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Original research article

Adsorption of Ammonia and Total Phosphorus in Surface Water Using Zeolite and Activated Carbon: Case Study of Chuat Man Canal, Samut Prakan

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ABSTRACT

Ammonia and phosphorus have been recognized as the cause of eutrophication in surface water. Chuat Man Canal is faced with water quality degradation problem due to the high concentrations of ammonia and total phosphorus in the water body that makes it unsuitable for fish ponds. Removal of ammonia and phosphorus by the adsorption process is simple and not requires chemical use. In addition, ammonia is well adsorbed by activated carbon and zeolite while phosphorus is adsorbed by zeolite. This research used zeolite and activated carbon for the adsorption of ammonia and total phosphorus. The results of laboratory experiments at 30 ˚C 200 rpm 60 minutes, revealed that adsorption of ammonia using zeolite correlated with Freundlich isotherm ($R^2 = 0.9031$). For ammonia adsorption using activated carbon, it correlated with Langmuir ($R^2 = 0.9596$) and Freundlich ($R^2 = 0.9113$) isotherms, respectively. For field experiment, 9 zeolite and activated carbon adsorbent pads with ratio of 1.6:1 by weight were placed across the canal sections. Each pad had 2 openings and each opening contained its adsorbent volume of $1.0 \times 0.015 \times 0.6$ m^3 (width × length × height). The front opening contained 5 kg of activated carbon while the back part contained 8 kg of zeolite. During the study, water flow velocity at surface of water was ranged from 0.022 - 0.027 m/s. Concentration of ammonia in influent and effluent was ranged from 1.755- 8.817 mg/L and 1.473-7.063 mg/L, respectively while that for total phosphorus was ranged from 0.045 - 0.095 mg/L and 0.042 - 0.089 mg/L, respectively. The maximum removal efficiency occurred 20 and 43 minutes after installation of the adsorption pads which were 6.73% for total phosphorus and 23.17% for ammonia, respectively.

Keywords: Zeolite; Activated carbon; Adsorption; Canal; Ammonia; Phosphorus; Surface water

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1. Introduction

High concentrations of nitrogen and phosphorus in still water can cause eutrophication resulting in deterioration of water quality, rapid growth of aquatic plants and subsequent reduction of dissolved oxygen concentration. Chuat Man Canal is the still water with low flow canal facing the deterioration problems due to the high concentrations of ammonia (0.720-5.470) mg/L) [1] and total phosphorus (1.980-15.530 mg/L) [1] that are unsuitable for fish ponds (ammonia shall be not higher than 0.5 mg-N/l for fish ponds) [2,3]. Removal of ammonia and total phosphorus could be achieved by various processes. However, the adsorption process is simple method, not requires chemical use, no sediment, and no waste generation for further treatment. Activated carbon and zeolite are mostly used for the adsorption process and able to adsorb ammonia in water; moreover, zeolite is able to adsorb phosphorus. This research was designed to use zeolite and activated carbon for ammonia and total phosphorus adsorption of surface water in the canal. The research was separated into 2 parts: (1) the laboratory experiment to investigate the adsorption isotherms for ammonia and total phosphorus and the suitable ratio of zeolite and activated carbon for ammonia and total phosphorus adsorption in Chuat Man Canal and (2) the field experiment to investigate the arrangement patterns of the above suitable ratio of adsorbent and the adsorption efficiency for ammonia and total phosphorus in the canal with continuous water flow condition.

2. Materials and Methods

For water quality analysis, research grade of chemical was used. The analytical method used the standard method for examination of water and wastewater. Parameters included pH using electrometric method (4500- pH value B), electrical conductivity (EC) using laboratory method (2510B), ammonia using titrimetric method (4500-NH_3 C) and total phosphorus using ascorbic acid method (4500-PE). Adsorbents used in this study were clinoptilolite zeolite with a diameter of 3.00- 5.00 mm and granular activated carbon (GAC) produced from coconut shells with a diameter of 2.36-4.75 mm. These adsorbents after purchasing from the manufacturers were immediately used without cleaning or activating in order to be easy and convenient for the community to use further for ammonia and total phosphorus removal in the canal.

The research was separated into 2 parts as follows.

2.1 Laboratory experiment

2.1.1 Characteristics of zeolite and activated carbon

This section was to study the characteristics of zeolite such as pH, EC, pH_{PZC}, acidic functional group, basic functional group and cation exchange capacity (CEC) and also the characteristics of activated carbon such as pH, EC, pH_{PZC}, acidic functional group, basic functional group, iodine number and specific surface area. Details of the analytical method are indicated in Table 1.

For the study of adsorption isotherm, surface water from Chuat Man Canal was collected by grab sampling for further investigation in the laboratory experiment.

2.1.2 Adsorption behaviors of activated carbon and zeolite for nutrient adsorption

The laboratory experiment was performed using batch process. Amount of adsorbents ranging from 0-50 g/100 mL was mixed with surface water at 200 rpm at 30 °C for 60 minutes. Then the water was filtered by using Whatman grade 1 with a diameter 11 cm. Filtered water (after adsorption) was then analyzed for pH, EC, ammonia and total phosphorus in the same way as the surface water before adsorption process. Results were used to construct the adsorption isotherm equations for ammonia and total phosphorus as indicated in Eq. (2.3) and Eq.

(2.5) Calculation methods for nutrient adsorption are indicated as follows.

Table 1. Analytical method for adsorbent characteristics.

Parameters	Analytical methods	References
pH^1	pH meter	[4,5,6]
EC^1	EC meter	[7]
pH$_{PZC}$^{2}	titration	[8,9]
acidic functional group^3	Boehm titration	[5,9]
basic functional group^4	titration	[5,9,10]
CEC	Determination of CEC at pH 7 with ammonium acetate	[11]
iodine number	Determination of iodine number of activated carbon	[12]
specific surface area	Nitrogen adsorption, S$_{BET}$	[5,6,13]

2.1.3 Calculation of nutrient adsorption

$$q_e = \frac{(C_0 - C_e)\times \forall}{W} \tag{2.1}$$

where q_e is nutrient adsorption capacity (mg/g)
C_0 is initial concentration of nutrient in water (mg/L)
C_e is equilibrium concentration of nutrient in water (mg/L)
$\forall$ is volume of water (L)
W is weight of adsorbent (g).

2.1.4 Langmuir adsorption isotherm equation of nutrient adsorption

$$q_e = \frac{bq_mC_e}{(1+bC_e)} \tag{2.2}$$

where q_e is nutrient adsorption capacity (mg/g)
C_0 is equilibrium concentration of nutrient in water (mg/L)
b is Langmuir constant at a certain temperature
q_m is maximum adsorption capacity for forming mono layer (mg/g).

b and q_m can be obtained from Eq. (2.3).

$$\frac{1}{q_e} = \frac{1}{bq_mC_e} + \frac{1}{q_m} \tag{2.3}$$

where is 1/ q_m the interception on y-axis and $1/bq_m$ is the slope of Eq. (2.3)

2.1.5 Freundlich adsorption isotherm equation of nutrient adsorption

$$q_e = K_f C_e^{\frac{1}{n}} \tag{2.4}$$

where q_e is nutrient adsorption capacity (mg/g)
C_e is equilibrium concentration of nutrient in water (mg/L)
K_f is Freundlich constant of nutrient adsorption at equilibrium (mg/g)
n is constant of adsorption intensity.

K_f and n and can be obtained from Eq. (2.5).

$$\log q_e = \log K_f + \frac{1}{n}\log C_e \tag{2.5}$$

where $\log K_f$ is the interception on y-axis and $1/n$ is the slope of Eq. (2.5).

2.1.6 Study of adsorbent ratio

The experiment was conducted in the same way as described in earlier section. Amount of adsorbents ranging from 0-40 g/100 mL was used. The suitable ratio of adsorbents by weight was considered from the reduction of ammonia and total phosphorus concentrations per adsorbents used.

2.2 Field experiment (Chuat Man Canal)

The ratio of adsorbents obtained from the previous experiment was used to design for arrangement of adsorbent pads in the canal to investigate ammonia and total phosphorus removal in the actual and continuous flow of surface water. Time spent for the field experiment was 1,925 minutes. The velocity at the surface of water was measured during the study. Surface water in the canal was collected before and after passing through the adsorbent pads (as influent and effluent water, respectively) with the total of 8 times by grab sampling method at the water depth of adsorbent pads from the water surface (30 cm). Collection time for water sampling after passing the adsorbent pads was calculated from water flow velocity, length of adsorbent pads and initial collection time (before passing the adsorbent pads). Then, samples were analyzed for pH, EC, ammonia and total phosphorus. The obtained results were used further to calculate for nutrient loading (Eq. (2.6)).

$$Loading = Co \times v \times \mathrm{A} \qquad (2.6)$$

where *loading* is nutrient loading (g/s)

Co is initial concentration of nutrient in water (mg/L)
v is velocity of water (m/s)
A is area of water flowing through adsorbent pads (m^2)

3.Results and Discussion

Results obtained from the laboratory and filed experiments can be elaborated and discussed as follows.

3.1 Laboratory experiment

3.1.1 Characteristics of adsorbents

The characteristics of zeolite were summarized as follows: pH (7.68-8.26), EC (5.3 µS/cm), pH_{PZC} (6.34-6.85), acidic functional group (2.225 mmol/g), basic functional group (0.850 mmol/g) and CEC (146.60 cmol/g). The characteristics of activated carbon were summarized as follows: pH (10.50-10.85), EC (292-299 µS/cm), pH_{PZC} (7.68-7.77), acidic functional group (2.512 mmol/g), basic functional group (0.750 mmol/g), iodine number (920 mg/g) and specific surface area (644.942 m^2/g). The pH of activated carbon was ranged in alkali condition. The alkali of activated carbon may cause from Eq. (2.1) the production process; combustion in low air at 600-700 °C for activated carbon production could increase its alkalinity [14], Eq. (2.2.) metal ions such as magnesium bounded with carbon could produce hydroxide ion when dissolving in water [4] or Eq. (2.3) it could be possibly due to the alkalinity of oxygen functional groups bounded at the surface area of activated carbon [15].

3.1.2 Adsorption behaviors of adsorbents for nutrient adsorption

Adsorption behavior of zeolite for ammonia adsorption was fitted with Freundlich isotherms (Table 2). Adsorption behavior of activated carbon for ammonia adsorption was fitted with Langmuir and Freundlich isotherms (Table 2). In contrast, adsorption behaviors of zeolite and activated carbon for total phosphorus adsorption were not fitted with both Langmuir and Freundlich isotherms.

Table 2. Langmuir and Freundlich isotherm equations of zeolite and activated carbon for ammonia adsorption.

Langmuir isotherm	Zeolite	Activated carbon
R^2	0.8334	0.9596
q_m	0.0493	0.0837
b (l/g)	0.2392	0.2222
q_e (mg/g)	$q_e = \frac{0.0118\,C_e}{1+0.2392\,C_e}$	$q_e = \frac{0.0186\,C_e}{1+0.2222\,C_e}$

Freundlich isotherm	Zeolite	Activated carbon
R^2	0.9031	0.9118
K_f	0.0201	0.2907
1/n	0.8989	0.5846
q_e(µg/g)	$q_e = 0.0201\,C_e^{0.8989}$	$q_e = 0.2917\,C_e^{0.5846}$

3.1.3 Optimum adsorbent ratio

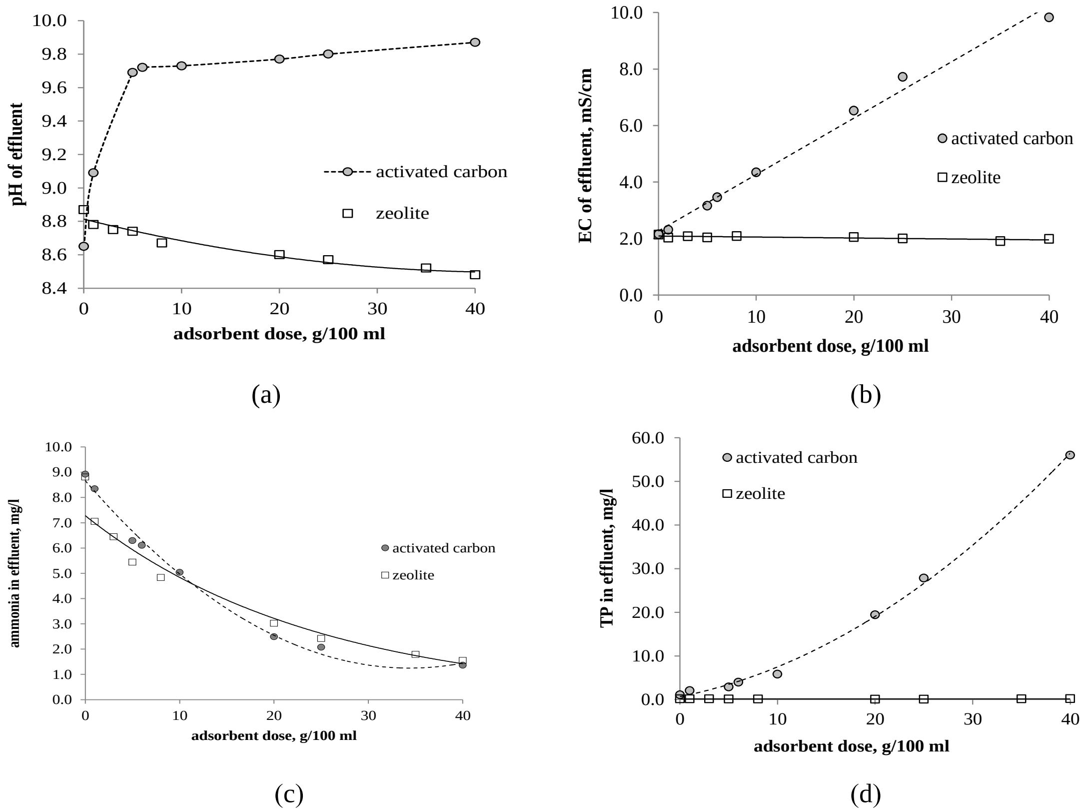
Results revealed that, for single adsorbent, activated carbon increased pH EC and total phosphorus in water. Zeolite slightly decreased pH and phosphorus in water (Fig. 1. (a), (b) and (d)). Both zeolite and activated carbon could reduce ammonia concentration in the comparable removal efficiency (Fig.1. (c)). Hydroxide ion from activated carbon increased pH and EC in the water [4, 14, 15]. The raw material for activated carbon was coconut shells which contained high amount of phosphorus [14]. Once dissolving in water, phosphorus could be released to generate high concentrations of total phosphorus and brought about an increase of EC in the water. Furthermore, it was possibly due to various anions presented in the surface water such as chloride, nitrate and sulfate that were competitive for adsorption of activated carbon with phosphate [20].

For zeolite, ammonia removal mechanism was elaborated accordance with its characteristics as follows. The pH of water (8.48-8.87) was less than pKa of ammonium (9.25) so that ammonium ion, which was cation, presented in the water. Zeolite was able to perform cation exchange 146.60 cmol/kg. The pH of water was above point of zero charge which created negative charge at the surface of zeolite. In addition, zeolite also had high acidic functional groups (2.225 mmol/g). The acidic functional groups such as carboxylic and lactonic acids had lower pKa (4.76 and 8.2, respectively) than pH of water which also created negative charge at the surface of zeolite. In summary, ammonia adsorption mechanism using zeolite consisted of cation exchange between zeolite and ammonium ion [17] and attraction from electrostatic force between ammonium and negative charge at the surface of zeolite.

For removal mechanism of ammonia using activated carbon, high iodine number of activated carbon represented the ability for removal of pollutants/adsorbates with a diameter less than 1 nm [18]. High specific surface area as 644.942 m^2/g could enhance adsorption efficiency. The pH of water ranged between 9.69-9.87 which was higher than pKa of ammonia (9.25). Hence, ammonia (NH_3) with molecular size of 0.326 nm was found in the water [19] which was adsorbed by activated carbon. In summary, major mechanism for ammonia removal by activated carbon was adsorption due to its surface area. Some amount of ammonia could be partially removed through chemical adsorption due to the reaction with acidic groups at its surface such as carboxylic, carbonyl and phenolic hydroxyl to produce the chemical compounds $(CO^-(NH_4)^+)$ at the surface of activated carbon [15].

To reduce ammonia and total phosphorus in the canal and to obtain the appropriate condition of pH and EC for fish cultures (pH and EC should be less than 9 [16] and 5.00 mS/cm [2], respectively), optimum range of zeolite and activated carbon could be between 8-20 g/100 ml and less than 6 g/100 ml, respectively.

Hence, suitable mixing ratio of zeolite and activated carbon for ammonia and total phosphorus could be ranged between 8:6 to 20:6 g/100 ml or ratio 1.3:1 to 3.3/1 by weight, respectively.



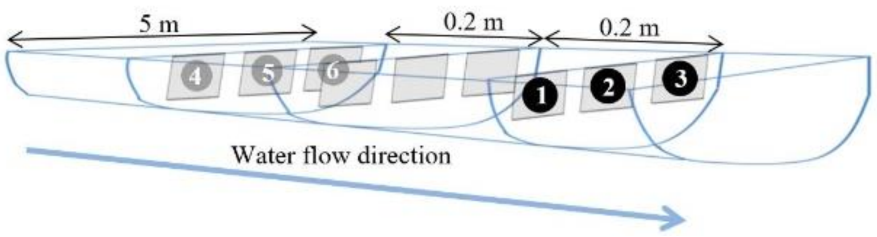
(a) (b)

(c) (d)

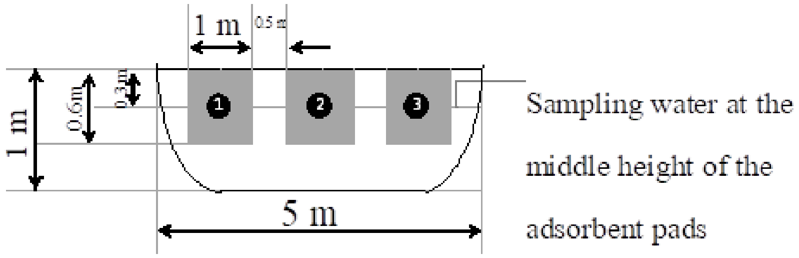
Fig. 1. Relationship between amount of zeolite and activated carbon, and (a) pH, (b) EC, (c) ammonia and (d) TP of surface water in Chuat Man Canal after adsorption process at 200 rpm, 30 °C and 60 minutes.

3.2 Field experiment (Chuat Man Canal)

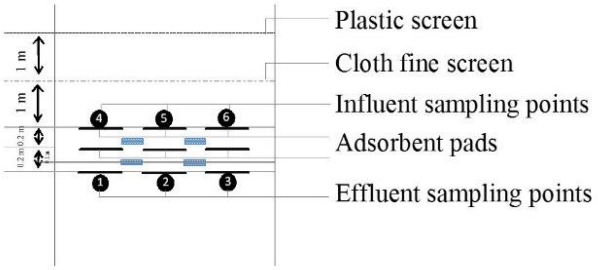
In the field experiment, ammonia and total phosphorus adsorptions were investigated using the adsorbent pads which were combined with zeolite and activated carbon in ratio of 1.6/1 by weight. The total number of 9 pads was placed in the canal along the cross section area. Each pad consisted of 2 opening and each opening contained its adsorbent volume of 1.0 × 0.015 × 0.6 m^3 (width × length × height). The front openings (which faced the up flow area) contained 5 kg of activated carbon while the back openings contained 8 kg of zeolite. The adsorbent pads were placed for 3 rows of 3 pads. The distance between each row was 0.20 m and the distance between each adsorbent pad was 0.50 m. The 4 baffles were also installed to prevent from shortcut of water flow. The baffles were placed for 2 rows along the cross section area of the canal and each row with 2 baffles in between the adsorbent pads at the middle between the rows (0.10 m). The baffles were put down into the water at the submerged depth of the adsorbent pads (0.60 m). The plastic screen and cloth fine screen were also installed along the cross section area of the canal at the inlet part of the experiment (at 1 m before the adsorbent pads) to remove debris contaminated in water that might inhibit the adsorption ability (Fig.2. and Fig.3.). The adsorbent pads were placed in the canal for 1,925 minutes. Water samples were collected before and after adsorption at inlet part (points 4, 5, 6) and outlet part (points 1, 2, 3), respectively (Fig. 3.) and at the specified times (Table 3). In the field experiment, water continuously flowed through



(a) side view



(b) cross section



(c) top view and sampling points of influent and effluent

Fig. 2. Layout of adsorbent pad arrangement: (a) side view, (b) cross section and (c) top view, and sampling points of influent and effluent (point 4-6 and point 1-3, respectively).

the adsorbent pads during the study period. The flow velocity of water was found to be ranged from 0.022-0.027 m/s. Contact time of the adsorbent and water was ranged from 1.1-1.36 seconds (Table 3). Ammonia and total phosphorus concentration of influent were fluctuated (Fig. 4.) that were different with the laboratory experiment. In the laboratory, contact time was controlled to be 60 minutes while all parameters included volume of water, temperature of water, ammonia concentration, total phosphorus concentration and other contaminants in water as well as mixing rate (rpm) were kept constant during the 60 minutes. Results of ammonia and total phosphorus adsorption in the canal (Table 3, Fig. 4. and Fig. 5.) found that pH and EC of water after the adsorbent pads were increased at the times between 20-249 minutes and 20-370 minutes, respectively. Increment of pH and EC was consistent with the laboratory results that using activated carbon increased pH and EC in surface water from the canal. In the field experiment at the times longer than 480 minutes, pH and EC of water were reduced that could be due to reduction of ions released from activated carbon after the longer period (Fig. 4 (a) and Fig. 4 (b)). Ammonia and total phosphorus loadings in different times were different depending on the influent concentrations of ammonia and total phosphorus and the flow velocity of water in each time. Ammonia concentration in the influent at the time between 20 – 191 minutes was reduced from 8.817 mg/l to be 3.830 mg/l while total phosphorus concentration in the influent at the time between 92 to 191 minutes was dramatically reduced from 0.090 mg/l to be 0.045 mg/l. Although the concentrations of ammonia and total phosphorus loading were fluctuated, the adsorbent pads were found to reduce the concentrations of ammonia and total phosphorus in the canal through all the study period (Table 3, Fig. 4 (c) and (d)).

The maximum removal efficiencies of ammonia and total phosphorus were 23.17% and 6.73%, respectively at 43 and 20 minutes, respectively. Later on, removal efficiencies of ammonia and total phosphorus were reduced depending on the increasing quantity of flowing water and times (Table 3). It could be explained that the available surface area of activated carbon and the available ion for ion exchange of zeolite were reduced. In addition, changes of reduction of ammonia concentration, reduction of total phosphorus concentration, ammonia loading, total phosphorus loading, ammonia adsorption rate and total phosphorus adsorption rate were indicated in Fig. 5. Although in the initial period (20-92 minutes) ammonia loading was higher than the latter period (191-1,925 minutes), reduction of ammonia concentration was lower in the latter period. It could be seen that changes of reduction of ammonia concentration and ammonia adsorption rate tended to slightly reduce with time and almost constant at the time longer than 480 minutes (Fig. 5 (a)). For phosphorus, changes of total phosphorus loading, reduction of total phosphorus concentration and total phosphorus adsorption rate followed the same trend as the ammonia (Fig. 5 (b)). In the initial period, cation at the surface area of zeolite and surface area of activated carbon were high and available for ion exchange and adsorption, respectively with ammonia.

Table 3 Sampling times of influent and effluent, water flow velocity and removal efficiency of ammonia and total phosphorus in Chuat Man Canal at different times.

water collection time, minutes		water flow velocity, m/s	contact time, s	%NH_3 removal	%TP removal
influent	effluent				
20	20:22	0.027	1.11	19.89	6.73
43	40:23	0.026	1.15	23.17	5.58
92	90:23	0.026	1.15	22.59	2.66
191	190:24	0.025	1.20	18.09	5.36
249	240:26	0.023	1.30	15.67	4.71
370	270:23	0.026	1.15	12.45	4.09
480	480:27	0.022	1.36	10.18	3.12
1,925	1920:24	0.025	1.20	16.07	2.22

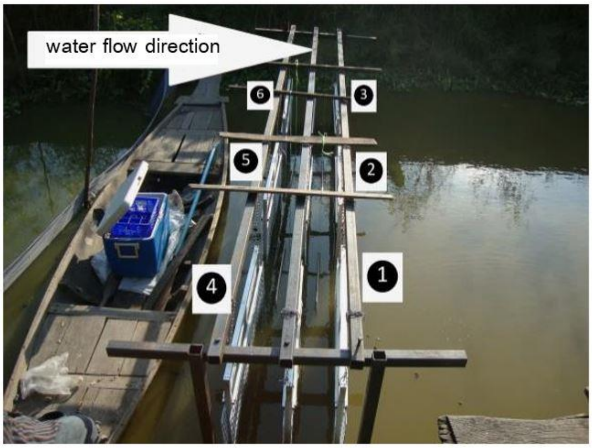


Fig. 3. Field experiment of combined zeolite and activated carbon adsorbent pads in Chuat Man Canal and sampling points of influent and effluent (point 4-6 and point 1-3, respectively).

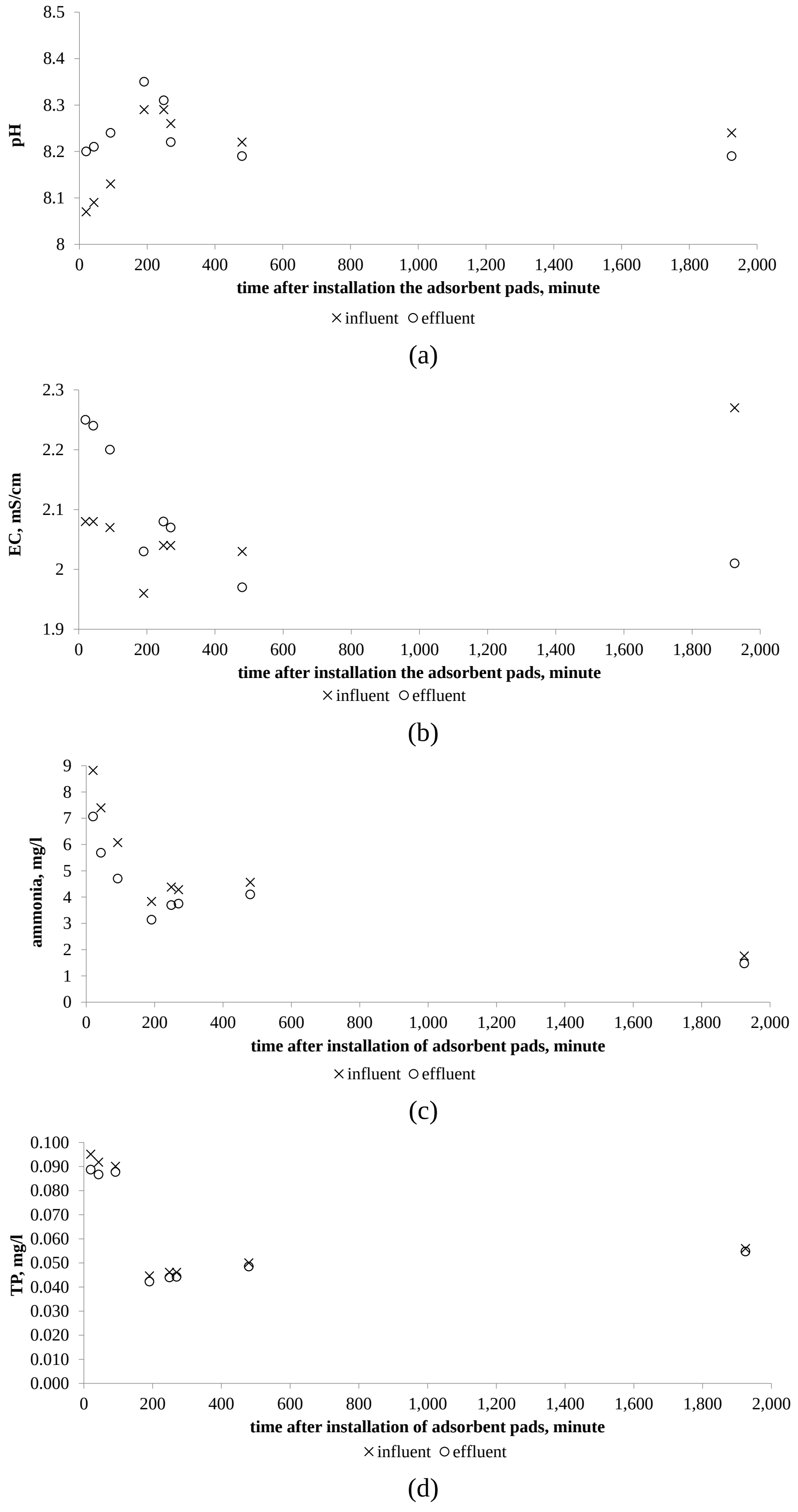
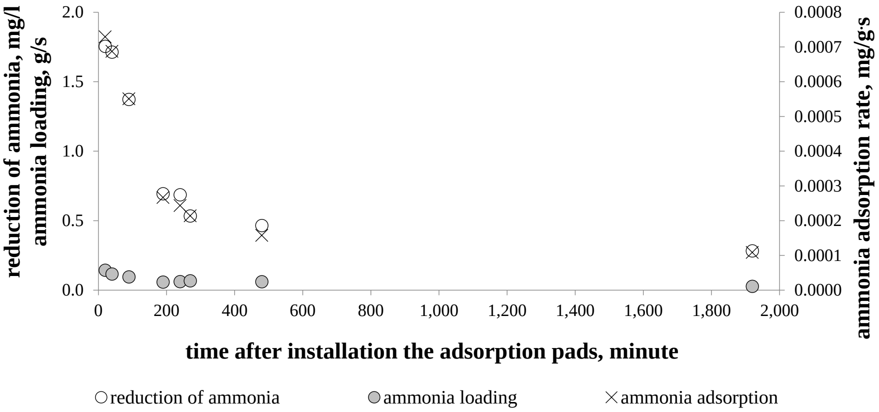
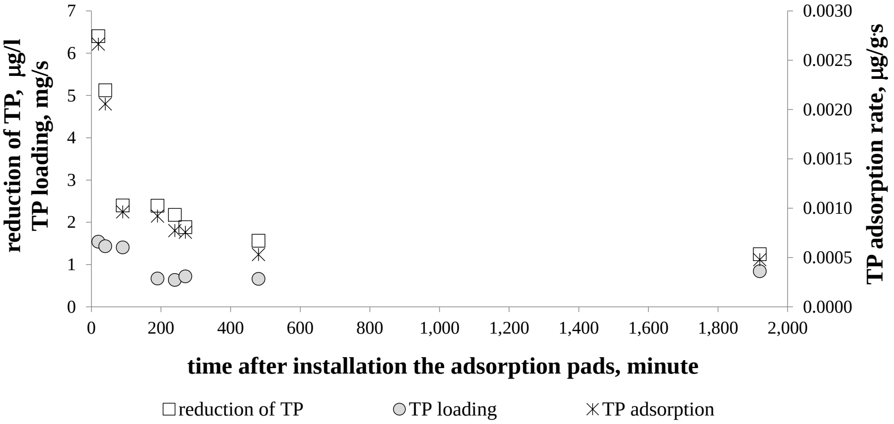


Fig. 4. Water quality before and after adsorption at different times: (a) pH, (b) EC, (c) ammonia and (d) total phosphorus (TP).



(a)



(b)

Fig. 5. Nutrient loading, reduction of nutrient concentration and nutrient adsorption rate different times after installation of adsorbent pads in the canal: (a) ammonia and (b) total phosphorus.

The longer the adsorbent pads were placed in the canal, cation of zeolite was reduced due to cation exchange with ammonia and available surface area of activated carbon was reduced due to ammonia adsorption. Meanwhile, partial concentration of phosphorus was adsorbed in the porous and surface area of activated carbon [21], was adsorbed by electrostatic attraction and ion exchange [22]. This process was time dependent mechanism. The ability for adsorption was reduced by increasing times. Finally, zeolite and activated carbon were less adsorb or ineffective at 1,925 minutes. Moreover, nitrogen in organic compounds adsorbed by activated carbon could be converted into ammonia that caused an increase of ammonia concentration [23]. Although adsorption efficiencies of ammonia and total phosphorus should be lowest at the time 1,925 minutes it was found that the adsorption efficiency of ammonia at 1,925 minutes was higher than that at 480 minutes.

This could be explained that ammonia concentration of influent at 1,925 minute was lower than 480 minute 2.6 times (4.560 and 1.755 mg/L, respectively) that would result in the higher efficiency.

4. Conclusion

When the single adsorbent was used, zeolite was able to adsorb ammonia and total phosphorus in surface water from Chuat Man canal. Activated carbon was able to adsorb ammonia while it increased total phosphorus, pH and EC of water. The ammonia adsorption of zeolite and activated carbon were fitted with Freundlich isotherm equations. Co- utilization of zeolite and activated carbon by making the adsorbent pads and placing along the cross section area of the canal can reduce concentrations of nutrient as ammonia and total phosphorus. The removal efficiencies of ammonia and total phosphorus were 10. 18- 23. 17% and 2.22-6.73%, respectively.

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