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Using powdered activated carbon to enhance atrazine removal in an anoxic/oxic (A/O) process

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ABSTRACT

Laboratory-scale experiments based on an anoxic/oxic (A/O) wastewater treatment process were conducted to investigate the effects of an spill of the emergent atrazine pollutant on the operation of wastewater treatment plant, as well as the feasibility of using powdered activated carbon (PAC) to improve the performance of the treatment system after the accident. Atrazine was added with a concentration varying from 2.5 to 20 mg/L at different experimental stages. The results showed that the performance of the A/O system, in terms of chemical oxygen demand (COD) and ammonia nitrogen (NH_4^+-N) removal, was negatively affected by the presence of atrazine. However, the system could recover a more than 80% of COD and 70% of NH_4^+-N removal within 6 d under an atrazine concentration of less than 10 mg/L. A 20 mg/L atrazine concentration showed irreversible effects on the treatment performance. PAC addition was found to be able to greatly improve the performance of the system at the atrazine concentration of 20 mg/L, as indicated by the oxygen uptake rate of microorganisms. The activity of the bacteria was proportional to the PAC dosage under experimental conditions, and a 300 mg/L of PAC dose was recommended.

Keywords: Emergent pollution accident; Atrazine; Anoxic/oxic (A/O) process; Powdered activated carbon (PAC); Oxygen uptake rate (OUR)

1. Introduction

Atrazine is synthetic organic herbicide with a triazine group in its molecular structure, and it is widely used in the agricultural and forestry industries. Around 70,000–90,000 tons of atrazine are applied annually across the world [1]. As a result, this herbicide is widespread in surface and ground waters [2,3] and is frequently detected in wastewater. As a type of endocrine disrupting chemical (EDC), atrazine is targeted for strict control in water treatment [4]. One of the most important concerns is the influence of atrazine on the reproduction of aquatic flora and fauna, which in turn impacts the community structure [5].

Methods for removal of atrazine from surface waters include advanced oxidation processes (AOPs) [6–8], biodegradation [9–11], and bioaugmentation [12–15]. However, these results always present only a modest efficiency of atrazine degradation due to

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complicated operation requirements, prolonged operation period, as well as high energy consumption and the need for additional chemicals in the treatment process. Furthermore, most of the methods are confined to decontamination of drinking water where the concentration of atrazine is typically low. For these reasons, the aforementioned methods are generally only applied to degrade atrazine in laboratory conditions for research purposes.

With rapid economic development and the stimulation of agricultural and industrial activities across China, pollutant spills have frequently occurred, resulting in the introduction of large amounts of industrial contaminants into wastewaters [16,17]. Consequently, the composition of wastewater reaching wastewater treatment plants (WWTPs) has become increasingly complicated and some components are not effectively removed by conventional treatment processes [12]. Therefore, it is considered a high priority to determine efficient treatment technologies that can be rapidly applied and efficiently operated to solve the problems caused by emergent pollutant spills in WWTPs.

Activated carbon is extensively used in various air purification and water treatment processes due to its outstanding characteristics of having enormous pore surface area and unparalleled surface chemistry. It is generally accepted that the application of activated carbon is one of the most effective techniques for removing organic matter from wastewater [18,19]. It has been reported that more than 98% of chemical oxygen demand (COD) and BOD can be removed at concentrations of 22 and 12 g/L, respectively, when a 4 g/100 mL PAC dose was used to treat coffee processing wastewater [20]. Furthermore, activated carbon has been proven to be one of the most effective methods for pesticide removal in drinking water treatment [21]. With larger specific surface area, the use of powdered activated carbon (PAC) showed better adsorption performance than granular activated carbon (GAC). Therefore, using PAC to enhance the removal of atrazine in the existing WWTPs may be an appropriate option for rapid treatment in case of emergent pollutant spills.

Previous research has shown that the presence of 15 mg/L atrazine had little effect on COD removal when an anaerobic/anoxic/aerobic (A^2/O) process was used to treat sanitary wastewater [22]. There is little information available on the enhancement of atrazine removal in a wastewater treatment process by activated carbon. This study was conducted to investigate the performance of a laboratory-scale anoxic/aerobic (A/O) process treating sanitary wastewater which has been subjected to shock loading of atrazine. The shock loading was to simulate a pollution spill into the system, and the removal performance of COD, ammonia, and atrazine was analyzed. The feasibility of using PAC to enhance atrazine removal and the effect of PAC on other pollutants were investigated.

2. Materials and methods

2.1. Reactor setup

A plug-flow A/O reactor made of plexiglass with a working volume of 52.5 L was used in the experiment. The experimental setup is shown in Fig. 1. The reactor consisted of six connected compartments, including two anoxic compartments followed by four aerobic compartments. The volumetric ratio of the anoxic zone to the aerobic zone was 1:2. All of the six compartments were connected together as a plug-flow A/O process. Electric stirrers were used in the anoxic compartments to keep the sludge suspended.

2.2. Process operating conditions

The feeding sludge used in the experiments was taken from a local WWTP in Harbin, China. Real sewage was used as the influent in the experiments. The COD concentration of the influent was between 350 and 450 mg/L, and the NH_4^+-N concentration was between 35 and 46 mg/L. The influent rate was 12 L/d, resulting in a hydraulic retention time (HRT) of 8 h. The sludge recycling ratio and the mixed liquor recycling ratio were 80 and 200%, respectively. The sludge retention time (SRT) was 18 d. The mixed liquor suspended solids (MLSS) concentration in the system was kept at 3,500–4,000 mg/L, and pH was in the range of 7.3–7.8. The process was operated at a room temperature of 20 ± 1°C, and the dissolved oxygen (DO) concentration in the aerobic compartments varied from 0.5 to 3.5 mg/L.

Coal-based PAC with an average particle size of between 80 and 160 μm was used in the experiment. The PAC had a specific surface area of 910 m^2/g, a micro-pore volume of 0.25 cm^3/g, and an iodine value of 850 mg/g. The PAC material passing the 100 mesh sieve but retained by a 200 mesh sieve was pre-dried at 101–106°C before being used as an adsorbent in the experiment.

The continuous operation experiments were divided into two stages. In the first stage, there were five run periods (Run I to Run V). Each run was defined based on the influent atrazine concentration. Run I to Run V were conducted to investigate the system performance when subjected to an atrazine shock. During the transition phase between each

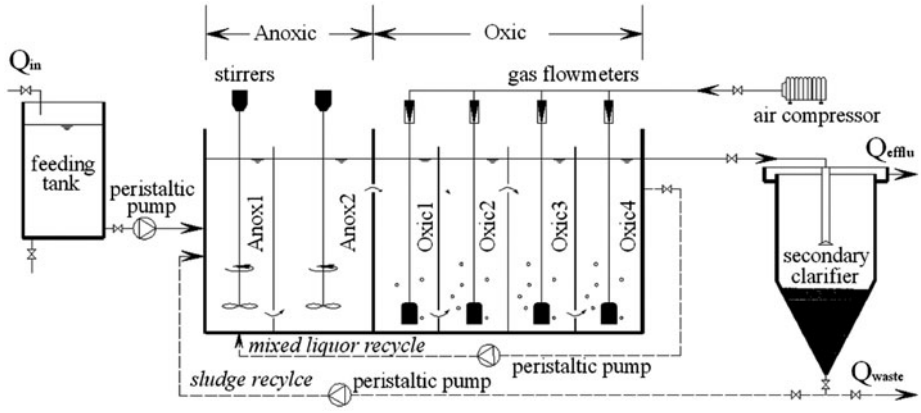

Fig. 1. Schematic diagram of the A/O process used in the experiment.

running period, 5 d of stable operation with simple sanitary wastewater were required to ensure the system performance had recovered to that in Run I, before the next run began. In the second stage, the operation was divided into 6 periods (Period I to Period VI). Each period was defined based on the doses of PAC and atrazine added, and each was conducted to investigate the effect of different PAC doses on pollutant removal at high atrazine concentration. The effect of PAC doses on the activity of microorganisms in terms of oxygen uptake rate (OUR) before and after a high concentration atrazine addition was also analyzed. The experimental conditions of all the operation periods are summarized in Table 1.

2.3. Analytical procedure

All samples were filtered through 0.45 μm filters prior to analysis. The COD, NH_4^+-N and total nitrogen concentration in the influent and the effluent were measured according to standard methods [23]. DO and pH were measured using a Multi 340i DO meter (WTW, Germany). Atrazine was measured in accordance with the description reported by Ma and Graham [6]. The OUR analysis of microbial activity was conducted in accordance with the description in Wang et al.'s work [24].

3. Results and Discussion

3.1. Effect of different atrazine loading on pollutant removal without PAC

The effect of different concentrations of atrazine on the performance of the A/O process was investigated in the experiment. The profiles of COD, NH_4^+-N and atrazine removal in different operation periods are plotted in Fig. 2.

The COD removal achieved during Run I (without atrazine) was over 95%. This represents a typical performance of the organics degradation using an A/O system. With the addition of different concentrations of atrazine, the COD removal efficiency dropped from 95% to below 70% on the day following the addition during Runs II, III, and IV, however, within the following 24 h, the COD removal efficiency recovered to 90.7, 84.1, and 83.6%, respectively. On the sixth day after the atrazine addition, the COD removal efficiency of Run II to Run IV recovered to the level measured in Run I.

This showed that the influence of atrazine on COD removal in the A/O system was temporary when atrazine concentration was less than 10 mg/L in the experiments. Wu et al. [22] presented a similar result, indicating that the removal of COD still remained at high levels when an atrazine

Table 1
Experimental conditions of each period during Stage One and Stage Two

	Stage One					Stage Two					
	Run I	Run II	Run III	Run IV	Run V	Test I	Test II	Test III	Test IV	Test V	Test VI
Atrazine concentration (mg/L)	0	2.5	5	10	20	0	0	0	20	20	20
PAC dosage (mg/L)	0	0	0	0	0	200	300	400	200	300	400

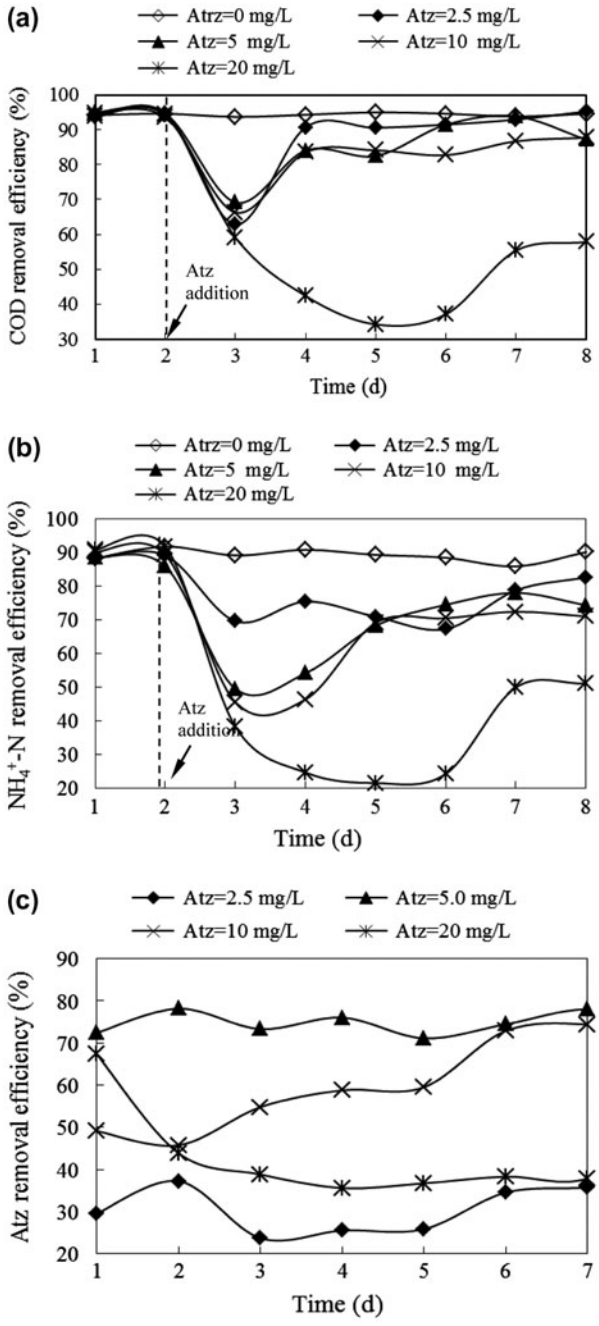

Fig. 2. Effect of different atrazine concentrations on removal of (a) COD, (b) NH_4^+-N, and (c) Atrazine.

concentration lower than 15 mg/L was fed into an A^2O system. This showed that the conventional activated sludge process has a relatively strong ability to remove organic matter. This can be attributed to the fact that in the activated sludge system, there was a large microbial diversity and a considerable amount of heterotrophs, which could resist the toxic substance to some degree.

However, when atrazine concentration in the influent was increased to 20 mg/L in Run V, the COD removal efficiency dropped to 59% on the day following the addition, it kept decreasing and reached the lowest value of 34% on the fifth day. The COD removal then started to increase and only recovered to 57.8% on the seventh day. This showed that atrazine can have a prolonged toxic effect on a system at higher concentration.

More than 85% of NH_4^+-N removal could be achieved during Run I. The removal efficiency dropped on the day following the addition during Runs II, III, and IV, and the NH_4^+-N removal efficiency recovered to 75.4, 54.1, and 46.3%, respectively, in the following 24 h. It seemed that compared to the COD removal, NH_4^+-N removal was more easily affected by atrazine. It took longer time (more than 6 d) for the NH_4^+-N removal to recover than for COD removal in Runs II, III, and IV. The worst situation occurred with an atrazine concentration of 20 mg/L in Run V, where the removal efficiency of NH_4^+-N dropped rapidly to 38.1% on the day after the addition, and fell to the lowest value of 21.4% on the fifth day. Nitrifying bacteria, which transform ammonia to nitrite or nitrate, are responsible for the removal of ammonia nitrogen in the activated sludge system. However, these bacteria belong to the class of autotrophic microbes and are more sensitive to poisonous substances. Moreover, the metabolites of atrazine contained amino groups, which may adversely affect the ammonia removal in the process. Boundy-Mills et al. [25] have observed that under aerobic conditions, *Pseudomonas* sp. strain ADP could rapidly metabolize atrazine to CO_2, NH_4^+-N, and Cl^-. Similar results were reported by Ghosh and Philip [9], who showed that when the aromatic ring on an atrazine molecule was broken (Fig. 3), amino groups could be converted into ammonia nitrogen to provide an additional nitrogen source to the microorganisms.

Besides the effects of atrazine on the performance of the A/O system, the removal of atrazine by biodegradation was also examined in the treatment system. Previous research by Wu et al. [22] on the

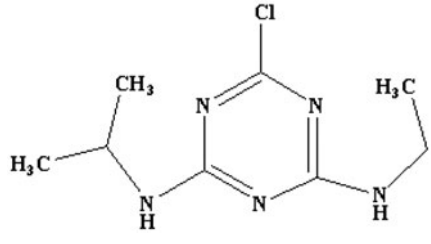

Fig. 3. Chemical structure of atrazine.

effect of short-term addition of atrazine on an A^2/O system indicated that there were no bacteria with the ability to biodegrade atrazine in the sludge of a conventional nutrient removal processes. The results in Fig. 2 show that the residual concentration of atrazine in the effluent of our experiment was proportional to the initial concentration in the influent. The maximum average atrazine removal efficiency of 74.2% was obtained when the atrazine concentration in the influent was 5 mg/L (Run III). The results indicated that atrazine can be partly biodegraded in the A/O treatment system without acclimation. Furthermore, the biodegradation was improved after a short-term exposure to a low level of atrazine concentration in Run II. However, at a high concentration, the microbial cells would irreparably suffer from atrazine and its biodegradation or biotransformation would be also inhibited.

The atrazine removal efficiency during Run IV firstly decreased and then slowly recovered to 72.8%. This verified that the inhibition of atrazine to the bacteria was reversible in this experiment at a concentration lower than 10 mg/L. The removal efficiency of atrazine during Run V decreased throughout the operation, indicating that an atrazine concentration of 20 mg/L was beyond the tolerance of the microbes. Overall, the results indicated that the bacteria in a conventional nutrient removal system can produce the inducible enzyme which acts as catalyst in the biochemical reaction required to degrade atrazine, especially after a short-term acclimation.

3.2. Effect of PAC doses on pollutant removal at high atrazine concentration

Since 20 mg/L atrazine showed an obvious inhibition to microorganisms in this experiment, PAC was added into the system in an attempt to improve the performance of the A/O system. The effect of PAC doses on the performance of the system in terms of COD, ammonia, and atrazine removal is shown in Fig. 4.

The PAC remarkably improved the performance of the A/O system when atrazine concentration in the influent was 20 mg/L. With a 200 mg/L PAC dose, the COD and ammonia removal rates on the fourth day were 58.3 and 33.1%, respectively. These values were higher than those obtained on the fourth day without PAC, when COD and ammonia removal rates were 34.2 and 21.4%, respectively (shown in Fig. 4(a), and (b)). Moreover, dosing of 200 mg/L PAC contributed to the recovery of the COD and NH_4^+-N removal efficiencies, which reached more than 85 and

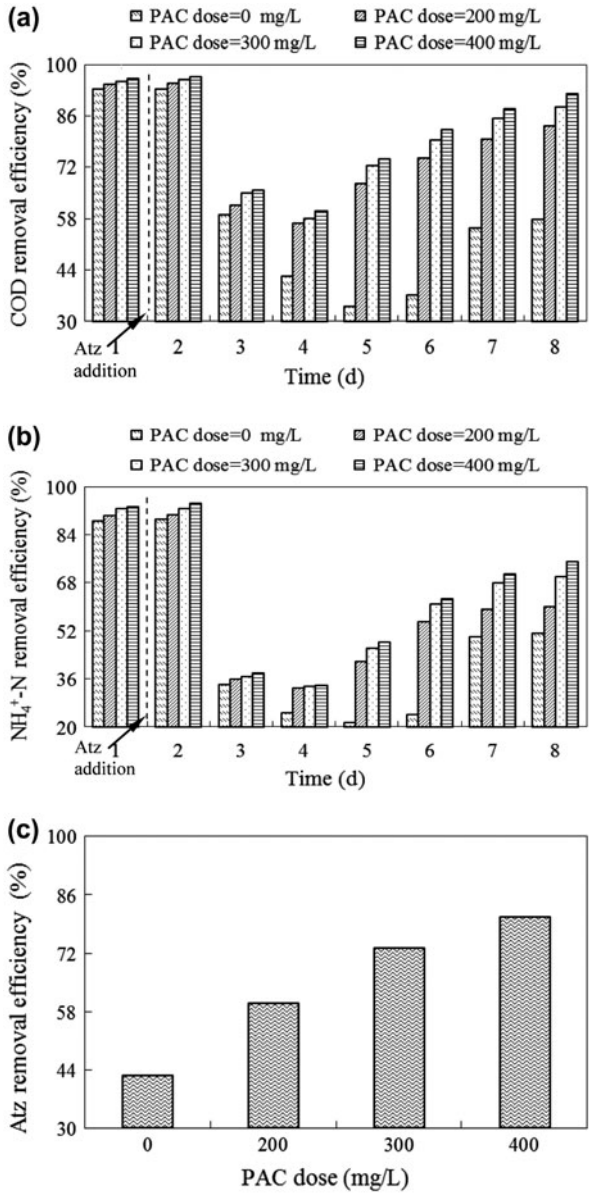

Fig. 4. Effect of PAC dose on removal of (a) COD, (b) NH_4^+-N, and (c) atrazine with a shock load of atrazine equal to 20 mg/L.

60%, respectively, at the end of each operation phase. The removal efficiency increased with the increase in the dosage of PAC. Atrazine removal can also be improved by the PAC addition, as shown in Fig. 4(c). The average removal efficiency of atrazine in the system increased from 42.6% without PAC to 60.1% with 200 mg/L PAC, and the maximum average removal efficiency of atrazine was 80.6% when 400 mg/L of PAC was added. The results indicated that adsorption on PAC had played an important role in atrazine removal [26]. This was attributed to an increase in the

adsorbent concentration, which increased the available surface area and adsorption sites [27]. It is readily understood that the number of available adsorption sites increases with the increase of the adsorbent dosage, consequently leading to an increase in the amount of adsorbed atrazine. However, the relationship between the PAC addition and atrazine removal was not linear, an increasing addition of PAC led to a reduction in the improvement of atrazine removal. For example, as the PAC dose increased from 200 to 300 mg/L, there was a 17.5% increase in the level of atrazine removal. However, as the PAC dosage increased from 300 to 400 mg/L, the additional atrazine removal was just 7.0%. Based on these findings, the optimum PAC dosage was considered to be 300 mg/L under the experimental conditions.

3.3. Effect of PAC on OUR of microorganisms

The effect of PAC on the OUR of the activated sludge in the A/O system was also investigated. The PAC dosage varied from 200 to 400 mg/L, the OUR without atrazine and with a 20 mg/L atrazine loading was investigated. The results are presented in Fig. 5.

In the absence of atrazine ("Atz-0" in the *X* axis in Fig. 5), increase in PAC dose resulted in a decrease in the OUR of total bacteria (TB). The OUR decreased from 49.3 to 42.3 $mgO_2/gVSS/h$ (VSS, volatile suspended solids), when PAC dosage increased from 200 to 400 mg/L. This was mainly caused by the decrease in the OUR of nitrifying bacteria (NB). The PAC addition had no effect on the OUR of heterotrophic bacteria (HB). It is well known that NB are very susceptible to environmental conditions, and it is postulated that the NB growth was restricted due to the substrate and DO concentration [28]. Baldauf [29] reported that acid and alkaline compounds as well as DO in solution could be adsorbed onto activated carbon. Therefore, the decrease of ammonia nitrogen and DO concentrations in the system after PAC addition may be responsible for the decrease in the OUR of NB.

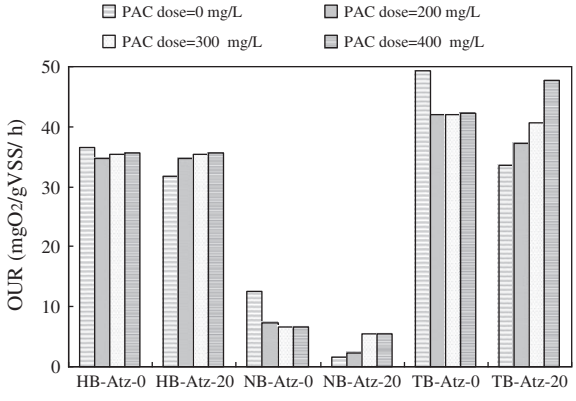

Fig. 5. Effect of PAC dosage on the OUR of microbes at 0 and 20 mg/L of atrazine load. HB: carbon oxidizing bacteria; NB: nitrobacteria; TB: total bacteria; Atz: atrazine; Atz-0: 0 mg/L of atrazine; Atz-20: 20 mg/L atrazine.

In the presence of atrazine at 20 mg/L, the OUR of NB dropped even lower with the addition of PAC. When the PAC dose was 200, 300, and 400 mg/L, with no atrazine, the OUR of NB was 7.3, 6.6, and 6.7 $mgO_2/gVSS/h$, respectively. When atrazine was present at 20 mg/L, the OUR of NB dropped to 2.4, 5.4, and 5.5 $mgO_2/gVSS/h$, respectively. This dramatic reduction contributed to the decrease of the OUR of TB in the system; however, the impact on the HB from atrazine was limited (shown in Fig. 5). Wu et al. [22] reported a negative effect on the activity of autotrophic bacteria from a short-term atrazine addition. The effect was overcome in that experiment by a prolonged oxidation time and raised DO concentration. However, in practice, the shock of contaminants may last for a few days in an emergent situation, which makes this effect potentially fatal to the treatment system. Fortunately, according to the results shown here, the addition of PAC to the treatment system can increase the OUR of NB from 1.7 $mgO_2/gVSS/h$ (atrazine 20 mg/L, no PAC) to 5.5 $mgO_2/gVSS/h$ with 400 mg/L PAC. Aktaş and Ceçen [30] found that addition of PAC favored the function of NB in an activated sludge treatment system treating landfill leachate. They pointed out that PAC adsorbed inhibitory organic matter and alleviated the negative effect of leachate constituents on nitrification. This research showed similar results, PAC adsorbed the high concentration of atrazine in the system and thus regulated the concentration of atrazine to which microorganisms were exposed. Moreover, it was reported that during the course of PAC activation, the aromatic functional group usually formed [31], and atrazine molecules tended to be incorporated into the structure of the organic matter [32]. Therefore, with an increase in PAC dose, the concentration of free atrazine in the system would be expected to fall, facilitating the recovery of NB activity.

4. Conclusions

This study simulated the effect of atrazine shocks from emergent pollutant spill events on WWTPs with an A/O process. It showed that atrazine concentrations lower than 10 mg/L can cause a deterioration in COD and ammonia removal of the treatment system,

but this deterioration can be recovered within 6 d. However, an atrazine concentration of 20 mg/L led to an irreversible effect on COD and ammonia treatment. PAC addition was found to be able to greatly improve the performance of the system subjected to a high (20 mg/L) atrazine concentration, which suggests that PAC addition in a conventional activated sludge system is an effective technique to reduce the negative effects caused by the presence of atrazine in wastewater and may improve the performance of WWTPs exposed to a shock load of atrazine following an emergent pollutant spill.

Acknowledgements

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