



## Do NH<sub>3</sub> and chemical oxygen demand induce continuous release of phosphorus from sediment in heavily polluted rivers?



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### ABSTRACT

While phosphorus (P) is a vital element in freshwater systems, excessive P loads will induce eutrophication. Large inputs of various pollutants, including P, to rivers in semi-arid regions result in complex environmental problems. In this study, we investigate dissolved oxygen (DO), ammoniacal nitrogen (NH<sub>3</sub>), chemical oxygen demand (COD<sub>Cr</sub>), soluble reactive P (SRP) in surface water and pore water, and sediment P in the Niuwei River, within the Hai River Basin. From our results we developed and tested a theory for P release in semi-arid regions driven by DO. The results show that NH<sub>3</sub> and COD<sub>Cr</sub> can cause variations in DO in surface water. The presence of pollutants at high concentrations caused seasonal hypoxia. Hypoxic river water induced adsorption and hydrolysis of NaOH-Pi and labile organic P (L-Po), which then resulted in increased SRP concentrations in pore water and surface water. Overall, our results illustrate that NH<sub>3</sub> and COD<sub>Cr</sub> may be important for P adsorption and desorption and for management of water quality problems in rivers in semi-arid regions.

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### 1. Introduction

Eutrophication of freshwater systems, as one of the most serious environmental issues in semi-arid region, is mainly caused by phosphorus (P) (Carpenter, 2008; Pernet-Coudrier et al., 2012). Rapid socioeconomic development, huge increases in population, and the systematic damming of rivers induce deterioration of freshwater ecosystems (Li et al., 2012; Pernet-Coudrier et al., 2012; Shan et al., 2012; Taylor et al., 2015). Over the last 30 years, numerous nutrient management schemes have been initiated in China, with the aim of improving water quality. The Chinese government is very concerned about controlling pollution inputs, and so regulations and projects have been implemented, such as the Chinese National Water Pollution Control and Technology Management Project. Despite recent sustained efforts to improve water quality, eutrophication still persists in rivers in semi-arid regions, and the water quality has deteriorated to such a degree that it is not suitable for any use (Han et al., 2013). Although external P inputs to rivers have been decreasing, internal release of P from sediment provides

sufficient reactive P to sustain harmful algal blooms (Read et al., 2014). Studies that provide information about the mechanisms and drivers of P adsorption and desorption in fluvial sediment in semi-arid regions are therefore urgently needed (Kraal et al., 2013; Ahlgren et al., 2005).

Biogeochemical cycles are generally driven by reducing and oxidizing conditions (Nielsen et al., 2010). Excessive nutrient inputs will consume oxygen and induce hypoxia in the bottom waters, with the result that P is readily remobilized from sediments. This internal source of P will fuel high primary productivity and oxygen consumption; this is particularly relevant for the slow flowing rivers with numerous dams that are common in semi-arid regions of northern China (Gustafsson et al., 2012). Iron (Fe) and sulfur (S) may influence P adsorption and desorption. The reductive dissolution of Fe-oxides induces P release under seasonal hypoxia (Cowan et al., 1996; McManus et al., 1997). In summer, the lower oxygen levels in bottom water induce high rates of sulfate reduction, with the result that there are fewer iron oxides to bind P when sediment iron oxides are being converted to iron sulfides (Jensen et al., 1995; Rozan et al., 2002). In re-oxygenated conditions, the iron sulfides can be oxidized, at which point they create iron oxides that have the ability to sequester P (Dale et al., 2013). In addition, sulfur-oxidizing bacteria can accumulate P, which will be released from

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seasonally hypoxic sediment during summer (Schulz and Schulz, 2005). Recently, it was discovered that a filamentous bacterium from the *Desulfobulbaceae* family (cable bacteria) could control iron-P dynamics in sediments in a coastal hypoxic basin (Pfeffer et al., 2012; Seitaj et al., 2015; Sulu-Gambari et al., 2015).

The Hai River Basin (HRB) experiences severe water resource shortages, and water quality was seriously deteriorated. Excessive quantities of pollutants from agricultural activities and industrial development have been discharged into the system, to the degree that the river suffers from a mixture of pollutants at high concentrations. P biogeochemical cycling is much more complex in this type of river. Through long-term monitoring, we discovered that ammoniacal nitrogen ( $\text{NH}_3$ ) and chemical oxygen demand (COD<sub>Cr</sub>) were important components of the pollutant load in surface water. We also found that there were relationships between  $\text{NH}_3$ , COD<sub>Cr</sub>, and P. In this contribution we present field data from a tributary of the HRB, the Niuwei River, and demonstrate the link between seasonal changes in concentrations of  $\text{NH}_3$ , COD<sub>Cr</sub>, dissolved oxygen (DO), P in surface and pore water, and P fractions in sediment. The results from this study will be very significant for pollutant control, especially P, in rivers in semi-arid regions.

## 2. Materials and methods

### 2.1. Study area

The Niuwei River is the most polluted part of the Fuyang River and is contaminated by a wide range of pollutants. It flows through a fertile plain that easily supports intensive agriculture, mainly wheat, corn, and vegetables (Li et al., 2012). This region has a temperate continental monsoon climate and most of the rainfall falls during summer; precipitation in June, July, and August accounts for 70% of the annual precipitation (Chun et al., 2009). The water flows from south west to north east. There are dams and gates in this region that lead to fragmentation and instability of the river system. The Niuwei River was chosen for this study as it is representative of the conditions in the HRB in northern China.

### 2.2. Field sampling

The sampling sites were located on the main channel of the Niuwei River. In general, sampling sites should not be in dead water,

backwater, or discharge outfall areas, and should be, as far as possible, on straight sections of the river where the flow is gentle and the channel is wide. We selected five sampling sites, the locations of which are presented in Fig. 1. We collected the sample at the middle of every-month from Jan-2014 to Dec-2015, which continue 2 years. Five sample sites distribute along the Niuwei River. The S1 and S2 were at the urban area of Xingtai City and Ren County. S3 was located at the transition area between the city and suburban region, and S4 and S5 were at suburban district. As a transition site, sediment was also collected at S3 in this research. We collected the water samples (200 mL) with an organic glass sampler (WB-PM, Beijing Splitter Instrument Co., Ltd.) from the middle of the river. The samples were stored at 4 °C in a refrigerator in our vehicle and were immediately transported to the laboratory, where analyses were carried out within two days. DO concentrations were measured in situ with a portable device (YSI ProPlus).

Surface sediments (about 2 cm) were collected with PVC sampling tubes, the diameter and length were 6.3 and 60 cm. There were three subsampling sites at each sampling site. Two subsamples collected at the same site were pooled and homogenized to obtain a representative sample. One sediment sample was used for extracted the pore water sample. Sediment samples for P analysis were kept in sealed plastic bags at 0 °C during transport to the laboratory. Samples were freeze-dried at –50 °C (FD-1 freeze-dryer) on arrival at the laboratory. Dried samples were ground and passed through a 100-mesh sieve. Sediment sample for extracted pore water was deal as soon as possible. Quartered samples were taken as representative samples, and were stored in sealed plastic bags at room temperature until analysis. All the samples were collected from January 2014 to December 2015.

### 2.3. Sample analysis

#### 2.3.1. SMT fractions

P fractions were determined as outlined in the Standards Measurements and Testing (SMT) protocol (Ruban et al., 2001a,b). Sediment P was classified into total P (TP), inorganic P (Pi), organic P (Po), P associated with Ca (HCl-Pi), and P bound to Fe, Al and Mn oxides and hydroxides (NaOH-Pi). To determine TP, 0.2 g dry sample calcined at 450 °C for 3 h. The cool ash was transferred to a centrifuge tube, to which 20 mL of 3.5 mol L<sup>-1</sup> HCl was added. The tube shaken overnight (16 h) and then was centrifuged at 2000g for

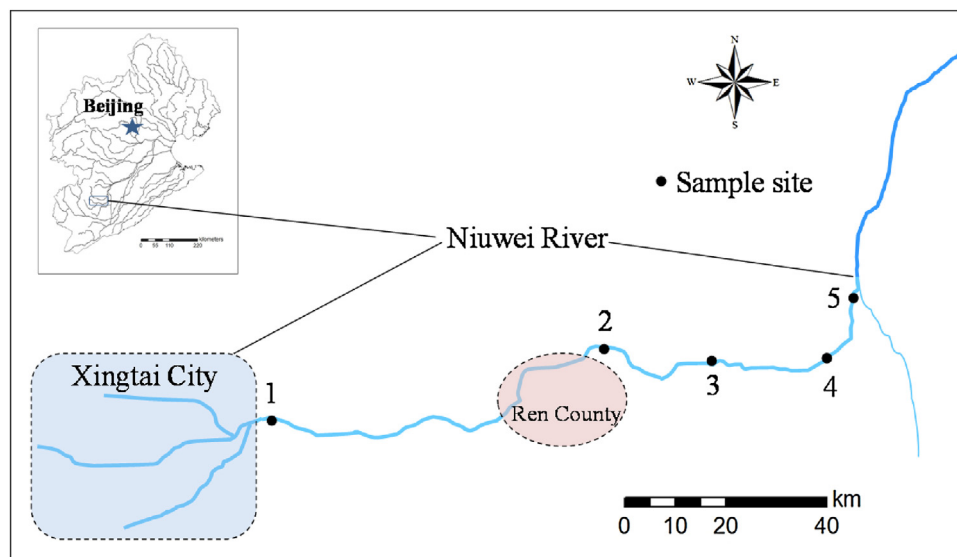


Fig. 1. The sample sites in Niuwei River.

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