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Article *in* Australian Journal of Soil Research · January 1998

DOI: 10.1071/S97084

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Aust. J. Soil Res., 1998, **36**, 411–21

Relationship between clay mineralogy and exchangeable Al in red and yellow soils from the Islands of Okinawa and Java

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Abstract

Red and yellow soils from tropical regions are generally more mature than their subtropical counterparts. Most of these soils contain kaolin as the dominant clay mineral. Exchangeable aluminium (Al) generally balances permanent negative charges and occupies strongly acidic exchange sites of the soil clay. The objective of this study was to identify those clay minerals that are most highly implicated in contributing exchangeable Al to red and yellow soils collected from the Islands of Okinawa and Java.

All soils exhibited an acid reaction but varied in their exchangeable Al content and clay mineralogy. Clay content was high in all Javan soils but varied in those from Okinawa. Javan soils were dominated by kaolinite, and Okinawan soils by an association of illite and halloysite. However, 2 : 1–2 : 1 : 1 intergrades were significant components in both the Oku red soils (Okinawa Island) and the Pamagersari red soils (Java Island). Javan soils were characterised by a more mature (advanced) state of weathering than those from Okinawa. The source of exchangeable Al was halloysite in Okinawan soils and 2 : 1–2 : 1 : 1 intergrades in Javan soils.

Additional keywords: 2 : 1–2 : 1 : 1 intergrade minerals, clay content, halloysite, kaolinite, weathering process.

Introduction

Red and yellow soils in the tropics differ from those in subtropical regions with respect to the maturity of soil development. In the tropics, the soils are clay-textured and acidic in nature, and the weathering process at its most mature stage results in clay minerals of the 1 : 1 group (Goenadi and Tan 1989; Hirai *et al.* 1991). Some 2 : 1 group minerals, however, may also occur in these soils (Bennema 1962).

The 2 : 1 type minerals possess a permanent negative charge resulting from isomorphic substitution in the octahedral (Mg^{2+} for Al^{3+}) and/or tetrahedral (Al^{3+} for Si^{4+}) sheet of the clay lattices. Under acidic conditions aluminium (Al) is mobilised and, having a positive charge, will be attracted to negatively charged sites in the clay structure to form exchangeable Al. Marshall (1949) and Pratt and Bair (1961) postulated that the permanent negative charge of clay minerals contributes to the occurrence of exchangeable Al in soils. Minerals of

0004-9573/98/030411$05.00

the 2 : 1 type also have an ability to fix Al in their interlayer space. Intercalation of Al in the interlayer region of swelling 2 : 1 silicates leads to the blocking of the interlayers and the resultant formation of 2 : 1–2 : 1 : 1 intergrades similar in characteristics to Al-chlorite (Dixon and Jackson 1962; Carstea *et al.* 1970). This type of interlayering is very common in most naturally acid soils, not only in those from the tropics (Uehara and Gillman 1981). Generally 2 : 1–2 : 1 : 1 intergrades constitute a resource of exchangeable Al in acid soils. Goenadi and Tan (1989) reported that Oxisols and Ultisols, which contain hydroxy-interlayered vermiculite, have a high exchangeable Al content.

Data from Hirai *et al.* (1991), however, show that red and yellow soils from subtropical regions that are dominated by kaolin also have a high exchangeable Al content. Shamshuddin and Ismail (1995) reported that Malaysian Ultisols, which contain kaolinite and mica-chlorite as the dominant clay mineral, had a higher content of exchangeable Al than Oxisols, which were dominated by kaolinite, gibbsite, and geothite. M. Nurcholis *et al.* (1997) have developed a simple method to predict the content of exchangeable Al in red-yellow soils with pH on a power regression. In that study interesting characteristics were displayed in the buffer curve, particularly in the relationship between exchangeable Al and clay content/clay mineralogy. It was the objective of this study to report those clay minerals that are associated with the occurrence of exchangeable Al in tropical and subtropical red-yellow soils.

Materials and methods

Samples of 5 red soils and 5 yellow soils formed under a subtropical climate were collected from Okinawa Island. According to the National Land Agency (1977), red and yellow soils in the northern part of Okinawa Island developed from sandstones or phyllites and slates. The soils collected for the present study were representative of this precursor material. Seven of the soils had developed from phyllitic parent rock, 2 soils (Sokonia and Sosu) from Kunigami gravel, and 1 (Arume) from sandstone (Table 1). Samples of 3 red soils and 2 yellow soils were collected from Java Island, which has a tropical climate. All Javan soils formed from volcanic material (Table 1). Dudal and Soepraptohardjo (1960) and Tan and Troth (1982) reported that soils from Java were generally influenced by volcanic activities, both past and recent. Surface (0–15 cm) as well as subsurface (20–35 cm) samples were collected at each site. Only samples that showed an acidic reaction were included in this study.

The soil samples were air-dried and ground to pass through a 2-mm sieve. Subsamples of soils were used to determine pH(H_2O) and exchangeable Al. The pH was measured in a 1 : 2·5 soil–solution suspension. Exchangeable Al was extracted with 1 M KCl (McLean 1965), and quantified colorimetrically using ferron (Davenport 1949). Soil texture was determined by mechanical analysis after removing the organic matter with hydrogen peroxide. The soil was dispersed ultrasonically and adjusted to pH 10 with NaOH; a clay fraction (<2 μm) was then syphoned after sand and silt fractions had settled.

The clay fractions were treated with citrate bicarbonate dithionite (CBD; (Mehra and Jackson 1960) and their mineral composition was determined by X-ray diffraction using an oriented specimen after a treatment involving saturation, solvation, and heat: Mg^{2+} saturation air-dry and glycerol-solvated; K^+ saturation air-dry and heated to 100, 300, and 550°C. Preliminary X-ray diffraction analysis showed peaks for neither chlorite nor smectite in all samples. Therefore, a semiquantitative analysis of the clay mineralogy was conducted by measuring the relative intensity (I) of each mineral species:

$$I_K = I_{7\mathring{A}(Mg)} - (1/8{\cdot}5)(I_{14\mathring{A}(Mg^{2+})}) \qquad (1)$$

Table 1. Locality, soil colour, parent material, and classification of the soils investigated

Location of soil samples	Soil colour	Parent material	US Soil Taxonomy
Subtropical region: Okinawa Island, Okinawa prefecture			
Red soils			
Oku, Kunigami village	2·5YR5/8	Phyllite	Typic Paleudults
Taira, Higashi village	2·5YR4/8	Phyllite	Typic Dystrochrepts
Arume, Higashi village	2·5YR4·8	Sandstone	Typic Paleudults
Sokonia, Nago city	2·5YR5/8	Kunigami gravel	Typic Hapludults
Toyohara, Ginoza village	2·5YR5/8	Phyllite	Typic Paleudults
Yellow soils			
Sosu, Kunigami village	7·5YR6/8	Kunigami gravel	Typic Dystrochrepts
Takae, Higashi village	10YR7/8	Phyllite	Typic Dystrochrepts
Miyagi, Higashi village	7·5YR7/6	Phyllite	Typic Hapludults
Teima, Nago city	7.5YR5/8	Phyllite	Typic Paleudults
Mahirabaru, Ginoza village	7·5YR7/8	Phyllite	Typic Paleudults
Tropical region: Java Island, West Java province, Bogor regency			
Red soils			
Pamagersari, Jasinga subdistrict	5YR4/6	Tuff (or claystone)	Typic Paleudults
Cigudeg, Cigudeg subdistrict	5YR4/6	Volcanic rock (?)	Typic Haplortoxs
Singajaya, Jonggol subdistrict	2·5YR5/6	Tuff	Tropeptic Eutrorthoxs
Yellow soils			
Cikopomayak, Jasinga subdistrict	7·5YR6/6	Tuff	Typic Paleudults
Singasari, Jonggol subdistrict	7·5YR6/6	Tuff	Dystropeptic Tropudults

where I_K is the relative intensity of kaolin and 1/8·5 is a correction factor for the intensity of the second peak of vermiculite (Wada 1966);

$$I_I = I_{10\text{Å}(Mg^{2+}\text{-gly})} \quad (2)$$

where I_I is the relative intensity of illite;

$$I_V = I_{10\text{Å}(K+)} - I_{10\text{Å}(Mg^{2+}\text{-gly})} \quad (3)$$

where I_V is the relative intensity of vermiculite;

$$I_{V\text{-}I} = I_{[10-14\text{Å}(Mg^{2+}\text{-gly})]} \quad (4)$$

where I_{V-I} is the intensity of the vermiculite/illite mixed layer and $I_{[10-14\text{Å}(Mg^{2+}\text{-gly})]}$ is the intensity of the peak between $I_{10\text{Å}}$ and $I_{14\text{Å}}$ in Mg^{2+}-gly;

$$I_{2:1-2:1:1} = I_{14\text{Å}(Mg^{2+}\text{-gly})} - I_V \quad (5)$$

where $I_{2:1-2:1:1}$ is the relative intensity of these intergrade minerals.

Relative amounts of phyllosilicates were calculated from the integrated intensity fraction (Harris *et al.* 1989)

$$I_a(\%) = I_a/(I_a + I_b + ...I_n) \times 100\% \quad (6)$$

where I is the peak intensity of each mineral type $(a, b, \ldots\ n)$.

To differentiate between halloysite and kaolinite, formamide was added to the air-dried Mg^{2+}-saturated clay specimen, which was then x-rayed after 30 min (Churchman *et al.* 1984).

Results and discussion

Physicochemical and mineralogical properties

Soils from Okinawa, both red and yellow, were characterised by a high degree of textural variability, with clay contents being highest in the red soil of Arume B (49·0%) and lowest in the yellow soil of Takae B (8·0%; Table 2). Generally illite and halloysite were the dominant clay minerals. Soils Oku A and B and Taira A and B were an exception. However, the Oku samples were dominated by 2:1–2:1:1 intergrades and the Taira samples by illite and kaolinite. The soil reaction was acidic in all samples with pH values ranging from 5·9 to 4·2. The highest content of exchangeable Al was recorded for the red soil of Arume B (52·49 mg/100 g clay), which contained halloysite as the dominant clay fraction, while the lowest content of exchangeable Al was found in the yellow soil of Takae A (0·53 mg/100 g clay), which was predominantly illitic.

All soils from Java had a pH <4·81 and a high clay content (Table 2). Kaolinite dominated the clay fraction in most samples except for the red soil of Pamagersari B, which was dominated by 2:1–2:1:1 intergrades. Exchangeable Al was highest in the red soil of Pamagersari B (214·83 mg/100 g clay), which had

Table 2. Selected textural, chemical, and mineralogical characteristics of the soils investigated

Based on the clay content: Vt, vermiculite; Int, 2:1–2:1:1 intergrade minerals; Vt/It, vermiculite/illite mixed layer minerals; It, illite; K, kaolin minerals; H, halloysite; Kt, kaolinite

Soil samples	Sand	Silt (%)	Clay	pH_{H_2O}	Exch. Al (mg/100 g clay)	Clay mineral associations Vt	Int	Vt/It	It	K
Oku A	9·4	45·6	45·0	4·21	23·98	3	18	6	6	12 (H)
Oku B	15·1	36·1	48·8	4·81	21·70	4	18	10	7	9 (H)
Taira A	26·0	54·6	19·4	4·51	4·25	0	0	0	10	10 (Kt)
Taira B	17·1	64·3	18·6	4·20	6·86	0	0	0	11	7 (Kt)
Arume A	30·3	24·0	45·7	4·21	47·90	3	2	7	16	17 (H)
Arume B	28·4	22·6	49·0	4·23	52·49	2	3	8	17	19 (H)
Sokonia B	42·6	21·9	35·5	5·74	3·60	5	4	5	10	12 (H)
Toyohara A	27·5	32·2	40·3	5·90	2·27	3	4	7	12	14 (H)
Sosu B	43·9	31·0	25·1	4·77	4·49	1	2	3	8	11 (H)
Takae A	59·4	25·3	15·3	5·10	0·53	0	0	0	9	7 (H)
Takae B	76·2	15·8	8·0	4·67	0·59	0	1	1	3	4 (H)
Miyagi A	36·8	36·1	27·1	4·49	13·55	1	0	1	16	9 (H)
Miyagi B	39·0	34·8	26·2	4·59	14·46	1	0	2	15	8 (H)
Teima A	37·7	31·0	31·3	5·29	17·19	0	2	6	13	11 (H)
Mahirabaru A	46·0	22·0	32·0	4·63	13·07	2	1	4	14	10 (H)
Mahirabaru B	49·2	22·0	28·8	4·33	12·78	2	0	3	14	9 (H)
Pamagersari A	7·1	16·2	76·7	4·62	130·98	12	50	0	5	10 (H)
Pamagersari B	8·2	13·6	78·2	4·60	214·83	6	56	0	6	10 (H)
Cigudeg A	5·7	7·1	87·2	4·54	38·34	0	5	0	0	82 (Kt)
Cigudeg B	6·0	7·1	86·9	4·56	45·94	0	4	0	0	83 (Kt)
Singajaya A	10·6	11·0	78·4	4·72	3·92	0	0	0	0	78 (Kt)
Singajaya B	10·7	10·6	78·7	4·81	6·37	0	3	0	0	75 (Kt)
Cikopomayak A	40·0	17·5	42·5	4·62	7·76	3	2	3	3	28 (Kt)
Cikopomayak B	37·3	17·0	45·7	4·52	19·71	4	1	4	3	32 (Kt)
Singasari A	23·6	21·8	54·6	4·62	22·60	5	14	0	3	33 (Kt)
Singasari B	19·4	21·1	59·5	4·43	64·59	4	15	0	4	37 (Kt)

a clay content of 78·3% and 2:1–2:1:1 intergrades dominating the clay fraction (56%). Lowest exchangeable Al levels were recorded for the red soil Singajaya A (3·92 mg/100 g clay), which had an almost identical clay content but was dominated by kaolinite (78%). In contrast to the Okinawa soils, exchangeable Al and clay content in the Java subsurface soils were higher than in the surface horizons (Table 2). These results are in line with findings by Goenadi and Tan (1989) and Funakawa (1993).

Generally speaking, kaolin minerals reflect an advanced stage of weathering. The 2:1–2:1:1 intergrades may form through Al interlayering and transform to 2:1–1:1 interstratifications (Wada and Kakuto 1983). This hypothesis is supported by Araki (1992), who regarded hydroxy-interlayered vermiculite as a transitional phase in the pedogenic transformation of 2:1 to 1:1 minerals.

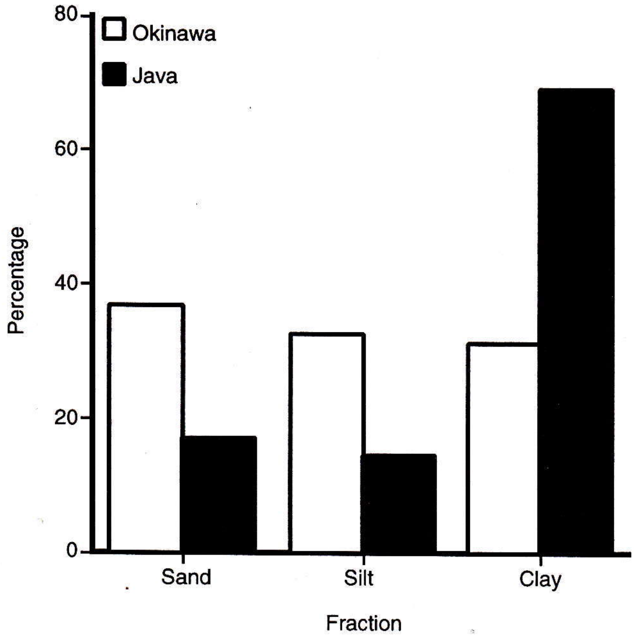

Fig. 1. Textural differences between Okinawa and Java soils.

The dominant clay mineral in tropical Javan soils is kaolinite, while the subtropical soils of Okinawa are dominated by an association of illite and halloysite (Table 2). Kaneko and Nagatsuka (1984) also described halloysite (7 Å) from red and yellow soils from Okinawa. Uehara and Gillman (1981) defined the stages of weathering as young, mature, and senile, based on the presence of the following minerals: rock-forming minerals, mica or montmorillonite, kaolinite or gibbsite, and gibbsite nodules or iron stone, respectively. A more highly weathered soil is also characterised by a higher clay content. Average particle size characteristics are presented in Fig. 1. Okinawan soils have approximately equal amounts of sand, silt, and clay particles. Javan soils, by contrast, are clay-textured. The weathering process of Okinawan soils is therefore inferred to be still close to the mature stage which shows illite preserved, while Javan soils are on the verge of reaching the senile stage, as indicated by the dominance of kaolinite and the very low illite content.

Relationship between clay mineralogy and exchangeable Al

Okinawan soils

In the present study a positive relationship between the amount of clay (X) and exchangeable Al (Y) has been established:

$$Y = -13 \cdot 14 + 0 \cdot 91X \qquad (r = 0 \cdot 722^{**}, \quad n = 16)$$

This relationship indicates that the permanent negative charge and the related number of strongly acidic exchange sites increases with the amount of clay, and that under strongly acidic conditions, Al may occupy these charged sites as well as exchange sites in an exchangeable form.

The relationship between the nature of the clay fraction and exchangeable Al content in Okinawan is shown in Table 3. Exchangeable Al increased with increasing amounts of kaolin minerals (significant at $P = 0 \cdot 01$) and illite and vermiculite/illite mixed-layer minerals (both significant at $P = 0 \cdot 05$), while no correlation could be established with vermiculite and 2:1–2:1:1 intergrades.

Table 3. Relationship between the nature of the clay fraction (X) and exchangeable Al (Y) in red and yellow soils from the Okinawa and Java Islands

Variable (X)	Okinawa ($n = 16$)	Java ($n = 10$)
Vermiculite	$Y = 3 \cdot 02X + 9 \cdot 91$ $r = 0 \cdot 308$	$Y = 11 \cdot 12X + 17 \cdot 78$ $r = 0 \cdot 625$
2:1–2:1:1 intergrade minerals	$Y = 0 \cdot 65X + 12 \cdot 76$ $r = 0 \cdot 247$	$Y = 3 \cdot 10X + 9 \cdot 09$ $r = 0 \cdot 948^{***}$
Vermiculite/illite mixed layer minerals	$Y = 3 \cdot 00X + 3 \cdot 18$ $r = 0 \cdot 615^{*}$	$Y = -14 \cdot 19X + 65 \cdot 53$ $r = -0 \cdot 313$
Illite	$Y = 1 \cdot 97X + 7 \cdot 26$ $r = 0 \cdot 512^{*}$	$Y = 22 \cdot 00X + 2 \cdot 66$ $r = 0 \cdot 739^{*}$
Kaolin minerals	$Y = 3 \cdot 05X + 17 \cdot 25$ $r = 0 \cdot 742^{**}$	$Y = -1 \cdot 37X + 119 \cdot 92$ $r = -0 \cdot 600$

$^{*}P < 0 \cdot 05$, $^{**}P < 0 \cdot 01$, $^{***}P < 0 \cdot 001$.

Halloysite, described by Yoshida (1979), had a CEC (m.e./100 g)* of 24, 75% of which was derived from strongly acidic exchange sites, which had the ability to adsorb Al in an exchangeable form when 0·33 M $AlCl^{3+}$ was added. Kaolinite has strongly acidic exchange sites, which amounted to 61% of the total CEC of 4·9 m.e./100 g. According to Zhou and Gunter (1992), the negative charge of kaolinite is associated not only with edge sites, but also with basal surfaces. Results from the present study demonstrate that the clay mineral which was most prominently associated with the occurrence of exchangeable Al in Okinawan soils was halloysite, although vermiculite/illite interstratifications and illite also contributed to exchangeable Al. In acid red and yellow soils from subtropical regions, Al was the dominant exchangeable cation blocking permanently charged sites (Hirai *et al.* 1991). These soils had clay fractions

* 1 m.e./100 g = 10 mmol(+)/kg

dominated by kaolin minerals, vermiculite, 2:1–2:1:1 intergrades, or vermiculite/illite interstratifications.

Present results show that halloysite contributed most significantly to the exchangeable Al pool, while the amount of clay was essential in providing the negative sites required for cation exchange.

Javan soils

Javan soils had a higher clay content than soils from Okinawa (Table 2), but no correlation could be established between clay (X) and exchangeable Al (Y) content:

$$Y = 29 \cdot 11 + 1 \cdot 23X \qquad (r = 0 \cdot 304, \quad n = 10)$$

The relationship between clay mineralogy and exchangeable Al content in Javan soils is presented in Table 3. A close relationship could be established between the amount of 2:1–2:1:1 intergrades and exchangeable Al (significant at $P = 0 \cdot 01$). Exchangeable Al was less significantly related to illite ($P = 0 \cdot 05$), while no relationship could be established with vermiculite, vermiculite/illite, and kaolin minerals.

In general, 2:1 clays have a high CEC and resultant strongly acidic exchange sites (Yoshida 1979) and may fix Al in their interlayer space (Shen and Rich 1962; Hsu and Bates 1964). This interlayer Al may then form hydroxy-Al islands (Dixon and Jackson 1962) or gibbsite layers (Barnhisel and Rich 1963). The amount of exchangeable Al decreases with increasing degree of interlayering (Matsue and Wada 1988). Results from this study show that 2:1–2:1:1 intergrades correlate most significantly with exchangeable Al (Table 3).

The following interpretation of these results is proposed. There was a considerable degree of variability in the negative charge and strongly acidic ionic exchange sites between the different soil samples, so that an increase in the clay content did not necessarily lead to an increase in the negative charge. The clay fractions of the red and yellow soils from Java (tropical region) were dominated by kaolinite, which contributed little to the exchangeable Al pool. In Okinawan soils (subtropical region), in contrast, halloysite (7Å) was the dominant clay mineral. This species possessed strongly acidic exchange sites in abundance.

Combination of Okinawan and Javan soils

The relationship between the nature of the clay fraction and exchangeable Al content is depicted in Fig. 2. A linear correlation was established for exchangeable Al and 2:1 minerals as well as 2:1–2:1:1 intergrades, which was significant at $P = 0 \cdot 01$. This relationship was particularly evident for Javan soils, where exchangeable Al was governed by 2:1–2:1:1 intergrades. The relationship was much less evident for Okinawa soils, where it was not only affected by the nature of the clay fraction but also by the clay content. In Javan soils, 2:1–2:1:1 intergrades contributed most to the exchangeable Al, while halloysite was most highly implicated in Okinawan soils (Table 3). In Javan as well as Okinawan soils, 2:1 phyllosilicates contributed to exchangeable Al.

From the results of the present study, it is concluded that minerals of 2:1–2:1:1 intergrades are clay minerals which may contribute to exchangeable Al in the

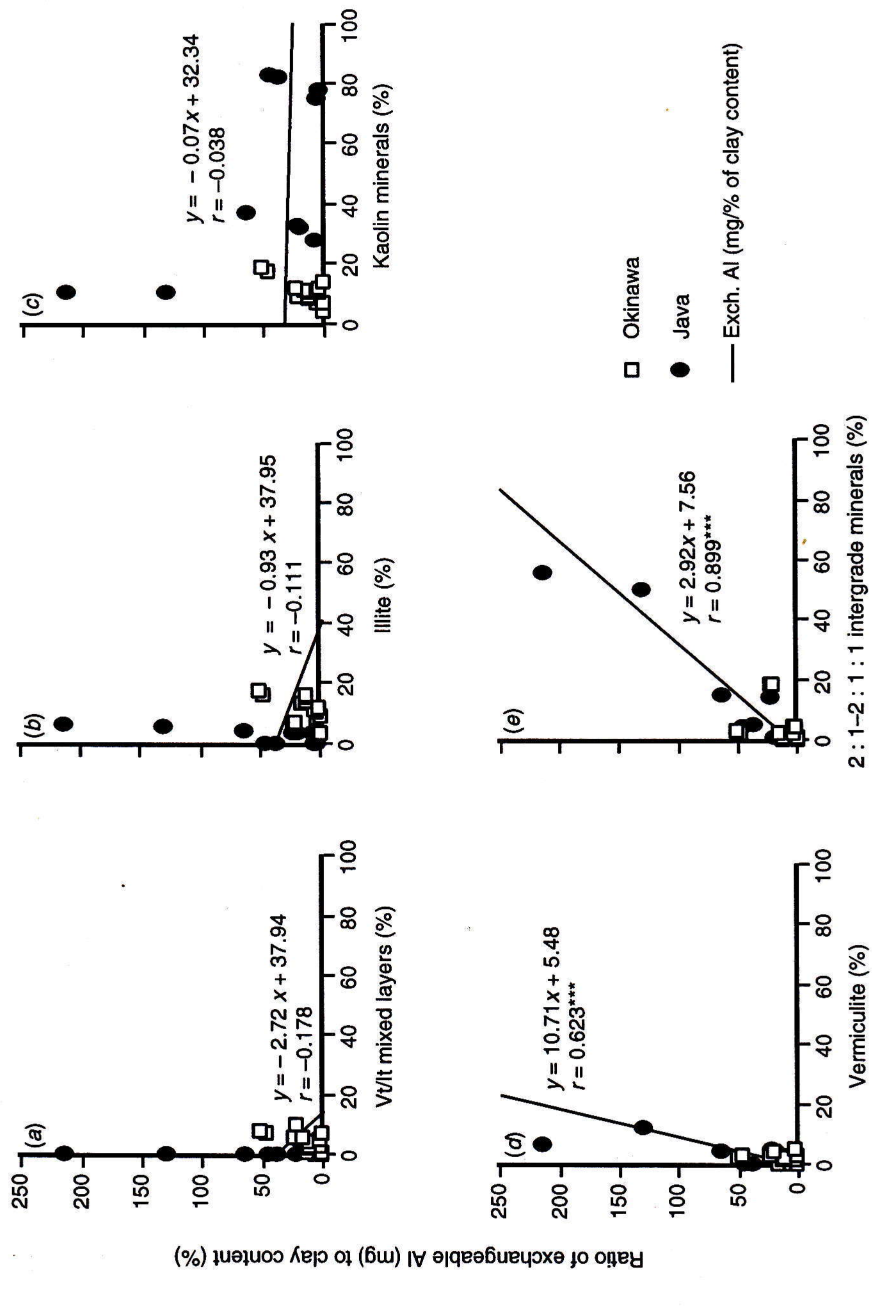

Fig. 2. Linear regressions of (*a*) vermiculite/illite mixed layer minerals, (*b*) illite, (*c*) kaolin minerals, (*d*) vermiculite, and (*e*) 2:1–2:1:1 intergrade minerals, and exchangeable Al in the red and yellow soils of Okinawa and Java Island.

Javan soils. The mineral that supplied most of the exchangeble Al in the Okinawan soils, however, was identified as halloysite.

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Manuscript received 20 August 1997, accepted 16 December 1997