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Biochars and hydrochars for the adsorption of organic contaminants from wastewater

Brigitte Mukarunyana



UMEÅ UNIVERSITY

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Wastewater bears our mark; biomass restores



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Institution eller motsvarande
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Abstract

The continuous release of pharmaceuticals, pesticides, polyphenols, and other organic contaminants into aquatic environments threatens water quality, public health, and ecosystem stability. The problem is especially severe in low-resource regions, where limited treatment infrastructure can lead to the direct discharge of untreated wastewater. This thesis focuses on the potential of biochars and hydrochars produced from locally available biomass residues as sustainable, low-cost adsorbents for reducing organic contaminants in various wastewaters. Through four interconnected studies, it examines the occurrence of organic contaminants in wastewaters and rivers in Rwanda, the performance of different chars, and the production factors that govern adsorption behaviour. High-resolution LC-MS/MS was used to analyse samples. In a study focused on hotspots, hospital effluent was shown to contain elevated concentrations of pharmaceuticals; one of the 28 pharmaceuticals investigated had a concentration up to 24000 ng/L. In the Nyabugogo River in Rwanda, 57 pharmaceuticals, five pesticides, and four polyphenols were detected, with average concentrations ranging from 960 ng/L for pharmaceuticals to 70 ng/L and 49 ng/L for pesticides and polyphenols, respectively. Spatial patterns showed that pharmaceuticals were most prevalent in urban areas, while pesticides and polyphenols were most abundant in the rural and agricultural regions of the Nyabugogo River. Temporal patterns revealed permanent pollution sources. To address these pollution challenges, adsorption experiments were performed using biochars and hydrochars produced from coffee pulp and husk, bagasse, softwood and bark under different thermochemical conditions. Cookstove biochars showed moderate pharmaceutical removal (14–66%). Hydrochars from coffee waste effectively removed polyphenols from coffee processing wastewater, achieving 100% removal, while higher-temperature biochars removed pesticides with 75% average removal. A systematic investigation of gasification conditions showed that increasing char conversion from 0% to 10% under a reactive gasification atmosphere produced the greatest improvement in surface area and removal efficiency, which plateaued at near-complete removal beyond this threshold; biochars produced under a nitrogen atmosphere were comparatively less effective. Bark-derived biochars consistently achieved high removal (>90%), while husk-derived materials mainly improved from 10% of char conversion. These findings indicate that biochars and hydrochars derived from locally available agro-industrial residues can act as adsorbents for organic contaminants adsorption with potential relevance for sustainable water treatment in resource-limited regions.

Abbreviations

ANOVA	Analysis of variance
ATC	Anatomical therapeutic chemical
BET	Brunauer–Emmett–Teller
CHUK	University Teaching Hospital of Kigali
COD	Chemical oxygen demand
CPWW	Coffee processing wastewater
DRIFTS	Diffuse reflectance infra-red Fourier transform spectroscopy
EDA	Electron donor acceptor
HLB	Hydrophilic-lipophilic balance
HPLC	High-performance liquid chromatography
HTC	Hydrothermal carbonization
Kow	Octanol- water partition coefficient
LC-MS/MS	Liquid chromatography coupled to tandem mass spectroscopy
LOQ	Limit of quantification
NMR	Nuclear magnetic resonance
OLS	Ordinary least squares
PAH	Polycyclic aromatic hydrocarbons
PBDEs	Polybrominated diphenyl ethers
PCBs	Polychlorinated biphenyls
PFAS	Per- polyfluorinated alkyl substances
PFO	Pseudo first order
PPCP	Pharmaceuticals and personal care products
PSO	Pseudo second order
RSD	Relative standard deviation
SPE	Solid phase extraction
TGA	Thermogravimetric analysis
TLUD	Top-lit updraft
UHLPC	Ultra high-performance liquid chromatography
WHO	World health organization
XPS	X-ray photoelectron spectroscopy

Sammanfattning

Dagens kontinuerliga tillförsel av läkemedel, bekämpningsmedel, polyfenoler och andra organiska föroreningar till akvatiska miljöer hotar vattenkvalitet, folkhälsa och ekosystemens stabilitet globalt. Problemet är särskilt allvarligt i resurssvaga regioner, där tex begränsad reningsinfrastruktur kan leda till att orenat avloppsvatten släpps ut direkt i miljön. Denna avhandling undersöker potentialen hos biokol och hydrokol, framställda från biomasserester, som kostnadseffektiva adsorbenter för att minska halten organiska föroreningar i olika typer av avloppsvatten. Genom fyra sammanlänkade studier analyseras; förekomsten av organiska föroreningar i avloppsvatten och floder i Rwanda; hur olika kolmaterial adsorberar; samt vilka produktionsfaktorer som styr deras adsorptionsbeteende. Högupplösande LC MS/MS användes för att analysera proverna. En av studierna fokuserade på hotspots och visade att sjukhusavloppsvatten innehöll förhöjda halter av läkemedel; ett av de 28 undersökta läkemedlen uppmättes till så höga koncentrationer som 24 000 ng/L. I Nyabugogofloden i Rwanda påträffades 57 läkemedel, fem bekämpningsmedel och fyra polyfenoler, med genomsnittshalter på 960 ng/L för läkemedel, 70 ng/L för bekämpningsmedel och 49 ng/L för polyfenoler. Studien visade att läkemedel var vanligast i urbana områden, medan bekämpningsmedel och polyfenoler dominerade i flodens jordbrukspräglade delar. För att möta dessa förorenings-utmaningar genomfördes adsorptionsförsök med biokol och hydrokol framställda från kaffebönrester och -skal, sockerrörrester, barrved och bark under olika termokemiska förhållanden. Biokol från enkla spisar visade måttlig avskiljning av läkemedel (14–66 %). Hydrokol från kaffeavfall avlägsnade däremot effektivt polyfenoler från kaffeprocess-vatten och uppnådde 100 % avskiljning, medan biokol producerat vid högre temperaturer avlägsnade bekämpningsmedel med cirka 75 % effektivitet i genomsnitt. En systematisk undersökning av gasifieringsförhållanden visade att en ökning av kolomvandlingen från 0 % till 10 % under en reaktiv gasifieringsatmosfär gav den största förbättringen i specifik yta och avskiljningsförmåga, varefter effekten planade ut. Gasifiering under inert N₂ atmosfär var betydligt mindre effektiv. Biokol från bark uppnådde konsekvent hög avskiljning (>90 %), medan material från kaffeskal främst förbättrades efter 10 % kolomvandling. Sammantaget visar resultaten att biokol och hydrokol, framställda från lokalt tillgängliga restströmmar från jordbruksaktiviteter, kan fungera som adsorbenter för organiska föroreningar och därmed har potential att bidra till hållbar vattenrening i resurssvaga regioner.

List of publications

1. **Mukarunyana Brigitte**, Boman Christoffer, Kabera Telesphore, Lindgren Robert, Fick Jerker, (2023). The ability of biochars from cookstoves to remove pharmaceuticals and personal care products from hospital wastewater. *Environmental Technology & Innovation* 32, 103391.
<https://doi.org/10.1016/j.eti.2023.103391>
2. **Mukarunyana Brigitte**, Sundberg Cecilia, Boman Christoffer, Kabera Telesphore, Fick Jerker. (2026). Coffee pulp and husk-derived hydrochars and biochars adsorb polyphenols and pesticides from wastewater. *Environmental Technology & Innovation*, 41, 104739.
<https://doi.org/10.1016/j.eti.2025.104739>
3. **Mukarunyana Brigitte**, Elias Nyandwi, Telesphore Kabera, Fick Jerker (2026). Occurrence and variability of pharmaceuticals, pesticides, and polyphenols in Nyabugogo River, Rwanda. *Manuscript submitted for publication*
4. **Mukarunyana Brigitte**, Strandberg Anna, Steinvall Erik, Kaium Abdul, Boman Christoffer, Fick Jerker (2026). Influence of the char conversion under gasification conditions on the adsorption performance for organic contaminants. *Manuscript*

Author's contributions

Paper I

The author planned the project under the supervision of the main supervisor (Jerker Fick), collected hospital effluents, pretreated the samples and analysed them with LC-MS/MS, characterized the surface of cookstove biochars, processed and interpreted the results, and led the manuscript writing.

Paper II

The author designed the project, carried out sampling, pretreatment, and chemical analysis, produced hydrochar, performed the characterization, conducted adsorption experiments, and analysed and interpreted the results. The author also led the writing of the manuscript.

Paper III

The author initiated the project and played a key role in project design and planning; planned and performed sampling; performed sample pretreatment and analysis; processed the results; and led the writing of the manuscript.

Paper IV

The author actively participated in the multi-disciplinary project design and planning, characterized all biochars, processed and interpreted the results, and led the writing of the manuscript.

1. Introduction

1.1. Organic contaminants in the aquatic environment

Across the world, aquatic environments are increasingly contaminated by a wide range of organic pollutants, including pharmaceuticals, pesticides, polyphenols, industrial chemicals, and personal care products [1-15]. These contaminants originate from municipal wastewater, hospital effluents, agricultural runoff, industrial discharges, landfill leachate, and stormwater [3, 6, 9-11, 15-18]. Their occurrence in the environment is caused by the combined effects of rising global consumption of chemicals, rapid industrialization, and inadequate wastewater treatment infrastructure [4, 5, 10, 11, 15, 16, 18-20]. Notably, emerging contaminants such as antibiotics, antidepressants, oestrogens, and flame retardants have been detected in rivers and lakes across Europe, Asia, South America, and Africa [1-3, 6, 11, 12, 15]. Certain organic pollutants are not removed by conventional wastewater treatment plants [13, 18, 21-23]. Additionally, as reported by WHO, 50% of wastewater remains untreated globally [24], thereby contributing to environmental pollution. Contamination of water bodies threatens biodiversity, disrupts endocrine systems, increases antimicrobial resistance, and leads to bioaccumulation in food chains, thereby posing major risks to human health and freshwater ecosystems [3, 4, 6, 8, 9, 20, 23]. According to international assessments, water pollution is one of the most significant ecological issues worldwide, affecting both developed and developing regions with important implications for public health, food security, and ecosystem functioning [1, 6, 13-15, 25]. Although organic contaminants are a global problem, the challenges are intensified in sub-Saharan Africa due to rapid urbanization, fast-growing populations, challenges with water scarcity, expanding agricultural activity, and limited wastewater treatment capacity. Numerous studies reported widespread contamination by pesticides, pharmaceuticals, and other emerging pollutants throughout the region [4, 11, 15, 19, 20, 25-27]. Pharmaceuticals are widely detected in aquatic environments across Sub-Saharan Africa, particularly in rivers receiving untreated or poorly treated municipal and hospital wastewater, with common compounds including antibiotics, analgesics, antiretrovirals, and antiepileptics at ng/L- µg/L levels [3, 4, 11, 15, 19, 23, 25, 28, 29]. By contrast, polyphenols (usually reported as phenolic compounds) are far less investigated, with sporadic detections linked mainly to agro-industrial and wastewater effluents, indicating a significant knowledge gap in their occurrence and ecological risk [23, 27]. A systematic review of pesticide exposure research in sub-Saharan Africa, written by Fuhrmann et al., identified 469 study sites across 37 countries,

confirming heavy use of both legacy and current-use pesticides and frequent detection of contaminants in water, sediments, and agricultural produce [15, 30]. Despite bans on organochlorine pesticides in many regions, they remain among the most frequently detected pollutants in African water bodies, demonstrating the persistence of historically used chemicals. Poor pesticide handling and limited awareness further exacerbate risks [20, 31-34]. Moreover, Mengistu et al. found that only 18-26% of farmers used appropriate protective equipment, and 51% stored pesticides in their homes, increasing the risk of environmental contamination and human exposure [20]. These unsafe practices, combined with limited awareness, exacerbate the risks associated with pesticide use [20, 26, 30]. Consequently, sub-Saharan African countries face increasing pressure from both old and emerging contaminants, while lacking the financial and technical capacity to implement advanced wastewater treatment systems [17, 28, 30]. Within sub-Saharan Africa, East Africa has recently emerged as a documented hotspot for organic contamination [11, 12, 15, 17, 29, 34, 35]. Pharmaceutical and personal care products (PPCPs), pesticides, and PFAS have been increasingly detected in water resources across Kenya, Uganda, Tanzania, and Ethiopia [11, 12, 15, 26, 29, 34, 35]. This has been driven by the rapid expansion of healthcare systems, increased pharmaceutical consumption, and widespread discharge of untreated wastewater from urban centres [12, 16, 25, 34]. Agricultural intensification in East Africa has also resulted in heavy pesticide use [20, 36, 37]. The ecological impacts of pesticides are already visible: Rwanda, Kenya, Uganda, Tanzania, and Ethiopia have reported mass mortality events among pollinators [37-40]. In Rwanda specifically, Nganso et al. reported that over 60% of bee colonies collapsed between 2021 and 2023, leading to a 40% decline in honey production, largely attributed to the use of highly toxic pesticides such as profenofos and malathion [38]. These pollinator losses threaten the production of key regional crops such as coffee and fruit, which are essential to local economies [20, 33, 36, 37]. The widespread use of such pesticides, combined with rapid urbanization, expanding agricultural zones, and limited availability of wastewater treatment facilities, exacerbates the problem of organic contamination in Rwanda. The on-site solutions mainly consist of septic tanks, pits and small decentralized units [41-43]. Rivers such as the Nyabugogo, which runs through Kigali City receive inputs from hospitals, residential areas, farmlands, and industrial zones, making them vulnerable to complex mixtures of pharmaceuticals, pesticides, and polyphenols [16, 41, 42, 44-46]. Localized monitoring efforts, including those described in this thesis, have begun to map these contamination patterns, highlighting the urgency of addressing organic pollution through affordable and sustainable treatment solutions.

1.2. Biomass carbonization, pyrolysis and gasification

Biomass carbonization encompasses a range of thermochemical conversion processes that transform organic biomass into carbon-rich solids, primarily biochar, hydrochar, and torrefied biomass, by heating the material under controlled, oxygen-limited conditions [47-55]. These processes remove moisture and volatile matter, thereby increasing the relative carbon content, energy density, and structural stability of the produced char materials [47-49, 53, 54, 56]. Carbonization has regained significant attention due to rising energy demands, environmental concerns, and the need to replace fossils with renewable biomass [47, 54, 56-58]. Since different biomasses vary significantly in composition, moisture content, and conversion behaviour, various types of carbonization exist, each adapted to specific feedstock properties and end-use objectives [48, 49, 58]. The main types of carbonization include pyrolysis, producing a solid carbonaceous material commonly called “biochar”, and other traditional carbonization. An overview of carbonization techniques is presented in Table 1. Other concepts related to pyrolysis include hydrothermal carbonization (HTC), which produces a carbonaceous material called “hydrochar”, and torrefaction, carried out at low temperatures, involving minor transformations of the organic polymers in biomass. The solid carbonaceous materials produced by these methods can be used as a valuable residual stream during gasification. The yield depends on the process conditions, which are designed to produce materials for specific applications in energy production, soil enhancement, pollutant removal, and materials engineering [47-49, 53, 54, 56, 58].

1.2.1. Pyrolysis

Pyrolysis is a thermochemical process that breaks down organic materials at elevated temperatures in the absence, or near absence, of oxygen, thereby preventing combustion and enabling the conversion of the biomass into valuable end products such as pyrolysis oil and biochar [48, 49, 53, 54, 56, 59-62]. The process is typically carried out in a controlled reactor system, where feedstock such as agricultural residues, wood, sewage sludge, or other carbon-rich materials is first dried to remove inherent moisture and then heated to a temperature range of 300 °C to 800 °C. An inert atmosphere, typically provided by nitrogen, is maintained to prevent oxidative reactions [51, 53, 56, 59, 61]. As the temperature increases, the complex polymeric structures of biomass, primarily cellulose, hemicellulose, and lignin, undergo a series of thermal

degradation reactions, including depolymerization, cracking, decarboxylation, and devolatilization [47, 49, 53, 54, 56, 61]. These reactions break down large organic molecules, producing three main fractions: a solid residue (biochar), a liquid condensate (bio-oil), and an energy-rich gaseous mixture (syngas). The relative distribution of these products is governed by process parameters such as heating rate, residence time, temperature, and pressure, as well as the physicochemical properties of the feedstock [48, 49, 53, 63, 64].

Pyrolysis can be broadly categorized into slow pyrolysis and fast pyrolysis, each distinguished by its operational conditions and product yields. Slow pyrolysis is characterized by low heating rates, typically below 10 °C per minute, long residence times ranging from minutes to several hours, and moderate temperatures between 300 °C and 800 °C [47, 49, 61, 63-65]. Under these conditions, gradual heating allows extensive secondary reactions of the evolved vapours, promoting the formation of solid carbonaceous material and resulting in higher biochar yields. Consequently, slow pyrolysis is commonly employed when the primary objective is to produce stable solid products for applications such as soil amendment, carbon sequestration, or material use [59, 61, 64-67]. By contrast, fast pyrolysis is designed to maximize liquid product yield and operates at significantly higher heating rates, often exceeding several hundred degrees per second, with very short vapour residence times of only a few seconds and temperatures typically in the range of 450 °C to 600 °C. The rapid thermal decomposition and immediate quenching of vapours limit secondary cracking reactions, thereby favouring the production of bio-oil [56, 64, 68-71].

Biochar is a carbon-rich solid consisting largely of fixed carbon, ash, and mineral components, and is noted for its porous structure and stability [48, 54, 59, 69, 72, 73]. Bio-oil is a complex liquid mixture containing water and a wide range of oxygenated organic compounds, including acids, aldehydes, ketones, phenolics, and sugars, which contribute to its high reactivity and relatively low stability compared to conventional fuels. Syngas is composed mainly of non-condensable gases such as carbon monoxide, carbon dioxide, hydrogen, methane, and light hydrocarbons, and can be utilized as a fuel for heat and power generation [49, 54, 57, 70, 71, 73, 74].

The applications of pyrolysis and its products are diverse and span multiple sectors. Biochar is widely used in agriculture to enhance soil properties, such as nutrient retention, water holding capacity, and microbial activity, while also serving as a long-term carbon sink [51, 60, 66, 71]. Bio-oil can be utilized as a source of renewable energy for

producing heat and electricity or further upgraded through catalytic processes to produce transportation fuels and high-value chemicals. Syngas is commonly recycled within the system to provide energy for the pyrolysis process itself, improving overall energy efficiency, or it can be utilized in gas engines and turbines for power generation [49, 54, 73, 74]. Beyond biomass conversion, pyrolysis is also applied in waste management for the treatment of plastics, rubber tyres, and municipal solid waste, enabling the recovery of fuels and materials while reducing environmental pollution. As such, pyrolysis represents a key technology in advancing sustainable resource utilization and supporting the transition towards a circular economy [48, 60, 74].

1.2.2. Hydrothermal carbonization (HTC)

Hydrothermal carbonization (HTC) is a thermochemical conversion process that transforms wet biomass into a carbon-rich solid known as hydrochar through thermal transformations and reactions in hot, pressurized water [56, 74-79]. Unlike dry thermochemical processes, HTC is specifically suited for high-moisture feedstocks such as sewage sludge, food waste, manure, and agricultural residues, eliminating the need for energy-intensive drying. The process is typically conducted at relatively moderate temperatures of 180–250 °C under autogenous pressures of approximately 2-10 MPa in a sealed reactor [51, 56, 76-78]. Under these conditions, water plays a dual role as both a solvent and a reactive medium, facilitating the breakdown and reformation of organic matter through a sequence of hydrothermal reactions [74, 77, 78].

The HTC process involves several key reaction pathways, beginning with the hydrolysis of biopolymers such as cellulose, hemicellulose, and lignin into soluble monomers and oligomers. These intermediates subsequently undergo dehydration and decarboxylation reactions, which remove oxygen as water and carbon dioxide, increasing the carbon content. Further polymerization, condensation, and aromatization reactions result in the formation of solid hydrochar particles with a relatively high degree of carbonization [53, 56, 65, 76, 79-82]. Compared to the original biomass, hydrochar exhibits enhanced energy density, improved hydrophobicity, and greater structural stability. However, it generally has a lower surface area and porosity than biochar produced via pyrolysis, although its surface is rich in oxygen-containing functional groups, which can be advantageous for certain applications [77, 80-84].

HTC produces three main product streams: solid hydrochar, a liquid phase commonly referred to as process water, and a minor gaseous fraction. Hydrochar is the primary product and consists of carbonaceous

material along with retained inorganic components such as ash and nutrients. The liquid phase contains a complex mixture of dissolved organic compounds, including organic acids, sugars, phenols, and nitrogen-containing compounds, as well as significant amounts of nutrients such as ammonium and phosphate. This process water is often characterized by high chemical oxygen demand (COD) and may require further treatment, although it also presents opportunities for resource recovery [80, 83, 85, 86]. The gaseous fraction is typically dominated by carbon dioxide, with smaller amounts of carbon monoxide, methane, and other light gases [75, 81, 85].

HTC has a wide range of applications due to its ability to process wet waste streams efficiently. Hydrochar can be used as a solid fuel due to its improved calorific value and coal-like properties, or as a soil amendment to enhance soil structure and nutrient retention [75, 86, 87]. It is also increasingly being investigated as a precursor for activated carbon and other engineered materials for environmental remediation. In water and wastewater treatment, hydrochar has shown potential as an adsorbent for removing heavy metals, organic pollutants, and nutrients, although its performance is often enhanced through chemical or thermal activation. Additionally, HTC is particularly attractive in wastewater treatment contexts because it improves sludge dewaterability and reduces sludge volume, thereby lowering disposal costs [75, 78, 79, 83, 86-88].

HTC represents a promising and sustainable approach for converting wet biomass and biobased waste streams into valuable products, complementing other thermochemical processes, and contributing to a more circular resource utilization.

1.2.3. Gasification

Gasification is a high-temperature thermochemical conversion process that transforms carbonaceous biomass into an energy-rich gaseous product stream known as synthesis gas (syngas). It operates under a limited oxygen supply (i.e. partial oxidation), distinguishing it from both combustion (excess oxygen, full oxidation) and pyrolysis (no oxygen added) [52, 53, 56, 60, 61, 66, 89-93]. In typical biomass systems, gasification occurs at 750–1200 °C, producing a gas mixture dominated by CO, H₂, CO₂, and CH₄ through partial oxidation and reduction reactions that unfold in several distinct thermal zones within the gasifier [56, 60, 89, 90, 93]. The process begins with drying, followed by pyrolysis (fuel devolatilization), during which volatile fuel breakdown. This is followed by partial oxidation, which provides heat, and finally reduction, where char reacts with CO₂ and steam to form CO and H₂ [52, 89-91, 93].

These stages determine the final distribution of products and the energy content of the resulting syngas. Gasification is therefore considered a versatile and efficient pathway for extracting energy from biomass, enabling its use for heating, electricity generation, and the synthesis of industrial chemicals and fuels [53, 60, 91]. Although syngas is the primary product, gasification may also yield other byproducts of significant environmental and industrial relevance [52, 53, 60]. Char, often referred to as *gasification biochar*, the residual carbonaceous solid, remains as an intermediate and final material due to incomplete conversion of biomass under sub-stoichiometric oxygen conditions. It can be collected and applied in soil amendment and carbon sequestration, owing to its persistent carbon structure and high mineral content [52, 60, 93]. More importantly, biochar is increasingly recognized for its potential as a low-cost sorbent in water and wastewater treatment. Research on related biomass-derived biochars and hydrochar produced via hydrothermal carbonization shows significant pollutant removal capabilities, demonstrating the value of carbon-rich adsorbents in environmental remediation applications [78, 79].

Table 1. Types of carbonization, operating conditions and final products

Process	Operating conditions	Final products	References
Torrefaction	Temperature: ~200–300 °C, low or no O ₂ , residence time: ~30–60 min	Solid, small amounts of gas	[53, 56, 74]
Slow pyrolysis	Temperature: ~400–600 °C, heating rate: low, residence time: long (hours)	Biochar (35%), bio-oil (30%), syngas (35%)	[53, 56, 74]
Fast pyrolysis	Temperature: ~500–1000 °C, heating rate: high, residence time: <2 seconds	Bio-oil (75%), some biochar (12%), syngas (13%)	[53, 58, 76]
Hydrothermal carbonization (HTC)	Temperature: ~180–250 °C, residence time: 1-16h, wet biomass	Hydrochar (70%), liquids (25%), gas (5%)	[76, 78, 94]
Gasification	Temperature: ~700–1200 °C, limited O ₂ /steam	Syngas (85%), biochar (10%) liquid (5%)	[56, 78, 91]

1.2.4. Feedstock for biochar and hydrochar production

There is a wide range of feedstocks for biochar and hydrochar production, as shown in Figure 1. The selection of these feedstocks is important

because it influences the physicochemical properties and adsorption abilities of biochars and hydrochars [94-98]. Various biomasses have been used to produce biochar and hydrochar, such as agricultural residues, forestry biomass, industrial and municipal waste, and aquatic biomass [53, 55, 57, 99, 100]. Agricultural byproducts such as coffee and rice husks, corn stover, wheat straw, sugarcane bagasse, and nut shells are often used as feedstocks because they are readily available, are cheap and have a high carbon content [55, 100]. Wet biomass feedstocks, like sewage sludge, agricultural residues (such as peels and pulp), animal manure, and food waste, are well-suited for producing hydrochar. Figure 1 indicates various biomass feedstocks used in thermal conversion processes. Hydrothermal carbonization allows direct processing of these materials, avoiding the need for a pre-drying step prior to some other thermal conversion processes (i.e. pyrolysis, gasification, or combustion) [53, 76]. Overall, the variety of biomass feedstocks provides more flexibility and sustainability in producing biochar and hydrochar [53, 76].

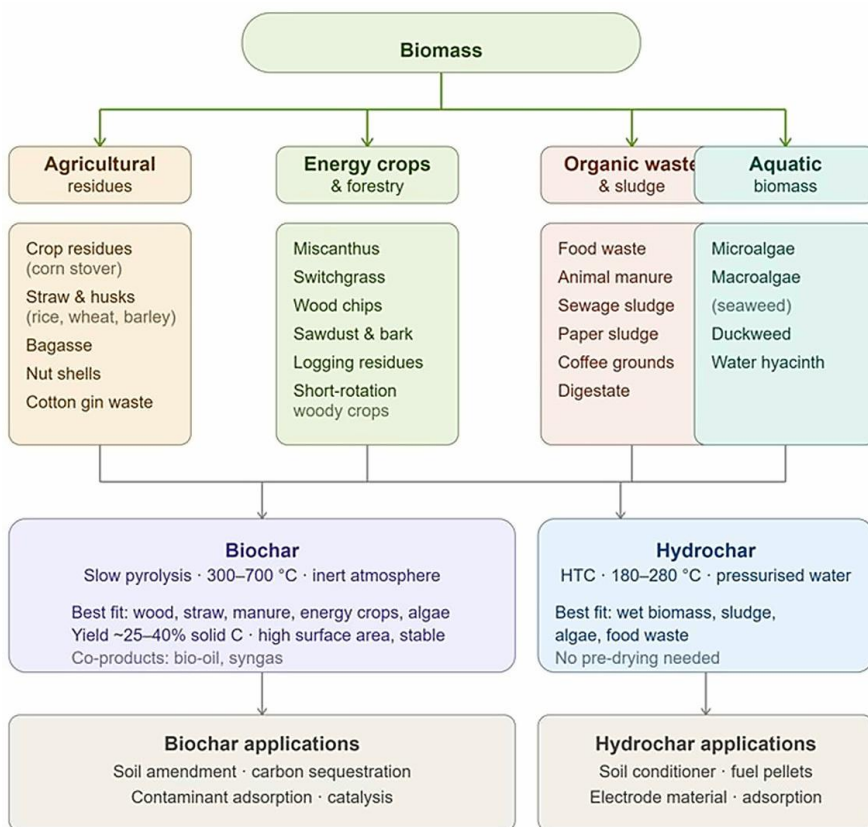


Figure 1. Biomass categories for biochar and hydrochar production.

1.3. Adsorption mechanisms of biochars and hydrochars

The adsorption of organic pollutants onto biochar and hydrochar is governed by a combination of physicochemical interactions arising from carbonaceous structures, surface functionalities, and pore sizes, as well as the properties of the pollutants. Although both materials originate from thermochemical conversion processes, their adsorption behaviour differs due to variations in surface area, degree of carbonization, and the abundance of functional groups [69, 94, 101]. Biochars, typically produced via pyrolysis at higher temperatures, can develop greater aromaticity, higher surface area, and more microporosity, whereas hydrochars, produced through hydrothermal carbonization, generally possess lower surface areas but higher densities of oxygen-containing functional groups such as hydroxyl, carboxyl, and carbonyl groups [77, 101, 102]. These differences influence the dominant adsorption mechanisms involved in the removal of organic contaminants from aqueous environments.

One of the primary mechanisms is *hydrophobic interaction*, which plays a significant role in the adsorption of nonpolar or weakly polar organic compounds such as polycyclic aromatic hydrocarbons (PAHs), some pharmaceuticals, and pesticides [103, 104]. The hydrophobic domains of biochar, formed by condensed aromatic carbon structures, preferentially attract and retain hydrophobic organic molecules, effectively partitioning them from the aqueous phase onto the solid surface [103, 105]. This mechanism is generally more pronounced in high-temperature biochars due to their increased aromaticity and reduced polarity. By contrast, hydrochars, which are more polar and less carbonized, tend to show weaker hydrophobic interactions unless further modified.

π - π *electron donor-acceptor* (EDA) interactions also contribute significantly to adsorption, particularly for organic pollutants containing aromatic rings [105, 106]. The graphitic structures in biochar can interact with the π -electron systems of organic molecules, facilitating adsorption via stacking interactions. Depending on the electron density of both the adsorbent surface and the adsorbate, these interactions may involve EDA complexes. Biochars produced at higher temperatures are especially effective in this regard due to their more developed aromatic structures, while hydrochars may exhibit weaker π - π interactions unless subjected to post-treatment to enhance carbonization [72, 106].

Another important mechanism is *hydrogen bonding*, which occurs between functional groups on the surface of the adsorbent and polar organic molecules. Both biochar and hydrochar contain oxygenated functional groups capable of acting as hydrogen bond donors or acceptors

[50, 72]. Hydrochars often show stronger hydrogen bonding capacity due to their higher content of surface functional groups. This mechanism is especially relevant for the adsorption of polar organic contaminants such as pharmaceuticals, dyes, and endocrine-disrupting compounds [50, 94].

Electrostatic interactions also influence adsorption, particularly for ionizable organic pollutants. The surface charge of biochar and hydrochar depends on factors such as pH and the presence of functional groups. Under certain pH conditions, the adsorbent surface may become positively or negatively charged, thereby attracting oppositely charged organic species [94, 105]. For example, negatively charged biochar surfaces can attract cationic dyes, while positively charged surfaces under acidic conditions may enhance the adsorption of anionic compounds. Hydrochars, with their higher functional group density, often show more pronounced pH-dependent surface charge behaviour [50, 94, 107].

Pore filling is another key mechanism, particularly for biochars with well-developed microporous and mesoporous structures [85, 108]. Organic molecules can diffuse into the internal pore network and become physically trapped, thereby increasing adsorption capacity. This mechanism is strongly influenced by the size and shape of both the pores and the pollutant molecules [85, 104, 108]. Biochars produced at higher temperatures typically have more developed pore structures, making pore filling a dominant mechanism, whereas hydrochars may require activation to enhance porosity.

Additionally, *surface complexations* contribute to adsorption, although they are generally considered weaker interactions compared to chemical bonding mechanisms [104, 109]. In modified or engineered biochars and hydrochars, additional mechanisms such as cation bridging or specific chemical interactions may also occur, particularly when metal ions or functional modifiers are introduced onto the surface [94, 109].

The adsorption of organic pollutants onto biochar and hydrochar is a multifaceted process involving both physical and chemical interactions, as illustrated in Figure 2. The relative importance of each mechanism depends on the properties of the adsorbent, the nature of the pollutant, and environmental conditions such as pH, temperature, and ionic strength. Understanding these mechanisms is essential for optimizing material design and improving the efficiency of carbon-based adsorbents in environmental remediation applications.

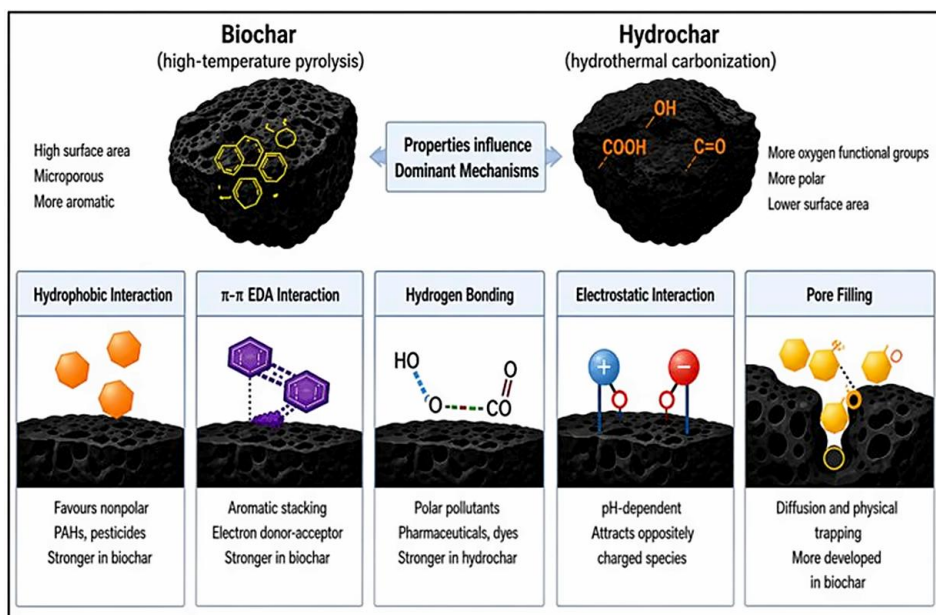


Figure 2. Adsorption mechanisms of biochars and hydrochars for organic pollutants.

1.4. Factors affecting the adsorption performance of biochar and hydrochar in wastewater

Adsorption of pollutants onto biochars is substantially influenced by both the surface characteristics of the biochars and the properties of the pollutants. The surface properties, in turn, depend on the type of feedstock and the techniques and conditions used during production. The main surface factors affecting the adsorption of organic pollutants onto biochars include surface area, pore-size distribution, charge, organic and ash content, chemical functional groups, and elemental composition. [71, 80, 103]. The molecular size, functional groups, and charge of pollutants also play essential roles in adsorption processes. Despite increasing research on biochar and hydrochar for wastewater treatment, major gaps remain in our understanding of what affects their adsorption performance and long-term stability. This makes it hard to compare findings across studies. Additionally, wastewater is a complex matrix in which factors such as pH, ionic strength, and the presence of competing ions or organic compounds can significantly affect adsorption processes [105, 106]. Investigation of the influence of these factors is therefore necessary to tailor biochar and hydrochar for use in real wastewater treatment applications. Coupling wastewater treatment with the production of

biochars and hydrochars to valorize biomass waste offers a promising path to improve resource management and to minimize environmental impacts [51, 59, 109]. However, such an approach requires a deep evaluation of biochar and hydrochar properties, treatment performance, and life-cycle implications.

1.5. Comparison between biochars, hydrochars and other conventional adsorbents

Biochars and hydrochars are cheaper, since they are usually made from abundant, renewable, and often waste-derived biomass sources, such as agricultural waste, forestry byproducts, and sewage sludge [48, 49, 68, 69, 94, 110]. This not only reduces reliance on non-renewable resources but also provides a sustainable means of waste valorization and carbon sequestration [56, 59, 62, 68, 111]. The production of non-activated biochars and hydrochars consists of simple processes that can save energy and reduce greenhouse gas emissions, whereas physical and chemical activation primarily requires high temperatures, extended processing times, or intense chemical treatments [56, 87, 110]. Additionally, producing non-activated chars prevents the use of activation agents and the subsequent washing and neutralizing phases by which secondary waste can be generated. It also reduces harmful chemicals that could remain in the produced chars [94, 110, 112]. Non-activated biochars and hydrochars yield higher mass because carbon loss during activation is avoided. Conventional adsorbents, including activated carbon, zeolites, and silica gel, are often made from raw materials such as coconut shells, wood residues, peat, and various agricultural wastes, which require extensive energy and are therefore not cost-effective [48, 67, 113, 114]. This makes them attractive for large-scale or local wastewater treatment systems. Their physical and chemical properties can also be easily changed by selecting different feedstocks and production conditions to optimize the adsorption capacity for a given pollutant [59, 68, 115].

Activated biochars, however, have higher surface areas and abundant reactive surface functional groups, which enhance their adsorption capacity and speed up pollutant removal compared to non-activated biochars [114, 116]. This makes activated biochars more effective for applications such as water treatment and contaminant remediation.

Non-activated biochars and hydrochars are better options for situations where ultra-high adsorption capacity is not critical and where reducing life-cycle environmental impacts is a main goal. The combination of low cost, renewable sourcing, tunable properties, and environmental benefits

makes biochars and hydrochars highly attractive and sustainable alternatives to conventional adsorbents. Their continued development and optimization are expected to play a crucial role in advancing green technologies for pollution control, resource recovery, and environmental protection.

1.6. Benefits of integrating organic waste into water treatment in Rwanda

Rwanda offers a very suitable environment for integrating biochar and hydrochar into water treatment systems because of its available agricultural resources, decentralized water infrastructure, and environmental policy framework [117]. These elements create positive conditions for the adoption of low-cost, sustainable, and locally adaptable treatment technologies that can work effectively at both household and community levels.

A key advantage is the country's *abundant biomass resources*, including coffee pulp and husks, rice husks, maize cobs, and banana residues. These agricultural by-products are widely available in rural districts and are often underutilized or disposed of by open burning, which contributes to environmental degradation [118, 119]. Converting these materials into biochar or hydrochar not only addresses waste management issues but also creates value-added products for water treatment. This approach reduces reliance on imported materials, such as activated carbon and chemical coagulants, thereby lowering operational costs and improving the sustainability of water treatment systems.

Regarding water quality challenges, Rwanda faces a dual burden influenced by its *socio-economic and geographic conditions*. In rural areas, water contamination is caused by agricultural practices, including pesticides, poor sanitation facilities, and dependence on untreated surface and groundwater sources [120, 121]. In urban and peri-urban areas, especially around Kigali, rapid population growth and industrial activities contribute to nutrient loading, organic pollution, and the accumulation of heavy metals in water bodies [41, 44, 46]. Biochars and hydrochars are particularly effective in this setting because of their high surface area, porous structure, and adjustable surface chemistry, which allow the adsorption of a broad range of contaminants, including pathogens, dissolved organic matter, and toxic metals [59, 68, 94, 111, 113, 115, 122].

Another crucial factor is Rwanda's reliance on *decentralized and small-scale water supply systems*, particularly in rural and remote communities. Many households depend on point-of-use treatment

methods, such as boiling, chlorination, or simple filtration [123, 124]. Biochar can be easily integrated into existing technologies such as biosand filters, ceramic filters, and community filtration units without requiring significant infrastructure upgrades or additional energy inputs. In addition, coffee washing stations could benefit from valorizing coffee processing by-products, such as coffee pulp and husk, by producing biochars and hydrochars, and treating the coffee processing wastewater.

The integration of biochar and hydrochar also closely supports Rwanda's national sustainability agenda, including initiatives backed by the Rwanda Green Fund [125]. Additionally, once saturated, these materials can often be reused as soil amendments, enhancing soil fertility and water retention. This creates vital links between water treatment, waste management, and agricultural productivity, strengthening an integrated resource management approach. Furthermore, adopting biochar-based water treatment systems offers opportunities for *local economic growth and job creation*. Small enterprises and cooperatives can participate in biomass collection, char production, and filter manufacturing, especially in rural areas where employment options are scarce. This promotes community ownership and boosts the chances of long-term sustainability.

Aims of the thesis

The effective removal of organic contaminants from wastewater remains a global challenge, especially in regions that cannot afford advanced treatment technologies. Although biochar and hydrochar have attracted attention as affordable, sustainable adsorbents, several knowledge gaps prevent their widespread practical use. Most research has been conducted under simplified conditions with single compounds in pure water solutions, which do not reflect the complexity of real wastewater containing multiple interactive contaminants. Additionally, the links between biochar's physicochemical properties and adsorption performance are not yet fully understood, particularly for materials produced using low-cost or decentralized technologies. The effects of production parameters, such as feedstock type and char conversion conditions, have also not been systematically assessed across a broad spectrum of contaminants. Furthermore, there is limited knowledge regarding the occurrence and variability of organic contaminants in water systems, particularly in resource-limited regions. Addressing these gaps, this thesis aimed to assess the potential of biochars and hydrochars derived from locally available feedstocks as adsorbents for organic contaminants in wastewater, and to identify the key factors influencing their adsorption performance through the following specific objectives:

- To screen and quantify pharmaceuticals, pesticides, and polyphenols in the Nyabugogo River, Rwanda, to assess their spatial and temporal concentration patterns, and to identify dominant contamination sources **(Paper III)**.
- To evaluate the ability of cookstove-derived biochars from three different feedstocks to remove pharmaceuticals and personal care products from authentic hospital effluent **(Paper I)**.
- To assess the adsorption efficiency of hydrochars and biochars derived from coffee processing by-products for the removal of polyphenols and pesticides from coffee processing wastewater, and to elucidate the underlying adsorption mechanisms **(Paper II)**.
- To systematically investigate how char conversion level and gasification atmospheres influence biochar structure, surface functionality, and adsorption performance across a chemically diverse range of organic contaminants **(Paper IV)**.

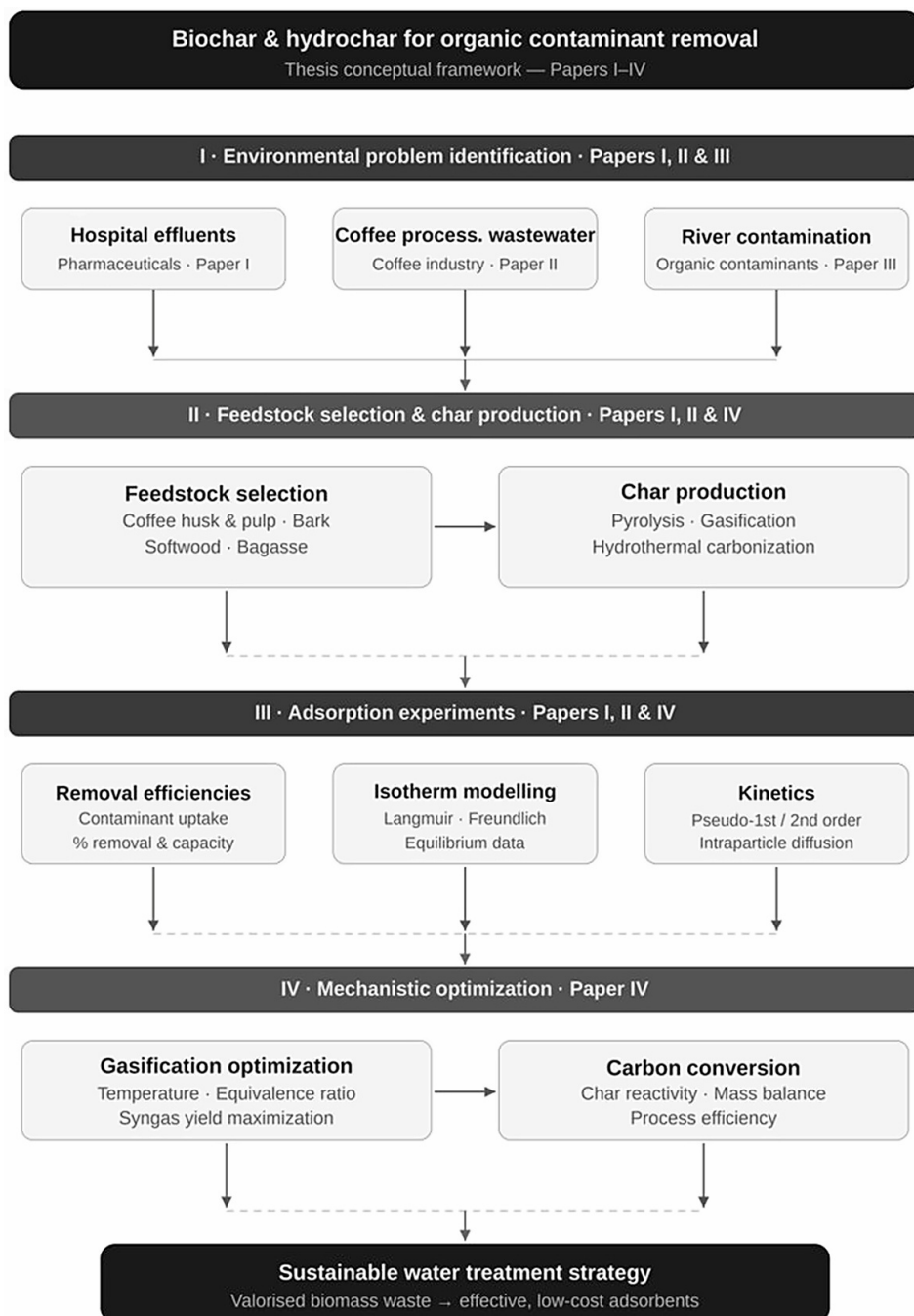


Figure 3. Conceptual illustration of projects conducted and presented in this thesis

2. Materials and methods

2.1. Chemicals

The pesticides investigated in this thesis represent a selection of the compounds most frequently applied in coffee production, including acetamiprid, azoxystrobin, carbendazim, cyproconazole, dimethoate, imidacloprid, metalaxyl, tebuconazole, and triadimefon. In addition to pesticides, polyphenols naturally present in coffee, including caffeic, chlorogenic, and ferulic acids, as well as catechin, were also examined. Figure 4 presents the molecular structures of some important pharmaceuticals, pesticides and polyphenols analysed in **Papers I-IV**. Internal standards, pesticide standards, and pharmaceutical standards used in this thesis were of analytical-grade purity (>95%). Twenty-eight chemicals in **Paper I**, 12 in **Paper II**, 158 in **Paper III**, and 145 in **Paper IV** were investigated, respectively, and all analytes are used in sub-Saharan countries. Stock solutions of each compound were prepared with methanol and kept at $-18\text{ }^{\circ}\text{C}$. Methanol and acetonitrile of HPLC grade were provided by Fisher Chemicals in Loughborough, UK, to create standard solutions, and LC-MS grade methanol (LiChrosolv) was provided by Merck in Darmstadt, Germany. Formic acid (Fluka), which was utilized as an additive in the mobile phase, was sourced from Sigma-Aldrich in Steinheim, Germany. Ultrapure water was produced using a Merck Millipore Advantage A10 system equipped with a Q-Pod unit.

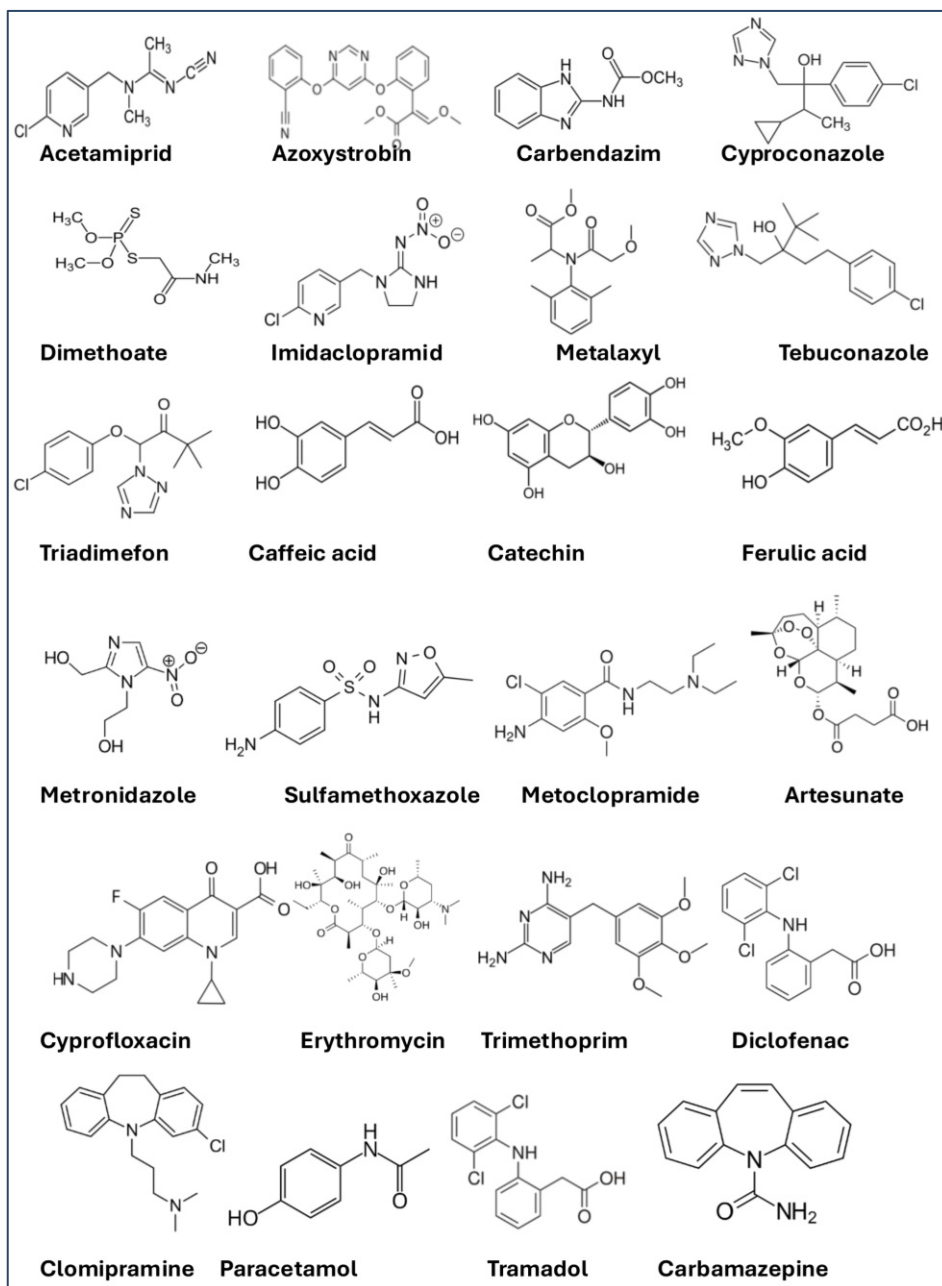


Figure 4. Molecular structure of some of the organic chemicals assessed in Papers I-IV.

2.2. Study area descriptions and sampling

Hospital wastewater samples for **Paper I** were collected from the University Teaching Hospital of Kigali (CHUK), the largest referral hospital in Rwanda, located in Nyarugenge District in central Kigali City. Wastewater generated from various hospital services is first directed into enclosed tanks for anaerobic treatment, after which the partially treated effluent is transferred to an open tank for aeration and chlorination. The final effluent is discharged into the adjacent canal, which flows into the Mpazi stream before joining the Nyabugogo River. For **Paper II**, coffee processing wastewater and coffee pulp and husks were collected at a coffee washing station in Rwanda and used in adsorption experiments. Water samples for screening for pharmaceuticals, pesticides, and polyphenols were collected from the Nyabugogo River (**Paper III**). This river is approximately 42.7 km long, extending from the outflow of Lake Muhazi to its confluence with the Nyabarongo River [126]. The Nyabugogo River passes through rural, peri-urban, and urban zones, each with distinct land uses. Rural areas are dominated by agricultural production and small-scale mining, while peri-urban areas are characterized by quarrying, local industries, markets, and health facilities. Urban areas are characterized by numerous industries concentrated in Kigali City, along with food markets, hospitals, schools, and other facilities. The Nyabugogo River crosses the Nyabugogo wetland, a major drainage sink that receives stormwater runoff, solid waste, sediments, and both treated and untreated wastewater from surrounding residential and industrial areas. The river is fed by multiple tributaries and channels, such as the Mpazi stream, which drains densely populated discharges. For **Paper IV**, wastewater used in adsorption experiments was collected from the Umeå wastewater treatment plant (WWTP) in Sweden.

For Paper I, influent and effluent wastewater from the CHUK treatment plant was collected over three days within one week. Samples were collected twice a day, with three subsamples collected at 30-minute intervals and pooled into composite influent and effluent samples, yielding six composite samples in total, which were stored frozen until analysis.

For Paper II, coffee processing wastewater, pulp, and husks were taken from a coffee washing station. **For Paper III**, 45 sites were sampled along

the Nyabugogo River system, including 26 river sites, ten tributaries, and nine channels spanning rural to urban areas. For **Paper IV**, wastewater was collected from the effluents from Umeå WWTP. Water was collected at 15-60 cm below the surface using polyethene bottles prerinsed with samples. After collection, water was kept in cooler boxes at approximately 4 °C during transport to the University of Rwanda and stored at -18 °C before analysis at Umeå University. For **Paper III**, additional sampling at site N21 was conducted over five consecutive days at 8 a.m., 1 p.m., and 5 p.m. to monitor the variability in the concentrations of pharmaceuticals, pesticides, and polyphenols.

2.3. Biochar and hydrochar preparations

Biochars and hydrochars were produced from forestry and agro-industrial residues using different thermochemical conversion techniques. In **Paper I**, softwood (SW), bagasse (BG), and coffee husks (CH) were pelletized into 8 mm pellets and converted into biochars using three improved TLUD gasifier cookstoves: two natural-draft stoves (N1 and N2) and one forced-draft stove (F). The stoves, as shown in Figure 5, varied in their airflow characteristics: N2 generated a stronger natural draft due to its longer combustion zone, whereas the forced-draft stove used a fan to control airflow. Each combination of feedstock and stove produced a distinct type of biochar, yielding nine distinct biochars (N1SW, N1BG, N1CH, N2SW, N2BG, N2CH, FSW, FBG, and FCH).

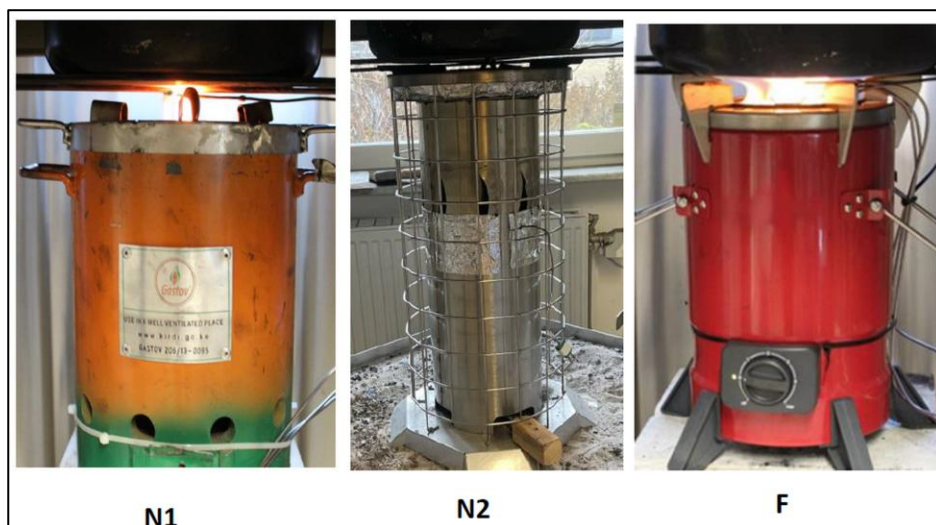


Figure 5. Cookstoves that produced biochars used in Paper I. N1 and N2 are natural-draft stoves, and F is a forced-draft stove.

In Paper II, coffee pulp and coffee husks collected from a washing station in Rwanda were processed into hydrochars through hydrothermal carbonization at 180 °C or 250 °C for four hours in sealed reactors with added water, followed by filtration, washing, drying, grinding, and sieving to 0.5 mm. Subsequent pyrolysis of these hydrochars at 600 °C and 800 °C for 30 seconds generated the biochars, which were homogenized to 0.5 mm. Hydrochars were also produced from coffee husk by first adding water to moisten it. Biochars from coffee husk were produced at 600 °C and 800 °C as well. In total, this process produced four hydrochars and six biochars derived from both coffee pulp and husks. Together, these methods generated a diverse set of chars suitable for evaluating their adsorption performance.

In Paper IV, biochars were produced from pine bark and coffee husk pellets using a laboratory-scale thermogravimetric analyser (TGA) equipped with continuous mass monitoring and a rapid nitrogen-quenching system. Pellets were heated to 800 °C under either a gasification atmosphere (65% N₂, 20% CO₂, 15% H₂O) or pure N₂ for pyrolysis, then quenched at specific stages of char conversion. Quenching times were determined by identifying the devolatilization endpoint, measuring ash content, and constructing a conversion curve from replicate mass-loss data. Samples were collected at 0%, 10%, 25%, and 50% conversion (or equivalent times in N₂), with multiple replicates to obtain sufficient biochar for analysis, yielding a total of 14 distinct biochar samples: seven from bark and seven from coffee husks, CCo, CC10, CC25 and CC50 in a gasification atmosphere, and CCo_N₂, CC10_N₂, and CC25_N₂ in nitrogen. Table 2 summarizes the feedstocks, the production conditions, and the biochars and hydrochars discussed in this thesis.

Table 2: Feedstocks, production conditions and biochars and hydrochars used in studies

Feedstock	Cookstove	HTC		Pyrolysis		Gasification	
		180°C	250°C	600°C	800°C	Gas	N2
Bagasse	N1, N2, F	-	-	-	-	-	-
Softwood	N1, N2, F	-	-	-	-	-	-
Coffee pulp	-	CP180	CP250	CP600 ^a CP600 ^b	CP800 ^a CP800 ^b	-	-
Coffee husk	N1, N2, F	CH180	CH250	CH600	CH800	CC0-CC50	CC0-CC25
Bark						CC0-CC50	CC0-CC25

N1, N2 and F are cookstoves with which N1SW, N2SW and FSW biochars are produced from softwood, N1BG, N2BG and FBG are made from bagasse, while N1CH, N2CH and FCH are coffee husk-derived biochars (Paper I). CP180 and CP250 are hydrochars made from coffee pulp at 180 °C and 250 °C, respectively. CH180 and CH250 are hydrochars made from husk at 180 °C and 250 °C, respectively. CH600 and CH800 are coffee husk-derived biochars produced at 600 and 800 °C, respectively. CP600^a and CP800^a are biochars produced by pyrolyzing coffee pulp-derived hydrochars produced at 180°C at 600 and 800°C, whereas CP600^b and CP800^b are biochars produced from coffee pulp hydrochars produced at 250°C (Paper II). CC0-CC50 are biochars produced during gasification at different degrees of char conversion. CC0-CC25 are their corresponding biochars produced in a nitrogen atmosphere (Paper IV).

2.4. Surface characterization of biochars and hydrochars

Characterizing the surfaces of biochar and hydrochar is critical for determining their physicochemical properties and potential applications. The materials were examined in this thesis utilizing a combination of spectroscopic and adsorption-based techniques to determine their functional groups, surface chemistry, and textural characteristics. Diffuse Reflectance Infrared Fourier Transform Spectroscopy (DRIFTS) was used to identify surface functional groups (**Papers I, II and IV**). Samples were mixed with spectroscopic-grade KBr and recorded under vacuum using a Bruker IFS 66 v/S apparatus. Complementary structural information was obtained by Raman spectroscopy using a Bruker Bravo device; the spectra were then trimmed, normalized, and smoothed. To determine the elemental composition on the biochar and hydrochar surfaces, X-ray photoelectron spectroscopy (XPS) was carried out using a Kratos Axis Ultra DLD spectrometer, with energy calibration referenced to the C1s peak at 285 eV (**Papers I and II**). Additionally, the specific surface area, pore volume, and pore-size distribution of the biochars and hydrochars were determined from nitrogen adsorption-desorption isotherms using the Brunauer-Emmett-Teller (BET) method (**Papers I,**

II and IV). Together, these analytical approaches provide a comprehensive understanding of the surface characteristics that influence the performance of the biochar and hydrochar.

2.5. Adsorption experiments (Papers I, II and IV)

Adsorption experiments were performed to determine the removal of pharmaceuticals from hospital effluents and of pesticides and polyphenols from coffee processing wastewater (CPWW) using biochars and hydrochars under controlled laboratory conditions. All experiments were performed in triplicate using plastic tubes, with adsorbent masses of 25 mg, and sample volumes of 5 mL. The adsorbent was added to each tube containing wastewater. The mixtures were subsequently agitated at room temperature (approximately 22°C) for 30 minutes in the **Paper I** experiments with hospital effluents and for 1 hour in the **Papers II and IV** experiments. Following agitation, samples were centrifuged at 4000–4500 rpm for 5 to 15 minutes. The resulting supernatants were transferred into vials, internal standards were added, and all samples were analysed within 24 hours using LC-MS/MS. Quality assurance and control procedures included the preparation of triplicate blank samples containing Milli-Q water without adsorbent, and tube-wall adsorption tests in which analytes were spiked into Milli-Q water at either 500 ng/L (**Papers I and IV**) or 1 µg/L (**Paper II**) in the absence of adsorbent. Additional control samples, consisting of hospital effluents, CPWW, or Umeå WWTP effluents, were agitated under identical conditions without an adsorbent to account for analyte losses unrelated to adsorption processes. Removal efficiency (R%) was calculated using equation (1).

$$R\% = \frac{(C_o - C_f)}{C_o} \times 100 \quad (1),$$

C_o and C_f were initial and final concentrations in ng/L or µg/L.

For adsorption experiments in **Papers II and IV**, solutions of targeted compounds were prepared by spiking CPWW with a mixed stock solution at 40 µg/L and 1 µg/L for the initial adsorption tests using 6 biochars and 4 hydrochars, and 14 biochars, respectively. Based on removal efficiency outcomes, a single hydrochar (CP250) was selected for further kinetic and isotherm investigations (**Paper II**). For these experiments, solutions at concentrations of 10, 30, 50, 75, and 100 µg/L were prepared, and 75 mg of CP250 was added to 5 mL of each solution. Supernatants were collected at 5, 10, 20, 30, 60, 90, and 120 minutes to evaluate adsorption kinetics. Additional experiments using Milli-Q water were performed to examine potential changes in the surface chemical functional groups of CP250 before and after adsorption. The adsorption capacities at equilibrium and

at time t , q_e (mg/g) and q_t (mg/g), were calculated using standard adsorption equations 2 and 3, respectively.

$$q_e = \frac{(C_o - C_e) \times V}{M} \quad (2)$$

$$q_t = \frac{(C_o - C_t) \times V}{M} \quad (3)$$

C_o (mg/L), C_e (mg/L), and C_t (mg/L) refer to the initial, equilibrium, and final concentrations, respectively. V (L) represents the volume of the solution, while M (g) denotes the mass of the adsorbent.

2.6. Sample Pre-treatment and LC-MS/MS analysis

Pretreatment of water samples analysed in this thesis was conducted using solid-phase extraction (SPE) prior to liquid chromatography tandem mass spectrometry (LC-MS/MS) analysis. In **Paper I**, samples were thawed at room temperature, centrifuged (3500 rpm, 5 min), and a 5 mL aliquot of each sample was spiked with 5 ng of internal standards. Automated online SPE was performed using a Dionex UltiMate 3000 UHPLC system (Thermo Scientific) equipped with quaternary and binary pumps and an autosampler. Samples were injected onto a 1 mL loop and loaded onto an Oasis HLB SPE column (2.1 × 20 mm, 15 μm), after which retained analytes were transferred to a Hypersil GOLD analytical column (50 × 2.1 mm, 5 μm) utilizing a gradient of 0.1% formic acid in Milli-Q water and acetonitrile. Chromatographic separation was performed at 25 °C. Detection was achieved using a TSQ Quantiva triple quadrupole mass spectrometer (Thermo Scientific) that features a heated electrospray ionization source functioning in positive mode. Quantification was conducted in selected reaction monitoring mode, using one transition for quantification and a second for confirmation. For papers **II**, **III** and **IV**, solid-phase extraction (SPE) was performed using Oasis HLB cartridges (Waters, Milford, MA, USA) mounted on a vacuum SPE manifold. Methanol (LC-MS grade) and Milli-Q water were used for cartridge conditioning, sample loading, and elution. Prior to extraction, samples were spiked with 5 ng of isotopically labelled internal standards and mixed thoroughly. HLB cartridges were conditioned with 5 mL of methanol followed by 5 mL of Milli-Q water to activate the sorbent. Samples were loaded to the cartridges at a low flow rate of approximately 0.1 mL /s to ensure efficient retention of analytes on the stationary phase. Elution of analytes was carried out with 5 mL of methanol. The eluates were evaporated to approximately 25 μL and reconstituted with methanol to a final volume of 1 mL prior to LC-MS/MS analysis.

Peak identification was performed using Xcalibur™4.3 (Thermo Fisher Scientific) by matching sample signals to their corresponding internal standards. A calibration curve of seven points was used for quantification. To make a calibration curve, concentrations ranged from 10 to 1000 ng/L for **Papers I, III and IV**, and from 1 to 100 µg/L for **Paper II** were utilized. Linearity assessment and quantification were carried out using these calibration intervals.

2.7. Quality control and quality assurance

To detect potential contamination and carry-over of samples, procedural and instrumental blanks were injected at regular intervals throughout the analytical sequence. Carry-over was further evaluated by injecting standards at 500 ng/L, followed by three injections of mobile-phase blanks. Quantification was based on comparing two transitions for each analyte, ensuring that their transition ratio remained within ±30% of the ratio observed in the corresponding calibration standard. The retention times of the analytes had to be within ±2.5% of those observed in the calibration standards. The limit of quantification (LOQ) was set as the second-lowest concentration within the linear range of the calibration curve.

2.8. Adsorption models

To elucidate the adsorption behaviour of the studied system in **Paper II**, several isothermal and kinetic models were applied. The experimental data were assessed using the Langmuir, Freundlich, and Temkin isotherms, while the kinetic characteristics were examined using the pseudo-first order (PFO), pseudo-second order (PSO), and intraparticle diffusion models.

2.8.1. Isotherm models

Langmuir isotherm

The Langmuir isotherm describes adsorption occurring at specific homogeneous sites on the surface of an adsorbent, ultimately forming a monolayer coverage. This model assumes uniform adsorption energies and no interaction between adsorbed molecules [127]. The Langmuir equation (4) was used to determine model parameters.

$$q_e = \frac{K_L q_{max} C_e}{1 + K_L C_e} \quad (4)$$

In this model, C_e (mg/L) represents the equilibrium concentration of the adsorbate, while q_e and q_{max} (mg/g) denote the equilibrium adsorption capacity and the maximum monolayer adsorption capacity, respectively. The constant K_L (L/mg) reflects the affinity between the adsorbent and the adsorbate. Fitting the experimental data to this model allowed quantification of maximum adsorption capacity and adsorption strength.

Freundlich Isotherm

The Freundlich isotherm (Equation 5) is an empirical model commonly employed to characterize adsorption on heterogeneous surfaces. It assumes multilayer adsorption and variable adsorption energies across different sites [128, 129].

$$q_e = K_F C_e^{1/n} \quad (5)$$

In this model, q_e (mg/g) is the equilibrium adsorption capacity and C_e (mg/L) is the equilibrium concentration. The constants $K_F[(\text{mg/g})(\text{L/mg})^n]$ and n indicate adsorption capacity and adsorption intensity, respectively. The values of these parameters were derived from the slope and intercept of the graph plotting $\log q_e$ versus $\log C_e$. A value of $1/n < 1$ typically signifies favourable adsorption.

Temkin Isotherm

The Temkin isotherm accounts for interactions between adsorbate molecules and presumes that the heat of adsorption decreases linearly with increasing surface coverage. By neglecting extremely low and high concentration ranges, the Temkin model (Equation 6) provides a more realistic description of adsorption energies.

$$q_e = B_T \ln(A_T C_e) \quad (6)$$

In this model, A_T denotes the Temkin binding constant, B_T refers to the heat of adsorption, and q_e and C_e represent the equilibrium adsorption capacity and concentration, respectively.

2.8.2. Kinetic Models

Pseudo-First Order and Pseudo-Second Order

Kinetic data were analysed utilizing the pseudo-first order and pseudo-second order models, which are expressed by Equations (7) and (8). These models describe the progression of adsorption with time (t),

$$\text{Pseudo-first order: } q_t = q_e(1 - e^{-k_1 t}) \quad (7)$$

$$\text{Pseudo-second order: } q_t = \frac{q_e^2 k_2 t}{1 + k_2 q_e t} \quad (8)$$

where k_1 and k_2 represent the rate constants for the PFO and PSO, respectively, while q_t and q_e denote the adsorption capacities at time t and at equilibrium. The performance of the models was investigated utilizing the coefficient of determination, R^2 , as a measure of best fit.

Intraparticle Diffusion Model

To further investigate mass transfer processes, the intraparticle diffusion model based on the Weber–Morris equation (Equation 9) was applied. This model analyzes the correlation between adsorbate uptake and the square root of contact time ($t^{1/2}$) [130].

$$q_t = K_{id} t^{1/2} + C \quad (9)$$

The parameter K_{id} ($\text{mg/g}/\text{min}^{1/2}$) represents the intraparticle diffusion rate constant, while C (mg/g) reflects the boundary-layer effect. For each initial concentration, q_t was plotted against $t^{1/2}$. The values of K_{id} and C were determined utilizing the slope and intercept of the linear sections. The assessment of fit, using the coefficient of determination (R^2), indicated whether the rate-limiting phase was intraparticle diffusion.

2.9. Statistical analysis

Statistical analyses were performed to evaluate variability in contaminant concentrations and differences in adsorption performance across experimental conditions. Descriptive statistics, including mean (\bar{A}_v), standard deviation (SD), and relative standard deviation (RSD), were calculated to summarize the data (**Papers I-IV**).

Inferential analyses were conducted at a significance level of $\alpha = 0.05$. Analysis of variance (ANOVA) was applied to assess statistically significant differences between groups. One-way ANOVA was used to compare removal rates of biochars produced from different feedstocks (**Papers I and II**) within the same cookstove and across different cookstoves from the same feedstock. In **Paper III**, independent t-tests and ANOVA were performed to evaluate differences in contaminant concentrations across spatial categories (rural, peri-urban, and urban) and temporal factors (time of day and sampling days). In **Paper IV**, a two-way ANOVA based on an ordinary least squares (OLS) model was employed to assess the effects of char conversion degree, gasification atmosphere, and their interaction on adsorption efficiency. The model included both main effects and interaction terms, and significance was evaluated using Type II sums of squares; F-statistics and corresponding p-values were reported. The relationship between the compound hydrophobicity (log Kow, obtained from PubChem and EPISuite) and adsorption behaviour was assessed using multiple linear regression, with log Kow as the dependent variable and biochar-specific removal efficiencies as predictor variables.

3. Results and discussions

This chapter presents key findings from 4 research projects conducted during these studies, along with a discussion and comparison with reported studies. The detailed results are outlined in the original **Papers (I-IV)**.

3.1. Surface characterization of biochars and hydrochars

Surface characterization was performed on biochars produced using three types of household cookstoves, hydrochars, biochars derived from coffee pulp and husks, and biochars produced by gasification from bark and coffee husk. To assess functional groups, carbon structure, elemental composition, and surface properties, a variety of analytical methods were used, including DRIFTS, Raman spectroscopy (**Papers I, II and IV**), X-ray photoelectron spectroscopy (**XPS**) (**Papers I and II**), and Brunauer–Emmett–Teller (BET) analysis (**Papers I, II and IV**). Together, these datasets provide a comparative understanding of how feedstock type and carbonization temperature govern the chemical and structural properties of carbonaceous materials.

3.1.1. DRIFTS and Raman analysis

Across all four studies, the DRIFTS and Raman results demonstrated several consistent spectral patterns and shared transformation pathways, despite differences in feedstock (bark, coffee husk, coffee pulp, softwood, bagasse), production method (cookstove, hydrothermal, pyrolysis, gasification), and temperature. One similarity between materials was the progressive loss of oxygenated and aliphatic functional groups and the accompanying growth of aromatic structures, a trend observed across all studies regardless of production technique. In **Papers I and II**, O–H stretching bands in the 2000–3500 cm^{-1} region (typically broad peaks at 3270–3520 cm^{-1}) and aliphatic C–H stretching bands at 2800–3000 cm^{-1} [68, 98] consistently weakened or disappeared with increasing temperature or char conversion. Also, carbonyl peaks around 1740 cm^{-1} , present in hydrochars and low-temperature biochars, were reduced systematically as carbonization intensified, as shown in Figure 6. This common reduction of oxygen-rich structures demonstrates dehydration, decarboxylation, and demethylation of feedstocks during thermal conversion [82, 95].

A second similarity is the consistent appearance and strengthening of aromatic features across all materials. In **Papers I, II, and IV**, aromatic

C=C stretching bands in the 1500–1600 cm^{-1} region intensified as temperatures increased or as char conversion progressed, indicating formation of condensed aromatic domains. In every study, peaks between 700–900 cm^{-1} , associated with aromatic ring deformation, were observed once the materials reached moderate temperature. These recurring spectral features demonstrate that aromatic ring formation is a core structural result of all carbonization pathways examined in the thesis, independent of feedstock or processing environment. The 1030–1260 cm^{-1} region, associated with mixed C–C, C–O, C–N, and C–X vibrations, was observed, although its intensity decreased with increasing temperatures. This suggests that mid-temperature chars retain a transitional mixture of partially degraded lignocellulosic structures before fully reorganizing into aromatic structures [95, 98]. Even cookstove biochars in **Paper I**, despite uncontrolled conditions, showed the same pattern in this region, indicating intermediate chemistry like that of biochars produced under more reproducible conditions in **Papers II and IV**.

The Raman spectra also showed notable shared patterns. In **Papers II and IV**, and implicitly in **Paper I**, where aromaticity was confirmed by DRIFTS, the D band at 1350 cm^{-1} and G band at 1580–1600 cm^{-1} showed an increase in D-band intensity (indicating greater disorder and defect formation) accompanied by the gradual sharpening of the G band (indicating growth of sp^2 -bonded aromatic carbon) [97, 98]. This same pattern of development appeared in both bark and coffee husk (**Paper IV**) and in the hydrochar-to-biochar transitions (**Paper II**), demonstrating that the change from mixed aliphatic–oxygenated structures to disordered, then increasingly ordered, aromatic domains was common throughout the thesis. While these similarities indicated shared underlying chemistry between biochars and hydrochars, the differences between feedstocks also showed consistent patterns. Coffee husk biochars in **Papers I, II, and IV** all showed strong mineral-related bands in the 900–400 cm^{-1} region, attributed to Si–O and metal–oxygen vibrations, confirming the inherently higher ash content of this feedstock, which was further highlighted across various production systems. The high ash content of coffee husk has been reported in various studies [62, 131–134]. Bark, softwood, and bagasse materials lacked these intense mineral peaks, reinforcing that feedstock chemical composition, rather than processing method, was the main factor influencing the mineral-dominated spectral characteristics.

The Gasification process (**Paper IV**) consistently accelerated the loss of O–H, C–H, and C–O bands and promoted more rapid increases in aromatic C=C and Raman D band intensity compared to nitrogen pyrolysis, as also seen by Bikane et al. [135]. This same acceleration of

aromatic development was implicitly observable in the biochars from **Paper II** produced at higher temperatures, which resembled the high conversion biochars of **Paper IV**.

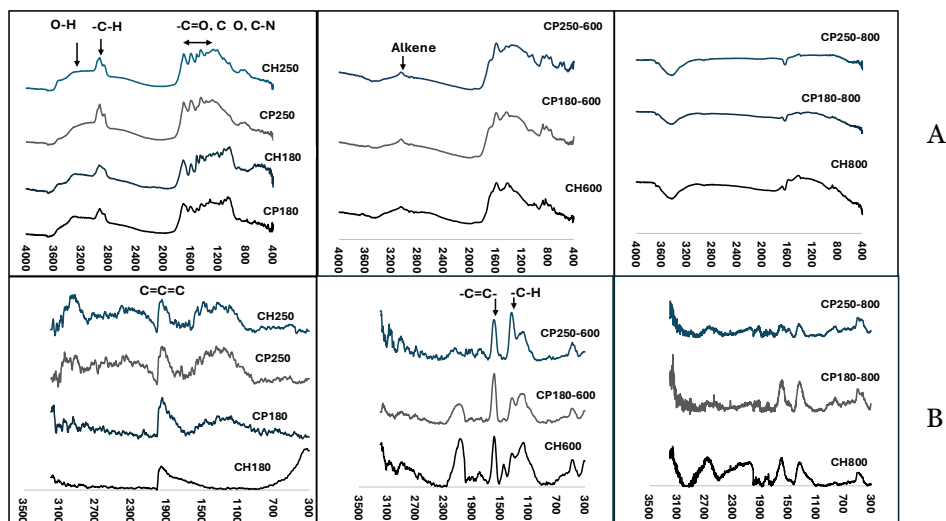


Figure 6. Spectra obtained from DRIFTS (A) and Raman (B). (A) displays the primary chemical functional groups, including O-H, C-H, C=O, C-O, and C-N, on the surfaces of hydrochars and biochars derived from coffee pulp at temperatures of 180 and 250°C (CP180, CP250, CH180, CH250), along with biochars produced at 600 and 800°C from both coffee pulp and husk (CP180–600, CP250–600, CP180–800, CP250–800, CH600, and CH800). (B) illustrates the intensities of the aliphatic C-H and aromatic C=C groups (**Paper II**)

3.1.2. X-ray photoelectron spectroscopy (XPS)

XPS analysis provided a detailed assessment of the elemental surface composition. Results showed that carbon content increased and oxygen content decreased with increasing carbonization temperature, as also indicated by DRIFTS spectra. In **Paper I**, softwood biochar had the highest carbon percentage, indicating a higher degree of carbonization compared with coffee husk and bagasse biochars. This was consistent with the pronounced aliphatic and aromatic peaks observed in the DRIFTS and Raman spectra for softwood.

In **Paper II**, hydrochars produced at 180°C exhibited relatively low carbon (72-78%) and high oxygen (20-26%) contents on their surfaces as indicated in Figure 7, reflecting a partially decomposed structure. Carbon content increased in biochars produced at 600°C and 800°C, reaching values above 79%, whereas oxygen levels decreased to below 8% at the

highest temperature, lowering the O/C ratios. These reductions in O/C ratios may indicate a progression towards hydrophobic and aromatic-dense biochars [63, 80]. The H/C ratios showed a similar trend; hydrochars exhibited higher H/C values, whereas high-temperature biochars displayed lower H/C ratios, consistent with greater aromatic condensation, as also reported by Al-Wabel et al. during the pyrolysis of cococarpus at different temperatures [136].

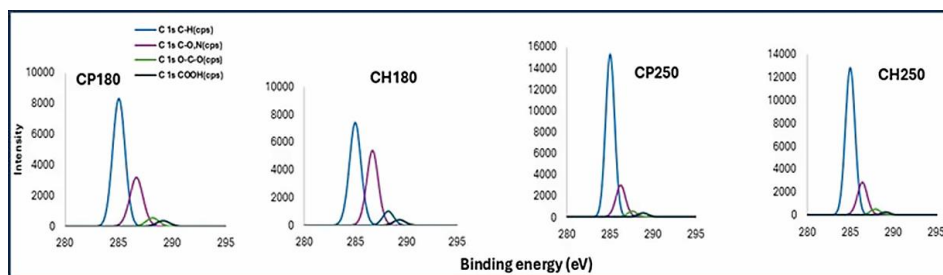


Figure 7. X-ray graphs of hydrochars adapted from **Paper II**. CP180 and CH180 are hydrochars produced at 180°C from coffee pulp and husk, respectively. CP250 and CH250 are hydrochars produced at 250°C from coffee pulp and husk, respectively

Minerals such as K and Ca were enriched in the coffee husk biochars, whereas Al and Si were characteristic of bagasse-derived biochars. In **Paper II**, K and Ca concentrations increased with temperature, indicating higher inorganic levels during pyrolysis. These minerals increased biochar pH [136, 137].

3.1.3. Surface area and porosity

BET analysis showed that biochars developed substantially higher specific surface areas and pore volumes than hydrochars, primarily because of carbonization and gasification. In **Papers I and II**, biochars showed higher surface areas (up to 289 m²/g in **Paper I** and 21-34 m²/g in **Paper II**), with biochars far exceeding hydrochars (4.7-15.7 m²/g). This increase in surface area was attributed to the removal of volatile components during carbonization, which generated new pores and exposed aromatic structures [65, 70, 90, 135]. **Paper IV** further demonstrated pronounced feedstock-dependent differences in pore development. Bark-derived biochars showed exceptionally high, progressively increasing surface areas (479-761 m²/g) and pore volumes, indicating robust formation of both micro- and mesopores during char conversion [90, 93, 135]. Coffee husk-derived biochars showed lower initial porosity and a rapid but early-plateauing increase in surface area (12-425 m²/g), suggesting limited structural stability for continued pore

development. Samples carbonized under N₂ showed minimal pore development, confirming that gasification reactions rather than heat alone drive porosity formation, as also argued by Pariyasamy et al. [138]. Figure 8 compares the surface areas of hydrochars and biochars derived from coffee pulp and husk, and bark for **Papers I, II and IV**.

Results show a clear increase in surface area with greater carbonization degree, progressing from low-temperature hydrochars (CH180, CP180) to intermediate-temperature pyrolysis biochars (CH600, CP-biochars), and reaching maximum values under a gasification atmosphere and char conversion.

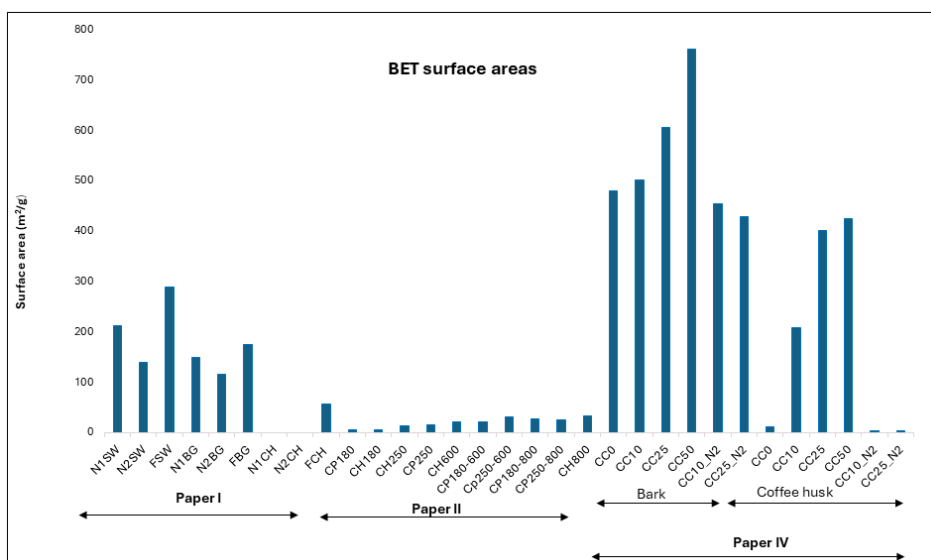


Figure 8. Comparison of BET surface areas of hydrochars and biochars from coffee pulp and husk produced in **Papers I, II and IV**. Samples include hydrochars generated at 180 °C and 250 °C, and biochars produced at 600 °C, 800 °C, and under gasification quenching conditions (CC0–CC50). Bars represent the measured BET surface area (m²/g) for each material.

While BET gas-phase surface area provides valuable information on the textural development and dry porosity of carbon materials, it does not necessarily represent the surface area that is functionally accessible under aqueous conditions. In water, pore accessibility is strongly constrained by pore hydration, competitive adsorption, and solvation effects, and many micropores that readily adsorb N₂ at -196 °C may be partially or completely inaccessible to water molecules or hydrated solutes [106, 139-141]. In addition, surface chemistry plays a dominant role in aqueous

systems, including functional groups, surface charge, ash content, etc., which can govern interactions independently of surface area [106, 140]. As a result, biochars with high BET surface areas do not necessarily correlate to higher sorption capacities in water, particularly for ions or larger organic molecules, as discussed in **Paper I**. Consequently, BET surface area alone should be regarded as a structural descriptor rather than a predictor of aqueous sorption behaviour, and its interpretation is best complemented by surface chemical analyses when evaluating sorption performance in aqueous environments.

3.2. Occurrence and distribution of organic contaminants in water systems in Rwanda

The results from both the hospital wastewater and Nyabugogo River studies showed a complex distribution of organic contaminants driven by various pollution sources. In the CHUK hospital wastewater, 28 pharmaceuticals and personal care products (PPCPs) were detected, dominated by analgesics/NSAIDs, antibiotics, antiretrovirals, antimalarials, and psycholeptics, with paracetamol, ciprofloxacin, abacavir, lignocaine, and caffeine showing the highest concentrations, as shown in Figure 9 (**Paper I**). Levels in the influent were 244,000 ng/L for paracetamol and remained substantial in the effluent, confirming hospitals as major point sources of pharmaceutical pollution [142, 143]. The high variability observed for several pharmaceuticals (up to 128% RSD) reflects fluctuating patient load, disease prevalence, and prescription patterns, which aligns with trends observed internationally [142, 144].

The Nyabugogo River demonstrated a much broader contaminant profile, with 57 pharmaceuticals across 21 ATC classes, along with pesticides and polyphenols originating from diverse urban, peri-urban, and rural activities (**Paper III**). Pharmaceutical concentrations in the river were 740 ng/L on average and a maximum of 4200 ng/L. The presence of compounds such as albendazole, paracetamol, triclosan, tetracycline, and nevirapine demonstrated significant diffuse contamination. Of the nine pesticides examined, five were detected, and all four polyphenols of interest were detected. Metalaxyl, a fungicide, was the pesticide detected at the highest average concentration of 70 ng/L and a maximum of 130 ng/L, which was within the range of 60-4800 ng/L previously reported by Houbraken et al. for surface waters in Rwanda [145]. This is attributed to the extensive agricultural use of metalaxyl in Rwanda, where it is applied alone or with other pesticides to control diseases in crops such as maize, potatoes, vegetables, and coffee, as described by Houbraken et al. [145].

Regarding polyphenols, caffeic acid was the most abundant compound, with an average concentration of 49 ng/L and a maximum of 160 ng/L. Spatially, concentrations of pharmaceuticals showed a pronounced increase in urban segments of the river, as shown in Figure 10, particularly at sites receiving untreated domestic wastewater, market runoff, industrial discharges, and effluents from the Mpazi tributary, before declining downstream due to dilution by the Yanze tributary and natural attenuation processes [146]. In contrast, pesticides and polyphenols did not follow a clear spatial trend, indicating strong hydrological mixing from multiple wastewater channels that homogenize their distribution along the river continuum, as indicated by Figure 11. Comparatively, while hospital wastewater contained extremely high concentrations of a limited number of clinically linked pharmaceuticals, the river reflected cumulative pollution from numerous sources, including large-scale albendazole administration campaigns, agricultural runoff, industrial waste (e.g., sugar-processing effluents introducing caffeic and ferulic acids), and disposal of food and plant-based residues from markets, restaurants, and processing facilities, as shown in figures 9-11 [120, 147, 148].

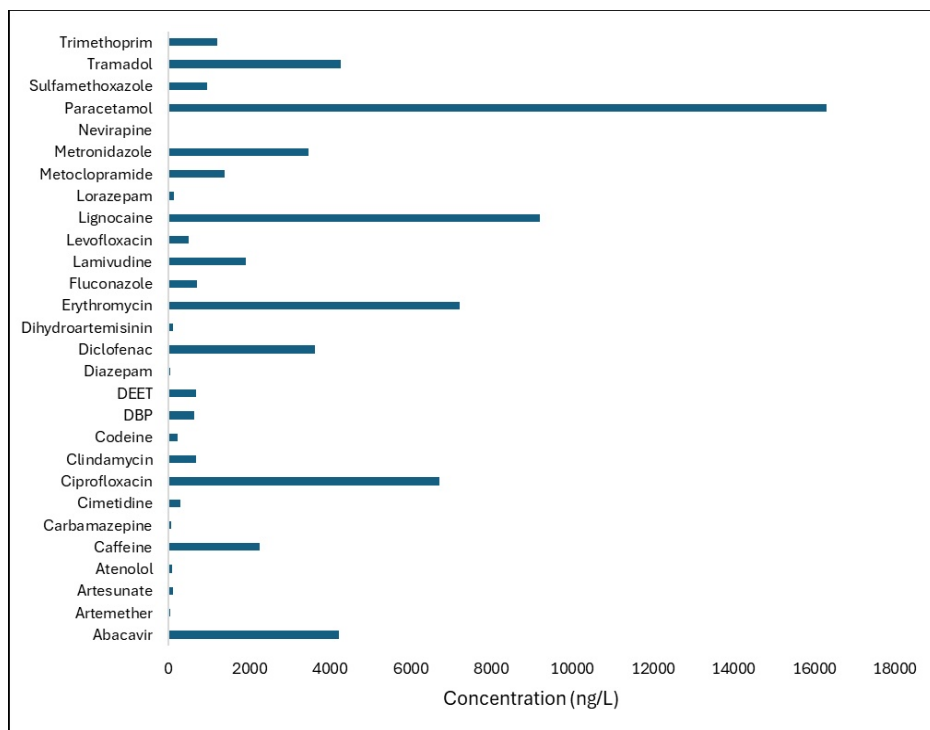


Figure 9. Pharmaceuticals and personal care products detected in hospital effluents and their concentrations (ng/L) (**Paper I**).

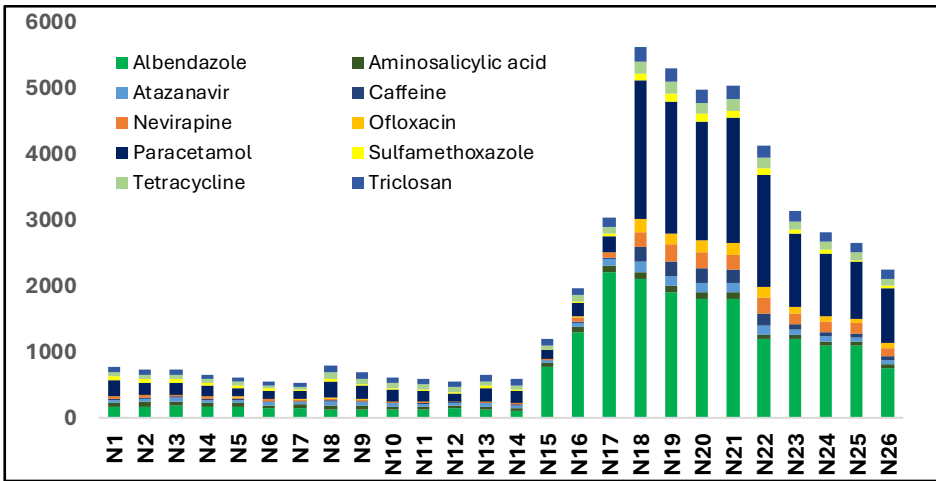


Figure 10. Concentration of the top ten pharmaceuticals for 26 sampling sites in the Nyabugogo River

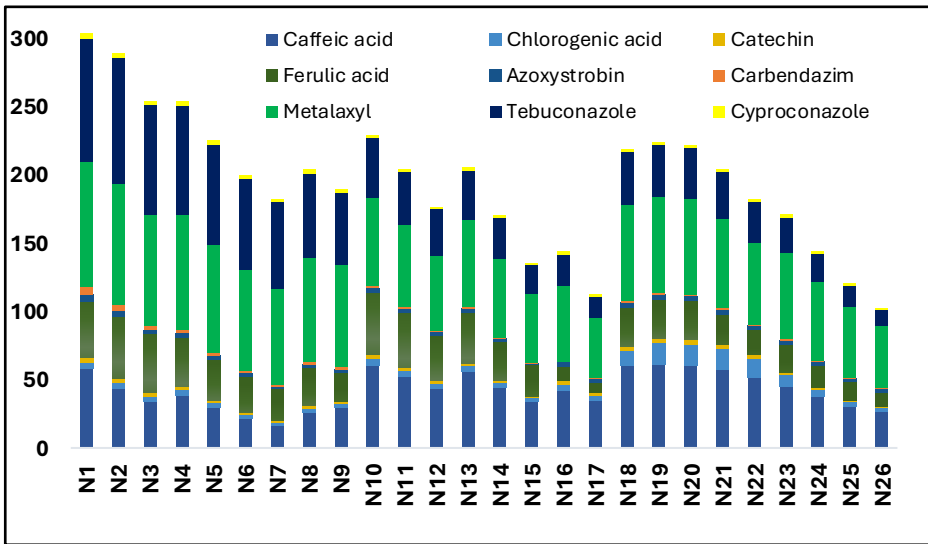


Figure 11. Concentration of pesticides and polyphenols in 26 sampling sites of the Nyabugogo River

These patterns demonstrate that pharmaceuticals are predominantly linked to urban wastewater discharges, whereas pesticides and polyphenols are influenced largely by agricultural activities and vegetation-related inputs in rural and peri-urban areas, with localized contributions from urban commercial and industrial sources [16, 45]. The results show the combined influence of land use, wastewater practices,

and human activities on contaminant variation across the Nyabugogo River system and highlight the need for management strategies that address both diffuse and point-source pollution.

3.3. Adsorption of organic contaminants by biochars and hydrochars

In three studies included in this thesis (**Papers I, II and IV**), biochars and hydrochars derived from various lignocellulosic and agro-industrial feedstocks such as softwood (SW), bagasse (BG), coffee husk (CH), coffee pulp (CP), and bark were assessed for their ability to remove organic contaminants from different wastewaters. The target contaminants included pharmaceuticals and personal care products (PPCPs) from hospital effluents (**Paper I**), polyphenols and pesticides from coffee processing wastewater (CPWW, **Paper II**), and a wide range of 137 organic contaminants, including pesticides, pharmaceuticals, and industrial chemicals (**Paper IV**). Together, these studies demonstrated that carbonaceous adsorbents showed broad removal capacity, while their efficiency was shaped by adsorbent structure and chemistry, production conditions, and contaminant physicochemical properties.

3.3.1. Removal efficiency of produced biochars and hydrochars

In **Paper I**, average removal rates (RRs) among nine SW, BG and CH-derived biochars ranged from 14.2% to 65.5%, with the five best-performing biochars (N1BG, FSW, FBG, N1SW, N2SW) exceeding 50% RR for at least 14 PPCPs. Metoclopramide was removed at RR > 95% by N1BG, FSW, and FBG; diazepam at RR > 56.6% by all nine biochars; and some antibiotics like ciprofloxacin, erythromycin, levofloxacin, clindamycin, and trimethoprim were removed at 54.4–93.3% by the top three biochars. Also, antibiotics, such as metronidazole and sulfamethoxazole, were removed at lower rates (25.9–43.6%), and artesunate was not detectably adsorbed by any biochar. Considering the hydrophobicity ($\log K_{ow}$) of analytes, a low coefficient of determination ($R^2 = 0.21$) and a non-significant outcome ($P=0.86$) from statistical analysis indicated that adsorption efficiency could not be reliably explained by $\log K_{ow}$ of analytes, emphasizing substantial unexplained variability resulting from the heterogeneity of the biochar properties and interactions specific to each analyte.

In **Paper II**, hydrochars preferentially removed polyphenols (caffeic acid, ferulic acid, catechin), while biochars were substantially more effective for pesticides (azoxystrobin, cyproconazole, tebuconazole, triadimefon, metalaxyl). The adsorption efficiency observed in **Paper II** indicates that

multiple interaction mechanisms govern the removal process, rather than hydrophobicity alone. This is supported by the regression analysis, which showed that log Kow had very limited predictive ability ($R^2 = 0.03$) and was not statistically significant at the 95% confidence level ($p = 0.08$). In contrast, the regression model improved when the type of adsorbent was included ($R^2 = 0.46$), indicating that the relationship between analyte hydrophobicity and adsorption efficiency varied depending on the adsorbents.

In **Paper IV**, removal rates across 14 biochars and 137 organic contaminants ranged from 10.4% to 100%. Statistical analysis using gasification biochars produced under varying char conversion levels and atmospheres showed that hydrophobicity cannot explain the variability in adsorption efficiency despite a highly significant regression model ($R^2 = 0.35$, $p < 10^{-6}$) and a wide log Kow range (-1.6 to >7) as seen in Figure 12. In the statistical analysis of biochar types, the significance of CC50 may have been due to its ability to remove almost all analytes, pushing results close to 100% and reducing data variability (a ceiling effect). This consistent removal pattern can affect the regression analysis, as described by Wang et al., who state that when many scores cluster near the maximum, the assumption does not hold, leading to reduced slopes and inaccurate predictions [149].

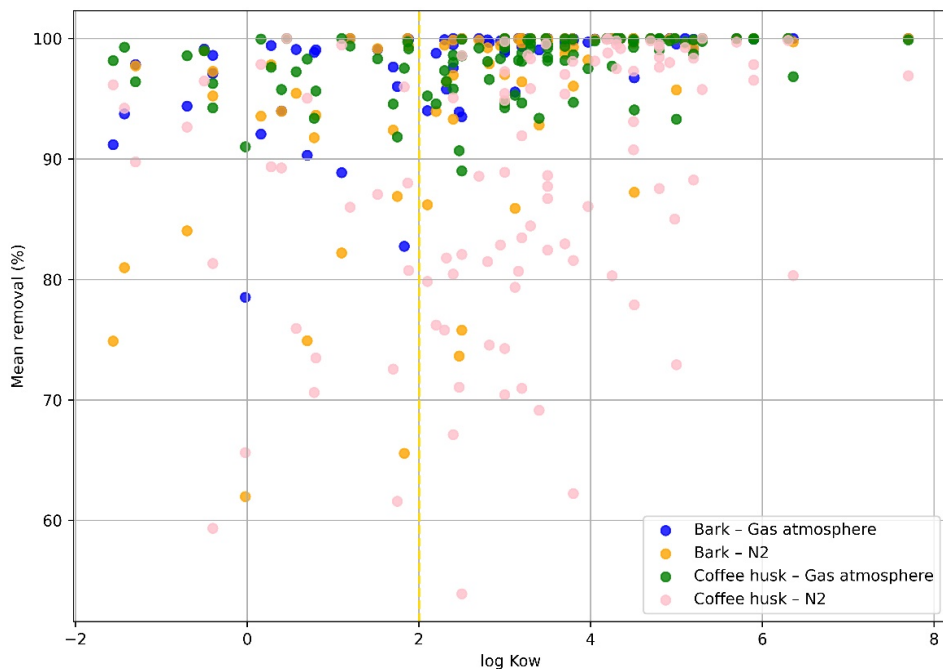


Figure 12. Removal efficiency of organic pollutants as a function of hydrophobicity (log Kow) for biochars produced under different gasification conditions. Bark biochar under gasification atmosphere, bark biochar under nitrogen atmosphere, coffee husk biochar under gasification atmosphere, and coffee husk biochar under nitrogen atmosphere. Each point represents an individual compound (137) (**Paper IV**).

Across all three studies, high and near-complete removals were observed for both hydrophilic and highly hydrophobic compounds within the same material categories, demonstrating that adsorption efficiency is primarily controlled by biochar structural development, surface functionality, pore accessibility, and char conversion conditions. Consequently, the collective statistical evidence from **Papers I, II, and IV** confirms that log Kow is insufficient as a standalone predictor of adsorption efficiency for biochars and hydrochars. Future research should use multivariate mechanistic models that combine adsorbate properties (charge, ash content, pKa, and size) with detailed adsorbent characteristics (surface chemistry, porosity, and production conditions) to better predict adsorption behaviour.

3.3.2. Adsorption Mechanisms

Several adsorption mechanisms operated concurrently in these studies for the removal of organic contaminants. π - π electron donor-acceptor interactions can explain the adsorption of aromatic compounds. DRIFTS

and Raman spectroscopy in **Paper I** confirmed that SW and BG-derived biochars were rich in aromatic carbon carrying excess π -electrons, enabling interactions with PPCPs containing electron-acceptor atoms (O, N, P, Cl, S) [85, 150]. The same mechanism can contribute to aromatic compounds undergoing π - π stacking on carbonized biochar surfaces in **papers II and IV**. The hydrophobic partitioning mechanism can explain the adsorption of organic chemicals onto biochars with low O/C ratios (< 0.2), as described by Zhao et al., during adsorption studies of atrazine on biochars, arguing that low O/C favoured hydrophobic mechanisms. [151]. The O/C ratio of biochars and hydrochars was calculated in **Papers I, II and IV**.

Hydrogen bonding can govern polyphenol adsorption onto hydrochars in **Paper II**: Hydrochars retain a high abundance of oxygen-containing functional groups, e.g., hydroxyl and carboxyl groups (see Figure 6 of the DRIFTS and Raman analysis), which promote adsorption through hydrogen bonding and electrostatic interactions, particularly for polar or ionizable compounds. Various studies reported that the strength of these interactions is strongly influenced by HTC conditions and pH, as higher carbonization temperature reduces oxygenated groups and elevated pH weakens hydrogen bonding through deprotonation, highlighting hydrogen bonding as a central mechanism controlling hydrochar reactivity and functionality [77, 86, 151, 152]. Pore filling and van der Waals forces can also contribute to adsorption, particularly for molecules with high molecular weight on high-surface-area biochars [98, 150, 153]. This mechanism may be involved in the adsorption described in **Paper IV**. The consistent contributions of multiple mechanisms confirm that biochar adsorption is inherently multi-factorial, with no single interaction governing removal efficiency.

In **paper II**, adsorption kinetics and isotherms were examined to identify the mechanisms governing the adsorption interactions of polyphenols and pesticides from CPWW with CP250, a hydrochar produced from coffee pulp at 250°C. Equilibrium modelling indicated that adsorption predominantly occurred on a heterogeneous surface, with the Freundlich model providing the best fit for most analytes. Kinetic modelling revealed a multi-stage adsorption process characterized by an initial rapid uptake driven by boundary-layer (film) diffusion, during which analyte molecules were quickly transferred from the solution to the external surface of CP250. This was followed by a slower intraparticle diffusion stage as molecules migrated into the hydrochar's pores. The applicability of both pseudo-first-order and pseudo-second-order models suggested that physical interactions, chemisorption, and surface reactions contributed to the overall rate behaviour, as indicated by the coefficients of

determination (R^2) for the models. In addition, the multi-linear trend of intraparticle diffusion plots, none passing through the origin as indicated in Figure 13 of caffeic acid and metalaxyl (one of the analytes assessed in **Paper II**), confirmed that intraparticle diffusion was not the sole rate-limiting step; instead, adsorption was controlled by a combination of film diffusion, pore diffusion, and gradual saturation of active sites.

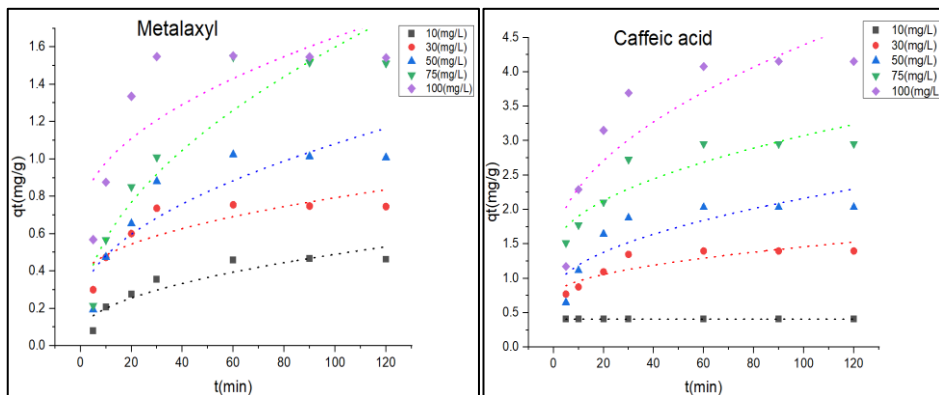


Figure 13. Intraparticle diffusion model graph for caffeic acid (polyphenol) and metalaxyl (pesticide) representing analytes adsorbed onto CP250 in **Paper II**. No graph passes through the (0,0) point. q_t is the adsorption capacity (mg/g), t is the time (min).

Collectively, these findings demonstrate that CP250 showed a kinetically complex adsorption process, governed by sequential transport steps and diverse interaction mechanisms consistent with its heterogeneous surface characteristics.

3.3.3. Influence of production conditions of biochars and hydrochars and contaminant hydrophobicity on removal efficiency

Across all studies in this thesis, the production conditions of the biochars and hydrochars proved decisive in shaping their adsorption performance. In **Paper II**, HTC at 180–250°C and pyrolysis at 600–800°C controlled the selectivity of the resulting adsorbents, with hydrochars maintaining oxygenated surface functionalities that favour polyphenols, while biochars developed hydrophobic, aromatic surfaces that favour pesticide adsorption. In **Paper IV**, both char conversion and gasification conditions exerted major effects. The transition from CCo to CC10 produced the largest improvement in adsorption efficiency for both feedstocks treated under a gasification atmosphere, rising from 90.5% to 99.5% for coffee husk and from 93% to 99.6% for bark, coinciding with a

sharp increase in BET surface area from 12 ± 4 to $208 \text{ m}^2/\text{g}$ for coffee husk. The abrupt increase in adsorption efficiency, therefore, reflected a threshold effect, in which early-stage char conversion dramatically enhanced the physicochemical properties of biochars.

Biochars produced under gasification atmosphere consistently outperformed N_2 -derived biochars, as shown in Figure 14. N_2 -derived coffee husk biochars showed declining efficiencies with increasing conversion (86.4% at CC_0 to 81.0% at CC_{25}), indicating that inert conditions limit or even degrade the development of adsorption-relevant surface properties.

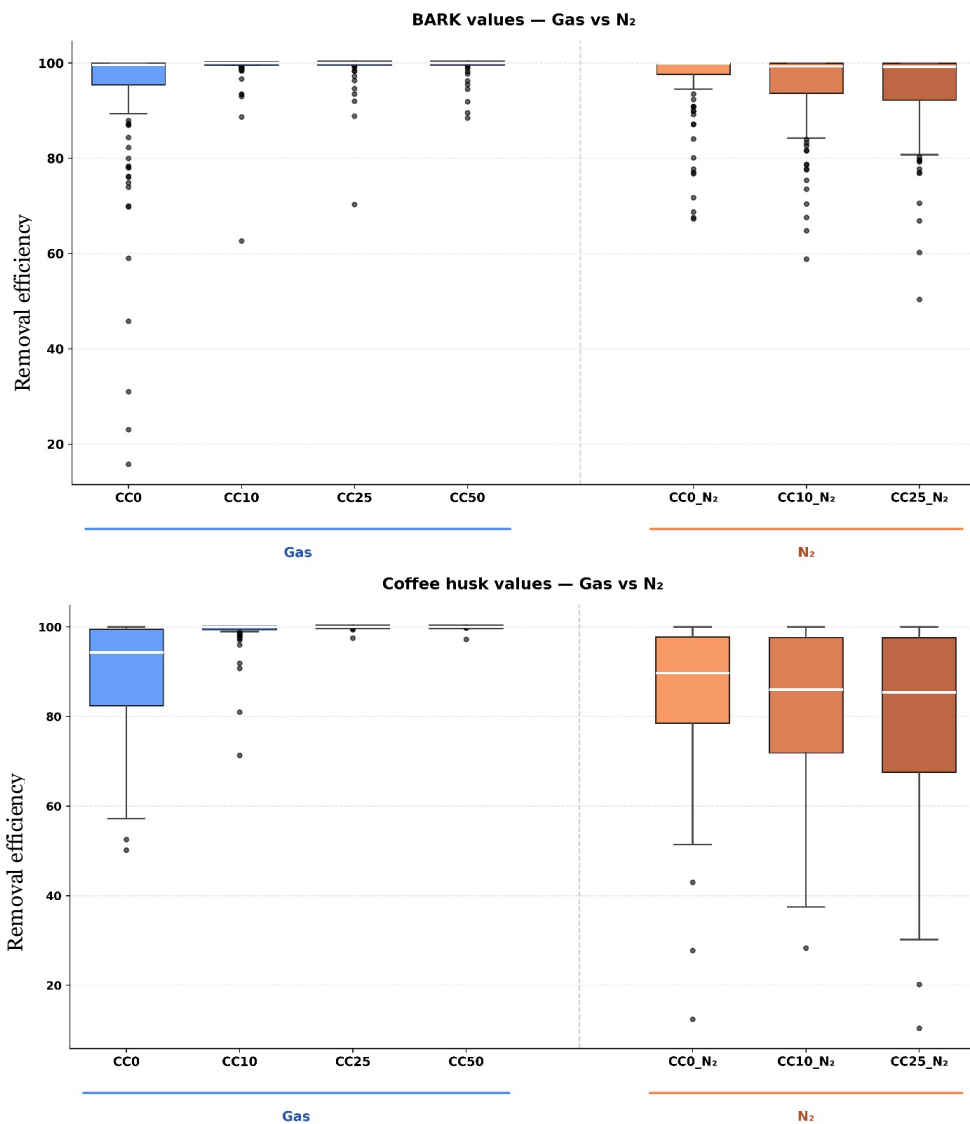


Figure 14. Distribution of removal efficiencies throughout all analytes (137) for bark-derived biochars (7) and coffee husk-derived biochars (7) produced by gasification and nitrogen atmospheres assessed in **paper IV**.

Surface area alone was not a universal predictor of removal performance. In **Paper I**, BET results showed N1BG and FSW with surface areas of $150 \pm 1.4 \text{ m}^2/\text{g}$ and $289 \pm 0.7 \text{ m}^2/\text{g}$, respectively, achieved comparable removal rates despite their large difference in surface area, suggesting that surface chemistry and specific interactions can compensate for structural differences, and that surfaces that are determined using BET have limitations, as discussed in the BET results section 3.1.3 of this thesis.

The role of contaminant hydrophobicity was similarly conditional; in **Paper II**, regression analysis showed that the removal efficiency was not associated with log Kow of contaminant ($p=0.08$); in **Paper IV**, a similar analysis showed $R^2 = 0.35$, $F = 4.66$ and $p < 10^{-6}$ only for bark biochar at CC50 under a gasification atmosphere. These values may be explained by a ceiling effect: this biochar removed almost all contaminants, achieving nearly complete removal, which influenced the analysis, as discussed in section 3.3.1 of this thesis. Under N₂, coffee husk biochars showed opposing log Kow relationships at CC10 (positive coefficient, $p = 0.02$) and CC25 (negative coefficient, $p = 0.02$), demonstrating the complexity of adsorption selectivity under inert conditions. Collectively, these findings showed that high removal efficiency requires a minimum threshold of structural development achievable at low char conversion under reactive conditions, and that adsorbent design, including gasification atmospheres, carbonization methods and feedstock selection, must be tailored to the target contaminant class.

3.3.4. Integrated discussion of adsorption performance

Across Papers **I**, **II** and **IV**, a consistent pattern emerged: the degree of char conversion and resulting structural development were the dominant factors controlling adsorption efficiency. Both high-temperature pyrolysis (**Papers I and II**) and the transition from low to higher char conversion during gasification (**Paper IV**) led to increased aromaticity, improved pore structure, and greater surface stability. These changes produced biochars capable of strong π - π interactions, hydrophobic partitioning, and pore-filling, which collectively favoured the adsorption of the assessed chemicals. Once biochars achieved sufficient structural development, such as at 600-800 °C in pyrolysis or CC10 and beyond in gasification, the influence of feedstock differences largely disappeared, indicating that thermal conversion was more decisive than biomass composition. These studies also demonstrated that surface chemistry often played a more critical role than surface area alone, with highly aromatic and chemically uniform biochars outperforming some materials with higher surface area, particularly when produced under reactive atmospheres that facilitated carbon restructuring.

At the same time, the findings demonstrated that not all sorbents benefit from extensive carbonization, particularly when targeting contaminants that rely on specific surface functionalities. Hydrochars, which retain significant oxygen-containing functional groups, performed exceptionally well for polyphenols in **Paper II**, where hydrogen bonding might play a major role in adsorption. This contrasted with their lower performance for some pesticides. Together, these results highlight that while biochars

engineered through a high degree of carbonization provided broad, high-efficiency adsorption for diverse contaminants, hydrochars offer distinct advantages for contaminants requiring polar interactions, such as phenolic compounds. The combined studies therefore emphasize that adsorption efficiency depends on the alignment between sorbent chemistry and contaminant properties, and that optimal water treatment strategies may require selecting or tailoring carbonized materials based on the specific functional groups and dominant interaction mechanisms relevant to each contaminant type.

4. Conclusion

This thesis demonstrated that biochars and hydrochars derived from locally available agro-industrial feedstocks, including softwood, bagasse, coffee husk, coffee pulp, and bark, can adsorb a range of organic contaminants from wastewater matrices. Screening for organic contaminants in the Nyabugogo River (**Paper III**) confirmed that urban Rwandan waterways are contaminated by pharmaceuticals, pesticides, and polyphenols, driven by inadequately treated and/or untreated wastewater discharges and agricultural runoff, highlighting the urgent need for affordable treatment solutions in the region. Across the adsorption studies (**Papers I, II, and IV**), production conditions, particularly the carbonization method, temperature, and gasification conditions, were important factors in determining removal performance. Surface chemistry was associated with adsorption efficiency, which was influenced by multiple co-existing mechanisms whose relative contributions shifted with both adsorbent and contaminant properties. These findings suggest that carbonaceous adsorbents derived from locally available residues have potential as tools for removing organic contaminants from real wastewater in resource-limited regions. Future research should focus on scale-up, regeneration, and integration with existing water treatment infrastructure in Sub-Saharan Africa to translate these results into practical environmental and public health benefits.

Coffee husk and coffee pulp served as common feedstocks throughout the thesis, demonstrating their versatility in producing adsorbents with distinct adsorption behaviours under different processing conditions. In low-temperature hydrothermal treatment (**Paper II**), these feedstocks yielded hydrochars rich in oxygenated functional groups, which can enable strong hydrogen-bond interactions and thus highly effective polyphenol removal. When subjected to higher temperature pyrolysis or gasification (**Papers I, II, and IV**), coffee husk produced biochars with greater aromaticity and pore development, resulting in high adsorption of chemicals, such as PPCPs and pesticides. Collectively, the studies show that coffee husk and pulp can be tailored through appropriate conversion pathways to target specific contaminant classes, reinforcing their value as abundant and flexible resources for sustainable water treatment applications.

Future perspectives

Paper III provided the first systematic multi-contaminant screening of the Nyabugogo River in Rwanda. However, the spatial and temporal scope of organic contaminant monitoring across East Africa remains very limited relative to the scale of the contamination problem. Future research should extend monitoring to other major Rwandan river systems, including the Akagera and Rusizi, and to lake systems that serve as drinking water sources for large populations. Cross-country comparative studies spanning Kenya, Uganda, Tanzania, and Ethiopia would enable the identification of regional contamination patterns and the dominant pollution drivers. Longitudinal monitoring over multiple years is needed to detect trends in contamination levels associated with population growth, changing agricultural practices, and expanding pharmaceutical consumption.

This thesis demonstrated that production conditions, carbonization method, temperature, and gasification atmosphere are among the most influential factors of adsorbent performance. However, the parameter space explored was necessarily limited. Systematic optimization studies using response surface methodology or machine learning approaches could identify optimal production conditions for specific contaminant classes or wastewater types. The potential of co-pyrolysis or co-hydrothermal carbonization of mixed feedstocks, combining, for instance, coffee husk with sewage sludge or bagasse with agricultural residues, could also be explored to produce adsorbents with synergistic properties.

All adsorption experiments in this thesis used authentic wastewaters, including real hospital effluent (**Paper I**), coffee processing wastewater (Paper II), and municipal wastewater treatment plant effluent (**Paper IV**), which was a significant strength of this work compared to studies relying on synthetic matrices. However, experiments were conducted under controlled laboratory conditions, with fixed contact times, controlled temperatures, and static batch systems. Future studies should therefore focus on transitioning from laboratory batch experiments to continuous-flow fixed-bed column systems using the same authentic wastewaters to evaluate breakthrough performance, bed exhaustion rates, and operational lifespan in environments that more accurately simulate real-world conditions. Pilot-scale field trials in Rwanda and neighbouring East African countries, ideally integrated with existing cookstove or gasification infrastructure, would be an important step towards real-world implementation.

While this thesis identified the dominant adsorption mechanisms involved in different adsorbent-contaminant interactions, the relative contribution of each mechanism under varying conditions was not always fully resolved. Advanced spectroscopic and computational approaches could provide deeper mechanistic insight. Techniques such as solid-state nuclear magnetic resonance (NMR) and synchrotron-based methods could reveal how specific functional groups interact with individual contaminant classes at the molecular level.

A critical but underexplored aspect of biochar and hydrochar application is their regeneration potential and performance over repeated use cycles. The economic and environmental sustainability of these materials depends heavily on whether they can be effectively regenerated through thermal treatment, solvent washing, chemical oxidation, or other means and reused without significant loss of adsorption capacity. Future work should evaluate regeneration efficiency across multiple cycles for the best-performing biochars identified in this thesis, especially those produced using gasification, and assess changes in surface chemistry, pore structure, and functional group composition following regeneration. The safe disposal or valorization of spent adsorbents loaded with organic contaminants is an equally important consideration, particularly in low-income settings where hazardous waste management infrastructure is limited.

Biochar and hydrochar adsorption alone are unlikely to achieve complete removal of all contaminants in complex wastewaters. Combining adsorption with complementary treatment processes offers a promising pathway to more robust, comprehensive contaminant removal. Potential synergistic combinations include biochar filtration coupled with constructed wetlands, which are already used in several East African countries and could benefit from enhanced contaminant interception at the inlet stage. Solar-driven advanced oxidation processes, including photocatalysis and solar photo-Fenton reactions, are particularly attractive for tropical regions, given the high solar irradiance available and could be used downstream of adsorption to mineralize desorbed or residual contaminants. Biological treatment systems incorporating biochar as a carrier medium for microbial biofilms represent another possibility, as biochar has been shown to enhance microbial diversity and activity in bioreactors. Evaluating these integrated systems under field conditions in Rwanda and neighbouring countries should be a priority for applied future research.

Beyond the scientific dimensions, the findings of this thesis have important implications for environmental policy and public health

governance in Rwanda and the broader East African region. The detection of pharmaceuticals, including antibiotics, antiparasitics, and psychoactive compounds in the Nyabugogo River at concentrations that may exert ecological effects underscores the need for national monitoring programmes, effluent discharge standards for organic compounds, and stronger regulation of agricultural pesticide use. Future interdisciplinary research connecting environmental chemists, public health experts, engineers, and policymakers is needed to translate scientific findings into regulatory action. Capacity building initiatives, including training of local scientists in advanced analytical techniques such as LC-MS/MS, and establishment of regional reference laboratories, are essential to ensure that monitoring and research efforts can be sustained beyond individual thesis projects. Engaging local communities, farmers, healthcare workers, and water utility managers in the design and implementation of treatment solutions will be critical to ensuring their adoption and long-term success.

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