

WEATHERING AND ORIENTATION IN TRIASSIC CLAY SEDIMENTS OF NEW JERSEY

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Abstract—The composition of clay mineral suites derived from Triassic argillite and shale were compared with those of the consolidated parent sediments. Lockatong argillite, near the weathered zone, is composed mainly of illite, chlorite, and feldspar. In the clay horizon immediately above it, illite replaces the feldspar and kaolinite increases with distance from the contact until, near the top of the weathered zone, it is almost the only clay mineral present. A similar study was carried out on Brunswick shale. Here the parent rock consists mainly of illite, and some kaolinite, montmorillonite, quartz, and feldspar. The kaolinite increases gradually as one progresses upward through the weathered zone, montmorillonite decreases, but approximately 30 per cent illite is still found in the top layers. Chlorite in the argillite near the weathered zone is randomly oriented while illite shows strong preferred orientation, probably indicating that chlorite formed in place during weathering. Illite in the Brunswick shale shows strong preferred orientation.

These two Triassic sediments are less than 5 miles apart and one may assume that they had a similar post-depositional environment. The difference in diagenetic development is striking and must be attributed mainly to the chemical composition and lithology of the parent sediment.

The orientation of clay particles in the solid rock was studied with the aid of an X-ray diffraction technique based on the analysis of cylindrical samples. Three-dimensional intensity data thus obtained were presented by vectors whose ends form a surface which is characteristic of the orientation of the particles. Generally the vectors generate hemispherical normal distributions whose principal vector coincides with the normal to the bedding plane.

INTRODUCTION

RESIDUAL clays derived from Triassic argillites and shales of New Jersey show noteworthy differences in their weathering characteristics. The residual clays that were studied developed on the surface of the Triassic age sediments, before the deposition of the overlying Raritan formation (Cretaceous).

A number of recent papers deal with the weathering and diagenesis of clay minerals. The weathering mechanisms of some sedimentary rocks are discussed in an excellent monograph by Keller (1962). Fanning and Jackson (1962) have studied the alteration and weathering of clay minerals in soils. Heydemann (1966) determined rates of dissolution of reference clays under controlled laboratory conditions at pH between 3 and 10. Mica-type clay minerals, after having been subjected to intensive weathering under natural conditions, were

studied by Guven and Kerr (1966). Harriss and Adams (1966) investigated the weathering of granitic rocks and related soils in terms of equilibrium relationships between soils and surface waters, and on that basis could characterize the soils as having reached maturity or as still undergoing change. Patterson (1964) compared the diagenetic processes resulting in saprolite and an adjacent underclay derived from the same parent material.

An attempt was made to combine the study of chemical and mineralogical changes through the weathering zones with an investigation of preferred orientation of the clay particles. The significance of preferred orientation studies in conjunction with chemical weathering lies in the fact that ionic replacement processes should have little or no effect on the orientation of the affected particles. Similarly, crystallographic overgrowths on nuclei should result in no significant changes in particle

orientation. If, on the other hand, clay minerals form as a result of ion migration to new centers, such as the surfaces of feldspar particles, one should not expect to find a tendency toward preferred orientation. The method used to study and characterize preferred orientation is based on the discovery that the orientation of particles in the parent rock may be characterized in terms of hemispherical normal distributions (Pincus, 1953). Because of the large number of uncertainties regarding fundamental chemical and structural characteristics of the clay minerals in a natural assemblage, the investigation of preferred orientation was based on X-ray intensity data collected from various surfaces of the specimens, rather than on ratios of intensities of reflections from different crystallographic planes obtained from one sample surface (Martin, 1966; Brindley and Kurtossy, 1961; Meade, 1961; Brindley, 1953).

THE PARENT ROCKS

Composition and environment of deposition of the Brunswick shale and the Locketong argillite are described in detail in three recent papers by Van Houten (1964, 1965*a*, *b*). In the locality studied, the argillite is a tough, dark brown, fine-grained and apparently homogeneous mudstone and marlstone. The remarkable hardness of these rocks cannot be due to silicification because of their low silica content. Probably the major contributors to the hardness are recrystallized carbonates and alumino-silicates. The argillite is lacustrine in origin and was deposited in many complicated detrital and chemical cycles. These rocks are rich in sodium, either as feldspar or as analcime. Quartz is almost absent. Rocks of the Brunswick formation are reddish-brown feldspathic shales (mudstones) and siltstones. Quartz, unlike in the argillites, is abundant in these rocks. Both Na and K Feldspars are present. These sediments are believed to have accumulated on mudflats, in an environment less saline than that of the argillite (Van Houten, 1965*b*). The red shale of the Brunswick formation weathers rapidly on direct exposure to the atmosphere, while the argillite is very resistant to weathering. Although the boundary between the consolidated parent sediments and the residual clays is well defined, one finds some evidence of alteration in the parent sediments close to the contact. Up to 16 ft of residual clays are found on top of the consolidated rocks, and these, in turn, are overlain by Cretaceous sediments. The stratigraphic relations of the residual clays are described by Servilla (1960).

METHODS OF MINERALOGICAL AND CHEMICAL ANALYSIS

Channel samples of residual clays were collected from the Cretaceous boundary downward to the consolidated parent sediment, in the following three localities. (see map, Fig. 1).

Almasi Pit, Woodbridge, N. J.; Brunswick shale and residual clay.

Wright Pit, Milltown, N. J.; Locketong argillite and residual clay.

Nixon N.J., Brunswick shale and residual clay.

For the purpose of identification the usual X-ray diffraction analytical methods were employed. The 14Å minerals were identified by first removing soluble iron and replacing all exchangeable ions by either magnesium or potassium in duplicate samples. To differentiate the minerals, samples were glycerated and heated to 300 and 550°C (Jackson, 1965). Kaolinite and chlorite were separated by treatment with warm dilute HCl (Brindley, 1951). The major elements were determined with a Norelco Vacuum Spectrometer after preparing pellets containing La_2O_3 as a heavy absorber (Rose *et al.*, 1962). For Na and Mg analyses, a thin-window flow counter and pellets without a heavy absorber were used. For orientation studies the undisturbed, moist clay specimens were indurated with Carbowax 6000 (Martin, 1966).

MINERAL AND CHEMICAL COMPOSITION

Table 1 and Figs. 2 and 3 show the changes in mineral composition from the top of the weathered zone to the consolidated parent sediments, both for the Locketong argillite and for the Brunswick shale. Chlorite, feldspar and illite are the major constituents in the Locketong parent rock; quartz was not detected.* Goethite found accounts for a minor amount of the iron. Some iron is present in chlorite and montmorillonite, but most of it is thought to be colloidal. The major constituents of the parent Brunswick shale are illite, quartz, kaolinite; mixed-layer clays, montmorillonite and feldspar are found in lesser amounts.

In Table 2 the corresponding major elemental composition as well as H_2O^+ and H_2O^- content are given. In the Brunswick suite the only sharp

*When the material is treated with NaOH (Methods of Soil Analysis, Jackson, 1965), the 14 Å mineral does not expand after glyceration. It collapses almost completely to 10 Å when heated to 300°C, and also when treated with KCl. These are characteristics of non-expanding Al-vermiculites; however, not enough H_2O is present in this mineral to allow for the presence of vermiculite. The treatment apparently altered the chlorite and made it unstable at elevated temperature.

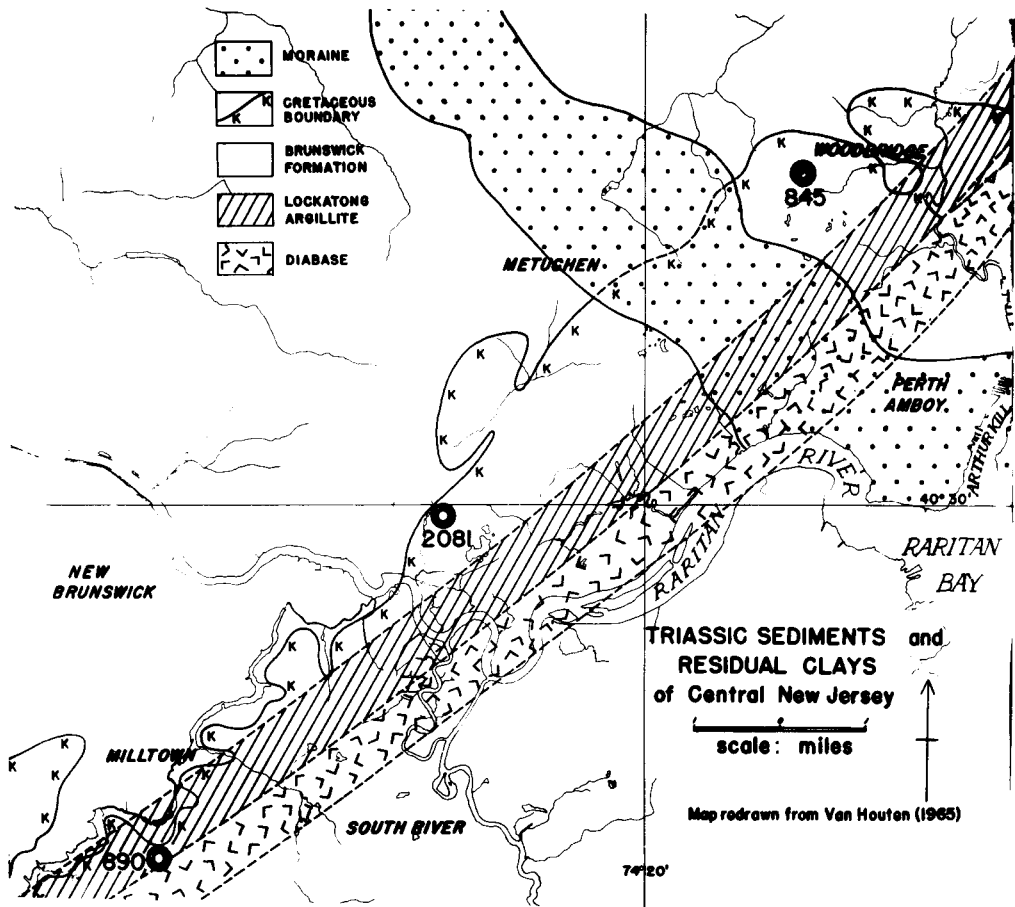


Fig. 1.

Table 1. Clay mineral suites derived from triassic shale and argillite

Mineral	Brunswick shale Sample 845				Lockatong argillite Sample 890			
	A	B	C	D	A	B	C	D
Mx	2	2	6	6	—	—	—	—
Mo	—	—	5	5	—	—	—	—
I	27	27	28	52	3	55	55	20
K	55	46	45	15	93	30	10	1
Q	18	22	12	16	—	—	—	—
F	3	3	3	5	2	2	3	32
Ch.	—	—	—	—	2	12	30	45

Key: Ch. = chlorite; Mx = mixed-layer clays; Mo = montmorillonite; I = illite; K = kaolinite; Q = quartz; F = feldspar.

break in composition is in the iron content between horizons C and B; CaO decreased to trace amounts between D and B. H₂O content increased steadily from D to A. The alkalis, MgO, SiO₂ and Al₂O₃, remained fairly constant through the whole section, except for some loss of Mg as one passes from D to C. The Lockatong suite is significantly different from the Brunswick suite. Na₂O and CaO decrease sharply from D to C, MgO content shows a sharp break as one proceeds from C to B, while K₂O and Fe₂O₃ decrease sharply from B to A. Alumina and water of hydration increase steadily upward.

The changes in chemical composition through the sections correspond well with the changes in mineral composition. In the Brunswick suite the content of illite, the most abundant mineral in the D horizon diminishes to one half in C, and then

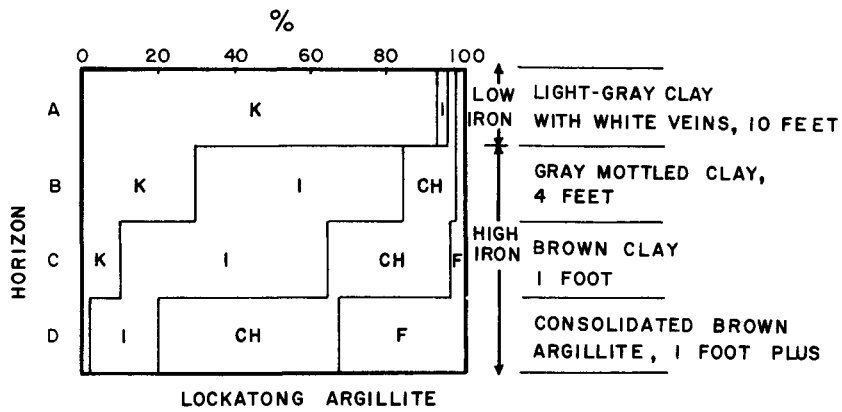


Fig. 2. Clay mineral suites derived from triassic argillite and shale.

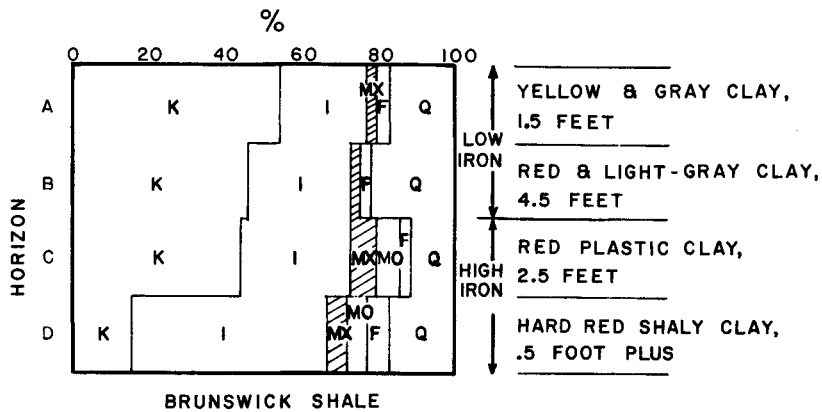


Fig. 3. Clay mineral suites derived from triassic argillite and shale.

Table 2. Chemical composition of the weathering suites derived from triassic shale and argillite

Per cent	Brunswick shale Sample 845				Lockatong argillite Sample 890			
	A	B	C	D	A	B	C	D
SiO ₂	53.0	55.0	46.6	46.0	42.0	34.2	32.0	41.2
Al ₂ O ₃	25.6	26.25	23.25	23.95	34.75	31.8	28.0	18.7
K ₂ O	4.10	4.05	3.75	3.60	0.20	4.15	4.30	3.66
Na ₂ O	0.90	0.95	1.3	1.25	0.85	1.25	1.40	5.50
CaO	Tr.	Tr.	0.5	1.1	Tr.	0.15	0.11	4.10
MgO	1.35	1.25	1.10	2.00	0.1	0.5	6.0	5.4
Fe ₂ O ₃	2.95	2.30	10.75	12.75	1.2	12.25	15.2	11.0
(total iron)								
TiO ₂	0.39	0.40	0.45	Tr.	0.12	0.35	0.37	0.34
H ₂ O ⁺	8.02	7.88	7.11	5.93	15.25	9.16	7.15	4.88
H ₂ O ⁻	0.04	0.31	0.48	1.37	1.64	1.48	0.85	0.43

diminishes only slowly upward; in the top horizon of this suite we still find 22 per cent illite. Feldspar is present in much smaller amounts than in argillite. The illite-feldspar ratio is roughly constant. The mixed-layer clays diminish between C and B, and montmorillonite was found only in D and C. The content of kaolinite increases from D to A.

In the Lockatong suite most of the feldspar found in D has been altered mainly to illite and some kaolinite in zone C. Chlorite is seen to decrease steadily upward, while kaolinite increases, until it constitutes 93 per cent of the A horizon.

JOINTING

Horizon C in the Lockatong is a brown clay which shows the argillite jointing and thin shiny colorless crusts on some breakage surfaces. These crusts were too thin to be separated from the clay, and attempts to identify them by optical microscopy and X-ray diffraction were unsuccessful. Similar crusts are described by Patterson (1964) who was able to identify them as aluminous silicate gels. The presence of such gels just above the feldspar-rich D horizon is not surprising.

Horizon B is a gray clay with whitish mottles. These mottles seem to represent the nuclei of the next alteration product leading to the white clay forming horizon A. Jointing planes can be seen throughout the residual clays above the Lockatong argillite; bedding planes can be barely discerned. Joint cracks are filled with white and greenish clay black blebs also present are melanterite and aggregates of very fine-grained iron sulphides. Mixed-layer clays and poorly crystalline aluminum silicates showing only a broad X-ray reflection between 6 and 14 Å are the clay minerals in these joint cracks. The joint cracks in the argillite proper, near the weathering boundary are bright-green when fresh, discoloring soon to yellow on exposure to air. They are composed of siderite, in addition to poorly crystalline aluminum-silicates (6–14 Å peak) and mixed-layer clays (41, 23 and 13 Å peaks).

The Brunswick horizon D is composed of shale particles in a matrix of residual deep red clay. Shale jointing is still visible. Both the Lockatong and the Brunswick residual clays are remarkably low in particles larger than clay size (less than 5 per cent coarser than 2 μ).

PREFERRED ORIENTATION OF THE CLAY PARTICLES

From our previous work (Sturm and Lodding, 1966) it was known that the clay minerals in the parent rocks were preferentially oriented. Preferred orientation can be dealt with quantitatively

in terms of vectors drawn from a common origin whose lengths are proportional to the number of particles in the direction of vectors (Chayes, 1949; Pincus, 1953, 1956). The ends of the vectors form a boundary surface, previously referred to as the orientation vector surface, or OVS (Sturm and Lodding, 1966), whose shape characterizes the degree and nature of the preferred orientation. For preferred orientation of particles where one cannot differentiate between the (001) and the (00 $\bar{1}$) planes, the function for the distribution of the vectors is periodic, with the period being 180°. In the general case, the vectors generate a hemispherical normal distribution.

To determine the nature of the distribution of the clay particles in the argillite, two cylindrical samples, or cores, were cut from each specimen. The cores were oriented so that their axes were perpendicular to each other and parallel to the bedding plane. With the aid of a special sample holder attached to a wide-angle goniometer, intensity data for the 10 Å lines for twenty angular positions, spaced over 180° of arc, were obtained for each specimen. Because of the cylindrical symmetry of the distribution, it is sufficient to obtain data for a central section only, that is, a section containing the axis of cylindrical symmetry; which, generally, is equivalent to the vector that is perpendicular to the bedding plane. The intensity data are then used in standard procedures to determine the particular semicircular normal distribution (Krumbein and Graybill, 1965; Pincus, 1953).

Our data show clearly that the orientation of the particles can be described in terms of the semicircular normal distribution (Sturm and Lodding, 1966); therefore, the characteristics of the particular distribution may be determined from X-ray diffraction data obtained from two surfaces of a sample block. Laboratory procedures have been established and the method has been verified on many samples. Both are the subject of a forthcoming paper.

Table 3 summarizes results of orientation studies on the argillite and shale obtained by the OVS method. X_1 , X_2 , X_3 are the relative X-ray intensities of the 7, 10 and 14 Å reflections, determined on three perpendicular planes. K -values, which are a measure of randomness or preferred orientation, were computed only for the strong reflections, such as the 10 Å line in the Lockatong argillite, and the 7 Å line in the residual Lockatong clay. The 10 Å reflection in the Lockatong argillite gives a k -value of 1.0 which indicates a moderate amount of preferred orientation, while the 7 Å line in the residual clay has a k -value of 0.4 which is almost random. The k -values found in the Brunswick shale and in the residual clay are both higher than in the

Table 3. Preferred orientation of clay minerals in Lockatong and Brunswick sediments

Lockatong argillite					Brunswick shale			
dA	X_1	X_2	X_3	k	X_1	X_2	X_3	k
Horizon D								
14A	5	4	—	—	59	24	29	1.5
10A	67	24	—	1.0	108	26	22	1.8
Horizon A								
14A	—	—	—	—	—	—	—	—
10A	—	—	—	—	199	155	100	0.75
7A	2266	1380	—	0.4	345	222	142	0.80

Intensities X_1 , X_2 , X_3 normalized for background.

— Intensities were not obtained.

k —measure of concentration in semi-circular distributions (equivalent to degree of preferred orientation).

Lockatong argillite. Although the precision of these measurements is not high, due to the difficulty of preparing suitable samples from specimens of non-indurated clay, they agree well with other observations made on these samples and thus constitute useful information.

DISCUSSION

Apparently the weathering sequence of these two Triassic sediments proceeded along different lines and as a result, the end products differ. In the argillite the decomposition of the abundant soda feldspar at the consolidated-unconsolidated boundary released Na and Ca ions and some silica. Movement of fluids was likely upward and lateral. These solutions must have had a rather high pH, and they in turn altered the over-lying strata. The net effect was the removal of Mg-ions in the B horizon and finally of K-ions in the top layer. Here the decomposition of iron sulphides often found in the Lockatong may have caused a reduction of the pH and in turn the removal of iron oxides from clay. In the weathered zones overlying the Brunswick shale, the chemical changes are less extreme than those associated with the Lockatong argillite. This is probably due to the lower cation concentration in the ground water. Consequently, the removal (and exchange) of cations in the overlying zones of weathered material is less complete, and we still find 22 per cent illite in the top layer of the weathered zone derived from the Brunswick shale. The most apparent change observed here is the removal of iron oxides between horizons C and B. This is likely to be due to the decomposition of pyrite and the reduction in pH values of the ground water. Although pyrite is not ordinarily found in the Brunswick shale, it is abundant in

the overlying Raritan formation and often concentrated at the boundary.

One may assume that post-depositional conditions in these localities, which are only a few miles apart, were quite similar. The differences in the weathering sequence of these two suites can be attributed mainly to the different chemical composition of the parent sediments.*

Intensity data from the 10 Å material of the residual Lockatong were quantitatively insignificant and could not be computed with reasonable accuracy. The kaolinite particles were almost randomly oriented with k being approximately 0.4. It is possible that the kaolinite which formed from feldspar is randomly oriented while the fraction which formed at the sites formerly occupied by illite inherited its orientation to some degree. In the parent Lockatong argillite only the orientation of the 10 Å material was computed because the intensities of the 14 Å clay reflections in the undisturbed slab were too low. The fact that the 10 Å clay shows only a moderate degree of preferred orientation can be readily explained by the high salinity of the environment of deposition which tended to result in the deposition of the clays as floccules, and, perhaps, by the simultaneous deposition of other minerals with a non-platy morphology, which tended to inhibit the settling of mica platelets in one preferred plane.

Higher degree of preferred orientation in the Brunswick shale is accounted for by a much less saline environment (Van Houten, 1965*b*) which let the clay minerals deposit from a partly-dispersed suspension. The residual clay from the Brunswick Formation likewise shows less preferred orientation of the clay minerals than the parent shale, which may be indicative of the mode of clay mineral transformation that took place in this weathering sequence.

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*Although the Wisconsin terminal moraine runs between the Almasi and the Milltown pit its effect on the weathering of the Triassic sediments was small due to the protective cover of Raritan clays. This assumption was confirmed by work on split-spoon cores recently obtained from the residual clays from Nixon, N. J. (2081). Unlike the Almasi pit the Nixon locality is south of the terminal moraine, and less than 5 miles from the Milltown pit. Yet the weathering sequence and the clay minerals found in these cores are almost identical to those of the Almasi pit north of the terminal moraine.

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Résumé—La composition de suites de minéraux argileux dérivés d'argillite et de shale triasiques, est comparée à celle des sédiments consolidés d'origine. L'argillite Lockatong, près de la zone de dégradation, est composée principalement d'illite, de chlorure et de feldspath. Dans l'horizon argileux, immédiatement au-dessus, l'illite remplace le feldspath et le kaolin augmente avec la distance à partir du contact jusqu'à ce que, près du sommet de la zone de dégradation, il n'y ait presque plus que des minéraux argileux. Une étude similaire a été faite sur le shale de Brunswick. Ici la roche parenté consiste principalement en illite, et quelque kaolinite, montmorillonite, quartz et feldspath. Le kaolinite s'accroît graduellement au fur et à mesure que l'on progresse en montant vers la zone de dégradation, le montmorillonite décroît, mais on trouve encore environ 30 pour cent d'illite dans les couches supérieures. Le chlorure qui se trouve dans l'argillite près de la zone de dégradation, est orienté au hasard tandis que l'illite montre une orientation déterminée, indiquant probablement que le chlorure s'est formé sur place pendant la dégradation. Dans le shale de Brunswick, l'illite montre aussi une orientation fortement déterminée.

Ces deux sédiments triasiques sont à moins de cinq miles l'un de l'autre et on peut supposer qu'ils étaient dans un milieu similaire après le dépôt. La différence dans le développement diagénétique est évident et il faut l'attribuer principalement à la composition chimique et à la lithologie du sédiment mère.

L'orientation des particules d'argile dans la roche solide a été étudiée avec l'aide d'une méthode de diffraction des rayons X, basée sur l'analyse du prélèvements cylindriques. Les données tridimensionnelles d'intensité ainsi obtenues ont été représentées par des vecteurs dont les extrémités forment une surface caractéristique de l'orientation des particules. Généralement, les vecteurs engendrent des distributions normales hémisphériques dont le vecteur principal coïncide avec la perpendiculaire au plan du lit.

Kurzreferat—Die Zusammensetzung von Tonmineralfolgen aus triassischen Argilliten und Schiefnern wurde mit derjenigen der verdichteten Muttersedimente verglichen. Lockatong Argillit in der Nähe der verwitterten Zone setzt sich in der Hauptsache aus Illit, Chlorit und Feldspat zusammen. Im

unmittelbar darüber liegenden Tonhorizont wird der Feldspat durch Illit ersetzt, und der Kaolingehalt wird grösser je weiter man sich von der Berührungsstelle entfernt und stellt in der Nähe des Scheitelpunktes der verwitterten Zone beinahe das einzige Tonmineral dar. Eine ähnliche Untersuchung wurde an Brunswick Schiefer durchgeführt. Hier besteht das Muttergestein grösstenteils aus Illit mit etwas Kaolinit, Montmorillonit, Quarz und Feldspat. Der Kaolinit erhöht sich in dem Masse als man sich durch die Verwitterungszone aufwärts bewegt, der Montmorillonit nimmt ab, doch wird in den obersten Schichten immer noch 30 Prozent Illit angetroffen. Chlorit im Argillit in der Nähe der verwitterten Zone ist unregelmässig orientiert, während Illit deutlich bevorzugte Orientierung aufweist, was vermutlich darauf hindeutet, dass der Chlorit während der Verwitterung an Ort und Stelle gebildet wurde. Illit in Brunswick Schiefer zeigt sehr deutlich eine bevorzugte Orientierung.

Diese beiden triassischen Sedimente befinden sich weniger als fünf Meilen voneinander entfernt, und es kann angenommen werden, dass sie nach der Ablagerung in ähnlicher Umgebung existierten. Die unterschiedliche diagenetische Entwicklung ist bemerkenswert und muss in erster Linie der chemischen Zusammensetzung und Lithologie des Muttersediments zugeschrieben werden.

Die Orientierung der Tonteilchen im Festgestein wurde mit Hilfe von Röntgenbeugungsmethoden auf der Grundlage der Analyse zylindrischer Proben untersucht. Die so erhaltenen dreidimensionalen Intensitätsdaten wurden durch Vektoren dargestellt, deren Enden eine für die Orientierung der Teilchen charakteristische Oberfläche formen. Im allgemeinen bilden die Vektoren hemisphärische, normale Verteilungen, deren Hauptvektor mit der Senkrechten auf die Schichtfläche übereinstimmt.

Резюме—Состав свит глинистых минералов, полученных из триасового аргиллита и сланца сравнивался со свитами получаемыми из уплотненных осадочных исходных пород. Локатонг аргиллит, поблизости зоны выветривания, состоит преимущественно из иллита, хлорита и полевого шпата. В находящемся сразу над ним глиноносном горизонте иллит замещает полевой шпат, а количество каолинита возрастает по мере увеличения расстояния от контакта до тех пор, пока при самом верхе зоны выветривания присутствует почти только глинистый минерал. Сходное исследование проводилось на брунswickском сланце. В этом случае исходная порода состоит главным образом из иллита и содержит также некоторое количество каолинита, монтмориллонита, кварца и полевого шпата. Каолинит постепенно увеличивается по мере продвижения вверх через зону выветривания, а монтмориллонит уменьшается, но прибл. 30% иллита все еще находят в верхних слоях. Хлорит в аргиллите близ зоны выветривания беспорядочно ориентирован, в то время как иллит показывает сильную предпочитаемую ориентировку, вероятно указывая, что хлорит образовался на месте в ходе эрозии. Иллит в брунswickском сланце показывает сильную предпочитаемую ориентировку.

Эти два триасовые отложения на расстояние менее 8 км (5 миль) друг от друга и можно предположить, что они оба имели сходную окружающую среду после отложений. Разница в диагенетическом развитии очень заметная и следует приписать ее преимущественно химическому содержанию и литологии исходного отложения.

Ориентировка частиц глины в коренной породе исследовалась, применяя методы дифракции рентгеновских лучей, основанной на анализе цилиндрических образцов. Полученные таким образом данные трехмерной интенсивности изображаются векторами, концы которых образуют поверхность, что и характерно для ориентировки частиц. В общем, векторы генерируют полусферические нормальные распределения, главный вектор которых совпадает с нормалью к плоскости напластования.