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# High sulfide production induced by algae decomposition and its potential stimulation to phosphorus mobility in sediment

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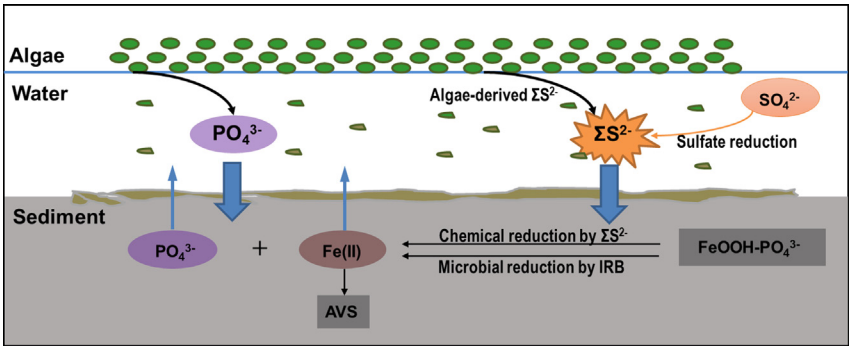
## HIGHLIGHTS

- Algae blooms significantly affect sulfide production and P mobility in sediments.
- Abundant $\Sigma S^{2-}$ can be produced from algae decomposition (termed "algae-derived $\Sigma S^{2-}$").
- $\Sigma S^{2-}$ diffused from water column to sediments can affect Fe and P cycles.
- Iron reduction in sediments can be promoted by the chemically mediated of $\Sigma S^{2-}$.
- P mobility in sediments can be dramatically influenced by the reduction of iron oxides.

## GRAPHICAL ABSTRACT

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## ABSTRACT

This study is devoted to addressing the effects of algae blooms on sulfur cycle and the consequent phosphorus mobility in the sediments of freshwater lake ecosystems. A mesocosm experiment was conducted to investigate these effects through monitoring the dynamics of sulfur (S), iron (Fe) and phosphorus (P) in water and sediments, and their diffusion fluxes at the sediment-water interface (SWI). In addition, the abundance of sulfate-reducing bacteria (SRB) in the water column was also detected. The addition of the algae lead to an increase of SRB, a drastic decline of sulfate and a significant increase of total dissolved sulfide ($\Sigma S^{2-}$, the peak value of near 3.0 mmol/L on day 6) in the water column. These results suggest the sulfate reduction was dramatically promoted during algae decomposition. Indeed the $\Sigma S^{2-}$ was 2 to 3 times of $SO_4^{2-}$ initial concentration, and higher $\Sigma S^{2-}$ was produced with higher algal biomass. Moreover, the diffusive flux of $\Sigma S^{2-}$ at the SWI was negative, indicating that diffusion of $\Sigma S^{2-}$ from water column toward sediment was occurring. These results indicated that algae decomposition might also be another important source of $\Sigma S^{2-}$ (termed "algae-derived $\Sigma S^{2-}$") in addition to sulfate reduction. The increase of Fe(II) in surface sediment pore-water was slightly delayed compared to the $\Sigma S^{2-}$ generation in the water column, which illustrated that Fe oxyhydroxides in sediments were transformed into Fe(II) through chemical reduction of $\Sigma S^{2-}$. Concomitantly, the vertical distribution of $PO_4^{3-}$ in high amounts algae group suggested that desorption and release of iron oxides-bound $PO_4^{3-}$ occurred in sediments. Collectively, algae bloom can boost the lake eutrophication not only through direct release of nutrients but also through the high production of $\Sigma S^{2-}$ and indirect promotion of phosphorus mobility in sediment.



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## 1. Introduction

Over the last decades, rapid population and economic growths has led to excessive nutrient inputs to freshwater ecosystems and an increase in the frequency and intensity of algae blooms as well as the large accumulation of nutrients in sediments (Paerl and Paul, 2012; Paerl et al., 2011; Yan et al., 2017). Even when external anthropogenic inputs are effectively cut down, the internal loads of nutrients, especially phosphorus in sediment, can still continuously contribute to water eutrophication (Wang et al., 2016; Yu et al., 2017). It has been reported that in eutrophic lakes the phosphorus released from sediments under certain environmental conditions is usually linked to the algae accumulation and decomposition (Chen et al., 2016; Chen et al., 2014; Chen et al., 2018). As a result, phosphorus mobility in sediments of eutrophic lakes caused by algae blooms has recently drawn significant attention.

Phosphorus release from sediments in eutrophic lakes has been attributed to phosphate desorption caused by microbial reduction and dissolution of Fe oxyhydroxides in sediments under anaerobic conditions (Amirbahman et al., 2003; Jensen et al., 1992; Roden and Edmonds, 1997). Briefly, when the algae accumulate to a large scale on the lake surface, an extremely anaerobic environment is gradually formed driven by the respiration and microbial degradation of algae (Chen et al., 2018; Ding et al., 2018; Fan, 2015). Under such circumstances, the Fe oxyhydroxides in sediments are reduced to Fe(II) by microorganisms like iron-reducing bacteria (IRB), and sulfate in the overlying water also can be reduced to soluble sulfides ($\Sigma S^{2-}$). Then, the solid iron sulfides will be formed and buried in sediment, leading to desorption and diffusion of phosphate from Fe oxyhydroxides to overlying water (Chen et al., 2016; Gunnars and Blomqvist, 1997; Shen et al., 2016). Apparently, the anaerobic environment is an essential prerequisite for P release from sediments. Besides, iron oxides reduction, $\Sigma S^{2-}$ production and further formation of solid iron sulfides are all indispensable direct causes of P release. Accordingly, the effect of $\Sigma S^{2-}$ on phosphorus mobility in sediments is crucial and should not be ignored.

It has been found that sulfate reduction and its product $\Sigma S^{2-}$ play a vital role in the release of phosphorus from marine sediments (Caraco et al., 1989; Jensen et al., 2003; Lehtoranta et al., 2009; Roden and Edmonds, 1997; Sulu-Gambari et al., 2016). For eutrophic marine systems, dissimilatory sulfate reduction is the dominant microbial mineralization pathway. The iron oxides in sediment can be chemically reduced to Fe(II) by the mediation of $\Sigma S^{2-}$ and buried in sediments as FeS and $FeS_2$, which significantly weakens the adsorption and fixation of phosphorus and results in phosphorus release to the overlying water (Lehtoranta et al., 2009). For freshwater lakes, however, it is commonly considered that sulfate reduction could not be sustained due to the low sulfate level (like Lake Taihu with about 1 mM of $SO_4^{2-}$) (Hansel et al., 2015). Thus, sulfate reduction in freshwater lake systems has always been deemed to have little effect on iron reduction in sediments, so phosphorus release from sediment has been mainly attributed to the direct dissimilatory reduction of iron oxides. Therefore, the influence of sulfate reduction and its product $\Sigma S^{2-}$ on phosphorus mobility in the sediment of freshwater lakes has not been throughout studied.

However, the effect of sulfate reduction and its product $\Sigma S^{2-}$ on iron reduction in sediment and the consequent phosphorus mobility in freshwater systems may be more important than previously recognized. Kwon et al. (2014) found that the reduction rate of iron oxides was significantly affected by sulfate concentration and was relatively limited under low sulfate concentration (0.2 mM) while increased by ten times when sulfate level increased to 10 mM. Hansel et al. (2015) further confirmed that even when the sulfate concentration was as low as 0.2 mM, the anaerobic metabolism of organic matters was still dominated by sulfate reduction, and the chemical reduction mediated by $\Sigma S^{2-}$ was the primary pathway of iron reduction. Thus, in freshwater lakes, sulfate reduction does not directly release phosphorus from iron oxides, but indirectly promotes phosphorus mobility and reduces the phosphorus retention capacity through blocking Fe cycling (Lehtoranta et al., 2009). In addition, sulfate concentrations are increasing in freshwater environments all over the world (Chen et al., 2016; Kumaresan et al., 2017; Yu et al., 2013). Under this context, the effects of sulfate reduction and sulfur cycle on phosphorus mobility in sediments of freshwater eutrophic lakes are more prominent. Chen et al. (2016) reported that increasing sulfate concentration caused high $\Sigma S^{2-}$ production and phosphorus release from sediment during algae decomposition. However, at present, the relationship between algae decomposition and $\Sigma S^{2-}$ production and the related impact on the mobility of phosphorus in sediments are mostly unknown.

In this study, laboratory incubation experiments were carried out to reveal the effect of algae decomposition on sulfur cycling and the phosphorus mobility of sediments in freshwater ecosystems. The impact of algae accumulation on the $\Sigma S^{2-}$ production in the water column and the migration behavior of S, Fe, and P at the sediment-water interface (SWI) during algae decomposition were investigated. The results are helpful to clarify the contributions of algae bloom decomposition to $\Sigma S^{2-}$ production and its effect on the mobility of phosphorus in freshwater sediments, as well as in highlighting the influence of algae bloom on the phosphorus release from sediments and even on the eutrophication processes of freshwater lakes.

## 2. Materials and methods

### 2.1. Sample collection and preparation

Samples of water, sediment and algae were collected from the eutrophic Lake Taihu in July 2016. Lake Taihu (31°10′ N, 120°24′ E), one of the largest shallow freshwater lakes in China, is situated at the south of the Yangtze River delta with a mean depth of 1.9 m and an area of 2340 $m^2$ (Qin et al., 2007). Lake water was collected into several 50-L closed plastic barrels near the Zhushan Bay (31°24.293′ N, 120°0.697′ E). Sediment cores from this site were also collected using a gravity core sampler. The sediment cores were then sectioned into 4 cm intervals, and the same layer samples were stored together. Algae bloom scum was collected and concentrated by sieving water through fine mesh plankton (250 meshes) and immediately stored in polyethylene bottles. Sediment and algae samples were kept in a portable refrigerator and delivered to the laboratory immediately. In the laboratory, the freshly collected algae were flushed with lake water and centrifuged at 1500 r/min for 5 min by a CT15RT versatile refrigerated centrifuge (China). Additionally, all sediment samples of the same layer were mixed and homogenized in a large container. Large particulates were carefully removed by hand. All samples were kept at 4 °C for no longer than 24 h before using them for the experiments.

### 2.2. Experimental mesocosms

Each layer of sediment was transferred into 40 Plexiglas tubes (Φ90 mm, length 500 mm) to reach a thickness of 20 cm. The lake water was gently added to the sediment surface to a thickness of 15 cm using intravenous needles. The 40 test-tube mesocosms were preincubated for two weeks at 30 °C in a water bath to stabilize the interaction between sediment and water. Then the mesocosms were divided into four groups of ten (Fig. S1): three treatments and one control. Different amount of algae bloom scum was added to each treatment in order to achieve low, moderate and high algal biomass concentration. Specifically, each tube of treatment B, C and D received 17 g, 50 g and 100 g of algaeto achieve a concentration of algae of 2500 g/$m^2$, 7500 g/$m^2$ and 15,000 g/$m^2$, respectively. The remaining group served as the control (group A). All mesocosms were incubated at 30 °C for 23 days in the dark. Sampling time was set on days 0, 1, 2, 4, 6, 8, 10, 14, 18 and 23. One tube from the four corresponding groups was sampled each time. Water samples from the water columns were collected 5 cm