

Proposal of a standard experimental model to determine the contaminant removal rate constants in subsurface flow constructed wetlands

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Abstract: The objective of this study was to establish a standard experimental model to determine the removal rate constants of wastewater pollutants in subsurface flow–constructed wetlands. Such the rate constants of removal processes of removal processes of COD, BOD, NH_4^+ , total nitrogen (TN), etc., are important parameters for calculating the size of wetlands, yet the data published in the international textbooks and research articles fluctuate in very wide ranges, that make it challenging to design constructed wetland accurately. By evaluating influencing factors and referencing available models, we proposed the experimental wetland model of cylindrical shape ($\text{Ø}18\text{cm}$ and 30cm height), with the type of material and plant, which must be specified for each experiment determining the rate constants. The experimental procedure has been given, including setting up the system, running experiments to collect data, and processing data to calculate the rate constant. The initial experiments with small gravel and common reed plant, determining the removal rate constants of COD, NH_4^+ and TN, provided good repeatability results, and the values are within a reasonable range with the published values in the world. This result shows the applicability of the proposed experimental model and procedure to determine the contaminant removal rate constants in constructed wetlands uniformly. Eventually, a complete set of more converging rate constants data can be obtained, which improves the accuracy of the constructed wetlands setup.

Keywords: Subsurface flow constructed wetland; Rate constant; Ammonium; COD; Total nitrogen.

1. Introduction

Constructed wetlands are systems that follow the idea of natural wetlands to carry out wastewater treatment processes. In constructed wetlands, the pollutant transformation is accomplished through an integrated combination of biological, physical, and chemical interactions between the plants, the media, and the inherent microbial community [1]. Wetland treatment systems are generally classified into two categories: free-water surface wetlands (FWS), which are shallow basins with water on the surface, and subsurface flow wetlands (SSF), which are the bed with water flow under the surface. According to the flow direction, subsurface flow systems are further divided into horizontal and vertical subsurface flow. The construction of these systems can be obtained at a relatively low cost due to the simple materials and equipment used. The removal efficiencies are high, and the treated

effluent can meet the standard for secondary or tertiary biological wastewater treatment. In addition, the constructed wetland has low operation and maintenance costs owing to its natural energy's employment at work [2]. Constructed wetlands have become an increasingly popular option for wastewater treatment.

The treatment efficiency of the wetland can only be obtained when it is appropriately designed. Nowadays, many textbooks and researchers agree that the first-order rate constant (k) of the pollutant removal process is the primary tool in wetland design [1]. The pollutants of concern in wetland treatment are organic matter and nutrients; accordingly, the removal rate constants of COD, BOD, NH_4^+ , NO_3^- , or TN are required. In addition, constructing a wetland requires the accurate value of those rate constants.

In some textbooks, the constant k of each parameter has a definite value, depending only on the type of wetland and the temperature rather than the type of media and plant. For example, at 20°C, the BOD removal rate constant, k_{BOD} , is 0.678 d^{-1} in the design of the FWS wetland; and in the design of the SSF wetland, it is 1.1 d^{-1} [3–4]. At a temperature other than 20°C, the value of k can be determined based on the Arrhenius equation with the temperature coefficient $\theta = 1.06$ [3–4]. However, this oversimplification makes the design calculation of wetlands unreasonable and unconvincing.

On the other hand, different research papers have given a wide range of values of the removal rate constants. For example, as Magdalena Gajewska summarized from many previous publications [5], the k_{BOD} value ranges from 0.071 to 6.11 d^{-1} , making it difficult for the designer. That wide range of results is understandable since each author conducts experiments to determine k under very different conditions. Although a completed study on the influence of all factors on the value of removal rate constant in wetlands has yet to be published, a number of publications have evaluated the effect of several factors. Those factors can be listed as the type of wetland media, type of plant, design shape, type of wastewater, and operational mode.

The media used in wetlands are divided into three principal groups: natural materials (soil, sand, gravel, zeolite...), industrial by-products (slag, rubble, bark...), and artificial materials (activated carbon, synthetic materials, ceramite...) with a total of dozens of different materials [6]. Many studies have confirmed that the treatment efficiencies of the same type of pollutant in wetlands using different materials are not the same [7–9]. Abdelhakeem et al. [9] conducted a study to determine and compare the constant k of experimental wetland systems with the same size and type of vegetation with two different materials: gravel and vermiculite. As a result, the COD removal rate constant of gravel, k_{COD} , is 2.64 d^{-1} , while that of vermiculite is 2.95 d^{-1} ; the NH_4 removal rate constant, k_{NH_4} , of gravel is 0.66 d^{-1} , and that of vermiculite is 0.96 d^{-1} .

Plant species commonly used in the wetland are *Phragmites spp.* (*Poaceae*), *Typhaspp.* (*Typhaceae*), *Scirpus spp.* (*Cyperaceae*), *Iris spp.* (*Iridaceae*), *Juncus spp.* (*Juncaceae*) and *Eleocharis spp.* (*Spikerush*), of which the reed (*Phragmites australis*) is the world's most frequently used plant species [6]. Like the material, the pollutant removal rate in the SW using different plant species also varies. [10] conducted a study to determine the COD treatment rate constant of the wetland with four different plant species, *Phragmites australis*, *Lythrum salicaria*, *Cladium mariscus*, *Iris pseudacorus*, giving constant results k are 0.22, 0.37, 0.35 and $0.55 \text{ (d}^{-1}\text{)}$, respectively.

Experimental models to determine the constant rate k are primarily designed with the rectangular box shape and operated in continuous mode to resemble wetlands in reality [6, 8, 11]. However, each model is designed with a very different dimension; even the length, width and depth are not the same. For example, the experimental model in the study [10] is 2.5 m long, 0.65 m wide, i.e., the ratio length: width = 3.8:1; depth is 0.6 m. [11] used the experimental model with the size of $12 \text{ m} \times 1.6 \text{ m} \times 1.1 \text{ m}$, i.e., the ratio of length: width = 7.5:1; 1.1 m depth. In addition, many other studies have experimental wetland models of

different sizes. Currently, there is no research to evaluate the influence of the size of the empirical model's shape on the k -constant results. However, [10] studied the constant value of k_{COD} at different depths and concluded that the constant k result depends on the depth of the wetland.

The wastewater used in the studies to determine the constant k has two types: natural wastewater [9, 11–13] and synthetic wastewater [10, 14–15]. The advantage of using natural wastewater is that it has a composition of pollutants (both macro- and micro-) and a realistically rich micro-organism. However, natural wastewater has a very high fluctuating component concentration, which is difficult to control. Synthetic wastewater has the advantage of being able to actively control the concentration of components, ensuring uniform characteristics of wastewater composition. The composition of microorganisms to be treated as natural wastewater can be provided by soaking the experimental wetland system with raw sewage to create the necessary microflora.

In terms of operational modes for wetlands studies, there are two categories: continuous operation [9–11, 13, 15] and batch operation [12, 14]. Most of the studies were carried out in the continuous regime to simulate a typical wetland system in nature. However, when changing different types of material and plant (due to different porosity), if operated continuously, the pumped wastewater flow rate needs to be adjusted to ensure water retention time, leading to process complexity. With batch operation, it is possible to control the exact contact time of aqueous solution in the wetland systems.

Thus, determining the constant k based on wetland systems with different designs, materials and plants and implementation procedures will always result in a wide range of k values. To uniformly determine the constant k , it is necessary to establish an experimental model with a uniform design and procedure called the standard experimental model. On this basic model, it is possible to conduct experiments to determine the constant values of k for different pollutant parameters in a uniform manner.

To determine the removal rate constant of the treatment process, the experimental wetland is considered as a simple reactor in this study. Thus, an experimental model with a compact size while ensuring that all wetland components present is entirely representative of a wetland system. Accordingly, we propose an experimental wetland model with a cylindrical shape with minimum diameter and depth, sufficient to ensure space for plant roots to develop. Experiments were then conducted to determine the treatment rate constants of TN, NH_4^+ , and COD. The obtained constant k results were compared with published results to confirm the experimental model and procedure.

2. Methods and data

2.1. Experimental wetland setup

The experimental wetlands were set up in plastic containers with a diameter of 18 cm and a height of 30 cm with a drain valve at the bottom (Figure 1). In preparation, small gravels (5–10 mm diameter) were washed thoroughly with tap water several times and dried at ambient temperature before being used as wetland media. Young *Phragmites australis* reeds were taken from the Red riverbank (Hanoi, Vietnam), and the roots were washed to remove mud and dirt. After that, the reeds were planted so that the roots were close to the bottom of the gravel bed, with a density of two plants in each bed. Then, the plants were raised for two months to adapt to the system. Wastewater source collected at Kim Nguu River (Hanoi, Vietnam) and diluted 1:1 with tap water was used to feed the plants. The feed water in the wetland beds was discharged and refilled twice weekly to ensure that the plants could acclimatize well. In addition, as mentioned above, the application of raw wastewater also helped to inoculate natural microorganisms in the systems.



Figure 1. Experimental wetland systems.

2.2. Feeding, sampling and analysis

This study investigates the applicability of the proposed SSF wetland model for determining the rate constants of COD, NH_4^+ and TN removal processes. Therefore, different experiments treating wastewater with specific targets were conducted. A set of experiments with the binary synthetic solution containing COD and TN, COD and NH_4^+ , and COD and NO_3^- were carried out in March, April, and December, respectively. It should be noted that owing to the lack of NO_3^- analysis, only the COD removal rate constant was studied in the third test using COD and NO_3^- artificial solution. The feed solution was prepared before each experiment using analytical chemicals purchased from Merck Co., Germany. Glucose ($\text{C}_6\text{H}_{12}\text{O}_6$) was utilized to prepare the desired COD initial concentrations. Potassium nitrate KNO_3 was used for NO_3^- , ammonium sulfate $(\text{NH}_4)_2\text{SO}_4$ for NH_4^+ , and urea $\text{CH}_4\text{N}_2\text{O}$ for the TN component. KH_2PO_4 was used as the phosphorus source. Additional compounds used in the synthetic wastewater included: 28 mg/L CaCl_2 , 52 mg/L $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, 11 mg/L K_2SO_4 , 0.03 mg/L $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, 0.08 mg/L ZnCl_2 , and 1.7 mg/L $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ following [16]. Details about the concentrations of contaminants in each batch test are given in Table 1.

Table 1. Experimental conditions

Test	Month	Solution	COD concentration (mg/L)	$\text{NO}_3^- / \text{NH}_4^+ / \text{TN}$ concentration (mg/L)
1	March	COD & TN	400	20
2	April	COD & NH_4^+	400	20
3	December	COD & NO_3^-	400	20

Before the batch experiments, the experimental wetland was washed with tap water once to remove the inherent water and contaminants. Specifically, the rinsing process was carried out gently by draining the water through the bottom drain valve. When the water had drained completely, the valve was closed, and clean tap water was added slowly from the top until the water filled the reservoir. Next, the wetland was kept consistently for 10 minutes to ensure that the water entered all the pores in the wetland, then it continued to drain. Repeat washing the wetland with synthetic wastewater three times to ensure that all sites in the system are filled with the wastewater homogeneously. Afterward, the experiments were conducted by feeding the synthetic wastewater into the wetland systems. The water level inside the container was maintained at the level of 5 cm under the surface of the gravel bed to create a

subsurface flow constructed wetlands configuration. After 10 minutes for stabilization, the first sample was taken from the drain valve at the bottom. Effluent samples were taken twice daily for five days or until no further removal performance could be obtained. Water temperature was immediately measured as samples were taken. COD, NH_4^+ , and TN were analyzed by using HACH reagents, of which methods are compliant with US EPA methods. Effluent samples were measured by a DR6000 laboratory spectrophotometer (HACH Company, Colorado, United States).

2.3. Data analysis

2.3.1. Determination of the removal rate constant

To investigate the rate constant of the contaminant removal, first rearrange the equation $dC/dt = -kC$ to $dC/C = -kdt$, then perform integration; the linearized equation of this model can be obtained as below:

$$\ln[C] = -kt + b \quad (1)$$

where C is the concentration of pollutant (mg/L), k is the rate constant (h^{-1} or d^{-1}), t is the time (hour or day), and b is the constant.

During experiments, the concentration C of COD, NH_4^+ , and TN were determined as a function of time. By plotting $\ln[C]$ versus time t , the constant rate k can be obtained from the slope of the Y axis. The correlation coefficient (R^2) was used to evaluate the reliability of the results.

2.3.2. Determination of the Arrhenius equation

The effect of temperature on the rate constant k is demonstrated by the Arrhenius equation:

$$k_T = k_{20} \cdot \theta^{(T-20)} \quad (2)$$

where k_T is the rate constant at temperature T , k_{20} is the rate constant at 20°C , T is the temperature ($^\circ\text{C}$), and θ is the Arrhenius temperature coefficient.

Take the natural logarithm of both sides and bring the equation to the linear form:

$$\ln(k_T) = \ln(\theta) \cdot (T-20) + \ln(k_{20}) \quad (3)$$

When attaining data of k_T values at various temperatures T , plot $\ln(k_T)$ values according to $(T-20)$ on the graph, interpolate the linear equation, then the value of θ and k_{20} can be defined.

3. Results and discussion

3.1. TN removal rate constant

To determine the TN removal rate constant, synthetic wastewater containing COD of 400 mg/L and TN of 20 mg/L was fed to the experimental wetlands for treatment. Samples were taken twice daily at 10 am and 4 pm for four consecutive days. Effluent TN concentrations of all the samples are presented in Table 2. These data were processed to obtain the natural logarithm of concentration as a function of time, which are coordinates of the graph in Figure 2. Since the time has either unit of hour or day, two graphs were plotted to obtain the two different TN removal rate constants in terms of unit.

Table 2. Results for TN analysis.

Time	10h, 26/3	16h, 26/3	10h, 27/3	16h, 27/3	10h, 28/3	16h, 28/3	10h, 29/3	16h, 29/3
TN (mg/L)	19.1	17.7	11.5	10.7	9.7	9	9	–

The rate constant is the slope of the linear equation obtained from the graph (Figure 2). It can be seen in Figure 2a that the k_{TN} is 0.0111 h^{-1} , corresponding to 0.2664 d^{-1} ; while

the direct result in Figure 2b show k_{TN} is 0.2428 d^{-1} . The two results are only 10% different, with equal correlation coefficients.

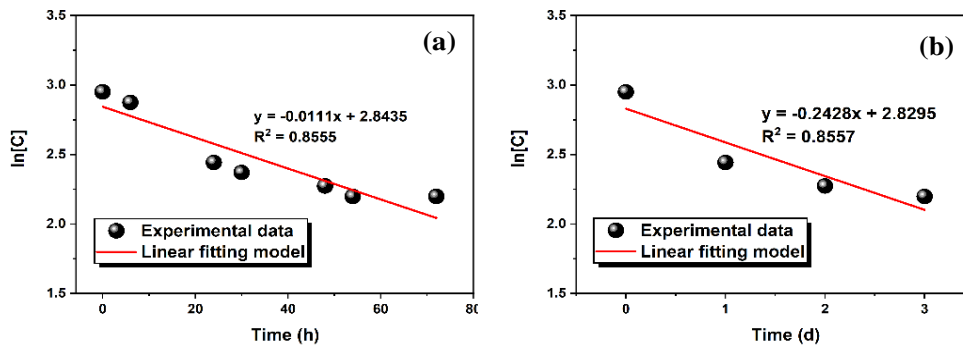


Figure 2. Plotting natural logarithms of TN concentration versus time to identify TN removal rate constant: (a) Time axis is in hour unit, k_{TN} (h^{-1}); b) Time axis is in day unit, k_{TN} (d^{-1}).

This k_{TN} value is equivalent to the result in the study [14] with $k_{TN} = 0.246 \text{ d}^{-1}$. Although the experimental model and plant species are different, the retention time is longer than 7.5 days, and the temperature range is higher ($25\text{--}30^\circ\text{C}$). [14] also conducted the study in batch mode, using synthetic wastewater.

3.2. Ammonium removal rate constant

Synthetic wastewater containing COD of 400 mg/l and NH_4^+ of 20 mg/l was fed to the experimental wetlands for treatment. Samples were taken twice daily at 10am and 4pm for four consecutive days. NH_4^+ concentrations of all the samples are presented in Table 3. These data were processed to obtain the natural logarithm of concentration as a function of time, which are coordinates of the graph $\ln[C]$ vs. time (Figure 3). Similarly, two graphs were plotted to obtain the two NH_4^+ removal rate constants in different units.

Table 3. Results for NH_4^+ analysis.

Time	10h, 8/4	16h, 8/4	10h, 9/4	16h, 9/4	10h, 10/4	16h, 10/4	10h, 11/4	16h, 11/4
NH_4^+ (mg/l)	21.8	14.7	10.9	9.4	7.8	7.7	7.4	7.2

The rate constant can be determined from the slope of the line; thus, it can be seen in Figure 3a that the k_{NH_4} is 0.0125 h^{-1} , corresponding to 0.3 d^{-1} , while the k_{NH_4} value obtained from Figure 3b is 0.3576 d^{-1} . In this case, the graph plotting $\ln[C]$ vs. day unit gives a better correlation, which is not the same with k_{TN} . Thus, the number of data points does not contribute to a better correlation between the experimental data and the model. For simplicity, the data processing method with day unit is proposed to calculate the rate constant.

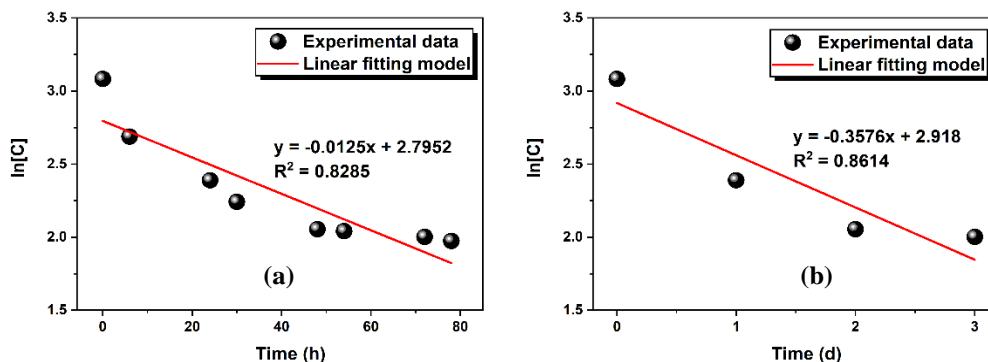


Figure 3. Plotting natural logarithms of NH_4^+ concentration vs. time to identify NH_4^+ removal rate constant: (a) Time axis is in hour unit, k_{NH_4} (h^{-1}); (b) Time axis is in day unit, k_{NH_4} (d^{-1}).

3.3. COD removal rate constant

Experiments to determine the COD removal constant rate were conducted at three different times of the year: March, April, and December. The initial concentrations of COD in the three experiments were kept similar, around 400 mg/l. COD concentrations of all the samples are presented in Table 4. These data were processed to obtain the COD removal rate constants in Table 4.

Table 4. Results for COD analysis.

Month	Temperature (°C)	COD removal over time								
		Time	10h, 26/3	16h, 26/3	10h, 27/3	16h, 27/3	10h, 28/3	16h, 28/3	10h, 29/3	16h, 29/3
March	20–23	COD (mg/L)	395	255	75	72	63	65	55	55
		Time	10h, 8/4	16h, 8/4	10h, 9/4	16h, 9/4	10h, 10/4	16h, 10/4	10h, 11/4	16h, 11/4
April	28–29	COD (mg/L)	390	121	79	34	19	17	16	14
		Time	12-Dec	13-Dec	14-Dec	15-Dec	16-Dec	17-Dec	–	–
December	15–18	COD (mg/L)	387	284	140	96	44	35	–	–
		Time								

For the reason that all of the experiments were conducted under natural conditions, and the recorded water temperatures during the experiment fluctuated. To explore the relationship between the constant k and the temperature, the average temperature value was chosen to correspond to the obtained constant k value. The temperature has a marked effect on the processing rate; as the temperature increases, the k_COD value increases (Table 5).

Table 5. k_COD values on the temperature.

Time	Average temperature	k_COD (d ⁻¹)	R ²
December	16.5°C	0.51	0.9835
March	21.5°C	0.61	0.7297
April	28.5°C	1.1	0.9145

Represent the points on the graph of the relationship between the values of ln(k_T) and (T–20) (Figure 4). The obtained linear equation showed that the COD removal rate constant at 20°C (i.e. k_COD₂₀) is 0.61 d⁻¹ and the temperature coefficient θ = 1.07.

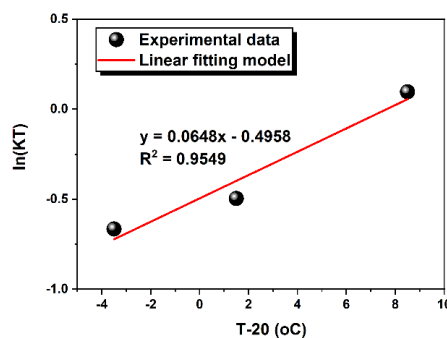


Figure 4. Plotting ln(k_T) vs. temperature difference (T–20).

This k_COD₂₀ result is higher than 0.22 d⁻¹, which is the value determined [10]. Since the two studies used the same plant as reed and gravel material of similar size, the difference was due to the experimental design. The study conducted by [9] also had reeds and gravels of the same size and depth, resulting in k_COD = 2.64 d⁻¹, which is much higher than the results of this study. This result may be explained by the fact that the experiment conducted

by Abdelhakeem in Giza, Egypt, located in a hot desert climate with extremely high temperatures of 40–45°C during the summer.

Regarding the temperature coefficient of the Arrhenius equation of COD, there are currently no published studies yet. Relative comparison with BOD removal parameters ($k_{20} = 1.1 \text{ d}^{-1}$ and $\theta = 1.06$) [4], the temperature coefficient θ of COD in this study has the same value.

4. Conclusion

This study conducted a set of experiments in batch mode using synthetic wastewater to investigate the contaminant removal rate constant in subsurface flow constructed wetlands system. According to the obtained data, it can be concluded that:

An experimental pilot was successfully set up and operated for a long period to simulate the SSF constructed wetlands in natural conditions.

The removal rate constant (k) could be calculated and presented in different units (i.e., h^{-1} or d^{-1}) with a negligible difference and equal correlation coefficients (R^2).

The total nitrogen and ammonium removal rate constant was 0.2428 and 0.3576 d^{-1} , respectively. Furthermore, the k values for COD elimination increased along with the rise of the water temperature, which was 0.51, 0.61, and 1.1 d^{-1} at 16.5°C, 21.5°C, and 28.5°C, respectively.

The Arrhenius temperature coefficient θ for COD removal in this study was 1.07.

These results demonstrate the feasibility of the proposed model to ascertain the value of the pollutant removal rate constant in the SSF CW system. From there, an accurate and complete data set of constant k can be obtained, improving the wetland design's accuracy and reliability.

Due to the lack of experimental conditions and time, this study still has some limitations, including the repeatability of the data set and the number of data points. Further studies aim to enhance the reliability by conducting the test with several reactors simultaneously, along with utilizing the actual wastewater sample as the water source instead of the synthetic one to investigate the performance of the SSF CW system in natural conditions.

Author contribution statement: Developing research ideas: T.P.P.; Data processing: D.Q.T., D.T.L., V.Q.N.; Building mathematical models: T.P.P., D.Q.T.; Writing draft articles: T.P.P., D.Q.T.

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