

GEOCHEMISTRY, MINERALOGY AND MICROFABRIC OF SOILS

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ABSTRACT

The modern characterisation of soils is based primarily on the type of pedogenesis and on their behaviour resulting from the pedoclimatic conditions. This general evolution results from the imbalance of the lithosphere's rocks when the primary minerals become exposed to both the atmosphere and the biosphere: a new state of equilibrium that must correspond with the current conditions of the surface environment.

This adaptation of pedological covers to their environment assumes two aspects: change in chemical and mineralogical constitution, and development of a characteristic organisation (fabric) at all levels, especially at the microscopic level corresponding to the plasma.

Constitution: The new constitution is essential due to geochemical processes (weathering) leading to the formation of characteristic minerals (clays and hydroxides). These new components (secondary constituents), which can vary with environmental conditions, are of small size and have a cryptocrystalline nature. They make up the so called "plasma", a divided phase having an expanded surface area that demonstrates all types of physico-chemical interface phenomena such as ion exchange and hydration with consequences particularly on the microscopic fabric.

Micro-organisation: As the plasma is made up of many particles and forms a "solid-water" system, problems concerning particle arrangement play a major role, especially in the domains of texture and microstructure. Particle arrangement depends on the mineralogical characteristics of the solid phase and on the properties of the contact solution which can play a role as a result of its composition, concentration, or hydric suction.

A review is given of the major evolutions likely to occur at the earth's surface for the two aspects above. The perfect harmony developing between pedoclimatic conditions and the characteristics of soil constitution and microstructure is shown.

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INTRODUCTION

Pedogenesis embraces all processes of soil formation and evolution at the Earth's surface. It results from the imbalance of any rock of the lithosphere when the latter is exposed and comes into contact with the atmosphere and the biosphere, which implies the search for a new state of equilibrium that must agree perfectly with the current conditions of the superficial environment.

Therefore, any soil results first from a constant adaptation of the Earth's surface to its environment and the review presented here aims initially at determining this adaptation and moreover, at showing that all aspects of soil evolution are perfectly adjusted, particularly the geochemical, mineralogical and microstructural components.

The resulting evolution involves geochemical and crystallochemical phenomena which lead to a new chemical composition and to a new mineralogical constitution. They are the general weathering phenomena (dissolution, neoformation, microdivision, transformation ...) leading most often to the development of new minerals which make up the soil plasma and whose clays are the most typical elements along with the humic constituents in the surface horizons.

The most general system is represented in Figure 1.

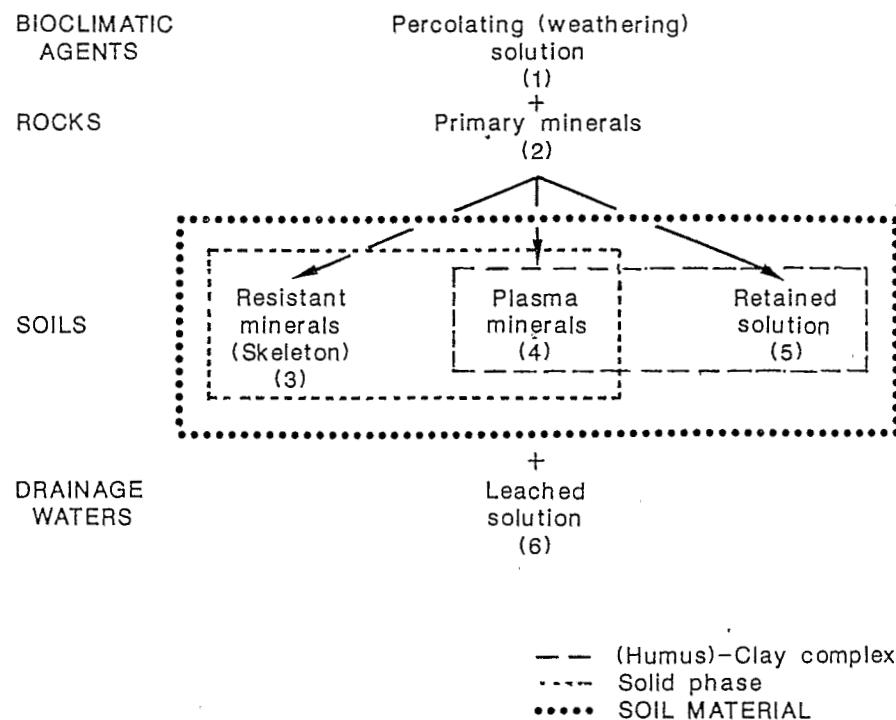


Figure 1 General representation of pedological system

The main processes observed in relation to geochemical weathering (petrographic parameters of the parent rock and bioclimatic parameters of the contact solution) are reviewed and the main types of plasmas (minerals or parageneses) likely to be found in the great pedological covers of the Earth are characterised.

The first subject deals mainly with the geochemistry-mineralogy (crystallochemistry) relations (Fig. 1 - Interactions between 1, 2, 4 and 6).

A second element must be considered due to the morphological and dimensional properties of the usual constituents of the plasma. The latter and particularly clays and oxides-hydroxides are most often characterised by:

- a lamellar facies
- a small size ($\phi < 2 \mu\text{m}$)
- a cryptocrystallinity and even an amorphous constitution in a number of environments, (showing not very pronounced mineralogical features). It results merely from the fact that the soil crystallogenesis occurs at low pressures (atmospheric pressure) and at not very high temperatures (about 300°K) - under low-energy conditions and therefore rather low kinetic rates. It is more rapid to produce small particles than large crystals.

Soft materials are formed composed of finely divided minerals (great number of "particles"), with a higher contact surface and a superficial electrical charge because the particles are smaller (Fig. 2 and 3). Therefore, they are likely to react easily with the liquid and gaseous phases observed in the environment and leading to surface tension and capillary

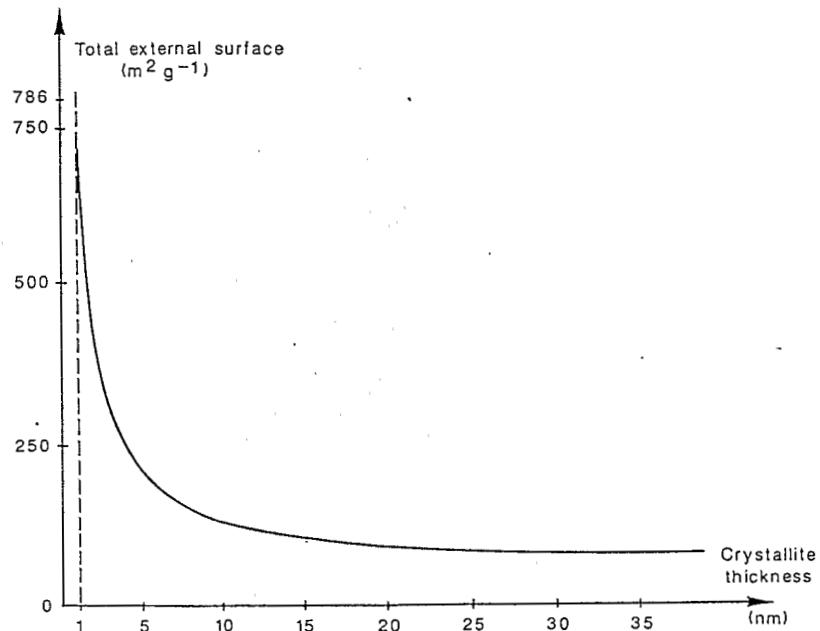


Figure 2 Variation in the external surface formed by clays as related to the crystallite thickness (number of layers) - Square particles of $0.025 \mu\text{m}$

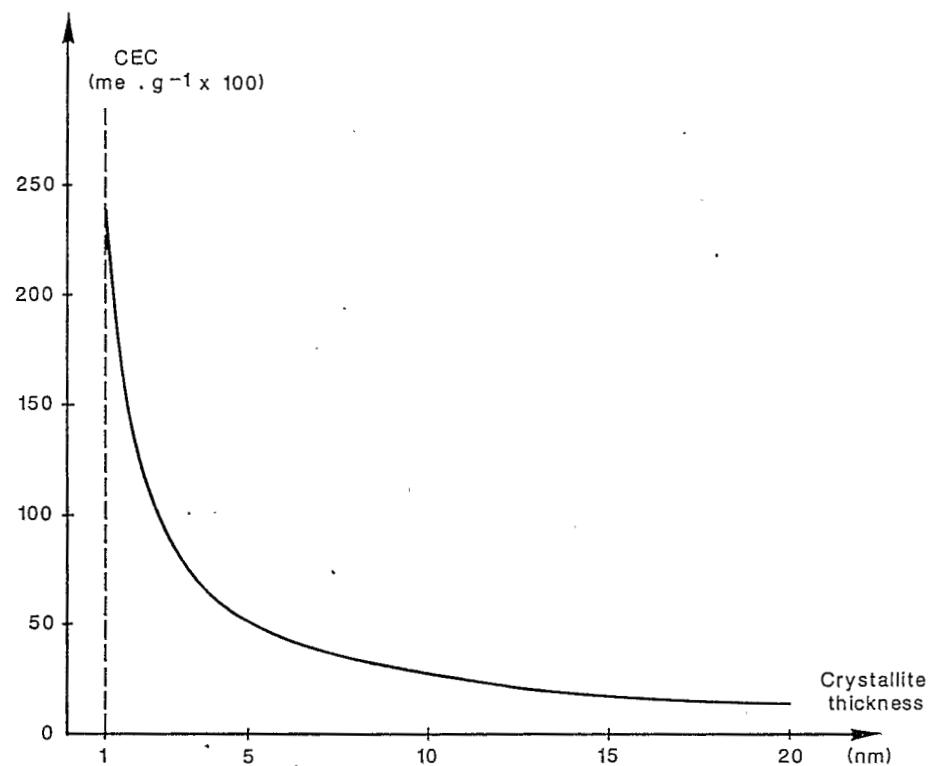


Figure 3 Variation in the CEC of micaceous clays (layer charge $z = 0.9$) as related to the crystallite thickness

phenomena. The latter play a major role in the arrangement of solid particles and therefore, they govern the geometry of the microfabric.

Thus, the relations between mineralogy and microfabric are established and they correspond to the interactions existing between the elements 4 and 5 of Figure 1.

On the whole, under given bioclimatic conditions, soil evolution leads to the development of soil materials characterised by:

- a given geochemical and mineralogical constitution
- a well defined microfabric

These two elements can be preserved as long as the environmental conditions remain the same.

These two points will be studied successively.

GEOCHEMICAL AND CRYSTALLOCHEMICAL PROBLEMS

The major types of weathering will be studied along with their related plasmic constituents (clays).

Geochemical and Crystallochemical Characteristics of the Main Soil Processes

In all cases, the biogeochemical phenomena of weathering lead to a partial or total release of the elements of parent rocks and their transfer into solution, in such a way that the chemical composition of the weathered horizon is completely different from that of the initial rock. Two can be distinguished: elements released stay independent or recombine to produce specific secondary minerals (clays s.l.) which will then constitute the "plasma"; the unweatherable or unweathered primary minerals which form the "skeleton" (Pedro, 1983).

Aplasmogenic weathering: The evolution does not lead to the formation of secondary minerals (plasma) and even provokes the destruction of secondary constituents which exist initially in the rock. The atmospheric evolution appears in this case as a simple solubilisation so that the A horizon includes unweathered primary minerals (for example: quartz). So it does not really correspond to an "alterite" but to a weathering residue and leads finally to soil profiles with strongly differentiated horizons. The weathering process, which corresponds in this case to a preponderant physico-chemical action, can be characterised according to the nature of the chemical attack. At the Earth's surface, there are three main types, which correspond to three great soil types (Table 1):

Table 1 Weathering processes related to the neoformation of plasmic minerals

Parent rock	Geochemical type		Processes	Typical secondary constituents
Crystalline rocks	Total hydrolysis (total desilicification and desalkalinization)		Allitization (Al)	Hydroxide (Gibbsite)
	Partial hydrolysis	Total desalkalinization	Monosialitization (Si-Al)	1/1 clays (Kaolinite)
		Partial Desalkalinization	Bisialferrization Si-(Al, Fe)-M	2/1 clays Montmorillonite (smectite)
Volcanic rocks (with vitreous phase)	Hydrolysis		Allophanization	Allophane (amorphous)

- A highly acid and complexing attack under the influence of organic acids ("acidocomplexolysis"), e.g. in podzols.
- A highly alkline attack, mostly due to an intervention of sodium carbonate solutions, with a preferential dissolution of aluminium and silica ("alkalinolysis"), e.g. in the solods formation.
- An attack of mineral constituents, likely to occur at the interface of clay minerals, in alternately flooded or ultra dry environments; resulting from the action of highly acidifying cations such as Fe ("ferrolysis" - Brinkman, 1970), e.g. in the planosols.

Plasmogenic weathering: In this case weathering leads to the formation of new and characteristic minerals constituting the soil plasma: the clays. The weathering horizon forms, in this case, a real "alterite". The problem consists of determining the different ways leading to the individual development of the main plasmic constituents (Pedro, 1966, 1968, 1979, 1980), taking into account simultaneously the degree of weathering, which can be massive or partial (Pedro et al, 1976); and the clay crystallogenetic origin, which can be neoformation or transformation.

Massive weathering - formation of neogenesis clays

In tropical humid climates, weathering is strong, under the action of water more or less charged with carbon dioxide (hydrolysis); this leads to the rapid destruction of weatherable minerals and a massive neogenesis of new minerals, clays and ferric hydrates. Of course, different minerals can form depending on the degree of hydrolysis and the nature of the materials undergoing this evolution. From this point, it becomes easy to characterise the main soil types, taking additionally into account the accessory constituents and the nature of superficial cations (Table 2).

Partial weathering - individualisation of transformation clay

In the climates where hydrolysis is less aggressive, either because the conditions are humid and colder or hot but dry, the weathering is of partial type with the accumulation of weatherable minerals in the horizons and discrete individualisation of clays. In this case, the clay plasma essentially comes from the inherited parent rock phyllites, and can be affected by transformations even in a solid state. Thus, the neoformation is only minor (Pedro, 1976) and only concerns the independent ferric hydrates, humic constituents and calcium carbonates.

Due to the frequent presence of inherited 2/1 phyllites, the differentiation can only occur in accordance with the ionic composition of the interlayer (their nature: Ca, Na, H, AlOH; and the degree of hydration) and the appearance of non-silicate minerals, for example CaCO_3 .

In "hydrolytic" environments, with accumulation "in situ" of iron and aluminium, it is possible to find the following when going towards the driest environments:

- [(Phyllite 2/1)¹ Ca]
- [(Phyllite 2/1) Ca]-CaCO₃
- [(Phyllite 2/1) (Na, Ca)]CaCO₃

This involves "apparent bisiallisation" (Paquet, 1970) which can be related to several processes and soil types (Table 2).

Table 2 Main processes of plasmogenic weathering and their corresponding soils

Processes		Typical minerals and paragenesis	Soil main types	
Hydrolytic weathering Neof ormation plasma	Allitization	Gibbsite - ferric hydrates	Oxidic soils	Ferralsols
	Monosiallitzation	(I/I) Kaolinite - ferric hydrates	Ferralsitic soils s.l.	(Oxisols)
	Sialferrization	Kaolinite - 2/I smectite Fe (2/I Smectite Al-Fe)	"Sols bruns eutrophes" (Inceptisols) Vertisols	
	Andosolization	Allophane (halloysite)	Andosols	
	Apparent Bisiallitzation	(Phyllite 2/I) Ca - Humus (Phyllite 2/I) Ca - ferric hydrates (Phyllite 2/I) CaCO ₃ (Phyllite 2/I) Na - Sel	Brown soils - Inceptisols Ferralsitic soils Monllisols (Tchernozem) Halomorphic soils	
	Aluminosiallitzation	(Phyllite 2/I) AlOH - Fe ₂ O ₃ n H ₂ O	Podzolic soils	

Partial acidolysis leads to a redistribution of aluminium and individualisation of aluminised clays (Vermiculites A1 - Intergrades ...):

- [(Phyllite 2/1) AlOH]

It is the "aluminosiallitzation", which characterises the process of "feraluminisation" and the soils, as podzolic and brown acid soils.

¹ Phyllite 2/1 comprises illites, vermiculites, smectites of transformation, interstratified structures.

Geochemical Evolution and General Bioclimatic Condition

The previous data show that surface weathering can occur following three major processes depending mainly on the climatic and pedoclimate conditions which govern:

- the temperature of the weathering solutions,
- their composition,
- their volume and rate of circulation,
- the continuity or discontinuity of phenomena: uniformly dry or humid climates; climates with well defined seasons (drought or frost) ...

In this field, in order to give a general outline, it is advisable to refer to simple parameters, namely:

- the average annual temperature ($T^{\circ}\text{C}$) on which depends either the degree of hydrolysis or the composition and concentration of solutions (acidifying vegetation under cold conditions or high evapotranspiration under hot climatic conditions) or frost;
- the dryness index of de Martonne

$$\text{Indice d'Aridité } I_A = \frac{P}{T+10}$$

(where P and T are respectively the amount of rainfall and the average annual temperature) which distinguishes climates using the humidity-dryness criteria, for it takes account both of the amount of rainfall and of the duration of the dry season. Thus, it follows that:

for $I_A > 20$: uniformly humid and highly percolating environments geochemically characterised by aluminium and iron (Pedalfers of Robinson, 1949)

for $5 < I_A < 20$: environments with well-marked seasons geochemically characterised by basic cations (Pedocals), given that $I_A < 5$ corresponds to desert areas without weathering.

Table 3 shows that outside the desert areas, the temperature factor is a determining parameter concerning the types of weathering and the specific minerals.

Aplasmogenic evolution of acidolytic type (podzolisation) characterises mainly the cold regions.

Total hydrolytic weathering with neoformation of clays occurs in tropical regions provided that the seasons are humid. Thus, the nature of clays (kaolinite or smectite) depends on the value of the rainfall parameter.

When the temperature is intermediate, hydrolytic weathering is moderate and the neoformation phenomena are discrete. Evolution affects the pre-existing phylliteous phases and corresponds to a transformation process.

The great soil associations are distinguished by the humidity-dryness criteria. They go beyond the usual mineralogical aspects (based on the crystallochemistry of the solid phase) and are based on the whole "solid-water" system. Therefore, this phenomenon must be considered since it is the basic element of the relations between mineralogy and (micro) fabric.

Table 3 Main types of weathering and soil processes according to the climatic conditions.
(excluding desert areas and hydromorphic conditions)

$T^{\circ}C$	I_A $I_A > 20$ Humid environments (PEDALFERS)	$I_A < 20$ Dry environments (PEDOCALS)
$< 5^{\circ}C$ (Cold) Alteration Aplasmogenic weathering	Podzolization	—
$5-20^{\circ}C$ (Temperate regions) Partial weathering with transformation plasma	Feraluminization 2/I - AlOH	Fersiallitzation Calcosiallitzation Halosiallitzation 2/I - Ca/Na...
$> 20^{\circ}C$ (Tropical regions) Massive weathering with neoformation plasma	Ferrallitzation I/I - Kaolinite (gibbsite) - Iron hydroxydes	Sialferrization 2/I Smectite

MINERALOGICAL AND MICROSTRUCTURAL PROBLEMS

General Presentation

The water held within a divided solid such as a soil material is not free water. It is bound water due to the application of various forces, among which surface tension-forces being at the basis of capillary phenomena have a considerable influence.

This means that:

The water contained in the "solid-liquid" system must be considered as water subject to tension. The value of this tension which depends on the sizes of minerals and on the environmental conditions can be defined using what is called the "matrix potential".

According to Kelvin's law, the curvature of the water surface which is observed at the solid-liquid interface is a function of that tension. Thus, there is a strict relation between the state of water and the geometry of the "solid-liquid" system.

However, the microfabric characteristic of the soil materials is based on this geometry. But in this case, the classical mineralogical methods (for instance, x-ray diffraction) are not adequate to characterise a material; other mineralogical methods must be used (scanning electron - SEM - and transmission - TEM - microscopy; HRMET; small angle x-ray scattering (SAS) and applied to the humid materials in order to take account of the characteristics of the liquid phase.

If samples are taken from the three most frequent soil clays: kaolinites, illites and associated minerals, and smectite and if they are in contact with atmospheres whose relative humidities are variable, it can be shown that their water contents at equilibrium vary to a large extent with both the type of clays and the relative humidity (Fig. 4).

Water ratio (θ)

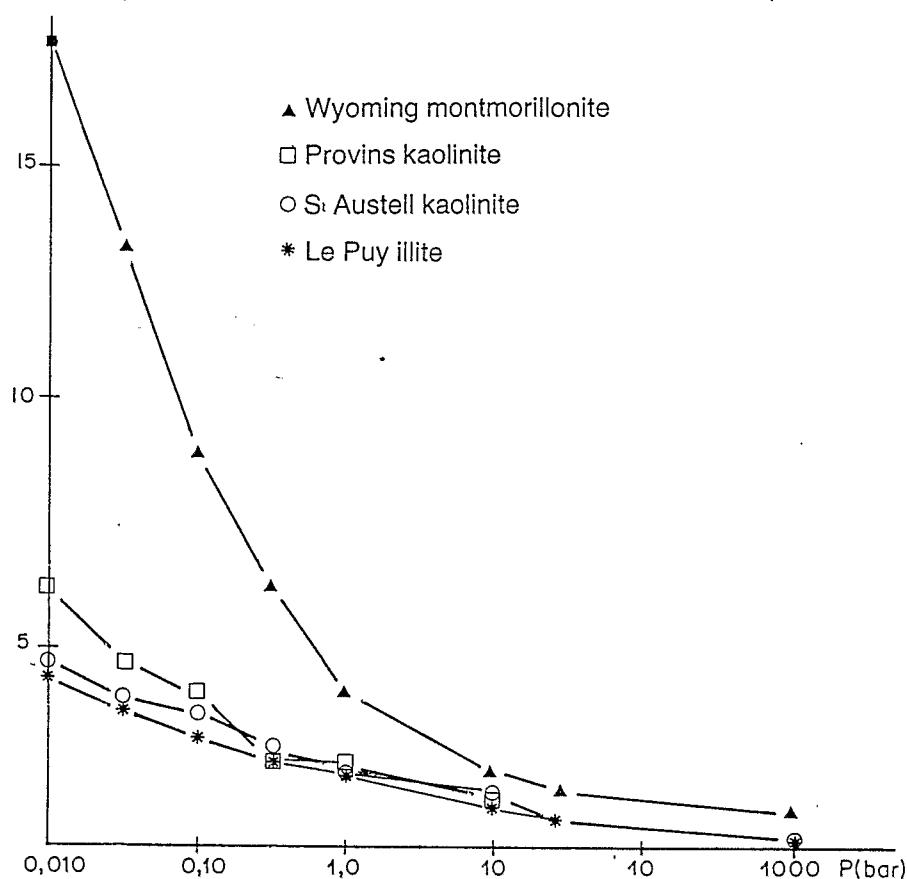


Figure 4 Water content of different clay minerals prepared with $\text{NaCl } 10^{-3}\text{M}$ at 32 mbar ($\text{pF } 1.5$) - First drying - with $\theta = \frac{V_w}{V_s}$ (V_w = water volume; V_s = solid volume)

However, the microscope studies made recently (Tessier, 1984; Oades, 1986) showed that:

The particles from the various clays have neither the same constitution nor are they of the same size (Fig. 5).

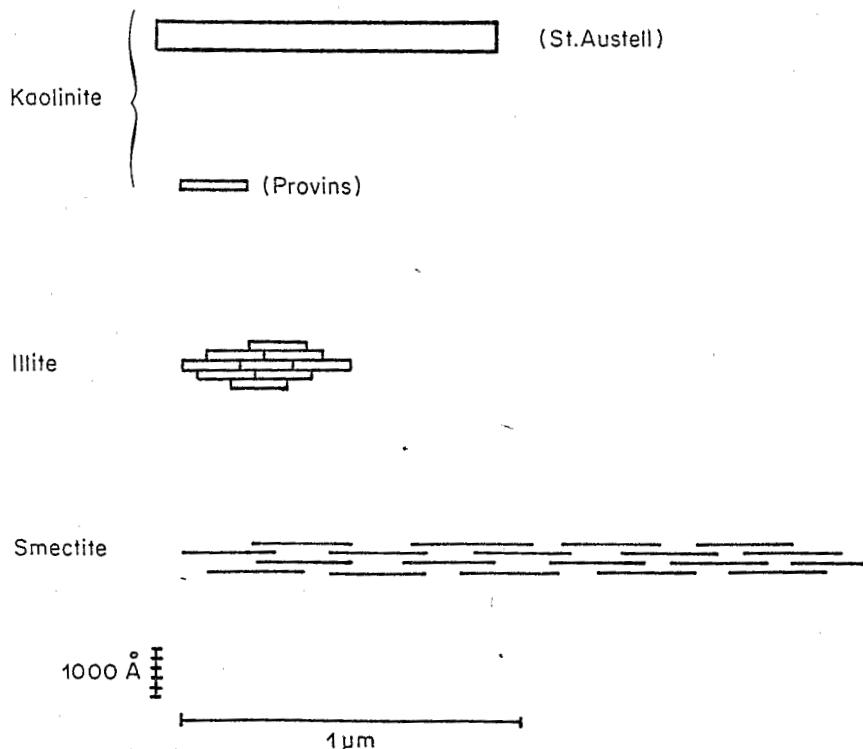


Figure 5 Aspect of the particles characteristic of the main clays

They do not show the same type of assemblage (Table 4).

- For kaolinites, particles are often isolated (crystallites)²; in the case of illites, particle aggregates are often observed (microdomains). The presence of an anhydrous interlayer leads to rigid particles.

- In the case of smectites, the basic element is quite different; it is a tactoid when layers are separated by a water film and it is a quasicrystal (Aylmore and Quirk, 1960) when the interlayer water is arranged round cations. Nevertheless, particles are flexible which leads to the face to face arrangement and to overlap (Fig. 6).

² The size of the crystallite can vary to a large extent with the soil genesis cf. Fig. 5 - Kaolinites from St Austell and Provins.

Table 4 Constitution of the main soil clays

Layer	Type of clay		Characteristic particle	Arrangement
I/I	Kaolinite		Crystallite (monocrystalline unit)	Cardhouse
2/I anhydrous	Illites (K) Aluminous intergrades		Microdomain (polycrystalline unit)	
2/I hydrated	Smectites	Ca	Quasicrystal	Network
		Na	Tactoid	

Quasicrystal (Smectite)

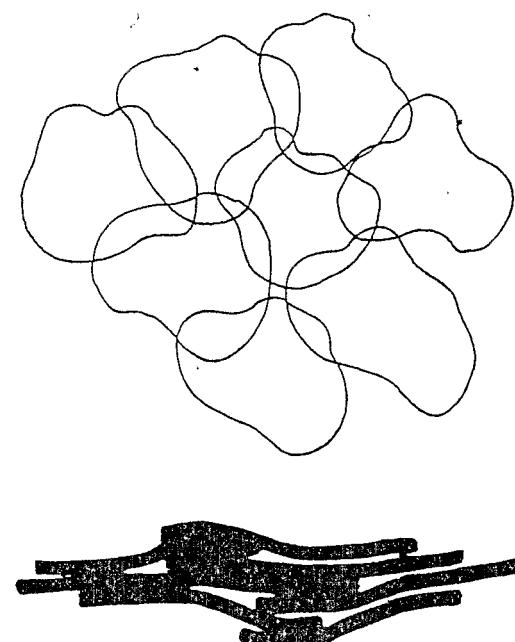


Figure 6 Schematic representation of a quasicrystal (Smectite)

The three-dimensional arrangement of these units in the "solid-liquid" system can be:

- either a discontinuous cardhouse juxtaposition as is the case of kaolinites and illites (Fig. 7) (Plate 1)

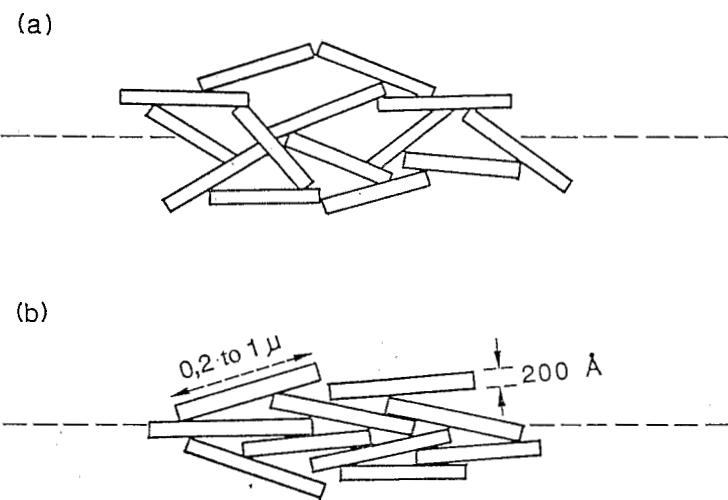


Figure 7 Type of kaolinic clay fabric
a) hydrated sample b) after drying (shrinkage limit)

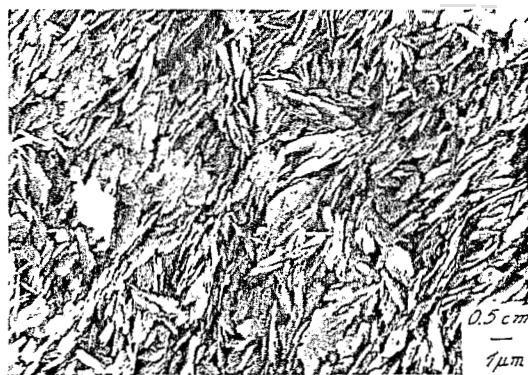


Plate 1 Scanning Electron Microscopy (SEM) for kaolinite of Saint-Austell
(Ca - $\text{CaCl}_2 10^{-3}$ M) at 1 bar ($pF = 3$)

- or a continuous arrangement for smectites (Fig. 8) which looks like a kind of "sponge", thus accounting for the large amounts of water likely to be held in this type of material (Plate 2).

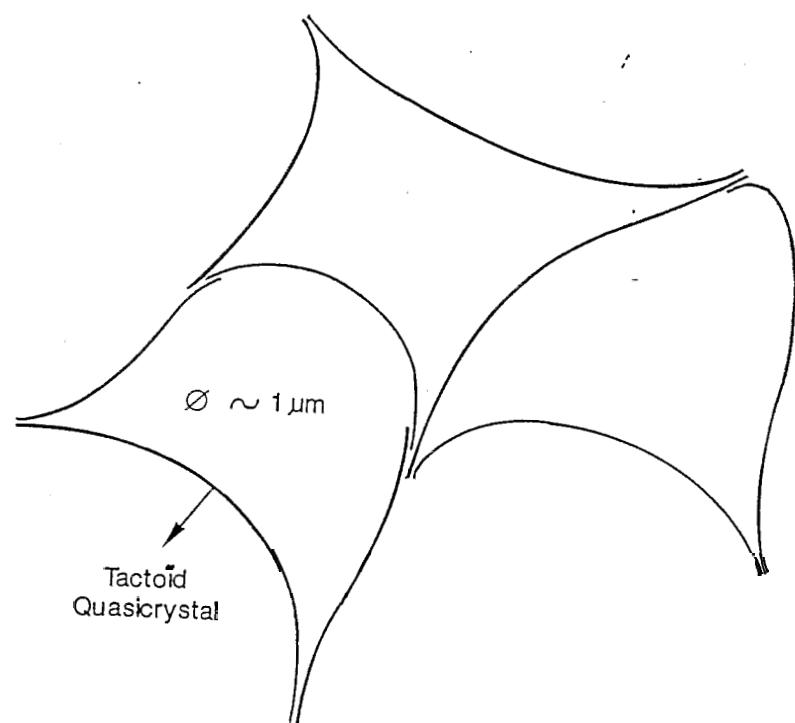


Figure 8 Arrangement of quasicrystals and aspect of a three-dimensional network in smectites

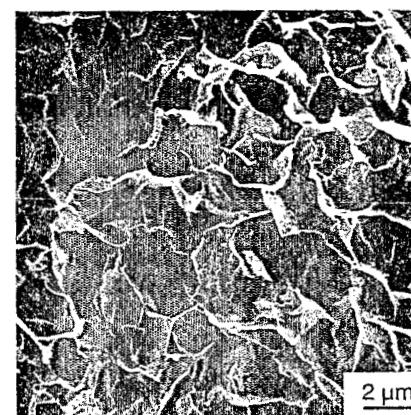


Plate 2 Scanning Electron Microscopy (SEM) for Na-Greek montmorillonite (NaCl_1M) at 0.032 bar ($\text{pF} = 1.5$)

However, all these arrangements result from two types of parameters which are linked respectively to the properties of solid and liquid phases.

Role of the Parameters linked to the Plasmic Solid Phase

They are of two types:

- either internal in relation to the electrical characteristics of the layers forming the elementary clay particles and to their crystallochemistry:

- or superficial, linked to the electrical characteristics of the particles observed in the soil samples.

Significance of the electrical characteristics of the layers: The electrical parameters involved are linked to the crystallochemical characteristics of the layers forming the elementary crystallites. From this point of view, the three clays studied correspond to three cases:

- in kaolinites, layers are neutral, ($z=0$); structural unit (U.S.) = 1/1.

- in illites and associated minerals, layers are charged electrically ($z > 0.6$), but the charge deficiency is balanced by cations (K) which are well fixed between the crystallite layers and are, therefore, non exchangeable; the structural unit (layer + interlayer) behaves as if it was still neutral; U.S. = 2/1 - K or 2/1 - Al OH.

- finally, in smectites, layers are charged electrically ($z < 0.6$), but the charge deficiency is balanced by hydrous and exchangeable cations, thus leading to the strong physicochemical reaction of the layer surface

$$U.S. = 2/1 - M(H_2O)_x$$

with M exchangeable basic cation: Na, Ca ...

and $(H_2O)_x$ water held in the interlayer and arranged by the field of forces of the layer.

In addition to these three types, in a number of temperate and subtropical regions there are 2/1 interstratified minerals which are intermediate between illites and smectites in which part of the balancing cations is fixed and the other is exchangeable.

Significance of the surface electrical characteristics of plasmic particles: All the real particles forming the solid phase show surface electrical charges due to the unbalanced (basal faces) and broken valences (lateral faces) linked to the limited particle size (while the layer shows theoretically an indefinite extent). These surface electrical charges are obviously neutralised by ions in solution which are generally interchangeable³, but they can

³ With basic cations; on the contrary, the hydroxy-cations Al and Fe are rapidly strongly fixed, thus leading to particles which are not very charged electrically.

be more or less abundant according to the type of clays: for instance, they are low for kaolinites with natural layers and they are higher for smectites and rather strong for illites.

General evaluation: The role played by the solid phase in the behaviour of the plasma-interstitial solution system is summed up in Table 5 which shows clearly that the surface hydrous reactivity is low in kaolinites; only slightly higher in illites and associated constituents; but it is very high in smectites.

Table 5 Characteristics of the interfaces in the three clays

Type of clay	Structural unit characteristic of the elementary crystallites (*)	Surface characteristics of plasmic particles		
		Electrical density	Contact area	Hydrous reactivity
Kaolinite	1/1	Very low	Low	Very low
Illite	2/1 - K	High	Low	Low
Smectite	2/1 - <u>Ca</u> (H ₂ O) _x Na	Mean	Very high (**)	High

(*) non exchangeable K; exchangeable Ca or Na

(**) external and internal surfaces

(i) with basic cations; on the contrary, the hydroxy-cations Al and Fe are rapidly strongly fixed, thus leading to particles which are not very charged electrically.

Therefore, the influence of the physicochemical and energetic characteristics of the solution is still to be considered on the hydration and arrangement of the different clay systems.

Role of the Parameters Linked to the Solid-liquid Interface and to the Interstitial Liquid Phase

These elements must be linked to the two components of any solution: water and its constituent aqueous solutions (Fig. 9). It is necessary to study successively:

- the significance of the chemical composition of the interface and of the solution,

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- the influence of the energy levels likely to be exerted on the liquid phase by studying only a few clay minerals among which smectites and kaolinites are the most characteristic.

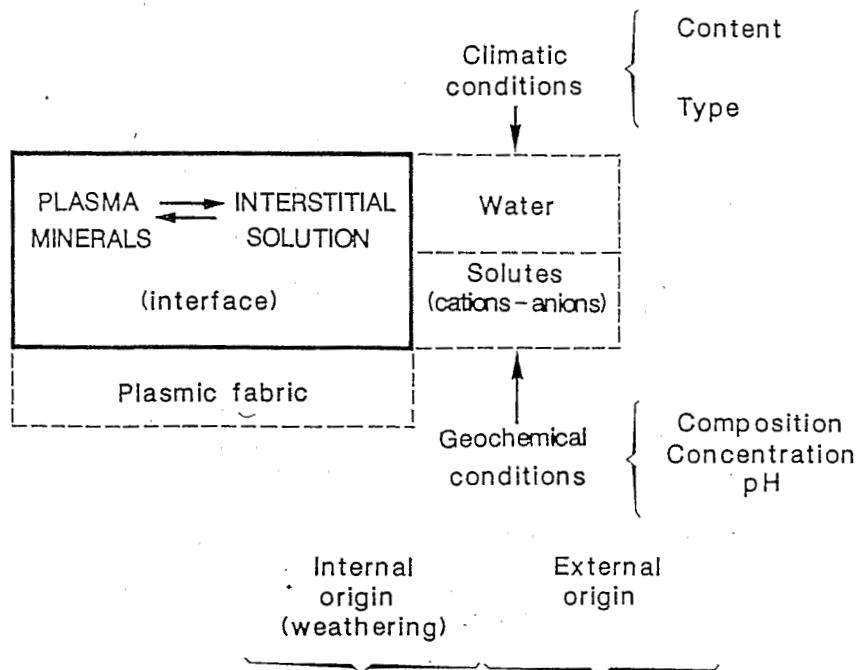


Figure 9 Influence of the various parameters on the plasmic microfabric

Significance of the chemical composition of the interstitial solution: Two cases can be distinguished depending on whether solutions are not acid and include basic cations such as Na, Ca, Mg, K or solutions are acid and include Al and Fe ions which are more or less hydroxylated and polymerized.

In the case of normal solutions (hydrolytic soil systems), two elements can be observed:

- the nature of the prevailing cation which will, therefore, balance the surface electrical charges;
- the concentration of the saline solution.

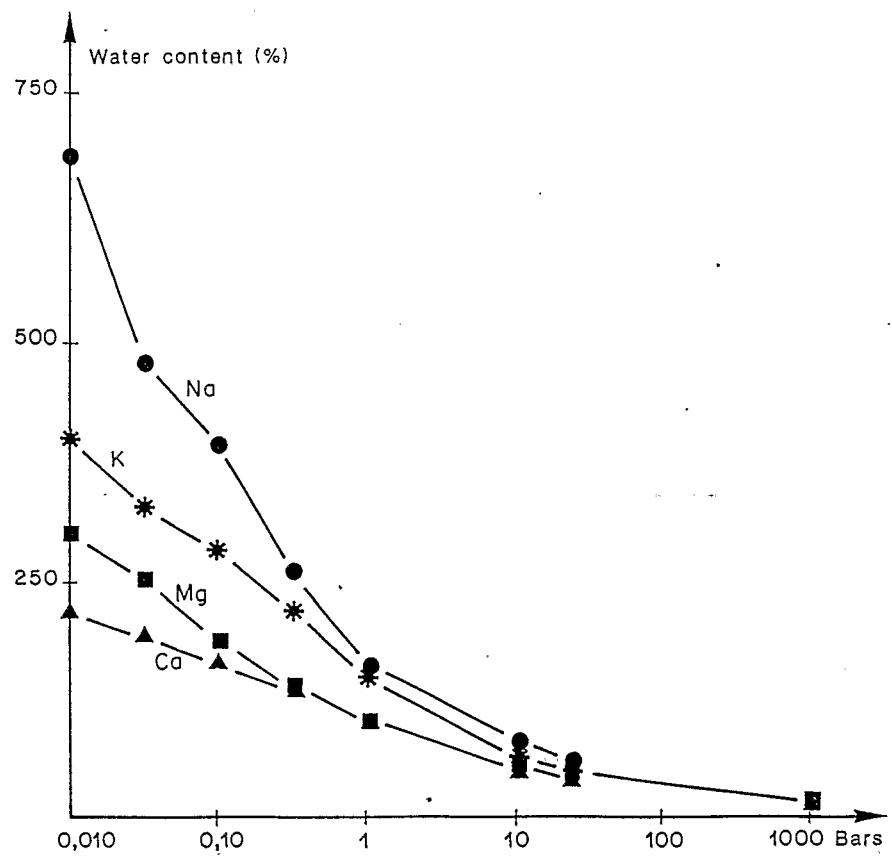


Figure 10 Evolution of the water content of smectites Na, K, Mg and Ca under dilute conditions ($10^{-3}M$) and during the first drying
Case of Béthonvilliers smectite

The first element is shown in Figure 10 (Tessier, 1984) where the Na clay samples are the most hydratable and the most swelling, while the Ca materials are the least hydrated (the hydration series $\text{Na} > \text{K} > \text{Mg} > \text{Ca}$ is obtained). The comparative observations made with SEM and TEM (Fig. 11) as well as the measurements made with small angle x-ray scattering (SAS) (Table 6) explain this situation: the Na samples show a network whose walls are composed of particles with a small number of layers arranged face to face ($M = 8$ layers), but with very thick films of interlayer water (diffuse layer) ($d_{001} = 85 \text{ \AA}$) (tactoid) (cf. Plate 3); to the contrary, the network of Ca smectites under the same conditions (diluted environment = $10^{-3} \text{ Mol.} 0.032 \text{ bar} = \text{pF} 1.5$) is composed of thick particles ($M = 55$ layers), where interlayer water arranged into layers ($d_{001} = 20$) (quasicrystal) (cf Plate 4).

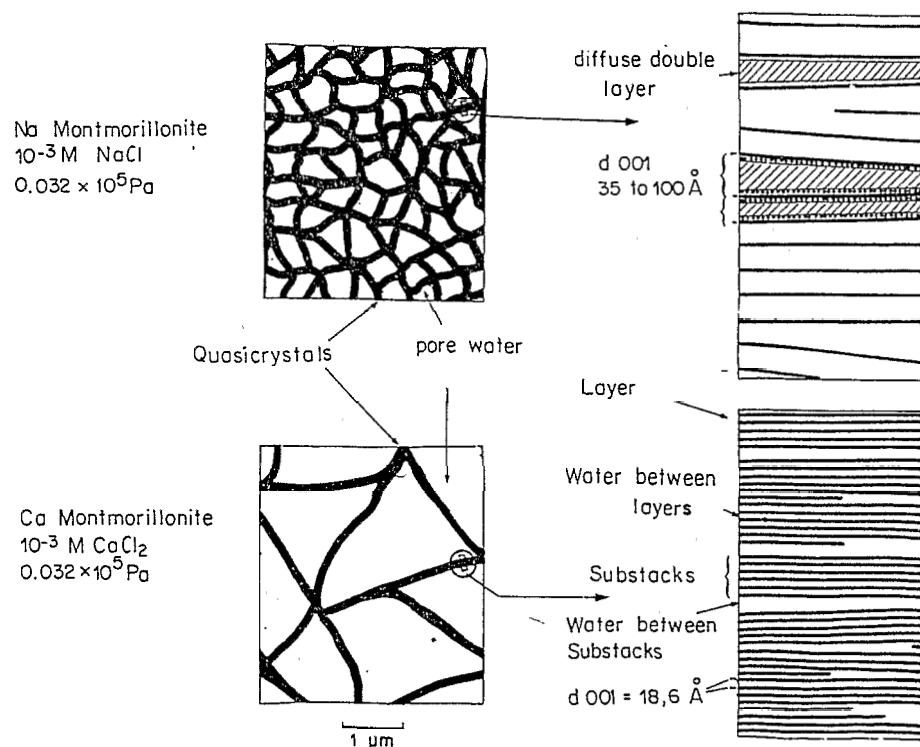
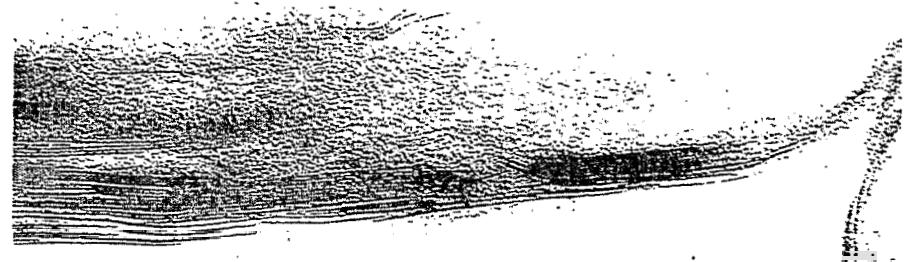


Figure 11 Schematic representation of the microstructure of Na and Ca Smectites prepared with dilute solutions (10^{-3} M)

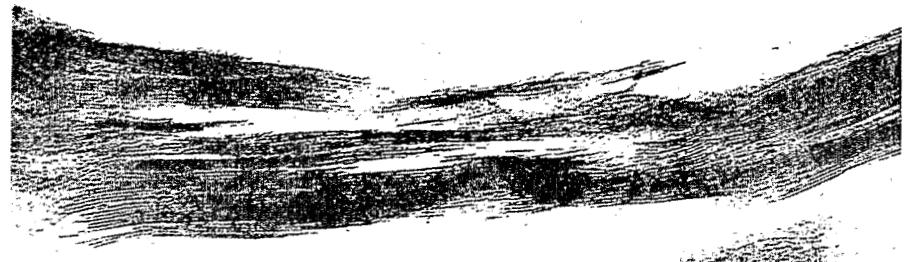
Table 6 Characteristics of hydrated smectites Na and Ca identified with S.A.S.
(first desiccation)

Samples		Mean number of sheets in the characteristic particle M	Equidistance d (001) \AA	Parameter $\frac{2}{d}$	Characteristic particles
10^{-3} M	Smectite Na	8	84.73	0.101	Tactoid
	Smectite Ca	55	20.09	0.046	Quasicrystal
M	Smectite Na	20	18.30	0.085	



50nm

Plate 3 High Resolution Transmission Electron Microscopy (HRTEM) for Na-Greek montmorillonite ($\text{NaCl}_2 \cdot 10^{-3}\text{M}$) at 0.032 bar ($\text{pF} = 1.5$)



50nm

Plate 4 High Resolution Transmission Electron Microscopy (HRTEM) for Ca-Greek montmorillonite ($\text{CaCl}_2 \cdot 10^{-3}\text{M}$) at 0.032 bar ($\text{pF} = 1.5$)

The second element is the concentration of the solution. The example selected (Fig. 12) shows that smectite Na at a high concentration (M) is much less hydrated and expanded than the same smectite Na in contact with a diluted solution, thus approaching smectite Ca. It is then characterised (Table 6) by networks composed of quasicrystals ($M = 20$) and showing an interlayer with 3 water sheets ($d_{001} = 18.3\text{ \AA}$).

However, the composition and concentration of the interstitial solution plays a significant role in the state of hydration of the clay systems. It is particularly obvious in smectites where these parameters have an influence on the size and the internal reactivity of particles: presence of tactoids with diffuse layers being highly evolutive for smectites Na under diluted conditions: development of thicker units with a film of interlayer water (quasicrystals) for smectites Na in concentrated solution or for smectites Ca.

In the case of acid solutions acidolytic or leaching hydrolytic soil systems, the cations characteristic of the liquid phase are aluminium and iron. But, they are hydrolyzable cations so that the pH of the solution becomes a significant parameter. The higher the pH is, the more hydroxylated and polymerised ions are. Therefore, there is between the simple

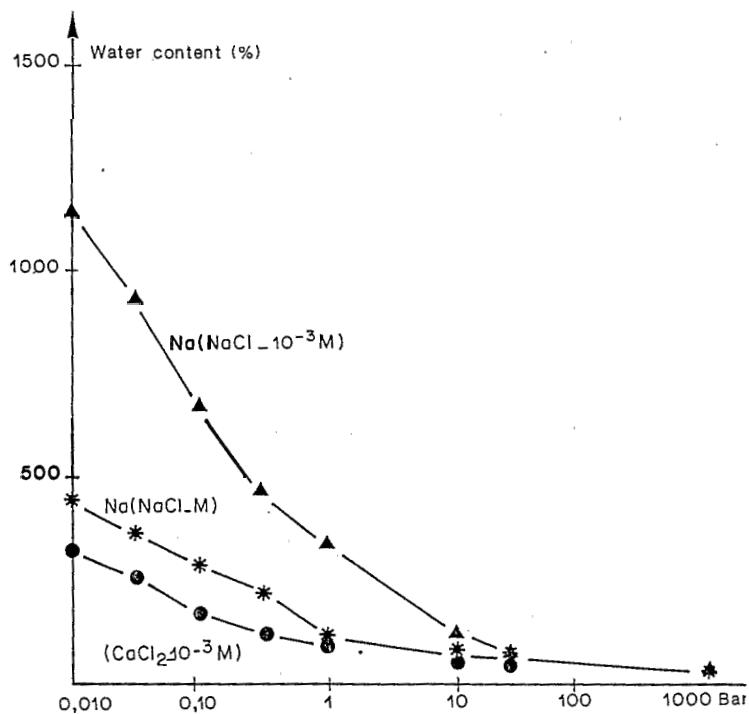
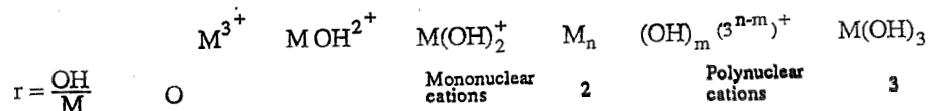


Figure 12 Role of the solution concentration (NaCl) on the evolution of the water content

hydrated cations and the precipitated hydrates the whole series as follows:



The experimental results obtained by Robert et al (1983) are given in Figure 13. Thus, one can note in smectites as compared to the sodic samples that:

- hydration is reduced with simple and mononuclear ions ($r < 2$) which are interlayered and not very hydratable;
- it increases with polycations which are fixed only on the external surfaces; but they are very hydratable in their transition phase.

Such results are interesting in so far as, under similar conditions, they permit comparisons with samples saturated with basic cations. Nevertheless, they are not adequate to account for what really occurs under natural conditions due to the fact that these different types of ions develop easily in soil leaching and tend to give rise to hydrophobic and not very hydrated structures easily in soil leaching and tend to give rise to hydrophobic and not very hydrated structures.

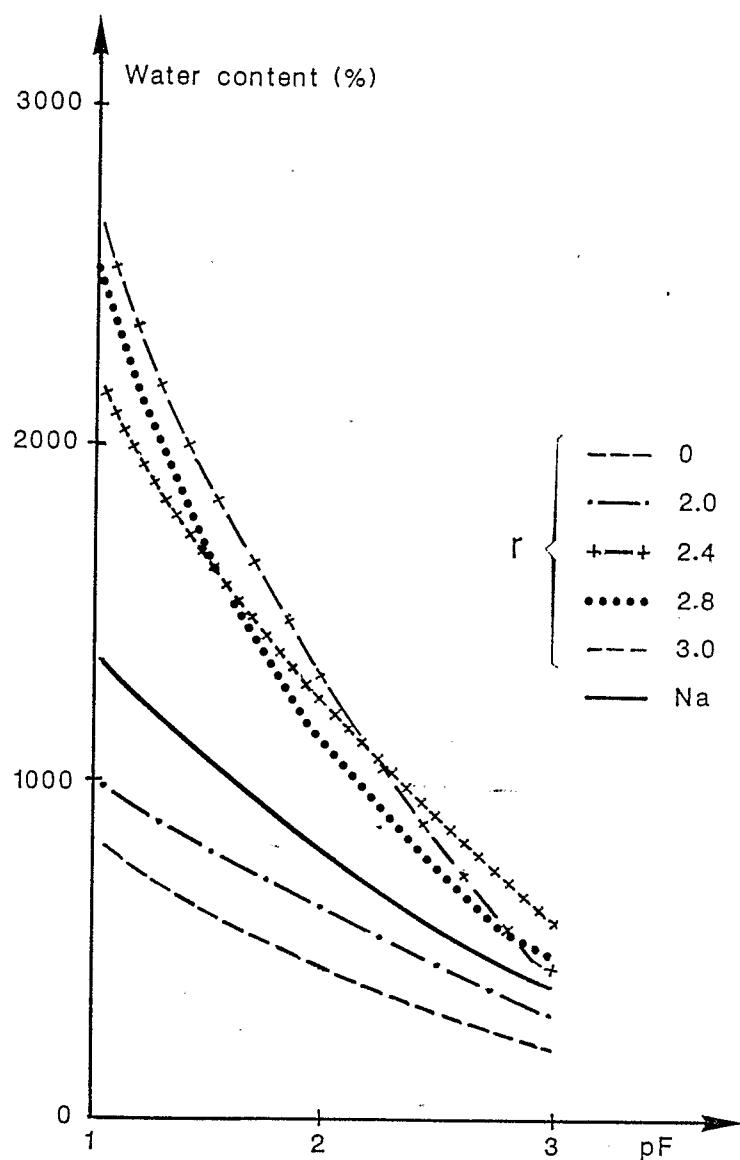


Figure 13 Evolution of the water content of smectites Fe for different values of the ratio $r = \frac{OH}{Fe}$ (First drying)

For instance, smectites A11-OH (intergrades) are composed of particles which are more similar to the illitic microdomains than to quasicrystals and they are, therefore, more rigid and less active.

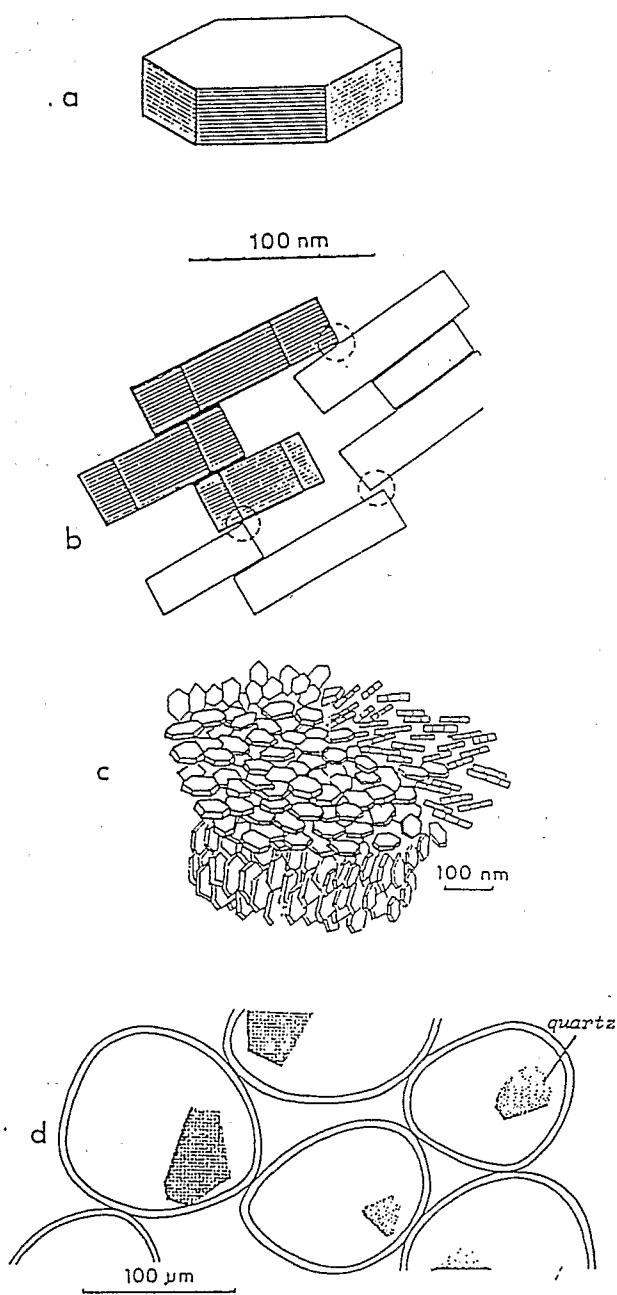


Figure 14 The different levels of the kaolinite in an oxisol from Casamance (Senegal)
a) crystallite b) polycrystalline particle c) oriented microdomain
d) micro

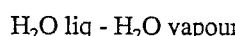
This phenomenon is still obvious with ferrized kaolinites which are found in oxisols. Then, there are no isolated crystallites as in the case of kaolinites Na and Ca, but as was shown by Cambier (1986), there are polycrystalline particles (a few crystallites linked in a parallel direction to each other and bonded by Fe units) which form larger units called oriented microdomains (1 μm). The latter form the microaggregates (100 μm) characteristic of the farinaceous structure of these oxisols; each micronodule being, in fact, formed by the isometric juxtaposition of disorganized microdomains (Fig. 14).

Significance of the stresses likely to influence the aqueous phase of the interstitial solution.

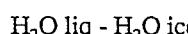
Origin of stresses

The stress levels which affect the soil aqueous phase (soil water potential) result:

- either from physical phenomena linked to the reactions of change in the state of interstitial water:



and



and which are generally expressed by the notion of matrix potential;

- or from osmotic phenomena related to the ionic strength of the interstitial solutions and which are at the origin of the osmotic potential.

These two components of the water potential are generally measured in the same units: bars, centimeters and pF (logarithm of suction expressed in cm). But they can also be expressed by the activity of the soil water (a_w), since the latter is thermodynamically linked (cf G. Bourrie and G. Pedro, 1979) to:

- the relative humidity of the ambient atmosphere
- its freezing point
- and the osmotic pressure of the solution

Table 7 sums up the relations between the energetic characteristics of the soil water on one hand and the climatic data (relative humidity - freezing point) on the other. Table 8 includes the values of the osmotic stresses linked to a number of salts likely to be present in soils.

Role of stresses on the hydration and microfabric of plasmic samples and their influence.

Table 7 Relations between the stress or the water activity with the relative humidity at 25°C and the freezing point.

Stress expressed by			Relative humidity (%)	Freezing point (°C)	Water activity a_w
Bars	Cm of water	pF			
0.010	10	1	99.99	-0.01	0.99999
0.1	100	2	99.99	-0.08	0.99992
1	1000	3	99.93		0.99927
10	10000	4	99.29		0.9927
15.8	16000	4.2	98.88	-1.1	0.9888
100	100000	5	93.12	-10	0.9312
		5.52	78.98		
1000	10^6	6	49.03		0.4903
		6.2	32.32	-90	
		6.4	16.69		
10000	10^7	7	0.08		0.0008

Table 8 Osmotic pressure of the various saline solutions observed.

Compound	Concentration	Ionic force	Osmotic pressure (pF)
CaCO ₃	10^{-4} M (saturation)	0.0004	1.14
CaSO ₄	10^{-2} M (saturation)	0.04	3.14
CaCl ₂	10^{-3} M M	0.003 3	2.13 4.92
NaCl	10^{-3} M 10^{-1} M M 6M (saturation)	0.001 0.1 1 6	1.68 3.67 4.67 5.51

This role already has been dealt with implicitly (Fig. 5, 11, 13) since consideration has been given above to the variation in the hydration and the microfabric in relation to the relative humidity of air. Therefore, it is sufficient to describe the "climatic" (relative humidity of the ambient atmosphere - temperatures lower than 0°C) and "osmotic" conditions in order to define the amount and state of water present at equilibrium in any plasmic material.

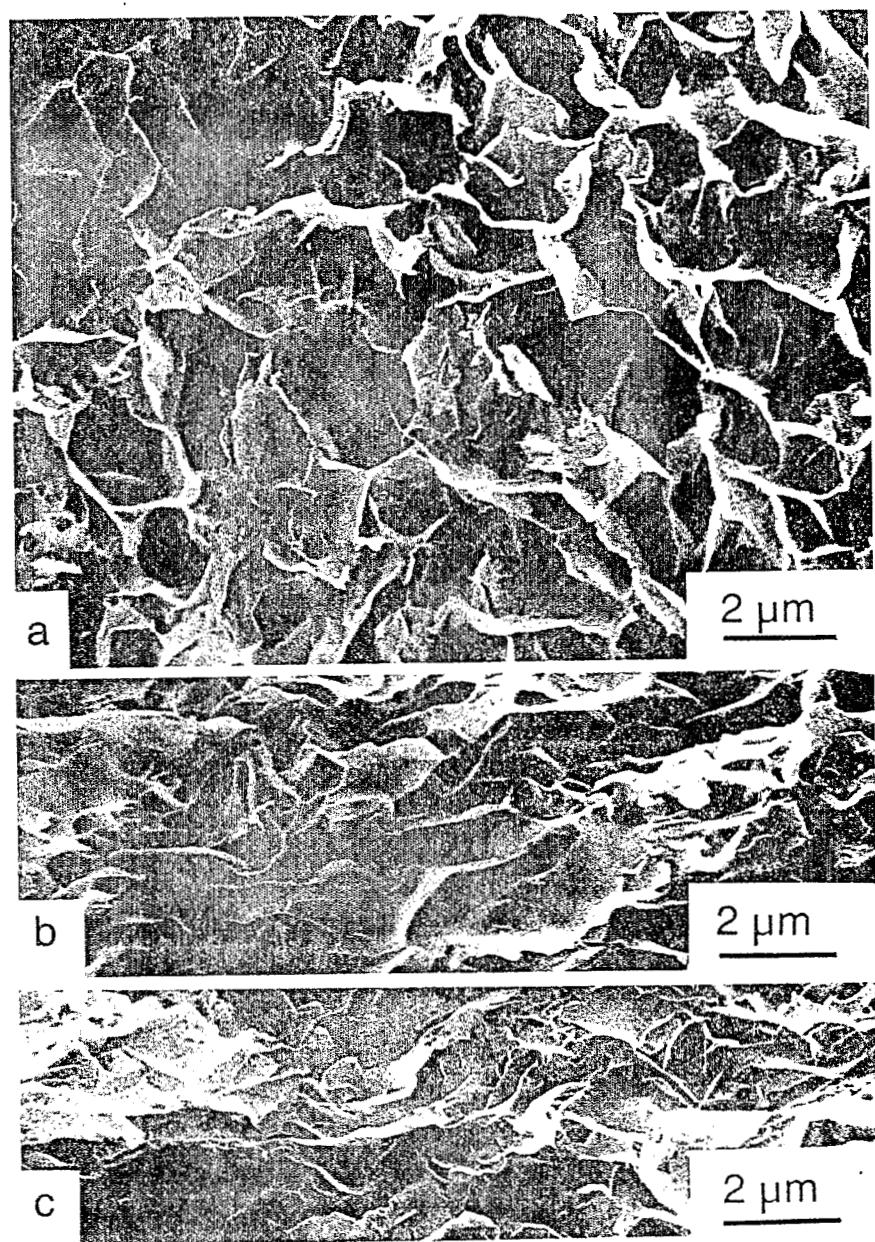


Figure 15 Evolution of the volume and the bulk density during drying
smectite Ca

V = Sample volume (cm^3g^{-1}) W = Water content (%)

a) 0.032 bar ($\text{pF} 1.5$), $W = 369\%$, $V = 4 \text{ cm}^3/\text{g}$

b) 1 bar ($\text{pF} = 3.0$), $W = 114\%$, $V = 1.5 \text{ cm}^3/\text{g}$

c) 10 bars ($\text{pF} = 4.0$), $W = 82\%$, $V = 1.2 \text{ cm}^3/\text{g}$

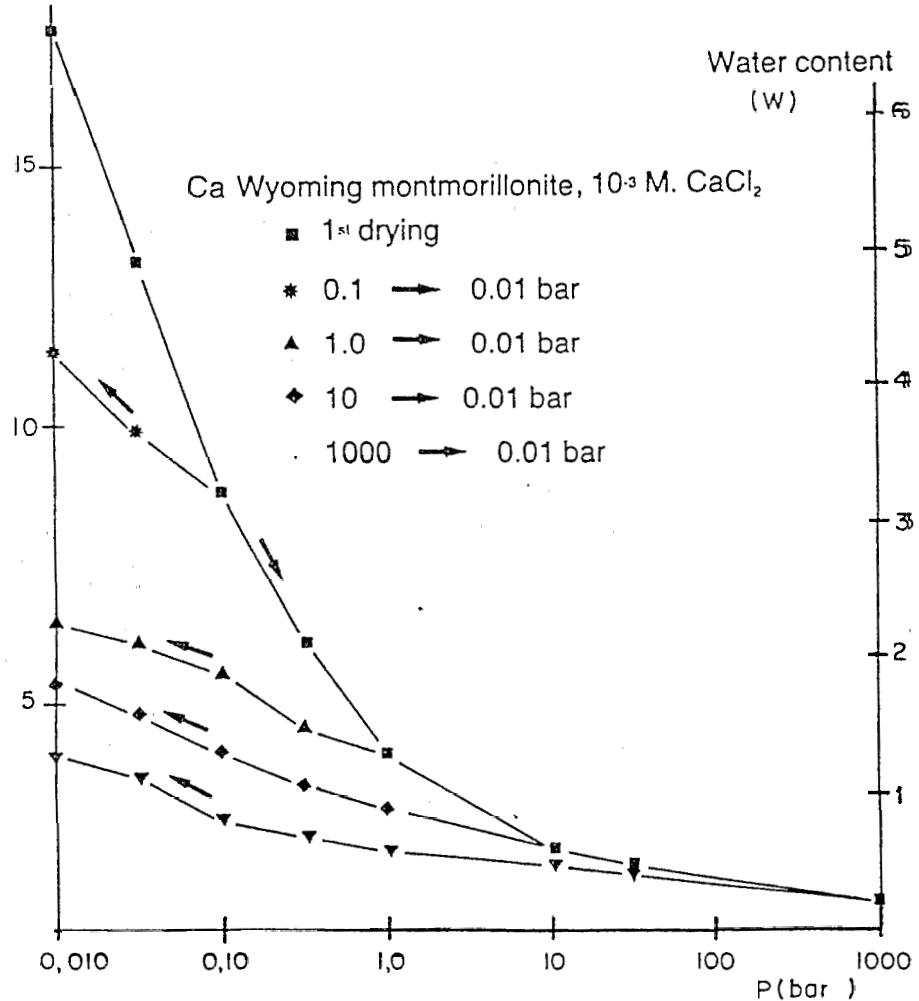
Water ratio (δ)

Figure 16 Hydration - Dehydration behavior of Wyoming Ca montmorillonite in 10^{-3} M CaCl_2 - with $\delta = \frac{V_w}{V_s}$ (V_w = water volume; V_s = solid volume)

Figure 15 (Tessier, 1984) illustrates clearly this problem.

The energetic data (intensity, desiccations, alternation between desiccation and wetting) have also a considerable influence. Figure 16 shows, for instance, that the intensity of desiccation is strong before any rehydration for a smectite Ca. This can, be applied to the other clay minerals - kaolinites and illites. Each desiccation led to a partially "irreversible" process in the clay system by aggregating, for instance, the initial particles of the hydrated material (Tessier, 1980). The data obtained with SAS (Table 9) enabled measurement of

Table 9

Characteristics of Wyoming montmorillonites
(first desiccation and after rehydratation)

Echantillons	pF			d ₂
Montmorillonite Ca (10 ⁻³ M CaCl ₂)	3	55	20.09	0.046
	6-3	170	20.15	0.066
Montmorillonite Na (10 ⁻³ M NaCl)	3	8	84.73	0.101
	6-3	20	46.55	0.085

these regroupings for smectites which can be illustrated schemitically, as is the case in Figure 17.

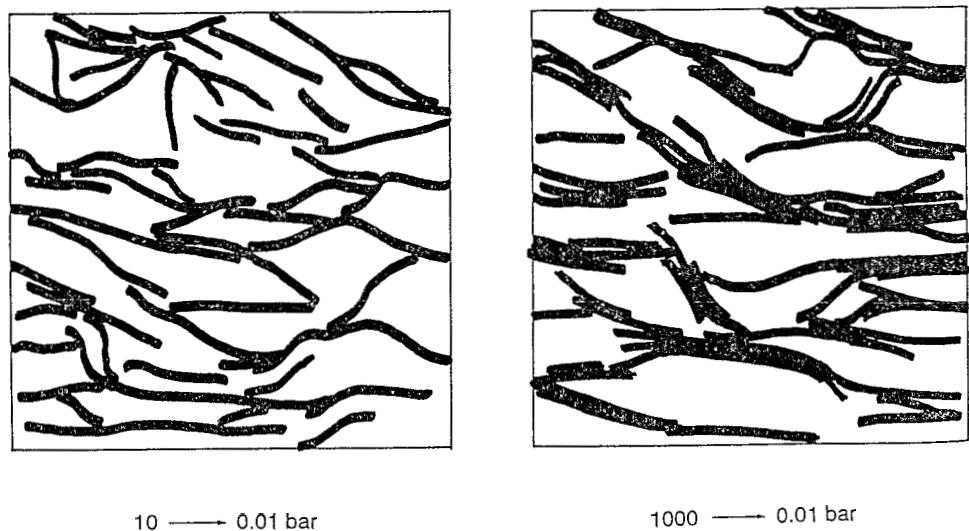


Figure 17 Type of rearrangement of a smectite Ca network during dehydration

Microstructural Evolution and General Bioclimatic Conditions

The relations between the identification of a particular type of microfabric and environmental conditions are much less obvious than those concerning the geochemical problems. The researches in this field are more recent and much remains to be done. It was shown above (Table 3) that the great soil systems could be related to the humidity-aridity criteria which are, in fact, the key parameters in the energetic characterisation of plasmic systems.

If one refers to the data of Table 3 which are defined according to the value of the "Indice d'aridité", it is now possible to account for the energetic specificity of each of the two great systems:

- The first system ($I_A > 20$) gathers the soils which show low energy levels (pF 1-2) due to the still high relative humidities of air and to the temperatures generally exceeding $0^{\circ}C$. Therefore, they are, under non hydromorphic conditions, highly percolating humid soils where solutions are diluted, highly acid and where the elements Al and Fe play an essential role ("Pedalfers"), mainly in the interfaces (microfabric) due to their hydrophobic character.

Particles combine to produce arrangements with fixed geometry which generally appear in the form of volumes having minimum areas and sizes consistent with the prevailing physical phenomena (surface tension), thus accounting for the formation of micro-aggregates.

- spherical (or subspherical), the sphere being the volume with the least surface area for a given amount of matter,

- with a size close to $100 \mu M$.

In these low-energy environments, there is always a convergent evolution in the microfabrics, since they are observed in ferrallitisation (kaolinite-ferriferous compounds): oxisols, in feraluminisation (minerals 2/1 - $AlOH$): acid brown and cryptopodzolic soils (Aurousseau, 1985; Van Oort, 1984) and in andoxolisation (alophane): andosols (Rosello, 1984).

- The second system concerns the high-energy environments ($I_A < 20$) due to the existence of well-marked seasons (variation of pF between 0 and 6 over the year) or of saline concentrations (halomorphic zones). They are "Pedocals" characterized by more concentrated solutions and basic cations: Na, K, Mg and Ca at the interfaces resulting in the development of arrangements with variable geometry, with shear strengths due to the swelling observed at each new seasonal wetting and with coarse structures (macrostructures).

Therefore, the distinction between the two great soil-systems based on the aridity-humidity criteria seems to be, nowadays, essential concerning the pedogenic phenomena.

CONCLUSION

This paper aimed at showing that it was possible to give a coherent outline of pedogenic problems. But it becomes necessary to consider:

- all the environmental factors and all the climatic parameters, and not only the classical "climatic factors" (P_{mm} , $T^{\circ}C$) but also the "climatic stresses" (dry season or frost, variation in the relative humidity of air);
- all the characteristics of the solid-solution system, namely
 - the mineralogical and biogeochemical characteristics of the solid phase
 - the physico-chemical characteristics of the liquid phase and the interface,
 - the morpho-structural characteristics of the whole system.

Thus, it is clear that, under given climatic conditions, evolution leads to a quite coherent mineralogical constitution (plastriaa) and microfabric.

Any variation in the environmental conditions must lead to a change in the system; however, the result depends on whether energy stresses are involved or how the usual major parameters P and $T^{\circ}C$ are modified.

- If the energy stresses vary little, the microfabric remains stable; therefore, the internal composition of the solid phase (crystallochemistry) will tend to change with the intensification of the thermohydric variables;

- On the contrary, if the climatic stresses are subjected to variations in one direction or in another, it is possible to change easily the microfabric and therefore, the interfaces without causing considerable crystallochemical changes. For instance, this phenomenon has been observed in the tropical zones where the transformation of a humid tropical climate into a dry tropical climate with well marked seasons leads to the transformation of a kaolinitic oxisol (sol ferrallitique) into a kaolinitic ultisol (sol ferrugineux tropical) (Chauvel, 1976).

It is obvious that much remains to be done in this field of research; nevertheless, the "coherence" of the soil phenomena seems, nowadays, to be very promising.

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