

# MINERALOGICAL AND GEOCHEMICAL STUDY OF CLAY MINERAL TRANSFORMATIONS IN THE SEDIMENTARY TRIASSIC JURA BASIN (FRANCE)

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**Abstract**—A detailed study of sediments from the Triassic of the French Jura shows that clay minerals vary continuously with the facies. Near the coasts of the Jura Sea, sediments consisted of sandy, continental detritus and the minerals are poorly crystallized. At the center of the basin, in what was a cation-rich chemical environment, they are well crystallized. A progressive mineralogical variation can be observed from degraded illite to well-crystallized chlorites passing through the intermediate stages of more-or-less regular mixed-layer structures. This variation is due to a true crystalline transformation.

Similarly, chemical variations in clay minerals and whole-rock samples are related to the paleogeography of the basin. The distribution of major and trace elements is a function of distance from the Jura Sea coastline and also a function of the mineralogical composition of the fine fraction. The most obvious relationships are: (1) An increase in the concentration of MgO and the percentage of ignition-loss water from the coast toward the deep sea; (2) A decrease in the concentration of SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, and TiO<sub>2</sub> as well as that of trace elements such as vanadium, gallium, and cobalt from the coastal regions to the center of the basin; (3) A lack of statistically significant variations in the concentration of Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>3</sub>O<sub>4</sub>, B, and Ni throughout the basin.

The authors conclude that: (1) Transformations (aggradations) observed by means of X-ray diffraction methods are in agreement with the chemical analyses; (2) Transformations are contemporaneous with the sedimentation; they are not diagenetic; (3) Clay minerals play an important role in the geochemical balance of sedimentary basins.

## INTRODUCTION

AT THE 10th National Conference on Clays and Clay Minerals in 1961 G. Millot presented preliminary results on transformations of clay minerals in the Triassic sediments of the Jura Basin (Millot, Lucas, and Wey, 1963). Since then, two studies on this basin have been completed, one on the mineralogy (Lucas, 1962) and the other on the geochemistry of these sediments (Ataman, 1967). This paper synthesizes these investigations.

### SUMMARY OF THE PALEOGEOGRAPHICAL EVOLUTION OF THE TRIASSIC JURA BASIN

The sedimentary evolution of the basin was reconstructed from analyses of samples from eight drill holes. Three of the drill holes are located on the eastern side of the Jura chain; Fraignot 1 and 2 ( $F_1$  and  $F_2$ ) and Montcoy<sub>1</sub> ( $My$ ). The other five, Montbouton<sub>1</sub> ( $Mb$ ), Lons<sub>3</sub> ( $L$ ), Toillon<sub>1</sub> ( $T$ ), Valempoulières ( $V$ ) and Laveron

( $La$ ) are in the Jura proper. The thickness of total Triassic sediment increases from Fraignot, where it is 100 m, to Laveron, where it exceeds 1400 m.† The drill holes nearest the Massif Central are the most detrital and sandy. Toward the center of the Basin clay, carbonate and evaporite facies increase in abundance and become predominant at the Laveron drill hole where salt formations are about 600 m thick.

The topography of the Jura region during the Triassic must be reviewed in order to understand the sedimentological history of the basin. More detailed paleogeographic reconstructions may be found in Ricour (1962) and Lucas (1962). Jura Basin sedimentation occurred sufficiently after the Hercynian Orogeny that continental relief was minimal. The Triassic sea encroached upon a continental shelf bounded by relatively smooth and gently rolling coasts. The Jura region was

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†A figure of the basin with the location of the drill holes is given in Lucas (1962), Millot, Lucas, and Wey (1963), Ataman (1967).

covered by a shallow epicontinental sea bounded on the western side by the Massif Central and on the east by the edge of the continental slope. This epicontinental sea faced the deep Mesogean sea, the location of the present Alps. The Jura Basin was separated from the Alpine Sea by a shallow sill or bar at the edge of the Continental Shelf (Fig. 1).

supplied with detrital sediments from the Massif Central to the west and with evaporite deposits caused by supersaturation of evaporating sea water.

#### STUDY OF CLAY MINERALS (LUCAS, 1962)

In the sandstone facies, adjacent to the Triassic coast line, the clay minerals are principally

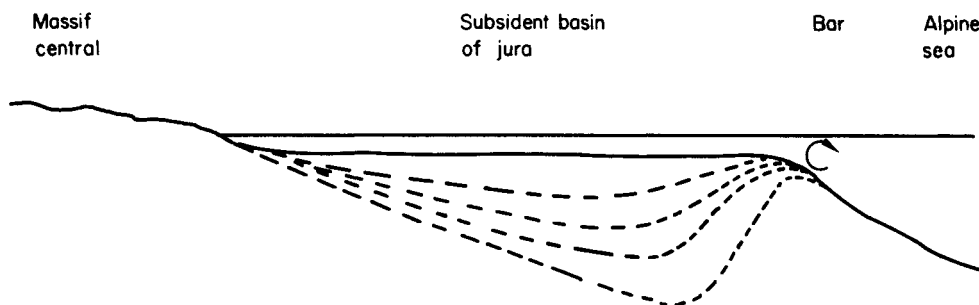


Fig. 1. Section across reconstructed Triassic Jura Basin.

Principally, carbonate sediments of the Alpine facies were deposited in the deep sea while on the continental shelf, sedimentary deposits of the Germanic facies accumulated. The thickness of the formations shown by the drill holes demonstrates that an important subsidence in the vicinity of Laveron affected the Continental Shelf, but it is likely that sedimentation took place constantly under only shallow water conditions.

In this topographic context, it is certain that the only detrital continental source of sediment was the Massif Central on the western flank of the basin. On the other side of the basin, the epicontinental sea communicated freely with deep sea. It is possible that in some places a submarine shelf separated the two environments, but it is more probable that the edge of the continental plateau itself, where the bar acted as a sill, constituted a sufficient barrier.

Under the warm, although certainly not arid, Triassic climate, evaporation was intensive and the water supplied to the continent was insufficient to replenish the amount lost by evaporation; thus a permanent current was created from the open sea toward the sea of the continental plateau. This process of increasing salinity is similar to that described by Ochsenius (1877) and by Sloss (1953). On the western side of the basin, the salt water was diluted by fresh waters draining from the continent which precluded precipitation of salt deposits.

In summary, Jura Basin sediments were deposited on a subsiding continental shelf and were

degraded illite which, in X-ray diffractograms, show a  $10 \text{ \AA}$  reflection, broad and open toward small angles. This illite is accompanied by small percentages of kaolinite and chlorite, also poorly crystallized. Electron micrographs of these clays reveal very small particles without apparent crystalline shapes and concentrated in rounded aggregates. We conclude that these minerals represent the detrital contribution originating in the exposed rocks of the Massif Central.

Going basinward from the coast, the facies become less sandy and the composition of clay minerals progressively changes. Mixed-layer minerals occur, as evidenced by broadening of the illite reflection toward small angles. Progressing basinward, a diffuse reflection appears between  $10$  and  $14 \text{ \AA}$ , and then the reflection tends to become distinct nearer  $14 \text{ \AA}$  and forms a mixed-layer mineral of the  $(14_c-14_M)$  type.

Still farther from the coast, this mixed-layer mineral becomes more regular, resulting in an excellent corrensite. But the evolution is not finished because corrensite gives way to a new mixed-layer mineral, more like chlorite, and of a  $(14_c-14_c)$  type. Then, in the drill hole at Laveron, in the thickest part of the sedimentary basin, the mixed-layer minerals give way to a well-crystallized chlorite which has strong, sharp reflections. Paralleling this progression of the  $14 \text{ \AA}$  population, X-ray reflections of illite also become sharper and approximate those of a well-crystallized mica.

This evolution may be summarized as follows: Degraded illite and chlorite  $\rightarrow$  irregular  $(14_c-14_M)$

mixed layers → corrensite → irregular (14<sub>c</sub>-14<sub>c</sub>) mixed layers → well crystallized chlorite.

The mineralogical evolution is also well defined by electron microscopy because, in the central parts of the basin, the predominant chlorite and illite constituents reveal well-formed hexagonal crystals exceeding 1  $\mu$  in length. This evolution which takes place from the coast toward the center of the basin is also more-or-less completely observed from the bottom to the top of the drill holes, the oldest or deepest sediments being less well crystallized than the youngest or topmost sediment. A similar relation between the nature of clays and the petrology of sediment has been pointed out by Millot, Lucas, and Wey (1963).

This variation of clay minerals across the basin is explained by a process of continual transformation. If, in the center of the basin, chlorite and illite appear in well-formed, large crystals, they could not have originated directly as detritus from the continent. Near the Massif Central, the only detrital source for the basin, clay minerals are very small and poorly crystallized. As the particles progress into the basin, they enter an environment increasingly rich in dissolved cations. The least-degraded illites pick up potassium, improving their crystallinity. The illites which are too degraded to be thus reconstituted, take up magnesium, becoming mixed-layer minerals and, ultimately, well-crystallized chlorite. This is not a true neof ormation [or authigenic formation] from a solution, but a transformation from a degraded mineral into a different, well crystallized one, the mixed layering constituting intermediate stages in the process. This inverse process of degradation is called "aggradation" (Lucas, 1962).

#### CHEMICAL STUDY OF SEDIMENTARY ROCKS (ATAMAN, 1967)

More than one thousand complete major and minor element analyses by direct reading emission spectrometry have been made upon the total rock samples and upon the extracted clay fraction (Ataman, 1963; Ataman and Besnus, 1965). We shall not detail here the distribution of these elements in the basin as this has been summarized in Ataman and Lucas (1967). Rather we shall discuss the evolution of some typical elements by considering variations in the average concentrations of the elements or their chemical compounds.

Figure 2 shows the variations between drill holes in the average concentrations of MgO, Al<sub>2</sub>O<sub>3</sub>, and SiO<sub>2</sub>. It is obvious that the silica content of the total rock decreases considerably from the coast to the open sea. This is easily explainable since the concentration of detrital minerals,

especially quartz, the most silicious component, decreases in the same direction. But silica, which is a constituent of clay minerals, also decreases in a regular, progressive manner.

The variations of Al<sub>2</sub>O<sub>3</sub> in the total rock and in clays, are very similar. This proves that alumina is principally contained in the clay minerals and that the clays are less aluminous toward the open sea than near the coasts.

Contrary to the trend of silica and alumina, MgO increases strongly in the clay fraction from the coast toward the open sea, but remains almost constant in the whole rock. Now in the most seaward drill holes, carbonate and salt facies are more abundant, and the absolute amount of clay decreases. However, because the clay minerals are more and more magnesian, despite their decreasing abundance, the MgO content of the whole rock remains almost constant.

A comparison of the chemical composition of the clay fraction in different facies is given in Fig. 3. Note that silica and alumina decrease between detrital and chemical facies, whereas magnesia increases strongly. These chemical variations in the clays across the basin agree perfectly with the mineralogical variations described above.

Moreover, variations in trace element concentrations also reflect the same evolution. Thus, vanadium, gallium, and cobalt decrease from the coast toward the open sea, while chromium, zinc, copper, and lead increase. Nickel and boron remain approximately constant across the basin. We shall not attempt here an explanation of all these variations but will limit the discussion to boron. Minor element variations have been statistically correlated with major element variations elsewhere (Ataman, 1967).

The boron content of the clays as opposed to whole rock remains approx. 300 ppm across the basin despite an inferred, regular increase in salinity from the coast to the open sea. Boron has often been described as an indicator of salinity (Goldschmidt and Peters, 1932; Landergren, 1945, 1958, 1959; Degens *et al.*, 1957; Tourtelot *et al.*, 1961; Walker, 1963; Harder, 1964). There is thus in the sediments of the Jura Basin an anomaly which requires explanation.

Calculations of statistical correlations between different major and minor elements (Ataman, 1967) show a positive dependence between aluminum and boron. Because, as we have noted, the aluminum content decreases regularly toward the center of the basin, the boron content should also decrease, but this is not the case. Under the influence of increasing salinity, however, the ratio of boron to aluminum does increase, so it is because aluminum decreases that boron remains

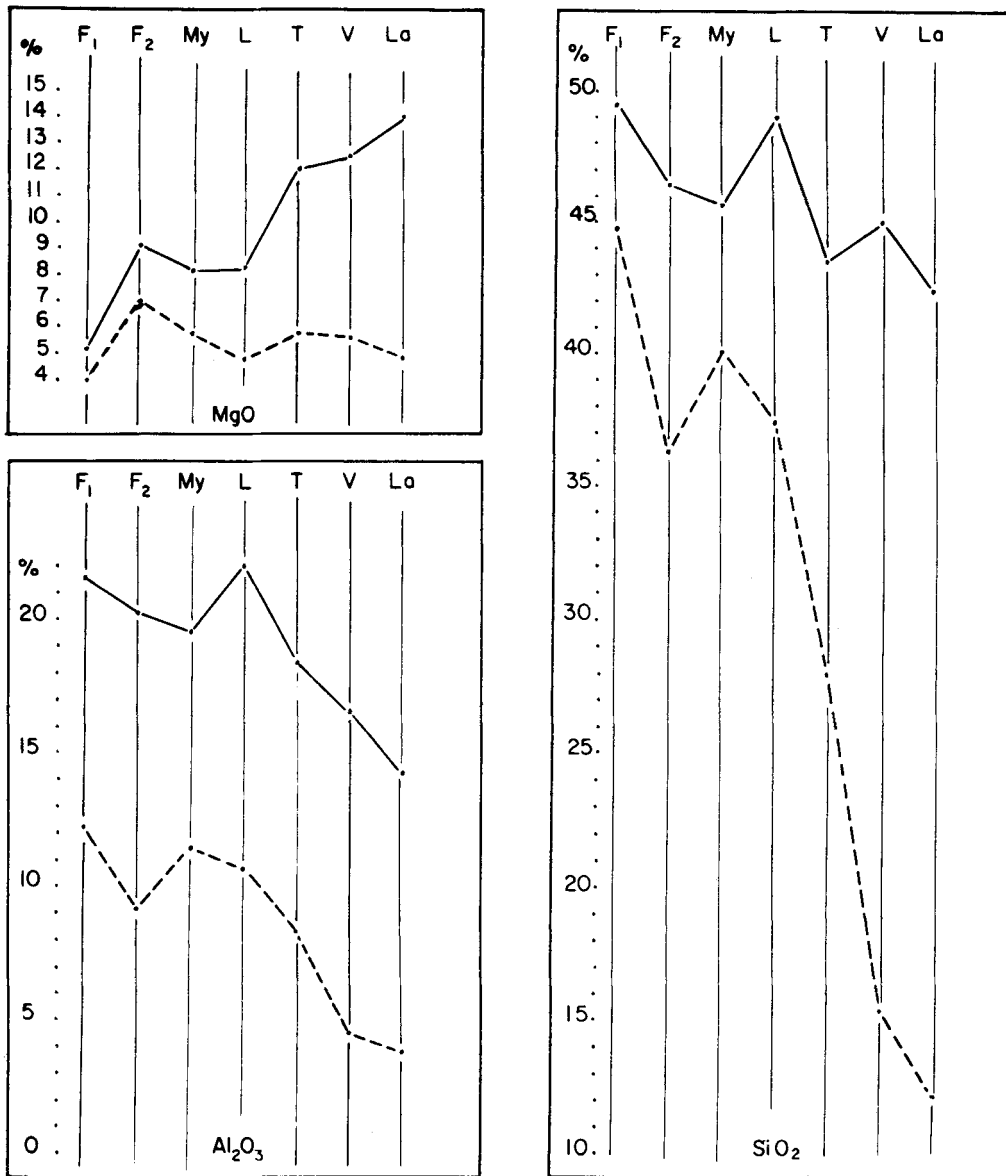


Fig. 2. Variations in the chemical composition of the clay fraction (—) and of the whole rock (-----).

constant. Hence, the concentration of boron does reflect an increase in salinity, but its correlation with aluminum disturbs the phenomenon. One may thus use the boron content of clays as an indicator of salinity provided that the mineralogical composition of samples to be compared remains constant.

The chemical results confirm the phenomenon of clay mineral transformations in the Triassic Jura Basin and permit its more precise definition. The detritus supplied to the basin consists princi-

pally of degraded 2:1 micaceous clay particles, Tet-Oct-Tet, i.e. degraded illite. Interlayer positions of these particles are largely stripped of cations by continental weathering, leading also to a degradation of the silicate layer itself. Upon arrival in an environment of increased dissolved cation concentration, these interlayer positions tend to refill, especially with magnesium ions for which clay minerals seem to have the strongest affinity. Thus, as magnesium is fixed, degraded lattices are progressively rebuilt to form mixed

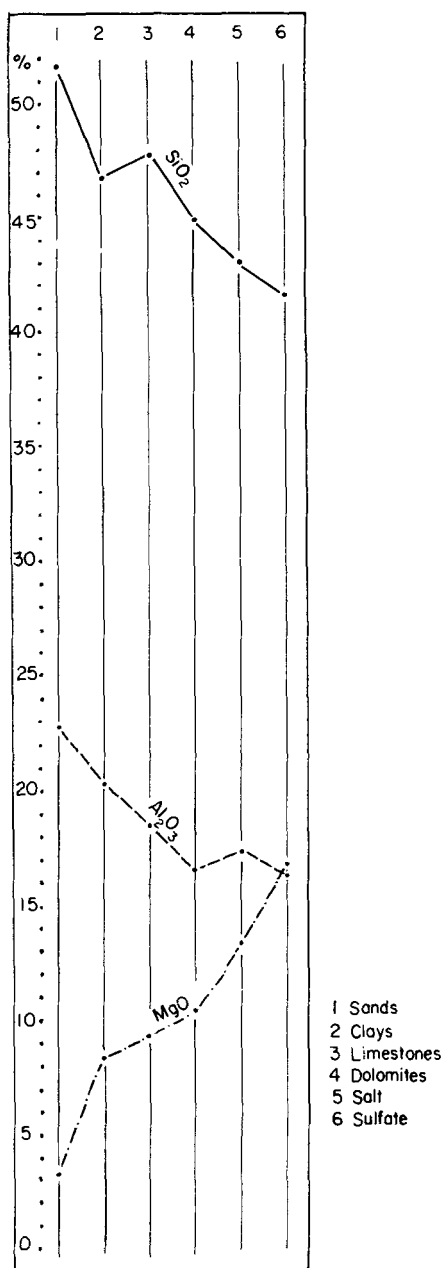


Fig. 3. Variations in the chemical composition of the clay fraction of different sedimentary facies.

layers, first irregular, then regular (corrensite), and result finally in a well-crystallized chlorite. This transformation is accompanied by an increase in crystallite size.

The increase in the amount of magnesium contained in the clays is accompanied by a decrease

in aluminum and silicon. It is not, however, because of an increase in dissolved magnesium that this transformation occurs, since whole rock analyses show that magnesium remains constant across the basin, but is rather a function of supply and demand. Near the coast, degraded particles are numerous and the concentration of available, dissolved magnesium is too low to fill all the potential magnesium sites. Farther from the coast, the number of particles decreases and each of them has available more magnesium. But clay minerals are not the only sites possible for magnesium; it is incorporated into carbonates also. This explains why in carbonate facies, especially dolomitic facies, clay minerals are transformed only slightly. On the other hand, in saline environments where cation concentrations are not sufficient for highly-soluble magnesium chloride or sulfate to form, magnesium remains entirely available for clays which can thus undergo extreme changes. But even in these super-saline environments, the competition for magnesium, though not vigorous, is none the less important. In the drill hole at Laveron, alternate layers of massive salt and clay correspond to alternate populations of either corrensite or chlorite respectively (Lucas, 1962; Millot, Lucas, and Wey, 1963). When salts precipitate, they remove magnesium and clay minerals do not alter beyond the corrensite stage. When clays are the principal materials deposited, all the magnesium is available to them and the clay minerals reach the final aggradation stage of chlorite.

#### DISCUSSION OF MINERAL TRANSFORMATIONS

##### *Transformation or neof ormation*

One may possibly conceive of the minerals found in the Triassic clays as resulting from neof ormation, i.e. precipitated from ions in solution. Two arguments, however, oppose this idea.

1. The clay minerals form a continuous sequence across the Jura Basin, passing smoothly from one type to another. Neof ormation would require imagining precipitation of irregular mixed-layer minerals in a precarious equilibrium, across a changing environment.

2. The neof ormation of well-crystallized illite and chlorite in a sedimentary basin does not appear possible. In fact, these minerals consist of highly-charged silicate layers. They have been synthesized by many workers under high temperature and pressure, but never under pressure and temperature conditions which exist at the earth's surface. Thermodynamic conditions at the surface do not permit the neof ormation of minerals with electrically-unbalanced charges on their layers, which require compensating cations for electric

neutrality. In addition, illite and chlorite require aluminum in order to form. But aluminum, almost insoluble and very immobile, would not be available in solution in the waters of sedimentary basins. The only minerals actually recognized as certainly "neoformed" in sedimentary basins, or synthesized under normal temperature and pressure conditions, are neutral or approximately neutral. Examples are attalugites, sepiolites, and probably some trioctahedral smectites of weak, or completely-balanced electrical charges. Stevensite may be such an example. They are essentially magnesium minerals. Magnesium, very soluble in water, is used in the place of aluminum in neoformation.

#### *Transformation and diagenesis*

Many arguments exist against the interpretation that these transformations have occurred during diagenesis, i.e. after the deposition of the sediments. Four which appear to us demonstrative will be considered.

1. Transformations and crystallinity of minerals do not correlate with depth of burial. In a single drill hole, if transformation were diagenetic, a deterioration of crystallinity ought to be found from the bottom to the top. The opposite is in fact observed; the best crystallized minerals occur in the highest levels of the Triassic sediment.
2. Diagenesis by compaction should result in a decrease in water loss on sample ignition, formation water having been squeezed out by increasing overburden pressure. On the contrary, the better-crystallized minerals contain more water in the Triassic Jura sediments.
3. The correlations between certain of the trace and major elements (especially vanadium, nickel, and cobalt with magnesium oxide) are opposite to those observed in metamorphism. In addition, the behaviour of boron is readily explained by its introduction into clay minerals during—not after—sedimentation. It is difficult to imagine a diagenetic mechanism which could homogenize the boron content of the sediments on such a large scale.
4. The corrensite-chlorite mineralogical association is not only found in the French Jura. It is frequently observed in Permian sediments and in European Triassic sediments of Germanic facies: England (Honeyborne, 1951); Germany (Fuchtbauer and Goldschmidt, 1959; Lipmann, 1956); France (Lucas, 1962); Spain (Martin-Vivaldi and MacEwan, 1957; Lucas, 1962); in the Permian of North America (Grim *et al.*, 1960; Fournier, 1961; Peterson, 1962; Tooker, 1962). This association was also found recently in the laboratory at Strasbourg in sediments of similar age from South America, in the formation of Karroo age. Such a distribution seems certainly to be the result of a sedimentary process characteristic of a geological period, and not the result of a diagenetic phenomenon.

In conclusion, it appears that clay minerals, degraded by weathering on the continents, are very sensitive to the sedimentological environment and reflect, by chemical variations, changes in their environment. Their ability to fix ions from solutions gives them an important role in the geochemical balance of sedimentary basins. In addition, their susceptibility to transformation makes them a useful tool in paleogeographic reconstructions.

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**Résumé**— Une étude détaillée de dépôts du triassique dans le Jura montre que les minéraux argileux varient de manière continue avec le faciès. Près des côtes de la Mer du Jura, les dépôts se composaient de détritux continentaux sablonneux et de minéraux peu cristallisés. Au centre du bassin, dans un milieu chimique qui était riche en cations, ils sont bien cristallisés. On peut noter une variation minéralogique progressive qui s'étend de l'illite dégradée à des chlorites fortement cristallisées, en passant par des stades intermédiaires de structures à couches mélangées plus ou moins régulières. Cette variation résulte d'une véritable transformation cristalline.

De même, les variations chimiques des minéraux argileux et des échantillons de rocs entiers se rapportent à la paléogéographie du bassin. La distribution d'éléments majeurs et d'éléments de trace est fonction de la distance de la côte de la Mer du Jura et aussi de la composition minéralogique du pourcentage en éléments fins. Les rapports les plus évidents sont les suivants: (1) Augmentation de la concentration en MgO et du pourcentage d'eau perdue au feu à mesure que l'on s'éloigne de la côte; (2) Diminution de la concentration en SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, et TiO<sub>2</sub> ainsi qu'en éléments de trace tels que le vanadium, le gallium et le cobalt à mesure que l'on s'approche du centre du bassin; (3) Pas de variations statistiquement vérifiables dans les concentrations en Fe<sub>2</sub>O<sub>3</sub>, Mn<sub>3</sub>O<sub>4</sub>, B, et Ni, dans tout le bassin.

Les auteurs arrivent aux conclusions suivantes: (1) Les transformations (remplissages par déposition) observées grâce à des méthodes de diffraction de rayons X concordent avec les analyses chimiques; (2) Les transformations ont lieu à la même époque que la sédimentation; elles ne sont pas diagénétiques; (3) Les minéraux argileux jouent un rôle important dans l'équilibre géochimique des bassins sédimentaires.

**Kurzreferat**— Eine ausführliche Untersuchung von Sedimenten aus der Trias der französischen Jura zeigte, dass sich die Tonminerale fortlaufend mit der Fazies verändern. In der Nähe der Küste der Jurasee bestehen die Sedimente aus sandigem, kontinentalem Detritus und die Minerale sind mangelhaft kristallisiert. In der Mitte des Beckens, wo ein kationenreiches chemisches Milieu vorhanden war, sind sie gut kristallisiert. Eine fortlaufende mineralogische Veränderung kann beobachtet werden, ausgehend vom erodierten Illit bis zu wohl ausgebildeten Chloriten über die Zwischenstufen von mehr oder weniger regelmässigen Mischformationen. Diese Veränderung beruht auf einer echten kristallinen Umwandlung.

Gleichermassen stehen die chemischen Veränderungen in Tonmineralen und Vollgesteinproben in Beziehung mit der Paläogeographie des Beckens. Die Verteilung der Haupt- und Spurenelemente ist eine Funktion des Abstandes von der Küstenlinie der Jurasee und ebenfalls eine Funktion der mineralogischen Zusammensetzung der Feinfraktion. Die offensichtlichsten Beziehungen sind: (1) Eine Zunahme der Konzentration von MgO und des Prozentanteils von Glühverlust Wasser von der Küste

gegen die Seetiefe hin; (2) Eine Abnahme der Konzentration von  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  und  $\text{TiO}_2$ , sowie der Spurenelemente wie Vanadium, Gallium und Cobalt in der Richtung von den Küstengegenden zur Mitte des Beckens; (3) Eine Abwesenheit statistisch geltender Änderungen in den Konzentrationen von  $\text{Fe}_2\text{O}_3$ ,  $\text{Mn}_3\text{O}_4$ , B und Ni im Bereiche des Beckens.

Die Autoren folgern, dass: (1) Umwandlungen (Aufschüttungen), die durch Röntgenbeugungsmethoden beobachtet wurden, mit der chemischen Analyse übereinstimmen; (2) Umwandlungen mit der Sedimentation zeitlich zusammenfallen; sie sind nicht diagenetisch; (3) Tonminerale eine wichtige Rolle im geochemischen Gleichgewicht sedimentärer Becken spielen.

**Резюме**—Подробное исследование отложений триасской системы французской Юры показывает, что глинистые материалы постоянно меняются с фациями. Вблизи побережья Юрского моря отложения состояли из песчаных континентальных наносов, а минералы слабо кристаллизованы. В центре бассейна, там где существовала окружающая среда с крупным наличием катионов, они хорошо кристаллизованы. Наблюдалось постепенное минералогическое изменение, начиная с деградированного иллита и кончая хорошо кристаллизованными хлоритами, проходя через промежуточные стадии более или менее регулярных структур смешанного слоя. Изменение это является следствием настоящего кристаллического превращения.

Сходным образом, химические изменения глинистых минералов и породных образцов связаны с палеогеографией бассейна. Распределение крупных или малых элементов является функцией расстояния от береговой линии Юрского моря, а также функцией минералогического состава мелкой фракции. Наиболее очевидные соотношения нижеследующие: (1) Повышение концентрации MgO и процент воды с потерями от прокаливания, от берега по направлению больших глубин моря; (2) понижение концентрации  $\text{SiO}_2$ ,  $\text{Al}_2\text{O}_3$  и  $\text{TiO}_2$ , а также рассеянных элементов, как ванадий, галлий и кобальт из береговых районов к центру бассейна; (3) отсутствие статистически значительных изменений в концентрации  $\text{Fe}_2\text{O}_3$ ,  $\text{Mn}_3\text{O}_4$ , B и Ni по всему бассейну.

Авторы пришли к заключению: (1) что превращения (береговые отложения), наблюдаемые пользуясь методами дифракции рентгеновских лучей, соответствуют химическим анализам; (2) что превращения происходят одновременно с осадкообразованием и не являются диagenетическими; (3) что глинистые минералы играют важную роль в геохимическом равновесии осадочных бассейнов.