



## Microorganism-induced weathering of clay minerals in a hydromorphic soil

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### Abstract

In order to improve the understanding of factors influencing weathering in hydromorphic soils, the clay mineral and chemical compositions, iron (hydr)oxides, organic compounds, and Sr and Nd isotopic compositions, of hydromorphic soils on the banks of the Liangzi Lake, Hubei province, south China, were investigated. The B horizon in the lower profile exhibits a distinct net-like pattern, with abundant short white veins within the red-brown matrix. Their various  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{143}\text{Nd}/^{144}\text{Nd}$  isotopic compositions showed only small variations of 0.7270–0.7235 and 0.51200–0.51204, respectively, consistent with the composition of Yangtze River sediments, indicating that the soils were all derived from alluvium from the catchment. The white veins contained notably more  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$ , and mobile elements relative to the red-brown matrix, while they both showed similar values for the chemical index of alteration of 86.7 and 87.1, respectively, and displayed similar degrees of weathering. The clay minerals in A, AE, and E horizons of the soil profile were illite, kaolinite, and mixed-layer illite–smectite. These same three clay minerals comprised the white net-like veins in the soil B horizon, whereas only illite and kaolinite were observed in the red-brown matrix. Iron (hydr)oxides in A, AE, and E horizons of the soil profile were hematite and goethite, whereas in the red-brown matrix of the B horizon they were hematite, goethite, and ferrihydrite. Different organic compounds were observed for the white vein and the red-brown matrix in the soil B horizon: an 18:2 fatty acid biomarker for fungi in the net-like vein, but not in the red-brown matrix. Compared with the red-brown matrix, the white net-like vein also clearly contained more mono-unsaturated fatty acids, which are sometimes associated with bacteria that have the capacity to reduce Fe (III). Thus, migration of iron and the formation of the net-like veins involved the participation of biota during the hydromorphic pedogenesis process, as ferrihydrite is usually formed with the assistance of biota. Redoximorphic processes will chemically affect phyllosilicates by changing their charge, and Fe(III) in illite may serve as electron acceptor for Fe(III)-reducing microorganisms. This leads to an increase in negative charge in the illite structure which could be neutralized by electrostatic displacement of interlayer cations and adsorption of protons, thereby promoting transformation of illite to mixed-layer illite–smectite and dissolution of the pre-existing illite mineral. Our results suggest that microorganisms brought about iron leaching and dominated the alteration of clay minerals by redox processes, and different mineral volumes formed in the net-like B horizon of the hydromorphic soil were attributed to the microorganism-induced weathering process.

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## 1. INTRODUCTION

Hydromorphic soils are characterized by a period of aquic conditions in normal years in one or more horizons within 50 cm of the mineral soil surface (Soil Survey Staff, 2014). The soils have regions where water table oscillations result in the presence of aquic soil regimes, and alternating of anaerobic and aerobic conditions along the soil profile (van Breemen and Buurman, 2002). Hydromorphic soils occur widely in the world and usually exhibit redoximorphic features resulting from reduction and segregation of iron, giving rise to typically pale gray or white zones or veins in the soil profile (e.g. Brinkman, 1970; Ambrosi and Nahon, 1986; Zhu, 1988; Nahon, 1991; Aleva, 1994; Beauvais, 1999; Rosolen et al., 2002; Hong et al., 2010; D'Amore et al., 2015; Gangopadhyay et al., 2015). Although hydromorphic soils and processes affecting the redox reactions of iron have been extensively studied, only very few observations have provided comprehensive information and understanding concerning the soil-forming process (van Breemen and Buurman, 2002; Nawaz et al., 2014). Brinkman (1970) proposed that in the hydromorphic soil-forming process, the cyclical alternation of redox conditions termed ferrolysis and associated acid attack of clay minerals would result in the leaching of iron and manganese and a decrease in clay abundance in the reduced, light-colored material. Rosolen et al. (2002) investigated mottled soils in the Amazon basin, and their results showed that the gray spots were quartz-rich, the yellow spots consisted of kaolinite, illite, and goethite, whereas the red matrix was iron-enriched. They concluded that the oxidized mottles were formed by intense depletion of Fe-oxides from an initially homogeneous Fe-rich soil during the pedogenic process. After investigation of the formation of redoximorphic features in different hydrologic regimes, Vepraskas et al. (2006) suggested that iron reduction was probably caused by bacteria. They believed that dead root tissue may foster growth of bacteria around it as the tissue was decomposed, giving rise to anaerobic conditions as the bacteria depleted the soil water of oxygen, which finally resulted in iron reduction as the growing bacterial colony used ferric iron as an electron acceptor. Based on observation of the mottled soils in North Carolina and South Carolina, USA, Fimmen et al. (2008) proposed that formation of the hydromorphic soils with different mineral, chemical and organic matter compositions at quite fine scales was due to so-called “rhizogenic” Fe–C redox processes, in which oxidation of organic matter concentrated in the rhizosphere was coupled with reduction of redoxactive species such as iron. These rhizogenic redox-reactions complementing acid- and ligand-promoted reactions represent an integrated biological, chemical, and physical system controlling the mineral weathering of the soils.

Iron is the fourth most abundant element in the earth crust, and in soils it occurs widely in iron-(hydr)oxides and iron-bearing clay minerals (Stucki and Kostka, 2006). Iron-(hydr)oxides are affected not only by inorganic reactions but also by bacterially mediated reduction and oxidation processes. Additionally, Fe-containing sheet silicates may undergo redox reaction with surrounding redox-active

species, resulting in changes in the chemical and physical properties of the clay mineral and its surrounding matrix (Stanjek and Marchel, 2008; Pentrakova et al., 2013). Therefore, iron redox processes in natural environments may couple with the biogeochemical cycles of C, N, S, and P, and are considered as a biogeochemical driving force (Kappler and Straub, 2005; Li et al., 2012). Organisms capable of reducing Fe(III) are ubiquitous in hydromorphic soils, and microbial Fe(III) reduction in Fe-rich clay minerals during the redox cycles is associated with transformation of Fe oxides and clay minerals (Stanjek and Marchel, 2008; Shelobolina et al., 2012; Pentrakova et al., 2013). Fe(III) in sheet silicates may be used by bacteria as an electron acceptor and thus plays a key role in the transformation of soil minerals (Kostka et al., 1996; Stucki and Kostka, 2006). Bioreduction of Fe(III) in smectite clay minerals usually induces the formation of biogenic mixed-layer illite–smectite; however, the experiments with bacteria always had sufficient  $K^+$  in solution coming from the nutrient medium (Kim et al., 2004; Zhang et al., 2012). Although Fe(III) in illite is less reducible than that in smectite, illite exhibits bioreducible property to certain extent due to the variable charge in illite layers and the positive charges at clay edges (Sposito and Prost, 1982; Seabaugh et al., 2006). Upon bioreduction, illite morphology changed dramatically from fibrous needles to plates, even though the structural change was unclear (Dong et al., 2003). Huggett and Cuadros (2005) suggested that in redoximorphic environment in gley soil, Fe(III) reduction in octahedral sites of illite may lead to increased layer charge, and the uncomplete reoxidation of reduced Fe probably caused the illite to smectite transition.

Hydromorphism may involve factors such as soil oxygenation, organic matter, activity and type of microorganisms, temperature, and soil composition (Sorokin et al., 2006; Nawaz et al., 2014). Nevertheless, the influence of different factors on alteration and transformation of the soil minerals during the weathering process are still not well understood, and mechanisms proposed to interpret the formation of these soils are still largely in debate (Boixadera et al., 2003; Lai et al., 2005; Fimmen et al., 2008; Hong et al., 2010; Nawaz et al., 2014; Gangopadhyay et al., 2015). Lack of convincing evidence makes it difficult to understand the alteration and transformation processes. In this paper, we present the results of comprehensive clay mineralogical, iron (hydr)oxide, geochemical, and fatty acid methyl-ester studies of a hydromorphic soil on the banks of Liangzi Lake, Hubei province, south China. Our objective is to characterize the clay mineralogy, Fe-(hydr)oxides, geochemistry, and microorganism activity of the contrasting pedogenic horizons of a hydromorphic soil, and therefore, to shed more light on mechanisms of iron migration and alteration and transformation of clay minerals in these sorts of soils.

## 2. MATERIALS AND METHODS

### 2.1. The soil profile and sampling

The Liangzi Lake bank soil profile (114°26'E, 30°34'N) is located in southeast Hubei, south China, in the zone of

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