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## Rapid Clay Mineralogical Analysis for Forensic Science Investigation—Clay Mineralogy Over the Short Distances

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**ABSTRACT:** Rapid clay mineralogical analysis using a particle size fraction less than 0.05 mm without extraction of a clay fraction (<0.002 mm) is applied to granitic soil samples collected from an area within a range of about 25 km with variation of topography. This area is located in a lucidophyllus forest climatic zone corresponding to a zone of yellow-brown forest soils. Clay mineralogical compositions obtained by this method showed different characteristics closely related to the topographic positions from where the soil samples were collected. It was confirmed that this method is effective for the discrimination of soil samples, and further for determining the original site of the soil samples as follows. In soils of hilly regions under well drained conditions, vermiculite is mixed with a large amount of gibbsite except for soil on flat places on ridges, which is characterized by the presence of a large amount of kaolin with a smaller amount of gibbsite. Soils under wet conditions such as valley-bottom plain and alluvial plain are characterized by interstratified clay minerals of chlorite and chloritized vermiculite. They are discriminated from each other in that the former are more abundant in gibbsite and the latter contain smectite.

**KEYWORDS:** forensic science, soils, minerals, soil discrimination, clay minerals, granite, topography, X-ray diffractometry, infrared absorption spectrometry

As minerals are major components of soils except in special cases such as peat soils, mineralogical examination is the most important technique for forensic soil discrimination. Soil minerals are divided into primary minerals and secondary ones. The primary minerals indicate rock forming minerals derived from parent rocks or materials (geological origin). The secondary ones are newly formed through soil forming processes (pedological origin). The former are contained mainly in coarser (sand and silt, > 0.002-mm) fractions, the latter in the finer (clay, < 0.002-mm) fractions. For forensic soil discrimination it is necessary to compare the mineralogical compositions in both mineralogical fractions between soil samples. However, the traditional technique used in forensic science laboratories has been restricted to petrological examination. Only slight consideration has been paid to clay mineralogical analysis by a few investigators [1,2].

The amount of soil samples obtained in criminal investigations is often so small that it is

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rare to obtain enough of the clay fraction which is generally used in clay mineralogical analysis. Concerning this problem, the authors have proposed to apply a rapid clay mineralogical analysis in which a particle size fraction less than 0.05 mm (<0.05-mm fraction) is used. The <0.05-mm fraction, which is the residue after extraction of sand samples (0.05 to 0.2 mm) for petrological examination by polarized microscope, is often discarded in many forensic science laboratories. In the previous paper [3], it was shown that the results obtained by the rapid method were in good agreement with those of the usual clay mineralogical analysis using the clay fraction. It has not been confirmed, however, whether the clay mineralogical analysis is applicable to case studies in forensic science investigations. Actually, in many cases soil samples are collected at a distance of several kilometres or hundreds of metres. Frequently, soil minerals show homogeneity over several kilometres or more, but in a region where the landscape factor such as topography or humidity varies, clay mineralogical composition also varies, even though the soils are formed from the same parent materials.

In this paper, validity of clay mineralogical analysis for forensic soil comparison over relatively short distances was investigated. An area was chosen where granite is widely distributed, and consequently, soil discrimination is difficult when the petrological examination of primary minerals is only used. Soil samples were collected from sites with different topographical conditions. The rapid clay mineralogical analysis was applied to the soil samples, and the clay mineralogical compositions of the samples were compared with each other.

## Materials and Methods

### *Soil Samples*

Large-scale intrusion of granitic rocks occurring during the Hiroshima disturbance in the Cretaceous are widely distributed in "Chugoku (Central) Provinces," the western part of the Main Island of Japan. The area chosen in this study is a basin of the Saba River in a range about 25 km from north to south, covering Hofu City and its adjacent region of Yamaguchi Prefecture. Location of sampling sites and topography are shown in Figs. 1 and 2. As the Saba River basin is surrounded by granite hills, soil covering here is formed from granite or its alterations. The bioclimatic condition of the area belongs to the warm-temperate lucidophyllus forest climate under which yellow-brown forest soil as a zonal genetic soil type in Japan is widely distributed. Climatic data are shown in Table 1.

Sampling Sites G2 to G6 are located on a hill called Shiroishi-yama at about 600-m altitude covered with lucidophyllus forest. G2 is a site on a northern foot of the hill. G3 is situated on a downslope where the moisture condition is wet from percolating water from the upslope. G4 is on a flat place similar to a terrace on the ridge running toward the north under well drained conditions. G5 is on a summit of the hill, and G6 is on a southfacing slope. The sampling sites except G3 are all under well drained conditions.

Sampling Sites G1 and G7 are located on narrow valley-bottom plains incised in the hilly region, Shiroishi-yama, running to the north from the hill, and eroding the southern foot of the hill to the east, respectively.

Sampling Sites G8 and G9 are located on an alluvial plain formed by fluvial sedimentation after the Jomon transgression about 6000 years ago, but G9 is on land reclaimed about 150 years ago at the issue of the Saba River. The sampling sites on the plains including the valley-bottom plains (G1, G7 to G9) are paddy fields under wet conditions.

All soil samples were collected from the surface horizon.

### *Identification of Clay Minerals*

The <0.05-mm fractions were prepared according to the procedure described in the previous paper [3] followed by X-ray diffractometry (XRD) and infrared absorption spectrometry.

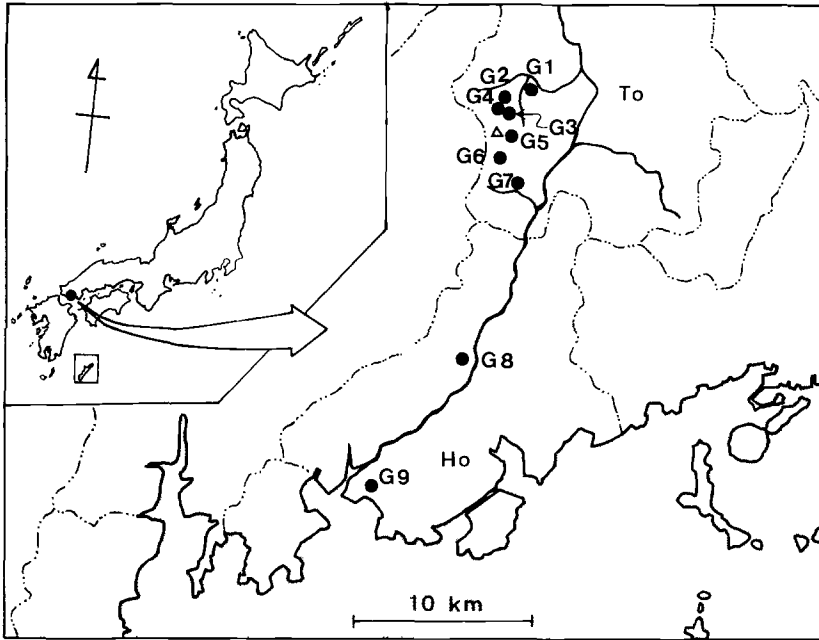


FIG. 1—Sampling sites of soil samples (G1–G9). Ho: Hofu-shi and To: Tokuji-cho, Yamaguchi Prefecture.

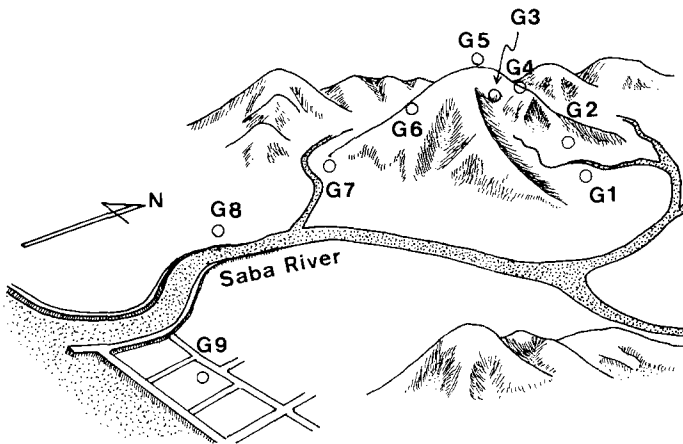


FIG. 2—Topography of the sampling sites of the soil samples. Altitude: G1, 220 m; G2, 260 m; G3, 420 m; G4, 460 m; G5, 600 m; G6, 440 m; G7, 280 m; G8, 20 m; and G9, 6 m.

try (IR). Thermal analysis was excluded in this study, since it requires relatively large amounts of samples.

XRD was carried out with a Rigaku-Denki Geigerflex X-ray diffractometer using a nickel filtered copper  $K\alpha$  on the oriented specimens of air-dried magnesium saturated clay (Mg-clay), Mg-clay solvated with glycerol, air-dried potassium saturated clay (K-clay), heated K-clay at 300°C, and heated K-clay at 600°C. When the clay films of the oriented specimens

TABLE 1—*Climatic data of the examined area.*

	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.	Annual
Temperature, °C <sup>a</sup>	3.8	4.3	7.4	13.3	17.8	21.6	25.6	26.3	22.5	16.4	10.7	5.5	14.6
Precipitation, mm	77	89	134	210	167	266	302	175	162	101	75	57	1816

<sup>a</sup>Mean temperature at Yamaguchi Meteorological Station during 1951 to 1980.

peeled off during heat treatment, they were fixed by spraying collodion solution (10% in isoamylacetate). Clay minerals were identified on the basis of the appendix in the previous paper [3].

IR was carried out using a Jasco Model A-701 infrared absorption spectrometer equipped with a beam condenser on micro discs (2 mm $\phi$ ) prepared from a mixture of the <0.05-mm fraction and cesium iodide with lead spacers. IR spectra were available to identify kaolin minerals and gibbsite, which show sharp doublet absorption bands in the OH stretching region, 3700·3620<sup>-1</sup> cm and 3530·3450 cm<sup>-1</sup>, respectively.

Abbreviations of clay minerals used in this paper are shown in Table 2.

## Results

Figure 3 shows the results of XRD for the granite soil samples. Major clay minerals in the area examined are Al whose Mg-clay shows 1.4-nm basal spacing, progressively collapsing toward 1.0 nm by heating, and Kn whose basal spacing shows 0.7 nm and disappears when the specimens are heated at 600°C. Ch and Mc, whose basal spacings are, respectively, 1.4 and 1.0 nm, never shift by any treatment, but the basal spacing of Ch shows a tendency to be stronger by heating at 600°C. They also exist generally in all soil samples, but their amounts are not as large. In the XRD patterns of G2 and G4 to G6, increase of the intensity of 1.0-nm basal spacing by potassium saturation accompanied by diminution of the intensity of 1.4-nm basal spacing shows contraction of the basal spacing of Vr by potassium fixation. The 1.2-nm basal spacing observed in heated K-clay specimens at 600°C of Sites G1, G3, and G7 to G9 is due to contraction of the basal spacing of randomly interstratified clay minerals of Ch and Al (Ch/Al) by heating, which behaves similarly to that of Al, except the contraction from 1.4 to 1.0 nm by heating is incomplete showing the intermediate 1.2-nm basal spacing. The 1.8-nm basal spacing observed in glycerol solvated Mg-clay specimens of G8 and G9 is due to swelling of the 1.4-nm basal spacing of Sm by solvating treatment.

Figure 4 shows the IR spectra in the OH stretching band. Two couples of sharp doublet absorption bands, respectively 3700·3620 cm<sup>-1</sup> of Kn and 3530·3450 cm<sup>-1</sup> of Gb, are observed in the spectra. IR spectra obtained here are divided into four types that consist of: (1) very strong doublet of Kn with very weak one of Gb (G4), (2) strong doublet of Kn with remarkable one of Gb (G2, G5, and G6), (3) strong doublet of Gb and weak one of Kn (G1, G3, and G7), and (4) weak doublet of Kn with very weak one of Gb (G8 and G9).

## Discussion

Both chloritized vermiculite and smectite, which have aluminum ions in their interlayer region, were represented as Al in this study, because the treatment for aluminum removal [4] was eliminated for simplification of analytical procedure. The Al observed here is presumed

TABLE 2—Abbreviations of clay minerals.

Clay Mineral	Abbreviation	Clay Mineral	Abbreviation
Chlorite	Ch	chloritized smectite	
Smectite	Sm	or vermiculite <sup>b</sup>	Al
Vermiculite	Vr	interstratified mineral	
Mica	Mc	of Ch and Al	Ch/Al
Kaolin minerals <sup>a</sup>	Kn	interstratified mineral	
Gibbsite	Gb	of Mc and Al	Mc/Al

<sup>a</sup>Metahalloysite or kaolinite.

<sup>b</sup>Aluminum interlayered clay minerals.

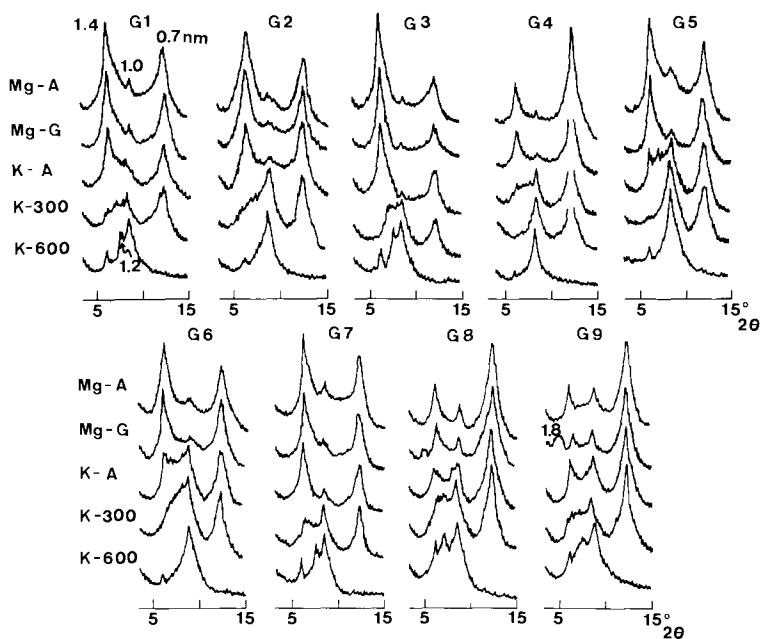


FIG. 3—X-ray diffractograms of the granitic soil samples. Mg-A, Mg-clay air-dried; Mg-G, Mg-clay solvated with glycerol; K-A, K-clay air-dried; K-300, K-clay heated at 300°C; and K-600, K-clay heated at 600°C.

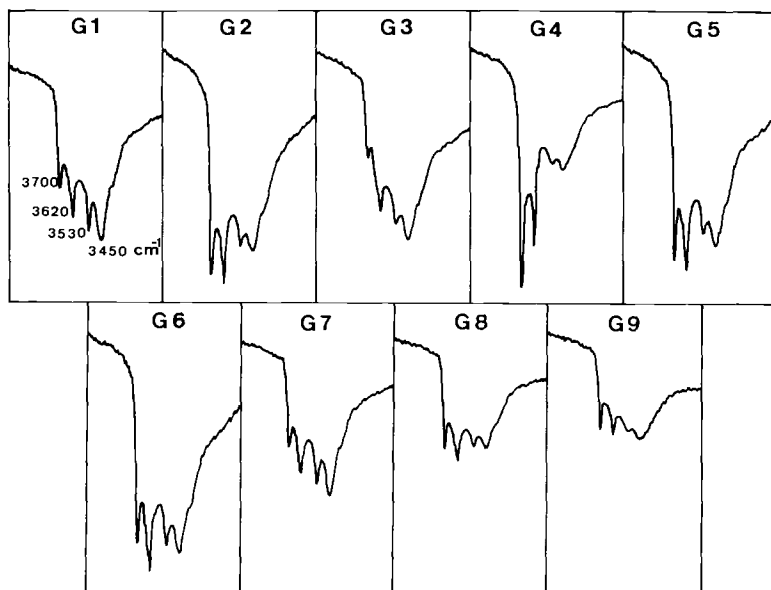


FIG. 4—IR spectra of the granitic soil samples. Absorption band: kaolin minerals, 3700 · 3620  $\text{cm}^{-1}$  and gibbsite, 3530 · 3450  $\text{cm}^{-1}$ .

to be chloritized Vr, because many investigators have reported its presence in granitic soils in Southwest Japan. Kato [5,6] reported chloritized Vr as trioctahedral aluminum-vermiculite in red soils of Aichi Prefecture, and Nagatsuka [7] and Iwasa [8] also reported it in yellow-brown forest soils of Mie Prefecture and Yamaguchi Prefecture, respectively. In granitic soils of Southwest Japan, chloritized Vr seems to be one of the common weathering products.

A very strange thing is observed in the examination of the soil samples from the alluvial plain. Whereas the XRD peak of Kn (0.7-nm basal spacing) is very strong, its absorption bands in IR spectra are very weak. A similar tendency is observed in the soil samples from the valley-bottom plains. Furthermore, this inconsistency has been observed in the author's other investigations of alluvial soils. This inconsistency between the results of IR and XRD, therefore, should be one of the important characteristics of alluvial soils to be distinguished from other soils such as residual soils. As apparent in XRD patterns, in fact, Kn is abundant in the alluvial soil samples, and the indistinctness of the absorption bands in IR spectra is probably a result to some extent of vacant OH in the crystal lattices. But its origin remains equivocal here. It is necessary to conduct more detailed study about crystalline structure to explain this fact.

Clay mineralogical compositions of the soil samples are summarized in Table 3. Although all of them originate from granite or its sediments, there are obvious differences among the samples.

Clay mineralogical characteristics of the samples from the hill under well drained conditions (G2, G4 to G6) are the presence of Vr with the lack of Ch/Al showing strong absorption bands of Kn and Gb in IR spectra. But G4, which is from a flat place similar to a terrace on the ridge, shows different characteristics of a very large amount of Kn with a very small amount of Gb, rather resembling red soil observed in Japan as a relic soil which had been derived from highly decomposed red weathering crusts under warmer climatic conditions during transgression in late Pleistocene [9]. Among the samples (G2, G5, and G6), Vr is more abundant in the upper part of the hill (G5 and G6) than the foot (G2).

The soil samples collected from the alluvial plain (G8 and G9) are characterized by the presence of Sm with abundant Kn, which shows only indistinct absorption bands in IR spectra, a relatively small amount of Al, and a very small amount or absence of Gb. Sm increases with the approach to the issue of the river.

TABLE 3—Clay mineralogical compositions of soil samples.<sup>a</sup>

Sample	Sm	Ch	Vr	Al	Al/Ch	Mc	Kn	Gb <sup>b</sup>
HILL								
G5(summit, 600 m) <sup>c</sup>		+	++	++		+—	++	+
G4(flat on ridge, 460 m)		+—	+	+		+—	+++	+—
G6(slope dry, 440 m)		+—	++	++		+—	++	+
G2(foot of hill, 260 m)		+—	+—	++		+—	++	++
G3(slope wet, 420 m)		+		++	+	+—	+	++
PLAIN								
G1(valley-bottom, 220 m)		+		++	+	+—	++	+
G7(valley-bottom, 280 m)		+		++	+	+—	++	+
G8(alluvial plain, 20 m)	+—	+		+	+	+	+++	+—
G9(alluvial plain, 6 m)	+	+—		+	+	+	+++	+—

<sup>a</sup>Abundance: + + +, abundant; + +, medium; +, little; and + —, very little.

<sup>b</sup>Identified by IR spectra.

<sup>c</sup>Topography and altitude.

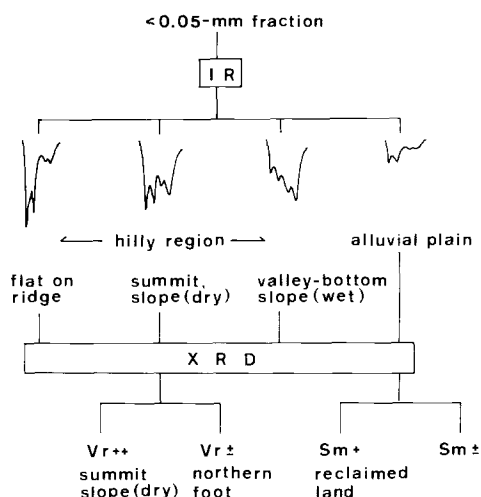


FIG. 5—Estimation of the sites where the soil samples originated.

The soil samples from the hilly region under wet conditions (G1, G3, and G7) show intermediate clay mineralogical compositions between the above regions, namely, a remarkable amount of Gb is present, Vr is absent, and Ch/Al is present.

As mentioned above, the clay mineralogical compositions of these soil samples show different characteristics closely related to topographic positions. Therefore, it is possible to discriminate soil samples and further to estimate the sites in the region from which they were taken as shown in Fig. 5. Primarily, IR is applied to the <0.05-mm fractions. According to the patterns of IR spectra, the soil samples are classified into four topographic regions. After that, XRD makes possible further detailed estimation of sites.

The above estimation is, of course, applicable only to the soil samples in granitic areas under warm-temperate lucidophyllus forest climate. However, if further research is conducted on various areas of different parent materials and different bioclimatic conditions, clay mineralogical analysis will be one of the most effective methods not only for forensic soil discrimination but also for estimation of the site where the soil sample originated.

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