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APPLICATION OF HIGH RESOLUTION ELECTRON MICROSCOPY TO SOIL CLAY ORIGIN AND ORGANIZATION IN A TEMPERATE CLIMATE

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Abstract: Soil clay most commonly encountered in temperate regions can be defined as a complex assemblage resulting from combined degradation and aggradation phenomena during paleoalteration and present day weathering. High resolution and analytical electron microscopy have been used to investigate clay texture and crystallochemistry of primary minerals and their alteration by products. Illites occur under two main facies: micromica and illite *stricto sensu.* Micromicas of clay size come from the microdivision of former larger micas, involving both mechanical and chemical alteration processes. Their composition is close to that of micas with a high K content (around 10 %). Illite *stricto sensu* is from sedimentary origin, typically barrel shaped, with 5 - 7 layers and heterogeneous in composition with a moderate K content (around 7 %). Mixed layer clays are also of two different types. The structural type, either ordered or semi-ordered is produced by alteration of micas. It displays an irregular opening of interlayer spaces due to the leaching of K. The textural type is disordered and formed by subparallel aggregation of small size units (1 - 4 layers) into quasicrystals. Clay organization appears to be closely related to the origin of the clay minerals.

Key words: clay minerals, electron microscopy, ultrastructure, soils, sediments.

Introduction

Extended studies have been carried out by soil scientists in order to correlate clay content, mineralogy and soil properties. But the interpretations fail to explain the mechanism of evolution of clay minerals and predict further changes.

We know that clays can originate in soils through three different mechanisms: **a** - inheritance from parent material, **b** - neogenesis/neoformation and **c** - transformation of pre-existing layer silicates. As temperate regions located in the northern part of Europe and the USA have relatively low precipitation and temperature, weathering is considered moderate and clay genesis mainly governed by bisialitisation (Pedro & Siefferman 1979). In other words 2 : 1 phyllosilicates are either stable inherited material or neoformation products, but their origin and the processes of evolution are still difficult to understand, since clear criteria are lacking.

Transmission electron microscopy (TEM) has been available since the forties, but most studies of soil clays have been based on data obtained by classical methods such as X-ray diffraction (XRD) and wet chemistry. The interpretations are very rarely supported by electron microscopic data. The study of the organization at a micrometric scale of randomly oriented clay minerals started with embedment in resin. Primarily developed for curing biological samples (Spurr 1969), the embedding technique for hydrated samples has been used by mineralogists and clay scientists and can be applied either to clay fractions, soil lumps or rock chips. This type of preparation can also include the fixation of organic components (Villemin & Toutain 1987).

In this study, high resolution and analytical electron microscopy (HRTEM and AEM) have been used to investigate *in situ* transformation processes at a nanometric scale. This method essentially provides pinpoint information on some genetic associations which can be correlated to more general data obtained by XRD, electron diffraction, or wet chemistry. Some criteria based on the clay organization at the layer scale have been selected to trace the evolution of 2 : 1 minerals from their origin to their present day state. The same criteria could be useful to describe the mechanisms of transformation and predict the general trends of evolution.

The soils investigated cover a large range of the most widespread bedrocks present on the French territory, which are essentially of acidic type, plutonic or from detritic origin. The soils were developed from granites or from clayey and silty sedimentary rocks. Soils developed from basic bedrocks such as basalts were not investigated.

Methodology

Material

The sampling concerns surface horizons developed from different bedrocks: clayey sedimentary rocks (Triassic to Quaternary formations), silty sedimentary rocks (mainly from Quaternary eolian deposits) and granitic rocks from the Alps and Central Massif. Samples come from A and B horizons of soils. Some arenes, coarser fractions or C horizons were also studied for comparison. Two glauconite-type illites formed in lacustrine environments were taken as references.

Clay fractions have been separated from coarser fractions by dispersion in pure water with mild mechanical agitation without any chemical dispersive agent. Extraction of the $<2\,\mu m$ fraction was done by sedimentation after Stokes' law at room temperature followed by centrifugation (Robert & Tessier 1974). The day pastes were randomly oriented and conserved hydrated.

Sample preparation

An agar coating is recommended as a preliminary step before further treatments and permits to handle the sample with tweezers without deformation. The sample is dipped in 2 % agar around the point of solidification, usually below 40 °*C*. The agar cake constitutes a porous net around the sample and permits all kinds of exchanges with solutions at the same ion diffusion rate than in pure water. The coated sample is then put in a pF tube for 24 to 48 hours for equilibration at a given water potential (Tessier & Pedro 1987). The agar coating preserves the sample fabric during rehydration, avoiding splitting and dispersion phenomena which tend to happen if the cohesion forces are drastically modified during changes of osmotic and matricial potentials.

The next step consists in the dehydration and impregnation of the sample. Dehydration is the replacement of all the water molecules by acetone or alcohol, and impregnation the replacement of the dehydrating agent by resin. In order to ensure rapid and complete substitution, the sample volume should be of the minimum size required for further microscopic study, i.e. 1 *mm*, and the agar coating reduced to a thin shell. As a consequence, the resin penetrates all interlayer spaces of swelling minerals as well as the intra-aggregate microporosity and the inter-aggregate macroporosity. The impregnated sample is then embedded in a block of resin.

After polymerization of the resin, the sample is sliced in ultrathin sections in a REICHER ULTRACUTE microtome; the maximum acceptable thickness for HRTEM is 50 *nm*.

High resolution and analytical electron microscopy

HRTEM and AEM data were obtained in a PHILIPS EM 420 microscope equipped with a LINK AN 10000 microanalyser. The device permits the acquisition of HRTEM and AEM data sequentially on the same area of interest, that is on individual particles or domains of 0.1 to a few micrometers.

One dimensional HRTEM has been performed, selecting (001) reflections and bright field lattice fringes at 120 *kV* and a 40 *μm* aperture eliminating lattice periodicities smaller than 0.35 *nm*. In the proper orientation, electron diffraction patterns display symmetric dots of equal intensity along the (001) row while the lattice fringes of silicate layers parallel to the optical axis of the microscope show a maximum of contrast.

Photographs have been taken in underfocus conditions, the optimal defocus being determined between 100 and 150 *nm* in order to visualize both 1.0 and 1.4 *nm* periodicities simultaneously with a favorable transfer efficiency in the microscope. Zero focus is obtained on the carbon support.

The thickness and number of interlayers inside sets of strictly parallel lattice fringes can be measured with a 10 % relative error on negatives using a binocular with a micrometric scale. Stacking

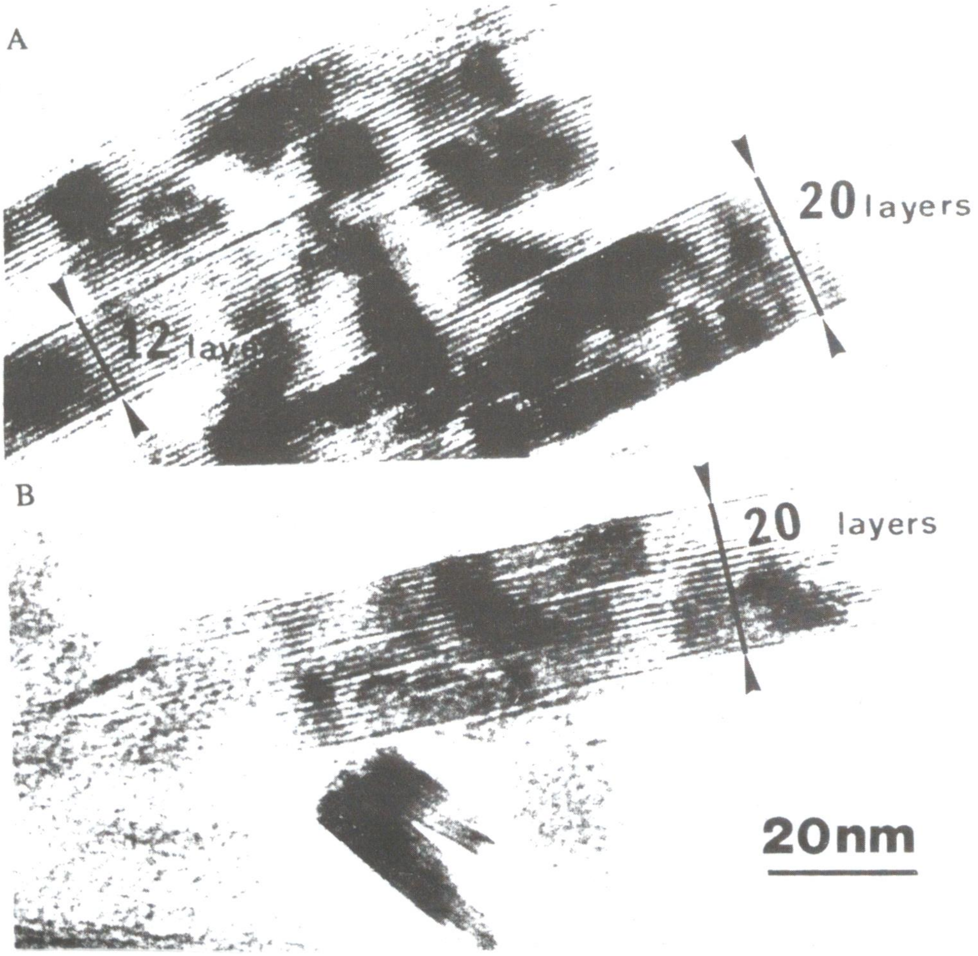

Fig. 1. Micromica in coarse fraction of silt from Ménard (France). **A** - detail of bands in a 10*μm* grain; **B** - clay particles in a process of microdivision.

periodicity is calculated from measures made between two intensity maxima corresponding to the center of layers. After impregnation with resin as described above, swelling interlayers of smectite type will be filled with one sheet of resin and display 1.36 *nm* periodicity of layers against 1.0 *nm* for collapsed interlayers (Środoń et al. 1990, 1992).

Elemental composition of individual particles or domains were obtained by X-ray analysis using an energy dispersive system (EDS) with a windowless Si/Li detector. The analyses were performed in transmission mode and convergent beam, using two criteria for minimizing irradiation damages, i.e. ion mobility and mass loss. The spot size was adapted to the selected area and the beam intensity reduced to the value required to get a count rate not exceeding 1000 counts/sec., which combined with an acquisition time of 100 sec. gives the minimum counting statistics corresponding to an acceptable relative error of 10 % for major elements.

Results

In all types of surface formations, the mineralogy of the clay fraction can be depicted as a mixture in variable amounts of the three main types of phyllosilicates: kaolinites, 2 : 1 non-swelling minerals and 2 : 1 swelling minerals.

Kaolinites

Kaolinites are widespread in soils formed on sedimentary rocks. They are abundant in Tertiary sediments (flint clays or ancient alluvia) as inherited weathering products formed under a tropical climate. Unlike reference kaolinites described in literature which exhibit typical platey and well developed rigid morphologies with hexagonal boundaries, they generally are of small size of about 100 *nm* in diameter with smoothed edges and wavy sections of 2 to 10 layers. They always contain traces of Na or K as impurities (Robert et al. 1991a).

In recent formations such as arenes developed from crystalline rocks they may form micrometric aggregates of small crystals resulting from the microdivision of larger particles also containing traces of Na and K. On recent weathering we can often notice the formation of halloysite and not of kaolinite.

Non-swelling 2 : 1 phyllosilicates

Non-swelling 2 : 1 phyllosilicates present a large suite of morphologies and chemical compositions. They can be classified into two groups (Robert et al. 1991a) with respect to their crystallochemical properties: micromicas and illites stricto sensu (AIPEA Nomenclature Comitee report, Bailey et al. 1984).

Micromicas: From the point of view of crystallochemistry, micromicas present in the studied samples are most of time dioctahedral (muscovite type) more rarely trioctahedral (biotite type), but always with a high K_2O content (around 10 %). Micromicas are abundant in the coarse clay fraction of soils developed from granitic rocks and in soils developed from silty rock, as well preserved particles with large coherent electron scattering domains of up to 100 parallel layers. They usually display bands of 12 to 20 layers which may be interpreted as primary zonation or growing defects (Fig. 1a). Micromicas also occur as smaller particles of 12 to 20 layers, distributed within an amorphous mineral groundmass (Fig. 1b). These may originate from the disaggregation of the larger banded micromicas.

Micromicas are also found in lower amount in soils developed from sedimentary rock as particles of less than 1 *µm* in diameter, with very sharp edges and a thickness of at least 10 layers. They often display surface indentation and channels across the particles which can be interpreted as dissolution features (Fig. 2a). Such features are widespread in soils derived from alluvium of acidic composition.

Illites *stricto sensu*: The chemical composition of illites present in the samples varies from one sample to another and within one sample, particles have different compositions. The mean K_2O content is around 7.5 %. Such illites occur in almost all soils developed from sedimentary rocks. They are of less than 0.5 *µm* in diameter, most of the time less than 0.1 *µm*, with a small number of layers usually not exceeding 5 layers in coherent domains. HRTEM pictures of perpendicular sections of these particles reveal a considerable density of stacking faults of corner type and/or random shift or rotation defects in the stacking direction (Fig. 2b). The barrel shape is another typical feature. This shape does not permit coherent stacking of several particles either by face to face or edge to edge grouping. These illites stricto sensu are abundant in Keuper formations and more recent lacustrine or marine sediments of Upper Tertiary or Quaternary age.

Their barrel shape morphology is similar to those of illites from lake sediments like the illites from Le Puy (France) and Muloorina (Australia) (Fig. 2c). The chemical composition of these lake illites (Duplay 1988; Norrish & Pickering 1983) compares well with illitic chemistry as determined by Weaver & Pollard (1975) and Caillere et al. (1982). As the illites found in soils, they have a high amount of stacking faults. These illites were determined for many years as 1Md polytype by XRD, meaning random disorder in the stacking direction. The lack of parallelism induced by stacking faults of corner type corresponds to what used to be called "poor crystallinity".

Swelling 2 : 1 phyllosilicates

Mixed layer 2 : 1 clay minerals containing illitic non swelling interlayers (I type) and smectitic swelling interlayers (S type) can be observed in all types of soils. They can be classified into two groups: structural mixed layer clay and textural mixed layer clay according to their stacking mode.

Mixed layer clays of the structural type (interstratified *stricto sensu)* are mainly found in the coarse clay fraction of soils developed from granitic rock. This structural type has been defined by Robert et al. (1991a) when the three dimensional order of the particles seems to be preserved through the transformation process. The alteration of 10 *Å* minerals induces the opening of some interlayer spaces, starting at one edge of the particle, progressing laterally and leading to mixed layer crystals of Mac Ewan type (Fig. 3a). The sequences of layers in the crystals are either ordered with regular alternance of I and S interlayers (R1 type or rectorite type) or semi-ordered with random sequences of I and S interlayers, S type interlayers being always surrounded by two I. In both cases the layers are strictly parallel over large distances and in all directions. At this stage it is not possible to distinguish between low charge vermiculite and smectite interlayers. The three dimensional order is considered to be preserved when the sixfold symmetry of the (hk0) diffraction patterns testify to coherent orientation of illitic domains with respect to a and b axes. With this approach, n60° or n120° rotational defects as well as +/- b/3 translational defects will not be detected.

In the same samples, loose particles of illite are also observed (Fig. 3b). They have a facies similar to the illitic domains contained in the mixed layer particles, and may origininate from the disaggregation of these larger particles. Such loose particles have been found in acid brown or podzolic soils on granite. In the coarse fraction, the bilayers represent the last step of physical breakdown. In the fine fraction, thin illite coexists with monolayers apparently released by larger particles. Thus smectite could be produced by extreme physical breakdown.

In acid conditions (in the case of granites or even some degraded luvisols on eolian deposits) opening of micas give 1.4 *nm* which can be interpreted as Al-hydroxy vermiculites.

Mixed layer clays of the textural type are common in sedimentary rock soils (Robert et al. 1991) and also widespread in silty rock soils (Fig. 3c). Coagulation of thin illites of 2 to 5 layers gives rise to mixed layered coherent domains when the stacking of crystallites is strictly parallel to the (a, b) plane. Such particles can include monolayers of smectite, thus containing several consecutive S interlayers (which never happens in the structural type). Even if coalescence along the stacking direction can be obtained in rather thick domains, mixed layer particles rarely exceed 10 layers. The lateral extent of the parallel domains is very reduced, especially in sedimentary rock soils where it concerns less than 0.2 *µm* illites which are only partially overlapping.

Microdiffraction patterns of such kind of mixed layer particles along the c* axis correspond to a 10 *Å* lattice. Microdiffraction patterns in the (a, b) plane are of polycrystalline type and suggest a turbostratic stacking mode of illites inside the particles. Moreover we can observe that the interlayer spacings of S type appear most often irregular on HRTEM pictures, not constantly corresponding to 1.36 *nm* as expected, even when tracing along the same interlayer inside a particle.

Discussion

Illite is a general term for non-swelling 2 : 1 minerals of clay size, usually identified by their basal reflexions in XRD patterns. As these minerals generally occur in soils as a mixture with diverse fine grained minerals, precise chemical composition and detailed identification of their unit cell polytypism are difficult to accomplish by classical global methods.

Several authors have pointed out the difference between micromicas and sedimentary illites (Gabis 1963 ; Porrenga 1968). HRTEM and AEM observations seem to confirm this distinction and bring new clues about the genetic origin of the two main species.

Micromicas present in clay fractions of soils may be considered as chips produced by the physical breakdown of bigger micaceous particles. As a matter of fact, micas formed under high temperature (T) and/or pressure (P) conditions such as those prevailing in plutonic rock crystallization, deep diagenesis or metamorphism will be submitted to destabilization factors when put in low P/T and oxidizing environment. Thus they will be highly sensitive to surface weathering, leaching and acidity (Ross et al. 1982; Righi & Meunier 1991).

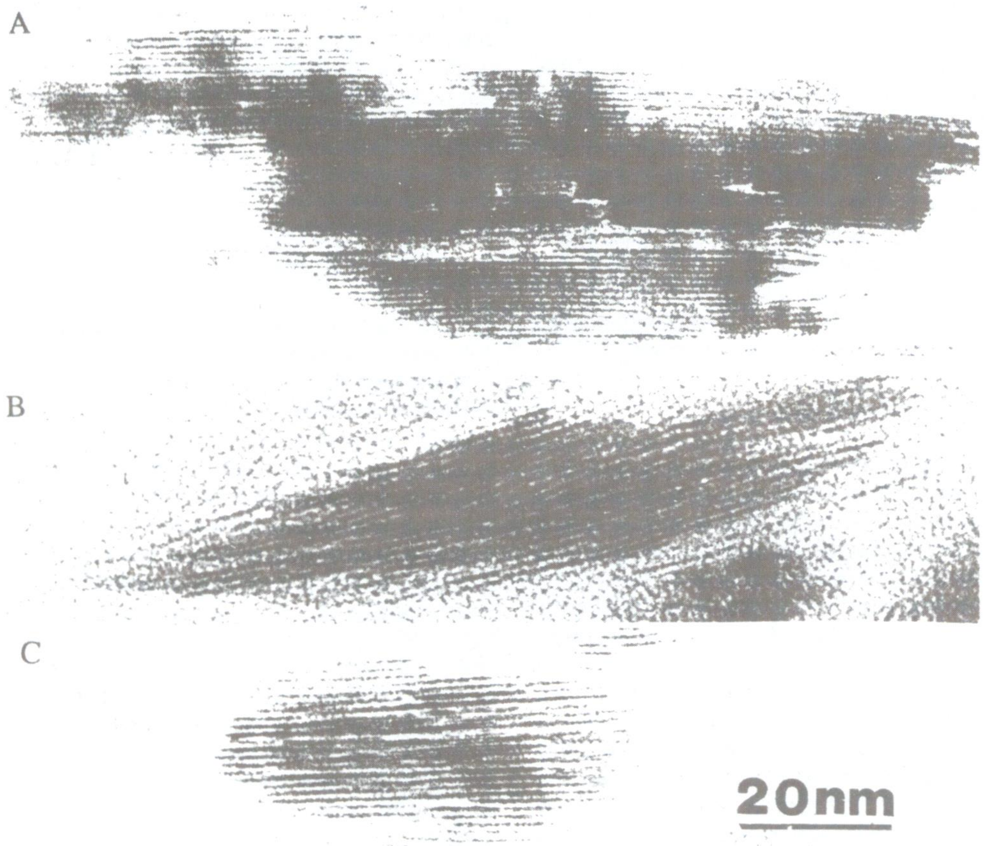

Fig. 2. Micromica and illites in sedimentary rocks.
A - dissolution features in micromica from Thianges (France); **B** - illite from Thianges displaying a lot of staking faults; **C** - lake illite from Muloorina (Australia).

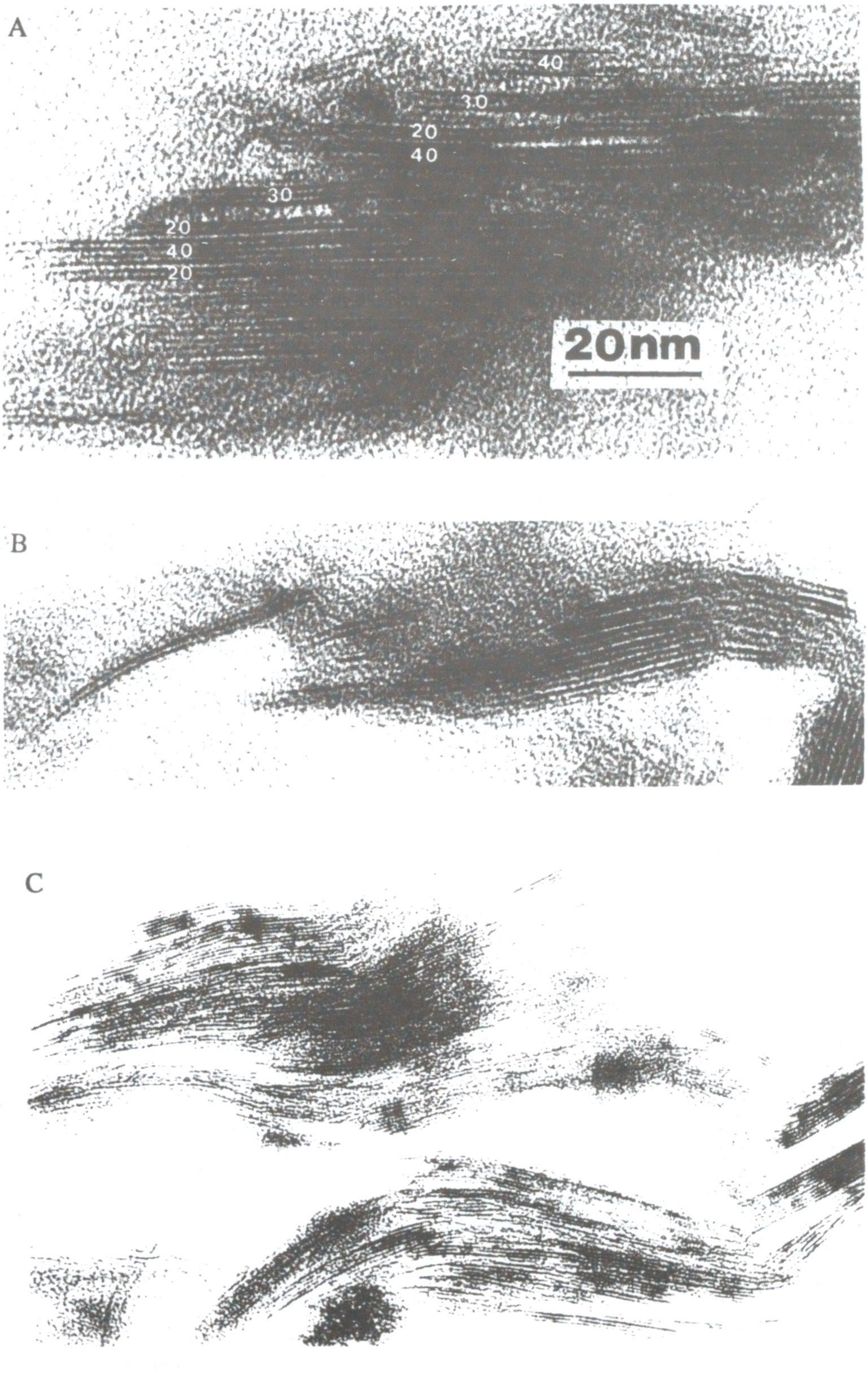

Fig. 3. Mixed layer clays.
A - structural type in a soil developed from granite in Central Massif (France); **B** - in the same sample, disaggregation in loose bilayered illites; **C** - textural type in a clayey soil developed from sedimentary rock in Thianges (France).

Leaching of K leads to swelling of interlayers and producing of S interlayers that can be observed by HRTEM. The so-called structural mixed layer minerals are of ordered or disordered type along the c* axis and ordered or semi-ordered in the (a,b) plane. Such structural mixed layer particles remain stable through very dispersive preparation methods (Righi & Jadault 1988). They correspond to MacEwan crystals in the sense of Altaner & Bethke (1988). The release of individual units from larger mixed layer particles could originate by mechanical scaling or when the layer charge has been sufficently lowered by K leaching to allow infinite swelling (Robert & Barshad 1972). When infinite swelling can be reached after Na saturation and in very dilute suspensions, illite units will behave as fundamental particles in the sense of Nadeau (1984)

Illite *stricto sensu* is widespread in clayey soils. Their barrel shape might be mistaken for eroded clastic micromicas, but, unlike micromicas, they display abundant stacking defects of corner type. Determination of the polytypism (Sakharov et al. 1990) describes these minerals as belonging to the glauconite type, which can be either ferric or aluminous. They contain a variable amount of randomly distributed n60° rotational stacking defects and no expandable layers. The authors suggest that these minerals were formed, regardless of the age and type of the rocks, according to one and the same mechanism, which is probably a solid phase transformation of smectite. When a high content of stacking defects can be observed by HRTEM in the cross sections of some of these minerals, it speaks in favor of a solid state transformation, i.e. the coagulation of smectitic layers of irregular shape and size followed by aggradation with K fixation. But some glauconitic illites have well-defined shapes and straight edges and are therefore unlikely to have formed from transformed smectite.

Geothermometry deduced from the poles contribution to the solid solution (Duplay 1988) indicates for illite from Le Puy a poor correlation with 25 and 200 °*C* models, so the temperature of formation should be intermediate. As a comparison, the best correlation for glauconite from Odin is obtained for 25 °*C*. So the illite from Le Puy is apparently formed from smectite which has undergone a diagenetic evolution.

Textural mixed layering appears to be built up by the aggregation of small particles of highly various size, shape and composition. Coagulation mainly occurs in a stepwise manner and is not coherent over long distances so that XRD gives no clear evidence of interstratification. Particles made by such type of aggregation would be better described as quasicrystals in the sense of Aylmore & Quirk (1971) and Tessier (1991). As the surfaces in contact are very limited and the S interlayers are uneven, the quasicrystals will have a poor cohesion under dispersive conditions.

In soils, the matrix is a continuous solid phase made of clay minerals organized in a porous network. In the case of textural mixed layering, the matrix network can be very loose and display unexpectedly high swelling properties even when no smectite *stricto sensu* is present.

Conclusion

High resolution electron microscopy (HRTEM) and analytical electron microscopy (AEM) show that whereas the global clay content and crystallochemistry of phyllosilicates is relatively stable within the horizons of soils of temperate regions, the clay organization is very diverse and strongly dependant on parameters such as crystallochemistry, shape and size, related to the genesis of the clay.

Such studies about clay genesis must take into account the whole chain of events starting from the origin with the type of parent material (acidic or mafic) and P/T forming conditions. Most of the bedrocks are reworked deposits in which clay minerals are essentially inherited 2 : 1 minerals formed or transformed by paleoweathering (clays, silts, sands or limestones). These minerals might have undergone several cycles of severe alteration by mechanical agents (transport, glaciations, burial) and/or chemical agents (leaching, acidity). The last event in the alteration sequence is the present day weathering, which is considered moderate. As concluded in most studies concerning mineralogical evolution in soils, no striking differences in the mineralogy between fine and coarse clay fractions or between different horizons reveal any strong present day weathering (Jamagne et al. 1984). Reactions such as cation exchange, diffusion and oxido-reduction are preponderant and 2 : 1 minerals undergo a solid state transformation (Robert & Tessier 1991).

The main process governing mineralogical genesis which can be observed by HRTEM involves inheritance followed by microdivision and opening of interlayers until pervasive physical breakdown is achieved, leading to bilayered illites of rectorite type and even to monolayers of smectite type. Neogenesis after dissolution, due to present day weathering, could be evidenced in soils by HRTEM only in the absence of paleoalteration, that is rankers (or saprolites) developed from plutonic or eruptive rocks (Romero et al. 1991).

The clay organization, pore size distribution and rheological properties will be determined essentially by the geometry of the particles, and the way of aggregation to build up the clay fabric. The geometry, shape, size and curvature are linked to the mineralogy and more precisely to the stage of physical breakdown. The clay layer extension and particle thickness decrease with increasing weathering. No marked global change in the mineralogical composition will be detected by XRD, but this geometric evolution will correspond to a completely different organization.

This study focused on 2 : 1 primary phyllosilicates as being the main source for 2 : 1 clay minerals. Feldspars are either relatively stable or transformed into 1 : 1 clay minerals when not subjected to complete dissolution. Chlorite was not found in sufficient amounts or was already too much altered to permit clear observations of its alteration process in such types of clay genesis and weathering conditions.

In the samples studied, soil clay composition is dominated by transformed inherited 2 :1 minerals under the moderate weathering conditions of temperate regions. Evolution of 2 :1 minerals is expected to be still reduced from a chemical point of view in arid or semi-arid areas, whereas it will lead to much more drastic and irreversible changes in equatorial or subtropical areas which are controlled by monosialitisation processes.

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References

Altaner S. P. & Bethke C. M., 1988: Interlayer order in illite/smectite. *Am. Miner.*, 73, 766 - 774.

Aylmore L.A.G. & Quirk J. P., 1971: Domains and quasicrystalline regions in clay systems. *Soil Sc. Soc. Amer. Proc.* 35, 652 - 654.

Bailey S. W., Brindley G. W., Fanning D. S., Kodama H. & Martin R. T., 1984: Report of the Clay Mineral Society Nomenclature Committee for 1982 and 1983. *Clays Clay Miner*., 32, 329.

Caillere S., Hénin S. & Rautureau M., 1982: Minéralogie des argiles. *Masson*, Paris.

Duplay J., 1988: Géochimie des argiles et géothermie des populations monominérales de particules. These Univ. Strasbourg, 1 - 284, in *Sci. Géol. Mém*., 82, 1989, 176.

Gabis V., 1963: Etude minéralogique et géochimique de la série sédimentaire oligocene du Velay. *Bull. Soc. Franc. Minéral. Crist*., 86, 315 - 354.

Jamagne M., De Coninck F., Robert M. & Maucorps J., 1984: Mineralogy of clay fractions of some soils on loess in northern France. *Geoderma*, 33, 319 - 342.

Nadeau P. H., Wilson M. J., McHardy W. J. & Tait J. M., 1984: Interstratified minerals as fundamental particles. *Science*, 225, 923 - 925.

Norrish K. & Pickering J .G., 1983: In: Soils, an Australian point of view. Clay minerals. *Division of Soils CSIRO*, Melbourne, 281 - 408.

Pedro G. & Sieffermann G., 1979: Weathering of rocks and formation of soils. In: Siegel (Ed.): *Review in modern problems of geochemistry*, UNESCO, 39 - 55.

Porrenga D. A., 1968: Non-marine glauconitic illite in the lower Oligocene of Aardeburg, Belgium. *Clay Miner.*, 7, 421 - 430.

Righi D. & Jadault P., 1988: Improving soil clay minerals studies by high gradient magnetic separation. *Clay Miner., 23*, 225 - 232.

Righi D. & Meunier, 1991: Characterization and genetic interpretation of clays in acid brown soil, dystrochrept) developed in a granitic saprolite. *Clays Clay Miner.,* 39, 519 - 530.

Robert M. & Barshad I., 1972: Sur les propriétés et la détermination des minéraux argileux 2 : 1 expansibles (vermiculites smectites) *C.R. Acad. Sci*., 275, 1463 - 1465.

Robert M., Hardy M. & Elsass F., 1991a: Crystallochemistry, properties and organization of soil clays derived from major sedimentary rocks in France. *Clay Miner.*, 26, 409 - 420.

Robert M., Hardy M., Elsass F. & Righi D., 1991b: Genesis, crystallochemistry and organization of soil clays derived from different parent materials in temperate regions (France). Conference invi tee Symposium Commission VII 2. *Occurence, Structure and Properties of Soil Minerals in the Temperate Regions. 14th Int. Conf. Soil Sci.*, Kyoto.

Robert M. & Tessier D., 1974: Méthode de préparation des argiles des sols pour les études minéralogiques. *Ann. Agron*., 25, 859 - 882.

Robert M. & Tessier D., 1992: Incipient weathering: some new concepts on weathering, clay formation and organization. In: Martini & Chesworth (Eds.): *Weathering, Soils and Paleosoils. Elsevier Publ.,* 71 - 106.

Romero R., Robert M, Elsass F. & Garcia C., 1992: Evidence by electron microscopy of weathering microsystems in soils developed from crystalline rocks. *Clay Miner.*, 27, 21 - 33.

Ross G. J., Wang C., Ozkan A.I. & Rees H.W., 1982: Weathering of chlorite and mica in a New Brunswick podzol developed on till derived from chlorite-mica shist. *Geoderma*, 27, 255 - 267.

Sakharov B. A., Besson G., Drits V. A., Kameneva M. YU., Salyn A. L. & Smolliar B. B., 1990: X-ray study of the nature of stacking faults in the structure of glauconites. *Clay Miner*., 25, 419 - 435.

Spurr A. R., 1969: A low viscosity epoxy resin embedding medium for electron microscopy. *J. Ultrastruc*., 26, 31 - 43.

Środoń J., Andreoli C., Elsass F. & Robert M., 1990: Direct high-resolution transmission electron microscopic measurements of expandability of mixed layer illite/smectite in bentonite rock. *Clays Clay Miner.*, 38, 373 - 379.

Środoń J., Elsass F., McHardy W. J. & Morgan D. J., 1992: Chemistry of illite-smectite inferred from TEM measurements of fundamental particles. *Clay Miner*., 27, 137 - 158.

Tessier D. & Pedro G., 1987: Mineralogical characterization of 2 : 1 clays in soils: Importance of the clay texture. *Proc. Int. Clay Conf., Denver*, 78 - 84.

Tessier D., 1991: Behaviour and microstructure of clay minerals. In: De Boodt et al. (Eds.): *Soil Colloids and Their Associations in Aggregates. Plenum Press*, New York, 387 - 415.

Villemin G. & Toutain F., 1987: Méthode de fixation d'échantillons organo-minéraux de sols pour la microscopie électronique a transmission. *Micromorphologie des sols- Soil micromorphology*, 43 - 48.

Weaver C. E. & Pollard L. D., 1973: *The chemistry of Clay Minerals.* Developments in Sedimentology, 15. *Elsevier Publ.*, Amsterdam.