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Development of Fe-rich clay minerals in a weathering profile derived from serpentinized ultramafic rock in Nui Nua massif, Vietnam

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ABSTRACT

We investigated the chemical and mineralogical characteristics of a weathering profile of the Nui Nua serpentinized ultramafic-mafic massif (Thanh Hoa Province, Vietnam) using TEM-EDX, XRD, and FT-IR methods to understand the effects of monsoon climate conditions on weathering processes and clay minerals, especially transformation and neoformation of clay minerals. Clay minerals of this profile were characterized by a temporariness of serpentine clay minerals though chlorite seemed to be stable and increased from the bottom layer to the top layer. The nature of Fe changed rapidly from ferrous Fe (close to parent rocks) to ferric Fe and additionally it was incorporated into the octahedral sheet of dioctahedral smectite. Consequently, a Fe-rich smectite formed at the beginning of the weathering process and was detected as a dominant phase in the clay fraction of the profile. This phase was later transformed progressively to Al-rich smectite because of Fe-oxidation and corresponding Si-mitigation. Chemically, the contents of Fe and Mg reduced from the bottom layer to the top layer but Al developed vice versa. Magnetite in top layer was formed by bacterial activities. The transformations of clay minerals were controlled strongly by leaching conditions in the weathering system and by the effects of oxidation of Fe ($Fe^{2+} \rightarrow Fe^{3+}$) on clay minerals paragenesis.

1. Introduction

Rocks in ophiolite sequences are composed of ultramafic (Fe, Mg-rich) minerals such as olivine and pyroxene. Serpentinite is formed during the serpentinization of ultramafic rocks. The reaction of serpentinite with water leads to the formation of a series of Mg- and Fe-rich clay minerals (Velde and Meunier, 2008). These rocks are rich in Fe and Cr, which primarily occur in oxides (magnetite, Cr-magnetite, and ferrichromite) and sheet silicates as secondary products of later transformations (Frost and Beard, 2007). The weathering products of ultramafic rocks under monsoon climate conditions have been intensively studied over recent decades especially due to the considerable economic interest in ore deposits (Golightly, 1981; Gaudin et al., 2004). Serpentine soils derived from the weathering of ultramafic rocks are typically characterized by high concentrations of four metals – nikel (Ni), chromium (Cr), manganese (Mn), and cobalt (Co) (Bonifacio et al., 1997; Brooks, 1987; Caillaud et al., 2009; Horen et al., 2014; Kataba-Pendias and Pendias, 1984; Lee et al., 2004; Schwertmann and Latham, 1986). Geochemistry and pedochemistry of paddy soils derived from serpentinites have been the subject of many studies over the past few years with the aim of understanding the potentially toxic forms of these metals (Caillaud et al., 2009; Hseu and Iizuka, 2013).

Caillaud et al. (2004, 2006) and Nguyen-Thanh et al. (2014) studied the weathering products of serpentinite rocks which released Fe-oxides and especially Fe-rich clay minerals. They also found a relationship between the weathering process under tropical climate condition and the neoformed clay minerals. Moreover, the factors controlling the transformation rates and the neoformation phases of serpentinite during weathering have been reported in many studies (Chang et al., 2013; Cheng et al., 2011; Hseu et al., 2015; Lessovaia et al., 2012). Alexander et al. (1989), Graham et al. (1990), and Rabenhorts et al. (1982) found a relationship between mineralogical and chemical properties of serpentinite and vegetation. Cleaves et al. (1974) and Clemency (1975) compared the weathering products of different parent rocks in the same climate and topographic conditions and found that the serpentinite soils were composed of the 14 Å clay mineral and

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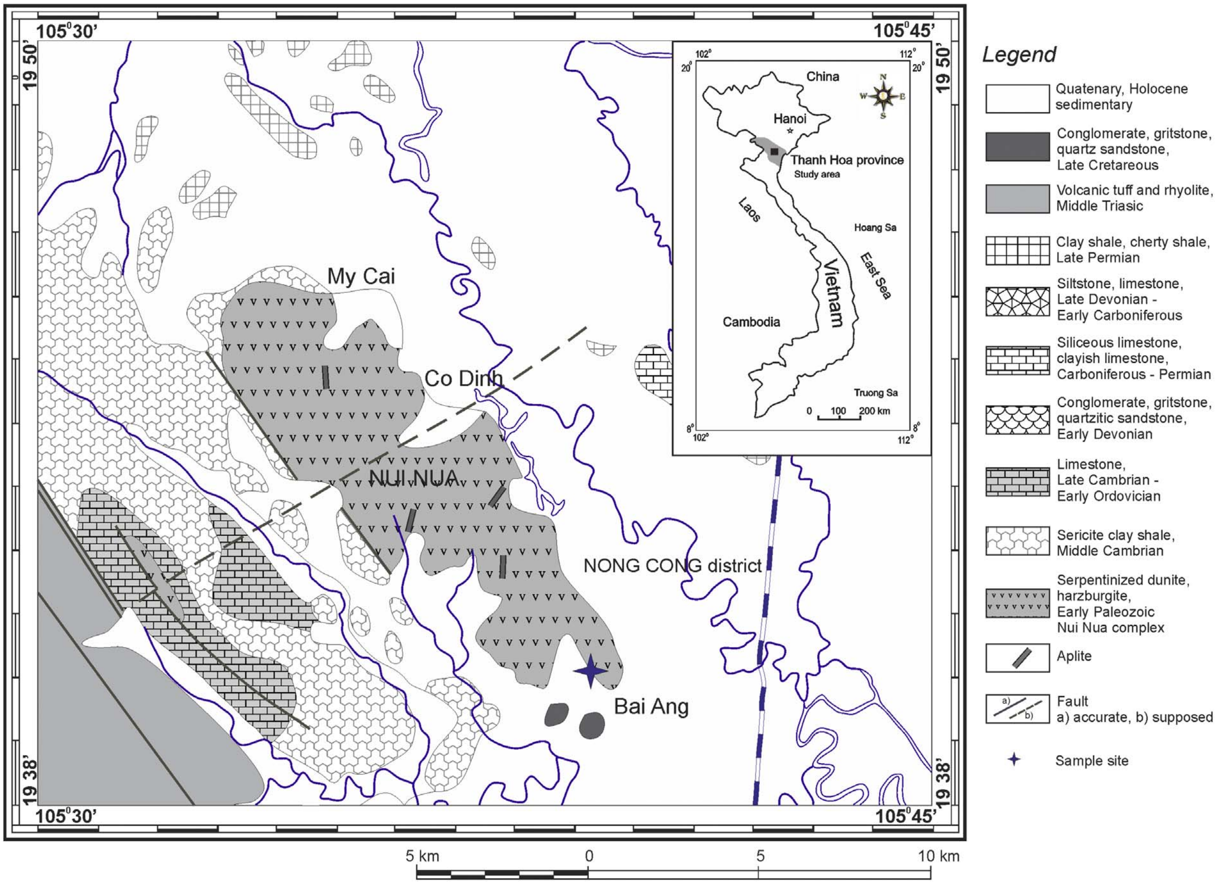

Fig. 1. Geological map of Nui Nua massif and location of Bai Ang area.

quartz. Otherwise, schist or gneiss soils contained clay minerals, gibbsite, and reddish kaolinite though greenish ferric iron-rich smectite was found in the weathering crust of amphibolite. Moreover, the thickness of the saprolite layer and the type or the mineralogical composition of soils derived from serpentinite rocks depended on the drainage (Bonifacio et al., 1996; Ducloux et al., 1976; Istok and Harward, 1982). The well-drained soils consisted mainly of 7 Å clay minerals including serpentinite and chlorite (Istok and Harward, 1982) or normal to low-charged vermiculite (Bonifacio et al., 1996). Otherwise, in poorly-drained soils, other clay minerals, such as beidellite (Berre et al., 1974), nontronite, chlorite, chlorite-vermiculite interstratifications (Ducloux et al., 1976), or low-charged smectite (Bonifacio et al., 1996), could be formed. In addition, Fe-rich smectites including nontronite and Fe-rich montmorillonite were mostly found from weathered serpentinized ultramafic rocks (Caillaud et al., 2004, 2006; Ducloux et al., 1976; Lee et al., 2003; Meunier et al., 2010; Seki and Yurdakoç, 2007; Wildman et al., 1968, 1971).

The Nui Nua massif is located in the Thanh Hoa province in the northern central region of Vietnam (Fig. 1). This massif is the largest ultramafic-mafic massif in Vietnam and is well known as a part of the Song Ma ophiolite zone (Bach et al., 1982; Chuong et al., 2001; Findlay and Trinh, 1997; Hung, 1999; Hutchison, 1975). The rock is composed mainly of harzburgite, lherzolite, dunite, gabbro, and diabase of Cambrian and Ordovic ages (Chien, 1964; Tri et al., 1986; Vuong et al., 2006) and has been altered to serpentinite and schist of actinolite and talc-actinolite containing talc and serpentine minerals (Chien, 1964; Son, 1975; Tong-Dzuy and Vu, 2011). Although the chemical and mineralogical properties of the Nui Nua clay have been published (Nguyen-Thanh et al., 2014), the weathering process was not well understood. Little is known about the weathering effects on the transformation and neoformation of sheet silicates (Fe-smectite) in weathering profiles, which developed on serpentinite parent rock and under a tropical climate, especially with respect to behaviour of Fe. This work presents some properties and the evolution of Fe-rich dioctahedral smectite in the Nui Nua weathering profile as a case study of the specific dissolution potential of Fe-smectite in a high drainage system under monsoon conditions. An Fe-pathway is described based on TEM-EDX measurements, which leads to a better definition of the geochemical structural evolution of sheet silicates according to weathering sequences of Fe containing phases. The particle-wise analysis by TEM-EDX mirrors the alteration in chemical composition of the interlayer, octahedral and tetrahedral sheets of the investigated particles. TEM-EDX is very sensitive to the transformation processes of clay minerals that occur in response to geological processes such as tropical weathering, hydrothermal alteration or diagenesis, or technical processes, such as pyrophyllitization, kaolinitization, illitization, saponitization, vermiculitization.

2. Materials and methods

2.1. Area description and sample collection

The study site is located in the Bai Ang area, in the south eastern part of the Nui Nua massif (Fig. 1), where serpentinite has been exploited (open cuts). The altitude of this area is around 160 m above sea level (ca. 19°40′N, 105°39′E) with a steep (18–20°) slope towards the east. The slope was covered by subshrub but it was easy to see some coarse fragments of serpentinite rocks which can be up to half of meter in diameter. It was assumed that the fine material in the profile is entirely the product of the local ultramafic weathering but the mineralogical data demonstrated that there is some allochthonous material. According to Köppen's climate classification system, this area has a