



Effect of reclamation on C, N, and P stoichiometry in soil and soil aggregates of a coastal wetland in eastern China

Caiyao Xu^{1,2} · Lijie Pu^{1,2} · Jianguo Li³ · Ming Zhu^{1,2}

Received: 19 January 2018 / Accepted: 3 September 2018 / Published online: 12 September 2018
© Springer-Verlag GmbH Germany, part of Springer Nature 2018

Abstract

Purpose The elements carbon (C), nitrogen (N), and phosphorus (P) in soil are crucial to all biological processes. Reclamation in coastal wetlands would alter the balance of multiple chemical elements in soils and sediments. This study aimed to investigate the effect of reclamation on total organic carbon (TOC), total nitrogen (TN), total phosphorus (TP), and C:N:P stoichiometry in bulk soil and soil aggregates of a coastal wetland in eastern China.

Materials and methods The space-for-time substitution method was used to determine the impact of reclamation on soil properties. Field investigation and soil sampling were conducted in September 2012 and April 2014. Four experimental sites were reclaimed in 1951, 1974, 1982, and 2007. The tidal flat was regarded as the control. We sampled soils based on the random uniform grid method to ascertain the basic trends in soil physicochemical properties for the whole study area. After identifying two areas with significant differences, we mainly selected the seasonally typical land use/cover types to analyze the effects of land use/cover on TOC, TN, TP, and C:N:P stoichiometry in bulk soil and soil aggregates (macroaggregate, > 250 μm , microaggregate, 53–250 μm , and silt + clay fraction, < 53 μm).

Results and discussion Soil TOC and TN content significantly increased, and EC and pH decreased after reclamation. C:N decreased from 19.997 to 9.527, while C:P and N:P increased from 4.441 to 8.101 and 0.222 to 0.850 respectively over the reclamation time. The main aggregate fractions were microaggregates in the natural wetland and macroaggregates after reclamation. TOC and TN were more likely to be reserved in macroaggregates, while TP was mainly stored in the silt + clay fraction. Compared with the sediments in the tidal flat, the effects of land use/cover types on TOC, TN, TP, and C:N:P stoichiometry in both bulk soil and soil aggregates after the 63-year reclamation were not significant.

Conclusions Long-term chronosequence reclamation could radically improve soil properties in coastal wetlands. Soil physicochemical properties tended to reach a steady state 32 years after reclamation. TOC and TN were mainly reserved in macroaggregates, and increasing the proportion of macroaggregates can contribute to increase TOC and TN content. Soil development owing to the long-term reclamation played a relatively more important role in the soil nutrient stoichiometry than land use/cover change.

Keywords C:N:P stoichiometry · Coastal reclaimed area · Eastern China · Soil aggregates

Responsible editor: Chengrong Chen

✉ Lijie Pu
ljpu@nju.edu.cn

¹ School of Geographic and Oceanographic Sciences, Nanjing University, Nanjing 210023, China

² The Key Laboratory of the Coastal Zone Exploitation and Protection, Ministry of Land and Resources, Nanjing 210023, China

³ School of Geography, Geomatics and Planning, Jiangsu Normal University, Xuzhou 221116, China

1 Introduction

Coastal wetlands are located at the interface between terrestrial and marine environments and play a vital role in the sustainability of regional development. They provide enormous direct and indirect ecosystem services, including nutrient cycling, carbon storage, and tourism and recreation (Ma et al. 2014). Over the past decades, the reclamation of coastal wetlands for agricultural and architectural land use is a common practice worldwide and has become an important way in China to accommodate the increasing demand for space for

living and development in China (Wang et al. 2014a; Tian et al. 2016). Coastal reclamation, including the embanking and filling of coastal wetlands and construction of seawalls, levees, and dikes along the shoreline (Tian et al. 2016), usually occurs in large river estuaries and coastal areas with rapid-speed sediment, such as Jiangsu, eastern China (Li et al. 2014). Previous studies showed that with reclamation, land use/cover types (LUTs) gradually transformed from natural wetlands to artificial areas (Xu et al. 2016), and the landscape had higher fragmentation but lower diversity (Li et al. 2010). Research indicated that the heterogeneous soil, to some extent, affected the landscape pattern and vegetation distribution (Mayes et al. 2014). Soil properties varied significantly in coastal wetland following reclamation. The study of evolutionary processes in soil is essential for the sustainability of coastal wetlands following reclamation (Iost et al. 2007; Sun et al. 2011; Xu et al. 2014).

The elements carbon (C), nitrogen (N), and phosphorus (P) are the main nutrients in soil and are crucial to all biological processes. C:N:P stoichiometry predicts how the relative ratios of these quantities can have dramatic effects on biological processes. Redfield (1958) reported that there is a constrained C:N:P ratio (106:16:1 M ratio) in planktonic and ocean waters. Researchers summarized the C:N:P stoichiometry in plants and soil to discover a similar rule at different scales, such as the national (Tian et al. 2010) and the global scale (Cleveland and Liptzin 2007; Xu et al. 2013). C:N:P stoichiometry is considered to be a powerful heuristic and predictive tool for analyzing ecological systems (Elser et al. 2000; Sterner and Elser 2002). Anthropogenic activity alters the equilibrium among soil C, N, and P in coastal wetland (Guo and Gifford 2002). Reports showed that N:P in both soil and plants was well-constrained in the reed-dominated wetlands of the Yellow River Delta (Qu et al. 2014), and the biogeochemical processes of coastal wetlands were more limited by N and P in the Yellow River Delta (Qu et al. 2014) and Laizhou Bay, Bohai Sea (Cao et al. 2015). However, C:N:P stoichiometry in soil has yet to be fully described (Zhang et al. 2013).

Further, to a large extent, soil functioning is defined by its structure, and soil structure is regarded as an important regulator of many soil physical and biological processes (Mikha and Rice 2004). Approaches based on the physical fractionation of soils, which permit soils to be separated into several size fractions, may provide information on soil functioning. Soil aggregates not only physically protect soil organic matter (Tisdall and Oades 1982) but also affect the microbial community (Schutter and Dick 2002) and determine nutrient cycling (Wang et al. 2001). The aggregation process often occurs in a hierarchy of three main size classes: free primary particles (<2 μm in diameter), microaggregates (2–250 μm), and macroaggregates (> 250 μm), which were first proposed by Tisdall and Oades (1982) and then improved by Oades (1984). Afterwards,

the main aggregates became macroaggregates (> 250 μm), microaggregates (53–250 μm), and the silt + clay fraction (< 53 μm) (Elliott 1986; Six et al. 2002; Six et al. 2000; Six and Paustian 2014). C:N:P stoichiometry in soil aggregates has been explored since the 1980s (Elliott 1986). Studies showed that smaller aggregates were more sensitive to fertilization (Jiang et al. 2017) and contained more C, N, and P than larger ones (Elliott 1986; Jiménez et al. 2008). Hence, the variations of C, N, P, and C:N:P stoichiometry in soil aggregates could further contribute to describing the mechanisms of soil property changes. However, little is known about C, N, and P stoichiometrical characteristics in the soils aggregates of coastal wetlands.

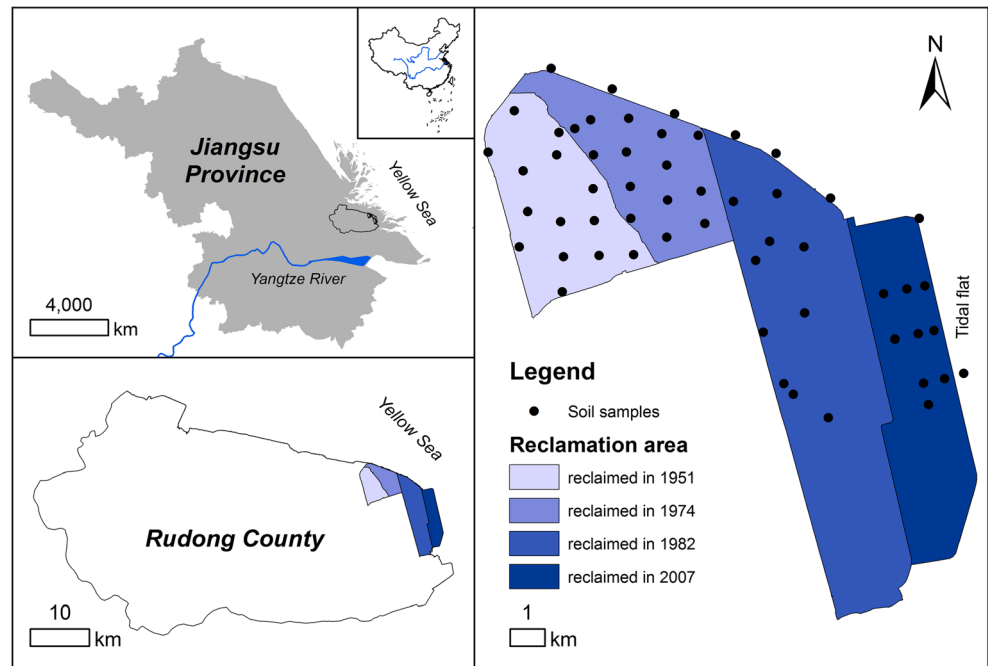
The objectives of this study were to identify the effects of coastal wetland conversion to artificial land on C, N, P, and C:N:P stoichiometry of bulk soil and soil aggregates and to explore the linkage among the nutrients in bulk soil and aggregates during long-term reclamation. We hypothesized that (i) long-term reclamation would have significant impact on soil structure linked to nutrient storage and these effects would be more pronounced in larger soil structures (e.g., macroaggregates), and (ii) different LUTs had less influence on the aggregate size and nutrient distribution than year of reclamation. To test these hypotheses, we used soil samples collected from a chronosequence of reclaimed areas and selected the oldest (63-year-old) reclaimed area to analyze the effect of LUTs on soil C, N, P, and C:N:P stoichiometry.

2 Materials and methods

2.1 Study area

This study was conducted at the reclaimed area of Rudong County, Jiangsu Province, eastern China (32° 14' 23"–32° 38' 10" N, 120° 41' 22"–121° 35' 25" E) (Fig. 1). This area is situated in the coastal plain of the Yangtze River Delta, and its north and east borders are the south Yellow Sea. Rudong, the largest alluvial coastal zone in the mid-latitude region, has a total land area of 1872 km² with an average elevation of 3.5–4.5 m (Zhang et al. 2016a). The climate is affected by subtropical and marine monsoons with a mean annual temperature and precipitation of 15 °C and 1028.6 mm, respectively. The tidal flat is currently expanding seaward at a rate of 25–30 cm per year. There is a history of over 60 years of reclamation. Since 1951, 302.09 km² of tidal flats have been reclaimed under a series of projects (Xu et al. 2014; Zhang et al. 2016a). The *Jiangsu Coastal Exploitation National Strategy* has been implemented since 2009 (Jiangsu Coastal Areas Development Office 2009). Therefore, reclamation activities in Rudong County are intensively conducted, which makes it a typical area for our study.

Fig. 1 The location of study area and soil samples



2.2 Field investigation and soil sampling

The space-for-time substitution method was used to determine the variation in TOC, TN, TP, and C:N:P stoichiometry following reclamation. The first and most important step was to identify the homogeneity of the soil sources observed in the different areas by trace metal analysis (specifically, the ratio of Ti to Zr). The results were presented in Li et al. (2018a) and verified that the soils of the different reclaimed areas in study area can be considered to share the same source. Field investigation and soil sampling were conducted in September 2012 and April 2014. We sampled soils according to reclamation time based on the random uniform grid method to document the basic trend of soil properties in study area in September 2012. The number of sampling points on the tidal flat and reclamation areas reclaimed in 2007 (7-year-old), 1982 (32-year-old), 1974 (40-year-old), and 1951 (63-year-old) was 8, 10, 10, 14, and 14, respectively (Fig. 1). After identifying two significant stages of reclamation (0-year-old and 63-year-old reclaimed areas), we mainly selected the seasonally typical LUTs (tidal flat, TF; aquaculture ponds, AP; rape, RP; wheat, WT; and broad bean, BB) to analyze the effects of land use on TOC, TN, TP, and C:N:P stoichiometry in bulk soil and soil aggregates (macroaggregates, > 250 μm , microaggregates, 53–250 μm , and the silt + clay fraction, < 53 μm) in April 2014. The number of sampling points for TF, AP, RP, WT, and BB was 10, 6, 7, 8, and 7, respectively. Soil profiles consisted of two soil layers at depths of 0–20 cm (topsoil) and 20–40 cm (subsoil). In the study area, the cropping system was mainly a wheat/broad bean/rape-corn rotation. The depth of plowing was 0–20 cm. NPK compound fertilizer is the main nutrient

input, and the amount applied to each type of field can be found in Li et al. (2018b). The aquaculture ponds here were located mostly near the 40-year-old reclaimed area; thus, their age was approximately 40 years and their water depth was around 1 m. Crop residues or litter on the soil surface were carefully removed before soil sampling. The soil samples for laboratory tests were air-dried, divided, and screened through a 2-mm sieve.

2.3 Soil physical and chemical analyses

Soil aggregates (macroaggregates, > 250 μm , microaggregates, 53–250 μm , and the silt + clay fraction, < 53 μm) were obtained by dry-sieving with nested flat sieves according to the procedures reported by Larney (1993) and Beauchamp and Seech (1990). We measured the weight of each aggregate fraction and the content of TOC, TN, and TP in soil aggregates. Soil pH and electrical conductivity (EC) were measured respectively, with a pH meter (Mettler Toledo, FiveEasy Plus, Switzerland) and a conductivity meter (Mettler Toledo, FiveEasy Plus with Conductivity Sensor LE703, Switzerland), using a 1:5 soil to deionized water ratio. Soil bulk density (BD) was determined by cutting a ring in undisturbed soil at each soil sampling point. TOC was determined by the dichromate redox colorimetric method (Heanes 1984; Larney 1993). TN was measured using dry combustion with a CNS Analyzer (Elementar Vario MICRO, Germany). TP was measured by perchloric acid digestion followed by the molybdenum-antimony anti-spectrophotometric method (O'Halloran and Cade-Menun 1993).

2.4 Data analysis

Soil C:N:P stoichiometry contained the ratios TOC:TN (C:N), TOC:TP (C:P), and TN:TP (N:P), which were expressed as mass ratios. The effects of years of reclamation on soil physicochemical properties and the effects of LUTs on TOC, TN, TP, and C:N:P stoichiometry in bulk soil and soil aggregates were assessed with one-way ANOVA using the Student–Newman–Keuls post hoc test or the Games–Howell test with significance defined at 0.05. The homogeneity of variance was examined by Levene’s test before ANOVA. The Pearson correlation coefficient was used to analyze the relationships among TOC, TN, TP, and C:N:P stoichiometry in bulk soil and soil aggregates. The data in Fig. 6 were log₁₀-transformed before analysis. ANOVA was performed using SPSS18.0 for Windows (Statistical Graphics Crop, Princeton, USA). Figure 1 was created with ArcGIS (version 10.2; Environmental Systems Research Institutes, Inc., Redlands, CA, USA). R (version 3.4.3; R Core Team) was used to conduct Pearson correlation analysis and construct other figures.

3 Results

3.1 Basic soil properties and C:N:P stoichiometry in bulk soil over time

Soil properties in the coastal reclaimed wetland varied markedly depending on reclamation years (Fig. 2). The

concentration of TOC, TN, and TP significantly increased from 3.019 g/kg, 0.151 g/kg, and 0.680 g/kg to 6.337 g/kg, 0.651 g/kg, and 0.782 g/kg respectively ($p < 0.05$) and EC and pH decreased from 21,171.272 to 783.772 $\mu\text{S}/\text{cm}$ and 8.621 to 8.261, respectively, after reclamation ($p < 0.05$). Soil BD initially decreased and then increased slightly after reclamation. Soil C:N decreased from 19.997 to 9.527, while soil C:P and N:P increased from 4.441 to 8.101 and 0.222 to 0.850, respectively, over time. Soil nutrients tended to accumulate after reclamation. Based on ANOVA, there was no significant difference between the 7-year-old reclaimed area and the natural wetland for all the soil properties in Fig. 2. Moreover, no significant differences were found among 32-year-old, 40-year-old, and 63-year-old reclaimed areas for TOC, TP, EC, and BD, which indicated that soil properties reached a relatively stable state after 32 years of reclamation.

3.2 Soil C, N, P, and C:N:P stoichiometry in bulk soil under different LUTs

LUTs in coastal wetlands changed sharply along with reclamation time. As in the analysis above, only natural wetlands and the 63-year-old reclaimed area were selected to analyze the features of soil TOC, TN, TP, and C:N:P stoichiometry under different LUTs and aggregate fractions. TOC, TN, and TP were generally higher in AP, RP, WT, and BB compared with TF ($p < 0.05$, Fig. 3). TOC content

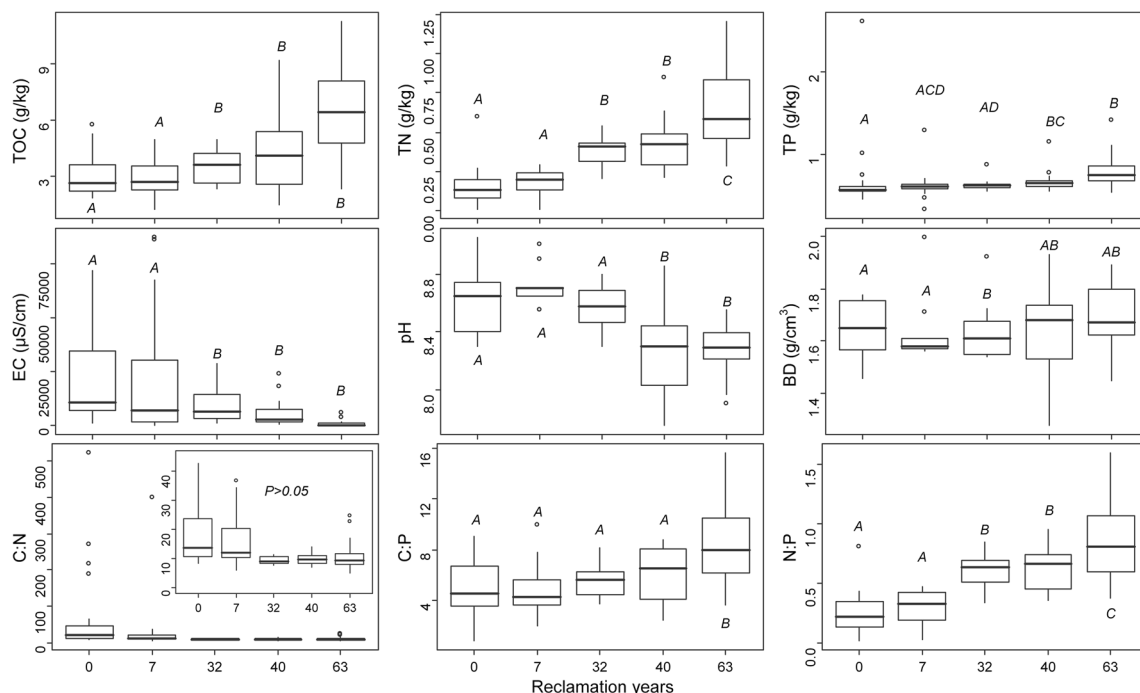


Fig. 2 Basic characteristic and C:N:P stoichiometry (C:N, TOC:TN; C:P, TOC:TP; N:P, TN:TP) in bulk soil (0–20 cm) over time. Different letters represent significant differences at $p < 0.05$

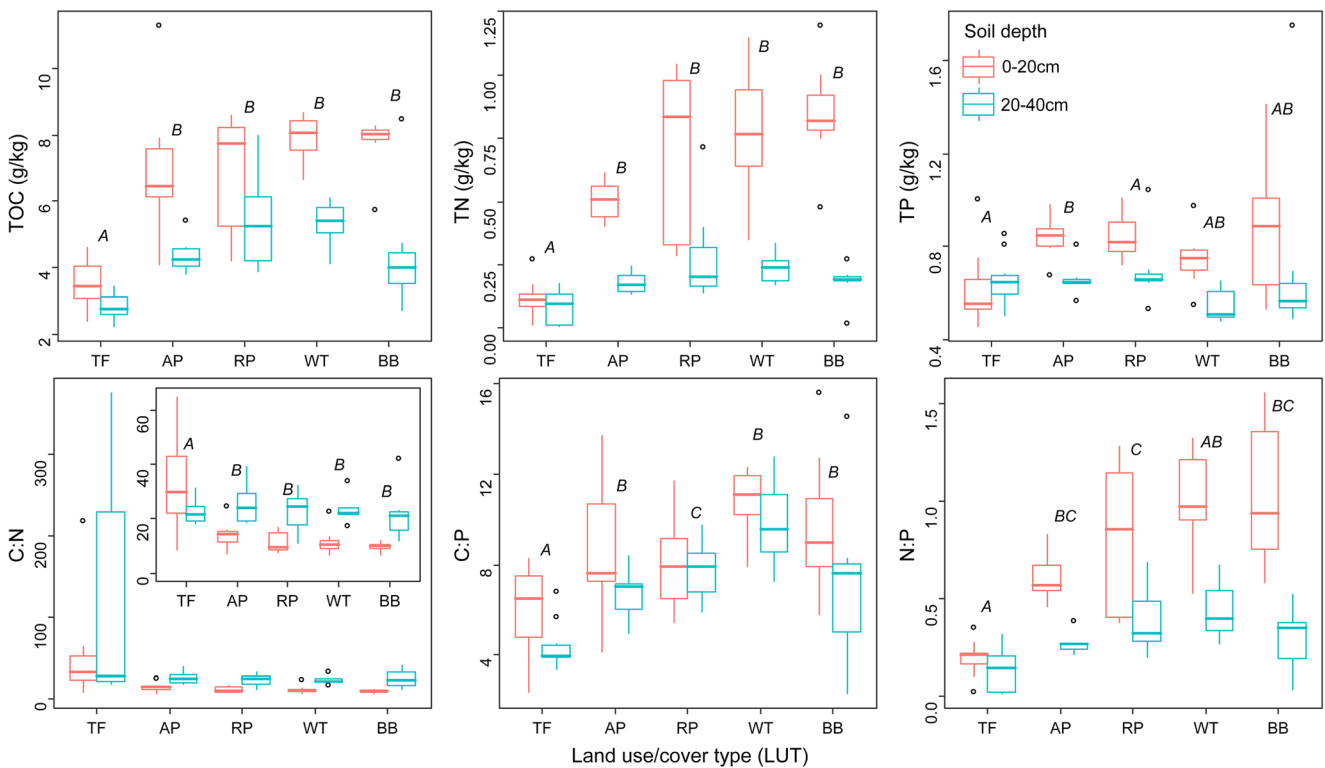


Fig. 3 Soil TOC, TN, TP, and C:N:P stoichiometry (C:N, TOC:TN; C:P, TOC:TP; N:P, TN:TP) in bulk soil with different LUTs (tidal flat, TF; aquaculture ponds, AP; rape, RP; wheat, WT; and broad bean, BB). Different letters represent significant differences at $p < 0.05$

was the lowest in TF, and the highest TN and TP in topsoil were found in BB.

Soil C:N was higher in subsoil than that in topsoil while C:P and N:P had the opposite pattern (Fig. 3). Soil C:N values for topsoil in TF, AP, RP, WT, and BB ranged from 8.7 to 218.5, 7.3 to 24.6, 8.3 to 17, 6.7 to 22.6, and 6.5 to 12, respectively. Soil C:P values of topsoil in TF, AP, RP, WT, and BB ranged from 2.4 to 8.3, 4.2 to 13.7, 5.4 to 11.7, 7.9 to 12.3, and 5.8 to 15.6, respectively. Soil N:P values of topsoil in TF, AP, RP, WT, and BB ranged from 0.02 to 0.35, 0.46 to 0.83, 0.37 to 1.28, 0.88 to 1.32, and 0.58 to 1.56, respectively. Specifically, C:N was the highest in TF. The analysis of variance indicated that there were significant differences between TF and other four LUTs, while in most cases, no significant differences were observed among these four types.

3.3 Aggregate size distributions

The distribution of soil aggregates changed significantly after reclamation (Fig. 4). The aggregate size distribution was dominated by microaggregates in TF, while macroaggregates became the major fraction in AP, RP, WT, and BB. The mass proportion of the silt + clay fraction was always accounted for the lowest ($p < 0.05$). There were no significant differences among AP, RP, WT, and BB.

3.4 Soil C, N, P, and C:N:P stoichiometry in soil aggregates under different LUTs

As seen in Fig. 5, at 0–40 cm, TOC content of each aggregate fraction remained relatively stable under different LUTs. TP content increased in macroaggregates and microaggregates in topsoil and decreased in subsoil after reclamation, but no significant differences were found among LUTs ($p > 0.05$). TOC decreased from macroaggregate > silt + clay fraction > microaggregate and TN from macroaggregate > microaggregate > silt + clay fraction, which indicated that TOC and TN was more likely to be stored in macroaggregates. TP tended to be stored in the silt + clay fraction. LUTs had no significant effect on TOC, TN, and TP of soil aggregates in the 63-year-old reclaimed area.

C:N:P in soil aggregates declined after 63 years of reclamation, which was similar with that in bulk soil. C:P in soil aggregates had a larger range of values than that in bulk soil. C:P in the silt + clay fraction was the lowest. N:P in soil aggregates increased after reclamation and was higher in topsoil than that in subsoil.

3.5 Relationships among C, N, P, and C:N:P stoichiometry in bulk soil and soil aggregates

Generally, the relationships of TOC and C:P, TN and C:N, TN and N:P, and C:N and N:P were similar in both bulk soil and

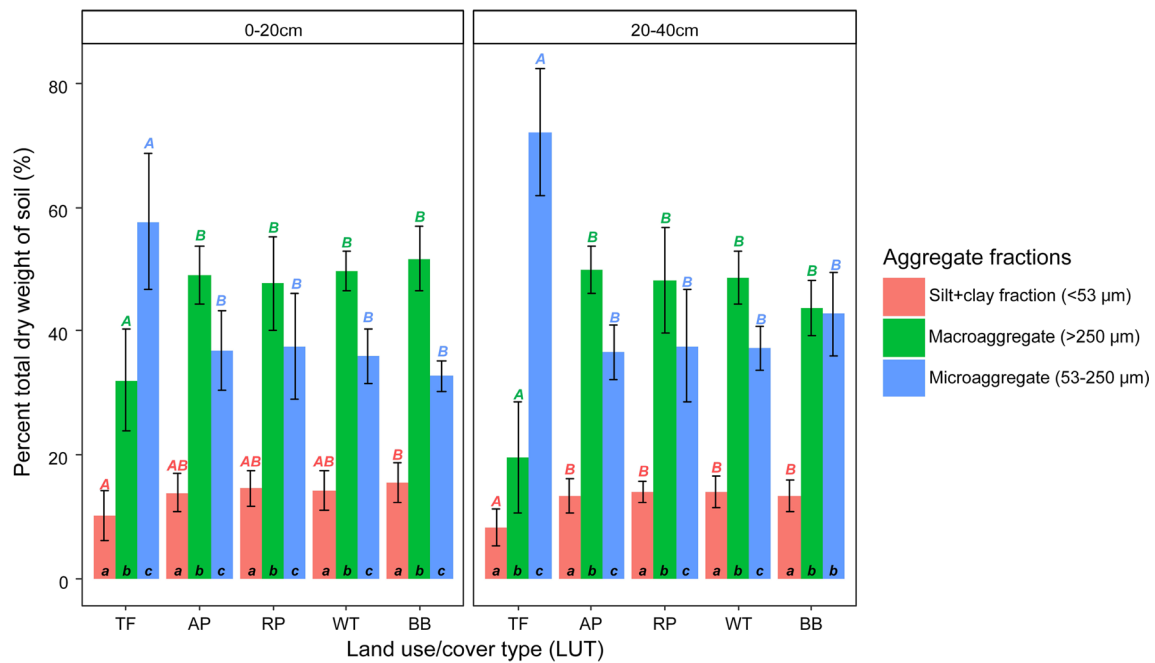


Fig. 4 Soil aggregate fraction distribution over time and under different LUTs (tidal flat, TF; aquaculture ponds, AP; rape, RP; wheat, WT; and broad bean, BB). Different uppercase letters represent significant

differences among LUTs in each aggregate fraction at $p < 0.05$. Different lowercase letters represent significant differences among aggregates under each LUT at $p < 0.05$

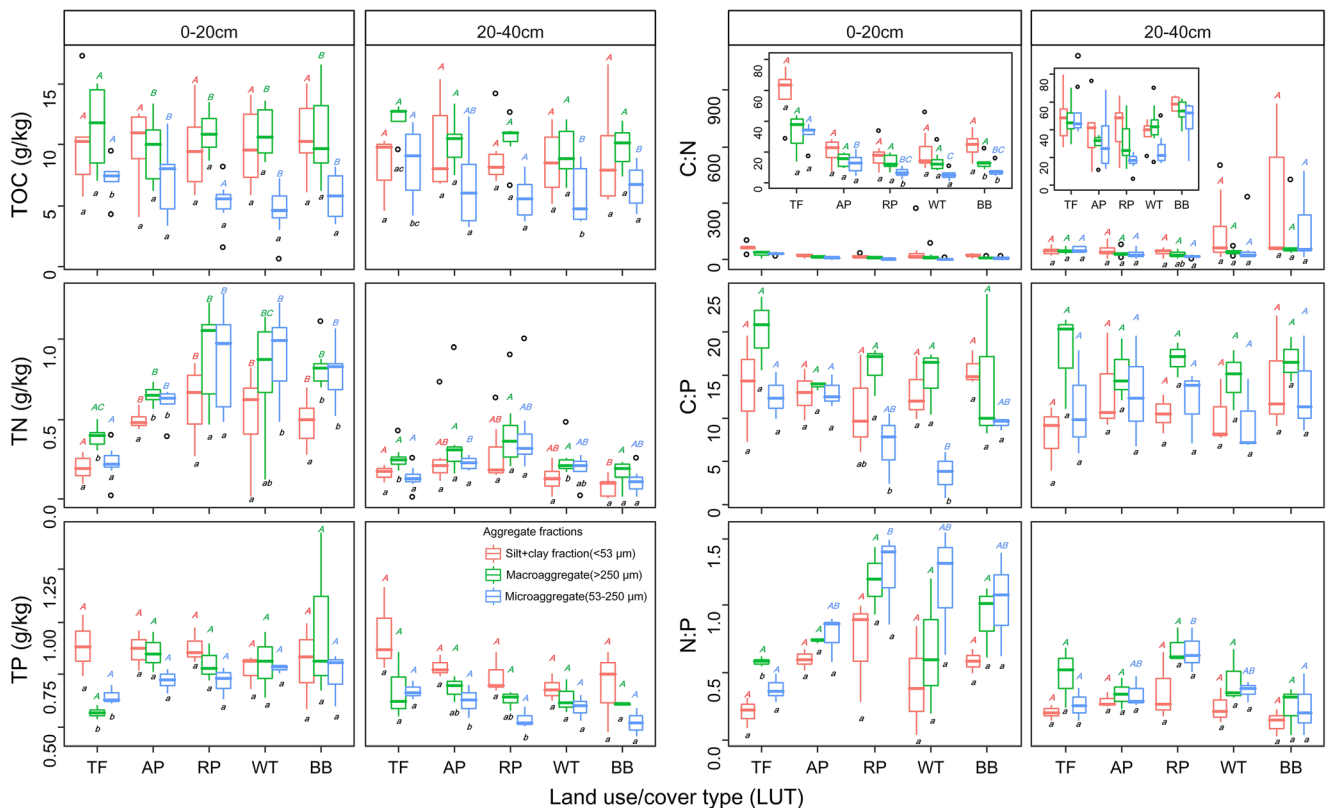


Fig. 5 Soil TOC, TN, TP, and C:N:P stoichiometry (C:N, TOC:TN; C:P, TOC:TP; N:P, TN:TP) in soil aggregates under different LUTs (tidal flat, TF; aquaculture ponds, AP; rape, RP; wheat, WT; and broad bean, BB). Different uppercase letters represent significant

differences among LUTs in each aggregate fraction at $p < 0.05$. Different lowercase letters represent significant differences among aggregate fractions under each LUT at $p < 0.05$

aggregates. However, there were many differences between bulk soil (Fig. 6a) and soil aggregates (Fig. 6b) in terms of the relationships among soil TOC, TN, TP, and C:N:P. There was a remarkably positive relationship between TOC and TN in bulk soil ($p < 0.001$), but there was no such significant relationship in soil aggregates ($p > 0.1$). TOC was found to be negatively related to C:N ($p < 0.05$) in bulk soil, while TOC and C:N displayed positive relationships in soil aggregates ($p < 0.001$). There were positive correlation between TOC, C:P, and N:P in bulk soil ($p < 0.001$), but their relationships were negative in soil aggregates ($p > 0.1$).

4 Discussion

4.1 Effects of coastal reclamation on soil properties in bulk soil

Soil physicochemical properties could be generally improved with reclamation (Li et al. 2013; Li et al. 2018a). In this study, we confirmed that soil TOC and TN content significantly increased and EC and pH decreased after reclamation (Fig. 2), which were consistent with other researches in coastal reclaimed areas, such as the Yangtze River Estuary (Sun et al. 2011), Hangzhou Bay (Iost et al. 2007), and Atlantic estuaries (Fernández et al. 2010). There were three main factors resulting in the accumulation of nutrients and the decalcification process. First, the seawalls blocked the influence from seawater, which has high salinity. Second, the surface elevation in the reclaimed area tended to increase with the increasing ages of reclamation, resulting in the decline of the groundwater level. Third, the input of fertilizer and manure and the decomposition of plant residues introduced a large amount of nutrients. Xie et al. (2017) showed that long-term reclamation had significant effects on soil enzyme activities in the reclaimed area of Rudong

County. In addition, bacterial communities in coastal saline soil changed radically after a 10-year reclamation (Fu et al. 2012). Therefore, coastal reclamation obviously changed the physical, chemical, and biological properties of soil. Furthermore, there was no significant difference in soil bulk density, TOC and EC over the 32-year reclamation (Fig. 2). Thus, soil properties tended to reach a steady state after being reclaimed for approximately 32 years, which is consistent with the researches in the Yangtze River Estuary (Sun et al. 2011) and Shangyu County of Zhejiang Province, China (Fu et al. 2014).

On the other hand, we found that even with 63 year of reclamation activity, the average content of TOC and TN was lower than that in China (Tian et al. 2010) and at the global scale (Cleveland and Liptzin 2007; Xu et al. 2013), while TP content was approximately the same. The different sources of soil C, N, and P determined the variations: the main source of C and N is the decomposition of plant litter, and their content is greatly influenced by vegetation, climate, and human activities; P primarily derives from rock weathering, and therefore its content depends on the soil parent material to a greater extent and experiences less influence from anthropogenic activity. It has been reported that tillage exposes physically protected organic material and enhances decomposition of soil organic matter (Mikha and Rice 2004). Considering the low content of TOC and TN in the sediments of the coastal wetland, although there was an input of nutrients, the intensive disturbance from human activity would accelerate the rate of nutrient decomposition.

Soil C:N:P ratios are good indicators of the condition of soil nutrients. High C:N ratio (> 25 on a mass basis) indicates that the accumulation of organic matter is faster than decomposition (Wei et al. 2009; Zhao et al. 2015). C:P < 200 suggests a net mineralization, C:P > 300 implies a net immobilization, and C:P between 200 and 300 reveals little change in the soluble P concentrations (Paul 2006). Our study revealed

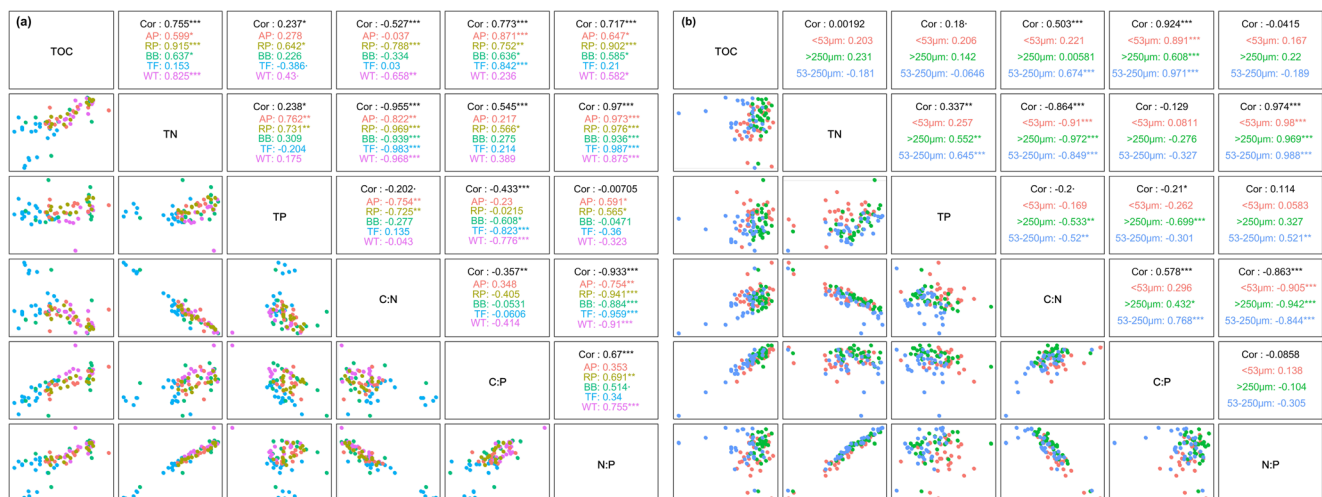


Fig. 6 Pearson correlation matrix among TOC, TN, TP, and C:N:P stoichiometry (C:N, TOC:TN; C:P, TOC:TP; N:P, TN:TP) in bulk soil (a) and soil aggregates (b). The statistical significance was denoted as for $p < 0.1$, * for $p < 0.05$, ** for $p < 0.01$, and *** for $p < 0.001$

that soil C:N decreased from 19.997 to 9.527 while soil C:P increased from 4.441 to 8.101 and N:P increased from 0.222 to 0.850 after the 63-year reclamation, which implied a net mineralization of nutrients. Soil C:N in this study was higher than the values in the Yellow River Delta (Qu et al. 2014), Laizhou Bay, Bohai Sea (Cao et al. 2015) and Chinese wetlands (Zhang et al. 2016b). C:P in the coastal wetland was similar to the results in Laizhou Bay, Bohai Sea (Cao et al. 2015), while both were far less than that in Chinese wetlands (Zhang et al. 2016b) and global wetlands (Xu et al. 2013). Moreover, C:P at the scale of study area was much lower than the average level in China (Tian et al. 2010) and worldwide (Cleveland and Liptzin 2007; Xu et al. 2013). The average level of N:P in China and globally was six to eleven times as high as the level in our study area. The results of high C:N and low C:P and N:P were also proven by Zhang et al. (2013). The comparative results above could mostly be attributed to the low content of soil C and N, especially the N content. N is often considered to be the most limiting element to net primary production in terrestrial ecosystems (Vitousek and Howarth 1991). This phenomenon also appeared in the Minjiang River Estuary (Wang et al. 2014b), the Yellow River Delta (Qu et al. 2014), and Laizhou Bay, Bohai Sea (Cao et al. 2015).

4.2 Effects of long-term reclamation on C, N, P, and C:N:P stoichiometry in soil aggregates

Soils in reclaimed area are cultivated and generally matured from the sediments in coastal wetland with long-term reclamation. Aggregate size distributions changed significantly after reclamation, and the proportion of macroaggregates markedly increased, consistent with the results of Elliott (1986) and Six et al. (2002). This finding suggested that macroaggregates contributed greatly to the variation in soil structure and that cultivation produced an immediate increase in the silt + clay fractions. Studies showed that cropping system (e.g., fertilization and straw application) promoted macroaggregate formation (Angers and Mehuys 1988; Green et al. 2005). This result may be attributed to the effect of a large number of plant residues, roots, and associated micro- and macro-organic activity (Li and Pang 2010).

Some researchers have shown that nutrient contents in soils are greater in macroaggregates than in microaggregates (Bhatnagar and Miller 1985; Kocyigit and Demirci 2012), whereas others have found greater nutrient concentrations in microaggregates than in macroaggregates (He et al. 1995; Maguire et al. 1998; Wan and El-Swaify 1998). In the present study, TOC and TN were mainly concentrated in macroaggregates, while TP was primarily concentrated in the silt + clay fraction. John et al. (2005) also indicated that increasing TOC concentrations were closely associated with the formation of macroaggregates. With respect to the difference between TOC, TN, and TP, He et al. (1995) demonstrated that the

method (dry-sieving or wet-sieving) used to obtain aggregate size classes impacted the results. Green et al. (2005) indicated that the differences in land use and management contributed to the discrepancies of nutrient distribution. However, in this study, TOC, TN, and TP were analyzed from one particular soil sample, so that we could exclude the above factors. Wan and El-Swaify (1998) demonstrated that unlike C, P movement from soil solution into the interior of aggregates is a diffusion process that can be chemically retarded due to the strong affinity of P to the oxisol studied. Therefore, the low TP concentration in large aggregates can be understood in a context of the combined effect of physical blockage and chemical retardation for P diffusion into aggregates.

C:N in macroaggregates and microaggregates was higher than found in Six et al. (2001), mainly because of the low N content in this study. The low TN content in bulk soil resulted in the similar condition in soil aggregates. Therefore, we confirmed that N was the most limiting factor in terms of soil C:N:P stoichiometry. The synergic relationship between TOC and TN has been proven in the bulk soil of this study and many others (Tian et al. 2010; Yang and Luo 2011; Zhang et al. 2012), but this relationship was not observed in soil aggregates (Fig. 6). This phenomenon should be further studied with many more soil samples.

4.3 Effects of land use on C, N, P, and C:N:P stoichiometry after long-term reclamation

Land use/cover changes during reclamation affect natural vegetation, crops and human structures that cover the land surface and alter soil physical, chemical, and biological properties as well as environmental conditions, thus affecting soil ecological processes. In the present study, TOC, TN, and TP were significantly higher in AP, RP, WT, and BB than in TF, and TN in BB was the highest because of N-fixing bacterial species associated with broad bean roots. N-fixing species have been widely proven to increase the soil TOC and TN concentrations, N-cycling and availability, and primary productivity (Uselman et al. 2000; Rice et al. 2004; Wang et al. 2010). However, our study revealed that there was no significant difference between AP, RP, WT, and BB, implying the weak effect of land use on TOC, TN, and TP. For soil C:N:P stoichiometry, Li et al. (2012) revealed that different LUTs had diverse soil C:N:P ratios due to the differences in elevation, vegetation type, and land management practices, and Zhang et al. (2012) also reported that soil C:N:P stoichiometry differed between two wetlands covered with different vegetation. Our analysis showed that C:N in soil covered with broad bean was lower, which could be attributed to biological nitrogen fixation. On the other hand, soil C:N:P ratios under different LUTs had no statistically significant differences. The distribution of soil aggregates also had no statistically significant differences among LUTs when reclaimed for 63 years. The TOC,

TN, TP, and C:N:P stoichiometry in soil aggregates basically followed similar patterns. Compared with the sediment of a natural wetland, soil from the 63-year-old reclaimed area proceeded through a long-term maturation process, and its properties reached a stable status, which exceeded the influences of vegetation and land use management. It has been demonstrated that soil development (e.g., substrate age and weathering intensity) can change nutrient availability and C:N:P stoichiometry in soils (Tian et al. 2010; Bing et al. 2016). Mayes et al. (2014) also suggested that soil type mediates the effects of land use on soil carbon and nitrogen. Sun et al. (2011) indicated that in the reclamation area of the Yangtze River Estuary, soil physicochemical properties were mainly influenced by reclamation time and then land use types. Many authors reported that the temporal trends in land use intensity, soil properties, and the biodiversity during reclamation would reach a relatively stable state after being reclaimed for 30 to 35 years (Shen et al. 2006; Sun et al. 2012; Li et al. 2013). Thus, except for land use/cover change, soil development plays an important role in soil nutrient stoichiometry.

5 Conclusions

This study comprises the comprehensive analysis of the impact of reclamation on soil C, N, P, and C:N:P stoichiometry in bulk soil and soil aggregates in a coastal wetland of Jiangsu Province, eastern China. Long-term chronosequence reclamation obviously changed the physicochemical properties of the coastal saline soil, and soil properties reached a steady state after being reclaimed for approximately 32 years. The main aggregate was microaggregates in natural wetland and was transformed to macroaggregates after 63 years of reclamation. Soil TOC and TN were mainly reserved in macroaggregates; thus, increasing the proportion of macroaggregates could contribute to increases in TOC and TN content. Compared with the sediments in tidal flats, the effect of land use/cover on TOC, TN, TP, and C:N:P stoichiometry in both bulk soil and soil aggregates after 63 years of reclamation was not significant. The results of this study may improve the understanding of nutrient cycling in coastal wetlands with intensive human activity. Our data also provide supplementary information for C:N:P stoichiometry in global terrestrial ecosystems.

Acknowledgements We thank the anonymous reviewers and the editors of the journal who provided constructive comments and suggestions on the manuscript.

Funding information This work was supported by the National Natural Science Foundation of China (41230751) and the program B for Outstanding PhD candidate of Nanjing University (201701B020).

References

- Angers DA, Mehuys GR (1988) Effects of cropping on macro-aggregation of a marine clay soil. *Can J Soil Sci* 68:723–732
- Beauchamp EG, Seech AG (1990) Denitrification with different sizes of soil aggregates obtained from dry-sieving and from sieving with water. *Biol Fert Soils* 10:188–193
- Bhatnagar VK, Miller MH (1985) Sorption of carbon and phosphorus from liquid poultry manure by soil aggregates of differing size. *Can J Soil Sci* 65:467–473
- Bing HJ, Wu YH, Zhou J, Sun HY, Luo J, Wang JP, Yu D (2016) Stoichiometric variation of carbon, nitrogen, and phosphorus in soils and its implication for nutrient limitation in alpine ecosystem of eastern Tibetan Plateau. *J Soils Sediments* 16:405–416
- Cao L, Song JM, Li XG, Yuan HM, Li N, Duan LQ, Wang QD (2015) Geochemical characteristics of soil C, N, P, and their stoichiometrical significance in the coastal wetlands of Laizhou Bay, Bohai Sea. *Clean-Soil Air Water* 43:260–270
- Cleveland CC, Liptzin D (2007) C: N: P stoichiometry in soil: is there a “Redfield ratio” for the microbial biomass? *Biogeochemistry* 85: 235–252
- Elliott ET (1986) Aggregate structure and carbon, nitrogen, and phosphorus in native and cultivated soils. *Soil Sci Soc Am J* 50:627–633
- Elser JJ, Sterner RW, Gorokhova E, Fagan WF, Markow TA, Cotner JB, Harrison JF, Hobbie SE, Odell GM, Weider LW (2000) Biological stoichiometry from genes to ecosystems. *Ecol Lett* 3:540–550
- Fernández S, Santín C, Marquín J, Álvarez MA (2010) Saltmarsh soil evolution after land reclamation in Atlantic estuaries (Bay of Biscay, north coast of Spain). *Geomorphology* 114:497–507
- Fu Q, Ding N, Liu C, Lin Y, Guo B (2014) Soil development under different cropping systems in a reclaimed coastal soil chronosequence. *Geoderma* 230–231:50–57
- Fu Q, Liu C, Ding N, Lin Y, Guo B, Luo J, Wang H (2012) Soil microbial communities and enzyme activities in a reclaimed coastal soil chronosequence under rice-barley cropping. *J Soils Sediments* 12: 1134–1144
- Green VS, Cavigelli MA, Dao TH, Flanagan DC (2005) Soil physical properties and aggregate-associated C, N, and P distributions in organic and conventional cropping systems. *Soil Sci* 170:822–831
- Guo LB, Gifford RM (2002) Soil carbon stocks and land use change: a meta analysis. *Glob Chang Biol* 8:345–360
- He ZL, Wilson MJ, Campbell CO, Edwards AC, Chapman SJ (1995) Distribution of phosphorus in soil aggregate fractions and its significance with regard to phosphorus transport in agricultural runoff. *Water Air Soil Poll* 83:69–84
- Heanes DL (1984) Determination of total organic-C in soils by an improved chromic acid digestion and spectrophotometric procedure. *Commun Soil Sci Plan* 15:1191–1213
- Iost S, Landgraf D, Makeschin F (2007) Chemical soil properties of reclaimed marsh soil from Zhejiang Province P.R. China. *Geoderma* 142:245–250
- Jiang M, Wang X, Liusui Y, Han C, Zhao C, Liu H (2017) Variation of soil aggregation and intra-aggregate carbon by long-term fertilization with aggregate formation in a grey desert soil. *Catena* 149(Part 1):437–445
- Jiangsu Coastal Areas Development Office (2009) Jiangsu Coastal Development Planning. http://yhb.jiangsu.gov.cn/art/2013/5/21/art_12318_2879348.html. Accessed July 28 2018 (in Chinese)
- Jiménez JJ, Lal R, Russo RO, Leblanc HA (2008) The soil organic carbon in particle-size separates under different regrowth forest stands of north eastern Costa Rica. *Ecol Eng* 34:300–310
- John B, Yamashita T, Ludwig B, Flessa H (2005) Storage of organic carbon in aggregate and density fractions of silty soils under different types of land use. *Geoderma* 128:63–79

- Kocyyigit R, Demirci S (2012) Long-term changes of aggregate-associated and labile soil organic carbon and nitrogen after conversion from forest to grassland and cropland in northern Turkey. *Land Degrad Dev* 23:475–482
- Larney FJ (1993) Dry-aggregate size distribution. In: Carter MR, Gregorich EG (eds) *Soil sampling and methods of analysis*, Second Edition edn. CRC Press, Taylor & Francis Group, USA, pp 821–831
- Li G-L, Pang X-M (2010) Effect of land-use conversion on C and N distribution in aggregate fractions of soils in the southern Loess Plateau, China. *Land Use Policy* 27:706–712
- Li J et al (2014) Evolution of soil properties following reclamation in coastal areas: a review. *Geoderma* 226–227:130–139
- Li J, Yang W, Li Q, Pu L, Xu Y, Zhang Z, Liu L (2018a) Effect of reclamation on soil organic carbon pools in coastal areas of eastern China. *Front Earth Sci* 12:339–348
- Li J, Yang W, Wang Y, Li Q, Liu L, Zhang Z (2018b) Carbon footprint and driving forces of saline agriculture in coastally reclaimed areas of eastern China: a survey of four staple crops. *Sustainability-Basel* 10:928
- Li XZ, Sun YG, Mander Ü, He YL (2013) Effects of land use intensity on soil nutrient distribution after reclamation in an estuary landscape. *Landsc Ecol* 28:699–707
- Li Y, Wu J., Liu S., Shen J., Huang D., Su Y., Wei W., Syers J.K. (2012) Is the C:N:P stoichiometry in soil and soil microbial biomass related to the landscape and land use in southern subtropical China? *Global Biogeochem Cy* 26 <https://doi.org/10.1029/2012gb004399>
- Li Y, Zhu X, Sun X, Wang F (2010) Landscape effects of environmental impact on bay-area wetlands under rapid urban expansion and development policy: a case study of Lianyungang, China. *Landscape Urban Plan* 94:218–227
- Ma ZJ, Melville DS, Liu J, Chen Y, Yang H, Ren W, Zhang Z, Piersma T, Li B (2014) Rethinking China's new great wall. *Science* 346:912–914
- Maguire RO, Edward AC, Wilson MJ (1998) Influence of cultivation on the distribution of phosphorus in three soils from NE Scotland and their aggregate size fractions. *Soil Use Manage* 14:147–153
- Mayes M, Marin-Spiotta E, Szymanski L, Akif Erdoğlan M, Ozdoğan M, Clayton M (2014) Soil type mediates effects of land use on soil carbon and nitrogen in the Konya Basin, Turkey. *Geoderma* 232–234:517–527
- Mikha MM, Rice CW (2004) Tillage and manure effects on soil and aggregate-associated carbon and nitrogen contribution. *Soil Sci Soc Am J* 68:809–816
- O'Halloran IP, Cade-Menun BJ (1993) Total and organic phosphorus. In: Carter MR, Gregorich EG (eds) *Soil sampling and methods of analysis*. Second Edition edn. CRC Press, Taylor & Francis Group, United States of America, pp 265–273
- Oades JM (1984) Soil organic matter and structural stability: mechanisms and implications for management. In: Tinsley J, Darbyshire JF (eds) *Biological processes and soil fertility*. Springer Netherlands, Dordrecht, pp 319–337
- Paul EA (2006) *Soil microbiology, ecology, and biochemistry*. Academic press, London
- Qu F et al (2014) Influences of anthropogenic cultivation on C, N and P stoichiometry of reed-dominated coastal wetlands in the Yellow River Delta. *Geoderma* 235–236:227–232
- Redfield AC (1958) The biological control of chemical factors in the environment. *Am Sci* 46:221–230A
- Rice SK, Westerman B, Federici R (2004) Impacts of the exotic, nitrogen-fixing black locust (*Robinia pseudoacacia*) on nitrogen-cycling in a pine-oak ecosystem. *Plant Ecol* 174:97–107
- Schutter ME, Dick RP (2002) Microbial community profiles and activities among aggregates of winter fallow and cover-cropped soil. *Soil Sci Soc Am J* 66:142–153
- Shen J, Hu R, Li M, Ding P, Yu M, Ding B (2006) Influence of reclamation on plant diversity of beach wetlands in Hangzhou Bay and Yueqing Bay in east China. *Journal of Zhejiang University Sciences Edition* 33:324–328,332 (in Chinese with English abstract)
- Six J, Callewaert P, Lenders S, de Gryze S, Morris SJ, Gregorich EG, Paul EA, Paustian K (2002) Measuring and understanding carbon storage in afforested soils by physical fractionation. *Soil Sci Soc Am J* 66:1981–1987
- Six J, Elliott ET, Paustian K (2000) Soil macroaggregate turnover and microaggregate formation: a mechanism for C sequestration under no-tillage agriculture. *Soil Biol Biochem* 32:2099–2103
- Six J, Guggenberger G, Paustian K, Haumaier L, Elliott ET, Zech W (2001) Sources and composition of soil organic matter fractions between and within soil aggregates. *Eur J Soil Sci* 52:607–618
- Six J, Paustian K (2014) Aggregate-associated soil organic matter as an ecosystem property and a measurement tool. *Soil Biol Biochem* 68:A4–A9
- Sterner RW, Elser JJ (2002) *Ecological stoichiometry: the biology of elements from molecules to the biosphere*. Princeton University Press
- Sun Y, Li X, He Y, Jia Y, Ma Z, Guo W, Xin Z (2012) Impact factors on distribution and characteristics of natural plant community in reclamation zones of Changjiang River estuary. *Chin Geograph Sci* 22:154–166
- Sun YG, Li X, Mander Ü, He Y, Jia Y, Ma Z, Guo W, Xin Z (2011) Effect of reclamation time and land use on soil properties in Changjiang River estuary, China. *Chin Geograph Sci* 21:403–416
- Tian B, Wu WT, Yang ZQ, Zhou YX (2016) Drivers, trends, and potential impacts of long-term coastal reclamation in China from 1985 to 2010. *Estuar Coast Shelf S* 170:83–90
- Tian H, Chen G, Zhang C, Melillo JM, Hall CAS (2010) Pattern and variation of C:N:P ratios in China's soils: a synthesis of observational data. *Biogeochemistry* 98:139–151
- Tisdall JM, Oades JM (1982) Organic matter and water-stable aggregates in soils. *J Soil Sci* 33:141–163
- Uselman SM, Qualls RG, Thomas RB (2000) Effects of increased atmospheric CO₂, temperature, and soil N availability on root exudation of dissolved organic carbon by a N-fixing tree (*Robinia pseudoacacia* L.). *Plant Soil* 222:191–202
- Vitousek PM, Howarth RW (1991) Nitrogen limitation on land and in the sea: how can it occur? *Biogeochemistry* 13:87–115
- Wan Y, El-Swaify SA (1998) Sediment enrichment mechanisms of organic carbon and phosphorus in a well-aggregated oxisol. *J Environ Qual* 27:132–138
- Wang F, Li Z, Xia H, Zou B, Li N, Liu J, Zhu W (2010) Effects of nitrogen-fixing and non-nitrogen-fixing tree species on soil properties and nitrogen transformation during forest restoration in southern China. *Soil Sci Plant Nutr* 56:297–306
- Wang W, Liu H, Li YQ, Su J (2014a) Development and management of land reclamation in China. *Ocean Coast Manage* 102(Part B): 415–425
- Wang W, Sardans J, Zeng C, Zhong C, Li Y, Peñuelas J (2014b) Responses of soil nutrient concentrations and stoichiometry to different human land uses in a subtropical tidal wetland. *Geoderma* 232–234:459–470
- Wang X, Yost RS, Linquist BA (2001) Soil aggregate size affects phosphorus desorption from highly weathered soils and plant growth. *Soil Sci Soc Am J* 65:139–146
- Wei X, Shao M, Fu X, Horton R, Li Y, Zhang X (2009) Distribution of soil organic C, N and P in three adjacent land use patterns in the northern Loess Plateau, China. *Biogeochemistry* 96:149–162
- Xie X, Pu L, Wang Q, Zhu M, Xu Y, Zhang M (2017) Response of soil physicochemical properties and enzyme activities to long-term reclamation of coastal saline soil, eastern China. *Sci Total Environ* 607–608:1419–1427

- Xu CY, Pu LJ, Zhu M, Li JG, Chen XJ, Wang XH, Xie XF (2016) Ecological security and ecosystem services in response to land use change in the coastal area of Jiangsu, China. *Sustainability-Basel* 8:816
- Xu XF, Thornton PE, Post WM (2013) A global analysis of soil microbial biomass carbon, nitrogen and phosphorus in terrestrial ecosystems. *Glob Ecol Biogeogr* 22:737–749
- Xu Y, Pu LJ, Zhu M, Li JG, Zhang M, Li P, Zhang J (2014) Spatial variation of soil salinity in the coastal reclamation area, eastern China. *J Coastal Res* 30:411–417
- Yang Y, Luo Y (2011) Carbon: nitrogen stoichiometry in forest ecosystems during stand development. *Glob Ecol Biogeogr* 20:354–361
- Zhang RS, Pu LJ, Li JG, Zhang J, Xu Y (2016a) Landscape ecological security response to land use change in the tidal flat reclamation zone, China. *Environ Monit Assess* 188:1
- Zhang Z, Lu X, Xue Z, Liu X (2016b) Is there a Redfield-type C:N:P ratio in Chinese wetland soils? *Acta Pedol Sin* 53:1160–1169 (in Chinese with English abstract)
- Zhang ZS, Lu XG, Song XL, Guo Y, Xue ZS (2012) Soil C, N and P stoichiometry of *Deyeuxia angustifolia* and *Carex lasiocarpa* wetlands in Sanjiang Plain, Northeast China. *J Soils Sediments* 12:1309–1315
- Zhang ZS, Song XL, Lu XG, Xue ZS (2013) Ecological stoichiometry of carbon, nitrogen, and phosphorus in estuarine wetland soils: influences of vegetation coverage, plant communities, geomorphology, and seawalls. *J Soils Sediments* 13:1043–1051
- Zhao FZ et al (2015) Land use change influences soil C, N, and P stoichiometry under 'Grain-to-Green Program' in China. *Sci Rep-Uk* 5:10195