Using PAC-modified clays to control black-bloom-induced black suspended matter in Lake Taihu: deposition and resuspension of black matter/clay flocs

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ABSTRACT

Flocculation using modified clays is a technique widely applied in the management of harmful algal blooms (HABs). Polyaluminum chloride (PAC) modified clay is an efficient flocculating agent in HAB control; however its effectiveness in black bloom management is still largely unknown. In the present study, PAC-modified clay was used to flocculate a black bloom under simulated flows. The deposition and resuspension of the black matter/clay flocs and the impact of the spreading of quartz sand to the flocs were quantitatively studied. The results showed that a dosage of 1.8 g/L PAC-modified clay (0.8 g/L PAC and 1 g/L diatomite) could reduce turbidity by more than 90% in 1 h. The resuspension of flocs could be generated by a threshold bed shear stress of 0.045 N/m². The addition of quartz sand inhibited the resuspension of flocs. We suggest that quartz sand can be used to effectively inhibit floc resuspension caused by waves and flow currents as the subsequent treatment of black bloom flocculation in Lake Taihu.

Key words | black bloom, modified clay, polyaluminum chloride, simulated flow experiments

INTRODUCTION

The Lake Taihu drinking water crisis in 2007, caused by a cyanobacteria-induced black bloom, attracted worldwide attention (Qin et al. 2010). Black bloom events take place in eutrophicated areas of lakes where large amounts of decaying algal biomass and benthic sludge organic matter have accumulated, heavily consuming oxygen (Lu & Ma 2009). The dissolved oxygen (DO) levels were depleted, and as a result of extensive anaerobic reactions, lots of nutrients (such as ammonia nitrogen and phosphate) were released into the water body, generating a strong malodor (Yang et al. 2008; Lu & Ma 2010). Similar events have been reported for other lakes, mainly in the deeper areas (Duval & Ludlam 2001; Pucciarelli et al. 2008).

Flocculation using modified clay has been proven an effective method of controlling harmful algal blooms (HABs) (Anderson 1997; Beaulieu et al. 2005; Li & Pan 2013). Pan and co-workers found that local soil modified with chitosan is effective in removing HAB cells, and this technique has been successfully used in the management of HABs in Lake Taihu (Pan et al. 2006a, 2006b, 2011a, 2011b,
Recently, the modified clay flocculation technique was also used as an emergency application to control black bloom (Shang 2015; Huang et al. 2015), based on the suspended particle composition of black matter, the size range of which fully matched with the functional region of modified clays in flocculation procedures (Shen 2011). Chitosan and polyaluminum chloride (PAC) are both superior clay modifiers in controlling HABs; however, PAC usually costs less clay compared with chitosan to remove the same quantity of HABs (Yu et al. 2017; Sengco et al. 2016). Although chitosan-modified clay has been effectively used in black bloom management, the efficacy of PAC-modified clays in controlling black bloom is much less understood. As an inorganic coagulant, PAC increases the chemical affinity of clay surfaces (Pan et al. 2014b). However, the PAC-clay/algae flocs take longer to settle and are resuspended more easily than the clay/algae flocs (Beaulieu et al. 2015). The opposite result showed that the amount of PAC was three times that of chitosan to reduce the same proportion of turbidity of black bloom (Dai et al. 2016). The exact deposition and resuspension behavior of black matter/PAC-modified clay flocs is currently unknown. Flow conditions greatly affect the size of flocs, deposition rate, and removal efficiency (Jackson & Lochmann 1993; Sengco et al. 2001). The removal efficiency of *Heterocapsa triquetra* was found to be lower in rapid flow than in low flow (Archambault et al. 2003), and a high flow condition results in smaller floc sizes and slower deposition (Hill 1998). The aim of the present work was to evaluate the efficacy of PAC-modified clay in flocculating suspended black bloom matter under simulated flow conditions, by quantitatively studying the deposition and resuspension of the black matter/clay flocs and the effect of the addition of quartz sand on resuspended flocs.

**MATERIALS AND METHODS**

**Black bloom simulation**

Cyanobacteria, water, and sediments were sampled from the west bank of Meiliang Bay, Lake Taihu (Figure 1 site A: N31°25′45.14″, E120°7′44.85″, Jiangsu Province of China). The sediments were collected using a self-made shallow-water original surface-sediment sampler backed with a soft cloth (Chinese Invention Patent Application No. 201310184563.6). The water and cyanobacteria were transported to the laboratory within 24 h. The simulation was carried out as described by Liu (2009). The initial properties of the black bloom water were as follows: pH = 6.83 ± 0.10; chlorophyll a = 4.6 mg/L; DO = 0.53 mg/L; and turbidity = 660 NTU.

**Optimal dosage of PAC-modified clay under simulated flow conditions**

Diatomite (ShengzhouHuali Diatomite Products Co., Ltd, China) was used as clay and PAC (Gongyi Hengrun Water Treatment Material Co., Ltd, China) was used as the modifying agent. The dosage of diatomite was set to 1 g/L, and the dosage of PAC was chosen as 0.5, 0.6, 0.7, 0.8, and 0.9 g/L, based on preliminary experiments and other reports (Sengco et al. 2001; Liu et al. 2010). Thus, the cumulative dosage of PAC-modified clay (diatomite + PAC) was 1.5, 1.6, 1.7, 1.8, and 1.9 g/L.

The sediment (20 cm) was placed at the bottom of the tank, and 50 L of black bloom water was slowly poured into the tank. The motor rotation speed was set at 100 rpm. The relationship between motor rotation speed
(\(n\)) and the bed shear stress (\(T_b\)) can be expressed as:
\[
T_b = 2.07 \times 10^{-7} n^2 + 2.84 \times 10^{-5} n - 0.00281
\]
(Huang et al. 2015).

Subsequently, 5 L of modified clay solution was sprayed into the tank at a rate of 1 L/min. When the flow speed was stabilized, 20 mL of the sample was collected at a distance of 45 cm above the sediment at 5, 15, 30, 60, 180, and 480 min. Experiments for each dose were performed in triplicates (Figure 2).

**Deposition and resuspension of flocs under simulated flow conditions**

When the optimal dose was decided, flow simulation experiments were conducted to examine vertical changes in turbidity. Flow conditions and dosing of modified clay were as mentioned above. Water samples (20 mL) were collected at distances of 5, 10, 25, 45, and 70 cm above the sediment surface, at 10 min intervals for 70 min.

After the deposition experiments, the motor rotation speed was initially adjusted to 100 rpm, and increased by 100 rpm at 20 min intervals, to a final speed of 900 rpm. Samples were collected each time before the rotation speed was increased. The sampling heights and volumes were similar to those in the deposition experiments.

**Inhibition of floc resuspension using quartz sand**

After the resuspension experiments, the motor rotation speed was adjusted to 100 rpm and maintained for 8 h. One kilogram of 60-mesh quartz sand (Dai et al. 2015) (Shanghai Bo Tong Chemical Co., Ltd, China) was added to the tank. The rotation speed was initially adjusted to 100 rpm, and then increased by 200 rpm at 20 min intervals, with a final speed of 900 rpm. Samples were collected each time before the rotation speed was increased. The sampling heights and volumes were similar to those in the deposition simulation experiments.

The turbidity of all the samples was measured by the HACH2100Q (USA) turbidity meter.

**RESULTS AND DISCUSSION**

**Efficiency of removal of black suspended matter by PAC-modified clay**

The turbidity of black bloom water decreased rapidly within 60 min of adding PAC-modified clay, and gradually stabilized after 3 h (Figure 3(a)). The turbidity removal rate was positively correlated to the doses applied within the first 60 min, during which turbidity was reduced by 90% in all treatments after the first 30 min. However, the removal rates for all doses of PAC-modified clay, except 1.5 g/L, exceeded 95% after 8 h. The turbidity removal rates were not improved significantly (\(P = 0.05\)) when the dose of PAC-modified clay exceeded 1.8 g/L.

The dosage of PAC-modified clay used in our experiments was much higher than that reported previously (0.15 g/L, Dai et al. 2015). There are two possible reasons for this. Firstly, in the jar-test, particle contact was improved by rapid stirring, but the low speed of the simulated experiments made particle contact inefficient (Liu et al. 2010). As a result, the size of flocs did not increase as easily as in the jar-test, hence, we used higher doses of PAC-modified clay to flocculate the black matter. Secondly, the dosage of PAC can be easily affected by water conditions such as pH, ionic strength, suspended particles, and the coexistence of other ions or organic materials. In other words, the in-situ flocculation efficiency of PAC is largely dependent on highly variable water conditions (Li & Pan. 2013).
Flocculation and deposition at the optimal dosage

According to the results of the simulated flow experiment, PAC-modified clay effectively removed suspended black bloom matter. Turbidity decreased rapidly in the first 50 min, especially in the first 10 min by approximately 50%, while after 50 min the turbidity removal rate stabilized (Figure 3(b)). Moreover, visible turbidity stratification occurred at the bottom of the tank in association with increasing water depth and flow speeds.

Lake Taihu is a large shallow lake under the disturbance of stormy waves throughout the year, and is rarely completely static. The differences in air temperature and bottom terrain may also induce the formation of chaotic currents (Wang et al. 2014). Owing to horizontal and vertical flow speeds, the flocs cannot fully settle down, and a small part of the flocs will be scattered at a certain depth in the water body. This stratification of turbidity becomes obvious with increased flow speed.

Floc resuspension and its prevention

The simulated flow gradually increased with increasing motor rotation speed. When the rotation speed exceeded 300 rpm, water turbidity at all layers gradually increased. When the rotation speed exceeded 400 rpm, water turbidity suddenly increased from 217 to 620 NTU at the bottom, and from 25.8 to 59.2 NTU at the surface. These results indicate that this rotation speed provided the critical shear stress for resuspension of the black suspended matter/clay flocs. The bed shear stress was computed to be 0.024 and 0.045 N/m² at 300 and 400 rpm, respectively (Figure 4(a)).

In a large shallow lake like Taihu, both waves and flow currents generate shear stress at the water–sediment boundary which subsequently induces floc resuspension (Qian et al. 2011). In our previous study, the shear stress due to flow currents at a wind speed of 6.5 m/s was 0.034 N/m² (Wang et al. 2014), about three-fourths of the value mentioned above to resuspend the flocs. It has also

Figure 3 | (a) The turbidity removal rate variation with different modified clay dosages. The motor rotational velocity was 100 rpm, equivalent to the flow at 2.07–2.39 m/s wind speed (Huang et al. 2019). The modified clay dosages were 0.5, 0.6, 0.7, 0.8, and 0.9 g/L PAC plus 1 g/L diatomite. (b) The vertical profile of turbidity in the simulated flow experiment with modified clay dose of 1.8 g/L, 0.8 g/L PAC plus 1 g/L diatomite, motor speed 100 rpm. The sampling positions were 5, 10, 25, 45, and 70 cm above the surface of the sediments.

Figure 4 | Resuspension of black matter/clay flocs (a) without and (b) with a covering of quartz sand at different rotational velocities. The rotational velocities ranged from 100 to 900 rpm. The quartz sand dose was 14.15 kg/m².
been documented that the shear stress due to waves in Lake Taihu is of a magnitude higher than that due to flow currents (Qin et al. 2004). It is thus speculated that waves and currents may have different impacts on the aggregation and stability of flocs, and subsequently differ in the threshold values to generate resuspension. The individual and coupling effects of waves and currents on the onset of black matter/floc resuspension are therefore worth being investigated.

After adding quartz sand, visible resuspension of black matter/clay flocs occurred at 700 rpm. At this rotational speed, the bed shear stress was calculated as 0.109 N/m² (Figure 4(b)). However, the average wind speed did not exceed 3.56 m/s during the three stages of black bloom in Lake Taihu (Wang et al. 2011), which means visible resuspension would not occur during the process of black bloom. Thus, we suggest that quartz sand was effective in inhibiting resuspension of flocs. However, the effective inhibitory duration of quartz sand for resuspension has not been determined, and further research needs to be carried out on the feasibility of using quartz sand to inhibit resuspension of flocs during black bloom management.

CONCLUSIONS

It is concluded that: (1) PAC-modified clay is an effective agent to floculate the suspended matter during black bloom events; however, the flocs could not fully settle down due to waves and/or flow currents, and visible turbidity stratification occurred in the lower part of the simulation tank; (2) the floc resuspension experiments under simulated flow conditions suggest that floc resuspension is more likely to be induced by wave currents than by flow currents; (3) quartz sand can effectively inhibit the resuspension of black bloom matter/PAC-modified clay flocs; however, the inhibitory duration and the distinctive roles of waves and currents need to be determined.

ACKNOWLEDGEMENTS

This work was financially supported by the Special Foundation on Water Pollution Control and Treatment of China (2012ZX07101-010), the National Natural Science Foundation of China (51179053), Taihu Lake Water Environment Comprehensive Treatment Projects of Jiangsu Province (TH2014402), the Natural Science Foundation of the Jiangsu Higher Education Institutions of China (14KJB610007), the Jiangsu Provincial Talents Plan of Innovation and Entrepreneurship (2014–2016) and A Project Funded by the Priority Academic Program Development of Jiangsu Higher Education Institutions. The authors are grateful to Ms Feng Ji, Ms Jie Ma, Ms Xun Cao, and Ms Yue Ma for their valuable help in our research works.

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