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Removal of pharmaceuticals from wastewater: a review of different adsorptive approaches

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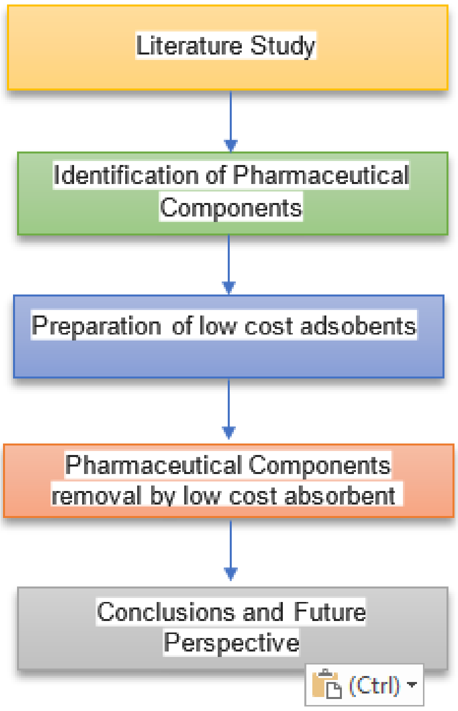
Graphical abstract



Abstract

Over the past few decades, pharmaceuticals have contaminated the marine ecosystem. The entry of prescription medications into the environment, their underlying causes and problems, and effective methods for treating such contaminated water are all evaluated in this study. Adsorption is becoming a more popular preferred treatment method because, compared to other post-secondary treatments, it has less strength requirements and is easier to use. Despite being extensively researched as a chemosorptive for prescription medications, industrial activated charcoal is heavily restricted due to its expensive cost. For the purpose of eliminating prescription drugs from water and wastewater, novel unconventional low-cost alternatives were looked into, and adsorbents based solely on clay, biochar, agricultural wastes, industrial wastes, and metal-natural frameworks were discussed in a number of research studies. This second study examines prominent articles that address the problem, covering the continuous fixed-bed process, regeneration capabilities, historical, economic, and practical aspects, as well as adsorption performance in terms of kinetics, equilibrium, and thermodynamics. Chemicals known as pharmaceuticals pollute the environment. This paper discusses the prevalence of pharmaceuticals in the environment, their main causes and effects, and proactive steps to remove them from the contaminated medium. This summary explains how the pollutant is mixed with the aquatic environment, along with its fundamental causes, insinuations, and effective methods for eliminating it. Compared to other therapies, adsorption offers advantages including less strength depletion and easier functioning conditions, making it a promising therapeutic approach.

Keywords: Adsorption, low-cost adsorbents, emerging contaminants pharmaceuticals removal

1. Introduction

Of late, the world has seen substantial growth in the devouring and utilization of prescription drugs that caused the liberation of massive quantities of many such poisonous chemical compounds within the surroundings. Numerous investigators stated the existence of pharmaceutical contaminants in various aquatic media and effluent treatment units (Villaescusa *et al*. 2018; Khazri *et al*. 2017). The majority of such composites are tenacious by nature and cause several damaging complications on people’s well-being and the atmosphere due to their bioaccumulation (Zhang *et al*. 2013). Although nation- of-the-art treatment procedures which include cutting-edge oxidation procedures, membrane bio-reactors, and so on can be employed for getting rid of those pollutants, excessive prices along with superfluous power needs save

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you the wide-ranging usage of such methods (Reynaud *et al*. 2011; Salem *et al*. 2016). Subsequently, the adsorption method gathers great recognition for eliminating numerous such contaminants it is economical and considerably easy in terms of usage. Bearing these in mind, several studies have suggested adsorption as a tertian method for relieving the effluent of pharmaceutical contaminants. Still, the overall expenditure of the adsorption procedure rests within the selection of adsorbent complete with its manufacturing cost, reusability, and recyclability, along with the comfort of usage both during and after the treatment. Over the years, investigators have worked towards the recognition / progression of economical, as well aseffective adsorbents concerned with nutrient, pharmaceuticals, and personal care products (PPCPs) elimination. Various investigations state the ability of several refuse-derivative adsorbents (activated carbon/bio-chars) formed following the synthetic/heat-related treatments using resources such as coconut shell, bamboo, pine wood, coffee wastes, fish waste, sewage sludge, etc for PPCPs removal (Reynel-Avila *et al*. 2015; Ferreira *et al*. 2017; Hasan *et al*. 2012). Such remedies substantially enhance floor characteristics along with the permeable formation of the substances. These inordinate synthetic and energy requirements prevent the huge-scale application of those substances. Some researchers also tried first-hand usage of Agro-business refuse for the adsorption of numerous contaminants, inclusive of medical waste contaminants. Specifically, the adsorption action of decided-on PPCPs over substances including mild-weight multiplied clay combination (LECA), exfoliated vermiculite, and cork granulate were substantiated within these preceding studies. (Peng *et al*. 2018; Azhar *et al*. 2016; Azhar *et al*. 2017; Salihi and Mahramanlıoğlu 2014).

Because of enlarged commercial traits, a huge amount of waste is generated all around the globe. Implementing waste materials as adsorbents should in all likelihood bring about more benefits of low fees, excessive profusion, and availability, along with a fractional discount of residue. Previous research has used waste substances inclusive of zeolites, industrial sludge, activated sludge, energetic alumina, bentonite, montmorillonite, brick debris, extended earth, quarry dust, biochar, steel slag, plastic and rubber substituting substratum in micro- organism investigations related to the elimination of organics, nutrients and heavy metals (Genç *et al*. 2013; Roca Jalil *et al*. 2015; Chang *et al*. 2009). Even though such investigations confirmed an affordable elimination of organics and nutrients, their ability for the elimination of PPCPs has not been researched broadly yet. Furthermore, adsorption in comparison to the substances is sturdily prompted using properties of the substances, physicochemical houses concerning the goal contaminants, ecological situations along with the pH, temperature, and electrolyte composition, etc. (Dordio *et al*. 2017). Therefore, at the same time as choosing a suitable sorbent concentrating on unique contaminant elimination, simple information on traits of the material, and the interaction between the pollutants and the cloth is crucial. In addition, interpreting the price along with the volume of the adsorption procedure is unavoidable for creating the remedy systems. With concern to herbal remedy structures which includes man-made wetlands, these permeable resources offer a large floor area for attachment of the microbial groups such that it facilitates the deterioration of the sorbed contaminants (Liu *et al*. 2012). Also, it is certainly thrilling to know that these permeable substances can potentially be utilizedby microbial companies as bio-improved deterioration structures. Therefore, an efficient choiceof cloth shall improve the adsorption potential with regard to the substances concerned with contaminant elimination. Selecting the material is in the main related to its capability for adsorption of the contaminant which is in turn motivated by the belongings of the substances (Zha *et al*. 2013). The key aim of this study lies in evaluating the sorption properties of the nutrients in addition to the three extremely recurrently noticed pharmaceuticals in India, viz. carbamazepine, diclofenac, and ibuprofen using economical sorbents comprising of natural and manufacturing residue by group investigations. Sand, natural zeolites (NZ), natural pyrite (NP), lightweight expanded clay aggregate (LECA), waste autoclaved aerated concrete (AAC) blocks, brickbats (BB), blast furnace slag (BFS) and wood charcoal (WC) are the materials that are utilised in this process. This study also tried to ingrain the connection between the properties of substances and the designated contaminant elimination, by association studies. The combative adsorption (antagonistic / synergistic) concerning the designated composites against the substances was scrutinized in individual and multi-contaminant situations. The percolation of adsorbent contaminants from the constituents was studied by desorption studies. Impacts due toconditions such as pH (2–12), organic substances, and nutrients were also examined. The solidity along with the rigidity of the wet materials was also tested. Therefore, this introductory study was principally concentrated on detect ingre sources that can be best utilized in a sorption structure or can be used as a medium in natural methods for treatment such as man- made marshlands or biologically improved deterioration systems, an effort was also undertaken to deliver perception of the contaminant treatment by various natural and waste materials.

2. Biochar from various feedstocks for removal of pharmaceutical components

PCs have been successfully removed from aqueous matrix using biochar. PC removal from diverse water matrices has attracted a lot of attention and use of biochar, a cheap and efficient adsorbent. Biochar made from plant remains Various biomasses derived from plants have been used as raw materials to create biochar for PC adsorption. Plant leftovers are naturally renewable, easily accessible, and affordable (Sun *et al*. 2017; De Oliveira *et al*. 2017; Carballa *et al*. 2004). The features and composition of the feedstock affect the properties of biochar. For example, most plant residues constitute lignin (27%), cellulose (43%), and hemicellulose (20%), which affect the elemental composition of biochar. Biochar produced from lignocellulosic biomass has a higher carbon content. Various plant residues like corncobs, pine needles, rice hulls, coconut shells, banana pseudostem, wood chips, oil palm fiber, pomelo peel, moringa seed powder, walnut shells, date stone seeds, mung bean husks, wood apple (Aegle marmelos) fruit shells, eucalyptus globulus wood, and hickory chips, have been used as feedstocks for the production of biochar for the adsorption of various PCs. Biochar made from animal waste leftovers Animal wastes such as pig dung, chicken feathers, cow bones, fsh scales, etc. have been utilized as feedstocks for the synthesis of biochar. These biochars made from animal manure have been effective for adsorbing PCs. For example, biochar made from cow's bones was used to absorb CAF. At 650 °C, the bone biomass was pyrolyzed and combined with clay minerals to form a composite. produced from MSW biochar The exponential rise in population and ineffective waste management and treatment practices is mostly to blame for the exponential rise in solid waste generation. Pyrolysis is one treatment approach that has gained a lot of interest from the scientific community since it uses heat to transform garbage into biochar. This biochar made from solid waste has been used to clean up the environment (Kwon and Rodriguez 2014). Indeed, studies have demonstrated the adsorption of different PCs on biochar made from MSW, food and garden waste, less waste, textile effluent treatment plant sludge, etc. modified and engineered biochar. To increase biochar's adsorption power, its structure has undergone numerous changes. The characteristics of biochar have changed significantly as a result of these modification techniques, which has an impact on the adsorption mechanism. For improved PC adsorption, various biochar modification techniques exist, including physical alterations, acid-base changes, ball milling, clay-biochar composites, and metal-biochar composites. physical arousal Biochar develops internal holes through the restriction and limiting of oxygen during pyrolysis (Oosterhuis and Sacher 2013). By exposing biochar to oxidizing agents—typically steam or carbon dioxide at various temperatures, it can be physically activated. Physicochemical characteristics and adsorption capacity are improved as a result of physical activation. It specifically improves the SSA, overall pore volume, average pore size, and surface chemistry of biochar. As an illustration, apple shell feedstock used to make biochar was steam activated. The SSA of the pure biochar was 4.4 m^2 g1. Steam activation, on the other hand, led to an SSA of 308 m^2 g1. Microspores were also produced by the steam activation, which also changed the chemical structure of the biochar. The pore volume calculations showed that these modifications had also occurred, with the pristine biochar having a pore volume of just 0.184 cm3 g1 compared to the steam-activated biochar's pore volume of 0.384 cm3 g1. Modification via acid-base Changes to the surface properties of biochar are made using acid or alkaline solutions (Vinayagam *et al*. 2022). The SSA and pore volume are both increased by chemical activation, and pores are also concentrated in a narrower range of pore sizes. One notable instance is the chemical activation of biochar made from swine dung, which raised the SSA from 227.56 m2 g1 to 319 m2 g1. Additionally, the post-activation pore capacity increased by 79% as a result of it. The most commonly used acids in acid activation are phosphoric (H_3PO_4), sulfuric (H_2SO_4), nitric (HNO_3), and hydrochloric (HCl) acids. For alkali activation, hydrogen peroxide (H_2O_2), potassium permanganate ($KMnO_4$), and ammonium persulfate [(NH_4)2S_2O8] are commonly used. The development of extra surface active sites on the biochar surface, primarily as a result of the removal of minerals and other silicates, is another major factor in the alterations in the acid-alkali-treated biochar (Topare and Wadgaonkar 2023). A ball mill A green modification technique for biochar engineering is the solid state grinding of biochar to nanometric size in a ball mill. Owing to an growth in SSA, opening of pore structures, and modification of surface functional groups, all of which encourage - and electrostatic interactions, ball milling improves the adsorption effectiveness of biochar. For instance, after being ball milled, biochar made from wood chips produced an SSA of 841 m2 g1. Also, by increasing the biochar-to-ball weight ratio to 1:5 and using a milling time of 5 h, the ideal ball milling parameters for the highest adsorption of PCs, including ACE, IBP, and SA, were identified. Additionally, the pore network changes as a result of ball milling. The proportion of mesopores typically increases by many folds while the amount of macrospores is decreased in biochar, which improves the diffusion of adsorbates (Tatarchuk *et al*. 2023). Ball milling consequently modifies the physicochemical characteristics of biochar, resulting in increased exterior and interior surface areas as well as more acidic surface functional groups than in virgin biochar. Biochar-clay composites Due to their synergistic effect, adding specific components to biochar, like clay, minerals, and metals, causes it to acquire those qualities. On biochar, MMT, kaolinite, and palygorskite are typically impregnated. The adsorption of several PCs has been accomplished with the use of biochar-clay composites. For instance, the improved adsorption of the MSW-derived biochar-MMT composite was caused by the improved active sites provided by the biochar and the clay material. CFX adsorption onto the composite was found to be up to 40% higher than it was for virgin biochar. The study found that biochar composite made from MSW may adsorb aromatic CFX molecules. Electron donor-acceptor interactions with the MSW-biochar are caused by the electrostatic attraction between CFX and MMT. Biochar-metal composites. In order to create metal-biochar composites, a variety of metals, their oxides (MgO, MnO, Al2O3, Fe_2O3, CaO), and hydroxides ($AlHO_2$, Mg $(OH)_2$) are frequently utilized. The negative surface charge of the biochar is changed to a positive surface charge by the impregnation. For instance, the biochar impregnated with Fe_2O_3 has an SSA of 786 m2 g-1. According to the study using energy dispersive X-ray spectroscopy, the material is composed of 68.75% carbon, 23.37% oxygen, and 7.23% iron. Several new functional groups appeared as a result of the iron being added to the biochar's surface. Additionally, the morphological and textural characteristics were different from those of pure biochar, which had a big impact on how well it worked as an adsorbent.

2.1. Environmental occurrence

Natural and artificial substances have been thought to affect an animal's hormonal interest for more than 80 years. The issue of complicated chemical substances developed for commercial, agronomic, and private use contaminating water supplies became a topic of discussion in the United States starting in the 1960s. In the 1990s, feminization and other procreant changes in aquatic species from downstream wastewater treatment plants (WWTPs) have been documented. The administration of synthetic oestrogen diethylstilbestrol (DES) to childbearing women for the prevention of stillbirths in the 1970s demonstrated the effects of estrogenic compounds on human fitness: DES recipients have been found to have vaginal cancer and womb-related abnormalities (Gkika *et al*. 2023). Even if the propensity of pharmaceuticals to stimulate the endocrine system is obvious, the first study that focused primarily on the discovery of such compounds in water and wastewater dates back to 1965. Prescription medications were reported as environmental pollutants at low amounts later in the 1990s. Utilising the U.S., pioneering movements were carried out. Listings of priority pollutants are provided by the Environmental Protection Agency and the European Union.

2.2. Concentrations in the environment

More expansive and thorough judgements on the presence of prescribed medications in their environment have been made possible by improvements in analytical techniques for locating them, even at lower quantities (ng/L-g/L). Recent research by the five regional offices of the United Nations revealed the prevalence of 631 distinct medicines and their metabolites in 71 nations (Khoshraftar *et al*. 2023). Every area had traces of sixteen pharmaceuticals, including diclofenac, carbamazepine, ibuprofen, sulfamethoxazole, naproxen, estrone, oestradiol, ethinylestradiol, trimethoprim, paracetamol, norfloxacin, and acetylsalicylic acid, in the groundwater, drinking water, and floor. Oestradiol and ciprofloxacin had notable average values ranging from 0.003 g/L to 18.99 g/L, respectively.

2.3. Environmental sources

The main ones are stated right here, however there are many other avenues and sources for learning about the prevalence of medicines in water media. Prescription medications that have been metabolised to a certain degree in human bodies and eliminated in faces and urine are fed to families and consumed in fitness facilities. Hospital wastewater may be treated one at a time before reaching WWTPs. Due to improper disposal, expired or rejected medications may also reach wastewater treatment plants (WWTPs), where a number of the prescribed medications and their metabolites are partially or completely degraded. As a consequence of this, a variety of microbial metabolites and chemicals are introduced into the surface water and ground water through the effluent discharge. (Kumar *et al*. 2023) During treatment in WWTPs using the sorption method, some pharmaceutical contamination may be converted to sewage sludge, which can subsequently be burnt, discarded, or applied to agricultural regions. Alternatively, the pharmaceutical contamination may remain in the water. In the most extreme situations, the aquatic environment may be supplemented with the sludge's leftover chemicals. Additionally, prescription medicines used as cattle treatments, growth boosters, or fish feed components in rural areas may contaminate food supplies. Additionally, drugs are continuously introduced into the environment by the addition of particularly contaminated pharmaceutical industry effluents. Take Dollar *et al*. as an example. Pharmaceuticals can appear in water media through a number of different channels and sources, and the most significant ones are covered below. Prescription medicines that have been partially metabolized in human bodies and eliminated in faces and urine are acquired by WWTPs and may be consumed in healthcare settings and by families. Health facility wastewater can be treated one at a time before reaching WWTPs. Due to off-site disposal, expired or rejected capsules may also reach wastewater treatment plants (WWTPs), where some pharmaceuticals and their metabolites are partially or completely degraded. As a result, a mixture of microbial metabolites and parent compounds enter the groundwater and floor through effluent discharge. A few pharmaceutical pollutants may be transferred to sewagesludge during the treatment process in WWTPs, which can then be landfilled, burned, or transported to agricultural fields. In extreme circumstances, the sludge's closing chemicals may be introduced to the aquatic environment. Additionally, medications utilised in agriculture as cow treatments, growth boosters, or fish feed components may contaminate food supplies.

2.4. Effects on the environment

Pharmaceuticals have a living, ongoing, and bio-accumulative biological nature. Despite being present in small concentrations (ng/L-g/L range), the presence of an increase in pollutants in the environment that share the same system of movement may also result in the aforementioned effects through additive introductions, such as endocrine disruption, Geno toxicity, adulteration of the marine ecosystem, and the development of resilient infectious microorganisms. Drugs are either released into the environment in their basically unmodified loose form or as their modification products, which can also have adverse repercussions due to the fact that their physiological activities may be greater than those of the parent component (Farah 2023). This is because their physiological activities may be higher than the parent component. Once they are present in aquatic media, the majority of these drugs are unable to flee the environment because of their polar and non-volatile properties. Even pills with exceedingly short half-lives are classified as highly tenacious contaminants in aqueous environments. This is because effluents from WWTPs and DWTPs are constantly being introduced into the system. In addition to the long-term effects on human health induced by consumption of those compounds found in drinking water, knowledge on the ecological affects of medicine and their transformation products inside of the environment is limited and unclear.

This is the case despite the fact that there are concerns regarding the long-term effects on human health caused by consumption of those compounds found in drinking water. This is due to the fact that tests for toxicologic effects are exceedingly complicated and on the rise, and at most they offer stern signs of previously measurably acute effects.

3. Methods of advanced treatment of pharmaceutical wastewater

In recent years, the main emphasis of the scientific research and engineering application has shifted to advanced treatment of pharmaceutical wastewater, which main method is physicochemical technology (Tawalbeh *et al*. 2023). It means that wastewater is treated by physical or chemical methods, like coagulation and sedimentation, flotation, activated carbon adsorption, advanced oxidation processes, membrane separation.

3.1. Coagulation and sedimentation

Coagulation is adding chemical agents to wastewater, dispersing by rapid mixing, then making stable pollutants into unstable and precipitable matters. The mechanism of coagulating is complex. For advanced treatment of pharmaceutical wastewater, the key is how to squeeze and remove bound water round hydrophilic colloid. So the character of flocculent is important, which related to the effect of coagulation. Inorganic metal salts and polymers are frequently used as flocculent. This method can remove SS, chromaticity and toxic organic matter (Bilal *et al*. 2022). Meanwhile, it can improve the biodegradability of pharmaceutical wastewater

3.2. Flotation except for sedimentation

Flotation can also remove suspended solids of secondary effluent.The technology characteristic is producing a large number of tiny bubbles by injecting air into wastewater, forming floating floc with smaller density than wastewater. And it can float to the surface of wastewater to separate.

3.3. Activated carbon adsorption

Activated carbon, as a kind of adsorbent, has many advantages. It has large specific surface area, multilevel pore structure, high adsorption capacity and stable chemical property. Therefore, it is widely used as adsorbent or catalyst carrier to remove pollutants (Peng *et al*. 2018; Azhar *et al*. 2016). In industrial effluents treatment, activated carbon is used for effluent, which is toxic and hard to achieve discharge standard. It is an important method of advanced treatment of pharmaceutical wastewater as well. Activated carbon adsorption can be classified as physical adsorption and chemical adsorption. Physical adsorption is reversible, and no selectivity to adsorbate. When activated carbon saturated by adsorbates, it is easy to desorb. To the contrary, chemical adsorption adsorbs only one or several specific adsorbates, which is irreversible and hard to desorption. For cyclic utilization, saturation of activated carbon restores its adsorption property by regeneration. This method is widely used for advanced treatment, because it can be recycled, its better treatment effect and wide suitability. But there are some disadvantages, such as high costs relatively, low efficiency of regeneration and complex operation, which limit application

3.4. Advanced oxidation processes

Advanced oxidation processes (AOPs), which can oxidize pollutants by forming free radicals. Those kinds of pollutants cannot be degraded by common oxidizing agent. There are many kinds of AOPs, such as wet air oxidation, supercritical water oxidation, Fenton reagent, photocatalytic oxidation, ultrasound oxidation, electrochemical oxidation and ozonation.

3.4.1. Wet air oxidation(WAO)

WAO has been put forward by F. J. Zimmer Mann in 1958, which was used for papermaking black liquid treatment. By using of air or oxygen as the oxidant, this method decomposes organic matter into inorganic or small molecules at high temperature (150-350 ℃) and high pressure (0.5-20 Mpa). WAO is generally used in pretreatment of wastewater advanced treatment. This method has wide range of applications, high efficiency of COD removal, which can even reaches more than 90 % under appropriate conditions, low energy consumption, less secondary pollution, and it is easy management.

3.4.2. Supercritical water oxidation(SCWO)

SCWO is chemical reaction between dissolved oxygen and organic pollutants in supercritical water. Organic matter, air, and supercritical water were complete mixed at 24 Mpa pressure and 400 ℃ temperature, becoming homogeneous phase. Under these conditions, organic compounds spontaneously initiate the oxidation reaction. With the increase of the reaction temperature, 99.9 % or more of the organic matter is rapidly oxidized into simple non-toxic small molecules in a period of time, achieving the purpose of removing pollutants. SCWO has high oxidation efficiency, will not cause secondary pollution, organic can be oxidized completely. However, this method has some shortcomings, such as it requires high operating conditions and high cost.

3.5. Membrane separation

Under certain driving force across the membrane, a component in water selectively permeates it by using permselective membrane separating media, which is called membrane separation. In this way, achieve the separation, purification, concentration of the target substance from the mixture. There are several membrane separation techniques in wastewater treatment. Such as microfiltration, ultrafiltration, reverse osmosis and electrodialysis.

3.5.1. Microfiltration (MF)

MF is based on static pressure as the driving force, and the separation process is performed by the action of the sieve separation of the membrane, which principle is similar to traditional filtration. The slight difference is that the pore size of MF is smaller. This method can effectively remove SS and microorganisms in wastewater.

3.5.2. Ultrafiltration (UF)

UF driving force is the pressure difference between the membranes on both sides, the filter medium is the ultrafiltration membrane. Under certain pressure, when water passes through the membrane surface, water, inorganic salts and small molecules penetrate, other macromolecules are trapped. This method is mainly used for the removal of macromolecules and colloids in wastewater. In the application of this method, it should be ensured that the membrane has adequate membrane flux and is easily disassembled, replaced, cleaned.

3.5.3. Reverse osmosis (RO)

There are two categories of RO membrane, cellulose ester and aromatic polyamide. Its component form includes tube, plate and frame, roll and hollow fiber type. RO process can remove a wide range of impurities, dissolved inorganic salts and a variety of organic matter. Meanwhile, it has a high efficiency of salt removal and water reuse rate. However, this method requires a high pretreatment of the feed water. With the development of RO, nanofiltration (NF) has been proposed as a new method.

3.5.4. Electrodialysis (ED)

ED is a combination of electrolytic and dialysis diffusion process. Under the action of the DC electric field, anions and cations of the dissolved salts in the wastewater are moved to the anode and the cathode respectively. In this way, the concentration of anions and cations in the intermediate compartment is gradually reduced, and the separation and recovery are achieved. This method has many advantages, such as less energy and pharmaceutical consumption, less environmental pollution, easy to operate and automate. But it can only remove the salt in water, and desalination efficiency is lower than RO.

3.6. Biological treatment

In theory, after secondary treatment, pharmaceutical wastewater should not be treated by biological methods, because of its poor biodegradability. However, we cannot ignore the advantages of biological treatment, such as low cost, stable treatment effect. It can be used as a way of pretreatment in advanced treatment.

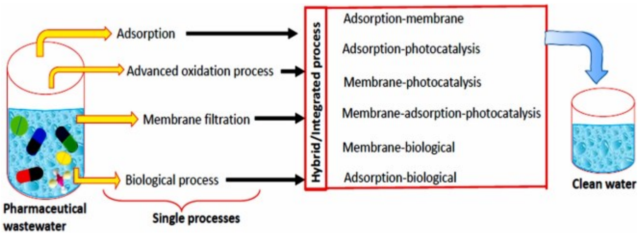

Figure 1. Different technologies used for treating pharmaceutical wastewater

4. Adsorptive process for pharmaceutical removal

Adsorptive approaches are recognized by humans on the grounds of antiquity. Early Egyptians, Greeks, Sumerians, Phoenicians, and Romans used sorbent substances including earthen, grit, and timber coal for the removal of salt from water, explanation of fats and oil, inaddition to the refinement of water for medical purposes. Sorption infers the transfer and gathering of sorptive molecules from the liquid section to the intersecting surface and may include physical and/or chemical interactions (Fakhri *et al.* 2023). While physical sorption instigates from inter particle thrusts, together with van der Waals, chemical adsorption indicates the chemical exchanges with the related switch of negative particles among the adsorbent and the adsorbate. Numerous elements decide the system competence, along with homes of the adsorbent, absorptive, and solvent, in addition to the pH, temperature, along with mixing speed during operational situations. The chemical properties, apparent net rate (additionally dependent on solute), and the permeable assembly of the sorbent fabric influence the sorption equipoise. For better strength in the separation methods, the fabric ought to be present on hand inner extent to the composites to be eliminated and exceptional technical houses. A lotof programs correspondingly involve the sorbent being renewed in a competent manner, without causing additional impairment to technical and sorptive homes. In the case of the sorptive and solvent, the sorption relies upon their assembly, contacts (mutual and with the stable floor), and the variances between physicochemical properties, such as their ability to dissolve, molar weight, and hydrophobicity.

5. Adsorbents for pharmaceutical compounds with a differential cost

5.1. Clays

Due to their layer shape, which is made up of tetrahedral and octahedral sheets in either a 1:1 or 2:1 ratio, clays are naturally occurring phyllosilicates. While 2:1 type clay minerals include propylite, talc, smectite, vermiculite, mica, chlorite, and sepiolite, 1:1 type clay minerals include kaolinite and serpentine. Because of their ion exchange capability, mechanical and chemical stability, and altitudinous specific surface area, clays are utilised as adsorbents. Dueto its chemical makeup and pore structure, clay has a great capacity for adsorption. Due to theirlow cost, clay materials are a beneficial choice of adsorbent. As an example, montmorillonite from the smectite group can be up to 20 times less expensive than activated carbon while also being just as effective—if not more so—as activated carbon at removing drugs.

5.2. Bio char

The thermal breakdown of biomass at low temperatures and in the absence of oxygen produces biochar, which has a high affinity and sorption capacity for natural pollutants like prescription medicines. Bio-oil, biochar, and syngas can be produced through this process, also known as pyrolysis. According to Lehmann and Joseph, pyrolysis is characterised by the presence of essential operational parameters such as residence duration and heating rates. Bio char may be produced at temperatures ranging from 300 to 700 degrees Celsius. Laird and co. Divide the thermo chemical age into four categories: gasification, quick pyrolysis, flash pyrolysis, and slow pyrolysis. Flash pyrolysis has the highest yield of bio char, typically producing 60% bio char, whereas slow pyrolysis has a bio char yield of up to 35%. These technologies are advantageous for the environment since they reduce CO_2 emissions and allow the use of by-products as feedstock. In addition, when compared to other typical adsorbents that are more costly, such as activated carbon, which is "activated" by steam or chemical compounds at higher temperatures, it can be a more cost-effective alternative (Gokulan *et al*. 2023; Ravindiran *et al*. 2019). Different materials, such as agricultural wastes and unconventional materials including waste tyres, plants, algae, municipal and industrial strong waste, bones, bioenergy residues, and food waste, may be pyrolyzed to produce biochar. Sludge from sewage and industrial wastewater treatment plants is mixed with organised bio chars.The authors confirmed that they are efficient gatifloxacin adsorbents, enabling for the removal of more than 96% of the drugs from the aqueous segment (Ravindiran *et al*. 2019). Table 4 gives figures for the greatest adsorption capacities from the Langmuir version (> 20 mg/g) achieved for two biochars made from sludge, even though Freundlich equipped all experimental data. For the single- and multi- solute adsorption of different prescription medications, Jung *et al*. employed bio chars made from torrefied loblolly pine chip. The maximal adsorption capacities for bio char pyrolyzed with pure nitrogen (N-bio char) followed the order diclofenac > naproxen > ibuprofen, according to single adsorption statistics.

5.3. Agro-industrial wastes

Agro-commercial waste is produced in significant amounts as a result of commercial activity. To increase the utility of these residues, a novel application as adsorbents has been suggested. Although they typically have lesser adsorption capabilities than activated carbon, their usage on a large scale is more economically advantageous. In addition, they provide environmental advantages over conventional adsorbents due to their regenerative nature and possibility for waste partial reduction. Agro-commercial wastes primarily consist of cellulose, which also makes up the majority of their adsorption properties. Hemicellulose, lignin, lipids, proteins, simple sugars, water, hydrocarbons, and starch are additional important constituents. There are various research addressing the use of agricultural and commercial wastes as adsorbents of pollutants, including pharmaceutical chemicals. Activated carbons and bio chars can be made from agricultural and commercial wastes. The components can be exploited either in their original form or after undergoing either a physical or a chemical transformation. For instance, Portinho *et al*. made use of grape stalk from the Vitisvinifera species in both its raw form and after being treated with phosphoric acid flow, in addition to using it in the form of activated carbon. The Freundlich model provided a good match for the equilibrium data, which points to a multilayer adsorption of diclofenac sodium over Isabel grape bagasse. According to the thermodynamic study, the process is of an exothermic character and is followed by a reduction in randomness at the interface between the solid and the solution. The modified version demonstrated superior capacity for the absorption of caffeine compared to the original form. The adsorption of paracetamol onto wastes generated during the production of wine (grape stalk), yohimbe extraction (yohimbine bark), and tap manufacturing (cork bark) was another topic that Villaescusa and colleagues investigated.

6. Adsorbent replacement

The recovery of adsorbents after usage is a crucial element when taking the financial effectiveness of the industrial adsorption process into consideration. The adsorbate can be desorbed to regenerate the adsorbent, especially in the case of interactions between the adsorbent and adsorbate that are sensitive, like dispersion-van der Waals. Thermal reactivation is frequently used to regenerate pricey adsorbents like granular activated carbon (GAC), however this is not always the best option because it can result in weight losses and changes to the porous structure, both of which can affect adsorption capacity. Due to this, PACs (Powdered Activated Carbon) are frequently used in batch adsorption as one-way adsorbents and eliminated through cremation or landfilling (Ganthavee *et al*. 2023).

Only a few research have looked at the regeneration and repurposing of used non-traditional, inexpensive adsorbents following the removal of medicines, but these materials are also not frequently recycled. Several research on the regeneration of used ACs following drug uptake employing a variety of methodologies, including thermal regeneration, chemical desorption, and catalytic ozonation, can be found in the literature. However, getting new, inexpensive adsorbents, including those made from waste products, may be more cost-effective than recycling the used ones when taking the total adsorption system's economic performance into account (Ravindiran *et al*. 2023). In order to lessen the environmental risk associated with the disposal of used adsorbents, ball milling has also been suggested as an efficient approach to decompose adsorbed contaminants.

7. Environmental and economic benefit evaluations

According to the literature described above, biochar offers a tremendous deal of promise for application in cleaning up industrial pollutants. To increase the economic viability of the process, certain parameters, such as the type of biochar, the accessibility of the feedstock locally, the temperature of the pyrolysis, the accessibility of industrial-scale reactors, the regeneration technique, and the lifespan, must be optimised before large-scale implementation. According to a recent statement the International Biochar Initiative and the European Biochar Certificate are currently defining standards for the enhancement of biochar. In general, over the past several years, there has been extensive discussion on the use of biochars for the removal of pollutants. In order for biochar to be used on a broad scale for the removal of pollutants, further research will need to be done in the future to examine the wide price fluctuations in the biochar market and their financial benefits. By lowering GHG emissions, biochar has the potential to be an effective technique for decreasing the effects of climate change (Davoodi *et al*. 2019). Studies have shown that using biochar-modified soil can reduce CO_2 and N2O emissions. It can be used to immobilise carbon from the active cycle, converting it to an inactive cycle (Thakur *et al*. 2022). In order to mitigate the effects of climate change and enhance environmentally friendly sustainable farming practises, biochar can be a valuable tool. This technology's practical application encompasses four complimentary and frequently synergistic social and economic uses, namely soil enhancement (to increase productivity and decrease pollutant toxicity), climate change mitigation, energy production, and waste management. In order to explore the biochar's potential for carbon sequestration in the soil, it is necessary to research the interaction between the preparation process and conditions, the feedstock, the microstructure, and the biochar's stability over time. Several techniques, such as infrared spectroscopy, biomarkers, nuclear magnetic resonance, and others researchers can be utilised in order to evaluate the mineralization of biochar throughout the course of time (Pal *et al*. 2022; Mangla *et al*. 2022; Elamin *et al*. 2021). In addition, it is essential to take into consideration and extensively examine any potential ecotoxicological implications brought on by the mineralization of the biochar itself. These impacts might have a significant negative effect on the environment.

8. Applications of adsorption processes in real life

The majority of papers on the adsorption of medicines utilising different low-cost adsorbents are currently just batch (kinetics and equilibrium) studies, according to literature reviews. Only regeneration research and bonded mattress studies—which are crucial for mimicking or forecasting dynamic performance—have received much attention. Additionally, for the purpose of investigating adsorptive capacity and absorption mechanisms, the vast bulk of studies is limited to laboratory size. Adsorptive techniques for the removal of pharmaceuticals have practical applications, particularly in the business sector, although little is known about them. In order to achieve full-scale design, this necessitates switching from laboratory adsorption column settings to pilot plant operations in small diameter fixed beds. Numerous methods, such as computer simulation, rapid-small scale column tests, and response surface methodology, can be used to carry out scale-up research. According to researcher (Lima *et al*. 2021), full-scale field testing conducted under various continuous-flow circumstances are the most effective method for predicting the long-term performance of adsorption systems. In order to optimise crucial operating parameters including breakthrough curves, conduct and mass transfer zones, pilot scale column studies should be carried out. Furthermore, to prevent any potential issues like hydrodynamic barriers or column fouling, adsorbent parameters like form, size, and density should be taken into consideration. While downhill flows are frequently used, solution feeding should also be taken into account when designing columns since upward flows can enable more uniform pollutant distribution, minimise pressure gradients, and lower the risk of fouling adsorbent. In addition, it is possible to employ adsorbent with smaller particle sizes, use less of it overall, and obtain higher adsorption rates when the solution is fed at the bottom of the column. To the authors' knowledge, there is no work that assesses the removal of pharmaceuticals at pilot scale utilising unconventional, affordable adsorbents (Wilkinson *et al*. 2022). Even less study has been done on pharmacological adsorption in its entirety. The fact that an adsorption system is an unsteady-state process presents the fundamental difficulty in scaling it up. As a result, even in pilot size investigations, it is still uncommon to use adsorbents for pharmaceutical removal in actual adsorption systems for water purification. Nevertheless, the rise in laboratory-scale research on this subject over the past five years suggests that testing of unconventional, inexpensive adsorbents in large-scale applications is just a question of time (Pal *et al*. 2022; Cusioli and Quesada 2020; Spessato *et al*. 2021). It is difficult to successfully transfer adsorption techniques employing low-cost adsorbents from lab-scale to commercial products for a number of reasons, including the following: (i) the sorption properties of low-cost adsorbents can be highly dependent on their substrate; (ii) the demand at an industrial level can limit the availability of the resource for industrial users; (iii) the physical and chemical pre-treatment methods that could enhance the sorption capacity of the materials may not be cost-effective at a large scale or environmentally friendly; and (iv) the efficacy of the treatment is dependent on the type of low-cost adsorbent being used, (v) the performance of adsorption is related to process variables (such as temperature, pH, concentration of adsorbate and adsorbent, ionic electricity, contact time, particle size of adsorbent, etc.); (vi) discrepancies in data presentation (experimental conditions, batch or continuous systems) prevent comparison of different materials for adsorption; (vii) limited data is available on the adsorption of pollutants from mixtures and real industrial effluent.

9. Future outlooks

Due to its twin advantages of water treatment and waste control, adsorption for water treatment utilising inexpensive adsorbents is a promising field. Interest in the field of water treatment has been sparked by research into the conversion of various waste materials into inexpensive adsorbents. Therefore, it is expected that low-cost adsorbents will be used in more water treatment applications in the near future. Another draw for the promising future of low-cost adsorbents is their ubiquitous and affordable character. It is anticipated that both industrialized and developing nations would use them even more frequently (Hernandez-Maldonado *et al*. 2022; Hethnawi *et al*. 2020; Mahmoud *et al*. 2020). Despite this, there are certain difficulties with the effective application of inexpensive adsorbents in the near future. The handling of used adsorbent is a significant issue that has not yet been fully resolved. Regarding the problem of managing removed pollution, there is no report available, but in our opinion, the removed pollution should be recycled or deeply contaminated. The pollutants that have been removed must be stored in metal containers and handled similarly to nuclear waste. There is a pressing need to create adsorbents that can function at pH 7.0, consistent temperature, and short contact times because some adsorbents are ineffective when used in their natural environment. At mg/mL concentrations, inexpensive adsorbents are often effective at removing pollutants (Bollinger *et al*. 2022). In order to reduce potential dangers to the environment, these adsorbents should be utilised responsibly. Even though adsorption in batch mode is covered in a number of studies, further study is needed on water treatment at the pilot and industrial scales (Lima *et al*. 2020). This necessitates the creation of efficient columns that can treat water on a vast scale. Additionally, there is a need to create low-cost adsorbents for water treatment that are more effective, selective, affordable, and environmentally friendly. In conclusion, it is necessary to change inexpensive adsorbents to make them more cost-effective, effective, and environmentally acceptable.

10. Conclusions

This assessment article began by briefly addressing environmental pharmaceutical contamination and showing that, despite being a reality that can result from industrial and human activities, it is still not fully understood how pharmaceuticals in water could affect human health over the long term and how that might affect the environment. In addition to traditional methods (such as coagulation, flotation, and lime softening) and other cutting-edge technologies (such as membrane filtration, ozonation, and advanced oxidation processes), the focus of this review is on adsorption technology for the removal of pharmaceuticals from water and wastewaters. The use of non-traditional, inexpensive adsorbents in place of commercial activated carbons has recently become more popular. After careful analysis, it has been determined that materials like clays, biochar, chitosans, agro-commercial wastes, and metallic-organic frameworks (MOFs) can be used to effectively adsorb a variety of prescription drugs. A variety of variables, including pH, temperature, and the adsorbent's affinity for the pollutant, are known to affect each type of adsorbent's efficacy. The pseudo- second-order model accurately describes the majority of the investigated adsorption processes, which suggests that chemisorption is probably taking place. The most frequently investigated technique is batch adsorption; however, studies on continuous adsorption and full- or pilot- scale pharmaceutical adsorption utilising these materials are lacking. Furthermore, it is challenging to do an exhaustive comparison of these materials because little is known about their cost-benefit and regeneration potential. Therefore, additional research is required, including an analysis of regeneration efficacy and cost benefit, to determine the economic and environmental significance of adsorption methods. In conclusion, there is currently a dearth of information regarding the actual use of unconventional low-cost adsorbents for pharmaceutical removal despite the rising research in this area. Additional research is needed to investigate topics including constant-bed adsorption, multi-component adsorption, treatment of actual wastewaters, continuous adsorption, and adsorption regeneration. Furthermore, as the majority of existing studies use synthetic or mono-component solutions at concentrations higher than those generally found in the environment, studies that model real systems should be carried out. Therefore, additional study is necessary to allow for industrial application.

References

Azhar M.R., Abid H.R., Periasamy V., Sun H., Tade M.O. and Wang S. (2017). Adsorptive removal of antibiotic sulfonamide by UiO-66 and ZIF-67 for wastewater treatment. *Journal of Colloid and Interface Science*, **500**, 88–95.

Azhar M.R., Abid H.R., Sun H., Periasamy V., Tadé M.O. and Wang S. (2016). Excellent performance of copper-based metal organic framework in adsorptive removal of toxic sulfonamide antibiotics from wastewater. *Journal of Colloid and Interface Science*. **478**, 344–352.

Bilal M., Ihsanullah I., Shah M.U.H., Reddy A. V.B. and Aminabhavi T.M. (2022). Recent advances in the removal of dyes from wastewater using low-cost adsorbents. *Journal of Environmental Management*, **321**, 115981. https://doi.org/10.1016/j.jenvman.2022.115981.

Bollinger J.-C., Tran H.N. and Lima E.C. (2021). Comments on Removal of methylene blue dye using nano zerovalent iron, nanoclay and iron impregnated nanoclay – a comparative study by M. M. Tarekegn, R. M. Balakrishnan, A. M. Hiruy and A. H. Dekebo, RSC Advanced., **11**, 30109, *RSC Advanced*. **12**, 5769–5771, https://doi. org/10.1039/d1ra08212d.

Carballa M., Omil F., Lema J. M., Llompart M.a., Garcıa-Jares C., Rodrıguez I., Gómez M. and Ternes T. (2004). Behavior of pharmaceuticals, cosmetics and hormones in a sewage treatment plant. *Water Resources*, **38**, 2918–2926.

Chang P.-H., Jean J.-S., Jiang W.-T. and Li Z. (2009). Mechanism of tetracycline sorption on rectorite. Colloids Surf. *Physicochemical. Engineering Aspects*, **339**, 94–99.

Cusioli L.F., Quesada H.B., de Brito Portela Castro A.L., Gomes R.G. and Bergamasco R. (2020). Development of a new low-cost adsorbent functionalized with iron nanoparticles for removal of metformin from contaminated water, *Chemosphere* **247**, 125852, https://doi.org/10.1016/j. chemosphere.2020 .125852.

Davoodi S., Dahrazma B., Goudarzi N. and Gorji H.G. (2019). Adsorptive removal of azithromycin from aqueous solutions using raw and saponin-modified nano diatomite, *Water Science and Technology*. **80** 939–949, https://doi.org/10.2166/ wst.2019.337.

De Oliveira T., Guégan R., Thiebault T., Milbeau C.L., Muller F., Teixeira V., Giovanela M. and Boussafir M. (2017). Adsorption of diclofenac onto organoclays: Effects of surfactant and environmental (pH and temperature) conditions. *Journal of Hazardous Materials*, **323**, Part A, 558–566.

Dordio A.V., Miranda S., Prates Ramalho J.P. and Carvalho A.J.P. (2017). Mechanisms of removal of three widespread pharmaceuticals by two clay materials. *Journal of Hazardous Materials*, **323**, 575–583.

Elamin M.R., Abdulkhair B.Y., Algethami F.K. and Khezami L. (2021). Linear and nonlinear investigations for the adsorption of paracetamol and metformin from water on acid-treated clay, *Scientific Reports*. **11** 1–13, https://doi.org/10.1038/s41598- 021-93040-y

Fakhri V., Jafari A., Vahed F.L., Su C. and Pirouzfar V. (2023). Polysaccharides as eco-friendly bio-adsorbents for wastewater remediation: Current state and future perspective. *Journal of Water Process Engineering*, **54**, 103980. https://doi.org/10.1016/j.jwpe.2023.103980.

Farah A. (2023). The use of agro-waste-based adsorbents as sustainable, renewable, and low-cost alternatives for the removal of ibuprofen and carbamazepine from water.

Heliyon, **9**(6), e16449. https://doi.org/10.1016/j.heliyon.2023.e16449.

Ferreira C.I.A., Calisto V., Otero M., Nadais H. and Esteves V.I. (2017). Fixed-bed adsorption of Tricaine Methane sulfonate onto pyrolysis paper mill sludge. *Aquacultural Engineering* **77**, 53–60.

Ganthavee V. and Trzcinski A.P. (2023). Removal of Pharmaceutically Active Compounds from Wastewater Using Adsorption coupled with Electrochemical Oxidation Technology: A Critical Review. *Journal of Industrial and Engineering Chemistry*, **126**, 20–35. https://doi.org/10.1016/j.jiec.2023.06.003.

Genç N., Can Dogan E. and Yurtsever M. (2013). Bentonite for ciprofloxacin removal from aqueous solution. *Water Science and Technology*, **68**, 848–855.

Gkika D.A., Mitropoulos A.C., Kokkinos P., Lambropoulou D.A., Kalavrouziotis I.K., Bikiaris D.N. and Kyzas G.Z. (2023). Modified chitosan adsorbents in pharmaceutical simulated wastewaters: A review of the last updates. *Carbohydrate Polymer Technologies and Applications,* **5**, 100313. https://doi.org/10.1016/j.carpta.2023.100313.

Gokulan R., Avinash A., Prabhu G.G. and Jegan J. (2019). Remediation of remazol dyes by biochar derived from Caulerpa scalpelliformis—An eco-friendly approach, *Journal of Environmental Chemical Engineering*, **7**, (5), https://doi.org/10.1016/j.jece.2019.103297.

Hasan Z., Jeon J. and Jhung S.H. (2012). Adsorptive removal of naproxen and clofibric acid from water using metal-organic frameworks. *Journal of Hazardous Materials*. 209-210, 151–157.

Hernandez-Maldonado A.J., Atkinson J.D., Hashisho Z., Saleh N., Deng Y., Bae S., Frank Xiao F. and Georgi A. (2022). Current and future trends in adsorption for environmental separations, *Journal of Hazardous Materials*. **433**, 128776, https://doi. org/10.1016/j.jhazmat.2022.128776.

Hethnawi A., Alnajjar M., Manasrah A.D., Hassan A., Vitale G., Jeong R. and Nassar N.N. (2020). Metformin removal from water using fixed-bed column of silicaalumina composite, Colloids Surface. *A Physicochemical Engineering Aspects*. **597**, 124814, https://doi.org/10.1016/j.colsurfa.2020.124814.

Khazri H., Ghorbel-Abid I., Kalfat R. and Trabelsi-Ayadi M. (2017). Removal of ibuprofen, naproxen and carbamazepine in aqueous solution onto natural clay: equilibrium, kinetics, and thermodynamic study. *Applied Water Science*, **7**, 3031–3040.

Khoshraftar Z., Masoumi H. and Ghaemi A. (2023). On the performance of perlite as a mineral adsorbent for heavy metals ions and dye removal from industrial wastewater: A review of the state of the art. *Case Studies in Chemical and Environmental Engineering*, **8**, 100385. https://doi.org/10.1016/j.cscee.2023.100385.

Kumar N., Pandey A., Rosy and Sharma Y.C. (2023). A review on sustainable mesoporous activated carbon as adsorbent for efficient removal of hazardous dyes from industrial wastewater. *Journal of Water Process Engineering*, **54**, 104054. https://doi.org/10.1016/j.jwpe.2023.104054.

Kwon J.-W. and Rodriguez J.M. (2014). Occurrence and Removal of Selected Pharmaceuticals and Personal Care Products in Three Wastewater-Treatment Plants. *Archives of Environmental Contamination and Toxicology*, **66**, 538–548.

Lima E.C., Dehghani M.H., Guleria A., Sher F., Karri R.R., Dotto G.L. and Tran H.N. (2021). Adsorption: fundamental aspects and applications of adsorption for effluent treatment, *Green Technology*. *Defluoridation, Water*, https://doi.org/10.1016/c2020-0-02666-6.

Lima E.C., Sher F., Guleria A., Saeb M.R., Anastopoulos I. and Tran H.N. (2020). A. HosseiniBandegharaei, Is one performing the treatment data of adsorption kinetics correctly, *Journal of Environmental Chemical Engineering*, 104813, https://doi.org/10.1016/j. jece.2020.104813.

Liu N., Wang M.-x., Liu M.-m., Liu F., Weng L., Koopal L.K. and Tan W.-f. (2012). Sorption of tetracycline on organo-montmorillonites. J. Hazard. Mater, 225–226, 28–35.

Mahmoud M.E., El-Ghanam A.M., Saad S.R. and Mohamed R.H.A. (2020). Promoted removal of metformin hydrochloride anti-diabetic drug from water by fabricated and modified nanobiochar from artichoke leaves, Sustain. Chem. Pharm. 18, 100336, https://doi.org/10.1016/j.scp.2020.100336.

Mangla D., Annu A. and Sharma S. (2022). Ikram, Critical review on adsorptive removal of antibiotics: present situation, challenges and future perspective, *Journal of Hazardous Materials*. **425**, 127946, https://doi.org/10.1016/j.jhazmat.2021.127946.

Ndoun M.C., Elliott H.A., Preisendanz H.E., Williams C.F., Knopf A. and Watson J.E. (2021). Adsorption of pharmaceuticals from aqueous solutions using biochar derived from cotton gin waste and guayule bagasse, *Biochar* **3,** 89–104, https://doi.org/10.1007/s42773-020-00070-2.

Oosterhuis M. and Sacher T.L. (2013). Prediction of concentration levels of metformin and other high consumption pharmaceuticals in wastewater and regional surface water based on sales data. *Science Total Environment*, **442**, 380-388.

Pal S., Ahamed Z. and Pal P. (2022). Removal of antibiotics and pharmaceutically active compounds from water environment: experiments towards industrial scale up, *Separation and Purification Technology*. **295**, 121249, https://doi.org/10.1016/j. seppur.2022.121249.

Pal S., Ahamed Z. and Pal P. (2022). Removal of antibiotics and pharmaceutically active compounds from water environment: experiments towards industrial scale up, Separation and Purification Technology **295**, 121249, https://doi.org/10.1016/j. seppur.2022.121249.

Peng Y., Zhang Y., Huang H. and Zhong C. (2018). Flexibility induced high-performance MOFE based adsorbent for nitroimidazole antibiotics capture. *Chemical Engineering Journal*, **333**, 678–685.

Ravindiran G., Ganapathy G., Jegan J. and Alagumalai A. (2019). A Critical Insight into Biomass Derived Biosorbent for Bioremediation of Dyes. *ChemistrySelect*, ***4***(33), 9762–9775. https://doi.org/10.1002/slct.201902127.

Ravindiran G., Jeyaraju R.M., Nandipati G., Seerangagounder S., Al-Zaqri N., Boshaala A. and Hayder G. (2023). Prevention of groundwater contamination from the pollutants released from dyeing industries using biochar produced from palm shell, *Urban Climate*, **49**, https://doi.org/10.1016/j.uclim.2023.101515.

Ravindiran G., Prabhu G.G. and Jegan J.A. (2019). novel sorbent *Ulva lactuca* -derived biochar for remediation of Remazol Brilliant Orange 3R in packed column. *Water Environment Research*, ***91***(7), 642–649. https://doi.org/10.1002/wer.1092.

Reynaud F., Tsapis N., Deyme M., Vasconcelos T.G., Gueutin C., Guterres S.S., Pohlmann A.R. and Fattal E. (2011). Spray-dried chitosan-metal microparticles for ciprofloxacin adsorption: Kinetic and equilibrium studies. *Soft Matter*, **7**, 7304–7312.

Reynel-Avila H.E., Mendoza-Castillo D.I., Bonilla-Petriciolet A. and Silvestre-Albero J. (2015). Assessment of naproxen adsorption on bone char in aqueous solutions using batch and fixed- bed processes. *Journal of Molecular Liquids*, **209**, 187–195.

Roca Jalil M.E., Baschini M. and Sapag K. (2015). Influence of pH and antibiotic solubility on the removal of ciprofloxacin from aqueous media using montmorillonite. *Applied Clay Science*, **114**, 69–76.

Salem N.A. and Yakoot S.M. (2016). Non-steroidal Anti-inflammatory Drug, Ibuprofen Adsorption Using Rice Straw Based Biochar. *International Journal of Pharmacology*, **12**, 729–736.

Salihi E.Ç. and Mahramanlıoğlu M. (2014). Equilibrium and kinetic adsorption of drugs on bentonite: Presence of surface-active agents effect. *Applied Clay Science*, **101**, 381–389.

Spessato L., Duarte V.A., Viero P., Zanella H., Fonseca J.M., Arroyo P.A. and Almeida V.C. (2021). Optimization of Sibipiruna activated carbon preparation by simplexcentroid mixture design for simultaneous adsorption of rhodamine B and metformin, *Journal of Hazardous Materials*, **411**, 125166, https://doi.org/10.1016/j. jhazmat.2021.125166.

Sun K., Shi Y., Chen H., Wang X. and Li Z. (2017). Extending surfactant-modified 2:1 clay minerals for the uptake and removal of diclofenac from water. *Journal of Hazardous Materials*, **323**, 567–574.

Tatarchuk T., Soltys L. and Macyk W. (2023). Magnetic adsorbents for removal of pharmaceuticals: A review of adsorption properties. *Journal of Molecular Liquids*, **384**, 122174. https://doi.org/10.1016/j.molliq.2023.122174.

Tawalbeh M., Mohammed S., Al-Othman A., Yusuf M., Mofijur M. and Kamyab H. (2023). MXenes and MXene-based materials for removal of pharmaceutical compounds from wastewater: Critical review. *Environmental Research*, **228**, 115919. https://doi.org/10.1016/j.envres.2023.115919.

Thakur A.K., Singh R.C., Pullela R.T. and Pundir V. (2022). Green adsorbents for the removal of heavy metals from Wastewater: A review. *Materials Today: Proceedings*, **57**, 1468–1472. https://doi.org/10.1016/j.matpr.2021.11.373.

Topare N.S. and Wadgaonkar V.S. (2023). A review on application of low-cost adsorbents for heavy metals removal from wastewater. *Materials Today: Proceedings*, **77**, 8–18. https://doi.org/10.1016/j.matpr.2022.08.450.

Villaescusa I., Fiol N., Poch J., Bianchi A. and Bazzicalupi C. (2018). Mechanism of paracetamol removal by vegetable wastes: The contribution of π–π interactions, hydrogen bonding and hydrophobic effect. *Desalination*, **270**, 135–142.

Vinayagam V., Murugan S., Kumaresan R., Narayanan M., Sillanpää M., Vo D.V.N., Kushwaha O.S., Jenis P., Potdar P. and Gadiya S. (2022). Sustainable adsorbents for the removal of pharmaceuticals from wastewater: A review. *Chemosphere*, ***300***, 134597. https://doi.org/10.1016/j.chemosphere.2022.134597.

Wilkinson J.L., Boxall A.B.A., Kolpin D.W., Leung K.M.Y., Lai R.W.S., Wong D., Ntchantcho R., Pizarro J., Mart J., Echeverr S. and Garric J. (2022). A. Chaumot, P. Gibba, I. Kunchulia, S. Seidensticker, G. Lyberatos, J.M. Morales-salda, H. Kang, Pharmaceutical pollution of the world' rivers, PNAS 119, 1–10, https://doi.org/10.1073/pnas.2113947119//DCSupplemental.Published.

Zha S. x., Zhou Y., Jin, X. and Chen Z. (2013). The removal of amoxicillin from wastewater using organ bentonite. *Journal of Environmental Management*, **129**, 569–576.

Zhang Y.-L., Zhang J., Dai C.-M., Zhou X.-F. and Liu S.-G. (2013). Sorption of carbamazepine from water by magnetic molecularly imprinted polymers based on chitosan-Fe3O4. *Carbohydrate polymers*, **97**, 809–816.