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ORIGINAL PAPER

High removal of nitrogen and phosphorus from black-odorous water using a novel aeration-adsorption system

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Abstract

Black-odorous waters are an increasingly common phenomenon characterized by excessive levels of nutrients, the formation of metal sufide precipitates, volatile sulfurous compounds, low dissolved oxygen and high chemical oxygen demand. Black-odorous waters frequently occur in lake and river systems where inputs have restricted circulation. The key remediation issue is the removal of nitrogen and phosphorus. Here, we present a novel aeration-adsorption system using fiber balls and we study treatment parameters and removal mechanism. Kinetics and changes of the solid phase were followed using Fourier transform infrared spectroscopy, X-ray photoelectron spectroscopy, and X-ray diffraction. Results show complete removal of ammonia N, initially at 31 mg/L, and 92.8% removal of total nitrogen, initially at 29 mg/L, after a 24 h reaction time at pH 9.67. At pH 5.67 and 9.67, total phosphorus and phosphate could be significantly reduced by 90–92% at 3.2–5.2 mg/L after 24 h. Treatment met China's integrated wastewater discharge standards, demonstrating an effective and robust treatment capability. First-order and second-order kinetic models provided a good fit to the treatment data, indicating physical and chemical adsorption were involved in the treatment reactions. The reaction mechanism involved hydrogen substitution and binding to oxygen. These results present a cost effective and robust approach for the removal of N and P from black, odorous water, providing opportunity to abate environmental contamination.

Keywords Black-odorous water · Nitrogen · Phosphorus · Aeration · Adsorption · Water pollution

Introduction

Black odorous water is characterized by a low oxygen content, unusual color (such as black or grey-black), and odor, posing a potential threat to the ecological environment and human health (GB/T50824-2013, 2013). This harm includes: affecting drinking water resources, exacerbating the water supply crisis; causing human disease and posing a serious threat to people's lives and health; producing pervasive odors and having a significant impact on the ecological environment. Black odorous water, in particular, is the primary source of river pollution, resulting in widespread contamination of rivers, instability of water resources, slow or static flow, and loss of self-purification capacity, often associated with algal blooms (Song et al. 2019; Cao et al 2020). According to recent statistics, the total number of black odorous waterbodies in 295 Chinese cities in 2018 was 1861, while in 2019, it was 2100 (Liu et al. 2021). The majority are mild black odorous water bodies (transparency 10–25 cm, dissolved oxygen 0.2–2 mg/L, oxidation–reduction potential

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− 200–50 mV, ammonia nitrogen 8–15 mg/L) (Ministry of Housing and Urban–rural Development of China, 2015). However, 30 rivers in Beijing, 10 rivers in Shanghai, 31 rivers in Shenzhen, and 18 rivers in Zhanjiang have exceeded the severe black odor standard (transparency lower than 10 cm, dissolved oxygen lower than 0.2 mg/L, redox potential lower than − 200 mV, ammonia nitrogen higher than 15 mg/L). Despite the fact that the Ministry of Housing and Urban–Rural Development of China issued a report in 2015, black odorous water pollution is significant, the composition is complex, the range is broad, and the impact is widespread, so treatment options continue to face significant challenges. The phenomenon of black odorous water has also been observed in the United States, Europe, Asia, and Africa at specific locations such as the Homewood Canal in the United States, the River Thames in the United Kingdom, and Lake Biwa in Japan have faced severe challenges from continued pollution (Cao et al. 2020), and the application of effective technology is needed.

Currently, the most common treatment methods for black odorous water are either in situ or through external treatment (Morin-Crini et al. 2022; Sun et al. 2020). The in situ treatment technology employs physical, chemical, or biological measures to reduce the volume of pollutants as well as their content, and the solubility, toxicity and migration of pollutants. External treatment by and restoration involves the separation of sediment and water, so that the polluted water is transferred to water treatment facilities and then returned to the original water body. However, external treatment and restoration technology necessitates a significant investment in both financial and material resources. Dealing with the sediment that has been removed from the water body is also a difficult problem (Hong 2011). Therefore, in situ remediation technologies are the most cost-effective approach (Morin-Crini et al. 2022) and a variety of methods include: the management of the hydraulic cycle (Guo et al. 2020), enhanced flocculation (Luan et al. 2002), microbial remediation technology (Zheng et al. 2021), artificial floating islands (Lu et al. 2015), and ecological wetland technology (Cai et al. 2021). These methods, however, are not suitable for continuous and efficient water purification under complex conditions, and they are prone to secondary pollution. Adsorption technology, for example, can effectively remove the odor of water, dissolved organic matter, and micro-pollutants; however, some conventional adsorption materials are difficult to recover, and the removal of nitrogen and phosphorus is poor. Polyester fiber balls have good elasticity, high porosity, a fast filtration rate, are recyclable, low in cost, and have a high resistance to acid and alkali (Yang et al, 2019). They are commonly used as filter media to remove suspended solids from water (Geng and Wu 2010). Their application for pollutant removal from water by adsorption is rarely addressed. Some key issues require further investigation, such as the short contact time between pollutants and filter materials, insufficient adsorption, and how to enhance the adsorption effect. Nitrogen removal has always been problematic by adsorption as well as through filtration.

In this study, we developed a novel polyester fiber ball-derived aeration adsorption system and the impact on total nitrogen (total N) and total phosphorus (total P) removal was evaluated. The removal mechanisms for pollutants were analyzed using scanning electron microscopy, Fourier transform infrared spectroscopy, X-ray photoelectron spectroscopy, X-ray diffraction, and adsorption kinetics including the pseudo-first-order and pseudo-second-order kinetic models. At present, the application of fiber balls in water treatment and pollution control is limited and there is considerable scope to develop much needed cost effective strategies (Crini & Lichtfouse 2019). This technology has the potential to provide new opportunities for the application of effective water treatment for extensive and severe contamination problems.

Experimental

Materials

The materials used in this study were of analytical grade, which were as follows: hydrochloric acid (ZhuzhouXingKongHuabo Co., Ltd., China), sodium hydroxide (Aladdin Technology Co., Ltd., Shanghai, China), and ethyl alcohol (Tianjin Hengxing Chemical Reagent Co. Ltd). HACH test kit for measurement of total nitrogen was purchased from HACH Company, USA. Water sample was collected from those rural ponds around Hunan University of Science and Technology, showing a deep black color without further treatment. The photo was added to the supplemental material Figure S1. The raw water quality parameters were chemical oxygen demand 200–350 mg/L, ammonia 31–47.6 mg/L, total dissolved solids 312–381 mg/L, total N 57–211 mg/L, total P 2.75–5.2 mg/L, phosphate 2.75–3.85 mg/L, turbidity 40.2–48.2 mg/L, conductivity 1.06–1.47 S/m and UV254 0.203–0.888 cm^{-1}. Polyester fiber ball was purchased from Henan Lvjie Environmental Protection Co., Ltd, which was pretreated in laboratory using the following procedure: In brief, 500 g of polyester fiber ball was soaked in a 10% alkali solution at 30 °C for 4 h and then washed three times with ultra-pure water. Finally, the modified polyester fiber balls were dried in an oven at 60 °C to obtain the modified fiber balls. All aqueous solutions and standard solutions used in this study were prepared with deionized water. The glassware and other labware were

acid-washed, rinsed thoroughly with water, and dried prior to use.

Treatment system

The experiment was carried out with a 25 L experimental system, as shown in Figure S2, and includes the experimental rig (Figure S2a) and a flow chart of experimentation (Figure S2b). The experiments were carried out with a predetermined mass of adsorbent fiber balls and a water sample collected from an impacted water body. The maximum capacity of the fiber ball in the experiments was 20 g, derived from previous study to provide the adsorption best performance. The water sample was injected through an inlet. The solution pH was adjusted using 0.5 mol/L NaOH and 0.5 mol/L HCl. Gravity compression of the fiber balls was achieved through a porous steel plate. The experiment was carried out using a circulation aeration reaction system, which included an open water inlet and outlet, as well as an air pump, to ensure that the water sample had sufficient contact with the fiber balls. Under aeration conditions, the solution circulated from the water inlet to the tank and then to the water outlet. Water quality was measured periodically by samples taken from the tank.

Analytical method

Water analysis

Because ammonia is a determining factor of water quality of back and odorous water. Therefore, the primary parameters, including ammonia, total N, total P and phosphate, were tested using a UV–vis spectrophotometer (TU-1901) in this study. Ammonia and total P were measured according to Chinese Standards HJ 535–2009 and GB11893-89, respectively. Total N concentration was analyzed using HACH spectrophotometer (HACH DR2800, USA) and HACH test kit. Total dissolved solids were measured using total dissolved solids analyzer (CT-3061). A portable conductivity meter was used to measure the conductivity. The turbidity of the supernatant was measured with a turbidity meter (HACH2100Q, HACH Company, USA).

Characterization of adsorbent

We measured the Fourier transform infrared spectra on a Nicolet iZ10 Fourier Transform Infrared Spectrophotometer (Thermo Electron Co., USA) with potassium bromide (KBr) as the dispersant used to examine the chemical functional groups in the adsorbents and the electron microscopy images of the product were obtained using a JEOL JSM-6380LV electron microscope. The X-ray diffraction patterns of the samples were characterized on a Rigaku Ultimate IV D/max-r X-ray diffractometer, which was operated at 40 kV and 40 mA with Cu Ka radiation. Specific surface area of the sample was measured using nitrogen sorption isotherms through a standard Brunauer–Emmett–Teller (BET) analysis (Tristar II 3020, Micromeritics Instrument Corporation, and USA). X-ray photoelectron spectroscopy recorded on an ESCALAB 250 X-ray photoelectron spectrometer (Thermo-Scientific Co., Waltham, MA, USA) was used to analyze the distribution of elemental species.

Results and discussion

Evaluation of pollutant removal

Ammonia nitrogen is the key index to evaluate quality of black and odorous water. The ammonia nitrogen range for heavily polluted levels is defined as more than 15 mg/L, while the ammonia nitrogen range for minor pollution is defined as 8–15 mg/L. The treated black and odorous water in this study is heavily polluted as defined, with ammonia nitrogen content exceeding 15 mg/L and dissolved oxygen content less than 0.2 mg/L. The reaction system for removal of ammonia nitrogen was found to be effective, as after 16 H of reaction, the concentration of ammonia nitrogen decreased from 31 to 1.8 mg/L, under alkaline conditions. The experimental investigation studied the effect of the system on removal of ammonia using initial water sample pH of 5.67, 7.67, and 9.67 to treat an initial ammonia nitrogen concentration of 47.6, 45.28, and 31 mg/L, respectively. The reaction time of the unit was 24 h. The ratio of mass of fiber ball (g) to solution volume (L) was 20. The system was continuously aerated and the dissolved oxygen content was 8 mg/L. Figure 1a, d shows the results of residual ammonia in the system.

As illustrated in Fig. 1a, the residual ammonia in the system decreased as reaction time increased, especially for the reaction at pH 9.67. For the next 24 h, the concentration could not be tested. At pH 9.67 for 16 h of reaction time, the ammonia nitrogen at 31 mg/L decreased to 1.8 mg/l, with a 94% removal. At pH 5.67 for 24 h of reaction time, ammonia nitrogen at 47.6 mg/L decreased to 27.85 mg/L, achieving 41% removal. At pH 7.67 for 24 h of reaction time, the ammonia nitrogen at 45.28 mg/L decreased to 18.23 mg/L, achieving 66% removal. A strong alkaline environment such as pH 9.67 was more beneficial for the system to achieve the maximum treatment efficiency because ammonium ions can be converted into ammonia molecules according to the following equation: $NH_3 + H_2O \leftrightarrow NH_4 + OH^-$, equilibrium constant $(K_b) = 10^{-4.7}$ with subsequent volatilization from the water. The treatment accounted for the aeration and adsorption effects of the fiber ball. In contrast, an acidic environment appears to inhibit treatment because the

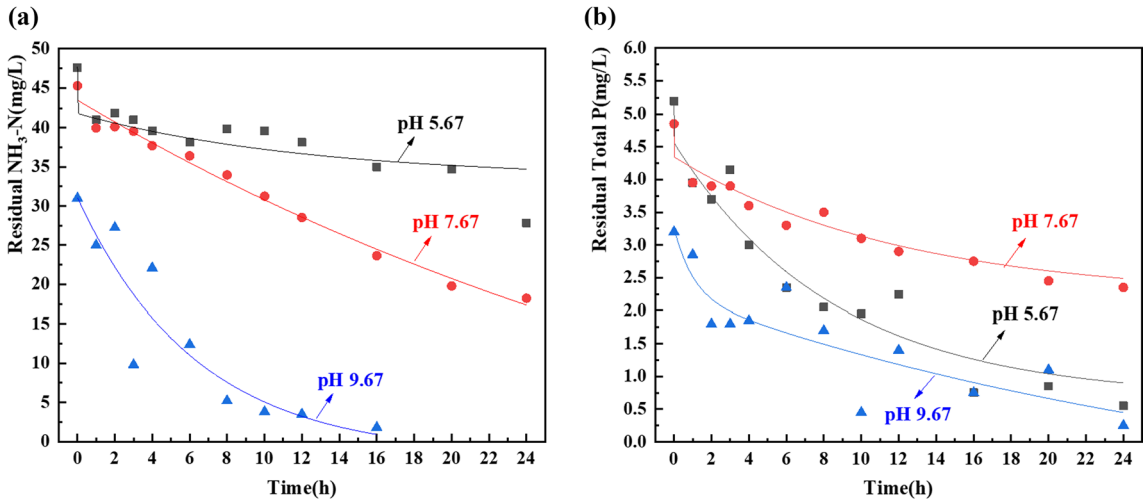

Fig. 1 Time and influence of pH on **a** residual ammonia and **b** residual total P in samples of black-odorous water treated by the fiber ball aeration system. The results show removal could be optimized by considering detailed reaction conditions

majority of ammonia exists in ionic form. The system's ability to remove ammonia ions was then primarily determined by the interface reaction between them and the fiber ball. As a result, ammonium removal was primarily dependent on both aeration and adsorption, and the best operating condition to achieve effective N-removal was by increasing the pH of the water sample. The system effect on removal of total N was identified. The experimental results for a 24 h reaction time with an initial total N concentration of 29 mg/L and a pH of 9.67 are shown in Figure S3. As shown in the figure, the total N concentration could be reduced to 2.1 mg/L for 18 h, with 92.8% removal, indicating good results. Furthermore, as we increased the operational time, the total N concentration was no longer detectable. In general, nitrogen removal was effective.

Total N has not been used as a quality indicator for black-odorous water. It has, however, been listed in other documents as an index to evaluate water quality, in wider catchment studies. The effectiveness of the system in removing total P was also evaluated. New experimental evaluation was conducted with the added mass ratio of fiber ball to solution volume of 20 to treat an initial total P concentration at 3.2 mg/L at pH 9.67 and 5.2 mg/L at pH 5.67, and 4.83 mg/L at pH 7.67. The system was continuously aerated and dissolved oxygen content was 8 mg/L. Figure 1e, h presents the results of the treatment. For 24 h of reaction time, the system had a maximum removal effect (Fig. 1b). The alkaline and acidic environments were both beneficial to the system in terms of total P removal. As illustrated in Fig. 1b, the initial total P concentration at 3.2 mg/L decreased to 0.25 mg/L at pH 9.67, showing the maximum removal efficiency of 92%, and initial total P at 5.2 mg/L decreased to 0.5 mg/L at pH 5.67, showing removal efficiency of 90%. This finding demonstrated that the system was effective for removal of total P, as well as specifically phosphate removal (Figure S4). The difference in the level of P reduction would be attributed to differences in chemical speciation at different treatment pH. At pH = 5.67, phosphorus is dominated by $H_2PO_4^-$ ions; at pH = 7.67, it was dominated by HPO_4^{2-} and $H_2PO_4^-$, and at pH = 9.67, the species was dominated by HPO_4^{2-}. The groups on the surface of the fiber ball were protonated in the acidic environment, possibly to bond to negatively charged dihydrogen phosphate. As the pH increases to 9.67, the HPO_4^{2-} may react with hydroxyl groups on the surface of fiber spheres and become bonded to the surface of the fiber spheres though oxygen binding. As seen from Fig. 1b, it seems that the acidic environment and higher degree alkaline condition were beneficial to the treatment.

Kinetics

The characteristics of the system in the removal of N or P was analyzed by the kinetics model including pseudo-first-order kinetics model (Eq. (1)) and pseudo-second-order kinetics model (Eq. (2) (Simonin 2016), which was evaluated by squared correlation coefficient (R^2).

$$q_t = q_e - q_e/(k_2 q_e t + 1) \tag{1}$$

$$q_t = q_e(1 - \exp(-k_1 t)) \tag{2}$$

where q_t (mg/g) is a sorption capacity of fiber ball corresponding to a specific reaction time (t, min); q_e(mg/g) is an equilibrium sorption capacity; k_1 (min^{-1}) is a

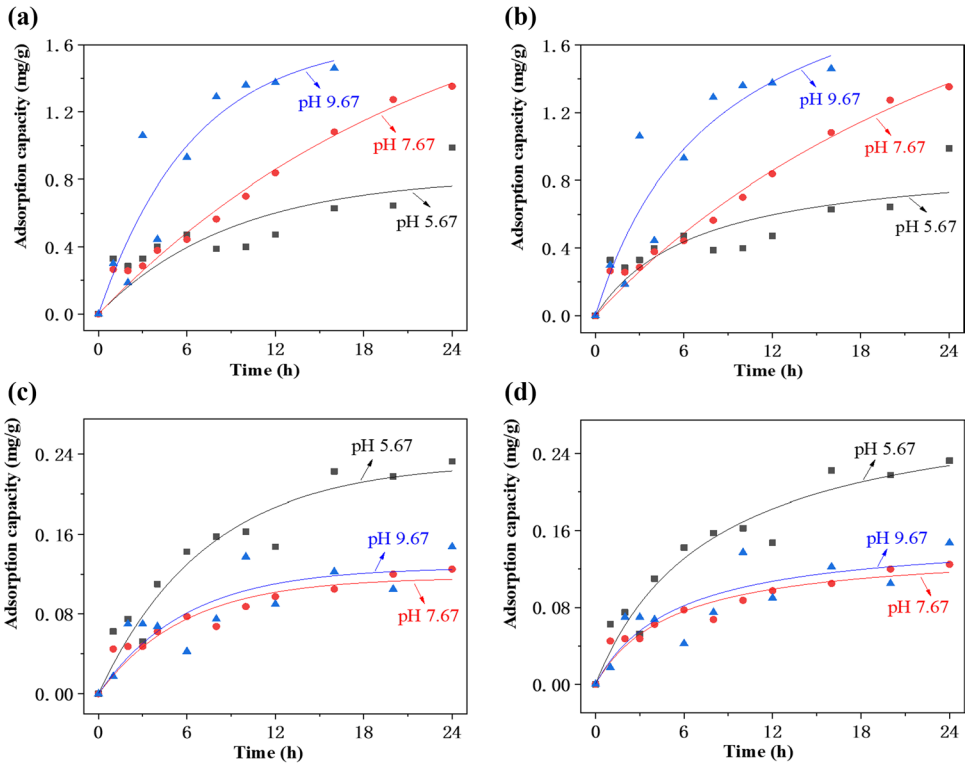

Fig. 2 Reaction kinetics for removal of ammonia and total P from samples of black-odorous waster: **a** pseudo-first-order kinetics for ammonia, **b** pseudo-second-order kinetics for ammonia, **c** pseudo-first-order kinetics for total P and **d** pseudo-second-order kinetics for total P. The solid lines are best fit curves. The results showed that the treatment data could be described by pseudo-first-order kinetics and pseudo-second-order kinetics, which implied both physical and chemical adsorption mechanisms controlled the treatment process

pseudo-first-order sorption rate constant; and k_2 (g/(mg·min)) is a pseudo-second-order sorption rate constant.

Figure 2 presents the plots of ammonia and total P adsorption capacity as a function of time and kinetics curves. Because it was not effective for the removal of ammonia nitrogen in an acidic environment, the result of the best fit was poor, with R^2 only 0.633 for a pseudo-first-order kinetics model and 0.685 for pseudo-second-order kinetic model (Fig. 2a, b). In contrast, there was a strong effect on the removal of N in an alkaline environment, resulting in a good fit for the two kinetic models (Fig. 2a, b). In comparison, the result at pH 7.67 was better than for pH 9.67. It demonstrated that, in addition to adsorption, aeration affected the removal of N, as indicated by a decrease in R^2 at pH 9.67 from 0.97 to 0.84. As seen from the R^2 results, the two kinetic models were similar. Data fitting for total N adsorption at pH 9.67 was consistent with the result for ammonia (Fig. 2a, b). This implies that environmental conditions affecting total N, and ammonia removal are similar. In addition, the system also had a good impact on the removal of total N (see Figure S5a-b), demonstrating effective nitrogen removal. In contrast to N adsorption, data fit for total P adsorption at pH 5.67 was better than for the other pH, with R^2 as high as 0.98. (Fig. 2c, d). The system was effective for removal of both phosphate and total P, with a best fit result at pH 5.67 (Table S1 and S2, Fig. 2c, d and Figure S5c, d) with relatively weaker fit under alkaline conditions. The pseudo-first-order or pseudo-second-order kinetics models were poor for adsorption under alkaline conditions. The pseudo-first-order and pseudo-second-order kinetic models produced similar R^2 values. For effective fitting, there will be physical or chemical interaction between N or P and surface of fiber ball, resulting in effective treatment. As seen from the kinetic parameters from the pseudo-first-order and pseudo-second-order kinetic models, the adsorption capacity for ammonia reached 2.14 and 3.47 mg/g, respectively, and that of total P was up to 0.23 and 0.43 mg/L, respectively.

Characterization

We used Fourier transform infrared spectroscopy (FT-IR), X-ray diffraction (XRD), and X-ray photoelectron spectroscopy (XPS) to characterize those fiber ball materials before and after adsorption to reveal possible mechanisms of aeration and adsorption system to remove nitrogen and phosphorous. The results are shown in Fig. 3. Although the specific surface area of the adsorbent is lower, there are many chemical groups that may lead to the adsorption system effectively removing pollutants. Fourier transform infrared spectroscopy data were used to investigate the potential chemical interaction between fiber balls and water pollutants. The Fourier transform infrared spectra were used to evaluate the potential adsorption mechanisms by revealing the chemical functional groups of the fiber ball and their variations before and after adsorption. Fourier transform infrared spectroscopy scan of fiber ball at 4000–400 cm^{-1} is presented in

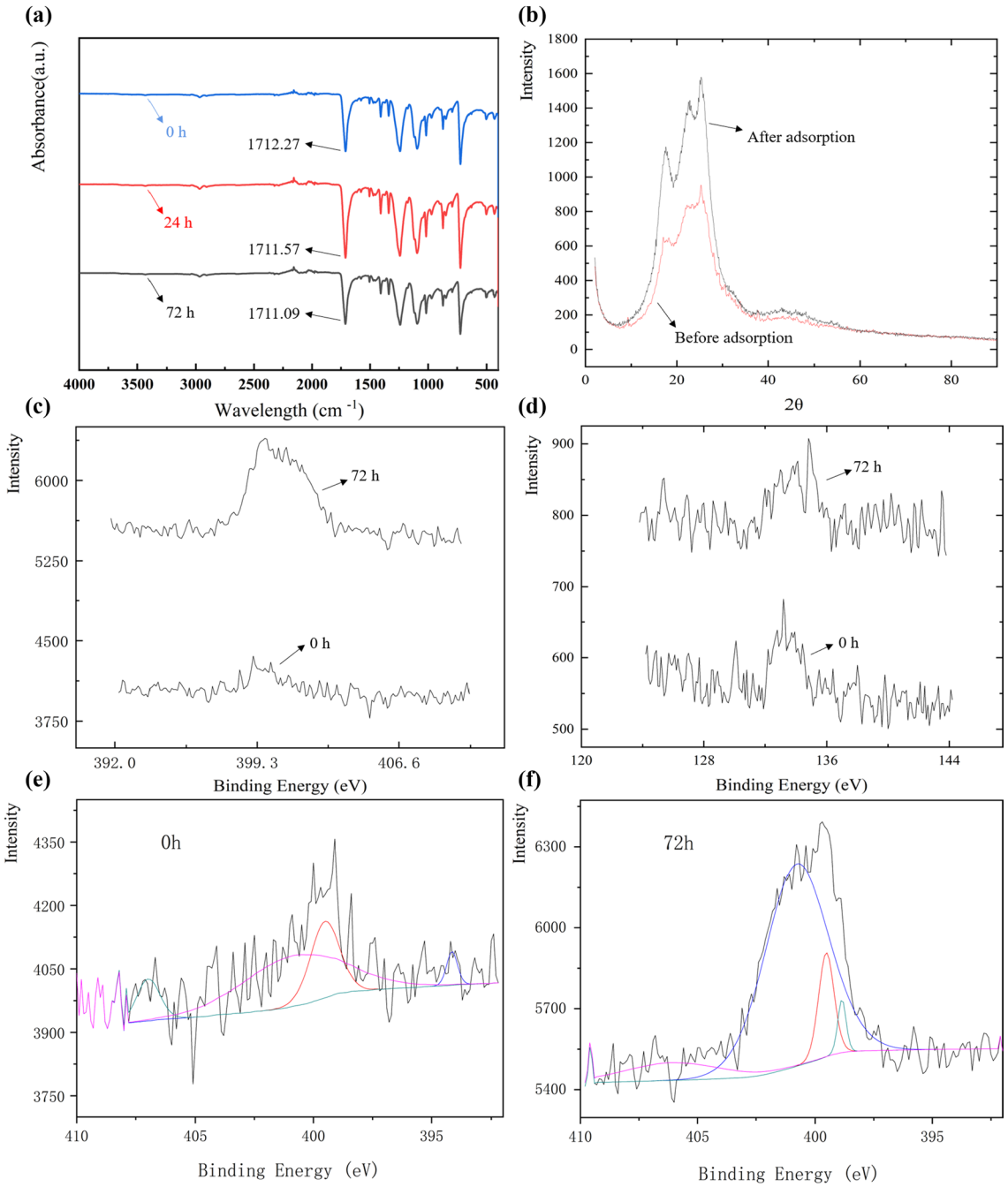

Fig. 3 A comparison of spectral analysis of fiber balls **a** Fourier transform infrared spectroscopy, **b** X-ray diffraction, **c** N 1*s* and **d** P 2p X-ray photoelectron spectrum, showing before and after adsorption. Results of N1s fitting composition diagram for **e** 0 h and **f** 72 h, respectively. These results support the kinetic data indicating physicochemical control on the removal of N and P

Fig. 3a. The chemical groups of the adsorbent before adsorption and after adsorption for 24 and 72 h were investigated. The potential functional groups (Rodrigues et al. 2015) included: –OH adsorption peaks at 3430.89 cm^{-1} for 0 h, 3430.5 cm^{-1} for 24 h and 3430.39 cm^{-1} for 72 h (Kong et al., 1999) and C=O adsorption peaks at 1712.27 cm^{-1} for 0 h, 1711.57 cm^{-1} for 24 h, and 1711.09 cm^{-1} for 72 h (Li, 2017). After adsorption, these adsorption peaks' positions had a slight blue shift. The absorption peaks at 3000–2800 cm^{-1} for 0 h (2968.25 cm^{-1} and 2907.00 cm^{-1}) are attributed to the N–H groups (Chai et al. 2010; Zhang et al. 2011), after which the adsorption shifted to higher wavelength number (2968.6 cm^{-1} and 2907.44 cm^{-1} for 24 h; 2967.9 cm^{-1} and 2907.38 cm^{-1} for 72 h). The adsorption peaks at

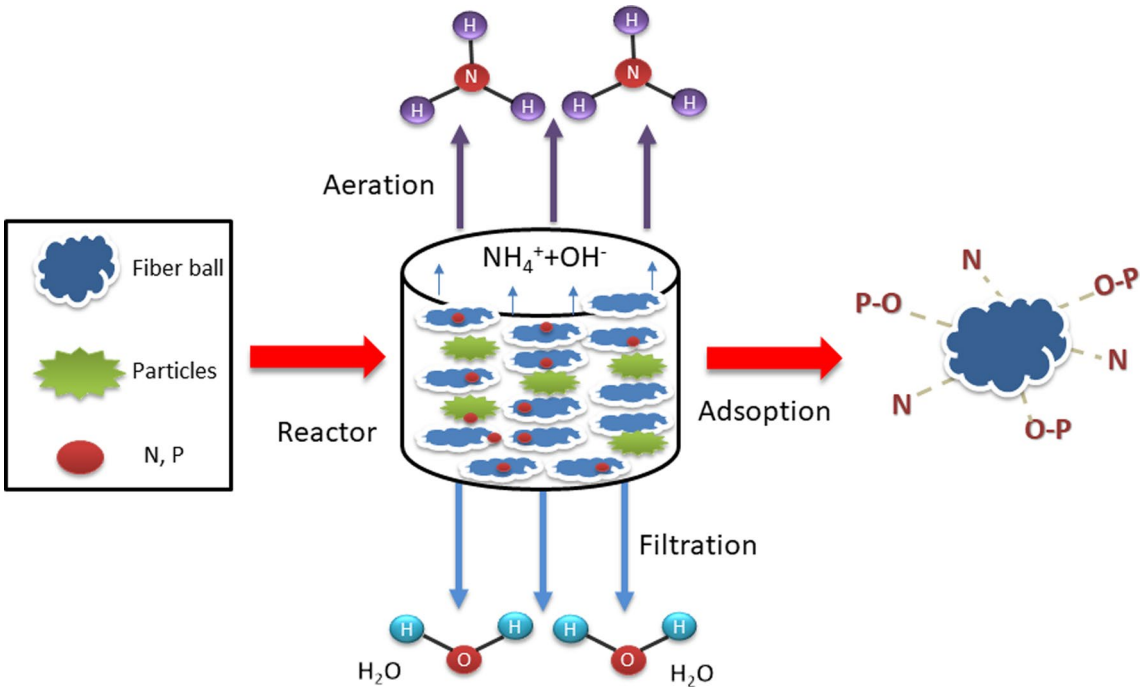

Fig. 4 Nitrogen and phosphorus removal by the gravity compression and aeration system

1338.54 cm^{-1} for 0 h corresponding to the O–H chemical groups (Wang et al., 2015) had a shift toward higher wavelength number at 1339.12 cm^{-1} for 24 h and 1339.27 cm^{-1} for 72 h. The adsorption peaks at 1176.23 cm^{-1} for 0 h corresponding to the C–O chemical groups of the sodium phthalate had a shift toward lower wavelength number at 1175.25 cm^{-1} for 24 h and 1175.1 cm^{-1} for 72 h (Hong et al., 2002). The shift is presumably due to the nitrogen and phosphorus absorption onto fiber ball, forming C–N functional groups and P=O functional groups due to combination of sodium terephthalate and amino groups, while P=O was from various forms of phosphorus.

The X-ray diffraction data confirmed the phase composition of the fiber ball before and after adsorption. The results of their patterns are shown in Fig. 3b. As seen from fiber ball shown in Fig. 3b, three typical peaks were included at 17.36°, 21.3°, and 25.2° belonging to (010), (111) and (100), which were characteristic amorphous peaks of polyester fiber (Dai et al, 2002, Huang et al, 1982). The possibility of a chemical interaction between the fiber ball and the pollutant existed because, following adsorption, the peak position shifted to higher values and the intensity increased. The primary cause of these changes would be the exchange of elements in the fiber ball with other external substances. In detail, the X-ray diffraction data confirmed that some crystalline phases existed in fiber ball after adsorption, such as calcium hydrogen phosphate hydrate (Ca $(HPO_3)_2(H_20)$, pyrazine ($C_4H_4N_2$), 2, 5-Dipyridine-2-yl-pyrazine-N-oxide ($C_{14}H10N_4O_2$), and a similar NH_4-containing crystal structure (Triammonium hydrogen bis (selenate (VI)) ($(NH_4)_3H(SeO_4)_2$)) (Figure S6). These findings demonstrated that the addition of an aeration adsorption system was far more effective at increasing nitrogen and phosphorous adsorption by the fiber balls.

The nature of pollutants removed was revealed through X-ray photoelectron spectrum result, is shown in Fig. 3 and shows the spectral results of N1*s* and P 2*p* before and after water treatment. It can be seen from the figure that the shape of the spectrum changed before and after the treatment of wastewater. Figure 3c, e and f shows the spectral of the N 1*s*. The N 1*s* peak occurred around 394 eV, of which intensity increased significantly after treatment. In Fig. 3d, the binding energy of the P 2*p* peak was 133 eV, which shifted to 134 eV after treatment, and is attributed to the interaction between P and pollutants. The binding energies corresponding to the peaks of four fitting components of N 1*s* occurred at: 394.14 eV accounting for 4.22%; 399.51 eV accounting for 25.67%; 400.84 eV accounting for 59.13%; and 406.98 eV accounting for 10.98%. At 72 h, the values changed to: 398.86 eV accounting for 2.68%; 399.53 eV accounting for 9.81%; 400.78 eV accounting for 77.09%; and 406.10 eV accounting for 10.42%. Compounds such as imines, heterocyclic C=N, and aromatic amines may be most adsorbed on the surface of the fiber ball, owing to their binding energy of 400.78 eV (Abe et al. 2005; Hua et al. 2018).

Based on the characteristics discussed above, three effective functions were involved in the removal mechanisms: filtration, adsorption and aeration. A simple scheme shown in Fig. 4 was used to describe the whole treatment process. We were able to pioneer the removal of pollutants and establish a promising technology for the remediation of contaminated and odorous water. A detailed review of application of the system is also shown in supplementary materials (see Text S1).

Conclusion

In this study, we developed a new aeration adsorption system utilizing fiber balls for the treatment of the nitrogen and phosphorus content black and odorous water. With the continued economic development across society, the associated emission of nitrogen and phosphorus into the natural environment results in a faster and more extensive deterioration of water quality. The increasing occurrence of black-odorous water is one of the visible signs. It promotes the growth of bacteria and production of harmful gases, both of which are detrimental to human health and the wider environment. We present a new system based on a cost-effective approach. Fiber ball materials are widely used in water treatment, and there is considerable scope for their further improvement; however, they are rarely used in the treatment of wastewater resources. We have demonstrated an effective treatment for N/P removal through adsorption and aeration. The inclusion of aeration increases the degree of nitrogen removal. Ammoniacal nitrogen can be completely removed, and phosphorus can be effectively removed up to 90%. In the case of severely polluted rivers, wider catchments and domestic wastewater, this system has considerable potential application with great opportunity due the low cost with high treatment efficiency.

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Author contributions Prof. ASH and Prof. GZ lead experimental activity and research direction and developed the manuscript. SZ, JC, HL and Dr. PW conducted experimental work under academic direction. Dr. ZZ and Prof. GF provided instruments for analysis of data.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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