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## Successful control of internal phosphorus loading after sediment dredging for 6 years: A field assessment using high-resolution sampling techniques

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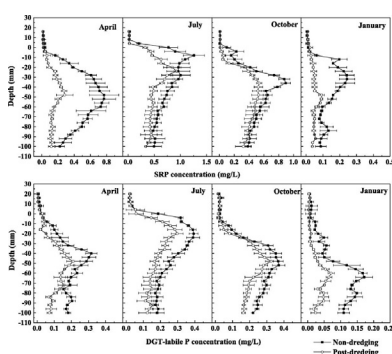
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### HIGHLIGHTS

- Decreased SRP and DGT-labile P concentrations evidenced the efficacy of dredging.
- Dredging effectiveness in winter and spring is higher than in summer and autumn.
- Fe redox cycling is key to regulating dredging effectiveness.
- Algal decomposition suppresses dredging effectiveness during summer and autumn.

### GRAPHICAL ABSTRACT



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### ABSTRACT

The effectiveness of sediment dredging for the control of internal phosphorus (P) loading, was investigated seasonally in the eutrophic Lake Taihu. The high-resolution dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) techniques were used to measure the concentrations of soluble Fe(II) and soluble reactive P (SRP) as well as DGT-labile Fe/P in the non-dredging and post-dredging sediments. The P resupply kinetics from sediment solids were interpreted using DGT Induced Fluxes in Sediments (DIFS) modeling. The results showed no obvious improvement in water and sediment quality after dredging for 6 years, due to their geographical proximity (a line distance of approximately 9 km). However, dredging significantly decreased the concentrations of soluble Fe(II)/SRP and DGT-labile Fe/P in sediments, with effects varying at different depths below the sediment-water interface; More pronounced effects appeared in January and April. The diffusive flux of pore water SRP from sediments decreased from 0.746, 4.08 and 0.353 mg/m<sup>2</sup>/d to 0.174, 1.58 and 0.048 mg/m<sup>2</sup>/d in April, July and January, respectively. DIFS modeling indicated that the P retention capability of sediment solids was improved in April in post-dredging site. Positive correlations between pore water soluble Fe(II) and SRP as well as between DGT-labile Fe and P, reflect the key role of Fe redox cycling in regulating dredging effectiveness. This effect is especially important in winter and spring, while in summer and autumn, the decomposition of algae promoted the release

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of P from sediments and suppressed dredging effectiveness. Overall, the high-resolution HR-Peeper and DGT measurements indicated a successful control of internal P loading by dredging, and the post-dredging effectiveness was suppressed by algal bloom.

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## 1. Introduction

Eutrophication is strongly associated with the enhancement of harmful algal blooms and has become a significant global environmental problem (Heisler et al., 2008; O'Neil et al., 2012). Phosphorus (P) has been established as the limiting factor for eutrophication (Carpenter, 2008; Schindler et al., 2016), with development of methods for the reduction of internal P attracting increasing attention. Sediments act as the sinks and sources of pollutants in aquatic environments, and P accumulated in the sediments can be released into the overlying water (Yang et al., 2017). Internal P loading from lake sediments can hinder the improvement of water quality and sustain the eutrophication for decades, even if external P loading source has been effectively reduced (Lepori and Roberts, 2017; Paytan et al., 2017; Søndergaard et al., 2013).

Typically, the techniques for internal loading control are either *ex-situ* or *in situ*. Sediment dredging is an *ex-situ* technique, commonly used worldwide as an engineering tool to permanently remove contaminated sediments from waterbodies (Bormans et al., 2016; Gustavson et al., 2008). The successful control of internal P loading by sediment dredging has been supported by laboratory scale studies (Chen C et al., 2016; Yu et al., 2017; Zhong et al., 2008). Successful implementation of sediment dredging in the field have been reported for Lake Xihu (Lou, 2007), Caohai of Lake Dianchi (Hu et al., 2010) and Gonghu Bay of Lake Taihu (Gu et al., 2016) in China; for Lake Uluabat in Turkey (Yenilmez and Aksoy, 2013); and in Lake Trummen in Sweden (Björk et al., 2010). In these cases, the level of eutrophication was reduced by dredging, with the oxygen penetration depth increased and internal P loading decreased afterwards. However, failure of dredging to control internal P loading has also been reported in the field in Lake Chaohu (Wang et al., 2014), northern part of Lake Taihu (Liu et al., 2016b) and some urban rivers in China (Weng, 2017); in Lake Vajgar in Czech Republic (Björk et al., 2010). In these cases, no obvious reduction of internal loading was found and the level of eutrophication remained relatively high. The inconsistent outcome of effects of dredging in field experiments makes it necessary to conduct further assessment in the field.

Most field studies performed assessments according to the change in chemical quality (e.g. total phosphorus (TP), total nitrogen (TN) and organic matter (OM)) in sediments and/or in solution phase (overlying water and pore water) (Weng, 2017; Yenilmez and Aksoy, 2013). In addition, chemical fractionation methods are commonly used to assess the mobility of P in post-dredging sediments (Jing et al., 2015; Wang et al., 2014; Xu et al., 2009). For example, Weng (2017) assessed the post-dredging effects based on TP, TN and OM contents in the sediment in urban rivers of Yangzhou and found the effects are not ideal and the eutrophication degree is still relatively high. Jing et al. (2015) conducted a Dongqian Lake field study following a three-year dredging period, finding that the easily-changeable P concentrations showed no significant change in comparison with P in non-dredging sediments, but metal (Fe, Mn and Al) bound P concentrations decreased. However, these methods based on the change in total mass in sediments or extracted mobile P fractions may not be sensitive enough to reflect dredging effectiveness, especially after long-term dredging implementation programs (e.g., several years to decades). Due to the limitation of these widely used conventional analytical approaches in elaborating the post-dredging effects, the mechanism underlying sediment dredging is still unclear and the critical success factors after dredging are still missing.

It is of note that there are significant vertical variations in the mobility and distribution of the mobile P fraction in sediments. This feature

has been studied intensively in recent years through high spatial resolution measurements using passive sampling techniques, such as high-resolution dialysis (HR-Peeper) and diffusive gradients in thin films (DGT) (Chen M.S. et al., 2016; Sun et al., 2017; Wang J.F. et al., 2016). Pronounced heterogeneity of labile P was observed in sediments at the two-dimensional, submillimeter level (Ding et al., 2015; Santner et al., 2015). The simultaneous, *in situ* measurement of labile P and Fe by DGT, has established the system of Fe redox-controlled remobilization of P, which is regarded as a primary mechanism responsible for internal P loading (Ding et al., 2016). It can be inferred that the change in mobile P concentration should vary according to depth in post-treated sediments (Lin et al., 2017a, 2017b; Wang et al., 2017). However, to date few studies have assessed the chemical heterogeneity of sediments when assessing dredging effectiveness.

In the present study, the effectiveness of sediment dredging on controlling internal P loading across four seasons was investigated in the eutrophic Lake Taihu. The concentrations of soluble Fe(II)/SRP and DGT-labile Fe/P were measured in the non-dredging and post-dredging sediments, with analysis to the millimeter scale performed using high-resolution dialysis (HR-Peeper) and ZrO-Chelex DGT. The kinetics of P resupply from sediment solids, were interpreted using the DGT Induced Fluxes in Sediments (DIFS) modeling.

## 2. Materials and methods

### 2.1. Preparation of HR-Peeper and ZrO-Chelex DGT probes

HR-Peeper and ZrO-Chelex DGT probes were provided by EasySensor Ltd. ([www.easysensor.net](http://www.easysensor.net)). They were used to measure soluble Fe(II)/SRP in the pore water and DGT-labile Fe/P in the sediment, respectively. The HR-Peeper was prepared according to the method described by Xu et al. (2012). The probe chambers were filled with deionized water and then covered by a Durapore® PVDF membrane (Millipore, 0.45 µm pore size, 0.10 mm thickness) with a plastic window having an open area of 1.8 cm × 15 cm. The ZrO-Chelex DGT device consists of a ZrO-Chelex binding layer (binding gel) and a diffusion layer (diffusion gel plus filter membrane). The ZrO-Chelex DGT has high capacities for measurements of P and Fe, and the values are 90 µg P/cm<sup>2</sup> and 75 µg Fe/cm<sup>2</sup> respectively (Xu et al., 2013). The diffusion gel and binding gel were prepared according to Ding et al. (2015) and when assembling the ZrO-Chelex DGT probe, the binding gel was covered in sequential layers of an agarose diffusive gel and a Durapore® PVDF membrane (Millipore, 0.45 µm pore size, 0.10 mm thickness). All HR-Peeper and DGT probes were soaked in 0.01 M NaCl solution and deoxygenated under a steady nitrogen flow for a minimum of 16 h, prior to their deployment in sediments.

### 2.2. Study sites and field sampling

Lake Taihu has an area of 2338 km<sup>2</sup>, a volume of 4.4 billion m<sup>3</sup> and is the third largest freshwater lake in China. Due to dramatic increases in nutrient loading due to urban and agricultural development within its watershed, Lake Taihu is highly eutrophic and suffers from increasingly severe, toxin producing cyanobacterial blooms during summer months (Xu et al., 2017). Meiliang Bay is in the northern part of Lake Taihu with an area of 132 km<sup>2</sup> and it is one of the most eutrophic regions of the lake, resulting in sediment contamination with nutrients and heavy metals (Yan et al., 2016; Zhang et al., 2017). From 2009 to 2010, dredging projects were applied to the northern part of Meiliang Bay (Fig. 1), with

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