



# Phosphorus storage dynamics and adsorption characteristics for sediment from a drinking water source reservoir and its relation with sediment compositions



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## ABSTRACT

Water and sediment samples were collected from a representative drinking water source reservoir to investigate the nutrient spatial distributions, phosphorus storage, adsorption features and its relation with sediment compositions. Monitored water TN, TP concentrations, sediment pH values exhibited a gradual decrease trend from the upstream to the downstream. Sediment AIT content (total aluminum oxides) was 2–4 times higher than FeT (total iron oxides) which mainly comprised with Fed (free iron oxides) and Feox (amorphous iron oxides). Phosphorus fractional analysis found that Ca–P was the major phosphorus component while Ex–P + Al–P + Fe–P contents accounted for no more than 20% to sediment TP. Phosphorus adsorption process followed the pseudo-second-order kinetic model and Langmuir isotherm equations better than other kinetic and isotherm models. Film-diffusion was the rate-limiting step and physic processes dominated phosphorus adsorption. Maximal phosphorus adsorption capacities could be significantly well simulated with sediment Feox, FeT and TP concentrations ( $r^2 = 0.96$ ).

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## 1. Introduction

The Danjiangkou Reservoir (32°36′–33°48′ N, 110°59′–111°49′ E) has a usual water surface area of 745 km<sup>2</sup> and water storage capacity of 17.45 billion m<sup>3</sup> (Li et al., 2008). Since 2002, the Chinese government has been implementing the Middle Route Project (MRP) of the South to North Water Diversion Project (SNWDP) transferring water from the Danjiangkou Reservoir to North China, especially to Beijing and Tianjin to mitigate the crisis of water resource shortages (Fig. 1). The Danjiangkou Reservoir will have a storage capacity of approximately 33 billion m<sup>3</sup> when the existing dam is heightened from 145 m to 175 m (Wang and Ma, 1999). It is planned to extracting 95–130 billion m<sup>3</sup> water a year from the Danjiangkou Reservoir since 2014, and good water quality thus determines the fates of the SNWDP.

Water eutrophication is the major risk for the Danjiangkou Reservoir (Li et al., 2009; Tang et al., 2012). As a centralized drinking water source, water quality of the Danjiangkou Reservoir

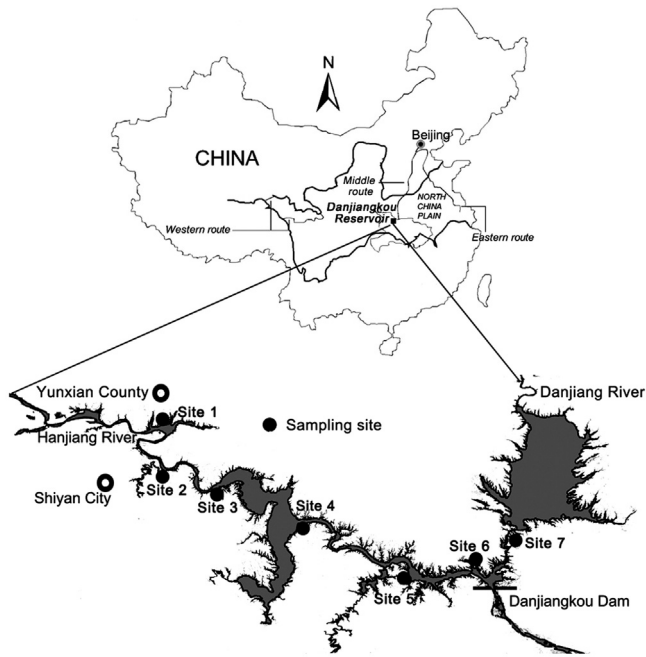
is required to match the Class II of the national standard GB 3838-2002 with total nitrogen (TN) and total phosphorus (TP) concentrations no more than 0.50 mg/L and 0.025 mg/L (Ministry of Environmental Protection, 2002), respectively. Anthropogenic inputs including agricultural runoff, urban domestics and industrial effluents contributes major nutrient loads to the Danjiangkou Reservoir. Algae blooms were already observed in 1992, 1998, 2000 and 2003 in some tributary estuaries and bays of the Danjiangkou Reservoir and phosphorus had been considered the limiting factor for algal production (Xiong and Liao, 2003). Between 2005 and 2006, water quality monitoring results further confirmed that phosphorus was the limiting nutrient factor for water eutrophication (Li et al., 2009). Increase in phosphorus concentrations implicates a great potential of algae bloom in the future considering that the water transfer project will enlarge the water storage capacity and sharply slow down the water velocity.

Release of nitrogen, phosphorus and other pollutants from submerged contaminated sediments will also potentially deteriorate water quality and threaten water supply safety. Previous studies have shown that sediment acts not only as a sink of nutrients but also as a source (Aigars and Carman, 2001). The nutrient release process has a significant impact on water quality and may result in continuous eutrophication in reservoirs, especially when external

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**Fig. 1.** Map of the Danjiangkou Reservoir showing (a) South to North Water Division Project (SNWDP) and (b) sampling sites of the water and sediment.

nutrient sources are under control. In 2010, sediment TP concentrations were firstly investigated from three representative sampling sites in the Danjiangkou Reservoir area, and recorded values ranged from 671 mg/kg to 953 mg/kg (Tang et al., 2012). High TP content in particular of loosely exchangeable phosphorus in the sediment is easily available to algae, and will therefore promote blooms after the completion of the dam heightening project.

The sorption of phosphorus on the sediments is one of the most important processes used to regulate the mobilization and dynamic behavior of phosphorus in the aquatic sediments (Appan and Wang, 2000). Mobilized sediment phosphorus may either be resorbed by other sediment components or released to the overlying water. Resorption or release mainly depends on the overall sediment compositions and phosphorus saturation (Istvánovics, 1994). It is demonstrated that phosphorus adsorption capacity is closely related to sediment composition, such as contents of organic matter, and Fe/Al oxides. In general, organic matter is positively correlated with phosphorus adsorption (Wang et al., 2009), while metals oxides play a key role in determining the phosphorus capacity because of the high specific surface of the Fe/Al hydroxides (Makris et al., 2005). Moreover, the maximal phosphorus adsorption capacity is also enhanced with the increase of the sediment total content of Fe + Al (Wang et al., 2009). For Danjiangkou Reservoir, however, the relationships between phosphorus adsorption performance and sediment compositions were not pronounced as in other freshwater bodies.

In the present study, seven representative water and surface sediment samples were collected from the Danjiangkou Reservoir to determine the nutrients in particular of phosphorus storage and spatial distribution characteristics. As a powerful method, sequential chemical extraction of phosphorus and Fe/Al oxides was conducted to explore the relationships between sediment composition variables and phosphorus storage and adsorption behavior. Finally, phosphorus adsorption process and mechanism was elucidated by batch adsorption kinetics and isotherm experiments and further mathematic model calculations.

## 2. Materials and methods

### 2.1. Water sampling and nutrient determination

In July 2012, water samples were collected from seven sampling sites (7 (Yunxian), 6 (Shendinghe), 5 (Xiaochuan), 4 (Longkou), 3 (Langhekou), 2 (Fenghuangshan) and 1 (Sigou), which were located from the upstream to the downstream of the Danjiangkou Reservoir (Fig. 1). Sampling bottles were soaked for 24 h in 10% hydrochloric acid and subsequently rinsed in pure water prior to use. According to Chinese Environmental Quality Standards for Drinking Water (GB3838-2002), 500 mL water samples were collected at an approximate depth of 30 cm with sampling sites depth of nearly 1.0 m. All water samples were analyzed on the same day for the following nutrient parameters: permanganate index ( $\text{COD}_{\text{Mn}}$ ), ammonia-nitrogen ( $\text{NH}_4\text{-N}$ ), nitrate-nitrogen ( $\text{NO}_3\text{-N}$ ), total nitrogen (TN), soluble reactive phosphorus (SRP) and TP according to the American standard methods (APHA, 1998).

### 2.2. Sediment sampling and composition analysis

Once finished the water sampling work, each surface sediment samples was immediately collected at the same water sampling sites (Fig. 1) with a grab style sampler. All the surface sediment samples were transported to the laboratory within sealed plastic bags, and natural air-dried and sieved with 100 mm-diameter meshes (obtained sub-sample with grain sizes less than 0.149 mm) before analysis. pH and organic matter contents for all the samples were determined according to the Chinese Soil Physical and Chemical Standard Methods (Bao, 2000).

#### 2.2.1. Phosphorus fractional analysis

Considering the similarities between sediment and soil, sequential extraction of phosphorus was undertaken to determine the phosphorus species according to the procedures recommended by The Chinese Soil Physical and Chemical Standard Methods (Bao, 2000). Exactly 25 mL of 1 mol/L  $\text{NH}_4\text{Cl}$  were added to 500 mg sediment (dry weight equivalent), and the suspension was shaken for 0.5 h. After shaking, the suspension was centrifuged at 3000 rpm for 10 min, and the supernatant was decanted. This process was repeated with 0.5 mol/L  $\text{NH}_4\text{F}$  (shaken for 1 h), 0.1 mol/L NaOH (shaken for 2 h, and shaken for 2 h again after standing for 16 h), 0.3 mol/L  $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 2\text{H}_2\text{O}$  (shaken for 0.5 h) and then with 0.5 mol/L  $\text{H}_2\text{SO}_4$  (shaken for 1 h). These fractions represented exchangeable P (Ex-P), P bound by Al oxides (Al-P), P bound by Fe oxides (Fe/P), occluded phosphorus (O-P) and calcium bound P (Ca-P), respectively. The TP concentration within the sediments was determined by treating the sub-sample at 720 °C, followed by 3 mol/L  $\text{H}_2\text{SO}_4$  extraction (Bao, 2000).

#### 2.2.2. Fe/Al fractional analysis

Free iron oxides (Fed), amorphous iron oxides (Feox) and iron present as pyrite (FeP) were analyzed by extracting with 0.3 mol/L  $\text{Na}_3\text{C}_6\text{H}_5\text{O}_7 \cdot 2\text{H}_2\text{O} + 1.0$  mol/L  $\text{NaHCO}_3$ , 0.1 mol/L  $\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O} + 0.2$  mol/L  $(\text{NH}_4)_2\text{C}_2\text{O}_4 \cdot \text{H}_2\text{O}$  and 0.1 mol/L  $\text{Na}_4\text{P}_2\text{O}_7 \cdot 10\text{H}_2\text{O}$ , iron content contained in the solution were then determined using phenanthroline spectrophotometric method (Bao, 2000). Exactly 10 mL of 1 mol/L KCl were added to 1000 mg sediment (dry weight equivalent), and the suspension was shaken for 0.5 h. After shaking, the suspension was centrifuged at 3000 rpm for 10 min, and the supernatant was decanted for aluminum contents determination using aluminum spectrophotometric method (Shao et al., 1998). This process was repeated with 0.2 mol/L HCl (shaken for 0.5 h). These fractions represent exchangeable aluminum oxides (ExAl) and adsorbed

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