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Original Research Paper

Effects of Peat and Mushroom Residues on Removing Ammonia-Nitrogen and Total Phosphorus in Wastewater

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ABSTRACT

Peat and mushroom residues were selected for adsorption isotherm and adsorption kinetics experiments and to comparatively discuss the effects of removing ammonia-nitrogen and the total phosphorus in wastewater. The results showed that the peat and mushroom residues for removing ammonia-nitrogen and the total phosphorus followed the Freundlich and Langmuir equations of the adsorption isotherm. By using a fitting equation, we obtained the static adsorption quantities of ammonia-nitrogen, which were 2.3679 mg/g and 2.1643 mg/g, and the static adsorption quantities of total phosphorus were 1.4363 mg/g and 1.3758 mg/g. A first-order kinetics equation was used to fit the reaction process of the two-substrate adsorption of ammonia-nitrogen and the total phosphorus. The correlation coefficients were all desirably above 0.990. Furthermore, the adsorption rates of ammonia-nitrogen were 0.0088 g/(mg·min) and 0.0084 g/(mg·min), and the adsorption rates of the total phosphorus were 0.001 g/(mg·min) and 0.0006 g/(mg·min). In a simulation of the soil percolation system, a volume ratio of 10% peat and 10% mushroom residues with different sizes of 0.5, 2 and 4 mm were mixed with the soil in different treatments (T1-T6), and the control group was the soil (CK). The concentration of ammonia-nitrogen in the wastewater was 54.45-78.96 mg/L, and the hydraulic loading was 0.08 $m^3/(m^2 \cdot d)$. The performance of seven types of experimental devices for removing ammonia-nitrogen in the wastewater was T2 > T3 > T1 > T6 > T5 > T4 > CK. The average concentration of ammonia-nitrogen in the effluent water was 65.58 mg/L, and the removal rate of ammonia-nitrogen and the total phosphorus reached 88.04-95.32% and 89.23-97.08%, respectively, which satisfied the additional minimum UWTD treatment performance for a discharge to a sensitive area.

INTRODUCTION

The main pollutants of wastewater include TN (total nitrogen), TP (total phosphorus), NH_4^+-N (ammonia-nitrogen), and COD (Chemical Oxygen Demand). The eutrophication process is closely related to the ratio of nitrogen and phosphorus. As reported in the literature, eutrophication occurs when the nitrogen content in water exceeds 0.2-0.3 mg/L or the concentration of the total phosphorus is greater than 0.02 mg/L (Wang & Gao 2006). It is generally believed that the total phosphorus is adsorbed by physical adsorption, and ammonia-nitrogen is adsorbed by physical adsorption and the microbial action in the soil filtration system (Bai et al. 2004). Currently, the most widely used substrates include soil, gravel, sand, zeolite, etc. (Shi et al. 2008, Qiao & Ren 2012), but these substrates are poor for the removal of ammonia-nitrogen and the total phosphorus; thus, the study of the physical and chemical properties of new substrates has received attention (Zhang et al. 2004). Peat and mushroom residues have good selective adsorption performance for pollutants in wastewater. As organic solid wastes, the use of peat and mushroom residues and other organics to remove ammonia-nitrogen, the total phosphorus, and other pollutants in wastewater is a promising method (Siegrist 1987), but there is little research on the impact of different particle sizes of peat and mushroom residues for removing ammonia-nitrogen and the total phosphorus in wastewater. This study systematically studied the effects of the peat and mushroom particle sizes on the adsorption, adsorption isotherms, and adsorption kinetics. A dynamic adsorption column simulation, which measures the soil permeability index and the change in carbon-to-nitrogen ratio (C/N) and monitors the removal efficiency of NH_4^+-N and TP, was used to analyse the influence of the particle sizes on the treatment effect. This analysis provided a theoretical basis for choosing the appropriate soil infiltration system for removing ammonia-nitrogen and total phosphorus.

MATERIALS AND METHODS

Materials: The physical and chemical properties of the peat and mushroom residues are summarized in Table 1. A volume ratio of 10% peat and 10% mushroom residues was mixed with the soil in different proportions, and mixtures with grain sizes of 0.5, 2 and 4 mm were used as the substrates (T1-T6) (Table 2). The control group (CK) was the soil.

Isothermal adsorption experiment: Six grams of the peat and mushroom residues with particle sizes of 0.5, 2 and 4 mm were each added to a 500-mL conical flask. NH_4CL for preparation of ammonia-nitrogen solution in 100mL with different concentrations (5-100 mg/L, pH = 7) were added to each conical flask and KH_2PO_4 (containing 0.01 mol/L KCl as a supporting electrolyte) for preparation of total phosphorus solution in 100mL with different concentrations (2.5-150 mg/L, pH=7) were added to each conical flask to do isothermal adsorption experiments respectively. The mixture was shaken for 24 h under the condition of constant temperature oscillation incubator at a speed of 150 rpm every minute, the temperature was set to 25°C, standing for 15 min and took supernatant to determinate balance concentration C_e of the total phosphorus. The adsorption of ammonia-nitrogen and total phosphorus for different particle sizes of peat and mushroom residues was calculated as follows:

$$q_e = (C_0 - C_e)\ \ V/m \quad ...(1)$$

Where C_0 is the initial concentration of the solution in milligrams per litre, C_e is the equilibrium concentration of the solution after adsorption in milligrams per litre, V is the solution volume, and m is the mass of the substrates.

It is generally believed that total phosphorus adsorption occurs by physical adsorption, and the adsorption of ammonia-nitrogen occurs by physical adsorption and microbial action (Qin et al. 2009). The Langmuir and Freundlich equations have usually been used as the isotherm adsorption equation to represent the physical process (Ye 2007). The Langmuir equation is a monolayer adsorption model derived from the adsorption equation; the Freundlich equation is established on the basis of the adsorbent in the multiphase surface adsorption of the adsorption equilibrium model. The construction of the two equations is based on certain assumptions, and the choice of which equation describes the solid medium depends on the adsorption of pollutants in the solution process and needs experimental data for fitting. According to the different adsorption formulas, it is necessary to use a mathematical simulation method to determine these equations (Cui et al. 2003). The adsorption isotherms were in accordance with the Langmuir and Freundlich equations within the scope of the research temperature. They are expressed as follows:

Langmuir equation:

$$q_e = bq^0C_e / \left(1 + bC_e\right) \quad ...(2)$$

q^0 and b are the Langmuir constants of the maximum adsorption capacity and the adsorption energy, respectively, and are related to the temperature and other factors.

Freundlich equation:

$$q_e = K_F C_e^{1/n} \quad ...(3)$$

K_F is the Freundlich constant of the homogeneous adsorbent, and n is related to the magnitude of the adsorption force and the adsorption energy distribution.

Adsorption kinetics experiments: Six grams of peat and mushroom residues with a particle size of 0.5 mm were added to a 500-mL conical flask. NH_4CL for preparation of ammonia-nitrogen solution in 100mL with different concentrations (5-100 mg/L, pH = 7) were added to each conical flask and KH_2PO_4 (containing 0.01 mol/L KCl as a supporting electrolyte) for preparation of total phosphorus solution in 100mL with different concentrations (2.5-150 mg/L, pH = 7) were added to each conical flask to do isothermal adsorption experiments respectively. The mixture was oscillated in the constant-temperature oscillation incubator at a speed of 150 rpm, and temperature was set to 25°C. After a certain time interval, we removed the Erlenmeyer flask, obtained the supernatant after centrifugation, and used Nessler's reagent colorimetric method and ammonium molybdate spectrophotometric method to measure the mass concentration of ammonia-nitrogen and the total phosphorus respectively in the supernatant. Every experiment was repeated three times.

Table 1: Physical and chemical properties of three substrates.

Substrates	Density (g/cm^3)	Total porosity (%)	Aeration Porosity (%)	Water holding porosity (%)	Water Content (%)	pH	CEC (cmol/kg)	Organic carbon content (%)	TN (g/kg)	TP (g/kg)	TK (g/kg)
Soil	1.73	40.32	32.96	8.94	2.93	7.31	3.46	1.09	0.39	0.47	1.9
Peat	0.17	78.8	31.25	40.67	36.45	5.1	123.0	58.21	2.38	0.073	1.64
Mushroom residue	0.18	84.2	23.34	37.58	24.7	7.61	78.2	45.83	3.2	0.035	1.02

The first-order kinetics equation can be used to fit the solid medium for the adsorption kinetics of the pollutants in the wastewater. In order to study the adsorption kinetics, false first-level and second-level equation-fitting adsorption kinetics data were used (Yan 2008).

Lagergren first-class equation:

$$\ln\left(q_e - q_t\right) = \ln q_e - k_1 t \qquad ...(4)$$

Lagergren second-class equation:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^{\ 2}} + \frac{1}{q_e} t \qquad ...(5)$$

q_t is the ammonia-nitrogen adsorption capacity of peat at the t^{th} moment (mg/g), k_1 is the adsorption process of the false level (min^{-1}), and k_2 is the fake secondary adsorption rate constant [g/(mg·min)].

Dynamic experiments: ***Experimental devices:*** A volume ratio of 10% peat and 10% mushroom residues with different sizes of 0.5, 2 and 4 mm were mixed with the soil in different treatments (T1-T6), and the control group was the soil (CK). A glass simulation soil infiltration system with a diameter of 10 cm, a height of 100 cm, and a wall thickness of 0.5 cm was used, as shown in Fig. 1. The top 10 cm of the glass tube was left unfilled, and nylon net was placed on the surface, which trapped suspended solids. The height of the filled substrate was 80 cm, and gravel with a particle size 0.5-1 cm padded the bottom 10 cm of the glass tube, which prevented the outlet pipe from clogging. The hydraulic load was 0.08 $m^3/(m^2 \cdot d)$, using intermittent watering twice a day at 10:00 and 22:00. Water samples were obtained once every 2 d at the bottom, and the mass concentrations of ammonia-nitrogen and the total phosphorus in water were measured. The experiments run 20 d.

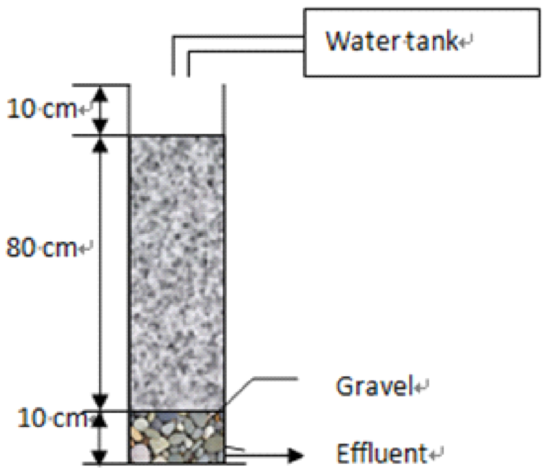

Fig. 1: Experimental matrix column of peat and mushroom residue sewage.

Table 2: Different treatments.

Group	Treatment
CK	Soil
T1	Soil with 10% peat (0.5mm)
T2	Soil with 10% peat (2mm)
T3	Soil with 10% peat (4mm)
T4	Soil with10% mushroom residue (0.5 mm)
T5	Soil with10% mushroom residue (2 mm)
T6	Soil with10% mushroom residue (4 mm)

***Sewage selection*:** In the experiment, the influent wastewater from a sewage treatment plant in Beijing was used and sampled once every two days to ensure water quality.

***Water quality*:** The NH_4^+-N concentration is 54.45-78.96 mg /L, the TP concentration is 7.28-8.98 mg/L, and the pH is 7-8.

Data analysis: Excel 2003 and Origin 8.0 were used for data analysis and mapping.

RESULTS AND DISCUSSION

Static Experiments

Isothermal adsorption curves of substances for NH_4^+-N and TP: The Langmuir and Freundlich equations were used to fit the adsorption of two substances for ammonia-N and the total phosphorus, respectively. The isothermal adsorption curves of three particle sizes of peat and the mushroom residue fitting for NH_4^+-N are shown in Figs. 2a-d. The fitting results are listed in Table 3. The isothermal adsorption curves of three particle sizes of peat and the mushroom residue fitting TP are shown in Figs. 2e-h. The results are listed in Table 4.

Figs. 2a-h. show that when the initial concentrations of ammonia-nitrogen and the total phosphorus increase, their adsorption also increased. This is consistent with the results of a predecessors' study (Su et al. 2007). On the basis of this study, by comparing the three particle sizes for the matrix, it is found that amount absorbed is greater than the mushroom residue, peat, and peat adsorption effect: 0.5mm > 2mm > 4mm and the mushroom residue adsorption effect 0.5mm > 2mm > 4mm. For a smaller particle size, the amount adsorbed is greater. The optimum particle size was 0.5 mm, and on the basis of the results of the study, it is necessary to perform adsorption kinetics experiments.

According to fitting constants of the Langmuir and Freundlich equations and a comparison of the correlation coefficient R^2, the Langmuir equation describes the adsorption behaviour of ammonia-nitrogen and the total phos-

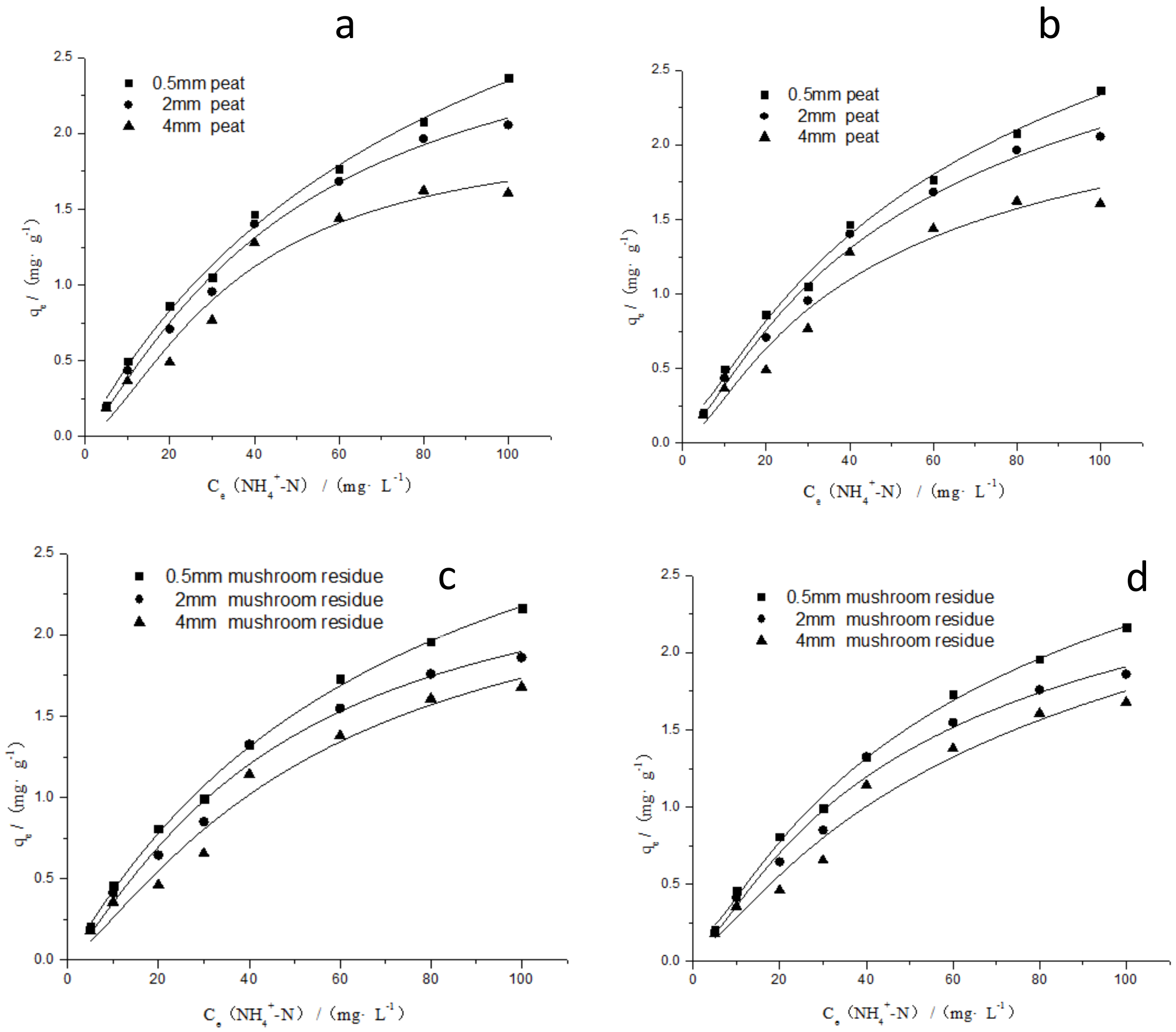

Fig. 2a-d. Langmuir and Freundlich isothermal adsorption curves of two substances for NH_4^+-N.

phorus in the peat and mushroom residues better than the Freundlich equation. This is consistent with the conclusions reported in the literature (Zacar 2003). According to the Langmuir equation, the amounts absorbed at saturation of peat and mushroom for ammonia-nitrogen were 2.3679 mg/g and 2.1643 mg/g, and the amounts adsorbed at saturation of peat and mushroom for total phosphorus were 1.4363 mg/g and 1.3758 mg/g. In the Freundlich equation, k is the adsorption capacity, and a larger value of k results in a stronger adsorption capacity of the adsorbent. The value of n reflects the adsorbent adsorption strength, and its value is less than 0.5, indicating that easy adsorption. When n is greater than 2, adsorption is difficult (Kostura et al. 2005). As can be seen from the value of n, the peat and mushroom residues are suitable for the adsorption of ammonia-nitrogen and the total phosphorus. The peat and mushroom residues can be used to remove ammonia-nitrogen and the total phosphorus in wastewater (Zou et al. 2010).

Adsorption Kinetics Experiment

Adsorption kinetics curves of two substances for NH_4^+-N and TP: The amount of ammonia-nitrogen adsorbed versus time is shown in Fig. 3. At the beginning of adsorption (2 h), the adsorption rate is similar. Then, the adsorption rate of the peat residue is higher than that of the mushroom residue. This is because the addition of the peat and mushroom residues provided more active adsorption sites, which lead to a high adsorption speed. The amount of the total phos-

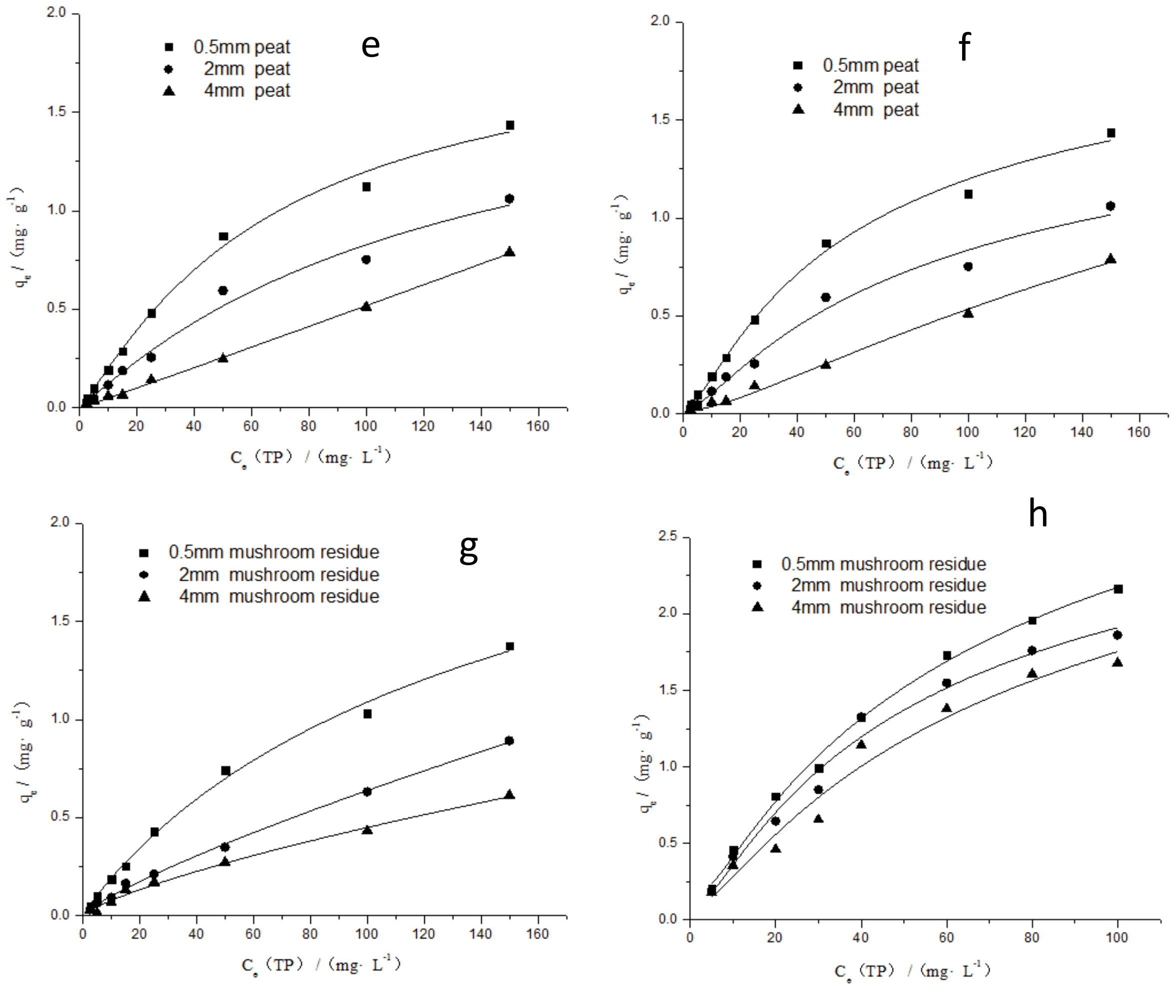

Fig. 2e-h: Langmuir and Freundlich isothermal adsorption curves of two substances for TP.

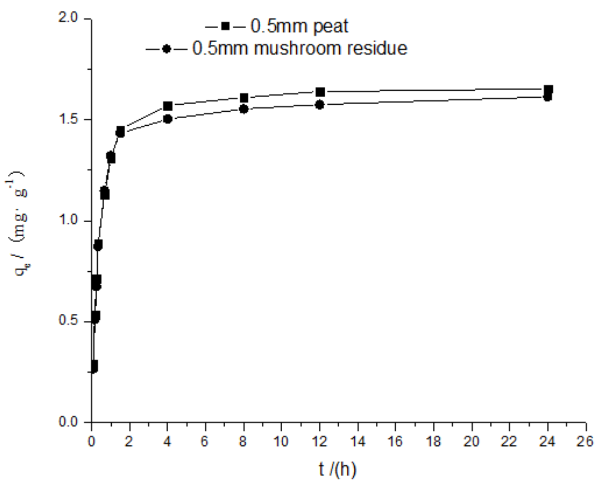

Fig. 3: Adsorption kinetics curves of two substances for NH_4^+-N.

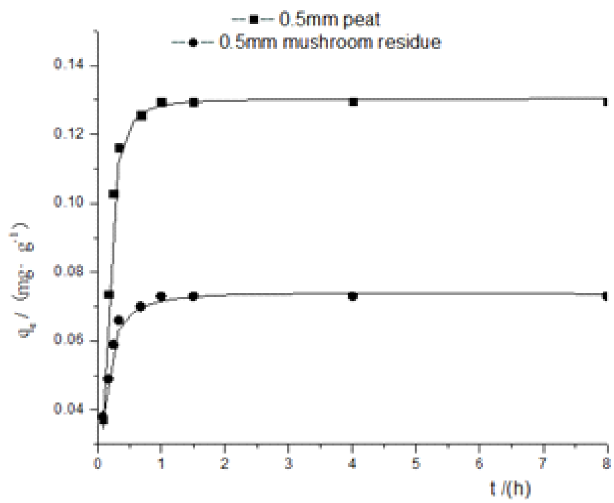

Fig. 4: Adsorption kinetics curves of two substances for TP.

Table 3: Results for the fitting adsorption isotherm of NH_4^+-N.

Group	Langmuir			Freundlich		
	q^0(mg/g)	b	R^2	K_F	n	R^2
T1	2.3679	0.012	0.993	5.001	0.804	0.992
T2	2.0572	0.008	0.988	3.853	0.886	0.986
T3	1.623	0.004	0.951	2.708	0.981	0.944
T4	2.1643	0.011	0.995	4.418	0.825	0.994
T5	1.8608	0.009	0.979	3.384	0.895	0.978
T6	1.6774	0.005	0.963	3.917	0.817	0.961

Table 4: Results for the fitting adsorption isotherm of TP.

Group	Langmuir			Freundlich		
	q^0(mg/g)	b	R^2	K_F	n	R^2
T1	1.4363	0.008	0.992	2.223	0.906	0.992
T2	1.0604	0.006	0.979	2.011	0.829	0.979
T3	0.7888	0.003	0.997	5.492	0.632	0.991
T4	1.3758	0.008	0.994	2.564	0.815	0.991
T5	0.8913	0.006	0.997	3.108	0.678	0.984
T6	0.6135	0.001	0.989	1.541	0.734	0.978

Table 5: Effects of different particle sizes of substrates on TOC, TN and C/N.

Group	TOC(g/kg)	TN (g/kg)	C/N
CK	6.32	0.39	16.2
T1	16.26	0.45	36.13
T2	12.95	0.43	30.1
T3	9.96	0.41	24.29
T4	15.4	0.49	31.4
T5	11.51	0.45	25.6
T6	9.43	0.42	22.5

phorus adsorbed versus time is shown in Fig. 4. At the beginning of adsorption (3 h), the adsorption rate of the peat residue is higher than that of the mushroom residue. Then, the adsorption rate of the peat residue is similar to that of the mushroom residue. After a period of time, the concentration of the peat residue is similar to that of the mushroom residue in wastewater, which leads to a small adsorption force and adsorption rate. Finally, equilibrium is reached at approximately the same time

The fitting correlation coefficient R^2 of the first-level equation is relatively lower than that of the second-level equation, and the second-level equation-fitting correlation coefficient is greater than 0.995. Further, $q_{e,\ cel}$ from the second-level equation is close to $q_{e,\ exp}$, and the experimental data for the second-level equation-fitting adsorption kinetics are better than those of the first-level equation (Liu 2004). The obtained adsorption kinetics curves of the two substances for NH_4^+-N are:

$y = (1.661 \times 3.744 \times x^{(1-0.145)}) / (1 + 3.744 \times x^{(1-0.145)})$ and

$y = (1.591 \times 4.632 \times x^{(1-0.275)} / (1 + 4.632 \times x^{(1-0.275)})$ respectively. The adsorption kinetics curves of the two substances for TP are $y = 0.129 \times x^{(0.004x^{-0.871})}$ and $y = 0.071 \times x^{(0.0053x^{-0..652})}$ respectively. The maximum adsorption rate was achieved at 2 h and reached adsorption equilibrium at 4 h. The ammonia-nitrogen adsorption rate of peat is slightly greater than that of the mushroom residue, and the total phosphorus adsorption rate is significantly faster than that of the mushroom residue.

Dynamic Experiments

Effects of different particle sizes of substrates: Tables 5-6 show that the soil infiltration system has a significant effect. Compared with the control group (CK), the hydraulic, total porosity, capillary porosity, and aeration porosity of the experimental group increased, whereas the permeability decreased. The removal rate is related to the total porosity and permeability with a significant correlation (Wang et al. 2009), and the improvement in C/N can shorten the start time for removing ammonia-nitrogen from the system.

Removal efficiency of NH_4^+-N: The water and soil were almost nearly and the water was slightly higher than the soil on the first day. As the literature shows, this may be due to the added organic substrate itself, which contains nutrients that were leached out (Yu et al. 2013). The removal rates of T1–T6 have improved significantly.

Fig. 5 shows that the removal rate of NH_4^+-N from the wastewater for the two kinds of matrices increased with time.

Table 6: Effects of different particle sizes of substrates on the soil infiltration system.

Group	Permeability (cm/h)	Bulk density (N/m³)	Retention ability(%)	Total porosity(%)	Capillary porosity(%)	Aeration porosity(%)
CK	43.8	1.73	4.89	40.32	8.94	32.96
T1	7.74	1.51	19.89	44.61	27.43	19.33
T2	17.6	1.45	15.7	53.2	23.9	27.8
T3	23.86	1.37	13.4	60.9	20.39	29.6
T4	8.43	1.54	17.2	43.83	24.59	21.2
T5	19.9	1.46	13.8	46.3	18.4	29.3
T6	21.46	1.39	12.3	57.2	16.7	30.4

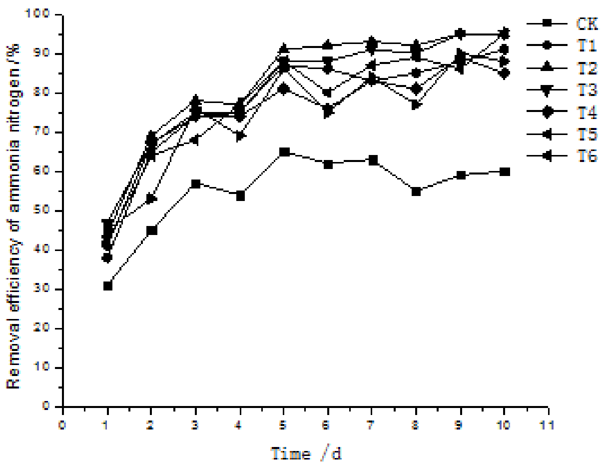

Fig. 5: Removal efficiency of NH_4^+-N.

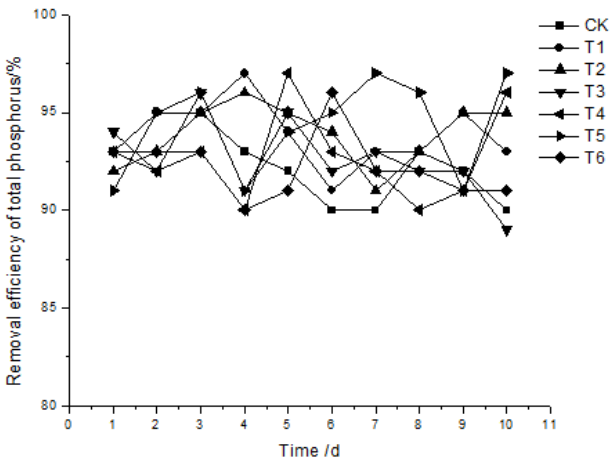

Fig. 6: Removal efficiency of TP.

In the first 10 days of operation, the amount removed of NH_4^+-N came to balance basically. The order of removal of each treatment on NH_4^+-N was T2 > T3 > T1 > T6 > T5 > T4 > CK, where the removal efficiencies of the peat and mushroom residue were 93.66% and 91.33%, indicating that peat provided stronger NH_4^+-N removal. The removal rates of the two substrates have similar trends and increase quickly before 5 days; they gradually stabilized 5 days later. As time increases, NH_4^+ moves into the internal gap and is exchanged with a cation. Diffusion of the internal cation in the liquid phase is a slow adsorption process (Ou et al. 2012). The removal rate of CK was volatile through 9 days, and the system startup time can be shortened by adding organics. The addition of 10% peat has the best effect.

Removal efficiency of TP: Fig. 6 shows that the total phosphorus removal rate is substantially above 90% with a longer run time. The effect of the peat and mushroom residues on removing the total phosphorus is relatively unstable owing to its loose texture. Water can become turbid after a certain period of time (Sakadevan 1998). In addition, during the experimental runs, the substrate column of the total phosphorus is susceptible to the influence of the inlet water quality and running cycle (Beltrán-Heredia & Martin 2011). Because total phosphorus adsorption occurs by physical adsorption, the adsorbed sewage would return to the water when the water quality changed and discharge with the effluent (Drizo et al. 1999). Thus, the addition of organic filler can improve the removal rate of nitrogen and phosphorus.

CONCLUSION

The Langmuir equation describes the adsorption behaviour of ammonia-nitrogen and the total phosphorus in the peat and mushroom residues better than Freundlich equation. When increasing the initial concentrations of ammonia-N and total phosphorus, the total amounts of adsorbed ammonia-N and the total phosphorus for the three particle sizes of the peat and mushroom residues increased overall. The optimum particle size was 0.5 mm, and on the basis of the results of the study, it is necessary to perform adsorption kinetics experiments.

The adsorption kinetics of the fitting calculation results show that the second-level equation fit the experimental data better than the first-level equation. The maximum adsorption rate was achieved at 2 h and reached adsorption equilibrium at 4 h. The ammonia-nitrogen adsorption rate of peat is slightly greater than that of the mushroom residue, and the total phosphorus adsorption rate of peat is significantly faster than that of the mushroom residue. It is generally believed that total phosphorus adsorption occurs by physical adsorption, the adsorption of ammonia-nitrogen occurs by physical adsorption and microbial action. Thus, the addition of organic filler can improve the removal rate of ammonia-nitrogen and total phosphorus.

In the simulation of the soil percolation system, the concentration of the wastewater was 3.43-8.43 mg/L, and the hydraulic loading was 0.08 $m^3/(m^2 \cdot d)$. The performance of seven kinds of experimental devices in removing ammonia-nitrogen in the wastewater was T2 > T3 > T1 > T6 > T5 > T4 > CK. The results showed that the addition of the peat and mushroom residues and other organic matter to the soil has many benefits. It can increase the soil porosity, improve the original soil, prevent blockage, increase the organic matter, and help increase microbial carbon, speed up the startup time of the overall wastewater treatment system, and increase ammonia removal.

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