Clay mineral composition of upland soils and its implication for pedogenesis and soil taxonomy in subtropical China

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Clay minerals are intermediate products generated during soil development, and their neoformation and transformation are closely related to pedogenesis. Here we aimed at identifying the difference in the clay mineral composition of upland soils derived from different parent materials and different soil-forming environments and exploring the importance of clay mineral composition in pedogenesis and soil taxonomy. We sampled 60 soil B horizons in Hunan Province of subtropical China by digging soils derived from granite (GR), slate and shale (SS), Quaternary red clay (QRC), limestone (LS), and sandstone (SDS). The clay mineral composition and its correlation with parent materials, elevation, micro-topography, and pedogenic processes were investigated using X-ray diffraction and Pearson’s correlation analysis. The clay mineral was dominated by kaolinite, followed by 2:1-type minerals (illite and vermiculite), and a small fraction of mixed-layer minerals. The composition of soil clay minerals varied with parent materials. Kaolinite was predominant in soils derived from GR and LS; mixed-layer minerals prevailed in QRC, whereas illite and vermiculite were prevalent in SDS. In addition, elevation and micro-climate could also explain the variations in clay mineral composition. Increase in elevation was associated with decreased 1:1 clay mineral content and increased 2:1 clay mineral content, especially in soils developed from LS. The composition and content of clay minerals indicated that Ferrosols, Ultisols, and Acrisols had undergone intense weathering; Primosols, Entisols, and Leptosols were characterised by weak weathering, and Plinthic Ali-Udic Cambosols, Plinthudults, and Plinthosols were characterised by strong redox status. This study suggests that clay mineral composition is related to the parent material, climate, and micro-topography, and that it can serve as an indicator of pedogenesis and soil type in subtropical China.
only one classification system, such as the World Reference Base for Soil Resources (WRB)\textsuperscript{17}, Soil Taxonomy (ST)\textsuperscript{18}, or Chinese Soil Taxonomy (CST)\textsuperscript{19}, to examine the variation in clay mineral composition among different soil types\textsuperscript{20–23}. A few studies have investigated the variation in the composition of clay minerals derived from diverse parent materials (more than three parent materials) and the effects of comprehensive environmental factors (e.g., elevation and micro-topography) on clay minerals in the subtropics. However, the dynamic changes in clay minerals among different soil types based on multiple soil classification systems (CST, ST, and WRB) have received limited attention.

Owing to its diversity of parent materials, complex topography, and changeable climate, the subtropical region of China has diverse soil types\textsuperscript{24,25}. Desilication and ferrallitisation are common in climates with high air temperatures and precipitation, and this results in the formation of a LAC-ferric horizon (lower-activity clay and free iron oxide rich) in Ferrsols. Clay migration is active in soils characterised by strong eluviation, and it occurs in soils originating from coarse parent materials. These soils easily develop an argic horizon (clay illuviation), which is necessary for Argosols formation. In areas with low air temperature, parent materials tend to be weakly weathered. The soils derived from these parent materials can form a cambic horizon (slight silica-illitisation), which is necessary for Cambosols formation. On steep slopes, Primosols are common due to the dynamic balance between soil formation and erosion\textsuperscript{25,26}.

Hunan Province is home to an ideal series of soils that enable investigation of the effects of parent materials (more precisely, five parent materials) and topography (plains, hills, and mountains) on the composition of clay minerals under warm and humid subtropical climate. The parent rocks or materials in the upland soils of the Hunan Province mainly consist of Quaternary red clay (QRC), granite (GR), slate and shale (SS), sandstone (SDS), and limestone (LS)\textsuperscript{27}. Thus, the pedogenic processes of upland soils vary widely because of the diversity of parent materials and the complex topography. It has been reported that the variation in clay mineral composition of Stagnic Anthrosols (paddy soil) is a consequence of diverse parent materials present in Hunan Province\textsuperscript{23}; however, there have only been a few studies on the upland soils (natural soils) in this area. Therefore, in the present study, we selected 60 upland soils derived from five parent materials in the Hunan Province and analysed the effect of different parent materials, soil environments, diagnostic horizons, and soil types on clay mineral types and contents using X-ray diffraction. The goals of this study were to (i) investigate the effects of diverse parent materials and environmental factors (elevation and micro-topography) on soil clay mineral composition and (ii) explore the evolution of clay minerals during soil development and its implications for soil taxonomy.

**Materials and methods**

**Study area and field sampling.** This study was conducted in Hunan Province, south-central China (Fig. 1). Hunan Province is located in the middle reaches of the Yangtze River, bordering Dongtinghu Lake in the north. It covers a land area of 211,800 km\(^2\), characterised by the presence of hills and basins in the centre, mountains in the east, south, and west, and plains in the north. Hunan has a mid-subtropical, seasonally humid climate characterised by moderate air temperature, abundant precipitation, and clear differentiation between wet and dry seasons. The annual sunshine duration is 1300–1800 h, and the annual average air temperature is 16–18 °C. The area is frost-free 260–310 days a year, and the annual precipitation ranges from 1200 to 1700 mm\textsuperscript{27}. Sixty sites were selected, covering elevations from 41 to 650 m a.s.l. (Table 1). Parent materials of the soils in the Hunan province mainly included granite (GR), slate and shale (SS), Quaternary red clay (QRC), limestone (LS), and sandstone (SDS). The formation period of GR consists of three geological times, namely...
| Profile | Horizon | Depth (cm) | Elevation (m) | Landform† | Parent material | Chinese soil taxonomy (soil subgroup) | Soil taxonomy (soil subgroup) | World reference base for soil resources (second level) |
|---------|---------|------------|--------------|-----------|----------------|-------------------------------------|-------------------------------|--------------------------------------------------|
| CS16    | Bw      | 50–90      | 126          | LH-MS     | Typic Argi-Udic Ferrosols | Typic Paleudults | Xanthic Acrisols (Dystric, Loamic) |
| XT04    | Bt      | 30–60      | 107          | LH-MS     | Typic Argi-Udic Ferrosols | Typic Paleudults | Dystric Chromic Sideralic Cambisols (Loamic, Ochric) |
| YIY02   | AC      | 20–60      | 107          | LH-BS     | Lithic Usti-Orthic Primosols | Lithic Ustorthents | Dystric Lithic Leptosols (Loamic, Ochric) |
| YZ01    | Bt      | 27–70      | 209          | HH-MS     | Xanthic Ali-Udic Argosols | Typic Paleudults | Xanthic Alisols (Clayic, Hyperdystric, Ochric) |
| YY08    | Bw      | 45–105     | 48           | LH-MS     | Typic Hapli-Udic Ferrosols | Oxic Dystrudepts | Hyperdystric Xanthic Sideralic Cambisols (Loamic, Ochric) |
| HH10    | Bt      | 30–65      | 231          | HH-MS     | Slate and shale (SS) | Typic Paleudults | Xanthic Alisols (Cutaenic, Loamic) |
| ZZ03    | Bw      | 20–50      | 87           | LH-LS     | Xanthic Ali-Udic Argosols | Typic Paleudults | Xanthic Alisols (Cutaenic, Differentic, Hyperdystric, Loamic) |
| YIY07   | BC      | 85–115     | 59           | LH-MS     | Typic Ali-Udic Cambosols | Typic Humudepts | Cambic Umbrisols (Chromic, Clayic, Dystric) |
| YY09    | Bw      | 30–80      | 103          | LH-LS     | Typic Ali-Udic Cambosols | Typic Dystrudepts | Hyperdystric Chromic Cambisols (Loamic, Ochric) |
| ZZ06    | Bw      | 25–65      | 190          | LS-MS     | Typic Ali-Udic Cambosols | Typic Dystrudepts | Hyperdystric Xanthic Cambisols (Loamic, Ochric) |
| YIY01   | Bw      | 56–97      | 226          | LH-BS     | Typic Ferri-Udic Cambosols | Typic Dystrudepts | Eutric Xanthic Cambisols (Loamic, Ochric) |
| CZ02    | BC      | 50–130     | 606          | LM-MS     | Typic Ferri-Udic Cambosols | Typic Dystrudepts | Xanthic Xanthic Cambisols (Loamic, Ochric) |
| CS02    | Bt      | 62–137     | 63           | LH-MS     | Typic Argi-Udic Ferrosols | Typic Paleudults | Rhodic Ferric Alisols (Clayic, Cutanic, Hyperdystric) |
| CS11    | Bks     | 33–71      | 41           | LH-MS     | Typic Ali-Udic Argosols | Typic Plinthudults | Aric Plinthosols (Clayic, Dystric, Ochric) |
| CS15    | Bw      | 40–90      | 109          | LH-LS     | Typic Ali-Udic Cambosols | Typic Dystrudepts | Dystric Rhodic Sideralic Cambisols (Loamic, Ochric) |
| ZZ02    | Bt      | 40–65      | 75           | LH-US     | Typic Ali-Udic Argosols | Typic Paleudults | Chromic Ferric Alisols (Clayic, Cutanic, Hyperdystric) |
| ZZ08    | Bt      | 25–95      | 105          | LH-LS     | Typic Argi-Udic Ferrosols | Typic Paleudults | Rhodic Acisols (Clayic, Cutanic, Hyperdystric) |
| XT03    | Bt      | 60–150     | 77           | LH-LS     | Typic Ali-Udic Argosols | Typic Paleudults | Chromic Alisols (Clayic, Cutanic, Hyperdystric) |
| YY04    | Bw      | 20–60      | 69           | LH-US     | Plinthic Ali-Udic Cambosols | Typic Dystrudepts | Plinthoferic Plinthosols (Clayic, Dystric, Ochric) |
| YY06    | Bw      | 55–95      | 56           | LH-LS     | Mottlic Ali-Udic Cambosols | Typic Dystrudepts | Dystric Chromic Cambisols (Clayic, Ferric, Ochric) |
| YY07    | Bks     | 60–120     | 83           | LH-MS     | Plinthic Ali-Udic Cambosols | Typic Plinthudults | Aloc Umbric Plinthosols (Clayic, Hyperdystric) |
| CD01    | Bw      | 55–120     | 58           | LH-US     | Typic Ali-Udic Cambosols | Typic Dystrudepts | Hyperdystric Chromic Cambisols (Clayic, Ochric) |
| CD06    | Bw      | 40–80      | 66           | LH-MS     | Typic Ali-Udic Cambosols | Typic Dystrudepts | Hyperdystric Chromic Cambisols (Clayic, Ochric) |
| HY05    | Bw      | 40–110     | 107          | LH-LS     | Plinthic Argi-Udic Ferrosols | Plinthic Paleudults | Rhodic Plinthic Acrisols (Cutanic, Differentic, Hyperdystric) |

 Continued
| Profile | Horizon | Depth (cm) | Elevation (m) | Landform† | Parent material | Chinese soil taxonomy (soil subgroup) | Soil taxonomy (soil subgroup) | World reference base for soil resources (second level) |
|---------|---------|-----------|-------------|-----------|----------------|----------------------------------|-------------------------------|------------------------------------------------|
| LY03    | Bt      | 60–120    | 179         | LM-MS     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Chronic Acrisols (Cutanic, Hyperdystric) |
| LY04    | Bt      | 49–107    | 482         | LM-LS     |                | Typic Ali-Udic Argosols          | Typic Paleudults               | Chronic Alisols (Cutanic, Dystric, Loamic)   |
| LY21    | Bw      | 11–51     | 650         | LM-MS     |                | Xanthic Ali-Udic Cambosols       | Typic Dystrudepts              | Hyperdystric Xanthic Cambisols (Loamic, Ochric) |
| XT02    | Bt      | 55–100    | 89          | LH-LS     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Chronic Acrisols (Cutanic, Dystric, Loamic)   |
| YY01    | Bw      | 40–90     | 287         | LH-MS     |                | Typic Ali-Udic Cambosols         | Typic Dystrudepts              | Hyperdystric Xanthic Cambisols (Loamic, Ochric) |
| YY05    | Bt      | 47–70     | 87          | LH-LS     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Acric Umbrisols (Clayic, Clastic, Hyperdystric, Sideralic) |
| YY10    | Bw      | 75–100    | 128         | LH-MS     |                | Typic Hapli-Udic Ferrosols       | Oxic Dystrudepts               | Sideralic Chromic Cambisols (Dystric, Loamic)   |
| YIY04   | Bw      | 40–120    | 68          | LH-MS     |                | Rhodic Hapli-Udic Ferrosols      | Oxic Dystrudepts               | Cambic Umbrisols (Clayic, Clastic, Hyperdystric, Sideralic) |
| CZ06    | Bw      | 25–75     | 350         | HH-MS     |                | Red Ferri-Udic Cambosols         | Typic Dystrudepts              | Dystric Cambisols (Loamic, Ochric)           |
| SY08    | Bw      | 60–120    | 416         | HH-LS     |                | Xanthic Hapli-Udic Ferrosols     | Oxic Dystrudepts               | Hyperdystric Xanthic Sidereal Cambisols (Loamic, Ochric) |
| HY07    | Bt      | 25–48     | 81          | LH-LS     |                | Typhic Hapli-Udic Ferrosols      | Oxic Dystrudepts               | Hyperdystric Xanthic Sidereal Cambisols (Loamic, Ochric) |
| CS18    | Bt      | 30–60     | 100         | LH-LS     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Hyperdystric Chromic Sideralic Cambisols (Loamic, Ochric) |
| ZZ05    | Bt      | 26–60     | 263         | LH-MS     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Acric Umbrisols (Clayic, Hyperdystric, Sideralic) |
| YZ03    | Bt      | 90–140    | 201         | LH-US     |                | Typic Argi-Udic Ferrosols        | Typic Paleudults               | Acric Umbrisols (Clayic, Hyperdystric, Sideralic) |
| YZ05    | Bts     | 60–100    | 228         | LH-MS     |                | Mottlic Argi-Udic Ferrosols     | Ulic Hapludalis                | Chronic Ferric Liixsols (Clayic, Cutanic, Differentic, Hyperpyric) |
| YZ06    | Bts     | 80–135    | 290         | HH-MS     |                | Mottlic Argi-Udic Ferrosols     | Aquic Paleudults               | Xanthic Ferric Liixsols (Clayic, Cutanic, Differentic, Hyperpyric) |
| YZ08    | Bts     | 60–130    | 380         | HH-MS     |                | Trunic Argi-Udic Ferrosols       | Typic Paleudults               | Chronic Ferric Acrisols (Clayic, Cutanic, Differentic, Hyperpyric) |
| CZ04    | Bt      | 75–110    | 404         | HH-MS     |                | Humic Ferri-Udc Argosols         | Ulic Hapludalis                | Lithic Umbrisols (Clayic, Hyperpyric, Rhodic) |
| CZ05    | Bt      | 100–160   | 255         | HH-MS     |                | Tyric Argi-Udic Ferrosols       | Typic Paleudults               | Acric Umbrisols (Clayic, Dystric, Sideralic) |
| SY04    | Bt      | 20–43     | 462         | LM-LS     |                | Red Ferri-Udc Argosols           | Typic Rhududals                | Placic Rhodic Ferric Luvisols (Clayic, Cutanic, Differentic, Hyperpyric) |
| SY07    | Bt      | 80–130    | 343         | LH-MS     |                | Mottlic Hapli-Udc Ferrosols     | Oxic Dystrudepts               | Dystric Xanthic Sideralic Cambisols (Clayic, Ferric, Ochric) |
| SY09    | Bt      | 50–100    | 320         | LH-US     |                | Xanthic Argi-Udc Ferrosols      | Typic Paleudults               | Chronic Ferric Acrisols (Clayic, Cutanic, Differentic) |
| LD01    | Bt      | 30–65     | 174         | LH-TS     |                | Tyric Argi-Udc Ferrosols        | Typic Paleudults               | Chronic Acrisols (Clayic, Cutanic, Differentic, Hyperdystric) |
| HY02    | Bt      | 50–105    | 92          | LH-MS     |                | Trunic Argi-Udc Ferrosols       | Typic Paleudults               | Chronic Ferric Acrisols (Clayic, Cutanic, Differentic) |

Continued

Granite (GR)  
Limestone (LS)
Table 1. Fundamental information about study sites and soil classification. †LH-TS: Low Hill-Top of slope; LH-US: Low Hill-Upslope; LH-MS: Low Hill-Mesoslope; LH-LS: Low Hill-Lower Slope; LH-BS: Low Hill-Bottom slopes; HH-US: High Hill-Upslope; HH-MS: High Hill-Mesoslope; HH-LS: High Hill-Lower Slope; HH-BS: High Hill-Bottom slopes; LM-MS: Low Mountain-Mesoslope; LM-LS: Low Mountain-Lower Slope.

| Profile | Horizon | Depth (cm) | Elevation (m) | Landform† | Parent material | Chinese soil taxonomy (soil subgroup) | Soil taxonomy (soil subgroup) | World reference base (for soil resources) (second level) |
|---------|---------|------------|--------------|-----------|----------------|--------------------------------------|----------------------------|-----------------------------------------------------|
| XT05    | BC      | 40–110     | 59           | LH-MS     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Dystric Chromic Cambisols (Loamic, Ochric)          |
| YIY03   | Bw      | 80–120     | 147          | LH-US     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Hyperdystric Chromic Cambisols (Loamic, Ochric)    |
| CD02    | Bt      | 65–110     | 197          | LH-MS     | Tybic Ferri-Udic Argosols        | Tybic Paleudults           | Aline Umbrisols (Eutric, Loamic)                   |
| YZ02    | Bw      | 35–75      | 238          | LH-US     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Hyperdystric Chromic Cambisols (Loamic, Ochric)    |
| YZ04    | Bw      | 15–60      | 216          | LH-US     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Hyperdystric Xanthic Cambisols (Loamic, Ochric)    |
| YZ07    | Bw      | 25–95      | 309          | HH-MS     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Hyperdystric Xanthic Cambisols (Loamic, Ochric)    |
| SY03    | Bw      | 45–110     | 317          | LH-MS     | Tybic Hapli-Udic Ferrorsols      | Oxic Dystrudepts           | Cambic Umbrisols (Hyperdystric, Loamic, Siderallic) |
| SY06    | Bw      | 12–48      | 375          | LH-MS     | Tybic Ali-Udic Cambisols          | Tybic Dystrudepts          | Hyperdystric Xanthic Cambisols (Loamic, Ochric)    |
| SY10    | Bt      | 80–160     | 370          | LH-MS     | Humic Ali-Udic Argosols          | Tybic Paleudults           | Xanthic Alisols (Cutanic, Differentic, Hyperdystric, Loamic) |
| SY01    | Bw      | 20–50      | 379          | HH-MS     | Humic Ali-Udic Cambisols          | Tybic Humudepts           | Cambic Umbrisols (Clayic, Hyperdystric)            |
| XX03    | Bt      | 20–60      | 421          | HH-MS     | Xanthic Ali-Udic Argosols        | Tybic Paleudults           | Xanthic Alisols (Clayic, Cutanic, Differentic)      |
| ZJJ05   | Bt      | 100–165    | 484          | HH-MS     | Humic Ferri-Udic Argosols        | Tybic Paleudults           | Xanthic Luvicosols (Cutanic, Differentic, Hyperpyric, Loamic) |

the Caledonian, Indosinian, and Yanshanian ages. The primary minerals in GR include feldspar, quartz, mica, and amphibole. SS are formed from moderately metamorphic slate and sedimentary shale. The main slate rocks are argillaceous slate, siliceous slate, and silt slate. Shale is mainly composed of carbonaceous slate, silt shale, and sandy shale. The LS in this region is mainly composed of marine sedimentary carbonate rocks, and its stratigraphic chronology mainly runs from the Devonian to the Permian period. With respect to minerals, LS is mainly composed of calcite and often contains mixed penetrations (sand, clay, dolomite, and silica). Soils developed from QRC, which is formed from glaciofluvial deposits dissolved in the Quaternary interglacial period of the Cenozoic era, are usually sticky due to intense mineral weathering and heavy material illuviation. The SDS in the area mostly consists of marine sedimentary clastic rocks and mainly includes siltstone, sandstone (quartz sandstone, feldspar quartz sandstone, and argillaceous sandstone), and conglomerate. SDS contained minerals, including quartz sand, feldspar sand, and iron siliceous cement.

We referred to The Manual of Soil Description and Sampling for collecting and describing the soil samples. The soils were classified as Ferrosols, Argosols, Cambisols, or Primosols using the CST classification system; as Ultisols, Alfisols, Inceptisols, and Entisols using the ST; and as Acrisols, Lixisols, Alisols, Luvisols, Plinthosols, Umbriols, Cambisols, and Leptosols according to WRB. These soil classifications are consistent with those of previous studies that investigated correlations among the CST, ST, and WRB systems. To avoid human interference and fully reflect the nature of the soil and the inheritance of parent materials, only the weathering B horizons in the soil profiles were sampled.

**Soil mineralogy.** The collected soil samples (clay fraction < 2 μm) were characterised using an X-ray diffractometer (Model D/Max-rA; Rigaku, Tokyo, Japan) with the following parameters: radiation, Ni-filtered CuKα; voltage, 40 kV; current, 40 mA. The diaphragm system was set as follows: divergence slit (DS) = anti-scatter slit (RS) = 0.3 mm. It measured from 3° to 30° 2θ at a scan rate of 2° 2θ min⁻¹ and a step size of 0.02° 2θ. For preparing samples for orientation sampling, the clay fraction samples were air-dried (AD), saturated with ethylene–glycol (EG) at 70 °C for 3 h, and heated (HT) to 450 °C or 600 °C for 2.5 h. The X-ray patterns were analysed using XPowder, a software package for powder X-ray diffraction analysis. Kaolinite was identified based on the presence of 0.72 and 0.358 nm peaks after being AD and saturated with EG, and the 0.72 nm peak disappeared after heating to 600 °C. Then, illite was identified based on the presence of a 1.00 nm peak after being AD and saturated with EG, and the peak persisted after heating. Vermiculite and chlorite were
distinguished by heat treatment. Shrinkage of the 1.42 nm peak to 1.00–1.03 nm after heating indicated the presence of vermiculite. I/S was identified based on the presence of a 1.45–1.54 nm peak, which slightly expanded after saturation with EG and shifted to 1.00 nm after heating. I/V was determined based on an unaltered D1 zone at 1.00–1.42 nm and appeared in the D3 zone at 0.50–0.47 nm; in the D2 zone, it shifted to < 1.0 nm after heating. The relative content of clay minerals was calculated based on the height of the diffraction peak.

### Soil physicochemical properties

Soil total K content was determined after digesting the samples with solid NaOH in a silver crucible at 450 °C for 15 min, and then gradually increasing the temperature. The concentration of K was determined using a flame photometer. Soil texture was determined using the pipette method and classified using the United States Department of Agriculture (USDA) classification standards. Soil organic carbon (SOC) was measured using the K2Cr2O7 wet oxidation method. Soil pH was measured using a glass electrode placed in a solution of soil and distilled water (1:2.5). The cation exchange capacity (CEC) and exchangeable Ca, Mg, K, and Na were determined using 1 M NH4OAc at pH 7. In each sample, the Fed was extracted using sodium dithionite-citrate. The total concentration of Fe, Al, and Si was determined by treating samples with Li2CO3-H3BO3 after heating at 900 °C for 0.5 h, and then, the solution was analysed by inductively coupled plasma atomic emission spectrometry (ICP-AES) (Table 2). CEC/clay was calculated using the following equation:

\[
\text{CEC/clay (cmol kg}^{-1} \text{ clay) } = \frac{\text{CEC (by NH4OAC pH7) (cmol kg}^{-1} \text{ soil)} \times 1000}{\text{clay (g kg}^{-1})}
\]

### Statistical analysis

The data were processed and analysed using SPSS 22.0 (IBM, Chicago, IL, USA). Before performing one-way ANOVA, the physicochemical properties of soils derived from different parent materials were tested for normal distribution and variance homogeneity using Kolmogorov–Smirnov and Levene test, respectively. Pearson’s correlation coefficient was used to test the correlation between clay minerals and the physical and chemical properties of the soil. The least significant difference test was used to test for significance. Significant differences are denoted by *P < 0.05, and extremely significant differences are indicated with **P < 0.01. All figures were created using Origin 9.1 software.

### Results

Composition and relative content of clay minerals in different parent materials. The clay minerals in upland soils were composed of 1:1-type clay minerals kaolinite (0.71–0.73 nm, 0.35–0.36 nm), 2:1-type clay minerals illite (1.0–1.03 nm), vermiculite (1.42–1.49 nm shifting to 1.0–1.03 nm after heating), and mixed-layer minerals illite/vermiculite (I/V) (determined by the unchanged D1 zone at 1.00–1.42 nm, appeared in the D3 zone at 0.50–0.47 nm and shifted to < 1.0 nm in the D2 zone after heating), and illite/smectite (I/S) (1.45–1.66 nm shifted to 1.0–1.02 nm after heating) (Fig. 2). The I/S mixed-layer minerals were only revealed in GR, whereas the 2:1-type illite (20%) and vermiculite (16%) content in LS were higher than those in GR. The upland soils in LS contained a small amount of I/V mixed-layer minerals (10%). Kaolinite (39%), illite (28%),
and I/V mixed-layer minerals (26%) were predominant in QRC, whereas vermiculite was less dominant (4%). In SS, the kaolinite and illite content varied substantially. The kaolinite content (43%) in SS was higher than that in QRC, but the content of illite (20%) and mixed-layer minerals (24%) was lower. In SS, the relative content of

Figure 2. X-ray diffraction patterns of the representative upland soil profiles derived from different parent materials (K: kaolinite; I: illite; V: vermiculite; I/S: illite/smectite mixed-layer mineral; I/V: illite/vermiculite mixed-layer mineral; Q: quartz; AD: air-dried sample; EG: ethylene–glycol saturated sample; HT 450 °C: sample heated at 450 °C; HT 600 °C: sample heated at 600 °C; SS: slate and shale; LS: limestone; QRC: Quaternary red clay; GR: granite; SDS: sandstone).
vermiculite (13%) was lower than that of other clay minerals. In SDS, the content of 1:1-type kaolinite (21%) was the lowest, and the content of 2:1-type illite (34%) and vermiculite (22%) were the highest. The content of I/V mixed-layer minerals (23%) in SDS was relatively higher than that in GR and LS.

Differences in clay mineral composition and relative content among diverse pedogenic environments.

Correlation analysis revealed that clay mineral composition was correlated with elevation (Fig. 4). For QRC, the range of elevation was small, and the elevation difference was less than 70 m; therefore, no correlation analysis was conducted. Apart from QRC, the kaolinite content in other parent materials was positively correlated with elevation (SS: r = −0.68, LS: r = 0.66, GR: r = 0.65, SDS: r = 0.66), but an opposite trend was observed for 2:1-type illite (SS: r = 0.74, LS: r = 0.65, GR: r = 0.67, SDS: r = 0.60). Nevertheless, the magnitude of variation at distinct elevations differed among the parent materials. The change in the slope of clay minerals in LS (k_{Kao} = −0.13, k_{2:1-type} = 0.08) was higher than that in the other three parent materials. This indicated that with increasing elevation, the kaolinite content decreased, whereas the 2:1-type illite content increased. Compared to the clay minerals of other parent materials, the clay minerals of LS were more sensitive to variations in elevation.

In addition to the correlation between elevation and the composition and relative content of clay minerals, we also observed a correlation between topographic position and clay minerals (Fig. 5). By comparing the topographic position with the composition and content of clay minerals, we found that the samples with high quantities of mixed-layer minerals and 2:1-type clay minerals were mostly collected from the slope foot or toe, whereas soil samples with a high 1:1-type kaolinite content were mostly collected from the slope shoulder or crest.

Differences in the composition and relative content of clay minerals at different diagnostic horizons and soil types (i.e., soil orders).

We identified 60 soil profiles with four diagnostic horizons based on the CST, i.e., LAC-ferric horizon, argic horizon, cambic horizon, and plinthic horizon19. The 60 soil profiles were classified as Ferrosols, Argosols, Cambosols, and Primosols in CST19; as Ultisols, Alfisols, Inceptisols, and Entisols in ST18; and as Acrisols, Lixisols, Alisols, Luvisols, Plinthosols, Umbrisols, Cambisols, and Leptosols in WRB17. The clay mineral composition was significantly different among the diagnostic horizons and soil types (i.e., soil orders) (Figs. 6, 7). The dominant clay mineral in the LAC-ferric horizon was 1:1-type kaolinite (63%), followed by illite (17%), I/V mixed-layer minerals (25%), and a small amount of vermiculite (8%). In the plinthic horizon, the relative content of mixed-layer minerals, including I/V (27%) and I/S (20%) mixed-layer minerals, was the highest (47%). The plinthic horizon also contained a high percentage of kaolinite (29%) and illite (23%) and a small amount of vermiculite (2%). The differences in clay mineral composition were negligible in the argic and cambic horizons.

Among all the soil types (i.e., soil orders), the content of 1:1-type kaolinite was the highest in Ferrosols (63%), followed by 2:1 clay minerals (I + V: 25%) and I/V mixed-layer minerals (12%). In Primosols, the content of 2:1-type clay mineral (I + V: 46%) and I/V mixed-layer mineral (36%) were the highest, and the content of 1:1-type clay mineral kaolinite (18%) was the lowest. The variation in clay mineral composition in Argosols and Cambosols was negligible. However, the content of mixed-layer minerals (I/V + I/S) was the highest in Plinthic Alu-Udic Cambosols (40%). Similar results were obtained using the ST and WRB classification systems. The content of 1:1-type kaolinite was the highest in Ultisols (51%) and Acrisols (63%), and the lowest in Entisols (18%) and Leptosols (17%). Mixed-layer minerals (I/V + I/S) were predominant in Plinthudults (47%) and Plinthosols (41%). This indicated that the clay mineral composition and relative content have a strong effect on Ferrosols,
Correlation between clay minerals and soil properties. Correlations between the composition and relative content of clay minerals with soil properties—such as SOC, CEC, soil texture, and pH—were determined (Fig. 8). Pearson's correlation analysis showed that the content of 1:1-type kaolinite was positively correlated with the sand content ($r = 0.27, P < 0.05$), significantly and positively correlated with aluminium oxide and exchangeable Na ($r_{Al} = 0.53, P < 0.01; r_{Na} = 0.34, P < 0.01$), and significantly and negatively correlated with silt content, SOC, and silicon oxide ($r_{silt} = -0.53, P < 0.01; r_{SOC} = -0.43, P < 0.01; r_{Si} = -0.34, P < 0.01$). The content of 2:1-type illite was negatively correlated with aluminium oxide and exchangeable Na ($r_{Al} = -0.32, P < 0.05; r_{Na} = -0.30, P < 0.05$) and significantly and positively correlated with silt content ($r = 0.42, P < 0.01$). Vermiculite content was

Ultisols, and Acrisols (high development degree); Primosols, Entisols, and Leptosols (low development degree); and Plinthic Ali-Udic Cambosols, Plinthudults, and Plinthosols (strong redox status).

Figure 4. Relationships between relative content of clay minerals and elevation for the studied soils (a,b: slate and shale; c,d: limestone; e,f: granite; g,h: sandstone).
positively correlated with SOC and pH (r_{SOC} = 0.59, P < 0.01; r_{pH} = 0.35, P < 0.01) and negatively correlated with potassium oxide and exchangeable Na (r_{K} = -0.37, P < 0.01; r_{Na} = -0.40, P < 0.01). The I/V mixed-layer mineral content was positively correlated with silicon oxide (r = 0.50, P < 0.01) and negatively correlated with aluminium oxide and iron oxide (r_{Al} = -0.53, P < 0.01; r_{Fe} = -0.46, P < 0.01). Additionally, 1:1-type kaolinite was significantly negatively correlated with 2:1-type illite, vermiculite, and I/V mixed-layer minerals (r_{I} = -0.74, P < 0.01; r_{V} = -0.47, P < 0.01; r_{I/V} = -0.62, P < 0.01). Illite showed a positive relationship with I/V mixed-layer minerals (r = 0.28, P < 0.05).

Pearson's correlations between the clay minerals and CEC/clay and Fe_d are shown in Table 3. Kaolinite was negatively correlated with CEC/clay and positively and significantly correlated with Fe_d, whereas 2:1-type clay minerals (illite and vermiculite) were negatively correlated with CEC/clay and positively correlated with Fe_d.

**Discussion**

**Effects of parent materials on the composition and relative content of clay minerals.** The type of parent material and the degree to which it is weathered have a major influence on the neoformation and transformation of clay minerals. These factors can result in variations in clay mineral composition and content in the same area due to differences in the mineral composition and texture of parent materials. GR is mainly...
Figure 7. Relative contents of clay minerals of different soil types (i.e., soil orders) (Kao: kaolinite; It: illite; V: vermiculite; I/S: illite/smectite mixed-layer mineral; I/V: illite/vermiculite mixed-layer mineral; CST: Chinese Soil Taxonomy; ST: Soil Taxonomy; WRB: World Reference Base for Soil Resources).

Figure 8. Correlativity of the clay minerals with some physico-chemical properties in studied soils.
composed of feldspar, quartz, biotite, and other primary minerals\textsuperscript{27}, and in the present study, the content of sand and K\textsubscript{2}O in GR was significantly higher than that in the other four types of parent materials (Table 2). Under acidic and strong leaching conditions, feldspar intensely weathers and hydrolyses, and kaolinite can be mass produced by neoformation\textsuperscript{37}. In addition, under conditions of high K content, vermiculite can be inhibited, and biotite can be oxidised and directly weathered to kaolinite\textsuperscript{38,39}. Therefore, the kaolinite content in the GR group was generally high (Fig. 3). QRC is composed of Quaternary fluvioglacial sediments, and during development, it is subjected to both sedimentary-weathering soil-forming processes and the glacial-interglacial cycle\textsuperscript{40}. Unstable factors in the sedimentary-weathering soil-forming process, strong cyclic climates, and environments that alternate between dry and wet seasons can promote illite alteration, a phenomenon that involves the transformation from single phase to mixed-layer minerals\textsuperscript{16,41,42}. Thus, in the present study, the content of mixed-layer minerals in the QRC group was generally high.

Previous studies have shown that the plinthic horizon of QRC was formed on flat, low-lying terrains in a climate that frequently alternated between dry and wet\textsuperscript{25}. Due to frequent fluctuations in groundwater, soil aggregates shrink when dry, a phenomenon that is conducive to the formation of cracks\textsuperscript{43}. When the groundwater level rises, the soil is in a reducing state in which Fe\textsuperscript{3+} can be transformed into soluble and mobile Fe\textsuperscript{2+}, thus promoting the leaching of Fe oxide from the soil. This process results in the formation of the plinthic horizon, which consists of uniform red soils with white veins and white spots\textsuperscript{44,45}. Smectite is generally formed in tropical and subtropical areas with dry and wet seasons, flat terrain, and poor drainage\textsuperscript{46}. In wet climates with poor drainage conditions, illite transforms into smectite by absorbing Mg\textsuperscript{2+}, but in dry conditions, K is fixed in the layer of smectite and promotes the formation of I/S mixed-layer minerals\textsuperscript{5,47}. Therefore, the environment is conducive to the transformation of illite to smectite, and the I/S mixed-layer minerals were found in the plinthic horizon.

In the present study, SDS was mainly composed of quartz sand, feldspar sand, and other iron and siliceous cements. The soils developed from SDS are characterised by high gravel content and low degree of development, and they occur at high altitudes (> 300 m)\textsuperscript{27}. Therefore, the contents of 2:1-type clay minerals and mixed-layer minerals were the highest in SDS. LS is mainly composed of marine sedimentary carbonate rocks, and its stratigraphic chronology mainly runs from the Devonian to the Permian period\textsuperscript{27}. The soils derived from LS are old and exhibit a high degree of weathering. Therefore, 1:1-type kaolinite was the main clay mineral in the soils derived from LS. The relative content of kaolinite in LS was lower than that in GR because of its high viscosity and weak leaching in LS.

The physical and chemical properties of soils developed from SS differ because the SS in the study area is composed of two kinds of parent rocks, i.e., low-grade metamorphic rock slate and sedimentary shale and as their components are relatively complex\textsuperscript{27}. Leaching was strong in soils developed from sandy slate with a low pH and high silt content, and therefore, the content of 1:1-type kaolinite was high in these soils. However, leaching was weak in soils derived from clay shale with a high pH and high clay content, and therefore, the content of 2:1-type clay minerals and mixed-layer minerals was high in these soils\textsuperscript{48}.

**Table 3.** Correlation coefficients between the clay minerals and CEC/clay and Fe\textsubscript{2+}. *Significant at the 0.05 level. **Significant at the 0.01 level.

| Parent material | Clay minerals | CEC/clay (cmolc kg\textsuperscript{-1} clay) | Fe\textsubscript{2+} (g kg\textsuperscript{-1}) | R     | P     | R     | P     |
|-----------------|---------------|-------------------------------------------|-----------------------------------------------|-------|-------|-------|-------|
| SS              | Kaolinite     | − 0.638*                                  | 0.026                                         | 0.626* | 0.030 |
|                 | 2:1-type      | 0.614*                                    | 0.034                                         | − 0.369 | 0.238 |
| QRC             | Kaolinite     | − 0.627*                                  | 0.029                                         | 0.642* | 0.024 |
|                 | 2:1-type      | 0.370                                      | 0.236                                         | − 0.370 | 0.236 |
| GR              | Kaolinite     | − 0.580*                                  | 0.048                                         | 0.593* | 0.042 |
|                 | 2:1-type      | 0.596*                                    | 0.041                                         | − 0.567 | 0.055 |
| LS              | Kaolinite     | − 0.722**                                 | 0.008                                         | 0.582* | 0.047 |
|                 | 2:1-type      | 0.767**                                   | 0.004                                         | − 0.358 | 0.253 |
| SDS             | Kaolinite     | − 0.684*                                  | 0.014                                         | 0.604* | 0.038 |
|                 | 2:1-type      | 0.513                                     | 0.088                                         | − 0.149 | 0.644 |

**Effects of pedogenic environment on the composition and relative content of clay minerals.** Warm and humid climatic conditions in subtropical regions promote soil weathering and development, and the eluviation and deposition of materials result in the development of acidic, fine-grained, strong-weathering soils characterised by desilication and ferrallitisation\textsuperscript{49,50}. These environmental factors mainly contributed to the development of 1:1-type clay mineral kaolinite, and to a certain extent, 2:1-type illite and vermiculite. The mixed-layer minerals were dominated by I/V (Fig. 3). These results were consistent with those of studies on upland soil clay mineral characteristics in other regions in southern China\textsuperscript{4,10,51}.

The regional pedogenic environments, especially the elevation and terrain, influence the transformation of clay minerals through temperature and precipitation or via the changes in hydrologic and thermal conditions. In high-altitude conditions with low temperatures, mica primary minerals can form a large amount of illite through depotassication in the mineral interlayer due to weak weathering\textsuperscript{32}. In weak acidic and strong leaching
illite were generally found in soil types with less intense weathering, such as Alfisols, Mollisols, and Aridisols. Clay minerals (smectite) were mainly found in the Vertisols that shrink and expand. Conversely, vermiculite and ST system, 1:1 clay minerals were mostly found in ultisols and oxisols with a high degree of weathering, and 2:1 clay minerals (illite) were often found in soils derived from LS, PL, and YS, where the weathering intensity was low. In the present study, the content of 1:1-type kaolinite in soils derived from LS, GR, SS, and SDS decreased with increasing elevation, whereas the content of 2:1-type clay minerals showed an opposite trend (Fig. 4). These results were similar to those observed for low mountains and hills (< 1500 m) but different from those found for subalpine or alpine zones (> 1500 m). However, the varying gradient of clay mineral content with changing elevation differed in diverse climatic zones, even in the same low mountain/hill area. Based on the findings of previous studies on clay minerals in different climatic regions, we established a series of regression models to analyse the influence of elevation on the composition and relative content of clay minerals (Table 4). In the present study, the decline in kaolinite content (k = −0.09) with elevation was greater than that recorded in tropical (k = −0.03) and temperate oceanic regions (k = −0.04). A possible reason for such a result is the relatively stable environmental and climatic conditions that exist in these two regions, which are responsible for the relatively slow material migration and mineral transformation in the soil.

Nevertheless, the magnitude of variation of clay minerals content in different elevations varies with the type of parent materials. For LS, its weak alkalinity (pH: 5.5 ± 0.5), high clay content (clay: 650 ± 152 g kg⁻¹), and slight weathering of minerals at high altitudes were conducive to the transformation of the primary mineral to illite via damouritisation and sericitisation. At low altitudes, the high temperature and old stratigraphic chronology of LS (Devonian to Permian) will facilitate the transformation of 2:1-type clay minerals into 1:1-type kaolinite through neoformation or mineral degradation. Therefore, the sensitivity of clay minerals in LS with the variation of altitude was greater than that of the other parent materials.

Micro-topography can affect the transformation of clay minerals in the subsurface horizon by redistributing soil hydrological conditions in the upland soils of the Hunan Province (Fig. 5). The present study showed that a wide distribution of kaolinite in the slope shoulder or crest soils also indicates strong leaching conditions, whereas a high content of 2:1-type clay minerals (illite, vermiculite) and mixed-layer minerals in the slope foot or toe soils suggests frequent wet–dry cycles and weak leaching conditions. These findings were similar to those obtained by Fang et al., who reported that kaolinite was most frequently found in divergent-site surface soils, whereas 2:1-type clay minerals mainly existed in convergent-site (water-collecting) surface soils. However, the present study showed that the influence of micro-topographical features on the transformation of soil clay minerals could extend to the subsoil.

**Dynamic changes in clay mineral composition among the different soil types (i.e., soil orders).** Soil classification systems, especially quantitative classification systems, such as the WRB, ST, and CST, are based on the theory of pedogenesis, diagnostic horizons, and diagnostic characteristics. Therefore, different soil types can often reflect different pedogenic processes and development stages. Clay minerals develop from primary minerals that have undergone weathering and pedogenesis during soil development. Their composition and relative content can reflect the strength of soil weathering and changes in the soil-forming environment. As the degree of pedogenesis increases (i.e., the CEC/clay content decreases and Fe₂O₃ content increases), the kaolinite content increases, whereas the content of 2:1 clay minerals decreases (Table 3). In the ST system, 1:1 clay minerals were mostly found in ultisols and oxisols with a high degree of weathering, and 2:1 clay minerals (smectite) were mainly found in the Vertisols that shrink and expand. Conversely, vermiculite and illite were generally found in soil types with less intense weathering, such as Alfisols, Mollisols, and Aridisols. Upon using the WRB, the 1:1-type kaolinite, which has low activity, was identified as the main clay mineral type in Ferralsols, whereas 2:1 clay minerals (such as chlorite, smectite, and vermiculite) were mostly found in Luvisols with a high activity. In the present study, the content of 1:1-type kaolinite was the highest in Ferrosols (CST), Ultisols (ST), and Acrisols (WRB) with moderate ferrallitisation and strong weathering. In contrast, the content of 2:1 clay mineral was the highest in Primosols (CST), Entisols (ST), and Leptosols (WRB) with slight siallitisation and weak weathering, and the content of mixed-layer mineral was the highest in Plinthic All-Udic Cambosols (CST), Plinthudults (ST), and Plinthosols (WRB) with a strong redox status (Figs. 6, 9).
**Conclusions**

The clay mineral composition at the study sites is representative of the upland soils in Hunan Province. Clay minerals mainly included 1:1 kaolinite, which contains a certain amount of 2:1-type clay minerals such as illite and vermiculite and a small amount of mixed-layer minerals. Our results suggested that the parent material, regional pedogenic environment (elevation and micro-topography), and degree of pedogenesis (soil type) play important roles in the transformation of clay minerals. Kaolinite dominated the mineral distribution patterns in GR (sandy) and LS (old), whereas 2:1-type clay minerals were dominant in SDS (clastic). The content of mixed-layer minerals was high in QRC (glacial-interglacial cycles and wet-dry cycles). In low mountains and hills (<1500 m), neoformation and transformation of clay minerals were determined by elevation and micro-topography. The influence of elevation on clay mineral transformation in the subtropical monsoon region is greater than that in tropical and temperate oceanic regions. Compared with the parent material, elevation showed a greater effect on the transformation of clay minerals in LS. Micro-topography can modulate the transformation of clay minerals in the subsurface horizon. The composition and relative content of clay minerals can efficiently indicate soil types with a high development degree (Ferrosols, Ultisols, and Acrisols), those with a low development degree (Primosols, Entisols, and Leptosols), and those with a strong redox status (Plinthic Ali-Udic Cambosols, Plinthudults, and Plinthosols).

The factors influencing clay mineral composition are complex and diverse. In the present study, only parent material, climate, and micro-topography were investigated, whereas the effects of vegetation type and pedogenic time were not explored. Future research should focus on these important factors to elucidate the mechanisms underlying clay mineral neoformation and transformation and to assess their significance in pedogenesis.

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