

RELATIONS OF COMPOSITION TO STRUCTURE OF DIOCTAHEDRAL 2:1 CLAY MINERALS

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Abstract—A ternary plot of the tetrahedral R^{3+} and octahedral R^{3+} populations for the dioctahedral 2:1 clay minerals shows a broad range of compositional variation within each of the major clay minerals. The clay minerals can be subdivided on the basis of total charge, location of the charge, and the relative amounts of Al and Fe^{3+} in the octahedral sheet. The division is natural and is controlled by the misfit between the tetrahedral and octahedral sheets and the need for tetrahedral rotation. The compositions of the tetrahedral and octahedral sheets are interdependent. Whereas muscovite has a lower limit of 1.7 Al octahedral occupancy, illite and montmorillonite have a lower limit of 1.3 Al; lower Al values result in the formation of a chain structure.

TERNARY PLOT

YODER and Eugster (1955) outlined the observed composition range of natural 2:1 clays in a triangular diagram in which the two variables were tetrahedral R^{3+} and octahedral R^{3+} . The diagram divides the various clays on the basis of the relative amounts of tetrahedral and octahedral charge.

A similar plot was made using 240 analyses in order to obtain some idea of how discrete the compositional limits of the various clay groups are. It is apparent, even excluding the extreme values, that there is nearly a complete overlap of fields (Fig. 1). All possible combinations of octahedral and tetrahedral charge, with the exception of the apex or extreme ratios, occur in the clay minerals, muscovite occurs at one extreme (all tetrahedral charge), pyrophyllite at another (no charge), and celadonite (all octahedral charge) at the third. Only one celadonite value approaching this end-member composition has been found; it might be considered questionable whether such an end-member actually exists.

Some of the spread is due to poor analyses and impure samples. Most of the analyses were taken from the literature and an effort was made to be selective. For example, montmorillonite formulas with octahedral occupancy values larger than 2.10 were excluded as few careful analyses have such large values (most samples had values less than 2.05); however, as values larger than 2.10 are commonly reported for the other dioctahedral clays, they were not excluded unless larger than 2.20. An attempt was made to use only data for samples for which the purity had been established by the authors with reasonable care. Most of the

structural formulas were recalculated by computer. A number of glauconite and celadonite samples could not be plotted on the triangle as the layer charge, calculated on the basis of 2.00 filled octahedral positions, was larger than 1.00.

For samples with an octahedral population larger than 2.00, the octahedral R^{3+} values are not a true measure of the amounts of octahedral charge ($2.00 - R^{3+} = \text{octahedral charge}$). These samples would have a lower octahedral charge than indicated by the present plot; however, as some of the cations in excess of 2.00 assigned to the octahedral layer actually belong in the interlayer position (Foster, 1951) the octahedral R^{3+} value may be more indicative of the true octahedral charge than is the calculated charge.

Figure 2 shows some general relations that can be inferred from these data. A layer charge of 0.7 (per $O_{10}(OH)_2$) serves to divide those clays that are predominantly expanded from those that are predominantly contracted. Another subdivision can be made on the basis of whether ferric iron or aluminum is the predominant octahedral cation. The Fe clays are concentrated in the diagonal zone cutting through the center of the triangle and flanked on either side by zones populated by Al clays. Only a few of the Fe-rich clays have a tetrahedral charge larger than 0.6. There is relatively little overlap of the Fe and Al minerals on the left side of the triangle—illite, glauconite, and nontronite.

There is considerable overlap between minerals named montmorillonite and nontronite. Many of the montmorillonite samples that overlap the nontronite field are relatively high in iron but

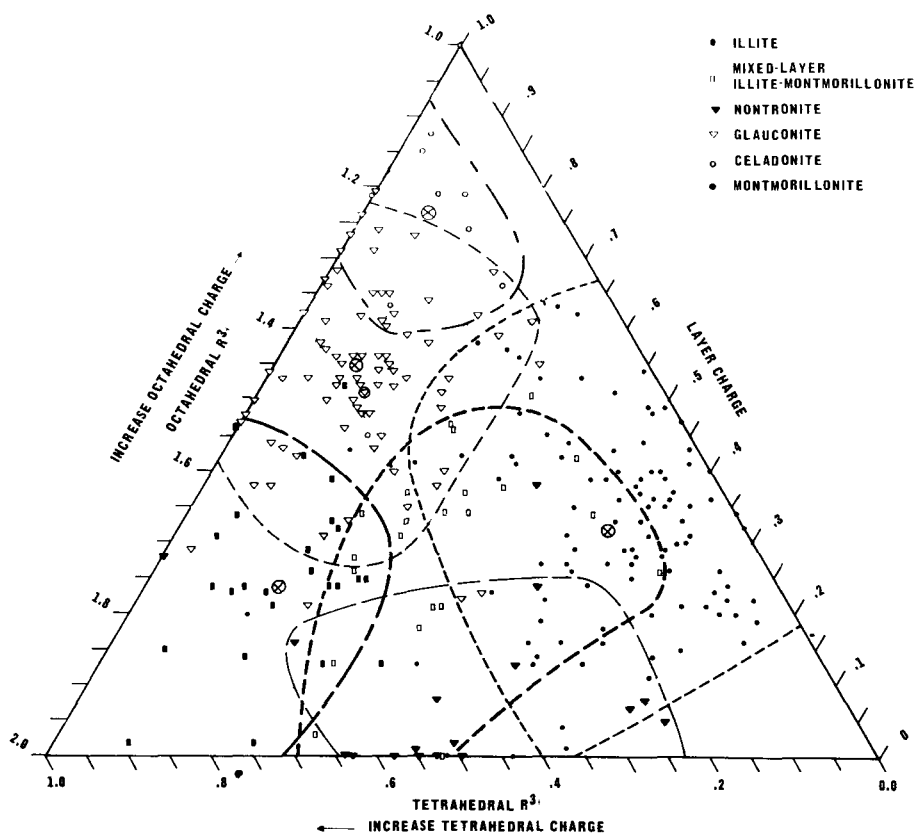


Fig. 1. Ternary plot of tetrahedral R^{3+} and octahedral R^{3+} for 240 dioctahedral 2:1 clay minerals. The layer charge is assumed to be due to tetrahedral and octahedral substitution. The octahedral sheet is assumed to have 2.00 cations per $O_{10}(OH)_2$. X = average composition of various clay minerals.

Montmorillonite analyses were obtained largely from Ross and Hendricks (1945), Grim and Kulbicki (1961) Osthaus (1955), Foster, (1954), Kerr, *et al.* (1950), Sawhney and Jackson (1958) and a scattering of single analyses. Nontronite analyses are from Ross and Hendricks (1945) and a selection of single analyses. Illite analyses include most of those in the literature up through 1965 and several unpublished analyses by the author. Glauconite analyses are from Hendricks and Ross (1941), Burst (1958), Warshaw (1957), Hallimond (1922), Tyler and Bailey (1961) and a selection of single analyses. The celadonite analyses are those compiled by Wise and Eugster (1964).

so are a number of samples that lie in the restricted montmorillonite zone. The structural formulas indicate that there is a complete gradation between montmorillonite-beidellite and nontronite so that any boundary is arbitrary. It might be noted that the average layer charge for these montmorillonites is 0.48 rather than the 0.33 value commonly used.

A zone near the middle of the triangle and within the less than 0.7 charge area has been labeled mixed. Relatively few minerals fall in this zone and this zone lies in the approximate center of the area in which the mixed-layer illite-montmorillon-

ites lie. This is the area in which the mixed-layering is well developed. The minor component is generally greater than 30 per cent. A few montmorillonite samples are in this area. It is quite likely that most of these samples have mixed layers and that a portion of the layers have a high enough charge to potentially contract (when potassium is available). The samples in the montmorillonite and the nontronite zones with charges greater than 0.6 and 0.5 respectively could also have some potentially contractable layers. The lower charge boundary (0.5) for potentially mixed nontronite is determined by the larger proportion of charge

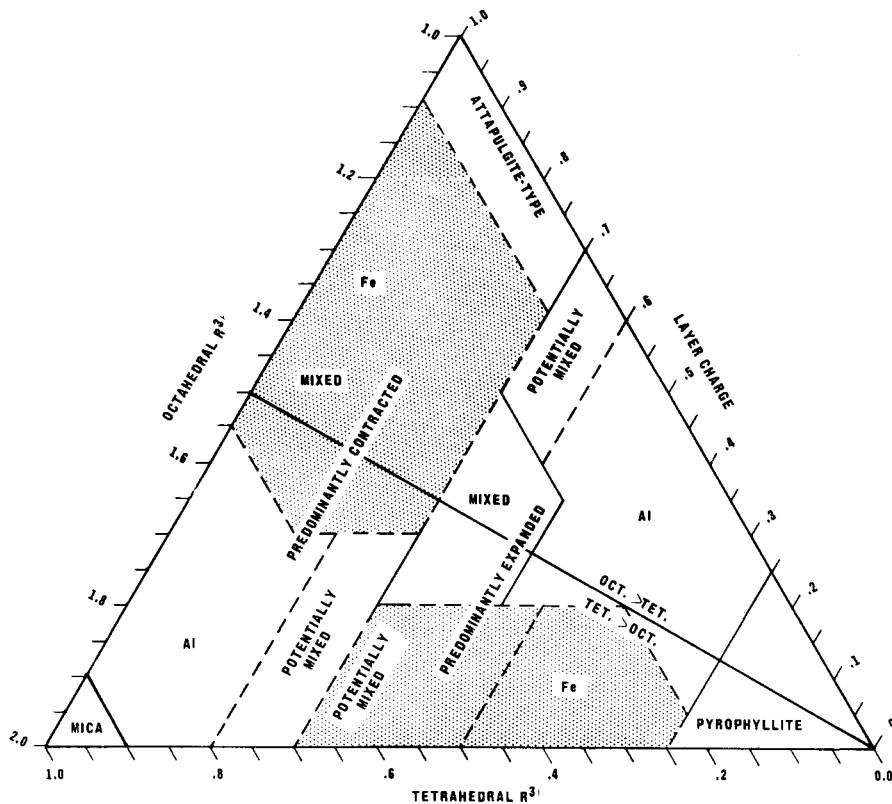


Fig. 2. Generalized classification of dioctahedral 2:1 clay minerals. Mixed refers to mixed-layering.

origination in the tetrahedral sheet. In the contracted Al and Fe zones, those samples with the higher layer charge are more likely to be pure mineral and as the charge decreases the percentage of the intergrown expanded layers increases. Completely contracted clays occur principally in the Al zone (illite) illustrating the more effective strength of the tetrahedral charge compared to the octahedral charge. Glauconites always contain some expanded layers.

Mixed-layer refers to intergrowths of layers that have a sufficiently high charge to contract to approximately 10 \AA and those that contain an interlayer of water (and usually a lower charge). This is gross, obvious interlayering. Even in those clays that have all layers contracted or all expanded, it is extremely likely that most are composed of two or more phases or types of layers.

The general vagueness of the various boundaries is due, in part, to the prevalence of mixed-layer clays. Contractable layers must have a charge range from 0.6 to 1.0 and expandable layers from 0.25 to 0.6 or 0.7. Many clays are a mixture of these two types of layers and a charge value of

0.7 for a clay may indicate that all layers have this charge or, more likely, that it represents a mixture of layers having values larger and smaller than 0.7.

Most of the celadonite samples lie in the area where some mixed-layering is to be expected. Though celadonite is commonly considered to be non-mixed, the literature suggests that little effort has been made to establish this. Of the 15 analyses examined, (Wise and Eugster, 1964) six reported H_2O -water and in the others it was not determined. In any event, the sheet structure of the celadonite is distinctly different from that of the other 2:1 dioctahedral clays (Radoslovich, 1963). It has a very thick octahedral sheet, all three octahedral positions are of equal size (in the other 2:1 dioctahedral clay, the two filled positions are smaller than the vacant position) and the interlayer separation is larger than in other contracted 2:1 dioctahedral micas (Radoslovich, 1963).

In Fig. 2, the dividing line between predominantly octahedral charge and predominantly tetrahedral charge approximately coincides with the

boundaries separating the Al and Fe clays (illite-glaucanite and montmorillonite-nontronite).

Actually a simplified division (Fig. 3) based on the 0.7 charge boundary and the boundary between predominantly octahedral and predominantly tetrahedral charge coincides well with the divisions based on the plotted data.

These diagrams indicate that when the total layer charge is less than 0.7, Al will be the dominant cation when the seat of the charge is largely in the octahedral sheet; as the predominant charge shifts to the tetrahedral sheet the larger Fe ion substitutes for Al in the octahedral sheet. Radoslovich (1962) found that montmorillonite was the only layer silicate in which tetrahedral Al caused the layer to increase in size in the *b* direction. He explains this by suggesting: "Suppose that in all the layer silicates, the tetrahedral layers exert a very small expansive force (when $\alpha > 0$). In kaolins, there is only one tetrahedral layer per octahedral layer, and in micas the inter-layer cation dominates the tetrahedral twist. But in montmorillonites the small force due to *two* tetra-

hedral layers per octahedral layer must just have a noticeable effect."

Thus, it appears that for the low charged dioctahedral 2:1 clays as tetrahedral Al increases and the tetrahedral sheets expand in the *b* direction there is a tendency for this expansion to be matched by substitution of the large Fe^{3+} ion in the octahedral sheet. That this latter substitution is effective in adjusting the difference in size between the two types of layers is suggested by the relatively small tetrahedral twist in nontronites.

This attempt at size adjustment between the two types of sheets is illustrated by the fact that very few Fe-rich 2:1 clays contain less than 0.2 tetrahedral R^{3+} . When increased Al in the tetrahedral sheets is compensated by substitution of Mg in the octahedral sheet layer, the octahedral charge increases and the overall layer charge is increased. This allows strong K-bonds to be developed and exert an influence on layer dimensions. Once the layer charge is larger than 0.7 and predominantly tetrahedral in origin, the Al dominated octahedral layer represents the stable phase. Such a clay

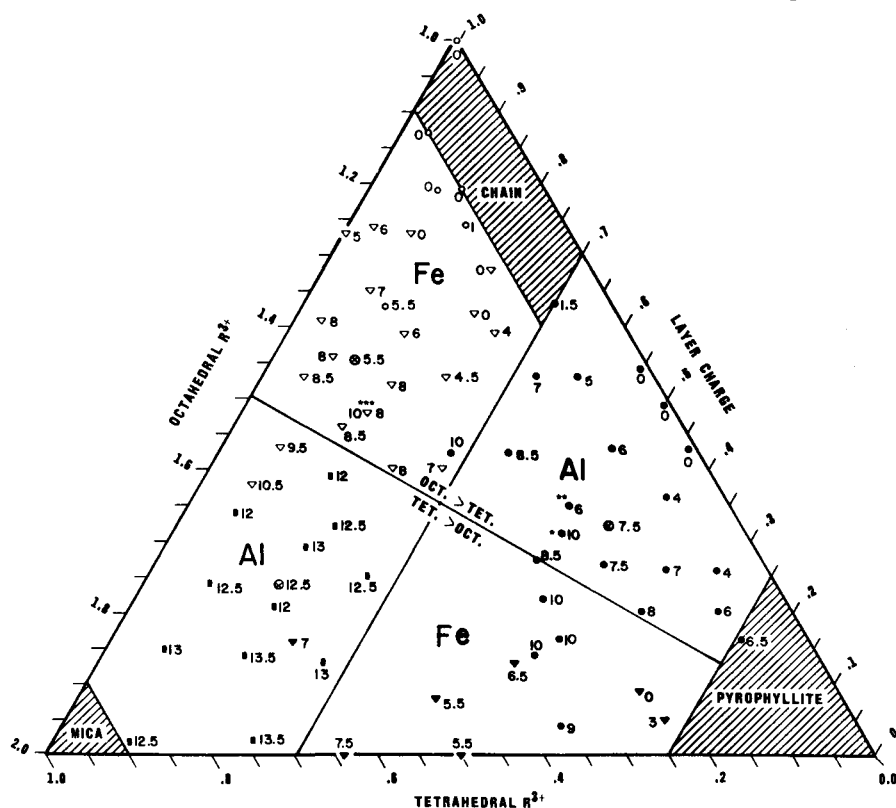


Fig. 3. Idealized subdivision of the dioctahedral 2:1 clay minerals. Numbers refer to amount of calculated tetrahedral twist. * = no octahedral Fe, ** = high octahedral Fe^{3+} , *** = high octahedral Al.

approaches muscovite in composition and charge distribution and its stability is determined by the same complex balance of interlocking strong bonds ascribed to muscovite by Radoslovich (1963).

When the tetrahedral charge is larger than 0.6 the octahedral sheet cannot adjust to the increased size of the tetrahedral sheet merely by increased size of the large Fe^{3+} ion. In this situation, it is not only necessary to substitute larger cations for octahedral Al but large divalent cations (Mg) so that an octahedral charge is created. The resultant layer charge allows interlayer potassium to aid in adjusting the size of the two types of sheets. When the tetrahedral charge is large enough (muscovite) the octahedral charge is not required.

As the predominant charge shifts from the tetrahedral to the octahedral sheet (illite to glauconite and celadonite) anion-anion repulsion is increased (octahedral sheet of celadonite 2.48 Å thick as compared to 2.21 Å for muscovite) and the larger Fe^{3+} ion in octahedral coordination represents the more stable phase.

As the relative proportion of large cations (Fe^{3+} , Fe^{2+} , Mg) in the octahedral sheet increases and the layer charge is large enough to cause the layers to be predominantly contracted, some Al substitution in the tetrahedral sheet is necessary to maintain the size difference between the sheets and allow for tetrahedral twisting. Radoslovich and Norrish (1962) have calculated that a celadonite with no tetrahedral substitution would have no tetrahedral twist and the K^+ would penetrate the oxygen sheets to such an extent that there would be "an impossibly close approach of successive layers." When tetrahedral Al values are larger than approximately 0.5–0.6 for these clays with a high octahedral charge the amount of tetrahedral twist may become excessive. To accommodate additional tetrahedral Al it is necessary to decrease the octahedral charge so the octahedral layer can expand in the *b* direction relatively more than the *c* direction.

When the tetrahedral charge is low and the $\text{R}^{3+}/\text{R}^{2+}$ ratio approaches unity (upper apex of triangle) the only way to adjust the size of the octahedral sheet so that the tetrahedrons can be induced to twist is to increase the amount of Al at the expense of Fe^{3+} . However, Radoslovich (1963) states: "If Al is substituted for Fe^{3+} then the average cation-oxygen bonds are correspondingly shortened, and the octahedral cations brought closer together—in fact, unduly close."

It is suggested that under these conditions the strain in the 2:1 layer is such that it is relieved by the inversion of the tetrahedra and the formation of a chain structure (attapulgitite-type). Part of the octahedral charge may be satisfied by OH proxying for oxygen. That this may happen is

suggested by the fact that in attapulgitite, the observed hydroxyl water is larger than the calculated hydroxyl water (4–7 per cent vs. 2.15 per cent—Caillere and Henin, 1961).

It is possible that some of the Al montmorillonites with a high layer charge and a high octahedral charge may be a mixture of chain and sheet structures of varying *b* axis widths. Bradley (1955) has suggested that such structural irregularities, which he termed faults, are common in the trioctahedral chlorites.

To obtain some additional support for the partitioning suggested in Figs. 2 and 3 an attempt was made to determine some relative tetrahedral twist values. Radoslovich and Norrish (1962) showed that the ratio:

$$\frac{\text{Fe}^{2+} + 0.853 \text{Fe}^{3+} + 0.455 \text{Mg} + 0.43 \text{Ti}}{\text{Al Tetrahedral}}$$

is a good measure of the ratio ${}^b\text{oct.}/{}^b\text{tet.}$ They showed a good linear relation existed between this value and the calculated layer separation, which is largely controlled by the amount of tetrahedral rotation.

Using their data for the dioctahedral clays and micas, a plot was made of calculated rotation vs. calculated ${}^b\text{oct.}/{}^b\text{tet.}$ using the above formula (Fig. 4). Though the absolute values may have appreciable error the relative differences appear to be real.

The data for the expanded and contracted minerals plot as two separate linear relations with contracted clays having larger tetrahedral rotation values for given ${}^b\text{oct.}/{}^b\text{tet.}$ values than the expanded clays. This is presumably due to the K which aids the tetrahedral rotation in the contracted clays.

Using the structural formulas that were used to plot Fig. 1, the ${}^b\text{oct.}/{}^b\text{tet.}$ ratio was calculated for a number of clays and the amount of tetrahedral rotation estimated from the graphs in Fig. 4. Some of these values are shown in Fig. 3. The montmorillonites with a low tetrahedral Al and low octahedral R^{3+} have high ratio values and presumably a low degree of tetrahedral rotation (0° to 1.5°). As the amount of octahedral R^{3+} increases, Mg decreases, the octahedral layer becomes smaller and the amount of tetrahedral rotation increases (6.5°).

As the amount of tetrahedral Al increases and the tetrahedral layer increases in size the amount of rotation systematically increases attaining a maximum value of approximately 10° .

As much of the octahedral Al is replaced by the larger Fe^{3+} (nontronite) the amount of rotation decreases. As the amount of tetrahedral Al increases, the amount of octahedral R^{3+} remains

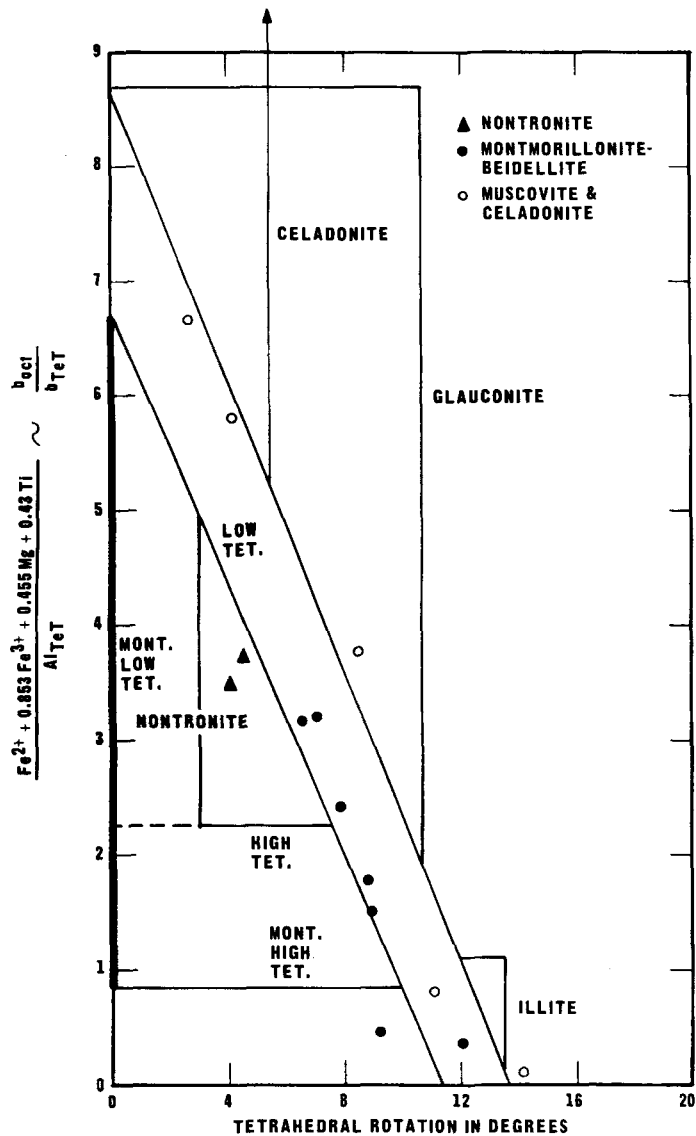


Fig. 4. Plot of calculated degree of tetrahedral rotation vs. calculated b_{oct}/b_{tet} values for the various clay minerals. Linear relations are based on data from Radoslovich and Norrish (1962). Triangles show range of calculated tetrahedral rotation for the various clay groups (See Fig. 3).

relatively constant and the tetrahedral rotation increases from 0° – 3° to 7.5° .

As tetrahedral Al becomes larger than 0.5–0.6 and the layer charge becomes strong enough to cause K-fixation and contraction (illite), the amount of tetrahedral rotation is nearly doubled (13°).

Along with the increase in tetrahedral Al there is an increase in the amount of octahedral Al and Mg at the expense of Fe^{3+} . The analyses suggest this is a continuous series and much of the abrupt increase in rotation is due to the increased layer charge and the fixation of the K ion. There appears

to be not good reason why Fe^{3+} illites could not exist. An illite-type clay with an octahedral sheet composed predominantly of Fe^{3+} ($\text{Fe}_{1.80}^{3+}\text{Al}_{0.10}\text{Mg}_{0.30}$ and $\text{Fe}_{1.30}^{3+}\text{Mg}_{0.20}$ with 0.6 tetrahedral Al) would have a tetrahedral rotation of approximately 11° – 9° , which is presumably larger than for most glauconites.

Present data indicate that Fe^{3+} -rich low charge clays increase their layer charge by increasing the Mg and Fe^{2+} content of the octahedral sheet at the expense of Fe^{3+} more so than Al. The average Al content of glauconite and celadonite is similar to that of nontronite, but the Fe^{3+} values are lower. With increased octahedral charge there is an increased anion–anion repulsion and the octahedral sheet increases relatively more in the *c* direction than the *b* direction, which also favors the large cations. Thus, relatively less tetrahedral Al is required to afford the sheet size differential to allow sufficient tetrahedral rotation to lock the K into place.

The calculated tetrahedral rotation for glauconites with high octahedral R^{3+} values ranges from 8 to 10° . There appears to be no overlap of the illite values (12° – 13.5°). As the amount of octahedral R^{3+} decreases, the octahedral sheet increases in size and charge and rotation values decrease. As the amount of tetrahedral Al decreases the layers become similar in size and the amount of rotation approaches zero and K cannot be locked in position to provide sufficient layer separation.

As octahedral R^{3+} decreases and the octahedral charge increases the amount of tetrahedral Al would have to increase to afford the size differential for tetrahedral twist. This would produce a layer charge larger than unity, which is presumably an unstable condition for low temperature layer structures. Further changes would require material changes in the makeup of the octahedral and/or tetrahedral sheets.

Layer structure clays have 4 tetrahedrons per 3 octahedral positions. When the limits suggested above are reached, there is little additional adjustment that can be accomplished by compositional changes. It is necessary for the tetrahedron to octahedron ratio to change. This is accomplished by increasing the tetrahedron to octahedron ratio to 8:5 (attapulgite). This structural change is apparently an over-compensation and to bring the sheets into closer accord, the amount of tetrahedral Al is decreased and though the amount of octahedral R^{2+} is increased the octahedral charge is decreased (favoring stretching in the *b* direction) by the addition of H^+ . However, the size differential caused by the 8:5 ratio is apparently too large to be adjusted by compositional changes

and the strain build up over an interval of five octahedral positions is sufficient to cause tetrahedral inversion.

When the relative amount of large cations (Fe^{3+} , Fe^{2+} , Mg) in the octahedral layer is increased (80–100 per cent of the octahedral cations) the layer strain is slightly reduced and the octahedral sheet can extend to 8 or 9 positions before forcing the tetrahedral inversion (sepiolite).

It seems likely that the octahedral layers of attapulgite and sepiolite have a much wider compositional range than indicated by the present limited analyses and that they occupy the intermediate position between the dioctahedral and trioctahedral clays.

CHAIN STRUCTURE

Unfortunately, there are few if any chemical analyses of pure attapulgite. The details of the structure are in doubt and structural assignments must be done subjectively (Caillere and Henin, 1961).

Attapulgites have less octahedrally coordinated alumina than is found in the montmorillonites. The magnesium content of the octahedral sheet is 2–4 times as abundant as in the montmorillonites. The iron contents are similar. The average $\text{Al}_2\text{O}_3/\text{MgO}$ ratio for 24 illites is 9.6, and for 101 montmorillonites 6.7. Attapulgite values range from 2.5 to 0.48. The average ratio octahedral Al/octahedral Mg are respectively 5.4, 4.3, and 1.8–0.4.

Radoslovich (1963) found that the 2 M muscovite structure required that a minimum of 1.7 of the octahedral sites be filled with Al. The lower limit of 1.7 Al is equivalent to 85 per cent of the two symmetrically related or occupied sites being filled in a stable muscovite structure. For 24 illites, an average of 1.53 Al per three octahedral sites was found. This is 77 per cent occupancy of the two occupied sites. Total trivalent ions ($\text{Al} + \text{Fe}^{3+}$) averaged 1.76 (88 per cent occupancy).

The average Al per three sites for 101 montmorillonite is 1.49 (74.5 per cent) and for Al and Fe^{3+} , 1.68 (84 per cent). Frequency distribution graphs of data from these two minerals groups indicate that the large majority contain between 1.3 and 1.7 Al per three sites. There are relatively abrupt decreases in frequency at these values (Fig. 5) suggesting that they are significant boundary values. The minimum value of 1.3 is equivalent to 65 per cent occupancy of the two occupied sites as compared to a minimum of 85 per cent for 2 M muscovite. The size of the cation is considered to be more important in controlling the size of the octahedral sheet than the charge of the cation. Thus, it is the amount of octahedral Al rather

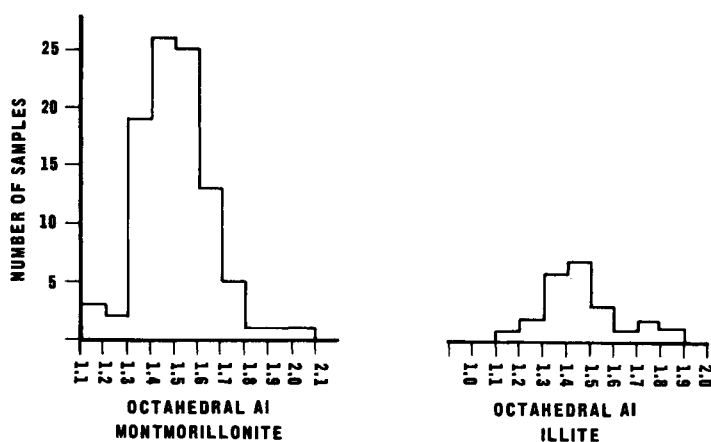


Fig. 5. Frequency distribution of octahedral Al per $O_{10}(OH)_2$.

than $Al + Fe^{3+}$ that most closely reflects the amount of strain in the octahedral layer.

In attapulgite, only four of the proposed five octahedral sites are occupied. The seven structural formulas calculated by Caillere and Henin (1961) indicate that Al fills from 1.13 to 2.34 of these five sites or between 28 and 59 per cent of the four occupied sites. ($Al + Fe^{3+}$ values range from 31 per cent to 62 per cent.) The amount of tetrahedral Al per four positions ranges from 0.0005 to 0.34 and averages 0.165. When four of the formulas were recalculated on the basis of the measured water loss in different temperature ranges, the average tetrahedral Al was 0.085 with two of the four showing no tetrahedral substitution. The amount of octahedral Al per four positions averaged 50.1 per cent.

On the basis of these admittedly few data, it would appear that the chain structure attapulgites are the most likely clay minerals to have an Al/Mg ratio near one. They also have relatively little tetrahedral substitution.

It would seem that in the dioctahedral 2:1 sheet structure silicates, when the occupied sites are more than 85 per cent occupied by Al, the structure can adjust to compensate for the internal strain and can grow to a considerable size. The Al octahedral occupancy values of muscovite (>1.7) and the 2:1 dioctahedral clays (1.3–1.7) indicate that there is little overlap. It is likely that the decreased amount of tetrahedral twist induced by increasing the size of the octahedral cations and octahedral charge (decreasing Al) determines that a clay-size rather than a larger mineral will form. The R^{3+} occupancy value can be less than 1.3 when the larger Fe^{3+} is substituted for Al. When Al occupancy values are less than 1.3 (65 per cent),

in the absence of appreciable iron, the internal strain is such that growth is in only one direction. The width of the layer is restricted to five octahedral sites. Sufficient lattice strain accumulates within this five-site interval that the silica tetrahedral sheet is forced to invert to accommodate the strain.

OCTAHEDRAL SHEET

Octahedral Mg, Al, and Fe^{3+} were totaled and the relative proportion calculated (atomic per cent). Octahedral Mg, Al, Fe^{3+} and Fe^{2+} were totaled and the per cent Fe^{2+} calculated. The distribution values for the 2:1 minerals are summarized in Fig. 6. For comparison purposes, values for attapulgite and some trioctahedral montmorillonites are also shown.

There is almost complete coverage of the lower portion (less than 40 per cent Mg) of the diagrams. The high Fe^{3+} octahedral layers have a higher maximum amount of Mg (50 per cent) than the high Al layers (35 per cent). The Fe^{3+} -rich layers also have a higher Fe^{2+} content than the Al-rich layers and in general the Fe^{2+} content increases as the Mg content increases. The average octahedral Fe^{2+} for the clays is: illite 1.7 per cent, mixed layer 0.6 per cent, montmorillonite 0.9 per cent, nontronite 0.7 per cent, glauconite 9.9 per cent, celadonite 10.0 per cent. Though there is considerable error in the Fe^{2+} values, the relative difference are real. If the Fe^{2+} values were added to the Mg values, the celadonite and glauconite fields would move appreciably closer to the Mg + Fe^{2+} apex.

The maximum amount of Fe^{2+} the octahedral layers of the 2:1 dioctahedral clays generally contain is 12 per cent (Fig. 7). This is equivalent

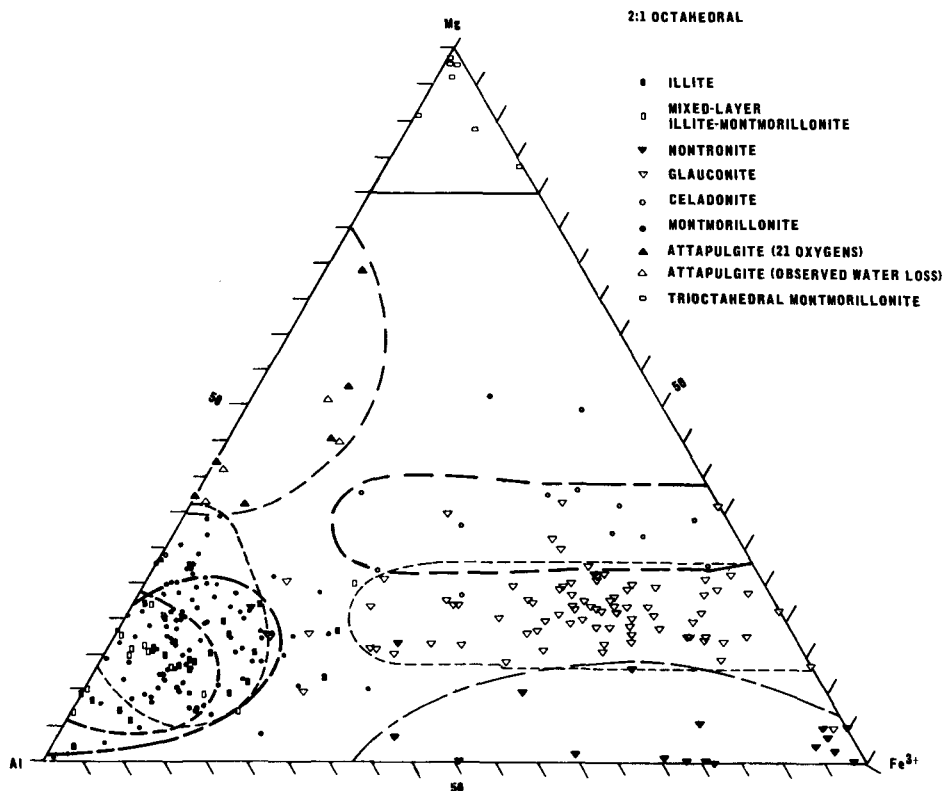


Fig. 6. Ternary plot based on the composition of the octahedral layer of the dioctahedral 2:1 clay minerals, attapulgite and trioctahedral saponite. Fe^{2+} was excluded.

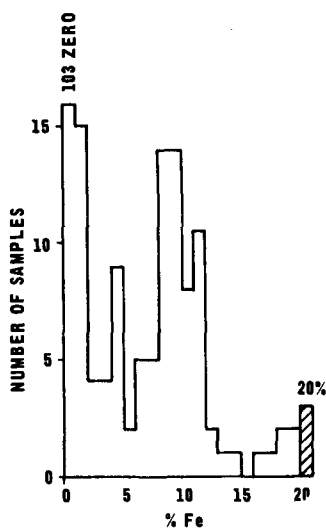


Fig. 7. Histogram showing the distribution of the per cent of Fe^{2+} in the octahedral sheet of the dioctahedral 2:1 clay minerals. No Fe^{2+} was reported for 103 samples and 3 samples had more than 30 per cent Fe^{2+} .

to approximately 0.25–0.30 octahedral positions. Due to the relatively large size of the Fe^{2+} ion, the structure can apparently adjust to only about half as much Fe^{2+} as Mg.

When the modal values are considered, there is relatively little overlap in the composition of the octahedral layers of the three Fe^{3+} rich clays; whereas, for the Al-rich clays, the illite and mixed-layer illite-montmorillonite fields fall within the montmorillonite-beidellite field.

Nomenclature is slanted in favor of Fe over Al. When the octahedral layer has more than 30 per cent Fe^{3+} the clay is usually given an Fe mineral rather than an Al mineral name. The data indicate that other than a restriction of less than 40 per cent Mg all combinations exist in nature; though most of the values are clustered in two areas—greater than 60 per cent Al; and greater than 40 per cent Fe^{3+} .

As long as the Mg and Fe^{2+} content of the octahedral sheet is below the limits stated, the tetrahedral sheet is able to adjust to the size and charge of the octahedral layer by cation substitution and tetrahedral twisting and can form a stable structure.

Though some tetrahedral–octahedral combinations are more stable than others, the relative abundance of certain octahedral compositions (Fig. 5) may be in part due to the restrictions imposed by the major natural environments. For example, the environmental conditions (and source material) under which most clays form is such that either Al or Fe may be relatively abundant, but seldom both.

The plot also shows that the octahedral sheet of attapulgite tends to be composed of approximately 50 per cent Mg and 50 per cent Al and their compositional field does not overlap that of the 2:1 dioctahedral and trioctahedral clays.

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Résumé—Un relevé ternaire des populations tétraédriques R^{3+} et octaédrique R^{3+} pour les minéraux argileux dioctaédriques 2:1, montre une gamme étendue de variations dans la composition de chacun des principaux minéraux argileux. Les minéraux argileux peuvent se subdiviser sur la base d'une charge totale, de la localisation de la charge et des quantités relatives de Al et de Fe^{3+} dans la feuille octaédrique. La division est naturelle et elle est contrôlée par l'échec entre les feuillets tétraédriques et octaédriques et le besoin pour la rotation tétraédrique. Les compositions des feuillets tétraédriques et octaédriques sont interdépendantes. Cependant, le muscovite a une limite plus faible de contenance de 1.7 Al octaédrique, l'illite et le montmorillonite ont une limite plus basse de 1.3 Al; des valeurs d'Al plus faibles résultent de la formation d'une structure en chaîne.

Kurzreferat—Ein Dreistoffdiagramm der tetrahedralen R^{3+} und oktahedralen R^{3+} Bestände für die dioktahedralen 2:1 Tonminerale zeigte einen weiten Bereich von Unterschieden in der Zusammensetzung innerhalb jedes der wichtigeren Tonminerale. Eine Unterteilung der Tonminerale ist möglich auf Grund der Gesamtladung, Lage der Ladung und der relativen Menge von Al und Fe^{3+} in der oktahedralen Schicht. Die Teilung ist eine natürliche Erscheinung und wird durch den Mangel an Anpassung zwischen tetrahedralen und oktahedralen Schichten, sowie durch den Bedarf für tetrahedrale Drehung bedingt. Die Zusammensetzungen der tetrahedralen und oktahedralen Schichten sind voneinander abhängig. Während Muscovit eine untere Grenze von 1,7 Al oktahedraler Besetzung hat, weisen Illit und Montmorillonit eine untere Grenze von 1,3 Al auf; niedrigere Al Werte geben Anlass zur Bildung von Kettenformationen.

Резюме—Третья кривая четырехгранных R^{3+} и восьмигранных R^{3+} плотностей издивосьмигранных 2:1 глинистых минералов указывает на крупное количество изменений по составу в каждом из главных глинистых минералов. Глинистые минералы подразделяют на основании общей нагрузки, местоположения нагрузки и относительного количества Al и Fe^{3+} в восьмигранном слое. Раздел естественный и регулируется отсутствием несоответствия между четырехгранными и восьмигранными слоями, а также необходимостью четырехгранного чередования. Составы четырехгранных и восьмигранных слоев взаимосвязаны. В то время, как мусковит имеет более низкий уровень восьмигранной занятости 1,7 Al, иллит и монтмориллонит имеют более низкий уровень 1,3 Al; более низкие значения Al влекут за собой возникновение цепной структуры.