# Comparative study about the performance of three types of modified natural treatment systems for rice noodle wastewater

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#### **Abstract**

In this study, three semi-pilot scale systems (vertical flow constructed wetland, multi-soil layering, and integrated hybrid systems) for treating real rice noodle wastewater were operated parallelly for the first time in a tropical climate at a loading rate of 50 L/(m²·d) for more than 7 months to determine the optimal conditions and to compare their treatment performance. The results demonstrated that these systems were appropriate for the removal of organics, suspended solids, and total coliform (Tcol). The highest reductions in chemical oxygen demand (COD<sub>Cr</sub>,73.2%), phosphorus (PO<sub>4</sub>-P, 54%), and Tcol (4.78 log MPN/100 mL inactivation) were obtained by the integrated hybrid system, while the highest removal efficiencies of ammonium (NH<sub>4</sub>-N, 60.64%) and suspended solids (80.49%) were achieved in the vertical-flow-constructed wetland and multi-soil layering systems respectively.

**Keywords:** constructed wetland; multi-soil layers; rice noodle wastewater; decentralized wastewater treatment system.

#### 1. Introduction

Untreated or incompletely treated wastewaters from the rice noodle villages in central Vietnam have been of great concern because they pose a serious impact on community health and ecological functions (MONRE, 2008). The production process of rice noodles releases a large volume of wastewater containing high concentrations of complex organic and nutrient compounds and high C/N ratios (Karmee, 2018; Siripattanakul-Ratpukdi, 2012; Suwan et al., 2014). The biggest challenge regarding a handicraft village wastewater is not only the technological solutions but also the costs of the treatment. Despite the significant advances in wastewater treatment technology, low-cost natural wastewater treatment methods have attracted more attention in recent years, especially in low and middle-income countries. These technologies are considered feasible tools for addressing the wastewater contamination in traditional handicraft villages, where centralized wastewater treatment plants might be impractical due to the high cost of construction, maintenance, and operation (Tang et al., 2019). Among the natural- wastewater treatment systems (WWTS), two types have generated considerable research interest: constructed wetland (CW) (Corbella et al., 2017; Luo et al., 2014; Marzo et al., 2018; Sgroi et al., 2018) and multi-soil layering (MSL) systems (Latrach et al., 2018; Shen et al., 2018; Song et al., 2018). Technically, both are designed based on mimicking natural processes, including filtration, adsorption, plant uptake, volatilization, and microbial degradation precipitation, in a controlled manner to neutralize pollutants in wastewater and enhance microbial activity in packed or filled beds (Jia et al., 2018; Luanmanee et al., 2001; Wei & Wu, 2018). The components and filter materials (sand, gravel, charcoal, zeolite, rice straw, soil, biochar, metal, and plants) constituting these systems are usually locally available and inexpensive (Ho and Wang, 2015). In addition, they are easily constructed and require simple operation and maintenance (Ho and Wang, 2015; Li et al., 2017).

Many studies have investigated CW (Uggetti et al., 2016; Valipour and Ahn, 2015; Vo et al., 2018; Vymazal, 2005; Wu et al., 2015) and MSL (Chen et al., 2009; Guan et al., 2015; Guan et al., 2018; Masunaga et al., 2007) for the removal of suspended solids, organic matter, nutrients, pathogens, and metals from various kinds of common wastewater (sewage, domestic wastewater, grey-water, river water, etc.) and have achieved a satisfactory removal performance (Bonanno et al., 2018; Licciardello et al., 2018). Recently, a study of Koottatepand co-workers (2018) integrated CW with MSL for treating septic tank effluent and called it novel multi-soil layer constructed wetland. However, there are currently very few or no reports comparing the effectiveness of these technologies in a similar context. In particular, no information is available on the evaluation and comparison of these technologies for rice noodle wastewater treatment (RNWWT) to date. Therefore, the comparison of data between studies with different experimental and operational conditions is often unsuitable and may lead to incorrect conclusions. For example, several studies concluded that MSL has more advantages and higher removal efficiency of pollutants than the other systems in terms of percentage, mass loading rate, and hydraulic loading rate (HLR) (Guan et al., 2012; Wakatsuki et al., 1993). However the application of MSL in practice is a less utilized method (Guan et al., 2012). Therefore, it is necessary to investigate these gaps about WWTS.

In this scenario, three semi-pilot scale systems, namely a vertical flow constructed wetland (VF-CW), MSL system, and integrated hybrid (IH) system, were parallel operated for a long period of time to compare their treatment efficiency for treating the wastewater from the rice noodle village under the same conditions of configuration in terms of size (height and volume), plants, and operational parameters. This experiment is also the first attempt to use of VF-CW, MSL, and IH systems for RNWWT. Furthermore, a parallel comparison at the same condition contributes to limit the bias in assessing WWTS which previous researches have done separately.

The specific objectives of this study were to determine optimal operating parameters and to evaluate and compare the performance of the VF-CW, MSL, and IH systems. Therefore, the results obtained could be used to plan appropriate technologies for treating rice noodle wastewater, which could allow greater versatility in choice of technology, design, installation, and operation to treat wastewater.

#### 2. Materials and methods

#### 2.1. Raw wastewater

The rice noodle wastewater used to feed the experimental systems in this study was randomly collected from a storage tank of a household producer in Cam Thach traditional noodle handicraft village, Cam Lo District, Quang Tri Province, north-center Vietnam. The wastewater obtained from the storage tank was pre-treated in a septic tank biogas basin. The characteristics of the rice noodle wastewater used in this study and effluent discharge requirements are depicted in Table 1.

Table 1. Characteristics of raw wastewater used in this study (n = 36).

Parameter	Units	Ri	ice n	oodle wa	- Vietnam's standard*			
		Mean	±	SD	Min	-	Max	vicinam s standard.
$COD_{Cr}$	mg/l	338.61	±	114.83	197.50	-	766.25	150
NH <sub>4</sub> -N	mg/l	72.10	$\pm$	30.95	24.55	-	135.35	10
NO <sub>3</sub> -N	mg/l	0.44	±	0.57	0.11	-	2.54	50
PO <sub>4</sub> -P	mg/l	16.70	$\pm$	3.25	8.56	-	24.20	10
TSS	mg/l	87.27	$\pm$	21.41	37.60	-	132.00	100
Tcol	MPN/100 ml	78.547	±	30.373	34.300	-	12.940	5.000
рН	-	7.29	±	0.11	7.02	-	7.45	5.5-9

SD: Standard deviation

<sup>\*</sup>Vietnam's standard includes the National Technical Regulation on Domestic Wastewater (QCVN 14:2008BTNMT) and Industrial Wastewater (QCVN 40:2011/BTNMT).

### 2.2. Experimental setup and description

Three parallel experimental systems were set up in similar rectangular steel tanks that were protected by double layer epoxy coating (Supplementary material). All three tanks were the same size with dimensions of 0.6 m in length (L), 0.4 m in width (W), and 0.6 m in height (H). Then, the systems were constructed and filled with bed media with different levels, sizes, and densities and marked as the VF-CW system, MSL system, and IH system respectively. The VF-CW system consisted of three layers of 10 cm of coarse gravel (3–5 cm in diameter) at the bottom, 12 cm of fine gravel (2–3 cm in diameter) in the middle, and 20 cm of coarse sand (1–2 cm in diameter) at the top. The MSL system was composed of soil mixture layers (SMLs < 2 mm diameter) and a permeable gravel layer (3–5 cm in diameter) with a height of 50 cm. The SMLs were composed of mixed local garden soil (70%), zeolite powder (12%), coconut activated charcoal (12%), and rice straw (6%) based on a dry weight basis. The obtained mixture was packed into rectangular burlap/canvas bags (40 cm L × 15 cm W × 5 cm H). Ten bags of SMLs were arranged into four layers in the form of a brick-like wall pattern, and alternating bags were filled with permeable river gravel layers (1–2 cm in diameter).

An IH system is a combination of MSL into a VF-CW system, and its structure was designed and installed comprising a 5-cm coarse gravel layer (3–5 cm in diameter) at the bottom, a 30-cm permeable gravel layer (2–3 cm in diameter) with two lines of SMLs in between, and 15-cm coarse sand layer (1–2 cm in diameter) at the top. The average porosity of the filter in the VF-CW system, MSL system, and IH system was 41%, 36%, and 37.5% respectively. *Colocasia esculenta* was planted in two rows on each filled bed of the VF-CW system and IH system, in which the plants were spaced in rows of 10 cm. A perforated drainage pipe with an internal diameter of 25 mm with the holes pointing down was installed at the bottom of the three tanks and covered by a coarse gravel bed of diameter 3–5 mm to easily collect and convey the treated wastewater from the system to discharge. In addition, all three tanks were equipped with a vertical tube to control the water level in the tank.

### 2.3. Operational procedures and conditions

The experimental procedures were the same in the three experimental systems. These systems were placed at room temperate (from 18 to 33 °C) and run in parallel for more than 7 months, including a 3-month start-up operation to establish proper growth in each tank, after which they were operated at design capacity for 4 months.

The start-up operation period of the systems was divided in two phases. In the first phase, the systems were operated at a constant HLR of approximately 66.67 L/(m²·d) for 5 weeks. Corresponding to operational weeks 1, 2, 3, 4, and 5, the systems were fed with rice noodle wastewater diluted with tap water at the rates of 100%, 75%, 50%, 25%, and 0% respectively. The observed results (e.g., plant growth parameters, the color of bubbles that appeared on the top surface of each tank, etc.) during the first 5 weeks indicated that the systems could achieve a stable operating level during week 4, which corresponded to 75% diluted rice noodle wastewater. The second phase prolonged about 6 weeks, which was from 6<sup>th</sup> week to 11<sup>th</sup> week and the systems were fed with rice noodle wastewater diluted to 25% with tap water at a HLR of 50 L/(m²·d). The dead and weakened plants in the VF-CW and IH systems were also replanted.

After the start-up period, the rice noodle wastewater was pumped intermittently into the three systems twice a day at 7:00 and 17:00 at an average inflow of 12 L/d (giving a HLR of 50 L/(m²·d)) during the 4 months of the experiment (August 14 to September 12), and the wastewater was waterlogged at 3 cm above the filter line. The influent wastewater was evenly distributed by horizontally perforated pipes that were installed on the top of each treatment tank. The hydraulic retention times (HRTs) of the VFCW, MSL, and IH systems were 3.3, 3.6, and 3.4 days, respectively. The treated wastewater from the three systems was manually drained prior to re-feeding wastewater to the systems.

#### 2.4. Removal mechanisms

The removal mechanisms and biochemical reactions are results of the interaction between multiple components in WWTS such as bacteria, filtration materials, plant, characteristics of influents, environmental parameters, operating conditions (hydraulic loading rate, influent fed regime, recirculation, organic carbon addition, etc.), etc. Each pollutant parameter may be removed by different mechanisms. For example, organic compounds of wastewater can be degraded aerobically and anaerobically by bacteria attached to plant underground organs (roots and rhizomes) and media area (Kadlec and Knight, 1996; Vymazal, 2005). However, nitrogen removal is quite complicated, including several pathways such as biological (e.g. ammonification, nitrification, denitrification, biomass assimilation, plant uptake), and physicochemical routes (i.e. ammonia volatilization, and adsorption) and others (i.e. anammox and canon processes) (Saeed and Sun, 2012). Whereas, the removal of fecal indicators happens by physical (e.g. filtration, sedimentation and sorption), chemical (e.g. oxidation), and biological factors (e.g. antimicrobial activity, predation and activity of lytic bacteria or viruses), in combination or alone. WWTS usually show both aerobic and anoxic conditions which depends on the depth and type of wetland (Nguyen et al., 2018). The abundant oxidant, electron donors (mainly organic matter and ammonia), are oxidized and this reduction of O2 to H2O is carried out by true aerobic microorganisms, and CO2 is evolved as a waste product. After oxygen is depleted, organisms capable of reducing  $NO_3^-$ ,  $MnO_2$  and  $SO_4^{2-}$  develop and other reactions might occur in response to the decrease in oxygen and the redox potential (Nguyen et al., 2018, Kadlec and Knight, 1996).

#### 2.5. Sampling and analyses

Total 36 wastewater samples were collected every 3 days or weekly from the effluent of the treatment tanks and influent of the storage tank. They were analyzed immediately after sampling without filtering

or pretreatment. The water quality parameters BOD<sub>5</sub> (5220D), COD<sub>Cr</sub> (5210B), NO<sub>3</sub>-N (4500 NO<sub>3</sub>-B), NH<sub>4</sub>-N (4500-NH<sub>3</sub> F), PO<sub>4</sub>-P (4500P-D), TSS (2540D), and Tcol (9221 B), were determined according to the standard methods (APHA/WEF/AWWA, 2005). Devices used to analyze the wastewater quality included water quality meter (Model: HQ40d, Hach Co., USA), spectrophotometer (Model: Carry 60 – Agilent, USA), incubator (Model: TC 135S - Aqualytic, Germany), water bath (Model: WNB 22 – Memmert, Germany), oven (Model: Won- 50: Daihan Scientific, Korean), colony counter (Model: ColonyStar - Funke Gerber, Germany), and Medical Clean Bench (SW-CJ-1EP, Airtech, China).

### 2.6. Statistical analysis

All statistical analyses were carried out using the R statistical environment (R Statistical Software Version 3.2.2). An analysis of variance (ANOVA) was used for the relationships between the treatment tanks and removal efficiencies, and a post-hoc test (Tukey's Honest Significant Difference (HSD)) was used for comparing the multiple means at a 95% confidence level. Post-hoc tests determined which levels were causing these differences.

## 3. Results and discussion

### 3.1. Wastewater characterization

In general, the concentrations of contaminants in rice noodle wastewater were high, unstable, and varied such as the COD<sub>Cr</sub>, BOD, and total Kjeldahl nitrogen concentrations, which varied from 4,200 to 29,000 mg/L, 5,400 to 23,200 mg/L, and 68.70 to 198.00 mg/L respectively (Jijai and Siripatana, 2017; Nanta et al., 2018; Siripattanakul-Ratpukdi, 2012). The key characteristics of the raw rice noodle wastewater are summarized in Table 1. According to a survey conducted by the Vietnam Ministry of Natural Resources and Environment on wastewater from traditional noodle handicraft villages, the average concentrations of COD<sub>Cr</sub>, BOD<sub>5</sub>, and Tcol for Cam Thach village were 3,550 mg/L, 500 mg/L

and 11.000 MPN/100mL respectively (MONRE, 2008), in which,  $COD_{Cr}$  and  $BOD_5$  were much higher compared to those of the wastewater used in this study (Table 1). This suggests that the septic tank contributed significantly to reduce the organic concentrations in the wastewater effluent from rice noodle manufacturing (338.6  $\pm$  114.8 mg/L  $COD_{Cr}$ ). However, nutrient concentrations remained relatively high in the effluent (72.1  $\pm$  24.5 mg/L  $NH_4$ -N and 16.7  $\pm$  8.6 mg/L  $PO_4$ -P), which must be further treated before being reused or discharged legally into the environment.

### 3.2. System performance

### 3.2.1. Organic matter removal

The overall results obtained from the three WWTS are shown in Table 2. To compare the treatment efficiency of the units, an ANOVA and Tukey's HSD analysis were applied to identify the statistical differences between the units. The ANOVA analysis indicated that there was a statistically significant difference between the three systems (p < 0.05, F = 4.13), whereas Tukey's HSD analysis indicated only a significant difference between the IH system and MSL system in effluent concentrations of  $COD_{Cr}(p < 0.05)$  (Table 2). It suggests that other similar conditions such as plants and media materials in the MSL and IH systems may influence the efficiency of  $COD_{Cr}$  removal. An indication of the difference was that the  $COD_{Cr}$  effluent concentration of the IH system (84.02  $\pm$  32.68 mg/L) was much lower than that of the MSL system (107.00  $\pm$  39.27 mg/L). In addition, the substantial mass removal for  $COD_{Cr}$  were noticed through the experimental systems with a mean of 11.58 to 12.73 g/(m²·d) (Table 2). These mass removal rates were lower than the results of the MSL system (Attanandana et al., 2000; Latrach et al., 2016) and higher than those of the VF-CW system (Wu et al., 2013). This difference may be explained because the height in MSL is usually larger and use better filter layers, while the plants in VF-CW are quite sensitive to high hydraulic rate and pollution load.

Table 2. The mean COD<sub>Cr</sub> effluent concentration, removal rate, and statistical analysis.

System Effluent		Removal rate $(g/(m^2 \cdot d))$	p value for Tukey's method		
MSL system	$107.00 \pm 39.27$	$11.58 \pm 5.36$	MSL vs. VF-CW	0.717	
VF-CW system	$100.58 \pm 34.14$	$11.90 \pm 5.62$	MSL vs. IH	0.017	
IH system	$84.02 \pm 32.68$	$12.73 \pm 5.27$	VF-CW vs. IH	0.115	
$MSL^a$	57	21.9			
$MSL^b$	140	42.9			
VF-CW <sup>c</sup>	7.2 - 75.5	1.51 - 56.07			
VF-CW d	19.7	10.9	2		

<sup>&</sup>lt;sup>a</sup> MSL for domestic water treatment (Latrach et al., 2016).

The variation in COD<sub>Cr</sub> influent and effluent concentrations and the COD<sub>Cr</sub> removal efficiency of each system during operation of experiments for 113 days are illustrated in Figure 2. The experimental results showed that there was significant variation in influent COD<sub>Cr</sub> concentrations (high fluctuations in the COD<sub>Cr</sub> loading rate in the influent) throughout the experiment. However, the effluent COD<sub>Cr</sub> concentrations decreased substantially through all the systems with an average reduction of more than 66%, which corresponded to COD<sub>Cr</sub> concentrations in the final effluent of less than 107 mg/L, which were much lower than that of the standard for waterbody discharges in Vietnam (150 mg/L, Fig. 1a), Italia (DLgs 152/2006) (160 mg/L) (Ghimpusan et al., 2017), Malaysia (200 mg/L) (NRE, 2009) and European Community (125 mg/L) (EU, 1991). The COD<sub>Cr</sub> removal percentage of the IH system (73.23%) was higher and more stable than that of the MSL system (66%) and VF-CW system (67.42%) (Fig. 1c), which corresponded to their average COD<sub>Cr</sub> concentrations remaining in the effluent of 84.02 mg/L, 107.00 mg/L, and 100.58 mg/L respectively. Conversely, previous studies conducted by Lu and co-workers (2015) and Luanmanee and co-workers (2001) concluded that the COD<sub>Cr</sub> reduction efficiency of the MSL system was higher than that of conventional filters or natural soil systems, such as sand filters and CWs.

<sup>&</sup>lt;sup>b</sup> MSL for food service wastewater treatment (Attanandana et al., 2000).

<sup>&</sup>lt;sup>c</sup> VF-CW for domestic and nitrified wastewater treatment (Chang et al., 2015).

<sup>&</sup>lt;sup>d</sup> VF-CW for domestic wastewater treatment (Wu et al., 2013),

Some outliers suggested that the removal efficiencies of the VF-CW and MSL systems were not stable (Fig. 1c). The higher  $COD_{Cr}$  treatment efficiency achieved by the IH system was attributed to the different media layer configurations with high absorbability and porosity, which adsorbed organic pollutants in wastewater onto the media and provided good environmental conditions that allowed easy decomposition by organisms and plant uptake (Ávila et al., 2015; Marzo et al., 2018; Nguyen et al., 2018).

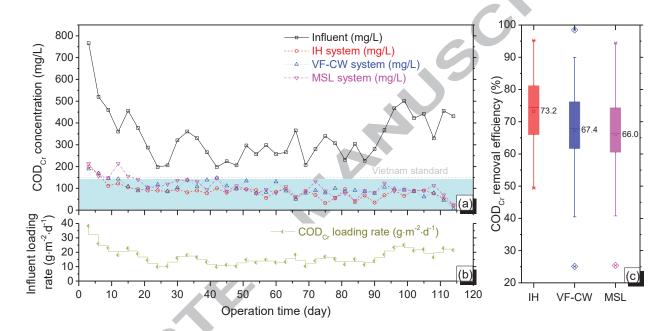


Figure 1. Temporal variations in  $COD_{Cr}$  concentrations (a), influent  $COD_{Cr}$  loading rate (b), and overall  $COD_{Cr}$  removal efficiency (c) in each system during operation.

### 3.2.2. Nutrients removal

The results obtained (Fig. 2a) indicate that although the concentration of NH<sub>4</sub>-N in the final effluent of the IH system remained at approximately  $35.93 \pm 20.01$  mg/L, which was slightly higher than that in the effluent of the MSL and VF-CW systems ( $31.59 \pm 16.56$  mg/L and  $30.95 \pm 24.94$  mg/L, respectively). In general, these values did not differ significantly based on post-hoc analysis and ANOVA (p > 0.05, F = 0.51). Similarly, this trend occurred with NH<sub>4</sub>-N removal rates in these

systems. This meant that the extent and process of removing NH<sub>4</sub>-N in the three systems were relatively similar. On the contrary, the effluent concentration of NO<sub>3</sub>-N increased slightly through all three tanks from  $0.40 \pm 0.57$  mg/L to  $1.50 \pm 1.51$  mg/L for MSL,  $1.33 \pm 1.03$  mg/L for VF-CW, and  $1.66 \pm 1.63$  mg/L for IH (Fig. 2b). It was expected because parallel processes occurred in the WWTS, including ammonium removal and nitrification, which were enhanced by intermittent water flow into the units through the perforated pipes. Nitrogen was reduced in these systems by the two main processes of nitrification and denitrification due to the simultaneous existence of aerobic and anaerobic conditions with various microbial abundances, which facilitated the conversion of NH<sub>4</sub>-N to NO<sub>3</sub>-N and nitrogen gas and the adsorption of NH<sub>4</sub>-N. This occurred mainly in SMLs (including zeolite, soil, and charcoal) (Chen et al., 2009; Guan et al., 2012; Zhang et al., 2015).

In addition, Figure 3a shows that the NH<sub>4</sub>-N concentration in the effluent of the three systems was still higher compared to Vietnam's standard (<10 mg/L) and Italy's standard (<15 mg/L) (Ghimpusan et al., 2017). This could be ascribed to the high concentration of influents (average of 72.1 ± 30.95 mg/L) that was beyond the inherent capacity of these WWTS. However, throughout the operation, the average NH<sub>4</sub>-N removal efficiency of the VF-CW system was 60.64%, which was higher than that obtained from the MSL system (53.1%) and IH system (49.3%) (Fig. 2d). These results were not in line with previous reports, that highlighted the nitrification and adsorption capacities of MSL in NH<sub>4</sub>-N reduction for treatment of municipal wastewater, domestic wastewater, and unsanitary landfill leachate as shown by Chen et al. (2009), Guan et al. (2012), Latrach et al. (2016), and Attanandana et al. (2000). The higher NH<sub>4</sub>-N removal efficiency reported in these studies (Chen et al., 2009; (50-70%), Guan et al., 2012; (82.4%), Latrach et al., 2016 (83%), and Attanandana et al., 2000) (61.2%)) could possibly be due to the characteristics of the material composition in MSL systems. It offers a number of advantages, such as larger porosity, high cation exchange and adsorption capacities, better oxygen

diffusion, which could help to enhance the wastewater distribution, microbial growth rate and activity, and, as a consequence, an increase in the removal of NH<sub>4</sub>-N.

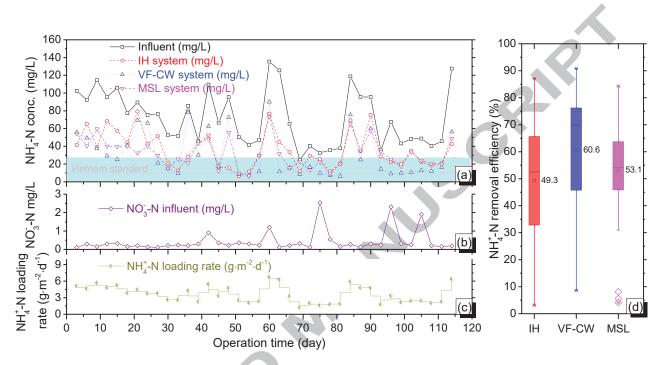


Figure 2. Temporal variation in concentrations of NH<sub>4</sub>-N in the influent and effluent. (a) Influent NO<sub>3</sub>-N concentration. (b) Influent NH<sub>4</sub>-N loading rate. (c) Overall NH<sub>4</sub>-N removal efficiency. (d) IH, VF-CW and MSL system during operation.

The PO<sub>4</sub>-P concentration in the influents and effluents and the PO<sub>4</sub>-P removal efficiency throughout the experiments are depicted in Fig. 3. These results showed that the average concentration of PO<sub>4</sub>-P remaining in the final effluent of the VF-CW system was high at  $12.35 \pm 2.01$  mg/L, which was higher than that obtained in the final effluent of the IH and MSL systems at  $7.26 \pm 1.99$  mg/L and  $8.92 \pm 1.63$  mg/L respectively (while Vietnam and Malaysia's phosphorus discharge standard is 10 mg/L). The PO<sub>4</sub>-P removal efficiencies in the VF-CW, IH, and MSL systems were 23.88%, 54.02%, and 44.73% respectively, regardless of the significant fluctuation in influent concentration in the range of 8.56 to 24.2 mg/L with a PO<sub>4</sub>-P loading rate in the range of 0.43 to 1.21 g/(m<sup>2</sup>·d). The higher removal

efficiencies found for  $PO_4$ -P in the MSL (44.7%; p < 0.0005) and IH (54%; p < 0.005,) systems may be due to the adsorption capacity in the SMLs of the MSL system, in which phosphorus could be adsorbed on the Al and Fe hydroxides in the soil (Chen et al., 2009). In contrast, the mechanism for reducing  $PO_4$ -P in the VF-CW system (23.9%, p < 0.0005) was limited may be because the gravel substrate was not capable of binding phosphorous (Arias and Brix, 2005; Brix et al., 2001). The results of the  $PO_4$ -P reduction obtained in this study were consistent with the experimental data of a VF system acquired by O'Hogain (2003) (39%) and lower than those of MSL systems studied by Latrach et al. (2016) (84%) and Masunaga et al. (2007) (56-85%). Moreover, the results of the statistical test indicated that the differences between the effluents of the three systems were statistically significant (p < 0.005, p = 70.1). It means that the SMLs in the IH and MSL systems contributed greatly to the  $po_4$ -P reduction in the experimental units.

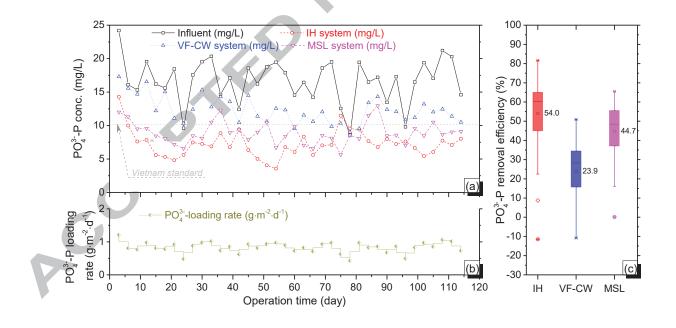


Figure 3. Temporal variations in PO<sub>4</sub>-P concentrations in influent and effluent. (a) Influent PO<sub>4</sub>-P loading rate (b) Overall PO<sub>4</sub>-P removal efficiency. (c) IH, VF-CW and MSL system during operation.

### 3.2.3. Total suspended solids and total coliform removals

During the operation of the systems, the TSS and Tcol concentrations in the influent and effluent of each system were measured (Fig. 4). The fluctuation in TSS concentrations in the influent ranged from 37.60 to 132.00 mg/L with an average value of  $87.27 \pm 21.41$  mg/L (Fig. 4a, e). Despite the large variations in TSS in the influent, the TSS removal efficiencies of the VF-CW, MSL, and IH systems were consistently stable and fairly high, and were  $75 \pm 16\%$ ,  $80 \pm 11\%$ , and  $72 \pm 15\%$  respectively. It corresponded to the average concentrations of TSS in the effluent of approximately  $20.76 \pm 9.52$  mg/L,  $15.85 \pm 7.16$  mg/L, and  $24.11 \pm 12.76$  mg/L respectively. These effluent concentrations were lower than the discharge limit of Vietnam and EU's standard for TSS of less than 150 mg/L and 60 mg/L respectively (Fig. 4a, e). In addition, post-hoc analysis and ANOVA test revealed that the TSS effluents did not differ significantly between the three systems (F = 0.55, p > 0.05).

The average Tcol inactivation efficiencies of the VF-CW, MSL, and IH systems were not high, with values of 4.57, 4.54, and 4.78 log MPN/100 mL respectively, which corresponded to the average coliform concentrations remaining in the effluent of  $2.63E+04\pm1.11E+04$  MPN/100 mL,  $3.61E+04\pm0.82E+04$  MPN/100 mL, and  $9.22E+03\pm3.48E+03$  MPN/100 mL respectively (Fig. 4c, d, f). The remaining coliform concentrations were still at high levels that exceeded the allowable discharge standard of Vietnam (5,000 MPN/100 mL). This was also reasonable because these systems only used biological methods. Thus, to achieve more effective disinfection, the further combination of other treatment techniques is encouraged, such as a tertiary treatment. Therefore, these systems are suitable for TSS reduction rather than coliform treatment, and the highest TSS removal efficiency was achieved in the MSL system.

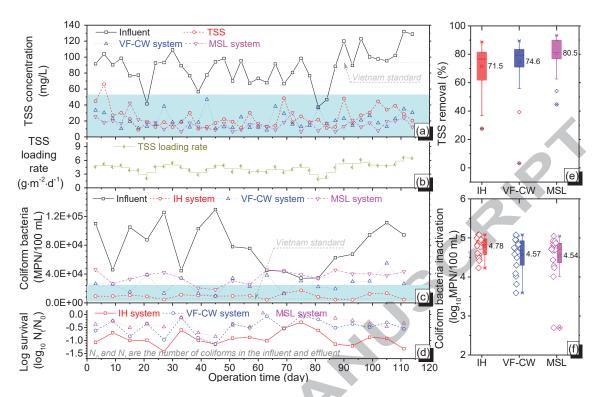


Figure 4. Temporal variations in total suspended solids (TSS) and total coliform (Tcol) concentrations in influent and effluent and their overall removal efficiency in each system during operation.

### 3.3. Correlations between parameters

Three main important parameters, namely COD<sub>Cr</sub>, NH<sub>4</sub>-N, and PO<sub>4</sub>-P, were monitored and further evaluated by analyzing the correlation between variables of the initial loading rate and removal loading rate or removal efficiency (organics and nutrients removal) during the operation of the systems, which could explain the relationship and influence between these parameters as well as predict the output of these systems.

The linear relationships between the outputs in terms of effluent quality, removal rates, and influent loading rates for  $COD_{Cr}$  and  $NH_4$ -N are presented in Fig. 5. The results indicate that there was a high correlation between removal loading rate ( $L_r$ ) and influent mass loading rate ( $L_i$ ) in the three systems, with a high coefficient of determination ( $R^2$ ) that varied from 0.88 to 0.92 (p < 0.05) (Fig. 6a). This

confirmed that the three equations in the plot could apply for predicting the output of systems or designing a new system. In addition, the correlations between effluent  $COD_{Cr}$  and  $L_i$  were relatively low (Fig. 5b), in which  $R^2$  ranged from 0.02-0.15, and were not statistically significant compared with the data obtained in the VF-CW system. It seemed that the  $COD_{Cr}$  concentration in the effluent was not influenced by  $L_i$ , and their correlation in the systems of this research did not follow first order linear equations.

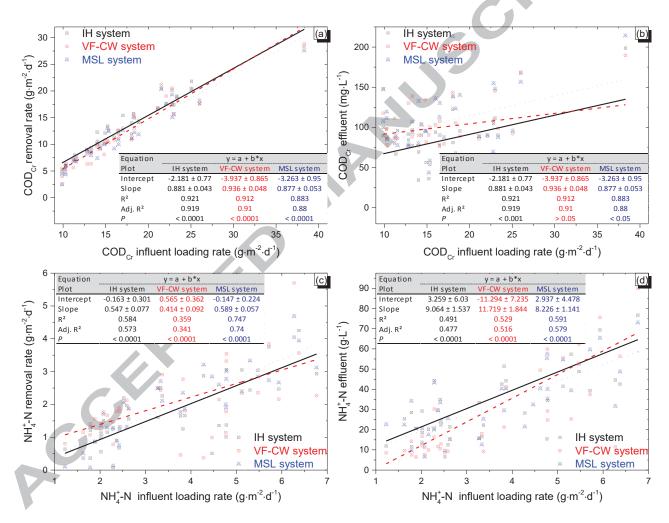


Figure 6. Correlations between  $L_i$ ,  $L_r$ , and effluent concentrations of COD<sub>Cr</sub> and NH<sub>4</sub>-N in each system.

For NH<sub>4</sub>-N, the relationship between NH<sub>4</sub>-N removal rate, NH<sub>4</sub>-N concentration in the effluent, and NH<sub>4</sub>-N influent loading rate was statistically significant (p < 0.0001) (Fig. 5c, d). Nevertheless, only the R<sup>2</sup> values above 0.5 were acceptable for the linear regression (Hijosa-Valsero et al., 2011). The correlations between NH<sub>4</sub>-N removal rate and NH<sub>4</sub>-N influent loading rate (Fig. 5c) and those between NH<sub>4</sub>-N concentration in the effluent and NH<sub>4</sub>-N influent loading rate (Fig. 5d) in the MSL system were clearer than those obtained in the IH and VF-CW systems. Weak relationships between L<sub>r</sub> and L<sub>i</sub> and between L<sub>i</sub> and the effluent NH<sub>4</sub>-N were observed. Therefore, they were not very meaningful in the analysis of the linear regression. In addition, the results of the correlation analysis for PO<sub>4</sub>-P indicated that there was no proof to reject the null hypothesis (p > 0.05) and suggested that the relationships between Li and Lr and between Li and effluent PO<sub>4</sub>-P were not statistically significant. This mean that the PO<sub>4</sub>-P concentration in the effluent might not be predictable through influent loading rates. In summary, the results showed that these technologies were suitable for the removal of organic matter and SS and could meet the regulation limits of Vietnam. However, the nutrients and Tcol concentrations were reduced modestly through all three treatment units, with removal rates of 49.3% to 60.6% for NH<sub>4</sub>-N and 23.9% to 54.0% for PO<sub>4</sub>-P. Consequently, a further study with two stages of tank might be required in the future. The IH system obtained the highest reduction with an average value of 73.2% for COD<sub>Cr</sub>, 54.0% for PO<sub>4</sub>-P, and 4.78 log MPN/100 mL inactivation for Tcol, while the highest removal efficiencies of NH<sub>4</sub>-N (60.64%) and TSS (80.49%) were achieved in the VF-CW and MSL systems respectively. Generally, the IH system is recommended for RNWWT due to higher treatment performance in comparison with the VF-CW and MSL systems. Moreover, the system with two stages of IH and a kind of WWTS such as the polishing pond, free water surface constructed wetland, which support to treat the nutrients and Tcol, might be needed for application of RNWWT to fulfill strictly the effluent regulations. Finally, this study also offers an additional choice and allow

greater versatility in choice of technology, design, installation, and operation to treat rice noddle wastewater.

#### 4. Conclusions

Three RNWWT systems using natural physical, biochemical, and microbiological processes were parallel operated in a tropical climate to compare and evaluate their treatment performance. The results showed that these technologies are suitable for the removal of organic matter and SS, and could meet the regulation limits of Vietnam. However, the nutrients and Tcol concentrations were reduced modestly through all three treatment units, further treatment might be required. Generally, the IH system is recommended for RNWWT due to higher treatment performance in comparison with the VF-CW and MSL systems.

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### **Graphical abstract**

