Review Paper:

Sustainable Approaches used in Pesticides contaminated Agricultural Waste Water Remediation

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Abstract

With the growing population of our global community and thereby increasing the demand for food has led the farmers to use more and more pesticides. Pesticides are chemical compounds used to kill pests. It is commonly used to control various agricultural pests that destroy crops and ultimately affect our agricultural productivity. Although the use of pesticides is a beneficial approach, but unfortunately it also has some drawbacks in terms of harming the environment and human health. Some pesticides pose significant hazards that ultimately affect human health systems. Contamination of agricultural land and agricultural wastewater by pesticides is a serious environmental problem and has a negative impact on biodiversity.

Most synthetic pesticides are not readily biodegradable, they accumulate in the environment and cause soil contamination. To overcome the environmental burden of pesticide-contaminated sites, various sustainable approaches can be effectively used to detoxify contaminants or transform them into harmless secondary compounds. The current review focuses on the role of microorganisms and lignocellulosic biomass and plants in the degradation of agricultural pesticides under sustainable approaches.

Keywords: Bioremediation, Pesecticide, Biosorption, Persistence, Lignocellulose, Biomass.

Introduction

Pesticides concentrations in water are increasing due to their widespread use in modern agriculture[197]. They are widely used worldwide for pest control as they are necessary to maintain the agricultural demand[84]. Soil is an important resource for natural sustainability and a major source of pollutants such as pesticides[93].

Globally, multiple human-induced activities such as rapid industrialization and modern gricultural practices are causing multiple ecological disturbances and ecological risks[230]. Various harmful pollutants including pesticides, are continuously released into the aquatic environment as well, severely impacting aquatic organisms[191,203]. Pesticide-contaminated soil can interact with heavy metals to further complicate the situation[276]. Microbial-mineral interactions serve as key factors in shaping the Earth's lithosphere. Microorganisms serve as valuable tools for soil remediation due to their ability to activate soil cycling, plant settlement and growth[151].

Soils have endogenous microbial activity indicated by basal respiration and microbial biomass[201]. Soil organisms respond to environmental changes primarily through carbon conversion, nutrient cycling and contaminant remediation[79]. Widespread application of chlorinated and organophosphorus-derived pesticides in agricultural practices increases the productivity of food crops while their high propensity for accumulation, persistence and bioaccumulation poses several threats to human health[227].

Soil remediation is typically uneconomical, labor intensive, costly and environmentally hazardous. An inexpensive and flexible way to restore ecosystem quality is the use of microorganisms, so-called bioremediation[22,133]. Microorganisms and their enzymatic systems play an important role in the remediation of contaminated soil[137]. In general, bioremediation-based methods primarily either rejuvenate indigenous microbiota that can utilize contaminants or introduce new efficient microbial isolates into pesticide-contaminated soils. Azotobacter fungi also play an important role in soil remediation[224]. For the development of sustainable and effective remediation techniques, it is of utmost importance to understand the mechanisms involved in microbial interactions and interactions with soil[232].

In India, agriculture is the one of the main sources of income for the majority of the population. Every time a crop is harvested, a large amount of biomass is produced, called "agrowaste." Several uses of waste biomass and its derivatives have recently been reported[243].

Adsorption is one of the most effective ways to adsorb impurities, namely pesticide residues, onto solid Sorbents[130]. This is a simpler, cheaper, simpler and universal method[256]. Lignocellulosic biomasses such as crops, agricultural waste[247], forest debris, vegetable and fruit peels, are sustainable and abundant resources for recycling into useful products and also act as robust material for the removal of various contaminants such as dyes and pesticides[44]. Pyrolysis of organic feedstocks produces thermal energy, bio-oil and solid products called biochar[172]. The composition of biochar (basically the amount of carbon, nitrogen, potassium, calcium etc.) depends on the starting materials used and the time and temperature of pyrolysis[174]. Therefore, biochar possesses high porosity, surface area, abundant functional groups and highly aromatic structure, which make it a promising candidate for adsorbing contaminants and reducing the bioavailability of pesticides in contaminated soils[143].

The adsorption properties of biochar are mainly determined by the relative amounts of cellulose components, mineral content, particle size and their structures[268]. It is important to use environmentally friendly and sustainable approaches to rehabilitate, restore and maintain soil quality. Plant-based technology (phytoremediation) is another strategy that can serve such applications.

Phytoremediation of contaminated soils mainly depends on the selection of suitable plant material and various soil factors involved[264]. This is another low-cost technology that uses plants to remove pollutants from the environment[22]. Interactions between microbial consortium systems and plant-microbe systems are used to improve the capabilities of actinomycetes in bioremediation[14]. Metal-tolerant plant growth-promoting (PGP) bacteria may enhance phytoremediation of contaminants from contaminated soil. These PGP bacteria aid plant growth and are involved in the uptake of metals by plants[21].

Whole microbial cell immobilization greatly enhances the bioremediation process[64]. In general, bioaugmentation with indigenous microorganisms appears to be more effective and environmentally friendly in soil remediation[112,274]. Better remediation strategies for treating contaminated waste and sites can be derived by considering several key principles: ability of pollutants to biotransform into less toxic products (biotransformation), its chemistry, accessibility of contaminants to microorganisms (bioavailability) and ability to optimize biological activity (bioactivity)[63]. This review highlights the usefulness of microorganisms, lignocellulosic biomass, biochar and plants in remediation of pesticides, widely used in agriculture.

Types of pesticides used in agricultural practices: Pesticides can be classified mainly according to their applications to the target organism viz. herbicides, insecticides, fungicides, rodenticides and pediculicides[5]. Chemical structures of various pesticides used in agriculture have been shown in fig. 1.

Effects of pesticides on biological systems: Ecosystems are adversely affected due to soil and water pollution by the overuse of pesticides[84]. Different chemicals, such as pesticides and heavy metals, can enter the soil through different routes and affect it at different levels[65]. Pesticide can occur at any point in the food chain[23]. Chlorine pesticides are converted or partially decomposed under suitable process conditions. Due to the highly persistent presence of 1,1,1-trichloro-2,2-bis[p-chlorophenyl]ethane (DDT) in the environment, plants, mammals, birds, fish and aquatic plankton consume DDT, which bioaccumulates readily [47], DDT and its decomposition products viz. DDE, TDE, keltan, chlorobenzilate, chloropropylate and acarol have been shown to inhibit both mitochondrial (HBHM) NADH oxidase and bovine centroid succinate oxidase enzymes[179]. The action of pesticides is based on their chemical composition.

The mechanisms of action of pesticides include lipid biosynthesis inhibitors, amino acid biosynthesis inhibitors, plant growth regulators, photosynthesis inhibitors, nitrogen metabolism inhibitors, pigment inhibitors, cell membrane disruptors, seedling growth inhibitors and PPO inhibitors, acetyl-coenzyme A carboxylase, acetolactate synthase (ALS) inhibitor, aromatic amino acid inhibitor, glutamine synthesis inhibitor[211]. Chronic exposure to OPP can cause neurological problems in both humans and animals[52]. The nervous system is affected when pesticides inhibit the enzyme acetylcholinesterase. Skin contact, inhalation and ingestion are likely routes of exposure to pesticides such as monocrotophos[130]. The study suggests that exposure to dichlorane (a fungicide) inhibits spore germination and produces more by-products that irritate the skin[261].

Azoxystrobin, a broad-spectrum fungicide is known to be highly toxic to freshwater fish and estuaries[231]. Methylparathion insecticides kill insects by affecting the stomach and respiratory system, methylparaoxones affect the nervous system by inhibiting cholinesterase and organophosphate pesticides (OPPs) inhibit acetylcholinesterase enzymes, thereby prolonging muscle contractions that kill insects[197]. Various health problems associated with pesticides are neurological[34], carcinogenic[24] and reproductive[38].

Remediation of pesticides through microbes

Soil microorganisms (bacteria, fungi etc.) are primarily responsible for breaking down pesticides. Microbes used pesticides as a carbon or phosphorus source for food and energy by microbes[68,218]. Microorganisms act as engines of remediation of contaminated environments that are essential for soil remediation[18,206,229]. Bioremediation agents are mainly bacteria (57%), enzymes (19%), fungi (13%), algae (6%), plants (4%) and protozoa[183]. *Sphingomonas* and *Porphyrobacter* have been of interest for bioremediation applications of lindane contamination[94]. A popular herbicide (Tordon) based on 2,4-dichlorophenoxyacetic acid (2,4-D) was effectively degraded by *Hydrocarboniphaga* sp., *Tsukamurella* sp. and *Cupriavidus* sp[175]. *Aspergillus sydowii* strain PA F-2 has been observed to be able to degrade trichlorfon (TCF) insecticide. The maximum clearance rate of TCF was about 55.52%[278].

A natural β-triketone herbicide (Leptospermone) was transformed by an isolate called *Methylophilus* sp. LS1[190]. The Carb-PV5 strain, identified as *Acremonium* sp.

(MK514615) was used to simultaneously remove carbofuran and carbaryl from soil[123]. *A. chroococcum* reported for 2,4-dichlorophenoxyacetic acid (2,4-D)[222]. *Pseudomonas aeruginosa* ITRC-5 was used to degrade chlorinated insecticide hexachlorocyclohexane (HCH) isomers in contaminated soil and found that bacteria readily degraded and detoxified HCH isomers[138]. *Pseudomonas* spp. also used for glyphosate degradation[282]. Alkalotolerant *Pseudomonas* sp. strain ISTDF1 was used for degradation of dibenzofuran[105].

Sphingomonas and *Xanthomonads* were found to be the dominant species in repairing trifluralin (TFL)[62]. It has been reported that *C. mexicana* can degrade the atrazine pesticides[116]. *L. crinitus* EF58 lineage resistance under (2,4-D) has also been reported[205]. Synthetic pyrethroid pesticides biodegradation was performed by the halotolerant *Enterobacter ludwigii DWT* [188] and its degradation metabolism was studied by Chen and Zhan[48]. *Bacillus* species. DM-1 converted parathion and methyl parathion to amino derivatives by reduction of the nitro group[267]. Isolated strain *Xanthobacter* spp. CP was able to degrade 2,4-dichlorophenoxyacetic acid (2,4-D). No by-products were detected after treatment[59]. *Pseudomonas* strains degrade 1,1,1-trichloro-2,2-bis(4-chlorophenyl) ethane (DDT) via intermediate formation of 2,3-dihydroxy-DDT, which leads to metacyclic cleavage and finally 4-chlorobenzoic acid is produced[118].

Degradation of bromoxynil octanoate has been demonstrated by *Acinetobacter* sp. strains XB2. Bromoxynil is further converted to 3,5-dibromo-4-hydroxybenzoic acid, finally forming 3-bromo-4-hydroxybenzoic acid[39]. 1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane (DDT) transforms to 4-chlorobenzoate by *alcaligenes eutrophus* A5 via a meta-ring fission product, catalyzed by monooxygenase or dioxygenase enzymes[164]. Cao et al[40] studied the detoxification of Atrazine by *Arthrobacter* sp. C2. Biodegradation of carbofuran by free and fixed cells of *Klebsiella pneumoniae* ATCC13883T was reported. The results showed that fixed cells based on alginate-bentonite PAC completely degraded carbofuranphenol[117]. *Pseudomonas putida* can degrade cadusaphos (organophosphate insecticide)[3].

The dangerous α- and β-isomers of the organochlorine insecticide lindane were effectively degraded by *Streptomyce*s sp. M7 after 7 days of incubation[216]. Degradation of the herbicide bentazone by *Phanerochaete chrysosporium* was also investigated. Manganese peroxidase (MnP) activity plays an important role in bentazone degradation[57]. *Trametes versicolor*, *Pleurotus ostreatus* and *Gloeophyllum trabeum* have been used to degrade organochlorine insecticides such as lindane and endosulfan. The bacterial protein CotA proved to be more efficient than fungal laccase in degrading lindane and endosulfan[246]. Alkalotolerant *Pseudomonas* sp. strain ISTDF1was used for degradation of dibenzofuran. Around 85% of dibenzofuran (200 mg $^{-1}$) was utilized within 36 h at 40°C[105].

Biodegradation of the herbicide diuron by *Lysinibacillus fusiformis* was reported by Reyes-Cervantes et al[189]. Lignin-degrader *Coriolus versicolor* was investigated for the degradation of diphenyl ether herbicides chloronitrofen. It catabolized herbicide into four metabolites: 2, 4, 6-trichloro-3-hydroxy-4'-nitrodiphenyl ether, 2, 4-dichloro-6-hydroxy-4'-nitrodiphenyl ether, NIP and 2, 4, 6-trichloro-4'-aminodiphenyl ether by several reactions such as hydroxylation, oxidative dechlorination, reductive dechlorination and nitro-reduction[97]. A newly isolated *Nocardioides* sp. strain DN36 was able to mineralize a variety of s-triazine herbicides[200].

Carbofuran degradation by *Novosphingobium* sp. KN65.2 (LMG 28221) was examined. Carbofuran was used as the sole source of carbon and nitrogen. The oxygenase gene cfdI and the transporter gene cftA play important roles in degradation[170]. The dibenzo-p-dioxin (DD) and dibenzofuran (DF)-degrading bacterium *Sphingomonas* sp. strain RW1 were marked by the insertion of a mini-Tn5 lacZ transposon. The labeled strain was able to degrade DF and DD at a concentration of 1 mg/g[156]. *Ralstonia eutropha* JMP 134 was able to grow on phenol and 2,4-dichlorophenoxyacetate at higher fixed substrate concentrations according to the nutristat principle[162] . *Pseudomonas putida* (ATCC 17484) was used to study carbazole degradation[273]. *Pseudomonas plecoglossicida* (TA3) strain utilized carbaryl, carbofuran and aldicarb as carbon and nitrogen sources[74].

A. Pseudomonas called IES-Ps-1 IES-Ps-1 strain can be used to treat environments contaminated with pesticides (malathion and cypermethrin)[110]. *Arthrobacter* W1 was able to degrade carbazole (CA)[214]. Biodegradation of the phenoxy herbicides MCPP and 2,4-D in fixed- film column reactors was carried out by Oh and Tuovinen[173].

P. aeruginosa degraded 96% of endosulfan after 288 h treatment. The enzymes such as dehydrogenase, arylsulfatase, dehalogenase enzymes are responsible for the degradation[167]. Biodegradation of diuron and other phenylurea herbicides by soil microbes was investigated. The rate of degradation was in the order of linuron>diuron>monolinuron≫metoxuron⋙isoproturon[113]. The biodegradation of organophosphorus pesticides by cyanobacteria was studied. *Aulosira fertilissima* ARM 68 and *Nostoc muscorum* ARM 221 grew best under organophosphorus pesticides (monocrotophos and malathion)[223]. An anaerobic endosulfan sulfate-degrading bacterium, strain *Rhodococcus koreensis* S1-1, also utilized endosulfan degradation[131]. A new dimethaclon-degrading strain was identified as *Brevundimonas naejangsanensis* J3. Whole cells and extracted enzymes of this strain were able to rapidly remove 75 mg/L dimethaclone in liquid media with over 90% degradation efficiency[275].

Bjerkandera adusta and *Oxysporus* sp were found to be good at degrading three phenylurea herbicides (chlortoluron, isoproturon and diuron)[128]. Strains *Stenotrophomonas acidophila* TD4.7 and *Bacillus cereus* TD4.31 were able to degrade diuron with efficiencies of 87% and 68% respectively[66]. The enantioselective degradation of the herbicide mecoprop[2-(2-methyl-4-chlorophenoxypropionic acid) was investigated by a consortium of bacterial cultures of *Alcaligenes denitrifier, Pseudomonas glycinea* and *Pseudomonas marginalis*. The culture mainly degraded the (R)-(+)-isomer of the herbicide whereas the (S)-(-)- enantiomer was unaffected[237].

Removal of organophosphorus pesticides through *Kosakonia oryzae* strain VITPSCQ3 biofilms was performed in a vertical flow fixed-bed biofilm biofilm reactor[153]. *Rhodopseudomonas capsulata* is used for biodegradation of carbaryl insecticides. MAPKKKs, MAPKKs and MAPKs gene induction were responsible for carbaryl hydrolase gene expression[258]. *Burkholderia* sp. IPL04 has been used for the biodegradation of a refractory man-made pesticide (γ- hexa chlorocyclohexane)[135]. Highly efficient biodegradation of hexadecane was performed by bacterial isolates (*Bacillus amyloliquefaciens, Staphylococcus epidermidis, Micrococcus luteus, Nitrotyreductor aquimarinus* and *Bacillus*). After 48 h of culture, all of them could degrade >80% of the initial hexadecane[76].

Genetically engineered microorganisms (GEMs) could be helpful in the rapid degradation of pesticide residues by adding large amounts of prepared inoculums[217]. Around 82% of dibenzothiophene (DBT) (40 mg^{l-1}) and 57% of carbazole (CBZ) (40 mg^{l-1}) were degraded by *Arthrobacter* sp. P1 strain in 14 days of incubation[204]. It has been observed that strains *Bacillus* sp. CBMAI 1833 and *B. cereus* P5CNB have a better methyl parathion degradation capacity[13]. Mixture of carbamates (CRBs) degraded by newly isolated *Ascochyta* sp. CBS 237.37 was performed[126]. A study on the biodegradation of methylparathion (an organophosphorus insecticide) by *Acinetobacter radioresistens* USTB-04 was conducted and it was observed that benzene ring C-C bond cleavage may be responsible for the biodegradation of MP by *A. radioresistens* USTB-04[72].

Biodegradation of the herbicide 2,4-dichlorophenoxyacetic acid (2,4-D) was performed in a sequence batch reactor. At steady-state operation, complete removal (>99%) of 2,4-D was achieved[147]. The isolated strain of *Agrobacterium tumefaciens* was able to degrade both α and β isomers of endosulfan with equal efficiencies without the accumulation of known toxic intermediates or end products[238]. *P. mendocina* NSYSU was able to completely degrade pentachlorophenol (PCP)[120]. The feasibility of a PAH- and phenol-degrading microorganism (*Pseudomonas putida* ATCC 17484) to degrade carbazole was investigated.

Carbazole can be completely degraded in the presence of 200 mg/l sodium salicylate[144]. *Pseudomonas aeruginosa* (KY781886), *Enterobacter ludwigii (*KX881423) and *Enterobacter cloacae* (KX881513) were used to degrade organophosphate pesticides (chlorpyrifos)[207]. A potent strain PYR-P2 with high pyrethroid degradation was isolated and identified as *Aspergillus* sp. It degraded up to 500 mg L^{-1} of pyrethroid mixture (cypermethrin (CYP), cyfluthrin (CYF), cyhalothrin (CYH)[125].

The natural bacterium *Brevibacillus panacihumi* C17 showed an efficient ability to degrade the toxic fungicide carbendazim (CBZ) (300 mg^{L-1})[119]. Predominantly laccases[32] and peroxidases can efficiently break the bonds in pesticide molecules converting it to lesser toxic metabolites[146]. Strain *Providencia stuartii* JD showed great potential for removing dangerous (dimethaclone) NDPS residues[278]. A novel carboxylesterase gene (estwx) was cloned from *Brevundimonas* sp. strain QPT-2 and overexpressed in *E. coli* BL21 which is responsible for the hydrolysis of ester bond cleavage of aryloxyphenoxypropionate (AOPP) herbicides to form the corresponding acid and alkyl side chain alcohol[262].

2,4-D degradation depends on the enrichment/substrate (E/S) ratio. An E/S ratio of 0.03 showed excellent performance in resolving 2,4-D[266]. *Bacillus thuringiensis* var. Kurustaki (Btk) is a microbial insecticide used for parallel degradation of dimethyl phthalate (DMP). DMP concentrations decreased to 1–3 mg/l with initial concentrations ranging from 100–500 mg/l after 48 h of incubation[37]. *Streptomyces* was able to remove the insecticide methoxychlor (MTX) with the highest removal rates (40% and 76%)[36].

Chlorella sorokiniana UUIND6 was able to degrade 100 ppm of malathion due to high carboxylesterase activity. Under such conditions, higher activities of superoxide dismutase (SOD), ascorbate peroxidase (APX) and catalase (CAT) were observed[165]. Biosorption of the 2,4-dichlorophenoxyacetic acid (2,4-D), 2,4-dichlorophenol (2,4-DCP), 4-chlorophenol (4-CP) and 2-chloro-ethylamino-6-isopropylamino-1,3,5-triazine (atrazine) has been carried out by the mycelium of *Emericella nidulans* and *Penicillium miczynskii*. Results showed rapid adsorption of toxic components on the fungal cell walls surfaces[28]. An aerobic carboxylation step has been identified in the lindane biodegradation pathway of *F. Verticillioides* (isolated from agave tequilana leaves)[87].

Biodegradation of methyl parathion by cells of the marine fungi *Aspergillus sydowii* and *Penicillium decaturense* was studied. *A.sydowii* CBMAI 935 was able to degrade various pesticides in 20 days whereas *P. decaturense* CBMAI 1234 took 30 days to fully decompose[12]. Triadimenol (a fungicide) inhibits fungal ergosterol biosynthesis and affects membrane integrity[212]. *Trametes versicolor* immobilized on Quercus ilex wood chips was originally proposed to remove pesticides (diuron and bentazone)[26].

Malathion biodegradation by *Acinetobacter johnsonii* MA19 was also reported by Shan et al[209]. *Aspergillus flavus* (AF), *Penicillium chrysogenum* (PC), *Aspergillus niger* (AN) and *Xanthomonas axonopodis* (XA) were utilized for degradation of alachlor (chloroacetanilide endocrine disruptor herbicide) with the efficiencies of 17.1%, 5.5%, 72.6% and 82.1% respectively, after 35 days of incubation[6]. Biodegradation of a herbicide mixture (ametrin and 2,4-dichlorophenoxyacetic acid) was performed in a compartmentalized biofilm reactor[41]. The highest carbofuran degradation of 95.40% by *Enterobacter* sp was found at a carbofuran concentration of 92.50 mg/L, a pH value of 6.0, at a temperature of 27.50 °C for the incubation period of 6 days[163]. Triclosan is widely used in personal care products as an antimicrobial agent but it is also considered as contaminant. *Geitlerinema* sp. and *Chlorella* sp. degraded 82.10% and 92.83% of 3.99 mg/L of triclosan at 10 days respectively[235].

95% of herbicide diuron degradation was achieved by *Streptomycetes* strain A7-9 after five days of incubation and no herbicide remained after 10 days[42]. Simultaneous degradation of phenol and n-hexadecane by *Acinetobacter* strains has been observed[226]. Among actinomycete strains isolated from soil, strain CCT 4916 was found is the most efficient and was able to degrade the herbicide diuron *in vitro* up to 37% of applied diuron (100 mg/ kg soil) in 7 days. Manganese peroxidase extracted from *Actinomycete* strain CCT 4916 was responsible for the degradation of diuron[70]. The ability of *Trametes versicolor*, *Pleurotus ostreatus* and *Gloeophyllum trabeum* for the degradation of two organochlorine insecticides (lindane and endosulfan) was investigated. Endosulfan was transformed to endosulfan sulphate by *T. versicolor* and *P. Ostreatus*[246]. Endosulfan biodegradation by *Pseudomonas fluorescens* was reported by Jesitha et al[109].

Freely suspended *Pseudomonas fluorescens* cells could degrade ndosulfan with an initial concentration of 350.24 ± 0.83 μg/L efficiently within 12 days. The photolysis of leptospermone was sensitive to pH. A bacterial strain able to degrade leptospermone was isolated from an arable soil named as *Methylophilus* sp. LS1. Major transformation was identified as Hydroxy-Leptospermone[190]. Chlorinated pesticides such as polychlorinated diphenyls, organophosphorus and polycyclic aromatic hydrocarbons are also detoxified by microbial systems[29]. The bioavailability and mobility of pesticides in soil mainly depends on absorption and desorption mechanisms[185]. Biological treatment is basically aerobic and anaerobic based. Aerobic treatment consists of oxidation, hydroxylation and the cleavage of bond[107]. The mechanisms involved in the microbial interactions that occur during the bioremediation process are of great importance[232].

Glyphosate [N-(phosphonomethyl)glycine CAS#1071-83-6] is one of the most commonly used organophosphate herbicides. Various microbial species have been reported to consume glyphosate as a nutrient for growth[220]. During herbicide degradation, several intermediates namely 4-(2,4-dichlorophenoxy)butyric acid (2,4-DB) and 4-(4-chloro-2-methylphenoxy) butyric acid (MCPB) and phenol were detected. This is probably due to side chain ether cleavage encoded by divergent tfd gene types[221]. Microbial catabolism is considered as the major pathway for the dissipation of herbicides in the environment[100]. Cytochrome P450 enzymes may catalyze the reduction, oxidation, or hydroxylation of the herbicide and thus provide functional groups for further metabolic steps[134].

Remediation of pesticides through Lignocellulosics: Lignocellulosics have wide range of utilization capability. Many lignocellulosic biomass such as bottle gourd peel[251] are used as raw materials for valuable product formation such as cellulases[248]. Agricultural residues can be used as an alternative source for bioethanol production[249]. On the other hand, lignocellulosic biomass is also used to remove toxic components such as pesticides. Verma et al[250] emphasized on lignocellulosic biomass-based dye decolorization. Lignocellulosic agricultural residues are used worldwide as adsorbents to produce activated carbon that removes a variety of pollutants. Adsorption mechanisms may be physical entrapment, strong covalent chemical bonding, weak Van der Waals binding, cation exchange, dipole-dipole exchange, and/or ion-dipole interactions[160].

Remediation of pesticides through lignocellulosic biomass: Adsorption is an effective and inexpensive purification method for removing some of the contaminants harmful to the environment and human health[10,222]. It is a fast, flexible and easy-to-use[20]. Adsorption-based techniques are well suited for the sequestration and chelation of multiple environmental contaminants (such as herbicides and pesticides)[161]. Agricultural and household wastes have also been used as activated carbon sources to adsorb insecticides such as banana peels, dates and coconut fibers[19,91,115]. Mesoporous and microporous rapeseed stem-derived activated carbon (CSAC) has been used to remove 2,4-dichlorophenoxyacetic acid (2,4-D)[17]. Chang et al[43] studied the adsorptive removal of the pesticide methomyl using hypercrosslinked polymers.

The combination of granular activated based adsorption and electrochemical oxidation in 3D electrochemical systems is potentially a very efficient process for the treatment of pesticides MCPA (2-methyl-4-chlorophenoxyacetic acid), MCPP (2-methyl-4-chlorophenoxypropionic acid) and the pesticide transformation product BAM (2,6-dichlorobenzamide)[83]. Coupling with an Actinobacteria consortium consisting of *Streptomyces* sp. for biostimulation and bioaugmentation of sugarcane bagasse A2, A5, A11 and M7 for lindane removal was used by Raimondo et al[184]. Best lindane removal was achieved in bioenhanced + biostimulated sandy soils.

KOH activated carbon sorbent made from the waste biomass of hemp fibers (*Cannabis sativa*) has also been used to remove pesticides[254]. Various lignocellulosic waste biomass that serves as sorbents for pesticide removal are summarized in table 1.

Remediation of pesticides through biochar

Biochar is formed from the pyrolysis of biomass at temperatures below 700 °C and in the absence of oxygen[240]. It was typically made from agricultural and forestry waste, sludge and biomass waste such as livestock and poultry manure. The resulting solids have high carbon content, large specific surface area, cation exchange capacity, nutrient retention capacity and stable structure[195,285]. Biochar contains many compounds (cellulose, lignin, hemicellulose etc.) and polyphenol functional groups that play an important role in binding to organic compounds and metal ions[15,16,132]. Biochar exhibited high thermal stability, an amorphous structure and a highly irregular surface composed primarily of carbonaceous bonds[95]. Biochar, a carbonaceous by-product of pyrolysis, has recognized potential to enhance environmental remediation/restoration.

The degradation of biochar-based pesticides in soil was investigated. Application of biochar to soil improves soil quality through long-term sequestration of carbon[111]. Today, it is used effectively to manage soil by removing pollutants, remediating contaminated soil, reducing greenhouse gas emissions and acting as a soil additive to improve soil health[15,78,149,177]. Biochar has emerged as a promising material for the absorption, desorption and degradation of pesticides in soil by adsorbing pesticides present in the soil and reducing the bioavailability of pesticides in contaminated soil. It is highly effective due to its high porosity, surface area, pH and large number of surface functional groups[143,225]. Biochar-based sorption of highly persistent and ionizable pesticides (imazamox, picloram, terbuthylazine) has also been reported[80]. Rice husk biochar (RHBC) was used as a sorbent for the herbicide metolachlor removal[257].

A dually modified (ultrasonic vibrations and alkali functionalization) novel biochar of *Trapa bispinosa* peel (UFBC) with improved adsorption capacity for chlorpyrifos (CPS) pesticides was synthesized[33]. Impact of hydrochar and biochar amendments on sorption and biodegradation of organophosphorus pesticides was evaluated by Isakovski et al[102]. Addition of hydrochars/biochars increases pesticide retardation 4 to 18 times. Various lignocellulosics biochars used for pesticides removal are summarized in table 2.

Remediation of pesticides through plant

Phytoremediation is a plant-based green technology that reduces the levels of toxic components in the polluted areas. Plant roots are involved in stabilizing contaminated soil, thereby inhibiting the mobilization of toxic metals[136]. There are distinctive types of phytoremediation such as (a) Phytostabilization (b) Rhizodegradation (c) Rhizofiltration (d) Phytodegradation (e) Phytoextraction (f) Phytoaccumulation (g) Phytovolatilization (h) Bioaumentation[56,182,192]. Diazotrophic cyanobacteria can fix nitrogen and carbon from the air, thereby increasing the fertility of contaminated soil. Improved and desirable traits of these microbes can be obtained by genetic engineering[101]. Heavy metal resistant-plant growth promoting bacteria (HMRPGPB) were also used in phytoremediation of HMs. The biochar-HMRPGPB-plant association could provide a promising green approach to remediate HM-polluted sites[54].

Utilization of inexpensive algal biomass (live-dead or immobilized) to remove contaminants has been used and serves as one of the most effective bioremediators[122]. Combined treatment with biochar and arbuscular mycorrhizal fungi (BC + AMF) serve as a better bioremediater for agroecosystem Management[88]. *Suaeda glauca* and *Arabidopsis* plants have been used for phytoremediation of contaminated soils [277]. Plant growth-promoting bacteria (PGPB) are commonly used in phytoremediation for soil and water decontamination[56]. Plants have the ability to absorb contaminated water and remove pesticides from the soil[234].

Remediation of toxic metals contaminated soils by metal-accumulating plants (hyperaccumulator). They perform such activity by the combined action such as metal solubilization in the soil, uptake of the metal by plant, detoxification/chelation and sequestration and volatilization[140]. Cyanobacteria based nanoparticles have been used successfully as a phytoremediator of heavy metals as well as a better nutrient solution for crops[178]. The green microalga *Chlamydomonas mexicana* is used to biodegrade the herbicide atrazine at low concentrations[116]. Phytoremediation of organophosphorus and organochlorine insecticides by *Acorus gramineus* plants was reported by Chuluun et al[50]. The biodegradation of 2,4-D (2,4-dichlorophenoxy acid) can be stimulated by structurally related plant secondary metabolites such as ferulic acid (FA)[158]. Rhizoremediation involves both plants and their associated rhizosphere microbes for remediation[82]. Rhizoremediation is the approach to select the best plant species for soil management and bioremediation.

Soil/Root volume ratio is mostly larger in rhizoremediation[67]. The root microbiome promotes plant growth and also regulates soil fertility[210]. Today, rhizome restoration is becoming a sustainable alternative to remediate contaminated environments by exploiting the symbiotic relationship between plants and associated soil microbes in the root zone[98]. The use of AMF (Arbuscular Mycorrhizal Fungi)-rhizosphere microflora combinations appears to be a promising strategy to enhance plant growth or protect plants against pathogens and other environmental stresses (pesticides accumulation)[259].

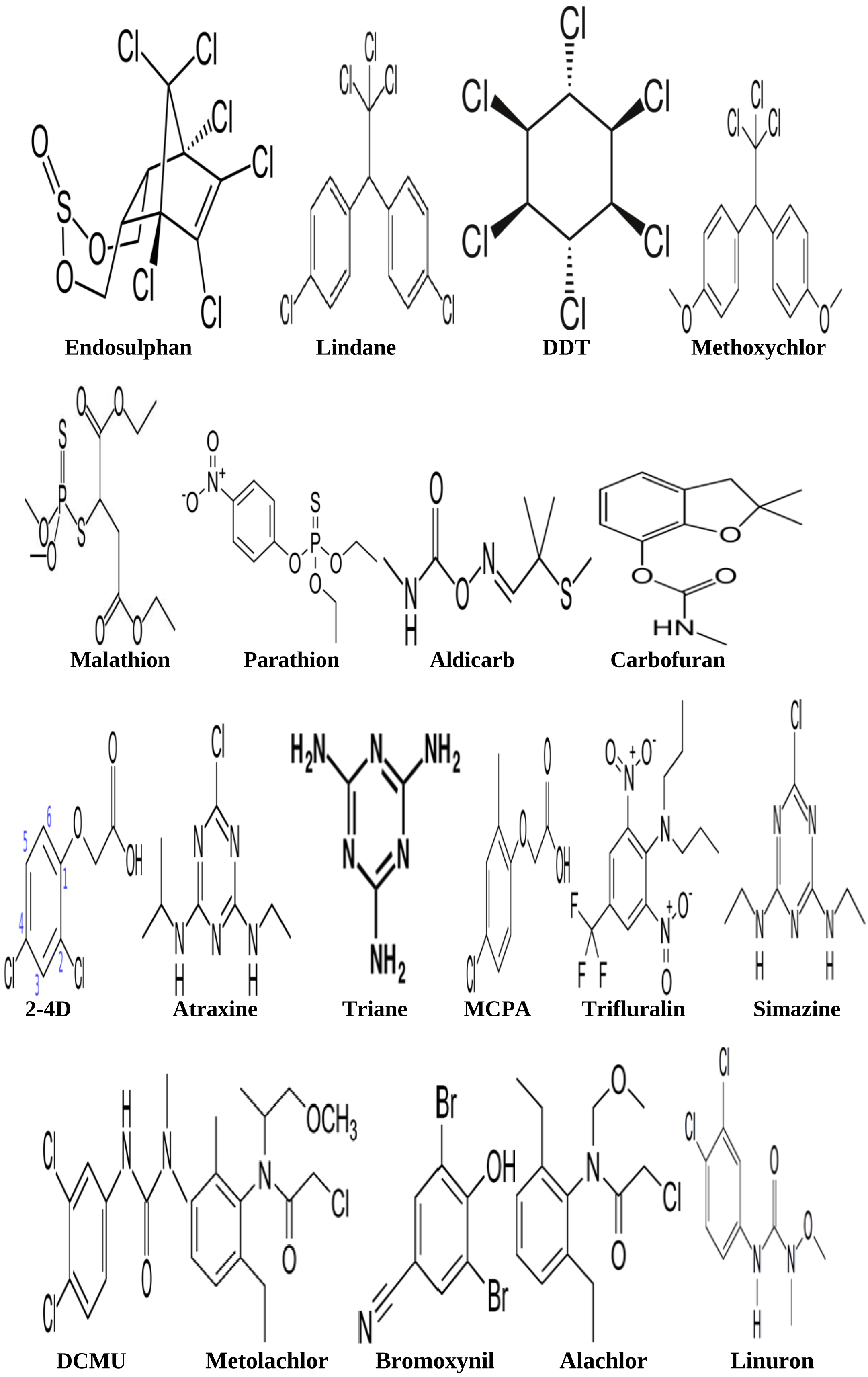

Fig. 1: Chemical structures of various pesticides used in agriculture

Table 1
Various lignocellulosic waste biomass that serves as sorbents for pesticide removal

S.N.	Microbial Biomass/Lignocellulosics	Pesticides	Type	Modification /Treatment	Adsorption	Removal %
1	White-rot shiitake mushroom, *Lentinulaedodes*[244]	2,4-dichlorophenol (DCP)		Vanillin as an activator		92%
2	Argan Nut Shell[273]	Diuron		Hydrothermal Carbonization		95%
3	Olive tree pruning[58]	Dimethoate		Biomixture with soil, vermicompost		(>73%)
		Imidacloprid				(>73%)
		Oxyfluorfen				(>73%)
4	Cucumber (*Cucumissativus*)[90]	Metribuzin		Modified with ZnO nanoparticles (CPZiONp-composite)	200 mg/g	
5	Composted sheep manure (CSM)[75]	Phenylurea	Herbicides			Effective
	Composted pine bark (CPB)					
	Spent coffee grounds (SCG)					
	Coir (CR)					
6	*Phoenix dactylifera*date stones[194]	Organochlorine pesticides (OCPs)		Pyrolysis-chemical activation with phosphoric acid -60%		70–100%
7	Sugarcane bagasse[239]	Carbofuran		Support of magnetite nanoparticles	175mg/g	Effective
		Iprodione			119 mg/g	
	Peanut shell	Carbofuran			89.3 mg/g	
		Iprodione			2.76 mg/g	
8	Pistachio nutshells[92]	Deltamethrin	Insecticide		162.6 mg/g	
9	Groundnut shell[242]	2,4-Dichlorophenoxyacetic acid		Activated carbon by chemical activation (GSAC)	250 mg/g	
10	Brown macroalga (Sargassumsp p)[187]	Chlordecone (CLD)				Effective
11	African Baobab[236]	Diuron	Herbicides		400mgg^{-1}	
12	*Lagenaria vulgaris*[35]	Ranitidine			315.5 mg/g	
13	Date-palm coir (DPC)[186]	2,4-Dichlorophenoxyacetic acid (2,4-DPA)	Herbicide	Carbonization-KOH activation	50.25 mg/g	98.60%
14	Spent substrate after *Pleurotuspulmonarius* cultivation[114]	Chlorothalonil				100%
15	Banana stalks (BSAC) waste[198]	Carbofuran	Insecticide			Effective
16	Maize cobs waste[77]	Carbofuran			149.15mg/g	
17	*Ayous* (*Triplochitonschleroxylon*) sawdust[166]	Paraquat				Effective
18	Coconut frond[171]	Carbofuran		H3PO4 modified		>80%
19	Rice husk[124]	Imazethapyr			0.158×10^3 - 0.636×10^3 μg g^{-1}	
		Imazamox			34.161×10^3 - 166.514×10^3 μg g^{-1}	
20	Date (*Phoenix dactylifera* L.) palm stones[7]	Pesticides			238.1mg/g	
21	Baobab seeds hulls (Andansoniadigitata)[270]	Diuron		Potassium hydroxide-activated	65.7 mg·g^{-1}	
22	Queen palm endocarp (*Syagrusromanzoffiana*)[199]	2,4-Dichlorophenoxyacetic acid (2,4-D)	Herbicide		367.77 mg g^{-1}	95.4%.

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23	Chickpea husk of black gram[8]	Triazophos (TAP)			3.5 ± 0.45 mmol g^{-1}	
		Methyl parathion (MP)			10.6 ± 0.83 m mol g^{-1}	
24	Short hemp fibers[253]	Pesticide		Potassium hydroxide activation		Effective
25	Coconut fiber–compost–soil biomixture[49]	Chlorpyrifos/Carbofuran	Insecticide/ Nematicides			Effective
27	*Trametesversicolor*immobilized onpinewood chips[27]	Diuron				61-94%
28	*Moringaoleifera*seed waste[89]	Chlorpyrifos		Bio-nanosrbent	25 mg g^{-1}	81%
29	*HeracleumPersicum*stems[157]	Paraquat (PQ)				82.20%
		Diquat (DQ)				93.20%
30	ArachisHypogaea (Groundnut) shell[157]	Dichlorvos				>98%
31	Chestnut shells[51]	Pirimicarb, imidacloprid, acetamiprid and thiamethoxam		Pretreated with citric acid		Effective
32	Biomixture composed of coconut fiber, compost and soil[152]	Triazines, triazoles and organophosphates				Effective
33	Activated Coconut Charcoal (AcCoC)[130]	Organophosphoruspesticide monocrotophos			103.9 mg g^{-1}	
34	Tangerine seed activated carbon (TSAC)[256]	Carbamate pesticides				Effective
	Rice straw[46]	Carbofuran		Potassium hydroxide (KOH) activation	296.52 mg/g.	
35	Cinnamon waste[71]	Chlorpyrifos		physical activation in the presence of carbon dioxide	12.37 mg/g	
36	Agriculture waste i.e. stigma maydis[60]	Organophosphorus		Pretreted with concentrated hydrochloric acid		Effective
37	Orange peel[2]	Prothiofos (*O*-2,4-dichlorophenyl*O*-ethyl *S*-propyl phosphorodithioate)			185.9 ± 1.8 mg/ g	91.70%
	Apricot kernel				145.8 ± 2.4 mg g^{-1}	86%

Constructed wetlands (CWs) are widely used for pesticide mitigation. The presence of plants improves pesticide retention in engineered wetlands. Biodegradation processes for four organophosphate pesticides (chlorpyrifos, diazinon, fenthion, dichlorvos) in wetlands have been investigated. The results of this study provide insight into the constructed wetland design for the mitigation of organophosphate pesticides[193]. *Proteobacteria*, *Chloroflexi*, *Acidobacteria*, *Planctomycetes* and *Bacteroidetes* are the dominant phyla and may be involved in the biodegradation of organic pollutants in integrated engineered wetlands.

Removal of pesticides by ICW resulted in an overall reduction in aquatic toxicity[142]. The effect of adding biochar to subsurface flow treatment wetlands (SSF TWs) and the performance of three macrophyte species (*Phragmites australis* subsp. *americanus*, *Scirpus cyperinus* and *Sporobolus michauxianus*) in chlorantraniliprole removal were investigated. Results showed that mesocosms with biochar were very effective in removing CAP mass (90 to 99%)[1]. Removal of four triazine pesticides (simazine, atrazine, terbuthylazine and metribuzine) was investigated by the free-swimming aquatic plant *Eichhornia crassipes* in microcosms of water sediments. Insecticide removal was significantly accelerated by the presence of *E. crassipes*, with removal efficiencies ranging from 66% to 79% after 30 days of treatment[255]. Rapid biodegradation of chlorpyrifos by the psychrophilic plant *Shewanella* sp. BT05 was evaluated. The isolated *Shewanella* sp.BT05 showed rapid CP degradation (94.3%) within 24 hours[86].

An artificial wetland system is used to remove terbuthylazine. A CW planted with common reeds showed the highest removal efficiency (73.7%)[84]. Five wetland plant species (*Viburnum*, *Phragmites australis*, *Iris pseudacorus*, *Juncus effusus* and *Berula electrica*) were planted in mesocosms to eliminate the pesticides imazalil and tebuconazole in saturated artificial wetlands (CW)[233].

Table 2
Biochar used in pesticides degradation

S.N.	Lignocellulosics Biochar	Pesticides	Type	Pyrolysis Temp	Modification /Treatment	Adsorption	Removal %
1	Potato peel biochar[219]	Chlorpyrifos	Organophosphate pesticide				72.06%
2	*Tenebriomolitor* frassbiochar[213]	Thiacloprid (THI)	Neonicotinoid pesticides (NEOs)	750°C	Activated with KOH	155.08 $mg \cdot g^{-1}$	
		Nitenpyram (NIT)				195.86 $mg \cdot g^{-1}$	
		Dinotefuran (DIN)				325.81 $mg \cdot g^{-1}$	
3	Grape pomace-derived[269]	Pesticide cymoxanil				161 mg CM/g	
	biochar (GP-BC)	(CM).				BC	
4	Coconut shell biochar[23]	Diazinon			Activated	9.65 mg/g	
					Phosphoric acid modified	10.33 mg/g	98.96%
	AzardirachtaIndica waste based biochar[181]	Bentazone	Insecticides			79.40 mg/g	
5	P doped biochar from[228]	Triazine				79.6 mg g^{-1}	(>96%)
	corn straw (CSWP)	pesticide (TRZ)					
6	Sugarcane bagasse-derived biochar[103]	chlorpyrifos			Biochar alginate beads	6.25 mg g^{-1}	86%
7	Rice straw biochar (RSBC)[145]	Atrazine					37.5–70.7%)
		Imidacloprid					39.9–77.8%)
8	Corn straw powder[141]	Organophosphorus		700 °C	Magnetization and		Effective
	biochar	pesticides			carbonization		
9	Walnut Shell Biochar (WSBC)[245]	Chlorpyrifos (CP)					86.64%
10	Neem (*Azadiracht a indica*) chip biochar[148]	Mancozeb	Fungicides	900°C		187.68 mg/g^{-1}	
11	Biochar from grapefruit[73] peel powder	Chlorpyrifos			Modified with Fe3O4 and CdS nanoparticles		97%
12	Bagasse based biochar[104]	Chlorpyrifos (CPS)				3.20 mg g^{-1}	89%
13	Biochar from sawdust of the wood forest[111] species *Cedrellafissilis*	Atrazine	Herbicides			7.68 mg g^{-1}	
14	Biochar derived from peanut shell[281]	Imidacloprid		700^0C			62.00%
15	Corn cob biochar[31]	2,4dichlorophenoxy acetic acid (2,4-D)	Herbicide	600°C			Effective
16	Greenwastebiochar[284]	Atrazine and Simazine	Triazine pesticides	450 °C .			Effective
17	Rice husk biochar (RHBC)[99]	Metolachlor.	Herbicide	750 C			Effective
18	Biochar from Bark residues of the forest species *Cedrelafissilis*[96]	Atrazine			Physically activated	3.44 mg g^{-1}	
					Zinc chloride (ZnCl2) activation	2.70 mg g^{-1}	
19	Biochar from *Prunusserrulata* bark[169]	Atrazine				63.35 mg g^{-1}	70%

20	Biochar from *Nepheliumlappaceum* (Rambutan) fruit peel waste[25]	Organochlorinepesticides (OCPs)			Fe0-Biochar nanocomposites		96–99%
21	Macauba endocarp biochar[252]	Atrazine	Herbicide		Activated with K_2CO_3		90–98%
22	Rice straw biochar (RSB)[55]	Monuron					41.90%
		Diuron					25%
		Linuron					56.80%
23	Wheat straw biochar[280]	Atrazine			$ZnCl_2$ or H_3PO_4 Pretreated		Effective

The removal efficiency of the pesticide chlorpyrifos (50 and 500 μg^{L-1}) of five wetland plant species (*Cyperus alternifolius, Canna indica, Iris pseudacorus, Juncus effusus and Typha orientalis*) was reduced in the recirculating vertical wetland system (RVFCW). Plants can enhance chlorpyrifos removal through enhanced biodegradation in the system[260].

Studies also point to the use of aquatic plants such as *Eichhornia crassipes*, duckweed and *Elodea canadensis* for wastewater treatment because of their photosynthetic activity, ease of harvesting and high rates of absorption of pollutants[45]. Liu et al[142] studied the removal of organophosphorus pesticides in constructed wetlands (CW). OPP removal in CW includes phytoremediation (plant uptake, plant accumulation, plant volatilization and plant degradation) substrate adsorption or sedimentation and biodegradation.

Future prospects

Another recent approach to degrade toxic pesticides in the ecofriendly way is the utilization of microbial enzymes in the process[229]. Poultry manure and green waste based compost used as nutrient supplements for soil reclamation. Composting can be a very practical approach to material management in terms of transport reduction[9,154]. For the yield improvement, crop quality improvement and to meet the global food demand, pesticides are widely used in agriculture but they pose potential risks to human and ecosystem health[168]. Municipal solid waste compost be used for improving physical, chemical and biological properties of both saline and sodic soils[155]. In surfactant-based remediation techniques, surfactants can improve desorption of contaminants from soil and bioremediation of organic matter by increasing the bioavailability of contaminants[150].

The degradation of nitroaromatic pesticides was studied using zerovalent iron powder. Finally, the dinitro groups of 2,6-dinitroaniline herbicides were reduced to the corresponding diamines. The integration of iron powder with hydrogen and quinone-producing microbial technology can be viewed as an efficient approach to remediation[127]. Thermal desorption is one of the commonly used methods to remediate contaminated soil[279]. Constructed wetlands (CW) and bioremediation systems (BPS) were used to remove the pesticide terbuthylazine from water bodies of agricultural origin[81].

Fenton-like (FL) hybrid nanoparticles (NPs) induced by microwave (MW) irradiation were applied for the degradation of methomyl pesticides. 91% of methomyl was removed in 8 min under MW irradiation based on C300-based MW/FL system[241]. ZnO/SnO_2 nanocomposite as a photocatalyst for the degradation of triclopyr removal was synthesized by Yadav et al[263]. A combined approach based on advanced oxidation process, anodic Fenton treatment (AFT) and a microbial consortia were used to degrade metribuzin [4-amino-6-tert-butyl-3-methylthio-1,2,4- triazin-5(4H)-one]a broad-use triazinone herbicide[202]. Solar simulator irradiation was used to degrade organophosphorus pesticides (OPs), namely malathion and parathion[283].

Abiotic degradation of 2,4-D is catalyzed by dissolved humic substances at neutral pH[180]. Humic acid graft copolymer (PSt-g-HA) was prepared for the biosorption of pesticides (parathion-methyl, carbaryl and carbofuran) by graft copolymerization of humic acid (HA) and styrene. The sorption capacity of organic pesticides on PSt-g-HA increased from 64.1% to 95.2%[265]. A synthetic nanocomposite CO_3O_4/MCM-41 based on green tea leaves has a large surface area, low bandgap energy (1.63 eV), typical spherical morphology and can be used for environmental remediation[196]. A hybrid photosensitizer (RB-HNT) consists of rose bengal (RB) incorporated into halloysite nanotubes and is used for the photolysis of phenol-based pesticides[30].

A green gamma (γ) radiation technique is used for the synthesis of quaternary amine-functionalized polymer (NPGMA) nanoadsorbent resins. The selectivity coefficient of NPGMA for malathion is 34- to 46-fold higher than that of phenol [271]. Removal of atrazine herbicide from water by adsorption onto iron nanocomposite materials was reported by Ali et al[11]. A novel green nanocomposite is based on silica gel containing rice husk and reducing agent (peach leaf green extract) for the application as a green nanocomposite photocatalyst in the degradation of acephate insecticides[4].

Sustainable remediation approaches understand the nature, composition, properties, sources of pollution, nature of the environment, fate, transport and distribution of contaminants, degradation mechanisms, interactions and relationships with microbes, influencing endogenous and exogenous factors[176] . Mixed microbial cultures can break down harmful organics into less toxic components by using them as the sole carbon source[129].

Advanced OMIC techniques, especially genomics and metabolomics analysis, help identify invasive microbes and discover mechanisms involved in interspecies interactions for the remediation[232]. Genetic engineered microbes are more likely to be repairable by various genetic approaches to get better output such as: better enzyme specificity, development of new metabolic pathways and improved bioprocesses[121]. Phytoremediation has emerged as an environmentally friendly, inexpensive and promising technology for recultivating contaminated land[159]. The extensive use of pesticides to increase agricultural productivity has resulted in soil and aquatic ecosystem contamination.

The use of plants to regenerate pesticide-contaminated sites (phytoremediation) is a new and proven method[139,208]. *In situ* and *ex situ* biodegradations have proven to be highly effective methods for remediation of organic pollutants from soils and aquifers[61]. Genetically modified plants have been used in phytoremediation strategies to improve the condition of contaminated soils and natural environments[192].

Aquatic macrophytes not only stabilize the function and structure of freshwater ecosystems, but are also important ecological components in many ecosystems that can remove pollutants from the aquatic environment[255]. Bioherbicides are used as an environmentally friendly alternative to synthetic herbicides (for weed control) because they have fewer side effects on human health and ecosystems[190].

Olive mill effluent (OMW) is a major source of pollution due to its high organic load, phytotoxicity and antimicrobial properties. A potential use of olive mill effluents as a biopesticide for crop protection[69]. Vermicompost is an alternative to biobed bioremediation systems (BBSs). Improved biodegradation of tebuconazole, oxyfluorfen, and metalaxyl was achieved by such bioaugmentation[42].

Conclusion

Modern agriculture uses pesticides to increase productivity and profits. Therefore, pesticide treatment from soil and water sources is an important area of research due to their high concentrations and refractory properties. Agricultural soil contamination has devastating consequences on a variety of ecological and environmental problems, leading to entry of contaminants into the food chain, soil degradation and stunting of plant growth. Soil and water pollution are the biggest problems and need sustainable solutions.

Adsorption by agricultural sorbents is highly effective in treating various pesticides at high concentrations. Microbial remediation and lignocellulosic waste biomass-based adsorption are promising technologies for remediation of pesticide-contaminated soils and water bodies. These environmentally friendly and effective removal strategies harness the amazing ability of microorganisms and lignocellulose to detoxify toxic contaminants (pesticides) or convert them into harmless secondary compounds.

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