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Abstract

The role of pesticides in enhancing global agricultural production is magnificent. However, their unmanaged use threatens water resources and individual health. A significant pesticide concentration leaches to groundwater or reaches surface waters through runoff. Water contaminated with pesticides may cause acute or chronic toxicity to impacted populations and exert adverse environmental effects. It necessitates the monitoring and removing pesticides from water resources as prime global concerns. This work reviewed the global occurrences of pesticides in potable water and discussed the conventional and advanced technologies for the removal of pesticides. The concentration of pesticides highly varies in freshwater resources across the globe. The highest concentration of α-HCH (6.538 μg/L, at Yucatan, Mexico), lindane (6.08μg/L at Chilka lake, Odisha, India), 2,4, DDT (0.90 μg/L, at Akkar, Lebanon), chlorpyrifos (9.1 μg/L, at Kota, Rajasthan, India), malathion (5.3 μg/L, at Kota, Rajasthan, India), atrazine (28.0 μg/L, at Venado Tuerto City, Argentina), endosulfan (0.78 μg/L, at Yavtmal, Maharashtra, India), parathion (4.17 μg/L, at Akkar, Lebanon), endrin (3.48μg/L, at KwaZuln-Natl Province, South Africa) and imidacloprid (1.53μg/L, at Son-La province, Vietnam) are reported. Pesticides can be significantly removed through physical, chemical, and biological treatment. Mycoremediation technology has the potential for up to 90% pesticide removal from water resources. Complete removal of the pesticides through a single biological treatment approach such as mycoremediation, phytoremediation, bioremediation, and microbial fuel cells is still a challenging task, however, the integration of two or more biological treatment approaches can attain complete removal of pesticides from water resources. Physical methods along with oxidation methods can be employed for complete removal of pesticides from drinking water.

Keywords: Wastewater; Pesticides; Biological treatment techniques; Physical treatment techniques; Chemical treatments techniques.

Abbreviation

µL: Micro litre, 2,4-D: 2,4-dichlorophenoxyacetic acid, 2,4-DP: 2,4-dichlorophenoxyacetic acid, AOTs: advanced oxidation technologies, BDL: below detectable limit, BIS: Bureau of Indian StandardsCNTs: carbon nanotubes, DDD: Dichlorodiphenyldichloroethane, DDE: Dichlorodiphenyldichloroethylene, DDT: Dichlorodiphenyltrichloroethane, DMDA: dimethyl dialkyl (C 14-C 18) amine, ECO: Electrochemical oxidation, ETZ: etridiazole, g/L: Gram per litre, h: hour, HCB: Hexachlorobenzene, HCH: Hexachlorocyclohexane, HRT: Hydraulic retention time, L: Litre, MCPA: 2-methyl-4-chlorophenoxyacetic acid, MCPP: methylchlorophenoxypropionic/Mecoprop, MFCs: Microbial Fuel Cells, mg: milli gram, MWCNTs: multi-walled carbon nanotubes, ND: non detectable, NF: Nano filtration, NOM: natural organic matter, OCPs: organochlorine pesticides, ODAAPS: aminopropyltriethoxysilane, OH: hydroxyl, PCP: Pentachlorophenol, PIP: piperazin, pm: Pico meter, SPE: solid phase extraction, SWCNTs: single-walled carbon nanotubes, TEA: trimethylamine, TFC: thin film composite, UV: Ultra violet, WHO: world health organization, CW: Constructed wetlands

1. Introduction

Fresh water is about 2.5% of the total water on the earth, out of which 1.2% is surface water and 30.1% is groundwater available for different usage. The water resources are severely polluted by anthropogenic activities such as agriculture, industries, urbanization, mining, etc. (Elfikrie et al., 2020; Amrish Kumar et al., 2019; Malyan et al., 2019; Zambito Marsala et al., 2020). Wastewater generated from homes, industries, hospitals, etc., is polluted with several toxic pollutants, and their removal is necessary to protect the life cycle of the water environment. Agricultural runoff and municipal wastewater may contain pesticides, and a high concentration of pesticides has been reported in urban and rural water (Malyan et al., 2019; Zambito Marsala et al., 2020). Pesticides are discovered to enhance the economic yield of crops by controlling the pest. Pesticides are used to control mosquitoes (Hustedt et al., 2020), agricultural weeds (Baghel et al., 2020; Cayuela et al., 2008), rodents (Swanepoel et al., 2017), nematodes (Cayuela et al., 2008), fungi (Cayuela et al., 2008), etc. (Md Meftaul et al., 2020). The application of pesticides in higher concentrations contaminates air, water, and soil (Kumar et al., 2019a; Yadav et al., 2015). Ingestion, inhalation, and dermal are the main routes of exposure to pesticides in humans. Vegetables, beverages, meat, and drinking water are key sources of pesticide exposure (Elfikrie et al., 2020; Nicolopoulou-Stamati et al., 2016). Pesticides are toxic to humans and may cause health issues such as cancer, asthma, Parkinson's disease, anxiety, skin allergies, eye irritation, and extreme weakness (Elfikrie et al., 2020; Yadav et al., 2015). Pesticides in low concentrations are also highly toxic and are responsible for the death of 0.2 million people per year at the global level, out of which, 95% of total death occurred in developing nations only (Md Meftaul et al. 2020). Thus, effective monitoring and removal of pesticides are imperative. Pesticides removal form wastewater and water can be achieved through constructed wetlands (Parlakidis et al. 2023; Malyan et al. 2021; Lyu et al. 2018), phytoremediation (Anand et al. 2019; Riaz et al. 2017), bioremediation (Nie et al. 2020; Spina et al. 2018), membrane filtration (Saleh et al. 2020; Hyllling et al. 2019), biochar (Vithanage et al. 2020), clay (Pal et al. 2001), etc., but efficiency and economical removal is still an area of investigation for the researchers. To our best knowledge, a review covering both the global pesticide distribution scenario in freshwater and their removal approaches is lacking. Most of the reviews on the pesticide distribution are region-specific and target specific removal technology. This study consolidates an overview of the global pesticide distribution in freshwater resources and various approaches involving physical, chemical, and biological processes for their elimination from the aquatic environment.

2. Pesticides contamination in global freshwater resources

Freshwater bodies such as ponds, lakes, rivers, and estuaries are contaminated with pesticides. When pesticide laden water reaches the aquifer, the groundwater is also polluted. The concentration of pesticides and their residues in the drinking water dictates the suitability of water for human consumption (Table 1). Extensive use of pesticides in agriculture and other systems pollutes ground and surface water resources.

2.1 Asia

Groundwater and surface water are mainly contaminated through anthropogenic sources such as agricultural runoff, domestic wastewater, industrial wastewater, etc. Pesticides include aldrin, atrazine, parathion, chlorpyrifos, dieldrin, Dichlorodiphenyltrichloroethane (DDTs), Hexachlorocyclohexanes (HCHs), lindane, malathion, endosulfan, imidacloprid, heptachlor, etc., have been detected in water resources of Asia region (Table S1). These pesticides have a detrimental effect on flora and fauna, and their monitoring and removal from water resources are necessary. A recent study in India by Nag et al. (2020) reported that Chilika lake of Odisha is contaminated with α-HCH, lindane, and 2,4 DDD, and their concentration ranged from 0.025 to 0.265 μg/L, 0.03 to 6.08 μg/L, and 8.99 to 23.4 μg/L respectively (Table S1). The groundwater of many regions of India, such as Haryana (Kaushik et al., 2012), Uttar Pradesh (Sankararamakrishnan et al., 2005), and Maharashtra (Lari et al., 2014) are contaminated with pesticide and their residues (Table S1). Punjab is an agricultural region of India. Chaubey et al. (2020) investigated the groundwater quality of Jalandhar City of Punjab and observed that the groundwater of the study area was contaminated with atrazine (0.0 to 0.9 μg/L), chlorpyrifos (0.0 to 0.03 µg/L), phorate (0.0 to 0.03 µg/L) and monocrotophos (0.0 0.14 µg/L). Pakistan is a neighboring country of India, and the surface and groundwater of Pakistan are also contaminated with pesticides (Table S1). Recently, (Taufeeq et al., 2021) reported that the surface water of Barandu River, Punjab, Pakistan, is contaminated with HCH (0.226 to 0.635 µg/L), aldrin (ND to 0.016 µg/L), DDT (ND to 0.023 µg/L), dieldrin (ND to 0.026 µg/L), heptachlor (0.082 to 0.308 µg/L), etc. (Table S1). Ara et al. (2021) investigated the groundwater quality of Potohar (Punjab region), Pakistan, and observed that groundwater is contaminated with HCH pesticides, and the highest concentration was 5.87 µg/L (Table S1). Similar findings were also reported by Ghaffar and Iqbal (2021) in Lahore, Pakistan (Table S1). Ghaffar and Iqbal (2021) observed that pesticides such as endrin (0.09 to 0.052 µg/L), dieldrin (0.012 to 0.089 µg/L), 1,2,3-Trichloropropane: TCP (0.0 to 0.031 µg/L), Dichloropropane: DCP (0.0 to 0.050 µg/L), DDT (0.0 to 0.051 µg/L) and dichlorodiphenyldichloroethylene: DDE (0.0 to 0.064 µg/L) were present in groundwater of Lahore, Pakistan. Pond and Canal water of the coastal region of the Bay of Bengal and Bangladesh are also contaminated with diazinon (0.320 to 0.631 µg/L), carbofuran (0.546 to 4.82 µg/L), and carbaryl (0.095 to 0.714 µg/L) (Bhuiyan et al., 2021).

Groundwater is the most important source of drinking water globally, and a wide range of pesticides was also detected in groundwater (Table S1), posing a threat to safe drinking water. The groundwater and surface water of China, Akkar, and Lebanon were polluted with α-HCH, β-HCH, 2,4-DDD, 4,4-DDT, 2,4-DDT, endosulfan, methyl-parathion, aldrin, dieldrin, endrin, etc. (Table S1) and making it unsafe for direct and indirect ingestion. The surface water of Guangdong, Guangxi, and Hainan Province of China is contaminated with α-HCH, β-HCH, lindane, 2,4-DDD, 4,4-DDT, 2,4-DDT, endosulfan, methyl-parathion, aldrin, dieldrin, and endrin pesticides (Wei et al., 2015). Wang et al. (2021) investigated the influence of pesticides contaminated irrigation water on groundwater and observed that the groundwater of the Jianghan Plain region of China is contaminated with methamidophos (BDL to 40.5 ng/L), dichlorvos (BDL to 15.5 ng/L), omethorate (BDL to 222.64 ng/L), phorate (BDL to 17.4 ng/L), dimethoate (BDL to 17.7 ng/L), diazinon (8.52 to 74.5 ng/L), methyl-parathion (BDL-10.2 ng/L), malathion (BDL-21.1 ng/L), parathion (BDL to 58.8 ng/L), and quinalphos (BDL-42.3 ng/L).

Table 1. Pesticide residue limits according to the World health organization (WHO, 2011), Bureau of Indian Standards (BIS, 2012), USEPA and other countries (Hamilton et al., 2003).

Name of Pesticide	**Limit [μg/L]**						
	WHO	**BIS**	**USEPA**	**New Zealand**	**Japan**	**Australia**	**Canada**
Alachlor	20	20	2	20	NGV	NGV	NGV
Aldrin	0.03	0.03	NGV	0.03	NGV	0.01	0.7
Atrazine	2	2	3	2	NGV	0.5	5
Chlorpyriphos	30	30	NGV	70	NGV	NGV	90
Endrin	0.6	NGV	2	NGV	NGV	NGV	NGV
Dieldrin	0.03	0.03	NGV	0.03	NGV	0.01	0.7
DDT (o, p and p, p-Isomers of DDT, DDE and DDD)	2	1	NGV	2	NGV	0.06	NGV
Endosulfan (alpha, beta and sulfate)	NGV	0.4	NGV	NGV	NGV	0.05	0.02
Malathion	NGV	190	NGV	NGV	NGV	NGV	190

Methyl Parathion	NGV	0.3	NGV	NGV	NGV	NGV	NGV
α-HCH	NGV	0.01	NGV	NGV	NGV	NGV	NGV
β-HCH	NGV	0.04	NGV	NGV	NGV	NGV	NGV
γ-HCH/Lindane	2	2	0.2	2	NGV	0.05	NGV
Delta-HCH	NGV	0.04	NGV	NGV	NGV	NGV	NGV
2,4-Dichlorophenoxyacetic acid	30	30	70	40	NGV	0.1	100 I
Ethion	NGV	3	NGV	NGV	NGV	NGV	NGV
Phorate	NGV	2	NGV	NGV	NGV	NGV	NGV
Isoproturon	9	9	NGV	10	NGV	NGV	NGV
Monocrotophos	NGV	1	NGV	NGV	NGV	NGV	NGV

NGV: No guideline value

2.2 Africa

Africa is a developing continent, and its dependence on surface water is more than any other continent. Surface water quality monitoring in such conditions is more critical. There are several scientific studies about freshwater contamination with pesticides in the region of Africa (Diop et al., 2019; Gakuba et al., 2018; Lawrence et al., 2015; Olisah et al., 2019; Twinomucunguzi et al., 2021; Yahaya et al., 2017; Khater, 2018). Ogbesse is the main river of Nigeria, and the water of this river is also contaminated with pesticides (Lawrence et al., 2015). Pesticide residues ranged from 0 to 0.43 µg/L in the Ogbesse river of Nigeria (Lawrence et al., 2015). IKpoda River is another major river in Benin City, Nigeria. Ogbeide et al. (2019) found that the water of the IKpoda river is contaminated with dieldrin (0.99 µg/L). It increases the ecological and human health risks in different parts of Nigeria due to direct and indirect exposures to pesticides (Lawrence et al., 2015; Ogbeide et al., 2019). Malawi Lake in the

Southern part of Africa is also contaminated with chlorinated pesticides, and the average annual concentration of pesticides in water was found to be up to 0.000022 µg/L (Karlsson et al., 2000). In another study, Gakuba et al. (2018) reported that the River Umgeni in South Africa is polluted with organochlorine pesticides (OCPs), and the total concentration ranged from 8.04 to 21.06 µg/L. Buffalo River in South Africa is also contaminated with 17 OCPs, and their highest concentration in the summer and autumn seasons reached up to 4.403 µg/L and 0.313 µg/L, respectively (Yahaya et al., 2017). The surface water of Egypt is also contaminated with different types of pesticides. In a study, it was reported that the water of the Muweis canal is contaminated with pesticides and ranges from 0 to 0.56 µg/L (Khater, 2018).

Dalvie et al. (2003) observed that the groundwater of Western Cape, South Africa, is contaminated with endosulfan up to the concentration of 3.16 µg/L, which is significantly higher than the permissible limit (0.10 µg/L) of that region. Recently, Twinomucunguzi et al. (2021) conducted an extensive study to monitor emerging contaminants in the groundwater of Uganda. They observed the presence of 59 different pesticides with the highest concentration of metalaxyl, 0.636 µg/L, at Bwaise, Uganda (Table S1). The groundwater of the Saïss aquifer of Morocco (North Africa) is contaminated with DDT (BDL to 0.0361 µg/L), DDE (BDL to 0.03323 µg/L), dicofol (BDL 0.24311 µg/L), diazinon (0.0148 to 0.04634 µg/L), malathion (BDL to 0.00064969 µg/L), carbendazim (BDL to 0.00595 µg/L)., etc and have high human health risk if consumed directly or indirectly (Berni et al., 2021). Based on the above findings, it can be concluded that the water resources of different regions of the African continent are contaminated with pesticides and therefore needs periodic monitoring and treatment for the removal of pesticides to safeguard human health.

2.3 Europe

Europe is the continent of developed nations of the world, and the freshwater of this region is also affected by several types of pesticides (Grondona et al., 2019; Karadeniz and Yenisoy-Karakaş, 2015; Lapworth et al., 2006; Mas et al., 2020; Rico et al., 2021). Recently, Rico et al. (2021) reported that the Iberian Peninsula is contaminated with pesticides. Out of 14600 samples, 21% were contaminated with chlorpyrifos. Köck-Schulmeyer et al. (2014) also found that the groundwater of Catalonia, Spain contaminated with twenty-two types of pesticides. The groundwater of Yorkshire, United Kingdom was also observed to be contaminated with twenty-two types of pesticides (Lapworth et al., 2006). Out of the boreholes samples analyzed, 78% were contaminated with pesticides, and 57% had higher concentrations than the permitted concentration value (Lapworth et al., 2006). The groundwater of the Netherlands is also vulnerable to pesticide contamination from different sources (Schipper et al., 2008). The monitoring of groundwater quality at 771 locations in the Netherlands indicated that 27% of samples were contaminated with pesticides and the pesticide concentration in 11% of samples exceeded the prescribed limits (Schipper et al., 2008). The shallow groundwater of Central Portugal is also contaminated with pesticides. Andrade and Stigter (2009) reported that the mean concentration (μg/L) of Alachlor, Atrazine, Metolachlor, Molinate, Propanil, and 3,4-dichloroaniline was 0.83, 0.49, 0.06, 3.62, 2.07, and 13.36 μg/L, respectively. Silva et al. (2006) investigated 171 groundwater samples in the Baixo Sado region (Portugal) for pesticide contamination. They found that 62% of samples used for domestic supply contain pesticides. Similar findings were also reported in the Beira Litoral and Ribatejo e Oeste agricultural area of Portugal (Batista et al., 2002), which is under cultivation of maize, vineyards, potato, apple, pear, etc. The authors reported atrazine in 70% of groundwater sites of this region having a maximum 11.2 μg/L concentration. Ghirardelli et al. (2021) reported that after banning Atrazine 27 years before in Italy, it is still found in the aquifer of Vicenza (Italy). However, the concentration reduces with time. Based on the above discussions, it can be concluded that groundwater in major areas of Europe is contaminated with pesticides, and its removal is one of the biggest challenges for the researcher.

2.4 Australia

Groundwater and surface water are usually contaminated through anthropogenic sources. Shishaye et al. (2021) monitored the pesticide contamination in the coastal alluvial plain agricultural area near the Great Barrier Reef, Queensland (Australia). They found that the groundwater is contaminated with major and minor pesticides (Table S1). A total of thirty-eight samples were collected from different locations. Among them, twenty-four samples contain major pesticides such as atrazine (0.0 to 0.01818 µg/L), desethyl-atrazine (0 to 0.10354 µg/L), bromacil (0 to 0.31533 µg/L), hexazinone (ND to 0.00359 µg/L), imazapic (ND to 0.01866 µg/L), diuron (ND to 0.00042 µg/L), imidacloprid (ND to 0.0008 µg/L), and simazine (ND to 0.00023 µg/L). The groundwater of the lower plain of Burdekin is also contaminated with pesticides (Table S1). Shaw et al. (2012) reported that the mean concentration of atrazine (0.019 µg/L), desethyl atrazine (0.070 µg/L), desisopropyl atrazine (0.021 µg/L), diuron (0.072 µg/L), hexazinone (0.019 µg/L), metolachlor (0.009 µg/L), and chlorpyrifos (0.3 µg/L) in groundwater of lower Burdekin, Australia. Similar findings were also reported by Schult and Schult (2016) in the Tindall aquifer (Table S1). The mean concentration (µg/L) of atrazine, desethyl atrazine, desisopropyl atrazine, tebuthuim, simazine, haloxyfop, imazapyr, imidacloprid, and 2,4 di-butylphenol is 0.007, 0.059, 0.024, 0.002, 0.002, 0.001, 0.004, 0.800, and 1.3 respectively. Based on the above discussion, it can be concluded that the aquifers in Australia’s agricultural region are contaminated with pesticides.

2.5 Americas

The groundwater of the American Continent is also contaminated with pesticides and their precursors (Table S1) (Acayaba et al., 2021; Bexfield et al., 2021; Munira et al., 2018; Thompson et al., 2021). Bexfield et al. (2021) investigated 1204 well samples for 109 pesticides and 116 pesticide degrades. Out of the total samples, 11% of samples were contaminated with active pesticides and 10% of samples were contaminated with the degrades (Bexfield et al., 2021). Bexfield et al. (2021) reported that atrazine and its metabolites were found commonly in the water samples. The midwestern region of the United States of America (USA) is under the cultivation of soybean and high corn. Pesticides sulfoxaflor and neonicotinoids are applied to these crops, and these pesticides were detected in the aquifers of this region (Thompson et al., 2021). Out of the total samples, 73% of samples have pesticide contamination, and Neonicotinoids and Clothianidin were detected commonly (Thompson et al., 2021). The groundwater of the Alberta region of Canada is also contaminated with pesticides (Munira et al., 2018). Munira et al. (2018) analyzed 105 groundwater samples for pesticides and detected pesticide contamination in 3% of the total samples. The groundwater of Ontario (Canada) is also contaminated with thiamethoxam, imidacloprid, and clothianidin (Browne et al., 2021). The maximum concentration of thiamethoxam, imidacloprid, and neonicotinoids clothianidin in groundwater samples was 0.46 µg/L, 0.7 µg/L, and 2.09 µg/L, respectively (Browne et al., 2021). The application of pesticides in rice cultivation contaminated the freshwater river system of Costa Rica, Latin America (Ramírez-Morales et al., 2021). Ramírez-Morales et al. (2021) reported that forty-two pesticides were present in groundwater. Pesticides such as diuron, oxyfluorfen, carbofuran, oxamyl, chlorpyrifos, carbendazim, and thiabendazole were commonly detected. The Sao Paulo State of Brazil is the largest sugarcane cultivation area globally, and pesticides are commonly applied for sugarcane crop management. Acayaba et al. (2021) conducted a study to assess pesticide contamination in the groundwater of Sao Paulo State (Brazil). They observed that groundwater is contaminated with tebuthiuron pesticides (0.107 µg/L). The groundwater and surface water of the Pampas region of Argentina is also contaminated with pesticides (Table S1). Vera-Candioti et al. (2021) reported contamination of atrazine (0.0 to 22.8 µg/L), azoxystrobin (up to 10.09 µg/L), acetochlor (up to 15.4 µg/L), glyphosate (up to 111 µg/L), and AMPA (up to 15.93 µg/L) in groundwater of Pampas region of Argentina. Brazil has the largest area under sugarcane cultivation worldwide, and pesticides are applied for crop management (Acayaba et al., 2021). Acayaba et al. (2021) observed that the surface water of the Brazilian sugarcane region is contaminated with several pesticides such as atrazine, carbofuran, malathion, imidacloprid, tebuthiuron, azoxystrobin, etc. Still, groundwater is contaminated with only tebuthiuron (ranging from 0.061 to 0.107 µg/L) (Acayaba et al., 2021). Based on the above discussion, it can be concluded that agricultural region aquifers in several countries of America are highly vulnerable to pesticides and their metabolites contamination.

2.6 Arctic and Antarctic

Freshwater resources such as surface water, glaciers, snow, etc. of the arctic and antarctic are also reported to be polluted with pesticides (Potapowicz et al. 2020; Casal et al. 2019; Cipro et al. 2017). Cipro et al. (2017) investigated pesticides contamination in the seasonal melting water and snow of George Island of Antarctica and observed cumulative concentration (pg/kg) of HCH, HCB, DDT, PCB, and chlordanes ranging from 1.46 to 4.17, 1.36 to 3.77, 4.32 to 24.4, 132 to 156, and 5.72 to 13.3. Casal et al. (2019) investigated the concentration of pesticides in the snow of Antarctica. Potapowicz et al. (2020) reviewed the contamination of OCP in the aquatic environment of the Antarctic and concluded that human activities are responsible for OCP contamination in polar regions. Atmospheric deposition of pesticides is one of the critical responsible factors for pesticide contamination in the snow of polar regions (Potapowicz et al. 2020; Casal et al. 2019). Long-range atmospheric transport of contaminated air has involved the movement of pesticides from pollutant areas to polar regions and cold environmental conditions of the polar regions restricted this movement which result in magnification of pesticides (Potapowicz et al. 2020; Casal et al. 2019; Bigot et al. 2016). The melting snow and glacier resulted in the release of these deposited pesticides into water and finally, it bioaccumulated in the food chain (Potapowicz et al. 2020; Chpora et al. 2011). Apart from the air deposition the biological system (migrated birds) also contributed to pesticide contamination in the freshwater ecosystem of the polar regions (Nagar et al. 2022; Evenset et al. 2007). Evenset et al. (2007) reported that seabirds contribute around 14% of total pesticide contamination in Lake Ellasjøen on Bjørnøya in the Barents Sea of the Artic region. Recently, Nagar et al. (2022) also reported that the seabird deposits organohalogen pesticides in the upper region of lake Polynya Lake in north west Greenland.

3. Holistic pesticides removal approaches

Pesticide remediation from water and wastewater system can be achieved through biological, physical, and chemical treatments (Figure 1). Pesticides can be removed through ozonation (Hua et al., 2006; King et al., 2020; Matsushita et al., 2018; Schmidt and Brauch, 2008), sorption (Mandal et al., 2017; Taha et al., 2014), nanofiltration (Karimi et al., 2016; Košutić et al., 2005; Nguyen et al., 2019), carbon nanotubes (Dehghani et al., 2019, 2017), coagulation-flocculation (Kouras et al., 1998; Ormad et al., 2008a; Shabeer et al., 2014), gamma radiations (Chu et al., 2018; Khedr et al., 2019; Mohamed et al., 2009), bioremediation (Ellegaard-Jensen et al., 2017; Malyan et al., 2020; Nousiainen et al., 2015), and electrochemical oxidation (Komtchou et al., 2017) from water resources (Table S2). The mechanism and removal efficacy of the treatment technologies for pesticide remediation are discussed in this section.

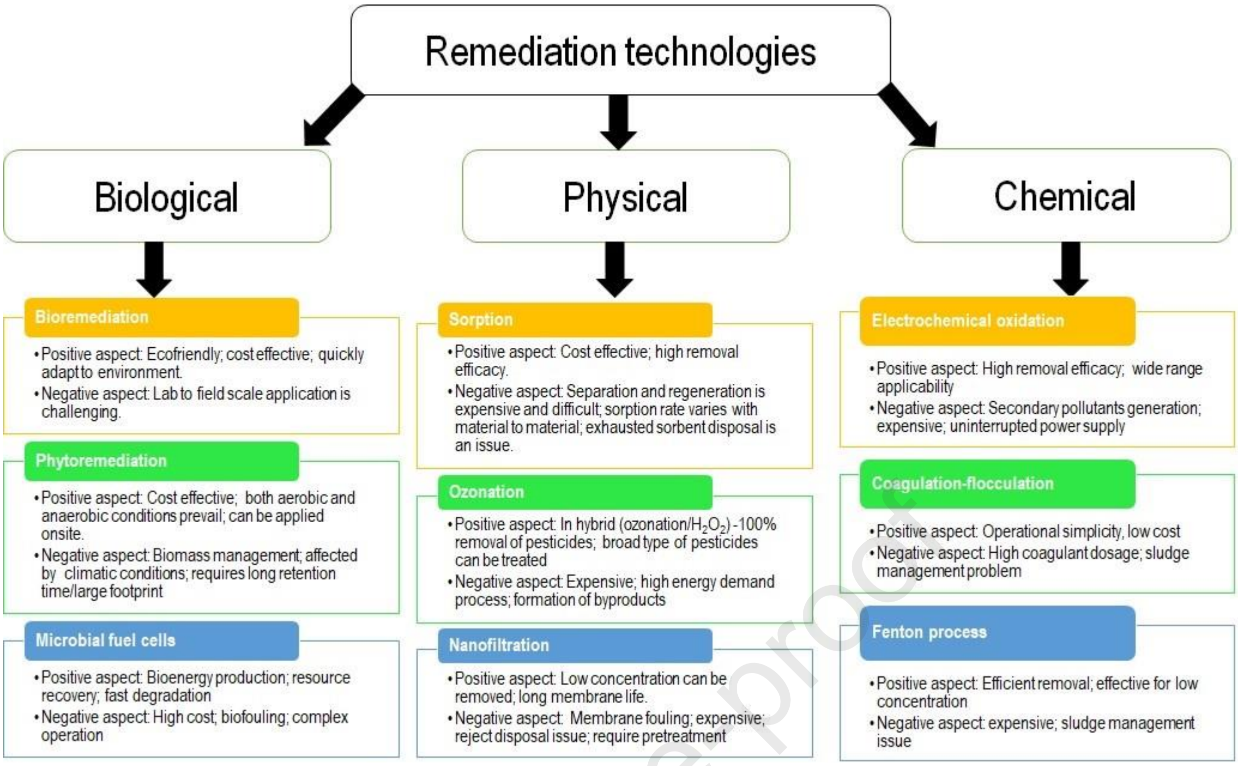

Fig. 1. Crucial biological, physical, and chemical techniques involved in pesticide remediation from water/wastewater.

3.1 Biological approaches for pesticides removal

3.1.1 Phytoremediation

Plants species uptake pesticides from water and degrade or transform them into less toxic compounds. Phytoremediation of pesticides consists of many processes such as phyto-degradation, phyto-volatilization, phyto-degradation, phyto-transformation, and rhizoremediation. Constructed wetlands are effective in pesticide removal from contaminated water. Several biotic and abiotic factors affect plant uptake of pesticides from water and their removal. Loffredo (2021) investigated the fungicide metalaxyl-M removal potential of Hemp (*Cannabis sativa* L.). They observed that 67% to 94% of metalaxyl-M was removed from wastewater in seven days. Transformation and plant uptake were the two crucial mechanisms involved in removing metalaxyl-M. Hillebrands et al. (2021) also concluded that the younger hemp plant removed pesticides more efficiently from the wastewater through an oxidation metabolized mechanism.

The application of aquatic plants for pesticide removal from water bodies has been reported (Alonso et al., 2021; Chander et al., 2018; Riaz et al., 2017; Xia and Ma, 2006). Riaz et al. (2017) investigated the phytoremediation potential of *Pistia stratiotes*, *Eichornia crassipes*, and algae. They studied the removal of pyrethroid and organochlorine in surface water. Cyanophos are widely used for pest control in Egypt. Romeh (2014) reported the Cyanophos remediation from the water stream through a broadleaf plant *Plantago major* L. (Table S2). The total Cyanophos was removed by 11-94.7% within hours owing to pesticide accumulation in the leaves and roots of the plants. Romeh (2010) investigated the removal of imidacloprid using *Plantago major* L. They observed a 55 to 95% accumulation of imidacloprid in roots and leaves within 1-10 days of treatment. The highest rate of pesticide accumulation was observed in roots (7.94 µg/g dry weight) of *Plantogo major* L. McKnight et al. (2021) investigated the removal of imidacloprid and azoxystrobin in three wetlands plants species of arrowhead (*Sagittaria latifolia*), pickerelweed (*Pontederia cordata*), and softrush (*Juncus effusus*) in 56 days controlled experiment. The highest imidacloprid removal efficacy of arrowhead, pickerelweed, and softrush was 79.3, 37.1, and 36.0%, respectively, compared to non-planted systems. The azoxystrobin removal was slightly lower in arrowhead, pickerelweed, and softrush, i.e., 51.7%, 24.9%, and 28.7%, respectively, over control. Thus, the aquatic and wetlands plant species can be effectively used in floating and subsurface flow constructed wetlands to remove pesticides from the aquatic medium.

3.1.2 Bioremediation

Bioremediation is a nature-based pollutant removal process in which microorganisms play a significant role (Dash and Osborne, 2022; Rani et al. 2022). Figure 2 shows the graphical representation of several bioremediation technologies. Bacterial species like Bacillus, Pseudomonas, Flavobacterium, and Azotobacter; algae like Chlorella; and fungi like Mycobacterium and Aspergillus are the common pesticides-degrading microorganisms (Bhalla et al. 2022). Mahajan et al. (2021) investigated the biodegradation of pesticides (profenofos) (initial concentration, 50 µg/ ml) by Bacillus and observed that 93% of pesticides degrade within 30 days at 28 °C and on pH 7. Recently, Fang et al. (2023) investigated the profenofos biodegradation through *Cupriavidus nantongensis* and observed that 88.82% of pesticide (initial concentration, 20 ppm) is degraded in just 48 hours on the optimal temperature range of 30-37 °C. Singh et al. (2021) investigated the bioremediation of monocrotophos (MCP) through three different strains of bacteria (*Rhizobium leguminosarum*, *Streptomyces* sp and *Bacillus subtilis*) isolated from arable field. Bacillus subtilis degrade highest MCP followed by Streptomyces sp. and *Rhizobium leguminosarum*. The degradation of MCP under these three bacterial strains range from 75-92%. (Singh et al. 2021). Degradation of malathion by bacillus sp. FYM31 isolated from drainage water of agriculture was investigated recently by Madbolly et al. (2022) and a 70.1% degradation in 12 days under optimal conditions (temperature, 37 °C) was reported. Bioreactors and biostimulation are the most widely used approaches to remove pollutants from wastewater (González et al., 2006). These technologies are used to remove pesticides from point sources such as agricultural farms and industrial wastewater. Membrane bioreactors effectively degraded phenoxyl herbicides (MCPA, MCPP, 2,4-DP, and 2,4-D) and removed up to 57% to 88% of total herbicides (González et al., 2006). Point source pesticide removal using membrane bioreactors is recently reported in several studies (Lopes et al., 2020). Lopes et al. (2020) investigated pesticide removal from industrial wastewater. They found that the membrane bioreactors significantly removed the pesticides, but complete removal of pesticides through solitary membrane bioreactors technology is not achieved. Similar findings were reported in another study where bioreactors treating agricultural runoff reduced 31%, 63%, and 76% of oxyfluorfen, chlorpyrifos, and bifenthrin, respectively, at an HRT of 21 hrs (Abdi et al., 2020). However, pesticides can be easily removed through hybrid membrane bioreactors and activated carbon technologies (Lopes et al., 2020). They observed that the sand bioreactor can degrade metaldehyde at the rate of 0.1 to 0.2 mg/ L. h and can remove 86% of the total metaldehyde. The sand bioreactors provide the media for microbial growth, which achieves the continued degradation of pesticides (Rolph et al., 2020).

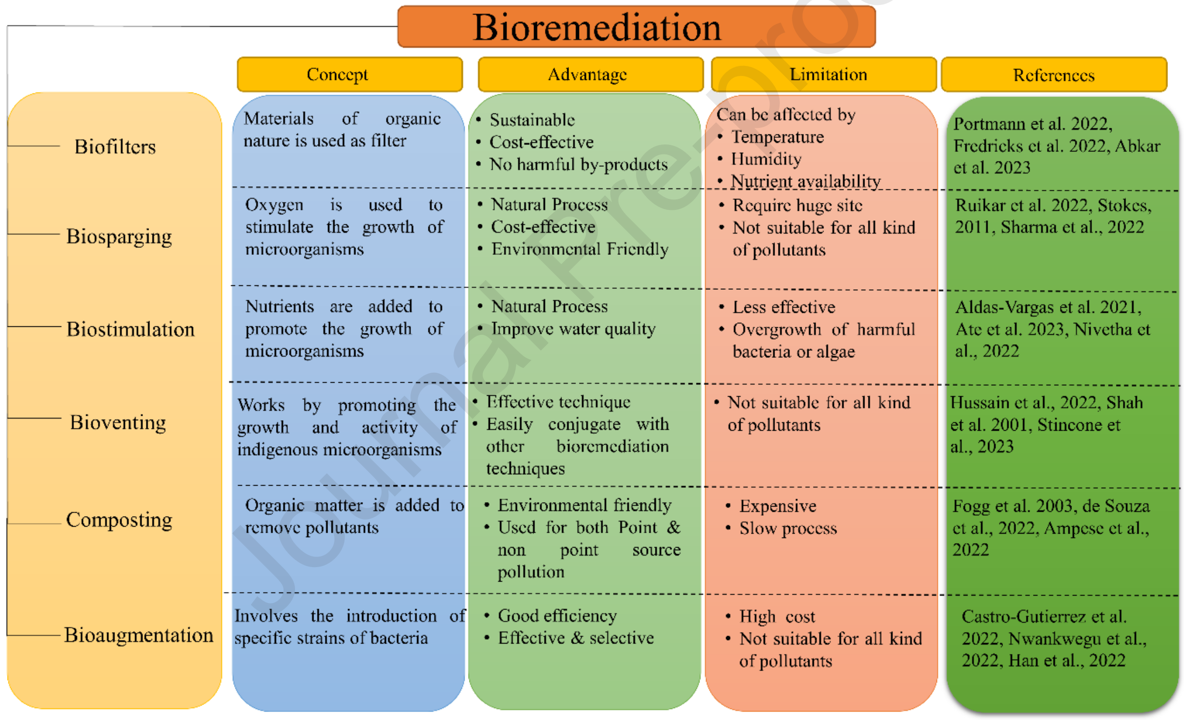

Fig. 2 Types of different bioremediation technologies

3.1.3 Mycoremediation

Fungi play a significant role in the biodegradation of pesticides (Table S2) and other environmental contaminants (Kumar et al., 2019; Maqbool et al., 2016; Spina et al., 2018). The biodegradation rate of pesticides is affected by nutrient availability, temperature, pH, oxygen level, etc. (Maqbool et al., 2016). Pesticides have high solubility and stability, which ensure their long half-life in water (Spina et al., 2018). Pesticide removal from water resources via fungi is gaining global attraction due to its eco-friendly and cost-effective aspect. Fungi quickly adapt to conditions, including pesticide stress, and grow even in water containing significant pesticide concentrations. Various fungal species combat pesticide pollution through biodegradation and biosorption processes (Oliveira et al., 2015).

Fungal species *Penicillium implicatum* and *Aspergillus viridinutans* were isolated from water drains. They were grown in a medium having pesticides lambda-cyhalothrin and chlorpyrifos with three different concentrations of 2.5, 5, and 10 ppm. *Penicillium implicatum* and *Aspergillus viridinutans* fungal species completely degraded 2.5ppm of lambda-cyhalothrin and chlorpyrifos pesticides in 14 days. However, pesticides were not completely removed at a higher pesticide dose of 10ppm (Abdel-Wareth and Abd El-Hamid, 2016). Dey et al. (2020) reported lindane removal through *Aspergillus* fumigatus fungus from water. *Aspergillus* fumigatus significantly reduced lindane concentration from 30ppm to 1.92ppm in 72 hrs. It indicates that fungus has the potential for the myco-remediation of pesticides from the aquatic system. The fungus *Fusarium proliferatum* isolated from agro pesticides contaminated site degraded up to 50 ppm of allethrin in 144 hrs (Bhatt et al., 2020b). Bhandari et al. (2021) isolated *Achromobacter sp*. from the agricultural field and investigated the carbendazim degradation potential. They observed that at pH 7 and temperature 30 ºC, around 76.2% of total pesticides were degraded within 20 days. Thus, mycoremediation can be applied for the effective degradation of pesticides from water medium. The efficiency of mycoremediation is affected by the temperature, pH, media, and concentration of pesticides.

3.1.4 Microbial Fuel Cells (MFCs)

Microbial Fuel Cells (MFCs) are emerging technologies that remove organic and inorganic pollutants from wastewater simultaneously with bioenergy production (Kumar et al., 2019b, 2019d, 2019c). Zhao and Zhang (2021) investigated pesticide removal through the electro-Fenton process coupled with MFC. They observed that mesotrione degradation under anodic biofilm of MFC reached up to 0.83 mg/ L.h (Table S2). In an anodic biofilm, the microbial population degraded complex aromatic compounds of pesticides through co-metabolism pathways, resulting in the removal of the pollutants from the wastewater (Zhao and Zhang, 2021). Hexachlorobenzene (HCB) removal through MFC was reported by Cao et al. (2015). Pentachlorophenol (PCP) is a toxic compound commonly used in organochlorine pesticides, and its degradation through MFC has been investigated (Khan et al., 2018). Pesticides can be degraded in both aerobic and anaerobic environments of the MFC. Plant-MFCs can be a useful technology to achieve sustainable development goals.

3.1.5 Constructed wetlands

Wetland ecosystems are well known for regulating ecosystem services such as water purification in nature. Constructed wetlands (CW) is engineered design for the treatment of wastewater and it is gaining popularity due to their low operating cost and environmentally friendly feature. The removal of pollutants in the CW system is governed by diverse microbial communities and different physiochemical processes (Malyan et al. 2021; Parlakidis et al. 2023; Lu et al. 2023). Pesticide removal through the CW system is recently reported by Pettigrove et al. (2023) and in several past studies also (Budd et al. 2009; Vymazal et al. 2015; Lv et al. 2016; Liu et al. 2019, Chen et al. 2022). Lv et al. (2016) investigated the removal of two pesticides (tebuconazole and imazalil) from wastewater through CW and observed that sorption along with the biodegradation pathway plays a significant role in the removal of pesticides. The removal of Imazalil was highest in the summer season in planted CW (95%), while in winter removal decreased to 60%. On the other hand, the removal of tebuconazole was also higher in summer (79%) as compared to winter (30%) due to enhanced microbial activity at higher temperature (Lv et al. 2016). Parlakidis et al. (2023) investigated the removal of triticonazole through horizontal subsurface flow CW and observed the removal in the range 49.2% - 88.4%. Budd et al. (2009) investigated the removal of pesticides from agricultural tailwater through CW and observed significant removal for pyrethroids (52-94%) and chlorpyrifos (52-61%), and non-significant removal of diazinon. The diazinon does not adsorb on the media surface of CW resulting in almost no removal (Budd et al. 2009). Tang et al. (2019) investigated the removal of chlorpyrifos through recirculating vertical flow CW using five plant species (*Juncus effusus*, Cyperus *alternifolius*, *Iris pseudacorus*, *Canna indica,* and *Typha orientalis)* and observed 94-98% of removal. Sorption (64-86.4%) and biodegradation (8.1-33.7%) are the main pathways involved in the removal of chlorpyrifos in CW (Tang et al. 2019). Chen et al. (2022) investigated the removal efficacy of integrated CW at larger scale and observed that the removal ranged from 49.99% to 84.96% during the different season. The highest removal of cumulative 19 different pesticides was observed in summer (84.96%), followed by autumn (61.80%), spring (57.02%), and lowest in winter (49.99%). Biotic degradation is one the important process in CW and it is effected by seasonal temperature change. Chen et al. (2022) observed that phyla of *Chloroflexi, Bacteroidetes, Proteobacteria, Planctomycetes, and Acidobacteria*, are associated with the biodegradation of pesticides in CW. Recently Hu et al (2022) investigated the impact of biochar in CW for the removal of atrazine and observed that addition of sulfuric acid modified biochar in media enhanced atrazine removal from 50% to 70%. On the basis of above finding, it can be concluded that CW has potential for the removal of pesticides contamination from water resources. The removal of pesticides is affected by environment conditions, microorganism diversity, plant species and substrate media used.

3.2 Physical approaches for the removal of pesticides

3.2.1 Sorption

The problem of contaminated water in the last few decades has emerged in developed and developing nations. Sorption is the easiest and most cost-effective technology for pesticide removal from water/wastewater. Adsorbents such as biochar, activated carbon, polymeric material, clays, agricultural by-product, industrial products, etc. (Srivastava et al., 2009) are used for the adsorption of pesticides (Kyriakopoulos and Doulia, 2006).

Biochar is carbon-rich material produced from diverse organic matter through pyrolysis under oxygen-free or limited oxygen concentrations (Yadav et al. 2023; Sakhiya et al., 2020). Biochar removes pesticides from an aqueous solution by holding active ingredients of pesticides on its surface (Malyan et al., 2021; Zheng et al., 2010). Zheng et al. (2010) reported that biochar produced from green-waste feedstock at 450 °C significantly removed atrazine (1158 mg/kg) and simazine (1066 mg/kg) from the aqueous medium at a biochar/contaminated solution ratio of 1:1000 (g/mL). The removal of bentazone pesticides from watershed systems through biochar has been documented recently by (Ponnam et al., 2020). Neem tree bark (*Azadirachta indica*) biochar synthesized at 300 ºC effectively adsorbed up to 79.40 mg bentazone /g biochar with 0.448 g/L of biochar dosage and 79.40 mg/L bentazone initial concentration (Ponnam et al., 2020). Biochar-based filters efficiently removed atrazine, naphthalene, phenanthrene, and anthracene by 37-97%, 49-93%, 100%, and 100%, respectively, from drinking water (Chan et al., 2020). The adsorption capacity for pesticides is significantly affected by the biochar feedstock (Mandal et al., 2017). Mandal et al. (2017) investigated the sorption capacity of six different feedstock (Eucalyptus bark, corn cob, rice husk, rice straw, bamboo chip, acid-treated rice straw) biochar for the atrazine and imidacloprid removal from contaminated aqueous solution (Table 2). The acid-treated rice straw biochar at 1 g/L dose removed 59.5 to 89.8% atrazine and 58.2 to 89.8% imidacloprid from the solution. The biochar produced from acid-treated feedstock enhanced the adsorption capacity of biochar for pesticides due to changes in pore volume, surface area, pore diameter, aromaticity, and polarity (Mandal et al., 2017). The corn straw biochar doped with P efficiently removed triazine pesticide (>96%) from the aqueous solution. The oxygen-containing and metaphosphates groups on the surface of corn straw biochar play a significant role in the adsorption of triazine pesticides (Suo et al., 2019).

Clay and clay-polymer also play a significant role in the removal of inorganic and organic contamination from water/wastewater, including pesticide removal. Montmorillonite clay has a surface area (600-760 m2/g) it makes it an interesting adsorbent material for pesticides (Helmy et al. 1999; Cosgrove et al. 2019). Vermiculite has a size of range 2-8 mm and it is a clay mineral composed of magnesium-aluminium silicates which used for the removal of pesticides. Vermiculite and other clay mineral has a high cation exchange capacity and can be used as substrate media in CW or in soil for the removal of pesticides (Cosgrove et al. 2019). Lemić et al. (2006) investigated the adsorption of atrazine, diazinone, and lindane from water through organic zeolite and the adsorption capacity for lindane, atrazine, and diazinone was 3.4 μmol/g, 2.0 μmol/g and 4.4 μmol/g respectively. The adsorption capacity of clay minerals can be enhanced through modification and intercalating Fe (III) polymer (Abate and Masini, 2005; Srinivasan, 2011). The intercalated montmorillonite clay shows 99.5% of sorption for atrazine which indicates its powerful sorbent nature after Fe (III) intercalating (Abate and Masini, 2005). Intercalated clay mineral sorption is affected by pH (Wu et al. 2021). Wu et al. (2021) investigated the effects of Cu (II) and Fe(III) intercalating in clay minerals for the sorption of atrazine and observed that the sorption of atrazine on smectite is reduced at pH 8 under Fe(III) preloading, while suppressed sorption was observed at pH 4.0 on smectite due to preloading of Cu (II). Fe(III) and Cu (II) has no suppression effects for atrazine sorption on kaolinite and illite clay mineral (Wu et al. 2021). Narayanan et al. (2020) found that biopolymer-nano-organoclay along with alum effectively remove atrazine (63.2 to 72.9%), butachlor (90.2 to 99.7%), carbendazim (64.1 to 73.1%), carbofuran (71.3 to 79.2%), imidacloprid (80.2 to 89.2%), isoproturon (89.3 to 98.2%), pendimethalin (91 to 100%), thiophanate methyl (91.1 to 100%), and thiamethoxam (83.2 to 92.4%). On the basis of above discussion it can be concluded that sorption different pesticides from water can be achieved through clay mineral, biochar and other material. Utilization of this sorbed material in CW as substrate media and in soil will help in combating pesticides problems.

Table 2. Pesticides removal efficiencies of different technologies

References	**Treatment**	**Pesticide**	**Removal efficiency**	**Experimental Conditions**			
Adsorption of pesticides via biochar				Scale of study	Initial Concentration (mg/L)	Contact Time (h)	Adsorbent dose (g/L)
Chen et al., 2022	Bagasse biochar	Imidacloprid	92 %	Lab	23.8	21	0.5-10
Alsherbeny et al., 2022	Corn cob biochar	Atrazine	85 %	Lab	0.5	1	10
		Chlorfenvinphos	96.3 %				
		Chlorpyrifos	97.6 %				
		Cyprodinil	98 %				
		Diazinon	90 %				
		Dimethoate	83.5 %				
		Diuron	95 %				
		Ethion	96.5 %				
		Malathion	94.5 %				
		Profenofos	97.5 %				
Chan et al., 2020	Woodchip biochar filter	Atrazine	37 to 97%	Lab	0.1	168	0-5
		Naphthalene	49 to 93%		0.05	168	0.05
		Phenanthrene	100%		0.025	168	0.05
		Anthracene	100%		0.05	168	0.05
Suo et al., 2019	Corn straw biochar	Triazine	Up to 96%	Lab	2	2	.005-0.12

Mayakaduwa et al., 2017	Rice husk biochar	Carbofuran	The highest adsorption capacity of 160.77 mg g-1	Lab	5-100	4	1
Mandal et al., 2017	Eucalyptus bark biochar	Atrazine	23.4 to 40.1%	Lab	1-10	24	1
	Eucalyptus bark biochar	Imidacloprid	14.7 to 28.4%				
	Corn con biochar	Atrazine	18 to 30.4%				
	Corn con biochar	Imidacloprid	5.9 to 20.1%				
	Rice husk	Atrazine	11.8 to 42.6%				
	Rice husk	Imidacloprid	28 to 42.6%				
	Rick straw	Atrazine	37.5 to 70.7%				
	Rick straw	Imidacloprid	39.9 to 77.7%				
	Acid-treated rice straw biochar	Atrazine	59.5 to 89.8%				
	Acid-treated rice straw biochar	Imidacloprid	58.2 to 89.5%				
	Bamboo chip biochar	Atrazine	12.3 to 26.9%				
	Bamboo chip biochar	Imidacloprid	16.9 to 35.7%				
Adsorption of pesticides via Clay							
Narayanan et al., 2020	Biopolymer-nano-organoclay composite*	Atrazine	63.8 to 72.9%	Lab	10	1	0.5-3
		Butachlor	90.2 to 99.7%				
		Carbendazim	64.1 to 73.1%				
		Carbofuran	71.3 to 79.2%				
		Imidacloprid	80.2 to 89.2%				
		Isoproturon	89.3 to 98.2%				

		Pendimethalin	91.0 to 100%				
		Thiophanate methyl	91.1 to 100%				
		Thiamethoxam	83.2 to 92.4%				
Dutta and Singh, 2015	Bentonite clay modified with hexadecyltrimethylammonium	Atrazine	55.2 to 58.8%	Lab	5.4	24	10
	Bentonite clay modified trioctylmethylammonium	Atrazine	64.7 to 72.4%				
	Bentonite clay modified stearylkonium	Atrazine	56.6 to 61.7%				
Other adsorption processes of the pesticides							
Soliman et al., 2022	Bagasse	Chlorfenvinphos, chlorpyrifos, diazinon, ethion, profenofos and cyprodinil	Up to 99%	Lab	0.2-1	0.16-2	10
Toledo-Jaldin et al., 2020	Magnetic sugarcane bagasse	Carofuran	175 mg/g	Lab	10-100	0.25-24	.002
	Magnetic peanut shell	Carofuran	89.3 mg/g				
	Magnetic sugarcane bagasse	Iprodione	119 mg/g				
	Magnetic peanut shell	Iprodione	2.76 mg/g				

Note:*Carboxy methyl cellulose and two nanoorganobentonites viz. DMDA (nanobentonite modified with 35–45 wt% dimethyl dialkyl (C14–C18) amine) & ODAAPS (nanobentonite modified with 15–35 wt% octadecylamine and 0.5–5 wt% aminopropyltriethoxysilane) in the ratio 1:2.5:2.5

3.2.2 Ozonation

Ozone, a powerful oxidant, was first used in the late 1800s to remove contaminants in water treatment. Extremely reactive species (OH radicals) released from ozone attack organic, inorganic, and other pollutants in water, resulting in water decontamination (Glaze et al., 1987). The amount of hydroxyl (OH) radicals produced in the ozonation process is directly proportional to contaminant removal efficiency (Glaze et al., 1987). Ozonation is globally used for the removal of pesticides (Hua et al., 2006; King et al., 2020; Matsushita et al., 2018; Schmidt and Brauch, 2008), phenols (Wang and Chen, 2020), pharmaceutical waste (Wang and Zhuan, 2020), metals (King et al., 2020), etc in water. Oxidation-reduction reaction, cycloaddition reaction, electrophilic substitution reaction, and nucleophilic reaction play a significant role in directly removing contaminants from water (Wang and Chen, 2020). Ormad et al. (2008) studied ozone's efficiency in removing 44 pesticides from drinking water in Spain. The pesticide removal efficacy of ozonation and integrated ozonation-activated carbon adsorption process was 70 and 90%, respectively (Ormad et al., 2008a). The degradation of pesticides in the ozonation process is affected by aqueous solution pH, and the same was demonstrated by Usharani et al., 2012. The removal efficiency of methyl parathion through the ozonation process at pH 3, 7, and 9 was 60%, 81%, and 98%, respectively (Usharani et al., 2012).

3.2.3 Nano-filtration

Nanofiltration (NF) is a physical process that requires a pressure-driven membrane that removes most of the organic molecules, viruses, natural organic matter, and a range of divalent salts. It is very similar to reverse osmosis but the major difference is that it offers higher flux rates and uses less energy than reverse osmosis (Košutić et al., 2005). Karimi et al. (2016) studied the pesticide (atrazine and diazinon) removal from water by using a nanofiltration membrane. They synthesized a thin-film composite (TFC) polyamide nanofiltration membrane and enhanced its performance with additive trimethylamine (TEA) and piperazine (PIP). Water flux and pesticide rejection show inverse relation with the increase in PIP and TEA amount. Further, the rejection of pesticides and change in water flux was observed to depend on both PIP and TEA concentration, thickness, and morphology. Karimi et al. (2016) indicated that the removal is due to the hydrophobic interaction between hydrocarbon (hydrophobic) segments of pesticide molecule and membrane, and the membrane prepared with a high concentration of PIP (2% w/v) and modified with TEA (2% w/v) was found to be the most appropriate Polyamide TFC Nanofiltration membrane for the removal of pesticides. Van der Bruggen et al. (2001) explored the removal of a mix of four pesticides (atrazine, simazine, diuron, isoproturon) and hardness from groundwater with the help of nanofiltration (NF) membrane and produce drinking water of very good quality. Here pesticide retention on the membrane was explained by molecular size and dipole moment. The largest molecule had the highest retention while a high dipole moment decreases retention because it interacts with the charge of the membrane. The authors concluded that groundwater flux is 5% lower than distilled water flux because natural organic matter (NOM) adsorbs and blocks the membrane, which results in flux decline. Tateoka et al. (2018) observed the partial and complete removal of carbamates in water by nanofiltration membrane and found the average removal efficiency for all pesticides (carbaryl, carbofuran, and methomyl) was about 95%.

3.2.4 Photodegradation

Photodegradation involves using light (UV) to alter or degrade chemical compounds. Pesticides are widely removed or altered into less harmful forms through photodegradation. Liu et al. (2009) investigated the photodegradation of etridiazole (ETZ) by UV at 254 nm and found the decomposition of ETZ into three organic byproducts, namely 5-ethoxy-3-

dichloromethyl-1,2,4-thiadiazole, 5-ethoxy-1,2,4-thiadiazole-3-carboxylic acid, and 5-ethoxy-3- hydroxyl-1,2,4-thiadiazole. The result showed that a minimum 40 mJ/cm2 dose of UV is required for the degradation. Shawaqfeh and Al Momani (2010) studied advanced oxidation technologies (AOTs) using UV light irradiated at a wavelength of 254 nm and 350 nm and solar radiation during March (Sol-Mar) and June (Sol-Jun) for treating pesticides (Vydine). The result indicated that photodegradation of Vydine in the presence of TiO7 followed the sequence UV(254) > Sol-Jun > Sol-Mar > UV(350). Kowalska et al. (2002) examined the photodegradation of organic pesticides in industrial wastewater by UV/H2O2/air system and found that the presence of hydrogen peroxide affects the efficiency of photodegradation.

3.2.5 Solid-phase extraction

It is an alternative to the liquid-liquid extraction method and one of the most convenient procedures for extracting pesticides from water samples (Kumar et al., 2010; Martínez Vidal et al., 2009). Inherently, it is used as a sample handling method because of its benefits like stability, higher environmental friendliness, and low cost. Nevertheless, modification occurred in traditional SPE to carry out extraction called dispersive SPE (dSPE) (Castillo et al., 2011; Zhao et al., 2012; Zhou et al., 2012). A new carbon-based sorbent material is used in SPE to increase extraction efficiency. Carbon nanotubes (CNTs) has outstanding extraction efficiency and excellent properties such as higher surface areas, large aspect ratios, and a broad range of application, etc., suggesting their great capacity for the sorption of organic compounds (González-Curbelo et al., 2013; Ravelo-Pérez et al., 2010; Zhou et al., 2006). CNTs can be divided into single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) based on the carbon atom layer in the wall of nanotubes. In recent years, MWCNTs have been used to extract pesticides and reported as an effective SPE material (Qin et al., 2015).

3.3 Chemical methods for the removal of the pesticide

3.3.1 Electrochemical oxidation

Electrochemical oxidation (ECO) is an efficient technology for removing several pollutants from wastewater (Dominguez et al., 2018a; Garcia-Segura et al., 2018; Kumar and Sharma, 2019). Pesticide removal from water resources through electrochemical oxidation is also documented (Dominguez et al., 2018b, 2018a). Lindane from groundwater through ECO using boron-doped diamond (anode) and carbon felt (cathode) has been reported by (Dominguez et al., 2018b). Dominguez et al. (2018a) found that the ECO processes remove 100% of lindane in just 4 hrs at 400 mA. HCH pesticides are generally degraded by three possible mechanisms in electrochemical oxidation processes (Fig. 3). β-elimination (net charge of the molecule remain unchanged), reduction due to dihaloelimination (two chlorine molecule is eliminated from the molecule) and hydrogenolysis (hydrogen atoms replace the chlorine atoms in the molecule), and free-radical elimination (addition of free-radical in the molecule), are the common mechanism involved in the HCH transformation (Wacławek et al., 2019). Renewable energy, such as wind energy (Souza et al., 2015), solar (Millán et al., 2019), etc., can also be used in electrochemical oxidation technology to remove pesticides. Souza et al. (2015) powered the boron-doped diamond electrochemical process with wind energy and observed that the 2, 4-D can be efficiently removed. Sustainable energy for the electrochemical anodic oxide for pesticide removal from wastewater has been reported (Millán et al., 2021, 2019). Millán et al.(2019) observed that 85% of clopyralid was removed at 25 ºC and 10 Ah dm-3.

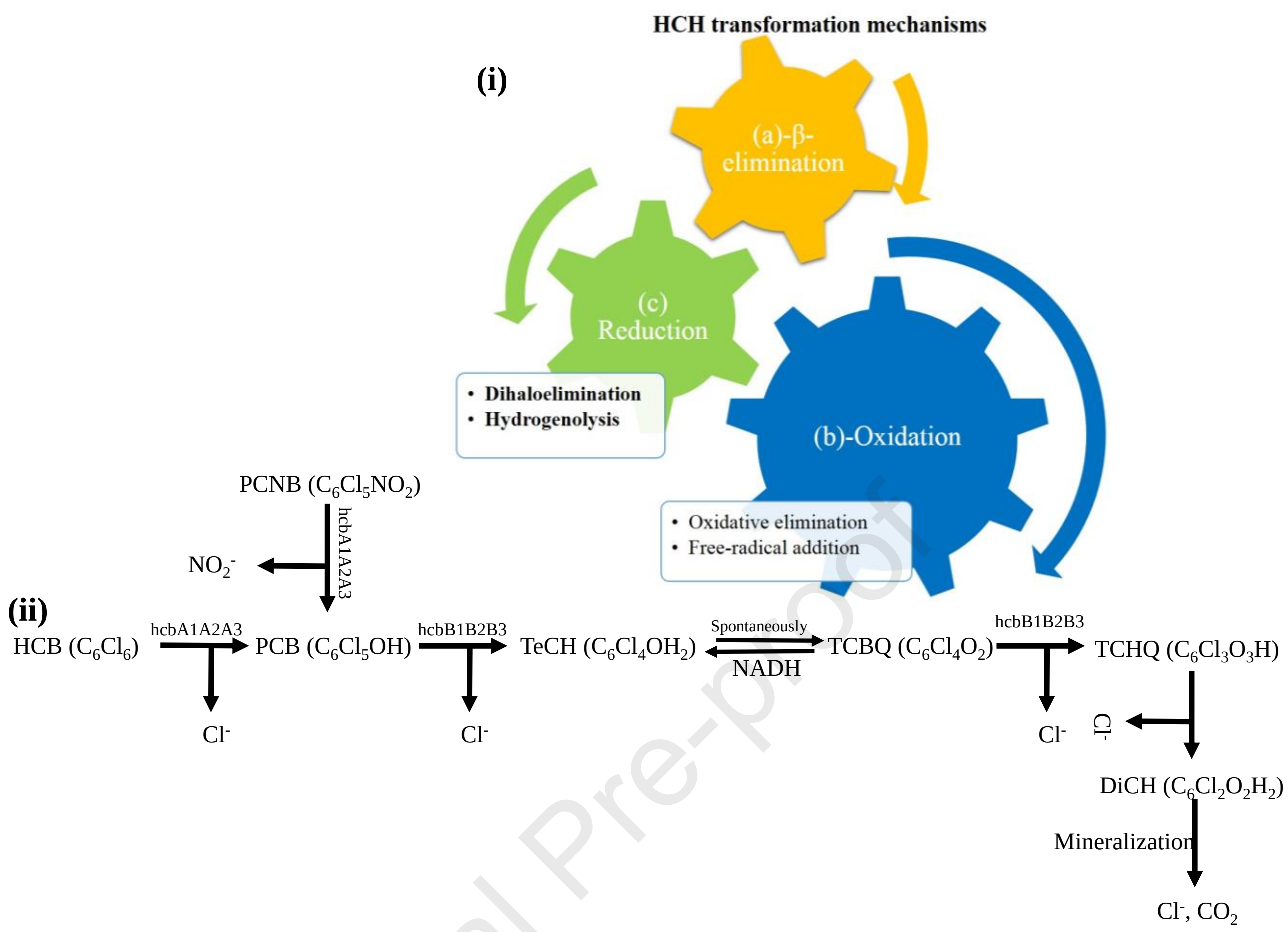

Fig. 3i HCH transformation mechanism in electrochemical oxidation processes (Image taken and adopted from (Wacławek et al., 2019) and, Fig. **3ii**-anaerabic degradation pathway of HCB as well as PCNB (Source: Takagi, 2020)

3.3.2 Coagulation –flocculation

Coagulation-flocculation is a practical approach for removing multiple pesticides from water (Campinas et al., 2021; Ormad et al., 2008b; Saini and Kumar, 2016; Shabeer et al., 2014). Saini and Kumar (2016) investigated the removal of pesticides through two coagulants, ferric chloride and alum. They optimized the dose and pH through RSM-CCD. Alum at 80 mg/L removed 82% and 64% of the chlorpyrifos and methyl parathion, respectively (Saini and Kumar, 2016). Ferric chloride at the optimum dose level (60 mg/L) resulted in the removal of 79% of the chlorpyrifos and methyl parathion (Saini and Kumar, 2016). Complete removal of multiple pesticides through the coagulation-flocculation process was not achieved. However, integrating two or more removal approaches can result in complete removal. The investigation conducted by Shabeer et al. (2014) supports integration approaches for removing multiple pesticides from the water medium. Shabeer et al. (2014) observed that the removal efficiency for the ten different pesticides through coagulation-flocculation ranged from 12 to 49%. Coagulation and adsorption-integrated approaches observed significantly higher removal efficacy (71 to 100%) (Shabeer et al., 2014). Integrated processes of powdered activated carbon before adding a coagulant to remove pesticides from water are also reported recently by Campinas et al. (2021) and Sánchez López et al. (2021). They reported an enhanced removal efficiency. Sánchez López et al. (2021) investigated the removal of the pesticide through dissolved air flocculation followed by adsorption through powdered activated carbon and observed a significant reduction. Kalantary et al. (2022) investigated the pesticide residues and their removal through coagulation-flocculation in Marun River of Bahbahan city of Iran. River water have 16 different pesticides which range in concentrations from 0.87 to 3.23 µg/l. The mean concentration of malathion, diazinon, chlorpyrifos, alachlor, 2-4-D, DDT, lindane endosulfan, dieldrin,, glyphosate, and aldrin in water was 2.743 µg/l, 3.229 µg/l, 3.163 µg/l, 2.44 µg/l, 0.87 µg/l, 0.03 µg/l, 0.021 µg/l, 0.03 µg/l, 0.026 µg/l, 2.55 µg/l and 0.03 µg/l respectively. Out of the total, 87% of the pesticides are removed from water through coagulation-flocculation and sand filtration. 100% removal of diazinon, malathion, lindane, heptachlor, endosulfan, dieldrin and glyphosate is achieved through the adsorption process. Kalantary et al. (2022) observed that most pesticides (except, 2,4-D) are hydrophobic in nature and hydrophobic micropollutants are easily removed through sorption in coagulation-flocculation.

4.0 Current Situation, Challenges, and Way forward

Pesticides are used for the control of pest and unsafe utilization of pesticides result in fresh water pollution. Yadav et al. 2015 reported pesticides consumption in Taiwan, China, Japan, Netherlands, USA, UK, and India around 17 kg/ha, 14 kg/ha, 12 kg/ha, 9.4 kg/ha, 7 kg/ha, 5 kg/ha, and 0.5 kg/ha respectively. The environment is also threatened by the use of pesticides in urban areas where the dose of the pesticides is generally very high (Md Meftaul et al., 2020), requiring immediate attention. Pimentel et al. (1995) reported that, out of total applied pesticides only 0.1% is utilized in pest control and remaining 99.9% become the part of environment. The contamination of water resources of Asia, Europe, Africa, America, Polar regions, and Australia due to pesticides has been reported (Yadav et al. 2015; Casal et al.2019; Potapowicz et al. 2020). The polar region (Arctic and Antarctic) of Earth is also contaminated with pesticides through biological system (migratory bird) and air deposition. Several pesticides are resistant to environmental degradation which makes them persistent resulting in biomagnification in the food chain and increased toxicity (Chopra et al. 2011). Human beings are exposed to pesticide contamination from several routes such as water, food, and air (Yadav et al. 2015), and majority of deaths due to pesticide poisoning have been reported from developing countries (Md Meftaul et al., 2020). High mortality in the developing countries may be due to improper handling of pesticides, paucity of funds for pesticide mitigation, lack of awareness, etc.

Pesticides retention time in biological system enhances its toxicity and therefore its removal from different components of environment is one the biggest challenges. Integrated pest management technology using biological and cultural interventions can play a prominent role in pesticide control in the environment, however economical and educational aspect limits its global adoption.

Biological pest control, one of important component of the integrated pest management practice can be options for dealing with the non-biodegradable pesticides. Phosphinothricin, Spinosads, Milbemycin, Azadirachta indica, Rotenone, Sabadilla, Nicotine, and Capsain are some of the natural pesticides and their sources are Streptomyces hygroscopicus, Bottle-brush plant, Saccharopolyspora spinosa, Streptomyces sp, Neem leaf and seeds, Lonchocarpus root, Sabadilla lily, Aqueous tobacco extract, and Capsicum peppers respectively (Sharma et al. 2020). Weed, insects, and fungus problem can be managed through these natural pesticides and they up to some extend can be used in place of chemical pesticides which reduces the pesticides pollution load of environment. Further, suitable formulations of easily biodegradable pesticides may be an option to avoid the pollution of water resources by persistent pesticides and research is needed in this direction.

Treatment of the water polluted with pesticides is needed to safeguard the health of humans and the ecology. Based on the literature, advanced tertiary treatment approach, nanofiltration in combination with ozonation, can be an option for the complete removal of pesticides from water. ECO can be also used for the removal of pesticides from the water and wastewater. Both the techniques have disadvantages that the pesticides present in the water is not completely eliminated from the environment and therefore, further research is required to get rid of pesticides from the water. Further, enforcement policies to restrict the sale of pesticides and usage in both urban areas and agriculture needs to be framed and strictly implemented along with social sensitization to minimize the water pollution.

5. Conclusions

Anthropogenic activities such as agriculture, industries, and urbanization are responsible for the pesticide pollution of water resources. Aquifers in agricultural activity zones are usually contaminated with pesticides. Significantly higher pesticide concentration was observed in Mexico, India, Argentina, and South Africa practicing intensive farming practices. Pesticides are highly toxic, and direct or indirect consumption of pesticide-contaminated water may cause health problems. Removal of pesticides from drinking water is the need of the hour. It can be achieved through biological, chemical, or physical treatment methods. Biological methods such as phytoremediation are environmentally friendly and technically feasible. But absolute pesticide removal through phytoremediation alone is difficult. Chemical methods such as coagulation-flocculation also have great potential for up to 90% removal of pesticides from water. Removal efficacy through physical processes such as adsorption through biochar (up to 100%), clay (up to 99.7%), and bagasse (up to 99%) is significantly higher as over coagulation-flocculation however their role at field scale still requires further investigation. Complete removal of pesticides at field scale through a single biological or chemical, or physical approach is challenging, however, integration of two or more methods results in the effective removal of pesticides from drinking water.

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RESEARCH HIGHLIGHTS

- Global distribution of pesticides in freshwater was reviewed.
- Pesticides are persistent in the groundwater even after 27 years of discontinuation.
- Biological, chemical, and physical removal approaches were discussed.
- Removal of pesticides up to safe levels through a single treatment approach is difficult.
- Hyphenation of two or more treatment approaches can achieve 100% pesticides removal.

Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: