



# Differences in major ions as well as hydrogen and oxygen isotopes of sediment pore water and lake water

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## Abstract

Isotopic and chemical compositions of pore water (PW) are highly relevant to environmental and forensic study. Five lake water (LW) samples and five sediment samples were collected to investigate the effects of pore sizes of sediments on PW chemistry and stable isotopes and determine mechanisms controlling their variations. Six pore water fractions were extracted from different-sized pores in each sediment sample at six sequential centrifugal speeds for chemical and isotopic analysis. The sediments consisted mainly of quartz, feldspar, and clay minerals. The hydrogen and oxygen isotopic compositions of PW are mainly controlled by the overlying LW, although the lag effect of exchange between overlying LW and PW results in isotopic differences when recharge of LW is quicker than isotopic exchange in PW. Identical isotopic compositions of PW from sediments with different pore sizes indicate that isotopic exchange of water molecules with different pore sizes is a quick process. The ratio of average total dissolved solid (TDS) concentration of PW to TDS concentration of LW shows a strong relationship with adsorption capacity of sediments, demonstrating that remobilization of ions bound to sediments mainly causes a chemical shift from LW to PW. Concentrations of  $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ , and  $\text{Cl}^-$  in PW remain unchanged, while concentrations of  $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{SO}_4^{2-}$  slightly increase with decreasing pore size. Chemical differences of PW from sediments with different pore sizes are governed by ion adsorption properties and surface characteristics of different-sized particles.

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**Keywords:** Pore size; Centrifugal extraction; Hydrogen and oxygen isotopes; Major ions; Remobilization

## 1. Introduction

The total volume of pore water (PW) occupying the pore spaces of sediments is around one-fifth of all the earth's water resources (Horn, 1964). PW is formed by infiltration and eventual trapping during sedimentation, where remobilization, mineral dissolution, degradation, diffusion, and the exchange

of PW with the overlying water column jointly determine chemical and isotopic compositions of PW, which play an important role in sediment-water exchange processes in aquatic ecosystems (Zhu et al., 2014) and also have the potential to yield important information on sediment quality (Vardy et al., 2015; Cleveland et al., 2017). Thus, the extraction and investigation of PW are particularly significant.

Currently, the most important extraction techniques used for PW extraction from sediment samples include centrifugation (Bath and Edmunds, 1981; Arnarson and Keil, 2000; Figueroa-Johnson et al., 2007; Delongchamp et al., 2010; Hong et al., 2011), pressure squeezing (Böttcher et al., 1997; Cyr et al., 2008; Kwong et al., 2015), dialysis (Mayer, 1976; Xu et al., 2012), and vacuum filtration (Concas et al., 2015;

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Sprenger et al., 2015). Of these techniques, centrifugation is most widely used for PW extraction due to its accuracy (Winger et al., 1998; Orłowski et al., 2016; Cleveland et al., 2017). Most studies have investigated chemical and isotopic characteristics of extracted PW. Watson et al. (1985) observed differences in PW chemistry upstream and downstream in the Tamar Estuary, and showed that these differences were controlled by re-oxidation of sulfide, denitrification, and waste dumping. Chemical variations of sediment PW from different depths were also investigated by Yang et al. (2003), illustrating that the local interaction of PW and fracture water plays a more important role in the evolution process of PW than diffusion or advection. Moreover, Kwong et al. (2015) studied stable isotopes of sediment PW from different depths in Cangzhou, on the North China Plain, and suggested that evaporation had been significant in the study area. In some other studies, seasonal variations in stable isotope and chemistry of sediment PW have been evaluated, showing seasonal differences to be governed by meteorological conditions such as temperature and precipitation (Karathanasis, 1991; Robertson and Gazis, 2006; Wildemeersch et al., 2015).

Other studies have investigated chemical compositions in PW from sediments with different pore sizes, but their results have been inconsistent. Chemical differences were observed in salinity, sodium, potassium, and calcium in PW from chalk by Edmunds and Bath (1976), but no chemical differences were observed by Kinniburgh and Miles (1983). Although Schön et al. (2016) showed isotopic differences in PW from sediments with three pore size gradients, the squeezing method used, which did not maintain constant temperature, might result in isotope fractionation during extraction. Therefore, studies on isotopic and chemical characteristics of PW from sediments with different pore sizes are necessary to expand our understanding of the evolution of PW along pore size gradients in sediments. To fill these knowledge gaps, this study intended to determine the impact of pore structure of sediments on stable isotopic and chemical compositions of PW.

The objectives of this study were to (1) identify the differences in chemical and isotopic compositions of LW and corresponding sediment PW, (2) characterize the effects of pore sizes (pore diameters) of sediments on chemical and isotopic compositions of PW, and (3) determine mechanisms controlling chemical and isotopic variations in LW from large-size pores to small-size pores during water-rock interaction.

## 2. Materials and methods

### 2.1. Materials

Five lake water samples and five sediment samples were collected from the Olympic Green (site O), the Summer Palace (site S), the Imperial Garden (site Y), Chaoyang Park (site C), and Yanqi Lake (site Q) in Beijing in November 2015 (Fig. 1). The four lakes at sites O, S, Y, and C had areas of 0.17, 1.9, 0.29, and 0.68 km<sup>2</sup>, respectively, and the maximum average water depth of 1.5 m was observed at site S (Zhang and Li, 2003). The lake area at site Q reached 2.3 km<sup>2</sup>, with a

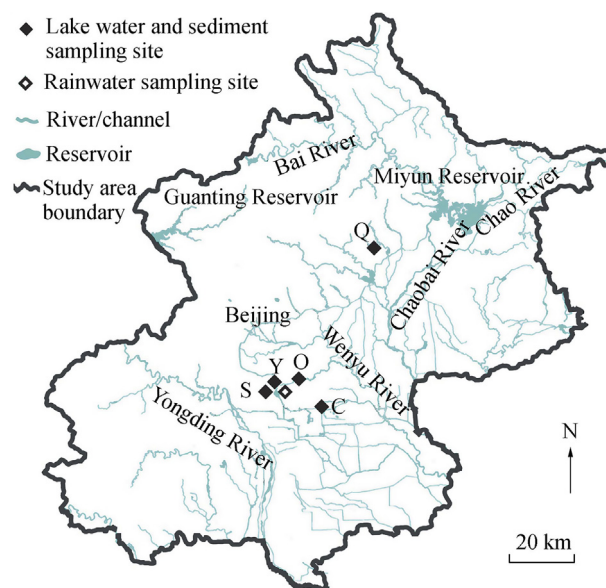


Fig. 1. Location of sampling sites of lake water, sediment, and rainwater in Beijing.

maximum water depth of 25 m. In addition, the annual average temperature of lake water at site Q was about 10°C, while at the other four sites it was around 13°C. At sites O and Y, the lakes are recharged by the Qinghe River, and the lake at site C is recharged by the Liangma River. The lake at site S is mainly recharged by local precipitation and adjacent springs in the monsoon season and by Miyun diversion water in the dry season, and the lake at site Q is mainly recharged by mountain rivers and local precipitation (Zhang and Li, 2003; Liu et al., 2013; Lan et al., 2016). All the water samples were filtered through 0.45- $\mu$ m membrane filters in the field for analysis in the laboratory. Water samples for major cation analyses were acidified to pH < 2 by the addition of 6 mol/L of ultra-pure HNO<sub>3</sub>. A subsample was left unacidified for anion determination. An aliquot of a sample was taken in an amber glass without headspace for oxygen and hydrogen isotope analysis.

Sediment samples were sealed in valve bags, and transported to the laboratory at 4°C. They were stored at -20°C in the laboratory for PW extraction within one week. In addition, three rainwater samples were collected for oxygen and hydrogen isotope analysis.

### 2.2. Methods

Predictions of the equilibrium partition model showed that the centrifuge method for PW extraction is the most accurate and precise for chemicals (Bufflap and Allen, 1995; Orłowski et al., 2016; Cleveland et al., 2017). Therefore, a high-speed refrigerated centrifuge (GL21M, Kaida) was used to extract PW in this study. The physics of water removal from porous materials by centrifugation are well understood, although the precise force distribution during the process is difficult to determine. Considering a column of water-saturated sediment under centrifugation, the applied tension

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