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ARGILLOSCOPY OF WEATHERING AND SEDIMENTATION

by

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"From a small piece one can judge about the whole thing"

CERVANTES

ABSTRACT

Throughout geological periods, argillogenesis - or the transformation of silicates and silicate-bearing rocks into mud - has always developed "uphill" from the sedimentary collector where mudcomposed sediments finally accumulated. The transformation of parent material into clay minerals appears to be a rather complex and intricate process, and depends on the interplay of a variety of both external and internal factors, called "stimuli", which cause the nearly infinite diversity in compositions, crystallinities, relative abundances and associations among clay minerals. Their birth, development and differentiation in various continental settings reflect thermodynamic and crystallochemical conditions prevailing at the surface of the lithosphere. The geochemical pathways and trends characterizing the formation of clay minerals are here reviewed and illustrated.

RESUME

A travers les âges géologiques, l'argilogenèse - ou transformation des silicates et des roches silicatées en argile - s'est toujours placée et développée en amont des bassins sédimentaires collecteurs de ces argiles. La transformation des matériaux parentaux en minéraux argileux apparaît plutôt comme relevant d'un mécanisme complexe et embrouillé dans ses itinéraires et ses phases minérales. Cette "métamorphose" est dépendante de l'interaction de stimuli extérieurs et intérieurs qui déterminent les compositions, les cristallinités, les abondances relatives et les types d'association presque infinis dans domaine des argiles. le L'occurrence, le développement et la différenciation des minéraux argileux au plan de leur gîtologie relfètent les conditions thermodynamiques et cristallochimiques présentes à la surface de la lithosphère. Les itinéraires et les tendances géochimiques caractérisant la formation des minéraux argileux sont rappelés et illustrés.

KEY WORDS

Argilogenesis, geochemical pathways, surficial weathering, sedimentogenesis, diagenesis.

MOTS CLE

Argilogenèse, itinéraires géochimiques, altération superficielle, sédimentogenèse, diagenèse.

INTRODUCTION

Because of the nearly infinite diversity in their composition, association, relative abundance, specific geological and pedological settings, stratigraphic and genetic conditions, clay minerals offer, geologically speaking, an attractive support in decoding the Earth's lithospheric history.

Argillogenesis is not an insignificant process among all the other processes that characterize the Weathering, erosion geological cycle. and sedimentation (of mud or clays) are events which, through physico-chemical and thermodynamic processes, convert diverse parent rocks into soft clayey sediments. This process takes place everywhere where the lithosphere is or comes into contact with the hydrosphere for a sufficiently long period to react with it. It is worth recalling that, through the whole stratigraphic column, claybearing sediments and sedimentary rocks constitute volumetrically more than 65 % of the whole stock of such materials, while detrital sediments and sedimentary rocks contain more than 70 % of minerals composed of clay minerals or related phyllosilicates. The abundance and ubiquitous occurrence of clayey materials within and at the surface of the lithosphere raise the question of the clay status.

In other words, why are silicate-bearing rocks at the surface regularly, if not necessarily doomed to transformation into clays? Why does this type of mineral "leprosy" affect the surface of the continents as well as the floor of deep oceanic waters?

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The reason is that the "clay" status is the normal status of any silicate-bearing rock or siliclastic mineral when cropping out at the surface. A mineral "leprosy" of this kind structurally conveys also the hallmark of the physico-chemical and thermodynamic equilibrium which the exposed materials try to reach. The lithosphere largely consists of silicates assembled into a variety of different parent rocks : granites, gneiss, micaschist, mudstones, lava. When exposed at the Surface and able to escape a (too) strong erosion, these silicatebearing rocks are normally doomed to weathering or destruction. The product of this destruction is Clay. Hence, the normal status of the Surface is the clay one in (nearly) steady thermodynamic condi-Clay minerals also have a particular tions. crystallo-chemical behaviour, because they are intermediate minerals between inert minerals (such as quartz), which remain practically insensible throughout their transfer into the superficial crust, and soluble minerals (such as carbonates, sulphates, salts) which are readily put into solution under favourable conditions (Millot, 1967).

Many clay minerals also bear the climatic hallmark of their genesis in their composition and structure prior to their sedimentation. Some are also sufficiently resistant so as to preserve, to some degree, and even during a long period, the physicochemical characters developed during their formation. Therefore, clay minerals are valuable auxiliaries in reconstructing (paleo)climates and hydrodynamic and paleogeographic Through mud-derived sediments and tectonic, events. pedologic materials, the lithosphere has always and everywhere accumulated clays in a variety of sedimentary collectors and repositories. However, the clay-machinery is not a simple device which readily transforms any non-clayey parent rock into a clay (mud) material. On the contrary, the processes leading to the transformation are usually gentle, progressive, easily submitted to many twistings as well as to reversals from the normal pathways or goal : the transformation into clay minerals".

"Clay minerals are the major solid phase present during the immense chemical transfer process which occurs at or near the Earth's surface. In clay environments, one finds the most extreme chemical segregation known in geological cycles. If we wish to interpret the past (geology) we must understand the reasons for the appearance of these clays which remain" (Velde, 1985).

In the present paper, the history and behaviour of clay minerals from their site of birht on the continent to their final setting in the sedimetary basin will be reviewed mostly graphically in honor of Dr. Eva Paproth, whose paleogeographic recontruction of many sedimentary basins throughout the world has always kept her interest and driven her imaginative dynamism.

The idea behind this paper is to synthesize the genetic links between argillogenesis and sedimentogenesis, with some references to diagenesis. The needs in Sciences sometimes arise from the will to substitute the explicative text by a more direct visualization of the parameters involved, in this particular case in clay geology. The whole story of the birth of a clay mineral, its development, its maturity, its "senility" and its final disappearance will be depicted.

1. THE PLACE OF ARGILLOGENESIS IN THE GEOLOGICAL ROCK CYCLE

In the geological cycle as traditionally displayed, several stages and subsequent mechanisms are presented within a cycle with a unidirectional pathway. For instance : sediments - sedimentary rocks metamorphic rocks - igneous rocks. Figure 1 depicts the geological cycle as a kind of infinite motion or continuum ; the pattern is inspired after the drawings of the well-known Dutch artist ESCHER. The cycles split near an important "interface" that separates what could be called the "dry" lithosphere from the hydrosphere (argillosphere) where weathering gives rise to clay minerals. A variety of rocks, including the sedimentary ones, may be brought to the surface by tectonic uplift and eventually be subjected to (pedi)peneplanation, thus to atmospheric and/or pedogenetic weathering.

(BIO) CLIMATE





Clay weathering, or the hydrolysis of silicates, is the paramount destructive mechanism that may affect all exposed geological materials. Hydrolysis necessitates the occurrence of (percolating) solutions through the silicate-bearing substratum, or the reaction of the latter with superficially circulating waters; however, the presence of water is not sufficient and other external as well as internal factors must intervene : the climate in general, precipi-

DEFINITION	STATUS	GEOCHEMICAL MECH- ANISMS
Minerals Ultra Teneous, Argillaceous Native of and in Transit at the Surface	Minerals Uninterruptedly, Thermodynamically Adapted to the New Transitory Situations	Minerals either Unchanged (herited) Transformed (degraded) Aggraded, Neoformed or Tumbled down in Solutions
or Minerals structurally organ- ized within the <i>Clay Integron</i>	or Minerals submitted to a <i>ion-chromatography</i> in and through the (micro)environments	or Minerals variably associated according to the <i>kaleidoscope</i> of genetic conditions and set- tings for their formation and accumulation

Table 1. : Definition of the clay minerals : the MUTANTS of the lithosphere.

tation and temperature, and drainage, either surficial or in the substratum. When weathering (with transformation into clays) has taken place during a sufficient time (this factor being of importance in argillogenesis), the clays are transported into the sedimentary basin. In addition to these continental weathering events, there also exist other ways and sites where clays may develop ; for instance, during halmyrolysis of volcanic materials delivered by surficial or submarine vents and fissures. At contact with sea water the volcanic substances (lava and ash) are transformed into clay minerals, and into smectites in particular (Fig. 2).



THE THREE MAIN MODES OF CLAY BEVESIS + HERIFAGE, TRANSFORMATION, VEOFORMATION.

Figure 2. : The rock-cycle, with emphasis on the possible split in the siliciclastic reservoir for the clay mineral genesis. Presented in another way, the definition of clay minerals comprises three different but interrelated levels of appreciation :

(i) a geological sense, referring to all unconsolidated (or deconsolidated) geological materials (such as a sediment) but also to all products of atmospheric, pedogenetic and halmyrolytic weathering ; by extrapolation, clay minerals can also be found in a series of consolidated sediments (cf. sedimentary rocks), provided that their minuteness and hydrated character have been spared through lapidification and diagenesis.

(ii) a specific grain-size of the minerals with a mean diameter around or less than 2 microns by international standards.

(iii) a mineralogic sense which may cover the association of diverse minerals and substances : fine-grained phyllosilicates (talc, chlorite, micas, ...); the clay minerals sensu stricto, either simple clay minerals like illites, chlorites, vermiculites, smectites, kaolinites or mixed layers (these may correspond to regularly interstratified minerals or to randomly interstratified structures) ; also sulphates, carbonates, salts, organic compounds, amorphous substances, and, among all these components, water or solutions bound at the surface of the clay particles and/or inside the interlayers.

The very "bricks" of any clay mineral structure are tetrahedra and octahedra. These are associated into tetrahedral and octahedral sheets respectively, themselves stacked one upon the other along the "c" axis" so as to provide two models named 1:1 and 2:1 (Fig. 3).

2. DEFINITION OF A CLAY MINERAL

A unorthodox definition of a clay mineral is presented in Table 1. Clay minerals are here named



Figure 3. : Schematic presentation of the tetrahedral and octahedral sheets combined into a layer.

MUTANTS of the lithosphere ; their status and driving geochemical mechanisms are also taken into account. The term MUTANT emphasizes the continuous mineralogical and thermodynamical "search" of clay minerals for a stability when offered to weathering agents : a progressive mutation intervenes which affects the occurrence, composition, abundance, and association of clay minerals.

This mutation is well supported by the nearly infinite variability of clay mineral species. Moreover, when breaking down letter by letter, the term *mutant* conveys the clay mineral definition, status and formative geochemical mechanisms.

These layers are separated by interlayered spaces (interlayers) which may either lack cation occupants, or be filled up by dry, or partially or fully hydrated cations or by a hydroxide supplementary layer (formerly the brucitic or the gibbsitic layer). Nature had built up clay minerals according to these two structural models, their classification would have been very simple. But the clay minerals differ greatly because of the existence of several crystallochemical parameters which modify their structure and composition. For instance : element subsitution in the sheets or within the interlayers (as substitution of Al for Si in the tetrahedral sheet, or Fe, Mg, Cr for Al in the octahedral sheet), irregularities in the stacking of the layers, ; or similar or dissimilar composition of the layers respectively in simple clay or in mixed layers.

Along the "c" axis", the stacked layers are organized in enlarged "items", comprising several layers, and called the "primary particle". Several primary particles are, on their turn, grouped into "morphologic units"; these are associated into clay particles, clay domains, crystallites, and may form the clay plasma (cement, matrix) of detrital sediment as well as the main mineral component in soils, or intervene as secondary fillings within the pores of a sediment. Such an integration of elements, differing in sizes and structures, leads to the concept of the *Clay Integron* (Thorez, 1985) (Fig. 4).



Figure 4. : The Clay Integron, or the way clay minerals are built up, from the Angström to the cubic Kilometre, in the landscape.

A clayey sample is collected from a section, itself part of a (clayey) landscape. From this sample, the clay fraction is extracted and prepared either as a random powder or an oriented aggregate to be analysed by XRD. The latter technique provides an XR pattern, a kind of diagram, on which reflections appear that can immediately be matched with the crystallographic and crystallochemic properties of the minerals, hence their nature, their occurrence in various relative proportions, etc.

3. THE CLAY MACHINERY

The natural manufacture of clay minerals roughly resembles a "liquid extractor" as presented in Figure 5 which associates different "ingredients" essential for initiation and progress of the production of clay minerals, either permanently or in a "jerky" manner with temporary stops. The reaction can be offered in a more classic way : clay minerals appear as the secondary products of such a reaction, but this does not imply that clay minerals could not already be parent material. The ion-enriched solutions may be trapped in the (micro)environment or escape into the landscape by lateral or downward drainage.

4. DIVERSITY OF ARGILLOGENETIC PATHWAYS

Whether submitted to atmospheric, or to pedogenetic or halmyrolytic weathering, parent silicate-bearing materials transform progressively



THE CLAY MACHINERY ANALOGOUS TO A LIQUID EXTRACTION REACTOR

Figure 5. : The Clay Machinery as an analogy of a liquid extractor.

PARENT-SILICATE	stable or still	SECONDARY	ION-ENRICHED
BEARING MATERIAL	residual	<pre>4 minerals (mostly CLAY MINERALS)</pre>	+ SOLUTIONS
OR MINERAL(S)	minerals		(escaping into
+ SOLUTION	residual		the landscape)

but continuously into clay substances provided that a (constant or temporary) contact exists with the percolating solutions. These are the driving factors for initiating and maintaining the hydrolysis and developing the variety of by - and end - products of the reaction, among which are clay minerals.

However, the geochemical and crystallochemical reactions and trends offer to the clay minerals a set of variable fates, either isolated or combined, or succeeding one antother : inheritance (heritage), transformation (degradation, degradationaggradation, aggradation), neoformation, then (re)crystallization, or/and solubilization.

During degradation, the system remains open allowing progressive withdrawal of ion-enriched solutions by hydrolysis. The system may become partially confined or confining at the standpoint of the double degradation-aggradation process (*see further*). Duration of the process must be long enough to allow hydrolysis to progress. However, erosion is a blocking process because it stops, definitevely or temporarily, the transformation into clay by removal of the already clay-transformed products from the site of formation. When erosion stops, hydrolysis is regenerated.

During weathering, ions may be selectively removed from the (micro)environment. Either all the ions leave the system, leading to a strict dissolution, or some of them are kept preferentially within the (micro)environment and favour the neoformation of some clay minerals (i.e. neoformation of kaolinite in the weathering profile by the blocking of Al and some Si, while all the other parent bases escape from the environment). Al and Si recombine themselves within the kaolinite ; the process can take place in situ, or down the relief, in low areas where the solutions are blocked again. A similar mechanism intervenes for the neoformation of some peculiar clay minerals such as smectite, palygorskite and sepiolite : ion-rich solutions reach a marine but evaporitic environment, and precipitate their contents as minerals (clays or carbonates or sulphates). Among all the geochemical processes, heritage, transformation and neoformation appear to be the driving factors in argillogenesis sensu lato.

Heritage means that, after transport from the site of its original formation to its final setting as clay sediment, a clay mineral preserves all its former erystallochemical characters : the parent and the "child" elay minerals are similar and cannot be distinguished from one another. Thus, heritage refers to the unchanged status of the mineral because no weathering has taken place, or because the mineral was in equilibrium with the (micro)environment. A typical example of herited clay minerals is a sediment which contains both illite and chlorite that already occurred in the parent substratum from where they have been derived through erosion and transport. The minerals are equally as fresh and intact as they are in the parent rock.

However, the couple illite-chlorite found in a sedimentary rock may derive from the effects of another, completely different process : diagenesis, in which case it does not correspond to the effects of a simple heritage. This genetic "mimetism" must be kept in mind when trying to reconstruct a paleogeographic and/or a paleoelimatic model for the elay minerals.

Neoformation depicts a geochemical process in which the clay minerals are entirely formed (neoformed) at the weathering site (i.e. a kaolinite in a latosol) or result from precipitation or crystallization from ion-bearing solutions which are trapped in the profile (i.e. Fe-beidellite in vertisols) or eoncentrated in the sedimentary collector (i.e. formation of smectite, palygorskite or sepiolite in evaporitic marine environments). Neoformation of clay minerals can also be generated from amorphous compounds.

Transformation is a process in which the parent clay minerals preserve their original layered structure despite chemical reactions. These latter develop either within the thetrahedral or the octahedral sheets or within the interlayers. Thus, the reaction mostly yields an ion-exchange and not a direct nor an entire layer modification (Lucas, 1962). Transformation can be differentiated into degradation, degradation-aggradation, and aggradation. Degradation concerns an ion substraction out of the layered structure. It may be combined with oxidation within the sheets (i.e. Fe of the octahedral sheet) and/or hydratation of the eations settled in the interlayered space that separates two directly superposed layers. A good example of degradation is that of a 10A-mineral (such as illite or biotite) which, in the course of the weathering (degradation), gives birth to a vermiculite-like structure. Smectite or secondary ehlorite can also become the by - or even the end - product of degradation. But these two cases weathering proceeds better through a double mechanism : degradation within the interlayers with the leaching of the K ions, and building-up of Al-pillars (cf. A1hydroxylization) at the same site as for the secondary ehlorite.

The passage of a chlorite into a smeetite also corresponds to a true "ion-chromatography" (Tardy, 1969); the process is generally "channeled" through a series of even labile intermediates which are represented by mixed layers. In the case of transformation of a chlorite into smeetite, the involved



Figure 6. : Schematic presentation of ion movement in degradation, degra- dation-aggradation, and aggradation processes (ion-substitution), at level of the layer and interlayer of a 10A clay mineral, showing three possible geochemical pathways : smectitization, vermiculitization and (secondary) chloritization.



Figure 7. : Evolution mechanisms, pathways and geochemical status (heritage, degradation, aggradation and neoformation) for clay minerals during their passage in the hydrosphere and at final sedimentation.

mixed layers are identified as chlorite-smectite intermediates. Other mixed layers exist such as (*illite-vermiculite*), (*illite-chlorite*), (*illite-smectite*), (*chlorite-vermiculite*).

Aggradation is a transformation by addition of ions. In other words, a former degraded (possibly herited) clay mineral is able to host some available ions provided by the (micro)environment or brought in (as in the sedimentary basin) by solutions. For instance, let us take the case of a degraded illite having its interlayers distended and partly depleted of their original Potassium.

The degraded illite, coming into contact with solutions enriched with this element, can rebuild an intact mineral (cf. a closed illite) : Potassium finds its way to the interlayers because of a sort of "structural memory" embedded within the affected illite.

The process of aggradation of an illite rarely occurs in weathering conditions (except for the A horizon of some soils), but generally develops during diagenesis, in a kind of confining or confined (micro)environment. However, the structural memory can sometimes fail ! An example is that of a highly degraded illite being placed in a sedimentary environment where Potassium is insufficiently available but where Mg is practically the sole ion concentrated in the marine waters. The degraded illite aggrades into a Mg-chlorite (Lucas, 1962). Degradation-aggradation is a mixed process. For instance, when a parent illite weathers into a smectite (cf. transformation smectite in opposition with a neoformed smectite) degradation develops through the leaching of the interlayered Potassium, while some silicification may occur in the octahedral layers (Fig. 6).

Degradation and aggradation may also develop, not at the surface, but during burial and diagenesis. A good example is the post-sedimentary weathering of illite and even of kaolinite within the pores of a clayey sandstone; even feldspar may be affected by weathering. The available cation "ingredients" serve for the *in situ* neoformation of a "hairy" secondary illite. This is built up within the pores. Aggradation also characterizes the passage of a smectite to an illite, via mixed layers (illitesmectite), during diagenesis. Such a case has been documented many times in the literature.

When comparing surficial degradation and burial aggradation effects, other factors offer a kind of symmetry for these two constrated geochemical environments (Dunoyer de Segonzac *et al.*, 1970). During *surficial weathering*, the grain-size of the parent minerals, including in particular that of the clay minerals, progressively decreases through fragmentation and edge-hydrolysis. Meanwhile, Al of the parent silicates passes from a coordinated status to that of a hexacoordinated one. Surficial weathering does not need high temperatures and takes place at surface pressure. The transformation into

clay minerals essentially comprises a cation leaching process within an open environment when this is visited by percolating but rather diluted solutions.

During *diagenesis*, following burial of the clay minerals, aggradation and possibly neoformation and recrystallization take place. Most, if not all, of the micrometric clay minerals are eventually doomed to become transformed into the wellcrystallized "bed-mates" phyllosilicates, mica and chlorite, which are ordinarily found in diagenetically matured mudstones and shales.

During diagenesis, grain-size increases through cicatrization, recrystallization and purification of the layered structures. Illite exchanges its Fe and Mg with the chlorite ; this in turn and simultaneously, frees its Al for the benefits of the new-formed "bed-mate" mica. The hexacoordinated Al as found within the original clay minerals turns into a tetracoordinated Al in both micas and chlorite.

During diagenesis, temperatures are somewhat higher, possibly reaching 250°; the same hold for the pressure. The (micro)environments are also characterized by relatively highly concentrated interstitial solutions resulting from the confining or confined qualities of the host-sediment (micro)environment.

5. EXTERNAL FACTORS IN ARGILLOGENESIS

As quoted above (Fig. 5), several external factors or stimuli are needed for (and during the) clay manufacture. At the surface of the lithosphere, these driving factors comprise at least climate, vegetation, relief, geomorphology, tectonics, erosion rate, and geological duration. All of these interfere while weathering develops.

The interplay of these factors also modulate the variability among clay minerals at the site of their formation before erosion, transport and sedimentation take place. This interrelation between the external factors, prior to sedimentation, is illustrated in detail in Figure 7. The influence of climatic factors on the trend of pedogenesis and the kind of clay minerals found in the soils are depicted in Figure 8.

6. THE KALEIDOSCOPIC PATTERN OF GEOCHEMICAL WEATHERING AND ITS HALLMARK IN PEDOGENESIS AND SEDIMENTOGENESIS

(Paleo)soils are the more typical fingerprints of the climate upon and during surficial weathering (Fig.



Figure 8. : Schematic "ecologic" conditions of weathering and subsequent characterization of clay minerals in some relevant soils.



Figure 9. : Geochemical mechanisms of weathering during pedogenesis and atmospheric alteration (modified after Pedro, 1985).

8). Various but contrasted geochemical and pedogenetic processes are involved in pedogenesis, namely : heritage, plasmogenic and aplasmogenic weatherings (Pedro, 1985) (Fig. 9). Here weathering results either from hydrolysis or from acidolysis ; at the level of the clayey by- and end-products and of the clay minerals, several geochemical mechanisms develop and differentiate themselves : aluminosiallitization, monosiallitization, bisiallitization, etc. (Fig. 10). Moreover, in each case, heritage, transformation, neoformation, and/or solubilization may intervene either by itself or in specific association, conditions which further on discriminate the pedogenic trends. Transformation, neoformation and heritage, but also solubilization and (re)crystallization form part of sedimentogenesis. The clay minerals in detrital sediments are, in some way, a kind of mirror of what occurred during the continental surficial weathering. The latter is depicted in Figure 10 for two contrasted, open and confined, systems.

For instance, smectite can be neoformed in soils as exemplified by Fe-beidellite in vertisols (Paquet et al., 1970). However, neoformed smectite, palygorskite and sepiolite are better confined to sedimentary environments (Trauth, 1977). During sedimentation, most of the clay minerals accumulate as a result of an "uphill" erosion, and transport to a "downhill" sediment collector.

Whatever geochemical processes have acted before the erosion, it clearly appears that the mud-derived sediment essentially bears the hallmark of heritage as far as nature, abundance and association of clay minerals are concerned. The sedimentary basin can roughly be compared with a kind of clay mineral "dustbin". The relationship between the continental (atmospheric and/or pedogenic) weathering and the sedimentary collector is shown in Figure 11. Beside the usual heritage, degradation, aggradation and neoformation, another geochemical process, rejuvenation, may possibly occur in the inherited sediments.

Rejuvenation can be illustrated as follows : for example, the sedimentary basin inherits a slightly degraded chlorite (among other clay minerals). Such a degraded chlorite is thus characterized by some distended interlayers due to partial removal of the original Mg-Fe oxidic layer and subsequent hydration. In the sediment, in which trapped solutions are somewhat enriched with Mg, the degraded chlorite finds back the "missing" cation and reincorporates it in its structure. The result is well-crystallized, fresh-looking Mg-chlorite, a which reveals a fresh, intact character as for the continental parent chlorite, but possibly with a higher amount of Mg than for the original Fe-Mg parent mineral.

7. DIVERSITY OF BY- AND END-PRODUCTS OF ARGILLOGENESIS

The transformation of a silicate-bearing parent material into a clay mineral is not a simple, undirectional nor a direct mechanism. Many highly intricate paths exist in Nature, which are best represented by a fan of mixed layers. These are the



- (+) ADDITION PROCESS
 - Figure 10. : Sketch displaying the three main modes of clay genesis : heritage, transformation and neoformation, and some conditions operating at the level of layers, with resulting typical clay minerals.



Figure 11. : Mechanisms of evolution of clay minerals in the hydrosphere on both sides of the continental weathering and sedimentary basin "interface" (modified after Lucas, 1962).

relays and the the bridge that separate two stages representing, for instance, the parent clay minerals and the end-products of transformation (through degradation, or aggradation).

It appears rather difficult to overview all the possible pathways. The only possiblity is that of a graphic presentation with four "poles" representing the parent minerals : illite (I) and chlorite (C), and as parent minerals or already end-products : vermiculite (V) and/or smectite (Sm) (depending on the local conditions) (Fig. 12). All the mixed layers, and possibly vermiculite and smectite, are then real transitory or intermediate mineral phases which occur and develop during weathering. For instance, illite (I) may weather towards vermiculite (V) by passing obligatory through a possibly (labile) mixed layer (illite-vermiculite) (10-14V) (in case of a randomly interstratified structure) or (I-V) (when the transitory phase is represented by a regularly interstratified mineral).

The geochemical mechanism corresponds to a *vermiculitization* of the parent illite. However, other pathways exist : weathering of illite into smectite (*transformation smectite* in opposition to the *neoformation smectite*); the transitory stages are represented by mixed layers (illite-smectite) (10-14Sm) or (I-Sm). The illite may, in reality, correspond mineralogically to a biotite. On the other hand, the smectite member of mixed layers can further become identified as a montmorillonite, beidellite, nontronite, saponite, etc. through specific post-treatments (i.e. Li-, K- and Mg-saturations).

However, this graphic representation does not reveal all the "tricks" of Nature. For instance, when sediment the C+in а association (14C-14Sm) + Sm is found, one can hardly decide whether the C has resulted from a strict aggradation process (starting from a "parent" smectite having passed through the intermediate mixed layer (14C-14Sm), or whether the reversal trend is the real pathway as a smectitization of a parent chlorite (the latter evolving into a smectitic end-product). In both cases the mixed layer (14c-14Sm) structurally preserves its genetic anonymity, or more appropriately, its structural "mimetism". Also shown in Fig.12 is the case of aggradation of a vermiculite into a secondary Al-chlorite. This process is here invariably unidirectional.

It is not because of occurrence of certain clay admixture that the geochemical trends are immediately recognized and possibly interpreted : field observations remain necessary for genetic interpretations as an XR pattern will always remain a "blind" investigation tool which cannot be separated from the geological background analysis. One should stress the fact that when indicating, for instance, the occurrence of a mixed layer such as (10-14V) that "bridges" the two end-members illite (I) and vermiculite (V), the passage from one to the other comprises in reality a complete series of intermediates. In these, illite-behaving layers are progressively repaced by vermiculite-behaving interlayers along a sequence which may be indicated as follows :

10-(10-14V)--(10-14V)--(10-14V)-14V



Figure 12. : Various clay mineral reactions and subsequent by- and/or products represented either by simple clay minerals or by randomly (or regularly) mixed layers. Parent clay minerals here are illite (1) and chlorite (C). Kaolinite (K) and smectite (Sm) are not taken into account because referring to neoformation processes.

Figure 12 shows that similar geochemical pathways (co)exist : for instance, illite and chlorite may become submitted to vermiculitization while the byand end-products differ in their structure and composition : (10-14V) for illite and (14C-14V) for chlorite. Respectively, the weathering of both illite and chlorite is not simultaneous, and is regularly dephased in Nature, with chlorite being more apt to weathering.

Table 2 shows the main combinations (two components) for regularly interstratified minerals and for the corresponding randomly interstratified structures. It does not take into account the relative abundance of the components or the relative regularity in the stacking of the layers and interlayers.

A. Regular mixed	layers				
I (illite)	C (Chlorite)	Csw (swelling)	V (Vermiculite)	Sm (Smectite)	
(I-C) (I-Csw) (I-V) (I-Sm)	(C-Csw) (C-V) (C-Sm)		(V-Sm)		
B. Random mixed	layers				
10	14c	14Csw 14V	14Sm	7(kaolinite)	
(10-14C) (10-14Csw) (10-14V) (10-14Sm)	(14C-14Csw) (14C-14V) (14C-14Sm)	14Csw) - (14Csw-14Sm) 14V) (14V-14Sm) 14Sm)		(7-14Sm)	

 Table 2 : Possible basic combinations for mixed layers.

Figure 13 offers a more detailed and complete overview of all the pathways incorporated in the different weathering sequences. In particular, the neoformation of kaolinite, the occurrence of the mixed clayer (C-Csw) and Csw, and the Al17 (a kind of smectite wherein Al-hydroxylized occur as pillars or islands), gibbsite and ions in solution complete the lay-out of Figure 12.

As a matter of fact one should also stress the fact that the sequence displayed in Figure 13 are related to complete sequences. In Nature, for various reasons, the sequential weathering may become interrupted at a particular stage without yielding the normal end-product. These breaks or changes of trends are induced, for instance, by a change of climatic *stimuli*, or because erosion has interrupted the process before being completed. During weathering, some kind of structural and compositional "mimetism" may arise, as mentioned above. This mimetism usually occurs when different parent minerals, associated in the original material, weather at the same moment.

For instance, Figures 14 and 15 provide the weathering (degradation, aggradation) trends for illite, biotite, chlorite, feldspars and ferromagnesians (Thorez, 1985, 1989). To ensure a simple graphic presentation, only the geochemical processes and the "poles" of the pattern are explicitly shown, whereas intermediate stages (ordinarily represented by mixed layers) are not represented. As shown for all weathering trends for these parent silicates (I, Bio, C, Feldspars and ferro-magnesians), a common end-product would



Figure 13. : Cartoon showing the diversity of geochemical processes and correlative sequences that eventually occur and develop, either partially or fully, during weathering of parent illite (l) and chlorite (CV).

be normally reached at the end of the weathering : kaolinite.

Moreover, some by-products can develop after degradation or aggradation of the parent minerals. For instance, smectite or vermiculite appear as intermediate stage mineral during transformation of illite, biotite, chlorite, and of even feldspars and ferro-magnesians. Hence, the necessity of petrographic investigation of the material and visual interpretation of the degree and stage of weathering for the associated of the by-products derived from the parent mineral.

In order to demonstrate the impact of climate (in terms of precipitation and temperature) on the final products of weathering, Figure 16 contrasts the weathering of a biotite under different climatic conditions.

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Al-Fe-clay minerals	Al-Mg-clay minerals	(Al)-Mg-clay mineral	ls Mg-clay minerals	
Wyoming-Sm (herited)	Cheto-Sm (aggraded)	Palygorskite (neoformed)	Sepiolite (neoformed)	
		Recrystallisation/epigeneis		
		Al-Saponite Mg (aggraded) (transf.	-saponite Stevensite by aggrad.) (neoformed)	

Table 3 : Sequence of aggraded (Cheto-smectite, Al-saponite), transformed by aggradation (Mg-saponite), neoformed by aggradation and precipitation (Stevensite), and neoformed Palygorskite and Sepiolite in a marine evaporitic carbonate environment (Trauth, 1974).

8. GEOCHEMICAL PATHWAYS FOR CLAY MINERALS DURING SEDIMENTATION

A sedimentary basin acting as the final collector, is a sort of "sink" for the eroded and transported clayey materials which were produced on the nearby continent, through physical (desagregation-heritage) and/or physico-chemical (transformation, possibly neoformation) processes. The materials feed the basin in the framework of a relation which can be described as an "uphilldownhill" movement of detrital particles and possibly of solution.

Although heritage characterizes the clay mineral stock in its downhill accumulation, syn- and postsedimentary aggradation or neoformation of clay minerals also takes place within the collector just after sedimentation or during early diagenesis. A characteristic example of such a process was described by Trauth (1974) (Table 3) in which smectites of different species occur in a sequence leading from a herited species to neoformed ones, with also palygorskite and sepiolite as end products.

In this example, only detailed mineralogical investigations discriminate the "global" smectite (this being usually obtained in XRD routine analysis). Trauth (19745) used both DTA and XRD data to reconstruct the really occurring sequence of smectitic minerals and provided clues to distinguish herited, aggraded and neoformed between smectites. In other words, this evaporitic sequence gives rise to an enchained series of smectite species or varieties. It clearly demonstrates that a routine XRD investigation overlooks the mineralogic, hence the genetic significance of smectite within a sediment even if the smectite component occurs associated with other clay minerals. Posttreatments (*i.e.* cation saturation) allow a mineralogic differentiation among the smectitic cation saturation) allow a species. Unfortunately, this aspect of clay mineralogy is too often overlooked and not sufficiently taken into consideration.

Although there exist many exceptions to the rules, a common pattern of genetic processes can be presented bridging tectonics (T), weathering (W), erosion (E) and sedimentation (S) to argillogenesis (Fig. 17). The influence of these various factors intervening between the "uphill" site of weathering and the "downhill" site of sedimentation can be expressed in an "intensity scale" (weak, moderate and intense effects). Moreover, all these relations can be graphically presented in a kind of "dart game" (Fig. 17).

The geochemical process that has initiated the weathering (heritage, transformation, neo-formation) may also appear in this overview. As a consequence, each "sector", with its above defined features, characterizes a certain trend for the clay minerals occurring in the sediment.

Of particular interest in this presentation is the possibility to investigate (through clay composition, relative abundance and kind of association) the story of the minerals prior their final accumulation as sediments. However, as Nature is highly diverse in its effects, it is impossible to take all cases into account (Fig. 17).

Another way to represent the geochemical pathways for clay minerals in the sedimentation realm, with reference to tectonics and erosion, is shown in Figure 18. As a matter of fact, the intensity of tectonics (which influences erosion) can be recorded within the clay mineral composition. For instance, in Figure 18, a vivid, strong tectonic effect at the site of clay "manufacture", associated with a strong erosion, will preferably remove and transport rather fresh, and thus intact, clay minerals into the sedimentary basin. Heritage will prevail over the other processes. At the other extreme, in a regime without tectonic disturbance but with a climate favouring deep weathering, the downhill sediment collector will receive a huge amount of ion-enriched solutions, with few detrital clay parti-



Figure 14. : Synthetic representation of weathering trends, geochemical paths and processes, and resulting mineral phases for illite, biotite and chlorite as parent clay minerals.

cles. If climatic conditions are downhill favourable (i.e. evapotranspiration favouring precipitation), the clay minerals will preferably bear the hallmark of neoformation. Until now emphasis was put on the strict relationship, mineralogically and genetically speaking, between the "uphill" continental weathering and the "downhill" marine *s.l.* environment. For instance, during the last decades, many investigations have



Figure 15. : Synthetic representation of weathering trends, paths and processes for feldspars and ferromagnesians.



Figure 16. : Climatic influence of chemical weathering of a parent (after Seddoh and Pedro, 1974).

been carried out on clay genesis and accumulation in oceanic basins, far from the direct influence of continental supply. This was possible thanks to the Deep Sea Drilling project. In particular, the problem of the "red clays' is now more clearly understood. These red clays actually appear as the product of a combined halmyrolysis of volcanic material and the dissolution of biogenic sediments. Characteristically, red clays are formed in the hydrosphere, but not in contact with the atmosphere. However, halmyrolysis operates in occanic waters but also in continental bodies of water, such



Figure 17. : The Clay Dart Game, or the possible genetic pattern linking, for argillogenesis, the direct influence of : tectonics, weathering trends, geochemical processes, erosion and sedimentation.



Figure 18. : Geochemical pathways for clay minerals in and during sedimentation (modified after Lucas, 1963).

as lakes and sea (as the case of tonsteins or kaolinized bentonites). At the bottom of the oceans, particularly within the framework of plate tectonics, magma may erupt and become ejected through vents and fissures. In oceanic environments, halmyrolysis produces transformation of magma into clay through a complex association of degradation, aggradation, neoformation. recrystallization and precipitation. Hoffert (1980) demonstrated how red clays are formed, recycled by erosion, translocated in deeper but quieter areas of the ocean and there evolve into a kind of "submarine soil" (Fig. 19).

The sequence of events could be presented as follows : (i) weathering of magma, and dissolution of the biogenic sediment below the CCD limit ; a red clay develops at the bottom of the ocean ; (ii) erosion and transport of the red clay by bottom currents ; the red mixed material is then transported to deeper and quietie areas ; (iii) early diagenesis operating on the finally accumulated sediment. For each site, in each sequence of deposit, a set of geochemical processes is at work either successively or simultaneously.

On the other hand, by its mineral products as well as through the involved geochemical processes, halmyrolysis shows some similarities with hydrothermalism processes affecting a variety of rocks, mainly igneous and volcanic. However, this feature of argillogenesis will not be considered here.

9. TECTONIC SETTINGS OF MUD-DERIVED SEDIMENTS

At least nine types of sediment collector can be distinguished (Fig. 20). Most of these incorporate mud-or clay-derived materials which accumulate or accumulated either separately from other kinds of sediments or mixed with these. Some of these sedimentary basins are still active. Five main categories are recognised : shelf to basin, transition, continental, deep oceanic, island arc and rift systems (Potter *et al.*, 1984).

The clay minerals (their occurrence is blackened in Fig. 20) reach sedimentary basins and record (in their composition, crystallinities, relative abundance and types of association) the hallmarks of the continental "uphill" argillogenesis. Thus, the clayey sediments are essentially derived from continental sources by a simple, overall heritage mechanism (if one excepts the occurrence of neoformed clay minerals within the collector). In other words, when carefully scrutinized in close relationship with other macroscopic and microscopic characters of the sediments, it is always possible, even with some difficulties, to subdivide the clay mineral stock into a part directly related to continental heritage, and a part, if existing, related to syn- and/or postsedimentary events (i.e. precipitation in evaporitic conditions, etc...). Moreover, every sedimentary basin evolves in the framework of plate tectonics at a certain moment in its existence. Consequently, a kind of cyclic evolution arises and develops along the sequence : extension-regression -- transgression -collision-diagenesis -- orogenesis-(metamorphism) -- peneplanation with tectonic uplifts -- opening-spreading of the basin (Fig. 21).

However, not every sedimentary basin necessarily follows the above complete sequence.

Some stages may be missing while others are occurring in relay without passing through all the stages, whereas some of these stages develop along another path. This synthetic model of geological events occurring in a sedimentary basin, displays the way in which clay minerals occur. For instance, during an extension-regression phase, due to hinterland tectonic uplift, the clay minerals delivered to the nearby basin by a prograding delta may typically contain a series of "antagonistic" clay components. "Antagonism" refers to clay components developed "uphill" in possibly contrasting environment(s), and assembled "downhill" into a single assemblage.

Let us consider, as in Figure 21, that deltaic mud deposits are composed by illite (I), chlorite (C), kaolinite (K) and mixed layers (X). From a quick genetic interpretation of such an admixture it can be concluded, at first sight, that the first two clay minerals, illite and chlorite, being still fresh, are the products of an "uphill" erosion affecting an unweathered basement. In turn, kaolinite (K) is a characteristic neoformed mineral, that develops in an open, leaching system, under relatively high temperature and abundant rain. Finally, the mixed layers (whatever their composition) ordinarily point to a relatively moderate weathering of the substratum.

At the site of the accumulation, these "antagonistic" clay minerals, because of their association, reveal the effects of tectonics, which, as a driving factor, induce reelatively deep erosion of the substratum. Thanks to erosion and transport, the various continental clay stocks, once well separated on the continent, merged into a single clay stock during sedimentation.

What can be deduced from such an occurrence of diverse clay minerals, is that a fresh substratum has been affected by erosion and the products of the latter, i.e. I and C, freed and later delivered to the sedimentary basin. The mixed layers imply, by their occurrence into the sediment, a moderate either athmospheric or pedogenic weathering affecting the continental substratum, in the hinterland. As for the kaolinite component, its occurrence in the sediment points to a deep weathering of the substratum, not necessarily all over the continent; so the kaolinic mantle can be limited in extension prior to erosion. However I, C and K, as well as its the mixed layers (X), may be generated by simple erosion of a substratum which already contained this assemblage (i.e. former sedimentary rocks). So, at each particular step of the interpretation of an argillogenic process, one has to be very





Figure 19. : The submarine sedimentary cycle of red clays, and their sequential characteristis at the level of geochemical processes and mineral occurrence (modified after Hoffert, 1980).

cautious, especially when no links can be traced between the "feeding" continent and the receiving basin.

Another kind of interpretative "trap" concerns the hallmarks left on the clay assemblages by burial diagenesis. This process is known to induce important crystallochemical changes, and hence mineralogical modifications of the original clay stock. Consequently, for consolidated sediments, it appears hardly possible, if not impossible, to trace back the complete quality of the original clay mineral spectrum once having been accumulated in the basin. For example, a Paleozoic shale rarely reveals other clay components than an ubiquitous illite (mica)-chlorite "doublet" ! Usually, the search for clay mineral source(s) will not yield positive results when the erasing effects of diagenesis have been at work !

During peneplanation (Fig. 21) and under favourable climatic conditions, weathering can be very extensive and can deeply affect the substratum. A thick "leprosy", comprising clay minerals and other matters (i.e. hydroxides, oxides), covers the substratum. Below the surface, one can find a horizon with predominantly kaolinite. Below this horizon, the substratum appears less affected by weathering and can display some residual parent minerals (such as illite and chlorite), but also mixed layers. These are the true representatives, the "mutants" of the weathering in the upper layers, where kaolinization represents the leaching process of all the original bases, and of a part of the silica. The ion-enriched solutions, being removed by gravity, reach the low lands where they may be stopped and concentrated.

If evaporitic conditions exist, neoformed minerals such as Fe-beidellite may develop in vertisols. Along an "uphill-downhill" transect, one can thus find kaolinite in the high reliefs, and in the low areas, a "contemporaneous" smectite. Both minerals are themselves superposed, directly or indirectly, on a fresh substratum containing its characteristic clay minerals (i.e. illite and chlorite).



Figure 20. : Schematic models of sedimentary basin, and the relationship between the types of basin and the geometric and paleogeographic characteristics of muddy (clayey) detrital materials (modified after Potter, Maynard and Pryor, 1984).



Figure 21. : Relationship between tectonics and clay sedimentation and evolution (modified after Chamley, 1986).



Figure 22. : Composition and association of clay minerals within a sedimentary basin as a direct consequence of a upward regressive and downward erosion into the nearby continent substratum (modified after Chamley, 1986).

When looking at the tectonic events which possibly affect such a transect, several possibilities exist (Fig. 22). The composition of the clay minerals accumulated in the nearby basin will depend on the erosion intensity. Erosion may affect separately the different sectors covered by the transect. It is either limited to nearshore areas where only the smectite cover is removed, or the smectite with the underlying phyllosilicates (illite and chlorite) are removed ; or the erosion develops essentially in the hinterland reliefs where kaolinite is accumulated. This mineral alone is eroded and brought to the basin with other clay minerals, in particular mixed layers and, possibly, with fresh illite and chlorite from the substratum.

A more general case is that of downward and upward erosion of the clay cover with the consequence that the sediment collector accumulates the complete lateral spectrum of clay minerals. These, after being accumulated in the basin, bear in their structure, composition, abundance and in particular their association, a direct "finger print" of the variable geochemical landscapes which gave birth to them separately in space, and possibly also in time.

In sedimentary basins older than the Cenozoïc, the original clay assemblage may become faded through diagenesis inducing more or less intense crystallochemical and compositional changes in the final clay assemblages. Aggradation, neoformation, and (re)crystallization in particular, can mask the former assemblages once these are accumulated at the surface of the basin. Simplified but herited assemblages occur or new ones may develop. For instance, an original kaolinite-bearing feldspathic sand may become transformed during diagenesis into a kaolinite-illite sandstone in which the final thermodynamic and crystallochemical stable state is achieved.

10. DIAGENETIC SINK OF CLAY MINERALS DURING BURIAL AND DIAGENESIS

Figure 23 schematizes, in a single comparative representation, the diagenetic fate or diagenetic sink that affects the clay minerals when these return to the depths of the lithosphere. This journey back to the depths encompasses the transformation of a mud into a clayey sedimentary rock. From the surface to the deep epizone, where clay minerals usually gain their ubiquitous status of a mixing of illite and chlorite (as found in shales and slates), the clay minerals are "channeled" into several zones step by step, namely early diagenesis, late diagenesis and anchizone, before reaching the epizone.

In parallel, with increasing temperature and pressure, the interstitial solutions are concentrated and induce the change from an open system into a close confining and then confined environment. Several minerals, such as illites and kaolinite, also change structurally by possibly modifying their polymorphs. Figure 23 depicts the occurrence of some hydrothermal events. These take place at great dephts or near the surface of the lithosphere. Their products may become exposed at the surface Their clay through tectonic uplift and erosion. minerals, oin their turn, eventually contribute to the stock of clay material that reaches the sedimentary basin through erosion and transport.

Also, it is worth bearing in mind that the very transformation of a mud into a mudstone or a shale implies a change of fabric that characteres the originally accumulating clay particles. Some compositional peculiarities of the original mud (clayey) material such as the presence or absence of organic, compound, yield either the mudstone or the shale acquisition. This petrographic and mechanic property was illustrated by Moon and Hurst (1984) and is schematized in Figure 24.

CONCLUSION

Argillogenesis or the acquisition of the clay status is a rather complex hydrosphere "machinery".

Its motion is driven by the interplay of a series of both external and internal factors or stimuli. Ionchromatography, either by substraction or by addition, modulates the transformation of the parent silicate-bearing materials (or minerals) into clay minerals. The transformation, which develops through degradation or aggradation, neoformation, etc., corresponds to a true *ion-chromatography* within the layered structure of the clay minerals.

The mixed layers appear as the real representatives of the transformation, inducing a kind of crystallochemical, and thus compositional, mutation. In general, weathering takes place at the and contact between the lithosphere the hydrosphere, mostly in continental settings. However, transformation into clay may also arise in deep oceanic waters and into hydrothermal environments. Neoformation of clay minerals can also occur in the sedimentary basin, provided that specific favourable conditions prevail. The weathered products are eventually eroded and transfered into the nearby sedimentary basins. Here, heritage of clay minerals largely prevails, whatever the real geochemical processes which produced them during their continental location may have been. This heritage acts uphill of burial.

The different geochemical mechanisms here reviewed also occur in the sediments and affect them when they are buried into the depths of the lithosphere and face diagenesis. The latter also evokes, through its effects on the clay assemblages, a kind of mineral "sink". Such a compositional "sink" is commonly represented in mudstones and shales or even slates, by the phyllosilicate "bedmates", illite and chlorite. The anonymity of the latter minerals always embarrasses the sedimentologists and above all the paleoclimatologist. In particular, diagenesis erases the mixed layers, the "finger prints" of argillogenesis.

However, - and this is important - the apparent disappearance of labile mixed layers might only be a crystallochemical "trap". Such may be the case of mud-derived sediments that had reached the collector but were originally composed only by original illite and chlorite because of unfavourable climatic conditions for their uphill, continental weathering. Such a hypothesis (especially in case of hard sedimentary rocks) can never be checked by hard facts. The products of weathering are represented by a nearly infinite combination of compositions, crystallinites, relative abundances and kinds of clay association, all of these being the hallmarks for silicates facing new thermodynamic and physico-chemical conditions or contraints once the parent materials reach the surface through tectonic uplift and, there, become exposed to (pedi)peneplanation and atmospheric or pedogenic weathering.



Figure 23. : The diagenetic fates for kaolinite, smectites, illites, chlorites and mixed layers (modified after Dunoyer de Segonzac, 1970, and Hayes, 1970).



Figure 24. : Cartoon depicting microstructural changes of clayey sediments, from sedimentation towards lithifaction, with emphasis on clay domain formation (modified after Moon and Hurst, 1984).



Figure 25. : The geochemical cycle represented as a continuum in the Earth's lithospheric crust, in the way of the Dutch artist Escher.

The various geochemical pathways developed or self-oriented along certain compositional goals can be engraved along a geochemical continuum (Fig. 25). On the split ribbons of this continuum, there occur two "sinks", one related to sedimentogenesis, the other to diagenesis. Through physical or physico-chemical weathering, the silicate reservoir is generating "uphill" various clay minerals through a variety of processes : heritage, degradation, aggradation, neoformation, precipitation, crystallization.

To sum up, let us quote Erhart (1956) : "A clay mineral found in a soil (or in a sediment) in general carries along a very long history which cannot be explained in a simple way". In this paper, a synthetic approach of the argillogenic realm has been attempted through graphic presentations, not unlike "strip-cartoons". It is the belief of the author that this way of presentation will offer some attractive effects to all those readers and in particular to Dr. Eva Paproth, allowing them to gain knowledge of the clay mineral responsible for the huge amount of detrital sediments and sedimentary rocks in the stratigraphy, which contain mainly clay minerals or their diagenetic derivatives.

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