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REVIEW

Improving Pharmaceuticals Removal at Wastewater Treatment Plants Using Biochar: A Review

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Abstract

The presence of pharmaceuticals within the environment poses serious threat to the health of humans and animals. Owing to the inability of existing wastewater treatment methods to completely remove pharmaceuticals when wastewater is treated at wastewater treatment plants, their effluent have been recognized as one of the main sources of pharmaceuticals into the environment. The negative effect of some of these pharmaceuticals in the environment has resulted in rising concern on how to improve wastewater treatment methods at wastewater treatment plants. Recently, adsorption process has been considered as an efficient method to complement the existing methods of wastewater treatment. This is because of the high affinity of suitable adsorbents for pharmaceuticals within wastewater. Nonetheless, the high price of prevalent adsorbent like activated carbon has been a major limitation. Biochar that possesses similar properties to activated carbon has recently been reported by different literature to be efficient in the removal of pharmaceuticals from wastewater and aqueous solution. Because of this, several literature were studied on pharmaceuticals adsorption with the use of biochar and a summary of our findings are presented in this review. In addition, a recent report in Estonia has shown considerable pharmaceuticals concentration above the limit of detection in the effluent streams of wastewater treatment plants. Based on the rate of human consumption data, The authors focused on three pharmaceuticals (1) Metformin, (2) Ibuprofen, and (3) Diclofenac which are part of the readily detected in wastewater treatment effluents in Estonia. In response to their inefficient removal, this paper offers the possibility of using adsorption, specifically with the use of biochar as an economical adsorbent for improving their removal. The findings in this review range from wastewater treatment methods, biochar production and characterization methods to the mechanisms involved in using biochar for the removal of pharmaceuticals. Lastly, the major challenges related with this possibility are highlighted, while recommendations for future research are also highlighted to hasten the implementation of adsorption process using biochar material as the adsorbent for improving pharmaceuticals removal from wastewater.

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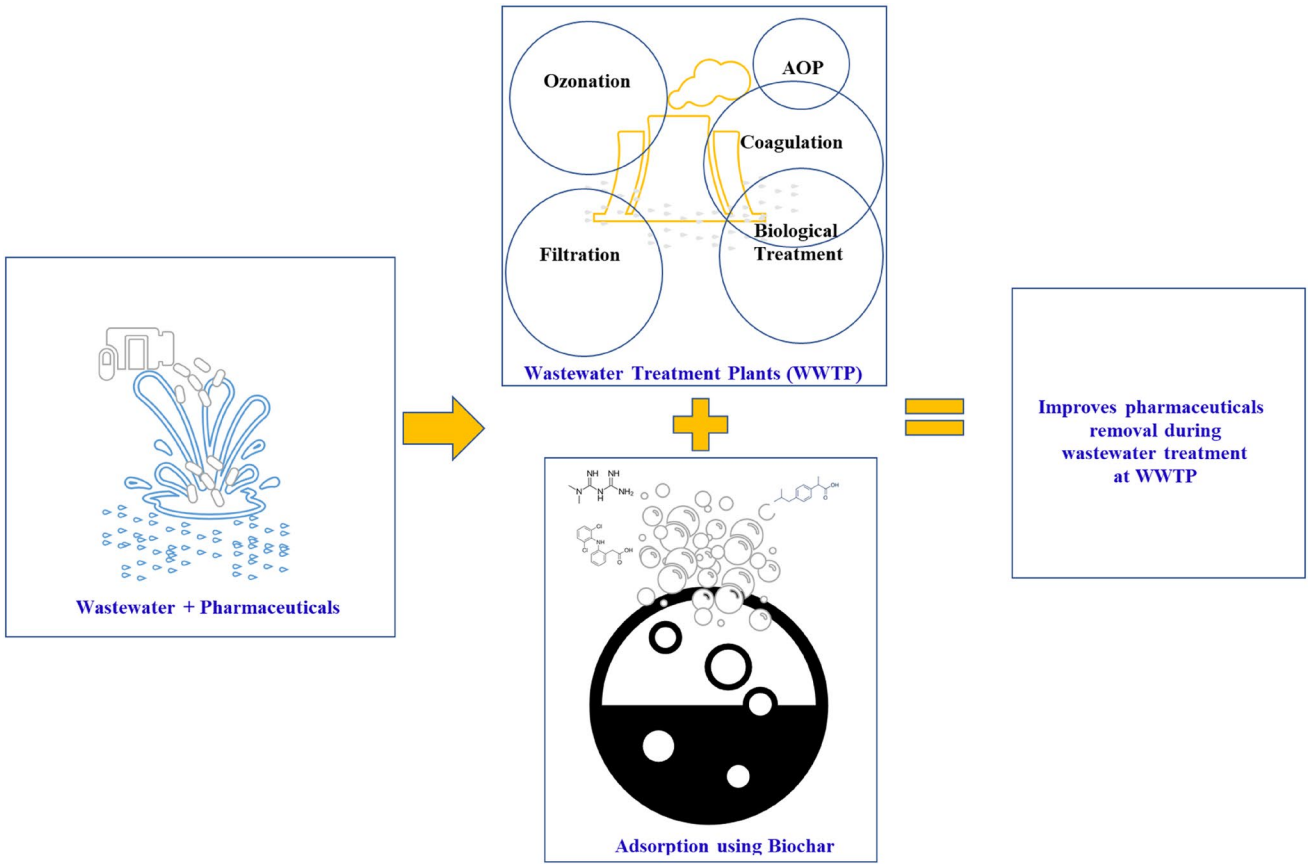
Graphical Abstract



Highlights

- **The presence of pharmaceuticals in the environment poses serious threat to the lives of human and animals**
- **The effluents of wastewater treatment plants have been identified as a major source of pharmaceuticals within the environment**
- **Concentrations of Diclofenac, Ibuprofen, and Metformin remain high in the effluents of wastewater treatment plants in Estonia**
- **Adsorption has the capacity to assist in improving pharmaceuticals removal during wastewater treatment at wastewater treatment plants**
- **Biochar possesses the desired features to replace high-cost adsorbents during an adsorption process**

Keywords Pharmaceuticals · Wastewater · Wastewater treatment plant · Adsorption · Biochar

Statement of Novelty

There have been various reports on the use of biochar for assisting in the adsorption of Phcs, however, reports on the utilization of biochar for improving Phcs removal during the treatment of wastewater at WWTP are limited. This synopsis aims to bridge that gap by specifically presenting the possibility of improving the removal of Phcs during wastewater treatment using biochar, with emphasis on diclofenac (DF), ibuprofen (IB), and metformin (MF) removal. Furthermore, Estonia is chosen as a case study owing to the limited research on biochar and its application within this region.

Introduction

Pharmaceuticals (Phcs) and Their Impact on the Environment

Pharmaceuticals (Phcs) are modified biologically active substance used to cure or prevent ailments in animals and humans [1, 2]. It includes analgesics, antibacterials, as well as antiepileptics [1, 3, 4]. Reportedly, the use of Phcs in the year 2020 was about 4.5 trillion dosages [5] and could increase in the future [6]. However, concerns about their presence in the environment and threat to aquatic lives have been raised by several authors [6–11]. The threats they pose to the environment include (1) The development of pathogens that are more resistant to treatment and threaten the health of lives within the environment [12–14] (2) Their bioaccumulation is poisonous to the environment [15] and (3) Their presence impacts the food web within the aquatic eco-system in an unfavorable manner [16, 17]. Reportedly, two of the major sources of Phcs within the environment is from the effluent of wastewater treatment plants (WWTP) [14, 18, 19] and untreated water [17]. Owing to this, the conventional methods of wastewater treatment have been faulted for their inability to completely eradicate Phcs, leading to their release into nearby rivers and coastal areas [20]. Other non-point sources of Phcs within the environment are connected to the inappropriate disposal of expired drugs, human feces, agriculture, and veterinary practices, leaching into surface and ground waters [10, 19, 21]. Some Phcs like paracetamol, ciprofloxacin, sulfamethoxazole, and caffeine are readily biodegradable and can be significantly removed during treatment [21], while some degrade slowly [22]. Furthermore, some could be stable in the environment and varying concentrations have been uncovered within the range of ngL^{-1} to μgL^{-1} in the effluents of WWTP, contaminating water bodies around the world [6, 19], [23–25].

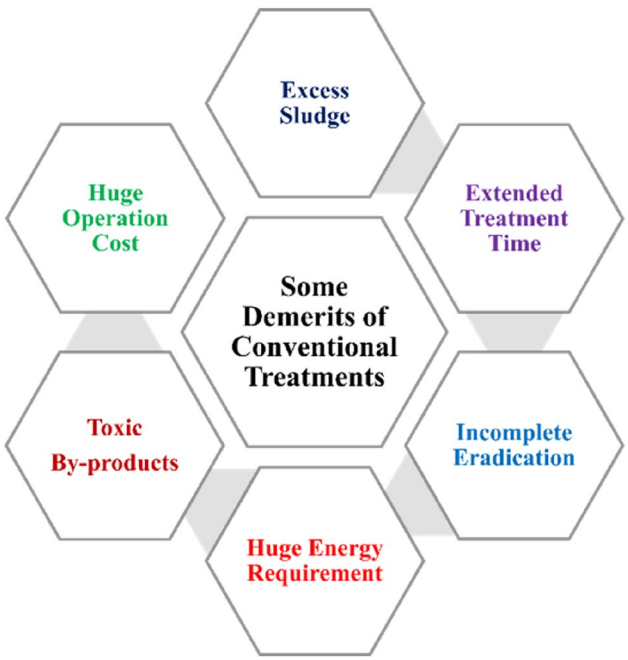


Fig. 1 Some demerits of conventional methods of wastewater treatment

General Wastewater Treatment Methods

In the past, different conventional methods have been considered for eliminating Phcs from wastewater, they include coagulation [26], advanced oxidative process (AOP) [27], biological method [28], ozonation and filtration [17, 27]. Coagulation is known for treating wastewater with the use of chemicals known as coagulants that bind pollutants until a huge mass that can be separated via settling is formed [29]. Common coagulants used are salts of aluminum and iron, although there are issues associated with the disposal of sludge generated from this method [29]. AOP also utilizes chemicals to remove inorganic or organic pollutants from wastewater, resulting in the formation –OH radicals [29]. AOP's potential to breakdown complex compounds present it as an efficient technique for removing Phcs from wastewater [30]. However, its enormous operation cost [27], huge energy consumption [31], and the need for a spacious set-up increases its process cost and hinders its use globally [27]. Biological technique utilizes the activity of microbes in the treatment of wastewater, it is an eco-friendly technique, nonetheless, it takes time and there are concerns on how to manage the sludge generated when it is used [29], besides, not all Phcs are removed using this technique [14]. Ozonation is beneficial in that it produces no sludge, nonetheless, it produces other by-products in WWTP effluents [29]. Reportedly, these by-products could be more toxic when compared to the initial Phcs contaminants, however, it remains a viable wastewater treatment option [31]. Filtration utilizes a membrane (Constro [32] attached to leaky support and is useful for taking out dissolved contaminants during the treatment of wastewater [29]. Nonetheless, for membrane-filtration, there could be a need for frequent membrane replacement [29], and the problem of fouling [28]. In themselves, each of the conventional techniques may not be sufficient for fully removing Phcs from wastewater [12, 14, 33], and a summary of their demerits is presented in Fig. 1. Hence, with their shortcomings, it is expedient to complement these methods to improve the percentage of Phcs being removed during treatment [34]. Of recent, the interest in the use of adsorption for improving the removal of Phcs from wastewater [17, 35, 36] is rising [37], and this is because it

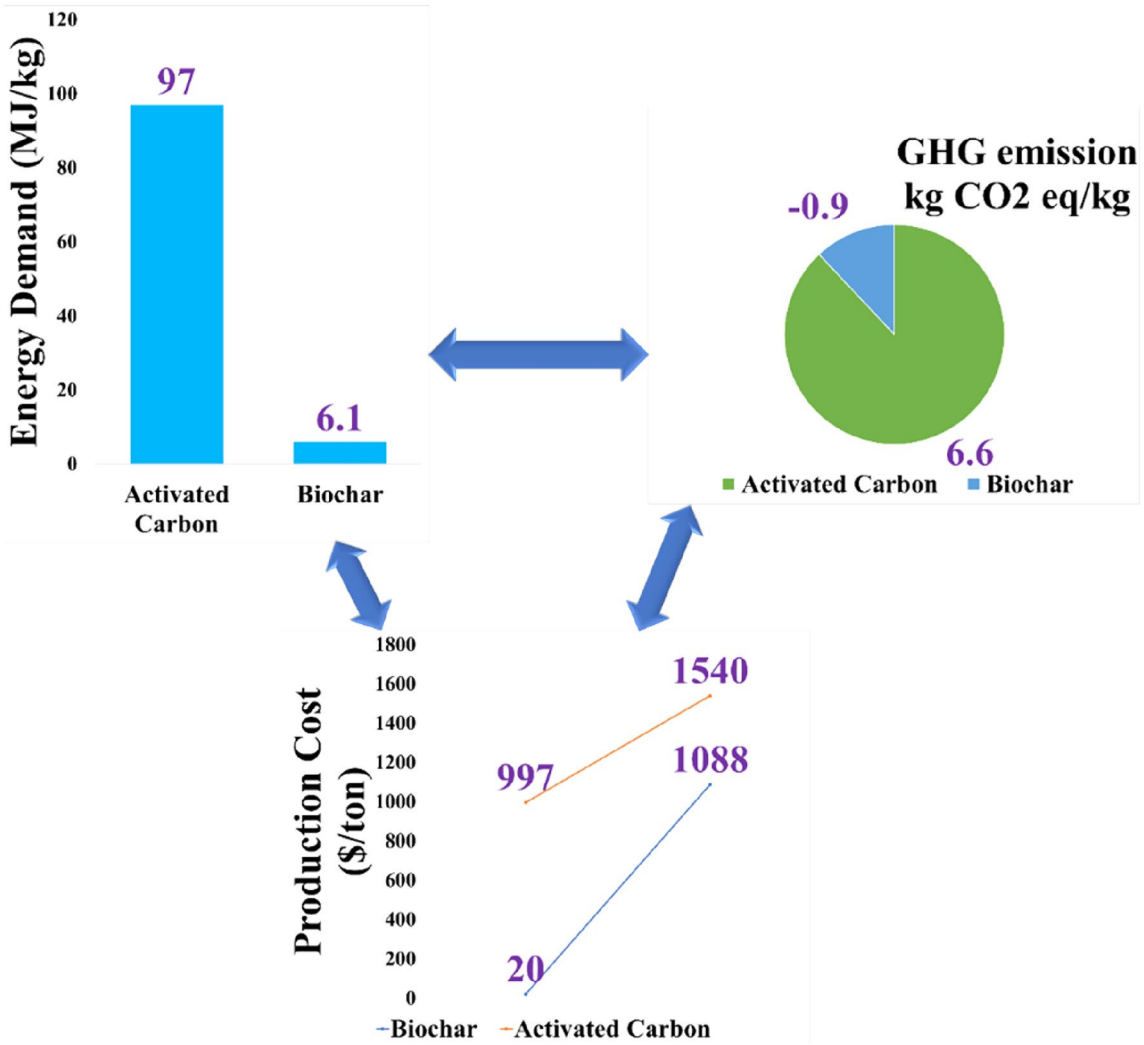


Fig. 2 Some performance metrics during biochar/activated carbon production. [29, 90–94] Source of information.

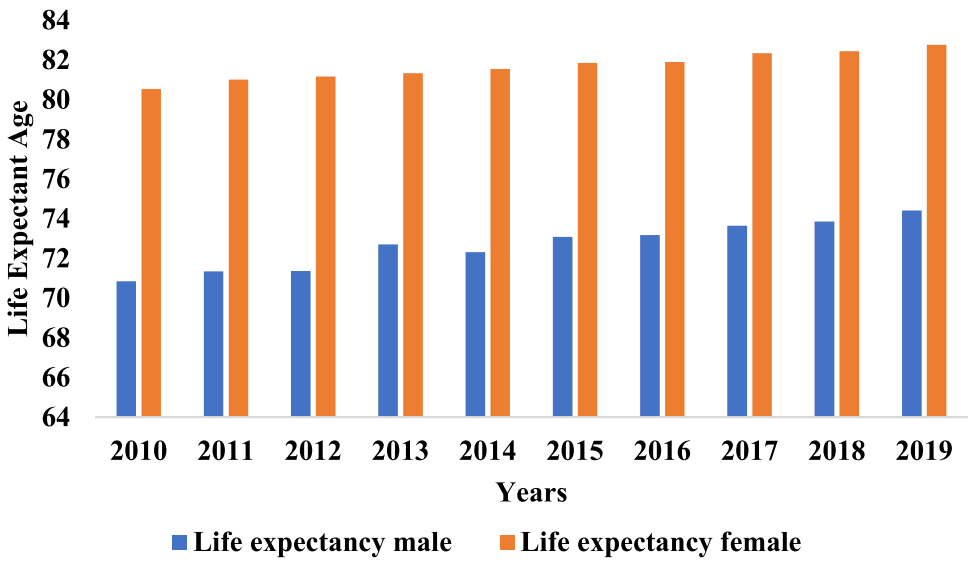


Fig. 3 Life expectancy graph for Estonia between 2010 and 2019. Source. [95]

is efficient [36], environmentally benign [38], and easily operated [39]. Albeit there are concerns about the huge cost of adsorbents like commercial activated carbon, paving way for more research on less-costly ones like biochar [40]. Biochar has been suggested suitable for removing Phcs in an adsorption process [17]. When compared to commercial-activated-carbon, the cost, energy-use, and greenhouse gas (GHG) emissions during the production of biochar is lesser. This is illustrated in Fig. 2.

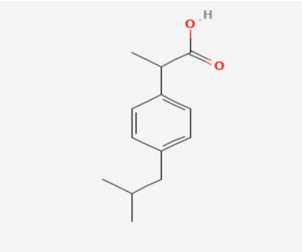
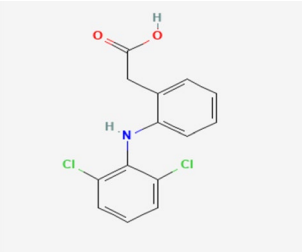
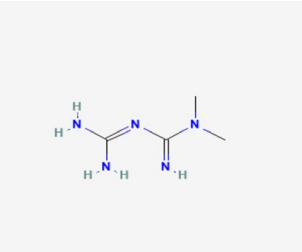
Estonia and IB, MF& DF Phcs

Estonia is one of the countries within the North-Eastern region of Europe. It lies within the latitude and longitudinal points of 59.5° N 28° E and 57.5° N 22° E respectively. Its vegetation zone is known as hemiboreal with a relatively flat landscape, albeit its south-eastern region is more mountainous [41]. There are four seasons in Estonia, spring starts from March–April, followed by summer from June–August, then Autumn from September–November and cold winter from December to February [41]. According to Statistics, Estonia's estimated population is 1.32 million [42], and the life expectancy of Estonians has shown a positive trend in the last decade as people are expected to live much longer; between 70–74 and 80–82 years for males and females respectively (Fig. 3). With the extended life expectancy, the consumption of IB and other medications to maintain a healthy lifestyle will increase [43]. Furthermore, according to the data available from Estonia Statistics on medicine, the consumption of different prescribed and over-the-counter drugs has risen significantly [2, 44]. Besides, three major Anatomical Therapeutic Chemical (ATC) groups of Phcs (such as alimentary tract drugs and nervous system pharmaceuticals ["non-steroidal anti-inflammatory drugs" (NSAIDs)] were frequently consumed among Estonians between 2016 and 2018, with MF, DF, and IB falling under this groups of Phcs [45, 46]. Hence, they are now frequently detected in municipal WWTP influent and effluent streams [47], resulting in their prevalence within the environment. Besides, with Estonia being one of the countries in the Baltic Area, data analysis on existing data confirmed that NSAIDs, epilepsy, diabetes and cardiovascular disease medicine were the most used, of which MF, IB and DF belongs to one of these class of drugs [18]. MF is an antidiabetic medicine [22] and is amongst the most detected Phcs in the effluent of WWTP [48]. According to statistics from the international diabetes federation, above 400 million persons worldwide live with diabetes [49], 59 million patients are in Europe and 58,700 cases in Estonia; which equals about 6.2% of

Table 2 Estonia's WWTP and their capacities

S/N	# of WWTP	Capacity (in PEs)
1	503	300
2	128	300–2,000
3	20	2,000–10,000
4	15	10,000–100,000
5	2	> 100,000

Table 1 Some properties of IB, DF, and MF

Property	IB	DF	MF
IUPAC name	2-[4-(2-methyl propyl) phenyl] propanoic acid	2-[2-(2,6-dichloroanilino) phenyl] acetic acid	3-(diaminomethylidene)-1,1-dimethylguanidine
Molecular formula	$C_{13}H_{18}O_2$	$C_{14}H_{11}Cl_2NO_2$	$C_4H_{11}N_5$
Molecular weight	206.3	296.1	129.2
Form	Crystalline and stable	Crystalline	Crystalline
pKa	5.3	4.15	12.4
log Kow	3.97	4.51	−2.64
Melting point	Between 75 and 77.5 ºC	Between 283 and 285 °C	Between 223 and 226 °C
Solubility	21 mg/L (at 25 °C)	2.37 mg/L (at 25 °C)	2 g/10 mL of water
2D-Structure			

Source: [6, 96–101]

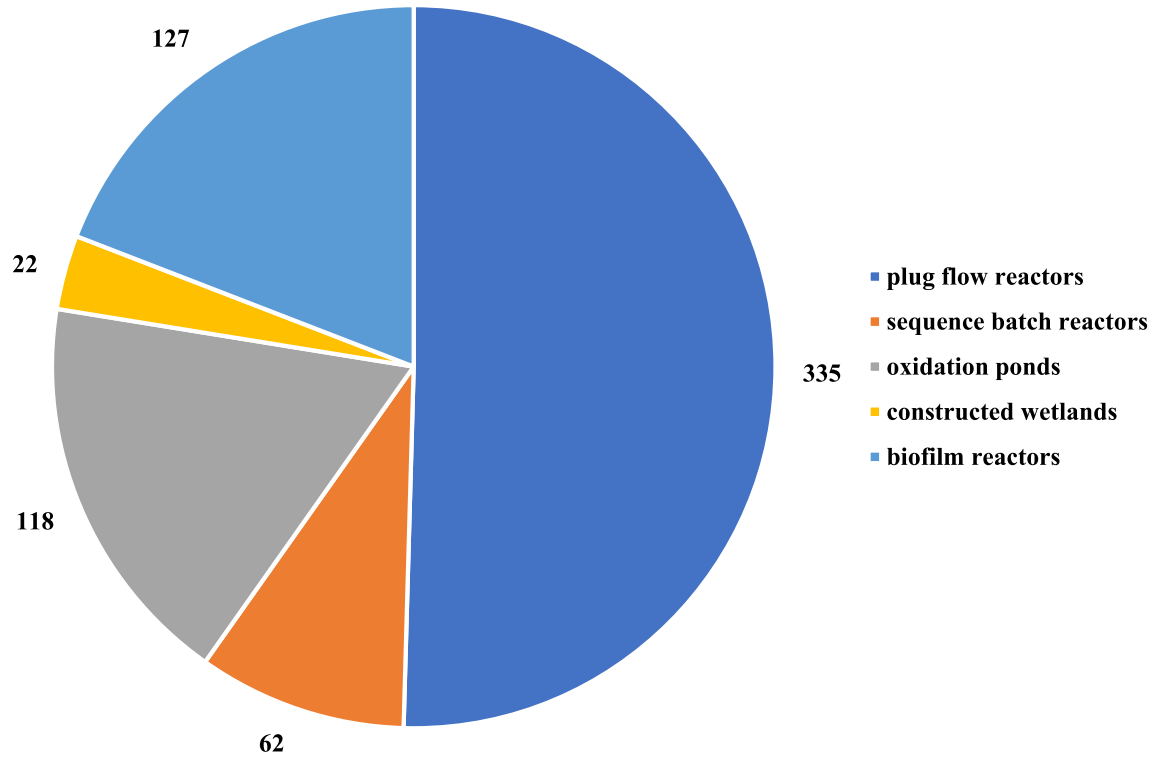


Fig. 4 Treatment technologies at Estonia's WWTP

the adult population in Estonia [50, 142]. MF's prescription has increased significantly owing to its efficiency in glucose removal via acting on metabolic paths to encourage catabolism and the elimination of glucose [48]. Furthermore, in Europe (including Estonia), the consumption of MF will likely increase, because over 68 million people would reportedly have this disease by 2045 [51]. A general report on the fate of Phcs across the Baltic States shows that MF's highest peak concentration exceeded 1 mg/l in WWTP influent while a peak optimum and average concentration of 0.92 mg/l and 0.16 mg/l respectively were detected in their effluent streams [47]. DF is a phenylacetic acid derivative, while IB is generally accessible as 2-(4-isobutylphenyl) propionic acid. Other properties of IB, DF and MF are presented in Table 1. Between 2006 and 2014, there was a significant 14.1% increase in IB consumption in Estonia [43], while the devastating side effects of DF (such as stroke and heart attack in a patient with certain cardiovascular risk factors) has prompted the European Medicine Agency to recommend that DF should be utilized at the lowest active dose [43]. Furthermore, the result of a survey in Europe found the peak absolute concentration of IB (48 µg/L), together with DF (11 µg/L) within the secondary effluent of WWTP [11]. DF has also been included as one of the emerging Phcs into the environment by the European commission, and has resulted in an effort to completely remove DF and other Phcs like MF and IB during the treatment of wastewater [14].

Estonia and WWTP

There are different reports on the number of WWTP in Estonia, however, from the literature that was found, we can conclude that the number varies from 664 to 730 [52–54]. Based on the report of [52], the capacity of each of the WWTP varies from 300 to > 100,000 PEs (person equivalents). The breakdown is presented in Table 2. Besides, the main wastewater treatment technologies utilized at Estonia's WWTP are presented in Fig. 4, where the use of batch and plug flow reactors fall under the activated sludge technology, oxidation ponds and wetlands fall under the natural solutions while others are the biofilm reactor technology. However, these technologies utilize a combination of physical, biological, and chemical treatment methods (including those mentioned in "General wastewater treatment methods" Section) that are deemed insufficient to totally take out Phcs in WWTP. This further buttresses the need to complement these existing methods (in "General wastewater treatment methods" Section) with adsorption using lower-cost biochar, which is cheaper when compared with activated carbon for improving the removal of Phcs.

Analysis of Phcs in Wastewater

To ensure the complete removal of Phcs during the treatment of wastewater, they need to be properly analyzed to ascertain

Table 3 Some common techniques for the analysis of Phcs in wastewater

S/N	Analytical methods	Description	Phcs analyzed	References
1	HPLC	It is utilized for identifying, separating and quantifying Phcs within different component mixture [102]. Besides, it is referred to as the contemporary application of LC [103], and is the choicest technique for the analysis of compounds with low volatility [104]	Fluoxetine	[30]
			Paracetamol	[105]
			Naproxen	[106]
			IB	[107–109]
			DF	[110]
2	UHPLC	It encompasses LC-separations that uses CEL that have smaller sizes than 2.5 to 5.0 µm [111]. Besides, it utilizes high pressure to improve its performance in identifying Phcs in a mixture of various components, with swifter form of analysis at better resolutions (EAG [112]	Acetaminophen	[31]
			Valsartan	[31]
			Cephalexin	[31]
3	UPLC	It is reported to create a novel path for LC owing to its sensitivity and improved resolution [113]. Furthermore, it is an improvement to HPLC with elevated pressures and offers a better throughput and efficiency at a lesser time of analysis (Creative [114, 115]. Lastly it uses particles whose diameter is lesser than 2 µm ([116]	Fluoroquinolone antibiotics	[117]
4	UV–Vis	It is a widely utilized method and works based on the quantity of distinct wavelengths of noticeable light that is absorbed or diffused by a sample. It is effective in the analysis of both organic and inorganic components of a mixture [118, 119]	Ofloxacin	[120]
			DF	[92, 121]
			Levofloxacin	[121]
			IB	[108, 109, 122, 123]
			Cephalexin	[92]
			Ketoprofen	[123]
			MF	[124]

HPLC High performance liquid chromatography, *UHPLC* Ultra-high performance liquid chromatography, *UPLC* Ultra performance liquid chromatography, *UV–Vis* Ultraviolet visible spectrophotometry, *LC* Liquid chromatography, *CEP* columns enclose particles

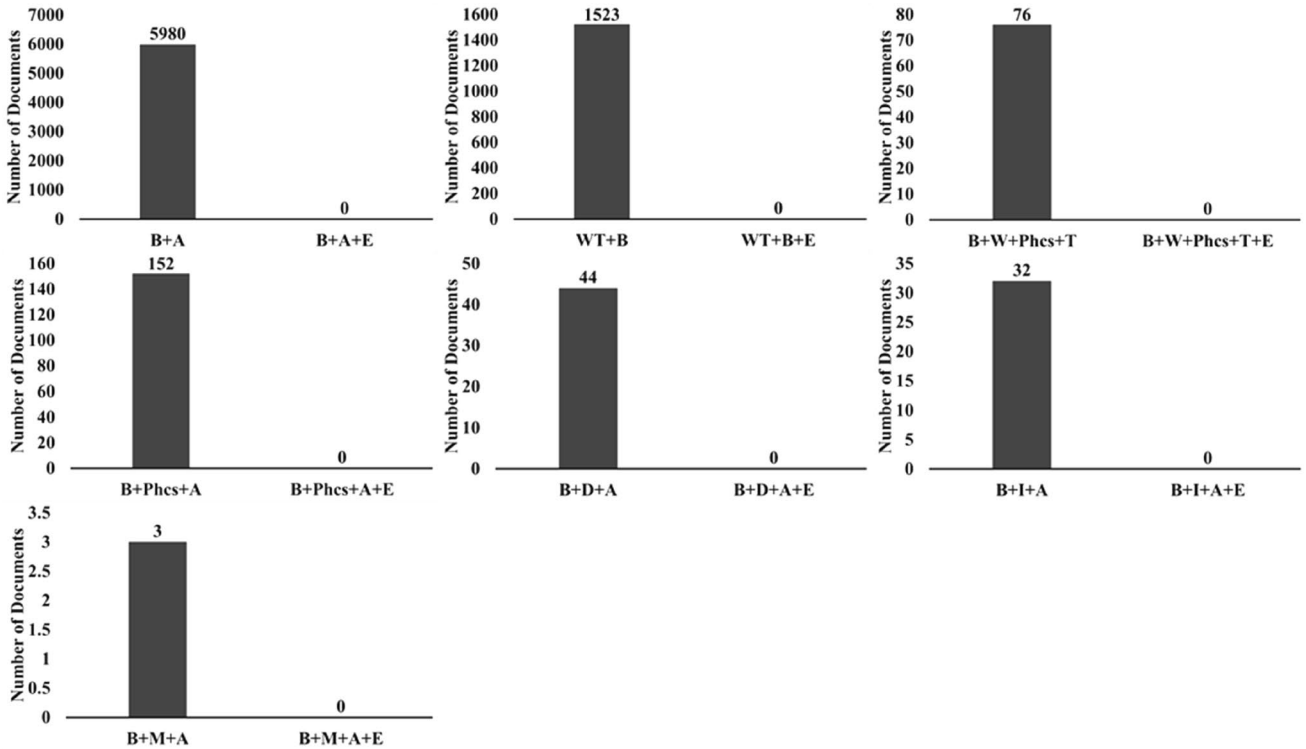


Fig. 5 Database analysis graph. *B* Biochar, *A* Adsorption, E Estonia, *WT* Wastewater treatment, *W* Wastewater, *Phcs* Pharmaceuticals, *T* Treatment, *D* Diclofenac, *I* Ibuprofen, *M* Metformin

Fig. 6 Suitable biochar properties as an adsorbent for Phcs uptake

their actual concentration within the influent and effluent streams of WWTP. Some of the common techniques for the analysis of Phcs are presented in Table 3.

Methodology

For database analysis, the Scopus database was used to establish the need for a synopsis on the possibility of using biochar to augment the removal of Phcs in Estonia's WWTP. Several searches were conducted using different keywords. The first search was steered at establishing the rising interest in the utilization of biochar for adsorption. Using biochar+adsorption as the main search word, bounded using the title of publications, abstracts, and keywords between 2005 and 2021. 5980 research documents were found. When Estonia was added to the main search words (biochar+adsorption+Estonia) under similar search conditions, no document was found. Although there might be a few documents on other databases, however, this result reflects the state of biochar research in Estonia. The second search was carried out using wastewater treatment+biochar as the main words under similar search conditions, and 1523 documents were found. This is lower than the result of the first search, and based on this, we concluded that the research into using biochar in wastewater treatment is just budding. Like the first search, when Estonia was added to the main search words (wastewater treatment+biochar+Estonia), there was no document found. Other searches were conducted to study the extent of research into using biochar

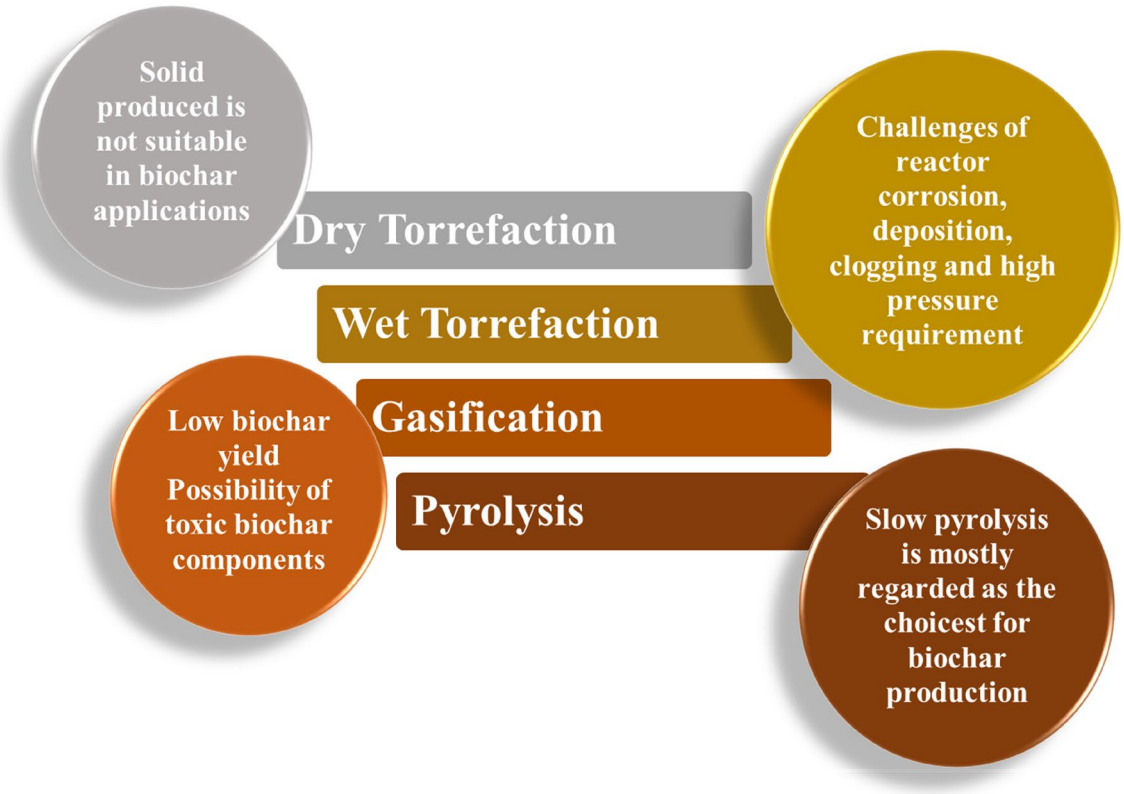


Fig. 7 Common thermochemical techniques for biochar production

for the adsorption of IB, DF, and MF, and the results are shown in Fig. 5. From the results, there was a confirmation of the need to present a summary of the potential of biochar as an adsorbent for Phcs uptake. This was later narrowed down to three of the most prevalent Phcs (IB, DF, and MF) in Estonia.

Biochar and Production

Biochar is rich in carbon and produced via the pyrolysis of various feedstocks, ranging from virgin to waste biomass. This comprises of agricultural plantings, manures, farm wastes, and municipal wastes [40, 55]. Some of biochar's remarkable properties as shown in Fig. 6 present it as a suitable material in diverse applications, including being used as an adsorbent in treating wastewater. Furthermore, the possibility of utilizing low-cost and easily accessible biomass wastes as its precursor cements biochar as a sustainable adsorbent that can be used during wastewater treatment [56].

Biochar is produced via several thermal-chemical processes [57]. It includes torrefaction, and gasification, together with pyrolysis (Fig. 7). The choice of thermochemical process and process parameter is vital during biochar production [40]. Generally, torrefaction is used in converting biomass feedstock into a stable solid that can be used as an energy fuel [58], it could either be a dry or wet process [59]. Dry torrefaction is used to pre-treat biomass [60]. During this process, biomass is warmed-up between 200 and 300 °C, in an inert setting for about 0.5 to a few hours [61]. Although dry torrefaction produces a biochar-like substance (known as torrefied biomass) [62], however, it is best to say that torrefaction is for improving some of the properties of raw biomass before being used in energy applications [59].

Wet torrefaction, on the other hand, is also known as hydrothermal carbonization [61]. It is utilized for charring wet biomass like forestry residue and animal manure [63]. The temperature at which hydrothermal carbonization takes place is relatively low (typically between 180 and 265 °C) [59]. Furthermore, its feedstock does not require drying, thus, reducing energy consumption and production cost [61]. The process takes place within the saturation vapor pressure of water (2–10 mega-pascal) [61] and at time intervals of 5 min [59] to hours [64]. The products of hydrothermal carbonization include hydrochar (slurry-char), bio-oil (liquid), and a small amount of gas (majorly CO_2) [62], [61]. Nonetheless, the problems of reactor corrosion, deposition, clogging, and high-pressure requirements limit the utilization of wet torrefaction [61].

Table 4 Reports on biochar production, characterization, and utilization for Phcs adsorption

SN	Biomass	Conditions of pyrolysis	Some biochar characterization techniques				Adsorbed Phcs	Predominant mechanisms governing adsorption	Maximum adsorption capacity	References
			BET	PA	EA (%)	Modifying agent				
1	Fibre of Oil Palm	Temperature: 550 °C Residence Time: 30 min Rate of Heating: 5 °C/min	SSA: 76.050 m^2/g MSA: 62.564 m^2/g MPV:0.0359 cm^3/g	–	C:78.00 H:2.20 O:18.10 N:0.60 S:0.60	–	Cephalexin (CPH), Acetaminophen (APH), and Valsartan (VLT)	Optimum pH of 3.0 Hydrogen Bond between biochar and the Phcs	CPH: 7.9 mg/g APH: 7.3 mg/g VLT: 23.85 mg/g	[31]
2	Rice Bran	Temperature: 300–750 °C Residence Time: 120 min Rate of Heating: < 5 °C/min, Nitrogen Flowrate: 3 L/min	SSA: 120 m^2/g	–	C:27.01 H:1.23 N:3.15 S < 0.30 O:68.31	–	Fluoxetine	Further treatment of biochar in an autoclave prior to use as an adsorbent Electrostatic attraction	92.6% removal of Fluoxetine	[86]
3	Pharmaceutical Sludge	Temperature: 800 °C Residence Time: 90 min	SSA: 264.05 m^2/g (unmodified biochar) 534.91 m^2/g (modified biochar)	–	–	$ZnCl_2$	Levofloxacin	Hydrogen bond, pore filling, and surface complexation, together with $\pi - \pi$ bond	159.26 mg/g	([117]
4	Text Mill Sludge	Temperature: 400–850 °C Residence Time: 240 min Rate of Heating: 10 °C/min	SSA: 91.00 m^2/g	–	–	–	Ofloxacin	$\pi - \pi$ electron-donor–acceptor form of interaction, pore-filling together with Hydrogen bonding Optimum pH of 6	19.74 mg/g	[120]
5	Chaga Mushroom's Residue	Temperature: 300 °C Residence Time: 120 min Rate of Heating: 10 °C/min	SSA: 1676.78 m^2/g MPV: 1.87cm^3/g APS: 3.88 nm	–	–	$ZnCl_2$	Tetracycline	Hydrogen bonding, Pore filling, Electrostatic attraction, and $\pi - \pi$ Interactions	Tetracycline: 947.42 mg/g	[125]
6	Waste of Coffee Bean	Temperature: 350 and 550 °C Residence Time: 120 min Rate of Heating: 7 °C/min	SSA: 429.19 mm^2/g TPV: 0.392 cm^3/g	Moisture: 13.05% Ash: 3.42% Fixed Matter: 65.67% Mobile Matter: 17.87%	C:81.56 H:3.74 N:1.04 O:13.41 S:0.25	H_3PO_4	Levofloxacin	Electrostatic attraction Hydrophilic interaction $\pi - \pi$ Bonding	110.70 mg/g	[121]

Table 4 (continued)

SN	Biomass	Conditions of pyrolysis	Some biochar characterization techniques				Adsorbed Phcs	Predominant mechanisms governing adsorption	Maximum adsorption capacity	References
			BET	PA	EA (%)	Modifying agent				
7	Waste Peels of Pomelo	Temperature: 700 and 900 °C	SSA: 1033 m^2/g TPV: 1.074 cm^3/g	–	–	–	Paracetamol	Pore-filling Hydrogen bonding $\pi - \pi$ Bonding Van der Waals Bonding	147 mg/g	[105]
8	Douglas Fir	Produced via Gasification (Temperature: 900–1000 °C) Residence Time: 1 s	Raw biochar SSA: 468.2 m^2/g TPV: 0.193 cm^3/g PS: 1.45 nm Modified Biochar SSA: 322.0 m^2/g TPV: 0.120 cm^3/g PS: 1.32 nm	–	Raw Biochar C:76.28 H:1.85 N:1.82 O:13.22 Modified Biochar C:58.52 H:1.22 N:1.52 O:5.83	Magnetic Iron Oxide (Fe_3O_4)	Caffeine Ibuprofen Acetylsalicylic acid	Electrostatic interactions Dipole-Diploe interactions Hydrogen bonding $\pi - \pi$ interactions	Caffeine: 75.1 mg/g Ibuprofen: 39.9 mg/g Acetylsalicylic Acid: 149.9 mg/g	[126]
9	Anthriscus Sylvestris	Temperature: 300 °C Residence Time: 15 min Rate of Heating: 10 °C/min Nitrogen Flowrate: 1 L/min	–	–	Raw Biochar C:49.69 H:5.14 N:5.28 O:39.82 S:0.07 Modified Biochar C:69.34 H:1.17 N:0.88 O:28.53 S:0.08	NaOH	Cephalexin	Electrostatic interactions $\pi - \pi$ interactions Hydrophilic Interactions	724.54 mg/g	[92]
10	Kernels of Wild Plum	Temperature: 180–500 °C Residence Time: 35–60 min	–	–	–	KOH	Naproxen	Hydrogen bonding Electron-donor acceptor	73.14 mg/g	[106]
11	Tea Waste (Engineered)	Temperature: 700 °C	–	–	–	Steam	Caffeine	Nucleophilic interactions Electrostatic interactions	15.4 mg/g	[127]

Table 4 (continued)

SN	Biomass	Conditions of pyrolysis	Some biochar characterization techniques				Adsorbed Phcs	Predominant mechanisms governing adsorption	Maximum adsorption capacity	References
			BET	PA	EA (%)	Modifying agent				
12	White Pine and Norway Spruce	Temperature: 900 °C	–	–	–	–	Caffeine	–	11.85 mg/g for White Pine 9.27 mg/g for Norway Spruce	[89]
13	Digestate (40% corn silage and 60% wood)	Temperature: 470 °C Residence Time: 25 min	SSA: 495m^2/g MeSA: 156m^2/g MPV:0.173cm^3_{liq}/g	Ash: 11.03%	C: 84.03 H: 0.71 O: 4.00 N: 0.23 S: –	–	Diclofenac Cetirizine Fexofenadine Irbesartan Metoprolol Telmisartan Venlafaxine O-Desmethyl-venlafaxine Tramadol Carbamazepine	Electron exchange	For 250 mg/L of biochar, at 10 and 120 min, the sorption efficiency for all the Phcs is greater than 90%	[55]

EA Elemental analysis, *PA* Proximate analysis, *SSA* Specific surface area, *MeSA* Mesopore surface area, *MSA* Micropore surface Area, *MPV* Micropore volume, *APS* Average poresSize, *TPS* Total pore size, *PS* Particle Size, *TPV* Total Pore Volume

Gasification is another thermochemical process [65]. Unlike torrefaction, gasification requires partial oxygen [65], and an elevated temperature of 500–1400 °C [66] within a short residence time, typically between 10 and 20 s [67]. The main target of biomass gasification is to produce gas mixtures of methane (CH_4), alongside carbon monoxide (CO), and hydrogen (H_2), and a small amount of carbon dioxide (CO_2) [66]. Although it produces biochar as a byproduct at a scanty rate (typically at a yield of about 5–10%) [68]. Similarly, biochar made from biomass gasification contains various inorganic elements, together with polycyclic-aromatic-hydrocarbons (PAHs) [67, 69]. The PAHs produced are harmful, hence limiting the utilization of gasification's biochar for environmental remediation purposes [70].

Pyrolyzing biomass is mostly known as the breakdown of biomass raw material within a zero oxygen setting using heat [68]. It is the main technique for producing biochar, such that biomass disintegrates into vapor [condensable (bio-oil, tar) and non-condensable (syngas)], and solid (biochar) [71]. The temperature interval during pyrolysis usually lies between 300 and 1000 °C [71]. It is cost-effective and possesses the ability to produce a variety of products that can be used in different applications (syngas and bio-oil can be converted into energy fuels) [72, 73]. Based on the conditions of operation, pyrolysis can be mainly as fast and slow type [71]. Fast pyrolysis takes place at a temperature interval of 400–600 °C, high heating rate (typically greater than 300 °C/min), and little vapor-residence time (typically between 0.5 and 10 s). The main target of fast pyrolysis is bio-oil, although biochar is formed as a by-product (about 15–30 wt.% yield) [67, 71]. Conversely, the slow form of pyrolysis is the choicest for producing biochar. This is because it produces the highest amount of solid product (typically between 35 and 50 wt.%) [67, 71]. It occurs between 300 and 800 °C [71], with a heating rate of 5–10 °C/min, and a few minutes to hours of residence time [67, 74].

Biochar Characterization

In understanding the features and possible application of biochar, it is subjected to various characterizations. Some of the characterization techniques include elemental, proximate, thermogravimetric analysis (TGA), scanning electron microscopy (SEM), Brunauer–Emmett–Teller (BET), X-ray Diffraction (XRD), X-ray photoelectron spectrometer (XPS), Fourier transform infrared spectroscopy (FTIR), pH, and energy dispersive X-ray (EDX) analysis [75]. Elemental analysis is utilized in determining the carbon, hydrogen, nitrogen, oxygen (by difference) [76], and sometimes sulfur content of biochar samples [77]. Proximate analysis is influential in determining the volatile matter, moisture, and fixed carbon, together with ash [76]. TGA assists in understanding the thermal decomposition

Table 5 Biochar for MF uptake

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
1	Leaves of Artichoke	–	The possibility of utilizing biochar as an adsorbent for MF uptake was attested to, such that biochar made from the leaves of artichoke was modified using NaOH and performed well in the uptake of MF-hydrochloride (a drug for controlling diabetics). In a low oxygen atmosphere, artichoke leaves were pyrolyzed to produce nano-biochar that was modified with sodium hydroxide	Reportedly, different parameters affected MF-hydrochloride's removal. Different concentrations of MF- hydrochloride were adsorbed by the nano-biochar. However, the highest removal percentage for 10, 15, and 20 mg/L of MF-hydrochloride are 85.7, 84.8, and 82.7% respectively. Lastly, the kinetics was best defined using a *pseudo-second-order* model	10.0 15.0 20.0	1.43 2.28 3.46	[128]
2	Fruit shell of chicha-do-cerrado	–	Biochar made via the pyrolysis of the fruit shell of chicha-do-cerrado was activated (using physical and chemical techniques) and proved effective in the adsorption of MF from wastewater. MF's uptake by the activated-pyrolyzed biochar proved that biochar is effective in Phcs uptake during wastewater treatment	The surface area of the produced biochar was reportedly low. Hence, it was activated to improve this property. Chemically activated (using KOH) biochar's surface area increased to 1273.6 m^2/g, with the capacity for adsorption being 45.50 mg/L, while the physically activated biochar using CO_2 had a surface area and adsorption capacity of 397.9 m^2/g and 33.75 mg/L respectively	10–500	–	[124]
3	Alternanthera philoxeroides	–	Biochar made from specific biomass known scientifically as *Alternanthera philoxeroides* was modified using hydrogen peroxide and utilized in a MF-hydrochloride adsorption experiment. The experimental results proved that biochar properties could be improved and used as an apposite adsorbent for MF's uptake, even in wastewater applications	pH and ion interaction performed a huge role in MF's uptake, with a spontaneous and endothermic adsorption process. The kinetic model was best fitted using a *pseudo-second-order* model while the Freundlich model was the appropriate isotherm model	5E-2-3.2 mmol/L	–	[129]
4	Four different biomasses (sawdust from pine, rice husk, macro-algae, and micro-algae)	Temperature: 500 °C Rate of heating: 10 °C/min Residence time: 1 h	The produced biochar was utilized for the removal of MF	The properties of biochar were improved using hydrogen peroxide before adsorbing MF. Activated biochar made from a large portion of macro-algae had the highest adsorption potential, removing almost 76% of the pollutant at a pH of 7. Chemisorption was reported as the major mechanism of adsorption supported by the rich functional group on the modified biochar's surface together with electrostatic attractive force. The isotherm for adsorption was well-defined using the Freundlich model	At varying concentrations of MF and different pH, the removal percentage of MF varied from 0.33 to 76.66%		[130]

Table 6 Biochar for DF uptake

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
1	Anthriscus sylvestris	Raw biochar Temperature: 300 °C Rate of heating: 10 °C/min Residence time: 15 min Activated biochar Activation agent: NaOH solution Temperature: 600 °C Rate of heating: 3 °C/min Residence time: 2 h	Activated biochar made from *Anthriscus sylvestris* was utilized for the removal of DF	Using Langmuir model, DF was taken up by the activated biochar to an extent of 392.94 mg/g. Besides, π-π bonding, hydrophobic interaction, Lewis acid–base interactions, and electrostatic adsorption were key to the removal of DF onto the surface of the activated biochar	–	–	[92]
2	Syagrus coronata	Temperature: 400 °C Rate of heating: 10 °C/min Residence time: 2 h	Modified biochar produced by impregnating *Syagrus coronata* biochar with "MgAl/ layered double hydroxide" was utilized in an adsorption experiment for DF-sodium's uptake from water. The experiment's success affirms the possibility of using biochar as a DF adsorbent during wastewater treatment	The experiment proved successful with the removal of this compound in capacities of 168.04 and 138.83 mg/g at temperatures of 60 and 30 oC, respectively. Chemisorption was highlighted as the predominant mechanism for the uptake, while the process was exothermic. This confirms the potential of biochar composite to eradicate pharmaceuticals within water and wastewater	200	36	[131]
3	Pig manure and pine wood	Biochar from pig manure Temperature: 400 °C Residence time: 2 h Biochar from pine wood Temperature: 525 °C Rate of heating: 25 °C/min Residence time: 20 min	Micro-sized biochar made separately from pig manure and pine wood were utilized as adsorbents in a fixed bed adsorption column to adsorb DF. Besides, Laccase was immobilized on biochar's surface. The experiment presents biochar as a suitable adsorbent for the uptake of DF	The result proved positive with an adsorption capability of 4.10 mg/g at a pH of 6.5. Also, the uptake of DF depended on the adsorbent mass together with the column's height	500 µg/L	For 2 g of biochar from pig manure: 55 µg/L after 30 min For 2 g of biochar from pine wood: 200 µg/L after 30 min	[132]

Table 6 (continued)

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
4	Sewage sludge	Temperature: 500–700 °C	Biochar generated from sewage sludge was well utilized as an adsorbent to remove DF from water	Biochar showed signs of being an effective adsorbent for DF with an adsorption capacity of 92.7 mg/g. The mechanism of adsorption for diclofenac was governed by π-stacking exchange with the biochar's surface. This was best described using pseudo-second-order kinetics and best fitted using the Dubinin-Radushkevich isotherm model	–	–	[27]
5	Fique bagasse	Temperature: 650–850 °C Rate of heating: 1 °C/min Residence time: 2–3 h	Biochar formed using fique bagasse as feedstock was tested as an adsorbent under the variation of pH for the removal of DF	The result was promising and showed the possibility of using biochar for the removal of DF	20	–	[133]
6	Aquatic plants	Temperature: 350–600 °C Residence time: 2 h	Biochar made from different aquatic plants was used for eliminating DF-sodium from water	The experimental result presented biochar as an active adsorbent for removing DF-sodium from water. The adsorption onto the surface of biochar was mainly fostered by π-electron attraction, filling of pores, van der Waals force, together with π-π interaction. The kinetic model was well-fitted using a pseudo-second-order model with the Langmuir model being the finest isotherm model. Furthermore, the adsorption capacity for DF-sodium varied between 18.35 and 23.25 mg/g at 294 K	12	–	[110]

Table 6 (continued)

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
7	Sludge	Temperature: 500 °C Rate of heating: 10 °C/min Residence time: 2 h	The possibility of degrading DF-sodium in wastewater was confirmed using sludge biochar that was modified using bimetallic nanoparticles of iron/copper, together with ultraviolet light	This effectively acted as DF's degrading agent during treatment. Reportedly, almost 99.7% of DF was removed using a pseudo-first-order kinetic model	–	–	[134]
8	A mixture of sludge and leaf	Temperature: 200–500 °C Rate of heating: 6 °C/min Residence time: 1–5 h The ratio of sludge to leaf: 1:3	Biochar was co-produced from waste sludge and leaves, modified using HCl, and utilized for the elimination of DF	From the experiment, DF was swiftly removed at 25 °C, with a maximum adsorption capability of 877 mg/g. The kinetic model was well-fitted using a pseudo-second-order model while the Temkin isotherm was the best isotherm model	10–500	–	[135]
9	The waste of Coffee bean	Raw biochar Temperature: 350 or 550 °C Rate of heating: 7 °C/min Residence time: 2 h Activated biochar Temperature: 550 °C Rate of heating: 7 °C/min Residence time: 1.5 h	Biochar activated using H_3PO_4 was produced from the waste of Coffee beans and used for eliminating DF from water	DF was well adsorbed with a capacity of 61.17 mg/g. The mechanism of adsorption onto the surface of biochar was mainly fostered by the existence of π-π bond, electrostatic attraction, and hydrophobic interaction	10–200 mg/L	–	[121]

Table 7 Biochar for IB Uptake

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
1	Shell of Aegle marmelos	Raw biochar Temperature: 650 °C Rate of heating: 4.3 °C/min Residence time: 1 h Activated biochar Temperature: 800 °C Residence time: 1.5 h	Raw and activated (using steam) biochar made from the shell of *Aegle marmelos* were utilized in the uptake of IB from an aqueous solution	Both the raw and activated biochar attained a 90 and 95% uptake at temperatures of 15 and 20 °C respectively. The kinetic model was well-fitted using a pseudo-second-order model while Langmuir and Freundlich adsorption isotherms were the best isotherm models	1–45	–	[136, 137]
2	Bovine bone	Temperature: 600 °C Residence time: 2 h	A composite (Zn/Al) biochar) was utilized for the uptake of IB	At equilibrium, IB's uptake was adequately fitted using Henry's isotherm model. The adsorption capacity was 1032.81 mg/g. This study proved that biochar can be utilized as an adsorbent for IB uptake	25–125	–	[123]
3	Almond shell	Temperature: 550 °C Residence time: 1 h	Biochar made from activated almond shell was utilized to eradicate IB from an aqueous solution	From the experiment, we can conclude that biochar may be utilized as a low-priced adsorbent to eliminate IB from wastewater. The kinetic model was well-fitted using a pseudo-second-order model, with Langmuir isotherm being the choicest isotherm model	15	1.08	[138]
4	Date seed	Raw biochar Temperature: 700 °C Residence time: 1 h Physical Activation Temperature: 800 °C Residence time: 1 h	Two different seeded biochar date-seed-activated-biochar using steam (DSPB) and date-seed-activated biochar using chemical (DSCC) were utilized as adsorbents for the uptake of IB	Optimum IB removal of 87 & 96% was attained by steam and chemically activated biochar after 18 and 21 h respectively. The kinetic model was well fitted using a pseudo-second-order model while Langmuir adsorption isotherm was the best isotherm model. The experiment showed that activated biochar from date seed is a good adsorbent for IB uptake from water and wastewater	20	Using DSPB: 0.75 Using DSCC: 2.60	[122]
5	Shell of Cocos nucifera	Temperature: 450 °C Residence time: 1 h	Physical and chemical activated biochars were produced using *Cocos nucifera* shell. It was physically activated using steam and chemically activated using H_3PO_4	In the study, IB sorption was well defined by Langmuir isotherm and pseudo-first order kinetic models. The rate of adsorption of IB was 9.69 and 12.16 mg/g for physically and chemically activated biochars respectively	1–50	–	[107]

Table 7 (continued)

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
6	Sugarcane bagasse	Physically activated biochar Temperature: 500 °C Residence time: 1 h Chemically activated biochar Temperature: 400 °C Residence time: 1 h	Sugarcane bagasse was used for the preparation of biochar, activated, and used for the adsorption of IB	The uptake performance of the steam and chemically (using H_3PO_4) activated biochars are 82% and 91% after 18 and 12 h contact times respectively. Besides, the maximum adsorption capacity of these biochars are 11.90 and 13.51 mg/g respectively	1–50	–	[136, 137]
7	Chili seeds	Temperature: 400–600 °C Rate of heating: 10 °C/min Residence time: 2 h	At temperatures between 400 and 600 °C, chili seeds were utilized as precursors for producing biochar and utilized for the uptake of IB	The produced biochar was used for IB's removal. The uptake was 50 times higher with biochar made at 600 °C when compared with the raw seeds of chili. The mechanisms that governed the uptake of IB are hydrophobic, electrostatic, and π-acceptor interactions, together with an intraparticle form of diffusion	50–100	–	[139]
8	Pine wood	Temperature: 425 °C Residence time: 20–30 s	Under a rapid pyrolysis condition, biochar was made as a by-product from pine wood and used for IB's adsorption	The Langmuir adsorption capacity revealed that 10.74 mg/g of IB was adsorbed. Pseudo-second-order kinetics explained the removal process, while the presence of carboxylic, hydroxyl, and phenols favored IB's uptake	25–100	–	[140]
9	Parthenium hysterophorus	Raw biochar Temperature: 300 °C Residence time: 1 h Chemical activation Temperature: 500 °C Residence time: 1 h	Chemically modified biochar (using NaOH) from *Parthenium hysterophorus* was utilized for IB sorption from contaminated water	The peak removal efficiency of IB onto the modified biochar was realized to be above 99% at an adsorbent material dosage of 20 g/L and confirms the potential of biochar as a possible adsorbent of IB during wastewater treatment. The kinetic model was well-fitted using a pseudo-second-order model, while the adsorption data at equilibrium was best fitted using a Langmuir adsorption isotherm	20–100	–	[108, 109]

Table 7 (continued)

S/N	Starting material	Conditions of pyrolysis	Summary	Observation and mechanism of adsorption	Influent concentration (mg/L)	Effluent concentration in mg/L (optimal values only are reported)	References
10	The husk of mung bean	Raw biochar Temperature: 550 °C Rate of heating: 3.7 °C/min Residence time: 1 h Physical Activation Temperature: 650 °C Rate of heating: 3.7 °C/min Residence time: 1 h	Steam-activated biochar from the husk of mung bean has been verified to be highly efficient (> 99%) in the removal of IB from an aqueous solution	The kinetic model was best fitted using a pseudo-second-order model while the adsorption data at equilibrium was best fitted using a Langmuir adsorption isotherm. The study showed that this type of biochar can be a profitable, effective, and safe adsorbent for removing IB from wastewater	20	0.168	[108, 109]

[78], stability [79], and kinetic pattern during the pyrolysis of biomass to form biochar [80]. SEM is an analysis that is useful in detecting biochar's superficial morphology and structure [79]. BET is a valuable technique for determining the pore size together with the pore volume, and the surface area of biochar [79], XPS is functional in evaluating the elemental constituent (albeit at the surface alone) [79], while XRD is useful in determining the crystalline and amorphous phase present in biochar [81]. pH analysis detects the extent of acidity or alkalinity of a biochar material, and FTIR is beneficial in determining the functional groups that are on biochar's surface [75]. EDX is another method for checking biochar's elemental constituent [75], ThermoFisher [82]. Other characterization techniques include "inductively coupled plasma mass spectrometry (ICP-MS)" [83], electrical conductivity [84], and Raman spectroscopy analysis [85]. It is essential to note that the choice of characterization depends on the projected application and utilization of biochar. For using biochar as an adsorbent in WWTP, some of the reported analyses are mentioned in Table 4.

Phcs Adsorption Using Biochar

Globally, carbon-containing materials are used in the adsorption process, with activated carbon (AC) being the most prevalent [40]. However, the cost of its production is high and has resulted in the search for a low-cost alternative [30, 86]. Recently, biochar that possess comparable adsorbent properties to AC has been considered an economically viable option [30, 86]. The slow pyrolysis of biomass and biomass wastes under restricted oxygen condition produces biochar that has recently been described as an appropriate and low-cost adsorbent for improving Phcs removal from WWTP using adsorption [55, 87]. Furthermore, Oliveira et al., elucidated that biochar is a cheap substitute for activated carbon for removing diverse environmental contaminants like Phcs [88]. Some existing reports on the use of biochar for Phcs uptake are mentioned in Table 4 Further reports on the use of biochar for the uptake of MF, DF, and IB are presented in Tables 5, 6, and 7 respectively.

Adsorption is a surface phenomenon such that the adsorbate lies on the adsorbent either through a chemical or physical mechanism [89]. The possibilities of utilizing biochar for Phcs adsorption have been attested to at laboratory scale, with reports of several mechanisms that govern their adsorption onto biochar's surface. Some of these mechanisms are highlighted in Tables 4, 5, 6, and 7. From these, a summary of the prevalent mechanisms controlling the adsorption of Phcs including MF, DF, and IB onto biochar's surface is presented in Fig. 8.

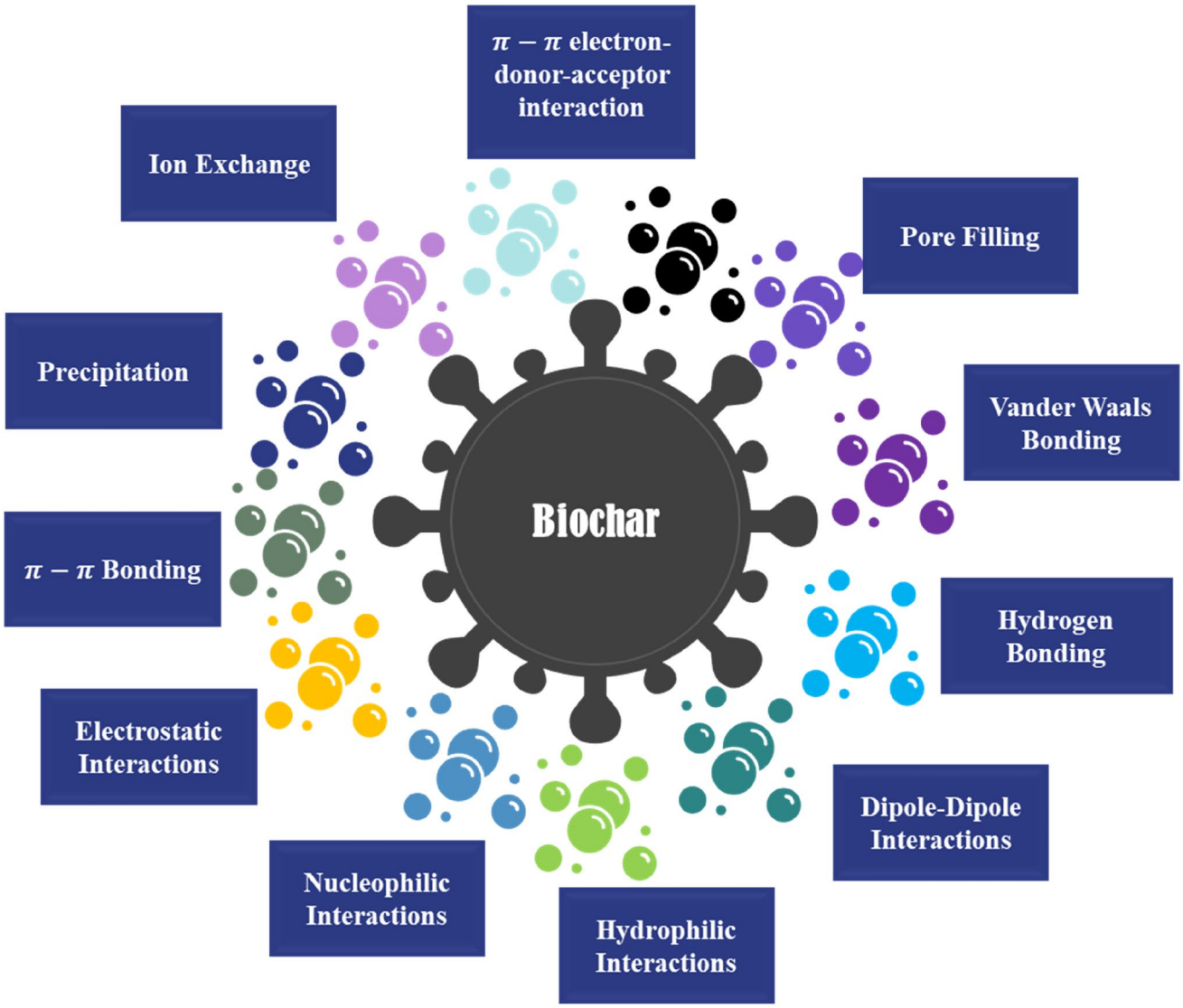


Fig. 8 Prevalent mechanisms governing Phcs (including MF, DF, and IB) adsorption onto biochar

Table 8 Some biochar regeneration techniques

S\N	Biochar regeneration mechanism	Comment	References
1	The electro-Fenton process with the use of an oxidizing agent (Peroxymonosulfate)	An innovative technology	[86]
2	Magnetic separation before thermal-induced regeneration	It proved effective with a total reduction of about 8% in efficiency during the removal	[120]
3	Electro-Fenton approach for biochar regeneration	The presence of iron within the biochar improved its regeneration	[141]

Recommendations

From the summaries provided, we can conclude that it is possible to utilize biochar for improving the removal of Phcs, including MF, DF, and IB during the treatment of wastewater at WWTP. Hence, there would need to be more publicity on this possibility. Furthermore, there is a need for proper education on Phcs disposal within the environment, alongside the danger it poses if it is improperly disposed of. One of the limitations of this study is the concern on the recovery and re-utilization of spent biochar during wastewater treatment, however, there are a few reports on

regenerating spent biochar during adsorption as reported in Table 8. Another concern is on augmenting biochar's physiochemical features to ensure that they perform extremely well as an adsorbent. Most of the studies reviewed had to improve biochar's features for better adsorption performance via activation using physical or chemical means. Hence, this could result in a rise in the overall cost of producing and utilizing biochar for Phcs adsorption. Nonetheless, a cost–benefit analysis should suffice in establishing the trade-off between the cost of biochar production and activation during Phcs' adsorption. For future study, a feasibility study should suffice to ascertain the cost-effectiveness of using biochar for improving the removal of Phcs at WWTP. To do this, a process model that reflects the operations of a WWTP would be very useful. Lastly, most biochar-adsorption experiments have been studied at laboratory scales and there should be a further step in trying it out at pilot and industrial scales. Lastly, asides the possibility of using biochar in an adsorption process, its production from biomass wastes pave a path for a bio-circular economy universally.

Conclusions

The danger of Phcs within the environment has been expressed in this synopsis, with emphasis on MF, DF, and IB in Estonia. The effluents from WWTP have been identified as one of the point carriers of Phcs into the environment, hence, the need to improve existing treatment methods for removing Phcs during wastewater treatment at WWTP. In contrast to the idea of replacing the existing methods of wastewater treatment, it is better to complement them with adsorption. This is because adsorption alone would not be sufficient for improving Phcs removal at WWTP. Owing to this, the possibility of using adsorption with biochar being the adsorbent for improving pharmaceutical removal has been stated in this work.

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Declarations

Conflict of interest The authors declare that there is no conflict of interest.

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