



Effect of water quality improvement on the remediation of river sediment due to the addition of calcium nitrate

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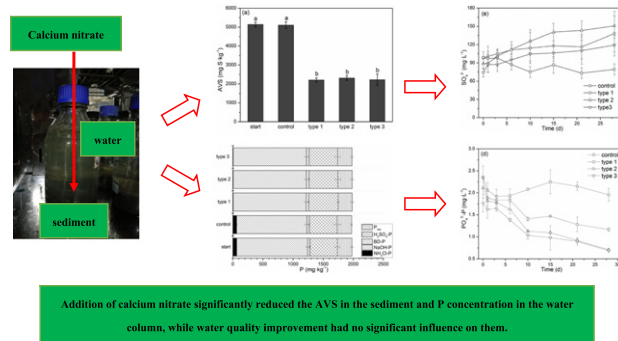
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HIGHLIGHTS

- AVS in the sediments decreased significantly with the addition of calcium nitrate.
- Calcium nitrate contributed to the reduction of phosphate in the water column.
- Water quality had effect on the interstitial nitrate and sulfate in the sediment.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
 Received 18 July 2016
 Received in revised form 17 September 2016
 Accepted 17 September 2016
 Available online 30 September 2016

Editor: Jay Gan

Keywords:
 Calcium nitrate
 Sediment
 Water quality
 Acid volatile sulphide
 Phosphorus
 Total organic carbon

ABSTRACT

In situ sediment remediation technique is commonly used to control the release of pollutants from sediment. Addition of calcium nitrate to sediment has been applied to control the release of phosphorus from sediments. In this study, laboratory experiments were conducted to investigate the effect of water quality improvement on the remediation of river sediment with the addition of calcium nitrate. The results demonstrated that the redox-potential of sediments increased from -282 mV to -130 mV after 28 days of calcium nitrate treatment. The acid volatile sulphide in the sediments significantly decreased (by 54.9% to 57.1%), whereas the total organic carbon decreased by 9.7% to 10.2%. However, the difference between these and water quality improvement was not significant. Due to the addition of calcium nitrate, low phosphate concentration in the water column and interstitial phosphate in the sediment were observed, indicating that the calcium nitrate was beneficial to controlling the release of phosphorus from river sediment. The decrease in phosphorus release could be attributed to the fixation of iron-phosphorus and calcium-phosphorus due to the addition of calcium nitrate. The addition of calcium nitrate to sediment caused the oxidation of sulphide to sulphate, hence resulting in high nitrate and sulphate concentrations in the water column, and high interstitial nitrate and sulphate concentrations in the sediment. The results also showed that only the water quality improvement had a significant effect on the interstitial nitrate and sulphate concentrations in the sediment.

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1. Introduction

Rivers have been the receivers of domestic wastewater effluent, rainwater runoff, agricultural runoff and industrial wastewater for

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years, and this has seriously damaged the river water quality (Scholes et al., 2008). Due to rapid increases in economy and population in China, the quantity of pollutants discharged into the rivers has also increased. The continuing development of pollution in water bodies causes them to become turbid and odourous after their carrying capacity of these pollutants has been exceeded. Water pollution mostly occurs in water bodies, which either have a low flow rate or are static. Therefore, water pollution of urban rivers is becoming one of the greatest threats to the aquatic ecosystem in China.

Large amounts of organic pollutants from wastewater are accumulated in river sediment, while excessive degradation of the organic matter consumes much dissolved oxygen (DO) resulting in hypoxia of the sediment. Low DO causes a low redox potential [oxidation reduction potential (ORP)], which leads to microbial reduction of sulphate (SO_4^{2-}) to hydrogen sulphide (H_2S) in the sediment (Berner, 1970). A decrease in the ORP of sediment can also cause the reduction of Fe and Mn, which results in the release of phosphorus (P) adsorbed on Fe and Mn oxide-hydroxide particles (Caraco et al., 1989; Schauser et al., 2006). Subsequently, Fe(II) in the water column and sediment reacts with H_2S to form iron sulphide (FeS) because of its low-solubility product and gets adsorbed on the organic particles, causing the water and sediment layer to turn black-coloured. Furthermore, toxic substances, such as ammonia, H_2S and other odourous gaseous sulphur compounds, are produced as a result of anaerobic respiration, which leads to odourous phenomenon along the rivers.

Water quality can be improved with the mitigation of external pollution sources and advanced treatment of wastewater. However, sediments can continuously release pollutants to the water column, thus delaying the recovery of eutrophic environments (Søndergaard et al., 2007). Previous research indicated that eutrophic ecosystems could be recovered using in situ technologies, such as addition of nitrate, Al, Fe and Ca to the sediment (Ripl, 1976; Foy, 1986; Cooke et al., 1993; Kleeberg et al., 2000; Xu et al., 2008; Yin and Kong, 2015). The addition of these compounds to sediments can improve the oxidation conditions and accelerate the denitrification activity (Shimizu and Nakano, 2009). Ripl (1976) found that the biochemical oxidation could be achieved for remediation purposes by injecting calcium nitrate into the sediment to control P release from the sediment, which would limit the growth of algae in the water body.

Apart from controlling the P release from sediment, nitrate is also used to abate the odours caused by sulphate-reducing bacteria in wastewater (Jenneman et al., 1986; Jefferson et al., 2002; Jiang et al., 2009). When nitrate is metabolised, a portion of the organic matter is used to reduce SO_4^{2-} to S^{2-} (Jenneman et al., 1986). The acid volatile sulphide (AVS) concept is defined as the sedimentary S, which is extracted by $1 \text{ mol} \cdot \text{L}^{-1}$ HCl solution, including porewater sulphides and metastable FeS (Berner, 1964). The AVS content in the sediment is strongly correlated with the colour intensity of the sediment, which could be used to assess the pollution of organically enriched sediment (Wilson and Vopel, 2012). Sulphur-containing odourous compounds also originate from AVS in the sediment, and to some extent, AVS can be considered as the source of sulphur-containing odour and potential production of H_2S . Thus, reducing AVS content in the sediment and controlling the release of sulphur-related odour to the water column are meaningful and urgent research tasks.

P loading is considered to have a key role in the formation of algal blooms and eutrophication (Yin et al., 2013). Since P release from anaerobic sediments usually makes up the major fraction of total P (TP) load in lakes or rivers, therefore the reduction of an external P load by controlling the influent water quality may be insufficient (Cooke et al., 1993). Consequently, a remediation planning to control the P sources should be a priority. Many amendment technologies, such as modified zeolite (Yang et al., 2014), magnetic microparticles (Funes et al., 2016) and thermally modified calcium-rich attapulgite (Yin et al., 2013; Yin and Kong, 2015) have been processed to immobilize both the soluble reactive P in porewater and mobile P in sediment.

Upgradation of wastewater treatment plants and the mitigation of external pollution sources have been considered effective in improving the lake and river water quality (Jeppesen et al., 2007; Fulton et al., 2015). However, both the improvement and remediation of polluted sediment are far slower than that of the water column. Meanwhile, sediment would continue to be an important internal source of nutrients to the water column for a long time, which affects the water quality of overlying water due to a larger nutrient concentration in the sediment porewater than in the water column (Liu et al., 2014). In the present study, the effect of water quality improvement on sediment remediation has been investigated, which is related to the variation in AVS due to the addition of calcium nitrate.

2. Material and methods

2.1. Water and sediment sampling

The sediment and water samples used in this study were obtained from the Shajing River ($22^\circ 46' 3''$, $113^\circ 49' 52''$) in Shenzhen City, Guangdong, China. This river is a tributary of the Maozhou River, which flows into the Pearl River Estuary. In the present study, the water and sediment samples were obtained from upstream, midstream and downstream of the Shajing River, and were mixed to present the characteristics of water and sediment. The river flows slowly at the depth of 1.5–2.0 m, and receives pollutants of rainfall runoff and occasional industrial and urban wastewaters from the riverside. The water samples were obtained from the middle zone of the river using a glass water sampler, while the sediment samples were collected from the top 20 cm layer of three sites using a piston column sediment sampler (purchased from Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences, China). The samples were homogenized and sieved to remove detritus, plant residues and plastic products. Then, the collected samples were kept cool in a box containing ice cake. These were transferred to the laboratory and stored at a temperature of approximately 4°C until further use. The water quality and sediment's characteristics of the sampling station are summarized in Tables 1 and 2.

2.2. Experimental design

The experiments were conducted using 1 L wide-mouth jars having screw top lids. Before the experiments, the jars were placed in a sterilizing pot at 121°C for 30 min. The sediment (250 g) and water column (800 mL) were placed in the jars, and subsequent treatment was performed in triplicate. The heights of sediment and overlying water were 5 and 10 cm, respectively. Four treatments were conducted. The first treatment did not receive calcium nitrate (control treatment). The sediments in the other three treatments were given the same dosage of calcium nitrate at three different water column compositions as shown in Table 3. The water column was replaced with pure water, which represented the treated water. Based upon the study of Chen et al. (2013), the application dosage was 1 g of calcium nitrate per 1 kg of wet sediment. Nitrogen gas was injected into the jars when the calcium nitrate was mixed with the sediment to avoid sediment oxidation. The jars were kept closed during the experiments, which were performed at 25°C in an incubator. The water samples were withdrawn on 0th, 1st, 3rd, 6th, 10th, 15th, 21st and 28th day of the experiment, while those of sediment were withdrawn on 0th and 28th day of the

Table 1
Characteristics of the Shajing River water investigated in this study.

Unit	pH	DO	Eh	NH_4^+-N	NO_3^--N	TN	$\text{PO}_4^{3-}-\text{P}$	COD
		$\text{mg} \cdot \text{L}^{-1}$	mV	$\text{mg} \cdot \text{L}^{-1}$	$\text{mg} \cdot \text{L}^{-1}$	$\text{mg} \cdot \text{L}^{-1}$	$\text{mg} \cdot \text{L}^{-1}$	$\text{mg} \cdot \text{L}^{-1}$
Mean	7.26	0.26	-68.6	14.6	0.10	18.2	6.2	84.5
Std	0.05	0.04	12.2	1.2	0.02	0.86	1.2	15.0

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