Environmental Science
Processes & Impacts

Accepted Manuscript

This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this *Accepted Manuscript* with the edited and formatted *Advance Article* as soon as it is available.

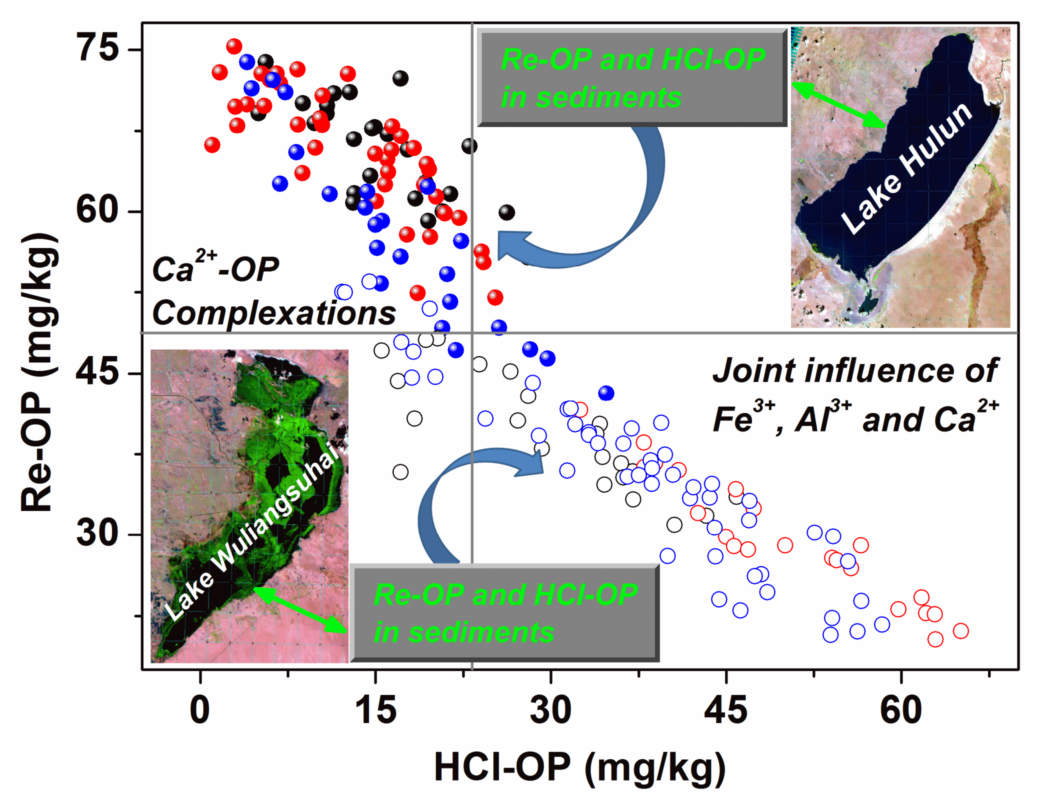
You can find more information about *Accepted Manuscripts* in the Information for Authors.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard Terms & Conditions and the Ethical guidelines still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.

ROYAL SOCIETY
OF CHEMISTRY

rsc.li/process-impacts

Graphic Abstract



Environmental Science: Processes & Impacts Accepted Manuscript

Environmental impact statement

The pattern of total OP (TOP) spatial distribution showed obvious decreasing trend from the west to the east lake districts in surface sediments from Lake Hulun (HLH). Whereas the spatial distribution pattern of dissolved OP (DOP) in overlying water, which presented different trend with TOP and total organic carbon (TOC) in surface sediments, indicated that the biological factors and terrestrial inputs showed joint influence on DOP distribution in HLH. The higher levels of Re-OP and lower levels of HCl-OP observed in HLH may reveal that calcium ions and their minerals were the key governing factors on OP fractionation in sediments from HLH. This work is helpful to understand the geochemical information archived in OP pools in lake sediments.

Environmental Science: Processes & Impacts Accepted Manuscript

Distribution characteristics of organic phosphorous in sediments from Lake Hulun, China

Changwei Lü[1, 2, 3], Jiang He[1, 2], Bin Zhou[3, 4], Rolf D. Vogt[3], Rui Guan[1], Weiying Wang[1], Le Zuo[1], Daohao Yan[1]

1. College of Environment and Resources, Inner Mongolia University, Huhhot 010021, China
2. Institute of Environmental Geology, Inner Mongolia University, Huhhot 010021, China
3. Department of Chemistry, University of Oslo, N-0315, Norway
4. Tianjin Academy of Environmental Sciences, Tianjin, 300191, China

Abstract

The amount of organic phosphorus (OP) and its distribution among different pools in lake sediments depend on biotic and abiotic processes driving the OP fractionation. Key abiotic factors governing these transformations processes between different OP fractions in sediments were studied on the basis of distribution characteristics of OP pools in sediments from Lake Hulun (HLH). The results showed that the order of size of OP pools was found in the surface sediments from Lake Hulun: Re-OP (residual OP) >> FAOP (fulvic acid fraction) ⩾ HCl-OP (OP extracted by HCl) > HAOP (humic acid fraction) >> LOP (labile OP); lower concentrations of LOP were found in lakes surface sediments with high pH (pH>9) of lakes overlying water indicating a governing role of pH in LOP degradation in aquatic environment.

Corresponding author, Email: lcw2008@imu.edu.cn; ndjhe@imu.edu.cn
Tel:+86-471-4992210
Fax: +86-471-4992210

Environmental Science: Processes & Impacts Accepted Manuscript

The pattern of total OP (TOP) spatial distribution showed obvious decreasing trend from the west to the east lake districts in surface sediments from HLH, which was attributed to the inputs of dust and dry grass driven by the prevailing wind and the finer grain size in the west lake districts. Whereas the spatial distribution pattern of dissolved OP (DOP) in overlying water, which presented different trend with TOP and total organic carbon (TOC) in surface sediments, indicated that the biological factors and terrestrial inputs showed joint influence on DOP distribution in HLH. The higher levels of Re-OP and lower levels of HCl-OP observed in HLH may reveal that calcium ions and their minerals were the key governing factors on OP fractionation in sediments from HLH. This work revealed the main abiotic process or mechanism driving the OP fractionations in aquatic environment helping to understand the geochemical information archived in OP pools in lake sediments.

Key words: organic phosphorus, fraction distribution, abiotic processes and factors, sediments, lakes

1 Introduction

As a major growth limiting nutrient in the global biosphere, phosphorus (P) always plays a key role in the biological productivity of aquatic ecosystems;[1] therefore elevated P inputs to the aquatic environment increase the risk of eutrophication and impacts on water quality. The predominant inorganic species is the most bioavailable P form and the emphasis for many decades has been on the study of its determination,[2, 3] abundance, bioavailability and transformation,[4-6] impacting mechanism on eutrophication.[7, 8] However in many waters and

Environmental Science: Processes & Impacts Accepted Manuscript

sediments, the organic phosphorus (OP) fraction, which are intrinsically linked within the aquatic environment, is at least as abundant as inorganic P.[9] In the last decade, the importance of OP is recognized gradually as a potentially large pool of bioavailable P. Therefore, there are many publications concerning on OP decomposition,[10, 11] fractionations [9, 12, 13] and OP compounds determination using ^{31}P nuclear magnetic resonance (NMR) spectroscopy,[14-18] which has largely elevated the acknowledged abundance of the OP fraction in environment. However, there still exists knowledge gaps' regarding the factors driving the underlying processes governing the quantitative and qualitative distribution of OP pools in lake sediments.

The accumulation and fraction distribution of sedimentary OP are shown to be greatly influenced by physical, chemical and biological factors and processes such as adsorption and desorption, complexation and precipitation, oxidation-reduction status (redox potential) and organic matter (OM) characteristics. In natural environment, in the presence of polyvalent cations (Fe^{3+}, Al^{3+}, and Ca^{2+}), the OP accumulation and distribution can be influenced by the formation of bridges between cations and adsorbents or co-precipitation with organic phosphate salts on the reacting surface.[19-22] Therefore, sorption onto sediment minerals, complexation and co-precipitation with polyvalent cations are listed as the main abiotic processes governing the accumulation, distribution and fate of OP in the environment.[9]

As mentioned above, previous works mainly concern on the OP abundance and its compounds determination in the surface sediments/soils, so far the studies on abiotic processes driving the OP fractionation are still fresh on the basis of the OP fractions distribution in sediments. The objects of this study were to (1) characterize OP fractions by

Environmental Science: Processes & Impacts Accepted Manuscript

sequential extraction in sediments collected from Lake Hulun (HLH), (2) discuss the distribution characteristics of OP pools and the governing factors, (3) reveal the main abiotic process or mechanism driving the OP fractionations in aquatic environment. This article aims to provide a better understanding of geochemical processes governing the distribution of OP pools in the Hulun Lake sediments.

2 Materials and Methods

2.1 Study Area

HLH is located in northeastern Inner Mongolia Autonomous Region, China. This is at the northeastern corner of the monsoon margin, and thus sensitive to small variations in the East Asian monsoon.[23, 24] 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 km^2 and an average depth of 5.7m. 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 shrank to 1770 km^2 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 (Figure 1). The Dalanolom River, connected to the lake in the northeast, was the channel used for water exchange between HLH and the Hailar River. The Hailar River are known as the upper reaches of the Argun River which is a part of the China-Russia border.

Environmental Science: Processes & Impacts Accepted Manuscript

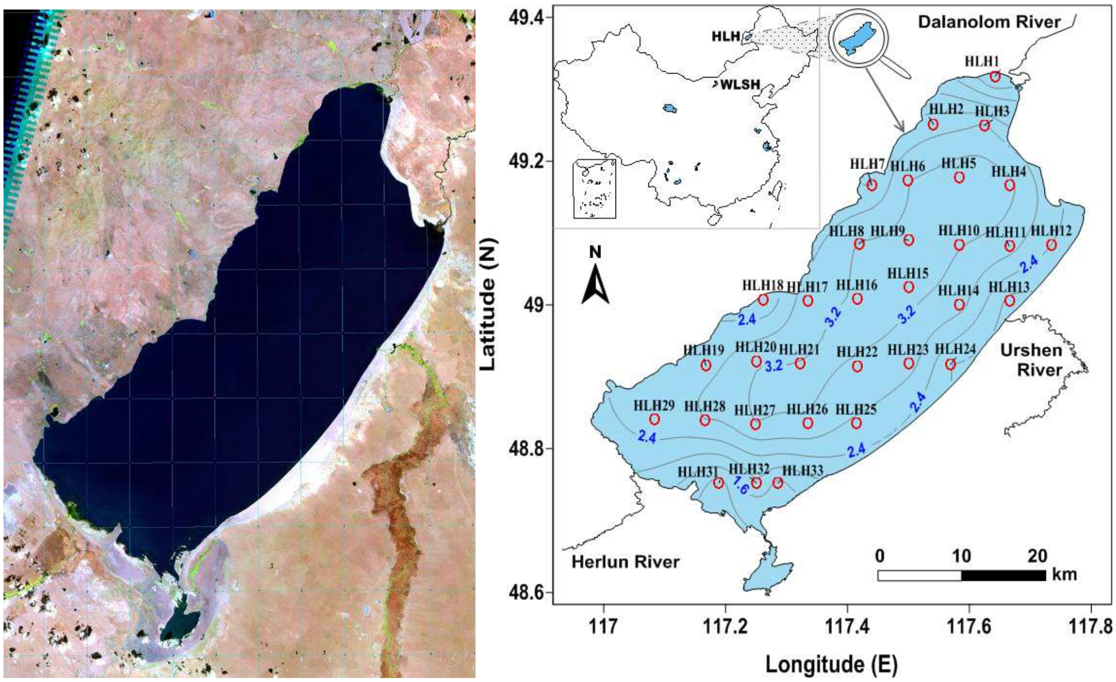


Figure 1 Remote sensing images and sampling sites in Lake Hulun

2.2 Sediment Sampling

Sediments from HLH were sampled in August 2011. The surface sediments (0-10cm) were collected using a KC collector mod B (Swedaq), while sediment cores were sampled applying a SA Beeker collector (Eijkelkamp). The sediment cores (HLH5, HLH15, HLH22, Figure 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 ºC). At the laboratory the sediments were freeze-dried, ground gently in a mortar and sieved through a standard 100-mesh sieve (150 µm) prior to analysis. Simultaneously, overlying water samples were collected for all sampling sites for determination of dissolved OP (DOP). These water samples were filtered through 0.45 mm polycarbonate filters, acidified to pH 1.0 with hydrochloric acid, and stored refrigerated in glass bottles. In addition, Lake Wuliangsuhai (WLSH), as a typical macrophyte-dominanted lake, is the second largest lake in the Inner

Environmental Science: Processes & Impacts Accepted Manuscript

Mongolia plateau, China.[25] Thus physiochemical data of sediments and overlying water that were sampled in a similar manner from WLSH were included in this study.

2.3 Experiments and methods

A sequential extraction procedure, modified by Ivanoff et al[12] and Zhang et al[26] (Figure 2) 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 M $NaHCO_3$ 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 M HCl (including HCl-OP, and IP), followed by 0.5 M 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 fraction (Re-OP) is determined by ashing the residue from the NaOH extraction at 550 °C for 1 h, followed by dissolution in 1.0 M H_2SO_4. The moderately labile OP (MLOP) and nonlabile OP (NLOP) are then calculated as the sum of HCl-OP and FAOP, and the sum of HAOP and Re-OP, respectively.

In all cases, P concentration in the extracts is determined colorimetrically by the phospho-molybdate method.[27] Acid or alkaline extracts are neutralized prior to P determinations. Total P (TP) in the extracts is measured after an aliquot is digested with 2.5 M H_2SO_4 and potassium persulfate ($K_2S_2O_8$), according to the method of Bowman,[28] as modified by Thien and Myers.[29] The OP in the extracts was calculated as the difference between TP and IP.

Environmental Science: Processes & Impacts Accepted Manuscript

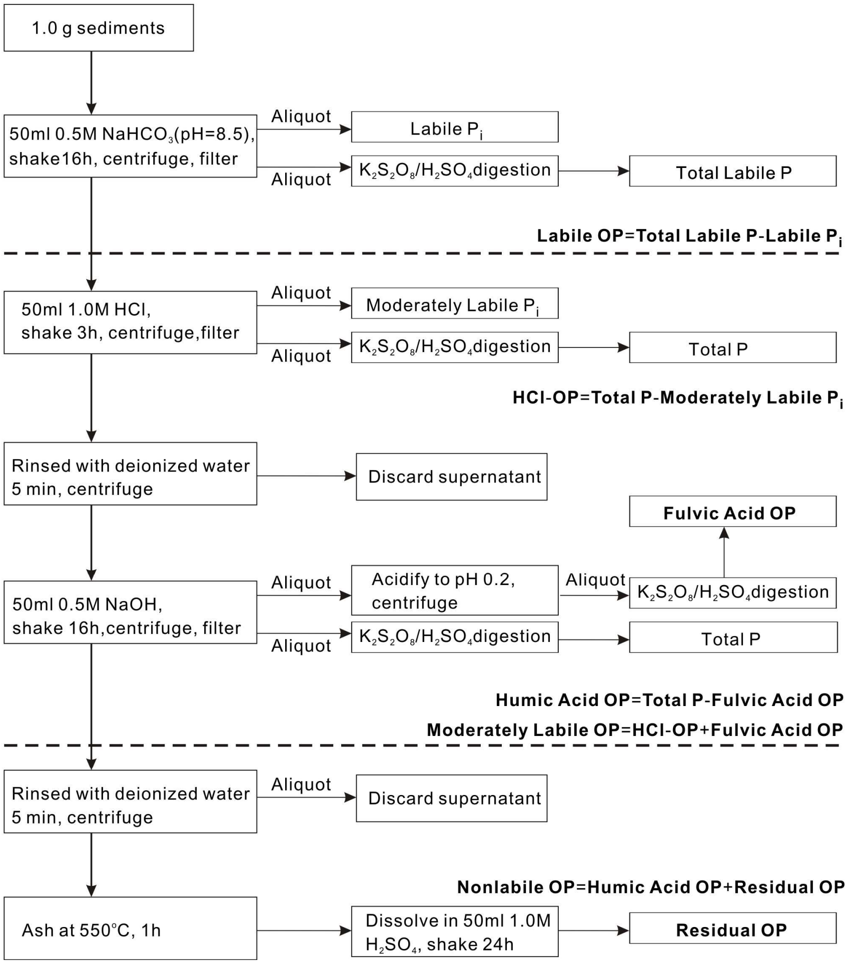


Figure 2 The sequential extraction procedure for OP

All filters, bottles and filtration apparatus were acid cleaned prior to use. All acids used were of guaranteed reagent grade. The water used for reagent preparation, dilution and washing was pure water (Milli-Q water). A standard deviation below 5% was achieved in all the analytical methods that were used.

Statistical analysis, such as spatial distribution and correlation analysis, was performed using Origin 8.5, Surfer 10.0 and SPSS Statistics 21.0.

Environmental Science: Processes & Impacts Accepted Manuscript

3 Results and Discussion

3.1 Fraction distributions of OP in surface sediments

As can be seen from Table 1, the concentration ranges of total extracted OP (TOP) were 72.2-210 mg/kg with the means of 146 mg/kg in surface sediments from HLH. The ranges of MLOP and NLOP were 14.7-92.6 and 54.1-133 mg/kg in surface sediments from HLH with the relative contribution of 19.3-56.7% and 41.6-80.2% to TOP, respectively. The average concentration of LOP found in surface sediments from HLH was 4.54 mg/kg with the average percentages of 3.04%. For WLSH, The concentration of TOP ranged from 91.2 to 169 mg/kg, with a mean value of 122 mg/kg in surface sediments; Though the mean concentration of LOP was only 2.02 mg/kg, constituting a mere 1.71% of the TOP. The ranges of MLOP and NLOP were 36.8-72.5 and 36.4-108 mg/kg in surface sediments from WLSH, respectively (Table 1). Summarily, the MLOP pool was made up of a relatively similar contribution of FAOP and HCl-OP, while the NLOP consisted of mainly Re-OP, with a minor amount of HAOP. The Re-OP pool therefore alone accounted for between 32.4-76.7% and 31.8-56.1% of the TOP in the surface sediments from HLH and WLSH, respectively. This means that the following order of size of OP pools was found in the surface sediments from the two lakes: Re-OP >> FAOP ≥ HCl-OP > HAOP >> LOP.

Environmental Science: Processes & Impacts Accepted Manuscript

Table 1 OP concentrations in surface sediments from Lake Wuliangsuhai and Hulun

OP fractions			**WLSH (n=10)**		**HLH (n=29)**	
			mg/kg	**%**	**mg/kg**	**%**
Labile OP (LOP)		**Min**	-	-	0.55	0.52
		Max	5.17	4.75	19.0	9.38
		Mean	**2.02**	**1.71**	**4.54**	**3.04**
Moderately Labile OP (MLOP)	**HCl-OP**	**Min**	4.37	4.66	2.34	1.44
		Max	51.6	41.0	54.9	37.0
		Mean	**25.9**	**21.6**	**22.5**	**16.0**
	Fulvic Acid-P (FAOP)	**Min**	19.9	16.6	4.14	5.12
		Max	43.8	34.6	75.5	37.3
		Mean	**28.7**	**23.8**	**34.9**	**22.0**
Non-labile OP (NLOP)	**Humic Acid-P (HAOP)**	**Min**	0.26	0.21	1.33	1.72
		Max	29.9	17.6	24.6	16.3
		Mean	**10.4**	**7.85**	**12.3**	**8.14**
	Residual OP (Re-OP)	**Min**	30.2	31.8	41.9	32.4
		Max	78.4	56.1	112	76.7
		Mean	**55.0**	**45.0**	**72.1**	**50.8**
MLOP (HCl-OP + FAOP)		**Min**	36.8	35.1	14.7	19.3
		Max	72.5	61.8	92.6	56.7
		Mean	**54.6**	**45.4**	**57.4**	**38.0**
NLOP (HAOP + Re-OP)		**Min**	36.4	33.5	54.1	41.6
		Max	108	63.9	133	80.2
		Mean	**65.5**	**52.9**	**84.3**	**58.9**
Total Extracted OP (TOP)		**Min**	91.2		72.2	
		Max	169		210	
		Mean	**122**		**146**	

Comparatively, WLSH lake is characterized as a shallow macrophyte-dominated eutrophic lake. There were much of root exudates, such as citric and malic acid, in the sediments due to widely distributed *Phragmites communis* Trirn. and *Potamogeton pectinatus* L.. These root exudates present the properties of organic acids which can extract Re-OP from sediment. Our previous studies showed that MBP (microbial biomass phosphorus) varied from 16.4 to 95.5 nmol/g·dw with the average of 58.1 nmol/g·dw in surface sediments from WLSH.[30] As a result of the joint influence of microbial degradation and rhizosphere effects,

Environmental Science: Processes & Impacts Accepted Manuscript

the average concentrations and percentages of Re-OP and NLOP in surface sediments from HLH were higher than those in WLSH, while the average percentage of HCl-OP and MLOP in surface sediments from HLH lower than those in WLSH (Table 1). It likely indicated that Re-OP may be transformed into labile P pools.[31, 32] This finding is helpful to find direct evidence for OP degradation in sediments and recognize the knowledge of OP bioavailability in aquatic environment.[31, 32] Furthermore, the average relative contribution of FAOP in surface sediments from HLH were equal to that of WLSH, similar as HAOP (Table 1), revealing the same degree of humification in both lakes.

Compared with other lakes of China [26, 33] (Figure 3), lower concentrations of LOP were found in Lake Hulun, Wuliangsuhai, Qinghai and Chenghai with the pH value of aqueous environments higher than 9.0, while higher LOP concentrations with lower pH in other lakes. Generally, pH is a key factor for molecular characteristics of dissolved OM in aquatic environment.[34, 35] The lower LOP found in lakes (pH > 9.0) may be attributed to the relatively high solubility of LOP and its strong interaction at the water-sediment interface especially in alkali aqueous environments,[36] which is as a result of the high solubility of OM in alkali aqueous environments. Furthermore, bacterial decomposition of OM is inhibited by low pH, nutrient poor conditions and low oxygen availability. [37, 38] Then more LOP was decomposed in aquatic environment with high pH such as Lake Hulun, Qinghai, Wuliangsuhai and Chenghai.

Environmental Science: Processes & Impacts Accepted Manuscript

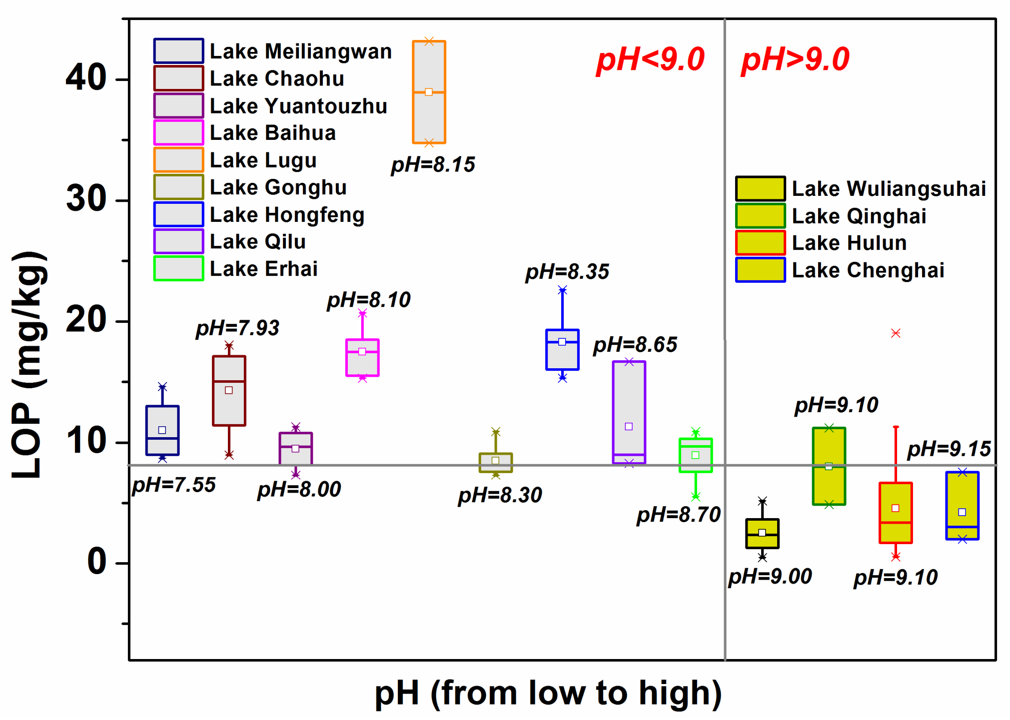


Figure 3 Relationship between LOP in surface sediments and water pH

3.2 Spatial distribution of OP pools

The pattern of TOP spatial distribution showed obvious decreasing trend from the west to the east lake districts (Figure 4), which was similar to the TOC spatial distribution in HLH surface sediments. Geomorphological, relatively hilly terrains and semi-arid grassland as main vegetation on the west bank of HLH lead to frequent soil erosion during rainy season. It is rationalized due to < 40% vegetational cover and the terrain topology characterized as high in the west and lower in the east. As a result, more import of suspended matter, associated OM and nutrients from the HLH west bank are anticipated. This organic matter and nutrients loads from non-point sources carried by overland runoffs are believed as one of the main nutrient sources of HLH. Furthermore, the prevailing wind direction is from the northwest to the southeast. The amounts of dust and dry grass deposited into HLH is estimated to be

Environmental Science: Processes & Impacts Accepted Manuscript

6.44×10^4 t and 3350 t each year, respectively, since the northwest lake shoreline is about 150 km. Previous studies have shown that silty clay is the dominated in the sediments from the northwest lake districts, while it is major sandy deposit on the south and east lake bottom.[39] Therefore, on the one hand, there are more inputs of dust and dry grass driven by the prevailing wind in the west lake districts; on the other hand, the inputs will be conserved effectively due to the finer grain size in these lake districts. The similar decreasing trends of TOC and TOP were found in the surface sediments from HLH.

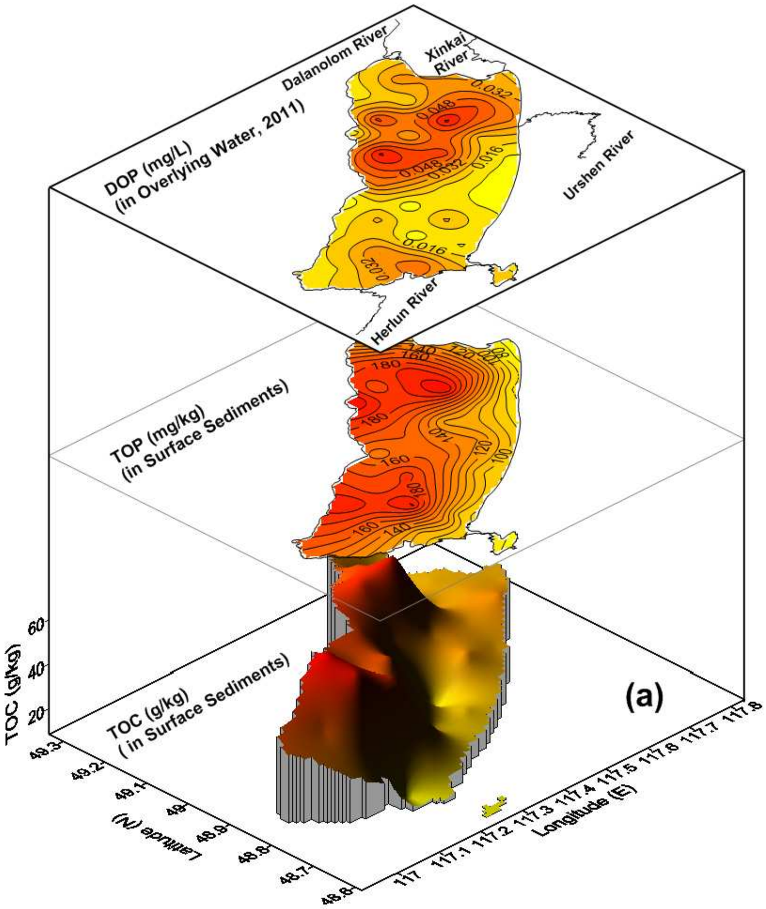


Figure 4 OP spatial distributions in surface sediments from HLH

Actually, DOP represents as a major reservoir of dissolved P in aquatic environments, comprising up to 75% of total dissolved P (TDP).[40-42] Over the past decades, DOP has been

Environmental Science: Processes & Impacts Accepted Manuscript

increasingly recognized as a critical driver to aquatic biological production and ecosystem structure and function.[43, 44] In overlying water from HLH, DOP concentrations accounted for 24-54% of TDP, suggesting its potential as major P source for the primary producers.[45, 46] In general, P levels in water column could be influenced by many factors such as elevated P inputs, internal loads release, hydrobios absorption and adsorption/desorption across the interface of the water-sediments. The spatial distribution pattern of DOP was different from the ones of TOC and TOP. From Figure 4, high levels of DOP were found in the areas near to the lake inlets, suggesting the governing role of the terrestrial inputs. Moreover, the midwest lake districts were also found with high level of DOP. This observation can be explained by higher level of measured biomass of phytoplankton in the midwest lake districts than in the southeast.[47] It has been reported before that DOP in aquatic environments may conceivably arise from the secretions and excretions of living bacteria, plants, and animals or from the decomposition of dead organisms.[48] Therefore, the biological factors and terrestrial inputs jointly governed the distribution pattern of DOP in overlying water from HLH.

3.3 Fraction distribution of OP in sediment cores

Calcite precipitation is known to induce co-precipitation of inorganic phosphate and thus, may potentially have important effects on the eutrophic status of aquatic environments.[49-51] Beside IP, calcium ions and their minerals can complex with OP in soils or sediments.[22, 52] For instance, precipitation of calcium salts with calcite occurs at even very low concentrations of *myo*-inositol hexakisphosphate [53] due to the simultaneous occurrence of adsorption and precipitation of insoluble calcium-phytate.[54] This explains why the OP content is positively correlated to the calcium content.[52] Furthermore, OP prefer binding with Ca^{2+} to

Environmental Science: Processes & Impacts Accepted Manuscript

monovalent cations even at low concentrations of calcium,[55] since Ca^{2+} cause a surface-excess of positive charge in aquatic environment. As can be seen from Figure 5, the concentrations of Re-OP in sediment cores from HLH were tested significantly higher than those in WLSH ($p<0.001$); while HCl-OP presented significantly lower levels in HLH than those in WLSH ($p<0.001$). Based on the sequential extraction procedure used in this work, OP extracted by 1.0 M HCl (HCl-OP) includes OP fractions adsorbed to Fe/Al oxides/hydroxides or OP salts co-precipitated with calcium ions;[56, 57] while Re-OP mainly include the complexations of OP with calcium ions and their minerals. Accordingly, the higher levels of Re-OP and lower levels of HCl-OP observed in HLH (Figure 5) may reveal that calcium ions and their minerals were the key governing factors on OP fractionation in sediments from HLH.

Oxides/hydroxides of Fe and Al [56, 57] can strongly adsorb OP on their surfaces in soils [20] and sediments,[21] even can play a governing role in most acidic soils and sediments. The Fe-bound P fraction (Fe-P) and Al-bound P fraction (Al-P) were extracted by a sequential extraction procedure[3, 58] in our previous work. Accordingly, the average concentrations of Fe-P and Al-P were 11.10 and 7.15 mg/kg in sediment cores from WLSH, respectively.[59] The pools of Al-P and Fe-P were much smaller in sediment cores from HLH, i.e. only 2.04 and 1.54 mg/kg, respectively. Meanwhile, significant correlations between HCl-OP and pools of Al-P and Fe-P were found in the cores from WLSH. These results therefore indicated that Fe and Al oxyhydroxides play a more important role in OP accumulation and fractionation in WLSH than in HLH due to that the amount of iron oxide in the substrate governs the extent of OP adsorption.[60, 61] Accordingly, HCl-OP in cores from WLSH presented higher levels and

Environmental Science: Processes & Impacts Accepted Manuscript

wider variation ranges (Figure 5) revealing the joint influence of Fe, Al and Ca on OP fractionation.

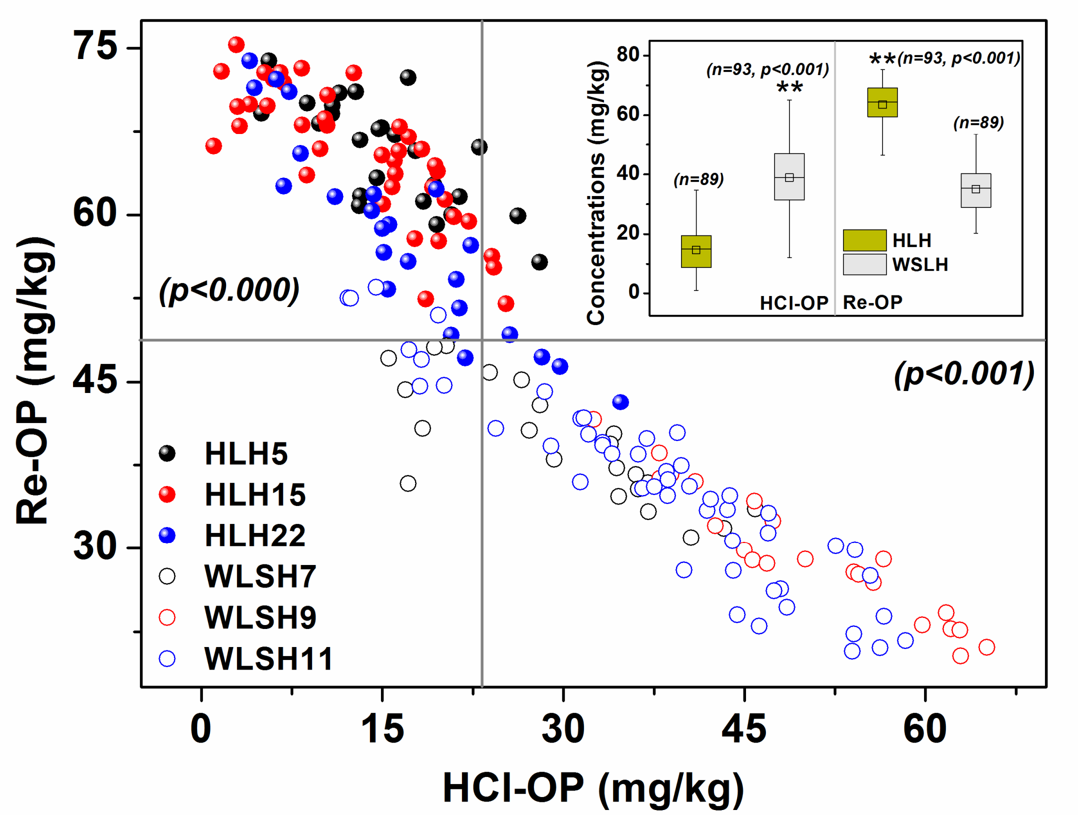


Figure 5 Distribution of HCl-OP and Re-OP in sediment cores

OP fractionations is governed by many processes and factors.[9] Thus abiotic process and factors may be the main driven forces for OP re-distribution in sediments due to the lack of obvious evidences on OP degradation in the profiles. Generally, NaOH-OP is closely related with OM, while HCl-OP with Fe/Al oxyhydroxides. Accordingly, transformations between NaOH-OP and HCl-OP were likely governed by the binding capacity changes of OM and Fe/Al oxyhydroxides.[31] In this work, whereas no significant correlations between the Fe/Al oxyhydroxides and HCl-OP were found in the cores from HLH; significant correlation between the ratio of Fe-P/TOC and the ratio of HCl-OP/NaOH-OP was found in the sediment

Environmental Science: Processes & Impacts Accepted Manuscript

cores from HLH (Figure 6). The OP transformation between NaOH-OP (including FAOP and HAOP) and HCl-OP might reflect the variation of binding capacity of OM and iron oxides for OP.[62] The ratio of Fe-P/TOC increased from 0.06 to 0.1 from top to bottom in sediment profiles from HLH (Figure 6) indicating the increasing binding capacity of iron oxides compared to OM.[31]

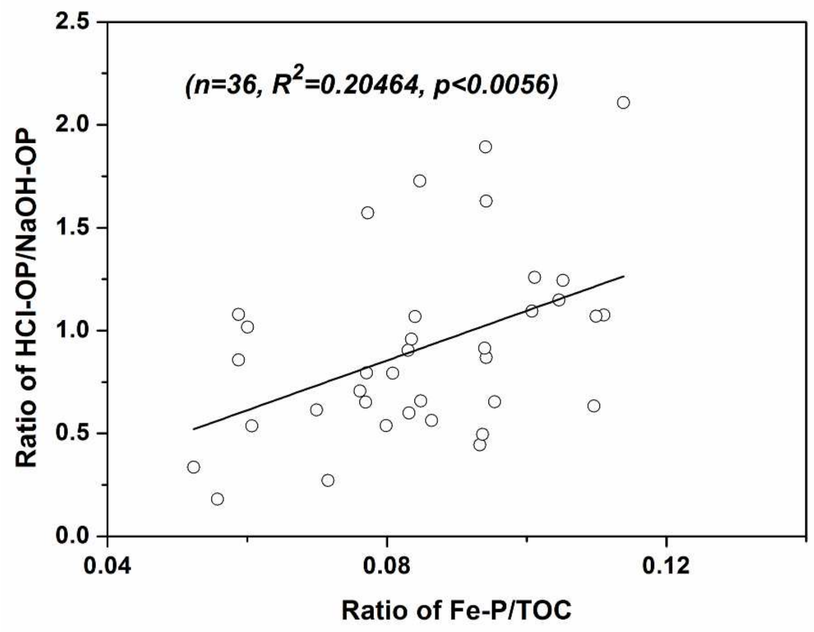


Figure 6 Correlations between the ratios of Fe-P/TOC and HCl-OP/NaOH-OP in HLH

4 Conclusions

The order of size of OP pools was found in the surface sediments from Lake Hulun: Re-OP >> FAOP ≥ HCl-OP > HAOP >> LOP. The distribution characteristics of LOP in surface sediments from 13 lakes in China indicated that high pH (pH>9) play a governing role in LOP degradation in aquatic environment.

Similar to TOC, the pattern of TOP spatial distribution showed obvious decreasing trend from the west to the east lake districts in surface sediments from HLH, which was attributed

Environmental Science: Processes & Impacts Accepted Manuscript

to the inputs of dust and dry grass driven by the prevailing wind and the finer grain size in the west lake districts. Whereas the spatial distribution pattern of DOP in overlying water, which presented different trend with TOC and TOP in surface sediments, indicated that the biological factors and terrestrial inputs showed joint influence on DOP distribution in HLH.

The higher levels of Re-OP and lower levels of HCl-OP observed in HLH may reveal that calcium ions and their minerals were the key governing factors on OP fractionation in sediments from HLH. The lower levels of Re-OP and the higher levels of HCl-OP indicated the joint influence of multi-factors such as Fe, Al, Ca, microbial degradation and rhizosphere effects on OP distribution characteristics in sediments from WLSH. This work revealed the main abiotic process or mechanism driving the OP fractionations in studied aquatic and surface sediment environment.

Acknowledgements

This work was supported by National Natural Science Foundation of China (No. 41003049, 40863003), Natural Science Foundation of Inner Mongolia Autonomous Region of China (2015MS0404) and assisted by the Research council of Norway (No. 209687E40).

References

1. P. J. Worsfold, P. Monbet, A. D. Tappin, M. F. Fitzsimons, D. A. Stiles and I. D. McKelvie, *Analytica Chimica Acta*, 2008, **624**, 37-58.
2. P. J. Worsfold, L. J. Gimbert, U. Mankasingh, O. N. Omaka, G. Hanrahan, P. C. Gardolinski, P. M. Haygarth, B. L. Turner, M. J. Keith-Roach and I. D. McKelvie, *Talanta*, 2005, **66**, 273-293.
3. K. C. Ruttenberg, *Limnology and Oceanography*, 1992, **37**, 1460-1482.
4. X. Jin, X. Jiang, Y. Yao, L. Li and F. C. Wu, *Science of the Total Environment*, 2006, **357**, 231-236.
5. P. Tallberg, P. Tréguer, C. Beucher and R. Corvaisier, *Estuarine, Coastal and Shelf Science*, 2008, **76**,

Environmental Science: Processes & Impacts Accepted Manuscript

85-94.

6. C. van der Zee, N. Roevros and L. Chou, *Marine Chemistry*, 2007, **106**, 76-91.
7. S. Wang, X. Jin, H. Zhao, X. Zhou and F. Wu, *Journal of Environmental Management*, 2008, **88**, 845-852.
8. B. A. Lake, K. M. Coolidge, S. A. Norton and A. Amirbahman, *Science of the Total Environment*, 2007, **373**, 534-541.
9. B. L. Turner, E. Frossard and D. S. Baldwin, *Organic phosphorus in the environment*, CABI Publishing, Cambridge, 2005.
10. S. Dyhrman, P. Chappell, S. Haley, J. Moffett, E. Orchard, J. Waterbury and E. Webb, *Nature*, 2006, **439**, 68-71.
11. E. G. Stets and J. B. Cotner, *Limnology and Oceanography*, 2008, **53**, 137.
12. D. Ivanoff, K. Reddy and S. Robinson, *Soil Science*, 1998, **163**, 36-45.
13. Y. R. Zhu, F. C. Wu, Z. Q. He, J. Y. Guo, X. X. Qu, F. Z. Xie, J. P. Giesy, H. Q. Liao and F. Guo, *Environmental Science & Technology*, 2013, **47**, 7679-7687.
14. B. L. Turner and A. B. Leytem, *Environmental Science & Technology*, 2004, **38**, 6101-6108.
15. X. Bai, S. Ding, C. Fan, T. Liu, D. Shi and L. Zhang, *Environmental Pollution*, 2009, **157**, 2507-2513.
16. R. Zhang, F. Wu, Z. He, J. Zheng, B. Song and L. Jin, *Journal of Environmental Quality*, 2009, **38**, 353-359.
17. J. Ahlgren, K. Reitzel, R. Danielsson, A. Gogoll and E. Rydin, *Water Research*, 2006, **40**, 3705-3712.
18. J. Ahlgren, L. Tranvik, A. Gogoll, M. Waldebäck, K. Markides and E. Rydin, *Environmental Science & Technology*, 2005, **39**, 867-872.
19. L. Celi and E. Barberis, *In: Turner B L, Frossad E, Baldwin D S. Organic phosphorus in the environment. Oxford University Press: Boston*, 2005, 113-132.
20. G. Anderson and E. Z. Arlidge, *Journal of Soil Science*, 1962, **13**, 216-224.
21. C. De Groot and H. Golterman, *Hydrobiologia*, 1993, **252**, 117-126.
22. W. A. House and F. H. Denison, *Science of the Total Environment*, 2002, **282**, 341-351.
23. Z. An, *Quaternary Science Reviews*, 2000, **19**, 171-187.
24. R. Wen, J. Xiao, Z. Chang, D. Zhai, Q. Xu, Y. Li, S. Itoh and Z. Lomtatidze, *Quaternary Research*, 2010, **73**, 293-303.
25. C. W. Lü, J. He, H. M. Sun, H. X. Xue, Y. Liang, S. Bai, Y. Sun, L. L. Shen and Q. Y. Fan, *Environmental Geology*, 2008, **55**, 1279-1289.
26. R. Zhang, F. Wu, C. Liu, P. Fu, W. Li, L. Wang, H. Liao and J. Guo, *Environmental Pollution*, 2008, **152**, 366-372.
27. J. Murphy and J. Riley, *Analytica Chimica Acta*, 1962, **27**, 31-36.
28. R. Bowman, *Soil Science Society of America Journal*, 1989, **53**, 362-366.
29. S. Thien and R. Myers, *Soil Science Society of America Journal*, 1992, **56**, 814-818.
30. H. F. Mao, J. He, C. W. Lü, Y. Liang, H. L. Liu and F. J. Wang, *Journal of Agro-Environment Science*, 2010, **29**, 2406-2412. (in Chinese).
31. C. Fan, Y. Zhou and Q. Wu, *The Sediment-water Interface of Lakes: Processes and Effects*, Science Press, Beijing, 2013.
32. D. Xu, S. Ding, B. Li, X. Bai, C. Fan and C. Zhang, *Journal of Environmental Sciences*, 2013, **25**, 637-644.
33. S. Huo, F. Zan, B. Xi, Q. Li and J. Zhang, *Journal of Environmental Monitoring*, 2011, **13**, 1088-1095.
34. V.-N. Roth, T. Dittmar, R. Gaupp and G. Gleixner, *Geochimica et Cosmochimica Acta*, 2013, **123**,

Environmental Science: Processes & Impacts Accepted Manuscript

93-105.

35. R. D. Vogt, J. Akkanen, D. O. Andersen, R. Brüggemann, B. Chatterjee, E. Gjessing, J. V. Kukkonen, H. E. Larsen, J. Luster and A. Paul, *Aquatic Sciences*, 2004, **66**, 195-210.

36. O. Oluyedun, S. Ajayi and G. Van Loon, *Science of the Total Environment*, 1991, **106**, 243-252.

37. E. Gorham, *Ecological Applications*, 1991, **1**, 182-195.

38. P. D. Moore, *Environmental Conservation*, 2002, **29**, 3-20.

39. L. Ji, W. Xia, L. Xiang and S. Wang, *Journal of Lake Sciences*, 1994, **6**, 227-232. (in Chinese).

40. C. R. Benitez-Nelson, *Earth-Science Reviews*, 2000, **51**, 109-135.

41. J. Wu, W. Sunda, E. A. Boyle and D. M. Karl, *Science*, 2000, **289**, 759-762.

42. E. Monaghan and K. Ruttenberg, *Limnology and Oceanography*, 1999, **44**, 1702-1714.

43. S. T. Dyhrman, P. D. Chappell, S. T. Haley, J. W. Moffett, E. D. Orchard, J. B. Waterbury and E. A. Webb, *Nature*, 2006, **439**, 68-71.

44. A. Paytan and K. McLaughlin, *Chemical Reviews*, 2007, **107**, 563-576.

45. C. Labry, A. Herbland and D. Delmas, *J. Plank. Res*, 2002, **24**, 97-117.

46. J. N. Boyer, S. K. Dailey, P. J. Gibson, M. T. Rogers and D. Mir-Gonzalez, *Hydrobiologia*, 2006, **569**, 71-85.

47. Z. F. Jiang, C. Y. Li, S. Zhang, K. L. Jia and X. H. Shi, *Journal of Agro-Environment Science*, 2011, **30**, 726-732. (in Chinese).

48. R. Johannes, *Limnol. Oceanogr*, 1964, **9**, 224-234.

49. A. Hartley, W. House, M. Callow and B. Leadbeater, *Water Research*, 1997, **31**, 2261-2268.

50. W. A. House and F. H. Denison, *Science of the Total Environment*, 1997, **205**, 25-49.

51. H. Jarvie, C. Neal, A. Warwick, J. White, M. Neal, H. Wickham, L. Hill and M. Andrews, *Science of the Total Environment*, 2002, **282-283**, 353-373.

52. A. F. Harrison, *Soil organic phosphorus: a review of world literature*, CAB International Wallingford, UK, 257 pp, 1987.

53. R. Celis, M. C. Hermosin and J. Cornejo, *Environmental Science & Technology*, 2000, **34**, 4593-4599.

54. L. Celi, M. Presta, F. Ajmore-Marsan and E. Barberis, *Soil Science Society of America Journal*, 2001, **65**, 753-760.

55. M. Presta, L. Celi and E. Barberis, *Competizione tra fosfato e inositol esafosfato su goethite. In: Proceedings of the XVIII SICA Congress, Catania, Italy, 22-24 September, pp. 100-108*, 2000.

56. E. Rydin, *Water Research*, 2000, **34**, 2037-2042.

57. K. Lukkari, M. Leivuori and A. Kotilainen, *Biogeochemistry*, 2009, **96**, 25-48.

58. G. W. Zhu, B. Q. Qin, L. Zhang and L. C. Luo, *Pedosphere*, 2006, **16**, 726-734.

59. C. W. Lü, J. He, H. M. Sun, X. D. Gao and H. X. Xue, *Acta Sedimentologica Sinica*, 2008, **26**, 508-512. (in Chinese).

60. G. Anderson, E. G. Williams and J. O. Moir, *Journal of Soil Science*, 1974, **25**, 51-62.

61. H. K. Pant, A. C. Edwards and D. Vaughan, *Biology and Fertility of Soils*, 1994, **17**, 196-200.

62. D. Xu, S. Ding, B. Li, F. Jia, X. He and C. Zhang, *Journal of Soils and Sediments*, 2012, **12**, 909-920.

Environmental Science: Processes & Impacts Accepted Manuscript