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# Biochar-Based Subsurface-Flow Wetland from Crumb Rubber Scrap in Treatment of Landfill Leachate

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# Abstract

Leachate resulting from the decomposition of organic waste is still a challenging problem, especially in landfill management. Constructed wetlands (CW) are effective, economical, and environmentally friendly options to treat landfill leachate. Biochar added into CW as a pollutant adsorbent in leachate treatment. This study aimed to determine the effectiveness of sub-surface flow CW amended with biochar in reducing leachate pollutants. Biochar was synthesized from crumb rubber scrap waste using pyrolysis. The variation biochar in CW i.e. CWo without biochar as a control system, CW1 10% biochar, CW2 20% biochar, and CW3 30% biochar. Leachate samples flowed into each CW reactor for 10 days of retention time. The scanning electron showed that the biochar pores ranged from 5-10 µm, containing elements of C, O, Ca, N, Mg, Al, Si, and Fe. Some elements have greater cumulative mass and atomic percentage values i.e. C 34.51%, O 26.54%, and Ca 21.85%. The result of CW treatment showed that the CW system was able to remove 76-88% BOD<sub>5</sub>, 70-87% COD, and 67-81% TSS. The addition of biochar in CW increased pollutant removal by 7-14%, showing that biochar is able to increase pollutant adsorption in wastewater and improve CW performance. Furthermore, converting organic waste into biochar is highly recommended as a sustainable way to generate useful resources.

Keywords: Biochar; leachate; constructed wetlands

## 1. Introduction

Leachate is generated from liquid precipitation due to the decomposition of organic waste. Landfill leachate treatment is a challenging problem due to its complex pollutant content (Saeed et. al, 2021; Joseph et. al, 2020). The composition of landfill leachate varies due to differences in moisture content, waste composition, weather, hydrology, rainfall, interaction with the environment, waste compaction, and others. Pollutants generated from municipal waste include organic and inorganic compounds, Xenobiotic Organic Compounds (XOC), and heavy metals from industry or households (Chen et al., 2021 & Luo et al., 2019). The analysis of landfill leachate water in Kudus Regency has a BOD (Biological Oxygen Demand) content of 230 mg/L, COD (Chemical Oxygen Demand) of 662.256 mg/L, and TSS (Total Suspended Solid) content of 64 mg/L (Ramadhani et al., 2020).

Conventional landfill leachate treatment with aerobic-anaerobic systems such as aeration ponds, anaerobic ponds, facultative, and maturation still leaves pollutants that exceed the standard before being discharged into the environment. The addition of physical-chemical processes with coagulation and

flocculation methods at the pre-treatment stage is considered to reduce COD, BOD, and TSS levels in leachate by 24-65% (Saeed et al., 2021). However, the application of this technology on a larger scale requires high costs. Therefore, a sustainable approach is needed in treating this landfill leachate more effectively and economically.

Constructed wetland (CW) is a system that has been widely used to treat various types of waste because it is considered very effective in reducing various pollutants in wastewater, low energy consumption, easy operation, and ecosystem services (Deng et. al., 2019 & Riyanti et. al., 2019). Wastewater degradation in constructed wetlands will occur through three processes, namely physical, chemical, and biological including nutrient adsorption by plants, filtration, sedimentation, and most importantly degradation by microbes (Perdana et al., 2020) Microorganisms that live in plant roots play a role in decomposing organic matter in wastewater with the assistance of oxygen. This constructed wetland system is a low-cost wastewater treatment in terms of construction and operation, eco-friendly, high sustainability, and aesthetic, so it can be an attractive alternative for landfill leachate treatment (Li et al., 2019).

The process in constructed wetlands occurs aerobic and anoxic/anaerobic where the anaerobic process is more dominant. Atmospheric oxygen diffuses into the media during dry periods, supporting nitrification and removal of organic matter in wastewater (Saeed, 2019). Microorganisms play an important role in transforming and removing contaminants in constructed wetlands and improving the operation of constructed wetlands (Jia et al., 2019). Biological processes in CW involve plant uptake, decomposition, denitrification, transformation, and nutrient storage. The reduction of pollutant concentration in CW systems is influenced by the number of plants and retention time.

Media selection is important in improving CW performance. Commonly used media in CW are sand, gravel, and soil. Recent studies have suggested that the addition of biochar such as sand/gravel-based CWs can increase nutrient removal and macrophytes growth (Deng et. al., 2019; Kasak et. al., 2018; Zhou et. al., 2018b). Biochar has the ability to remove contaminants from landfill leachate, including Volatile Organic Compounds (VOCs) (Jayawardhana et al., 2019), ammonia, COD, nitrate, phosphate, dyes, and heavy metals (Sandoval-Herazo et al., 2018).

Biochar is a porous carbon product derived from biomass with the abundance and chemical properties of the carbon components dependent on its source material. Biochar is an attractive option for environmental remediation because it has a large surface area, high porosity, and abundant function making it an effective adsorbent in removing various organic and inorganic contaminants in wastewater (Joseph et al., 2020). The use of biochar as an adsorbent and at the same time as a source of nutrients to improve soil fertility has been widely researched.

Biochar in CW can support microbial growth that is different from other media (Ji et al., 2020). The addition of biochar in constructed wetlands is effective in reducing pollutants in wastewater. Subsurface flow CW added with biochar provides removal efficiency for ammonium (49.69% - 63.51%) and total nitrogen (81.83% - 86.36%) (Deng et al., 2019). Constructed wetland using biochar with a retention time of 3 days has a BOD5 removal efficiency of 91.3% while the combination of biochar and soil has a BOD5 removal efficiency of 95% (Feng et al., 2023).

Rubber scrap waste is an organic solid waste resulting from the disposal of the crumb rubber industry that contains mostly sand, rubber wood chips, rubber leaves, and rubber. Crumb rubber scrap biochar is an effective adsorbent in reducing organic pollutants because it has a high mechanical strength and porosity. The use of this biochar in peat water filtration can neutralize the pH, reduce organic compounds to 83% and color 75% (Riyanti et al., 2023). With its high adsorbent capacity, crumb rubber scrap biochar will be effective for removing pollutants from landfill leachate.

This study discuss the performance of subsurface CW with biochar from crumb rubber scrap to reduce pollutants in landfill leachate in East Tanjung Jabung Regency, Jambi Province. Syngonium podophyllum will be used as plants in the CW which has a large root system so it is effective in absorbing pollutants.

## 2. Methods

## 2.1. Experimental Set Up

Biochar was prepared from solid waste of crumb rubber scrap. Biochar synthesis was carried out by pyrolysis method at 350°C for 1 hour. The pyrolyzed biochar was cooled and washed. After washing, the drying process is carried out using an oven with a temperature of 105 oC for one hour.

The plant to be used in the CW system is Syngonium podophyllum. Plant acclimatization is carried out in stages with an initial ratio of leachate: clean water as much as 1: 4, namely 2 liters of leachate: 8 liters of clean water. In diluting the leachate water, stirring was carried out so that it was homogeny us before being put into the CW and observed for 3 (three) days. After 3 (three) days, acclimatization was carried out again using a ratio of 3:2, namely 6 (six) liters of leachate water and 4 liters of clean water for 3 (three) days to ensure that the plants did not wither or die. The plants were neutralized again with clean water for 1 (one) day before the experiment was carried out (Riyanti et al., 2019).

The CW system used in this study is sub-surface flow wetland (SSF-Wetlands), because this system uses planting media such as sand or soil so it is more suitable for adding biochar. The planting media in CW uses sand, biochar and gravel at the bottom of the basin.

This study is a laboratory-scale batch experiment consisting of 4 CW reactors with dimensions of 30 cm high and 20 cm diameter. CWo serves as a blank with planting media in the form of sand and gravel without biochar. While CW1, CW2, and CW3 were added biochar was mixed with sand. The biochar variations in CW1 are as much as 10%, CW2 as much as 20%, and CW3 as much as 30%. The retention time in each CW reactor is 10 days.

## 2.2. Sampling and analysis

#### 2.2.1. Biochar analysis

Biochar from crumb rubber scrap is analyzed using SEM EDX Mapping to see the morphology and analyze the compound elements in biochar. SEM EDX Mapping will show the magnification of various shapes and sizes of pores as well as the number and distribution of compound elements contained in biochar.

#### 2.2.2. Leachate sampling and analysis

Samples of influent and effluent water in each CW reactor will be collected at days 10 and then analyzed for pollutant concentrations in the laboratory. The parameters observed are BOD<sub>5</sub>, COD, and TSS. The performance of CW treatment is assessed from the amount of pollutant removal in influent and effluent. This study also looks at the effect of varying the amount of biochar on reducing pollutant concentrations in CW.

## 3. Result and Discussion

## 3.1. Scanning Electron of Biochar

Biochar synthesized from crumb rubber scrap through pyrolysis was tested using the Sem EDX Mapping test to determine the size of the pores and the compounds contained therein. The morphological structure of the biochar produced is in the form of crystals arranged in clumps of different sizes and irregular patterns called amorphous. The pore size of the biochar formed ranges from 5-10 µm. The more pores in biochar, the better the absorption of pollutants. Scanning Electron Microscope (SEM) images of biochar at various magnifications are shown in Figure 1.

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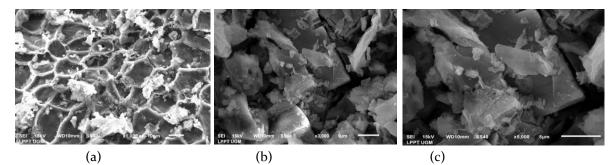


Figure 1. SEM image of with various pore sizes 5-10 µm in magnification (a) 1000x; (b) 3000x; (c) 5000x.

The results of scanning electron show that the elements contained in biochar are C, N, O, Mg, Al, Si, Ca, and Fe, where the highest element is the element C (Carbon), followed by O (Oxygen), Ca (Calcium) and N (Nitrogen), while other elements have smaller amounts.

Element	<b>Mass (%)</b>	Atom (%)	Cumulative (%)		
С	34.75	43.83	34.51		
0	35.55	33.66	26.54		
Ca	11.87	4.49	21.85		
Ν	15.71	17.00	14.15		
Al	0.67	0.37	0.87		
Fe	0.60	0.16	0.87		
Si	0.57	0.31	0.85		
Mg	0.29	0.18	0.32		
Total	100.00	100.00			

Table 1. Atomic mass of biochar

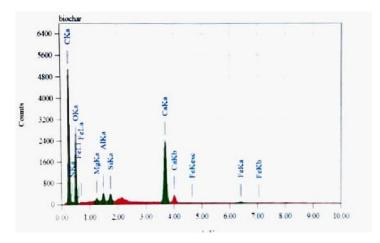


Figure 2. Distribution of elements in biochar

Based on Table 1 and Figure 2. there are four elements that have a greater cumulative mass and atomic percentage value. namely the C element of 34.51%. O of 26.54%. Ca of 21.85%. and N of 14.15%. While several other elements including Al. Fe. Si. and Mg are in lower amounts. The scanning electron EDX results show the distribution of elements in biochar which can be seen from the different colors of each element (Figure 3. 4. and 5).

Riyanti et al. 2024. Biochar-Based Subsurface-Flow Wetland from Crumb Rubber Scrap in Treatment of Landfill Leachate J. Presipitasi, Vol 21 No 1: 51-60

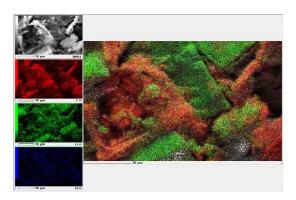


Figure 3. Distribution of C. O. and N

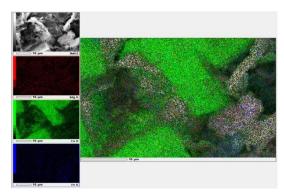


Figure 4. Distribution of Mg. Ca. and Fe

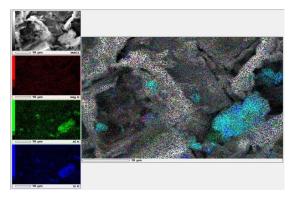


Figure 5. Distribution of Mg. Al. and Si

## 3.2. Removal pollutant in leachate by Constructed Wetlands (CW)

Initial leachate quality testing (influent) aims to determine the concentration of pollutants contained in leachate water before treatment. including  $BOD_5$ . COD. and TSS parameters (Table 2).

Parameter	Influent Concentration (mg/L)	Quality standard* (mg/L)				
BOD <sub>5</sub>	445.17	150				
COD	727.60	300				
TSS	274.12	100				
*Source: (PermenLHK Nomor 59/2016)						

 Table 2. Leachate characteristic (Influent)

The concentration of pollutants in the influent showed that the levels of COD. TSS. and BOD5 contained in the leachate water were far above the quality standards. Furthermore. the leachate water

flowed into the CW reactor with different amounts of biochar. The leachate water was allowed to stand with a retention time of 10 days and water samples in the effluent were taken on the 10th day for analysis. The results of pollutant concentrations in the effluent are listed in Table 3.

Table 3 shows that in the performance of constructed wetland with no biochar (CW o) as a control system. the concentration of BOD<sub>5</sub>. COD. and TSS decreased significantly. where BOD<sub>5</sub> decreased by 76.54%. COD 70.71% and TSS 89.73%. This shows that the removal of pollutants with CW system is effective even without biochar. Wastewater treatment with CW occurs through three mechanisms namely physical. chemical. and biological processes (Perdana et al., 2020) that occur between media. plants. and microorganisms. The physical process consists of filtration and sedimentation. After the leachate enters the CW. it flows slowly through the media in the CW. This slow flow keeps the solids suspended. filtration by plant roots and media against pathogens and organic matter. In CW o. sand media and Syngonium podophyllum plants were able to remove organic pollutants. turbidity. and color due to suspended solids.

Parameter	Effluent concentration in biochar variation								
	No biochar (CWo)	Removal (%)	Biochar 10% (CW1)	Remova 1 (%)	Biocha r 20% (CW2)	Remova 1 (%)	Biocha r 30% (CW3)	Removal (%)	
BOD <sub>5</sub>	104.41	76.54	83.16	81.31	74.62	83.23	52.12	88.29	150
COD	213.12	70.71	149.07	79.51	129.17	82.24	93.86	87.10	300
TSS	89.73	67.26	71.12	74.05	54.13	80.25	50.19	81.69	100

Table 3. Effluent concentration and removal rates of different biochar variation

CW system with no biochar is able to reduce the concentration of BOD5. COD. and TSS to meet the quality standard. The removal process occurs biologically through the role of Syngonium podophyllum plants. Organic compounds in wastewater. represented by COD and BOD5. can be effectively removed in CW systems through aerobic and anaerobic degradation (Saeed and Sun, 2017 and Zhao et al., 2020). Organic matter contained in leachate is absorbed through plant roots and then distributed into plant tissues as nutrients for photosynthesis. Microorganisms that live in plant roots play a role in decomposing organic particles in water with the help of oxygen transfer by plants. The oxygen flows to the roots through the stem after diffusing from the atmosphere through the pores of the leaves (Sandoval-Herazo. 2018).

Syngonium podophyllum plants used in CW systems can grow well. have a root system that is able to adapt to polluted soil conditions in CW. and can survive until day 10. Plants in CW contribute greatly to the removal of pollutants mainly through assimilation and rhizofiltration. Plants create a suitable microenvironment for microbial communities through the release of oxygen and exudates from plant roots (Kumar and Dutta, 2019). Root exudates release carbon and various compounds needed by soil organisms. The microbes assist plants in providing nutrient requirements through their enzymatic activities.

While the chemical process occurs through the adsorption of pollutants by biochar. The greater the amount of biochar added. the lower the concentration of pollutants. The decrease in pollutant parameter levels can be influenced by the addition of biochar which acts as an absorbent (Chen et al., 2021). The scanning electron results show that biochar has an elemental content. namely carbon (C) of 34.5% which increases the ability to absorb pollutants.

The reduction of pollutants in CW also occurs due to the activity of microorganisms with plants in wetlands (Sandoval-Herazo. 2018). While oxygen is needed to decompose organic matter contained in leachate aerobically by microorganisms. Oxygen supply is obtained by microorganisms from the photosynthesis process of plants. water. algae. and biofilms. The presence of oxygen in biochar helps the degradation process of organic matter so that the removal of BOD5 and COD becomes greater. The greater the amount of biochar used. the more pollutant removal increases.

Zhou et al. (2019) mentioned that nitrogen and oxygen support in reduction of organic substances in water through filtration. Nitrogen forms proteins that can bind organic matter and also function to accelerate the formation of the biofilm layer. while oxygen is used for the respiration process. degradation of organic and inorganic materials. the process of metabolism. and exchange of substances that can help reduce organic substances in peat water. The greater the amount of dissolved oxygen. the better the water quality. It can be seen in Table 2 that only one sample met the quality standard for organic matter. However, the concentration of organic matter also decreased, indicating that each treatment variation was effective in reducing the concentration of organic matter. The filtration media, biochar and silica sand form the biofilm layer and absorb organic substances as well as sediment.

Naturally. wetlands function as sediment precipitators. The decrease in TSS occurs because solids in leachate water will be attached to plant parts and absorbed by biochar. The mechanism of action in CW shows the role of plants and biochar in the constructed wetland system. namely as pollutant catchers (Wimbaningrum et al., 2020). Organic compounds in leachate that react with OH radicals are then oxidized and produce simpler compounds. In addition. the distribution of elements in biochar can help reduce TSS is the element of oxygen (O). calcium (Ca), aluminum (Al), and Silica (Si) (Singh et al., 2020) where the biochar used contains O 26.5%. Ca 21.8%. Al o.87%. and Si o.85%. Oxygen and calcium elements form quicklime which increases TSS removal. While aluminum functions to form positively charged ions so that they are easily dissolved and silica helps purify water and remove suspended solids in water.

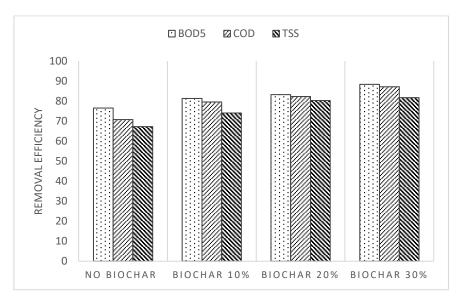


Figure 6. Removal efficiency of BOD5. COD dan TSS in biochar variations

The increase in pollutant removal in CW with the addition of biochar at CW1 10%, CW2 20%, and CW3 30% in Figure 6 shows that biochar is able to become an adsorbent in absorbing pollutants in leachate water and improving constructed wetlands performances. The efficiency of pollutant removal in leachate water increased by 7-14% with the addition of biochar in CW. The removal of BOD5. COD. and TSS increased to 81-88% in CW3 with 30% biochar. The more pores formed on biochar also increase the adsorption capacity of pollutants in wastewater.

Biochar has a high adsorption capacity for organic compounds and provides a heterogeneous surface with many pores for oxygen replenishment. Some researchers found that the addition of biochar in CW increased the abundance and metabolic activity of heterotrophic bacteria for organic degradation thus alleviating the clogging of the substrate in CW (Deng et al., 2019).

In addition. biochar enhances plant growth better than the control system without biochar (CW o) by releasing nutrients. such as K. P. N. Mg. and Ca. from the biochar (Kasak et al., 2018; Li et al., 2019) as well as increasing the porosity of the media which favors better oxygen conditions (Werner et al., 2018).

Better plant growth then releases more oxygen into the CW for aerobic degradation of organic compounds.

Toxic contaminants in wastewater in CWs can be absorbed by biochar. thereby reducing phytotoxicity (Mohanty et al., 2018). In addition. biochar media can increase root aerenchyma tissue and macrophytes porosity in CWs. favoring increased root oxygen release and thus enhancing aerobic microbial metabolism including organic degradation. nitrification. and methane oxidation (Huang and Gu. 2019). Biochar in CW promotes better plant growth in CWs which is beneficial for nutrient and organic matter removal through plant uptake. oxygen release. secretion of organic exudates. and providing more space for microbial attachment in the plant rhizosphere (Zhou et al., 2019). This makes CW systems an efficient green technology for wastewater conservation and remediation (Deng, 2021).

Physical observations showed that there was odor removal in the leachate every day until day 10. Meanwhile. the condition of singonium plants observed every day until day 10 did not wilt. On days 0 to 6. the plants still looked fresh. while on days 6 to day 10. there was growth of new leaf shoots on the plants. This shows that singonium plants are able to survive and grow well in the polluted environment of constructed wetlands. This plant is able to utilize the pollutants in the leachate water as nutrients to survive and grow.

Retention time influences the level of permeability or permeability of the media. Sufficient retention time will provide oxygen released by plant roots and increase the opportunity for contact between microorganisms and wastewater in degrading pollutants. Studies revealed that the removal efficiency of pollutants in constructed wetlands systems usually increases with increasing retention time. where the longer the retention time the greater the nutrient removal. The most effective retention time was reported from 4 to 15 days (Metcalf and Eddy. 1991). Removal of BOD5 and COD in CW with a retention time of 1 to 7 days has not provided a significant decrease in concentration. where the decrease in BOD and COD concentrations is not stable. experiencing up and down each day (Kasman et al., 2022). While BOD5 removal at 15 to 45 days of retention time in this study is a reasonable time to develop the bacterial colonies in CW rooting. which is essential for pollutant removal. The result showed that with 10 days retention time, the removal of pollutants in leachate water achieved up to 88% in the CW3, until the concentration of the three parameters meets the standard.

## 4. Conclusions

Synthesis of solid waste crumb rubber scrap into biochar through pyrolysis produces biochar with pores ranging from 50 µm to 5 µm. Scanning electron shows the structure of biochar in the form of crystal grains arranged in clumps of different sizes and with irregular patterns (amorphous). The element contents of biochar are C. O. Ca. and N with higher atomic mass. followed by Al. Fe. Si. and Mg.

In leachate treatment using sub-surface flow constructed wetlands without biochar was able to remove BOD5. COD. and TSS with an efficiency of 67-76%. The addition of biochar to the CW media can increase the pollutant removal value to 81-88% in CW with 30% biochar. In this study, a greater amount of biochar in CW media resulted in better pollutant removal. The high carbon content in this biochar can increase the adsorption of pollutants thus improving CW performance. The addition of biochar increases plant growth. which in turn increases oxygen release in plant roots that support the degradation process of organic pollutants. while the pores in biochar can purify water and increase suspended solid removal in leachate. Overall, addition biochar as a substrate in CW is promising to enhance pollutant removal in wastewater.

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