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Long-Xiu Cheng1, Pei Sun Loh1*, Zhang-Hua Lou1, Ai-Min Jin1, Xue-Gang Chen1, Chen-Tung Arthur Chen2, Shuangyan He1, Lin Yang1 and Hong-Wei Yuan1

Abstract: In this study, two sediment cores (C1 and C2) were collected from the Andong salt marsh southwest of Hangzhou Bay and analysed to determine their different sedimentary phosphorus (P) forms. The changes in the sedimentary P forms over the past 30 years and the implication of these changes on environmental risks were determined. The low mean total P (TP) contents along C1 (30.42 mg/kg) and C2 (68.60 mg/kg) indicate no P pollution in the Andong salt marsh. The sedimentary TP and all P forms decreased towards the present due to a reduction in particle size and an overall decrease in nutrient input. The sedimentary P contents were also affected by other factors, such as typhoons, bridge construction and aquaculture activities that facilitated erosion and increased P input, whereas drought caused a reduction in sedimentary P. Organic P (OP) represented more than half of the TP, and NaOH-P (P bound to Fe and Al oxides and hydroxides) represented more than half of the inorganic P (IP), indicating an overall high level of bioavailable P. Thus, the sediments in the Andong salt marsh could be releasing P during organic matter decomposition and under anoxic conditions.

Subjects: Earth Sciences; Geochemistry; Environmental Chemistry; Keywords: Salt marsh; sediment core; phosphorus fractions; bioavailable P

ABOUT THE AUTHOR
One of the research activities of the authors include studying the carbon and nutrient cycling in the Andong salt marsh at the southwest of Hangzhou Bay. The authors have investigated the sources and distribution of sedimentary organic matter and heavy metals along the surface sediments of the Andong salt marsh. The authors have also determined the impact of human activities on the salt marsh during the past 30 years by studying the changes of sedimentary organic matter and heavy metals along two sediment cores collected from the salt marsh. In this paper, we report trends of different sedimentary phosphorus forms in the salt marsh for the past 30 years.

PUBLIC INTEREST STATEMENT
Phosphorus pollution is a common phenomenon in coastal zones. This study investigated the different phosphorus forms in the sediments of a coastal salt marsh situated at the southwest of Hangzhou Bay, China. The total phosphorus in the sediments in the salt marsh was lower than the lowest limit indicative of phosphorus pollution (500 µg/g). The sedimentary phosphorus contents in the marsh during the past 30 years was affected by typhoon, bridge construction and aquaculture activities that facilitated erosion and increased phosphorus input, whereas drought caused a reduction in sedimentary phosphorus content.
1. Introduction
Salt marshes are regions that connect the ocean and the land. These wetlands have a characteristically high salinity and the presence of halophytic vegetation adapted to waterlogged soils. Salt marshes are considered to be efficient sinks for phosphorus (P) as well as a source of P (Coelho, Flindt, Jensen, Lillebo, & Pardal, 2004; Jordan & Correll, 1991). Salt marshes with vegetated sediments are an important P sink, as they can retain approximately four times more P than bare sediments (Freitas, Duarte, & Caçador, 2014). Nitrogen and P from the sediment interstitial water are taken up and incorporated into plant biomass, promoting nutrient sequestration and retention within the wetland system, thus reducing their availability in the water column and the chance of the occurrence of eutrophication (Sousa, Lillebo, Pardal, & Cacador, 2010). The role of salt marshes as P sinks is important in areas that are affected by eutrophicated water (Álvarez-Rogel, Jiménez-Cárceles, & Egea-Nicolás, 2007), mine waste and wastewater effluents (Jiménez-Cárceles & Álvarez-Rogel, 2008). However, sediments could also become a source of P and cause a drastic environmental impact, as this P could be released into the aquatic environment under suitable conditions (Jiménez-Cárceles & Álvarez-Rogel, 2008) and after land conversion (Huang et al., 2012). Thus, salt marshes could also pose a threat of increasing the eutrophication risk for coastal waters (Coelho et al., 2004).

The adsorption of P onto sediments and the release of this sedimentary P into water (Jansson et al., 1988) are affected by various physical, chemical and biological factors (Baturin, 2003; Caetano & Vale, 2002; Confield, Kristensen, & Bo, 2005; Ingall, Bustin, & Van Cappellen, 1993; Schenau & De Lange, 2001). Some P forms, such as loosely-bound, redox-sensitive and organic P (OP), are bioavailable and can cause eutrophication (Coelho et al., 2004; Fytianos & Kotzakioti, 2005; Nurnberg 1988). Hence, knowledge of sedimentary P forms is important, and many studies have determined sediment P forms to serve as indicators of the potential of sediment to release P, the bioavailability of P (Søndergaard, Windolf, & Jeppesen, 1996) and the potential of P to contribute to eutrophication (Wang et al., 2009).

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The Andong salt marsh is a typical salt marsh system that links Hangzhou Bay to the town of Andong in the southwestern part of the bay and is the sediment boundary between the Qiantang River and Hangzhou Bay. Hangzhou Bay is discharged by the Qiantang River to the west and by the Changjiang Estuary to the north, with most of the materials in Hangzhou Bay originating from the Changjiang River and less than 2% of these materials originating from the Qiantang River (Xie, Gao, Wang, & Pan, 2013). In recent years, the Changjiang River has been affected by the presence of reservoirs, which have reduced the discharge from the river by half (Dai, Chu, Stive, Du, & Li, 2011). The Andong salt marsh could also be affected by the reduced input from the Changjiang River. In this study, two sediment cores were collected from the Andong salt marsh, C1 from the lower marsh and C2 from the upper marsh. These sediments were determined for their $^{210}$Pb activities, bulk elemental composition and different sedimentary P forms (HCl-P or apatite P; NaOH-P or P bound to Al, Fe, or Mn oxides and hydroxides; IP or inorganic P; and OP or organic P). The objectives of this study were to determine the changes in the sedimentary P forms in the salt marsh over the past 30 years and to assess the risks of P pollution faced by the salt marsh.

2. Materials and methods
2.1. Study areas and sampling
Two sediment cores of 130 cm and 165 cm in length were successfully collected from the Andong salt marsh on October 14th and 15th, 2015. The cores were collected from two locations, 30°22′41″N, 121°10′19″E and 30°22′26″N, 121°12′20″E (Figure 1). These locations were at the fringe of the salt marsh nearest to the coastal water, which was accessible by foot during low tide. The location of C1 (130 cm core) was closer to the mouth of the Qiantang River, and C2 (165 cm core) was east of the first core. The sediment cores were collected using a gravity columna sampler lined with a 60-mm diameter polyvinyl chloride (PVC) core tube. In the laboratory, the PVC core tubes were sliced vertically into halves, and sediments were then sliced at 5 cm deep intervals. These sediments...
were dried at approximately 45°C for three days, ground with a mortar and pestle until homogenous and ready for the following analyses.

2.2. Analytical methods

Different sedimentary P forms were extracted using the “Standards, Measurements and Testing (SMT)” method by Ruban et al. (1999, 2001)). Precisely 200 mg of dry sediment was weighed into a centrifuge tube, and then 20 ml of 1 M NaOH was added. The sediment was extracted by shaking overnight for 16 h. On the following day, the sample was centrifuged at 2000 g for 15 min. Exactly 10 ml of the supernatant was collected. Four millilitres of 3.5 M HCl was added to the supernatant, and the solution was stirred vigorously for 20 s and allowed to stand overnight for 16 h. This solution was centrifuged at 2000 g for 15 min, and the supernatant was measured for NaOH-P. The sediment residue after the first extraction was washed with 12 ml of 1 M NaCl, stirred for 5 min, centrifuged at 2000 g for 15 min, and then the supernatant was discarded. This step was repeated once. Afterwards, 1 M HCl was added to the residue, and this mixture was stirred overnight for 16 h. The solution was centrifuged at 2000 g for 15 min on the following day, and the supernatant was measured for HCl-P. Another 200 mg portion of the dry sediment was weighed into a centrifuge tube, 20 ml of 1 M HCl was added, and the solution was stirred overnight for 16 h. On the following day, this solution was centrifuged at 2000 g for 15 min, and the supernatant was measured for IP. The residue was washed twice with distilled water and centrifuged, and the supernatant was discarded. The residue was then dried at 80°C for 3 h, followed by combustion at 450°C for 3 h. When cool, the ash was poured into a centrifuge tube. Precisely 20 ml of 1 M HCl was added to the ash, and the solution was stirred overnight for 16 h, followed by centrifugation at 2000 g for 15 min. The supernatant was then ready for OP determination. All P concentrations were determined colourimetrically as a molybdenum blue complex using a UV-visible spectrophotometer UV-8000 (METASH, Shanghai, China). The absorbance was measured at a wavelength of 885 nm.

The dry sediment was added to an excess of 1 M HCl to remove the inorganic carbon. The residues were then dried in an oven at 45°C for a few days. These residues were homogenized using a mortar and pestle, weighed precisely into tin foil and crimped into pellets. The total organic carbon (TOC) and total nitrogen (TN) were determined using a Vario EL III elemental analyser (Elementar, Germany). The standard reference materials used were BSCC (2.24% C, 0.24% N) and NIST2704 (3.34% C, 0.22% N). The average coefficients of variation based on replicate analyses of the samples were 2.32% for the % OC and 3.08% for the % N.
Particle sizes were measured using a Microtrac laser particle size analyser after H$_2$O$_2$ digestion and (NaPO$_3$)$_6$ dispersion (Microtrac, US; BT-9300Z, Dandong Baite Instruments Co., Ltd., China). The measurement range was 0.02–2800 μm, and the accuracy was 0.6% (Jin et al., 2016).

The activity of $^{210}$Pb was determined for the sediments collected at every 10 cm interval. This was carried out using a GWL series high-purity germanium low gamma spectrometer with a high-purity germanium coaxial well photon detector system (AMETEK-AMT Ortec Co., USA) at the Nanjing Institute of Geography and Limnology, Chinese Academy of Sciences. The method used was based on Chinese National Standard GB/T11743-2013 for the determination of radionuclides in soil gamma energy spectrum analysis. Excess $^{210}$Pb activity decreases exponentially with sediment depth according to:

$$A_x = A_0 e^{-xS}$$

where $A_x$ is the excess $^{210}$Pb activity (Bq/kg) at depth $x$, $A_0$ is the activity of $^{210}$Pb at the sediment surface, $\lambda$ is the $^{210}$Pb decay constant (0.0311/yr) and $S$ is the accretion rate (cm/yr) (Boyd, Sommerfield, & Elsey-Quirk, 2017; Tylman 2004). The mean accretion rates for C1 and C2 were estimated from the slope of the measured $^{210}$Pb activity versus depth profile (Figure 2) to be 5.03 cm/year and 4.89 cm/year, respectively. The R$^2$ value refers to the fitted lines in the graphs.

Figure 2. Profile of $^{210}$Pb activity versus depth for C1 and C2. The mean accretion rates for C1 (5.03 cm/year) and C2 (4.89 cm/year) were estimated from the slope of the $^{210}$Pb activity versus depth profile using the least-square fit method. The R$^2$ value refers to the fitted lines in the graphs.
5.03 cm/year and 4.89 cm/year, respectively. Hence, the total number of years represented by cores C1 (127.5 cm) and C2 (162.5 cm) were 25 and 33 years, respectively (Jin et al., 2016). The ages of the sediment layers were calculated by multiplying the mean accretion rates of each core by the depth of the respective sediment layers.

3. Results
The results of the sedimentary P forms along C1 and C2 are presented in the Appendix. The sedimentary P forms along C1 are in the following order: average NaOH-P = 7.16 mg/kg (ranging from 1.65 to 21.83 mg/kg) < HCl-P = 12.05 mg/kg (4.48 to 26 mg/kg) < IP = 12.73 mg/kg (5.51 to 27.53 mg/kg) < OP = 17.80 mg/kg (6.02 to 30.95 mg/kg). The sedimentary P forms along C2 are in the following order: HCl-P = 26.77 mg/kg (13.92 to 50.35 mg/kg) < NaOH-P = 29.82 mg/kg (9.25 to 55.32 mg/kg) < IP = 34.20 mg/kg (16.37 to 77.44 mg/kg) < OP = 34.40 mg/kg (17.49 to 69.59 mg/kg). All P forms showed higher concentrations in C2 than in C1, with the total P (TP) along C2 ranging from 33.37 to 137.45 mg/kg (average = 68.60 mg/kg), and the TP along C1 ranging from 14.97 to 49.26 mg/kg (average = 30.42 mg/kg).

The particle sizes in C1 ranged from 25.92 µm (in 2002) to 34.20 µm (in 1996), and the mean value was 30.4 µm. The particle sizes seemed to be larger before 1996 (ranging from 30.4 µm to 34.20 µm) and smaller after 1997 (ranging from 25.92 µm to 31.91 µm) and showed a general decreasing trend towards the present (Appendix). The particle sizes in C2 ranged from 7.76 µm (in 2003) to 33.03 µm (in 1986), with a mean value of 20.3 µm. The particle sizes in C2 also showed a gradual decrease towards the present, and in the years after 1998, the particle sizes were approximately 20 µm or less (Appendix).

The trends in the sedimentary P forms and the OC/OP molar ratios along both cores are also presented in Figure 3. In core C1, HCl-P and NaOH-P showed fluctuations between 1990 and 1996; the highest HCl-P and NaOH-P values of 26.00 mg/kg and 21.52 mg/kg, respectively, occurred in 1997, after which both fractions started to show a decreasing trend towards 2014. The IP, OP and TP also showed fluctuations before 2002; the highest values for IP, OP and TP occurred in 2000 (26.83 mg/kg), 2002 (30.95 mg/kg) and 2000 (49.26 mg/kg), respectively, after which all of the fractions decreased gradually towards the present (Figure 3(a)).

In core C2, HCl-P showed the highest value of 50.35 mg/kg in 1983, after which it decreased gradually to the lowest value of 15.94 mg/kg in 1997, fluctuated between 18.69 and 35.88 mg/kg between 1998 and 2009, and then started to decrease gradually from 2009 to the present. NaOH-P, IP, OP and TP showed smaller ranges and lower values prior to 1998. NaOH-P showed a drastic increase in 1998 (48.24 mg/kg) and became quite constant afterwards until a slight increase in 2015. The IP, OP and TP showed a gradual increase from 1998 onwards, with the highest IP (77.44 mg/kg), OP (69.59 mg/kg) and TP (137.45 mg/kg) occurring in 2004 and 2005, after which all three fractions showed a gradual decrease towards 2015 (Figure 3(b)).

The OC/OP molar ratios ranged from 332.3 (in 2010) to 853.6 (in 2012) in C1 and ranged between 145.8 (in 2004) and 618.1 (in 1991) in C2. The OC/OP ratios along C1 decreased from 812.8 in 1994 to 332.3 in 2010, increased drastically to 853.6 in 2012, and then decreased again to 388.7 in 2014. Overall, the OC/OP ratio in C2 showed great fluctuations (with the highest and lowest values) between 1982 and 2004 (ranging from 124.5 to 618.1), after which the OC/OP values were lower in a smaller range from 262.3 to 397.7 (Figure 3(c)).

4. Discussion

4.1. Implications of different sedimentary P forms in the andong salt marsh
The mean TP content in both C1 and C2 were 30.42 mg/kg and 68.60 mg/kg, respectively. According to Liu, Jin, and Jing (1999), sedimentary TP < 500 µg/g indicates no P pollution, TP within 500 µg/g < TP < 1000 µg/g indicates medium pollution, and TP > 1000 µg/g indicates pollution. The results obtained indicate no P pollution in the Andong salt marsh system. In addition, the mean TP in C1 and C2 were
relatively lower than those in other locations, such as the Min River Estuary marsh, where sediment TP ranged between 338–846 mg/kg in May 2013 and 353–932 mg/kg in August 2013 (Zhang, Zeng, Tong, Zhai, & Lin, 2015); the rivers discharging into the Bohai Sea, such as the Liao and Yongdingxin, which were composed of 218.96 mg/kg and 913.48 mg/kg TP, respectively (Shan, Li, Zhang, Di, & Jin, 2016); the Yellow River delta sediments, which contained 541.58 mg/kg TP (Cui, Xiao, Xie, & Zhang, 2018); and the TP in the sediments from ten major basins in China, which averaged 733 mg/kg (Yang, Gao, Hao, Zhou, & Lu, 2017). Other locations worldwide also showed wide ranges of TP: TP in soils in salt marshes in southeast Spain...
were 1123 mg/kg and 1076 mg/kg (Jiménez-Cárceles & Álvarez-Rogel, 2008); Lake Mariut sediments have TP values of 332–2171 mg/kg and El Mex Bay has a TP value of 249.6–825.6 mg/kg (Soliman, El Zokm, & Okbah, 2017); surface sediments in the south Caspian Sea have a mean TP of 535 ppm (Bastami et al., 2018); and the Gargalheiras and Cruzeta Reservoirs in the Piranhas-Açu watershed in Rio Grande, northeast Brazil have mean surface sediments TP of 3603.96 mg/kg and 4106.22 mg/kg, respectively (Cavalcante, Araujo, Noyma, & Becker, 2018).

The main inorganic P forms are as follows: (i) the easily releasable, loosely bound, labile and exchangeable P form; (ii) the P form that is associated with Al, Fe and Mn oxides and hydroxides; and (iii) the P form that is bound to Ca compounds, usually known as apatite-P or Ca-bound P (Ruban et al., 1999). The P forms extracted using Ruban’s SMT method are NaOH-P (P bound to Al, Fe, Mn oxides and hydroxides), HCl-P (Ca-bound or apatite-P), IP and OP. TP is the sum of the IP and OP. Thus, NaOH-P + HCl-P > IP in Ruban et al. (2001) and this study (Appendix) could be due to the extraction of some of the easily bound P in the NaOH-P fraction. In addition, the drying of sediments could interfere with P speciation, and the use of NaOH could precipitate some of the iron-bound P (Golterman, 1996). Nevertheless, the SMT method provided a fast, easy and economical means to gauge the different sedimentary P species.

Our results showed that OP was the predominant form in both C1 and C2, followed closely by IP. OP can be released into the environment from organic matter decomposition. In C2, OP and IP contributed almost equally, 50.1% and 49.9%, respectively, to TP. In C1, OP and IP contributed approximately 58.8% and 41.8%, respectively, to TP. There were significant correlations between the TOC with OP and TP in C1 and between TOC and most P forms in C2 (Table 1), indicating the adsorption of P with organic matter. This result is consistent with studies that have found that sites with agricultural activity that received long-term P fertilization also showed increased P accumulation in the sediments, as well as high NaOH-P and OP, but relatively stable HCl-P. These locations also showed a good correlation between the organic matter and P due to the increased sorption of P by organic matter (Tang, Shan, & Zhang, 2010).

HCl-P or Ca-bound P was the main IP in C1, as HCl-P (12.05 mg/kg) was higher than NaOH-P (7.16 mg/kg). However, both NaOH-P and HCl-P contributed almost equally to IP in C2, with NaOH-P (29.82 mg/kg) slightly higher than HCl-P (26.77 mg/kg). The higher NaOH-P fraction and the positive correlation between OP and NaOH-P in C2 (Table 1) indicated pollution at this site. High NaOH-P has also been found in sites affected by mine waste with high metal contents (Gonzalez-Alcarez et al. 2012). Another factor contributing to the differences in NaOH-P and HCl-P is the pH gradient. Elevated adsorption of P onto the Al and Fe oxides and hydroxides was observed at lower pH values, and higher HCl-P occurred at higher pH values or at seaward locations due to the precipitation of P with CaCO₃ compounds (Oxmann & Schwendenmann, 2015). Hence, C1 having a relatively higher HCl-P than NaOH-P could have been the result of C2 being located at a relatively upper marsh and C1 at a lower marsh.

Although the overall TP concentrations along both C1 and C2 indicate no P pollution, the fact that more than half of the TP was composed of OP and that more than half of the IP in C2 was the NaOH-P fraction suggests that more than 70% of the sedimentary P in the Andong salt marsh system was bioavailable. The loosely bound P, iron-bound P and OP are easily desorbed from sediments and released to the overlying water; hence, these P forms represent the P that is bioavailable to primary producers (Bastami et al., 2018; Cong et al., 2014). NaOH-P includes P bound to Al and Fe and may be released under anoxic conditions; HCl-P is relatively stable and considered nonbioavailable (Cavalcante et al., 2018). Under oxic conditions, phosphate can be adsorbed onto the iron oxyhydroxides to form Fe³⁺ precipitates or solid FeOOH·PO₄ complexes. Under anoxic conditions during flooding, ferric ions (Fe³⁺) are reduced to ferrous ions (Fe²⁺), thereby releasing P from the sediments. In addition, sulfate reduction occurs under reducing conditions, and sulfate is converted to soluble sulfides (H₂S and HS⁻), which react with Fe²⁺ to form sulfides, further releasing P (Bruland & DeMent, 2009; Jimenez-Carceles and Alvarez-Rogel...
As the NaOH-P form includes P that is adsorbed onto the Al and Fe oxides and hydroxides, this P form will likely be released from the sediment due to anoxic conditions probably caused by prolonged flooding.

4.2. Impact of human activities and extreme weather on the sedimentary P forms

The gradual decrease in particle size to the present value could be due to the presence of reservoirs upstream of the Changjiang and Qiantang Rivers that trap the larger particles (Wu et al., 2013). One reason for the relatively smaller particle size in C2 than in C1 is most likely because of the location of C2 at the upper marsh and C1 at the lower marsh. Previous studies have also found smaller particle sizes at the upper marshes and larger grain sizes closer to the sea in the salt marsh systems off Chongming Island (Zhou, Wu, Kang, & Zhang, 2007; Zhou, Wu, Zhang, Kang, & Liu, 2006) and the Andong salt marsh (Yuan et al., 2017). Thus, in addition to the possibility that C2 may be more polluted than C1 (Section 4.2), the overall higher concentrations of all P forms in C2 than in C1 were likely due to smaller particle sizes in C2 facilitating P adsorption. Correlation analysis (Pearson correlation) showed significant negative correlation coefficients between the particle size and most P forms (except HCl-P) and the TOC in C2 (Table 1), further indicating that smaller particles facilitate the adsorption of sedimentary TOC and different P species. In addition, the fact that C2 has a higher NaOH-P than C1 could also be attributable to the smaller particles, as studies have found that smaller particles facilitate the adsorption of Fe/Al-bound P (Andrieux-Loyer and Aminot 2001; Lair, Zehetner, Khan, & Gerzabek, 2009).

### Table 1. Correlations among sediment P fractions, TOC and particle size in C1 and C2

|       | C1 HCl-P | NaOH-P | IP | OP | TP | TOC | Particle size |
|-------|----------|--------|----|----|----|-----|---------------|
| C2    | HCl-P    | NaOH-P | IP | OP | TP | TOC | Particle size |
| HCl-P | 1        | 0.745**| 0.120| −0.263| −0.090| −0.227| 0.344 |
| NaOH-P|          | 1      | −0.105| −0.236| −0.231| −0.219| 0.237 |
| IP    |          | 1      | 0.057| 0.748**| 0.202| −0.173|     |
| OP    |          | 1      | 0.705**| 0.668**| −0.460**|     |
| TP    |          | 1      | 0.587**| −0.429|     |
| TOC   |          | 1      | −0.279|     |

Particle size (a) C2 (n = 33).

Pearson correlation is used to determine the relationships among the P fractions, TOC and particle size. Results of correlation coefficient (r) are presented in the table; n = sample size; * indicates p is significant to 0.05, ** indicates p is significant to 0.01, the numbers not marked by * or ** indicate no significant correlation between the two parameters. Abbreviations: HCl-P = apatite P; NaOH-P = P bound to Al, Fe, Mn oxides and hydroxides; IP = inorganic P; OP = organic P; TP = total P = IP + OP; TOC = total organic carbon.

2008; Sherman, Fohey, & Howarth, 1998). As the NaOH-P form includes P that is adsorbed onto the Al and Fe oxides and hydroxides, this P form will likely be released from the sediment due to anoxic conditions probably caused by prolonged flooding.
Many studies of sedimentary P forms in cores from other regions of China have found increasing sedimentary P towards the present; for example, in Chaohu Lake Valley (Tang et al., 2010), the Zhejiang coastal waters off the East China Sea (Cong et al., 2014), Sishili Bay (Zhang et al., 2016), Jiayzhou Bay (Kang et al., 2017) and Lake Hulun (Lü, He, & Wang, 2018). These increases in P towards the present were attributable to the influence of terrestrial organic matter on the OP pools (Lü et al., 2018) and to the rapid agricultural developments since the 1980s, resulting in fertilizers with increased P (Tang et al., 2010). The cores along C1 and C2 showed a continuous decrease in most P forms towards the present (Figure 3(a, b)). This result was partly attributable to the presence of upstream reservoirs trapping materials and nutrients, as well as to the successful management strategies reducing P input into the aquatic system.

Although assigning a precise year to each sediment section and relating events to specific years may not be exactly accurate, we found some possible environmental impacts on the sedimentary P forms. The sudden spikes in the HCl-P and NaOH-P concentrations in 1997 and the slightly higher IP, OP and TP concentrations in the following year along C1 and the sudden spikes in NaOH-P and IP in 1998 and the slightly higher HCl-P, OP and TP concentrations in the following year along C2 could be attributable to Typhoon Winnie causing an increase in erosion, thereby increasing the input of P (Figure 3(a, b)) and terrestrial organic matter (Figure 3(c)) into nearby areas such as the Andong salt marsh. Increased aquaculture activities in this area since 2000 (Guo, Li, & Huang, 2005) were reflected in C1 having the highest IP in 2001, the highest OP in 2000, the highest TP in 2000, and a continuous increase in all three fractions after 2001. The construction of the Hangzhou Bay Bridge resulted in an increase in erosion, as well as the input of nutrients such as P into nearby surrounding areas, hence the spikes in IP (in 2005), OP (in 2004) and TP (in 2004) in C2, and the higher HCl-P and NaOH-P (in 2003), spike in IP (in 2004) and higher OP and TP in 2006 in C1. All P forms (Figure 3(a, b)), as well as terrestrial organic matter (Figure 3(c)), in both cores started to decrease after the 2006 dry year, most likely due to reduced erosion and material discharge from the rivers.

5. Conclusions
In this study, two sediment cores from the Andong salt marsh southwest of Hangzhou Bay showed that the sedimentary P fractions were affected by human activities and extreme weather conditions, such as floods and droughts, over the past 30 years. The 1997 high tide due to Typhoon Winnie, the increased nearby aquaculture activities in approximately 2000, and the construction of the Hangzhou Bay Bridge from 2003–2007 caused an increase in the sedimentary P fractions in the salt marsh. The 2006 drought and overall decreased input of materials due to the trapping of materials within reservoirs in rivers could have resulted in a gradual decrease in the sedimentary P fractions from 2006 to the present. Although the overall low TP concentrations indicate no P pollution, the sediments in these locations were prone to release P due to the high amount of bioavailable P, as OP composed more than half of the TP in C1 and C2 and NaOH-P composed more than half of the IP in C2. The NaOH-P, in terms of Fe-bound P, was released under anoxic conditions such as prolonged flooding, and OP was released during organic matter decomposition when the marshes were exposed to the atmosphere.

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