Pollutants Biodegradation by Fungi

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Abstract: One of the major problems facing the industrialized world today is the contamination of soils, ground water, sediments, surfacewater and air with hazardous and toxic chemicals. The application of microorganisms which degrade or transform hazardous organic contaminants to less toxic compounds has become increasingly popular in recent years. This review, with approximately 300 references covering the period 2005-2008, describes the use of fungi as a method of bioremediation to clean up environmental pollutants.

1. INTRODUCTION

Pollution of the environment has been one of the largest concerns to science and the general public in the last years. Nowadays, the industrialized world is confronted with the contamination of soils, water sources and air with hazardous and toxic xenobiotics. While regulatory steps have been implemented to reduce or eliminate the production and release to the environment of these chemicals, significant environmental contamination has occurred in the past and will probably continue to occur in the future. The industrialization of agriculture, rapid growth in the chemical industry and the need to generate cheap forms of energy have all caused the continuous release of very organic chemicals into the biosphere. For example, in the United States alone an enormous amount waste is produced annually. In fact, approximately 300 million metric tons of hazardous wastes are produced each year.

Bioremediation is a process by which living organisms degrade or transform hazardous organic contaminants to less toxic compounds [1]. Microorganisms in the indigenous environment have been known to play key roles in the biodegradation of organic compounds. Unlike prokaryotes, eukaryotic fungi have shown diverse metabolic potential resulting in metabolites similar to those produced from mammalian metabolism. These metabolic properties may help us to directly elucidate the metabolic fates of organic compounds occurring in mammalian liver cells instead of using mammalian microsomal fractions or live organisms. Fungal metabolism also provides an easy preparative method for the production of metabolites in large quantity.

The use of fungi as a method of bioremediation provides an option to clean up environmental pollutants. Bioremediation using fungi has drawn little attention in the past two decades since most bioremediation research has focused mainly on the use of bacteria. Nevertheless, recently fungi have received considerable attention for their bioremediation potential which is attributed to the enzymes they produce. In addition, fungi have advantages over bacteria such as fungal hyphae that can penetrate contaminated soil to reach the pollutants [2]. This review, with approximately 300 references covering the period 2005-2008, will highlight the main applications of fungi to the biodegradation of organic contaminants to less toxic compounds in order to clean up environmental pollutants.

2. BIODEGRADATION OF CHEMICAL POLLUTANTS BY FUNGI

The revolutionized development of resources and technologies has produced more chemicals and compounds which has consequently increased the number of compounds identified as being potential environmental threats to living organisms. Pharmaceuticals and personal care products (PPCPs), surfactants, various industrial additives and numerous chemicals are purported to be pollutants. These pose challenges to the designers of future treatment plants and related methodology for their eradication [3].

These pollutants vary greatly in their form and mechanism of action. Thus, the identification and evaluation of these compounds from the environmental matrixes have provided a unique challenge. The methodologies used for degradation include biological and instrumental methods. The new advances in molecular biology and the isolation of new microorganisms from contaminated environments form the basis of bioremediation emerging as a clean and low-cost methodology for the future.

Aromatic Hydrocarbons

The biodegradation of aromatic hydrocarbons by fungi has traditionally been considered to be of a cometabolic nature. Recently, however, an increasing number of fungi isolated from air biofilters exposed to hydrocarbon-polluted gas streams have been shown to assimilate volatile aromatic hydrocarbons as the sole source of carbon and energy. The biosystematics, ecology, and metabolism of such fungi were reviewed, based in part on the re-evaluation of a collection of published hydrocarbon-degrading isolates obtained from authors around the world [4]. For example, the degradation performance of benzene, toluene, styrene and xylene by fungi was widely studied [5, 6]. Moreover, biodegradation of monomeric styrene by *Phanerochaete chrysosporium* KFRI

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1385-2728/09 $55.00+.00

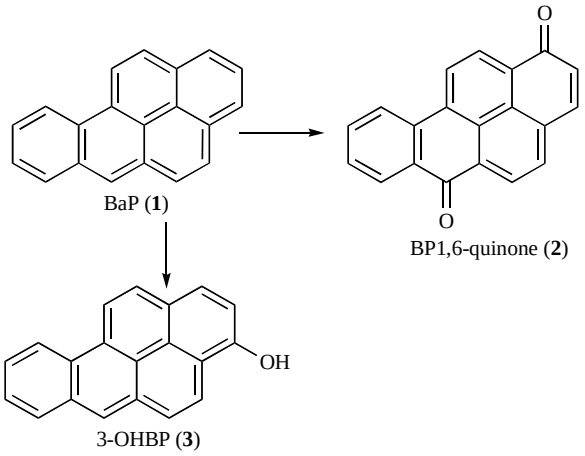
20742, *Trametes versicolor* KFRI 20251, and *Daldinia concentrica* KFRI 40-1 was carried out by Lee *et al.* [7], giving metabolites including 2-phenyl ethanol, benzoic acid, cyclohexadiene-1,4-dione, butanol and succinic acid.

Oil pollution has become an environment problem which has been paid wide attention in the world. Bioremediation technology applied to oil contaminated soil has become an important field in research for its advantages such as low cost, little environmental effect, simplicity and efficiency and has a bright perspective for the future. The main research fields relating to the bioremediation of oil contaminated soil were put forward employing fungi [8] such as *Trichoderma* sp. [9]. Thus, naphthalan petroleum was studied using fungi cultures to obtain degrader strains as *Penicillium* sp. 3n, *Fusarium* sp. 11a, *Cephalosporium* sp. 45a, and *Mucor* sp. 16 [10, 11]. Also, the fungus *Cladosporium* proved to have good aromatic-degrading ability to biodegrade diesel pollution in aqueous solution [12] and crude oil contaminated soils were used to test the degradation ability of *Pleurotus tuber-regium* where reduction of aromatics was appreciable in all the experiments [13]. Moreover, fuel oil A contaminated sites were investigated for degradation by *Fusarium solani* SZFWT02 showing a great biodegradation activity [14].

Polycyclic aromatic hydrocarbons (PAHs) are toxic pollutants that have accumulated in the environment due to a variety of anthropogenic activities. Bioremediation using various microorganisms is one of the approaches tested for the removal of PAHs from the environment. Fungi belonging to the genera *Aspergillus, Penicillium, Paecilomyces, Coriolus, Pycnoporus, Pleurotus, Fomitopsis,* and *Daedalea,* have been found to be responsible for degrading PHAs in soil and aquatic environments [15]. The degradation potential of white rot fungi belonging to the genera *Phanerochaete, Irpex, Polyporus, Stereum, Lentinus, Bjerkandera, Irpex, Pleurotus,* and *Phlebia* to remediate contaminated soils [16-22] is known. The most effective biodegradation of pyrene was obtained with *Coriolus versicolor, Trichoderma* sp., *Aspergillus niger,* and *Fusarium* sp. [1, 23]. Other lignolytic fungi, *Allescheriella* sp. strain DABAC 1, *Stachybotrys* sp. strain DABAC 3, and *Phlebia* sp. strain DABAC 9 were selected for remediation of naphthalene, dichloroaniline isomers, *o*-hydroxybiphenyl and 1,1'-binaphthalene [24]. For anthracene (AC) degradation, *Tetrahymena pyriformis* accumulated high amounts of AC without any transformation. In contrast, the fungi *Absidia cylindrospora, A. fusca, Cunninghamella elegans, Aspergillus terreus, Cladosporium herbarum, Penicillium chrysogenum, Rhodotorula glutinis,* and *Saccharomyces cerevisiae,* were able to transform AC to 1,4-dihydroxyanthraquinone as a product of biotransformation [25]. Biodegradation of phenanthrene by *Thrichoderma* sp. S019 afforded 1-hydroxy-2-naphthoic acid, salicylaldehyde, salicylic acid, and catechol as intermediates in the bioremediation process [26]. In addition, *Cunninghamella elegans* IM 1785/21Gp gave the metabolites *trans*-1,2,3,4- and 9,10-dihydrodiols, phenols, diphenols (diols), and glycoside conjugates of 1-,2-,3-,4-, and 9-phenanthrols [27]. Other experiments based on co-cultures were carried out with two fungi, *Aspergillus terreus* and *Penicillium* sp., and the bacterial strain *Rhodococcus* sp. IC10 [28]. Moreover, using *Fusarium solani*, a high degradation of phenantrene was obtained in free cultures and immobilized [29, 30, 31].

In the meanwhile, Li *et al.* [32] reported the biodegradation of 1,2,3,4-tetrahydronaphthalene (THN) by the marine fungus *Hypoxylon oceanicum* (326#) giving one major product, 3,4-dihydro-4-hydroxy-1(2*H*)-naphthalenone, and three minor products: 3,4-dihydro-1(2*H*)-naphthalenone, 1,2,3,4-tetrahydro-1-naphthalenol, and 1,2,3,4-tetrahydro-1,2-naphthalenediol.

Other PAHs were tested for fungi biodegradation such as benzo(α)anthracene, benzo(α)fluoranthene, benzo(α)pyrene (**1**), and chrysene, which were biodegraded by *Fusarium flocciferum, Trichoderma* species, *Tremetes versicolor,* and *Pleurotus ostreatus* [33]. Benzo(α)pyrene (BaP) (**1**) is a 5-ring polycyclic aromatic hydrocarbon and a large number of fungi were tested for its degradation such as *Trichoderma* sp., *Aspergillus niger, Mucor* sp., and *Fusarium* sp. [19, 34, 35, 36]. *Fusarium* sp. E033 was able to biodegrade 65-70% of the initial benzo(α)pyrene (**1**) provided giving two transformation products, dihydroxy dihydro-benzo(α)pyrene and benzo(α)pyrene-quinone [37]. Meanwhile, *Penicillium chrysogenum* SF04 had the highest degradation of BaP (**1**) (up to 71.31 %) [38]. Furthermore, benzo(α)anthracene was degraded by *Irpex lacteus* affording 2-hydroxymethyl benzoic acid or monomethyl- and dimethyl-esters of phthalic acid and 1-tetralone as final products [39]. However, a high degree of benzo(α)pyrene (**1**) degradation is undesirable for the bioremediation of BaP-contaminated soils because some of its accumulated metabolites still have severe health risks for humans such as benzo(α)pyrene-1,6-quinone (BP1,6-quinone) (**2**) and 3-hydroxybenzo(α)pyrene (3-OHBP) (**3**) (Scheme **1**) [40].



Scheme 1. Degradation of BaP (**1**) by fungi.

Other groups of important aromatic contaminants are phenols and derivatives. The major sources of phenol contamination are the chemical and petrochemical industries, agriculture (pesticides, containing hydrocarbons), wood processing as part of papermaking technologies, textile industry, etc. The ubiquitous nature of phenols, their toxicity even in trace amounts and the stricter environmental regulations make it necessary to develop processes for the removal of phenols from wastewaters. Biodegradation allows for the utilization of aromatic hydrocarbons by the biological agent and for their re-entrance into the carbon cycle [41].

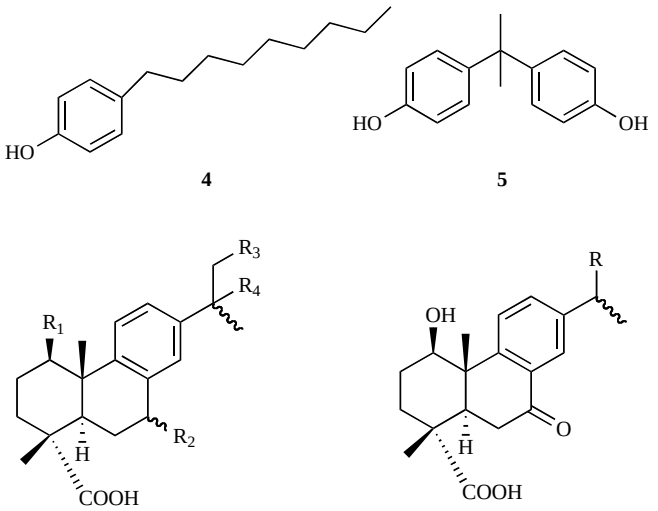
Along these lines, white-rot fungi have been shown to exhibit unique biodegradation capabilities for phenols [42, 43, 44]. For instance, the fungus *Panus tigrinus* CBS 577.79 was investigated for its ability to reduce the polluting load of olive-mill wastewater (OMW) with a significant presence of phenolic components [45].

On the other hand, strains from *Fusarium* were able to reduce aromatic components by 65% in olive-mill dry residue (DOR) [46]. Moreover, *Fusarium* sp. HJ01 was able to grow using phenol as the only carbon resource giving the intermediate catechol as a biotransformation product [47, 48]. The fungus *Trametes versicolor* was capable of decolouring and degrading phenol compounds from paper mill effluent [49, 50, 51].

As the symbiotic fungi and plant, mycorrhiza was able to degrade organic pollutants [52]. Some conifer ectomycorrhizae can degrade and detoxify water-solution phenolic compounds produced by the conifer *Kalmia angustifolia*. Thus, *Paxillus involutus, Laccaria laccata*, and *L. bicolour* were employed to degrade water leachates of *Kalmia* leaf and litter. Pure ferulic, *o*-coumaric, and *o*-hydroxyphenylacetic acids were degraded by 100, 98, and 79.5%, respectively, in the presence of *P. involutus* 211804 [53].

The biodegradation of polychlorophenols (PCBs) [54], an important group of phenols which have been used as fungicides, herbicides, insecticides, and in the synthesis of other pesticides, has been widely studied [55-58]. The white-rot fungus *Phlebia brevispora* was shown to be able to degrade PCBs obtaining *m*-methoxylated, *p*-dechlorinated and *p*-methoxylated metabolites [59]. 4,4'-Dichlorobiphenyl (4,4'-DCB) and its metabolites were added to cultures from *Phanerochaete* sp. and the metabolic pathway was elucidated by the identification of metabolites namely 2-hydroxy-4,4'-DCB and 3-methoxy-4,4'-DCB, 4-chlorobenzoic acid, 4-chlorobenzaldehyde, 4-chlorobenzyl alcohol, and 4-hydroxy-3,4'-DCB [60, 61, 62]. Fungi *Phlebia* sp., *Phanerochaete chrysosporium*, and *Mortierella* sp. were also selected to degrade different chlorobenzoic acids (CBA) [63] giving some aromatic metabolites from a hydroxylation pathway and a dechlorination pathway [64]. Furthermore, *Bjerkandera adusta*, *Anthracophyllum discolour*, immobilized and non-immobilized *Phanerochaete chrysosporium* [65], *Trametes versicolor* isolate HR131, and *Trametes* sp. isolate HR577 were studied in the degradation of pentachlorophenol [51, 66, 67, 68]. Fungi *Boletus edulis, Suillus luteus, Cortinarius russus, Suillus grevillei, Gomphidius viscidus, Laccaria bicolor, Leccinum scabrum, Xerocomus chrysenteron, Heboloma crustuliniforme*, and *H. longicaudum* were grown in media with different substrate concentrations of pentachlorophenol (PCP) to determinate their effect on fungal growth. No impact on the growth of the mycelia was observed at low ambient PCP levels. In addition, the high tolerance capability for pentachlorophenol may be related to their oxidoreductase activities and acidification effect [69].

Whereas, *Aspergillus awamori* NRRL 3112 degraded phenol, catechol, 2,4-dichlorophenol and 2,6-dimethoxyphenol as well as *Peniophora cinerea*, *Psilocybe castanella*, two strains of *Trametes villosa*, *Agrocybe perfecta*, *Trichaptum bisogenumand* and *Lentinus villosus* were able to colonize soil containing up to 4600 mg pentachlorophenol/kg soil. All fungi produced chloride ions during degradation, indicating dehalogenation of the molecule [70]. Moreover, Taseli *et al.* [71] studied the potential of the fungus *Penicillium camemberti*, which degraded pentachlorophenol (PCP), 2-chlorophenol and trichloroacetic acid.



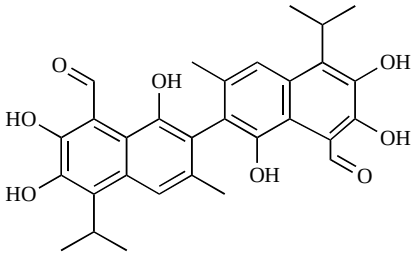
6: $R_1=R_2=R_3=R_4=H$
7: $R_1=OH, R_2=R_3=R_4=H$
8: $R_1=OH, R_2=\alpha\text{-}OH, R_3=R_4=H$
9: $R_1=R_3=OH, R_2=R_4=H$
11: $R_1=R_4=H, R_2=\beta\text{-}OH, R_3=OH$
12: $R_1=R_3=OH, R_2=\beta\text{-}OH, R_4=H$
14: $R_1=R_2=OH, R_3=\alpha\text{-}OH, R_4=H$
15: $R_1=R_2=OH, R_4=\alpha\text{-}OH, R_3=H$

10: R=Me
13: $R=CH_2OH$

Scheme 2. Fungal biotransformation products of dehydroabietic acid (**6**).

Other phenolic compounds with special activities are nonylphenols (**4**) and bisphenol A (**5**) (BPA), known as endocrine-disrupting compounds. Technical nonylphenol (t-NP) mixtures (**4**) were assessed using the mitosporic fungal strain UHH 1-6-18-4 and a strain of the aquatic hyphomycete *Clavariopsis aquatica*. All t-NP isomers were degraded to individual extents [72]. On the other hand, Soares *et al.* [73] showed that the fungi *Phanerochaete chrysosporium*, *Pleurotus ostreatus*, *Trametes versicolor* and *Bjerkandera* sp. BOL13 degraded nonylphenol (**4**) at an initial concentration of 100 mg/L. In addition, bisphenol A (**5**) was biodegraded with several white rot fungi (*Irpex lacteus*, *T. versicolor*, *Ganoderma lucidum*, *Polyporellus brumalis*, *Pleurotus eryngii*, *Schizophyllum commune*) isolated in Korea and two transformants of *T. versicolor* (strains MrP 1 and MrP 13) [74]. *Stereum hirsutum* and *Heterobasidion insulare* showed high resistance to BPA (**5**) [75].

Further results showed the potential of the fungi *Basidioradulum molare* and *Schizopora paradoxa* to degrade phenolic compounds such as 4-*tert*-octylphenol [76]. Moreover, van Beek *et al.* [77] reported the degradation of dehydroabietic acid (DHA) (**6**) from Scots pine wood by *Trametes versicolor* and *Phlebiopsis gigantea* in liquid stationary cultures, isolating some biodegradation products from *P. gigantea* cultures: 1β-hydroxy-DHA (**7**), 1β,7α-dihydroxy-DHA (**8**), 1β,16-dihydroxy-DHA (**9**), and tentatively 1β-hydroxy-7-oxo-DHA (**10**) and *T. versicolor* cultures, 1β,16-



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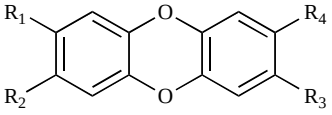
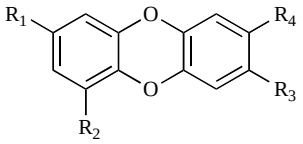
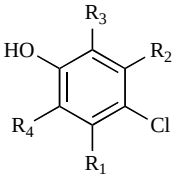


17: R_1=R_4=Cl, R_2=R_3=H
18: R_1=R_2=R_3=R_4=Cl



19: R_1=R_3=Cl, R_2=R_3=H
20: R_1=R_2=R_4=Cl, R_3=H



21: R_1=R_2=R_4=H; R_3=OH
22: R_1=Cl; R_2=R_4=H; R_3=OH
23: R_1=R_2=H; R_3=Cl; R_4=COOH
24: R_1=R_2=R_3=H; R_4=COOH

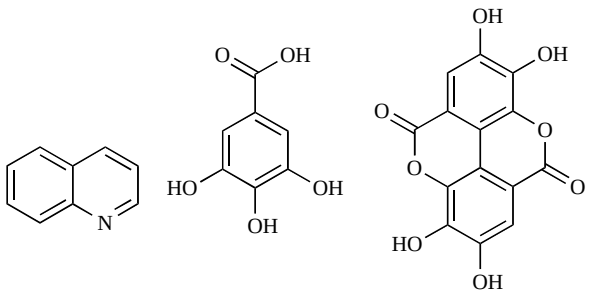
Scheme 3. Biodegradation of the polychlorinated dioxins **17-20**.

dihydroxy-DHA (**9**), 7β,16-dihydroxy-DHA (**11**), 1β,7β,16-trihydroxy-DHA (**12**), 1β,16-dihydroxy-7-oxo-DHA (**13**), 1β,15-dihydroxy-DHA (**14**), and 1β,7α,16-trihydroxy-DHA (**15**) (Scheme **2**). Also, *Candida tropicalis* was tested on the reduction of free gossypol (**16**), a polyphenol derived from the cotton plant. This biodegradation was evaluated through optimization of the parameters in order to operate under optimal conditions [78].

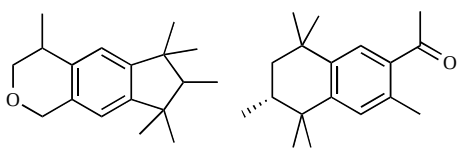
Indeed, polychlorinated dibenzo-*p*-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs), commonly known as dioxins (PCDD/Fs) [79], are toxic environmental pollutants formed from various sources. Removal of dioxins by biological degradation is considered a feasible method as an alternative to other expensive physic-chemistry approaches [80]. Different dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) were studied for degradation by fungi such as *Phlebia radiata* I-5-6 [81, 82, 83], *P. acerina*, *P. lindtneri* and *P. brevispora* which can hydroxylate and methoxylate PCDDs [84], and *Phanerochaete chrysosporium* DSM 6909, *P. chrysosporium* DSM 1556, *Irpex* sp. W3, *Trametes* sp. CH2, *Fusarium* sp. VSO7 [85], and *Pleurotus pulmonarius* [86]. Biodegradation of 2,8-dichlorodibenzo-*p*-dioxin (2,8-DCDD) (**17**), 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) (**18**), 2,7-dichlorodibenzo-*p*-dioxin (2,7-DCDD) (**19**), and 2,4,8-trichlorodibenzofuran (2,4,8-TCDF) (**20**) was conducted with two fungi (PL1 and 267) already screened from nature [87]. Furthermore, 2,8-DCDD (**17**) and 2,3,7,8-TCDD (**18**) were also degraded by those fungi, giving compounds considered to be intermediates, namely 4-chlorocatechol (**21**) and 4,5-dichlorocatechol (**22**) respectively [88, 89]. Chlorocatechol (**21**), 3,5-dichlorosalicylic acid (**23**) and 5-chlorosalicylic acid (**24**) were isolated from 2,7-DCDD (**19**) and 2,4,8-TCDF (**20**), respectively (Scheme **3**) [90].

Lastly, other aromatic pollutants have been tested for bioremediation. Quinoline (**25**) was biodegraded by *Pleurotus ostreatus* BP resulting in total mineralization and some fermentation products [91].

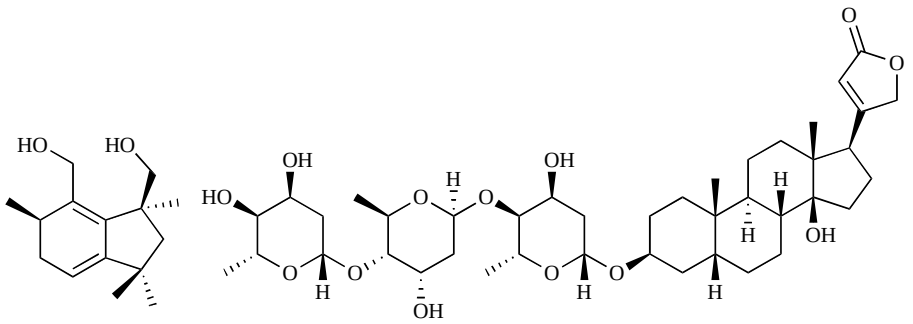
As the use of hydrocarbons by the microorganisms is associated with biosurfactant production, biodegradation by *Aspergillus niger* and *Penicillium nigricans* was also investigated [92]. Another *A. niger* strain namely PSH is capable of degrading tannins giving gallic acid (**26**) and ellagic acid (**27**) [93]. Martin *et al.* [94] reported the degradation of galaxolide (HHCB) (**28**) and tonalide (AHTN) (**29**), two micropollutants from aquatic environments, by *Myrioconium* sp. strain UHH 1-13-18-4 and *Clavariopsis aquatica*. The products obtained were the result of hydroxylations at different positions. *Polyporus brumalis* was applied to degrade dibutyl phthalate (DBP), the main product being phthalic acid anhydride as well as trace amounts of α-hydroxyphenylacetic acid, benzyl alcohol, and α-hydroxyphenylacetic acid [95].



25 **26** **27**



28 **29**



30 **31**

Additionally, degradation of refractory organic matter (OM) by the basidiomycete fungus *Schizophyllum commune* and white rot fungi was reported. Main products of the biodegradation were organic heavy metal complexes which can enter the environment [96, 97].

Moreover, indole degradation was studied by *Sporotrichum thermophile* and *Pleurotus ostreatus* with more than a 99% consumption rate of indole [98, 99]. Biodegradation of *p*-cresol by *Gliomastix indicus* was studied [100].

Aliphatic Hydrocarbons

Fungi can also degrade *n*-alkanes such as tridecane, tetradecane, pentadecane, hexadecane, heptadecane, octadecane (C13-C18) and crude Omani oil. Biodegradation by the fungi *Aspergillus niger*, *A. ochraceus*, *Trichoderma asperellum* strain TUB F-1067 (SA4), *T. asperellum* strain Tr48 (SA5), *T. asperellum* strain TUB F-756 (SA6), *Penicillium* species (P1), and *Aspergillus* species (P9) was studied [101]. Among these fungi, the P1 strain exhibited greater potential in degrading the aliphatic hydrocarbon compounds of used motor oil [2].

Some of the most important aliphatic hydrocarbon pollutants are *n*-eicosane, which was degraded by *Trichoderma* sp. S019 affording nonadecanoic acid, *n*-octadecane, hexadecanoic acid, oleic acid and stearic acid as reaction products [102]. Imidazolium compounds (ICs) and quaternary ammonium compounds (QACs) were degraded by two strains of *Gliocladium roseum*, *Penicillium brevi-compactum*, *P. funiculosum*, *Phialophora fastigiata*, *Verticillium lecanii* [103]. Carbon tetrachloride (CT) [104], trichloroethylene (TCE) and perchloroethylene (PCE), one of the most important groundwater pollutants, were tested for degradation by fungi. The aerobic degradation of PCE was reported for the first time by *Trametes versicolor*, giving 2,2,2-trichloroethanol and CO_2 as main byproducts from TCE degradation, and trichloroacetic acid (TCA) from PCE [105]. Moreover, *Ganoderma lucidum* and *Irpex lacteus* were able to degrade substantial levels of perchloroethylene (PCE) and trichloroethylene (TCE) in pure culture [106].

Another white-rot fungi, *Bjerkandera adusta*, was able to degrade hexachlorocyclohexane (HCH) isomers giving 1-(3-chloro-4-methoxyphenyl)ethanone and (2,4-dichloro-3-methoxy)-1-benzenecarbonyl chloride demonstrating the capability of *B. adusta* to produce these types of organochlorine compounds [107].

Compounds with different activities have been investigated for biotransformation including the sesquiterpene botrydienediol (**30**) by *Botrytis cinerea* affording three new sesquiterpenoids [108]. Wang *et al.* [109] studied the biodegradation by several fungi of digitoxin (**31**), a cardiac glycoside that is presumed to be effective in the treatment of heart failure. *Curvularia lunata* AS3.3589 and *Absidia coerulea* CICC40302 gave some biotransformation products.

Cyanide Compounds

Organic and inorganic cyanide compounds are widely distributed on the planet and they are among the most common corrosive pollutants. Most people associate the word cyanide with an extremely dangerous and fast-acting poison. However, there are several cyanide species, of varying toxicity, depending on the source of cyanide contamination. Free cyanide is the most toxic form and is easily and rapidly absorbed through inhalation, ingestion or skin contact. Thiocyanates are much less toxic than free cyanide and iron-complexed cyanides are only mildly toxic [110].

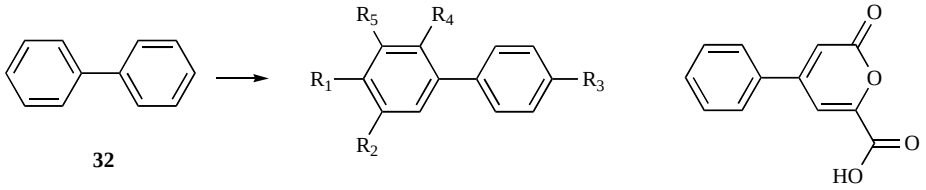
The degradation of simple cyanides has also been demonstrated in fungi [111, 112]. A fungal mutant of *Trichoderma koningii*, TkA8, constructed by restriction enzyme-mediated integration, was shown to have a high cyanide degradation ability [113]. As shown by Hossain *et al.* [114], *Trametes versicolor* ATCC 200801, *Phanerochaete chrysosporium* ME 496 and *Pleurotus sajorcaju* tolerated up to 500-ppm initial concentration of cyanide.

3. PESTICIDES

The agricultural industry's dependency on chemicals to sustain productivity in marginal landscapes has led to a global-scale contamination of the environment with toxic pesticides and nutrient fertilizers which are changing the course of biogeochemical cycles. They include fungicides, insecticides, and herbicides and are one of the causes of water pollution, and some pesticides are persistent organic pollutants contributing to soil contamination. Among the techniques employed to remove these contaminants, biodegradation is very effective, less contaminating and cheaper than others.

Fungicides

Biphenyl (**32**) and the monohydroxylated derivatives 2-hydroxy- and 4-hydroxybiphenyl are known to be fungistatic substances. These compounds are widely used for the con-

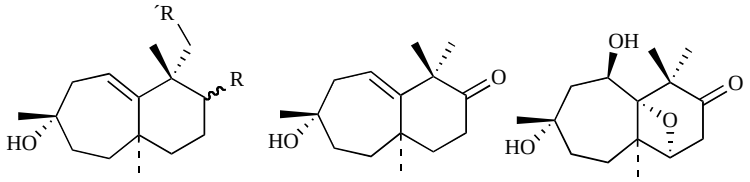


32

33: R_1, R_3=OH; R_2, R_4, R_5=H
34: R_1, R_5=OH; R_2, R_3, R_4=H
35: R_4=OH; R_1, R_2, R_3, R_5=H
36: R_2, R_4=OH; R_1, R_3, R_5=H

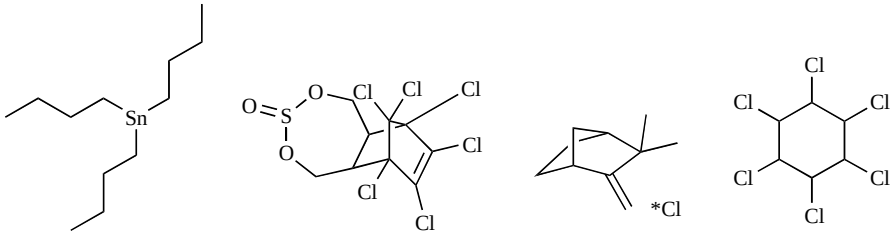
37

Scheme 4. Biodegradation of biphenyl (**32**) by *Talaromyces elicus.*



38:R=R´=H
39: R=β-OH, R´=H
41: R=α-OH, R´=H
42: R=H, R´=OH

40 **43**



44 **45** **46** **47**

servation of citrus fruits, even though biphenyl (**32**) is known for its toxic effects on humans. The filamentous fungus *Talaromyces helicus* oxidized biphenyl (**32**) to the hydroxylated derivatives 4,4´-dihydroxybiphenyl (**33**), 3,4-dihydroxybiphenyl (**34**), 2-hydroxybiphenyl (**35**), 2,5-dihydroxybiphenyl (**36**), and the ring cleavage product 4-phenyl-2-pyrone-6-carboxylic acid (**37**) (Scheme **4**) [115].

Tribromophenol (TBP) is used in wood preservation. *Trametes versicolor* and *Agaricus augustus* proved effective in decreasing TBP concentrations and *A. augustus* was also capable of biotransforming TBP to tribromoanisole (TBA) [116]. Other fungi employed for the degradation of this fungicide were *Laetoporeus sulfureus*, *Gloephyllum trabeum*, and *Ganoderma australe* in liquid culture, and were able to degrade TBP, degradation by *G. australe* being the most efficient (71% to 77%) [117].

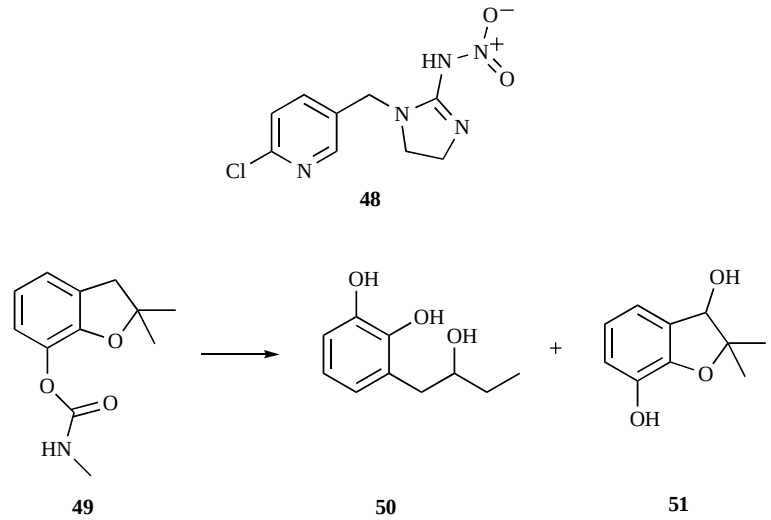
In addition, widdrol (**38**) has shown activity against the necrotrophic plant pathogen *Botrytis cinerea*. The biotransformation of **38** by *B. cinerea* and *Colletotrichum gloeosporioides* afforded four and one biotransformation products (**39**-**43**), respectively. Biotransformation with *C. gloeosporioides* yielding for the most part oxidation products at C-10: 10-oxowiddrol (**39**), 10β-hydroxywiddrol (**40**), 10α-hydroxywiddrol (**41**), and 14α-hydroxywiddrol (**42**). The biotransformation products were then tested against *B. cinerea* and found to be inactive [118].

Another group of pesticides with fungicide activity is tributyltin (**44**) compounds, a group of compounds containing the $(C_4H_9)_3Sn$ moiety. The filamentous fungus *Cunninghamella elegans* was able to degrade tributyltin chloride (TBT) giving less toxic compounds, dibutyltin and monobutyltin [119, 120].

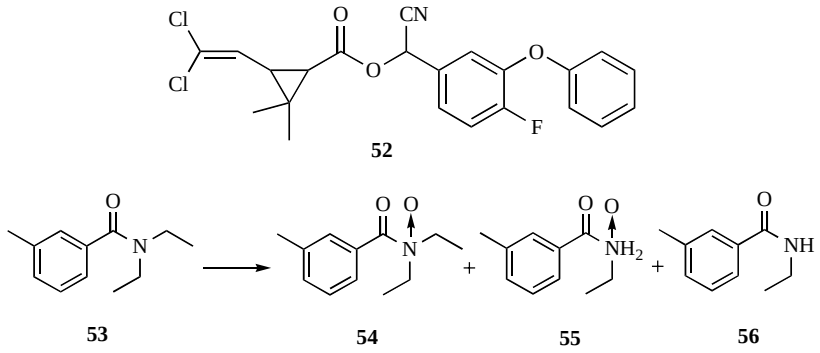
Insecticides

Some insecticides have been degraded by several fungi. For instance, endosulfan (**45**), widely employed as pesticide (insecticide and acaricide), was degraded by *Chaetosartorya stromatoides*, *Aspergillus terricola*, and *A. terreus* showing degradation rates of up to 75% [121]. In other studies using *A. niger*, various intermediates of endosulfan (**45**) metabolism including endosulfan diol and endosulfan sulfate were isolated [122].

Romero *et al.* [123] reported the biodegradation of toxaphene (**46**) in waste substrates by the fungus *Bjerkandera* sp. strain BOL13. One of the most important insecticides that is extensively used and toxic is lindane (**47**) [124], which was tested for biodegradation by nonwhite rot fungi [125] and white-rot fungi such as *Phanerochaete chrysosporium*, *Trametes hirsutus*, *Bjerkandera adusta*, and *Pleurotus* sp. [126, 127]. Also imidacloprid (**48**), a class of neuro-active insecticide modeled after nicotine, was degraded by *Calocybe indica* [128].



Scheme 5. Biodegradation of carbofuran (**49**) by *Mucor ramannianus.*



Scheme 6. Biodegradation of *N*,*N*-diethyl-*m*-toluamide(**53**).

Another example of insecticide biodegraded by several fungi is carbofuran (**49**), which was added to cultures of *Gliocladium* [129] and *Mucor ramannianus* affording 2-hydroxy-3-(butan-2-ol)phenol (**50**) and 3-hydroxycarbofuran-7-phenol (**51**) as transformation products (Scheme **5**) [130].

Brown-rot fungi were also investigated for their ability to degrade 1,1,1-trichloro-2,2-bis (4-chlorophenyl)ethane (DDT), as well as white-rot fungi [131] and ectomycorrhizal fungi [132]. For instance, *Gloeophyllum* genus, *Daedalea* genus, and *Fomitopsis* genus showed a high ability to degrade DDT affording 1,1-dichloro-2,2-bis (4-chlorophenyl) ethane (DDD), 1,1-dichloro-2,2-bis(4-chlorophenyl)ethylene (DDE), and 4,4-dichlorobenzophenone (DBP) as metabolic products [133].

β-Cyfluthrin (**52**), a synthetic pyrethroid insecticide, was biotransformed by *Aspergillus nidulans* and *Sepedonium maheswarium* to 4-fluoro-3-phenoxybenzaldehyde and 3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid [134].

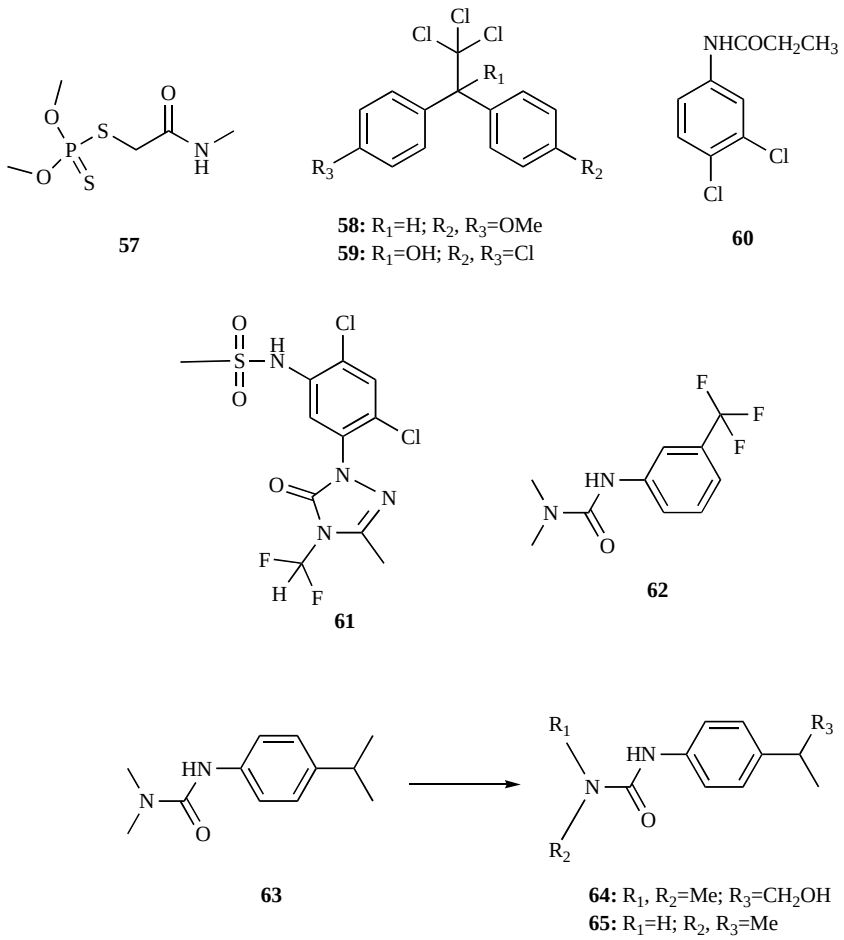
Seo *et al.* [135] reported the biotransformation of *N*,*N*-diethyl-*m*-toluamide (DEET) (**53**)**,** a topical insect repellent, by *Cunninghamella elegans* ATCC 9245, *Mucor ramannianus* R-56, *Aspergillus niger* VKMF-1119, and *Phanerochaete chrysosporium* BKMF-1767 to *N*,*N*-diethyl-*m*-toluamide-*N*-oxide (**54**), *N*-ethyl-*m*-toluamide-*N*-oxide (**55**), and *N*-ethyl-*m*-toluamide (**56**) (Scheme **6**).

The biodegradation by the strain *Aspergillus nomius* L3 of the insecticide dimethoate (**57**) as a cosubstrate was reported by Ai *et al.* [136]. In addition, methoxychlor (**58**) was converted to 2,2-dichloro-1,1-bis(4-methoxyphenyl)ethane, 2,2-dichloro-1,1-bis(methoxyphenyl)ethylene, 2-chloro-1,1-bis(4-methoxyphenyl)ethane, 2-chloro-1,1-bis(4-methoxyphenyl) ethylene, and 1,1-bis(4-methoxyphenyl)ethylene by *Stereum hirsutum* [137].

Lastly, Osman *et al.* [138] studied the biotransformation of dicofol pesticide (DCF) (**59**) by *Trichoderma viride* and *T. harzianum* with no intermediate or final degradation metabolites isolated. Fenitrothion breakdown product 3-methyl-4-nitrophenol (MNP), a newly characterized estrogenic chemist, was biotransformed by *Aspergillus niger* VKM F-1119 to 2-methyl-1,4-benzenediol, 4-amino-3-methylphenol, and two singly hydroxylated products [139].

Herbicides

Herbicides are widely used in agriculture to kill undesired plants. However, because of the large number of herbicides in use, there is significant concern regarding health effects which make their elimination from soil and water, an important focus of study.



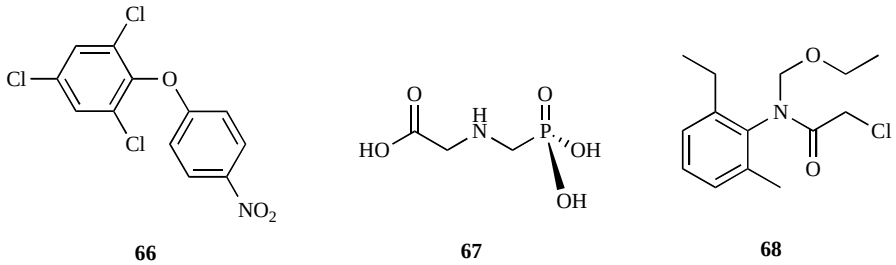
Scheme 7. Biodegradation of isoproturon (**63**) by fungi.

For example, ectomycorrhizal fungi from rice were evaluated for their applicability in studies of herbicide degradation. One of those strains was studied for its ability to degrade propanil (**60**) and its metabolite 3,4-dichloroaniline (3,4-DCA) [140]. Other fungi capable of degrading herbicides such as sulfentrazone (**61**) were *Chrysosporium* sp., *Eupenicillium* sp., and *Paecilomyces* sp [141]. Romeh *et al.* [142] investigated the degradation of fluometuron (**62**) herbicide by *Trichoderma viride*, *Metarhizium anisopliae*, and *Beauveria bassiana* showing biodegradation rates up to 85%.

Other herbicides were also employed to study their biodegradation by fungi, such as isoproturon (**63**), which was converted by *Phoma eupyrena*, *Mucor hiemalis*, and *Mortierella* sp. to hydroxylated metabolites *N*-(4-(2-hydroxy-1-methylethyl)phenyl)-*N*',*N*'-dimethylurea (**64**) and *N*-demethylated metabolite *N*-(4-isopropylphenyl)-*N*'-methylurea (MDIPU) (**65**) (Scheme **7**) [143]. Chlornitrofen (CNP) (**66**) was studied for biodegradation by *Phlebia brevispora* TMIC33929. In the degradation experiment using CNP (**66**) standard compounds, CNP (**66**) was transformed into several metabolites including monomethoxylated compounds and 2,4,6-trichlorophenol [144]. Another example of herbicide is glyphosate (**67**), a systemic non-selective herbicide, which was degraded by *Fusarium* sp. [145]; Zhu *et al.* [146] investigated the biodegradation by fungi of acetochlor (**68**) in soil. The effects of metsulfuron-methyl (**69**), a sulfonylurea herbicide, on soil microorganisms were evaluated in various experiments showing that fungi such as *Penicillium* sp. were highly tolerant [147, 148].

Metals

Soil contamination by toxic metals has become a serious problem, because of their long-term persistence and their diffusion into underground water. Heavy metal and non-



degradable chemical contamination of soil and water is a major environmental threat. In recent years, worldwide researchers are investigating new sustainable methods to mitigate such environmental contamination, such as biodegradation by fungi.

The common filamentous fungi can absorb heavy metals (Zn, Cd, Pb, Fe, Ni, Ag, Th, Ra & U) from aqueous solutions. The availability of a variety of fungi with different characteristics and metal binding potential makes it an economical and sustainable option for the removal and recovery of heavy metals [149].

Contaminated soil containing sulfide ore ashes and aromatic hydrocarbons could be treated with a metal-resistant strain BAS-10 of *Klebsiella oxytoca* and other fungi added to the soil, *Allescheriella* sp. DABAC 1, *Stachybotrys* sp. DABAC 3, *Phlebia* sp. DABAC 9, *Pleurotus pulmonarius* CBS 664.97, and *Botryosphaeria rhodina* DABAC P82. *B. rhodina* was the most effective fungus leading to the depletion of the most abundant contaminants [150].

Baldi *et al.* [151] developed a novel process combining sequential treatments of contaminated soil from the ACNA site (Cengio, Savona, Italy). The soil was leached to remove metals in the following order: Pb (74.2%) > Cu (72.6%) > Zn (40.2%) > Ni (55.7%) > Cd (41.5%) > Cr > (21.7%) Co > (19%) Fe (8.2%). The leachate was then incubated with the metal-resistant *Klebsiella oxytoca* strain BAS-10 and *Allescheriella* sp. DABAC 1 leading to a complete degradation of several organic contaminants.

Several metal ions were bioremediated by fungi. For instance, the treatment of the simulated lead-contaminated solid waste by composting with white-rot fungus was studied in the laboratory [152]. The white-rot basidiomycete *Phanerochaete chrysosporium* was very effective in the bioremediation of Pb-contaminated soil [153]. The Pb(II) biosorption potential of *Aspergillus parasiticus* [154] and the macrofungus *Ganoderma carnosum* [155] were also studied in a batch system and biosorption conditions were optimized showing that biosorption potential depended on physicochemical parameters. Tunali *et al.* [156] reported the biosorption of Pb(II) onto *Cephalosporium aphidicola* and the nature of the possible cell and metal ion interactions was examined by the FTIR technique. Moreover, the potential of *Botrytis cinerea* as a biosorbent for metal ions such as Zn(II) and Pb(II) was studied. Competitive biosorption experiments were performed with Zn(II) in the presence of Cu, Cd, and Ni ions simultaneously [157], demonstrating that other competing metal ions (Cu(II), Cd(II), and Ni(II) co-cations) reduced the biosorption capacity on Pb(II) and Zn(II) [158]. *Aspergillus flavus* and *Neurospora crassa* fungal biomass were also able to absorb Pb(II) and Cu(II) under optimum conditions [159, 160]. Cr(VI) biosorption with *Trichoderma viride*, *Aspergillus niger*, *A. sydoni*, and *Penicillium janthinellum* biomass was studied by Bishnoi *et al.* [161] showing that biosorption of Cr(VI) was pH dependent and the maximum adsorption was at pH 2.0. Cr(VI) removal was 91.03% using *A. niger* and 87.95% and 86.61% with *A. sydoni* and *P. janthinellum* [162]. Lastly, the brown-rot fungus *Lentinus edodes* was used as an efficient biosorbent for the removal of Cd from water and three kinds of adsorption models were applied to simulate the biosorption data [163].

Mercury is one of the most harmful ion metals and its biotransformation by fungi was investigated in *Hymenoscyphus ericae*, *Neocosmospora vasinfecta*, and *Verticillium terrestre* following the exposure of these fungi to environmentally relevant doses of Hg (II) ($HgCl_2$) in aerated pH-controlled cultures [164].

Industrial Dyes

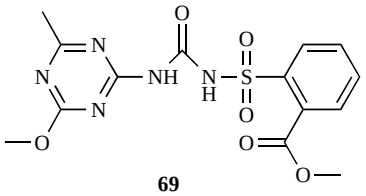
Dyes, originally obtained exclusively from natural sources, are today also produced synthetically on a large scale and represent one of the very mature and traditional sectors of the chemical industry.

Different dyes and pigments are extensively used in the textile, paper, plastic, cosmetics, pharmaceutical and food industries. Most of the earlier dye decolourization studies were based mainly on white-rot fungi such as *Phanerochaete chrysosporium*, *Trametes versicolor*, *Phellinus gilvus*, *Pleurotus sajor-caju*, *Pycnoporus sanguineus*, *Dichomitus squalens*, *Irpex flavus*, *Daedalea flavida*, *Polyporus sanguineus*, *Funalia trogii* ATCC200800, *Ischnoderma resinosum* [165], *Dichomitus squalens* [166], and *Ganoderma* sp [167-170].

Textile industries consume large amounts of water and their effluents contain a wide range of contaminants. These contaminants are dyes with strong colour, inorganic salts, as well as high pH. Textile wastewater containing significant concentrations of dyes cause substantial treatment problems. Most of the dye molecules have a polyaromatic structure with a high molecular weight and contain atoms of nitrogen, sulfur and metals making it very difficult to break them down [171]. Biological methods are emerging as an effective alternative for chemical approaches. For instance, fungi have shown a strong resistance to dye toxicity and it would therefore be a good idea to study fungal strains to identify the potential fungal candidates for dye removal and biodegradation [172-177].

White-rot fungi have been widely employed to biodegrade textile dyes [178-181]. Thus, Sukumar *et al.* [182] investigated the decolourizing ability of *Phanerochaete* sp. and *Trametes* sp., recording colour reduction of 82.01% and 76.07% respectively [183]. The potential of *Trametes villosa* and *Pycnoporus sanguineus* to decolourize reactive textile dyes was evaluated, *T. villosa* being the best degrader [184]. The characteristics of biodegradation of the classical triphenylmethane dyes such as Crystal Violet (**70**), Malachite Green (**71**), and Bromophenol Blue (**72**) were reported by white rot fungi in rice straw media [185, 186, 187]. Yan *et al.* [188] studied the decolourization by biosorption using dead white rot fungus *Pleurotus ostreatus* BP, whose decolourization rate for Remazol Brilliant Blue R (RBBR) (**73**) reached 82.35%.

Synthetic dyes are released in wastewater from textile manufacturing plants, and many of these dyes are genotoxic. For instance, *Irpex lacteus* was used for mutagenicity assays showing that all dyes except Congo Red (CR) (**74**) were mutagenic, indicating that the combined biodegradation process may be useful for reducing the mutagenicity associated with wastewater from textile industries [189]. The biodegradation of Methyl Orange (**75**), Yellow RR Gran, Congo Red (**74**), Bismarck Brown (**76**), Brilliant Red K-2BP (**77**), and the azo dye Remazol Red RR Gran in cultures of the



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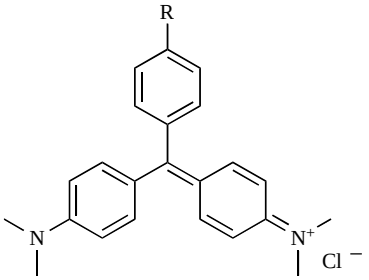
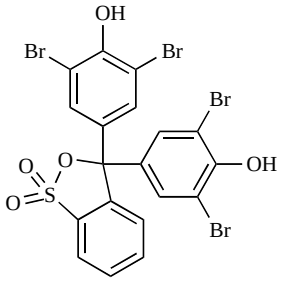


70: R=$N(CH_3)_2$
71: R=H



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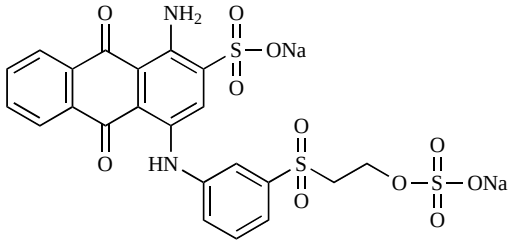

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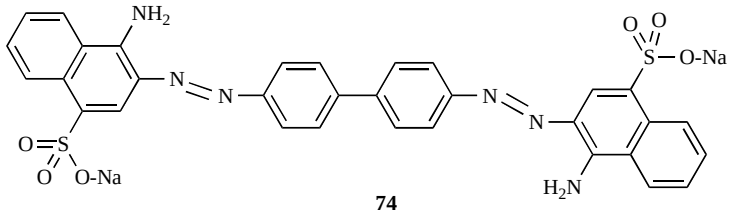

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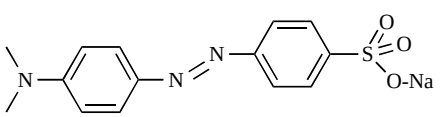

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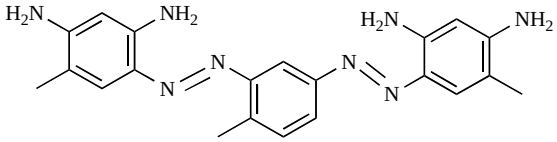

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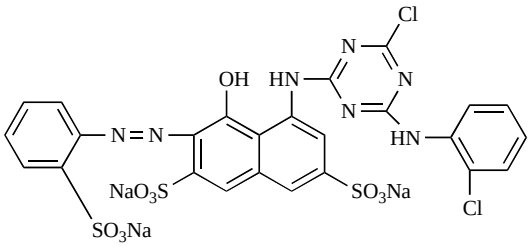

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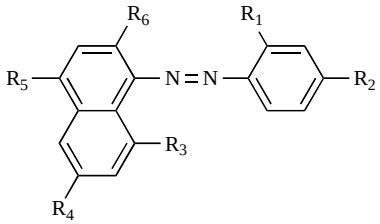
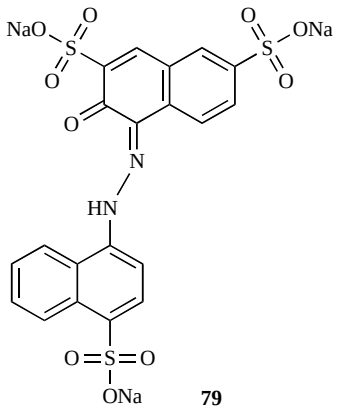


78: R_1=R_2=R_5=H; R_3=R_4=SO_3Na; R_6=OH
80: R_1=R_3=R_4=R_6=H; R_2=SO_3Na; R_5=OH
82: R_1=R_3=R_4=R_5=H; R_2=SO_3Na; R_6=OH
83: R_1=Me; R_2=SO_3Na; R_3=R_4=R_5=H; R_6=OH

white rot fungus *Phaenerochaete chrysosporium* was demonstrated by decolourization studies [190, 191, 192, 193]. Moreover, the ability to decolourize eight chemically different synthetic dyes (Orange G (**78**), Amaranth (**79**), Orange I (**80**), Remazol Brilliant Blue R (RBBR) (**73**), Cuphthalocyanin, Poly R-478 (**81**), Malachite Green (**71**) and Crystal Violet (**70**)) by the white rot fungus *Dichomitus squalens* was evaluated showing high decolourization capacity for all dyes tested, but not to the same extent [194].

Asgher *et al.* [195, 196] investigated the indigenous white rot fungi *Pleurotus ostreatus* IBL-02, *Phanerochaete chrysosporium* IBL-03, *Coriolus versicolor* IBL-04, *Ganoderma lucidum* IBL-05, and *Schizophyllum commune* IBL-06 for decolourization of several textile dyes. The results



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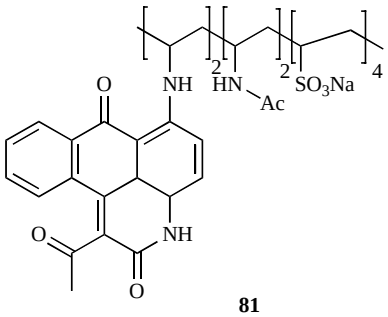

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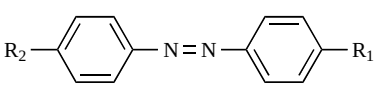
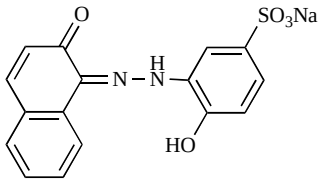


84: R_1=OH; R_2=SO_3Na
86: R_1=NH_2; R_2=NO_2
87: R_1=NHPh; R_2=NO_2
88: R_1=N(Et)(CH_2CH_2OH); R_2=NO_2



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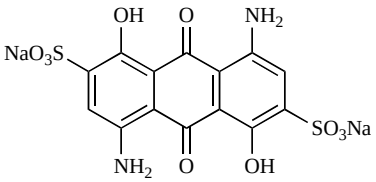

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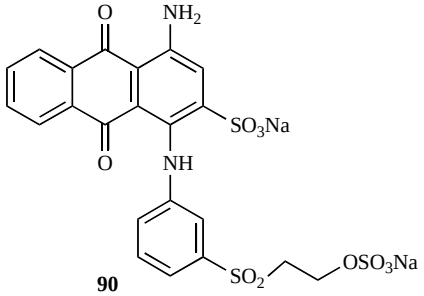

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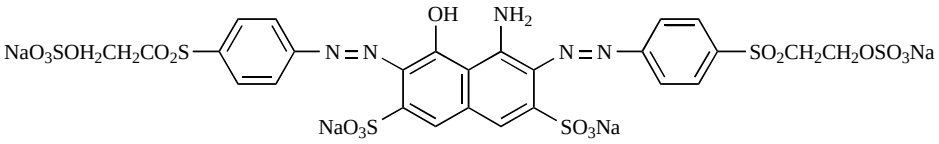
showed that *P. chrysosporium* could decolourize all the dyes tested, and *C. versicolor* IBL-04 degraded all the dyes, except Drimarene Orange K-GL. Moreover, *P. ostreatus* also showed good decolorization efficiencies on all dyes, except Remazol Brilliant Yellow. However, the rest of the strains showed poor decolourization potential. In addition, *S. commune* IBL-06 and *G. lucidum* IBL-05 were able to degrade Solar Golden Yellow R [197], Solar Orange RSN [198], and Cibacron Red FN-2BL [199].

Moreover, Machado *et al.* [200, 201] studied the potential of basidiomycetous fungi isolated from tropical ecosystems to remove Remazol Brilliant Blue R (RBBR) (**73**) dye. *Trogia buccinalis* showed the highest RBBR (**73**) decolourization.

Removal of water-solution sulfonated azo dyes from textile industry effluents is a major issue in wastewater treatment [199]. Biodegradation of sulfonated azo dyes was studied using white-rot fungi affording 4-hydroxy-benzenesulfonic acid, 3-methyl-4-hydroxy-benzenesulfonic acid, benzenesulfonic acid, 1,2-naphthoquinone-6-sulfonic acid, and 3-methyl-benzenesulfonic acid, as major biotransformation products for Orange I (**80**), Acid Orange 7 (**82**), Acid Orange 8 (**83**), Great Acid Red, and 4-[(4-hydroxyphenyl)azo]-benzenesulfonic acid (**84**) [202, 203, 204]. The degradation of Mordant Violet 5 (**85**) by *Pleurotus ostreatus* gave benzenesulfonic acid, 4-hydroxybenzensulfonate, and 1,2-naphthoquinone [205, 206]; Disperse Orange 3 (**86**), Disperse Orange 1 (**87**), and Disperse Red 1 (**88**) were degraded to nitrobenzene, 4-nitrophenol and 4-nitroaniline [207, 208]. Furthermore, 1-methoxy-4-nitrobenzene, 1,2-dimethoxy-4-nitrobenzene, and 2-methoxy-4-nitrophenol were found to be produced from 4-nitrophenol [218].

Decolourizing by non white-rot fungal species such as *Aspergillus flavus*, *A. niger* [209, 210], *Helminthosporium* sp, *Mucor* sp, *Penicillium* sp., *Trichoderma viride*, *Myrothecium* sp. IMER1 [211], and *Fusarium* sp. isolated from textile effluent, was investigated for Remazol Yellow, Remazol Orange [212], Remazol Brilliant Blue R (RBBR) (**73**), Neutral Brilliant Blue GL, and Acid Blue B (**89**), obtaining decolourization rates ~100% [213, 214, 215]. Also, *Trichophyton rubrum* LSK-27 was able to decolourize 83% of Remazol Tiefschwarz, 86% of Remazol Blue RR (**90**) and 80% of Supranol Turquoise GGL in liquid cultures [216].

The mushroom fungi *Lentinus conatus*, *Ischnoderma resinosum*, and *Ganoderma lucidum* KMK2 were studied for this purpose [217]. The results showed that *I. resinosum* was able to decolourize all textile dyes tested (Reactive Black 5 (**91**), Reactive Blue 19 (**73**), Reactive Red 22 (**92**), and Reactive Yellow 15 (**93**)) [218] and *G. lucidum* decolorized anthraquinone dye Remazol Brilliant Blue R (RBBR) (**73**) and



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diazo dye Remazol Black-5 (RB-5) (**91**) with biodegradation levels up to 90% [219]. Furthermore, *Coprinellus xanthothrix*, a new fungal strain isolated from a polyphenol polluted soil in Greece, was tested for its ability to degrade a polyaromatic dye Poly R-478 (**81**). The fungus showed biosorption and biotransformation as removal mechanisms [220].

Dyehouse effluent treatment has become inevitable because of the presence of dyes which originate from harmful chemicals. *Galactomyces geotrichum* and *Trametes* sp. isolated from contaminated soil were employed to degrade these dyes, finding a new strain with the high rate of 87.21% color reduction [221, 222]. In addition, *Trametes versicolor* decolourized the mono-azo-substituted naphthalenic dye Amaranth (**79**) [223, 224].

Lignin and Cellulose Degradation

Cellulose is the main polymeric component of the plant cell wall, the most abundant polysaccharide on Earth, and an important renewable resource. Basidiomycetous fungi are among its most potent degraders because many species grow on dead wood or litter which are rich in cellulose. For the degradation of cellulose, basidiomycetes utilize a set of hydrolytic enzymes typically composed of endoglucanase, cellobiohydrolase and β-glycosidase [225, 226, 227]. For instance, *Pleurotus ostreatus* produces the cellulolytic and hemicellulolytic enzymes endo-1,4-β-glucanase, exo-1,4-β-glucanase, 1,4-β-glucosidase, endo-1,4-β-xylanase, 1,4-β-xylosidase, endo-1,4-β-mannanase and 1,4-β-mannosidase and ligninolytic enzymes Mn-peroxidase and laccase during growth on wheat straw [228].

Lignin is a complex chemical compound most commonly derived from wood, and an integral part of the secondary cell walls of plants and some algae. Several fungi have been studied for the biodegradation of lignin. Thus, agro-industrial wastes containing lignocellulose can be upgraded by solid state fermentation [229] other than biopulping during which the selective conversion of lignin is required. Several fungi (e.g. *Pleurotus* sp., *Schyzophyllum* sp., *Tremetes versicolor*, *Lentinus crinitus*, *Aspergillus fumigatus*, *Stemphylium verruculosum*, *Paecilolomices carneus*, *Ceriporiopsis subvermispora*, and species of the genus *Phlebia*) were able to grow on different agro-industrial wastes, obtaining high biodelignification [230-237]. Moreover, fungi belonging to genus *Aspergillus*, *Trichoderma*, *Phanerochaete* and *Coprinus* are known to decompose paddy straw, corn straw, wheat straw and horticultural wastes, whereas *Pleurotus sajor-caju*, *P. platypus* and *P. citrinopileatus* are known to colonize coir fibre, cotton stalks and sorghum stover. These fungi may be specific for each substrate and can be used as an effective tool for in situ degradation of lignin residues [238-243].

Trametes versicolor contributed to improving the biodegradability of Norway spruce chips from the paper industry [244] and gave better results in the removal of poplar chips than the other fungi tested, *Phanaerochaete chrysosporium* and *Pycnoporus sanguineus* [245]. Thus, Elissetche *et al.* [246] studied the biodegradation of *Drimys winteri* and *Nothofagus dombeyi*, two native Chilean wood species, by *Ganoderma australe*, which is responsible for a unique field biodegradation process resulting in completely white-rotted logs known as "palo podrido" in southern Chile.

Nyochenbeng *et al.* [247, 248] studied edible white rot fungi for selective plant biomass transformation and recycling in a sustainable ecological advanced life support (ALS) needed for extraterrestrial expeditions, such as the mission to Mars. *Pleurotus ostreatus* ('Grey Dove'), *P. pulmonarius*, *P. eryngii*, and four shiitake mushroom (*Lentinula edodes*) strains were used in the study on processed residues.

Selective degradation of lignocellulose by bamboo white rot fungi was initially studied. Zhang *et al.* [249, 250] investigated the degradation of bamboo residues by *Coriolus versicolor* B1 and *Trametes* spp. B1, having apparent degradation selectivity for hemicellulose and lignin. *Echinodontium taxodii* 2538 and *Trametes versicolor* G20 were selected for the biological pretreatment of bamboo culms (*Phyllostachys pubescens*), increasing the sugar yield of bamboo culms [251]. White-rot fungi were also capable of degrading the effluent from *Eucalyptus* chemithermomechanical pulp (CTMP) [252] and david poplar wood living on broad-leaf trees. For instance, *Funalia gallica*, *Lenzites tricolor*, *Phellinus igniarius*, *Polyporellus brumalis*, *Pseudotrametes gibbosa*, and *Pycnoporus sanguineus* reduced phenolic acids in primitive david poplar wood and wood degradation [253].

A strain of non white-rot fungi isolated from soil, *Penicillium simplicissimum*, showed different ligninolytic ability from white-rot fungi. The lignin degradation by *P. simplicissimum* happened mainly during the primary metabolism and it was greatly influenced by the pH of the media, the concentration of Cu^{2+} and Mn^{2+} [254]. *P. simplicissimum* was also tested with *Aspergillus niger* to test its capacity to decompose hydroxybenzene and nonhydroxybenzene lignin compounds of low molecular weight. Five different enzymes, lignin peroxidase, manganese peroxidase, laccase, cellulase and hemicellulase, were believed to be the most important catalysts in biodegrading process, and they always worked synergistically [255].

Lastly, biodegradation by brown-rot fungi is quantitatively one of the most important fates of lignocellulose in nature. *Gloeophyllum trabeum* and *Fomitopsis* sp. IMER2 were investigated for the biodegradation of different samples of lignin. *G. trabeum* resulted in a marked, non-selective depletion of all intermonomer side-chain linkages in the lignin [256], and *Fomitopsis* sp. IMER2 was used for the treatment of black liquor by biological acidification for the pre-

cipitation of alkali lignin [257]. Furthermore, *Piptoporus betulinus*, a common wood-rotting fungus parasitic for birch (*Betula* species), was able to degrade the lignin of birch wood [258, 259].

Polymers

The increasing consumption of plastics has generated environmental problems because it takes more than a hundred years for a discarded polymer to degrade. The ideal plastic should present desirable industrial properties and be degradable within a satisfactory time period [260].

The biodegradation ability of fungi is being investigated for polymers and plastics [261]. One example is the biodegradation of plasticized polyvinyl chloride (pPVC) by *Penicillium janthinellum* and *Doratomyces* spp. in grassland soil. The incorporation of biocides into pPVC was also studied affecting both fungal growth and the richness of species isolated [262]. Moreover, *Gloeophyllum trabeum* were used to degrade poly(vinylalcohol)(PVA) films [263, 264]. Alariqi *et al.* [265] investigated the effect of sterilization on the biodegradation by *Aspergillus niger* of polyolefins which are widely used as part of biomedical devices and food packaging after sterilization.

Attending to the biodegradability of several polymers, the degradation of a blend of the copolymer poly(hydroxybutyrate-hydroxyvalerate), PHB-HV, which is a natural, biodegradable and biocompatible thermoplastic, was studied by a mixed culture of *Phanerochaete chrysosporium* and *Talaromyces wortmannii*. The results showed that the biodegradation of the blend was a function of time, with the appearance of terminal carboxylic groups [266]. In addition, carboxymethylchitosan-g-medium chain length polyhydroxyalkanoates polyhydroxyalkanoates (mcl-PHA) were biodegraded by *Aspergillus fumigatus* 202 with a 93% weight loss of the graft [267].

Some studies were done to elucidate the microbial communities responsible for the decomposition of poly-(ε-caprolactone) (PCL), poly-(butylene succinate) (PBS), poly-(butylene succinate and adipate) (PBSA), and poly-lactide (PLA) [268, 269, 270]. Fungi isolated from various soil environments were investigated to biodegrade poly(butylene succinate) (PBSu), such as KTF003, KTF004, and NKCM1001 strains, which have also been reported to be P(3HB)-degraders. Mesophilic strain NKCM1003 exhibited the highest poly(alkylene succinate) (PESu) hydrolytic activity among all the isolates [271, 272]. Biodegradation of polyethylene (DPE) was reported by white-rot fungi *Phanerochaete chrysosporium*was, *Talaromyces wortmannii*, and *Penicillium frequentans* showing high degradation [273, 274, 275].

Cosgrove *et al.* [276] studied the biodegradation of polyester-polyurethane (PU) showing that *Geomyces pannorum* and a *Phoma* species were the dominant species in soil fungal communities involved in the biodegradation of PU. Another recalcitrant synthetic polymer polyamide-6, generally known as nylon-6 was tested for biodegradation by fungi: five *Fusarium* spp., two Phanerochaete chrysosporium strains, four *Aspergillus* spp. and *Penicillium* spp., three *Cladosporium* spp. and *Ulocladium* spp., two *Trichoderma* spp., and one strain each of *Gliocladium roseum*, *Pithomyces chartarum*, *Trichotecium roseum*, and *Mucor hiemalis*. The study showed that only white rot fungi are able to break down nylon-6 [277]. On the other hand, the biodegradation of the aliphatic polyester resin Bionolle was carried out by the filamentous fungi *Aspergillus niger* and *Penicillium funiculosum*, and *Chlorella* sp., *Lemna minor*, *Brassica rapa*, *Daphnia magna* and *Allium cepa*. The products of hydrolytic degradation did not negatively affect the organisms living in the environment [278]. The aerobic biological degradation by fungi of the synthetic aliphatic-aromatic co-polyester Ecoflex (BASF) was also studied. Weight loss was not as obvious as visual degradation and suggested broader types of microbial attack [279].

Other Industries

Fungi are capable of mineralizing a wide variety of toxic xenobiotics due to the non-specific nature of their extracellular enzymes. For instance, anaerobically digested molasses spent wash (DMSW) is a dark-brown-coloured recalcitrant effluent which has a high chemical oxygen demand (COD) and high pollution potential. Fungi such as *Aspergillus*, *Rhizopus* and *Fusarium* were able to effectively degrade DMSW [280].

Wastes from the agricultural activities were used for biodegradation. The biodegradation of untreated fertilizer industry effluent using native fungus *Aspergillus niger* and non-native, white-rot fungus *Phanerochaete chrysosporium* was studied [281].

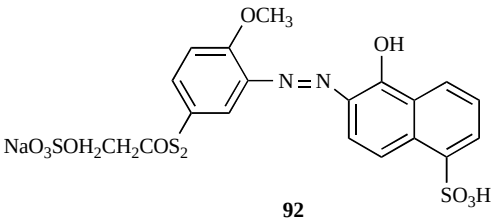
Kalyankar *et al.* [282] investigated the degradation of India dyes by fungi from the poultry industry in Maharashtra State. Of the fungi isolated from the soil of poultry farms, *Chrysosporium tropicum*, *C. keratinophilum*, *Microsporium cannis*, *Trichophyton verrucosum*, and *T. equinum* were found to be the most dominant. *Aspergillus* sp. was the most efficient microorganism in removing ammonia from the natural sources-poultry farm and agricultural fields [283].

Penicillium sp. P6 was isolated from coal mine soil at the Qiantong colliery, Liaoning Province, North-west China, as was able to degrade Chinese lignite effectively [284]. Deuteromycete *Neosartorya fischeri*, degraded coal in the Witbank coal mining area of South Africa [285].

Biodegradation of sugar industry wastewater using the fungi *Aspergillus niger* and *Phanerochaete chrysosporium* is an effective pollution abatement solution for wastewater treatment. The fungus can degrade 98.92 % of COD and 99.86 % of biochemical oxygen demand (BOD5) in 168 h of incubation at optimum biological process conditions. The color removal of the effluents is 99.34 % at optimum incubation time with optimum bioprocess parameters [286, 287].

One of the foremost environmental concerns in developing countries today is Solid Waste Management. In India, degradation of fruit waste was investigated by aerobic composting [288]. Garbage biodegradation was also studied by white-rot fungi [289]. Sludge degradation and bioflocculation were studied using pellet-forming filamentous fungi isolated from municipal wastewater sludge [290].

Tannery wastewater is a powerful pollutant especially due to its high chemical oxygen demand (COD). *Trametes versicolor* was examined to remove color from secondary treated tannery wastewater giving a maximum color removal efficiency of 64% [291]. *Fusarium culmorum* and *Muscodor*



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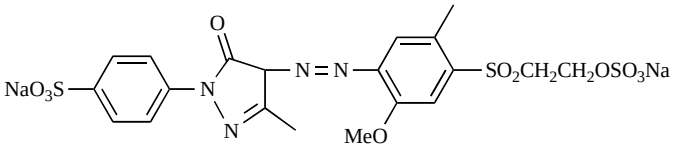

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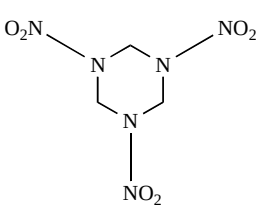

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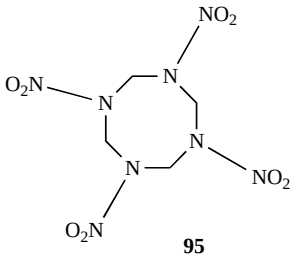

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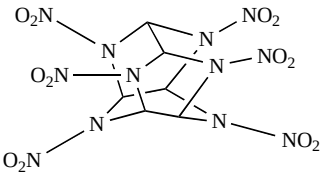

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albus were used to treat human and/or animal waste products with a good results [292].

Lastly, the accumulation of munitions wastes in the environment has damaged many ecosystems because of their explosive properties and these compounds are biological poisons. Biodegradation by fungi is being investigated. For instance, the biodegradation of diazodinitrophenol (DDNP) wastewater was carried out by a white rot fungus cultivated and domesticated at a laboratory *in situ*. The removal of aniline compounds and nitro compounds were over 99.9%, reaching the National First-degree Wastewater Discharge Standard [293].

Undersea deposition of unexploded ordnance (UXO) constitutes a potential source of contamination of marine environments by hexahydro-1,3,5-trinitro-1,3,5-triazine (RDX) (**94**) and octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazozine (HMX) (**95**). Using sediment from a coastal UXO field, Oahu Island, Hawaii, four novel aerobic RDX-degrading fungi HAW-OCF1, HAW-OCF2, HAW-OCF3 and HAW-OCF5 were isolated and tentatively identified as members of *Rhodotorula*, *Bullera*, *Acremonium* and *Penicillium*, respectively. The four isolates mineralized 15-34% of RDX (**94**) [294]. Royal Demolition Explosive (RDX) (**94**) was also degraded by white-rot fungi, showing that the removal efficiency in wastewater could reach 87% under optimum conditions [295, 296].

Degradation of the emerging contaminant CL-20 (2,4,6,8,10,12-hexanitro-2,4,6,8,10,12-hexaazaisowurtzitane) (**96**), was reported using *Phanerochaete chrysosporium*. Another ligninolytic fungus, *Irpex lacteus*, was also able to degrade CL-20 (**96**), but as for *P. chrysosporium*, no early intermediates were observed. The intermediate was thus tentatively identified as a doubly denitrated CL-20 (**96**) product [297].

CONCLUSIONS

Biocatalysis has been demonstrated to be a powerful tool for the pollutants degradation, which is a priority for scientists in the current industrialized world.

Chemical methods have several disadvantages in the degradation of pollutants because they usually use contaminant catalysts and their use in large-scale contaminated field sites is difficult. On the other hand, biocatalytic reactions can be carried out at ambient temperature and atmospheric pressure, under safety, health, environmental, and economical conditions. In this regard, fungi have been shown to possess a variety of catabolic activities that can be harnessed to transform contaminants in less toxic compounds for the environment.

Most of the research on fungal bioremediation has been conducted on laboratory scale and conditions, so further work is required to study these capacities taking into account the natural variables and their applicability in large-scale contaminated fields.

In addition, the screening of new fungal strains with interesting enzymatic activities is necessary for the degradation of the new pollutants from the increasing industry contamination. This microorganism screening, in combination with current biotechnologies such as genetic engineering, will pave the way to the future use of fungal whole cells and enzymes for bioremediation.

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