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Application of Vertical Flow Constructed Wetland for Organic Pollutant Removal from Petroleum Refinery Wastewater

Amelia Eka Lestari [1*)], I Nyoman Widiasa[1]

[1)]Department of Chemical Engineering, Faculty of Engineering, Universitas Diponegoro
Jl. Prof. Soedarto, SH, Tembalang, Semarang

[*)] Corresponding author: ameliaekalestari83@gmail.com

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Abstract

This study examined the application of a pilot-scale vertical flow constructed wetland (VFCW) system for secondary oil refinery effluent treatment at the Centre for Human Resources Development of Oil and Gas (PPSDM MIGAS), Indonesia. The VFCW technique, known for its simplicity, minimum operational cost, and environmental friendliness, was used to reduce organic pollutants (BOD and COD) level satisfying Indonesian environmental safety standards. The system construction composed of a closed pond planted with Typha angustifolia and supported by gravel and sand substrates. It was evaluated under hydraulic retention times (HRT) of 3, 4, and 5 days. The results showed BOD removal efficiencies of 52.9%, 54.4%, and 53.6%, while the COD removal efficiencies were 35.7%, 49.1%, and 47.2% for HRT of 3, 4, and 5 days, respectively. Statistical analysis (ANOVA) showed no significant difference ($p > 0.05$) in BOD removal efficiencies and COD removal for 4 and 5 days along the studied HRTs. These findings implied diminishing benefits after 4 days of VFCW operations. The limited BOD and COD removal was due to the short acclimatization time (7 days) for Typha angustifolia to drive oxygen sufficiency and form biofilm. These findings underline the capability of the VFCW system to sustainably and economically reduce wastewater contaminants in tropical areas. A 4-day HRT is recommended for practical applications in refinery wastewater treatment with pollutant loads up to complement. Extended acclimatization duration and improved operational settings are recommended to enhance the performance of the VFCW. This study illustrates the feasibility of VFCW as a scalable and sustainable solution for wastewater treatment, especially in the petroleum industry.

Keywords: acclimatization, organic pollutants, removal efficiency, retention time, VFCW.

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INTRODUCTION

The Centre for Human Resources Development of Oil and Gas (PPSDM MIGAS) - Indonesia has petroleum refinery facilities that produce harmful contaminants containing wastewater (Jain et al., 2020). Some of the contaminants can be expressed in terms of organic matter level, viz. biological oxygen demand (BOD) and chemical oxygen demand (COD). Refinery wastewater is usually treated using the American Petroleum Institute (API) and Corrugated Plate Interceptor (CPI) units as the primary process to achieve permitted pollutant levels prior to disposal. Because the primary processing is often insufficient to achieve the targeted levels, further processing is needed. For that reason, biological processes that utilize microbial activity can be applied as the secondary processes (Jain et al., 2020), one of these being the constructed wetland.

Constructed wetland (CW) is an engineered system designed and built to achieve particular quality results from wastewater by utilizing natural processes involving wetland vegetation, soil, and microbes in a controlled environment (Vymazal, 2010). Therefore, it is environmentally friendly with minimal impact losses (Hassan et al., 2021) and a low-cost technology due to its low-maintenance operations (Waly et al., 2022). One type of CW is a vertical flow constructed wetland (VFCW), which is generally a layer (bed) of a porous substrate (Vymazal, 2022), where large amounts of wastewater is fed to its top. The wastewater flows downward through the media (substrate) with air entering to the pores of the media (Retta et al., 2023). It requires smaller dimensions, good oxygen supply, simple hydraulic conditions, but offers high operational performance (Gorgoglione & Torretta, 2018).

Previously, Mustapha et al. (2018) have applied VFCW to treat petroleum refinery wastewater in a tropical climate area in Kaduna, Nigeria. They reported that a VFCW unit with a two-day residence time can reduce BOD and COD levels by 76.0% and 72.7%, respectively. In addition, there is a pilot-scale VFCW study at the Faculty of Engineering, Delta State University, Abraka, Oleh Campus, Nigeria by Aggary et al. (2018), which supported and unsupported plant VFCWs to treat refinery wastewater. Compared to the unsupported one, the BOD and COD removal efficiency of the VFCW supported by plants produced better pollutant removal efficiency by 37.3% - 94.6% and 41.3% - 80.2%, respectively for BOD and COD in two to ten days retention time operations.

Considering the earlier research outcomes, it is necessary to simulate a pilot scale VFCW application at PPSDM MIGAS to provide an overview of its specific performance when it is designed using local Indonesian vegetation and under appropriate operating conditions according to the specific characteristics of its refinery wastewater. The results of this study will be useful as a basis for the development of a full-scale constructed wetland in Indonesia while expanding the scope of understanding and providing a stronger basis for generalizing previous findings or implementing VFCW on a multi-site.

MATERIALS AND METHOD

Vertical Flow Constructed Wetland

A Pilot-scale VFCW was built near the primary refinery wastewater treatment outlet with coordinates 7°08'39.9"S 111°36'10.4"E, using *Typa Agustifolia* plant species, and sand and gravel substrates. The plant and substrates were selected based on their availability in Indonesia and their potential for use in CW to achieve a sufficient performance.

The VFCW dimension design is based on wastewater feed discharge data of 0.326 m^3/d (Q), which was 1/500 times the actual wastewater average discharge from PPSDM MIGAS during January 2024. The thickness of the substrate layer according to the reference (UN-HABITAT, 2008) 0.7 m high (h), the type was 5-40 mm gravel and 1-5 mm sand with an average porosity of 30% or 0.3 (ε), with the configuration illustrated in Fig.1. Five days (τ) as the maximum HRT was determined because based on several previous studies, HRT under 5 days was able to provide an efficiency of removing organic substances over 50% (Varma et al., 2021). The equation (1) calculated the surface area of the VFCW was 7.61 m^2 (A). Furthermore, the length (L) and width (W) of the pond were determined using the recommended L/W ratio of 3:1 (Rahman et al., 2020), which was approximately 5 m: 1.5 m.

$$\tau = \frac{V_{active}}{Q} = \frac{\varepsilon \times h \times A}{Q} = \frac{\varepsilon \times h \times L \times W}{Q} \quad (1)$$

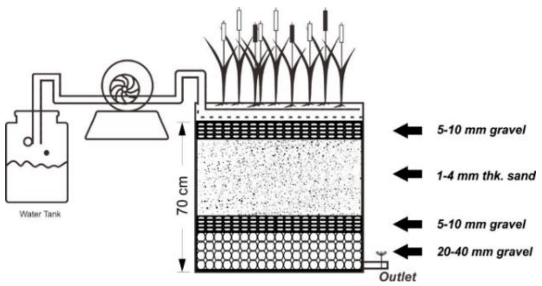

Figure 1. VFCW Configuration

The first preparation was filling the feed tank with secondary refinery wastewater. It was followed by irrigating the VFCW pond with secondary refinery wastewater up to the top surface of the substrate with the outlet valve was perfectly closed. *Typha Angustifolia* was planted on the gravel media with a planting density of one plant per 0.25 m^2 in VSCW and left in the pond for several days as an acclimatization stage.

Operation Process and Sample Collection

Since the acclimatization stage passed, the wastewater was entirely discharged through the outlet valve. The VFCW is rinsed using clean water by filling it vertically and draining it out of the pool completely. Next, the outlet valve was closed again, and the VFCW operation treatment simulation was executed using wastewater fed intermittently. The feed was continuously delivered according to Table 1 at a specific rate for one day (24 hours) using a dosing pump. Because the operation for three HRT variables using the same pond, the daily feed discharge (Q) and the hydraulic loading rate (HLR) would have three variations if calculated using equation (1). As seen in Table 1, the dosing pump worked in the range of 0.315 m^3/d to 0.525 m^2/d or 13 L/hour to 22 L/hour for 3 days, 4 days, and 5 days HRT variables. The process was operated simultaneously from June until July 2024, starting from the 3-day HRT operation.

During the 24-hour feed flow process, wastewater samples were taken from the feed tank/water tank every less than 3 hours to determine the characteristics of the feed. After feeding for 24 hours, the VFCW was allowed to work to degrade pollutants according to the HRT design. The outlet valve from the bottom of the VFCW was opened later than the HRT time had reached. The bottom valve opening was adjusted equal to the feed discharge. Like the feed, every less than 3 hours effluent samples were taken during the 24-hour draining period.

Table 1. Variable Independent

No	HRT (days)	Q (m^3/d)	HRL (m/s)
1.	3	0,525	0,069
2.	4	0,394	0,052
3.	5	0,315	0,041

Analysis and Evaluation

The influent and effluent wastewater were analyzed for their organic levels, viz. their BOD and COD. The analysis of COD levels in samples was performed refer to the APHA 5220 B standard method, while the testing of chemical oxygen levels in water was carried out using a closed reflux device. Furthermore, the BOD analysis was carried out based on the SNI 6989.72.2009 standard method that based on the difference of dissolved oxygen before and after incubation for 5 × 24 hours at a temperature of 20^0C.

The effect of the length of HRT processing operation on VFCW performance was evaluated through the removal efficiency (RE) rate, which is the percentage of output concentration in mg/L (C_{out}) to input concentration in mg/L (C_{in}) (Minakshi et al., 2022; Weerakoon et al., 2020).

$$RE = \frac{C_{in} - C_{out}}{C_{in}} x\, 100\% \quad (2)$$

The data set from laboratory analysis and %RE must be selected using Z-Score to eliminate outliers from the normal distribution by using Xi as data at i, $\bar{X}$ as average data, and S as standard deviation. Data with a Z-Score value of more than 1.96 is an outlier (Kallner, 2014).

$$z = \frac{X_i - \bar{X}}{S} \quad (3)$$

Although the constructed wetland ecosystem is a complex living system (Swarnakar et al., 2022), which is not easy to formulate, but a kinetic modeling approach can be used to predict the results of CW. Several model approaches have been applied in previous studies including the first-order plug flow reactor (PFR) (Aleissa & Bakshi, 2021; Cuong et al., 2022) with k_1 as a reaction rate constant, the first-order continuously stirred tank reactor (CSTR) (Kadlec & Wallace, 2009; Weerakoon et al., 2020) with a reaction rate constant k_2, and a combination of Monod continuously stirred tank reactor with a reaction rate constant k_3 (CSTR) (Nguyen et al., 2018). All constants use output concentration in mg/L (C_{out}), input concentration in mg/L (C_{in}), and q as hydraulic loading rate in m/s. Specific for combination Monod CSTR model kinetic there is Monod half saturation (Ch) constant in g/m^2L.

$$k_1 = (lnC_{in} - lnC_{out})\, x\, q \quad (4)$$

$$k_2 = \frac{C_{in}-C_{out}}{C_{out}} x\, q \quad (5)$$

$$k_3 = \frac{(C_{in}-C_{out})\, x\, (C_h+C_{out})}{C_{out}} x\, q \quad (6)$$

The accuracy of the obtained rate constant was then tested for statistical accuracy using root mean square error (RMSE) and normalize objective function (NOF), where the modeling was declared valid when the NOF value was in the range of 0.0 - 1.0 (Gikas et al., 2011). The formulas need data A_i as the actual value, P_i as the predicted value, N as several data, and $\bar{X}$ as average data.

$$RMSE = \sqrt{\frac{\sum(A_i-P_i)^2}{N}} \quad (7)$$

$$NOF = \frac{RMSE}{\bar{X}} \quad (8)$$

RESULTS AND DISCUSSION

Acclimatization

The initial stage of VFCW construction was carried out on June 3, 2024, followed by planting vegetation after the VFCW pond was ready. Furthermore, they enter the acclimatization phase where the plants are allowed to adapt to the new environment and the wastewater to be processed so that biofilms are formed (Vera-Puerto et al., 2021; Wibowo et al., 2023). Acclimatization was considered as sufficient when the plant growth was visible, characterized by new leaves with a height between 5 and 10 cm starting to appear on the seventh day.

Figure 2. *Typa angusifolia* leaves growth

Data Analysis

There were variations in the analysis data for both influent and effluent samples at the three different HRTs. Influent and effluent data were sorted from the smallest value and then processed using Equation 2 to express the removal efficiency. To accurately determine the performance of VFCW, each set of data distribution needs to be selected using Z-Score (Equation 3) to eliminate outlier data (Z-Score > ± 1.96) that do not represent the actual characteristics and removal efficiencies. Table 2 shows the average analysis results and removal efficiencies in data that passed the Z-Score selection.

Table 2. Average of Selected Analysis Results and Selected Removal Efficiencies

HRT	Influent (mg/L)	Effluent	Removal Efficiency (%)
BOD			
3 days	36,4	17,3	52,9
4 days	23,2	11,4	54,4
5 days	26,0	12,4	53,6
COD			
3 days	82,5	52,0	35,7
4 days	66,1	34,6	49,1
5 days	68,8	33,6	47,2

BOD Removal

Biological Oxygen Demand (BOD) refers to the amount of oxygen microorganisms need to decompose organic matter (Weiner, 2008). The removal of organic materials occurs rapidly at the start of the treatment process using VFCW to address the BOD levels (Minakshi et al., 2022; Stefanakis et al., 2014). The amount of oxygen that helps the degradation of organic materials is provided by VFCW through the feed contact mechanism with free air just before entering the pond. Additionally, when wastewater is fed intermittently, it gradually descends by gravity through the CW substrate, allowing BOD loading to be proportional to the availability of oxygen entering the feed (Stefanakis et al., 2014).

The VFCW treatment operation in removing BOD with HRT 3, 4, and 5 days resulted in an average removal efficiency of 52,9%, 54,4%, and 53,6%, respectively. The data was tested using ANOVA as in Table 3, the *p*-value is 0.745, greater than alpha (*p*> 0,05), and F 0,30 is smaller than F crit 3,47. This concludes that the treatment operation with HRT 3, 4, and 5 days using VFCW in this study did not differ in BOD removal performance.

The removal efficiency for all HRTs was approximately at around 50%. This aligns with a study by Mburu et al. (2019), which simulated a VFCW to treat wastewater from animal slaughterhouses. Mburu et al. (2019) concluded that increasing HRT from 3 to 5 days did not significantly change the removal efficiency. This observation contradicts the general expectation that HRT has a linear relationship with removal efficiency, as noted by Hassan et al. (2021) and Kataki et al. (2021), where a longer HRT extended wastewater contact and the constructed wetland components, lead to a higher treatment efficiency.

Table 3. ANOVA Study of BOD Removal Efficiencies

Sources of Variation	F	*p*-value	F crit
Between Groups	0,29	0,745	3,47

The removal efficiency with a value of 50% was smaller than the CW research data that treated previous oil refinery wastewater. Hasan et al reported in their review journal that refinery wastewater treatment using VFCW could remove BOD above 90% (Hassan et al., 2021). BOD removal primarily occurs in the initial stages of the treatment process, where aerobic microorganisms decompose organic matter by consuming oxygen (Stefanakis et al., 2014). In this study, the VFCW treatment was conducted during the early stages of plant growth, meaning that the roots had not yet adequately developed to create pores in the soil, which would allow more oxygen to reach the bottom (Rasool et al., 2023). Well-established plants enhance oxygen diffusion from photosynthesis into the surrounding environment, including the root zone. This ensures that aerobic bacteria at the bottom of the pond have enough oxygen to degrade pollutants effectively (Kataki et al., 2021). The amount of oxygen in artificial wetlands determines the success of biological degradation by aerobic microorganisms (Aslam et al., 2007). Providing adequate oxygen can increase BOD removal in wetlands by up to 20% (Varma et al., 2021).

Organic matter in CW can be physically removed through filtration and sedimentation mechanisms on the substrate or from plant roots, but biological removal by microbes is more dominant (Mahmoudi et al., 2024). Aerobic microbes can rapidly degrade organic matter, as evidenced by the BOD value at the start of the treatment process, while anaerobic degradation occurs at a slower pace (Stefanakis et al., 2014). Microbes can work individually or form colonies (biofilms) that adhere to the substrate and plants (Kataki et al., 2021). The presence of plant roots supports the development of microbes forming biofilms, which ultimately affect the efficiency of CW (Agarry et al., 2018). The Acclimatization gives plants time to adapt to the environment (Vera-Puerto et al., 2021; Wibowo et al., 2023) and related to the time required for biofilm formation requires 3 to 4 weeks (Rasool et al., 2023; Vera-Puerto et al., 2021).

This study's acclimatization period ended on the 7th day when *Typa Angustifolia* produced new leaves. Inadequate growth might have limited its ability to promote sufficient oxygen and microbial growth necessary for the breakdown of organic matter. The BOD removal efficiency in VFCW for refinery wastewater by Agarry et al in a 2020 study was slightly higher, up to 60% for 4 days HRT when an acclimatization period was set up to 28 days (Agarry et al., 2018). In another study, the maturity of VFCW, which is visually indicated by the formation of dense roots at the end of the processing operation, given an adaptation period of 3 months, was able to remove up to 76% BOD with a 2-day HRT (Mustapha, Gupta, et al., 2018). Although VFCW has yet to reach its full potential because of the minimum acclimatization period, a 3-day HRT is proven capable remove BOD to meet Indonesian regulatory quality standards.

COD Removal

The COD removal efficiency for HRT 3, 4, and 5 days in this research was 36.7%, 49.1%, and 47.2%, respectively. The removal efficiency data set was studied using ANOVA display in Table 4. The results of the ANOVA study concluded that HRT 3 days, 4 days, and 5 days had significantly different removal rates. Furthermore, it is concluded that HRT 4 days had better removal efficiency than 3 days. While in 4 days compared to 5 days, the conclusion was not significantly different. Hence, if the processing aims to reduce the COD level, the optimal condition is at HRT 4 days, where the time is not too long, but has the same performance as 5-day HRT.

Table 4. The ANOVA Analysis for COD %RE

	3, 4, 5 days	3 and 4 days	4 and 5 days
alpha	0,05	0,05	0,05
p-value	$6.1x10^{-5}$	$6.1x10^{-6}$	0,54
F	15,58	45,35	0,40
F crit	3,44	4,54	4,54
Conclusion	*p*-value < alpha F > F crit SD	*p*-value < alpha F > F crit SD	*p*-value > alpha F < F crit NSD

SD = Significantly different
NSD = Not significantly different

The COD removal increased with the increase in HRT from 3 to 4 days, which shows different behavior with the BOD removal. This may be the results on rapid decomposition of the organic matter at the beginning is biodegradable organic leaving other materials that are more difficult to decompose and require longer contact times (Stefanakis et al., 2014). Traces of organic materials contained in refinery wastewater include GRO (gasoline range organics), DRO (diesel range organics), and BTEX (benzene, toluene, ethylbenzene, xylenes) (Skrzypiecbcef & Gajewskaad, 2017). These organic materials can be aliphatic, aromatic, and halogenated organic compounds contributing to COD levels (Keramati & Ayati, 2019). As an illustration, a small amount of DRO in refinery wastewater may be present as straight-chain n-alkanes that are easily biodegradable. However, some iso-chain hydrocarbons or cycloalkanes degrade more slowly (Weiner, 2008).

Compared with several previous studies, as presented in Table 5, the COD removal in this study was slightly lower and no significant difference was observed when the HRT was increased from 4 to 5 days. This condition is possible because most of the organic matter decomposed rapidly at the beginning through aerobic mechanisms (Mustapha, 2018), while the anaerobic process develops gradually and tends to be slower (Stefanakis et al., 2014). This is related to the discussion of BOD removal where VFCW that is not mature enough cannot provide aerobic decomposition. Treatment operations carried out at the beginning of the development of the VFCW environment tend to rely on atmospheric oxygen to degrade organic matter, generally obtained from the atmosphere. Oxygen from the air comes into direct contact with wastewater fed intermittently. Oxygen can reach the inside of the VFCW with limitations because the porosity of the substrate is not yet supported by the development of plant roots that reach depths far below the surface. Additional oxygen from plant growth is also not obtained optimally. Dominant anaerobic conditions at the wetland bottom may limit organic matter removal despite increased HRT (Skrzypiecbcef & Gajewskaad, 2017).

Table 5. Comparison of removal efficiencies for petroleum refinery wastewater treatment using VFCW

Reference	HRT (d)	Removal Efficiency COD (%)
(Mustapha et al., 2015)	2	63 - 65
(Mustapha, van Bruggen, et al., 2018)	2	72,7
(Agarry et al., 2018)	2	41,3
	4	51,3
	6	66,3
(Bafrani et al., 2024)	2	60
	4	78
	6	82
This Research	3	35,7
	4	49,1
	5	47,2

Kinetics Model

Referring to equations (4), (5), and (6), the influent and effluent concentration data were used to calculate the reaction rate constants k_1, k_2, and k_3. In the Monod CSTR combination modeling, equation (6) requires another constant, namely the Monod Half Saturation (C_h) constant which takes data from other studies. The C_h value for BOD is 20 mg/L, and COD is 60 mg/L (Nguyen et al., 2018). Furthermore, the resulting constant was used to predict effluent concentration. The comparison of the results of the actual effluent/output concentration compared to the expected concentration is presented in Figures 3 and 4.

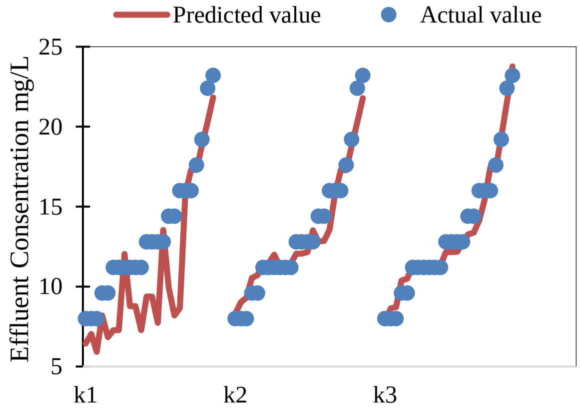

Figure 3. Predicted Value Vs Actual Value of BOD

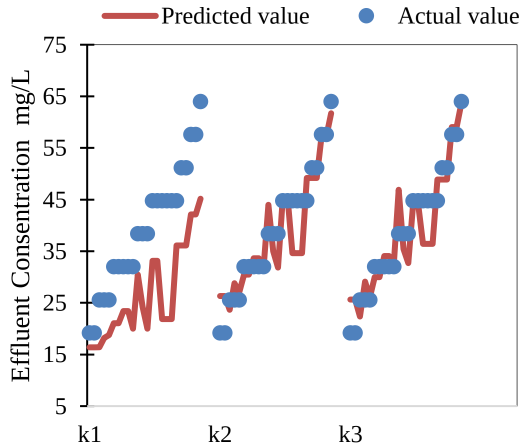

Figure 4. Predicted Value Vs Actual Value of COD

Table 6. The Statistically Tested Accuracy

	Constant	NOF
BOD		
k_1 (m/d)	0,04152	0,76680
k_2 (m/d)	0,06285	0,08438
k_3 (g/m^{2}L)	2,10261	0,04127
COD		
k_1 (m/d)	0,03182	**5,04561**
k_2 (m/d)	0,04515	0,61751
k_3 (g/m^{2}L)	4,35852	0,53120

Figures 3 and 4 show that the actual output concentration compared to the predicted concentration in some conditions has a significant difference. The statistically tested accuracy is summarized in Table 6, showing that the first-order CSTR and Monod CSTR modeling have a better similarity value compared to the other models because the NOF value is 0,0 – 1,0.

CONCLUSION

This study focused on wastewater management at PPSDM MIGAS, where petroleum processing organic contaminants containing produced wastewater. The VFCW system was applied with varying HRTs for 3, 4, and 5 days, to meet environmental regulations and minimize pollutant exposure. The system achieved organic removal as BOD and COD with maximum removal efficiencies of 54.4% and 49.1%. Statistical analysis using ANOVA showed no significant difference ($p > 0.05$) in BOD removal performance across the three HRTs, and COD removal across 4 and 5 HRTs, indicating that extending the HRT beyond 4 days did not proportionally increase treatment efficiency. The observed lower removal efficiencies compared to previous studies could be attributed to the short acclimatization period of 7 days given to the VFCW to establish an optimal ecosystem. This limited acclimatization may not have provided sufficient time for *Typa Angustifolia* to develop optimally, thus promoting oxygen diffusion to the pond bottom and providing space for microbial biofilm formation, all of which being essential for effective organic matter degradation. Further improvement in the performance of VFCWs can be achieved by providing a longer acclimatization period and ensuring optimal plant and microbial development. These findings demonstrated the relationship between HRT and pollution removal effectiveness as VFCW can reduce BOD and COD levels. These results are consistent with the potential of constructed wetlands to reduce wastewater quality costs sustainably and reasonably in tropical environments such as Indonesia. Furthermore, the simplicity, low operating costs, and environmental friendliness of VFCW underline its feasibility as a scalable option for like-minded businesses. Future studies can investigate the long-term performance of the system for more general use and further optimize the operational parameters.

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