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Research Article

Phosphorus Buildup and Release Risk Associated with Agricultural Intensification in the Estuarine Sediments of Chaohu Lake Valley, Eastern China

Phosphorus (P) accumulation and the risk of P release were studied in the estuarine sediments of the Chaohu Lake Valley in Eastern China. Rapid agricultural intensification has occurred in this area since the 1980s, resulting in the extensive use of phosphate fertilizers. This extensive use of phosphate has affected P processes throughout the valley and resulted in the total phosphorus (TP) contents in the substrates and surface sediments increasing from 374 to 537 mg/kg, on average. Of all the forms of P evaluated in this study, residual P (Res-P) was present at the highest levels and accounted for 19.8–74.0% of the TP. Conversely, the lowest contents were observed for KCl-extracted P (KCl-P), which accounted for only 0.602% of the TP. Additionally, aluminum/iron-bound P (NaOH-Pi), which accounted for 16.0–53.1% of the TP, was an important factor that caused the TP to increase due to long-term P fertilization. However, calcium-bound P (Ca-P) was maintained at a relatively stable level. Furthermore, the degree of P saturation (DPS) and the P-induced lake eutrophication risk index (ERI) were investigated to determine the risk of release of P from the sediments. The average DPS and ERI values were found to range from 3.92–21.4% and from 12.6–33.6%, respectively. These results suggest that almost half of the estuarine sediments posed a potential risk of eutrophication. Moreover, these findings indicate that estuarine sediments that have been enriched with P are likely the sources of P that lead to the formation of algal blooms in Chaohu Lake.

Keywords: Agricultural intensification; Chaohu Lake; Estuarine sediments; Risk

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1 Introduction

Phosphorus (P) is an essential element for all life forms, but the loading of rivers and lakes with P has been a global problem since the 1960s [1–3]. Application of P in excess of plant requirements can lead to the buildup of P in the environment and saturation of sediment sorption sites with P. In such cases, there is a high potential for P to be released into overlying water from the sediments present [4]. In Eastern China, agricultural activity has intensified greatly since the 1980s, which has resulted in the extensive application of fertilizers to farmlands [5]. Fertilizer application, which is the primary source of P in agricultural regions, accelerates the eutrophication of downstream waters [6]. In addition, agricultural intensification has led to widespread P enrichment in sediments, which has caused a wide variety of environmental problems associated with

eutrophication at regional and local levels [7, 8]. As a result, the regulation of fertilizer application may be important for the control of eutrophication in lakes since most lakes are P limited [9].

It is well known that Chaohu Lake in the Yangtze-Huaihe region has undergone eutrophication [10]. Unlike industrial areas with high levels of pollution loading [11], there were few environmental problems in the agricultural estuaries of Chaohu Lake Valley in the past. However, due to the intensification of agricultural activity in the valley, non-point source pollution from agricultural land has become an important factor in recent years [12]. Therefore, an improved understanding of the response of P dynamics to anthropogenic changes is essential to the formation of effective strategies for the management of agricultural cultivation and lake eutrophication [13–16].

Lake estuaries are semi-enclosed lacustrine bodies of water that contain one or more rivers and a free connection to an open lake. Lake estuaries are often characterized by high productivity and sedimentation due to terrestrial runoff and runoff from the lakefront [17]. Estuaries are dynamic environments in which properties such as pH, sediment type, biological habitats, localized depositional and transport processes and water levels change. Although changes in these properties depend on the rivers that feed the estuaries and lakes, they are especially sensitive to human activities, which can modify the physicochemical characteristics of an entire system [18].

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Abbreviations: Ca-P, calcium-bound P; DPS, degree of P saturation; ERI, P-induced lake eutrophication risk index; KCl-P, KCl-extracted P; NaOH-Pi, aluminum/iron-bound P; NaOH-Po, organic-bound P; OM, organic matter; P, phosphorus; PSI, P sorption index; Res-P, residual P; TP, total phosphorus

Figure 1. The sediment sampling stations in the agricultural estuaries of Chaohu Lake Valley.

Estuaries are usually the important conduits of pollutants into lakes, and may become the headstream of algal blooms. Accordingly, numerous studies have been conducted to evaluate the physical, chemical and biological characteristics of water and sediments in coastal estuaries [19, 20]. However, little is known about these characteristics in lake estuaries, especially with regard to P and the risk of its release from estuarine sediments in agriculture regions.

The processes involved in P dynamics are important drivers of biological activity in sediments that must be understood to avoid the risk of eutrophication in response to agricultural intensification [8]. In this study, all ten estuaries located in the agricultural region of Chaohu Lake Valley were evaluated to determine if changes in the estuary P dynamics had occurred due to agricultural intensification. Sediments were collected from the estuarine region to: (I) Investigate the accumulation of P in the estuarine sediments, and (II) Evaluate the risk of eutrophication associated with the estuarine sediments.

2 Materials and Methods

2.1 Study Area

Chaohu Lake ($30^{\circ}58' - 32^{\circ}06' \text{ N}$ and $116^{\circ}24' - 118^{\circ}00' \text{ E}$), which is one of the five largest freshwater lakes in China, is located in the center of Anhui province and flows into the Yangtze River. Chaohu Lake is a typical, large, shallow, subtropical freshwater lake that covers an area of 780 km^2 and has a mean water depth of 2.69 m. The valley in which the lake is located has a traditional agricultural-based economy and a centuries-old history of cultivation [21, 22]. In the last few decades, agricultural cultivation in the valley has increased in response to a steady increase in population and relative

scarcity of land [23]. Consequently, non-point source pollution has increased and Chaohu Lake has become as one of the three most polluted freshwater lakes in China [24, 25].

For this study, the effects of agricultural activities on P buildup in all ten estuaries located in the agricultural region of Chaohu Lake Valley, Fig. 1, were evaluated. The estuaries were located at the end of the Changlin, Qianxi, Liucha, Shengli, Huatang, Laoshu, Jiaozhi, Xiao, Baishan and Xin rivers (hereafter denoted as a to j, respectively). The length of these rivers ranges from approximately 5–15 km, and their width ranges from approximately 6–30 m. All of these rivers are primary agricultural rivers in the Chaohu Lake Valley, and the estuaries of these rivers are all subject to non-point source inputs from agricultural activities [26]. These characteristics have resulted in spatial consistency in the agricultural region of Chaohu Lake Valley; and therefore, they were deemed to result in similar sedimentation processes among the estuaries. Traditional agricultural hydraulic systems known as multi-pond systems are typically located in the watersheds in the study area [27]. It has been shown that the ponds that comprise these systems can effectively retain nutrients from agricultural land and reduce diffuse pollution [27, 28]. Despite this, a high level of nutrients has recently accumulated in the estuarine sediments in the study area due to the intensification of agriculture.

2.2 Sample Collection

Sampling was conducted in November 2007 at each of the ten stations shown in Fig. 1, using hand-held PVC corers with a diameter of 8 cm and a length of 150 cm. Five sediment columns were taken at random from each station in the studied areas in which the sediments had not been disturbed. The sediment columns were divided

Figure 2. Contents of total phosphorus and phosphorus fractions versus depth in the estuarine sediments of the agricultural region of Chaohu Lake Valley.

into 5 cm slices in sequence. The samples ($n = 300$) were initially collected into self-provided bags. Upon arrival in the laboratory, the samples were air-dried to reduce the water content, after which they were dried in an oven at 40°C , and then ground and passed through a 100-mesh sieve prior to analysis.

2.3 Analytical Methods

The organic matter (OM) content was determined by residual titration of $\text{K}_2\text{Cr}_2\text{O}_7$ [29]. The total phosphorus (TP) of the sediment samples was determined by the $\text{HClO}_4\text{-H}_2\text{SO}_4$ digestion method [29]. P fractionation was determined following the procedure described by van Eck as modified by Moore and Reddy [30, 31]. Repeated measurements of standard samples revealed that the analytical accuracy of P was 10%. The extracted P fractions were exchangeable P (KCl-P, extracted using 1 M KCl), aluminum/iron-bound P (NaOH-Pi, extracted using 0.1 M NaOH and then evaluated directly by spectrophotometry), organic-bound P (NaOH-Po, extracted using 0.1 M

NaOH, the difference between digestion and non-digestion), calcium-bound P (Ca-P, extracted using 0.5 M HCl), and residual P (Res-P, calculated based on the difference between TP). For analysis of the P fraction and OM, the variations within replicates were $<10\%$.

Estimates of the P sorption capacity were made using the degree of P saturation (% DPS), which uses an acid ammonium oxalate extraction to relate the molar concentration of P (as a percentage) to half the sum of Al and Fe [32]. The P sorption index (PSI, mg P/100 g sediment) was determined using the method described by Bache and Williams [33]. In addition, the P-induced lake eutrophication risk index (ERI) was utilized to assess the risk of eutrophication [34–36].

2.4 Statistical Analysis

The experimental data were analyzed using SPSS 13.0 for Windows. Spearman's correlation was used to assess the relationship between the TP and P fractions and OM. A one-sample t test ($p = 0.05$) was used

Figure 3. Organic matter contents versus depth in the estuarine sediments of the agricultural region of the Chaohu Lake Valley.

for analysis of variance. Origin Pro 7.5 was used to plot the experimental data.

3 Results

3.1 Spatial-vertical Distribution of TP and P Fractions in the Estuarine Sediments

The TP contents in the estuarine sediments of Chaohu Lake Valley did not exhibit spatial variability ($p > 0.05$), except within the small watershed of the Changlin River. However, there was a prominent vertical increasing trend from the substrate to the surface sediments in all estuaries as shown in the data presented in Fig. 2. Specifically, the average TP contents of sediments in the ten estuaries increased from 374 to 537 mg/kg from the substrate (25–30 cm) to the surface (0–5 cm).

The labile and non-labile P contents in the estuarine sediments were also evaluated, Fig. 2. In all sediments, Res-P, which included

immobile organic P and resistant apatite, was the most abundant form of P (accounting for 19.8–74.0% of the TP). In addition, these forms of P showed an increasing vertical trend similar to that of TP. Res-P comprised not only refractory organic P, but also a major organic fraction that will eventually be decomposed and released. Therefore, the increasing vertical trend observed in this study was likely due to the contemporary sedimentation of organic bound phosphorus as phytoplankton or macrophytes and its accumulation in the upper sediments [37]. The level of NaOH-extracted P, including NaOH-Pi and NaOH-Po, also increased from the substrate to the surface sediments. NaOH-Pi, which represents the P associated with Fe and Al oxyhydroxides [38], was the next most abundant form of P, accounting for 16.0–53.1% of the TP. NaOH-Po, which represents the P associated with humic and fulvic acid and is considered to be resistant to biological breakdown [38], accounted for 3.11–26.5% of the TP. NaOH-extracted P is a potential source of P that could be released into the water column under certain physicochemical environments. In fact, the level of NaOH-extracted P in the study area rose so dramatically that it has become the dominant P fraction in

Figure 4. Degree of phosphorus saturation of the estuarine sediments in the agricultural region of Chaohu Lake Valley.

the sediments of some estuaries, such as those of the Liucha and Xiao rivers. Ca-P, which represents P bound to calcium, accounted for 2.21–17.7% of the total P and showed little change with sediment depth. KCl-P, which represents the labile inorganic P [38], accounted for only 0.602% of the TP. Despite its low levels, KCl-P is one of the most important forms of P in the system.

3.2 Relationship between P and OM

As shown in Fig. 3, the OM contents obviously increased from the substrate to the surface sediments in all ten estuaries, with the average values increasing from 12.5 (25–30 cm) to 21.3 g/kg (0–5 cm). In addition, the relationship between P and OM was analyzed by Pearson's correlation and the data is summarized in Tab. 1. KCl-P, NaOH-Pi, NaOH-Po, Ca-P and Res-P were found to be positively correlated with TP, as demonstrated by correlative coefficients of 0.589, 0.735, 0.317, 0.594 and 0.636, respectively. In addition, TP was positively correlated with OM ($p \leq 0.01$, $r = 0.567$) in all estuarine sediments. Finally, significant positive correlations were observed between OM and NaOH-Po ($p \leq 0.01$, $r = 0.401$).

OM was positively correlated with the TP and P fractions, and all of these factors increased from the substrate to the surface sediments in the ten estuaries evaluated in this study. The presence of

OM in sediments is known to have a strong effect on P sorption [39, 40]. As the OM contents increased, sediments had higher capacity to adsorb organic P [41]. This finding explains the positive correlation that was observed between the OM content and NaOH-Po and Res-P. Overall, these results indicate that nutrient matter (P and OM) was subject to a synchronous sedimentation process in the estuarine sediments.

3.3 Risk of P Release from Estuarine Sediments

In order to evaluate the risk of P release from the estuarine sediments, the DPS, PSI and ERI were determined and the results are summarized in Figs. 4 and 5. Similar to the trends in the TP and OM contents, the sediment DPS in most estuaries increased from the substrate to the surface. Specifically, the average DPS was 9.41% at a depth 25–30 cm (minimum 3.92%), but this value was 12.7% at 0–5 cm, with values as high as 21.4% being observed. Higher DPS values were observed on the southern bank of Chaohu Lake (Xiao River, Baishan River and Xin River), with the highest values being observed in the Baishan River. The ERI (%) of the surface sediments (0–10 cm) was determined by dividing the DPS (%) by the PSI (mg P/100 g). The ERI values ranged from 12.6–33.6%, with the highest values being observed in Baishan River and the lowest values being observed in

Table 1. Pearson correlation coefficients for the relationship between phosphorus and organic matter in the estuarine sediments ($n = 300$).

	TP	KCl-P	NaOH-Pi	NaOH-Po	Ca-P	Res-P	OM
TP	1	–	–	–	–	–	–
KCl-P	0.589 ^{a)}	1	–	–	–	–	–
NaOH-Pi	0.735 ^{a)}	0.557 ^{a)}	1	–	–	–	–
NaOH-Po	0.317 ^{a)}	0.076	0.355 ^{a)}	1	–	–	–
Ca-P	0.594 ^{a)}	0.498 ^{a)}	0.708 ^{a)}	0.026	1	–	–
Res-P	0.636 ^{a)}	0.265 ^{b)}	–0.022	–0.144	0.051	1	–
OM	0.567 ^{a)}	0.029	0.275 ^{b)}	0.401 ^{a)}	0.279	0.255 ^{b)}	1

a) Correlation is significant at the 0.01 level (2-tailed)

b) Correlation is significant at the 0.05 level (2-tailed).

Figure 5. Phosphorus-induced lake eutrophication risk index of the surface estuarine sediments (0–10 cm) in the agricultural region of Chaohu Lake Valley.

Changlin River. There were four estuaries in which the ERI values exceeded the risk value of 25.0%, Fig. 5. Therefore, nearly half of the estuarine sediments had the potential to induce eutrophication.

4 Discussion

4.1 P Buildup in Response to Agricultural Intensification

In the Chaohu Lake Valley, agriculture has played a major role in shaping the landscape. As a result, the ten estuaries evaluated in this study have similar sedimentation processes [21, 22]. To determine the effects of agricultural intensification on the P dynamic processes in the estuaries, the Liucha River Watershed, which has an area of 6.92 km², was evaluated. The average linear sedimentation rate in the estuary of the Liucha River was found to be 1.20 cm/year [5]. Therefore, in the present study 30 cm sediment cores were evaluated, which represent an approximately 25 year history, during which time agriculture has intensified rapidly in Eastern China.

In the Liucha River Watershed, land use has remained fairly constant over the past fifty years, with the use of phosphate fertilizer becoming common at the end of the 1960s. As agricultural activity in the region intensified from the 1980s to the year 2000, the amount of phosphate fertilizer usage increased from 0.40 kg/km² to 2.70 kg/km² [5]. It has been reported that 85–90% of phosphate fertilizer is carried from the land into runoff and/or subsurface flow

[5]. As a result, the lost P causes marked changes in the nutrient load of the watershed in which it is applied. Multi-pond systems typically located in the watershed evaluated in this study have acted as sinks for most of the particulate phosphorus that has runoff from fields. However, much of the dissolved phosphorus has also been transported to estuaries, where it has been assimilated by hydrophytes and eventually entered the estuarine sediments [5, 28]. Estuarine sediments serve as a record of this process and clearly demonstrate that P loading has increased due to the increased levels of agricultural activity. Indeed, the changes in the TP contents in the estuarine sediments of Liucha River over the last 25 years were similar to the changes in phosphate fertilizer dosage that occurred over the same period, as seen in Fig. 2. These results clearly demonstrate that agricultural intensification is the primary source of P accumulation in sediments. In addition, because the land use and phosphate fertilizer dosage were similar in the other nine watersheds evaluated in this study, it can be assumed that the pattern is consistent among all of the estuaries in the agricultural region of the Chaohu Lake Valley.

There were 5 P fractions evaluated in the sediments, and correlation analysis revealed that each of these was correlated with TP. Generally, KCl-P is easily desorbed from the sediments and released to the overlying water [25]. However, NaOH-Pi, which was found to be the second most common form of P in the estuarine sediments, can also act as an important indicator of the P-release potential of sediments in certain physicochemical environments [42]. When the amount of P added during fertilization exceeds the amount removed by crops, the excess P is primarily converted to NaOH-Pi [37, 43], which can act as a source of P for plants. The high level of NaOH-Pi observed in the sediments in this study reflected the increase of TP in the estuarine sediments in response to vast input of fertilizer P. Other fractions, such as NaOH-Po, can also act as a source of available P under certain conditions [37].

4.2 The Risk of P Release from Estuarine Sediments

The release of P from sediments is related to the P sorption and desorption processes, which usually control the risk of lacustrine eutrophication [44]. According to the Chinese environmental dredging common standard, when the TP contents in lake sediments is greater than 500 mg/kg, the region is considered to have an increased risk of P release and the sediments should be dredged [45]. In addition, DPS and PSI are also used to describe the P sorption ability and potential for release [34–36]. Specifically, a higher ERI indicates a higher risk of P release, and ERI values higher than 25.0% indicate a high eutrophication risk [35, 46].

The TP contents in the surface sediments (0–5 cm) were greater than 500 mg/kg in the estuaries of the Baishan River, Huatang River, Xin River and Xiao River. In addition, the ERI values were greater than 25.0% in the Xin River, Huatang River, Baishan River and Xiao River, reaching 33.6%, 30.7%, 28.2% and 26.9%, respectively, Fig. 5. With the exception of the Huatang River, which is near a small factory that discharges P, these rivers are in the southern portion of the Chaohu Lake Valley. These findings may indicate that there are more agricultural areas in the southern portion of the valley than in the northern portion, where there are more mountains present and the land is less suited to intensive agricultural activity [22]. The ERI results were consistent with the TP values, and almost half of the estuaries were found to be at risk of eutrophication. The DPS values of the estuarine sediments were also found to increase over the

last 25 years, Fig. 4. Indeed, the high sediment P level is approaching the P sorption capacity of the estuarine sediments as the development of agriculture intensifies. The high DPS and ERI values observed in this study suggest that the estuarine sediments could become the P source. The transformation of estuarine sediments into the source of P could result in the estuaries becoming the head-streams of algal blooms, which has recently been observed in the study area. Moreover, the harvest of hydrophytes for feed has decreased as the economy has improved. As a result, there are now an increased number of water plants growing in the rivers and estuaries. The death and decomposition of these plants may change the conditions at the water-sediment interface, which could result in the release of P from surface bed sediments into the overlying water. Therefore, the mechanism of this phenomenon is presently being studied by the current authors. Overall, these findings indicate that the export of P in response to agricultural intensification, as well as the associated water deterioration and the risk of P release should be considered during the formation of strategies to manage the water environment of the valley.

5 Conclusions

Agricultural intensification, which has developed rapidly in Eastern China since the 1980s, may be an important factor affecting the nutrient processes of the Chaohu Lake Valley. Indeed, the increasing use of phosphate fertilizer has likely resulted in a dramatic increase in the TP and P fractions in the estuarine sediments in the valley over the last few decades. The contents of TP and most P fractions were higher in the uppermost 5 cm sediments, and were found to be dependent on sediment depth. The increase in P contents indicated that there was the potential to overwhelm the sorption capacity of the sediments, causing continuous water quality deterioration. Furthermore, significant positive correlations were observed between OM and P (TP, NaOH-Pi, NaOH-Po, and Res-P), which implies that nutrients in estuarine sediments are subject to a synchronous sedimentation process. Moreover, the high DPS and ERI values demonstrated that almost half of the estuarine sediments posed a potential eutrophication risk. Taken together, the results of this study imply that P loading from agricultural intensification is an important factor in the nutrient input of Chaohu Lake.

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