

Article

Chemical Speciation and Preservation of Phosphorus in Sediments along the Southern Coast of Zhoushan Island

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Abstract: This study investigated the distribution of sedimentary phosphorus (P) species along an area of a rapid current at the southern coast of Zhoushan Island. The objective of this study was to improve the understanding of P cycling in a zone of rapid water cycling. Results showed that the average percentage of each P form to total P (TP) was in the following order: apatite P (Ca-P; 52%) was found in the most abundant, followed by organic P (OP; 16%), exchangeable-P (Ex-P; 14%), detrital P (De-P; 11%), and iron-bound P (Fe-P; 7%). Ca-P showed a trend of an increasing concentration from a location at the west (ZS1 has mean Ca-P = 45.6 mg kg⁻¹) toward the east (ZS2 has mean Ca-P = 82.69 mg kg⁻¹) and south-east (ZS3 has mean Ca-P = 82.17 mg kg⁻¹); De-P also increased from 15.12 mg kg⁻¹ at ZS1 to 22.53 mg kg⁻¹ at ZS2 and 27.45 mg kg⁻¹ at ZS3, but the three bioavailable P species, OP, Ex-P, and Fe-P, decreased from the west toward the east of the coastal area. Results along the cores showed the occurrences of sediment P adsorption and release throughout the time span from the 1930s to the present, with an overall trend of decreasing Ca-P and TP from the bottom to surface sediments. There was a tendency of Ca-P formation at the expense of Ex-P and OP release during transport and organic matter decomposition. The likely impact of climate change in the coastal zone would be an increased temperature resulting in elevated organic matter decomposition and P release.

Keywords: coastal sediments; phosphorus fractions; environmental health risk; archipelago



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1. Introduction

Phosphorus (P) is an important ingredient for primary production in the oceans. The main source of P is from fluvial input, which is delivered to the oceans via coastal zones. P is removed from the aquatic environments through biological uptake, incorporation into organic matter and phosphorites, the sedimentation of biogenic calcium carbonate, and precipitation with clays, iron hydroxides, and Al and Fe oxides [1]. P release from sediments could contribute to P pollution in the aquatic environments, but total P (TP) in sediments could not serve as an indicator of the potential of the sediments to release P because only certain sedimentary P species, termed bioavailable P (BAP), which includes organic P (OP), exchangeable P (Ex-P), and the iron-bound P (Fe-P), can be released to the aquatic environments. Different sedimentary P forms also provide information on the sources, early diagenesis, and potential bioavailability of P [2–4]. Thus, many studies have been carried out to better understand the biogeochemistry of sedimentary P species in coastal areas [5–7]. For instance, industrial and domestic waste has resulted in increased P species, OP, and nutrients, causing hypoxia and enhancing sedimentary P release in a subtropical estuary in Brazil [8]. High sedimentation rates control the sedimentary P speciation in the Eastern Arabian Sea continental shelf [9], and the sedimentary P species in

other coastal archipelago areas were affected by multiple factors, such as dissolved oxygen, mobile P fractions, and the sedimentary grain size and organic matter [10]. A study in the Southeast Arabian Sea found that hypoxic bottom water conditions enhanced the release of Fe-P and OP, resulting in low concentrations of Ex-P, Fe-P, and OP, high C/P and N/P, and a low C/N ratio in these sediments [11].

The Yangtze River serves as the major contributor of materials in the East China Sea and the Zhejiang-Fujian Mud Area [12]. Materials from the Yangtze River also flow into Hangzhou Bay, which then mix with materials delivered from the Qiantang River and flow out of the bay [13]. The Zhoushan Island is the largest archipelago in China (Figure 1), and these islands receive input from the Yangtze and Qiantang Rivers [14]. The construction of dams in the Yangtze River has caused a decrease in the sediment load from the river [15,16] and the retreat of the Zhejiang-Fujian Mud Area [17,18], resulting in increased relative contributions from the Qiantang River. There have also been increased contributions of older and finer grain sizes and rock materials from the Zhoushan Archipelago into these coastal zones [19]. The Zhoushan coastal areas have high tidal currents [20], as water passing an island will be diverted to both sides of the island, resulting in increased velocity and intensity of the water movement and inducing sediment resuspension [21]. To our knowledge, there has been no study that determined the dynamics of sedimentary P species under rapid water movement.

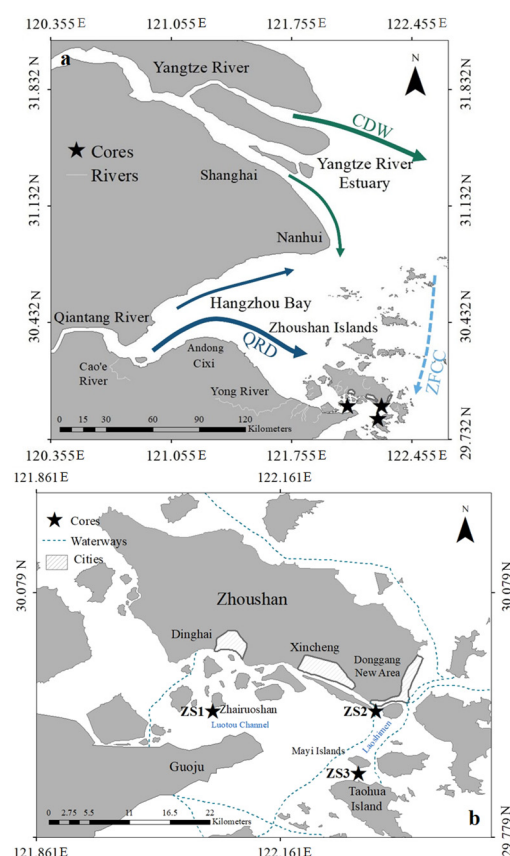


Figure 1. (a) Map showing the sampling locations, with arrows showing input from the Yangtze River, Qiantang River, and Hangzhou Bay; (b) magnified map showing the locations of the three cores, namely ZS1, ZS2, and ZS3, along the southern coast of the Zhoushan Island. Note: QRD = Qiantang River Discharge, CDW = Changjiang (Yangtze) Diluted Water, ZFCC = Zhejiang-Fujian Coastal Current. This figure shows southward flow of ZFCC in the winter. The major cities in the main Zhoushan Island, namely, Dinghai, Xincheng, and Donggang New Area, are shown as the perimeters with shaded lines.

Investigations of sedimentary P species have been conducted along Chinese coastal areas, such as in bays [3,4,22] and in the Yangtze River estuary and the East China Sea [23–25]. Studies of surface and core sediments across transects from nearby Zhejiang coastal water have found an influence of the particle size [26] and input from river discharge affecting the sedimentary P species [27] and redox conditions, such as suboxic environments, resulting in increased P bioavailability in these coastal areas [24,28]. Seasonal variations have been observed to affect the P speciation in the sediments in the East China Sea [25], while other studies have found that P released from the Ex-P and OP fractions has become the P in the Fe-P and Ca-P fractions [29,30]. Less is known about the sedimentary P dynamics along the coastal zones surrounding the Zhoushan Archipelago. Hence, three short sediment cores (each core was about 20 cm in length) were collected from the southern coastal area of the Zhoushan main island and determined for ^{210}Pb activities (in order to determine the ages of the sediment cores) and total organic carbon (TOC) (in order to determine the ratios of TOC/OP and TOC/ $\text{P}_{\text{reactive}}$). A sequential extraction method of Ruttenberg [31] was used to elucidate the sediments into different P forms, namely the Ex-P, Fe-P, calcium-bound or apatite P (Ca-P), detrital P (De-P), and OP. The aim of this study was to determine the biogeochemical cycling of sedimentary P species in a dynamic coastal area.

2. Materials and Methods

2.1. Sediment Sampling

Sediment cores were collected on 22 July 2019, from the zone off the southern coast of the main island of the Zhoushan Archipelago (hereby known as the southern coast of Zhoushan Island), on board the RV Zijingang. A gravity columnar sampler that was lined with a polyvinyl chloride core tube (of 60 mm diameter) was used for sample collection. Three sediment cores were successfully collected, namely a 21 cm core obtained from near Zhairuoshan Island (hereby known as ZS1; 29.93 N, 122.08 E), a 22 cm core was obtained near the port of Shenjiamen (ZS2; 29.93 N, 122.32 E), and a 15 cm core was collected at the south of Mayi Island (ZS3; 29.84 N, 122.25 E; Figure 1). These locations were protected by some nearby islands and were situated near the shores so that it was easier to collect sediment cores under less rapid water movement and shallower conditions compared to the relatively more exposed middle part of the coastal zone. The sediment samples were sliced at every 2 cm intervals along the cores. These sediment slices were dried at 45 °C for two days to a constant weight and homogenized using a mortar and pestle.

2.2. ^{210}Pb Dating

First, dry sediment samples were loaded into test containers. The containers with samples were weighed and then sealed and kept for 10 days for the ^{210}Pb activities to achieve equilibrium. The activities of ^{210}Pb were determined using a high-purity germanium coaxial well proton detector system (AMETEK-AMT ORTEC Co. GWL Series HPGe, Berwyn, PA, USA). The constant initial concentration model was used to calculate the exponential decay of excess ^{210}Pb activities. Calculation details have been presented elsewhere [32].

2.3. Particle Size Determination

For the particle size determination, sediments were subjected to H_2O_2 digestion, followed by $(\text{NaPO}_3)_6$ dispersion. Particle size was determined using a Malvern Mastersizer 2000 laser particle size analyzer (Malvern Panalytical, Malvern, UK). The detection range of the particle size analyzer was between 0.02 μm and 2000 μm . The calibration standards were SiO_2 standard particle samples, which have particle sizes that range between 15 μm and 120 μm .

2.4. Bulk Elemental Composition

Approximately 1 M HCl was added into dry sediments in order to remove the inorganic carbon fraction in the sediments. The HCl was then removed using a pipette, and the

residues were dried in oven at 45 °C for two days to a constant weight. The dry sediment was homogenized and weighed onto tin foil, and the TOC was analyzed with a vario III Elemental Analyzer (Thermo Fisher, Bremen, Germany).

2.5. Sequential P Extraction

The sequential P extraction steps described by Ruttenberg [31] were used in this study. This method has been widely used, thus enabling us to compare our results with other studies. Precisely 0.5 g of dry sediment was weighed and added to 20 mL of 1 M MgCl_2 (which was pre-adjusted to pH 8 with NaOH) into a 50 mL centrifuge tube. The solution was extracted for 2 h. This was followed by two washings with MgCl_2 and one with H_2O , followed by centrifugation. The supernatant from this step was saved to determine the Ex-P content. After that, 20 mL of a citrate–dithionite–bicarbonate solution (which was made up of 32.4 g of $\text{C}_6\text{H}_5\text{Na}_3\text{O}_7 \cdot 2\text{H}_2\text{O}$, 42 g of NaHCO_3 , and 2.87 g of $\text{Na}_2\text{S}_2\text{O}_4$ in 500 mL of distilled water) was added to the residue, and the solution was extracted for 8 h. The content was centrifuged and washed with MgCl_2 and H_2O , and the supernatant was determined for Fe-P. In the next step, the residue was added to 20 mL of pH 4 acetate buffer, shaken for 6 h, centrifuged, and washed twice with MgCl_2 and once with H_2O . The supernatant was to be determined for Ca-P. Next, the residue was added to 1 M HCl and extracted for 16 h. The content was then centrifuged, and the supernatant was saved to be determined for De-P. In the final step, the residue was dried for one day at 80 °C and combusted at 550 °C for 5 h. Approximately 1 M HCl was added to the residue, followed by extraction for 16 h. The supernatant was determined for the OP fraction. A flow diagram showing the extraction steps has been presented elsewhere [33]. All extractions were carried out through shaking at room temperature. All P concentrations were determined using the molybdenum blue complex method. The absorbance was measured at an 885 nm wavelength with a UV-8000 UV-visible spectrophotometer (METASH, Shanghai, China). Ex-P, Fe-P, Ca-P, and De-P contribute to the sum of inorganic P (IP), and hence, the sum of IP and OP is the total P (TP). The sum of Ex-P, Fe-P, and OP is known as bioavailable P (BAP). Ex-P, Fe-P, Ca-P, and OP contribute to P_{reactive} .

3. Results

3.1. Sedimentation Rates

The slopes of $^{210}\text{Pb}_{\text{xs}}$ activities along the three sediment cores are shown in Figure 2. The constant initial concentration model was used to establish linear sedimentation rates along the cores. In ZS1, the average sedimentation rate from 0 cm to 18 cm layers was 0.36 cm year^{−1}, and the sedimentation rate from the 20 cm to 26 cm layers was 0.29 cm year^{−1}. Along ZS2, the sediment layers from 0 cm to 8 cm layers had an average sedimentation rate of 0.25 cm year^{−1}, whereas the layers from 10 cm to 16 cm had a rate of 0.30 cm year^{−1}. These rates corresponded to deposition ages between 1933 and 2016 along ZS1 and between 1968 and 2015 along ZS2. A strong correlation between $^{210}\text{Pb}_{\text{xs}}$ and time existed along ZS3, resulting in a constant accumulation rate of 0.15 cm year^{−1} and years along the core between 1932 and 2012. The multiple sedimentation rates along the three locations could be due the dynamic coastal area and because ZS3 was more protected by other small islands compared to ZS1 and ZS2. More details on the sedimentation rates have been presented elsewhere [32]. The ages along the sediment cores are presented in Appendix A Table A1.

3.2. Particle Size

Particle size was divided into clay (<4 µm), silt (4–63 µm), and sand (63–2000 µm) fractions. Particle sizes along ZS1 and ZS2 ranged between 14.70 µm and 33.07 µm, with particle size at ZS1, ZS2, and ZS3 ranging from 14.70 µm to 33.07 µm, between 25.10 µm and 28.84 µm, and between 18.15 µm and 31.94 µm, respectively. Particle sizes showed fluctuations along the sediment cores and showed an overall decrease from the bottom of the cores toward the surface sediments along ZS1 and ZS3 (Appendix A Table A1).

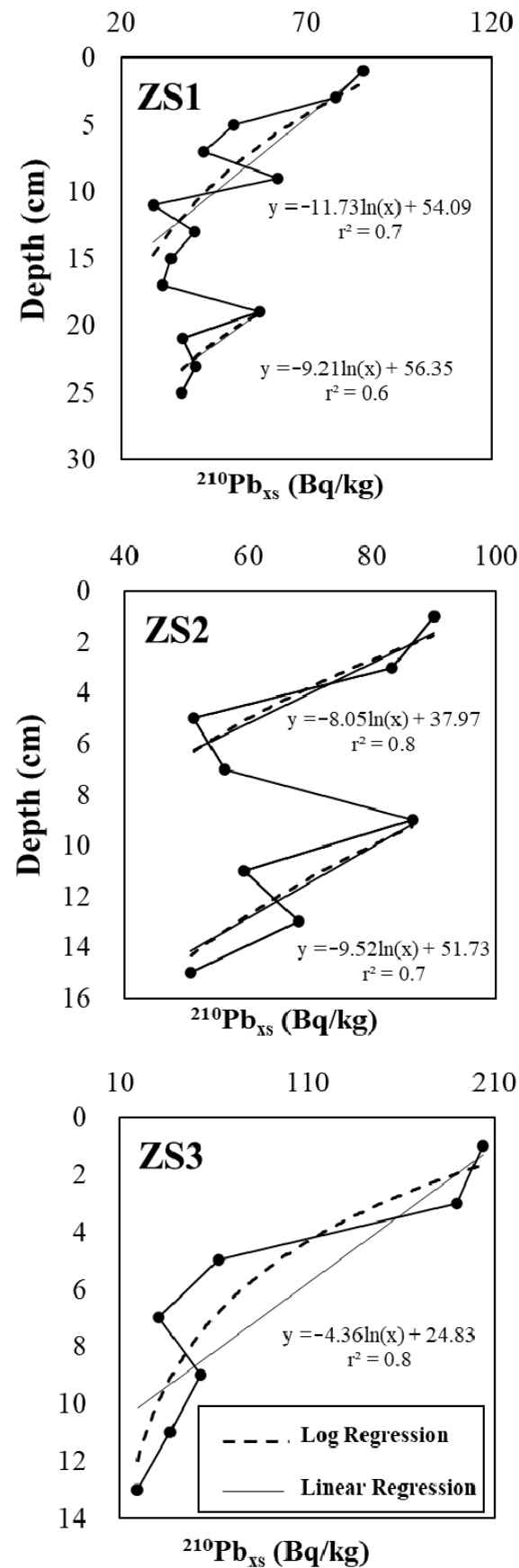


Figure 2. The decay slopes and the linear regression accumulation rate, where $x = ^{210}\text{Pb}_{\text{xs}}$ activity and $y = \text{depth}$.

3.3. TOC Contents

Results showed that TOC ranged between 0.38% and 0.62%. TOC along ZS1, ZS2, and ZS3 ranged from 0.47% to 0.62%, 0.52% to 0.59%, and 0.38% to 0.60%, respectively. The average TOC percentages at ZS1, ZS2, and ZS3 were 0.54%, 0.56%, and 0.48%. Overall, TOC contents increased from the bottom of the cores toward the present (Appendix A Table A1).

3.4. Sedimentary P Forms

Complete results and the means and ranges for sedimentary P species are presented in Appendix A. The TP concentrations along the three sediment cores ranged from 122.32 mg kg⁻¹ to 371.82 mg kg⁻¹. The ranges of concentrations of each P species were Ex-P (19.37–49.03 mg kg⁻¹), Fe-P (7.44–25.28 mg kg⁻¹), Ca-P (34.14–274.89 mg kg⁻¹), De-P (7.44–34.26 mg kg⁻¹), and OP (14.92–62.83 mg kg⁻¹). Thus, Ca-P was present in the most abundance, accounting for an average of 32%, 46%, and 66% of TP in the cores of ZS1, ZS2, and ZS3, respectively. Fe-P, on the other hand, was present in the lowest abundance, accounting for 5% to 9% of TP in these cores. The average abundance of the different sedimentary P species in ZS1 was in the following order: Ca-P > OP > Ex-P > De-P > Fe-P; in ZS2, it was Ca-P > Ex-P > OP > De-P > Fe-P; and in ZS3, it was Ca-P > OP > De-P > Ex-P > Fe-P. The overall average abundance of different sedimentary P species along the three cores was Ca-P (which accounted for 52% of TP) > OP (accounting for 16% of TP) > Ex-P (comprising 14% of TP) > De-P (representing 11% of TP) > Fe-P (comprising 7% of TP; Figure 3).

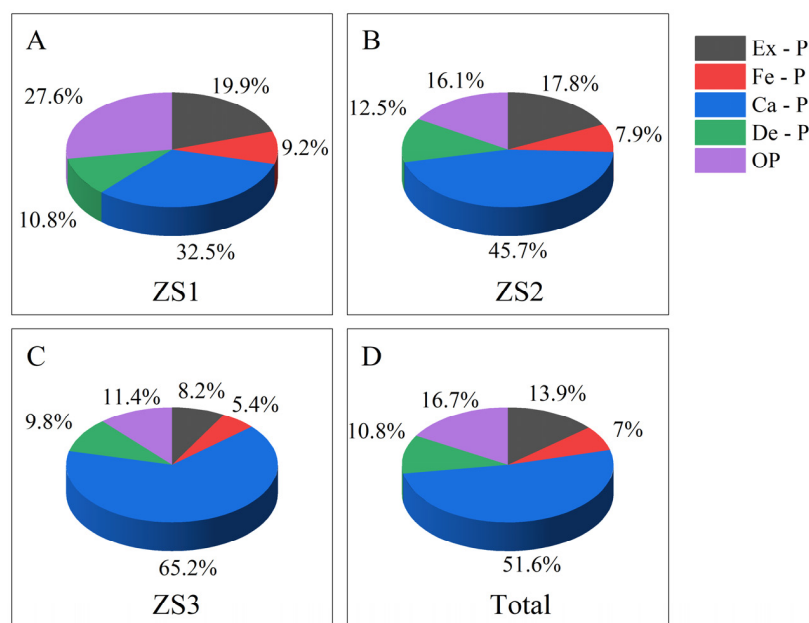


Figure 3. Pie charts showing the percentages of each P species relative to TP along (A) ZS1, (B) ZS2, (C) ZS3, and (D) the overall average values for all the locations.

Both Ex-P and OP showed a slight decrease from ZS1 (the location at the western part of the coastal area) to ZS3 (which was situated at the southeastern part compared to ZS1 and south of ZS2), whereas Fe-P increased slightly from ZS1 to ZS3. Ca-P, on the other hand, showed a drastic increase from ZS1 (mean = 45.60 mg kg⁻¹) to ZS2 (82.69 mg kg⁻¹) and ZS3 (182.17 mg kg⁻¹). Because Ca-P contributed importantly to IP and TP, both IP and TP also showed a drastic increase from ZS1 to ZS3. TP showed an increase from core ZS1 (122.31–185.20 mg kg⁻¹; average 140.31 mg kg⁻¹) to ZS2 (range 126.22–247.96 mg kg⁻¹; average 180.80 mg kg⁻¹) and ZS3 (172.86–371.82 mg kg⁻¹; average 279.39 mg kg⁻¹; Appendix A Table A2). Vertical distribution-wise, Ex-P, Fe-P, and OP showed much fluctuation across the cores (Figure 4). As Ca-P contributed significantly

to TP (Appendix A Table A3), both Ca-P and TP showed similar patterns of a decreasing proportion from bottom to surface sediments (Figure 4).

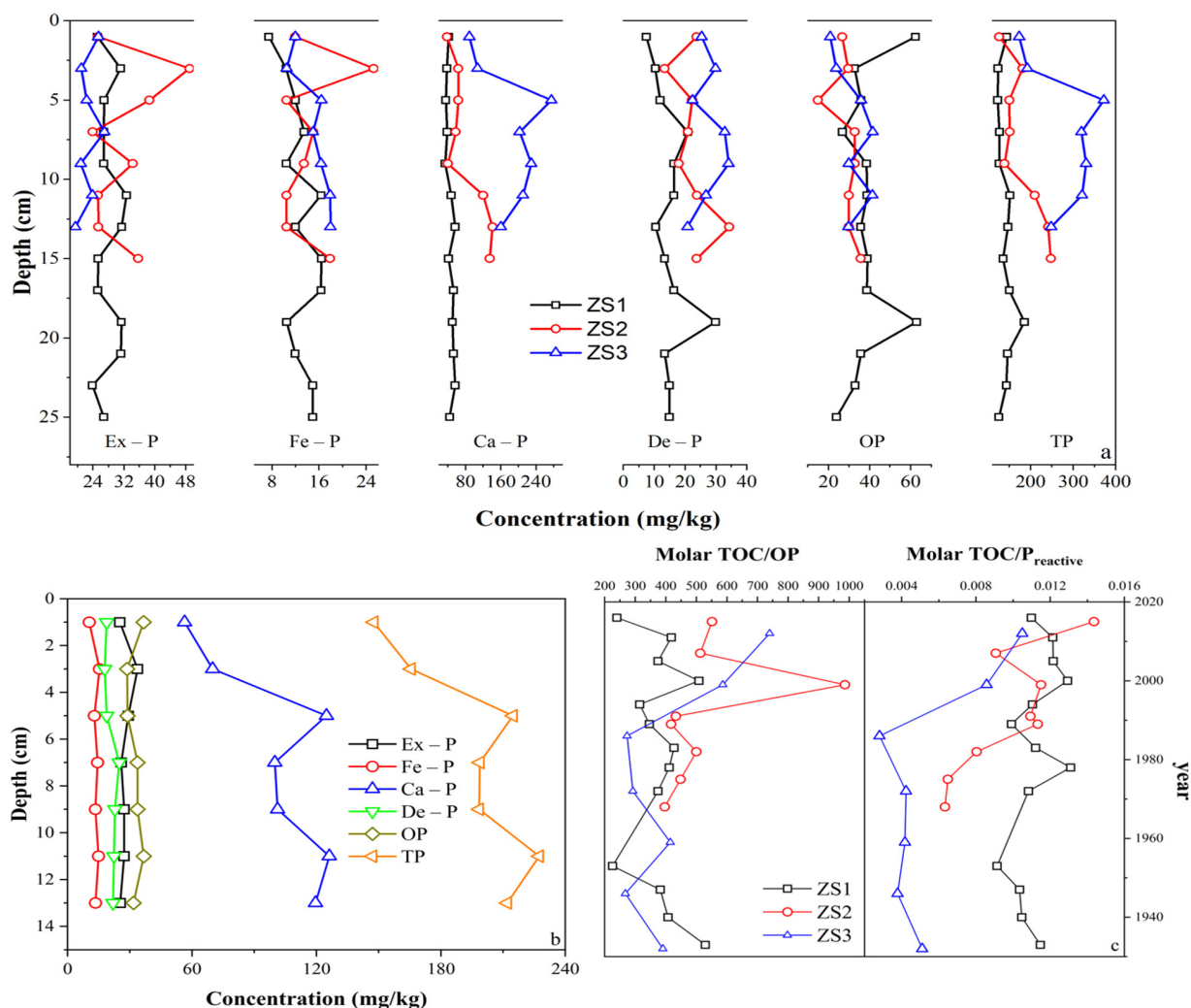


Figure 4. Vertical profile of (a) different sedimentary P forms along ZS1, ZS2, and ZS3, (b) mean values of sedimentary P forms for all locations, and (c) OC/OP and OC/P_{reactive} molar ratios along the cores.

The TOC/OP molar ratios at ZS1, ZS2, and ZS3 ranged from 225 to 529, 397 to 986, and 268 to 740, respectively. The average TOC/OP molar ratios were 382, 531, and 424 at ZS1, ZS2, and ZS3. The TOC/OP molar ratios along ZS2 and ZS3 showed an increasing trend from the bottom to surface sediments. On the other hand, the TOC/P_{reactive} ratios were very low and ranged from 0.009 to 0.013, 0.006 to 0.012, and 0.003 to 0.010 along cores ZS1, ZS2, and ZS3. Both TOC/OP and TOC/P_{reactive} increased from the bottom to the surface of the cores (Figure 4).

4. Discussion

4.1. Sources of Sedimentary P Species

Coastal zones affected by pollution and wastewater normally have high levels of BAP [3,4,34]. Hence, a high percentage of BAP relative to TP has been observed in coastal areas impacted by pollution (>50% of TP) [8] and high population (70–95.9% of TP) [35]. The percentage of BAP relative to TP along the sediment cores in this study was 37%, which is within the percentage of BAP relative to TP in the Zhejiang coastal zone (20–34% of TP) [27] and East China Sea (29.7% of TP) and within the lower limit of percentage of BAP relative to TP in the Yangtze Estuary (15.6–58.5%) [24], indicating that the Zhoushan coast

is not directly impacted by wastewater input, but there is potential for the sediments along this coastal area to release P into the water.

The De-P fraction, which originates from rock materials due to weathering and erosion processes, does not decompose easily, is relatively stable, and is indicative of land sources [28,36]. Sediments with De-P accounting for the highest portion of TP have been found nearer land, which were influenced by rivers [26], bays [37], and at the central part of the ocean receiving atmospheric input [38]. Previous studies have found a predominance of De-P, while Ca-P accounted for the second or third lowest portion of TP in the sediments in the nearby water off Zhejiang province [23–25]. The low De-P concentrations along the southern coast of Zhoushan Island could be due to a lesser contribution from riverine materials, which is in agreement with the Zhoushan islands being located farther from the influence of rivers from the mainland; this coastal area of rapid water movement did not facilitate the accumulation of materials and was due to the rapid transformation of other sedimentary P species, resulting in a difference in the composition of P species compared to the adjacent coastal areas.

Ca-P can be present in the forms of biogenic and authigenic apatite [39]. Ca-P can also originate from marine origins, like shell materials [40]; thus, locations near shellfish farming usually have high amounts of sedimentary Ca-P [41]. The highest composition of Ca-P in the Zhoushan coastal area could be attributable in part to the Zhoushan Archipelago being an important fishing ground [14]. On the other hand, the high concentration of Ca-P in this study area could also be attributable to the formation of Ca-P at the expense of the release of other P species from the sediments, as discussed in the following section.

4.2. Transformation of Sedimentary P Species

The TP values along the three sediment cores (ranging from 122.32 mg kg^{−1} to 371.82 mg kg^{−1}) fall within the lower limit of the TP concentrations of nearby coastal areas, such as the surface sediment (451.60–1140.14 mg kg^{−1}) and sediment core of the Yangtze River Estuary (529.96–760.41 mg kg^{−1}) [23] and the TP of the surface sediments of the Yangtze River Estuary and East China Sea, which ranged from 464.61 mg kg^{−1} to 662.84 mg kg^{−1} [24]. The TOC contents along ZS1 (0.49–0.62%), ZS2 (0.52–0.59%), and ZS3 (0.38–0.60%) were also lower compared to the adjacent coastal areas, such as the Yangtze River Estuary (0.60–0.98%), Hangzhou Bay (0.84–0.98%), and the East China Sea (0.17–0.92%) [42]. The high tidal currents [20], velocity, and intensity of the water movement [21] could have resulted in the rapid transportation and decomposition of materials and further decomposition of the resuspended particles, all of which could have resulted in less material accumulation along the coast. Similarly, there are other studies that found that strong tidal currents increased aerobic respiration and organic matter decomposition and reduced the burial of OC and N [43].

Sediments can easily release the Ex-P, Fe-P and OP to the aquatic environments, with Ex-P released to overlying water during environmental changes via desorption processes and used by phytoplankton and OP released from the sediments during organic matter decomposition [7,28,36,44]. Thus, a decrease in the Ex-P concentrations from ZS1 (mean = 28.02 mg kg^{−1}) and ZS2 (mean = 32.18 mg kg^{−1}) toward ZS3 (mean = 22.78 mg kg^{−1}) and OP decreasing slightly from ZS1 (mean = 38.74 mg kg^{−1}) to ZS2 (mean = 29.06 mg kg^{−1}) and ZS3 (mean = 31.87 mg kg^{−1}) could indicate the release of Ex-P and OP from the sediments during transportation across this coastal zone, Ex-P due to physical disturbances of the water column, and OP through organic matter decomposition. P is adsorbed by iron oxides and hydroxides under oxidized conditions as Fe-P, and Fe-P is released to the water column under anoxic conditions due to the reduction of iron oxides [45,46]. Thus, the slight increase in Fe-P from ZS1 (mean = 12.84 mg kg^{−1}) to ZS2 (mean = 14.35 mg kg^{−1}) and ZS3 (mean = 15.13 mg kg^{−1}) indicates no Fe-P release across the coastal area; instead, it indicates P adsorption by the sediments, as the iron-bound fraction occurred due to the likely oxygenated condition along the coastal area.

The P mobilized from sediments can be utilized in the formation of apatite or be absorbed by iron oxides [47]. Hence, a high Ca-P portion could be due to its formation

from the P released from the Ex-P, Fe-P, and OP fractions [24,28]. The drastic increase in Ca-P, coupled with a slight decrease in Ex-P and OP from ZS1 to ZS3 (Figure 5) could indicate a contribution of some Ex-P and OP to Ca-P formation during transportation along this coastal area. The significant correlation of TOC with Ca-P ($r = -0.66, p < 0.05$) and IP ($r = -0.63, p < 0.05$) and with TP ($r = 0.63, p < 0.05$) could be associated with the increased formation of Ca-P due to increased organic matter or TOC decomposition. Thus, besides the contribution from the Zhoushan fishing ground, the Ca-P along this coastal area was formed at the expense of these bioavailable P species. Decreasing trends in both TOC/OP and TOC/ P_{reactive} from the surface toward the bottom of the sediment cores indicate increased organic matter decomposition with an increasing depth. The vertical distribution also showed a contribution of P species to Ca-P formation with an increasing depth, in this case from Ex-P and Fe-P (as indicated by the weak negative correlations between Ca-P with Ex-P and Fe-P in the ZS2 core; Table A3). Slight anoxic conditions could occur with an increasing depth; thus, some Fe-P was released from the sediments and contributed to the formation of Ca-P. Other studies have also shown the transformation of sedimentary P species; for example, Coelho et al. [48] found that BAPs, such as Ex-P and OP contributed to the formation of Ca-P; Hou et al. [23] demonstrated the post-depositional reorganization of P via the conversion of OP and Fe-P to Ca-P; and Oxmann and Schwendenmann [49] showed an increase in Ca-P at the expense of adsorbed P under an increasing pH condition.

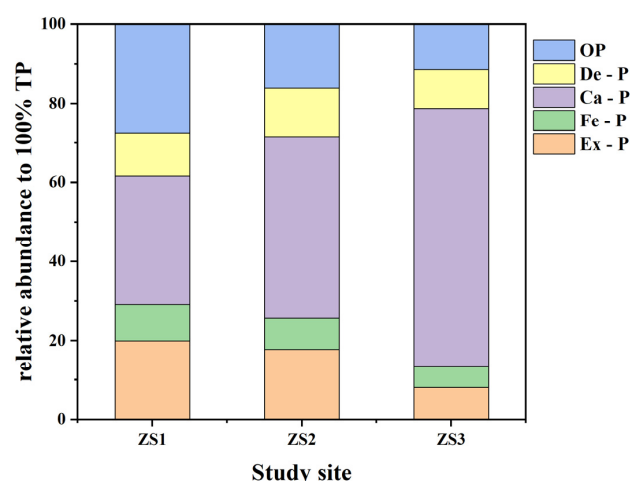


Figure 5. Comparison of the mean abundances of the sedimentary P species along ZS1, ZS2, and ZS3 along the southern coast of the Zhoushan Island.

The TOC/OP molar ratio can be used as an indicator of organic matter sources and diagenesis, as fresh phytoplankton has an TOC/OP ratio of about 106 [50], and a TOC/OP ratio higher than 106 can indicate the presence of land-derived organic matter [51] or that OP is released preferentially to OC during organic matter decomposition [39], and an OC/OP ratio lower than 106 can indicate the presence of bacterial biomass and/or extensive decomposition characterized by a low OC content [51]. The high TOC/OP molar ratios along the southern Zhoushan coast indicates P lost during early diagenesis, which is consistent with the extensive organic matter decomposition occurring along this coastal area. The extremely low values of TOC/ P_{reactive} due to high abundance of Ca-P further support the extensive formation of Ca-P along this coastal area.

4.3. Impacts of Climate Change and Human Activities on P Dynamics

In the seasonally hypoxic Changjiang Estuary, increased organic matter and nutrient discharge and increased stratification have resulted in decreased dissolved oxygen or hypoxia [52], which in turn resulted in increased P cycling in the coastal area [29]. As for the Zhoushan Island coastal areas, the overall decreasing trends of Ca-P and TP from bottom to surface sediments, coupled with the rapid flowing water, indicates that the coastal area is not

likely to be subjected to increased P pollution from human activities in the future. On the other hand, the decreasing trend of particle size from the bottom to surface of the sediment cores in this study is in accordance with similar trends observed in downstream of the Yangtze River [53] and in the coastal wetland ecosystem off Hangzhou Bay [54] as a result of the decreased sediment load from the Yangtze River [16]. The trend of decreasing particle size is associated with an overall decrease in sediment discharge and turbidity. Decreased turbidity, coupled with increased nutrient discharge from the rivers, could result in increased algal blooms and subsequent organic matter decomposition [32], thus releasing OP that could be contributing to the formation of Ca-P, a phenomenon that could be corroborated in this study based on the significant positive correlation between particle size and OP and the significant negative correlation between the particle size and Ca-P (Appendix A Table A3).

Higher temperatures promote microbial activity, which accelerates oxygen consumption and reduces redox potential. As a consequence, P was mobilized to the overlying water due to the enhanced release of the Fe-P fraction due to the reduction of Fe(oxyhydr)oxides [55]. Thus, an increased temperature has been associated with anoxia and P release due to the mobilization of the sediment Fe-P pool [56]. A study has found that summer hypoxia facilitated P release through organic matter decomposition and from the dissolution of Fe-P, but a decreasing temperature and the disappearance of stratification caused the autumn water to return to oxygen-rich conditions, which promoted the formation of Fe-P via precipitation with Fe/Mn oxides [57]. Studies have also found that high C and N can result in P release due to organic matter decomposition [58] and cyanobacterial blooms, resulting in increased pH and a decreased redox potential and subsequent increase in P mobility [59] or algal blooms causing the accelerated mineralization of sedimentary OC and OP [60]. Because the coastal area of Zhoushan Island was likely oxygenated due to the rapid water movement, we presume that the likely impact of climate change on the P cycling in the coast would be in terms of the increasing temperature causing increased organic matter decomposition and OP release at the surface sediments and increased organic matter decomposition causing anoxic conditions at sub-surface sediments that resulted in release of the Fe-P fraction.

5. Conclusions

This study provides new insights into P cycling in a zone of high currents and rapid water cycling. High water velocity and the occurrence of sediment resuspension have facilitated organic matter decomposition and resulted in low P accumulation across this coastal area. Instead, Ca-P was formed from the Ex-P and OP released from sediments during transportation across this coastal area, and sediments released Ex-P under increased water perturbations and released OP during organic matter decomposition. Besides its formation at the expense of Ex-P and OP, the high abundance of Ca-P along this coastal area was also likely contributed to by the Zhoushan fishing ground. Fe-P did not contribute to Ca-P formation in this likely oxygenated coastal zone, but Fe-P might contribute to Ca-P formation with an increasing sediment depth. The very high TOC/OP (ranging from 226 to 986) and low TOC/P_{reactive} (ranging from 0.003 to 0.014) provide further evidence of increased organic matter decomposition and the extensive formation of Ca-P during early diagenesis. The most probable impact of climate change on the P dynamic in the Zhoushan coast would be an increasing temperature resulting in increased organic matter decomposition and OP release or higher temperature causing increased anoxic conditions and Fe-P release at sub-surface sediments. More studies should be conducted to determine the impacts of climate change and human activities on the biogeochemical cycling of sedimentary P species in the coastal zone and also in zones of rapid current or water movement.

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Appendix A

Table A1. Characteristics of sediments based on sedimentary P species, TOC, particle size, and OC/OP and OC/P_{reactive} molar ratios.

(A) ZS1													
Depth (cm)	Year	Concentration (mg kg ^{−1})								TOC (%)	Particle Size (μm)	TOC/OP	OC/P _{reactive}
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	BAP	TP				
−1	2016	25.15	7.44	41.4	7.44	62.33	81.42	94.92	143.75	0.58	16.04	240	0.011
−3	2011	31.09	10.4	38.48	10.4	32.7	90.37	74.19	123.07	0.53	17.09	418	0.012
−5	2005	26.83	11.95	35.75	11.95	35.83	86.48	74.61	122.31	0.52	18.85	375	0.012
−7	2000	26.78	13.42	38.66	20.84	26.89	99.69	67.09	126.58	0.53	16.58	509	0.013
−9	1994	26.73	10.43	34.14	16.36	38.52	87.65	75.68	126.17	0.47	18.64	315	0.011
−11	1989	32.67	16.36	47.5	16.36	38.78	112.89	87.81	151.67	0.52	17.19	346	0.010
−13	1983	31.39	11.99	56.75	10.5	35.66	110.63	79.04	146.29	0.59	14.70	427	0.011
−15	1978	25.26	16.37	41.58	13.4	38.92	96.6	80.55	135.52	0.62	16.80	411	0.013
−17	1972	25.2	16.32	53.3	16.32	38.59	111.15	80.11	149.74	0.56	16.54	375	0.011
−19	1953	31.34	10.48	50.71	29.85	62.83	122.37	104.65	185.2	0.55	17.49	226	0.009
−21	1947	31.15	11.9	53.37	13.38	35.78	109.8	78.83	145.57	0.53	18.25	382	0.010
−23	1940	23.82	14.91	56.49	14.91	32.95	110.13	71.68	143.07	0.52	17.24	407	0.010
−25	1933	26.79	14.91	44.62	14.91	23.9	101.23	65.6	125.13	0.49	33.07	529	0.011

(B) ZS2													
Depth (cm)	Year	Concentration (mg kg ^{−1})								TOC (%)	Particle Size (μm)	TOC/OP	OC/P _{reactive}
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	BAP	TP				
−1	2016	25.25	11.91	38.59	23.77	26.69	99.53	63.85	126.22	0.57	27.25	551	0.014
−3	2011	49.03	25.28	63.87	13.41	29.68	151.59	103.99	181.27	0.59	26.44	513	0.009
−5	2005	38.64	10.44	63.87	22.32	14.92	135.27	64	150.18	0.57	26.53	986	0.012
−7	2000	23.9	14.96	58.19	20.92	32.78	117.97	71.64	150.75	0.55	28.84	433	0.011
−9	1994	34.29	13.45	40.24	17.92	32.78	105.9	80.52	138.68	0.53	26.51	417	0.011
−11	1989	25.32	10.46	120.46	23.84	29.94	180.08	65.72	210.02	0.58	26.01	500	0.008
−13	1983	25.33	10.46	141.32	34.26	29.94	211.38	65.73	241.32	0.52	25.10	448	0.006
−15	1978	35.63	17.84	134.97	23.77	35.78	212.19	89.25	247.96	0.55	26.27	397	0.006

(C) ZS3													
Depth (cm)	Year	Concentration (mg kg ^{−1})								TOC (%)	Particle Size (μm)	TOC/OP	OC/P _{reactive}
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	BAP	TP				
−1	2016	25.34	11.95	89.3	25.34	20.93	151.93	58.22	172.86	0.6	18.15	740	0.010
−3	2011	20.92	10.49	107.38	29.87	23.76	168.66	55.17	192.42	0.54	22.31	587	0.009
−5	2005	22.34	16.4	274.89	22.34	35.86	335.96	74.6	371.82	0.38	31.94	273	0.003
−7	2000	26.9	14.97	202.9	32.87	41.51	277.64	83.38	319.14	0.47	25.55	292	0.004
−9	1994	20.78	16.34	229.5	34.1	29.87	300.71	66.99	330.58	0.48	23.30	415	0.004
−11	1989	23.79	17.85	210.67	26.75	41.42	279.06	83.06	320.48	0.43	23.49	268	0.004
−13	1983	19.37	17.89	160.56	20.86	29.72	218.68	66.98	248.41	0.45	29.20	391	0.005

Table A2. Mean and ranges (in bracket) of sedimentary P species along cores ZS1, ZS2, and ZS3.

P Species	Concentration (mg kg ^{−1})		
	ZS1	ZS2	ZS3
Ex-P	28.02 (23.82–32.67)	32.18 (23.90–49.03)	22.78 (19.37–26.90)
Fe-P	12.84 (7.44–16.37)	14.35 (10.44–25.28)	15.13 (10.49–17.89)
Ca-P	45.60 (34.14–56.75)	82.69 (38.59–141.32)	182.17 (89.30–274.89)
De-P	15.12 (7.44–29.85)	22.53 (13.41–34.26)	27.45 (20.86–34.10)
OP	38.74 (23.90–62.83)	29.06 (14.92–35.78)	31.87 (20.93–41.51)
IP	101.57 (81.47–122.37)	151.74 (99.53–212.19)	247.52 (151.93–335.96)
TP	140.31 (122.31–185.20)	180.80 (126.22–247.96)	279.39 (172.86–371.82)
IP (%)	72% (57–81%)	84% (76–90%)	89% (87–91%)
OP (%)	28% (19–43%)	16% (10–24%)	11% (9–13%)

Table A3. Correlation analyses among the sedimentary parameters.

		(A) ZS1						
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	TP
Ex-P	Pearson correlation	1						
	Significance							
	N	13						
Fe-P	Pearson correlation	−0.162	1					
	Significance	0.597						
	N	13	13					
Ca-P	Pearson correlation	0.2	0.312	1				
	Significance	0.512	0.299					
	N	13	13	13				
De-P	Pearson correlation	0.191	0.152	0.132	1			
	Significance	0.532	0.621	0.668				
	N	13	13	13	13			
IP	Pearson correlation	0.421	0.453	0.816	0.613	1		
	Significance	0.152	0.12	0.001	0.026			
	N	13	13	13	13	13		
OP	Pearson correlation	0.084	−0.541	0.066	0.216	0.037	1	
	Significance	0.786	0.056	0.83	0.478	0.904		
	N	13	13	13	13	13	13	
TP	Pearson correlation	0.359	−0.035	0.632	0.586	0.745	0.694	1
	Significance	0.228	0.911	0.021	0.035	0.003	0.008	
	N	13	13	13	13	13	13	13

Table A3. Cont.

		(B) ZS2						
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	TP
Ex-P	Pearson correlation	1						
	Significance							
	N	8						
Fe-P	Pearson correlation	0.734	1					
	Significance	0.038						
	N	8	8					
Ca-P	Pearson correlation	−0.189	−0.172	1				
	Significance	0.654	0.765					
	N	8	8	8				
De-P	Pearson correlation	−0.66	−0.677	0.652	1			
	Significance	0.075	0.065	0.08				
	N	8	8	8	8			
IP	Pearson correlation	0.015	0.049	0.977	0.54	1		
	Significance	0.972	0.908	0	0.167			
	N	8	8	8	8	8		
OP	Pearson correlation	−0.194	0.367	0.29	−0.034	0.272	1	
	Significance	0.645	0.371	0.486	0.937	0.514		
	N	8	8	8	8	8	8	
TP	Pearson correlation	−0.012	0.096	0.972	0.511	0.992	0.394	1
	Significance	0.978	0.821	0	0.196	0	0.334	
	N	8	8	8	8	8	8	8

Table A3. Cont.

		(C) ZS3						
		Ex-P	Fe-P	Ca-P	De-P	OP	IP	TP
Ex-P	Pearson correlation	1						
	Significance							
	N	7						
Fe-P	Pearson correlation	−0.21	1					
	Significance	0.651						
	N	7	7					
Ca-P	Pearson correlation	−0.043	0.72	1				
	Significance	0.928	0.068					
	N	7	7	7				
De-P	Pearson correlation	0.282	−0.273	0.045	1			
	Significance	0.54	0.554	0.924				
	N	7	7	7	7			
IP	Pearson correlation	0.01	0.708	0.997	0.116	1		
	Significance	0.983	0.075	0	0.804			
	N	7	7	7	7	7		
OP	Pearson correlation	0.35	0.668	0.744	0.12	0.768	1	
	Significance	0.441	0.101	0.055	0.798	0.044		
	N	7	7	7	7	7	7	
TP	Pearson correlation	0.046	0.72	0.992	0.119	0.998	0.81	1
	Significance	0.921	0.068	0	0.8	0	0.027	
	N	7	7	7	7	7	7	7

Table A3. Cont.

(D) Results for all cores												
		All Cores										
		Fe-P	Ca-P	De-P	OP	IP	BAP	TP	TOC	Particle Size	TOC/OP	TOC/P _{reactive}
Ex-P	Pearson correlation	0.292	−0.36	−0.295	−0.0472	−0.269	0.55	−0.276	0.392	−0.0106	0.252	0.238
	Significance	0.131	0.0599	0.127	0.812	0.166	0.00244	0.155	0.0391	0.957	0.196	0.222
	N	28	28	28	28	28	28	28	28	28	28	28
Fe-P	Pearson correlation		0.314	0.000632	−0.135	0.371	0.33	0.352	−0.201	0.336	−0.126	−0.364
	Significance		0.103	0.997	0.493	0.0519	0.0864	0.0662	0.306	0.0808	0.523	0.0568
	N		28	28	28	28	28	28	28	28	28	28
Ca-P	Pearson correlation			0.635	−0.0424	0.992	−0.123	0.986	−0.66	0.473	−0.208	−0.936
	Significance			0.000281	0.83	3.79×10^{-25}	0.532	9.31×10^{-22}	0.000133	0.0109	0.289	2.78×10^{-13}
	N			28	28	28	28	28	28	28	28	28
De-P	Pearson correlation				−0.159	0.677	−0.279	0.654	−0.289	0.368	0.105	−0.601
	Significance				0.42	7.71×10^{-05}	0.15	0.00016	0.135	0.0539	0.593	0.000714
	N				28	28	28	28	28	28	28	28
OP	Pearson correlation					−0.0672	0.76	0.073	−0.0432	−0.399	−0.809	−0.111
	Significance					0.734	2.66×10^{-06}	0.712	0.827	0.0355	1.88×10^{-07}	0.573
	N					28	28	28	28	28	28	28
IP	Pearson correlation						−0.0816	0.99	−0.628	0.5	−0.169	−0.941
	Significance						0.68	9.52×10^{-24}	0.000348	0.00677	0.389	9.04×10^{-14}
	N						28	28	28	28	28	28
BAP	Pearson correlation							0.025	0.102	−0.235	−0.577	−0.079
	Significance							0.899	0.604	0.228	0.00131	0.69
	N							28	28	28	28	28
TP	Pearson correlation								−0.634	0.444	−0.283	−0.956
	Significance								0.000295	0.018	0.145	2.01×10^{-15}
	N								28	28	28	28

Table A3. Cont.

(D) Results for all cores												
		All Cores										
		Fe-P	Ca-P	De-P	OP	IP	BAP	TP	TOC	Particle Size	TOC/OP	TOC/P _{reactive}
TOC	Pearson correlation Significance N									−0.411	0.424	0.657
										0.0296	0.0247	0.000147
										28	28	28
Particle size	Pearson correlation Significance N											−0.427
												0.0235
												28
TOC/OP	Pearson correlation Significance N											0.348
												0.0696
												28

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