Goldschmidt 2026 Abstract
https://doi.org/10.46427/gold2026.36647

Fraction-Specific Mineralogical and Geochemical Variability of Tropical Soils in the Critical Zone: Controls on High-Resolution Soil Discrimination

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Since 2021, the Forenso Project (https://forenso.uff.br/) has been developing a geochemical and mineralogical database of soils from the Baixada Fluminense region (Rio de Janeiro, Brazil) to support forensic investigations while also providing a framework to examine small-scale variability in highly weathered tropical environments. This region is among the most violent in the country (35 murders per 100,000 inhabitants), approximately 70% of which remain unsolved. The project currently comprises 87 sampling sites distributed over a 500 km² area and includes topsoil samples representing diverse soil classes, geological settings, relief conditions, and crime indicators. Geochemical (XRF), mineralogical (XRD Rietveld), and FTIR analyses were conducted on the silt–clay fraction and, at selected sites, on the clay fraction to compare their discriminatory potential. Six sites separated by distances ranging from 0.5 to 13 km were selected for detailed evaluation. The silt–clay fraction revealed two statistically distinct groups: (a) a primary mineral–dominated group, characterized by feldspars (8–36 wt%), quartz (18–33 wt%), kaolinite (36–60 wt%), and minor amounts of mica and goethite; and (b) a secondary mineral–dominated group, defined by higher kaolinite contents (>60 wt%) and goethite (6–13 wt%), associated with elevated Fe_2O_3 contents (>10 wt%). The clay fraction exhibits high kaolinite abundance (>85 wt%) and three distinct groups defined by the kaolinite crystallinity index and by the presence or absence of mica, goethite, and anatase. FTIR spectra of both fractions are generally similar; however, the 1100–900 cm^{-1} region of the silt–clay fraction provides the greatest discriminatory potential, reflecting differences in the relative proportions of kaolinite, quartz, and feldspars. In contrast, variability within the clay fraction is mainly associated with iron chemical removal and corresponding spectral changes related to goethite, amorphous Fe phases, and structural Fe incorporated into kaolinite. Despite offering additional discriminatory variables, the clay fraction is not always sufficient to distinguish geographically close samples and requires substantially greater analytical effort. Therefore, the silt–clay fraction captures most of the small-scale geochemical variability, highlighting its relevance for high-resolution studies of tropical Critical Zone processes and for discriminating nearby soil samples in forensic applications.