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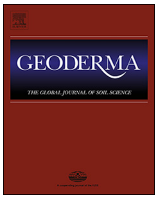

Surficial weathering of kaolin regolith in a subtropical climate: Implications for supergene pedogenesis and bedrock argillization

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ABSTRACT

Regolith, or in situ weathered material overlying bedrock, develops through pedogenic processes such as clay-mineral formation (“argillization”). Research on products of argillization, such as kaolin, is commonly focused on its economic value rather than on an integrated understanding of the pedological, mineralogic, and geochemical processes taking place in the regolith. Here, we analyzed three kaolin-regolith drillholes from a subtropical climate zone in southern China using X-ray diffraction (XRD), major and trace element analyses, H and O isotopes, differential thermal-thermogravimetric analysis (DTA-TG), and scanning electron microscopy (SEM). Many kaolinite aggregates within regolith have fan-shaped stacks, a morphology that is closely associated with transformation of muscovite plates. Hypogene indicators such as a shallow level of granite emplacement, representative structural controls on kaolinization, mineral zoning, and hypothermal minerals cannot be observed. Mineral assemblages and morphologies, elemental binary plots (e.g., Zr vs. TiO_2 and P_2O_5 vs. SO_3), as well as chemical profiles all show characteristics typical of surfical weathering. The $\delta^{18}O$ and $\delta^{2}H$ values of the clay fractions range from $+16.8$ to $+18.7‰$ and from -70 to $-51‰$, respectively, suggesting that supergene weathering has played a key role in forming the kaolin regolith. Dominance of kaolinite over halloysite implies a near-surface, freely draining environment. Adopting the underlying weakly altered granite (saprolith) as the parent material, the kaolin regolith exhibits four elemental profile patterns as revealed by mass transfer coefficients: (1) depletion (e.g., Na), (2) depletion-enrichment (e.g., Al), (3) enrichment (e.g., Mn), and (4) biogenic (e.g., Ca). These profiles reflect a combination of chemical, geologic, and biologic processes that are typical of relatively thin, in situ regolith profiles, and that is not necessarily similar to those typically associated with deep (thick) granite weathering profiles. We propose that supergene kaolin regolith is intrinsically more similar to shallow, biologically active residual soil deposits, rather than deeply weathered granite-hosted regoliths.

1. Introduction

Regolith is defined as in situ weathered material overlying bedrock, and it is present to varying degrees across the Earth (Wilford et al., 2016). It represents the entire unconsolidated and secondarily recemented cover that overlies relatively intact bedrock, and it is formed by erosion, transport/deposition, and weathering of pre-existing material (Anand and Paine, 2002; Keeling et al., 2003). The regolith is host to many of the resources that maintain our society and living standards. It is also in the upper part of the regolith that our soils are formed; the regolith is thus the host for life and agriculture (Taylor and Eggleton, 2001). The distribution and nature of regolith are controlled by various factors, reflecting long-term interactions among the atmosphere, pedosphere, lithosphere, biosphere and hydrosphere (Brantley and White, 2009; Buss et al., 2017). Regolith develops through a variety of processes such as argillization, i.e., the formation of clay minerals in the

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soil environment (Gilkes et al., 1973), and clay minerals commonly provide insights into the nature of these processes (Dill, 2017; Lopez et al., 2018).

One type of argillization process is kaolinization, which generates the mineral kaolin (Ekosse, 2001; Anand and Paine, 2002; Dill, 2017). Kaolin deposits can be formed in situ by hydrothermal processes during late-stage pluton cooling (hypogene), by (near-)surface weathering reactions (supergene), or by a combination of these processes (Dudoignon et al., 1988; Murray, 1988; Dill et al., 1997; Galán et al., 2016). Hypogene processes play a prominent role in alteration of granitic rocks intruded at relatively shallow depths, and it is commonly associated with high acidity and high temperatures (Marfil et al., 2005). In supergene deposits, chemical weathering is the dominant process of formation of kaolin minerals (e.g., kaolinite, halloysite, and dickite) from a variety of parent rocks (e.g., granite, gneiss, and slate; Chorover and Sposito, 1995; Scott and Bristow, 2002; Keeling, 2015; Hong et al., 2016). With time, regolith composition diverges progressively from that of the parent material owing to the cumulative influences of vegetation, biota, topography, and, in particular, climate (Thanachit et al., 2006; Zhao et al., 2017). In the present study, “kaolin regolith” is defined as a residual kaolin deposit that has undergone substantial chemical leaching and physical weathering processes such as regolith creep/flow and erosion by water (Taylor and Eggleton, 2001).

Kaolin genesis has a direct relation to its industrial applications, with supergene kaolins generally being more concentrated and having higher economic value than hypogene ones (Ekosse, 2001; Njoya et al., 2006; Fernandez-Caliani et al., 2010). As a raw material for chinaware, kaolin has substantial economic value, but its economic significance is broader in that kaolin is found in many products for daily use, including paper coating, ceramics, plastics, rubber, cosmetics, and pharmaceuticals (Zhou and Keeling, 2013; Lu et al., 2016; Pruett, 2016). Relatively systematic work on kaolin argillization has been undertaken in Australia (Gilkes et al., 1973; Anand and Paine, 2002), USA (Schroeder et al., 2004), and Europe (Galán et al., 2016). China represents one of the most important producers of kaolin (especially for ceramics, sanitary ware and porcelain; Wilson, 2004). Indeed, the first use of kaolin in pottery manufacture emerged in Jingdezhen, Jiangxi Province, China over 2000 years ago (Schroeder and Erickson, 2014), and the first mining site for *gaoling tu* (kaolin clay) was the Gaoling area, located 50 km northeast of Jingdezhen. Despite its cultural importance, only limited research concerning the formation of kaolin regolith particularly related surficial weathering process has been conducted in China.

A detailed study of kaolin deposits in China has the potential to improve our understanding of mineral alteration and elemental migration during supergene processes and kaolin accumulation worldwide. The Chongyi kaolin regolith in Jiangxi Province, southern China, is located in a subtropical climate zone with warm temperatures and high rainfall, contributing to its intensely weathered condition. Here, we investigate the Chongyi kaolin regolith using an integrated approach based on XRD, SEM, XRF, ICP-MS, DTA-TG, and H and O isotopic analyses. The goals of this study were to: (1) systematically characterize the kaolin regolith using a variety of mineralogic and geochemical techniques; (2) gain insights into the geochemical behavior of major and trace elements during supergene kaolin argillization; (3) determine the genetic relationships between kaolin regolith and the typical residual soil deposit; and (4) propose a model for kaolin argillization in Earth's surface environments.

2. Materials and methods

2.1. Study location and climatic setting

The Chongyi kaolin regolith is located in Chongyi County, southwestern Jiangxi Province, southern China (Fig. 1). The landscape consists of low mountains and hilly terrane. Within Chongyi County, elevations fall from northwest (~1000 meters above sea level - masl) to southeast (~500 masl), with rivers thus flowing in a southeasterly direction. The study area is within a subtropical monsoonal climate zone that is characterized by large seasonal variations in air temperature, wind direction, and rainfall, with > 80% of precipitation falling during the period from May to September. Mean annual temperature (MAT) is 17.6 °C, and mean annual precipitation (MAP) is 1600 mm. The Chongyi kaolin regolith is located within the Nanling metallogenic belt, which was generated during the Jurassic-Cretaceous (~200–134 Ma) Yanshanian magmatic event. The Yanshanian event resulted in NE–SW-trending faults and associated pluton emplacement in the study area (Zhou et al., 2006; Wang et al., 2018). The bedrock of the kaolin regolith is a muscovite granite of early Yanshanian age (~200–170 Ma), and the original argillization process may have been driven by magmatic heat.

According to layering of typical regolith profiles by Taylor and Eggleton (2001) and Anand and Paine (2002), the Chongyi regolith can be divided into a lower saprolith layer and an upper pedolith layer (Fig. 2): (1) the saprolith contains saprorock below and saprolite above; and (2) the pedolith contains a plasmic zone, a mottled zone, and an overlying surface layer. The regolith can be further vertically subdivided into four units based on differences in color and lithology (Fig. 2; listed from top to bottom): the surface layer, the mottled kaolin layer, the homogenous kaolin layer, and the saprolith.

2.2. Field sampling

Geological field work included mapping in outcrop and description and sampling of drillcore and drill-cuttings. In each of three drillcores, five samples were collected: one in each regolith layer (surface layer, mottled kaolin, homogenous kaolin, and saprolith) with a second sample taken in the homogenous kaolin layer (the two samples of the homogeneous layer are designated “upper kaolin” and “lower kaolin”). This yielded a total of 15 samples for geochemical analyses (Fig. 2). Clay fractions ($< 2\,\mu m$) were extracted from 12 samples (all but the three saprolith samples) by conventional sedimentation and centrifugation methods. The $< 45\,\mu m$ fractions were extracted from three kaolin samples (i.e., CY 1–3, CY 2–3, and CY 3–3). The grain-size fraction of 63–600 μm was used for analysis of heavy minerals.

2.3. X-ray diffraction (XRD)

The samples of bulk kaolin (including mottled kaolin and homogeneous kaolin), fine fractions ($< 45\,\mu m$ and $< 2\,\mu m$), and heavy minerals were mineralogically analyzed by XRD. These samples were pretreated to remove organic matter with 30% H_2O_2 at 60 °C. Air-dried oriented clay mounts for the $< 2\,\mu m$ fractions were prepared by dispersing the clay slurry onto glass slides. XRD patterns were recorded using a D/max-3B X-ray diffractometer with Cu Kα radiation at 40 kV, 35 mA (resolution ratio = 0.02°2θ; scan rate = 4° 2θ/min; scan range = 3–65°2θ). The formamide test (Churchman et al., 1984) was carried out for the $< 2\,\mu m$ fractions to distinguish halloysite from kaolinite. Bulk-rock mineral contents were determined on randomly oriented powders using reference intensity ratio(RIR) together with the so-called 100% approach (Hillier, 2000). RIR is also known as I/I_{cor}, which represents the ratio of intensities (areas) of specific XRD peaks (mineral: corundum = 1:1). The term 100% approach means that the sum of all mineral phases identified is 100%. Five minerals (muscovite, feldspar, quartz, kaolinite, and gibbsite) that were involved in content calculation have RIR values (refer to corundum 113 peak) of 0.23 (~10.0 Å), 2.07 (~3.20 Å), 0.89 (~4.25 Å), 0.53 (~3.57 Å), 1.91 (~4.85 Å), respectively. Considering this is a semi-quantitative method with a calculation error of 5–10%, we used ‘—’ (not detected), ‘*’ (< 15%), ‘**’ (15–40%), and ‘***’ (> 40%) to describe different mineral contents in order to present more reasonable data (Table S1).