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Q5 Spatial and historical distribution of organic phosphorus driven 2 by environment conditions in lake sediments

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90 A R T I C L E I N F O

11 Article history:

12 Received 17 June 2016

13 Revised 10 December 2016

14 Accepted 9 January 2017

15 Available online xxxx

37 Keywords:

38 Organic phosphorus

39 Spatial and historical distribution

40 Environmental conditions

41 Sediment

42 Lake

43

A B S T R A C T

The chemistry of sedimentary organic phosphorus (OP) and its fraction distribution in 16
 sediments are greatly influenced by environmental conditions such as terrestrial inputs 17
 and runoffs. The linkage of OP with environmental conditions was analyzed on the basis of 18
 OP spatial and historical distributions in lake sediments. The redundancy analysis and OP 19
 spatial distribution results suggested that both NaOH-OP (extracted by NaOH-OP) and Re-OP 20
 (residual OP) in surface sediments from the selected 13 lakes reflected the gradient effects of 21
 environmental conditions and the autochthonous and/or allochthonous inputs driven by 22
 latitude zonality in China. The lake level and salinity of Lake Hulun and the runoff and 23
 precipitation of its drainage basin were reconstructed on the basis of the geochemistry 24
 index. This work showed that a gradient in weather conditions presented by the latitude 25
 zonality in China impacts the OP accumulation through multiple drivers and in many ways. 26
 The drivers are mainly precipitation and temperature, governing organic matter (OM) 27
 production, degradation rate and transportation in the watershed. Over a long temporal 28
 dimension (4000 years), the vertical distributions of Re-OP and NaOH-OP based on a dated 29
 sediment profile from HLH were largely regulated by the autochthonous and/or allochtho- 30
 nous inputs, which depended on the environmental and climate conditions and 31
 anthropogenic activities in the drainage basin. This work provides useful environmental 32
 geochemistry information to understand the inherent linkage of OP fractionation with 33
 environmental conditions and lake evolution. 34

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Published by Elsevier B.V. 36

49 Introduction

50 In the last decade, the importance of organic phosphorus (OP)
 51 has been gradually recognized as a potentially large pool of
 52 bioavailable P due to the fact that the OP fraction is at least
 53 as abundant as inorganic P in many waters and sediments
 54 (Turner et al., 2005). Therefore, many publications are available
 55 concerning OP decomposition (Dyhrman et al., 2006; Stets and

Cotner, 2008), fractionation (Ivanoff et al., 1998a; Turner et al., 56
 2005; Zhu et al., 2013) and OP compound determination using 57
³¹P nuclear magnetic resonance (NMR) spectroscopy (Ahlgren 58
 et al., 2005; Turner and Leytem, 2004), which has greatly 59
 elevated the acknowledged abundance of the OP fraction in 60
 the environment. However, knowledge gaps still exist regarding 61
 the factors driving the underlying processes governing the 62
 spatial and historical distribution of OP pools in lake sediments. 63

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Lakes generally have high rates of sedimentation and primary production holding information on organic carbon (OC), nitrogen and phosphorus, so short-term processes that affect organic matter (OM) delivery and burial are amplified in lake sediment records (Meyers, 2003), preserving valuable *in situ* historical information about the past condition of lakes and their environmental states (Zhang and Shan, 2008). Therefore, vertical concentration profiles of total OC (TOC) archived in lacustrine sediments are widely used as a tool to reconstruct the paleoenvironment and paleoclimate of the lakes' basins (Lü et al., 2008; Lüniger and Schwark, 2002; Meyers, 2003; Meyers and Ishiwatari, 1993; Wu et al., 2007) and monitor the effects of environment, climate change and/or anthropogenic activities on the environment (Bränvall et al., 2001; Herczeg et al., 2001; Meyers, 2003; Renberg et al., 2002; Routh et al., 2004). Usually, significant correlations are found between natural OM and OP, because OP is one of the main components of OM, and OM acts as the main carrier for OP in the environment. Accordingly, the chemistry of sedimentary OP and the distribution of its forms in sediments is also greatly influenced by environmental and climate conditions such as terrestrial inputs and runoffs. The geochemical phases of OP are important parameters in understanding the burial, diagenesis, bioavailability and environmental geochemical significance of OP in sediments.

As mentioned above, previous works mainly concern the OP abundance and the determination of its compounds in the surface sediments/soils, and some works are beginning to focus the distribution, degradation, diagenesis, transformation and their factors for OP in sediment profiles using fractionation techniques and/or ³¹P NMR (Ahlgren et al., 2006; Coelho et al., 2004; Ding et al., 2013; Edlund and Carman, 2001; Penn et al., 1995; Reitzel et al., 2007; Xu et al., 2013; Zhang et al., 2013) due to the environmental significance of OP. In an attempt to investigate the relationship between OP fractions in sediments and environmental conditions, this work hypothesized that the trends of OP fractions in surface sediments, which were collected from different lakes spanning a wide latitude range in China (spatial distribution), were driven by the environmental conditions of the lake basins, which were next in importance to latitude effects. Then the corresponding relationship of OP fractions to environmental conditions and lake evolution over a long time span (4000 years) was investigated based on a dated sediment profile from Lake Hulun (HLH). Accordingly, the objectives of this study were to (1) present the trends and factors of OP fractionation in surface sediments from 13 lakes on the basis of redundancy analysis and spatial distribution, (2) reconstruct the lake level and salinity and the runoff and precipitation of the lake basin, and discuss the relationship of OP fractionation with environmental conditions and climate changes according to the historical distribution of OP fractions in a dated sediment profile, (3) reveal the main processes or mechanism driving OP fractionation in the aquatic environment. This work provides some useful information on OP fractions archived in sediments and shares the novel concept and results with the environmental science community, especially the response of OP fractionation to environmental conditions over such large spatial and temporal dimensions.

1. Materials and methods

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1.1. Study area

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Lake Hulun (HLH) is located in northeastern Inner Mongolia, China. This is at the northeastern corner of the monsoon margin, and thus is sensitive to small variations in the East Asian monsoon (An, 2000; Wen et al., 2010). The highly variable climate is generally arid with an annual mean precipitation of 290 mm, of which three-fifths falls between July and August, an annual mean theoretical evaporation of 1600 mm, and an annual mean air temperature of -0.2°C . It once was the fifth largest lake in China, with an area of 2339 km² and an average depth of 5.7 m. During the last 35–50 years the lake has experienced a dramatic reduction in its size and water depth due to variations in climate. In 2011, its area had shrunk to 1770 km² with the mean depth of 3.0–3.5 m. Two major permanent rivers, the Herlun (Kerulen or Kelulun) and Urshen (Orxon or Orshun) River, feed the lake from the southwest and east, respectively (Fig. 1). The Dalanolom River, connected to the lake in the northeast, used to be an intermittent river. Since 1971 this river has been a channel used for water exchange between Lake Hulun and the Hailaer River. For much of the year, however, the water is stagnant, and occasionally, when its water table above sea level is lower than 545.1 m, there is a reverse flow of waters into the lake (Sun, 2010).

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1.2. Sediment sampling

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Sediments from HLH were sampled in August 2011. The surface sediments (0–10 cm) were collected using a KC collector mod B (Swedaq), while sediment cores were sampled applying a SA Beeker collector (Eijkelkamp). The sediment cores (HLH15, Fig. 1) were sectioned into 2-cm slices immediately after collection and stored in sealed polyethylene bags. Samples were transported to the laboratory in iceboxes ($<4^{\circ}\text{C}$). At the laboratory the sediments were freeze-dried, ground gently in a mortar and sieved through a standard 100-mesh sieve prior to analysis. In addition, the physicochemical data of sediments that were sampled in a similar manner from Lake Wuliangshuai (WLSH) are included in this study, which is detailed in previous work (Lü et al., 2016a).

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1.3. Experiments and methods

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A sequential extraction procedure modified by Ivanoff et al. (1998a) and Zhang et al. (2008) was used to fractionate the OP pools in the surface sediments and sediment cores. Following the procedure, OP in sediments is fractionated into a labile pool, a moderately labile pool, and a nonlabile pool. The labile pool is extracted with 0.5 mol/L NaHCO₃ at pH 8.5. The extracted P includes both OP (LOP) and inorganic P (IP) in pore water and sorbed on sediments. The moderately labile pool is extracted with 1.0 mol/L HCl (including HCl-OP, and IP), followed by 0.5 mol/L NaOH. The NaOH extract is acidified with concentrated HCl to separate the nonlabile fraction (humic acid fraction, HAOP) from the moderately labile fraction (fulvic acid fraction, FAOP). Finally, the highly resistant, nonlabile

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