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*Review Article*

# Role of Microbes and Nanomaterials in the Removal of Pesticides from Wastewater

**Arpita Roy**,[1] **Madhura Roy**,[2] **Saad Alghamdi**,[3] **Anas S. Dablool**,[4] **Angham Ahmed Almakki**,[5] **Ismat H. Ali**,[6] **Krishna Kumar Yadav**,[7] **Md. Rabiul Islam**,[8] **and Marina M. S. Cabral-Pinto**[9]

[1]*Department of Biotechnology, School of Engineering & Technology, Sharda University, Greater Noida, India*
[2]*Centre for Translational and Clinical Research, School of Chemical and Life Sciences, Jamia Hamdard University, India*
[3]*Laboratory Medicine Department, Faculty of Applied Medical Sciences, Umm Al-Qura University, Makkah, Saudi Arabia*
[4]*Department of Public Health, Health Sciences College at Al-Leith, Umm Al-Qura University, Makkah, Saudi Arabia*
[5]*Department of Laboratory and Blood Bank, Security Forces Hospital, Makkah, Saudi Arabia*
[6]*Department of Chemistry, College of Science, King Khalid University, P.O. Box 9004, Abha 61413, Saudi Arabia*
[7]*Faculty of Science and Technology, Madhyanchal Professional University, Ratibad, Bhopal 462044, India*
[8]*Department of Pharmacy, University of Asia Pacific, Dhaka, Bangladesh*
[9]*Geobiotec Research Centre, Department of Geosciences, University of Aveiro, 3810-193 Aveiro, Portugal*

Correspondence should be addressed to Arpita Roy; arpita.roy@sharda.ac.in and Md. Rabiul Islam; robi.ayaan@gmail.com

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Pesticides are a class of xenobiotic compounds that are recalcitrant and show persistence in the environment for a longer period of time. Research studies have linked their potential for mutagenicity, teratogenicity, and carcinogenicity. The accumulation of pesticides in water sources due to runoff from agricultural lands has posed a serious threat to the biota of the water ecosystem as well as to the human population. Long-term exposure to pesticides can cause neurological disorders, reproductive complications, cancer, immunological, and pulmonary diseases. The use of pesticides has dramatically surged in agricultural as well as nonagricultural practices. Tons of pesticides are applied in the fields, but a limited amount reaches to the target organism while the rest is wasted and gets accumulated in soil or ends up in water sources like groundwater or river, which results in eradication of nontarget organisms. A variety of pesticides are used for pest management, such as organochlorine (DDT), carbamates (carbaryl), organophosphates (malathion), and pyrethroids (pyrethrins). These chemicals are highly toxic to flora and fauna because of their nonbiodegradable and persistence nature. Biomagnification of pesticides usually leads to cause various problems in human beings. Organochlorines like DDT have been banned in many developed countries due to these reasons. Therefore, the removal of pesticides from wastewater and natural water sources is of utmost importance. Conventional methods possess various limitations; therefore, there is a requirement of an alternative method which can efficiently remove these pollutants from the wastewater. In this review, environmental impacts and health-related complications of pesticides and microbial remediation approaches and use of different nanomaterials in the pesticide removal have been discussed.

## 1. Introduction

In agriculture, crops are attacked by various pests which include organisms such as insects, fungi, bacteria, unwanted crops (weeds), and some animals. If the pests remain uncontrolled, they would destroy and damage the crop and thus lead to substantial economic loss for the agronomist. Pesticides have come to aid in the agricultural practices as they are able to eliminate, repel, mitigate, and control the pests attacking the crops. There is a diverse range of synthetic as well as natural compounds which fall under pesticides, and depending upon the target organism, they fall under insecticides, herbicides, etc. The largest demand of pesticides is generated by agriculture which accounts for 85% of the whole production worldwide, and the rest is used for other purposes like control of seasonal vector-borne diseases and pest removal in domestic settings as well as for industrial setups [1]. A huge growth in the human population worldwide has tempted the excessive use of pesticides to produce the large appetite for food crops, but compromising the quality of the product due to the toxicity of pesticides and their detrimental effects on human health. Since the Indian economy is highly dependent on agriculture, the consumption of pesticides in India is the highest among South Asian countries [2].

Pesticides consist of a broad range of synthetic and natural compounds that are widely used for pest control. These are organochlorines, organophosphorus, phenoxy derivatives, pyrethrins and pyrethroids, triazines, carbamates, dipyridyl derivatives, glycine derivatives, chloroacetanilide, dithiocarbamates, benzimidazoles, and other miscellaneous compounds. The composition of some pesticides with their exposure and health effects is elaborated in Table 1.

Conventional methods of removal of pesticides have some limitations, like that they are suitable for only some specific substrates, they are highly dependent on chemical properties such as charge, hydrophobicity, size, and molecular weight of the chemical compound, which limits their selectivity, higher machinery cost, and difficult in situ remediation, and also, process of removal requires energy input and other chemicals, making it worse for the environment as well. Microbial remediation does not possess this kind of limitations as microbes can be acclimatized to a number of chemicals by various means. Microbial remediation is cheaper if done in situ and one microbe can be used for different pesticide removal, thus increasing the range of chemicals which can be removed. Therefore, in this review, a discussion of the effects of pesticides on human health as well as the environment has been done. Further various conventional methods, their limitations, and microbial remediation approach for removal of pesticides have been discussed.

Nanotechnology is one of the most emerging and widely studied fields because of its extensive applications. Nanoparticles (NPs) are the most basic constituents of nanotechnology. The size of NPs varies between 1 and 100 nanometers (nm). NPs are usually made up of metal or metal oxides, carbon, and other organic substances [3]. NPs demonstrate distinctive biological, physical, and chemical characteristics as compared to the relative bulk particles, due to the increased mechanical strength, and the enhanced ratio of surface area to volume, and they also demonstrate better stability or reactivity in a chemical reaction. As a result of the aforementioned characteristics of NPs, they have varied applications. NPs exist in different shapes, sizes, and dimensions [4]. The different types of dimensions of NPs are (a) zero-dimensional: like nanodots, length, breadth, and height have a specified single point; (b) one-dimensional: these types of NPs have only a single parameter, like graphene; (c) two-dimensional: in such NPs, they possess both length and breadth, like carbon nanotubes; (d) three-dimensional: these types of NP possess length, breadth, and height, as in gold NPs.

NPs can be of different shapes and forms such as conical, spherical, hollow, tubular, spiral, cylindrical, and flat. They can have a regular or uneven surface. Various NPs can be either crystalline or amorphous with unbound or clustered single or multicrystal solids [5]. Various methods are employed for the synthesis of NPs in order to either develop or enhance their characteristics and make their production cost-effective. Few methods are altered to enhance the mechanical, chemical, optical, and physical characteristics of NPs [4]. Thus, NPs are widely used in biotechnology and pharmaceuticals, electronics and communication, automobiles and machinery, agriculture, chemical processes, and environmental testing. Nanotechnology provides the potential for a future that is both clean and renewable.

## 2. Environmental Impacts of Pesticides

The application of pesticides has been associated with merits as well as demerits. They increase crop production as well as yield by eradicating the pests feeding on the crops. Without pesticides, crop production would decrease by 40% worldwide due to the pest attacks. They also help in the reduction of insect-mediated aflatoxin contamination of crops which is a carcinogenic leading to liver cancer, reduction in immune response, and lesser growth in children. They also help in prevention of disease outbreaks (e.g., malaria), protect the farm land and forests from invasive pests, and help in controlling the population of bugs. Despite all these benefits associated with them, their production and use in many countries have been banned because of many complications associated with them.

An ideal pesticide should only mediate killing of its target pest, but nontarget organisms usually get affected by them as well. Pesticides after their application can be taken up by the target organisms or they might get dissipated into the groundwater which could ended up in the surface water bodies such as rivers, lakes, or ponds, volatilize into atmosphere, or taken up by the soil and finally reach to nontarget organisms. Their usage poses a serious threat to flora and fauna biodiversity, disturbs the food webs, and has a serious implication on the ecosystem. Usually, pesticides are applied by sprayer in the form of volatilized material which evaporates into the air, increasing the area in which it could spread and, in this way, they can affect nontarget organisms as well. Herbicides, insecticides, etc. are applied in this way, and thus, they cause more harm to nontarget organisms than to target organisms. A variety of aquatic as well as terrestrial animals and plants are under serious threat due to the unrestricted use of pesticides. Some exotic species such as the bald eagle, osprey, and peregrine falcon have experienced survival threats due to overconsumption of pesticides [6]. Moreover, soil, air, and water are subjected to pollution and toxicity as they are the end point where these chemicals ultimately runoff. The bioaccumulation of pesticides in the case of lipophilic pesticides and the seepage of pesticides into groundwater indicate a major issue of their removal from

Table 1: Pesticides with their composition, health effects, and associated properties.

| Pesticide | Composition | Exposure | Effects on human health | Physical and chemical properties |
|---|---|---|---|---|
| Organophosphorus | Organic carbonaceous and phosphoric acid derivative | Easily absorb by the skin, lungs, gastrointestinal tract (GI), and conjunctiva and metabolized by cytochrome P450 in the liver | Muscarinic syndrome, nicotine syndrome, effects on the CNS, teratogenic and carcinogenic | Most are polar, highly stable, and water soluble |
| Organochlorines | Organic carbonaceous compound with cyclodiene ring | Absorption via skin, GI tract, and lungs, ingestion of contaminated food, and inhalation | It has been linked to diabetes, cancer, asthma, and growth disorder in children | Lipophilic, polar, and show high persistence with long half-life |
| Carbamates | Organic compound with general chemical formula RHNCOOR, a derivative of carbamic acid | Absorption through the gastrointestinal tract, lungs, and skin | Lesser CNS symptoms, abdominal pain, behavioral change, diarrhea, vomiting, urinary incontinence, dyspnea, bronchospasm, bradycardia, hypoxemia, etc. | Polar compound, water-soluble, and have potential chemical reactivity |
| Pyrethrins and pyrethroids | Natural compounds extracted from *Chrysanthemum cinerariaefolium* and pyrethroids are synthetic derivatives of pyrethrins, chemical structure contains an acidic and alcohol moiety and an ester bond in the center | Show little cytotoxicity but hyperexcitablity, target voltage-gated chloride, sodium and calcium channels, nicotinic receptors, GABA-gated chloride channels, metabolized by CYP450 | They have known to cause asthma and rhinitis (act as an allergen to respiratory system) as well as contact dermatitis | Readily degradable in the presence of light (pyrethrins) |
| Triazines | Derivatives of a six-membered heterocyclic compound (s-triazine) with substitution at positions 2, 4, and 6. | — | Human poisoning is rare and might produce local irritation | — |
| Dithiocarbamates | Synthetic derivatives of S-containing dithiocarbamates (either dimethyldithiocarbamate or ethylenebisdithiocarbamate) in conjugation with metallic salts of manganese, ferric, or zinc | Absorption is slow | Exposure for longer period might produce adverse effects; metabolites are carcinogenic | Less phytotoxic and have more stability |
| Phenoxy derivatives | Consist of an aliphatic carboxylic acid group in conjugation with either an aromatic ring (methyl substituted) or chloride | Absorption in GI tract, lungs, negligible in skin | CNS damage, teratogenic, shows hepatotoxicity and carcinogenicity, renal failure, hyperthermia, birth defect, etc. | Half-life is less (1-3 days), are easily hydrolyzed and decomposed |
| Dipyridyl derivatives | Dipyridylium quaternary ammonium derivatives | Tissue damage in the lungs, liver, and kidneys | Induction of pulmonary injury, hypoxemia, and edema hemorrhage, necrosis in the GI tract, liver, kidney tubules, and lungs | Highly toxic, exposure results in the production of radicals which damage lipid membranes. |

these sources, as ultimately the human race would face the consequences of this pollution.

Because of the lower biodegradability and more stability of these chemicals, they usually tend to accumulate in the ecosystem as well as in the food webs posing a daunting hazard for the higher-order organisms in the food webs, i.e., predators. The aquatic ecosystem has suffered a lot of damage due to the excessive use of these chemicals. Pesticides usually end up in the aquatic ecosystem by drift or leaching through the soil, contaminating ground water or by direct application into the water source like to control waterborne pests. Fish and aquatic plants are affected due to the toxicity of these chemicals, as well as the dissolved oxygen level is decreased, leading to oxygen stress. Exposure of these chemicals could occur by three [6]: dermally (absorption through skin), orally (entry via mouth), and breathing (entry via gills). The runoff of herbicides in rivers causes the eradication of aquatic plant life, which is solely responsible for 80% of the oxygen supply in this ecosystem, leading to the death of various fish and animals [6]. Atrazine, a triazine derivative, is highly toxic to some species of fish, and it has been observed that it might also secondarily affect the immune system in amphibians. Loss of reproductive potential [6], habitat loss for various amphibians, high mortality of aquatic organisms, and changes in abundance and composition of planktons are some of the potential impacts of pesticides.

Terrestrial biodiversity is also under threat due to the overconsumption of pesticides. Nontarget plants are the worst affected by the sprayed herbicides. Plants exposed to glyphosate has increased vulnerability to diseases and also poor quality of seeds. Reduction in the productivity of nontarget plants, wildlife, and forest plants has been observed even at lower doses of sulphonamides, imidazolinones, and sulfonylureas [7]. Broad-spectrum pesticides such as organophosphates, pyrethroids, and carbamates have affected the population of beneficial insects such as beetles and bees that are crucial for growth in plants and pollination. Insecticides like neonicotinoids have resulted in a decline in demography of honey bees leading to lesser crop production as well as a sharp decline in the production of honey and bees wax which is consumed commercially in various cosmetic products. Consumption of pesticides like DDT, organophosphates, has led to accumulation of these chemicals and its metabolites in birds resulting in high mortality as well as reproductive loss in them. Plants act as a source of food for humans and are the consumer of carbon dioxide, and as the draining of pesticides in the soil affects plant production as well as the microbiota of the soil, which is essential for the requirement of plant nutrients, there are serious complications associated with this. The reduction of soil fertility and degradation of soil quality due to the shrinking population of soil-dwelling microbes is an important environmental impact of pesticides.

Fungicides like dinitrophenyl and chlorothalonil have been reported to obstruct the nitrification and denitrification process done by microbes. Symbiotic or cooperative relationship among mycorhizal fungi and plants roots are also inhibited by pesticides [8]. The friend of the farmer, earthworms have also seen a huge decline due to the pore water of pesticides in the contaminated soil. The neurotoxic effects of some fungicides and insecticides have not spared earthworms, and there has been a substantial loss of soil fertility due to a smaller population of earthworms in farmland soils [9]. It has been observed that neonicotinoids could accumulate in the soil and destroy the population of earthworms such as *Eisenia foetida* from the soil.

Pesticides have also found to trigger or cause bronchial hyperactivity or asthma. These xenobiotics may cause inflammation, irritation, endocrine disruption, or immunosuppression leading to exacerbation of asthma in exposed individuals. An investigative research by Raanan et al. [10] concluded that organophosphates exposure in early life could lead to childhood asthma [10]. Herbicide pendimethalin and insecticide aldicarb were reported to positively worsen the asthma in active patients.

Farmers in Iran reported having a substantial high risk of developing acute leukemia due to exposure to pesticides. Prenatal exposure or exposure in pregnancy to household pesticides was found to be positively linked to development of childhood leukemia. Risk of neurobehavioral defects or cognitive effects were reported to be higher (3 fold increase) in individuals who had these three metabolites hexachlorobenzene, transnonachlor, and p,p′-DDE in their plasma [11]. Other effects of pesticides such as organophosphorous could be impairment of male reproductive system and reduction in fertility by inhibition of spermatogenesis, decreasing sperm count, density and reduction in sperm motility or viability, mass reduction in testis, DNA damage in sperm, and morphological abnormality in sperm.

## 3. Conventional Methods of Pesticide Removal

Pesticide removal is the process by which the contaminated soil or water is purified in which these chemicals are degraded or filtered or extracted out. In conventional methods, chemical oxidation, ozonation, Fenton oxidation, photocatalytic degradation, photochemical degradation (such as UV radiation), coagulation and flocculation, electrocoagulation, incineration, electrooxidation, reverse osmosis, adsorption, nanofiltration, membrane distillation, and electrodialysis are used.

Advanced oxidation process (AOP) or chemical oxidation process takes advantage of highly reactive hydroxyl radical for pesticide degradation. The hydrogen is extracted from the organic substrates by the free hydroxyl radicals or in case of double-bond electrophilic addition takes place [12]. There is another reaction of free radicals with the oxygen molecule to form a peroxyl radical, which ultimately leads to complete mineralization of the pesticide by undergoing various stepwise oxidative degradation reactions. Moreover, phenols substituted with halogens could also be attacked by hydroxyl radicals.

*Ozonation* deploys ozone gas to oxidize and degrade xenobiotics. Ozone is a strong oxidizing agent that attacks an organic substance at low pH, or it can also be used to generate hydroxyl radicals by modifying pH, and thus, hydroxyl radicals can do their part [12]. A recent study on the removal of pesticide residues from rice grains reported that residues of deltamethrin (92.7% removal) and bifenthrin (91.1% removal) residues were effectively removed from stored rice grains using ozone as a degrading agent [13].

Hydroxyl radicals can also be produced for the oxidation of organic pesticides at acidic pH using *Fenton's reagent*, which represents a mixture of hydrogen peroxide and ferrous or ferric ions [12]. Fenton oxidation was investigated to remove methyl parathion, and various parameters (mentioned them) were studied to identify their role in degradation [14]. Fenton oxidation was reported to be able to remove 74-92% of methyl parathion.

*Photocatalytic* process deals with the use of a catalyst such as $TiO_2$, $Fe_2O_3$, ZnO, ZnS, and CdS, which gets activated by absorption of photon and further helps in oxidation of organic compounds. Heterogeneous $TiO_2$ in combination with UV has been mostly studied for its degradative potential. Phenoxy derivatives 2,4-dichlorophenoxyacetic acid and 2,4-dichlorophenoxypropanoic acid were reported to be effectively degraded using heterogeneous $TiO_2$/UV at

283 nm, and degradative potential was observed to be directly proportional to the concentration of Ti in pillared clay [15]. Mepiquat chloride was reported to be completely degraded by using heterogeneous $TiO_2$P-25 where UVA was at pH 3, and degradation rates were higher as compared to homogeneous photocatalytic process.

*Coagulation* is a destabilization process in which a coagulant is added such that it destabilizes a stable charged particle.

*Flocculation* is the subsequent step of mixing that promotes accumulation of microflocs leading to the sedimentation of coagulated particles. In a study, four pesticides (dieldrin, aldrin, bentazon, and atrazine) were taken, and their removal by coagulation-flocculation was investigated using different concentrations of aluminum sulphate as a coagulant. Removal of pesticides was below 50% for all the investigated doses, and hydrophobicity of organic compound was observed to be directly related to the removal efficiency of treatment. A combinatorial approach of coagulation and fenton oxidation was reported to be more effective in wastewater treatment of nonbiodegradable pesticides. Polyferric chloride was deployed as the coagulant which was used as the first step of treatment, resulting in removal of 58% chemical oxygen demand and also lesser hydrogen peroxide requirement in the consecutive fenton oxidation step [16].

*Electrocoagulation* is a modified coagulation process in which an electrocoagulation cell (like an electrochemical cell) is used to destabilize an emulsified, suspended, or dissolved contaminants in wastewater by passing an electric current into it. In this process, electrodes of aluminum or iron are used, and wastewater acts as an electrolyte. Hydrolysis of water takes place at cathode resulting into the formation of molecular hydrogen and hydroxyl groups, and simultaneously, dissolution of metal from the anode takes place to produce coagulant in situ [17]. Released metal ions initiated the coagulation by neutralizing surface charges on the suspended solids [17]. The electric field also helps in electrolysis, ionization, hydrolysis, and free radical formation which also helps in decontamination of pollutants.

*Reverse osmosis* (RO) technology has been used for ground water treatment contaminated with organochlorines. Membranes used were of RO98pHt polyamide, and removal rate was observed to be 98.4-99.7% for pesticides, and dissolved solids were reduced from $1.35\,g/dm^3$ to less than $0.05\,g/dm^3$ [18]. Coupling of electrodialysis to desalinate wastewater and nanofiltration could improve the retention of pesticides on NF membranes [19]. Other conventional methods like electrooxidation and membrane distillation are also used for the removal of pesticides.

## 4. Microbial Remediation of Pesticides

Despite the advances in the conventional approach, researchers are discovering other means to degrade the pesticides in the environment. Microbial remediation uses microorganisms to eradicate these compounds from the contaminated source. Certain bacterial species and their strains, actinomycetes, fungi, and algae, have been documented to show degradation of pesticides. Microorganisms utilize these chemicals as a source of their energy by metabolizing them and directing their intermediates into energy generation pathways like Krebs cycle. These microbial strains have been enriched, cultured, isolated, and screened from various rivers, sewage, and soil [20]. For degradation of endosulfan, Kafilzadeh et al., [21] isolated bacterial strains from water samples, and they found five bacterial genus *Acinetobacter*, *Alcaligenes*, *Klebsiella*, *Bacillus*, and *Flavobacterium* capable of degrading endosulfan [21]. Various bacteria show the ability to degrade pesticides, which include *Pseudomonas*, *Bacillus*, *Alcaligenes*, *Flavobacterium*, *Klebsiella*, *Thiobacillus*, *Escherichia coli*, *Bacillus licheniformis*, and *Clostridium*. Various algae that are capable to degrade pesticides include Diatoms, *Chlamydomomas*, green algae, and microalgae, and fungi include *Anthracophyllum*, *Cladosporium*, *Rhizopus*, *Aspergillus fumigatus*, *Aspergillus*, *Penicillium*, *Mucor*, *Fusarium*, *Mortierella* sp., and *Trichoderma* sp. [20].

Bacteria are in the forefront for bioremediation purposes, as they are highly adaptive and liable to undergo mutation rapidly, thus acclimatizing themselves according to the demand of the vicinity environment. Pesticides are used as a nutrient by microbes and go through enzymatic reactions to produce carbon dioxide and water. These chemicals are first taken up by the microbes, followed by sequential attacks of metabolic enzymes, and they are completely degraded into lesser toxic or nontoxic compounds. These enzymatic reactions involve oxidation (epoxidation, N/P/S-oxidation, hydroxylation reactions on aromatic ring, aliphatic chain, or N-hydroxylation, oxidative dehalogenation, oxidative deamination, and oxidative dealkylation), reduction (reductive dehalogenation, and reduction of nitro or quinone), hydrolysis of ester bonds, condensation, decarboxylation, dehalogenation, dehydrogenation, and others [22].

Microbes could perform mineralization of compounds or cometabolism for degradation. Mineralization refers to the conversion of organic compounds into inorganic one by microbial enzymatic systems. For a molecule to undergo mineralization, it should have resemblance to the natural ligand (should be analog to the natural compound) against which the microbes have enzymes to act upon it [20]. Microbes use this analog as the nutrient and convert them into nontoxic inorganic compound, carbon dioxide, and water. As most pesticides do not resemble the natural compound metabolized by the microbes, they could not undergo mineralization. Another degradative strategy is cometabolism in which an organic compound which could be used as a primary energy source is provided along with the compound to be degraded which helps in the degradation of pesticides. Enzymes or cofactors used for the consumption of energy yielding substrate help in the metabolism of pesticides.

The bioremediation of pesticides depends on environmental factors, as well as some intrinsic factors related to microbes or pesticide chemical structure. These factors include the following:

(i) Metabolic activity and adaptability capacity of the microbial strain

(ii) Molecular weight, substitution type, location and frequency, and spatial structure determine the rate and efficiency of pesticide degradation. Polymeric compounds are resistant to degradation. The bioavailability of pesticides is also an important factor in bioremediation

(iii) Environmental conditions such as temperature, pH, salinity, humidity, available nutrients, required concentration of substrate, surfactant availability, carbon dioxide, and aerobic or anaerobic conditions are some of the factors that influence the remediation process carried out by microbes

Bioremediation can take place in situ (biostimulation, composting, biosparging, bioventing, and liquid delivery systems) as well as ex situ (biofilters, bioreactors, and land farming). The microbes residing in the contaminated soil can be used for bioremediation by stimulating them with nutrient or electron donors. Microbes can be added intentionally to break down the pesticides if the native microbes are not capable of bioremediation of these chemicals.

## 5. Bacterial Remediation of Pesticides

Organochlorines have been shown to undergo bioremediation by bacterial genus *Bacillus*, *Micrococcus*, *Arthrobacter*, and *Pseudomonas.* The CS5 strain of *Achromobacter xylosoxidans*, which was isolated form activated sludge, was reported for its capability to degrade $\alpha$-endosulfan and $\beta$-endosulfan by more than 0.0248 g/L and 0.0105 g/L, respectively, after eight days of incubation [23]. Hexachlorobenzene was observed to be anaerobically dechlorinated by *Dehalococcoides* sp., and the trichlorobenzene reductive dehalogenase enzyme was reported to be crucial in degradation which was the product of the *cbrA* gene [24]. Two closely related species of *Citrobacter amalonaticus* were isolated, and their genome was sequenced by Chaussonnerie et al. [25], and they reported their potential of biotransformation and remediation of chlordecone by these two isolates [25]. Ozdal et al. [26] reported five bacterial species (*Pseudomonas aeruginosa*, *Acinetobacter lwoffii*, *Stenotrophomonas maltophilia*, *Citrobacter amalonaticus*, and *Bacillus atrophaeus*) that were isolated from cockroaches that resides in pesticide contaminated zones and reported that endosulfan bioremediation by these isolates was found to be 88.5, 80.2, 85.5, 56.7, and 64.4%, respectively [26]. Anaerobic mineralization of pentachlorphenol by various bacterial strains was studied by Li et al. [27]. They reported various dechlorinators (*Sulfospirillum Dehalobacter, Desulfovibrio*, and *Deslfitobacterium* spp.) and phenol degraders (*Syntrophus* and *Cryptanerobacter* spp.) and determined the responsible functional genes; *cprA* (chlorphenol reductive dehalogenase), *bamA* (benzoyl-CoA reductase), and seven variants of nitrogenase reductase genes ([27]).

More than 90% of chlorpyrifos has been reported to be degraded by *P. putida* MAS-1 in minimal salt concentration [28]. Several strains of microbes were able to remediate chlorpyrifos such as *Providencia stuartii*, *B. cereus*, *Actinobacteria* sp., *Xanthomonas* sp. *4R3-M3*, and *Pseudomonas* sp. *4H1-M3.* Sharma et al. [29] reported that a *Bacillus* sp. G2 was able to remediate cypermethrin, and a novel mechanism was discussed in which intermediates such as 4-propylbenzoate, phenol M-tert-butyl, 4-propylbenzaldehyde, and 1-dodecanol were produced [29]. In a study, *Pseudomonas stutzeri* SMK strain, under controlled conditions, was able to degrade dichlorvos effectively [2].

The treatment of acetamiprid by *Rhodococcus* sp. strain BCH2 and the explanation of its degradative mechanism were also found [30]. Madhuban et al. [31] reported the remediation of imidacloprid and metribuzin by the *Burkholderia cepacia* strain CH-9 (aerobic) and showed 69 and 86% removal of imidacloprid and metribuzin, respectively, after 20 days [31].

Carbofuran hydrolase (a product of the *mcd* gene) enzyme has been reported for carbamate degradation by hydrolyzing the methylcarbamate linkage in various microbial genera such as *Bacillus*, *Pseudomonas*, *Ralstonia*, *Mesorhizobium*, *Ochrobactrum*, and *Rhodococcus*. Shin et al. [32] reported various carbamate degrading bacteria which included *Spingomonas*, *Rhodococcus*, *Spingobium*, *Microbacterium*, and *Bosea* [32]. Using carbaryl as the main source of carbon and nitrogen, *Corynebacterium*, *Bacillus*, and *Morganella* were able to degrade carbaryl by 48.8%, 94.6%, and 87.3%, respectively [33].

Fuentes et al. [34] reported that some actinomycetes genera (*Streptomyces* and *Micromonospora*) were capable of degrading organochlorines (lindane, chlordane, or methoxychlor). *Streptomyces* have also been studied for removal of chlorpyrifos and pentachlorophenol [35]. It was reported that chlorpyrifos was effectively removed by *Streptomyces* sp. M7 (99.2%). In bioremediation of cypermethrin, two enzymes estearses and phosphatases were involved in the degradation of pesticide [36].

## 6. Fungal Remediation of Pesticides

Xiao et al. [37] conducted a study to evaluate fungal species capable of degrading heptachlor and its epoxide. The remediation of heptachlor was observed to be 71, 74, and 90% by *P. tremellosa*, *P. brevispora*, and *P. acanthocystis*, respectively, after incubating them for two weeks. The heptachlor was metabolized to heptachlor epoxide, 1-hydroxy-2, 3-epoxy-chlordene, and 1-hydroxychlordene, and the removal of heptachlor epoxide after 2 weeks was found to be 25, 22, 16, and 16% by *P. aurea*, *P. lindtneri*, *P. brevispora*, and *P. acanthocystis*, respectively [37]. The *lin* gene was found to be responsible for the uptake and metabolism of hexachlorocyclohexane in *Spingobium francense*, *Spingobium japonicum*, and *Spingobium indicum* [38]. Remediation of acetamiperid and thiacloprid by *Rhodotorula mucilaginosa* strain IM-2 was reported by [39]. *Aspergillus niger* has been reported to produce hydrolase for the hydrolysis of carbamates [40].

*A. niger* was investigated for its capability to degrade endosulfan, and it was observed that the culture was able to tolerate 400 g/L of technical grade endosulfan, and endosulfan was completely eliminated after 12 days of incubation

[41]. Likewise, white rot fungus *Trametes hirsuta* was also observed to degrade endosulfan and endosulfan sulfate via hydrolytic pathways [42]. *Lecanicillium saksenae*, *Fusarium oxysporum*, *Penicillium brevicompactum*, *Aspergillus oryzae*, and *Lentinula edodes* were shown to be able to degrade difenoconazole, pendimethalin (99.5% removal by *L. saksenae*), and terbuthylazine (maximum 80% removal by *A. oryzae*) [43].

## 7. Types of Nanoparticles

NPs are categorized into various types on the basis of size, shape, morphology, and chemical properties [44].

## 8. Carbon-Based Nanoparticles

The carbon-based NPs mainly include fullerenes and carbon nanotubes (CNTs).

*8.1. Graphene.* It is an allotrope of carbon and a 2D planar and hexagonal complex of honeycomb-like lattices. Graphene sheets generally have a thickness of 1 nm [45].

*8.2. Fullerenes.* They are composed of carbon atoms that are bonded to each other by sp2 hybridization, forming spherical molecules of carbon (C60). Fullerenes are composed of about 28-1500 atoms of carbon, the single layers have a diameter of 8.2 nm, and the multilayered ones have a diameter of about 4 to 36 nm [46].

*8.3. Carbon Nanotubes (CNT).* They are formed by the graphene nanofoil that has a honeycomb-like network of atoms that are embedded into hollow coils, resulting in CNTs. The single-layered carbon nanotubes are about 0.7 nm, and the multilayered ones are 100 nm. The size of CNTs can vary from several micrometers to a few millimeters. They have hollow ends or can be enclosed by half fullerene molecules [47]. CNTs can have a relatively same shape as rolled graphite [48]. The rolled sheets can be single-walled carbon nanotubes, double-walled carbon nanotubes, or multiwalled carbon nanotubes [49].

*8.4. Carbon Black.* They are spherical amorphous carbon substances and have a diameter that varies between 20 and 70 nm. Since the particles bound instantly, they form a clustered shape, and approximately 500 nm clusters are formed [50].

*8.5. Carbon Nanofiber.* They are formed in a similar manner to CNTs and nanofoils of graphene, except that they are coiled into a cone-shaped structure rather than cylindrical tubes [51].

## 9. Metal NPs

They are produced from metals by using constructive or destructive procedures at the nanoscale level. NPs can be generated from most of the metals [52, 53]. Some of the metals that are often used for the production of NPs are zinc, aluminum, copper, lead, cobalt, gold, cadmium, silver, and iron [52, 54, 55]. NPs have unique surface properties, such as a high ratio of surface area to volume, spherical structure, reactivity, pore size, color, surface charge density, sensitivity, and sizes varying between 10 and 100 nm [56, 57].

*9.1. Metal-Oxide NPs.* Several metal-oxide NPs have been synthesized for the electrochemical determination of biological compounds. Some of the examples include $Fe_2O_3$, ZnO, $Co_3O_4$, $MnO_2$, NiO, and $TiO_2$ [58]. Moreover, mixed metal oxides have been of great interest due to such characteristics. The CuO nanoparticles have distinct features, because of which they have multiple applications like sensors, catalysts, antibacterial, and superstrong materials [59, 60]. Metal-oxide NPs have the ability to interact with other NPs due to the high surface area to volume ratio [61, 62].

*9.2. Ceramics NPs.* They are inorganic and nonmetallic substances that are synthesized by heating and cooling. Ceramic NPs exist in several morphologies and sizes; they can be hollow, dense, amorphous, porous, and polycrystalline. Thus, they have various applications in imaging, catalysis, photocatalysis, and photodegradation of dyes [63, 64].

*9.3. Semiconductor NPs.* They demonstrate a broad variety of applications since they exhibit characteristics of both metals and nonmetals [65]. Since these NPs exhibit larger bandgaps, bandgap tuning causes important modifications in their characteristics. Consequently, they demonstrate significant applications in photooptics, electronic devices, and photocatalysis [66, 67].

*9.4. Polymeric NPs.* These are organic nanoparticles [68, 69]. Generally, they exist in the form of a capsule or sphere. The former is a solid mass that is enclosed in the particle, and the latter is a matrix substance with a solid mass [70]. Lipid nanotechnology is one of the emerging fields for the production of lipid NPs that have important applications in drug delivery and cancer treatment [71, 72].

## 10. Role of Nanomaterials in Pesticide Remediation

*10.1. Cerium Oxide (CeO2).* The CeO2 nanofibers are synthesized from the metal-oxide frameworks of the Ce(1,3,5-benzenetricarboxylate) (H2O)6 (Ce-BTC), and they have been extensively studied for their uses in the adsorption of pesticides from water bodies [73]. The metal-organic frameworks of Ce-BTC were formed by using the hydrothermal procedure, and the CeO2 nanofibers are synthesized using the calcination procedure from Ce-BTC NPs at 650 degrees Celsius for 3 hours. 2,4-Dichlorophenoxyacetic acid (2,4-D) was adsorbed from water using CeO2 nanofibers using a batch system at 308 K with the $q_{max}$ value of 95.78 mg $g^{-1}$. 2,4-D was adsorbed into CeO2 by diffusion into the particle and the boundary layer on the basis of isothermal and kinetic studies.

*10.2. Magnesium Ferrite (MgFe2O4).* The mesoporous MgFe2O4 is magnetically retrievable and has been investigated for the adsorption of chlorpyrifos from wastewater containing pesticides [74]. MgFe2O4 having a high surface area of 170 $m^2 \cdot g^{-1}$ was synthesized by employing benign initiating substances and urea as binary purpose intercessors in a single-step solvothermal procedure. Chlorpyrifos was significantly adsorbed on the MgFe2O4 adsorbent at a pH > 9 , as exhibited by the batch adsorption studies. Similarly, chemisorption was observed when the hydroxylated magnesium ferrite surface having electronegative atoms such as sulfur, oxygen, and chlorine reacted with the aromatic ring of chlorpyrifos, which caused degradation into small organic molecules. This experiment demonstrated that the mesoporous magnesium ferrite with effective adsorption properties can be used for the treatment of wastewater.

*10.3. Activated Carbon.* Several investigations were conducted to study the removal of pesticides from the water bodies by employing activated carbons, like paclobutrazol [75], carbendazim and linuron [76], 11 pesticides [77], 2,4-dichlorophenoxyacetic acid (2,4-D) [78], iodosulfuron [79], and carbendazim (T. [80]). There are 2 types of activated carbon that are commercially available (GAB; $V_{Mic} = 0.27$ c $m^3 \cdot g^{-1}$, $S_{Mic} = 580\,m^2 \cdot g^{-1}$, $pH_{PZC} = 7.46$ and CBP; $V_{Mic} = 0.04\,cm^3 \cdot g^{-1}$, $S_{Mic} = 99\,m^2 \cdot g^{-1}$, $pH_{pzc} = 4.76$) and were used for the adsorption of 2,4-dichlorophenoxyacetic acid (2,4-D; pKa = 2.73) and 4-chloro-2-methylphenoxyacetic acid (MCPA; pKa = 3.07) from aqueous solutions [81]. The microporous GAB demonstrated better adsorption abilities ($q_{max} = 367.15\,mg \cdot g^{-1}$) than CBP ($q_{max} = 273.07\,mg \cdot g^{-1}$). The data of the assessment of adsorption exhibited that pH and ionic strength were important parameters for analyzing the rate of adsorption, which was increased for both types of pesticides by enhancing the ionic strength. As per the pesticides pKa and the $pH_{PZC}$ of activated carbons, removal of 2,4-D and MCPA was decreased with an increase in pH (pH > 4.76). The results concluded that the mechanism of adsorption was determined by the electrostatic interaction between the surface of the activated carbon and pesticides.

*10.4. Graphene-Based NMs.* Lazarevic-Pasti et al. [82] studied the impact of graphene-based materials on the removal of dimethoate and chlorpyrifos from the aqueous medium. The outcomes demonstrated that the adsorption of the pesticides on the adsorbents based on graphene was significantly dependent on the structural characteristics of the sorbent and sorbate, and the surface area was not the primary determinant for the pesticide elimination capability. The chlorpyrifos was specifically eliminated with aromatic moiety in the graphene basal plane, which contains the p electron system and high structural order, while adsorption of the aliphatic dimethoate onto hydrophilic oxidized graphene surfaces took place. Both chlorpyrifos and dimethoate were eliminated due to the moderate proportion of oxygen functional groups on the surface of graphene-based adsorbent.

*10.5. Carbon Nanotubes (CNTs).* The CNTs that were produced from plastic wastes were used as an efficient adsorbent for the removal of diuron from the water bodies [83]. This study demonstrated that the CNTs that were produced from plastic wastes, with $q_{max} = 103.73\,mg \cdot g^{-1}$ can be applied as a significant adsorbent for the treatment of water bodies. The multiwalled CNT was synthesized using particle size varying between 10 and 40 nm of the compound Ni/MgO, having a surface area of 9.10 $m^2 \cdot g^{-1}$ [84]. The effective elimination of about 90.5% diuron from water was carried out using multiwalled CNT ($q_{max} = 132.5\,mg \cdot g^{-1}$) under specific conditions ($C_{diuron} = 100\,\mu g \cdot L^{-1}$, time = 60 minutes, pH = 7.0, temperature = 25°C, CMWCT = $2.0\,g \cdot L^{-1}$). The statistical adsorption results indicated instant exothermic adsorption into the multiwalled CNTs. Modeling readings and computing the energy and binding affinity of diuron with multiwalled CNTs were used to identify the supramolecular adsorption process. The bonding between multiwalled CNT and diuron includes p-p T-shaped bonds, p-donor hydrogen bonds, p-p stacked, and p-alkyl bonds. These findings revealed that 16 hydrophobic interactions and 2 hydrogen bonds were involved in diuron physical adsorption and the formation of the multiwalled CNT-diuron compound.

*10.6. Titanium Dioxide ($TiO_2$) NPs.* $TiO_2(s)/H_2O_2/UV$ and $TiO_2(c)/H_2O_2/UV$ systems were investigated for the disintegration of insecticides as a function of the irradiation time. $TiO_2(s)/H_2O_2/UV$ system demonstrated the most efficient disintegration rate of the insecticides. 100% disintegration of the insecticides was accomplished after irradiation for 320 minutes by the $TiO_2(s)/H_2O_2/UV$ system. The half-life values of the insecticides were 36.28 and 43.86 for methomyl and dimethoate under the $TiO_2(s)/H_2O_2/UV$ system. But the half-life values of the insecticides were 19.52 and 27.72 minutes for methomyl and dimethoate under the TiO2 (s)/H2O2/UV system, respectively. In comparison to light settings, the disintegration of the pesticides examined in the dark utilizing the various techniques was minimal. Under dark conditions, the disintegration percentages of the studied pesticides using the various techniques ranged from 0.30 to 1.6 percent [85].

*10.7. Nanopesticides.* Various pesticides can be enclosed or adsorbed in multiple nanomatrices such as lipid NP, polymers, CNT, silica, or graphene oxides [86]. The nanopesticides have several advantages; they enhance the solubility of the barely soluble components, avoid early disintegration of pesticides, enhance the solubility of barely soluble components, and cause stimulated production of the active constituents [87, 88]. Silver NPs (Ag) were synthesized using the extract of the stem of the cotton plant (*Gossypium hirsutum*) and demonstrated effective antibacterial functions against pathogenic bacteria (*Xanthomonas campestris pv. campestris* and *Xanthomonas axonopodis pv. malvacearum*) that invade the crops of *Brassicaceae* and *Malvaceae* [89]. Ag-based chitosan nanocomposites exhibit better fungicidal activities compared to conventional fungicides [90].

## 11. Challenges and Future Prospects

Population worldwide is increasing rapidly, so as to deliver the food requirements of this burgeoning population, pesticides have been integrated in the modern lifestyle. Overconsumption of pesticides have resulted in buildup of resistance (acquired resistance) in the targeted pests. Excessive consumption of pesticides has led to clearance of normal susceptible population but providing a selective advantage to resistant pests leading to increase in their population. The control of important pests in crop, livestock parasites, household pests, and vector-borne diseases has become extremely challenging due to the developed resistance in them. Another issue with pesticides is reappearance of a large number of pest populations after the application of a pesticide known as pest resurgence. Pest resurgence has been associated with killing of natural enemies, increase in reproductivity, and feeding of insect pests, by using pesticides at less lethal dosages, and sometimes, secondary pests become primary pests because of eradication of the primary pest. Elimination of nontarget organisms like pollinators and earthworms has a huge impact on agricultural output. Pollinator populations have seen a major reduction due to the application of pesticides which translates into the reduction of crop production. Soil fertility is highly dependent upon the earthworms present in the soil. Indiscriminate use of xenobiotics has reduced the population of earthworms in the soil, and thus, more fertilizers are required to maintain soil nutrient quality [91]. Pesticides disturb the balance in ecosystem and have serious implications on the environment and human health; most of them are nonbiodegradable and highly toxic, led to bioaccumulation and biomagnification, and cause pollution of air, water, and soil which are some other challenges offered by the use of pesticides.

Bacterial and fungal remediation as well as use of nanomaterials are the future of pesticides removal from the environment. It has been reported for various pesticides but extensive research is necessary to increase the range of chemicals it could remediate as well as to know about the mechanism behind the remediation process. Genetic engineering could be used for production of superbugs with various genes combined for a range of pesticides removal. Coexpression of mixed plasmids with different set of genes for elimination of pesticides is the new approach which is possible due to genetic engineering, and in the upcoming years, it may be used for environmental clearance of pesticides.

## 12. Conclusion

The bioremediation of pesticides is urgent as it affects the environment and human population. Conventional methods could be used for its removal but their inherent problems like cost and time have discouraged their use. Microbial remediation is a good alternative for conventional methods. Microbes isolated from contaminated environments like water, soil, or sewage have been shown to possess enzyme systems to degrade these chemicals. Persistent organic pollutants like DDT can be utilized and metabolized by these microorganisms. Bacteria and fungi are the most studied, and various enzymes and pathways have been reported for the remediation of pesticides. For efficient removal of pesticides, conventional and microbial treatment could be coupled to increase the efficiency of elimination. Apart from microbial remediation of pesticides, use of nanomaterials for the removal of pesticides is another potential method that can be utilized. Different conventional methods could be integrated, or different microbial genes could be coupled by novel techniques offered by genetic engineering to enhance the efficiency of remediation treatment. Similarly, different nanomaterials can be used along with microbes to enhance the remediation efficiency. Thus, microbial and nanoremediation is the future for removal of pesticides.

## Data Availability

The data used to support the findings of this study are included within the article.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

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