**Pesticides in Aquatic Environments and Their Removal by Adsorption Methods**

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**Abstract:** Pesticides are widely used in agriculture, industry and household and pose a risk to human health and ecosystem. Based on the target, the main types of pesticides are insecticides, herbicides, and fungicides, of which herbicides accounted for 46% of total pesticide usage worldwide . The movement of pesticides into water bodies occurred through run-off, leaching, sub-surface drainage and spray drift, which have negative impacts on aquatic environments and humans. During this study, we sought to define the critical factors affecting the fluxes of contaminants into receiving waters. Furthermore we aimed to specify the feasibility of using sorbants to remove pesticides from waterways. In this study, the occurrence of pesticides in water sources in several countries was investigated. The concentration of pesticides in some locations, such as Karun River in Iran (1.2 × 104 ng/L), was too high. Apart from that, the concentration of pesticides in fish’s muscles and body was studied. For example in this research, maximum concentration (ng/L) of pesticides in fish samples was detected at 3.2(104), for methoxychlor herbicide in *Perca fluviatilis* in Lithuania. During the last decades, researchers have focused on effective elimination of organic pollutants, such as pesticides, from aqueous solution. One the promising technique in removing pesticides is adsorption method. Among several adsorbents, nanoparticle-adsorbents and carbon-based adsorbents have revealed a high performance (near 100%) in removing pesticides from water bodies.

**Keywords:** Pesticides; Adsorption; Biochar; Graphene; Water; Wastewater

**1. Introduction**

Anthropogenic activity can result in the contamination of water bodies, which in turn, reduces ecosystem function and can pose a human health risk (Mojiri et al., 2019c). Water contamination might be described as any impairment in its native characteristics by the addition of anthropogenic pollutants to the extent that it either may not serve humans for drinking purposes and/or support the biotic communities, such as fish (Ekubo and Abowei, 2011). All water contamination affects plants and animals that live in these water bodies and in practically all cases the influence is damaging not only to the individual species and populations but also to the natural biological communities (Agrawal et al., 2010). Pesticides in water resources are very hazardous because of their adverse impacts on environment and human (Zhou et al., 2015). Pesticides are chemical substances applied on agricultural land, private gardens, and other public areas (Grube et al., 2011)to kill organisms that are harmful to cultivated plants or to animals. Pesticides are therefore required to maintain sufficient food production for humanity (Aktar et al., 2009). The usage of pesticides for crop protection is estimated to increase based on a growing world population and the need for more food supplies. However, pesticides may also kill non-target organisms, particularly if they are mobile in the environment (Özkara et al., 2016). While pesticides improve agricultural production, bioaccumulation over the food chain can ultimately become a danger to mammals because pesticides cause certain negative impacts (Jardim and Caldas, 2012). The movement of pesticides into water bodies can occur through run-off, leaching, sub-surface drainage and spray drift (Cosgrove et al., 2019). Therefore, the removal of pesticide residues from the environment has gained widespread attention. Several physical, chemical and biological treatment ways have been employed to remove pesticides from aqueous solutions (Mojiri et al., 2019a) but most of these techniques have drawbacks such as limited flexibility, high cost, low efficacy and probability of secondary contaminants production (Shamsollahi and Partovinia, 2019). One of the promising techniques to eliminate pesticides from aqueous solution is adsorption method. Adsorption is one of the effective procedures of advanced wastewater treatment, which industries apply to diminish hazardous organic/inorganic contaminants present in the effluent (Yagub et al., 2014). Adsorption can be greatly effective process because of simplicity of design, inexpensiveness, rapidity, ease of operation, and availability of different adsorbents without dangerous substances formation (Sadon et al., 2012). Since the adsorption procedure is a surface phenomenon, the physical and chemical properties of adsorbent and adsorbate may affect adsorption efficiency (Shamsollahi and Partovinia, 2019). Currently, the trend in the occurrence of pesticides in waterways and fishes around the world and the mechanism of pesticides removal by different adsorbents are not well understood.. Therefore, the article aimed to (1) study different pesticides and their usage, (2) review the concentration of pesticides in waterways, and (3) assess the pesticides removal via adsorption methods.

**2. Pesticides**

The rapid development of numerous agricultural activities to meet human food demand necessates the widespread application of many different pesticides. In 2016, the worldwide consumption of pesticides reached 4.1 millions of tons, of which 1.4% was used in Oceania, 2.2% in Africa, 11.8% in Europe, 33.3% in Americas, and 51.3% in Asia (Pérez-Lucas et al., 2018). However, it has been assessed that only 1% of applied pesticides reach the target and around 99% release to soil and water supplies (Ali et al., 2019). Pesticides can be categorized generally by considering two criteria: chemical classes and target organisms (Ahmad et al., 2010). Pesticides can be divided in four main groups based on their usages in agricultural production, including fungicides, herbicide, insecticide, and other (Huang et al., 2018). Other group contain miticides (to kill moths), algaecides (to kill algae), acaricides (to kill ticks) and rodenticides (to kill mice, rats, moles and other rodents). Balmer et al. (2019) reported that 4.1 million tonnes (Mt) of pesticides were applied worldwide in 2016 which herbicides were the largest proportion of pesticide usage (Fig. 1).

**2.1. Types of Pesticides** **based on** **Their Functions**

**2.1.1. Fungicides**

The Global Fungicides Market (Table 1) is counted to be $13.12 billion in 2016 and is estimated to touch $17.58 billion by 2022 (Zion Market Research, 2017). In 2016, 29% of total used pesticides (4.1 million tonnes) was fungicides. Fungicides are often used as a prophylaxis to prevent disease (Santísima-Trinidad et al., 2018). Hutchens et al. (2019) expressed that periodic fungicide applications are frequently essential to maintain crop with acceptable quality. Fungicides can be categorized based on the mode of action, general usage, and chemical composition (Baibakova et al., 2019). Based on the usage, they are classified as (1) foliar fungicides and (2) dressing fungicides. Foliar fungicides are employed as powders or liquids for the aerial green parts of plants, producing a defensive barrier on the cuticular surface and systemic toxicity in the developing fungus. Dressing fungicides are employed to the postharvest crop as dry powders or liquids to prevent fungal infestation, chiefly if stored under less than optimal conditions of humidity and temperature (Gupta, 2019). Based on the mode of action, they are categorized as (1) protectant, (2) therapeutant (systematic), and (3) eradicant. Protectant fungicides are prophylactic in their behaviour, and are effective only if used prior to fungal infection, e.g. Sulphur and Zineb (Koli et al., 2019). Systemic fungicides can be absorbed by the plant without damage and be transported to other tissues where they are toxic to fungi (Baibakova et al., 2019). Fungicide which is able to eradicate a fungus after it has resulted infection and there by curing the plant is named chemotherapeutant, e.g. Carboxin, Oxycarboxin antibiotics like Aureofungin. Frequently chemotherapeutants are systemic in their action and affect the deep-seated infection (Koli et al., 2019). Eradicant fungicides eliminate pathogenic fungi from an infection court (area of the host around a propagating unit of a fungus in which infection could feasibly occur, e.g. Organic mercurials, dodine, lime sulphur. They may remain effective on or in the host for some time (Parthiban et al., 2015). Fungicides are classified in nine main group based on their chemical composition. Table 2 shows the fungicides classification and their examples.

**Table 2:** Fungicides based on their chemical nature

**2.1.2. Herbicides**

The Global Herbicides Market (Table 1) is estimated to be $28.76 billion in 2017 and may reach $41.63 billion by 2023 (Pioneer Reports, 2018). In 2016, 46% of total used pesticides (4.1 million tonnes) was herbicides. Application of herbicides is a common way to control weeds in agricultural and non-agricultural systems. Herbicides can be employed in different forms such as liquids, granules and by fumigation (Harrington and Ghanizadeh, 2017). Herbicides are classified into 27 groups on the basis of their mode of action, of which 11 groups o are more widely used (Table 3). Apart from that, herbicides are categorized in terms of their chemical nature and mechanism (Table 4).

**Table 3:** Herbicides based on their mode of action

**Table 4:** Herbicides based on their chemical nature and mechanism

**2.1.3. Insecticides**

Insecticides are broadly applied to control insect pests, but concerns have occurred regarding their environmental safety (Mulé et al., 2017). The Global Insecticides Market (Table 1) is counted for $14.51 billion in 2016 and is estimated to touch $19.27 billion by 2022 (Markets and Markets, 2017). In 2016, 17% of total used pesticides (4.1 million tonnes) was insecticides. Pyrethroids are the most frequently employed insecticides for indoor spraying against mosquitoes worldwide, owing to their efficacy and safety (Liu, 2015). Most of the insecticides in common use today are toxic to people as well as well as insects (Eaton, 2017). Based on the chemical nature, insecticides are classified into six main groups (Table 5)

**Table 5:** Fungicides based on their chemical nature

**2.2. Types of Pesticides** **based on** **Their Chemical Nature**

Pesticides can be divided into 10 (Table 6) main groups based on their chemical nature, including: benzoic acid, carbamates, dipyridyl, organochlorines, organophosphates, phenoxyalkonates, phenylamides, phthalimides, pyrethroids, and triazines (Jayaraj et al., 2016).

**2.3.1. Benzoic acid (Herbicides)**

Benzoic acid and benzoate (benzoic acid sodium salt) were the first chemical preservative allowed in foods in the USA. It is still generally employed today for a large number of foods and soft drinks because of its antibacterial effects (Jorge, 2003). During 2013, the consumption of benzoic acid in the world was almost 500000 metric tons, with a subjected growth to just around 600000 metric tons by 2018 (Settle et al., 2017). Benzoic acid herbicides do not bind to soil particles (Kamrin, 1997). Groundwater half-life of 41 h has been recorded for benzoic acid (initial concentration 1–100 µg/L (WHO, 2000). Benzoic acid is a monobasic aromatic acid, mildly strong, soluble in alcohol and ether, white crystalline powder, and benzene, but poorly soluble in water. Benzoic acid has the advantage that it does not affect the odor or taste of the soft drink, if used in small quantities (Søltoft-Jensen and Hansen, 2005). The important examples of this kind of pesticides are 3-phenoxy benzoic acid (3-PBA), bromoxynil, dicamba, dichlorobenil, and tricamba (Jayaraj et al., 2016). Benzoic acid has been criticised anecdotally for possible allergenic effects (Ashurst and Hargitt, 2009). From the aquatic organisms examined so far, cyanobacteria (*Anabaena inaequalis*) demonstrated to be the most sensitive (WHO, 2000).

**2.3.2. Carbamates (Insecticide)**

Global consumption of carbamates insecticides was 4092.13 tonnes in 2014 (Zhang, 2018) with around 6% growth until 2018. Carbamates signify one of the chief classification of synthetic organic pesticides since their introduction into the agrochemical market in the 1950s and are applied annually on a large-scale worldwide (Morais et al., 2012). Carbamates are a class of insecticides mechanistically and structurally alike to organophosphate (OP) insecticides (Silberman and Taylor, 2019). Carbamate insecticides are adsorbed by co-ordination and/or protonation at the carbonyl oxygen by exchangeable cations of clays (Bansal, 2010). Mostly carbamates have a half-life around 25-30 days in soil (Vlcek and Pohanka, 2012). The important examples of this kind of pesticides are aldicarb, carbaryl, carbofuran, metalaxyl, methomyl, oxamyl, pirimicarb, and propoxur (Struger et al., 2016). Commonly, the vapour pressure of carbamates is low, though they can gradually evaporate or sublimate causing to volatilization from soil and water (Struger et al. 2016). They are remarked hazardous to the environment because the role of three carbamate insecticides (baygon, carbaryl, and carbofuran), as promising cancer chemotherapeutic agents (Morais et al., 2012). Toxic exposures to carbamates can occur via dermal, inhalational, and gastrointestinal (GI) exposures. Carbamate poisoning cases are most often related to intentional oral ingestion or dermal occupational exposure. In the developing nations, cases of large outbreaks from contaminated food and crops have been reported (Silberman and Taylor, 2019).

**2.3.3. Dipyridyl (Herbicides)**

It has widely used as effective herbicides (Dalefield, 2017). In agricultural fields, it is employed before seeding annual crops, during the dormant stage of perennial crops, or as a spray directed away from growing crops (Jang et al., 2019). Paraquat is the most vital of dipyridyl, and it is the most significant herbicide in veterinary toxicology (Dalefield, 2017). The paraquat molecule has the unusual feature of being a planar dication (i.e. with two positive charges), and it is strongly bound by clay particles in soil. The strong sorption of paraquat to soil avoids leaching and losses to surface water via drainage or surface run-off during heavy rainfall (Srinivasan, 2003). Its half-life in soil is 16 months to 13 years (Rao and Davidson, 1980). In several countries, paraquats remain the pesticide active ingredient responsible for more fatal poisonings than any other pesticides. The EU has banned paraquat as the potential exposure of workers is remarked too high. Paraquat has endocrine and immunotoxic effects (Public Eye, 2017). Kim et al. (2017) reported that paraquat resulted in 3206 deaths in 2010 in Korea.

**2.3.4. Organochlorines (Insecticide)**

Organochlorine pesticides (OCPs) have been applied during half century because of their effectiveness as insecticides. Several organochlorine compounds, such as DDT, are forbidden in developed countries since 1970s, but in underdeveloped countries are still applied particularly for the control of vector-borne diseases as malaria (Moreno et al., 2006). They are typically ubiquitous, hydrophobic, resistant and persistent to degradation (Taiwo, 2019). Aldrin, BHC, chlorobenzilate, chlordane, DDT, DDD, dicofol, dieldrin, eldrin, endosufan, heptachlor, isodrin, isobenzan, lindane, methoxychlor, and toxaphene, are the important types of OCPs (Jayaraj et al., 2016). The organochlorines have been described as environmentally persistent, and remaining long time in soils and sediments (Carvalho, 2017). The lipophilic ability of OCPs lets them attach to fatty tissues in human or animal bodies. OCPs can concentrate directly in fish tissues from surrounding water and diets, thus allowing the evaluation of the contaminant's transfer over the food chain difficult (Taiwo et al., 2019). They are among the major types of pesticides notorious for their high toxicity (Gbeddy et al., 2015).

**2.3.5. Organophosphates (Insecticide)**

Kumar et al. (2018) expressed that the organophosphate pesticides are among the commercially favored group of pesticides, with large application areas all over the world. Organophosphate pesticides have attained global popularity compared to organochlorine pesticides, which are persistent and more damaging to the environment (Kumar et al., 2018). Azamethiphos, azinphos-methyl, chlorpyrifos, diazinon, dichlorvos, fenitrothion, malathion, methyl parathion, parathion, phosmet, and tetrachlorvinphos are some wide used of OPPs (Adeyinka and Pierre, 2019). The organophosphorus pesticides, such as diazinon, dimethoate, and fenitrothion, can be easily degraded by microorganisms (Motoki et al., 2016). The organic toxins pass in the water bodies or food chain, directly or indirectly, and pose a threat to existence of the human being by displaying high potency to the enzymes in cholinergic synapses i.e. Acetylcholinesterases (AChE). It is a key enzyme accountable for the normal functioning of the central nervous system in humans (Patel et al., 2019).

**2.3.6. Phenoxy (Herbicides)**

These pesticides are mainly applied to control weeds in agriculture. Nearly all compounds of this group are degraded by microorganisms (Jayaraj et al., 2016). 2,4-D(2,4 Dichloro-phenoxy-acetic acid), 2,4 5 T(2,4 5 Trichloro-phenoxy-acetic acid), dichloroprop, mecoprop, and sesone are the main phenoxy (Kafkas et al., 2019). Phenoxy herbicides display a pattern similar to the benzoic acid herbicides in terms of persistence and mobility in soil (Kamrin, 1997). Derr et al. (2016) expressed that the most phenoxy herbicides cannot persist in soil for more than a month.

2,4-dichlorophenoxyacetic acid (2,4-D) is a selective herbicide, with highest toxicity to broadleaf plants. 2,4-D is simply adsorbed into the human organism from the alimentary tract and skin and is consequently excreted in the urine in almost unchanged form (Bukowska, 2006).

**2.3.7. Phenylamides (Fungicides)**

Phenylamide fungicides are systemic compounds that display potent eradicative anti-fungal activity. When applied to the soil, they enhance plant growth and yield. They are known to effect mitosis and cell division in target fungi (Jayaraj et al., 2016). They inhibit rRNA synthesis and may control pathogens through preventive, curative, and eradicative stages of the disease cycle, chiefly because of their systemic (acropetal) translocation in crop plants (Gisi and Ziegler, 2003). Phenylamide fungicides (PAFs) are a class of systemic compounds including benalaxyl (Galben), metalaxyl (Ridomil), ofurace (Patafol), and oxadixyl (Sandofan), that display excellent protective, curative, and eradicative antifungal activity and completely control diseases leaded by Peronosporales (Gisi and Cohen, 1996). In human, phenylamide pesticides affect nucleic acids by inhibiting the activity of RNA polymerase I system (Jayaraj et al., 2016).

**2.3.8. Phthalimides (Fungicides)**

The important examples of this kind of pesticides are captan, folpet (Kafkas et al., 2019). They represent about half the usage of the dithiocarbamates. The fungicides captan and folpet react with thiols such as cysteine and glutathione at acidic pH levels of 4.0 to 5.0 (Jayaraji et al., 2016). They are listed as sensitizers and strong irritants of the eyes, skin, and respiratory airways, and their analysis is performed in worker exposure, commodity residue, and environmental studies (Raina-Fulton, 2014).

**2.3.9. Pyrethroids (Insecticide)**

The Global Pyrethroids market is assessed at $2655.25 million in 2016 and is estimated to touch $4204.18 million by 2023 (Market Research Consulting, 2017). Synthetic pyrethroid insecticides were announced into widespread usage for the control of insect pests and disease vectors around three decades ago (Soderlund, 2012). According to their impact and chemical structure, pyrethroids divided into two types: Type one, allethrin, phenothrin, and tetramethrin, do not contain a cyano group, and type two, cyfluthrin, cyhalothrin, cypermethrin, fenvalerate, and tralomethrin, are the ones that are predominantly alpha-cyano-3-phenoxybenzyl alcohol esters and they result a longer period in the sodium current (Guven et al., 2018). Pyrethroids are employed in animal houses, fields, and greenhouses (Gajendiran and Abraham, 2018). Pyrethroid persistence under aerobic soil conditions is greatly variable, with half-lives ranging from 11.5 days for cyfluthrin to 96.3 days for bifenthrin. Photolysis is possibly a significant degradation pathway for pyrethroids in the soil (Palmquist et al., 2012). Their metabolism is a concern to disrupt a cell’s homeostatic machinery via reactive oxygen species (ROS) production. Significant pyrethroid residues have been recorded in drinking water, cow milk and human breast milk in a sample location of South Africa (Guven et al., 2018).

**2.3.10. Triazines (Herbicides)**

Triazines have been employed as selective herbicides in agriculture in the United States and other locations of the world for 50 years. Even after five decades of usage, certain of these triazine herbicides remain commercially and vital, particularly for the preemergent control of broadleaf weeds (Breckenridge et al., 2008). Triazine herbicides belong to a classification, which are categorized as persistent organic compounds since they resist chemical and biological degradation (Klementova and Keltnerova, 2015). The important examples of this kind of pesticides atrazine, ametryn, cyanazine, deethylatrazine, deisopropylatrazine, didealkylatrazine, prometon, prometryn, propazine, and simazine (Trimble and Lydy, 2006). These substances interfere with the human body´s endocrine system and resulting in adverse developmental, reproductive, neurological and immunological impacts in both humans and wildlife (Klementova and Keltnerova, 2015).

**Table 6:** Pesticides groups based on their chemical nature

**2.4. Presence of Pesticides in Waters and Wastewaters**

Pesticide residues have been identified in numerous water bodies (Derbalah et al., 2019). Pesticides are capable to transfer from one ecosystem to another through procedures such as transformation (degradation) and transfer (mobility) (Mensah et al., 2014). To better understand the mobility of pesticides, three main factors, including octanol-water partition co-efficient (Kow; which is usually shown by log Kow), the sorption coefficient (Koc; which is sometimes shown by log Koc), and acid ionization constant (pKa), should be described. Paschke et al. (2004) expressed that the octanol/water partition coefficient (Kow) is a vital physicochemical descriptor for evaluating transport pathways and environmental partitioning. In the aquatic systems, pesticides are simply absorbed by particulate organic matter because of their high Kow values, and can result in bioaccumulation and biomagnification in aquatic organisms (Arisekar et al., 2019). Pesticides with log Kow less than 3 are low lipophilic, with log Kow 3 to 5 are moderate lipophilic, and with log Kow more than 5 are highly lipophilic (Knauer et al., 2017). Material with very low lipophilicity might not be able to pass lipidous membranes, but materials with a high lipophilicity will accumulate in fat tissue and other lipophile phases (Hansen, 2004). The Kow is also a suitable guide of systemic mode of action of a pesticide. Pesticides with low Kow values (generally ≤2) signify the likely systemic translocation of such pesticides or their metabolites in the plants transvascular system (Zacharia, 2011). Boithias et al. (2014) stated that there are a strong relationships between Koc and Kow. The Koc is the described the coefficient between soil particles and water normalized to the OC content of the particles (Knauer et al., 2017). Pesticides with high Koc have the stronger the tendency to attach with soil. Apart from that, soil pH could affect the Koc of ionic and partially ionic pesticides (Motoki et al., 2014). Another important factor is pKa which displays the tendency of a non-ionized acid to separate into an ionized compound and a hydrogen ion (Pereira et al., 2016). Mendes et al. (2019) stated that the pKa of pesticides and pH of the soil solution affect the sorption behavior of the molecule.

The aquatic ecosystem comprises of different groups of organisms such as fish, plants, amphibians, invertebrates or microorganisms. Several researchers have been reported pesticides in aquatic environments, which are listed in the Table 7. Based on the Table 7, the concentrations of the pesticides (with 1% to 10% error) ranged between 6.8 ng/L to 121222 ng/L in rivers and lakes. In wastewaters, pesticides ranges from 23.2 ng/L to 3172 ng/L have been reported in literature. And finally pesticides concentrations in groundwater and drinking water have been ranged between 20.4 to 1060.0 ng/L, and 141 to 14629 ng/L, respectively.

**Table 7:** Most reported pesticides in water sources

**2.5. Effects on Fishes**

Water soluble pesticides become dissolved in water and enter ground water, streams, and lakes and rivers and then resulting in harm to untargeted species. In another word, fat soluble pesticides enter the bodies of animals by a process known as “bioamplification” (Mahmood et al., 2015). Jacquin et al. (2019) investigated the high temperature aggravates the impacts of pesticides in goldfish, their results revealed pesticide exposure caused higher genotoxic impacts (micronuclei rate) and irreversible cellular damage of the gills and liver (apoptosis, inflammation, necrosis) at 32 °C compared to 22 °C. Pérez-Parada et al. (2018) reported several kinds of pesticides, such as: OCPs and OPPs, in different fish species, such as: *Oreochromis mossambicus*, *Clarias gariepinus*, *Sarotherodon galilaeus*, *Clarias anguillaris* and *Cirrhinus molitorella*. Ernst et al. (2018) investigated the occurrence of pesticide residues in fish from South American agroecosystems. Their findings indicated that thirty different pesticides were identified at concentrations from <1 to 194 μgkg−1. Incidences of pesticides in fish were strongly related to: i) factors of the contaminant: (Kow, environmental persistence and mobility) and ii) intensity of usage of particular pesticides and land dedicated to agriculture. Trifloxystrobin, pyraclostrobin and metolachlor displayed the highest rates of occurrence. Shankar Murthy et al. (2013) expressed that pesticides are able to kill salmon and other aquatic life directly and within a short period of time. Pesticides leach into aquatic habitats may influence several physiological processes that may impact upon the larvivorous potential of fishes (Shankar Murthy et al., 2013). The bioaccumulation of pesticides in fishes’ samples is shown in Table A.2 (in appendix). The maximum concentration (26179.5 ng/g) of pesticides in fish’s samples was detected in Rohu Fish for accumulation of ∑Endosulfan in this fish in India (Arisekar et al., 2019)

**3. Removal Pesticides from Aqueous Solutions by Adsorption**

Extensive research has been concentrated on effective sequestering of organic contaminants from aqueous solution. A variety of methods have been adopted such as coagulation, chemical oxidation, membrane filtration, photocatalytic degradation, and adsorption (Khairy et al., 2018). All organic pollutants removal may be divided into two main groups, including biological and physical/chemical methods. Among several treatment methods, one of the key strategies for elimination of pesticides from water is the adsorption on different types of materials (Momić et al., 2016). Sophia and Lima (2018) stated that adsorption is the one relevant and promising technique for eliminating organic and inorganic contaminants., due to the important benefits like availability, profitability, ease of operation, and efficiency than other methods (Uddin, 2017). Many studies relating to the application of different adsorbents for organic contaminant elimination have been published over the last few decades (Sophia and Lima, 2018). Ariffin et al. (2017) stated that simplicity and flexibility of design, ease of operation and insensitivity to toxic contaminants are most advantages of adsorption method, and needing regeneration processes is the main disadvantages of adsorption method. Among several adsorbents, activated carbon (Gupta et al., 2011), graphene (Madej et al., 2018), biochar (Mojiri et al., 2019a), bentonite (Durán et al., 2019), zeolite (Vakili et al., 2019), chitosan (Mojiri et al., 2019b) and nanoparticle adsorbents (Boruah et al., 2017) have been mostly used in organic pollutants removal from aqueous solution. Table 8 displays the adsorption capacity of most reported adsorbents during adsorption of pesticides.

**3.1. Activated carbon (AC)**

Activated carbon is described as a carbonaceous material with a highly porous internal structure, which is frequently derived from the pyrolysis and chemical treatment of sources comprising wood, nutshells, coal and bamboo and other organic materials (Sweetman et al., 2017). Ligneris et al. (2018) stated that AC has been commonly applied against organic compounds and displayed high adsorption capacities for a range of initial contamination varying between 15 µg/L and 80 mg/L. Based on the literature review, the main advantages of AC are the high adsorption capacity (Ligneris et al., 2018), and large surface area and highly porousity (Abraham et al., 2018). Using AC is costly, disadvantage of AC. These reasons restrict the use of AC in a large scale. Frequently the reuse of AC impregnated with organic compounds is lower than 40% (Sophia and Lima, 2018). Salman et al. (2013) reported 164 mg/g adsorption capacity of insecticide (carbofuran) removal by AC. Based on their regeneration study, the regeneration effectiveness of spent AC was 90.0% to 96.4% by using ethanol. Chang et al. (2011) reported 296.92 mg/g as maximum adsorption capacity during carbofuran (insecticide) removal by AC. 99% of triazole (fungicide) was removed by AC based adsorbent (Crini et al., 2017).

**3.2. Graphene based adsorbents**

Graphene, a single-atom-thick planar sheet of sp2-bonded carbon atoms organized in a hexagonal crystalline structure, possesses unique physicochemical properties, remarkably the large specific surface area (theoretically 2630 m2/g), rapid heterogeneous electron transfer, high thermal conductivity, and outstanding mechanical strength (Xu and Wang, 2017). Usually, graphene gained from graphite exists in two shapes, i.e., graphene oxide (GO) and reduced graphene oxide (RGO). GO is water-soluble with low conductivity but RGO has good conductivity with poor solubility in water (Cao and Li, 2014). There are three promising sites in graphene for the adsorption of contaminants: (1) one is sites on the oxygen functional groups, (2) another is assessed to be sites of Cπ-electrons on graphene sheets, and (3) another is the active site on the outer surface of the decorated nanoparticlels on graphene sheets (Cao and Li, 2014). Lazarević-Pašti et al. (2018) listed advantages of graphene as: its excellent thermal conductivity (5000 W.m-1K-1), high specific surface area (2600 m2.g-1), high-speed electron mobility at room temperature (200000 cm2.V-1S-1), optical intriguing properties and extraordinary electrocatalytic activity. Nupearachchi et al. (2017) expressed that commonly the interactions with hydrogen bonds, hydrophobic impact, π-π bonds, electrostatic and covalent bonds have been found in carbon materials. These are supposed to be playing a main role in the adsorption of organic chemicals of carbon adsorbents. It was defined that the GO displayed higher adsorption capacity compared to graphite for organic compounds. Zhang et al. (2015) removed more than 95% of triazine pesticides by using cellulose/graphene composite in the optimum performance. More than 89% of glyphosate (herbicide) was removed by ferrite manganese/graphene, as reported by Yamaguchi et al. (2017).

**3.3. Biochar**

Biochar chiefly comprises of carbon, oxygen, hydrogen and nitrogen. The carbon content is in the range of 38–80%, and most of it is included in the alkyl and aromatic matters. The pH of biochar commonly varies from neutral to alkaline, while acidic biochar has been also reported (Yuan et al., 2019). Yuan et al. (2019) expressed that the biochar surface carries negative charges were because of the occurrence of organic groups, hence its cation exchange capacity (CEC) may be enhanced. Biochar characteristics comprise porosity (including several pore sizes), large surface area, pH, ash, CEC, EC and nutrient level. The pore size of biochar varies depending on the material applied for biochar production and commonly ranges from nano (<0.9 nm), micro (<2 nm) to macropores (>50 nm) (Shaaban et al., 2018). Graber et al. (2012) and Wang S et al. (2018) reported some advantages, such as high surface area, extensive raw material sources, low-cost, and high environmental stability, for using biochar for adsorption on pollutants. Suo et al. (2019b) expressed that usually the diameter of some pesticides are less than the pore size of the biochar. Therefore, pesticides ions can be caught in the mesoporous and micropores on the surface of biochar. Ćwieląg-Piasecka et al. (2018) removed 76.4%-84.3% of carbamates and 70.2% of metolachlor by biochar. Around 96% of six triazine was removed by a modified biochar (Suo et al., 2019b).

**3.4. Mechanism of removal of different pesticides by carbon-based adsorbents**

The pesticides were adsorbed on graphene due to its high specific surface area, as well as through noncovalent interactions, especially the π-π stacking interaction with the aromatic rings of the studied compounds, and the hydrophobic effect (Madej et al., 2018). Organic compounds can be adsorbed by graphene or graphene oxide (GO) through five electrostatic interactions including hydrophobic effects, covalent, hydrogen bonding and π-π stacking. The main interaction between organic ions and graphene are π-π stacking, πcation-π interaction (Cheng et al., 2013), dative bonding and hydrophobic effects. Cation polarization and electrostatic interaction between cations can cause formation of πcation interaction in the adsorption process (Huang et al., 2007(. GO can form H-bonding with organic compounds including oxygen and nitrogen groups. Meanwhile, graphene is suitable as adsorbent for extraction of aromatic compounds without polar functional group due to rich delocalized π-electron system that provides strong affinity via π-π interaction with carbon-based aromatic ring structure (Ibrahima et al., 2015). For example regarding chlorpyrifos (CPF) adsorption by graphene nanoparticle (GNP), a very strong adsorption of CPF on GNP is due to the aromatic moiety of CPF, which enables the π-π stacking interactions on the GNP surface. The aromatic ring of CPF can interact with the π electron system of graphene basal plane (Lazarević-Pašti et al., 2018).

Aromatic compounds adsorb on carbons by a donor–acceptor complex processes, with the carbonyl oxygen of the carbon surface acting as the electron donor and the aromatic ring of the adsorbate acting as the acceptor. Once the carbonyl groups are exhausted, the aromatic compounds form donor–acceptor complexes with the rings of the basal plane (Moreno-Castilla, 2004). For example for carbofuran adsorption, carbofuran adsorption phenomena could be explained by several mechanisms. There may be π−π electron donor-acceptor (EDA) interactions between protonated aromatic ring of the carbofuran molecule and π-electron rich graphene surface of the BCs. Moreover, the presence of -NH2 on the surface of biochars, -NH2 can combine with carbofuran through acid-base interaction as carbofuran is a weak monoprotic acid (Mayakaduwa et al., 2016).

**3.5. Bentonite and clay**

One of the effective and low cost adsorbents is clay minerals. Natural clay minerals due to their molecular sieve structure and high surface area are effective sorbents for organic pollutants of cationic or polar in character (Rashed, 2013). zabo et al. (2011) expressed that clay minerals such as sodium montmorillonite might be proficiently altered by cationic surfactants in water using ion exchange reactions. Permanent negative charges on the clay mineral sheets may be compensated by organic cations by replacing the sodium ions. The main advantages of clay minerals are inexpensive precursors, surface area, high cation exchange capacity, micro- and meso-porosity and swelling properties (Rathnayake et al., 2016). In some studies before using clay for adsorption of organic pollutants, some treatments would be done on clays, such as producing organoclay. An organic-based cation, such as quaternary ammonium salt, substitutes inorganic ions such as sodium, calcium, or magnesium on the surface of clay soil. These consequences in the detachment of clay layers from each other and increased interlaminar basal spacing. These changes modify the clay nature from hydrophilic to organophilic (Heidarzadeh et al., 2017). El-Geundi et al. (2012) reported the removal of 27.6% to 32.9% of insecticide (methomyl) by using natural clay. Around 99% of sulfentrazone, 99% of sulfosulfuron, 73%-93% of imazaquin, 95% of chlorotoluron, and 90% of acetochlor were removed by a modified clay (Polubesova et al., 2005). Ahmad and Yasin (2018) removed 98% of deltamethrin by using a modified bentonite.

**3.6. Zeolite**

Zeolites are economically and environmentally viable hydrated aluminosilicate materials with exceptional ion-exchange and sorption properties (Muir et al., 2017). The properties of zeolites vary by the proportion of silica and aluminium content, namely the silica to aluminium ratio (Si/Al ratio). Low-silica zeolites, with a Si/Al ratio less than 2, have excellent ion exchange capacity (Jiang et al., 2018). Muir et al. (2017) expressed that main advantages of zeolite are good ion-exchange capacity and a high affinity with the cationic form. The zeolite is categorized by a negative overall charge. Hence, all cations are freely adsorbed on their surface. Maximum 100% of bentazone (herbicide), 16.9% of clopyralid (herbicide), 99.7% of imidacloprid (insecticide), 100% of soproturon (herbicide), and 99.8% of metalaxyl-M (fungicide) were removed by different zeolites (Smedt et al., 2015). All carbamate was removed by modified zeolite (Arnnok and Burakham, 2014).

**3.7. Mechanism of different pesticides removal by aluminosilicate based-adsorbents**

Clay minerals have two kinds of surface charge: a net permanent surface charge σ0 and a variable charge σH. The net permanent charge results from isomorphic substitutions of the mineral of Si(IV) by Al(III) in the tetrahedral sheet or, indirectly, from isomorphic substitutions in the inner layers (Polati et al., 2006).

**3.8. Chitosan based adsorbents**

One of the biopolymers that is derived from chitin is chitosan, whichis made of glucosamine and N-acetylglucosamine units. The presence of N-acetylglucosamine and glucosamine units in chitosan structure contributes to its flexibility and its heterogeneity in the polymer. The presence of hydrophilic functional groups counting amino and hydroxyl groups cannot change chitosan’s hydrophobic nature, and support to make chitosan capable for adsorption and modification (Wang and Zhuang, 2017). The advantages of chitosan comprise its low cost, ease of polymerization and functionalization, and good stability (Mojiri et al., 2019a). Usually researchers have done some modification on chitosan to improve its adsorption, including physical modifications such as blending, and chemical modifications such as crosslinking and grafting (Wang and Zhuang, 2017). Hence, ethoprophos (insecticide) was removed by 85.6%-89.2% by chitosan (Abdeen and Mohammad, 2014). More than 90% of oxadiazon (herbicide) was removed by chitosan (Arvand et al., 2009).

**3.9. Composite adsorbents and nano-adsorbents**

Nowadays, more attention has been spent on nano-adsorbents and composite adsorbents, which have been frequently caused by their high specific surface area, as well as the lack of diffusion resistance (Dai et al., 2018). Abdelhameed et al. (2016) removed around 97% of ethion (insecticide) by Cu-BTC@Cotton composite. 82.5% of diazinon (insecticide) was removed at 60 min and pH (5.5) by chitosan/carbon nanotube (Firozjaee et al., 2017). 97.7% of λ-cyhalothrin (insecticide) by chitosan-zinc oxide nanocomposite (Shalaby et al., 2018). Rahmanifar and Dehghani (2014) removed 99% of permethrin (insecticide) by chitosan/silver oxide nanoparticles. Aminosilane modified magnetite nanoparticles removed 84% of diazinon (Naeimi et al., 2018). Bandforuzi and Hadjmohammadi (2019) reported 99%, 98% and 96% removal of diazinon, phosalone and chlorpyrifos by magnetic chitosan nanoparticles, respectively. 99% of permethrin (insecticide) was removed by chitosan–zinc oxide nanoparticles (Dehghani et al., 2014). Mansouriieh et al. (2015) removed 94.51% of profenofos (insecticide) via zero-valent bimetallic nanoparticles.

**Table 8:** Pesticides removal by adsorption methods

**3.10. Effects of pH on pesticides removal by adsorption**

Among the characteristics of the adsorptive that chiefly influence the adsorption procedure are its solubility, molecular size, pKa and the nature of the substituent if they are aromatic. The molecular size adjusts the availability to the pores of the carbon and the solubility determines the hydrophobic interactions. The pKa controls the dissociation of the adsorptive if it is an electrolyte. This parameter is closely related to the solution pH (Moreno-Castilla, 2004). Commonly, the pH is considered as a significant factor during the adsorption process. It has been accepted that pH affects the surface charge of the adsorbents. Therefore, during pesticides removal by adsorption, effects of pH should be monitored. Bahrami et al. (2018) expressed that initial pH affects the electrostatic interaction between pesticide molecules and the surface of adsorbents. Omri et al. (2016) reported that the optimum adsorption was defined at pH 3.5 during insecticide removal by activated carbon. Salman and Hameed (2010) reported that the equilibrium adsorption of carbofuran was determined to decrease slightly when the initial pH of the aqueous solution was increased from 2 to 12 during insecticide removal by activated carbon. Salman et al. (2011) expressed that adsorption capacity of activated carbon was decreased with increasing pH during removal of 2,4-D. Bahrami et al. (2018) stated that maximum removal of 2,4-D by using biochar and activated carbon was achieved at acidic pH. El-Nahhal and Safi (2008) reported that the maximum removal of bromoxynil (herbicide) gained at pH = 3 during bromoxynil removal by organobentonite (El-Nahhal and Safi, 2008). The explanation of these results is that at pH = 3, the majority of bromoxynil molecules are mainly neutral and/or protonated, they might interact with the organobentonite surfaces either as cations or as neutral molecules. For some pesticides, maximum removal has been reached at neural or alkaline pHs. Maximum removal of paraquat (herbicide) by biochar was reached at pH (7.5) (Tsai and Chen, 2013). Tsai and Chen (2013) expressed that the adsorbed amount of cationic paraquat improved in response to the increasing number of negatively charged sites that are available due to the loss of H+ from the surface. Maximum pesticides (atrazine and imidacloprid) removal by TRSBC (phosphoric acid treated rice straw biochar) was achieved at pH (6.9) (Mandal and Singh, 2017). Mandal and Singh (2017) stated that lack of deprotonation at lower pH might be suitable for the sorption of the ionisable compounds.

**3.11. Desorption processes**

Regeneration of adsorbents is a key procedure in adsorption process to diminish the processing cost. Numerous regeneration ways have been employed for desorption studies, comprising chemical regeneration and thermal regeneration. Nonetheless, it is vital to choose the applicable pH and desorbents (such as inorganic desorbents NaOH, HCl, and H2SO4, or organic desorbents ethanol, methanol, acetonitrile and acetic acid) for the chemical desorption procedure. Based on Table 9 for regeneration of adsorbents, mostly organic desorbents have been employed. Mojiri et al. (2019a) expressed that some organic pollutants are highly soluble in alcohols due to the presence of hydroxyl groups. Additionally, the low molecular weight alcohols might enhance the efficacy of organic pollutants desorption (Mojiri et al., 2019a).

**Table 9:** Desorption process after adsorption of pesticides

**4. Conclusions**

Monitoring as the occurrence of pesticides in water sources and their removal with highly efficient techniques have attracted attentions in the scientific community. In this paper, we tried to review recent research progress on the occurrence of pesticides in water sources and the application of adsorption method in removing these organic pollutants. The key conclusions of this research review are listed below:

* Maximum reported pesticide concentrations in water bodies were 121222.1 ng/L in Karun River, Iran.
* Carbon-based adsorbents such as activated carbon, and chitosan/silver oxide nanoparticles have shown the highest performance in removing pesticides with removal efficiency around 100%.
* pH is considered as a vital parameter during the adsorption of pesticides. Mostly maximum removal of pesticides by adsorption processes was occurred in acidic pH.
* For desorption and regeneration of adsorbents after adsorption of pesticides, organic desorbents, such as ethanol and methanol, have been mostly effective.

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