VUDP PROJEKTRAPPORT

Aktiv kul fremstillet af spildevandsslam og plantemateriale

VUDP
FORENINGEN

Vandsektorens forening til forbedring af vandsektorens effektivitet og kvalitet

FEASIBILITY OF REPLACING COMMERCIAL ACTIVATED CARBON WITH BIOCHAR MADE FROM SLUDGE AND WOOD PELLETS --- ASSESSMENT BASED ON REMOVAL AND CAPACITY

THE VUDP ASSOCIATION PROJECT REPORT

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1. Dansk sammenfatning

Projektets ide var at fremstille aktiv kul fra spildevandsslam og plantemateriale for at undersøge anvendeligheden af det aktive kul til rensning af miljøfarlige stoffer såsom lægemidler og perfluoralkylforbindelser (PFAS).

I projektet er undersøgt, om aktiv biokul fra slam kan bruges som erstatning eller supplement for konventionelt aktivt kul. Aktiv biokul refereres til som aktiveret biochar i denne rapport.

Aktiv kul filtrering, kaldet GAC, er en af de teknologier, som kan fjerne miljøfremmede stoffer fra renseanlægs udløbsvand.

Der er blevet fremstillet 4 kombinationer af aktiveret biochar med forskellige forhold af kul fra spildevandsslam og kul fra træpiller. Der er brugt kul fremstillet af 100% spildevandsslam hhv. 100% træpiller så vel som 70/30 og 30/70 blandinger af de to materialer og sammenlignet med en reference af kommerciel GAC (Brennsorb 1240).

De fire kombination af aktiveret biochar er testet i laboratorie skala kolonner. Forsøgene har vist at den aktiverede biochar har en lavere sorptionsevne sammenlignet med kommercielt aktivt kul.

Hvorvidt dette kan løses i den næste generation af aktiverede biochar produkter eller dette kan kompenseres ved mere hyppig udskiftning og regenerering af biokul bør afklares nøjere i efterfølgende udviklingsarbejde.

Biokul, som et cirkulært materiale, indeholder høje fosfat koncentrationer og er derfor også testet for udvaskning af fosfat. Biokul udvasker mere fosfat end reference GAC.

Relevansen af dette skal vurderes i forhold til den aktuelle udløbstilladelse for fosfat.

Table 1: Resume af kolonneforsøgene med fjernelse af lægemidler fra vandfasen. PNEC-værdierne for de undersøgte lægemidler og den beregnede udløbskoncentration af disse for de 5 materialer ved 2000 empty bed volumes. Hvis den beregnede koncentration er lavere end den kvantificerbare koncentration angives mindre end denne i tabellen.

Compound	PNEC	100% Slam	70% Slam	30% Slam	0% Slam	GAC (Brennsorb 1240)
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
4-Methyl-benzotriazole	8	0.083	0.012	<0.01	<0.01	0.024
Amisulpride	0.17	<0.001	<0.001	0.0094	<0.001	<0.001
Atenolol	128	<0.005	<0.005	0.012	<0.005	0.0056
Benzotriazole	19	<0.01	<0.01	0.014	0.013	00.19
Bicalutamid	0.1	<0.01	<0.01	0.010	<0.01	<0.01
Carbamzepine	0.5	<0.003	<0.003	0.013	<0.003	<0.003
Cetirizine	0.52	<0.003	0.013	0.073	0.056	0.040
Citalopram	0.51	0.0038	0.0044	<0.003	0.0048	0.005
Clarithromycin	0.06	<0.02	<0.02	<0.02	<0.02	<0.02
Clindamycin	0.0114	0.014	0.014	<0.01	0.016	0.018
Diclofenac	0.1	<0.01	<0.01	0.049	0.025	0.019
Erythromycin	0.04	<0.001	<0.001	0.0018	<0.001	<0.001
Furosemide	31	0.041	<0.03	<0.03	<0.03	<0.03
Gabapentin	100	0.019	0.97	0.66	0.41	0.7
Iohexol	1 000 000	0.23	1.4	0.18	0.39	3.0
Iomeprol	1 000 000	0.029	0.34	0.032	0.047	0.42
Iopamidol	1 000 000	<0.1	<0.1	<0.1	<0.1	<0.1
Irbesartan	704	<0.001	0.0058	0.013	0.0033	0.0025
Lidocaine	0.00261	<0.003	<0.003	0.006	0.0037	0.014
Losartan	331	0.0064	0.027	0.026	0.015	0.033
Mefenamic acid	3.9	<0.001	<0.001	<0.001	<0.001	<0.001
Metoprolol	0.1	<0.005	<0.005	0.018	0.010	0.0073
Oxazepam	0.0019	<0.003	<0.003	0.058	<0.003	<0.003
Propanolol	0.1	<0.003	<0.003	<0.003	<0.003	<0.003
Sertraline	0.1	0.010	0.010	<0.01	0.010	0.013
Sulfadiazine	4.6	<0.003	0.0033	0.0033	<0.003	<0.003
Sulfamethizole	2.54	<0.001	0.018	0.010	<0.001	0.028
Sulfamethoxazole	0.118	<0.003	<0.003	0.025	0.028	0.072
Trimethoprim	10	<0.001	<0.001	<0.001	<0.001	<0.001
Tramadol	2.25	<0.05	<0.05	<0.05	<0.05	<0.05
Venlafaxine	0.1	<0.005	0.075	0.119	0.063	0.14

2. Summary

In this project, it was explored whether activated biochar made from sludge or wood or in combination can be used as a replacement of conventional granulated activated carbon (GAC) to remove organic micropollutants such as pharmaceuticals or PFAS from effluent wastewater.

GAC is one of the available technologies which can remove micropollutants from wastewater.

The project assessed biochars made of 100% sludge and 100% wood as well as 70/30 and 30/70 mixtures of these materials in comparison to a commercial GAC.

In lab-scale, it could be demonstrated that all materials have considerable sorption for all tested compounds.

At the current state of development, the activated biochars have slightly lower affinity to the pollutants than the commercial GAC. – Whether this can be overcome in the next generation of activated biochars or whether this can be compensated by more frequent exchange and re-generation of biochars needs to be clarified in succeeding projects.

The biochars as materials of circular economy were made of materials with a high phosphate content and also tested for emissions of phosphate. They were emitting more phosphate than the comparison GAC. How relevant this is will depend on the respective discharge permit.

Table 2: Overview of column experiments for removing micropollutants. The PNEC for target micropollutants and the calculated micropollutant concentration of every material at 2000 bed volumes. If the calculated concentration is lower than the quantifiable concentration, the quantifiable concentration will be presented in the column.

Compound	PNEC	100% Sludge	70% Sludge	30% Sludge	0% Sludge	Brennsorb 1240 GAC
	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]	[µg/L]
4-Methyl-benzotriazole	8	0.083	0.012	<0.01	<0.01	0.024
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Oxazepam	0.0019	<0.003	<0.003	0.058	<0.003	<0.003
Propanolol	0.1	<0.003	<0.003	<0.003	<0.003	<0.003
Sertraline	0.1	0.010	0.010	<0.01	0.010	0.013
Sulfadiazine	4.6	<0.003	0.0033	0.0033	<0.003	<0.003
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Sulfamethoxazole	0.118	<0.003	<0.003	0.025	0.028	0.072
Trimethoprim	10	<0.001	<0.001	<0.001	<0.001	<0.001
Tramadol	2.25	<0.05	<0.05	<0.05	<0.05	<0.05
Venlafaxine	0.1	<0.005	0.075	0.119	0.063	0.14

3. Introduction

The presence of micropollutants is frequently detected in wastewater effluents (Undeman et al., 2022) and has become an increasing concern (Schwarzenbach et al., 2006; Yang et al., 2022). Micropollutants, including pharmaceuticals, personal care products, as well as per-and polyfluoroalkyl substances (PFAS) are commonly present in waters at trace concentrations (ng/L to µg/L) (Barbosa et al., 2016; Dasu et al., 2022). The existence of micropollutants in water raises public concerns. The major concerns pose on aquatic ecosystems and on human health involve endocrine disrupting effects (Jobling et al., 1998), possible synergistic effects between micropollutants, and chronic effects by a long-term exposure (Fent et al., 2006). Therefore, an effective micropollutant removal procedure needs to be identified and implemented. To facilitate the installation of effective treatment processes, Aarhus Water and Hillerød municipalities published micropollutant discharge guidelines for their wastewater treatment plants (WWTPs). Both utilities will be required to comply with these guidelines in the future. The suggested emission concentration in effluent wastewater is designed based on predicted no-effect concentration (PNEC) (Kisielius et al., 2023; Spilsbury et al., 2024). It is the concentration of a chemical which marks the limit at which below no adverse effects of exposure in an ecosystem are expected.

Various technologies were discussed to further improve the treatment performance of micropollutants in the final wastewater effluents (Kosek et al., 2020). For instance, advanced oxidation processes (AOPs) (Giannakis et al., 2015), ozonation (Lee et al., 2014) and activated carbon filtration (Kovalova et al., 2013). In the perspective of industrial application, the preferred technology should have features such as easy to implement, low energy input, low CO_2-footprint low maintenance and less generation of harmful byproducts. Therefore, among those technologies, granulated activated carbon (GAC) filtration is preferred as its main removal mechanism is sorption thus no metabolite will be generated in the activated carbon filtration process. Therefore, the application of GAC in reducing micropollutant concentrations in effluent wastewater will be discussed in this report.

To date, typical raw materials for commercial activated carbon production are materials with high carbon content such as coals, coconut shells, wood, peat and petroleum residues. The removal performance of activated carbon made from these raw materials are studied extensively (Ashoori et al., 2019; Favre et al., 2022; Hagemann et al., 2020). Despite these studies usually conducted

with real wastewater as matrix, the quantity of micropollutants monitored is often inadequate (15 compounds max) and the experimental time is relatively short (<1000 bed volumes). In addition, these studies selected either pharmaceuticals or PFAS as target contaminant which did not assess the performance of activated carbon holistically.

In the perspective of industrial applications, the purchasing and transportation cost of commercial GAC should be minimized. Sludge, a common byproduct generated during conventional activated sludge (CAS)-based WWTPs, is usually recognized as an ecological burden. Previous studies indicated that sludge can be regarded as a raw material for GAC production (Hadi et al., 2015). As the utilization of sludge for biochar production is in line with circular economy (Ihsanullah et al., 2022), efforts are made to investigate if the sludge-derived GAC performs well in terms of micropollutant removal (Regkouzas and Diamadopoulos, 2019; Zhang et al., 2023). However, their studies did not consider the long-term performance of GAC and thus unable to assess the capacity of the GAC material. To date little studies have been conducted to investigate the performance of hybrid (sludge mixed with wood pellet) biochar in terms of micropollutant removal and to evaluate its industrial application perspective.

In this project, four compositions of biochar are prepared with different ratio of sludge and wood pellet. Their removal performances and capacities in micropollutants (pharmaceutical and PFAS) mitigation were assessed against commercial activated carbon. In addition, this project also assessed the concentration of phosphorus leached from each biochar to evaluate its potential for industrial application if strict regulations are placed for phosphate after treatment.

Overall, this project has three milestones (M1-M3).

- M1: Laboratory column experiments with four types of biochar made with different ratio of sludge and wood pellet provided by AquaGreen and one commercial activated carbon. Column experiments with five bottom-up column reactors packed with aforementioned activated carbons and run with real effluent wastewater collected from Solrødgaard wastewater treatment plant (HCR Syd, Hillerød Forsyning). As the wastewater effluent naturally contains micropollutants, their concentration in the inflow and outflow of column reactors are constantly monitored to assess the removal performance as well as capacity of activated carbons over time.

- M2: Laboratory equilibrium experiments with aforementioned activated carbons (commercial activated carbon and 4 biochar with different ratio of wood pellet and sludge). The aim of this milestone is to conduct a rapid assessment of the filter media's capacity in terms of removing micropollutants (pharmaceuticals and PFAS). The equilibrium experiments for pharmaceutical and PFAS were conducted with real effluent wastewaters (collected from Hillerød and Fårevejle wastewater treatment plants, respectively). The effluent wastewater was spiked with pharmaceuticals and PFAS respectively to desired initial concentration.
- M3: Laboratory phosphorous leaching experiments with all the filter media mentioned. The aim is to investigate the phosphorous leaching of activated carbons and determined whether the phosphorous emission is below environmental standards.

Micropollutants of interest

Table 3: Micropollutants (excluding PFAS) included in this project and their respective PNEC in µg/L. The average concentration (µg/L) represents the concentrations of target compound in effluent water collected from Solrødgaard wastewater plant (HCR Syd) between January 2023 to March 2024.

Name	PNEC [µg/L]	Category	Average conc. [µg/L]
4-Methyl-benzotriazole	8	Corrosion inhibitor	2.99
Amisulpride	0.17	Antipsychotic medication	0.099
Atenolol	128	Beta blocker medication	0.08
Benzotriazole	19	Corrosion inhibitor	5.9
Bicalutamid	0.1	Antiandrogen medication	0.125
Carbamzepine	0.5	Anticonvulsant & antidepressant	0.054
Cetirizine	0.52	Antihistamine	0.41
Citalopram	0.51	Antidepressant	0.21
Clarithromycin	0.06	Antibiotic	0.061
Clindamycin	0.0114	Antibiotic	0.06
Diclofenac	0.1	Nonsteroidal anti-inflammatory drug	0.62
Erythromycin	0.04	Antibiotic	0.031
Furosemide	31	Diuretic medication	1.21
Gabapentin	100	Anticonvulsant	0.98
Iohexol	1 000 000	X-ray contrast media	9.02
Iomeprol	1 000 000	X-ray contrast media	1.13
Iopamidol	1 000 000	X-ray contrast media	0.98
Irbesartan	704	Hypertension drug	0.028
Lidocaine	0.00261	Anesthetic agent	0.24
Losartan	331	Hypertension drug	0.77
Mefenamic acid	3.9	Nonsteroidal anti-inflammatory drug	0.009
Metoprolol	0.1	Hypertension drug	0.98
Oxazepam	0.0019	Anxiety-treatment drug	0.16
Propanolol	0.1	Hypertension drug	0.05
Sertraline	0.1	Antidepressant	0.066
Sulfadiazine	4.6	Antibiotic	0.026
Sulfamethizole	2.54	Antibiotic	0.11
Sulfamethoxazole	0.118	Antibiotic	0.06
Trimethoprim	10	Antibiotic	0.14
Tramadol	2.25	Pain killer	0.47
Venlafaxine	0.1	Antidepressant	0.46

Table 4: Compounds included but not analyzed due to low initial concentration in wastewater. These compounds were not further followed up upon.

Name of compound	Initial concentration level (µg/L)
Phenazone	<0.01
Ibuprofen	<0.05
Sotalol	<0.05
Mycophenolic acid	<0.01
Olmesartan	<0.01
Candesartan	<0.01
Diatrizoic acid	<0.01
Codeine	<0.01
Ranitidine	<0.05
Gemfibrozil	<0.05
Miconazole	<0.01
Propyphenazone	<0.02
Rosuvastatin	<0.01
Simvastatin	<0.01

Table 5: PFAS included in this project.

Name	Full name	CAS	Chemical formula
PFOA	Perfluorooctanoic Acid	335-67-1	$C_8HF_{15}O_2$
PFOS	Perfluorooctanesulfonic acid	1763-23-1	$C_8HF_{17}O_3S$
PFHpA	Perfluoroheptanoic acid	375-85-9	$C_7HF_{13}O_2$
ADONA	Dodecafluoro-3H-4,8-Dioxanonanoic Acid	919005-14-4	$C_7H_2F_{12}O_4$
PFHxA	Perfluorohexanoic acid	307-24-4	$C_6HF_{11}O_2$
PFHxS	Perfluorohexanesulfonic acid	355-46-4	$C_6HF_{13}O_3S$

Role of partners

1. Aquagreen produced the required quantities of test materials and characterized them.
2. Aarhus University (AU) received the activated carbon, set up and operated laboratory columns and performed analysis for pharmaceuticals to assess the removal rate and efficiency of GAC filtration with the 4 selected compositions of sewage sludge and wood pellets against a reference with conventional activated carbon.
3. Odsherred Forsyning is the main applicant and project coordinator assisted by Hillerød Forsyning.
4. Vandcenter Syd and Aarhus Vand participated in the follow-up group and carried out quality assurance of the final report.

4. Relevance for the Danish Water Sector

A focus point in the Danish Water Sector is the removal of toxic pollutants, such as pharmaceuticals and PFAS from wastewater. This is a focus point in the rest of EU as well.

The Urban Waste Water Treatment Directive (UWWTD) (Commission, 2022) requires all EU countries to ensure wastewater is collected and treated to protect the environment from adverse effects of urban and industrial wastewater. The UWWTD is expected to be updated in October/November 2024 to include removal of micropollutants for WWTPs over a certain size. The renewed UWWTP, or "Byspildevandsdirektivet" in Danish, will therefore be implemented for Danish Wastewater Utilities depending on the size of their wastewater treatment plants.

The removal of micropollutants is known as the 4th treatment step. Well-known technologies are ozonisation, activated carbon (GAC) and sand filtration.

A popular choice for 4th treatment step is a combination of ozonisation and GAC. The disadvantage with this is the energy cost and amount of activated carbon needed.

The focus of this project has been to investigate if GAC made from pyrolyzed wastewater sludge (biochar) can be used as an alternative or supplement to conventional GAC in the 4th treatment step, thereby introducing a more circular and cost beneficial method with a lower carbon footprint.

Results show that GAC from pyrolyzed wastewater sludge (biochar) can be used as a supplement for removal of specific micropollutants. There is therefore a potential for use of biochar to replace commercial GAC but also the need for investigating the application in full scale.

4.1 Markets and applications

As it is still the early stages, application of GAC from biochar in full scale at different wastewater treatment plants (WWTP) is needed to determine the scope.

Preliminary results show the potential for Wastewater Utilities to use the GAC from biochar for removal of specific micropollutants, including 4-methyl-1H-benzotriazole, amisulpride, atenolol, benzotriazole, bicalutamid, clarithromycin, citalopram, diclofenac, erythromycin, furosemide, lidocaine, losartan, mefenamic acid, metoprolol, propranolol, sertraline, sulfadiazine, sulfamethizole, tramadol and trimethoprim.

The marked value is therefore not known at this point.

4.2 The next steps

The next step will be to apply for a new research project to further study the possibilities of biochar-GAC full scale in a wastewater treatment plant.

4.3 Dissemination

The preliminary results have been presented at the Danish Water Forum in January 2024.

An abstract with conclusions from the study have been submitted to Danskvand.

5. The project

5.1 Project goals and objectives

The goal of the project was to test whether and to what extent activated biochar made of sewage sludge can be used to remove organic micropollutants (MFS) from effluent wastewater.

The objectives of the projects were:

- To test the sorption of pharmaceuticals and PFAS on activated biochars made from different fractions of wood and biochar in comparison to a commercial granulated activated carbon (GAC)
- To assess removal for pharmaceuticals and PFAS with biochars made from different fractions of wood and biochar in comparison to GAC from effluent wastewater.
- To assess the capacity (how much water can be treated with one unit of activated carbon) while maintaining the target value in the effluent water.
- To test whether the biochars leach unwanted compounds such as phosphate to the effluent wastewater.

5.2 Output

Biochars produced from four different fractions of municipal sewage sludge and wood were produced by Aquagreen. They were activated and mixed in ratios 100 % wood biochar (B), 70 % wood biochar and 30 % sludge biochar (C), 30 % wood biochar and 70 % sludge biochar (D), and 100 % sludge biochar (E).

Columns packed with these four different materials plus one made of commercial GAC (Brennsorb1240) were packed and run by Aarhus University.

45 pharmaceuticals and 6 PFAS were analyzed by Aarhus University after contacting polluted water (effluent wastewater) with the respective materials.

The main output of this project is the final report enabling the decision in which cases biochars can be used for treating wastewater.

5.3 Production of activated biochars

AquaGreen produced activated biochars consisting of different ratios sludge/wood, i.e.; 0/100%, 30/70%, 70/30% and 100/0%. These were sieved and the fraction 525-1600 µm was used for the tests.

Production of biochar and activated biochar from sludge

On Fårevejle WWTP (Odsherred utility), the sludge was dried and pyrolyzed on a HECLA1000 plant with automatic drying and pyrolyzing processes for sludge.

Pyrolysis was conducted at 650 °C for 20 minutes, which are representative operation conditions.

The biochar was succeedingly activated by steam in an electric oven at AquaGreen. Activation was conducted at 600°C over 30 minutes. In total 63 kg biochar was activated over 8 h by applying access of steam continuously.

Figure 1:Picture of activation oven at AquaGreen and temperature/time diagram of the activation of sludge-biochar at AquaGreen

Production of biochar and activated biochar from wood

Wood pellets of 6 mm were pyrolysed in an electric pyrolysis oven with a screw conveyor at 650°C over 30 minutes. During the pyrolysis the system was continuously flushed by nitrogen both from the intake and the outlet side of the system. In total 100 kg wood pellets were pyrolyzed over a 10 h period.

Figure 2: Temperature/time diagram of the production of the wood-biochar at AquaGreen.

The wood-based biochar was successively activated by means of steam in an electrically heated oven with a screw conveyor at 760 °C over 30 minutes. In total 19 kg wood-based biochar were activated over a 4 h period, while steam was injected in excess to the system.

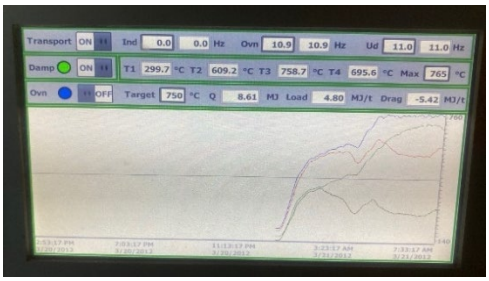

Figure 3: Temperature/time diagram of the activation of the wood-biochar at AquaGreen.

Table 6: Overview on pyrolysis and activation temperatures.

	Sludge	Wood pellets
Pyrolyze	650°C for 20 min	650°C for 30 min
Activation	600°C for 30 min	760°C for 30 min

Sieving of activated biochars

The sorption test was conducted with columns which required a selected particle range to be able to maintain hydraulic conductivity. Also to be comparable to the commercial GAC, the materials were sieved to a fraction 425 - 1600 µm.

The sieving was conducted on a Retsch vibrator AS200 with 1.4 mm/s^2 over 4 minutes.

From the produced sludge and wood based activated biochars, respective fractions were determined by weight and mixed to give the following ratios: 0/100%, 30/70%, 70/30% and 100/0%.

5.4 The removal of pharmaceutical and PFAS

The removal of pharmaceutical was assessed in two separate experiments (equilibrium and column). The column experiment is a long-term experiment used to assess the removal over time as well as the capacity. The column experiment took the effects of wastewater constituent and flow rate factor into consideration. On the other hand, equilibrium experiment is a quick method to predict the capacity of the material with a short-term experiment without considering other influencing factors.

Since real wastewater effluent without spiking will be used for the column experiments and the concentration of PFAS in wastewater effluent is around the LC-MS/MS' limit of quantification thus make it impossible to conduct the column experiment for PFAS removal. Therefore, column and equilibrium experiments will be used to investigate the removal of pharmaceuticals and only equilibrium experiment will be used to assess the removal of PFAS.

5.4.1 Pharmaceutical removal by column experiments

5.4.1.1 Experimental setup

As shown in Figure 1, the experimental setup consists of five columns (180 mL in volume and 30 cm in height) packed with different activated carbons - four biochars made from different compositions of sludge and wood pellets (Figure *4* and Table *7*) and one commercial activated

carbon (Brennsorb 1240) as reference. The columns are covered with aluminium foil to protect the column from direct sunshine.

Real wastewater effluent from Hillerød WWTP was regularly collected and transported to Aarhus university, Roskilde and stored in 4 ºC cold room until usage. Before usage, the wastewater was moved from cold room and stored in fridges (Figure *5*). Two 4-channel peristaltic pumps were used to deliver the wastewater to the column reactor (bottom-up direction) and the flow rate were calibrated at 3.66 mL/min (equivalent to 0.22 L/h). The outflow of column reactor was directed to a waste cannister. The volume of wastewater in each waste cannister was measured (in volume) and discarded regularly. The measured volume was recorded and was used to calculate the bed volume (the volume needed to fill the column reactor) that each column reactor has passed. The sampling was done by collecting the wastewater from the outflow (~ 10 mL) and store in a 20 mL glass vial. Wastewater from the inflow was directly sampled from the wastewater cannister in the fridge (kept at 4 ºC). The glass vial was placed in a freezer room after the sampling procedure is done.

Figure 4: Four types of biochar with different mixing ratio of sludge and wood pellet provided by AquaGreen.

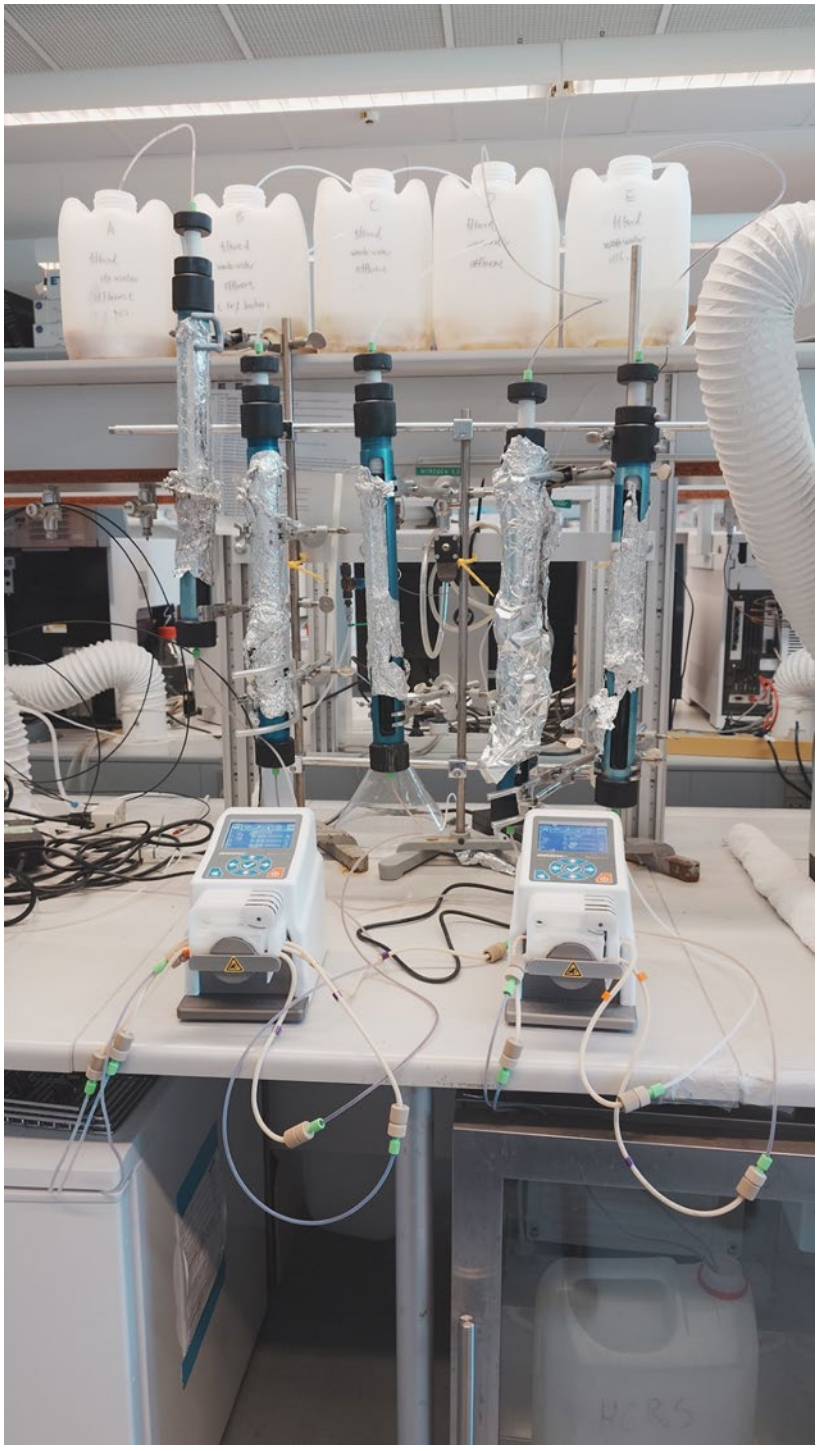

Figure 5: Experimental setup for the micropollutant column experiment.

Table 7: Composition of biochar provided from AquaGreen and their coding for the column experiment.

Code of columns	Brennsorb 1240 (Commercial GAC)	Biochar from wood pellet	Biochar from sludge
A (Brennsorb 1240)	100%	-	-
B (Wood)	-	100%	-
C (Wood/Sludge)	-	70%	30%
D (Sludge/Wood)	-	30%	70%
E (Sludge)	-	-	100%

Table 8: Key parameters for the column experiment.

Parameters	
Inflow	Effluent wastewater from Hillerød WWTP
Flow rate	0.22 L/h
Packing height	30 cm
Contact time	30 min
Flow direction	Bottom up
Sampling frequency	Every 50/100 bed volume

5.4.1.2 Data analysis and visualization

The removal of pharmaceuticals is expressed as R and calculated with the following equation:

$$R = \frac{C_{in} - C_{out}}{C_{in}}$$

Whereas C_{in} is the concentration of target compound in the inflow of the reactor in a specific bed volume (BV), C_{out} is its concentration in the outflow that was taken at the same day with the sample for C_{in}. The concentration of target compounds is the calculated concentration determined by LC-MS/MS.

The plotting of graphs was performed in GraphPad Prism 10 by plotting removal proportion of target pharmaceutical against the bed volume that had passed through the specific column reactor at that time point.

5.4.1.3 Removal

The appearance order of the micropollutants (excluding PFAS) is determined based on the order of suggested PNEC value (from low to high). The calculated concentration of every target compound at each specific bed volume (100, 500, 1000, 1500, and 2000 BV) is also given and the red font in the table means the calculated concentration is higher than the suggested PNEC value. If the calculated concentration at specific bed volume is below the limit of quantification (LOQ) of the LC-MS/MS, < the numerical value of LOQ will be marked in the table. A short summary of the column's performance and a brief discussion about whether the column can treat target compound to the concentration below PNEC level are listed in bullet points. Certain micropollutants (see table) (Union, 2022) are mentioned in the revised urban wastewater directive. These micropollutants are divided into two categories (category 1 and 2) with category 1 meaning substances that can be very easily treated while category 2 refers to substances that can be easily disposed of. At least 80% the listed micropollutants is required to be removed in the influent (required by the draft EU Urban Wastewater Directive (COM(89) 518 final).

For the removal of target compounds, the main removal mechanisms are adsorption onto GAC and absorption on the biofilm followed by biodegradation via microorganisms (Reungoat et al., 2010). Adsorption onto GAC is affected by the molecular charge, hydrophobicity and molecular weight (Mailler et al., 2015) as well as the functional groups (Delgado et al., 2012). Other influencing factors includes contact time (Snyder et al., 2007) and composition of effluent organic matter (EfOM) presented in effluent wastewater (Graham et al., 2000; Knappe et al., 1998). Biodegradation only happens after the formation of biofilm on the activated carbon. The formation of biofilms was caused by the microorganisms existing in the effluent wastewater and the microorganisms were colonizing the activated carbon during the operation period (Gibert et al., 2013). The formation of biofilm could result in clogging in the column which may lead to flow

issue (Gibert et al., 2013), which was observed in column C. The effect of flow issue in the column leads to less stable removal on certain micropollutants.

Table 9: Indicator micropollutants listed in the EU directive.

Name of micropollutants	Category number	Included in this project
Amisulpride	1	Yes
Carbamazepine	1	Yes
Citalopram	1	Yes
Clarithromycin	1	Yes
Diclofenac	1	Yes
Hydrochlorothiazide	1	No
Metoprolol	1	Yes
Venlafaxine	1	Yes
Benzotriazole	2	Yes
Candesartan	2	No
Irbesartan	2	Yes
4-Methylbenzotriazole	2	Yes
6-Methylbenzotriazole	2	No

Summary of micropollutant removal by column experiments

In this section, the removal of 31 micropollutants using activated carbons are summarized in the following table (details are given in section 1 in the appendix).

As sorption is the main mechanism for micropollutant removal in GAC and the occurrence of biodegradation is compound dependent, the table indicates if biodegradation occurred to specific compounds by +, if not an - is put. The average removal of micropollutants in biochars in the first 50 bed volumes is counted as initial removal%. After the biofilm is developed (if applicable) and the average removal of micropollutants in biochars in the final 100 bed volumes is given in the "avg. removal%" column. The last data point of biochars is used to check if the outflow concentration of micropollutants is higher than specified PNEC or not. If not, then "No" is put in the column and the required PNEC level is given in the bracket. If only one biochar composition failed to meet the PNEC requirement, its column is denoted in the bracket (ex. means except). When the removal by commercial GAC is better than biochars, "Yes" is used to indicate such differences in the last column.

Table 10: Overview of micropollutant removal in biochar materials.

Name	Possible involvement of biodegradation?	Initial removal%	Avg. removal%	End concentration lower than PNEC?	Does commercial GAC perform better?
4-Methyl-benzotriazole	-	>99	>99	Yes	No
Amisulpride	+	70	>99	Yes	No
Atenolol	-	>99	92	Yes	Yes
Benzotriazole	-	>99	>99	Yes	No
Bicalutamid	-	80	>95	Yes	Yes
Carbamazepine	+	95	90	Yes	Yes
Cetirizine	-	>99	80	Yes	Yes
Citalopram	+	97	90	Yes	No
Clarithromycin	+	90	>95	Yes	No
Clindamycin	-	>99	60	No (0.0114 µg/L)	No
Diclofenac	-	>99	90	Yes	Yes
Erythromycin	+	80	85	Yes	No
Furosemide	-	>99	>99	Yes	No
Gabapentin	-	35	30	Yes	Yes
Iohexol	+	50	60	Yes	Yes
Iomeprol	+	70	60	Yes	Yes
Iopamidol	+	70	60	Yes	Yes
Irbesartan	+	>99	60	Yes	Yes
Lidocaine	-	>99	>95	Yes	Yes
Losartan	-	>99	90	Yes	Yes
Mefenamic acid	+	73	>99	Yes	No
Metoprolol	-	>99	>95	Yes	No
Oxazepam	+	>99	80	No (0.0019 µg/L)	Yes
Propanolol	+	92	>99	Yes	No
Sertaline	+	80	90	Yes	No
Sulfadiazine	+	>99	90	Yes	Yes
Sulfamethizole	-	>99	90	Yes	No
Sulfamethoxazole	-	>99	95 (ex.E, 20)	Yes	Yes
Trimethromycin	-	>99	>95	Yes	No
Tramadol	-	>99	85	Yes	No
Venlafaxine	-	>99	80	No (ex. E, 0.1 µg/L)	Yes

5.4.2 Removal of micropollutants in equilibrium partition experiments

5.4.2.1 Experiment setup

The equilibrium experiment was conducted in an overhead shaker and effluent wastewater collected from Hillerød (for pharmaceutical) and Odsherred (for PFAS) are used as the matrix. 200 mL of effluent wastewater was added to 250 mL Polypropylene (PP) bottles and spike the stock solution to desired concentrations (10 µg/L for pharmaceutical or 10 ng/L for PFAS). The first sample was taken without the presence of activated carbon. Samples for pharmaceutical analysis were stored in amber LC vials and samples for PFAS analysis were stored in a PP vial to avoid sorption of PFAS from the LC vials. The predetermined activated carbon was transferred to the PP bottles and the bottle was mounted in the overhead shaker. The overhead shaker was started, and sample was taken every 2 hours in the next 48 hours. The sample vials were stored in a fridge (4 ºC) for analysis.

Figure 6: Overhead shaker used for equilibrium experiments.

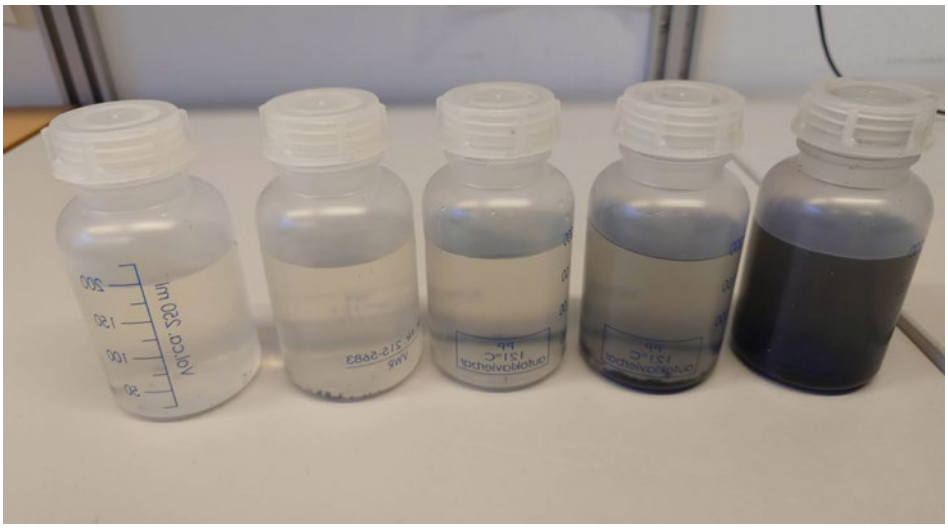

Figure 7: PP bottle with activated carbon after the equilibrium experiment. The dosage of carbon increases from left to right.

Table 11: Key parameters in the equilibrium experiment.

Parameter	Setting
Initial concentration	10 µg/L (pharmaceutical), 10 ng/L (PFAS)
Volume	200 mL
Matrix	Wastewater effluent (Hillerød for pharmaceutical, Odsherred for PFAS)
GAC weight	0,1, 0,3, 1, 3, 10 g/L
Rotation speed	~120 rpm
Duration	48 hours
Sampling frequency	Every two hours

5.4.2.2. Calculation of distribution coefficients

The distribution coefficient (K_d) of GAC in the equilibrium experiment is calculated as

$$K_d = \frac{C_S}{C_{eq}}$$

In this equation, C_{eq} values were directly determined by LC-MS/MS and C_s was obtained by the following equation:

$$C_s = \frac{(C_0 - C_{eq}) * V}{m}$$

where

C_0 (μg L^{-1}) corresponds to the initial concentration of pharmaceutical added to the suspension, V (mL) is the volume of solution and m (g) refers to the dry mass of the material.

The calculated value of K_d can be used to estimate the capacity of biochars, the higher the K_d is, the higher capacity certain composition of biochar is.

5.4.2.3 Results on pharmaceutical removal in partitioning experiments

The graphs of the micropollutant partitioning experiment are given in section 2 of the appendix.

Table 12: Table summarizing the micropollutant partitioning experiment.

Name	Minimum dosage (g/L) for 50% removal	Estimated bed volume for removal exceeding 50%	Difference in biochar composition?	Does commercial GAC (Brennsorb 1240) perform better?
Atenolol	1	1 000	Yes	Yes
Amisulpride	1	1 000	No	Yes
Benzotriazole	0.3	3 000	No	Yes
Candesartan	0.1	10 000	No	No
Carbamazepine	1	1 000	No	Yes
Cetirizine	1	1 000	No	Yes
Citalopram	1	1 000	No	Yes
Clarithromycin	1	1 000	Yes	No
Codeine	1	1 000	Yes	Yes
Diclofenac	1	1 000	No	Yes
Gabapentin	10	100	Yes	Yes
Iohexol	3	300	No	Yes
Iomeprol	3	300	No	Yes
Iopamidol	3	300	No	Yes
Irbesartan	1	1 000	No	Yes
Losartan	1	1 000	No	Yes
Mefenamic acid	1	1 000	No	Yes
Metoprolol	1	1 000	Yes	Yes
Mycophenolic acid	0.3	3 000	No	No
Olmesartan	3	300	No	Yes
Oxazepam	1	1000	No	Yes
Phenazone	1	1 000	Yes	Yes
Propranolol	0.3	3 000	Yes	Yes
Propyphenazone	3	300	Yes	Yes
Rosuvastatin	1	1 000	No	Yes
Sertraline	0.3	3 000	No	Yes
Sulfadiazine	1	1 000	No	Yes
Sulfamethizole	1	1 000	No	Yes
Sulfamethoxazole	1	1 000	No	Yes
Trimethoprim	1	1 000	No	Yes
Venlafaxine	1	1 000	Yes	Yes

It seems like the partitioning experiments under-estimate the performance of the columns considerably (see appendix 7.1 and appendix 7.2)

5.4.3. Removal of PFAS in partitioning experiments

5.4.3.1 Data analysis for removal

The removal for PFAS is expressed as R and calculated with the following equation:

$$R = \frac{C_0 - C_t}{C_0}$$

Whereas C_0 is the concentration of target PFAS at time 0 before the activated carbon was put in, C_t is its concentration after t hours of shaking. The concentration of target compounds is the calculated concentration determined by LC-MS/MS.

5.4.3.2 Result

The initial concentration of PFAS was set at 10 ng/L, which is a more environmental relevant concentration of PFAS in wastewater effluent (Lenka et al., 2021). The matrix for the PFAS equilibrium experiment is the wastewater effluent collected from Fårevejle, Denmark. The PFAS concentration was determined using either one or two transitions depending on the response from the LC-MS/MS. The PFAS concentration determined with two transitions are PFOS and PFHxS. The graphs of PFAS partitioning experiment are given in section 3 of the appendix.

Table 13: Table summarizing the PFAS partitioning experiment.

Name	Minimum dosage for 50% removal	Estimated bed volume for 50% removal	Does the difference in biochar composition make a difference?	Does commercial GAC perform better?
PFOA	1 g/L	1000	No	Yes
PFOS	1 g/L	1000	No	No
PFHpA	3 g/L	300	No	No
ADONA	3 g/L	300	No	Yes
PFHxA	3 g/L	300	No	No
PFHxS	1 g/L	1000	No	No

5.4.4 Phosphorus leaching

5.4.4.1 Experimental setup

As sludge contains phosphorus (Silva-Leal et al., 2021), the emission of phosphorus from biochar should be monitored in order to avoid possible eutrophication in the receiving water bodies. Phosphorus leaching experiment was conducted for this purpose. A 1L Schott bottle filled with 1 L deionized water and ~10 mL water was sampled as initial sample. The bottle was then dosed with pre-determined activated carbon and was under constant shaking condition on a horizonal shaker (~200 rpm). After 8 hours, another 10 mL sample was taken as the end sample. The sample was stored in a cold room before sending for analysis.

Figure 8: Platform shaker used in phosphorus leaching experiment. A Schott bottle containing 1 L deionized water and 10 g activated carbon was used for this experiment.

Table 14: Key parameter applied for the phosphorus leaching experiment.

Parameter	Setting
Volume	1 L
GAC weight	0,1, 0,3, 1, 3, 10 g
Matrix	Deionized water
Rotation speed	~200 rpm
Duration	8 hours
Sampling timepoints	Start and end points

5.4.4.2 Result

The calculation of leached phosphorus in the filter material C_{emit} is done by

$$C_{emit} = C_1 - C_0$$

Whereas C_1 is the phosphorus (PO_4-P) or total phosphate (TP) concentration (mg/L) from the sample taken after the leaching experiment is done and C_0 is the concentration before the experiment started.

The rate of phosphorus emission is obtained by plotting the linear line to specific carbon dosage (3 g/L or 10 g/L) and calculate the slope of the line.

The plotting of graphs was performed in GraphPad Prism 10 by plotting the increment of phosphorus (expressed as PO_4-P) or total phosphate (expressed as TP) concentration before and after the experiment for every composition of activated carbon at different activated carbon dosages.

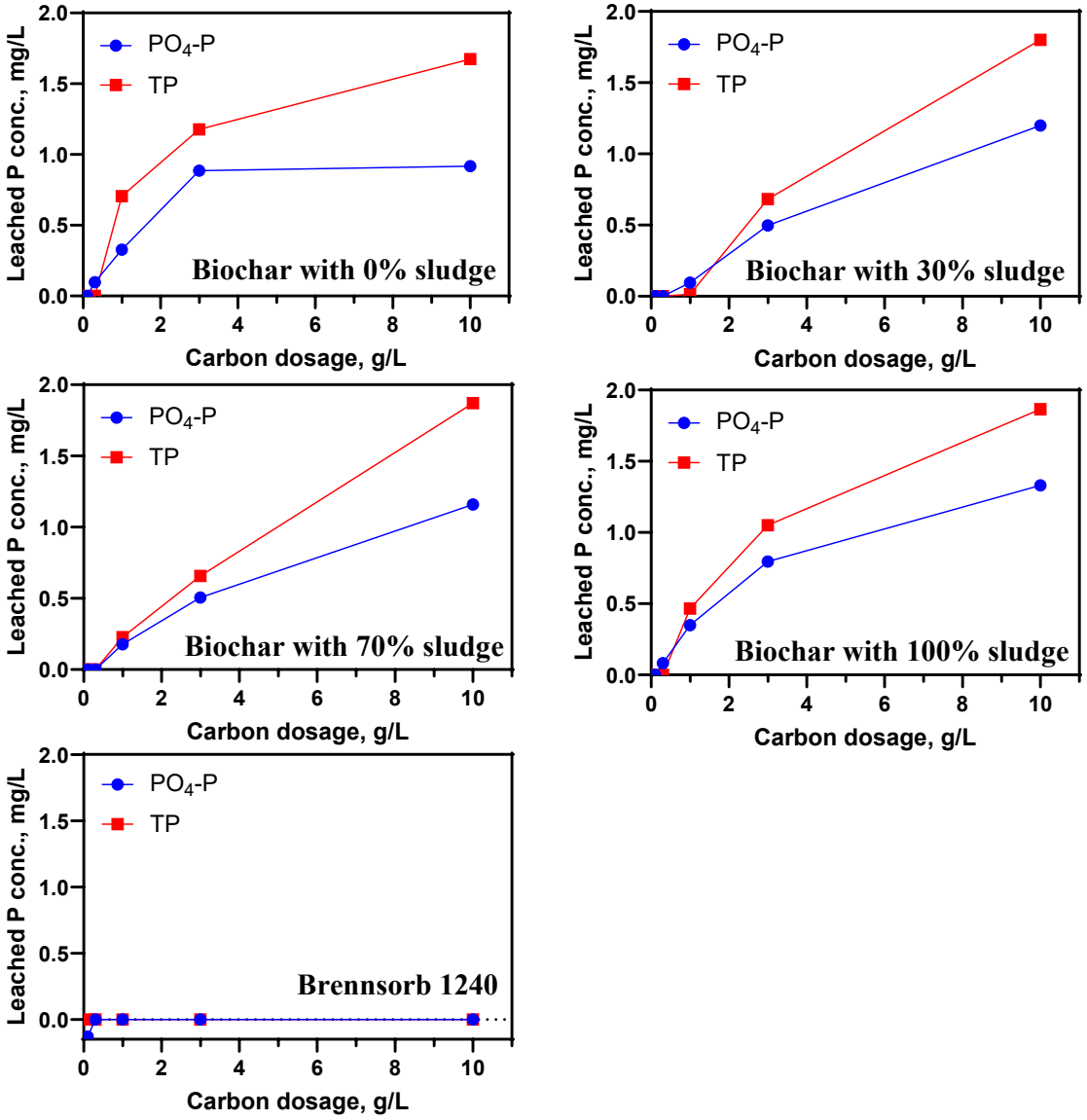

Figure 9: Comparison of phosphate leaching for different filter materials.

The commercial activated carbon (Brennsorb 1240) did not leach any phosphorus in the leaching experiment even at the carbon dosage of 10 g/L. In the contrast, all the biochars in this experiment leached phosphorus to deionized water after 8 h contacting time, indicating that the biochar leach phosphorus regardless the raw material (sludge or wood pellet). The leached amount is positively correlated with the amount of biochar dosed. When 1 g/L of biochar is dosed (mimicking operations over 1000 BV), the leached phosphorus level for all biochars is below 0.5 mg/L. The

leaching level of TP is higher than 0.5 mg/L when dosed with biochar made with 0% sludge at 1 g/L. As the plotted curve is not linear, it is suggested that in the experiments with the higher biochar/water ratios (exceeding 3 g/L) some kinetic hindering for the transfer of phosphate into the water occurred. The total phosphorus leaching level for all biochars is below 2 mg/L. With other words all biochars elute about 0.2-0.5 mgP per g biochar/L.

As the phosphorus tend to "saturate" in water at activated carbon level greater than 3 g/L, the calculation of phosphorus emission rate is conducted using two sets of result (leaching data up to 3 g/L and data up to 10 g/L). The obtained value indicated the difference in emission rate between biochar made with wood pellet and biochar made with sludge can be ignored (difference < 0.05 mg/L per gram of activated carbon)

Table 15: Phosphorus emission rate (mg/L) of each activated carbon up to 3 g/L and up to 10 g/L (shown in the bracket).

Type of carbon	**PO_4-P**	**TP**
Brennsorb 1240	0 (0)	0 (0)
Biochar with 0% Sludge	0,30 (0,08)	0,41 (0,16)
Biochar with 30% Sludge	0.17 (0,11)	0.23 (0,20)
Biochar with 70% Sludge	0,18 (0,12)	0,23 (0,19)
Biochar with 100% Sludge	0,27 (0,13)	0,37 (0,18)

6. Conclusions

- The biochars are able to remove micropollutants regardless of their compositions. The constituent of water, the types of target micropollutants and the development of biofilm seem to be the dominant factors in influencing the removal.
- In general, all biochars demonstrated good removal (>80%) for micropollutants especially after the biofilm is developed. Compared with commercial activated carbon (Brennsorb 1240), the removal achieved with the biochar is somewhat less.
- Removal of micropollutants can be achieved with biochars and the biochar made with less sludge content is having slightly higher removal for certain micropollutants. Using commercial activated carbon (Brennsorb 1240) in general results in higher removal of organic micropollutants than biochars.
- Removal of PFAS can be achieved with biochars and the difference in performance of the different biochars of different composition is neglectable.
- There is an indication removal of PFAS with activated biochars yield a better removal rate than commercial GAC.
- All biochars leached phosphorous. The highest leached P concentration in 1 L water is between 1 and 2 mg/L when mimicking operations with 100 BV. Emissions get lower, relatively speaking when operating at higher BV. The commercial GAC tested did not emit any phosphate.

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8. Appendix

8.1. Pharmaceutical removal in column experiments.

The figure showing the removal of micropollutants using different types of GAC is listed here.

Oxazepam (column experiments):

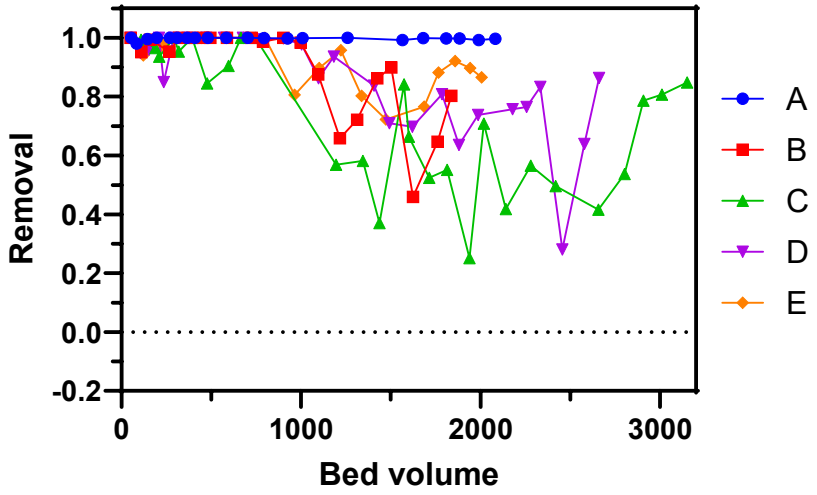

Figure 10: Removal of oxazepam in column experiment over bed volume.

Table 16: Concentration of oxazepam after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Oxazepam	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.003	<0.003	<0.003	<0.003	<0.003
B (Wood)	<0.003	<0.003	<0.003	<0.003	<0.003
C (Wood/Sludge)	<0.003	<0.003	0.048	0.053	0.058
D (Sludge/Wood)	<0.003	<0.003	<0.003	0.058	<0.003
E (Sludge)	<0.003	<0.003	0.046	0.042	<0.003

Over vast periods of the time, all materials resulted in removals exceeding 50%. In terms of removal over time, the commercial activated carbon achieved ~100% removal (SD = 0.004) and was able to reduce the oxazepam concentration to below LOQ (0.003 µg/L) throughout the experimental period. The performance of biochars started to decrease after eluting 1000 bed volumes of effluent wastewater and the maximum removal rate for the biochar was 80% after 1000 bed volumes. The stable performance of commercial activated carbon in removing oxazepam indicated towards this column is still far below its capacity and it is able to treat more wastewaters. On the other hand, the decrease in removal as observed for the biochars indicated the oxazepam had less affinity towards biochars. The PNEC for oxazepam is 0.0019 µg/L (Kisielius et al., 2023), which is lower than the LOQ of the LC-MS/MS, it is unlikely that any of the biochars can treat oxazepam to the concentration below PNEC level opposite to the commercial activated carbon.

Except for the commercial activated carbon and the biochar from 100% wood pellets all materials led oxazepam pass with concentrations exceeding the PNEC over significant periods of time. It can be assumed that the increasing removal around 2000 BV for material D&E could be due to starting biodegradation by biofilms growing on the materials.

Lidocaine (column experiments):

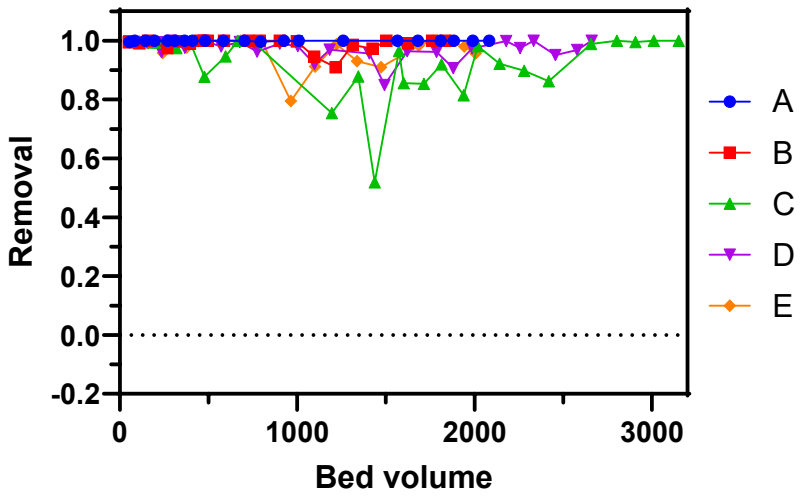

Figure 11:Removal of lidocaine in column experiment over bed volume.

Table 17: Concentration of lidocaine after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Lidocaine	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.003	<0.003	<0.003	<0.003	<0.003
B (Wood)	<0.003	<0.003	<0.003	<0.003	<0.003
C (Wood/Sludge)	<0.003	0.020	0.072	0.016	0.0060
D (Sludge/Wood)	<0.003	<0.003	0.0071	0.049	0.0037
E (Sludge)	<0.003	<0.003	0.10	0.013	0.014

Over vast periods of the time (14 months operation), all materials resulted in removals exceeding 90%. In terms of removal over time, the commercial activated carbon achieved ~100% removal throughout the experimental period and the performance of biochars have minor fluctuation after 1000 bed volume. As biofilm is likely to form in the first 1000 bed volume (discussed in the

oxazepam removal section), the formation of biofilm could result in clogging in the column which may lead to flow issue (Gibert et al., 2013), which was observed in column C. Other columns maintained good removal (>95%) in treating lidocaine in real effluent wastewater. The PNEC for lidocaine is 0.00261 µg/L (Kisielius et al., 2023), which is lower than the limit of quantification (0.003 µg/L) of the LC-MS/MS. It is unlikely that the activated carbon can treat lidocaine below PNEC level.

Clindamycin (column experiments):

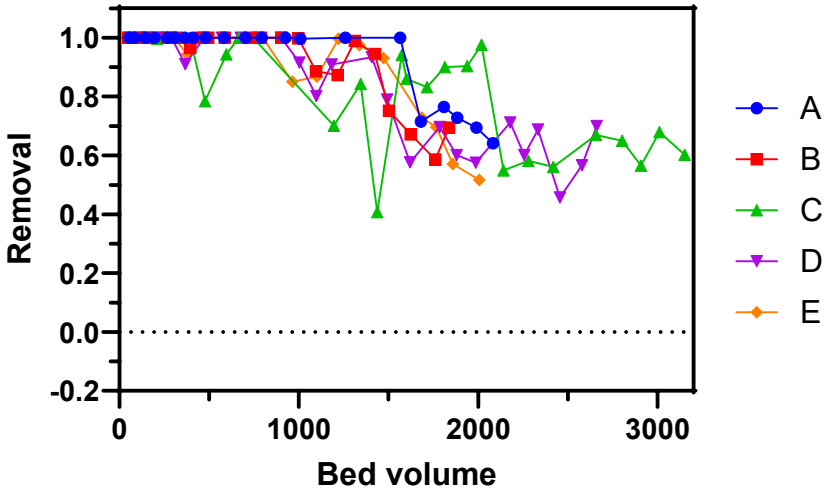

Figure 12: Removal of clindamycin in column experiment over bed volume

Table 18: Concentration of clindamycin after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Clindamycin	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.01	<0.01	<0.01	<0.01	0.014
B (Wood)	<0.01	<0.01	<0.01	0.014	0.014
C (Wood/Sludge)	<0.01	0.013	<0.01	<0.01	<0.01
D (Sludge/Wood)	<0.01	<0.01	0.010	<0.01	0.016
E (Sludge)	<0.01	<0.01	<0.01	<0.01	0.018

All activated carbon has stable removal for clindamycin in the first 1000 bed volumes except for material C has a minor decrease around 500 bed volumes. The removal of all activated carbon started to drop after 1300 bed volumes with the removal of column C started to decrease around 2000 bed volumes. Since the flow rate of column C (< 3.66 mL/min) is faster than other columns

and all the column reactors were using wastewater from the same batch, it is likely that the composition of EfOM in the effluent wastewater (effluent supplied for column A after 1500 BV and column C after 2000 BV) is hindering the removal of clindamycin. After 2000 bed volumes, all columns are able to remove 50% - 70% of clindamycin. The PNEC level found for clindamycin is 0.0114 µg/L (Kisielius et al., 2023) and only column C can reduce the clindamycin concentration to below PNEC level after 2000 bed volumes, even though after 500 BV also one effluent sample from column C occurred which exceeded the PNEC.

Erythromycin (column experiments):

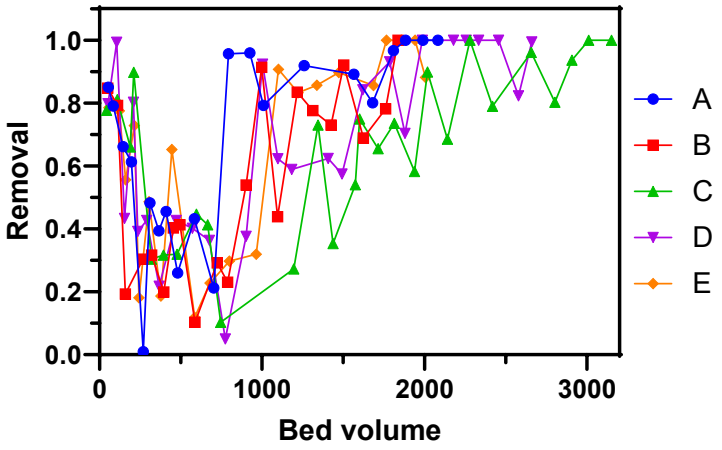

Figure 13: Removal of erythromycin in column experiment over bed volume.

Table 19: Concentration of erythromycin after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Erythromycin	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	0.0051	0.028	0.0033	0.0019	<0.001
B (Wood)	0.0051	0.036	0.0031	0.0017	<0.001
C (Wood/Sludge)	0.0059	0.030	0.033	0.0072	0.0018
D (Sludge/Wood)	<0.001	0.022	0.0051	0.0075	<0.001
E (Sludge)	0.0055	0.020	0.033	<0.001	<0.001

The removal of erythromycin in all columns decreased to 10% and then recovered to 90% in the first 1000 bed volumes, indicated that biofilm formed in that period can treat erythromycin and biodegradation was involved after 1000 bed volumes. The removal gradually increased after 1000 bed volumes and stabilized after 2000 bed volumes (~99% removal) except for column C required

more time to become stabilized due to flow issue. The PNEC level found for erythromycin is 0.04 µg/L (Kisielius et al., 2023) and all columns can reduce the erythromycin concentration to below PNEC level after 2000 bed volumes.

Clarithromycin (column experiments):

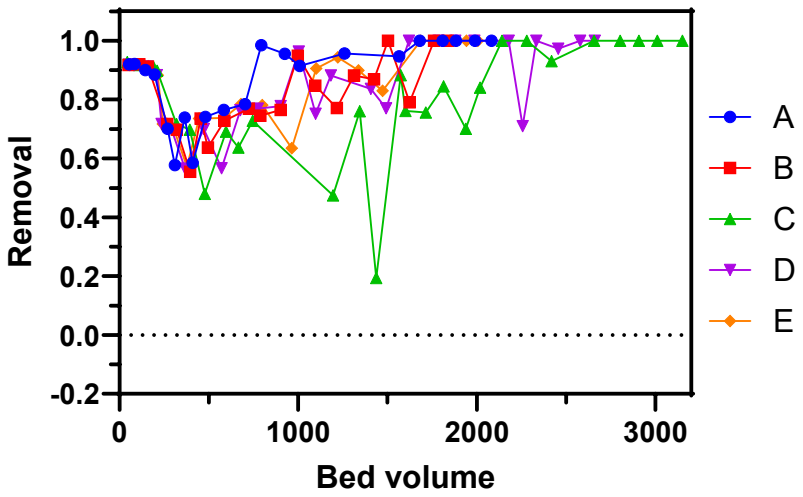

Figure 14:Removal of clarithromycin in column experiment over bed volume.

Table 20: Concentration of clarithromycin after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Clarithromycin	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.02	<0.02	<0.02	<0.02	<0.02
B (Wood)	<0.02	<0.02	<0.02	<0.02	<0.02
C (Wood/Sludge)	<0.02	<0.02	0.027	<0.02	<0.02
D (Sludge/Wood)	<0.02	<0.02	<0.02	<0.02	<0.02
E (Sludge)	<0.02	<0.02	0.023	<0.02	<0.02

The removal of clarithromycin in all columns decreased to 50% and then recovered to ~99% in the first 1000 bed volumes (except column C and E), indicated that biofilm formed in that period can treat clarithromycin and biodegradation was involved after 1000 bed volumes. The removal gradually increased after 1000 bed volumes and stabilized after 2000 bed volumes (~99% removal)

except for column C required more time to become stabilized. The PNEC level found for clarithromycin is 0.06 µg/L (Kisielius et al., 2023) and all columns can reduce clarithromycin concentration to below PNEC level after 2000 bed volumes.

All the materials are unable to achieve 80% removal of clarithromycin in the first 1000 bed volumes due to the development of biofilm. After 1000 bed volumes, all the materials can remove ~99% clarithromycin which satisfied the requirement of the revised urban wastewater directive.

Bicalutamid (column experiments):

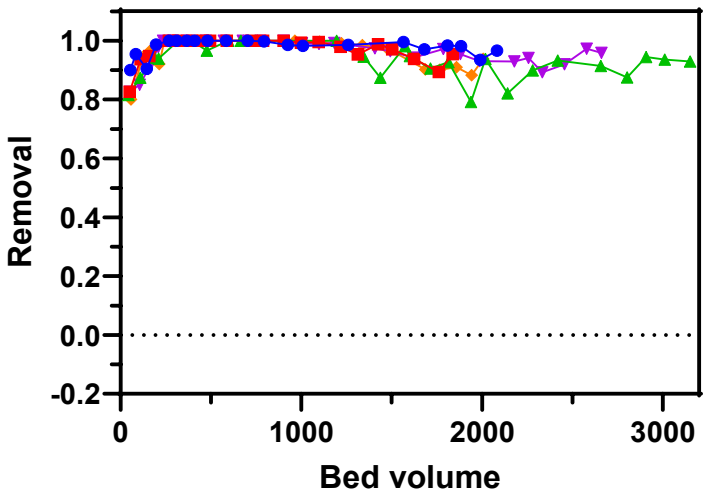

Figure 15:Removal of bicalutamid in column experiment over bed volume.

Table 21: Concentration of bicalutamid after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Bicalutamid	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.01	<0.01	<0.01	<0.01	<0.01
B (Wood)	<0.01	<0.01	<0.01	<0.01	<0.01
C (Wood/Sludge)	<0.01	<0.01	<0.01	<0.01	0.010
D (Sludge/Wood)	<0.01	<0.01	<0.01	<0.01	<0.01
E (Sludge)	<0.01	<0.01	<0.01	<0.01	<0.01

The removal of bicalutamid in all columns started from ~80% and then increased to ~99% in the first 250 bed volumes. The removal remained stable to 1500 bed volumes then the removal became less stable but remained >80%. The PNEC level found for bicalutamid is 0.1 µg/L (Kisielius et al.,

2023) and the average concentration of it in the reactor inflow is 0.125 µg/L. All columns can reduce bicalutamid concentration to below PNEC level after 2000 bed volumes.

All the materials can remove ~99% bicalutamid which satisfied the requirement of the revised urban wastewater directive.

Diclofenac (column experiments):

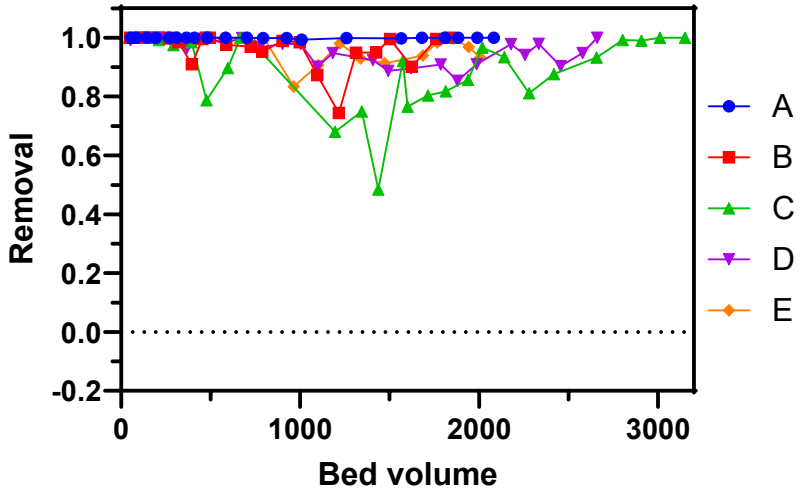

Figure 16:Removal of diclofenac in column experiment over bed volume.

Table 22: Concentration of diclofenac after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Diclofenac	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.01	<0.01	<0.01	<0.01	<0.01
B (Wood)	<0.01	<0.01	0.02	0.038	<0.01
C (Wood/Sludge)	<0.01	0.08	0.13	0.025	0.049
D (Sludge/Wood)	<0.01	<0.01	0.02	0.08	0.025
E (Sludge)	<0.01	<0.01	0.12	0.01	0.019

The removal of diclofenac in all columns are stable at ~99% in the first 1000 bed volumes except the removal in column C was once decreased to 80% and was recovered to ~99% in 400 – 600 BV. After 1000 bed volume, the removal in column A was stable throughout the experiment period (~2000 BV) and the removal in other columns were less stable but kept at ~90%. The PNEC level

found for diclofenac is 0.1 µg/L (suggested concentration received by Aarhus Vand) and all columns can reduce diclofenac concentration to below PNEC level after 2000 bed volumes.

All the materials (except column C due to flow issue) are able to achieve 80% removal of diclofenac which satisfied the requirement of the revised urban wastewater directive.

Sertraline (column experiments):

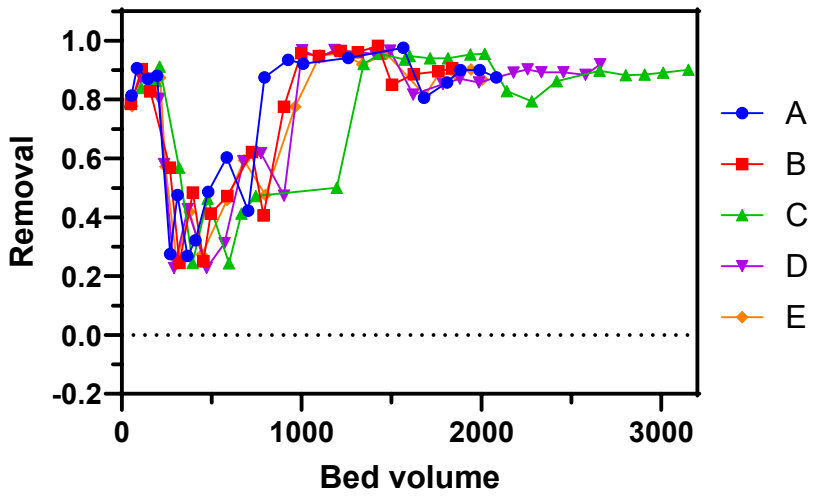

Figure 17: Removal of sertraline in column experiment over bed volume.

Table 23: Concentration of sertraline after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Sertraline	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.01	0.017	<0.01	<0.01	0.010
B (Wood)	<0.01	0.018	<0.01	0.010	0.010
C (Wood/Sludge)	<0.01	0.018	0.018	<0.01	<0.01
D (Sludge/Wood)	<0.01	0.019	<0.01	<0.01	0.010
E (Sludge)	<0.01	0.018	0.018	<0.01	0.013

The removal rate for all columns decreased in the beginning (300-700BV) and then recovered to close to 100% after the first 1000 bed volumes. This simultaneous behaviour could be explained by the development of biofilms on the activated carbons as the such development is regardless of

the type of material (Paredes et al., 2016). The formation of biofilms could be caused by the microorganisms existed in the effluent wastewater and the microorganisms were colonizing the activated carbon during the operation period (Gibert et al., 2013). The PNEC level found for marine water is 0.1 µg/L (Pivetta et al., 2020) and all the activated carbon is able to demonstrate >95% removal after 1000 bed volumes.

Metoprolol (column experiments):

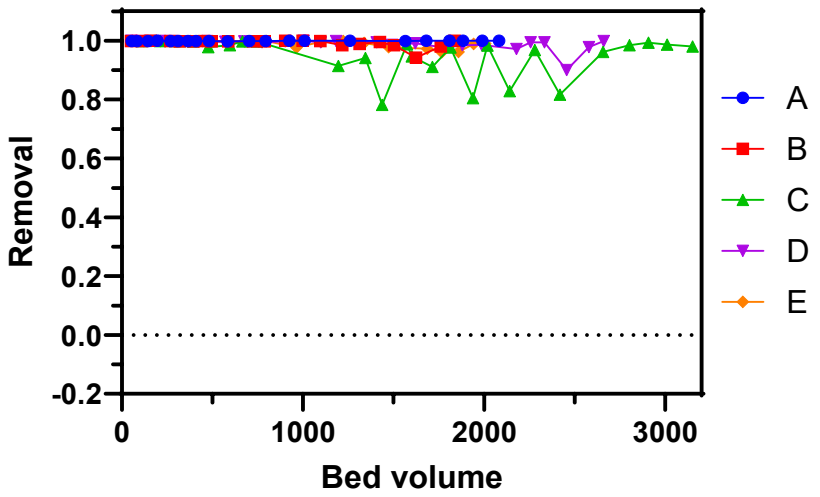

Figure 18: Removal of metoprolol in column experiment over bed volume.

Table 24: Concentration of metoprolol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Metoprolol	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.005	<0.005	<0.005	<0.005	<0.005
B (Wood)	<0.005	<0.005	<0.005	0.0127	<0.005
C (Wood/Sludge)	<0.005	0.014	0.035	0.024	0.018
D (Sludge/Wood)	<0.005	<0.005	<0.005	0,016	0.010
E (Sludge)	<0.005	<0.005	0.027	0.014	0.0073

The removal of metoprolol in column A are stable at ~99% throughout the experimental period (~2000 BV). The removal in other columns is able to achieve ~99% with less stable performance (especially column C with known flow issue). Since all the columns can maintain ~99% removal at the last sampling point, it indicates none of the column are close to the capacity of sorbing

metoprolol. The average concentration of metoprolol in effluent wastewater is 0.98 µg/L which is higher than the PNEC (0.1 µg/L) (Kisielius et al., 2023). As the columns can remove ~99% of the metoprolol from effluent wastewater, all columns can treat metoprolol to below PNEC level after 2000 bed volumes.

All the materials (except column C due to flow issue) can remove ~99% metoprolol which satisfied the requirement of the revised urban wastewater directive. For vast period of the experimental period, column C also meets the requirement with only four samples having less than 80% removal against metoprolol.

Propranolol (column experiments):

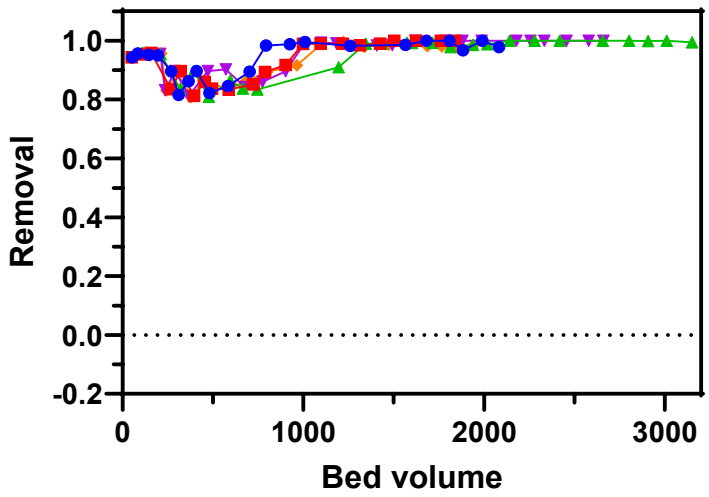

Figure 19: Removal of propranolol in column experiment over bed volume.

Table 25: Concentration of propranolol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Propranolol	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.003	0.0041	<0.003	<0.003	<0.003
B (Wood)	<0.003	0.0040	<0.003	<0.003	<0.003
C (Wood/Sludge)	<0.003	0.0039	0.0038	<0.003	<0.003
D (Sludge/Wood)	<0.003	0.0038	<0.003	<0.003	<0.003
E (Sludge)	<0.003	0.0038	0.0039	<0.003	<0.003

The removal of propranolol in all columns started at ~92% in the first 300 bed volumes and decreased to 80% afterwards within the first 1000 bed volume. After 1000 bed volume, the removal in all columns is stable throughout the experiment period (~3000 BV). The PNEC level found for

propranolol is 0.1 µg/L (suggested concentration received by Aarhus Vand) and all columns can reduce propranolol concentration to below PNEC level throughout the experimental period.

Venlafaxine (column experiments):

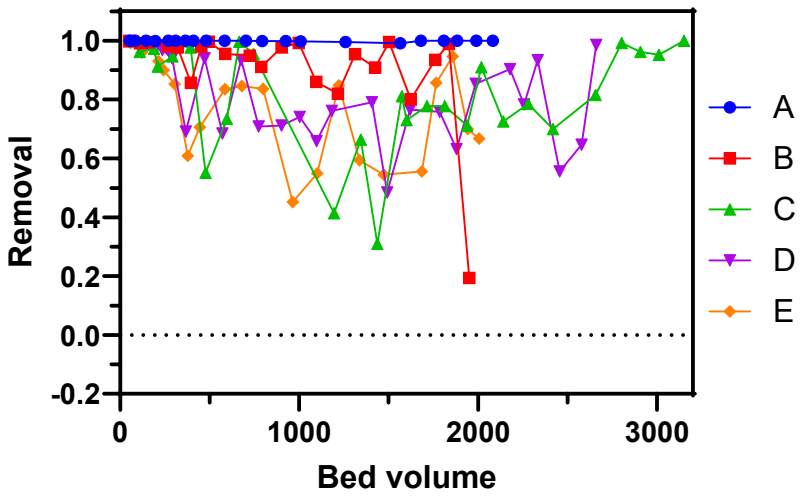

Figure 20: Removal of venlafaxine in column experiment over bed volume.

Table 26: Concentration of venlafaxine after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Venlafaxine	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.005	<0.005	<0.005	<0.005	<0.005
B (Wood)	<0.005	<0.005	0.007	0.046	0.075
C (Wood/Sludge)	0.018	0.059	0.032	0.046	0.119
D (Sludge/Wood)	<0.005	0.063	0.19	0.26	0.063
E (Sludge)	0.019	0.066	0.27	0.09	0.14

The removal of venlafaxine in column A are stable at ~99% throughout the experimental period (~2000 BV). The removal in other columns is varying between ~20% to ~99% after the first 200 bed volumes. Since the removal in column C and D is able to recover to ~99%, the varying in removal is mainly due to the EfOM in the wastewater rather than the limitation of capacity. The

average concentration of venlafaxine in effluent wastewater is 0.46 µg/L which is higher than the PNEC (0.1 µg/L, suggested concentration received by Aarhus Vand). At 2000 bed volumes, column C and E are unable to treat venlafaxine to below PNEC level while the other columns can.

For the vast period of the experiment, all the biochars (excluding the commercial activated carbon) are unable to achieve 80% removal of venlafaxine which did not meet the requirement of the revised urban wastewater directive. On the other hand, commercial activated carbon (column A) maintained stable removal (~99%) in mitigating venlafaxine in inflow wastewater.

Sulfamethoxazole (column experiments):

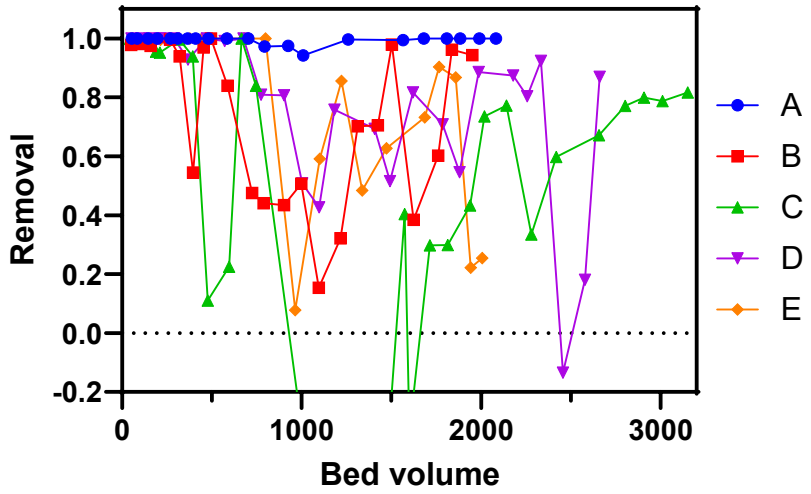

Figure 21: Removal of sulfamethoxazole in column experiment over bed volume.

Table 27: Concentration of sulfamethoxazole after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Sulfamethoxazole	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.003	<0.003	0.0050	<0.003	<0.003
B (Wood)	<0.003	<0.003	0.033	<0.003	<0.003
C (Wood/Sludge)	<0.003	0.032	0.030	0.051	0.025
D (Sludge/Wood)	<0.003	<0.003	0.037	0.045	0.028
E (Sludge)	<0.003	<0.003	0.025	0.016	0.072

The removal of sulfamethoxazole in column A are stable at ~99% (SD = 0.014) throughout the experimental period (~2000 BV) while the removal in other columns is varying between ~10% to ~95% after the first 200 bed volumes. Column C had the least stable performance (minimum removal is -122%) which is most likely due to flow issue in that column. Since the removal in

column C and D is able to recover to ~80% rather than 99%, the capacity of biochar may start to exhaust after 2500 bed volumes. Column A did not show any sign of exhaustion as its current bed volume is only 2000. The average concentration of sulfamethoxazole in effluent wastewater is 0.06 µg/L which is lower than the PNEC (0.12 µg/L, suggested concentration received by Aarhus Vand). All columns are able to treat sulfamethoxazole to below PNEC level throughout the column experiment.

Amisulpride (column experiments):

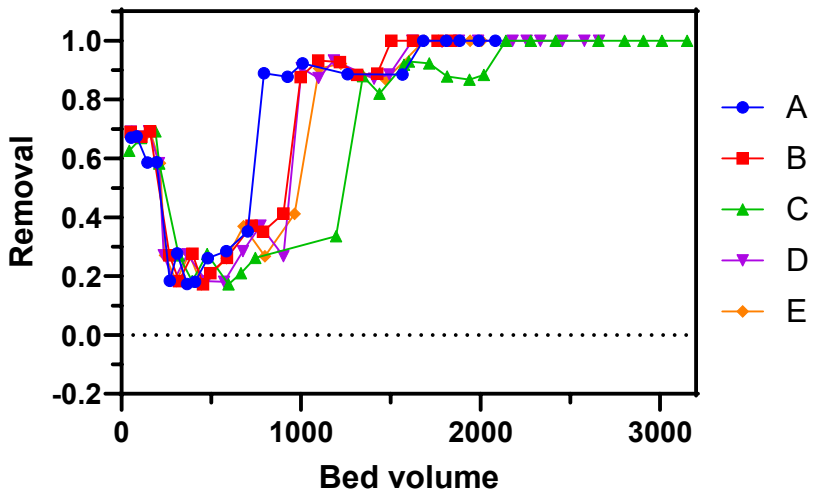

Figure 22: Removal of amisulpride in column experiment over bed volume.

Table 28: Concentration of amisulpride after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Amisulpride	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	0.045	0.089	0.0092	0.0093	<0.001
B (Wood)	0.045	0.089	0.0093	<0.001	<0.001
C (Wood/Sludge)	0.045	0.089	0.089	0.0096	0.0094
D (Sludge/Wood)	0.045	0.089	0.0093	0.0093	<0.001
E (Sludge)	0.045	0.089	0.089	0.0094	<0.001

The removal of amisulpride in all columns maintained at ~70% in the first 300 bed volumes and decreased to ~18% afterwards within the first 700 bed volume. The removal after 700 BV in all columns increased to ~90% at 1300 BV then further increased to ~99% at 1500 BV. Column C took longer time to reach ~99% removal due to flow issue. The graph indicates the biofilm formed

in the first 1000 BV participated in the biodegradation of amisulpride. The PNEC level found for amisulpride is 0.17 µg/L (suggested concentration received by Aarhus Vand) and all columns can reduce amisulpride concentration to below PNEC level throughout the experimental period.

All the activated carbon are unable to achieve 80% removal of amisulpride in the first 1500 bed volumes which did not meet the requirement of the revised urban wastewater directive. After 1500 bed volume, all the activated carbon has 80% removal in mitigating amisulpride in inflow wastewater which meets the directive.

Carbamazepine (column experiments):

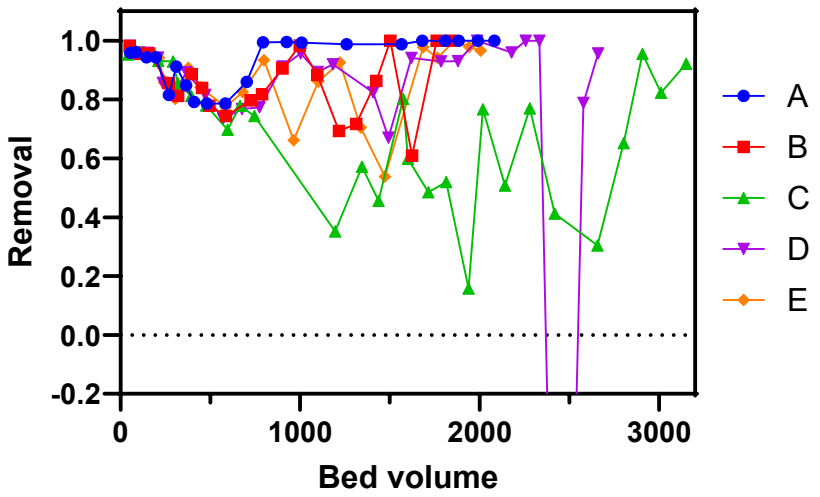

Figure 23: Removal of carbamazepine in column experiment over bed volume.

Table 29: Concentration of carbamazepine after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Carbamazepine	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.003	0.0038	<0.003	<0.003	<0.003
B (Wood)	<0.003	0.0039	<0.003	<0.003	<0.003
C (Wood/Sludge)	<0.003	0.0096	0.019	0.018	0.013
D (Sludge/Wood)	<0.003	0.0044	0.0037	0.018	<0.003
E (Sludge)	<0.003	0.0039	0.015	0.020	<0.003

The removal of carbamazepine in all columns started at ~95% and gradually decreased to ~70% in the first 500 bed volumes. The removal in column A recovered to ~99% and maintained throughout the experimental period. Other columns (except column C) have less stable removal (~50% - ~99%). Column C took longer time to reach >90% removal due to flow issue. The graph

indicates the biofilm formed in the first 1000 BV participated in the biodegradation of carbamazepine. The PNEC level found for carbamazepine is 0.5 µg/L (suggested concentration received by Aarhus Vand) and all columns can reduce carbamazepine concentration to below PNEC level throughout the experimental period.

All the activated carbon are able to achieve 80% removal of carbamazepine in the first 300 bed volumes which meets the requirement of the revised urban wastewater directive. After 300 bed volumes, all the biochar has varying removal in carbamazepine (<80%) and thus did not satisfy the requirement of the directive. The commercial activated carbon has >80% removal after the biofilm was developed (after 700 bed volumes).

Citalopram (column experiments):

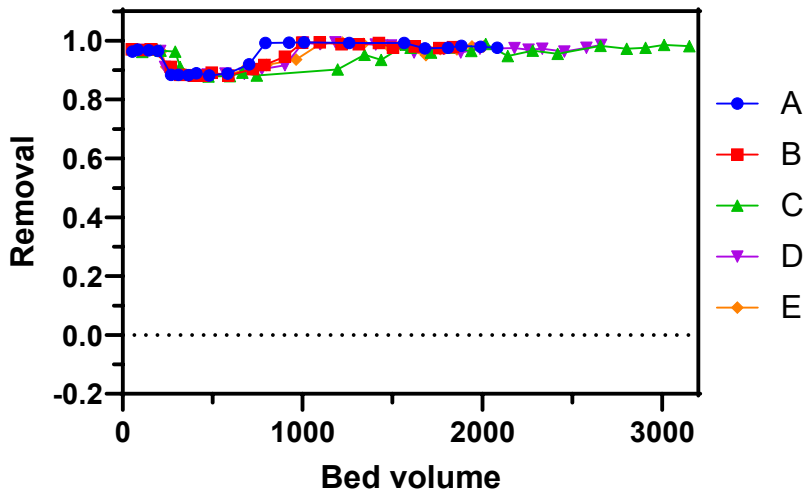

Figure 24: Removal of citalopram in column experiment over bed volume.

Table 30: Concentration of citalopram after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Citalopram	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	0.0074	0.014	<0.003	0.0018	0.0038
B (Wood)	0.0077	0.015	<0.003	0.0036	0.0044
C (Wood/Sludge)	0.0077	0.015	0.016	0.0046	<0.003
D (Sludge/Wood)	0.0076	0.015	<0.003	0.0039	0.0048
E (Sludge)	0.0076	0.015	0.017	0.0028	0.0050

The removal of citalopram in all columns started at ~98% and gradually decreased to ~88% in the first 750 bed volumes. The removal was then gradually recovered (with column A recovered first) to ~99% and maintained at this removal at all times. The graph indicates the biofilm formed in the first 1000 BV participated in the biodegradation of citalopram. The PNEC level found for

citalopram is 0.51 µg/L (suggested concentration received by Aarhus Vand) and all columns can reduce citalopram concentration to below PNEC level throughout the experimental period. All the materials can remove >80% citalopram which satisfied the requirement of the revised urban wastewater directive.

Cetirizine (column experiments):

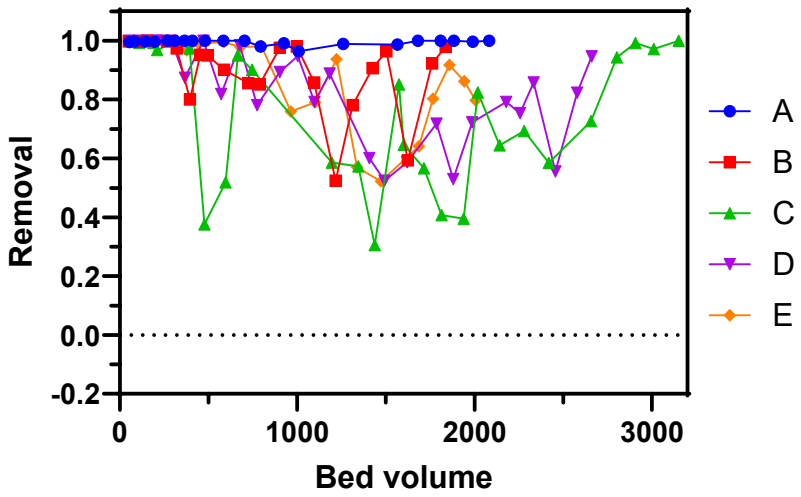

Figure 25: Removal of cetirizine in column experiment over bed volume.

Table 31: Concentration of cetirizine after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Cetirizine	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.003	<0.003	0.052	0.0054	<0.003
B (Wood)	<0.003	0.0056	0.020	0.0096	0.013
C (Wood/Sludge)	<0.003	0.082	0.11	0.21	0.073
D (Sludge/Wood)	<0.003	0.0089	0.048	0.20	0.056
E (Sludge)	<0.003	<0.003	0.16	0.13	0.040

The removal of cetirizine in all columns started at ~99%. Column A maintained ~99% removal of cetirizine throughout the experimental period (~2000 BV). The removal in other columns is varying between ~30% to ~99% after the first 400 bed volumes. Since the removal in column C

and D is able to recover to >90%, the varying in removal is mainly due to the EfOM in the wastewater rather than the limitation of capacity. The average concentration of cetirizine in effluent wastewater is 0.41 µg/L which is lower than the PNEC (0.52 µg/L) (Kisielius et al., 2023). All columns can reduce cetirizine concentration to below PNEC level.

Tramadol (column experiments):

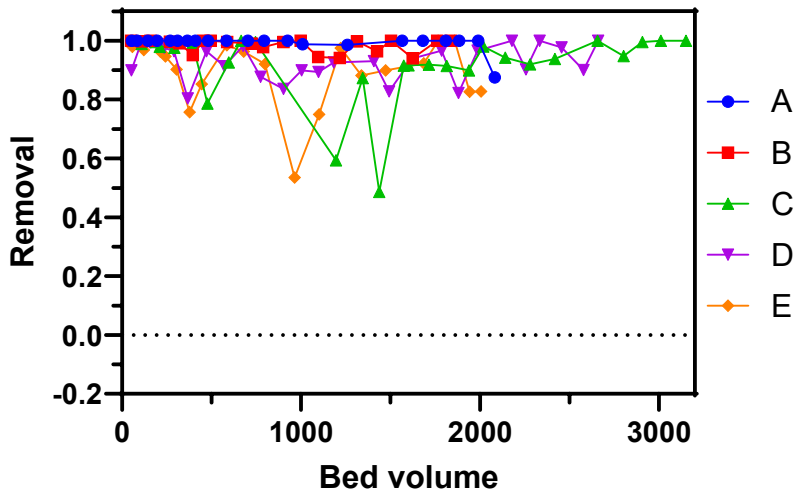

Figure 26: Removal of tramadol in column experiment over bed volume.

Table 32: Concentration of tramadol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Tramadol	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.05	<0.05	<0.05	<0.05	<0.05
B (Wood)	<0.05	<0.05	<0.05	<0.05	<0.05
C (Wood/Sludge)	<0.05	<0.05	<0.05	<0.05	<0.05
D (Sludge/Wood)	<0.05	<0.05	0.079	0.10	<0.05
E (Sludge)	<0.05	<0.05	0.10	<0.05	<0.05

The removal of tramadol in all columns started at ~99%. Column A maintained ~99% removal of tramadol throughout the experimental period (~2000 BV). The removal in other columns is varying between ~50% to ~99% after the first 400 bed volumes. Column C being the least stable column due to its flow issue. Since the removal in column B, C and D is able to recover to >95%, the

varying in removal is mainly due to the EfOM in the wastewater rather than the limitation of capacity. The average concentration of tramadol in effluent wastewater is 0.47 µg/L which is lower than the PNEC (2.25 µg/L) (Kisielius et al., 2023). All columns can achieve 90% removal of tramadol and reduce its concentration to below PNEC level.

Sulfamethizole (column experiments):

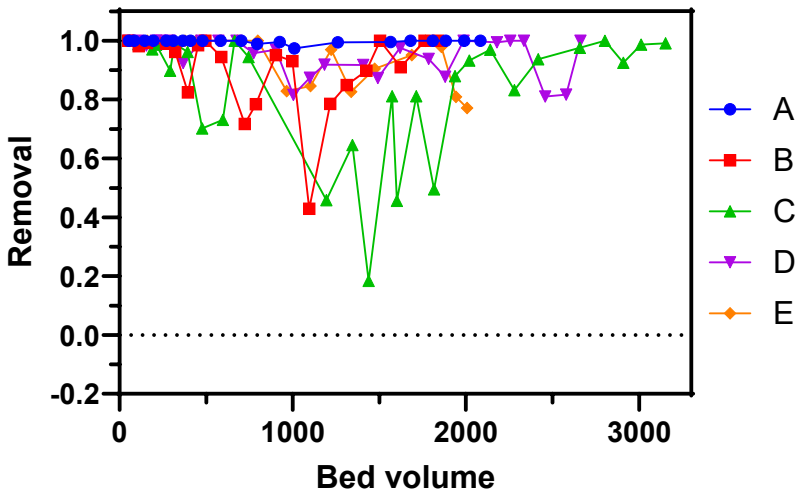

Figure 27: Removal of sulfamethizole in column experiment over bed volume.

Table 33: Concentration of sufamethizole after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Sulfamethizole	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.001	<0.001	0.0046	<0.001	<0.001
B (Wood)	0.0020	<0.001	0.015	<0.001	0.018
C (Wood/Sludge)	0.0011	0.014	0.018	0.034	0.010
D (Sludge/Wood)	<0.001	0.0027	0.023	0.020	<0.001
E (Sludge)	<0.001	<0.001	0.033	<0.001	0.028

The removal of sulfamethizole in all columns started at ~99%. Column A maintained ~99% removal of sulfamethizole throughout the experimental period (~2000 BV). The removal in other columns is varying between ~40% to ~99% after the first 300 bed volumes. Column C being the least stable column due to its flow issue. Since the removal in column B, C and D is able to recover to >95%, the varying in removal is mainly due to the EfOM in the wastewater rather than the limitation of capacity. The average concentration of sulfamethizole in effluent wastewater is 0.11 µg/L which is lower than the PNEC (2.54 µg/L) (Kisielius et al., 2023). All columns can achieve 90% removal of sulfamethizole and reduce its concentration to below PNEC level.

Mefenamic acid (column experiments):

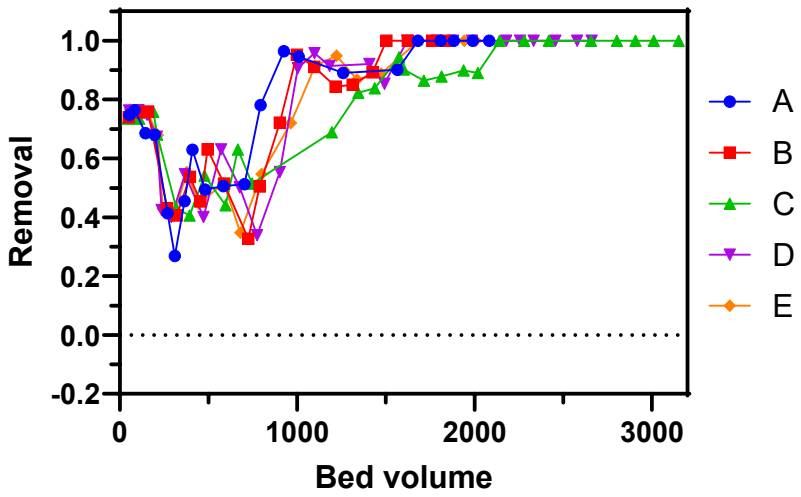

Figure 28: Removal of mefenamic acid in column experiment over bed volume.

Table 34: Concentration of mefenamic acid after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Mefenamic acid	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	0.0024	0.0051	<0.001	<0.001	<0.001
B (Wood)	0.0025	0.0061	0.0010	<0.001	<0.001
C (Wood/Sludge)	0.0026	0.0048	0.0048	<0.001	<0.001
D (Sludge/Wood)	0.0024	0.0046	0.0012	<0.001	<0.001
E (Sludge)	0.0025	0.0048	0.0048	<0.001	<0.001

The removal of mefenamic acid in all columns started at ~78% and gradually decreased to ~40% in the first 200 bed volumes. The removal was then varying between 40% to 60% in the range of 200 – 700 BV. All columns gradually recovered (with column A recovered first and column C being the last due to flow issue) to ~99% at 1500 bed volumes and maintained at this removal at

all times. The graph indicates the biofilm formed in the first 1000 BV participated in the biodegradation of mefenamic acid. The PNEC level found for mefenamic acid 3.9 µg/L (Kisielius et al., 2023) and all columns can achieve >99% removal of mefenamic acid and reduce its concentration to below PNEC level.

Sulfadiazine (column experiments):

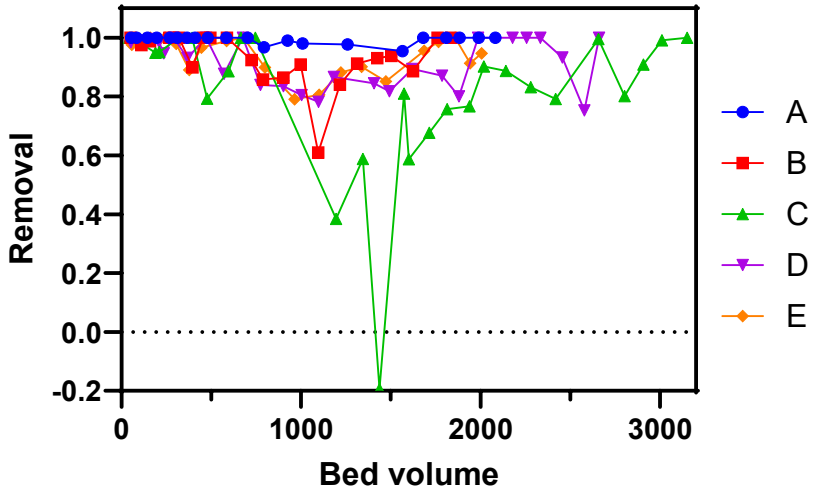

Figure 29: Removal of sulfadiazine in column experiment over bed volume.

Table 35: Concentration of sulfadiazine after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Sulfadiazine	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.003	<0.003	<0.003	<0.003	<0.003
B (Wood)	<0.003	<0.003	0.0036	<0.003	0.0033
C (Wood/Sludge)	<0.003	<0.003	0.0053	0.0079	0.0033
D (Sludge/Wood)	<0.003	<0.003	0.0078	0.0062	<0.003
E (Sludge)	<0.003	<0.003	0.0046	0.0031	<0.003

The removal of sulfadiazine in all columns started at ~99%. Column A maintained ~99% removal throughout the experimental period (~2000 BV). The removal in other columns is varying between ~60% to ~99% after the first 300 bed volumes. Column C being the least stable column (removal

varied from -20% to ~99%) due to its flow issue. Since the removal in column B, C, D and E is able to recover to >95%, the varying in removal is mainly because of the EfOM in the wastewater rather than the limitation of capacity. The average concentration of sulfadiazine in effluent wastewater is 0.026 µg/L which is lower than the PNEC (4.6 µg/L) (Kisielius et al., 2023). All columns can achieve 95% removal of sulfadiazine and reduce its concentration to below PNEC level.

4-Methyl-1H-benzotriazole (column experiments):

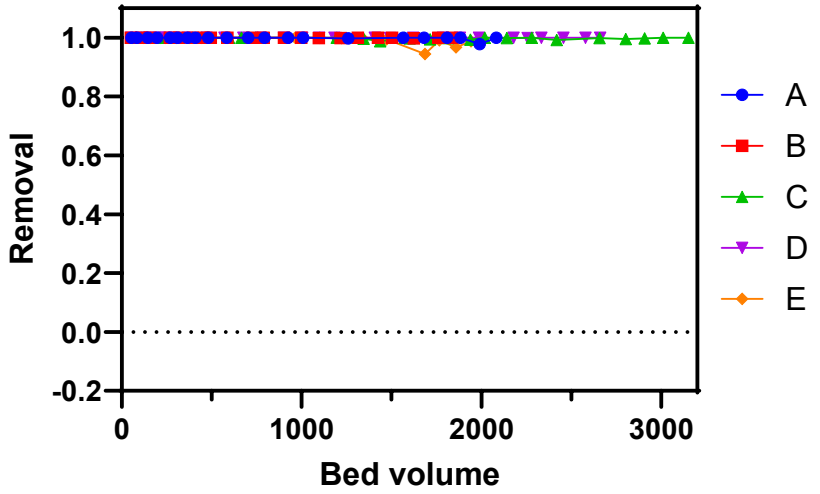

Figure 30: Removal of 4-methyl-1H-benzotriazole in column experiment over bed volume.

Table 36: Concentration of 4-methyl-1H-benzotrizole after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

4-Methyl-1H-benzotriazole	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.01	<0.01	<0.01	<0.01	0.083
B (Wood)	<0.01	<0.01	<0.01	<0.01	0.012
C (Wood/Sludge)	<0.01	<0.01	<0.01	<0.01	<0.01
D (Sludge/Wood)	<0.01	<0.01	<0.01	<0.01	<0.01
E (Sludge)	<0.01	<0.01	<0.01	<0.01	0.024

The removal of 4-methyl-1H-benzotriazole in all columns started and maintained at ~99% throughout the experimental period. All columns can effectively remove 4-methyl-1H-benzotriazole to below its PNEC level (8 µg/L) (NORMAN, 2024). All the materials can remove

>80% 4-methyl-1H-benzotriazole which satisfied the requirement of the revised urban wastewater directive.

Trimethoprim (column experiments):

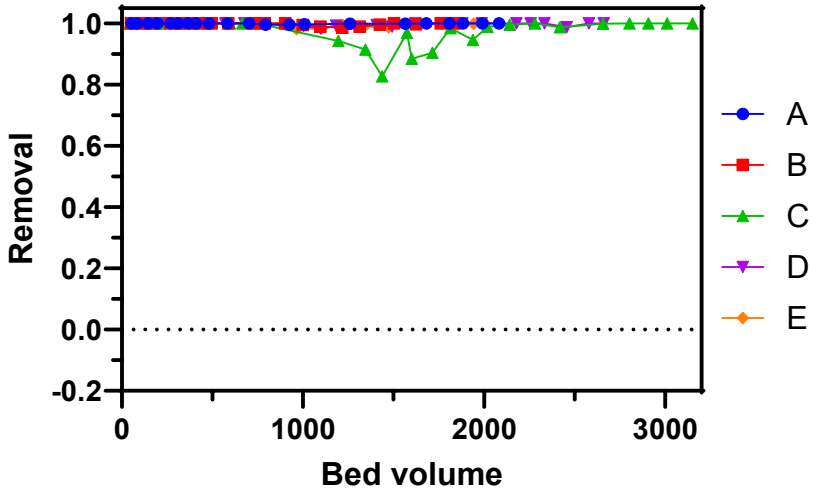

Figure 31: Removal of trimethoprim in column experiment over bed volume.

Table 37: Concentration of trimethoprim after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Trimethoprim	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.001	<0.001	<0.001	<0.001	<0.001
B (Wood)	<0.001	<0.001	<0.001	<0.001	<0.001
C (Wood/Sludge)	<0.001	<0.001	0.0014	0.0032	<0.001
D (Sludge/Wood)	<0.001	<0.001	0.0011	0.0018	<0.001
E (Sludge)	<0.001	<0.001	0.0015	0.0011	<0.001

The removal of trimethoprim in all columns started and maintained at ~99% throughout the experimental period with column C had less stable removal due to flow issue. All columns can effectively remove trimethoprim to below its PNEC level (10 µg/L) (Kisielius et al., 2023).

Benzotriazole (column experiments):

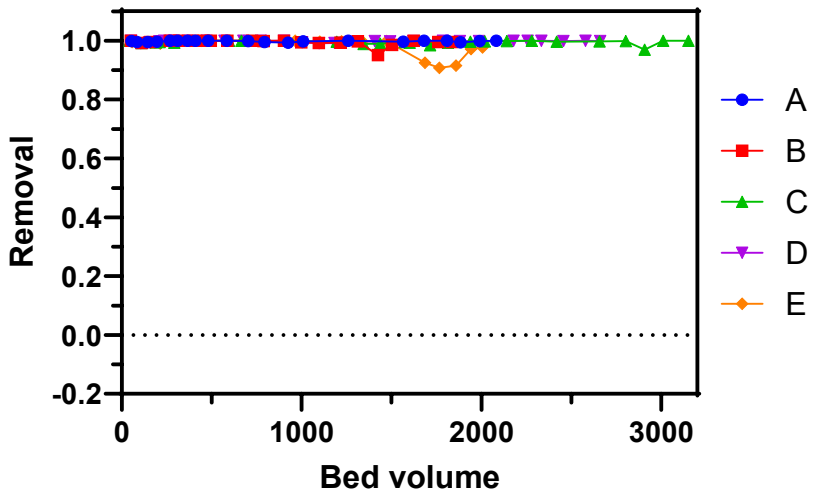

Figure 32: Removal of benzotriazole in column experiment over bed volume.

Table 38: Concentration of benzotriazole after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Benzotriazole	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	0.037	<0.01	0.021	0.017	<0.01
B (Wood)	0.052	<0.01	0.025	0.12	<0.01
C (Wood/Sludge)	0.041	<0.01	<0.01	0.031	0.014
D (Sludge/Wood)	0.058	<0.01	0.022	0.016	0.013
E (Sludge)	0.039	<0.01	<0.01	<0.01	0.19

The removal of benzotriazole in all columns started and maintained at ~99% throughout the experimental period. All columns can effectively remove benzotriazole to below its PNEC level

(19 µg/L) (NORMAN, 2024). All the materials can remove >80% benzotriazole which satisfied the requirement of the revised urban wastewater directive.

Furosemide (column experiments):

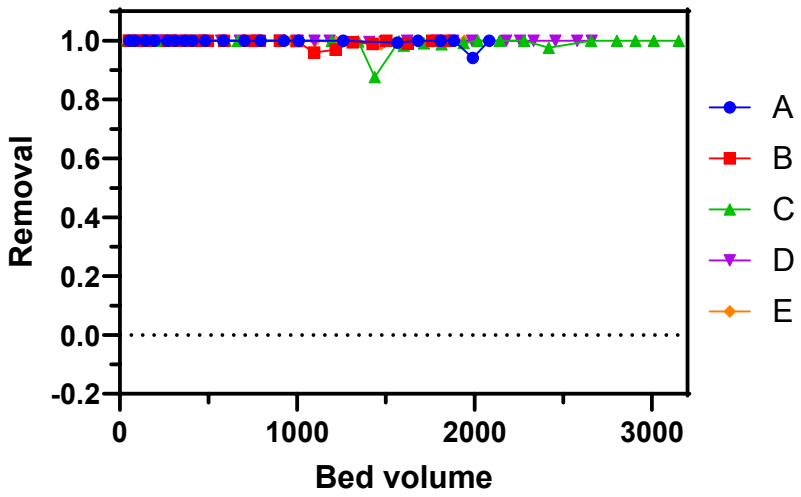

Figure 33: Removal of furosemide in column experiment over bed volume.

Table 39: Concentration of furosemide after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Furosemide	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.03	<0.03	<0.03	<0.03	0.041
B (Wood)	<0.03	<0.03	<0.03	<0.03	<0.03
C (Wood/Sludge)	<0.03	<0.03	<0.03	<0.03	<0.03
D (Sludge/Wood)	<0.03	<0.03	<0.03	<0.03	<0.03
E (Sludge)	<0.03	<0.03	<0.03	<0.03	<0.03

The removal of furosemide in all columns started and maintained at ~99% throughout the experimental period. All columns can effectively remove furosemide to below its PNEC level (31 µg/L) (Kisielius et al., 2023).

Gabapentin (column experiments):

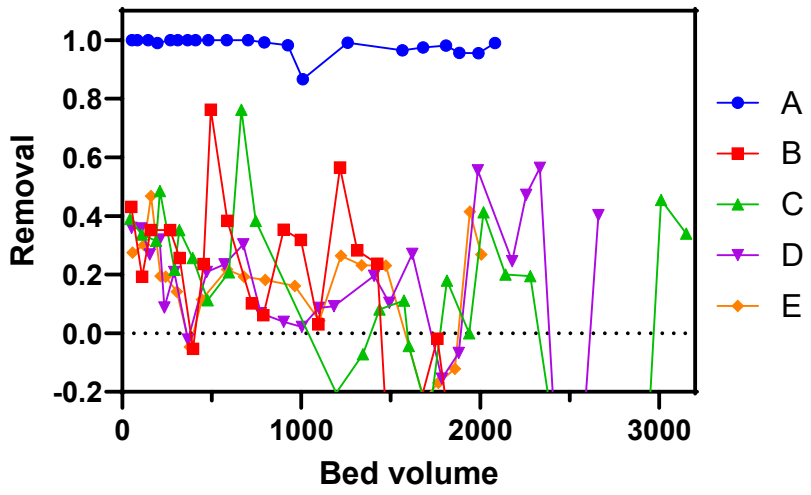

Figure 34: Removal of gabapentin in column experiment over bed volume.

Table 40: Concentration of gabapentin after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Gabapentin	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.003	<0.003	0.098	0.039	0.019
B (Wood)	1.7	0.14	0.63	0.84	0.97
C (Wood/Sludge)	1.3	0.43	0.46	0.65	0.66
D (Sludge/Wood)	1.4	0.49	0.74	1.0	0.41
E (Sludge)	1.5	0.50	0.39	0.82	0.70

The removal of gabapentin in column A started at ~99% and varied to a small degree (average: 98%, SD =0.03) over the experimental period (~2000 BV). The removal in other columns was being unstable (ranging from negative to ~80%) at all times. Since the removal in column A (commercial activated carbon) is much higher than the others (biochar), gabapentin has more

affinity towards the active site in the commercial carbon than in the biochar. The average concentration of gabapentin in effluent wastewater is 0.98 µg/L which is lower than the PNEC (100 µg/L) (Kisielius et al., 2023). Despite the biochar is having unstable in removing gabapentin, it can reduce its concentration to below PNEC level.

Atenolol (column experiments):

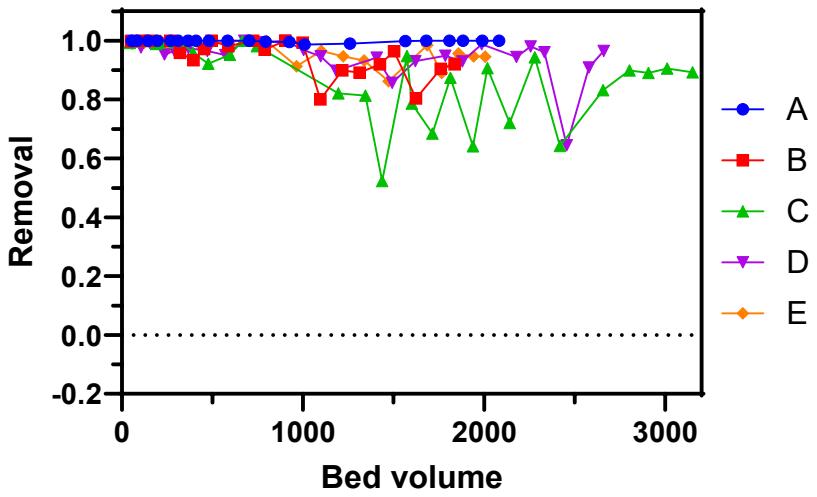

Figure 35: Removal of atenolol in column experiment over bed volume.

Table 41: Concentration of atenolol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Atenolol	**100 BV**	**500 BV**	**1000 BV**	**1500 BV**	**2000 BV**
A (Brennsorb 1240)	<0.005	<0.005	<0.005	<0.005	<0.005
B (Wood)	<0.005	<0.005	<0.005	<0.005	<0.005
C (Wood/Sludge)	<0.005	<0.005	<0.005	0.0059	0.012
D (Sludge/Wood)	<0.005	<0.005	<0.005	0.018	<0.005
E (Sludge)	<0.005	<0.005	0.0067	0.015	0.0056

The removal of atenolol in all columns started at ~99%. Column A maintained ~99% removal of atenolol throughout the experimental period (~2000 BV). The removal in other columns is varying between ~60% to ~99%. Column C being the least stable column due to its flow issue. Since the removal in column B, C, D and E is able to recover to >95%, the varying in removal is mainly due

to the EfOM in the wastewater rather than the limitation of capacity. The average concentration of atenolol in effluent wastewater is 0.08 µg/L which is lower than the PNEC (128 µg/L) (Kisielius et al., 2023). All columns can achieve >80% removal of atenolol at the last sampling time point.

Losartan (column experiments):

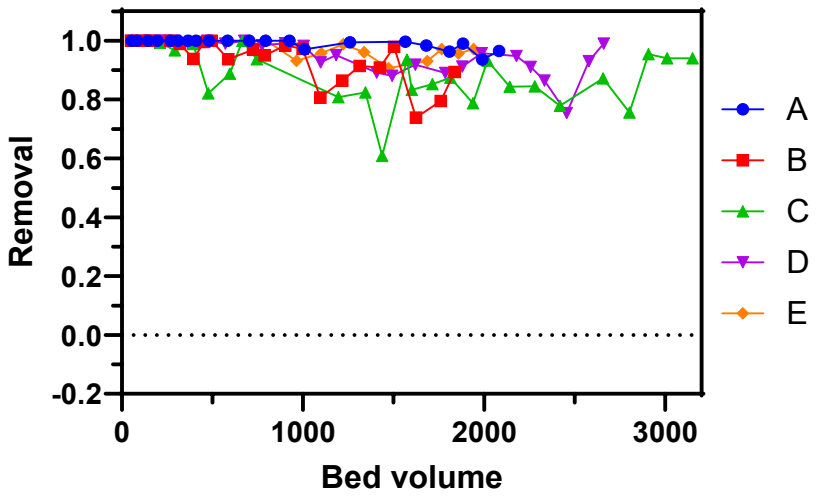

Figure 36: Removal of losartan in column experiment over bed volume.

Table 42: Concentration of losartan after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Losartan	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.003	<0.003	0.022	<0.003	0.0064
B (Wood)	<0.003	<0.003	0.024	0.0063	0.027
C (Wood/Sludge)	<0.003	0.13	0.12	0.049	0.026
D (Sludge/Wood)	<0.003	<0.003	0.020	0.045	0.015
E (Sludge)	<0.003	<0.003	0.054	0.026	0.033

The removal of losartan in all columns started at ~99%. Column A maintained ~99% removal of losartan throughout the experimental period (~2000 BV). The removal in other columns is varying between 60% to 99%. Column C being the least stable column due to its flow issue. Since the removal in column B, C, D and E is able to recover to >95%, the varying in removal is mainly due

to the EfOM in the wastewater rather than the limitation of capacity. All columns can achieve >80% removal of atenolol at the last sampling time point. The average concentration of losartan in effluent wastewater is 0.77 µg/L which is much lower than the PNEC (331 µg/L) (Kisielius et al., 2023).

Irbesartan (column experiments):

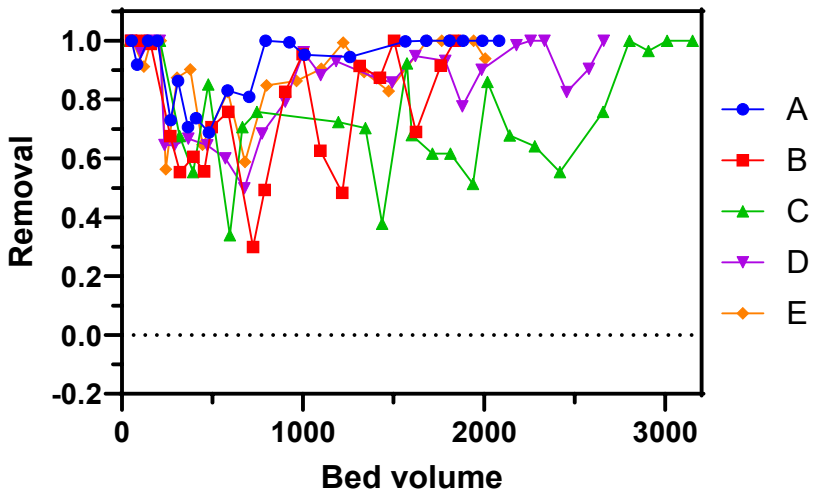

Figure 37: Removal of irbesartan in column experiment over bed volume.

Table 43: Concentration of irbesartan after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Irbesartan	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.001	0.0040	0.0036	<0.001	<0.001
B (Wood)	<0.001	0.0049	0.0023	<0.001	0.0058
C (Wood/Sludge)	<0.001	0.0021	0.0056	0.0058	0.013
D (Sludge/Wood)	<0.001	0.0043	0.0016	0.013	0.0033
E (Sludge)	<0.001	0.0033	0.0052	0.0055	0.0025

The removal of irbesartan in all columns started at ~99% and gradually decreased to ~50% in the first 500 bed volumes. The removal was then varying between 70% to 99% in the range of 700 – 3000 BV. All columns gradually recovered (with column A recovered first and column C being the last due to flow issue) to ~99% at 1800 bed volumes and maintained stable removal. The graph

indicates the biofilm formed in the first 1000 BV participated in the biodegradation of irbesartan. The PNEC level found for irbesartan 704 µg/L (Kisielius et al., 2023) and all columns can achieve >90% removal of irbesartan and reduce its concentration to below PNEC level at the last sampling point (between 2000 BV and 3200 BV, depending the column sampled).

All the activated carbon are able to achieve 80% removal of irbesartan in the first 200 bed volumes which meets the requirement of the revised urban wastewater directive. After 200 bed volumes, all the biochar (except column E, which consists of biochar made from wood pellets) has varying removal in irbesartan (<80%) and thus did not satisfy the requirement of the directive. Biochar made from wood pellets demonstrate relatively stable removal compared to other biochars and exhibited >80% removal after 700 bed volumes. The commercial activated carbon has >80% removal after the biofilm was developed (after 1000 bed volumes).

Iohexol (column experiments):

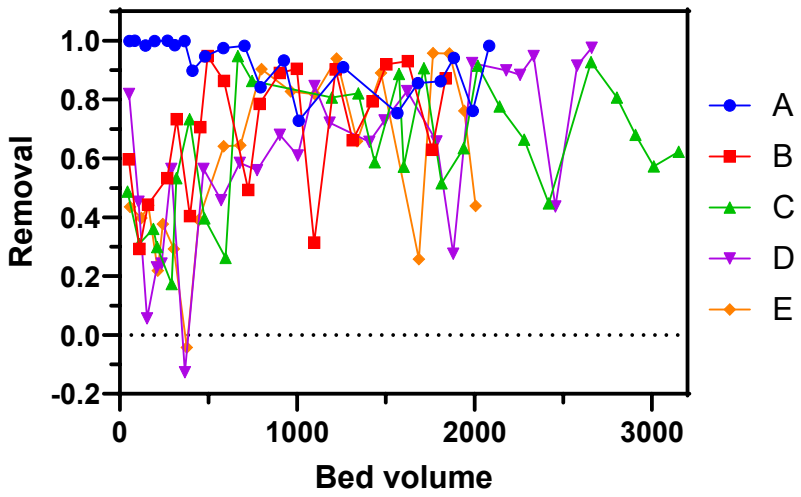

Figure 38: Removal of iohexol in column experiment over bed volume.

Table 44: Concentration of iohexol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Iohexol	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.015	0.51	1.6	0.52	0.23
B (Wood)	14	0.74	0.95	0.20	1.4
C (Wood/Sludge)	12	3.8	2.8	0.64	0.18
D (Sludge/Wood)	11	5.3	4.7	0.57	0.39
E (Sludge)	11	5.5	1.1	0.21	3.0

The removal of iohexol in column A kept at 99% in the first 500 BV and varied between 70% to 99% throughout the experimental period. On the other hand, the removal in biochars started at 90% maximum and decreased from ~50% to < 20% in the first 500 BV and gradually increased but varied between 30% to ~99% afterwards. The PNEC level found for iohexol 1 000 000 µg/L

(Kisielius et al., 2023) which is well above the inflow concentration of the column reactor (9.02 µg/L).

Iomeprol (column experiments):

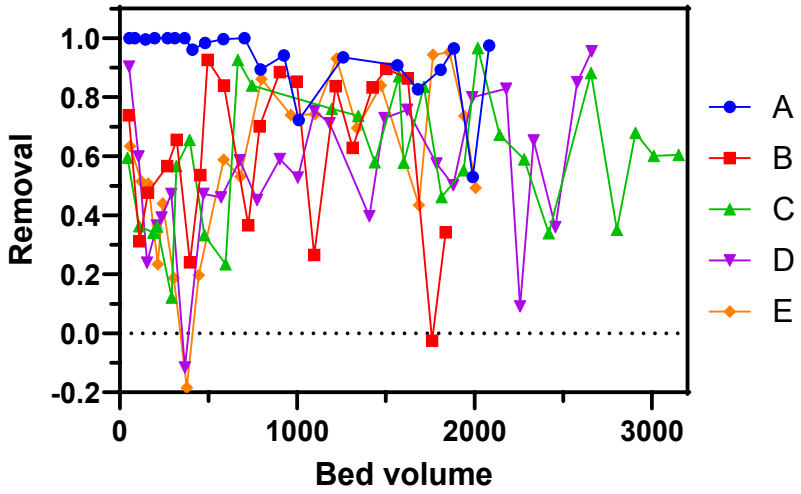

Figure 39: Removal of iomeprol in column experiment over bed volume.

Table 45: Concentration of iomeprol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Iomeprol	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.015	0.029	0.24	0.089	0.029
B (Wood)	1.5	0.15	0.17	0.057	0.34
C (Wood/Sludge)	1.3	0.56	0.62	0.11	0.032
D (Sludge/Wood)	0.87	0.56	0.56	0.26	0.047
E (Sludge)	1.1	0.69	0.36	0.019	0.42

The removal of iomeprol in column A kept at 99% in the first 600 BV and varied between 50% to 99%. The removal in biochars decreased from ~90% to 10% and negative removal was observed in the first 600 BV. The removal in the biochar increased after 600 BV but varied between 30% to

~99%. The PNEC level found for iomeprol 1 000 000 µg/L (Kisielius et al., 2023) which is well above the inflow concentration of the column reactor (1.13 µg/L).

Iopamidol (column experiments):

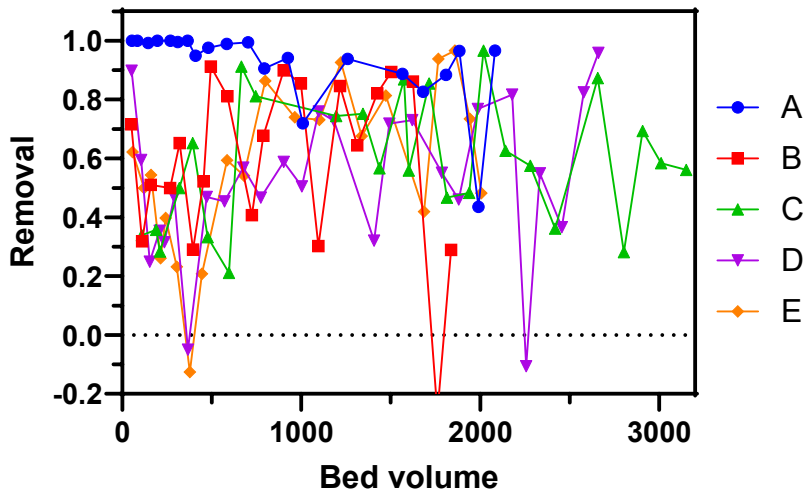

Figure 40: Removal of iopamidol in column experiment over bed volume.

Table 46: Concentration of iopamidol after eluting effluent wastewater (volume given in bed volumes (BV)) in five column reactors. (Red font indicates the concentration exceeds PNEC).

Iopamidol	100 BV	500 BV	1000 BV	1500 BV	2000 BV
A (Brennsorb 1240)	<0.1	<0.1	0.21	<0.1	<0.1
B (Wood)	1.2	0.14	0.14	<0.1	<0.1
C (Wood/Sludge)	1.2	0.51	0.53	<0.1	<0.1
D (Sludge/Wood)	0.74	0.51	0.47	0.24	<0.1
E (Sludge)	0.92	0.59	0.33	<0.1	<0.1

The removal of iopamidol in column A kept at 99% in the first 500 BV and varied between 40% to 99%. On the other hand, the removal in biochars decreased from 90% to 20% and sometimes negative removal was observed for column D and E in the first 500 BV. It gradually increased but varied between 50% to 90% afterwards. The PNEC level found for iopamidol 1 000 000 µg/L

(Kisielius et al., 2023) which is well above the inflow concentration of the column reactor (0.98 µg/L).

8.2 Micropollutant partitioning experiments

Oxazepam (Partitioning experiments):

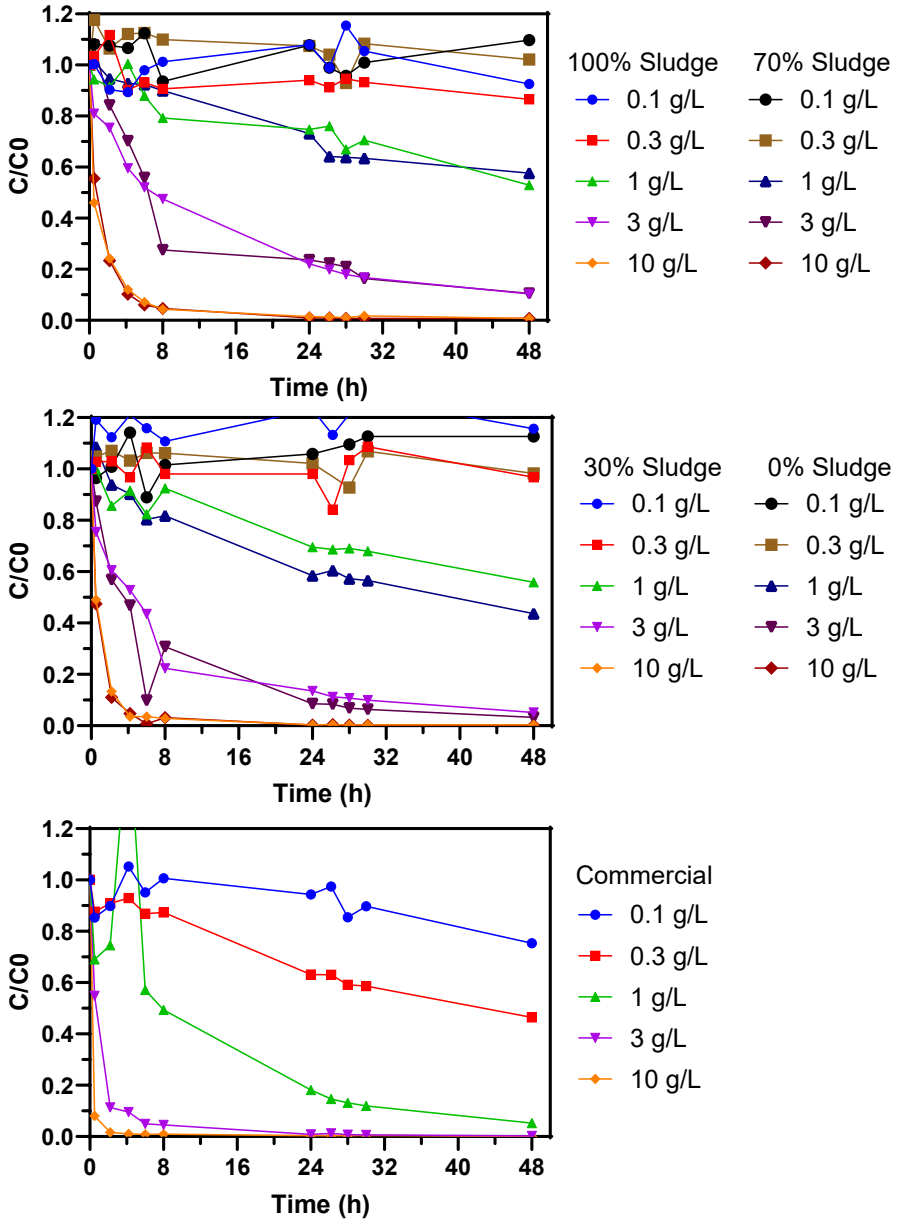

Figure 41: Comparison of oxazepam removal with different GAC in the partitioning experiment.

The partitioning experiment on oxazepam removal suggested biochar with dosage 0.3 g/L or less could only yield < 10% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% oxazepam. There's no noticeable difference in oxazepam removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 70% at 0.3 g/L dosage.

Sertraline (Partitioning experiment):

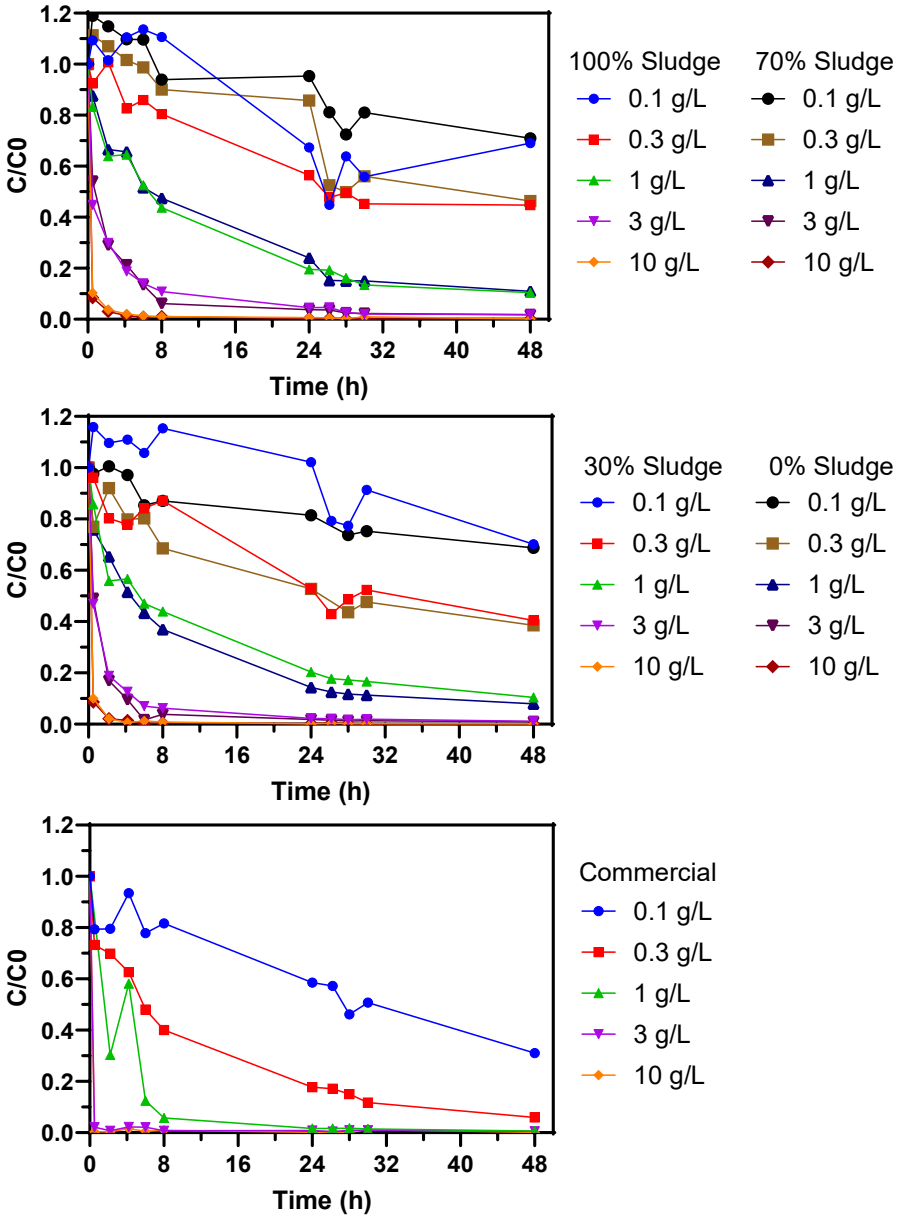

Figure 42: Comparison of sertraline removal with different GAC in the partitioning experiment.

The partitioning experiment on sertraline removal suggested biochar with dosage 0.3 g/L or less could only yield < 50% removal. With dosage at 1 g/L or higher, the biochar could remove > 85% sertraline. There's no noticeable difference in sertraline removal between different composition of

biochars. The commercial activated carbon, on the other hand, yielded 60% removal at the dosage of 0.1 g/L and the removal increased to 90% at 0.3 g/L dosage.

Clarithromycin (Partitioning experiment):

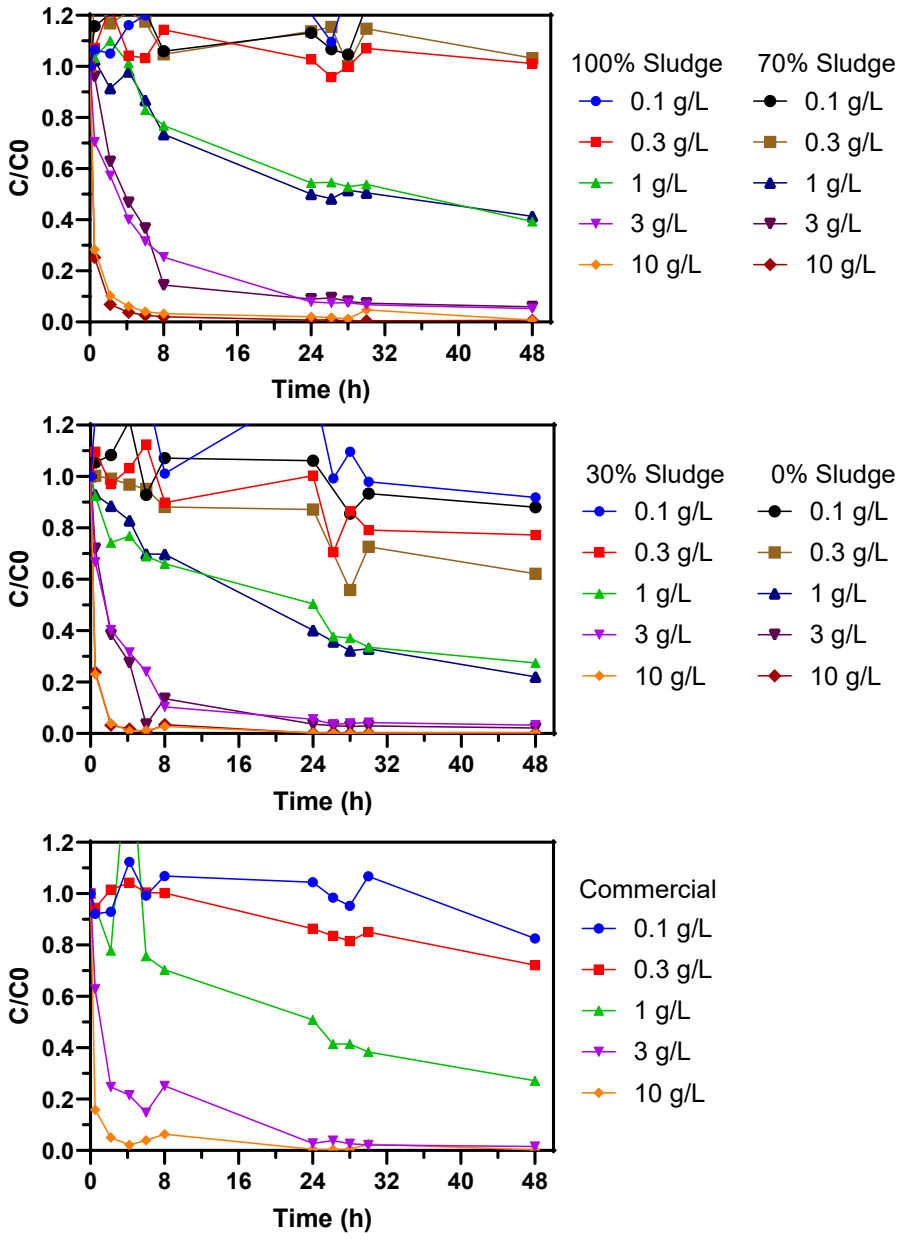

Figure 43: Comparison of clarithromycin removal with different GAC in the partitioning experiment.

The partitioning experiment on clarithromycin removal suggested biochar with dosage 0.3 g/L or less could only yield < 5% removal. With dosage at 1 g/L or higher, the biochar could remove > 60% clarithromycin. In comparison, biochar with less sludge content has better removal but the difference is not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 10% removal at the dosage of 0.1 g/L and the removal increased to 70% at 0.3 g/L dosage.

Diclofenac (Partitioning experiment):

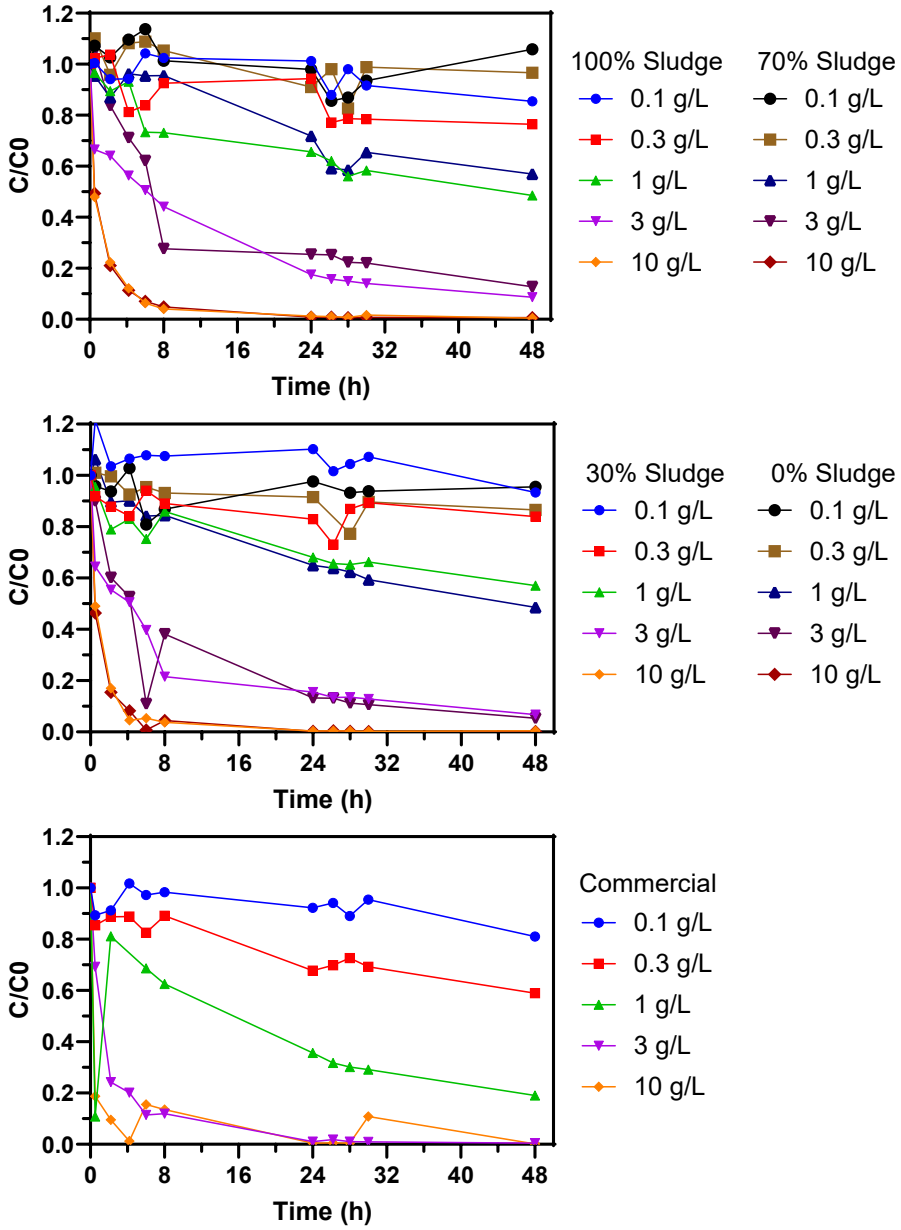

Figure 44: Comparison of diclofenac removal with different GAC in the partitioning experiment.

The partitioning experiment on diclofenac removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% diclofenac. In comparison, biochar with less sludge content has better removal but the difference

is not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 40% at 0.3 g/L dosage.

Metoprolol (Partitioning experiment):

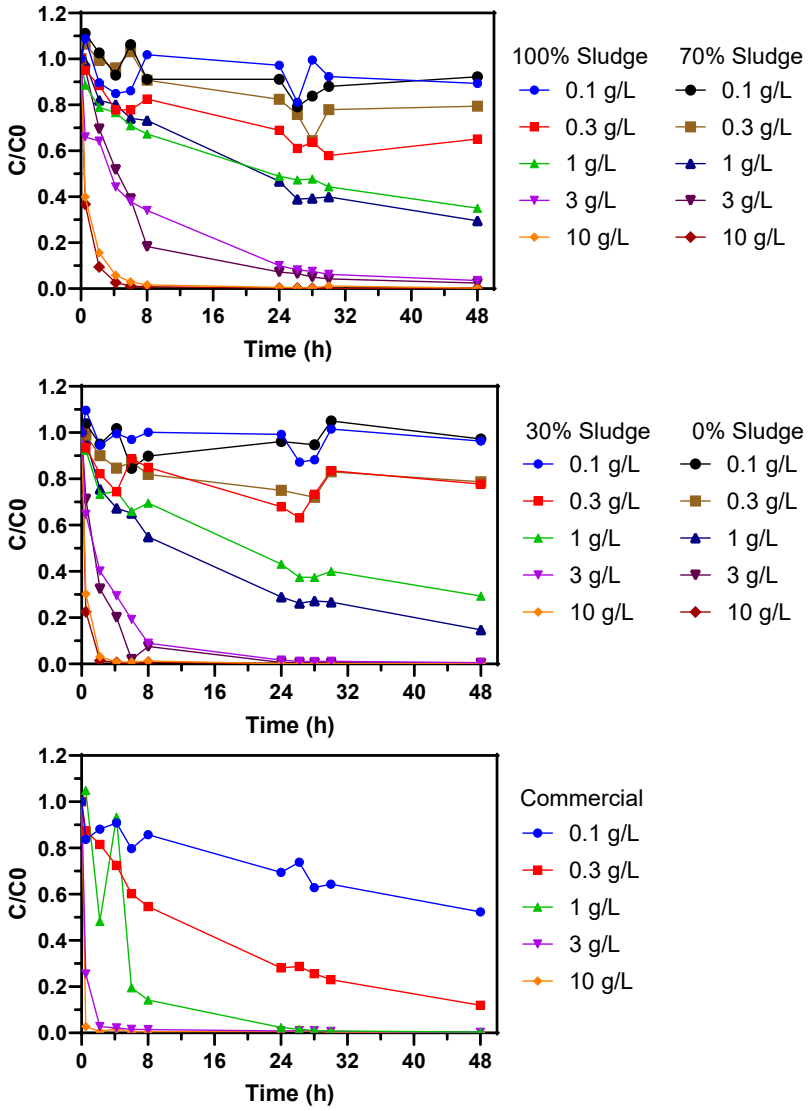

Figure 45: Comparison of metoprolol removal with different GAC in the partitioning experiment.

The partitioning experiment on metoprolol acid removal suggested biochar with dosage 0.3 g/L or less could only yield < 30% removal. With dosage at 1 g/L or higher, the biochar could remove > 60% metoprolol. In comparison, biochar with less sludge content has better removal but the difference is not significant (removal difference <5% at 48 h contacting time). The commercial

activated carbon, on the other hand, yielded 40% removal at the dosage of 0.1 g/L and the removal increased to 85% at 0.3 g/L dosage.

Propranolol (Partitioning experiment):

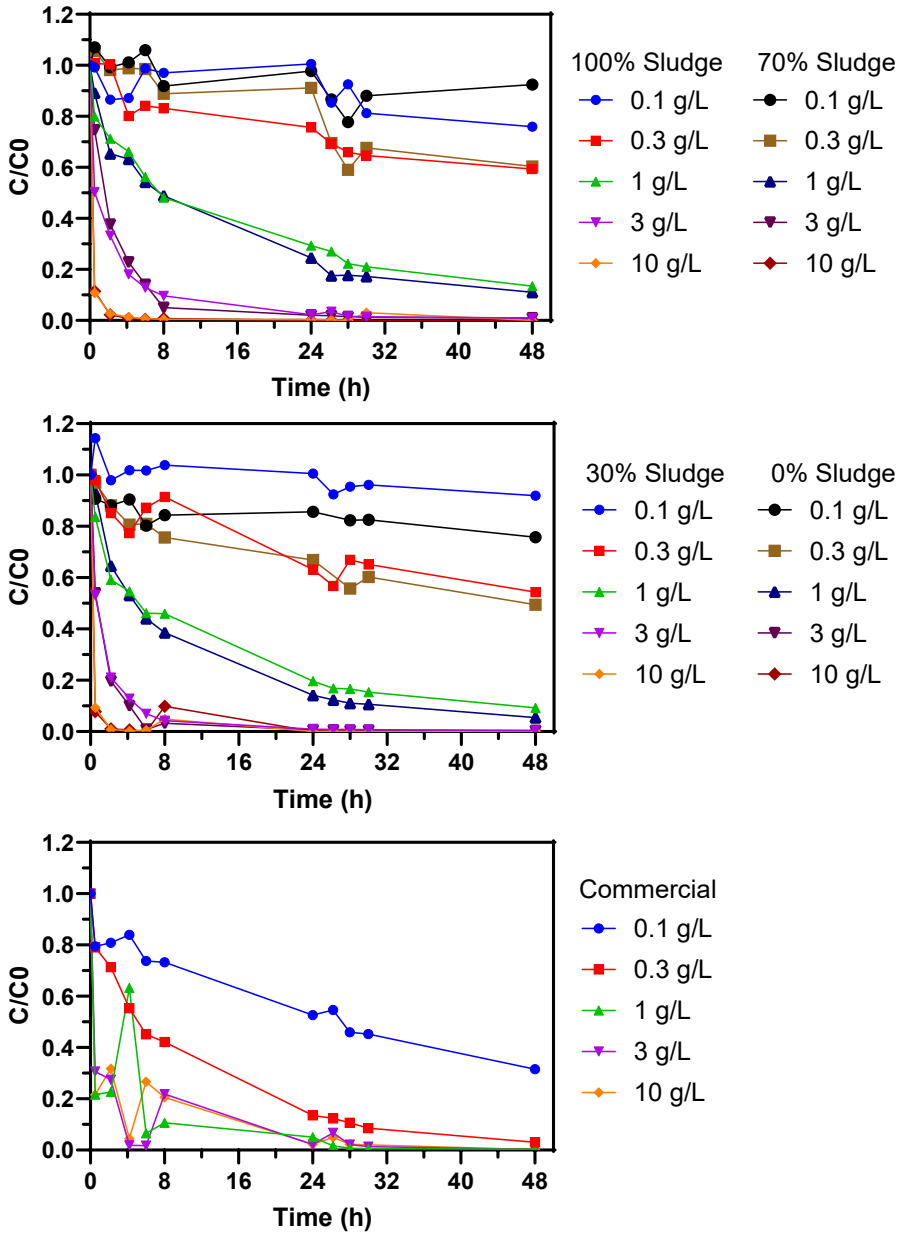

Figure 46: Comparison of propranolol removal with different GAC in the partitioning experiment.

The partitioning experiment on propranolol removal suggested biochar with dosage 0.3 g/L or less could only yield < 30% removal. With dosage at 1 g/L or higher, the biochar could remove > 80% propranolol. There's no noticeable difference in propranolol removal between different

composition of biochars. The commercial activated carbon, on the other hand, yielded 60% removal at the dosage of 0.1 g/L and the removal increased to 95% at 0.3 g/L dosage.

Venlafaxine (Partitioning experiment):

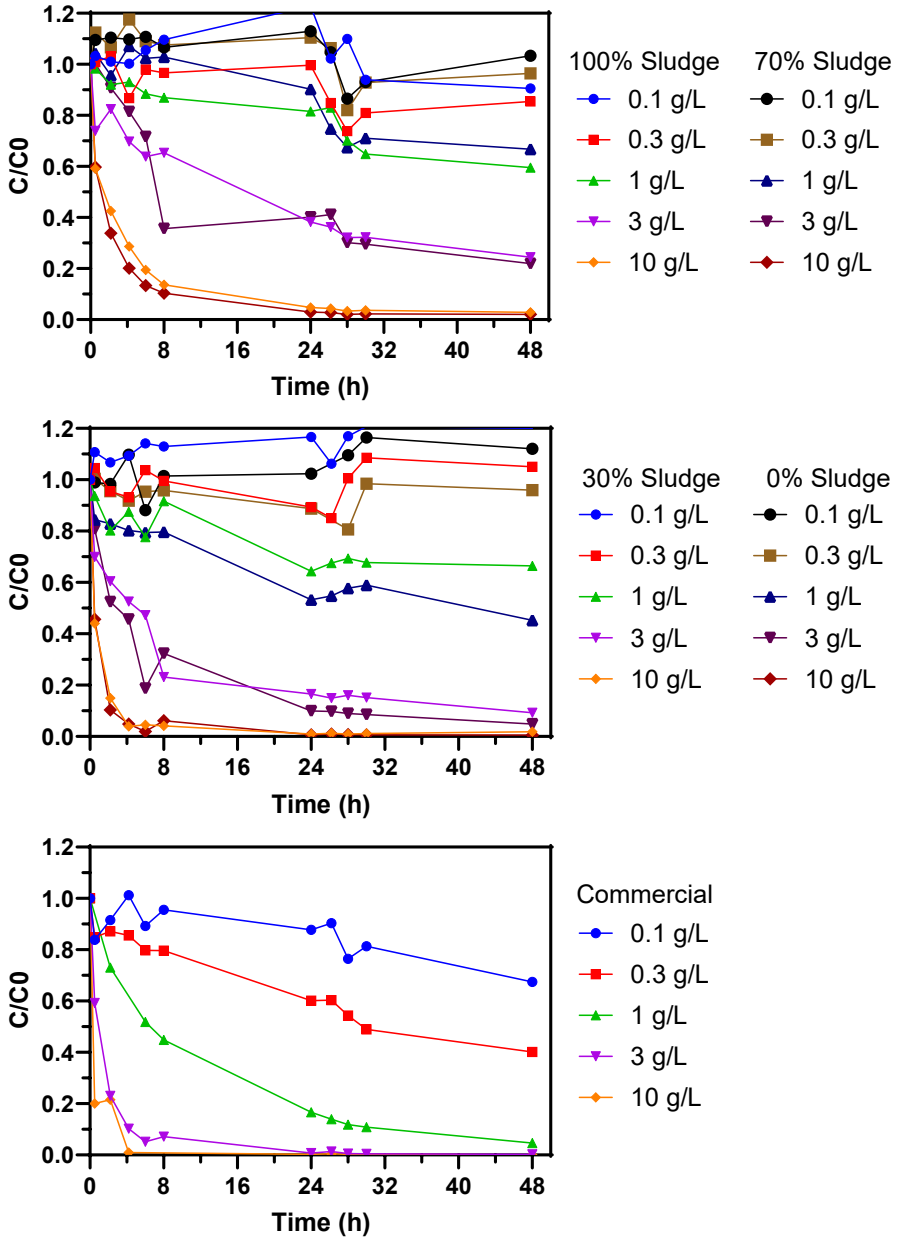

Figure 47: Comparison of venlafaxine removal with different GAC in the partitioning experiment.

The partitioning experiment on venlafaxine removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 3 g/L or higher, the biochar could remove > 80% venlafaxine. There's no noticeable difference in venlafaxine removal between different

composition of biochars. The commercial activated carbon, on the other hand, yielded 30% removal at the dosage of 0.1 g/L and the removal increased to 50% at 0.3 g/L dosage.

Sulfamethoxazole (Partitioning experiment):

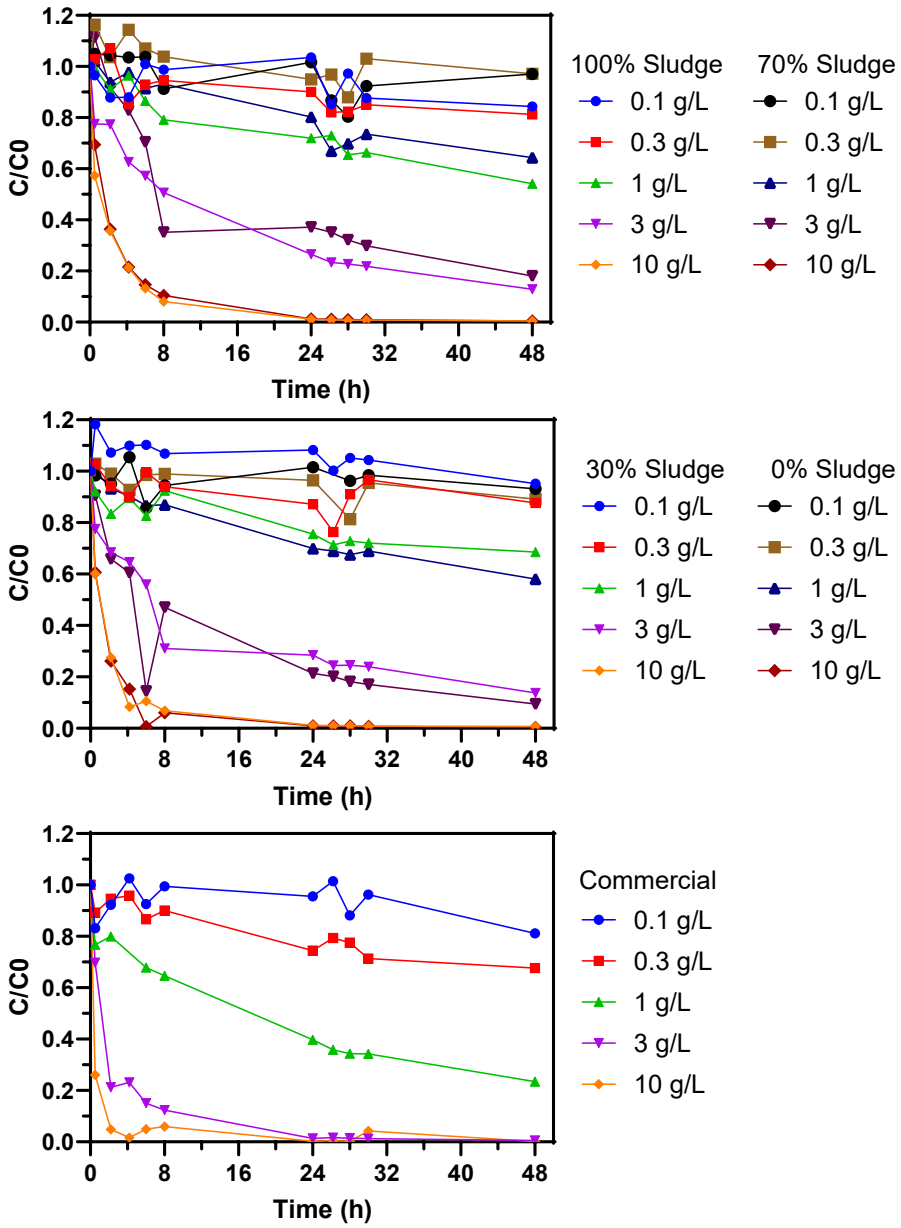

Figure 48: Comparison of sulfamethoxazole removal with different GAC in the partitioning experiment.

The partitioning experiment on sulfamethoxazole removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove

> 30% sulfamethoxazole. There's no noticeable difference in sulfamethoxazole removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 15% removal at the dosage of 0.1 g/L and the removal increased to 30% at 0.3 g/L dosage.

Candesartan (Partitioning experiments):

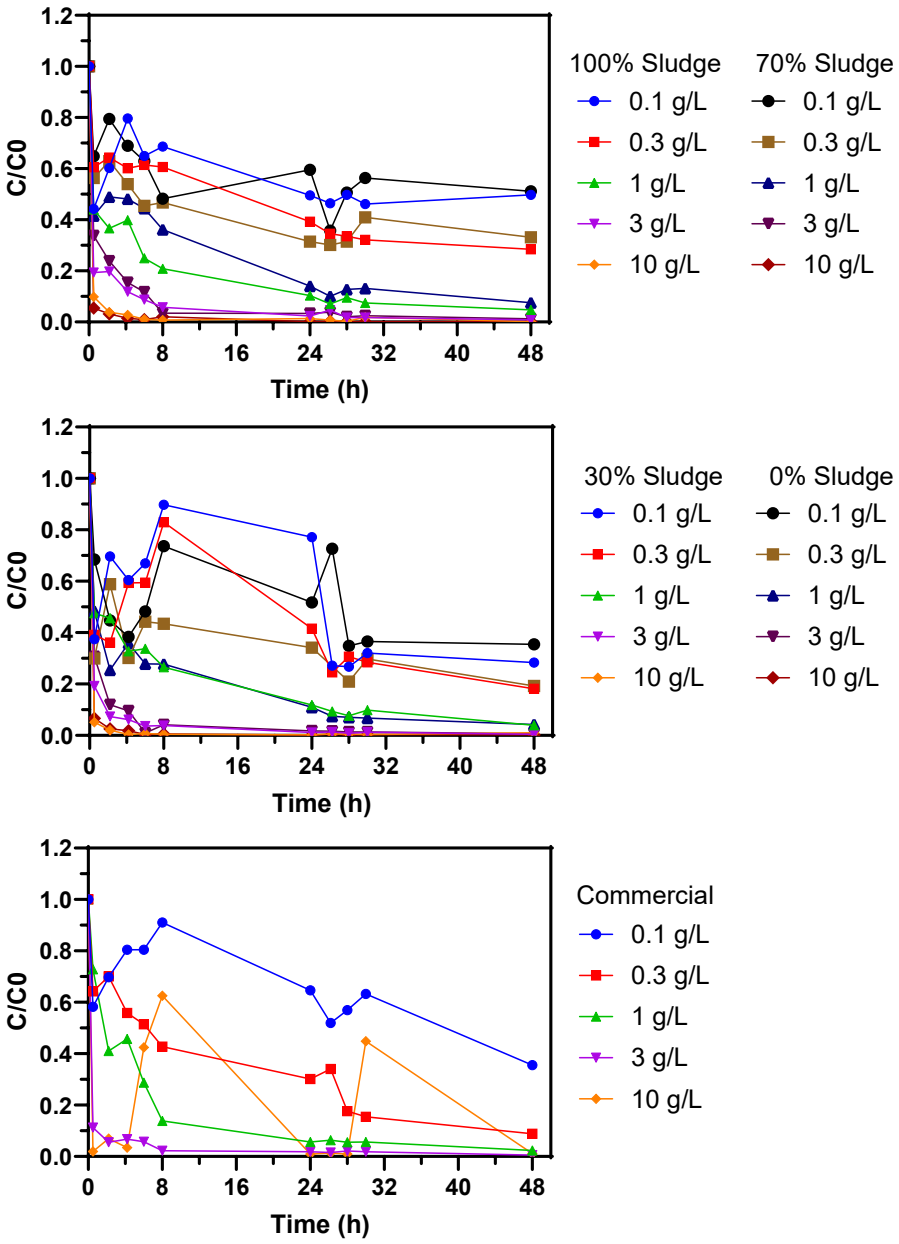

Figure 49: Comparison of candesartan removal with different GAC in the partitioning experiment.

The partitioning experiment on candesartan removal suggested biochar with dosage 0.3 g/L or less could only yield < 70% removal. With dosage at 1 g/L or higher, the biochar could remove > 90% candesartan. In comparison, biochar with less sludge content has better removal but the difference

is not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 60% removal at the dosage of 0.1 g/L and the removal increased to 90% at 0.3 g/L dosage.

Amisulpride (Partitioning experiments):

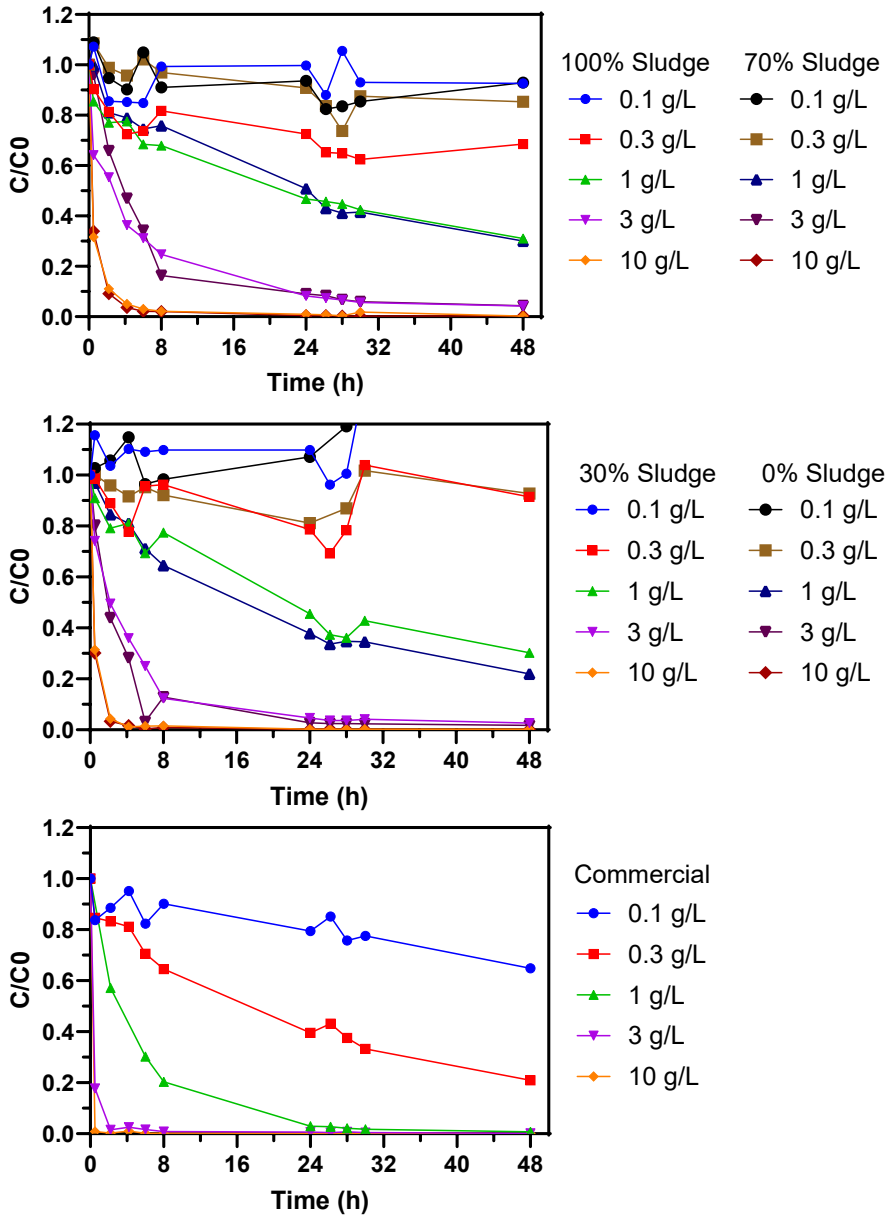

Figure 50: Comparison of amisulpride removal with different GAC in the partitioning experiment.

The partitioning experiment on amisulpride removal suggested biochar with dosage 0.3 g/L or less could only yield < 30% removal. With dosage at 1 g/L or higher, the biochar could remove > 70% amisulpride. There's no noticeable difference in amisulpride removal between different

composition of biochars. The commercial activated carbon, on the other hand, yielded 30% removal at the dosage of 0.1 g/L and the removal increased to 80% at 0.3 g/L dosage.

Carbamazepine (Partitioning experiments):

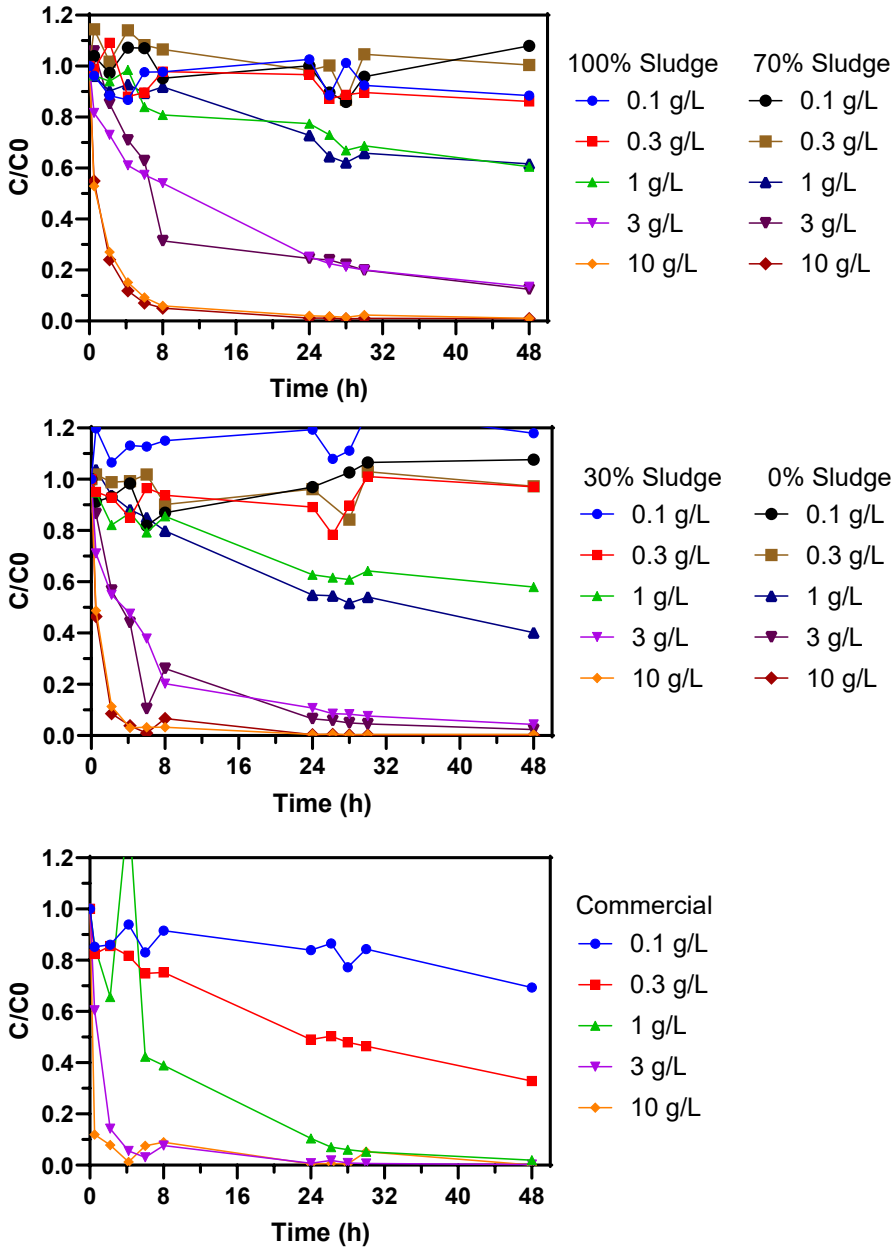

Figure 51: Comparison of carbamazepine removal with different GAC in the partitioning experiment.

The partitioning experiment on carbamazepine removal suggested biochar with dosage 0.3 g/L or less could only yield < 10% removal. With dosage at 1 g/L or higher, the biochar could remove > 30% carbamazepine. In comparison, biochar with less sludge content has better removal but the difference is not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 60% at 0.3 g/L dosage.

Citalopram (Partitioning experiments):

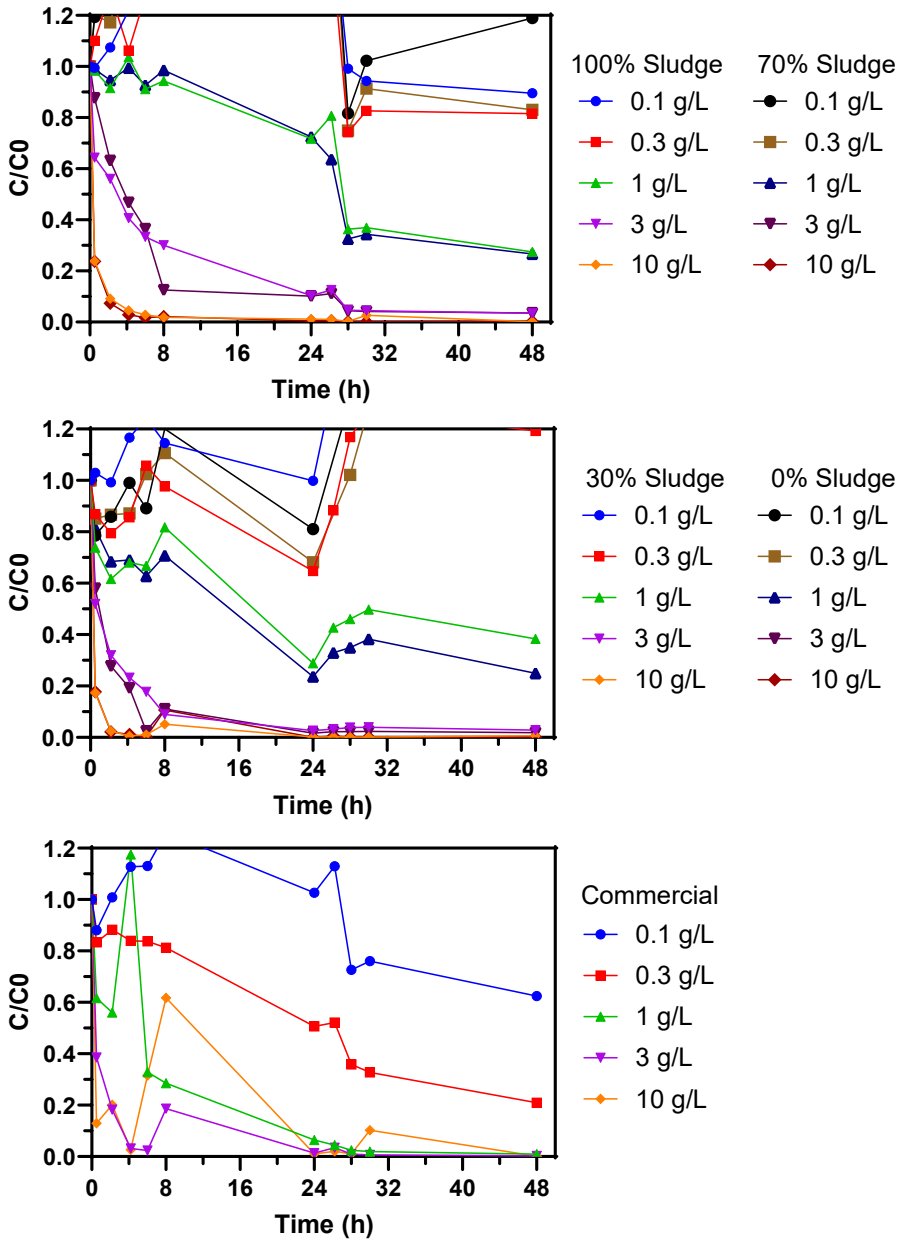

Figure 52: Comparison of citalopram removal with different GAC in the partitioning experiment.

The partitioning experiment on citalopram removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 70% citalopram. In comparison, biochar with less sludge content has better removal but the difference is not significant (removal difference <5% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 40% removal at the dosage of 0.1 g/L and the removal increased to 80% at 0.3 g/L dosage.

Cetirizine (Partitioning experiments):

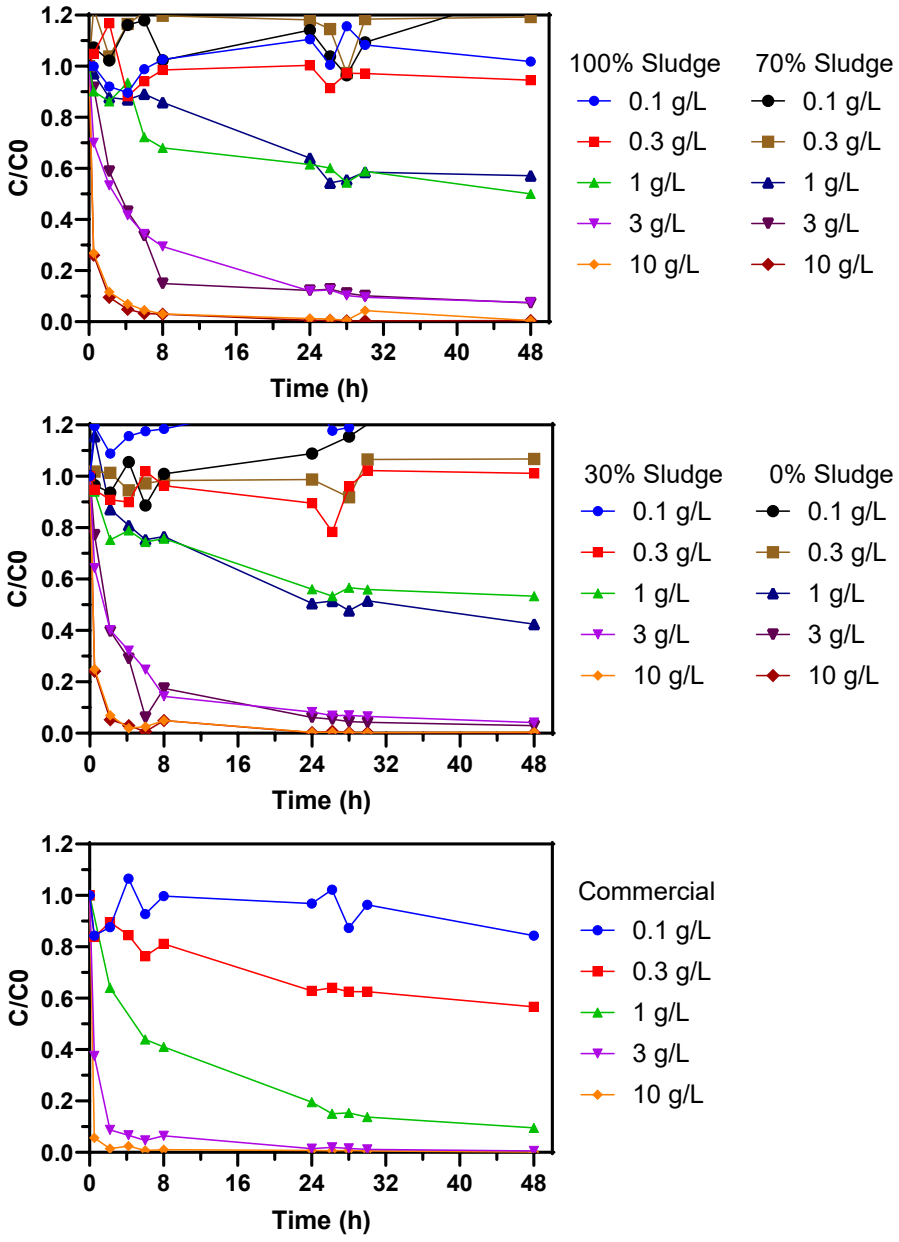

Figure 53: Comparison of cetirizine removal with different GAC in the partitioning experiment.

The partitioning experiment on cetirizine removal suggested biochar with dosage 0.3 g/L or less could only yield < 10% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% cetirizine. There's no noticeable difference in cetirizine removal between different composition of

biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 90% at 1 g/L dosage.

Sulfamethizole (Partitioning experiments):

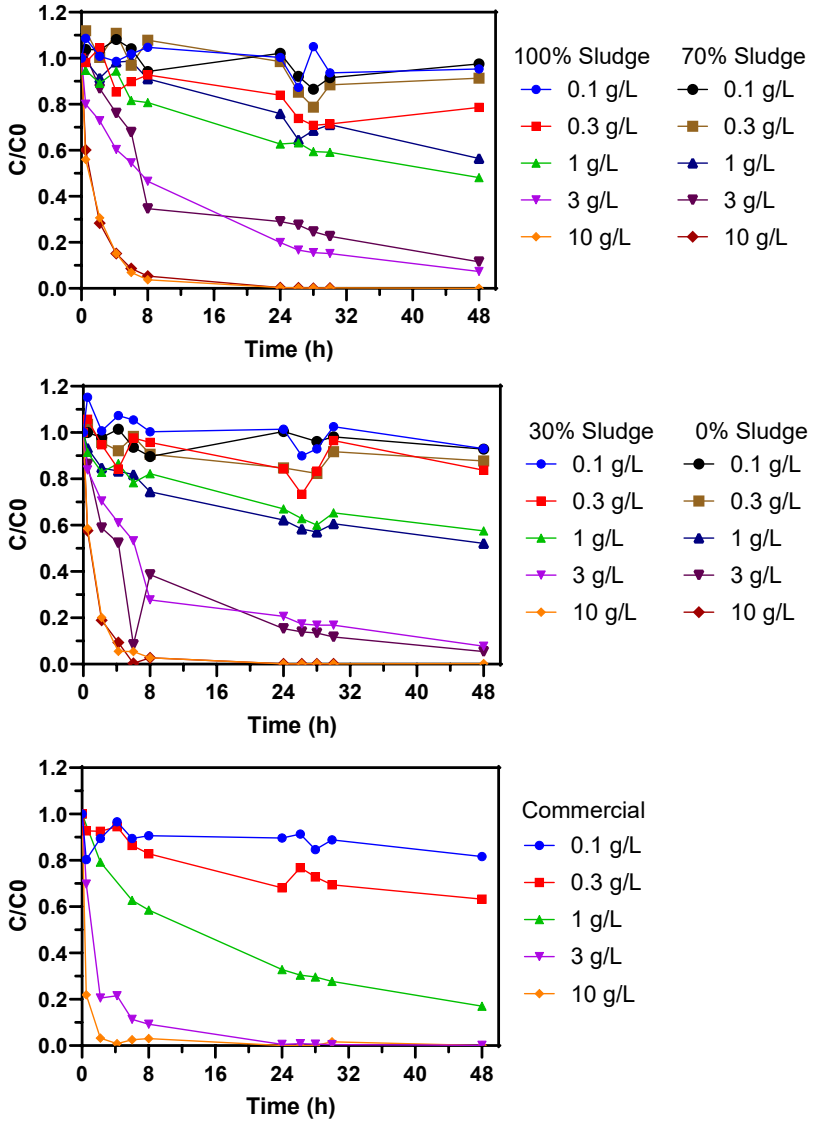

Figure 54: Comparison of sulfamethizole removal with different GAC in the partitioning experiment.

The partitioning experiment on sulfamethizole removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% sulfamethizole. There's no noticeable difference in sulfamethizole removal between different

composition of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 80% at 1 g/L dosage.

Mefenamic acid (Partitioning experiments):

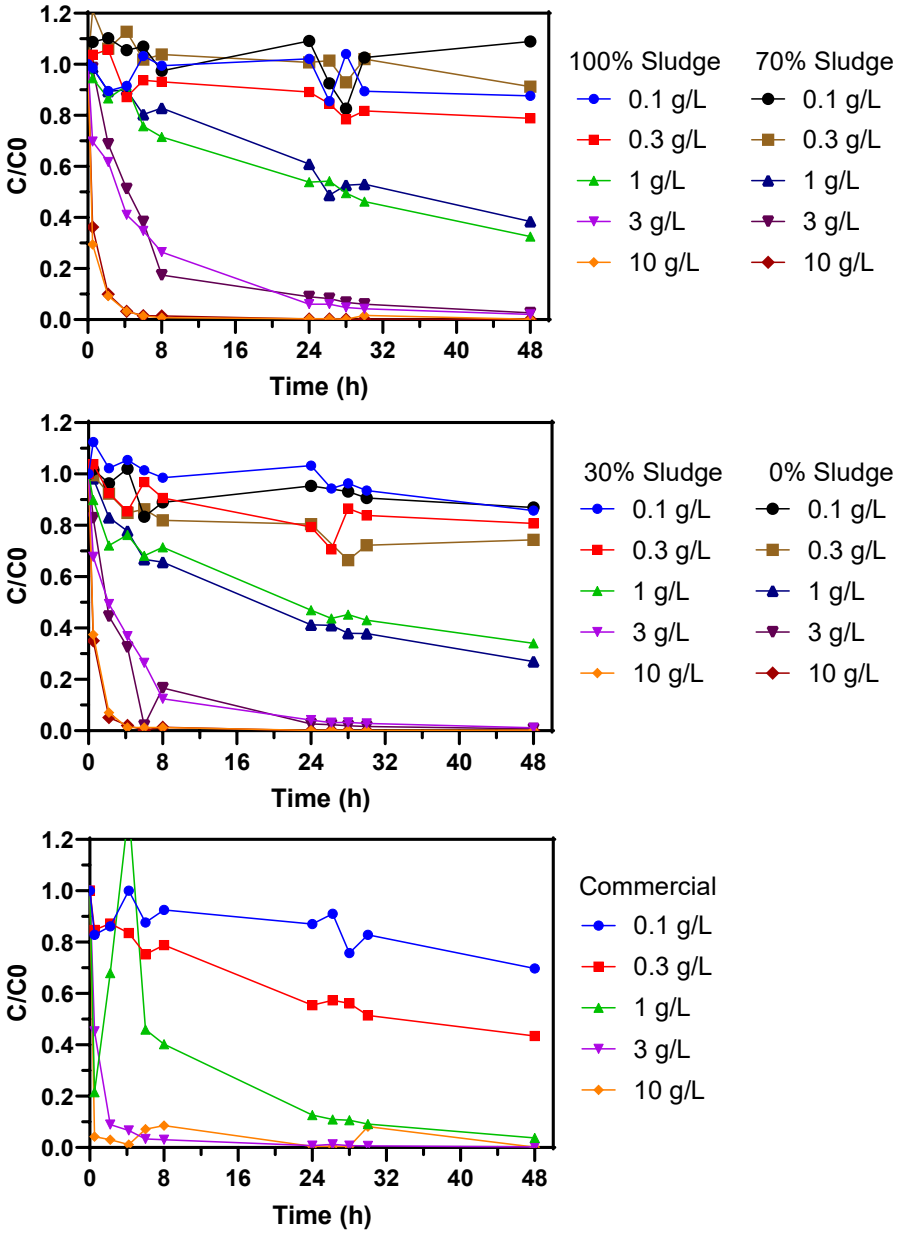

Figure 55: Comparison of mefenamic acid removal with different GAC in the partitioning experiment.

The partitioning experiment on mefenamic acid removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 50% mefenamic acid. There's no noticeable difference in mefenamic acid removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 50% at 0.3 g/L dosage.

Sulfadiazine (Partitioning experiments):

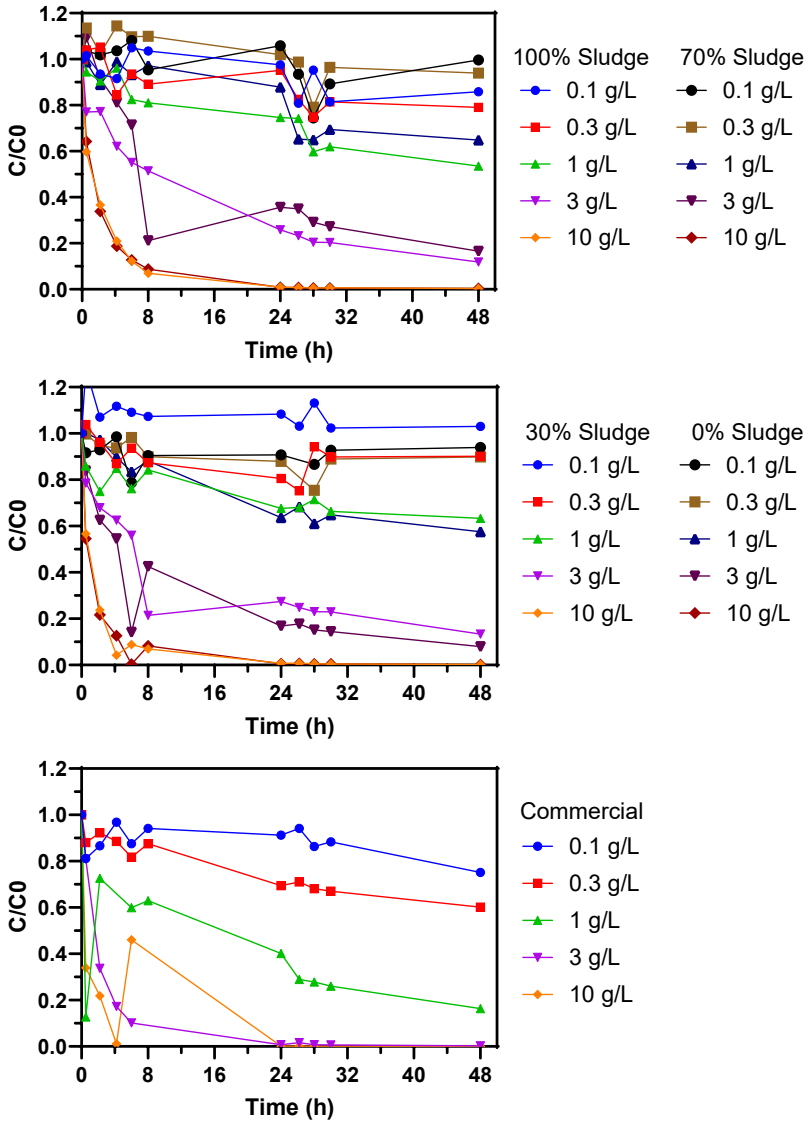

Figure 56: Comparison of sulfadiazine removal with different GAC in the partitioning experiment.

The partitioning experiment on sulfadiazine removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 3 g/L or higher, the biochar could remove > 70% sulfadiazine. There's no noticeable difference in sulfadiazine removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 80% at 1 g/L dosage.

Trimethoprim (Partitioning experiments):

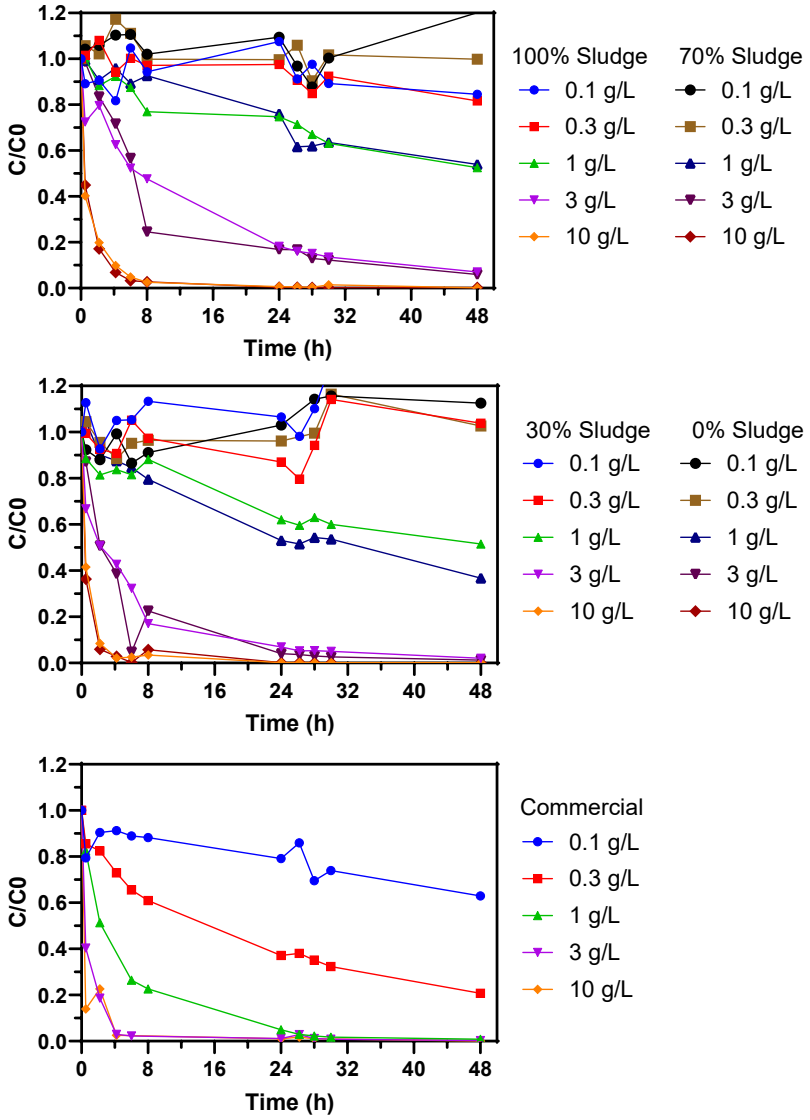

Figure 57: Comparison of trimethoprim removal with different GAC in the partitioning experiment.

The partitioning experiment on trimethoprim removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% trimethoprim. There's no noticeable difference in trimethoprim removal between different

composition of biochars. The commercial activated carbon, on the other hand, yielded 35% removal at the dosage of 0.1 g/L and the removal increased to 80% at 0.3 g/L dosage.

Benzotriazole (Partitioning experiments):

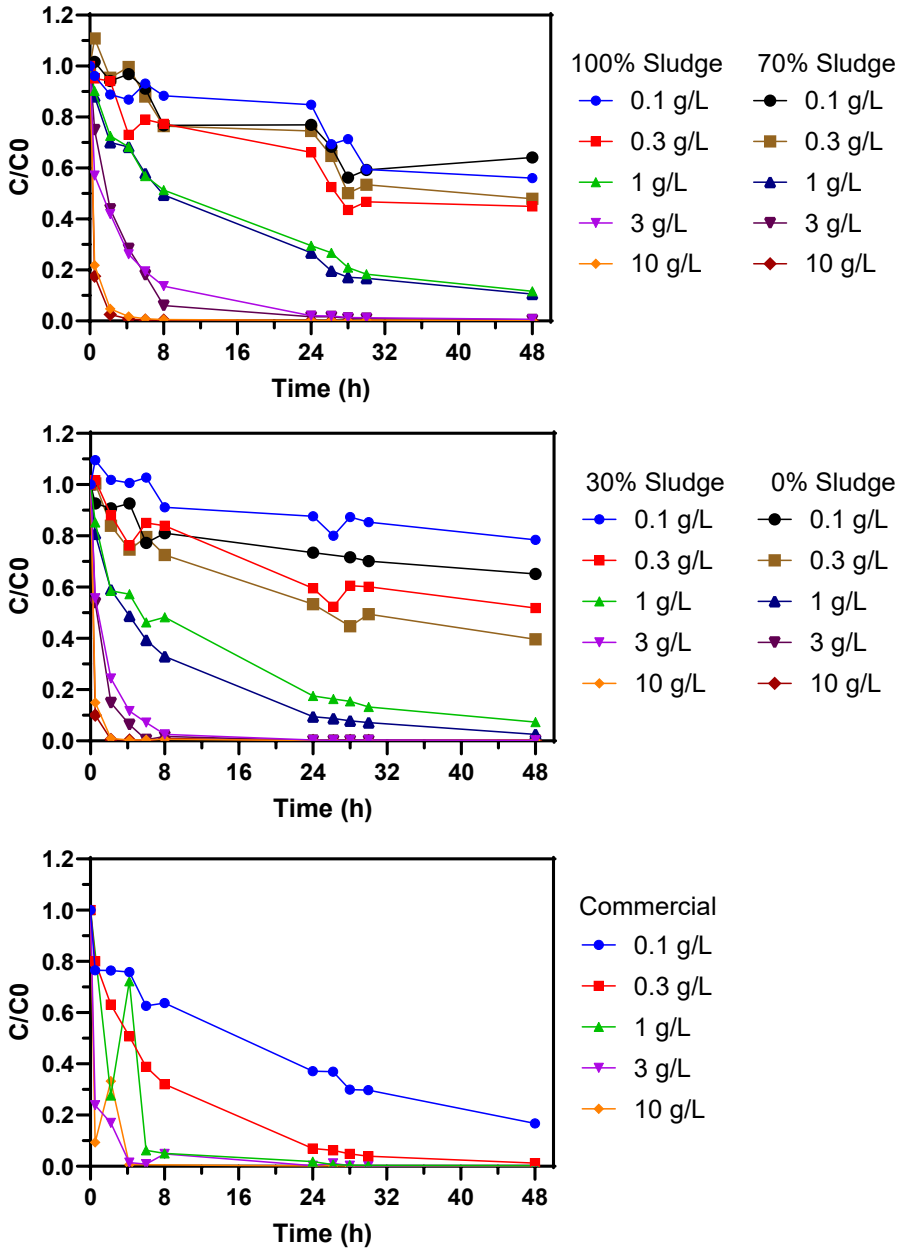

Figure 58: Comparison of benzotriazole removal with different GAC in the partitioning experiment.

The partitioning experiment on benzotriazole removal suggested biochar with dosage 0.3 g/L or less could only yield < 40% removal. With dosage at 1 g/L or higher, the biochar could remove >

90% benzotriazole. There's no noticeable difference in benzotriazole removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 80% removal at the dosage of 0.1 g/L and the removal increased to 95% at 0.3 g/L dosage.

Gabapentin (Partitioning experiments):

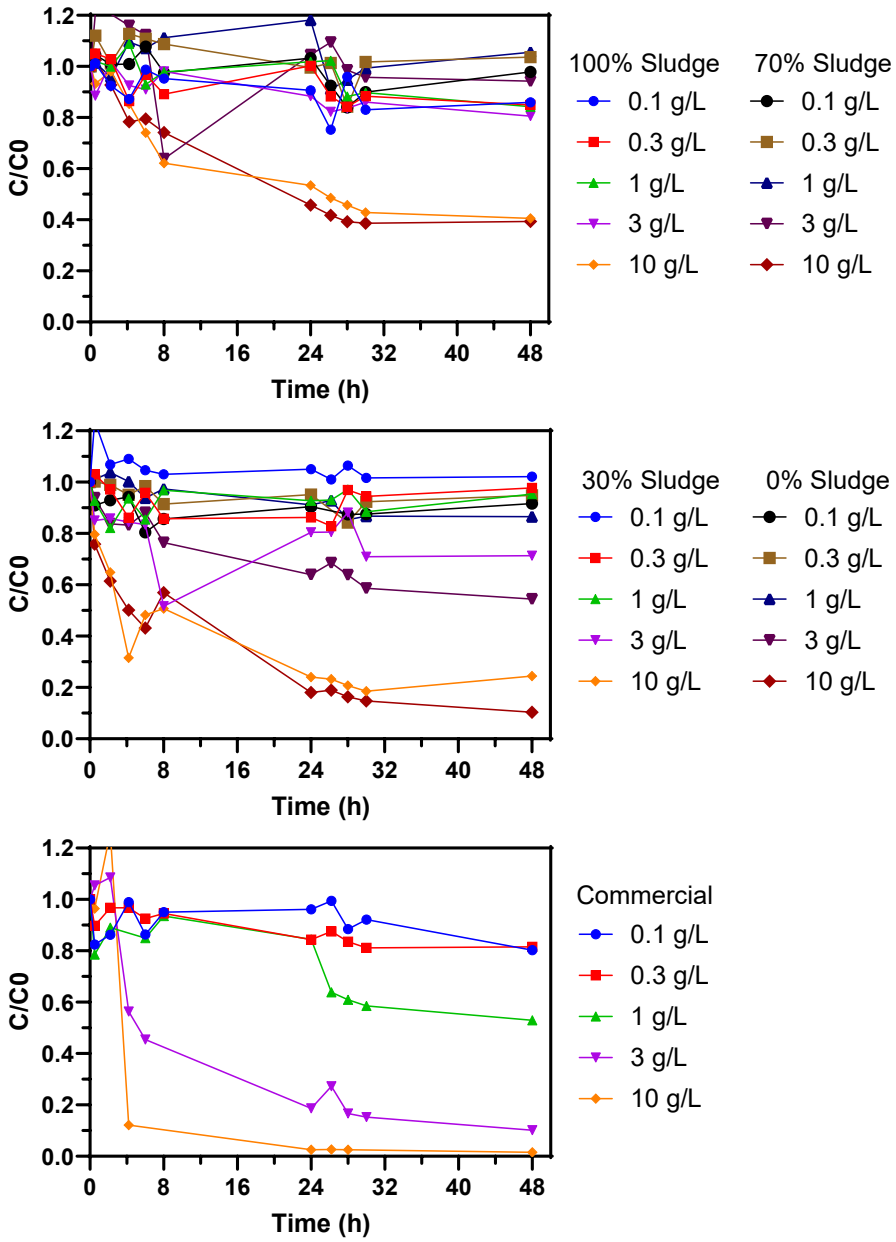

Figure 59: Comparison of gabapentin removal with different GAC in the partitioning experiment.

The partitioning experiment on gabapentin removal suggested biochar with dosage 1 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 30% gabapentin. In comparison, biochar with less sludge content has better removal but the difference

is not significant (removal difference <5% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 40% at 1 g/L dosage.

Atenolol (Partitioning experiments):

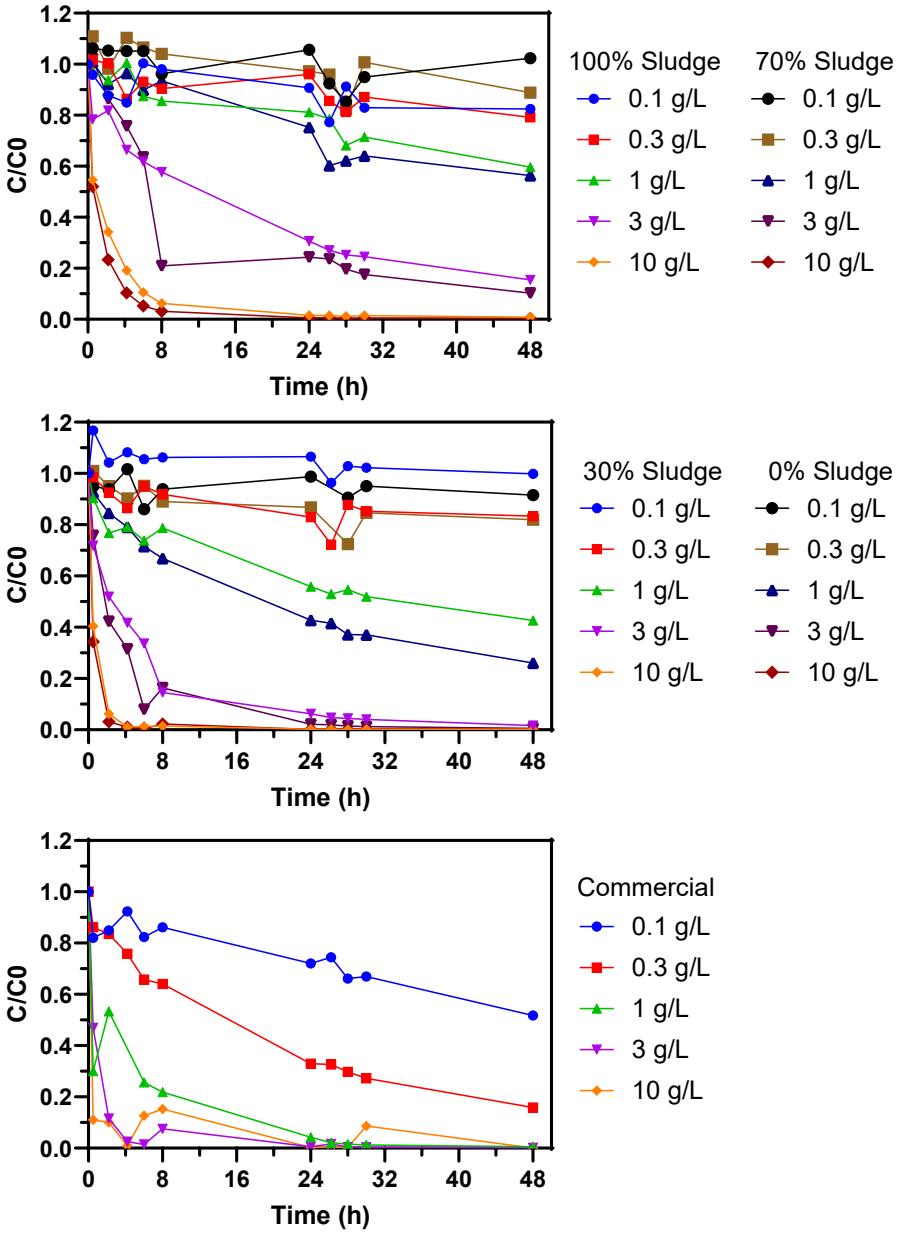

Figure 60: Comparison of atenolol removal with different GAC in the partitioning experiment.

The partitioning experiment on atenolol removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 40% atenolol. In comparison, biochar with less sludge content has better removal but the difference is

not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 40% removal at the dosage of 0.1 g/L and the removal increased to 80% at 0.3 g/L dosage

Losartan (Partitioning experiments):

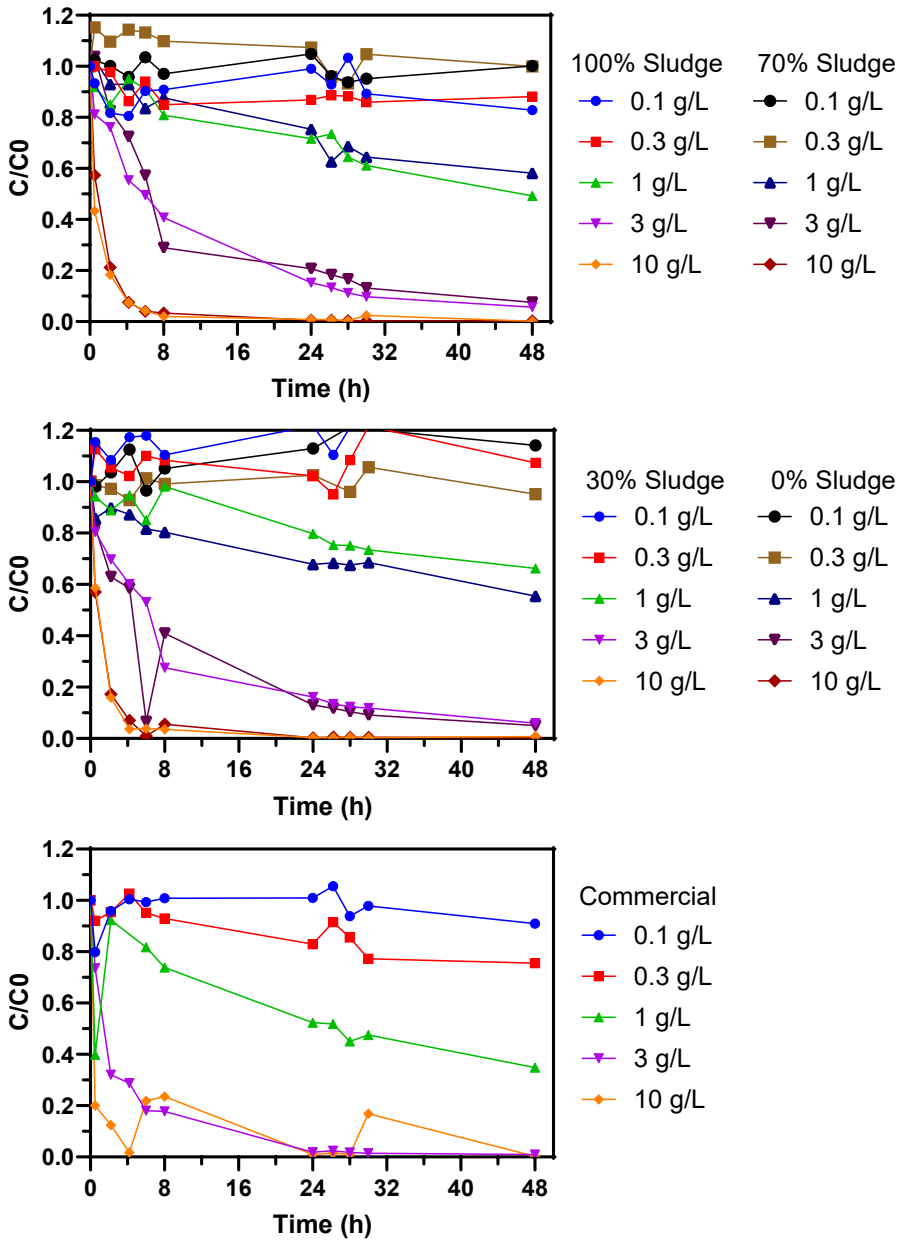

Figure 61: Comparison losartan removal with different GAC in the partitioning experiment.

The partitioning experiment on losartan removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 30% losartan. There's no noticeable difference in losartan removal between different composition of

biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.3 g/L and the removal increased to 60% at 1 g/L dosage.

Irbesartan (Partitioning experiments):

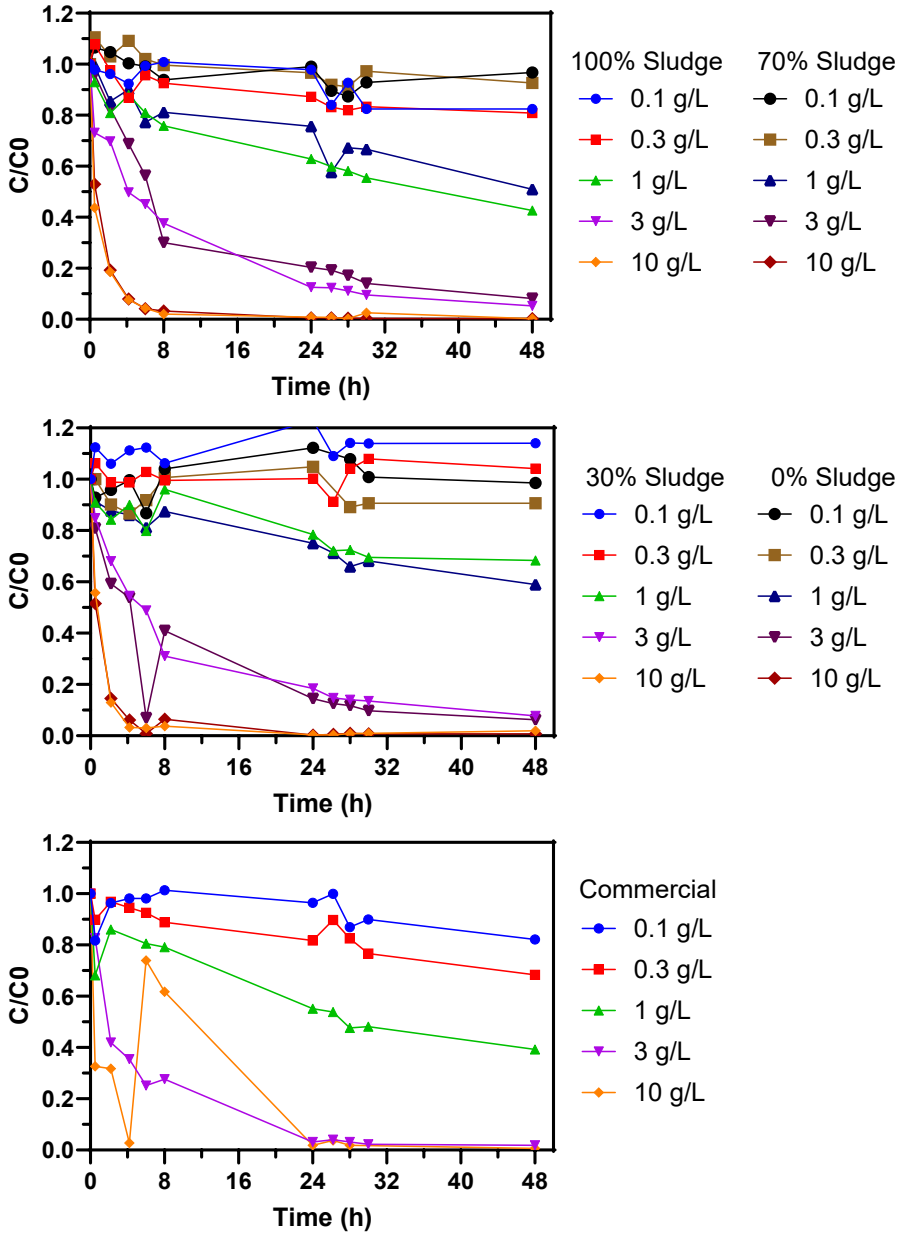

Figure 62: Comparison of irbesartan removal with different GAC in the partitioning experiment.

The partitioning experiment on irbesartan removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 30% irbesartan. There's no noticeable difference in irbesartan removal between different composition

of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 60% at 1 g/L dosage.

Iohexol (Partitioning experiments):

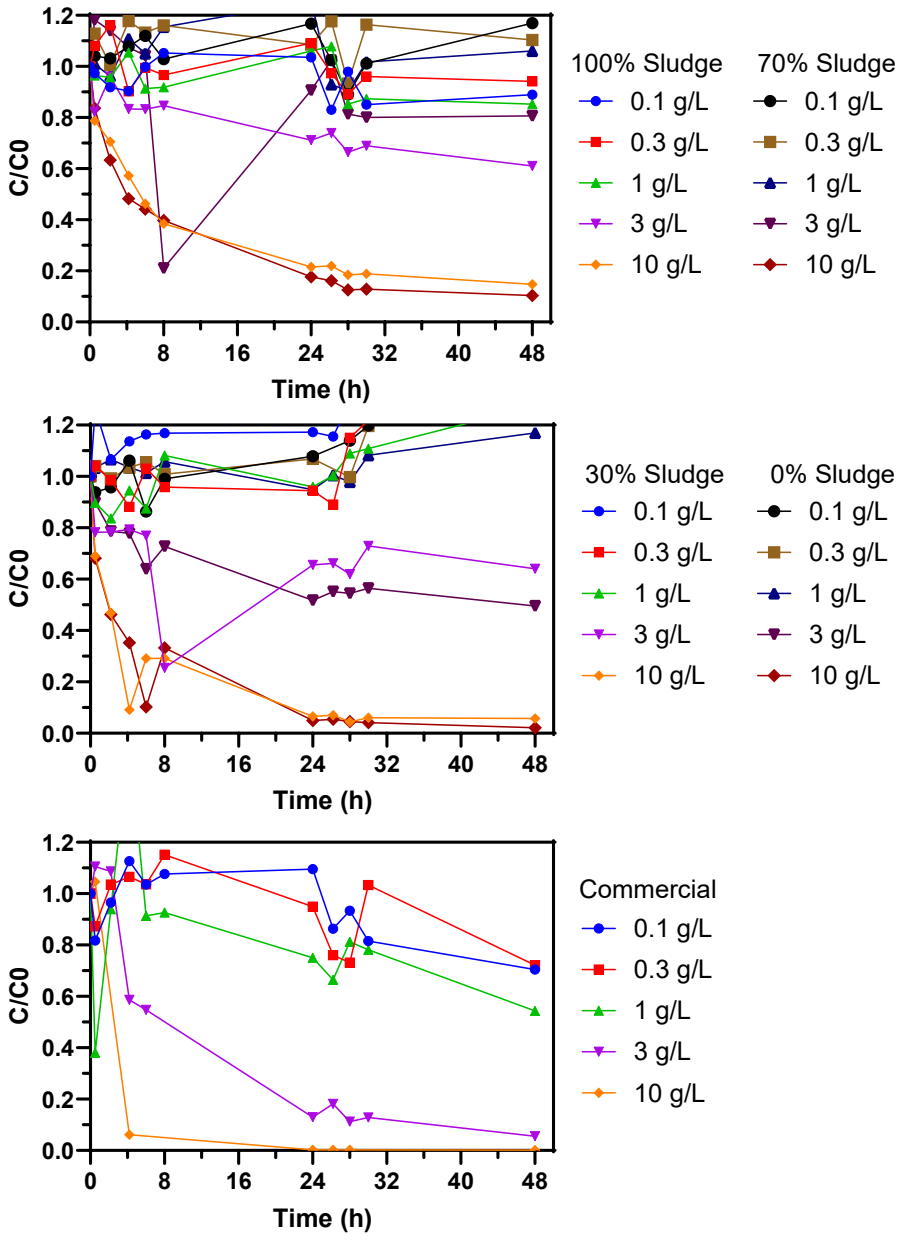

Figure 63: Comparison of iohexol removal with different GAC in the partitioning experiment.

The partitioning experiment on iohexol removal suggested biochar with dosage 1 g/L or less could only yield < 20% removal. With dosage at 3 g/L or higher, the biochar could remove > 25%

iohexol. There's no noticeable difference between biochar compositions in iohexol removal. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 40% at 1 g/L dosage.

Iomeprol (Partitioning experiments):

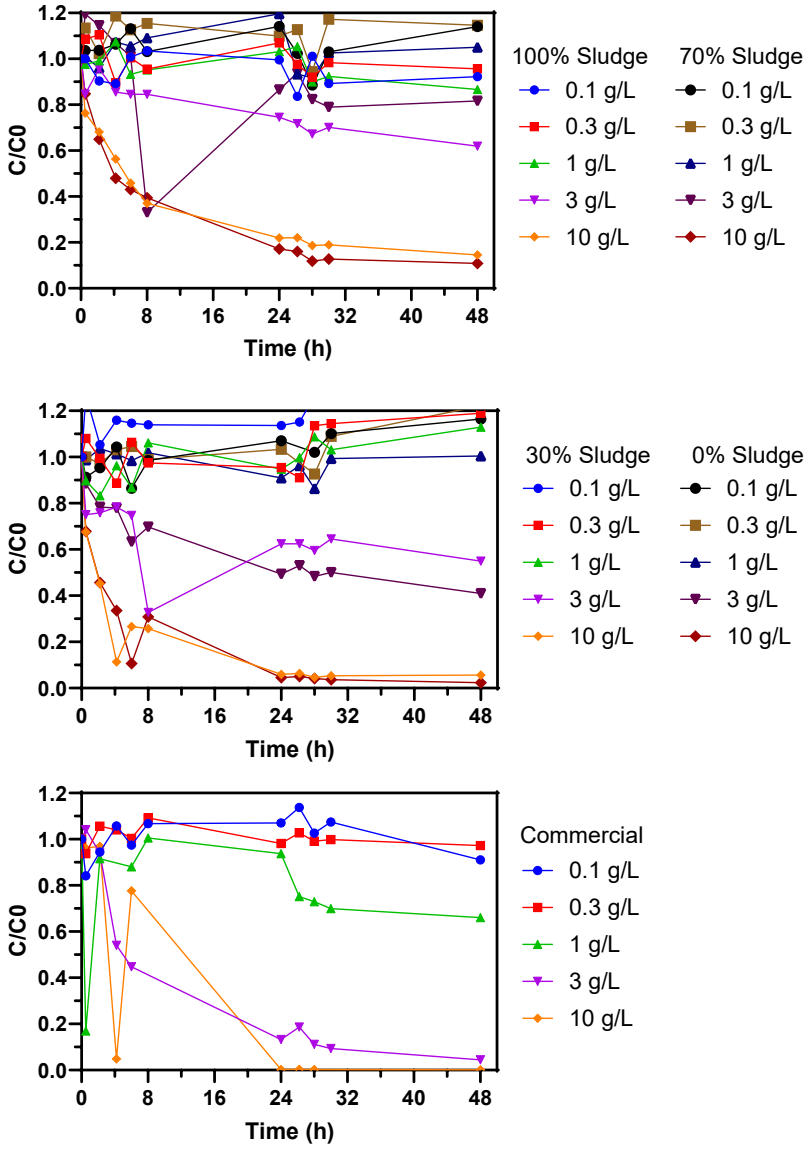

Figure 64: Comparison of iomeprol removal with different GAC in the partitioning experiment.

The partitioning experiment on iomeprol removal suggested biochar with dosage 0.3 g/L or less could only yield < 10% removal. With dosage at 3 g/L or higher, the biochar could remove > 20% iomeprol. In comparison, biochar with less sludge content has better removal but the difference is not significant (removal difference <5% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 10% removal at the dosage of 0.1 g/L which is the same with

biochars. The removal increased to 30% at 0.3 g/L dosage, which is higher than any of the biochar compositions.

Iopamidol (Partitioning experiments):

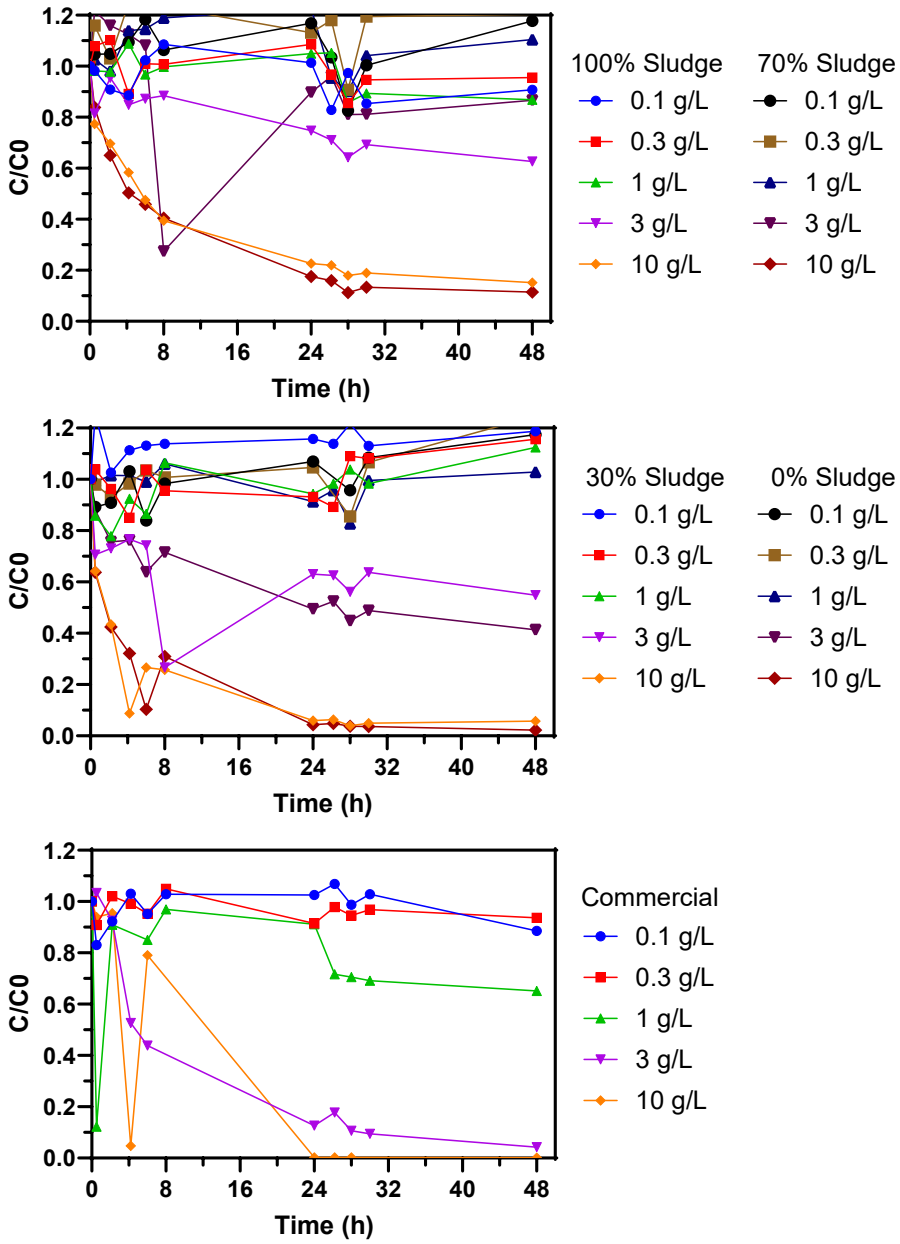

Figure 65: Comparison of iopamidol removal with different GAC in the partitioning experiment.

The partitioning experiment on iopamidol removal suggested biochar with dosage 0.3 g/L or less could only yield < 10% removal. With dosage at 3 g/L or higher, the biochar could remove > 20% iopamidol. In comparison, biochar with less sludge content has better removal but the difference

is not significant (removal difference <10% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 30% removal at the dosage of 0.3 g/L and the removal increased to 95% at 3 g/L dosage.

Codeine (Partitioning experiments):

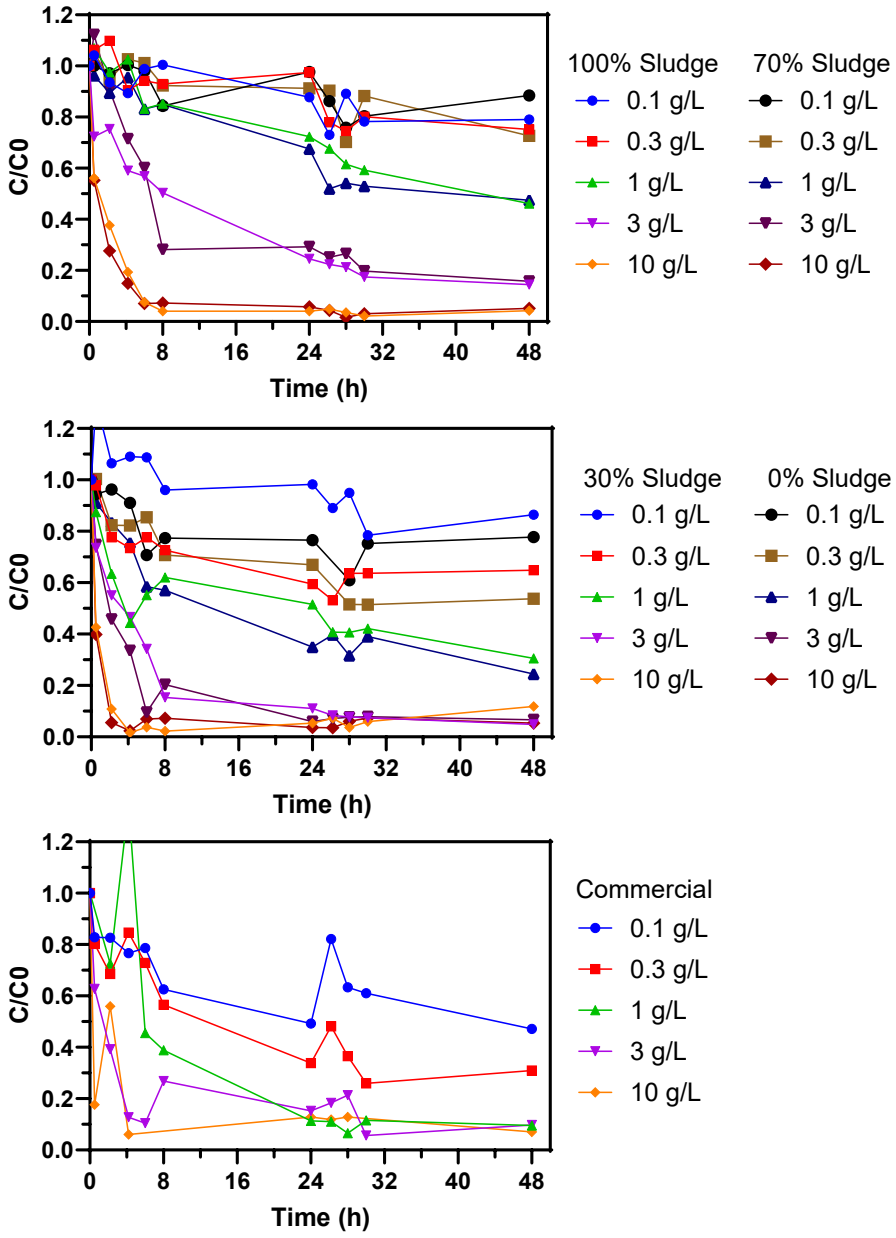

Figure 66: Comparison of codeine removal with different GAC in the partitioning experiment.

The partitioning experiment on codeine removal suggested biochar with dosage 0.3 g/L or less could only yield < 20% removal. With dosage at 1 g/L or higher, the biochar could remove > 50% codeine. In comparison, biochar with less sludge content has better removal but the difference is

not significant (removal difference <5% at 48 h contacting time). The commercial activated carbon, on the other hand, yielded 50% removal at the dosage of 0.1 g/L and the removal increased to 70% at 0.3 g/L dosage.

Mycophenolic acid (Partitioning experiments):

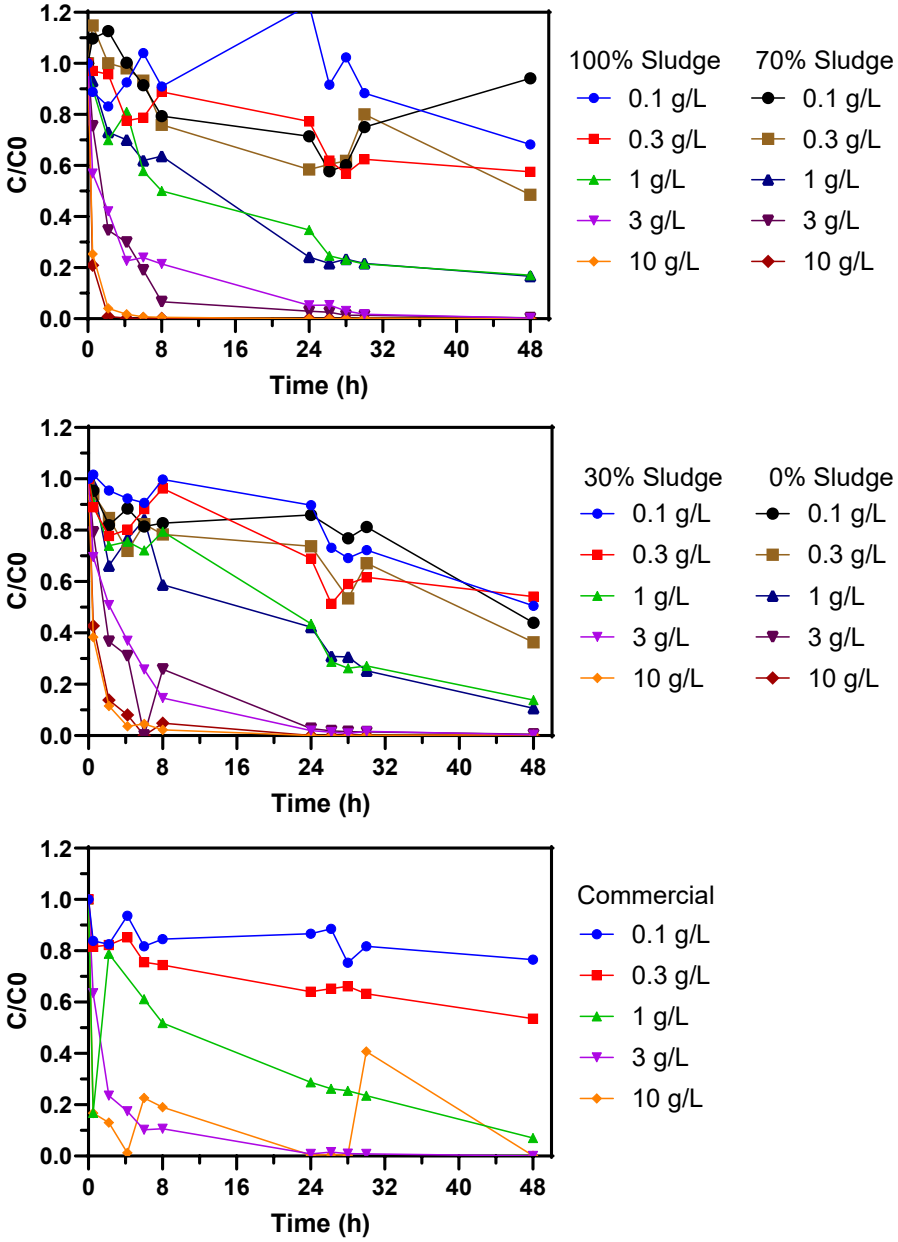

Figure 67: Comparison mycophenolic acid removal with different GAC in the partitioning experiment.

The partitioning experiment on mycophenolic acid removal suggested biochar with dosage 0.3 g/L or less could only yield < 40% removal. With dosage at 1 g/L or higher, the biochar could remove > 80% mycophenolic acid. There's no noticeable difference in mycophenolic acid removal between different composition of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 0.1 g/L and the removal increased to 40% at 0.3 g/L dosage.

Olmesartan (Partitioning experiments):

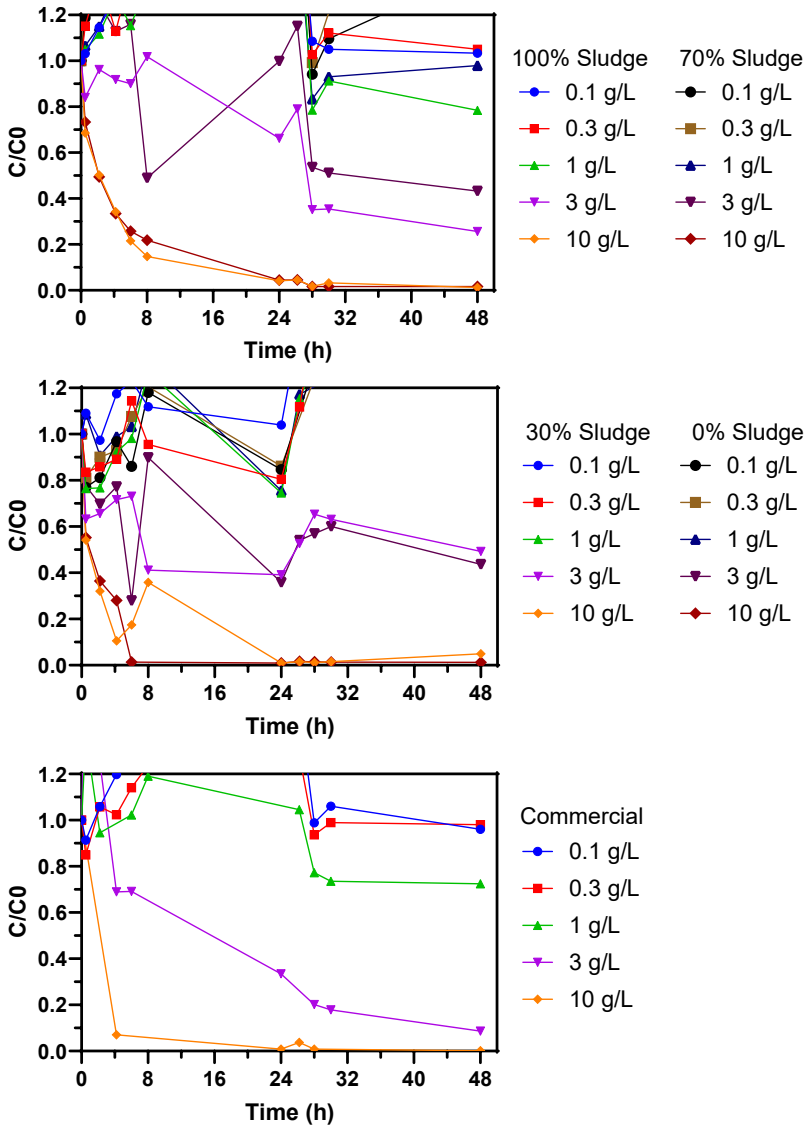

Figure 68: Comparison of olmesartan removal with different GAC in the partitioning experiment.

The partitioning experiment on olmesartan removal suggested biochar with dosage 1 g/L or less could only yield < 20% removal. With dosage at 3 g/L or higher, the biochar could remove > 50% olmesartan. There's no noticeable difference in olmesartan removal between different composition

of biochars. The commercial activated carbon, on the other hand, yielded 20% removal at the dosage of 1 g/L and the removal increased to 90% at 3 g/L dosage.

8.3 GAC removal on PFAS in partitioning experiments

The figure showing the removal of PFAS using different types of activated carbon is listed here.

PFOA (Partitioning experiments):

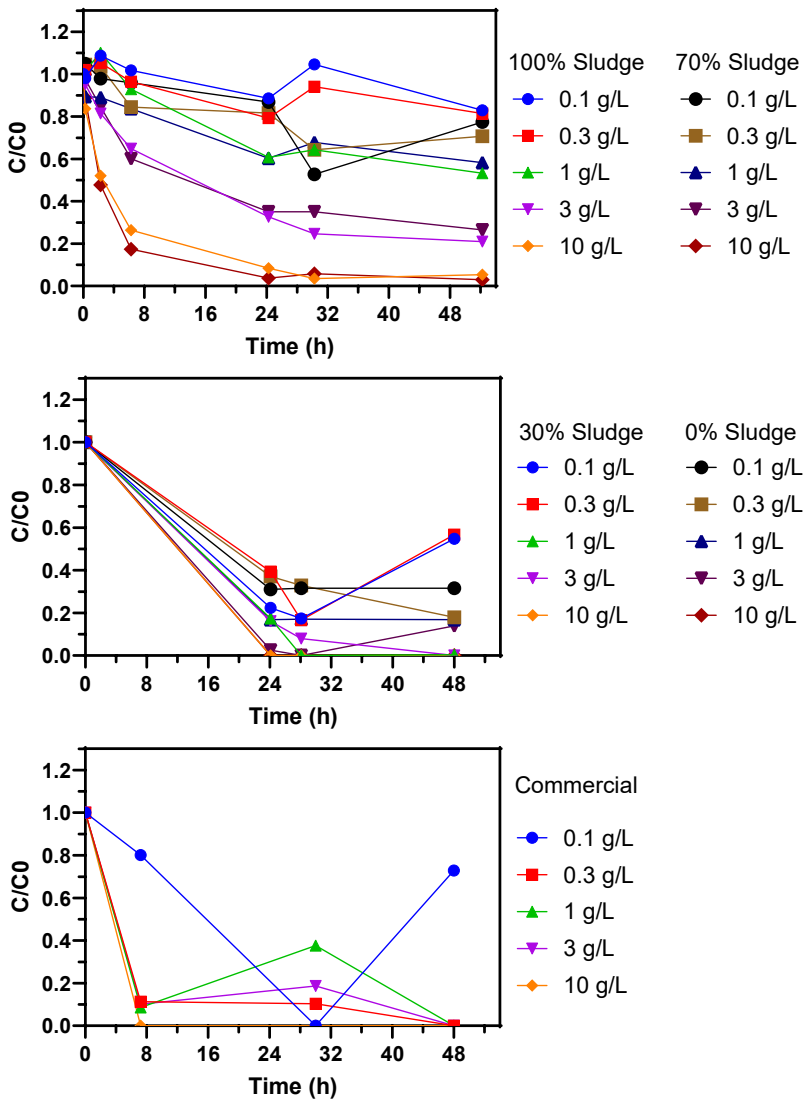

Figure 69: Comparison of PFOA removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing PFOA is similar as the remaining PFOA concentration in wastewater after 48 h contacting time for both

materials at the same dosage is very close (difference <0.05). Less than 20% PFOA was removed with 0.1 g/L biochar and less than 25% of it was removed when the dosage was increased to 0.3 g/L. For these two materials, more than 1 g/L of biochar is needed to remove 50% PFOA or more after 48 h (indicating towards a capacity of 1000 BV, if 50% removal is sufficient). When the dosage increased to 3 g/L, the removal became ~80% and the removal is >95% when the biochar was dosed at 10 g/L level (capacity 100 BV). In terms of the removal of PFOA by commercial activated carbon and biochars made from 30% sludge and 0% sludge, the biochar with 30% sludge removed less PFOA than the biochar with 0% sludge at lower dosage (0.1 and 0.3 g/L) and at higher dosage (1 g/L and above), the biochar with 30% sludge is able to remove ~99% PFOA (capacity 1000BV). For comparison, the commercial activated carbon can remove ~99% of PFOA with 0.3 g/L dosage after 48 h contacting time (indicating towards a capacity exceeding 3000 BV).

PFOS (Partitioning experiments):

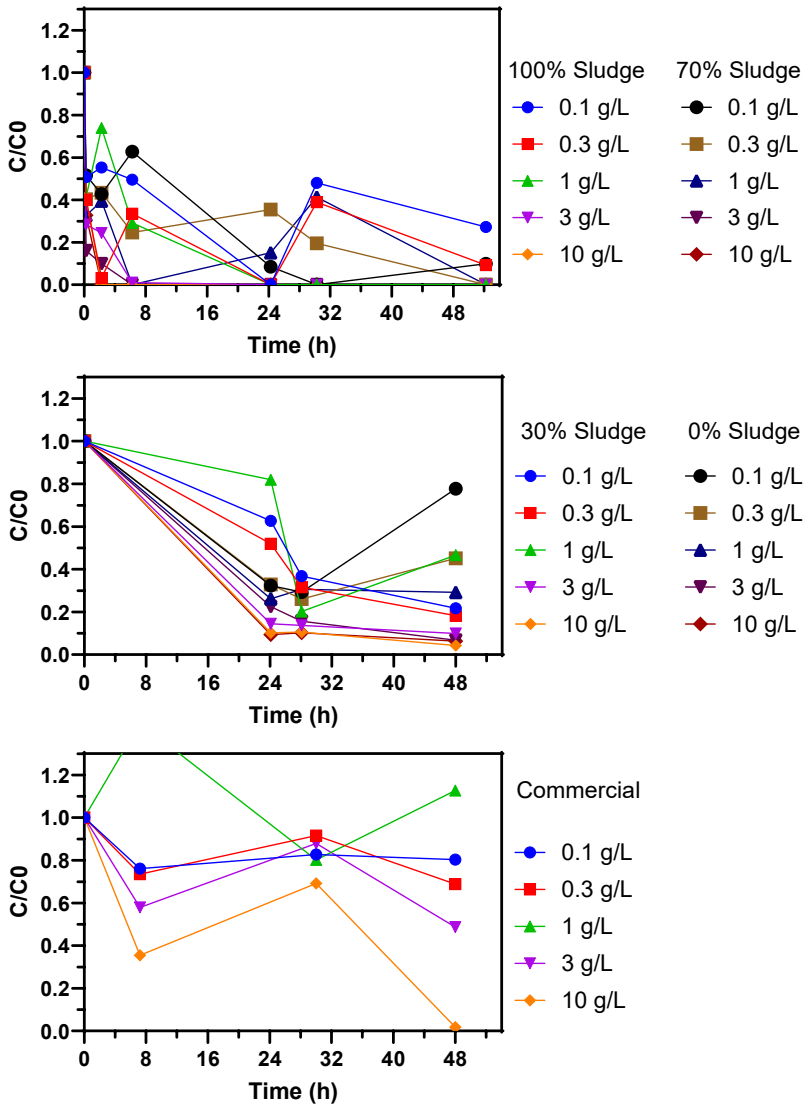

Figure 70: Comparison of PFOS removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing PFOS is similar as the remaining PFOS concentration in wastewater after 48 h contacting time for both materials at the same dosage is very close (difference <0.05) and they can achieve at least 60% removal of PFOS. In terms of the removal of PFOS by commercial activated carbon and biochars made from 30% sludge and 0% sludge, the biochar with 30% sludge and with 0% sludge is able

to remove ~95% PFOS (capacity 300 BV) at the dosage of 3 g/L or above. For comparison, the commercial activated carbon can remove ~70% of PFOS with 3 g/L dosage after 48 h contacting time (indicating towards a capacity less than 300 BV).

PFHpA (Partitioning experiments):

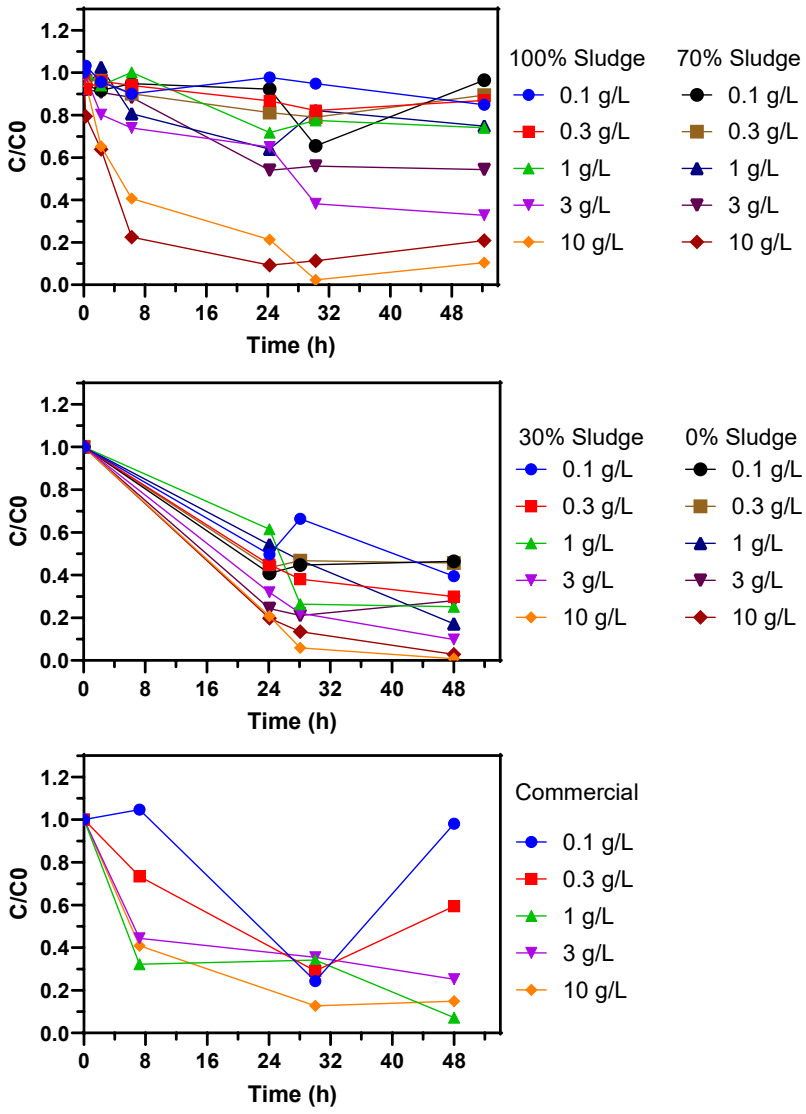

Figure 71: Comparison of PFHpA removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing PFHpA is similar as the remaining PFHpA concentration in wastewater after 48 h contacting time for both materials at the lower dosage range (0.1 – 1 g/L) is very close (difference <0.05). Less than 10% PFHpA was removed with 0.1 g/L biochar and less than 15% of it was removed when the dosage

was increased to 0.3 g/L. ~ 20% PFHpA was removed at 1 g/L biochar dosage after 48 h contacting time. The biochar made with 100% sludge removed more PFHpA at 3 g/L (~60% removal) compared to biochar with 70% sludge (~50% removal). Both materials can remove >80% PFHpA at 10 g/L dosage. In terms of the removal of PFHpA by commercial activated carbon and biochars made from 30% sludge and 0% sludge, no distinctive difference can be observed between the biochars made with 30% sludge and with 0% sludge. For comparison, the commercial activated carbon can achieve >40% PFHpA removal with 0.3 g/L dosage or above after 48 h contacting time.

ADONA (Partitioning experiments):

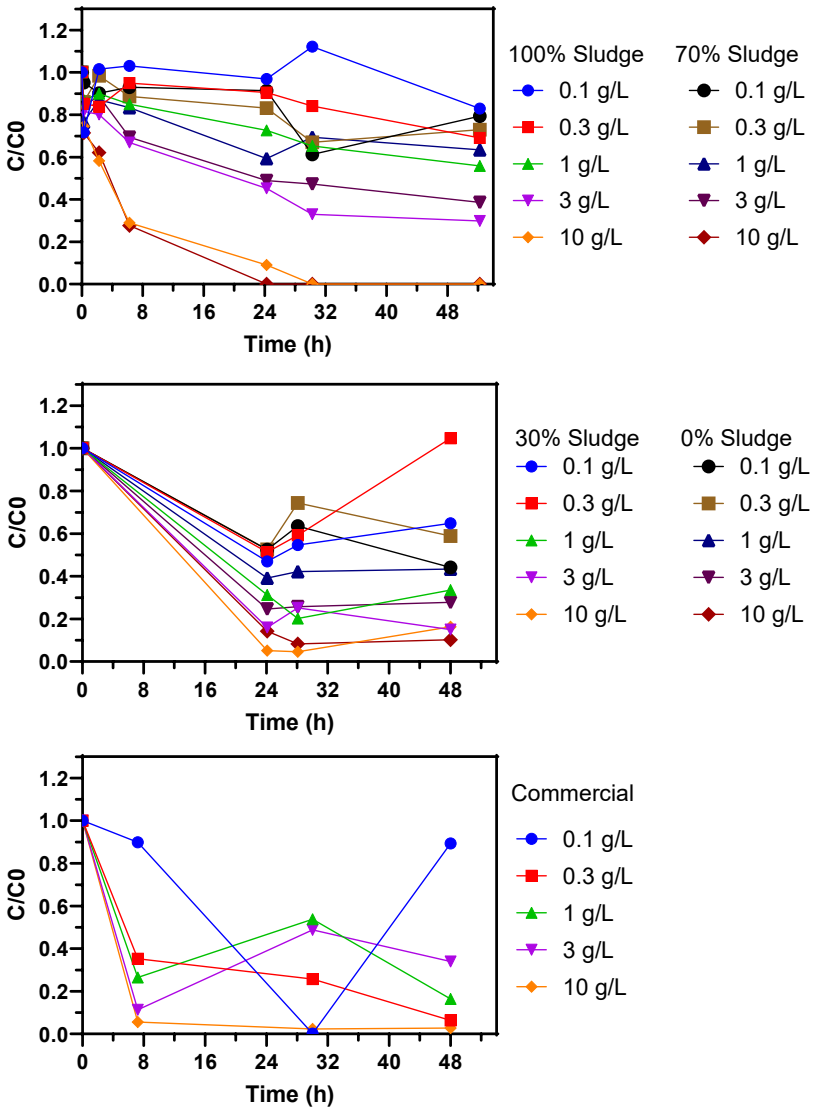

Figure 72: Comparison of ADONA removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing ADONA is similar as the remaining ADONA concentration in wastewater after 48 h contacting time for both materials at the same dosage is very close (difference <0.05). Approximately 10% ADONA was removed with 0.1 g/L biochar and ~20% of it was removed when the dosage was increased to 0.3 g/L. ~ 40% ADONA was removed at 1 g/L biochar dosage after 48 h contacting time. Both

materials can remove >50% ADONA at 3 g/L dosage and >99% at 10 g/L dosage. In terms of the removal of ADONA by commercial activated carbon and biochars made from 30% sludge and 0% sludge, no distinctive difference can be observed between the biochars made with 30% sludge and with 0% sludge. For comparison, the commercial activated carbon can achieve >60% ADONA removal with 0.3 g/L dosage or above after 48 h contacting time.

PFHxA (Partitioning experiments):

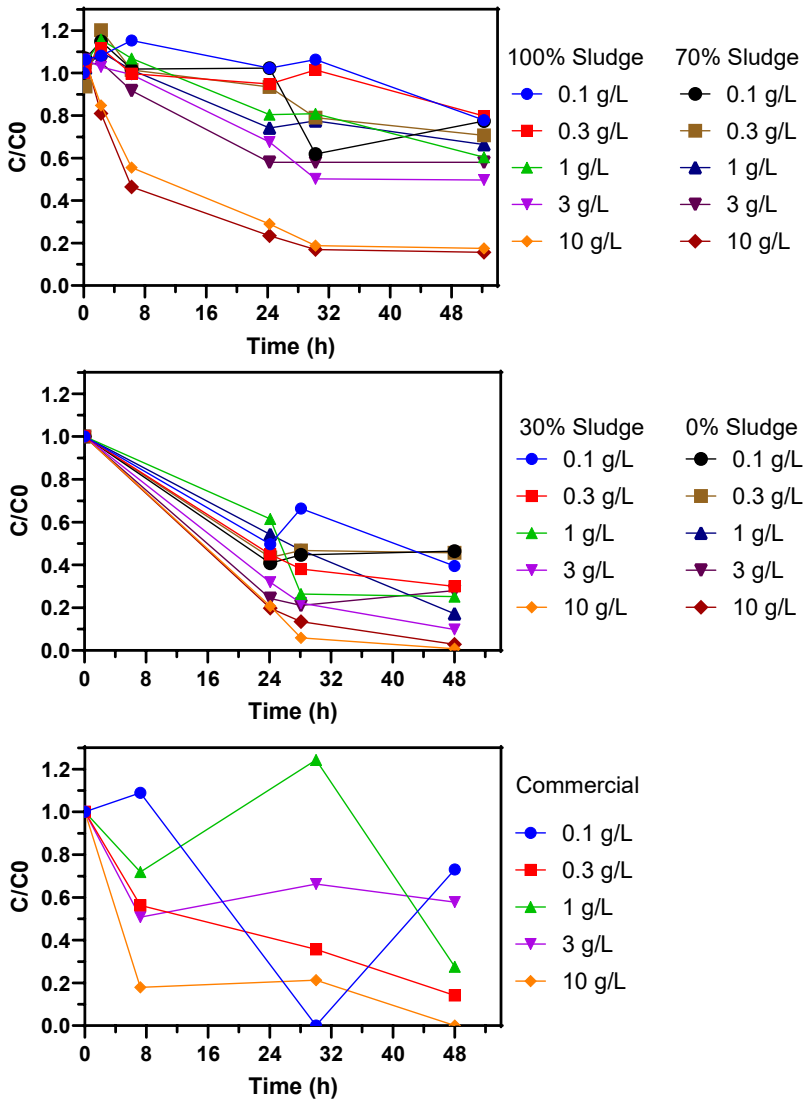

Figure 73: Comparison of PFHxA removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing PFHxA is similar as the remaining PFHxA concentration in wastewater after 48 h contacting time for both materials at the same dosage is very close (difference <0.05). Approximately 20% PFHxA was removed with 0.1 g/L biochar and ~25% of it was removed when the dosage was increased to 0.3 g/L. ~ 40% PFHxA was removed at 1 g/L biochar dosage after 48 h contacting time. Both materials

can remove ~50% PFHxA at 3 g/L dosage and ~80% at 10 g/L dosage. In terms of the removal of PFHxA by commercial activated carbon and biochars made from 30% sludge and 0% sludge, no distinctive difference can be observed between the biochars made with 30% sludge and with 0% sludge. For comparison, the commercial activated carbon can achieve >80% PFHxA removal with 10 g/L dosage after 8 h contacting time.

PFHxS (Partitioning experiments):

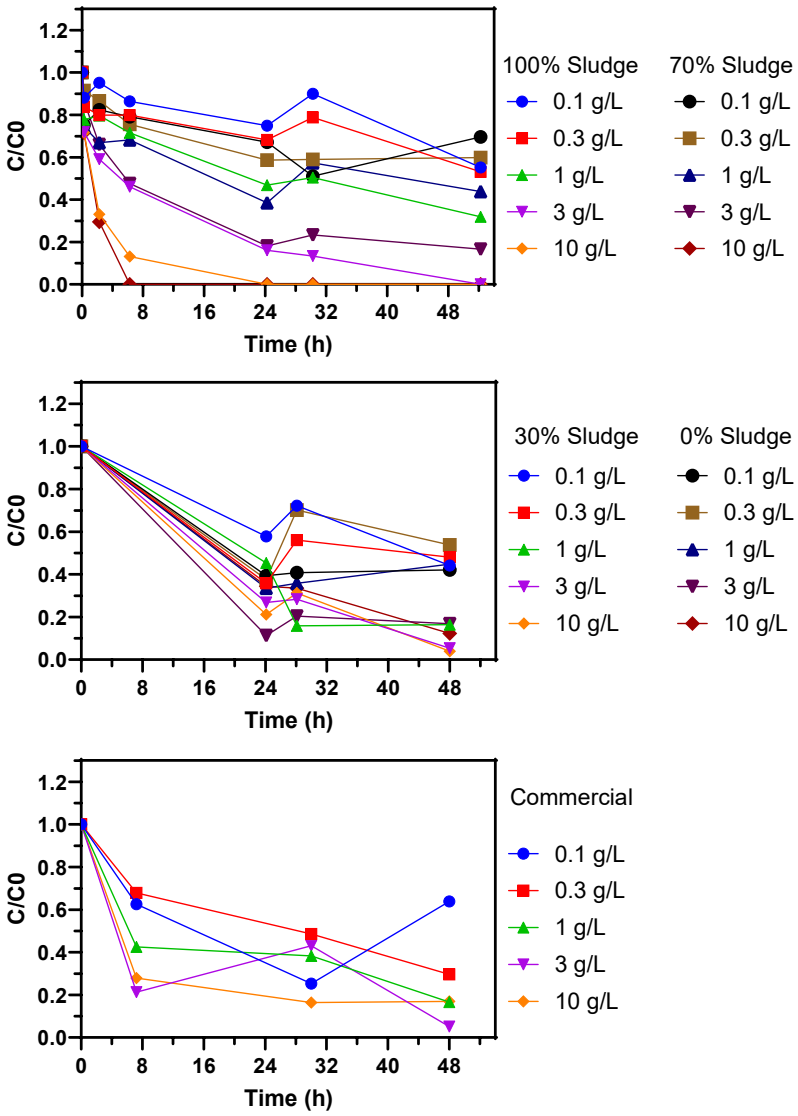

Figure 74: Comparison of PFHxS removal with different GAC in the partitioning experiment.

The performance of biochars made form 100% sludge and 70% sludge in removing PFHxS is similar as the remaining PFHxS concentration in wastewater after 48 h contacting time for both materials at the same dosage is very close (difference <0.05). Approximately 40% PFHxS was removed with 0.1 g/L biochar and ~50% of it was removed when the dosage was increased to 0.3

g/L. ~ 70% PFHxS was removed at 1 g/L biochar dosage after 48 h contacting time. Both materials can remove 80% PFHxS or more at 3 g/L dosage (after 48 h) and ~99% at 10 g/L dosage (after 24 h). In terms of the removal of PFHxS by commercial activated carbon and biochars made from 30% sludge and 0% sludge, no distinctive difference can be observed between the biochars made with 30% sludge and with 0% sludge. Both biochars (made from 30% sludge and 0% sludge) can remove >80% PFHxS after 48 h when the dosage is greater than 3 g/L. For comparison, the commercial activated carbon can achieve >60% PFHxS removal with 0.3 g/L dosage after 48 h contacting time.