



The role of organo-clay associations in limiting organic matter decay: Insights from the Dajiuhu peat soil, central China

Shuling Chen^a, Hanlie Hong^{a,*}, Xianyu Huang^{a,b}, Qian Fang^a, Ke Yin^a, Chaowen Wang^c, Yiming Zhang^a, Liuling Cheng^a, Thomas J. Algeo^{b,d,e,*}

^a School of Earth Sciences, China University of Geosciences, Wuhan 430074, China

^b State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, China

^c Gemological Institute, China University of Geosciences, Wuhan 430074, China

^d State Key Laboratory of Geological Processes and Mineral Resources, China University of Geosciences, Wuhan 430074, China

^e Department of Geology, University of Cincinnati, Cincinnati, OH 45221-0013, USA

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ABSTRACT

The mineral-bound organic matter (OM) in soils is of great importance for the global carbon cycle. Long-standing debate exists regarding the mechanisms of soil organic matter (SOM) preservation and, especially, the role of organo-clay mineral associations. Investigation of such associations is rarely undertaken in peat soils. In this study, we examine organo-clay relationships in the Dajiuhu peat soil in Hubei Province, central China, based on molecular markers, which allows for understanding of rates of OM degradation and the preservation of OM associated with clay minerals. We analyzed the major and trace element chemistry, clay mineralogy, and lipid compositions of both the coarse- and clay-sized fractions of the soil. The uniformity of trace element and REE distribution patterns in the soil (except for Layer 2) suggests that the source materials were well mixed. Chemical index of alteration (CIA) values of 70–85 indicate moderate to strong weathering processes. A significant correlation between $C_{org}:P$ and TOC may indicate that redox conditions were a significant control on OM preservation. The clay fraction of Dajiuhu peatland soils also has played an important role in preserving OM, especially in anoxic environments. Fourier transform infrared spectrophotometry (FTIR) and X-ray diffraction (XRD) analysis show that OM is absorbed mainly on the edges of illite grains rather than in interlayer sites. The amphoteric margins of illite crystals allow them to more effectively bind organic compounds. The high $R_{1/h}$ ratio, high carbon preference index (CPI), and high ratio of unsaturated-to-saturated fatty acids ($C_{18:2}/C_{18}$) in the clay-sized fraction provide further biomarker evidence that illite efficiently protects OM from biodegradation, thus enhancing the OM content of soils.

1. Introduction

Soil organic matter (SOM) contains approximately three times more carbon than the atmosphere, representing one of the largest and most active reservoirs of organic carbon on the global scale (von Lützow et al., 2006; Rumpel and Kögel-Knabner, 2011; Feng et al., 2014; Barré et al., 2017; Steffens et al., 2011). Peat soil, a unique type of soil, contains about one-third of the organic carbon in the pedosphere, even though peatland covers only 2–3% of total land area (Gorham, 1991). More than 90% of a soil's carbon inventory exists in close association with Fe-(hydr)oxide and/or minerals, especially clay minerals, which can protect organic matter (OM) from degradation in redox-active environments (Kaiser and Guggenberger, 2003; Mayer, 1994; Mikutta et al., 2009; Schrumpf et al., 2013; Zeng et al., 2016). Such associations

can significantly reduce the bioavailability of OM and slow down its remineralization rate (Conant et al., 2011), decreasing the amount of CO_2 emitted from soils to the atmosphere. Organic carbon stabilized by minerals has longer turnover times than particulate organic matter or organic carbon in aggregates (Trumbore, 2000). Thus, OM associated with minerals controls the long-term sedimentary sequestration of carbon and plays an important role in the global carbon cycle (Kalbitz et al., 2000; Matus et al., 2014; von Lützow et al., 2006).

Mechanisms of organic carbon stabilization by minerals have been extensively investigated but still remain controversial. A number of studies have demonstrated the significance of OM-mineral interactions in a wide range of environments, such as sedimentary rocks (Li et al., 2016; Löhr et al., 2015; Playter et al., 2017), marine and coastal sediments (Bianchi et al., 2016; Loehr and Kennedy, 2014), and soils

* Corresponding authors at: State Key Laboratory of Biogeology and Environmental Geology, China University of Geosciences, Wuhan 430074, China.
E-mail addresses: honghl8311@aliyun.com (H. Hong), thomas.algeo@uc.edu (T.J. Algeo).

(Mayer, 1994; Schmidt et al., 2011; Singh et al., 2017b). Also, synthesis of organo-clay composites has been conducted in laboratory experiments in order to better understand mechanisms of OM-mineral associations (Pronk et al., 2016; Zeng et al., 2016).

A number of different mechanisms of OM preservation have been proposed (Keil and Mayer, 2013; Kögel-Knabner et al., 2008; Matus et al., 2014; Mikutta et al., 2006; Sollins et al., 1996; von Lützow et al., 2006), including: (1) selective preservation due to differential inherent biological recalcitrance of OM (plant litter, rhizodeposits, microbial products, humic polymers and charred OM) (Vonk et al., 2010); (2) sequestration of OM in micro- or nano-pore spaces that reduce access by microbes and enzymes (Churchman and Tate, 1986); (3) formation of organo-mineral associations or organo-metallic ligands that reduce the rate of OM decomposition (Chorover et al., 2004).

OM associations with minerals depend on the characteristics of both the minerals and the OM. The large surface areas of clay minerals and Fe-(hydr)oxides and the wide occurrence of these minerals in soils probably contribute greatly to preservation of OM. The surfaces of phyllosilicates exhibit a high affinity for carbohydrates and aliphatic natural OM (Chorover and Amistadi, 2001). In addition, clay minerals with expandable layers can sequester large amounts of OM (Bu et al., 2017; Loehr and Kennedy, 2014). Polar and non-polar organics, ions, and water can get access to the expandable layers of smectite, and smectite is commonly considered as the most effective clay species to stabilize OM (Kennedy et al., 2014; Loehr and Kennedy, 2014). Furthermore, the negative charge of clay mineral surfaces attracts positively-charged cations or OM (Mortland, 1970), and cations can act as a bridge between OM and clay minerals. Positively charged Fe-(hydr)oxides can bind to both clay minerals and OM, acting as bridges between negatively charged OM and clay minerals. Kaolinite was shown to strongly interact with Fe-(hydr)oxides, promoting the formation of aggregates preserving OM (Goldberg, 1987; Kitagawa, 1983). At a molecular scale, OM is stabilized on mineral surfaces via multiple organo-mineral bonding processes, including ligand exchange, polyvalent cation bridges, hydrophobic interactions, H-bonding, and complexation (Arnarson and Keil, 2001; Theng, 1976). Intrinsic characteristics of OM, for instance, the degree of cyclization and existence of polar functionalities, enhance the sorption rate by promoting strong bonding (Ahmat et al., 2016). OM with higher molecular weight, acidity, and aromaticity tends to be preferentially absorbed (Kaiser and Zech, 1997). The relative importance of different bonding mechanisms depends on the specifics of organic functional groups and environmental conditions such as temperature, pH, and cationic concentrations.

The near surface layer of earth, where rock, soil, water, air, and living organisms interact, is now recognized as the critical zone (Xu and Liu, 2017). Complex interactions among physical, chemical and

biological processes control its evolution and structural development (Chorover et al., 2007; Rasmussen et al., 2011). Addressing the complexity of these interactions is essential for understanding fundamental earth-surface processes such as terrestrial carbon cycling, mineral-organic interactions, and the geologic and atmospheric controls on long-term landscape evolution (National Research Council, 2001). Monitoring of critical zones is more advanced in developed countries, but a critical zone observatory (CZO) monitoring program has just been activated in China in 2014 based on decades of study of the Dajiuhu peatland.

The Dajiuhu peatland is one of the largest sub-alpine peatlands in the subtropical region of China (Zhao et al., 1999), and it has been investigated extensively with regard to how the carbon cycle responds to climate changes (Huang et al., 2017). Earlier studies of the Dajiuhu peatland focused on environmental reconstructions (Huang et al., 2013a, 2013b; Xie et al., 2015), however, the relationship between clay minerals and OM in the Dajiuhu peatland has received inadequate attention to date. We propose that OM in the Dajiuhu peat soil is effectively protected from decay through interactions with clay minerals. In order to test this hypothesis, we combined weathering proxies, geochemical redox proxies, and biomarkers (1) to determine the distribution of OM relative to clay minerals; (2) to investigate the factors influencing total organic carbon content; and (3) to elucidate the possible role of clay minerals in the preservation of OM.

2. Site description

The Dajiuhu peatland (31°28'50" N, 110°00'09" E) is located in the Shennongjia Forest region, Hubei Province, central China (Fig. 1). It is covered with thick peat and is a unique subalpine peatland in a subtropical region, dominated by the East Asian summer monsoon. The average annual temperature is 7.2 °C, and the mean annual rainfall is 1560 mm (Huang et al., 2013a, 2013b, 2013c).

CZO monitoring points have been established at Yangluchang, Niangniangfen, Erhaoba, Dashuba, Lake 4, and Lake 5 in the Dajiuhu peatland (Huang et al., 2017). A 2.5-m-long core was retrieved at the No. 4 monitoring point (31°29'27" N, 109°59'44" E) at Yangluchang in June 2016. This core can be subdivided into seven layers based on color and compositional characteristics (Fig. 2, Table 1). The surface soil, Layer 1 (0–15 cm), consists of dark brown (5YR 1/2) peat with abundant plant detritus. The following layers can be divided into two groups, one being peat layers (Layers 2, 4 and 6) and the other being clay layers (Layers 3, 5 and 7). We have labeled samples using “L” for “layer”, e.g., sample “L2-1” represents the first sample (from the top downwards) in Layer 2.

The peat layers are thick and have a reddish- or yellowish-brown

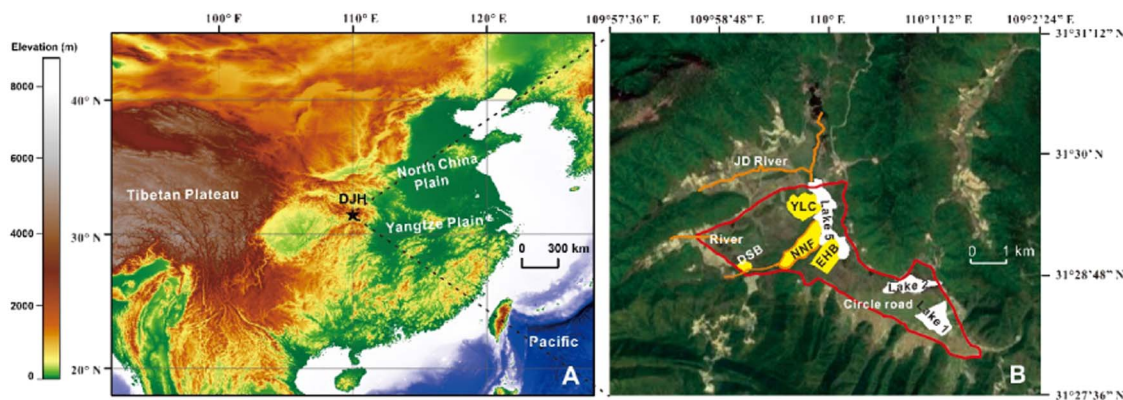


Fig. 1. (A) Location of Dajiuhu peatland, central China. The study area is a unique sub-alpine peatland in a subtropical region, dominated by the East Asian summer monsoon. The base map was downloaded from <http://topex.ucsd.edu/pub/archive/srtm/>. (B) Wetlands, rivers and lakes in the Dajiuhu peatland. The yellow patches and lakes are critical-zone observation points. YLC: Yangluchang; EHB: Erhaoba; NNF: Niangniangfen, DSB: Dashuba. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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