial concentration is not indicated; medium (Sabouraud dextrose broth or Nutrient broth) incubated for 5 days at 25 °C	not reported	Kumaravel et al. [25]
Penicillium miczynskii CBMAI 93	90%	culture medium of artificial salt water supplemented with 50 mg·L^{-1} of **dieldrin** incubated for 14 days at 32 °C and 130 rpm	no intermediate degradation products were detected, suggesting dieldrin mineralization of conjugation	Birolli et al. [26]

3. Degradation of Organophosphorus Pesticides by *Aspergillus* and *Penicillium* Species

The performances of strains belonging to *Aspergillus* and *Penicillium* genera in the biodegradation of organophosphorus pesticides in culture media are listed in **Table 2**. The summarized biodegradation pathways for methyl parathion and chlorpyrifos, the most studied organophosphorus pesticides, are depicted in **Figure 2** and **Figure 3**, respectively.

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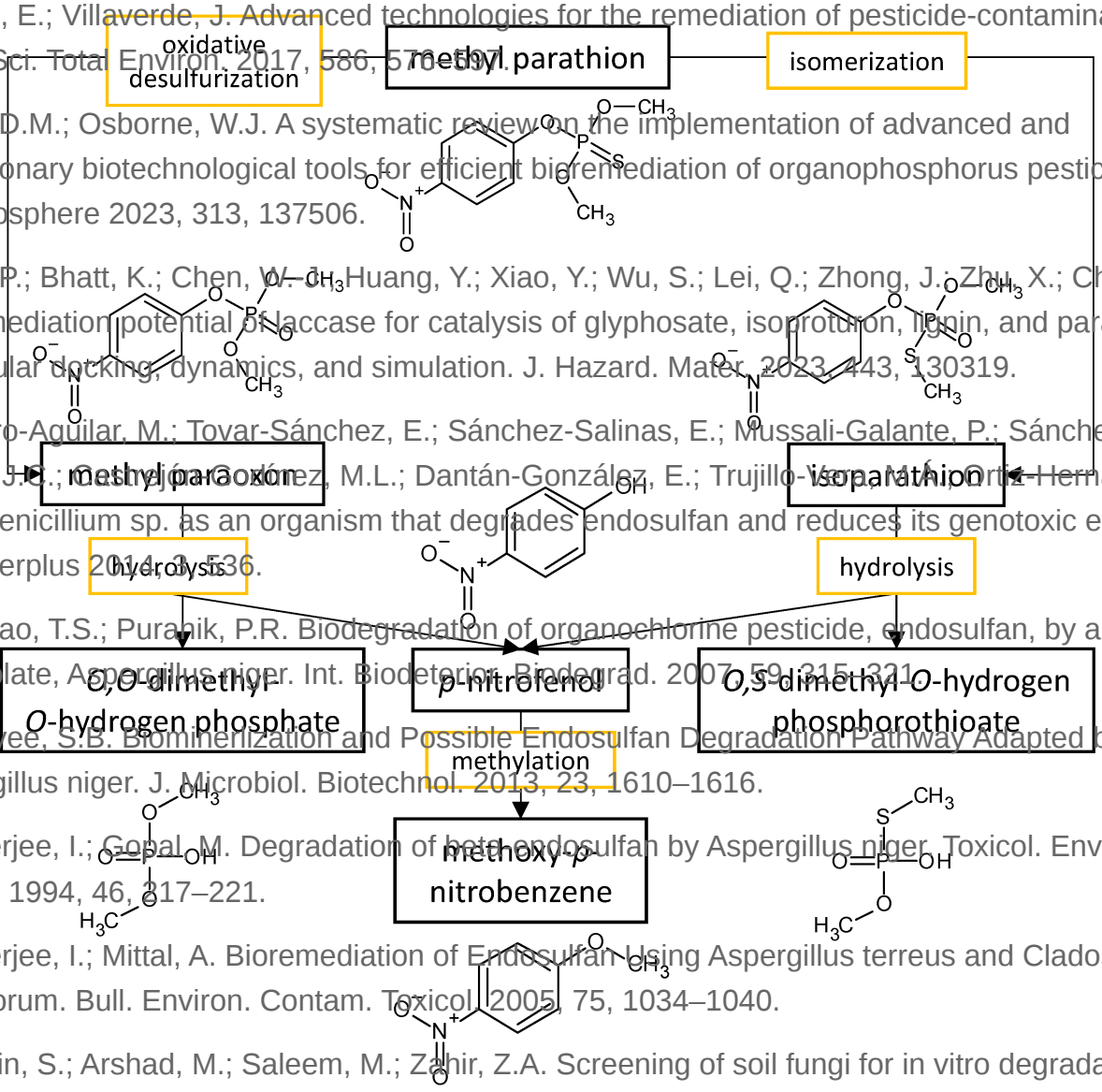

Figure 2. Proposed metabolic pathway for the degradation of methyl parathion by *Aspergillus* and *Penicillium* fungal strains.

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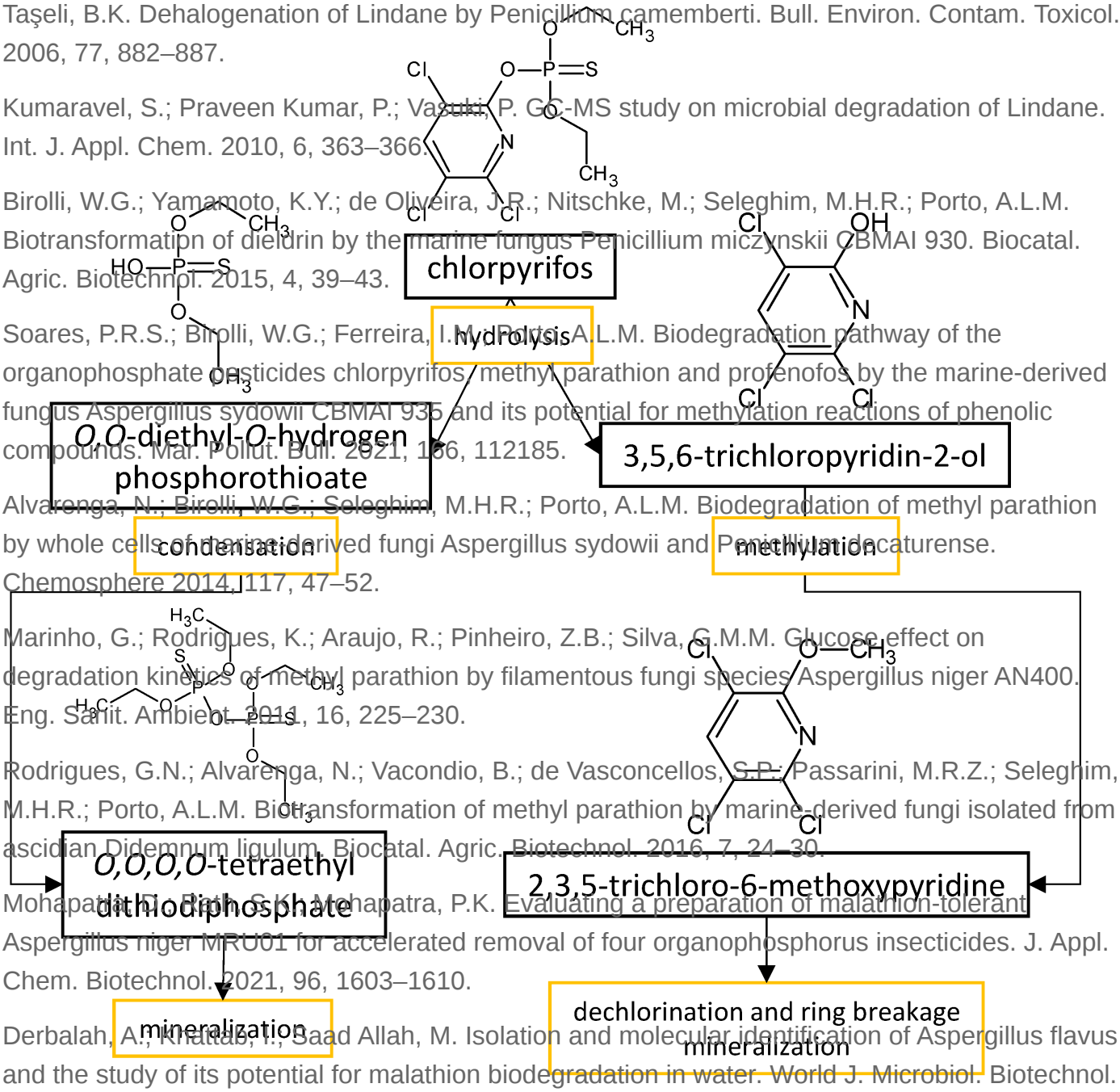

Figure 3. Proposed metabolic pathway for the degradation of chlorpyrifos by *Aspergillus* and *Penicillium* fungal strains.

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Table 2. Cultivation conditions and reported performances of filamentous fungi belonging to the genera *Aspergillus* and *Penicillium* in the biodegradation of organophosphorus pesticides.

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
Aspergillus sydowii CBMAI 935	32%, 80% and 52% of chlorpyrifos, methyl	2% malt liquid medium with 50 mg·L^{-1} of **chlorpyrifos,**	chlorpyrifos degradation resulted in tetraethyl dithiodi-phosphate and 2,3,5-	Soares et al. [27]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
	parathion, and profenofos, respectively	**methyl parathion** or **profenofos** incubated for 30 days at 32 °C and 130 rpm	trichloro-6-methoxypyridine; methyl parathion hydroloyzed into methylated phosphate and phosphorothioates, and 1-methoxy-4-nitrobenzen; profenofos degraded into 4-bromo-2-chloro-1-methoxybenzen and *O*,*O*-diethyl-*S*-proyl phosphorothioates	
Aspergillus sydowii CBMAI 935, *Penicillium decaturense* CBMAI 1234	80%	liquid mineral medium supplemented with KNO_3 and 100 mg·L^{-1} of **methyl parathion** incubated for 30 days at 32 °C and 130 rpm	*p*-nitrophenol	Alvarenga et al. [28]
Aspergillus niger AN400	2% (glucose-free treatment, initial concentration of methyl parathion was 19.1 mg·L^{-1}), 43% (glucose-treated medium, initial concentration of methyl parathion 24.9 mg·L^{-1})	glucose-free or glucose treated distilled water supplemented with Vishniac solution and up to 24.9 mg·L^{-1} of **methyl parathion** incubated for 27 days at 30 °C and 200 rpm	not reported	Marinho et al. [29]
Penicillium citrinum, *(Fusarium proliferatum)*	complete degradation (the biotic control had the same degradation efficiency)	3% malt liquid medium with 30 mg·L^{-1} of **methyl parathion** incubated for 30 days at 32 °C and 130 rpm	not reported	Rodrigues et al. [30]
Aspergillus niger MRU01	70%, 54%, 58%, and 68% of malathion, parathion, chlorpyrifos and	Czapek–Dox broth spiked with 500, 470, 260 and 680 µmol·L^{-1} (0.5%) of **malathion**, **parathion**,	not reported	Mohapatra et al. [31]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
	dimethoate, respectively	**chlorpyrifos,** and **dimethoate**, respectively, incubated for 5 days at 26 °C and 120 rpm (optimum at pH 4)		
Aspergillus flavus	complete degradation	mineral salt medium supplemented with 5 $mg \cdot L^{-1}$ of **malathion** incubated for 36 days at 30 °C (pH 7) on a rotatory shaker (optimized conditions)	not reported	Derbalah et al. [32]
Aspergillus sp. F1	over 90% (89% at an inlet load of 180 $mg \cdot L^{-1} \cdot d^{-1}$)	bioreactor supplemented with 300 $mg \cdot L^{-1}$ of **chlorpyrifos** as the sole carbon source incubated at 28 °C (pH 7) with dissolved oxygen concentration of 5.8 $mg \cdot L^{-1}$ (optimized conditions)	not reported	Yadav et al. [33]
Aspergillus fumigatus	99%	potato dextrose broth supplemented with **chlorpyrifos** (10%) incubated for 9 days at 25 °C (pH 7) and 180 rpm	not reported	Anggreini et al. [34]
Aspergillus fumigatus	95.9%	potato dextrose broth with **chlorpyrifos** (1.5%) incubated for 5 days at 25 °C (pH 7) and 180 rpm	not reported	Anggreini et al. [35]
Aspergillus terreus JAS1	complete degradation (after 24 h)	M1 medium supplemented with 300 $mg \cdot L^{-1}$ of **chlorpyrifos** as the sole carbon source incubated for 96 h	3,5,6-trichloropyridin-2-ol that was completely degraded after 48 h; no other metabolites were reported	Silambarasan and Abraham [36]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
		at 30 °C and 120 rpm		
Aspergillus oryzae strains AM1 and AM2	73% (AM1), 50% (AM2)	Czapek–Dox broth spiked with 20 mg·L^{-1} of **chlorpyrifos** incubated for 30 days at 25 °C and 60 rpm (optimum at pH 4)	not reported	Barberis et al. [37]
Aspergillus viridinutans, *Penicillium implicatum*	44.6% (*A. viridinutans*), 16.2% (*P. implicatum*)	potato dextrose broth with 20 mg·L^{-1} of **chlorpyrifos** incubated for 14 days at 28 °C	not reported; high losses of chlorpyrifos from culture medium were due to abiotic hydrolysis	Abdel-Wareth and Abd El-Hamid [38]
Aspergillus niger, *(Trichoderma viride)*	72.3%, (95.7%)	Czapek–Dox broth spiked with 1.25 mg·L^{-1} of **chlorpyrifos** incubated for 14 days at 30 °C and intermittent shaking (pH 6.8)	not reported; high losses of chlorpyrifos from culture medium were due to abiotic hydrolysis	Mukherjee and Gopal [39]
Penicillium citrinum, *Aspergillus niger*, *Aspergillus oryzae*	25.9% (*P. citrinum*), 64% (*A. niger*), 50.8% (*A. oryzae*)	Burkes mineral broth with 10 mg·L^{-1} of **chlorpyrifos** incubated for 15 days at 27 °C without shaking (pH 7.2)	not reported; high losses of chlorpyrifos from culture medium were due to abiotic hydrolysis	Abd-Alrahman and Mostafa [40]
Aspergillus niger	~80%	soil extract medium with 10 mg·L^{-1} of **chlorpyrifos** incubated for 30 days at 25 °C and 60 rpm	3,5,6-trichloro-2-pyridinol were detected below the concentration of 1 mg·L^{-1}	Karas et al. [41]
Aspergillus fumigatus, *A. flavus*, *A. niger*, *A. ochraceus*, *A. tamarii*, *A. terreus*, *Penicillium chrysogenum*, *P. brevicompactum*,	phosphor mineralization efficiencies ranged from 4 to 46% (Cyolan), from 9.5 to 26.8% (Malathion),	Czapek–Dox broth spiked with 100 mg·L^{-1} of **cyolan**, **malathion**, and **chlorpyrifos** incubated for 35 days at 28 °C without shaking	not reported; media and biomass were analyzed for phosphor and sulfur that mineralized from the degradation of insecticide	Omar [42]

Fungal Strain	Degradation Efficiency	Cultivation Conditions	Degradation Products	Reference
P. citrinum, *P. funiculosum*	and from 2.3 to 6.7% (Dursban)			
Penicillium oxalicum ZHJ6	complete degradation	mineral salt medium with 1% glucose supplemented with 1 $mg \cdot L^{-1}$ of **methamidophos** as sole nitrogen source incubated for 12 days at 25 °C and pH of 5.0 (most favorable conditions)	inorganic phosphor, CH_3SH, and CH_3OH are hypothesized being formed	Zhao et al. [43]
Aspergillus oryzae A-F02	~13.7%	fermentation medium with 1.5 $g \cdot L^{-1}$ of **glyphosate** incubated for 144 h at 30 °C and 150 rpm	aminomethylphosphonic acid and methylamine, the latter being further degraded	Fu et al. [44]

4. Biotransformation of Pesticide Belonging to Some Other Chemical Groups by *Aspergillus* and *Penicillium* Species

The biodegradation of three pesticides of the different chemical classes, difenoconazole, pendimethalin, and terbuthylazine, by *Penicillium brevicompactum* and *A. oryzae* was studied by Pinto et al. [45]. After an 8-day incubation, 99% of pendimethalin was degraded by both fungal strains. Fungi *A. oryzae* and *P. brevicompactum* were capable of degrading 88% and 92.7% of difenoconazole, while lower removal percentages were exhibited with terbuthylazine approximating 78% and 71%, respectively. More importantly, the authors have highlighted adsorption as a potential mechanism of pesticide removal, characterized by the fast initial removal rates, which may be an efficient mechanism utilized by the fungus to block the xenobiotic uptake.

Derbalah et al. [46] reported that approximately 93% of the initial famoxadone concentration was degraded within four weeks by strains of *A. niger* EB2 and *Penicillium* sp. EB3 and the negligible spontaneous degradation was observed in control experiments. The bioassay conducted with *Alternaria solani* using the spent medium with degradation products collected at the end of famoxadone treatment showed only slight antifungal activity, indicated by 5% growth inhibition of *A. solani*.