



# Glomalin-related soil protein deposition and carbon sequestration in the Old Yellow River delta

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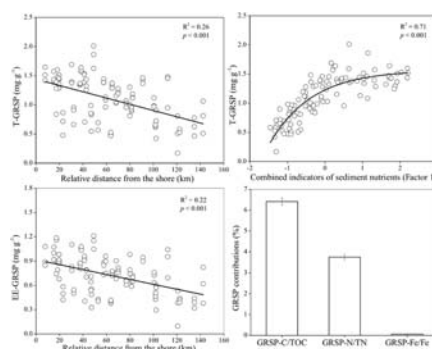
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## HIGHLIGHTS

- The coastal marine system in the Old Yellow River delta is an important sink of GRSP.
- The mean concentration of total GRSP was  $1.10 \pm 0.04 \text{ mg g}^{-1}$  ( $0.24 \text{ Mg C ha}^{-1}$ ), accounting for  $6.41 \pm 0.17\%$  of TOC and  $3.75 \pm 0.13\%$  of TN in the 0–10 cm marine sediments.
- GRSP was a new natural Fe fertilization, accounting for  $0.058 \pm 0.003\%$  of total Fe in marine sediments.
- GRSP enhanced marine C sequestration by its rapid deposition and Fe contribution.
- Co-deposited with nutrient elements, GRSP fractions were accumulated more in more fertile sediments but less in less fertile sediments.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Glomalin-related soil protein (GRSP), a particular terrigenous-derived carbon (C), is transported to the coastal oceans, where it accumulates in sediments. We hypothesized that terrigenous C (GRSP) accumulation could enhance marine C sequestration, and sediment fertility would increase the C stock in the marine ecosystem. In this study, we tested GRSP contribution to marine sediment C, nitrogen (N) and iron (Fe), and explored whether GRSP deposition varied with sediment fertility levels in the Old Yellow River delta. The mean concentration of total GRSP was  $1.10 \pm 0.04 \text{ mg g}^{-1}$  ( $0.24 \text{ Mg C ha}^{-1}$ ), accounting for  $6.41 \pm 0.17\%$  of total organic C and  $3.75 \pm 0.13\%$  of total N in the 0–10 cm marine sediments, indicating that the coastal marine system is an important sink of GRSP. GRSP also contained  $1.46 \pm 0.06\%$  Fe ( $20.7 \text{ kg Fe ha}^{-1}$ ), accounting for  $0.058 \pm 0.003\%$  of total Fe in marine sediments. Meanwhile, Fe-content in GRSP significantly decreased with distance from the shore, indicating that Fe was released with GRSP transfer and thus GRSP was a new natural Fe fertilization in marine environment. Furthermore, GRSP enhanced marine C sequestration by its rapid deposition and Fe contribution. Combined indicators of sediment fertility (factor 1) were significantly positively correlated with GRSP concentrations by Principal Component Analysis. Co-deposited with nutrient elements, GRSP fractions were accumulated more in more fertile sediments but less in less fertile sediments. GRSP, a mixture of co-existent multiple

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elements, can be used as a nutrient controlled-release agent in the marine ecosystem. GRSP fractions were responsive to marine sediment fertility levels and the understanding of their function in sediment C sequestration will provide new insights into the importance of terrestrial-marine linkages.

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## 1. Introduction

Although land-ocean linkage is quite important for marine ecosystem service, little is known about the transfer of terrigenous organic matter and its contributions in marine ecosystems. River deltas serve as sources, sinks and transporters of terrigenous materials on their way to the ocean, including sediment, minerals, carbon (C), nitrogen (N), and pollutants, thus affecting the biodiversity and productivity of coastal marine ecosystems (Wooldridge et al., 2006; Vörösmarty et al., 2009). Since 1855, the Old Yellow River delta has suffered from coastal erosion (Zhou et al., 2014), and the eroded materials are redeposited in the near-shore area, the Yangtze River delta, the Yellow Sea and south of Cheju Island in the East China Sea (Yang and Youn, 2007; Liu et al., 2010). Meanwhile, rivers have provided a large amount of terrigenous sediment into marine environment (Zhou et al., 2014). These terrigenous organic matters are stored and mixed with marine organic matter (Hedges et al., 1997), and thus approximately 90% of global organic C is buried in the coastal marine sediments, and the near-shore areas can affect global C cycle (Hedges and Keil, 1995).

Given the potential impacts of anthropogenic drivers and global climate change, coastal marine environments become a major focus of concern (Harley et al., 2006). Providing long-term storage of organic carbon, carbon accumulation in marine sediments has been referred to as “blue carbon” to distinguish it from carbon in terrestrial sinks (Greiner et al., 2013). In terrestrial ecosystems, plants absorb carbon dioxide (CO<sub>2</sub>) from the air, and C is converted through photosynthesis and stored in living biomass and soils (Mcleod et al., 2011). Arbuscular mycorrhizal (AM) fungi can generally mediate above- and below-ground nutrients by absorbing nutrients (e.g., phosphorus and nitrogen) which are essential for plants in exchange for photosynthate (Smith and Read, 2008). AM fungi can form symbiotic relationships with over 80% of plant families (Smith and Read, 2008) and utilize up to 45% of photosynthetically fixed C (Grayston et al., 1997). Subsequently, a substantial amount of carbon in the mycorrhizal tissues can be stored in soils (Treseder and Allen, 2000; Rillig et al., 2001; Lovelock et al., 2004a). Furthermore, AM fungi can contribute to nutrient storage and protect soil C from degradation by forming mycelial networks and improving soil structure (Wright and Upadhyaya, 1998; Rillig and Mummey, 2006). The above roles of AM fungi are in fact related to the production of a novel glycoprotein, glomalin (Wright and Upadhyaya, 1998; Rillig, 2004).

Glomalin or glomalin-related soil protein (GRSP), a recalcitrant compound that is stocked in soil with the senescence of hyphae and spores (Rillig, 2004; Treseder et al., 2007). Meanwhile, GRSP can be accumulated in soil, with its concentrations ranging from 2 to 15 mg g<sup>-1</sup> in temperate and tropical forest ecosystems (Treseder and Turner, 2007; Singh et al., 2013), and up to 60 mg cm<sup>-3</sup> in Hawaiian soils (Rillig et al., 2001). Furthermore, GRSP provides nutrients to soil through the molecule structure, which is composed of 36 to 59% C and 3 to 5% N, representing as much as 3–5% of soil C and N (Rillig et al., 2001; Lovelock et al., 2004a). GRSP also contains 1 to 9% iron (Fe) (Wright and Upadhyaya, 1998; Rillig et al., 2001). As a limiting micronutrient, Fe could enhance growth of phytoplankton and play a pivotal role in controlling carbon uptake in marine ecosystems (Coale et al., 2004; Blain et al., 2007). GRSP is present in the floodplain soils, rivers, ocean and coastal ecosystems (Harner et al., 2004; Adame et al., 2010, 2012; López-Merino et al., 2015), indicating that a significant quantity of GRSP can be transferred with water or deposited in sediment as an important component of organic carbon (Singh et al., 2017; Wang et al.,

2018). Those researches only proved that GRSP is transported from terrestrial sources to the ocean and investigated distribution of GRSP in the coastal zone. However, the deposition mechanism of GRSP and its contribution to the sediment carbon, nitrogen and iron, as well as the carbon sequestration are poorly understood in marine ecosystems, which are the focus of our study. In this study, we detected the pool sizes of the GRSP fractions, and made a first assessment of the relative contributions of Fe in the GRSP to the total Fe in the marine sediments. In addition, we measured the C and N contents in GRSP to calculate their contributions to marine sediment C and N pools and analyze GRSP contribution to C sequestration.

Total GRSP in soils is currently partitioned into easily extractable GRSP (EE-GRSP) and difficultly extractable GRSP (DE-GRSP) (Koide and Peoples, 2013; Wu et al., 2014). EE-GRSP is considered as the newly produced and relatively more labile fraction, whilst DE-GRSP represents the older fraction that is relatively more stable (Koide and Peoples, 2013; Wu et al., 2014). In addition, because of the sticky nature of GRSP, it binds together organic matter and many elements (K, Ca, Mg, Fe, Mn and Al) to form a more complex compound in terrestrial ecosystems (Zhang J. et al., 2017; Zhang Z. et al., 2017). Meanwhile, previous studies have shown that soil nutrients significantly affected GRSP accumulation and deposition in forest ecosystems (Treseder et al., 2007; Zhang et al., 2015; Lovelock et al., 2004a, 2004b). However, the knowledge about their different responses of these GRSP fractions to marine sediment nutrients is limited. In this study we characterized GRSP distribution and deposition in the marine sediments, and analyzed GRSP correlation with sediment fertility levels in 108 sediment samples within the Old Yellow River delta, China.

Here we tested the following hypotheses: (i) GRSP would be present in marine sediment and decrease with distance from the shore. With coastal erosion, terrigenous sediments are transported through rivers, tides and groundwater, so we expect that GRSP is differentially distributed in the surface sediments and accumulated with sediment deposition. (ii) GRSP could be a substantial contributor to marine sediment C and N pools, and enhance C sequestration in the marine sediments, as GRSP is a recalcitrant glycoprotein that contains Fe and is accumulated in soils (Rillig et al., 2001; Lovelock et al., 2004a). (iii) GRSP deposition is correlated with sediment fertility and provides nutrients for organisms in the marine ecosystem, because GRSP is glue-like in nature and can float to the surface (Harner et al., 2004). Our results provided the first evidence of the terrigenous C (GRSP) contributions and suggested deposition mechanisms of GRSP in marine environment.

## 2. Materials and methods

### 2.1. Site description and sampling

The Yellow River, with a total length of approximately 5464 km, flows into the Bohai Sea, China, and is the fifth longest river in the world. The Old Yellow River delta is the ninth superlobe of the Yellow River delta (Xue, 1993; Saito et al., 2000). During 1128–1855, the Yellow River flowed into the South Yellow Sea, and contributed to the formation of the subaqueous old Yellow River delta (Chen et al., 2012). In 1855, the river changed its channel and again flowed into the Bohai Sea (Xue, 1993).

In this study, 108 surface (0–10 cm) sediment samples in 37 sample sites were collected in the summer of 2015 in the Old Yellow River delta (Fig. 1). We collected three replicate samples at each site. However, due to excessive wind and strong turbulence which made it extremely

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