SEDIMENTOLOGY, PALEOCLIMATOLOGY, AND DIAGENESIS OF POST-HERCYNIAN CONTINENTAL DEPOSITS IN THE SOUTH-CENTRAL PYRENEES, SPAIN

BY

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ABSTRACT

The first chapter of the post-Hercynian geologic history of the South-Central Pyrenees is recorded in a sequence of fluvial and volcanic deposits which reach a total of added maximum thicknesses of more than 2300 m and date from the Westphalian D up to and including the Lower Triassic. The present study concerns the primary lithology, paleocurrents, depositional environment, paleoclimatology, and diagenesis of these deposits.
Five formations are distinguished:

(5) Bunter Formation	(Lower Triassic)
(4) Peranera Formation	(Permian)
(3) Malpas Formation	(Stephanian)
(2) Erill Castell Volcanics	(Stephanian?)
(1) Aguiró Formation	(Westphalian D)

The deposits of the Aguiró Formation are the fills of fossil valleys forming part of a pronounced paleorelief. The formation consists largely of conglomerates deposited by braided streams. One level, near Aguiró, contains coal-bearing, tuffaceous mudstone and tuff beds, the environment of deposition corresponding to that of a back-swamp. Most clastic components, many of wich are non-calcareous phyllite grains, can be recognized as stemming from the Paleozoic basement (Axial Zone of the Pyrenees, lying to the North of Aguiró).
The Erill Castell Volcanics, consisting of silicified and kaolinized andesitic tuffs and one intercalated basaltic andesite sheet, were deposited on a hilly land surface, since they locally overly fossil slope breccia and contain some fluvial channel-fills. The tuffs show evidence of penecontemporaneous pedogenesis.
The Malpas Formation contains coal-bearing mudstones and shales, bedded chert, limestones, and ferroan dolomite beds (back-swamp and lacustrine environments), sheets of upward-fining sandstones (the fills of mainly meandering streams), and one thick level of conglomerates (braided stream environment). The distribution of these deposits on the geological map still indicates the filling-in of a fossil valley. Nearly all the clastic components in the Malpas Formation stem from the directly underlying Erill Castell Volcanics, which had already been altered by weathering.
The Peranera Formation consists of caliche-bearing red beds. Depositional features point to the lowlands bordering alluvial fans. Many of the clastic components are similar to those in the Aguiró Formation, stemming from the Paleozoic basement.
The Bunter Formation, which is also composed of red beds, unconformably overlies all other deposits; its lower surface records a period of pediplanation. The Bunter sandstones consist mainly of quartz; they are the most mature in the entire sequence. The Peranera depositional environment was comparable to that of the present-day steppes, the Bunter environment to that of savannahs.
All fluvial deposits show paleocurrent directions indicating flow from either the West, the North, or the East.
The paleoclimate was of paramount importance for the primary lithology, depositional characteristics, and diagenesis of the deposits. Humid conditions would account for the deduced continuous flow during the deposition of the Malpas Formation and flooded back-swamps of the Aguiró and Malpas formations. In these formations, diagenetic parageneses of iron sulfides, kaolinite, and siderite, and the preservation of abundant vegetational debris, point to a weakly to strongly reducing and a neutral to acid milieu. A semi-arid climate is consistent with the intermittent flow deduced from channel-fill properties in the Peranera Formation. Flow in the Bunter channels was more continuous, but the channels dried up in the dry seasons. The instability of the vegetational debris in both formations (root traces are present but there is hardly any coalified material) points to lowered water-tables and the oxidizing conditions of sub-aerial diagenesis.
In spite of the similarity represented by a greyish-red stain, the Peranera and Bunter red beds differ strongly in primary lithology and diagenesis. The clastic composition of the Peranera Formation is to a great extent unstable; in the Bunter Formation it is highly stable; ferric oxides in the Peranera sandstones were largely brought in clastically; in the Bunter sandstones they are authigenic. Diagenesis in the Peranera deposits is mainly that of precipitation of and replacement by calcite (caliche); in the Bunter deposits in situ kaolinization of muscovite, possibly early diagenetic quartz (silcrete?), and authigenesis of dolomite can be observed. These differences are probably related in part to the morphologic setting (Peranera Formation: still some relief; Bunter Formation: deposits directly overlying wide pediments), but chiefly to the paleoclimate (Peranera Formation: steppe climate; Bunter Formation: savannah climate).
The composition of recent fluvial sands deriving from the Paleozoic core of the Pyrenees is treated in an Appendix. These sands are to a large extent comparable to the sandstones in the Aguiró and Peranera Formations.

CONTENTS

INTRODUCTION

AIM AND SCOPE OF THE STUDY

At the end of the Paleozoic, a mountain chain existed in the area now occupied by the Tertiary Pyrenees. These precursors of the Pyrenees were the product of the Hercynian orogenic period which had largely come to a close in the Upper Carboniferous. The present study concerns a sequence of strictly continental deposits that collected, geographically speaking, on the southern flanks and foot-plains of the former, or Hercynian, Pyrenees.

The sequence consists mainly of fluvially accumulated conglomerates, sandstones, and mudstones, intercalated with pyroclastics. These deposits, which reach a total of added maximum thicknesses amounting to more than 2300 m, range in age from the Westphalian D up to and including the Lower Triassic.

The complete story of a degrading mountain chain can be read from the sequence: the oldest deposits are highly immature and were formed under conditions of a persisting pronounced relief; the youngest fluvial deposits are mineralogically mature and were formed on extensive pediments. Conclusive evidence that by the time these youngest deposits had accumulated the

Hercynian Pyrenees had largely ceased to exist, is offered by the fact that they are overlain by transgressive shallow marine low energy deposits (evaporites, marls, and limestones) which show an almost constant facies in the entire region.

The area in which the post-Hercynian continental deposits were studied is shown in Fig. 1.

The sequence is subdivided by delineating lithostratigraphical units or formations, in the sense of the report of the International Subcommission on Stratigraphy and Terminology (1961) and the Code of Stratigraphic Nomenclature (American Commission on Stratigraphic Nomenclature, 1961)[1].

Fig. 2 shows the sequence schematically as well as the interrelationships of and the names given to the various formations.

The aim of the study was to analyse and compare the defined formations with respect to primary lithology, paleocurrents, depositional environment, paleoclimatology, and diagenesis.
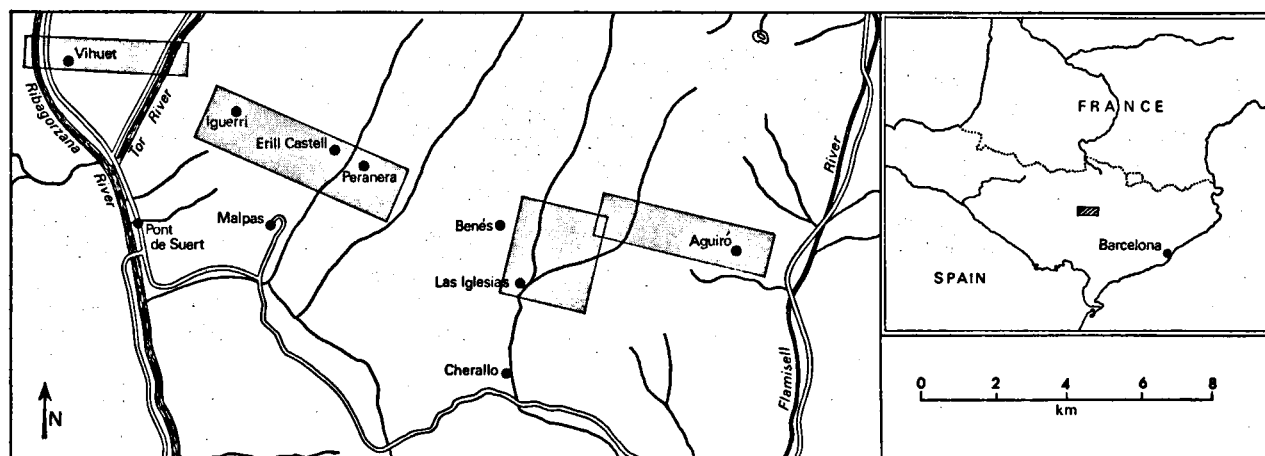
Fig. 1. Topographic map of the study area. The shaded rectangles indicate the areas for which geological maps are given in Figs. 3—6.
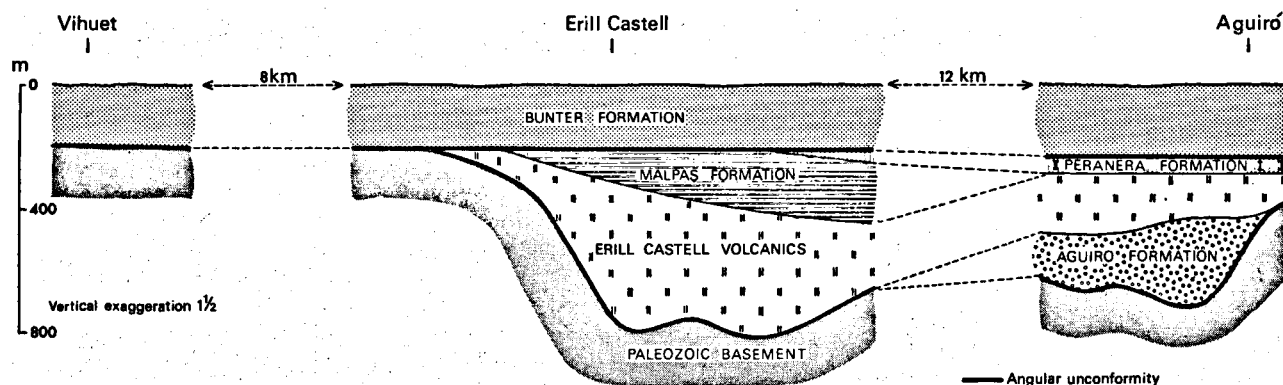
Fig. 2. Schematic E—W cross-section through the study area, showing the sequence of formations at Aguiró, Erill Castell, and Vihuet.

GEOLOGICAL SETTING

The sequence under study occurs in a monoclinal structure dipping 40° to 90° (locally overturned) SSW. The structure as a whole is slightly curved, its convex side facing southwards. The rocks reached this position during the Pyrenean phase of the Tertiary orogenic period (Misch, 1934).

In the so-called Axial Zone of the Pyrenees, which is located to the North of the study area, Paleozoic rocks occur that were strongly affected by the Hercynian and to a lesser degree by the Tertiary tectogenesis (Schmidt, 1931; de Sitter, 1964; Zwart & Mey, 1965, Mattauer & Séguret, 1966; Mey, 1967) (Fig. 70).

The investigated sequence forms the northern part of a narrow structural zone referred to as the Nogueras Zone. This zone also contains Triassic limestones, fine-grained dolomites, and evaporites, intrusions of green dolerites, and problematic up-thrusted or gliding structures consisting largely of Paleozoic rocks (Jacob et al., 1926; Misch, 1934; Nagtegaal, 1962; Séguret, 1964, 1966; Rijnsburger, 1967).

[1] A preliminary note on the newly proposed stratigraphy for the South-Central Pyrenees has recently been published (Mey et al., 1968).

METHODS

Careful analyses of the deposits in the field based on drawings of detailed sections, data on lithology and sedimentary structures, and measurements of current structures, made it possible to determine the various depositional environments and paleocurrent directions. Thin-section studies done by standard optical procedures and supplemented by X-ray analyses and staining were used to investigate primary lithology and diagenetic changes.

CHAPTER I

THE FORMATIONS: NOMENCLATURE AND AGES

AGUIRÓ FORMATION

Two separate, though not widely separated, occurrences of the Aguiró Formation are found in the Aguiró area (Fig. 3). The formation consists mainly of conglomerates, the remainder comprising fossil slope breccia, sandstones, tuffaceous, coal-bearing dark grey [1] mudstones and shales, and tuff beds.
A detailed stratigraphic section of the formation, taken directly to the West of Aguiró, is given as section 1 (sheet I). The occurrence to the West of Aguiró is the largest; in an East-West direction it measures 3 km and is maximally about 400 m thick. It is also well differentiated into the rock types mentioned, the occurrence to the East of Aguiró being much smaller and consisting mainly of fossil slope breccia irregularly interstratified with pebble-bearing horizons.
The top of the formation was drawn at the top of the upper level of conglomerates, in contact with the Erill Castell Volcanics. The base of the formation was drawn at the base of the fossil slope breccia or at the base of the lowermost level of conglomerate: the Hercynian unconformity (section 1, sheet I).
The age of the Aguiró Formation has traditionally been determined on the basis of the fossil flora. Fossils other than those of plants and trees are not known.

Roussel (1904) and Dalloni (1913, 1930) concluded that the sequence at Aguiró is, at least partially, of Westphalian age.
Because the Aguiró Formation contains the oldest deposits known to overlie the Hercynian unconformity, interest in its flora has not decreased. On the basis of a collection made by Stevens (1956), Stevens and A. C. Hopping concluded a Westphalian D age, in agreement with the earlier workers. Professor F. Stockmans (Institut Royal des Sciences Naturelles de Belgique, Brussels) was so kind as to examine collections made by Vink (1958) and Nagtegaal (1962). He confirmed the Westphalian D age, and listed the following imprints:

Dicksonites pluckeneti (Schlotheim)
Pecopteris unita Brongniart
Pecopteris lepidorachis Brongniart
Linopteris brongniarti (Gutbier)
Sphenophyllum sp.
Cordaites sp.
Dicksonites sterzeli (Zeiller)
Alethopteris grandini Brongniart
Mixoneura ovata (Neuropteris)
Sphenopteris sp.
Pecopteris sp.
Cardiocarpus sp.
Hexagonocarpus sp.

[1] The colours are named according to the Rock Color Chart distributed by the Geol. Soc. of America, New York, 1963.
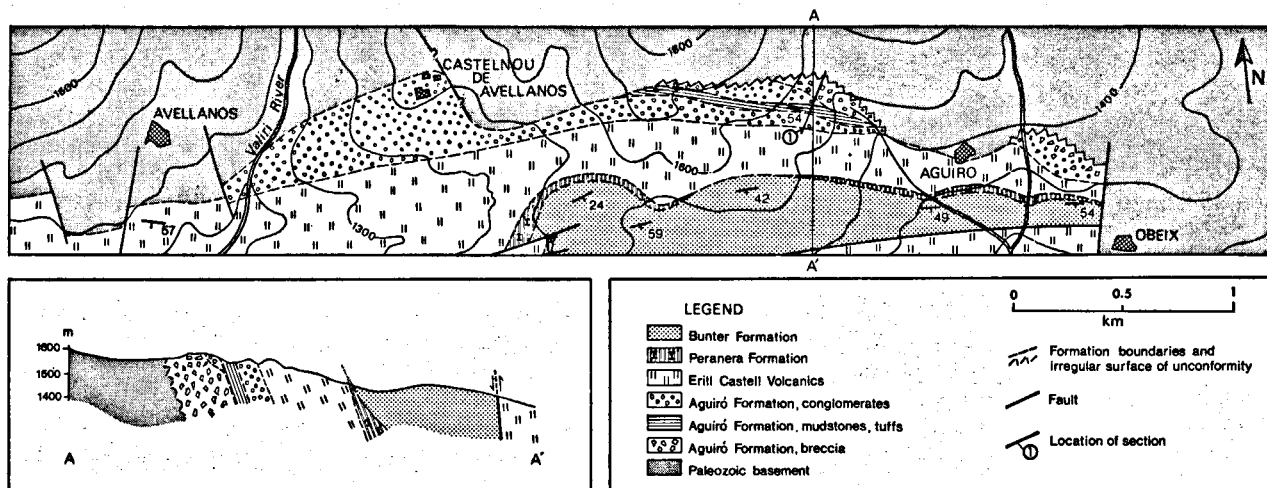
Fig. 3. Geological map of the Aguiró area. With modifications by K. Haagsma. For location, see Fig 1.
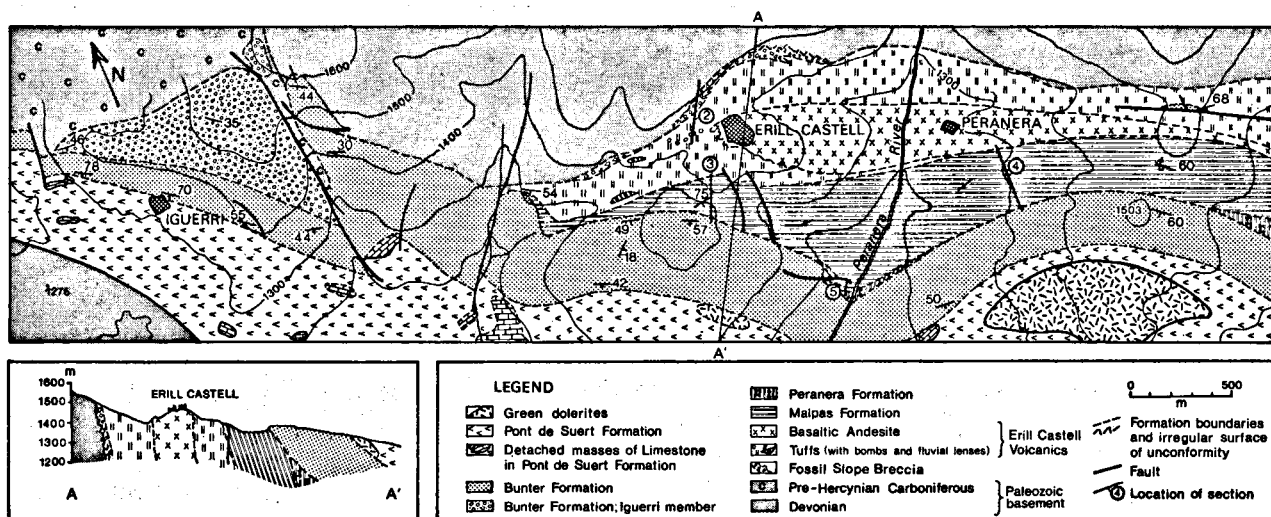
Fig. 4. Geological map of the Erill Castell area. For location, see Fig. 1.

ERILL CASTELL VOLCANICS

The Erill Castell Volcanics, which in the Erill Castell area reach a maximum thickness of about 550 m, are composed of light- and dark-coloured tuffs, tuff breccia, and a sheet of dark-coloured basaltic andesite (Fig. 4). In the Erill Castell area the lower boundary of the formation is drawn at the base of a wedge of fossil slope breccia. Although these slope breccia directly overlie the Hercynian unconformity and could be of Westphalian D age (see above, Aguiró Formation), for mapping purposes they are included in the Erill Castell Volcanics. The upper boundary of the formation is difficult to draw, because the transition into the fluvial deposits of the Malpas Formation is gradual. It is arbitrarily set at a level of a pedalfer paleosol (p. 190) (section 2, sheet II).

In the Aguiró area (Fig. 3) the Erill Castell Volcanics overlie the Aguiró Formation (which in the Erill Castell area was never deposited or had previously been eroded) and are overlain by the Peranera Formation

(the Malpas Formation being absent). In the Las Iglesias area (Fig. 5) the Erill Castell Volcanics occur again directly above the Hercynian unconformity; they are overlain by a wedge of the eastern termination of the Malpas Formation and by the Peranera Formation. A detailed stratigraphic section of an exposed part of the sequence at Erill Castell is given on sheet II.

The volcanics once occupied a wide area; in our region they can be followed from Erill Castell to Aguiró, over a distance of more than 12 km. Their initial extension must have been much greater: they are cut away by the unconformity underlying the Bunter Formation (Figs. 2, 3, and 4). The formation reappears to the West of the Ribagorzana River, and, towards the East, near Seo de Urgel (Mey *et al.*, 1968). Previous workers have included the Erill Castell Volcanics in the Stephanian (Dalloni, 1930; Misch, 1934). The formation was named after the picturesque hamlet lying on a steep hill representing the western termination of a basaltic andesite sheet (Fig. 4).

MALPAS FORMATION

The Malpas Formation is composed mainly of sandstones and coal-bearing shales and mudstones. Over its full exposed extent, the formation has as its lower boundary a level of a fossil pedalfer in contact with the Erill Castell Volcanics. To the SW of Erill Castell the formation overlies tuff and tuff breccia, but at the hamlet of Peranera the lower contact is with basaltic andesite (Fig. 4). Detailed stratigraphic sections are given on sheet III.

The contact of the Erill Castell Volcanics and the Malpas Formation is gradual and conformable. However, the deeply weathered horizon at the base of the Malpas formation suggests a slowing down of deposition or even a hiatus. Moreover, the clastic material in the Malpas sandstones was derived almost exclusively from the Erill Castell Volcanics, thus leaving room for at least a disconformity towards the source area, which lay to the North (p. 158).

The upper boundary of the Malpas Formation is drawn where the sediment abruptly changes colour, within a conformable sequence, from dark grey to the predominantly greyish-red of the Peranera Formation. To the West of Erill Castell, the Malpas Formation is cut away by the unconformity at the base of the Bunter (Fig. 4). In an East-West direction, the Malpas Formation can be followed over a distance of 8 km; its maximum thickness is about 500 m.

The formation has been named after the village that houses many of the coal miners working in the *Minas de Malpas* (M.I.P.S.A. Company, Spain).

Previous workers have referred to the Malpas Formation as the Stephanian (Dalloni, 1930; Misch, 1934; Virgili, 1960). Prof. F. Stockmans (p. 146) found the following species of plant imprints in a collection taken 132 m above the base of section 3 (sheet III):

> *Sphenophyllum angustifolium* (Germar)
> *Pecopteris feminaeformis* (Schlotheim)
> *Acitheca polymorpha* (Brongniart) nov. var.
> *Alethopteris zeilleri* Wagner

The level at 137 m above the base of section 4 (sheet III), yielded the following assemblage:

> *Annularia sphenophyoides* (Zenker)
> *Asterophyllites equisetiformis* (Schlotheim)
> *Pecopteris (Asterotheca) lepidorachis* Brongniart
> *Pecopteris villablinensis Wagner (= pseudobucklandi* Zeiller non Andrae)
> *Pecopteris integra* Andrae
> *Pecopteris polymorpha* Brongniart nov. var.
> *Pseudomariopteris* cf. *rbeyroni* (Zeiller)
> *Alethopteris grandini* (Brongniart)

These determinations and an inferred Stephanian age are in agreement with the findings of Dalloni (1930). The Malpas Formation is probably not entirely of Stephanian age: the transition to the Lower Permian (Autunian) could well lie somewhere in its upper part. A Permian age has not yet been recorded from the Malpas Formation in the area shown in Fig. 4, but it has been found in deposits which lithologically should be considered as the very top of the Malpas Formation, at Baro and Gerri (Pallaresa valley, Mey *et al.*, 1968) by Dalloni (1930, 1938).

PERANERA FORMATION

The Peranera Formation, which is composed of greyish-red mudstones and conglomeratic sandstones, conformably overlies the Malpas Formation (Virgili, 1960). Where the latter has wedged out, it overlies the Erill Castell Volcanics (Figs. 2, 4, and 5). A detailed stratigraphic section of the Peranera Formation at the locality where it reaches its maximum thickness (approximately 700 m), i.e. in the Las Iglesias area, is given on sheet IV.
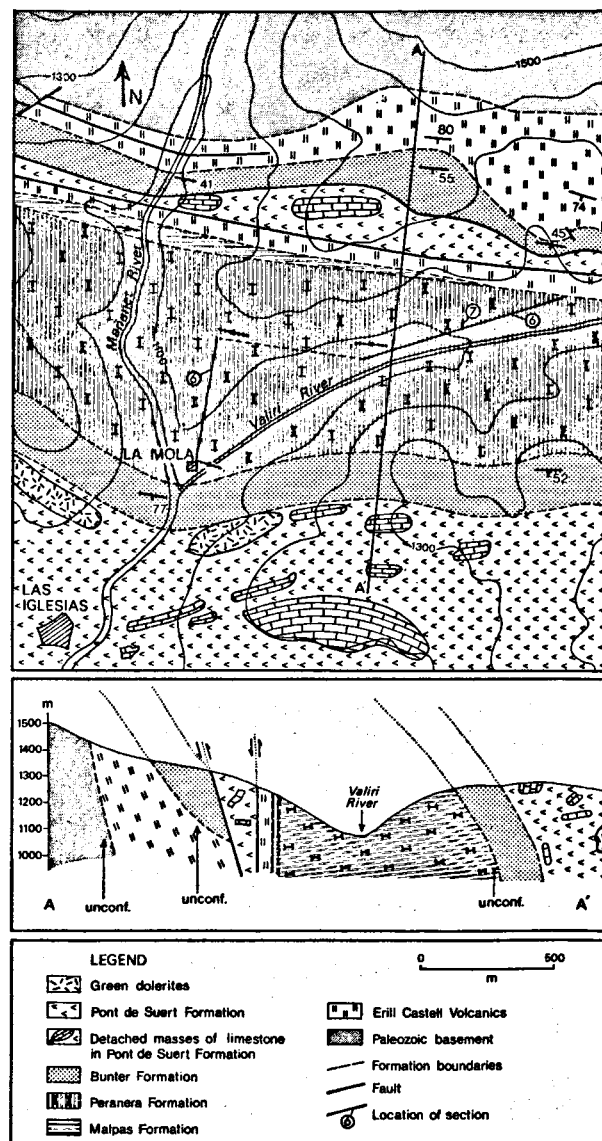

Fig. 5. Geological map of the Las Iglesias area. For location, see Fig. 1.

The Peranera Formation is widespread, occurring nestled against the flanks of the Hercynian core of the Pyrenees at many localities. The Formation is lithologically similar to the 'Série du Somport' and the 'Permian' in the Western Pyrenees (Mirouse, 1959; Lamare, 1931, 1937), to the units P 1 and P 2 in the Canfranc area (Van der Lingen, 1960), and to the 'Permian' at the eastern extremity of the Pyrenees (Guitard & Ricour, 1959). Its thickness reaches 1000 m South of Seo de Urgel (Hartevelt, 1969).

Everywhere in the area under study the Peranera Formation is truncated by the unconformity at the base of the Bunter Formation. Variations in thickness seem to be largely governed by the level at which the unconformity at the top is located. In our area the formation can be followed over about 13 km.

The formation was named after the Peranera River, in whose valley the relations with the conformably underlying Malpas Formation and the unconformably overlying Bunter Formation are clearly displayed (Fig. 4).

The Peranera Formation is considered to be of Permian age (Virgili, 1960) and probably largely was deposited in Saxonian times (Roger, 1965). The only plant imprints found by the present author belong to *Walchia piniformis (= Lebachia piniformis)*, according to determinations by Prof. F. Stockmans (p. 146).

BUNTER FORMATION

Throughout the area the Bunter Formation, which is composed, like the Peranera Formation, of mainly greyish-red mudstones and sandstones, has an angular unconformity as a lower boundary (Figs. 2 to 6). Going from East to West, the basal unconformity

LEGEND

[symbol] Green dolerites

[symbol] Pont de Suert Formation

[symbol] Detached masses of limestone in Pont de Suert Formation

[symbol] Bunter Formation

[symbol] Pre-Hercynian Carboniferous

—·— Formation boundaries

[symbol] Location of section

— Fault

0   0.5   1
km
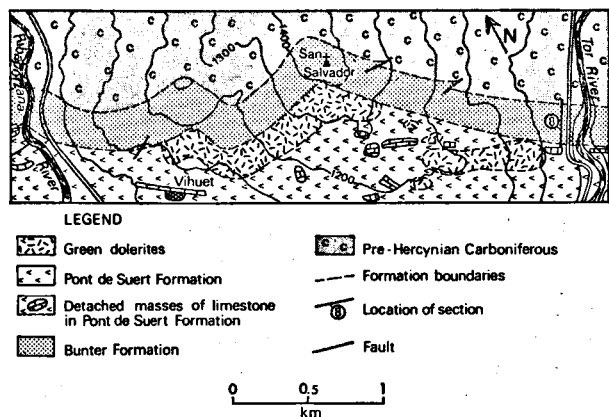
Fig. 6. Geological map of the Vihuet area. For location, see Fig. 1.

surface cuts progressively into lower stratigraphic levels until it coincides with, or rather cuts into, the Hercynian unconformity to the West of Erill Castell.

The Bunter Formation, which in our area has an average thickness of 200 m (ranging between 50 and 300 m), is very widespread, extending from the Mediterranean to the Atlantic coast.

Some authors, although aware of the presence of the unconformity surface, have treated the Peranera and Bunter formations jointly as the Permotriassic (Misch, 1934; Ashauer, 1934).

It should be emphasized that in the present study, 'Bunter' is used as the name of a rock unit, a formation. It has been generally assumed, however, that the deposits were formed in Lower Triassic times (summarized by Roger, 1965).

The extensiveness just referred to applies to the Bunter Formation as a whole. A local lower conglomeratic unit, the Iguerri member, has a restricted spread however, measuring 1.4 km in an East-West direction and approximately 500 m in thickness (Fig. 4).

There has been some confusion with respect to the stratigraphic position of the Iguerri member. On the basis of the pronounced relief of the lower boundary surface (the Hercynian unconformity) and the — albeit limited — content of unstable components such as phyllite and Devonian limestone, the present author has defended the view that the Iguerri member is just another occurrence of the Aguiró Formation. Judging from his schematic section, Dalloni (1930, p. 324) assigned the sequence at Iguerri an upper Eocene age. Misch (1934) saw the conglomerates at Iguerri as part of the Bunter Formation.

As can be concluded from a careful investigation in the field, the undisturbed position of the conglomerates at Iguerri below the overlying part of the Bunter definitely shows that the conglomerates are of Lower Triassic or older age.

Although the Iguerri member differs in many aspects from the overlying part of the Bunter, petrographic evidence now strongly suggests that Misch (1934) was justified in including the Iguerri member in the Bunter (p. 215).

A detailed stratigraphic section of the Bunter Formation in the Rio Tor valley (Fig. 6) is given on sheet V.

On top of the Bunter Formation are Triassic dark shales and limestones, fine-grained dolomites, marls, and evaporites, but these rarely, if ever, show an undisturbed position.

DEPOSITIONAL ENVIRONMENTS

AGUIRÓ FORMATION

Paleotopography

The Aguiró Formation occurs, as previously mentioned, in the form of two lenses (Fig. 3). In an East-West direction these lenses wedge out rapidly; their thicknesses vary with the undulations of the lower boundary surface.

This lower surface, the Hercynian unconformity, is of particular interest. At some localities it can be observed to be highly irregular; even over distances of a few metres there are irregularities corresponding to sharp, fossil crags with elevations of more than 5 m. At the eastern termination of the larger occurrence, directly to the West of Aguiró, it is often very difficult to pinpoint the unconformity surface because of its irregularity and because it is overlain by breccia, the fragments of which have the same composition as the Devonian phyllites below it (section 1; sheet I).

The breccias are fully angular, badly sorted, and devoid of stratification. The association of large undulations, a local craggy small-scale relief, and the overlying breccia, is interpreted as corresponding to an ancient land surface with scree breccia at the bases and on the flanks of valley walls. By turning the geological map in Fig. 3 upside down, one obtains a section of a fossilized valley system filled with mainly coarse clastic deposits.

The locations of the fossil valleys could be related to faults in the underlying Paleozoic basement, into which they were eroded. These faults, where they continue into the beds overlying the Aguiró Formation, must have been newly formed or have been reactivated during later tectogenesis (Fig. 3).

An alternative explanation, namely that the breccia is a fault breccia and that the inclined lower boundary surfaces are faults rather than old land surfaces, must be rejected, since it does not account for the considerable local thickness of the breccia and the absence of parts of the Aguiró Formation that should have been set off by the faults.

In the Aguiró area three fossil valleys can be distinguished: one to the East of the village, another to the West of it, and the deepest one at Castelnou de Avellanos (Fig. 3). Because these valleys occur so close to each other and seem to be separated by only minor divides, they probably belonged to one fluvial system. The biggest one, at Castelnou de Avellanos, will therefore be referred to as the trunk valley, and the other two will be called the first lateral valley (W of Aguiró) and the second lateral valley (E of Aguiró).

Modes of deposition

The conglomerates of the Aguiró Formation show a very coarse trough type of cross-stratification in some places. Internal scour structures are common. Large, flat pebbles and boulders usually lie in an imbricate position. Although it was not systematically measured, the orientation of imbrication is suggestive of flow from northern directions.

The thick fill of the trunk valley contains the coarsest conglomerates, many boulders reaching a diameter of 40 cm. Towards the East in the same conglomerate unit, the size of the boulders decreases regularly and cross-stratification in the intercalated sandstones becomes more frequent. The lower conglomerate unit in the first lateral valley, which cuts sharply into the thick lens of fossil slope breccia (section 1, sheet I), has a coarse facies similar to that at Castelnou de Avellanos.

On the basis of their general coarseness, internal scour structures, and an absence of intercalated shale or mudstone levels of appreciable thickness, the conglomerates are interpreted as having been deposited in braided streams.

The coal-bearing shales, the mudstones and siderite levels in the interval of 76 to 97 m in section 1 die out towards West; they do not occur in the fully conglomeratic valley-fill at Castelnou de Avellanos (Fig. 3). Since these fine-grained sediments rich in plant debris are laterally transitional, over a short distance, to braided stream deposits, they probably represent deposits formed in back-swamps.

The occurrence of tuff beds intercalated between the fine-grained sediments agrees with this view. Such beds could have been preserved in quiet back-swamps; tuff was dispersed to a varying extent, however, in the active current areas in the fluvial environment (p. 178).

Sequence of events

The story that can be read from the relations in the field and from the detailed section 1 is as follows: after the Hercynian uplift, erosion developed a pronounced relief which at Castelnou de Avellanos reached a relief energy of at least 340 m. Colluvial breccias were formed as scree on the flanks and at the bases of the valley walls. After this, deposition of conglomerates started in all three valleys. The accumulation of conglomerates continued in the trunk valley, but came to a stop in the first lateral valley. Here, marshy and lacustrine back-swamp conditions developed, possibly due to the large masses of conglomerates deposited in the trunk valley, which may have dammed off the first lateral valley. The possibility that deposition alone can form swamps in an environment such as the one as visualized here, has also been mentioned by Twenhofel (1950, p. 71).

As the observations made at 83 m above the base of section 1 suggest, masses of breccia having the same composition as the basal slope breccia but containing vegetational matter as well, were deposited in a slump-like fashion.

Finally, when the trunk valley was largely filled up, the braided streams in it moved eastwards, eroded the lacustrine and marshy sediments slightly, and built a sheet of conglomerate.

In summarization of the foregoing, it may be said that the Aguiró Formation overlies a pronounced, scree-carrying relief, consists largely of conglomerates deposited in braided streams, and contains one lens of fine-grained sediments rich in vegetational debris that was deposited in a back-swamp environment. Taken together, these points are strongly suggestive of a proximal piedmont environment of deposition. As argued elsewhere (p. 171, p. 178), the climate during the accumulation of the Aguiró deposits was probably humid, and there may have been relatively little evaporation [1].

In the Pyrenees, the sequence of occurrences deduced here for the Aguiró Formation was repeated in the Eo-Oligocene. At that time, again directly after a major orogenic period, fluvial deposition once more buried a pronounced relief and small lacustrine and marshy basins also developed. The climate, however, was semi-arid (Birot, 1937; Nagtegaal, 1966; Rosell & Riba, 1966).

Roundness of pebbles

The roundness of the pebbles in the conglomerates of the various fossil valleys differs. In Fig. 7 the frequency

[1] In paleoclimatology it is generally not possible to apply in detail the classifications available for present-day climates. Nevertheless, W. Köppen's classification of climates (1931), which combines temperature with precipitation values and therefore implicitly takes evaporation into account and, moreover, considers sedimentologically and petrographically recognizable environments, forms a sound basis for paleo-climatological work on continental deposits. The types of paleoclimate recognized in the present study are essentially Köppen's A, B, and C climates; in the deposits there is no evidence of cold climates. The definitions of the various climates have been slightly modified to allow their application to fossil continental deposits. The following terms are used for the paleoclimates:

Humid climates. — either tropical rain climates (Köppen's Af or Am climates) or warm-temperate rain climates (Cf climates). The distinction between A and C climates in the fossil deposits is difficult. In the humid climates the precipitation : evaporation ratios are too high to let alluvial plains fall dry for long, vegetational debris can be preserved in great quantities, and reducing conditions prevail in the surface layers of the deposits.

Savannah climates. — tropical rain climates with dry seasons (Aw or As climates). Precipitation : evaporation ratios are not high enough to support a dense vegetation. Alluvial plains fall dry seasonally; oxidation and marked *in situ* weathering of the deposits.

Steppe or semi-arid climates. — Dry climates, seasonally very dry, no cold seasons (Bsh climates). Precipitation : evaporation ratios not high enough to support a dense vegetation. Alluvial plains may fall dry for prolonged periods of time; oxidation and moderate *in situ* weathering of the deposits, and accumulation of salts (mainly $CaCO_3$) in soil horizons.
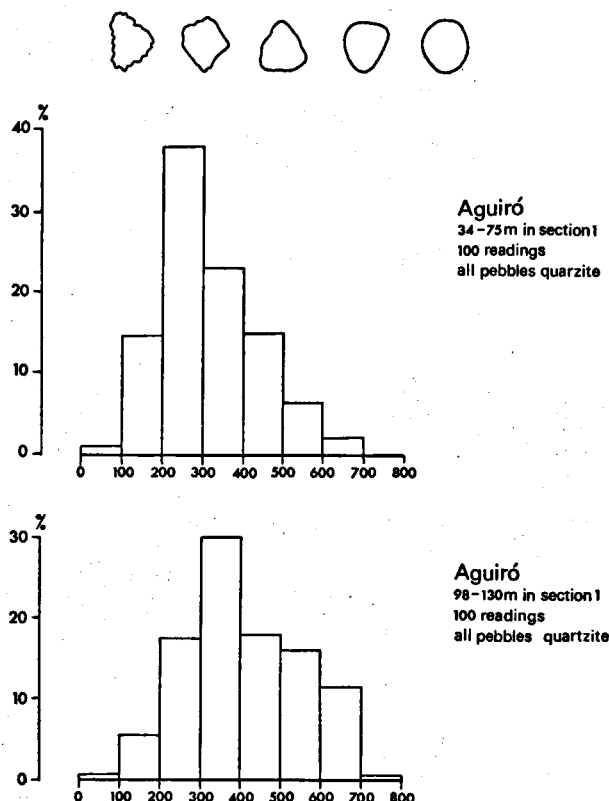
Fig. 7. Roundness of pebbles; Aguiró Formation.

distributions of the roundness indices ($\frac{2r}{L} \times 1000$) of two samples are given in histogram form. Comparison of the histograms (Aguiró 34—75 m and Aguiró 98—130 m above the base of section 1) shows that the quartzite pebbles in the upper conglomerate unit are more rounded than those from the lower unit. If it is assumed that the pebbles had a primary origin, those of the upper conglomerate unit, presumably deposited by streams stemming from the trunk valley at Castelnou de Avellanos, must have travelled over a greater distance than those of the lower conglomerate unit in the first lateral valley (Krumbein, 1941).

ERILL CASTELL VOLCANICS

Paleotopography

At Erill Castell, as at Aguiró, the morphology of the Hercynian unconformity (here in contact with the Erill Castell Volcanics) is believed to correspond to a fossil land surface with a marked relief. This surface, as at Aguiró, was later tectonically rotated into a vertical position. Turning Fig. 4 upside down again produces a section through a wide valley running 'below' Erill Castell and Peranera and, at Erill Castell, flanked by fossil slope breccia. Fluvial deposits of any appreciable thickness are not found on the valley floor, but rounded phyllite pebbles do occur between the fragments of the slope breccia (section 2, sheet II). The relief of the fossil valley at Erill Castell may, due

to a later tectonic movement, have become somewhat more pronounced than it was originally. After deposition of the Peranera Formation, the area to the West of Erill Castell was uplifted more than the area to the East of it, as indicated by the fact that the unconformity surface underlying the Bunter Formation progressively cuts through older strata in a western direction (Fig. 4; p. 149). This differential movement must have brought the Hercynian unconformity surface to the West of Erill Castell to a higher level, thus exaggerating the old topographic high directly to the West of the hamlet.

Sequence of events

The topographic low 'below' Erill Castell and Peranera was gradually buried under tuffs showing a wide textural variety. In section 2 (sheet II) a unit of 7.5 m of well-stratified tuffs is found on top of the fossil slope breccia. Similar units occur between 54

Fig. 8. Well-stratified tuff beds in the Erill Castell Volcanics; top toward the left. Photograph taken 36 m above the base of section 2 (sheet II).

and 70 m, and between 152 and 155 m above the base of the section (Fig. 8).

It is believed that the stratification of these units is a result of mainly slow, intermittant deposition and penecontemporaneous pedogenesis (p. 179). Since several tuff beds are lentiform and some have erosive bases, it is probable that colluvial processes such as short-distance reworking by means of rain-wash also played a part. Reworking of the tuffs, in the form of true channel-fills, is discussed below.

The fossil slope breccia at the base of the section reappears, in the form of a tongue, between 40 and 50 m. At this height the matrix of the breccia, unlike the basal breccia, contains fine-grained tuff.

Thick accumulations of non-reworked pyroclastics occur at 70.5 to 152 m and above 155 m above the base of the section.

The first of the thick units contains tuff breccia (angular, light-coloured fragments of tuff measuring up to 0.5 m, lying in a matrix of darker tuff), showing instances of well-developed grading, both fining and coarsening upwards (Fig. 9).

Grading of ejecta in volcanic deposits is a common feature. As Kuenen (1953) argued, upward-fining deposits may be formed due to a decrease in the size of the particles produced during an eruption or through selective settling, in air or water, of a mixture of particles. An instance of upward-coarsening graded beds of tuff breccia (most fragments smaller than 2 cm) is reported by Lauder (1962) from the Deborah Volcanic Formation, New Zealand. Lauder thinks that the upward-coarsening cycles are the result of gradual increases of the explosive force at the site of eruption. Selective settling must have played a part in the

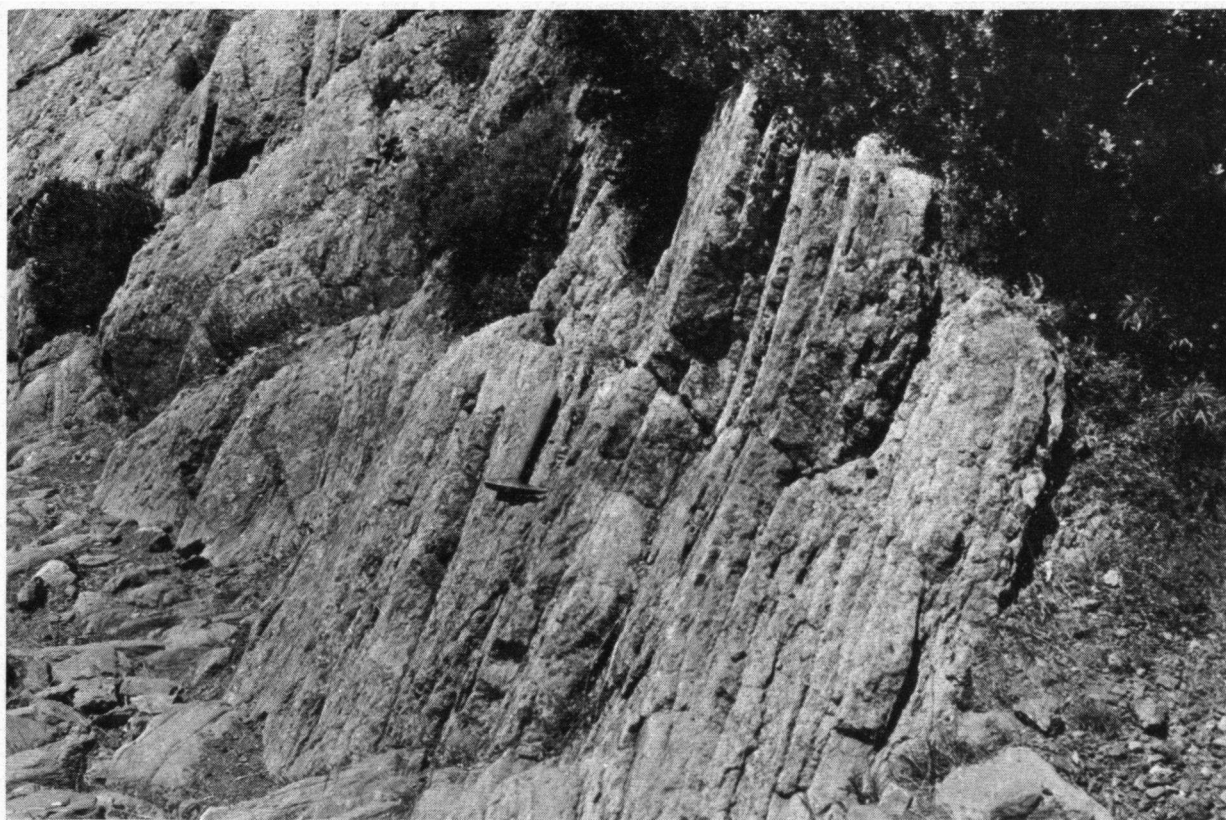Fig. 9. Upward-coarsening unit of tuff breccia in the Erill Castell Volcanics; top toward the left. Photograph taken 146 m above the base of section 2 (sheet II).

deposition of the Erill Castell tuff breccia. But it seems likely that, in accordance with Lauder's suggestion, variations in the explosive force of the volcanic source were the main cause of the observed gradations. The second thick non-reworked unit consists of a chaotic mixture of tuff breccia (similar to the first unit), large chunks of Devonian phyllite, and bombs of tuffaceous material measuring up to 1.5 m. After the accumulation of this deposit as well as of some largely unexposed tuffaceous deposits, a lava flow took its course through the presumably still only partially filled valley, thus forming the sheet of basaltic andesite on which the hamlets of Erill Castell and Peranera are now located (Fig. 4).

Reworking of the tuffs

The undisturbed character of the graded units and the sharp angularity of the fragments show that those tuff fragments are still *in situ* (Fig. 9). At several localities, however, small fluvial channel-fills (up to 5 m wide and 2 m deep) occur in the tuff breccia. These channel-fills consist of poorly sorted tuff sands. Good examples of such fluvial reworking of the Erill Castell pyroclastics

can be observed in an exposure 800 m (as the crow flies) to the SE of the hamlet of Castelnou de Avellanos (Fig. 3). Here, conglomeratic and coarse sandy cross-stratified channel-fills occur intercalated in the tuffs. Such channel-fills, which in addition carry abundant vegetational debris, can also be found 500 m SW of Erill Castell (Fig. 4).

MALPAS FORMATION

Composition

The Malpas Formation consists, in decreasing order of abundance, of: medium grey to black mudstones and shales, sheets of light grey, often conglomeratic sandstones, a 20 m thick continuous tuff bed near the base, thin continuous siderite beds and levels of siderite concretions, some ferroan dolomite and limestone beds, coals and chert. Vegetational debris abounds at all levels; coarse, conglomeratic sandstones contain fossilized logs up to 2 m long.

The Malpas Formation is composed entirely of freshwater, mainly fluvial, deposits, as will be demonstrated in the following pages.

Lateral and vertical variations in lithology

Three detailed stratigraphic sections of the formation are presented on sheet III (sections 3, 4, and 5). The locations of the sections are indicated in Fig. 4.

Comparison of section 3 (Erill Castell area) with section 4 (Peranera area) shows a number of differences which are expressed quantitatively as percentage values and mean thicknesses of lithologic units in Table I.

TABLE I

units	section 3		section 4	
	%	mean thickness	%	mean thickness
mudstone, shale	66.9	3.5	48.7	4.9
sandstone	18.8	2.2	31.3	4.2
tuff	9.7	–	7.1	–
conglomerate	0.0	–	11.7	–
siderite	3.3	0.2	1.1	0.1
limestone	1.0	–	0.0	–
coal	0.2	–	0.1	–
chert	0.1	–	0.0	–

Table I. Percentages and mean thicknesses (in m) of the Malpas lithologic units. Section 3, Erill Castell area and section 4, Peranera area.

The data of the two sections may only be compared with caution, because over their full height the sequences of sediment probably do not represent exact time equivalents: in the Erill Castell area the unconformity surface at the base of the Bunter Formation cuts somewhat deeper than in the Peranera area. It is clear, however, that the sandstone units are thicker in the Peranera area than in the Erill Castell area, and that the percentage of coarse deposits is considerably higher in the Peranera area. It may also be safely concluded that chemical precipitates such as siderite, limestone, and chert are

more abundant in, or are restricted to, the Malpas deposits at Erill Castell.

These differences between the Erill Castell and Peranera areas reflect environmental changes in a sense roughly perpendicular to the drainage direction of the flood-plain (see *Paleocurrents*, p. 158). In the field, a gradual lithologic transition between the two sections can be observed.

Compositional trends are present in the vertical sense as well. Fig. 10 shows the vertical variation of the percentages of the coarse units (sandstone and conglomerate) for the two areas under consideration. In both areas the percentages of the coarse units first increase and then decrease in the lower reaches of the formation; going upwards, two major trends are clearly expressed:

(1) the percentages of the coarse units increase markedly,
(2) concomitantly, grain-size increases, which is particularly well-expressed in the Peranera area.

These trends are reflected in the channel-fill characteristics, as will be shown below.

Fig. 12. Sandstone channel-fill built up mainly of tabular units of cross-stratification. Top toward the right. Photograph taken in the area between section 3 and section 4 (Fig. 4).

Modes of deposition

The various lithologic units show a regular distribution in the vertical sense. Most sandstone sheets start with an occasionally loaded, sharply erosive base, overlain by coarse, often pebbly sandstone showing tabular, semi-tabular, and through cross-stratification (McKee & Weir, 1953; Allen, 1963) (Figs. 11 and 12). On top of the cross-stratified members there are many occurrences of parallel-bedded, occasionally rippled, medium to fine-grained sandstones. Grain-size decreases upwards *via* sandy mudstones to mudstones and shales commonly containing siderite concretions and thin siderite beds. At a height of 138 m above the base in section 3, and of 130 m in section 4, the first signs of plant growth *in situ* in a mud and clay environment are recorded by well-developed seat-earths (Fig. 13), topped by a stringer of coal.

Fig. 10. Malpas Formation; vertical variation of percentage and coarseness of coarse rock types. Erill Castell area (section 3) and Peranera area (section 4, sheet III); computed for 30 m intervals.

Fig. 11. Malpas Formation. The hammer rests on the sharply erosive base of a sandstone channel-fill; in the foreground, black shales and chert beds. L = loadcasts; EB = erosive base of channel-fill; CH = chert beds; B SH = black shale. Photograph taken 86 m above the base of section 3 (sheet III).

Sequences such as those described here, but mostly without seat-earths and coal at the top, are characteristic of the Malpas formation in the reaches below approximately 220 m above the base of the sections (221 m in section 3; 217 m in section 4; sheet III). Depending on the criteria applied, 11 sequences can be counted in section 3, and 9 in section 4. It follows that the Malpas Formation shows a cyclic development, and may therefore be directly compared with many other cyclic fluvial deposits (summarized by Duff et al., 1967).

The sandstone units show a close analogy to those of recent point-bar deposits with respect to the upward decrease of grain-size and suite of sedimentary structures (Bernard & Major, 1963; Bernard & Leblanc, 1965; Visher, 1966).

Associated with the upward-fining sequences are instances of surfaces inclined 10 to 25 ° and having a strike parallel to the paleocurrent direction as inferred from cross-stratification. These surfaces probably record the depositional slopes of point bars. Several of such surfaces arranged *en échelon,* as found in a

Fig. 13. Seat-earth (underclay) in the Malpas Formation. Photograph taken 138 m above the base of section 4 sheet III)

number of cases, testify to the lateral accretion of the point bars and consequently to the meandering of the streams. Indeed, only lateral accretion could account for the spread reached by the sandstone sheets, which extend from approximately 50 to 500 m in a sence perpendicular to the main paleocurrent direction (p. 158).

The size of the sets in cross-stratification in the Malpas channel-fills indicates bed forms carrying large ripples. In laboratory experiments such ripples form in the lower flow regime. The upward change, in the channel-fills, to small ripples and parallel bedding in fine-grained sandstone, points to still lower current velocities (Allen, 1965, p. 110), which is in agreement with the suggested point bar origin of parts of the sandstone sheets.

Many sandstone sheets do not conform over their full

width to the structural build-up just described. Some parts of a sheet contain sandy mudstone even near the base; other parts show a sequence consisting entirely of semi-tabular units of cross-stratification, commonly separated by thin strata of mudstone showing parallel laminae. These latter cases may record variations of current velocity which led to alternating periods of deposition of bed load and, to some extent, deposition from suspension.

Although the composite paleocurrent diagrams of the Malpas sandstones show a rather wide spread of current directions, and even complete reversals of current direction, this is not the case in single exposures (Figs. 12 and 15). In the latter the paleocurrent pattern is strictly in one sense, corresponding to unidirectional flow. This means that in the Malpas channels there was no reversal of current

direction brought about, for instance, by tidal influence. The possibility that the current velocities were to some degree checked by tides (thin mudstone levels in the channel-fills) should be left open. The mudstone intercalations, however, could also easily result from seasonal changes in discharge. No direct evidence of the proximity of the sea, either in the form of beach deposits or in the form of a marine fauna, is found in the Malpas Formation.

Some small sandstone bodies (15 × 3 m in cross-section) occur in a form corresponding to narrow channels that apparently kept their courses. The vertical suite of textural and structural properties of the sandstones in these cases is similar to that of a point-bar deposit. They may represent abandoned channels in which a decrease of current velocity led to the deposition of sediments of a gradually decreasing coarseness (Oomkes, 1967).

A large abandoned channel contains a different fill. Approximately 1 km to the East of Peranera, at a height of 150 m in section 4, a unit of sediment is exposed which is more than 20 m wide and approximately 4 m thick. It consists of parallel-laminated, fine-grained muddy sandstone and mudstone, a unique

Fig. 14. Parallel lamination in fine-grained muddy sandstone and mudstone overlying coarse sandstone; interpreted as the fill of an abandoned channel. Malpas Formation.

type of deposit not yet observed anywhere else in the Malpas Formation (Fig. 14). No current structures occur, and therefore this unit could be interpreted as an abandoned channel, possibly an oxbow lake, which was slowly filled up by fine-grained material deposited from suspension.

As noted above, the trends in the uppermost part of the Malpas Formation (above a height of about 220 m) correspond to a change in channel-fill characteristics (Fig. 10, p. 154). Particularly in the coarse conglomeratic unit extending from 217 to 250 m in section 4, internal scour structures (small channel-fills, in fact) are the rule, and the stratification is lenticular. In the sandstones, cross-stratification is well developed in the interval between 221 and 231 m in section 3, but it is absent or very poorly developed above a height of 217 m in section 4. Boulders of up to 40 cm in an imbricate position occur, which is indicative of high current velocities. These observations show that the energy of these streams must have been higher than that of the streams in the middle and lower part of the formation; many of them were capable, at least at times, of transporting a very coarse bed load. The numerous internal scours and the lenticular stratification probably reflect deposition by braided streams. The deposits above a height of 225 m in section 4 still contain conglomerates in sharply erosive channel-fills, but the pattern is well differentiated into graded, upward-fining units. Possibly, the influence of the factors that caused the braiding in the underlying thick mass of conglomerates became weaker, and the streams slowly reassumed the meandering form. As mentioned by Leopold *et al.* (1964, p. 292), transitional types between braided and meandering streams exist. A complete return to very quiet conditions is shown in the very top of the Malpas Formation by lacustrine deposits lying in conformable contact with the overlying Peranera Formation (Fig. 4). The graded channel-fills in the upper part of section 4 are all of limited width, not exceeding 20 m. The grading therefore may correspond to decreases in current velocity in many abandoned meander loops, which generally may have formed a braided system.

The reasons for the sudden initiation of braided stream patterns are not entirely clear. An increase in channel slope (which would amount to tectonic movements, or to stream capturing in the hinterland) or a temporary change to a semi-arid climate or both may only have been components of a complex set of factors (Leopold & Wolman, 1957).

The petrographic analysis of the sandstones occurring in the braided systems reveals the effects of near-surface carbonate diagenesis. This suggests that the streams were at least ephemeral to the extent that temporary drying up of the beds occurred. Such features are absent in the underlying meandering stream deposits (p. 188).

The mudstones and shales are all dark grey to black. Coalified plant remains abound and often form thin
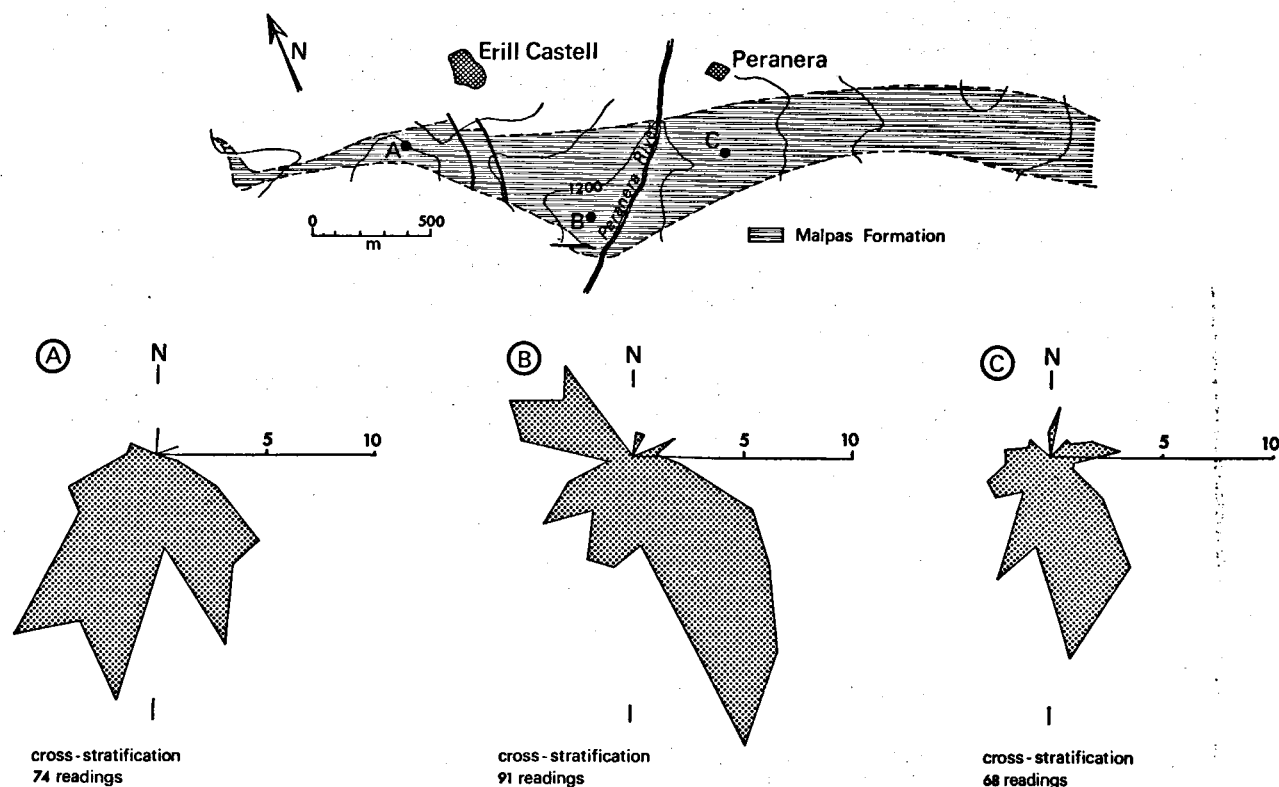
Fig. 15. Paleocurrent data of the Malpas Formation. Diagrams give the directions of dip in cross-stratified sets.

stringers. The structure of the mudstones is crumbly and apparently homogeneous. The fine grain of the deposits indicates low energy conditions. The deposits are interpreted, on the basis of their close association with fluvial channel deposits, as having been formed in a marshy to lacustrine back-swamp environment. The characteristic homogeneity of the fine-grained sediments is thought to result from the presence of many kinds of vegetation, e.g. floating, suspended, and substrate-attached types. No indications of homogenization by a burrowing fauna were found, in contrast to the Peranera and Bunter mudstones, where such indications are common (p. 159, p. 167). The possible general absence of burrowers in the Malpas mudstones may have resulted from a too toxic *milieu* (reducing, H_2S-carrying waters); in the aerially exposed Peranera and Bunter floodplains to the contrary, they could thrive.

The Malpas back-swamps were probably flooded most of the time. The levels of strongly homogenized seat-earths record situations in which the roots of a different vegetation could penetrate the muds to some extent; here the prevailing water-table may have been somewhat lower, although the environment was probably still waterlogged (Smith, 1964; Huddle & Patterson, 1961).

At two levels in section 3 (127 and 184 m above the base of the section), sandstones occur in an upward-coarsening sequence. They may represent crevasse-splay deposits formed by overflow and break-through

of natural levees, which permitted the introduction of increasingly coarser-grained material into the calm back-swamp environment.

The very top of the Malpas Formation, near the conformable contact with the Peranera Formation, consists of very fine-grained clastic sediment and fine-grained ferroan dolomite, and shows mud cracks (section 5, sheet III). Stratification is clear and parallel, and there is no evidence of scouring or of other current structures, which suggests fully lacustrine conditions.

Paleocurrents

The paleocurrent directions in the Malpas Formation were determined by measuring the strike and dip of the sets in tabular and trough types of cross-stratification. The directions of dip of the sets, graphically corrected for simple tectonic tilt, are given in Fig. 15. Although there is a considerable spread in the paleocurrent directions in each diagram, which could be accounted for by the meandering of the streams, it is clear that the streams must have flowed in southerly directions. The greater spread of directions at stations B and C as compared to station A may be the result of stronger meandering at these localities.

Meckel (1967) has shown that trough cross-stratification gives a greater spread in the diagram than tabular cross-stratification. The greater spread at localities B and C is however, as far as could be checked in the field, not due to higher frequencies of trough types of cross-stratification.

Paleotopography

The higher content of coarse deposits in the Peranera area as compared to the Erill Castell area points to greater current activity in that area (Table I; Fig. 4). This, and the probability that meandering may have been more intense at Peranera, suggests that the axes of the many successive flood-plains may have been closer to Peranera than to Erill Castell.

It seems that the location of the flood-plains in which the Malpas sediments accumulated was to some extent still conditioned by the topography of the valley occurring on the Paleozoic basement below Erill Castell and Peranera (p. 151). This valley may have been only partly filled with volcanic deposits by the time deposition of Malpas sediment started, as suggested by the limited lateral extension of the Malpas Formation in a direction perpendicular to the paleocurrent directions [1]. The eastern termination, in the form of a thin wedge, is a natural one, developed in almost solely sideritic and dolomitic mudstones and black shales (Fig. 5). To the West, the Malpas Formation is cut away by the unconformity at the base of the Bunter, but it may be assumed that the flood-plains may also have terminated hereabouts, as indicated by the increase in fine-grained material in this direction (Table I).

PERANERA FORMATION

Composition

From base to top, the Peranera Formation is developed in a fluvial (and probably partly eolian), red-bed facies. Three main rock types occur: (a) greyish-red mudstones (75 %), (b) greyish-red, commonly conglomeratic sandstones (24 %), and (c) greenish-grey or reddish tuff beds (1 %). A detailed section of the formation, taken just North of Las Iglesias, is presented on sheet IV (section 6, location of section in Fig. 5).

Conglomeratic sandstones

As in the Malpas Formation, the coarse deposits occur in the form of fluvial channel-fills. Some rare Peranera channel-fills show features similar to those of the Malpas Formation, and may correspond to point-bar deposits. They are of limited lateral extent, not exceeding a width of 40 m.

Most channel-fills, however, show a number of characteristics testifying to a very different mode of deposition. In their lower parts they contain flat lenses stretched parallel to the channel axes, poorly developed, commonly low-angle cross-stratification, occasionally primary current lineation, and deeply incised internal and basal scour-and-fill structures. The scour-and-fills at some channel-bases are similar to a type also seen at the bases of sheet-flood deposits in an alluvial fan facies (Nagtegaal, 1966) (Fig. 16). These features, taken together, suggest rapid flow.

[1] The sheet of basaltic andesite directly underlying the Malpas Formation also shows a limited lateral extent (Fig. 4). The shape of the sheet and its position in the geological map suggest flow of only slightly viscous lava through a valley partly filled with tuffs (p. 153).

Most of the channel-fills are not wider than 200 m, and many do not exceed 50 m.

The streams were ephemeral, as shown by the fact that various levels of rain prints and mud cracks occur *within* the channel-fills, starting locally at only 30 cm above the erosive base (section 7, sheet IV). Sudden decrease of current velocity in shallow streams is indicated by current-rippled surfaces on sandstone overlain by a mud-cracked thin veneer of mudstone (Fig. 17). Current ripples are the characteristic structure of the usually finer-grained, upper part of the channel-fills.

The characteristics of the Peranera channel-fills point to semi-arid conditions. The fact that the stream beds fell dry (intercalated mud cracks and rain prints) indicates that at the same time or shortly afterwards, the largest part of the alluvial plain must have fallen dry. These conditions stand in sharp contrast to those in the Malpas environment. There, the overbanks and the stream channels were probably almost continuously filled with water, with the exception of the intercalated member of braided stream deposits (p. 157; see also discussion of Bunter channel-fills, p. 167).

The clastic assemblage in the Peranera conglomeratic sandstones is immature, both texturally and mineralogically (p. 201).

Mudstones

The mudstones locally show current ripples and fine parallel lamination. Wherever present, these structures apparently escaped homogenization by burrowers and roots, whose signs are clearly displayed in most other mudstone units. The occurrence of root imprints *(Lebachia piniformis* (?), p. 149) indicates that, at least at times, some vegetation did occur in the Peranera environment. Although the amount of this vegetation is impossible to estimate, the mere presence of root levels excludes the possibility of continuously barren conditions. It is of interest to note in this respect that nowhere in the Peranera Formation were sandstone bodies of any appreciable thickness found that could be considered of eolian origin. An unknown, but possibly substantial amount of the mudstones however, may have been brought in as airborne dust (p. 163).

The homogeneity of the mudstones need not have resulted only from reworking *after* deposition; it may in part have been produced by thin covers consisting of low plants, the presence of which would prevent the development of such structures as parallel lamination and ripples. This effect of plants has been described, for example, by Van Straaten (1955) from tidal flat environments.

The occurrence of numerous levels of isolated to densely packed calcite nodules is a striking characteristic of the Peranera mudstones. The nodules are particularly well displayed and occur very frequently between 40 and 80 m above the base of section 6 (sheet IV). In various levels the nodules, which have a con-

cretionary origin (p. 207), are oriented with their long axes perpendicular to the stratification (Fig. 18); at most levels, however, the nodules are equidimensional or are oriented parallel to the stratification.
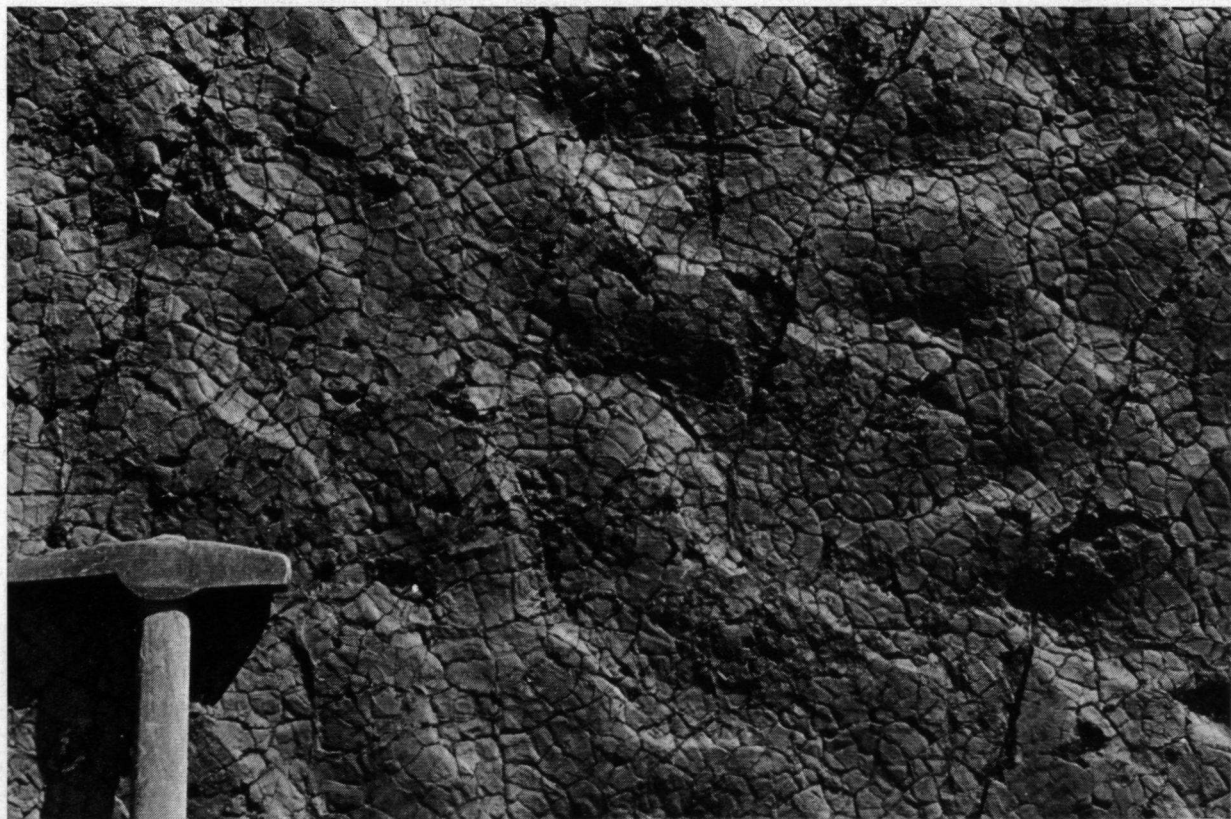
Nodules similar in habit and size to those in the Peranera mudstones are also common in recent caliche profiles. Fig. 19 illustrates such nodules from a caliche profile containing calcrete in its uppermost horizon. The photographs in Fig. 19 were taken in a region rich in caliche, previously studied by Rutte (1958;

eastern coast of Spain). Rutte concluded that the nodules are formed in the subsoil — within the first 2 m below the surface — by descending waters (illuviation). According to Rutte, the association with calcrete is common, but nodules also form in regions where no calcrete develops (e.g. in the vicinity of Barcelona). Where only nodules form, annual precipitation is 500 mm or somewhat higher, the optimal conditions for the formation of calcrete being in the order of 150 to 250 mm. In both cases rather high

Fig. 18. Greyish-red mudstone packed with vertically oriented calcite concretion in the Peranera Formation. Stratification is parallel to the handle of the hammer and perpendicular to the plane of the photograph. Photograph taken 72 m above the base of section 6 (sheet IV). Compare with Fig. 19.

temperatures, which favour evaporation in the surface layers, and the occurrence of dry seasons are the decisive factor in the generation of both limestone crusts and nodule levels.

Ramana Rao (1966) described recent nodular calcareous deposits of similar origin (kankar) from the Hyderabad area, India, where the conditions of formation are comparable. Depth below the surface:

Fig. 16. Scoured base of a greyish-red conglomeratic sandstone sheet in the Peranera Formation. Photograph taken 29 m above the base of section 6 (sheet IV).

Fig. 17. Mud-cracked thin veneer of greyish-red mudstone overlying current-rippled sandstone in the Peranera Formation. Photograph taken 201 m above the base of section 6 (sheet IV).

60 to 180 cm; average annual precipitation: 640 to 760 mm (largely restricted to the summer months); air temperatures: from 15° C in winter to 40° C in summer.

On the basis of the similarity of the Peranera calcite nodules to those found in recent caliche, it is concluded that both formed under similar conditions. This conclusion is corroborated by the similarity in calcite microtextures of the fossil and the recent caliche (Nagtegaal, 1969; p. 207).

It follows that the prevailing climate during the accumulation of the Peranera deposits must have been sufficiently hot to cause strong evaporation, that rainfall was markedly seasonal or intermittent (a fact which also follows from the ephemeral character of the streams as deduced from their types of fills; see above)

and may have exceeded the annual value of 500 mm. Temporarily, however, conditions may have been drier, as evidenced by a horizon of calcrete occurring at the height of 112 m above the base of section 6 (sheet IV). This is the only horizon of calcrete met with in the Peranera deposits under study.

The cited examples of recent caliche, in which the nodule levels do not exceed a thickness of approximately 2 m, occur on degrading land surfaces. The Peranera caliche, on the other hand, formed on a generally aggrading land surface. Only if this fact is realized can the considerable thickness of some Peranera nodule levels (over 30 m) be understood: nodule formation must have kept pace with the rate of deposition. Apparently, a temporarily slower rate of clastic deposition permitted the formation of calcrete when the climatic conditions were right, but in many cases the rate of deposition may have been too high to allow for the development of complete caliche profiles strongly enriched in calcite (Nagtegaal, 1969).

Extensive polygonal mud-crack patterns also occur in the mudstones, but they were only noted where they are overlain by a tuff bed, in that case as negatives on the base of the bed.

The numerous thin tuff beds provide an important clue concerning the environment of deposition. Some of these beds, intercalated between mudstone, could be traced over more than 1 km, suggesting that the deposition of tuff occurred on an even and essentially flat surface.

A similar significance should be attributed to two highly persistent levels of greenish shale (at 470 m and at 480 m above the base of section 6, sheet IV). The persistency of these beds, their extreme fine grain and probable chemically reduced state (no laboratory determinations) suggest that they record the temporary establishment of a playa environment.

In the uppermost part of the Peranera Formation (410 to 730 m; section 6, sheet IV), channel-fill deposits fail almost completely. Almost all vertical transitions in deposits predominantly consisting of parallel-stratified mudstones are gradual. Moreover, the fine-grained upper part of the Peranera Formation is similarly developed in widely separated areas, e.g. in the Segre valley and at Castellar de N'Hug (J. Bloemraad, pers. comm.).

These facts support an eolian origin in the sense that

Fig. 19. Recent caliche. Large photograph shows a complete caliche profile containing calcrete (light coloured lens, top part) and underlying densely packed calcite nodules, many of which are oriented horizontally. Predominant colour is yellowish-brown. The very top part of the profile, the loose soil overlying the calcrete, is missing, probably washed away by surface water.

Inset shows vertically oriented calcite nodules, occurring in the same road-side section. Photograph taken along the Barcelona-Valencia highway, at Ametlla de Mar, Spanish East Coast.

airborne dust accumulated but was not picked up again.

Environment of deposition

One gains the impression that the main body of coarse sediment was deposited by wide, shallow, rapid, and ephemeral streams having one or more high velocity paths transporting and depositing the coarsest bed load. Such types of streams, called stream-floods and sheet-floods (Blissenbach, 1954), have been described from recent alluvial fan depositional areas by various authors (summarized by Allen, 1965, p. 158). By analogy, it is thought that in the Peranera Formation, the coarse deposits form part of such a former alluvial fan environment.

The high proportion of mudstone (75%) and the persistent tuff and shale beds suggest, however, that in the exposed stretch of the Peranera Formation the main environment of deposition was that of the lowlands commonly bordering the alluvial fans at the downstream ends (Twenhofel, 1950, p. 69). The two main influxes of coarse material (see below) may represent temporary advances of the environment situated closer to the alluvial fans; the fans proper may have been situated still further to the North or Northeast (see under *Paleocurrents*, p. 164).

The indications of a relatively low-gradient topography, the ephemeral character of the streams, the presence of root levels, the common development of caliche, and the oxidized state of the deposits, all strongly suggest that the prevailing Peranera environment was, both morphologically and climatologically, similar to that of the present-day steppes occurring between belts of savannahs and deserts (Tricart & Cailleux, 1965, p. 279, Fig. 48) (see footnote on p. 151).

It is worth noting that an environment of deposition strikingly similar in many aspects to the one described here for the Peranera Formation, has been recorded for the New Red Sandstone found on the islands of Raasay and Scalpay off the southwest coast of Scotland (Bruck *et al.*, 1967).

Vertical lithological variation

Fig. 20 shows the vertical variation of the percentages of the coarse deposits (sandstones, conglomeratic sandstones) and the non- or little reworked, continuous tuff beds. The percentages were computed from section 6 (sheet IV) for 50 m intervals.

There are two main influxes of coarse material particularly rich in pebbles, the first lying between 100 and 150 m, and the second between 350 and 400 m. A lateral reconnaissance in the field showed that this phenomenon can be traced over the entire area of exposure of Fig. 5.

The sudden introduction of coarser-grained material may have been due to slight upheavals in the source area of the sediment or may correspond to periods of very heavy downpours. In the area studied, no corresponding unconformities occur in the Peranera Formation.

Fig. 20. Peranera Formation; vertical variation of percentage and coarseness of coarse rock types and of percentage of tuff beds. Computed for 50 m intervals. Section 6 (sheetIV).

The interrupted occurrence, in the vertical sense, of *in situ* tuff beds need not correspond to long-term cessations of volcanic activity. A more plausible explanation is that, as suggested by Fig. 20, during times of increased influx of coarse material tuff beds already deposited in the accumulation area ran a higher risk of being reworked and dispersed. Another point to be kept in mind is that the vertical axis in the figure does not represent a scale with equal time intervals; the rate of deposition probably varied with the percentages of coarse material.

Puddle deposits

An interesting feature of the Peranera Formation is the occurrence of what can be termed 'puddle deposits'. These are deposits formed in drying rain puddles or in residual puddles in the beds of dried-up streams.

The fossil puddle fills show, as do recent ones, one or more laminae consisting of fine-grained material often graded fining upwards. The total thickness of the deposits varies from less than 1 cm to about 40 cm probably depending on whether the puddles were isolated or formed a system interconnected by small streams which for some time continued to carry a supply of sediment. Two properties are significant in the recognition of puddle-fills: (1) restricted lateral extent, and (2) an uppermost lamina usually less than 1 mm thick corresponding to the finest fraction settled out of suspension. This uppermost lamina is often quite distinctive, showing a soft, silky gloss. The same gloss is observed in recent dried-up puddles; apparently, this feature is not destroyed during diagenesis. Rain prints, mud cracks, birdseye structure, many problematic tiny surface disturbances, and plant imprints *(Walchia piniformis)* are the features normally associated with the puddle deposits in the Peranera Formation. Puddle deposits also occur, although less frequently, in the Bunter Formation (p. 168), but were not found in the Aguiró and Malpas Formations.

Fig. 21. Paleocurrent data of the Peranera Formation. Diagrams give the directions of dip in cross-stratified sets and orientations of channel-fills and scour-and-fill axes.

Paleocurrents

The paleocurrent data for the Peranera Formation are presented in Fig. 21. Channel axes and the axes of

scour-and-fills run almost strictly North-South (data collected at stations A and C, at the height of the first coarse influx, p. 163). Cross-stratification (data originating from station B, at the height of the second coarse influx), however, indicates a WSW direction of flow. Because of the scarcit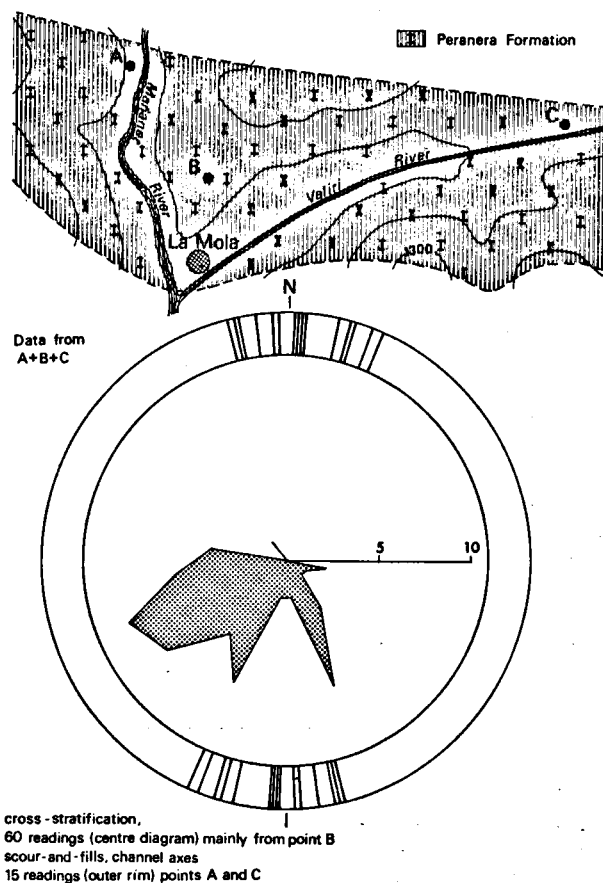y of paleocurrent information it is not possible to conclude more than that the main direction of flow may have been from an angle varying between North and East.

BUNTER FORMATION

Viewed on the scale of the Pyrenees as a whole, the Bunter Formation presents itself as an almost omnipresent, paper-thin sheet. Where the Bunter Formation is not found, as in the Llobregat valley (75 km East of the study area) and East of that locality, there is evidence, in the form of an unconformity surface, that its absence is due to erosion previous to the deposition of younger strata.

Composition
In this report, the term 'Bunter Formation' normally refers to the relatively thin, highly continuous formation. But at one locality, just North of Iguerri (Fig. 4), a thick unit of conglomerates and coarse-grained sandstones occurs which for petrographic reasons is considered as a member, the Iguerri member, of the Bunter Formation (p. 149).
The Bunter Formation (excluding the Iguerri member), like the Peranera Formation, is entirely developed in a red-bed facies. In the area under study it consists mainly of an alternation of two rock types: (1) pale-red to greyish-red, normally homogenized mudstones, and (2) pale-red to greyish-red, well-sorted, cross-stratified sandstones. These rocks make up 63 per cent and 31 per cent, respectively, of the section selected for detailed study (Tor River valley; section 8, sheet V). Conglomerates constitute a third lithological type; they frequently occur at the lower contact of the formation (not exposed in this valley) and in one or two levels at varying heights. In the Tor River valley they are also found near the top of the formation and they amount to 6 per cent of the total section.
No indications of other than fully terrestrial conditions were found in the Bunter Formation.

Paleotopography
In the entire area of the study, the lower boundary of the Bunter Formation is an angular unconformity. Where the Bunter Formation overlies the strongly folded pre-Hercynian Paleozoic rocks (Fig. 6), the angle of the unconformity varies between 0° and 90° and opens in many different directions. Where the Bunter Formation overlies older deposits (Erill Castell Volcanics, Malpas and Peranera Formations; Figs. 3, 4, and 5) the angle of the unconformity varies between 10° and 60° and constantly opens towards South.
Since the structure of the entire sequence is one large

monocline (see Geological Setting, p. 145), the unconformity is related to a southward tilt.
A remarkable feature of the Bunter unconformity is its general flatness. This is true also of many other regions in the Spanish peninsula (Virgili, 1968; p. 814; 1960a). Nowhere in the exposed stretch are there signs of any marked paleorelief, either in the morphology of the unconformity surface or in the directly overlying Bunter deposits. Locally, however, subdued highs and lows are suspected (see under *Paleocurrents;* p. 168). At distances of 2 to 5 km to the North, the unconformity surface shows some more pronounced ridges and many other irregularities corresponding to a small-scale relief, whereas directly overlying breccia reflect the local lithology (Mey, 1967). Similar relations exist in an area 20 km West of the Tor River, which was mapped in detail by Roger (1965), and between the Pallaresa and Segre rivers, East of the study area (Hartevelt & Roger, 1968; Hartevelt, pers. comm.). Paleotopographic highs appear to occur preferentially at localities where the basement contains rocks relatively resistant to erosion (commonly quartzites; occasionally Devonian limestones). The relief certainly did not reach the dimensions of the much older unconformity surface as at Aguiró: most of the highs are completely buried by the relatively thin Bunter deposits.

The shape of the unconformity surface below the Iguerri member is entirely different (Fig. 4), suggesting the presence of a fossil deep valley, now filled-in with mainly conglomerates. The valley-fill is of the dimensions of the valley-fills at Aguiró (p. 150). It is assumed that the fossil valley at Iguerri is an incidental remnant of the important erosion period that preceded the deposition of the Bunter Formation (see below).

If, in a graphic construction, the Bunter Formation is rotated back to its former near-horizontal position and the then southward-dipping underlying formations are extended in a straight line northwards, it becomes evident that enormous amounts of material should have been eroded prior to the deposition of the Bunter Formation. A certain degree of degradation is also in agreement with the fact that the granodiorites became exposed before the deposition of the Bunter Formation (p. 213).
There is another reason why it may be concluded that degradation before and during the deposition of the Bunter Formation must have reached an advanced stage: when the deposition of the Bunter Formation had come to a close, a wide-spread marine transgression set in and fine-grained dolomites, limestones, and evaporites were deposited (Pont de Suert Formation; Mey *et al.,* 1967). These rocks contain hardly any terrigeneous material, which also points to emerged lands with little relief.
It is probably not fully justified, however, to simply extend the formation boundary planes as straight lines northwards: the present cross-sectional picture could be accounted for equally well by a slight upheaval (or even a constant level) of the region to the North and a relative subsidence of the region to the South.

A major 'knick' would in that case have occurred at the height of the Nogueras Zone (see Geological Setting, p. 145; compare cross-sections in Figs. 3, 4, and 5). Also, the Aguiró, Malpas, and Peranera Formations must all have formed wedges diminishing in thickness and even ending toward the North, because their clastic material was derived from sources to the North (p. 150, 158, 164). In any case, a fair amount of erosion must have taken place, as indicated by the unconformity angles, which run up to 60°.

Because the relatively flat basal surface of the Bunter Formation is carved into the underlying rocks and has a wide extension, it can be considered to be a pediment surface. Paleocurrent directions in the Bunter deposits indicate that this surface must have (gently) sloped southwards (p. 168).

Although recorded from regions with a humid climate, pediments reach their maximum development under arid and semi-arid conditions (Leopold *et al.*, 1964), and a semi-arid climate prevailed previous to the deposition of the Bunter Formation, when the Peranera deposits accumulated (p. 171). It is probable that the pediment surface is the ultimate result of a type of erosion that already prevailed in Peranera times.

Conglomerates

The conglomerates of the Bunter Formation are quite different from those contained in the underlying Peranera Formation, and show in general the following properties:

(1) only a few, well-separated levels having a maximum thickness of about 30 m,
(2) sharply erosive, scoured bases and sharp to gradual, occasionally coarse sandy tops,
(3) internal scours, poor stratification, and rare imbrication,
(4) stable components; pebbles are quartzite, quartz, chert, and occasional Bunter mudstone chunks and flakes,
(5) high percentages of well-rounded pebbles (Fig. 22).

The Tor River section (sheet V) shows only one level of conglomerate, near the top of the section. These conglomerates, which are strikingly light-coloured, form a wide sheet of almost constant thickness traceable from the Tor valley westwards to the Baliera valley, a distance of approximately 9 km. A similarly continuous sheet in the Bunter Formation, mapped by Roger (1965), occurs at Las Paules. Roger found this sheet to extend over more than 8 km in an East-West direction.

The sandstones in the Tor River section contain no pebbles, not even at the bases of the channel-fills, and are well-sorted. This suggests that the level of light-coloured conglomerate corresponds to a major change in the fluvial regime. The formation of the more than 9 km wide sheet cannot have resulted from a single catastrophic event such as an extraordinary heavy cloudburst, but may well have been due to the establishment of a braided-stream system that migrated laterally over a relatively flat alluvial plain.

The conglomeratic deposits of the Iguerri member were not studied in great detail, and no section was drawn up. The deposits of the Iguerri member are better differentiated in conglomerate and sandstone levels than the conglomerates in the overlying Bunter Formation. Internal scours are also common; cross-stratification as well as parallel bedding were noted in the sandstones. The conglomerates are almost structureless, except for lateral and vertical gradations in pebble-size and the presence of scour structures. These features, and a general absence of intercalated mudstone and shale, suggest a prevailing braided-stream environment of deposition.

Sandstones

The sandstone units in the section studied vary in thickness from less than 0.5 m to 6.5 m, and in width from a few metres to an estimated 200 m. A downward-convex shape and erosive bases in mudstone show that most of the sandstone units are channel-fills. Most of the channels must have been very shallow in proportion to their widths.

The channel-fills almost all show good cross-stratification in tabular, semi-tabular, and trough types (Fig. 23). In single exposures, the cross-stratification is strictly unidirectional.

The internal structure of the Bunter channel-fills resembles that of the Malpas channel-fills. Both con-
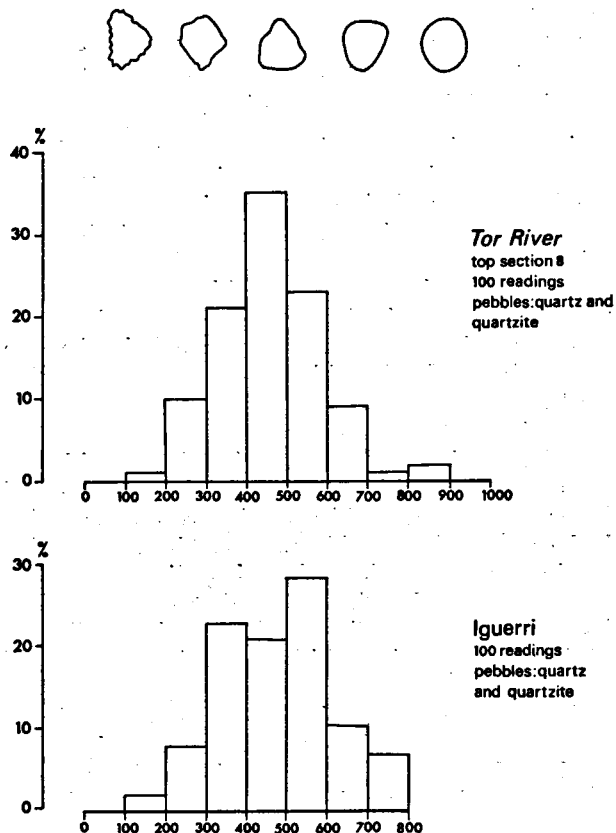

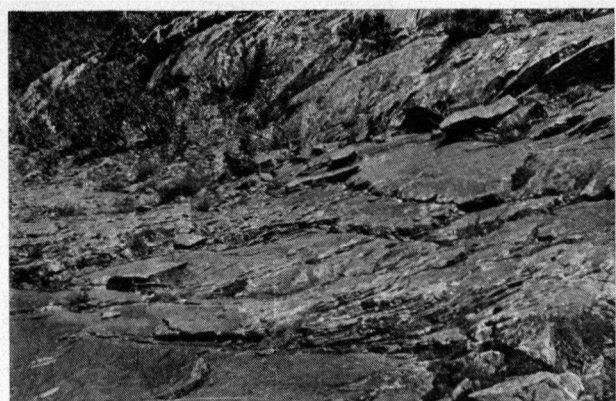Fig. 22. Roundness of pebbles; Bunter Formation.

Fig. 23. Trough cross-stratification in Bunter sandstones. Photograph taken in the area between Aguiró and Castelnou de Avellanos (Fig. 3).

tain cross-stratification resulting from large ripple bed forms indicative of the lower flow regime (p. 156). They are in marked contrast to the internal structure of the majority of the Peranera channel-fills, which probably resulted from rapid flow (p. 159).
In contrast to both Malpas and Peranera sandstones, the Bunter sandstones are well-sorted. The sand grains are, in the vertical sense, either of almost constant size throughout (many channel-fills) or decrease in size upwards as in the channel-fill starting at 11 m above the base of section 8 (sheet V). The very top of the channel-fills, however, is normally graded from fine-grained sandstone to sandy mudstone.

Flow in the Bunter channels must have continued long enough at a time to sort the sands (though a considerable degree of pre-sorting may have resulted from repeated intervals of transport somewhere upstream), to bring about the well-developed cross-stratification, and to prevent frequent drying up of channel beds (in contrast to the Peranera channels p. 159). But flow apparently did not persist long enough to induce much lateral channel migration, although the setting of the alluvial plain (low relief) might have been ideal for widespread meandering.
It is also highly unlikely that the many small-sized channels (most of the channel-fills being less than 1 m thick; Fig. 26) could have extended lengthwise over the whole alluvial plain. The duration of flow therefore was probably limited, because moderate to high precipation occurred only seasonally or at intervals, and strong evaporation and infiltration would cause losses of water. As in the Peranera deposits, flow in the stream channels was ephemeral. But once the channels were filled with water, flow must have continued longer.
Indications of eolian transport of sand were not found in the Bunter Formation.
The mudstones may have been deposited laterally, from suspensions overflowing the stream banks, and probably also at the downstream ends of dying channels. An unknown, but probably small amount of mudstone may have been brought in by air.

There is another observation suggesting that the Bunter channels were not filled with water all year round: i.e. the fact that the Bunter sandstones show petrographic evidence (as do the Peranera sandstones), in the form of a primary authigenic precipitate of ferric oxide, of having been completely oxidized. If the stream channels were continuously filled with water, concentration of some kind of vegetation would have occurred along the borders of the streams. In such circumstances much vegetational debris accumulates in the stream bed, thus bringing about reducing conditions in the sediment deposited in it. Examples of such an environment, with which the author is personally acquainted, are found in the savannahs of Surinam (S.A.). If these savannah deposits formed a generally aggrading system, they would probably ultimately show a sedimentary sequence in which the overbank deposits showed signs of oxidation and the channel-fills signs of reducing conditions. Deposits in the geological record to which these relations may apply are those of the Devonian Catskill delta (U.S.A.). Here, the overbank deposits are red, but the channel-fill sandstones are commonly grey or greenish-grey (Dunbar & Rodgers, 1958, p. 215; Friend, 1966) The absence of such relations in the Bunter deposits indicates that, after the channels had been filled in with sands, the ground-water table dropped sufficiently to let air penetrate between the grains.

It is believed that the differences between the Peranera and the Bunter channel-fill characteristics are chiefly related to a difference in climate in the sense that during the deposition of the Bunter Formation the climate was less dry than during the deposition of the Peranera Formation, and in part also to a difference in topographic setting in the sense that the topographic gradients were less steep (implying larger drainage areas) in the Bunter than in the Peranera environment. This point is also discussed on p. 171.

Mudstones

The mudstones, which by volume are approximately twice as frequent as the sandstones in the section studied, occur in continuous levels, locally 'washed out' by the sandstone channel-fills. Although because of their marked homogeneity the mudstones are less spectacular than the nicely-structured sandstones, they in fact constitute the dominant element in the Bunter environment. This applies even more strongly to the Peranera mudstones (p. 159).
At various levels in the mudstones there are 'ghost' structures of current ripples and parallel lamination; these structures, which apparently partly escaped destruction by homogenization, indicate that the mudstones were at least in part deposited from flowing water (section 8, sheet V).
The homogeneity of the mudstones, which is considered to be a highly significant property, could have been brought about by a number of processes: the influence of the presence of a cover of low plants, reworking by the roots of those plants, and reworking

by burrowers. Direct evidence in the form of root-imprint horizons and burrows is clearly displayed in the field. In thin section, too, the signs of reworking of the material are clear (p. 224).

As in the Peranera environment, indications of the formation of caliche are present. These indications include nodules which are oriented vertically at two levels but show no preferred orientation at other levels, and numerous spherulites. Both types of concretionary structures probably formed from calcite but were later dolomitized. Caliche in the Bunter deposits is, in comparison to that in the Peranera deposits, only very poorly developed, which may be due to a too humid climate and an unsuitable substrate (p. 227).

Compared to the Peranera deposits, interpreted as having accumulated in a prevailing steppe environment, the Bunter deposits show a number of similarities such as low gradients, oxidized state of the deposits, presence of root levels, and the (modest) development of caliche. Marked dissimilarities, however, lie in the fact that flow in the Bunter stream channels appears to have been more continuous and that the Bunter deposits show the effects of a much stronger chemical weathering (p. 222). For these reasons it is suggested that there was more precipitation in the Bunter environment and that the environment was similar to that of the present-day savannahs, located between the steppe and tropical rain-forest belts (Tricart & Cailleux, 1965, p. 286, Fig. 48) (see footnote on p. 151).

Puddle deposits

Some channel-fills, particularly the smaller ones, show phases of drying up during the last stages of infill with fine-grained sediment. A good example of this occurs at the top of a channel-fill exposed between Aguiró and Castelnou de Avellanos (Fig. 3). Here, at least 9 phases can be distinguished in a puddle deposit on the basis of successive levels of rain prints (Fig. 24). Puddle deposits are much less frequent in the Bunter than in the Peranera Formation.

Paleocurrents

The paleocurrent data for the Bunter Formation are presented in Fig. 25. These data were collected at 6 stations (A to F). For each station, the directions of dip of the sets in tabular, semi-tabular, and through types of cross-stratification, graphically corrected for simple tectonic tilt, are shown jointly.

The frequency distribution of the directions of dip of the cross-stratification sets in each separate channel-fill is unimodal and shows restricted spread (not shown). The wide spread seen in most of the diagrams results from the added values of many channels, which were measured at random in small circular areas having a diameter of no more than 200 m. It is therefore not known whether the diversity in the directions results from braiding or meandering or whether it is due to superimposed, differently oriented channels. In view of the environment of deposition, the latter two possibilities seem more likely as the main cause.

To the West of Erill Castell, maxima pointing to southwesterly directions are found (stations A, B, and C). Going toward stations D and E, the maxima rotate to the South; at F and G they are directed SE. It is not known, however, to what extent the orientation in diagrams F and G is influenced by movement along the fault occurring at this locality (Fig. 25).

The difference in direction found between the hamlets of Peranera and Vihuet may have originated from a local, subdued high then situated between Iguerri and Erill Castell. This high could have resulted from a difference in resistance to weathering of Devonian (relatively high resistance) and Carboniferous (relatively low resistance) rocks whose line of contact runs just North of Iguerri (Fig. 4). At present at least, a pronounced ridge of Devonian rocks occurs along this line of contact. But the high could also be related to the differential upheavel that occurred after the Peranera Formation was deposited (p. 152).

Fig. 24. Above: Thin-bedded puddle deposit showing sharp and blurry rain prints at successive levels. Note fine surface texture of the thin beds, typical of puddle deposits.

Below: Cross-section of the fragment lying in the centre of the upper photograph. The thin bed is graded, with upward fining, but contains appreciable amounts of clay near and at its base. The upper surface shows the imprint of a rain drop partly filled with greyish-red clay. To the left and right of the imprint there is a thin wedge of fine sand. Note also disturbance of the sediment at the site of impact of the rain drop, two sand-filled, thin burrows (top right) and many black spots consisting of void-filling sparry calcite (birdseye structure). Sample 1079; *negative print*, plain light.
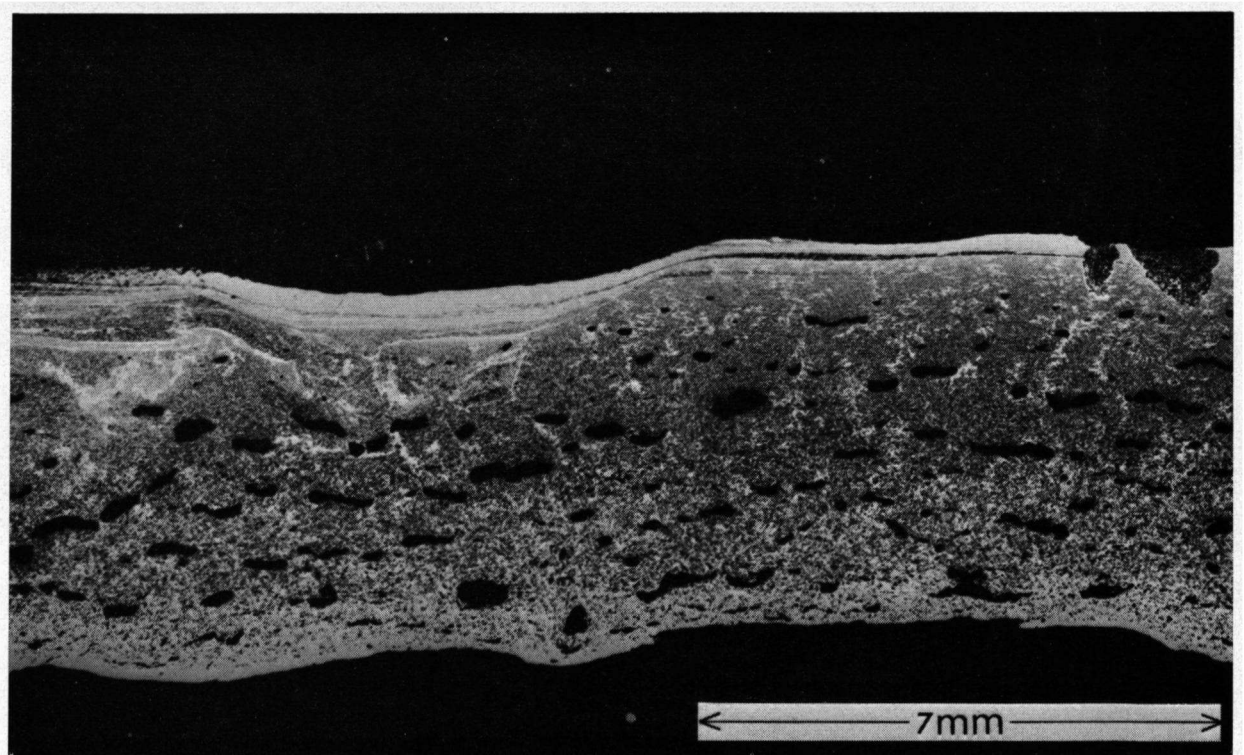
Fig. 25. Paleocurrent data of the Bunter Formation. Diagrams give the directions of dip in cross-stratified sets.

CHAPTER III

DEPOSITIONAL ENVIRONMENTS: SUMMARY, COMPARISON, AND CONCLUSIONS

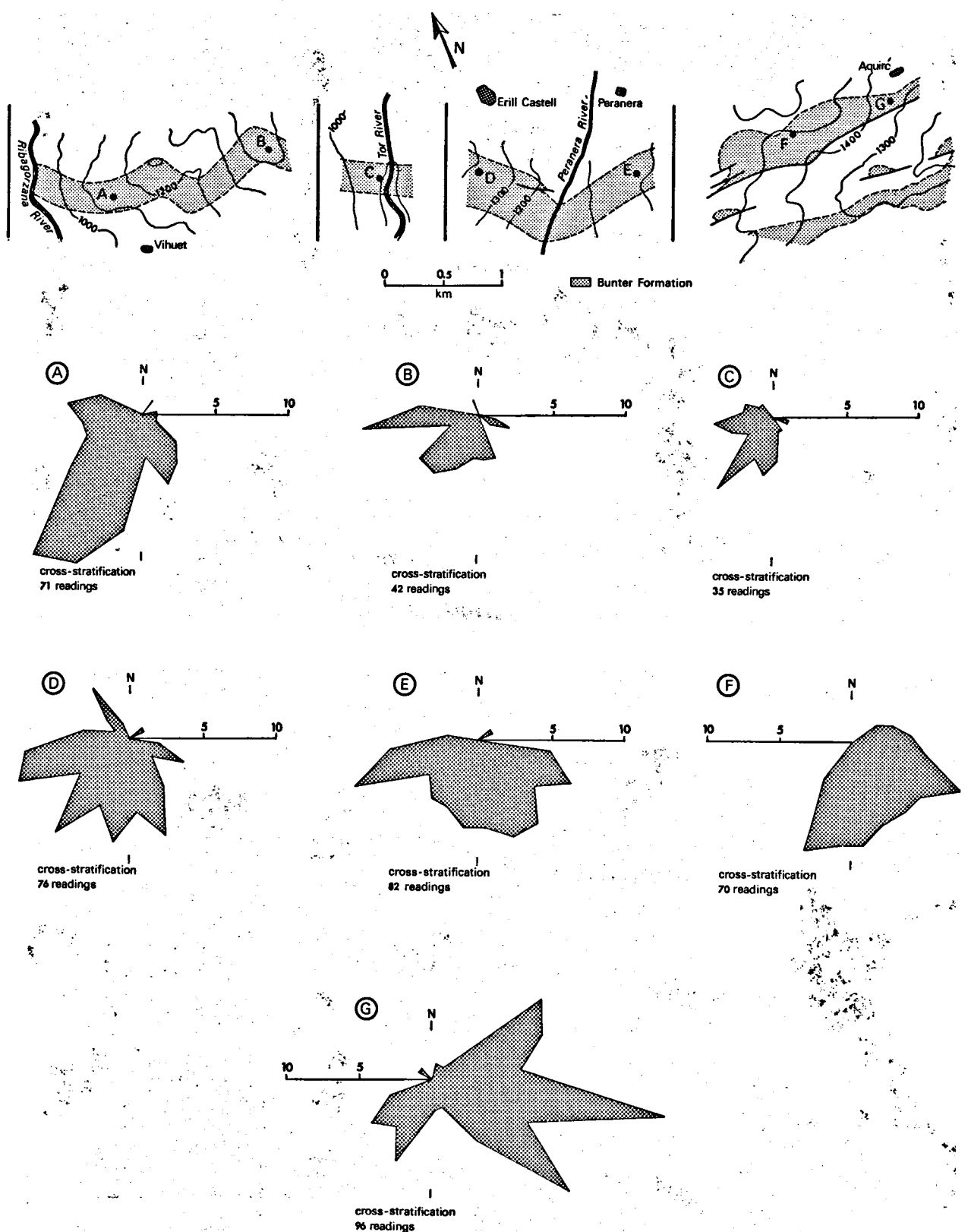Table II summarizes the main environmental characteristics of the Aguiró, Malpas, Peranera, and Bunter Formations. These four formations have in common that they were all, at least for the major part, deposited by means of fluvial processes, but they nevertheless differ in many aspects.

These differences are greatest between the Aguiró and Malpas Formations on the one hand, and the Peranera and Bunter Formations on the other. The decisive factor must have been the climate during the time of deposition.

In the case of the Aguiró and Malpas Formations the

TABLE II

	AGUIRO FORMATION	MALPAS FORMATION	PERANERA FORMATION	BUNTER FORMATION
Environment of deposition	fluvial	fluvial	fluvial	fluvial
Predominant colours	dark grey	dark grey	pale red greyish-red	pale red greyish-red
Paleotopography	high gradients	moderate gradients	moderate to low gradients	low gradients
Predominant types of streams	braided	meandering	ephemeral, moderate to high energy	ephemeral, low energy
Overbank deposits	flooded back-swamps	flooded back-swamps	altern. dry and wet	altern. dry and wet
Vegetational debris	abundant, coal beds	abundant, coal beds	imprints only	imprints and rare coalified fragments
Paleosols	not observed	some black seat-earth	caliche common	caliche present
Width alluvial plain	3 km	≫ 8 km	very extensive	very extensive
Puddle deposits (including rain prints, mudcracks)	not observed	not observed	frequent	present
Tuff beds	some	some	numerous	not observed

Table II. Summary of the main environmental characteristics of the Aguiró, Malpas, Peranera, and Bunter Formations.

precipitation : evaporation ratios were sufficiently high to keep the rivers flowing and to keep the back-swamps flooded most of the time, thus insuring subaqueous preservation of the abundant vegetational debris; the climate must therefore have been humid.

In the case of the Peranera and Bunter Formations the climate was warm and precipitation occurred seasonally. These conditions led to ephemeral flow in the stream channels, the generally dry condition of the flood plains, the oxidation of the deposits, the oxidation of organic matter at near-surface levels (root levels are present but there is no coalified vegetational debris except for rare occurrences in the Bunter Formation), and to the formation of caliche (common in the Peranera, present in the Bunter deposits).

The probably longer duration of flow in the Bunter

stream channels, and a much stronger chemical weathering (p. 222) suggest a climate with higher precipitation than that of the Peranera environment: the Peranera and Bunter environments can be considered as having been similar to those of the present-day steppes and savannahs respectively (see footnote on p. 151).

The frequency distributions of channel-fill thicknesses and mudstone and shale unit thicknesses are probably also related to the type of climate.

Fig. 26 shows these relations in histogram form for the Malpas, Peranera, and Bunter formations. The thickness distributions for the Peranera and Bunter deposits compare well; those for the Malpas deposits are quite different. The high percentage of thin channel-fills
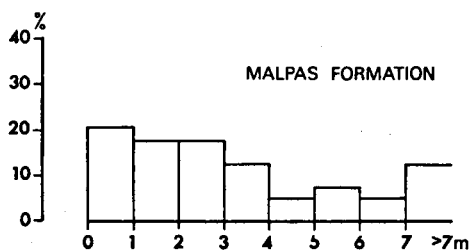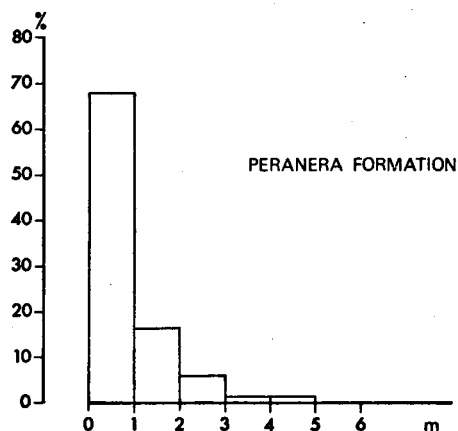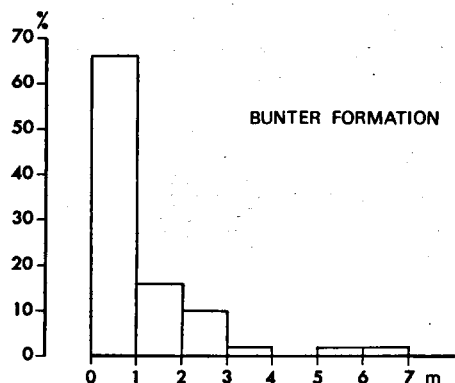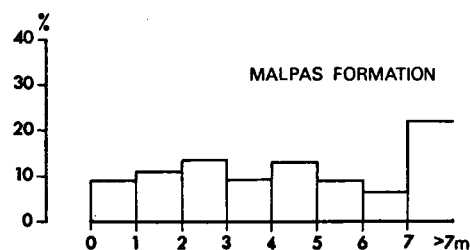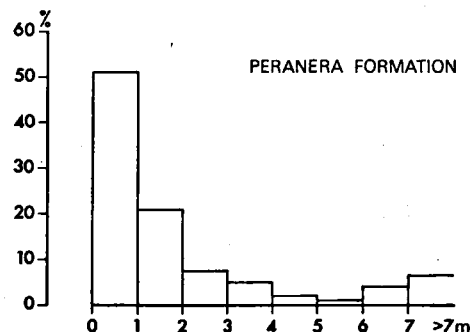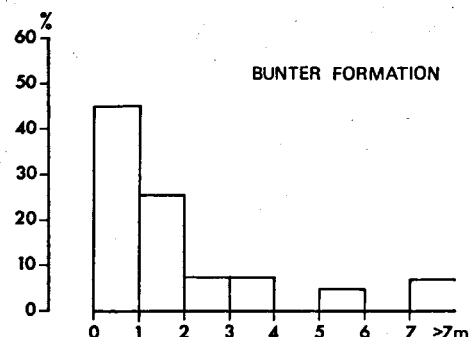
Fig. 26. Percentage frequence distributions of channel-fill and mudstone/shale unit thicknesses for the Malpas, Peranera, and Bunter Formations.

and thin mudstone levels of the Peranera and Bunter Formations agree with the suggested ephemeral flow; the higher percentages of thick channel-fills and mudstone/shale units in the Malpas Formation agree with larger channels of continuous flow and well-developed back-swamps.

Another factor was the morphologic setting resulting from tectonic movements. Relatively high gradients probably account for the fact that the streams in the Aguiró Formation were braided and carried a very coarse bed-load; less steep gradients were probably the cause of the near-equilibrium flow patterns of the Malpas Formation. Similarly, the relatively higher energy streams of the Peranera as compared to the

Bunter streams were probably in part a consequence of somewhat steeper reliefs.

The characteristics of the morphologic setting, in the course of geologic time and given relative tectonic stability, are governed by the climate. The observations of Watkins (1967) on environments in Queensland, Australia, illustrate this point. Watkins emphasized the distinction between what he calls (1) humid fluvial erosion, and (2) arid pediplain erosion (Fig. 27).

Under the conditions of humid fluvial erosion, according to Watkins, run-off water makes a considerable contribution to the ground-water table, which consequently lies at a high level. Ground-water sapping

PROFILE FOR HUMID CONDITIONS

PROFILE FOR ARID CONDITIONS
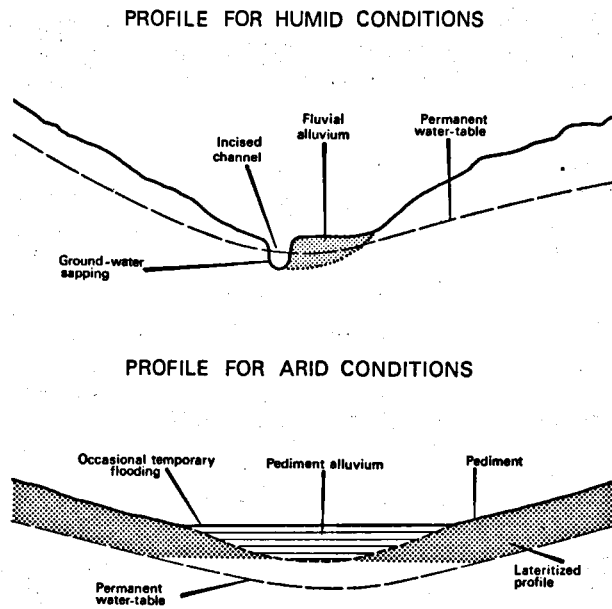
Fig. 27. Relief profiles for humid and arid conditions; after Watkins (1967).

occurs, and linear stream-flows form. Under the arid pediplain erosion conditions, run-off water contributes little to the groundwater table (high evaporation), the ground-water table is low, and no sapping occurs. Consequently, there is no tendency to form linear, continuously water-filled stream channels.

To some extent, the two contrasted types of morphologic setting are recognizable in our deposits. The Aguiró and Malpas Formations would fit into the scheme for the humid fluvial situation, and the Peranera and Bunter Formations into that for the arid

pediplain situation. This is evidenced by the following facts: (1) in contrast to the Aguiró and Malpas deposits, the Peranera and Bunter deposits show evidence of a low position of the groundwater table, and (2) the lateral spreads of the deposits (Aguiró and Malpas: restricted spread; Peranera and Bunter: very wide spread) correspond to the fluvial valley and pediment situation, respectively.

The morphologic settings distinguished could not be fully realized, however, because the region was far from being tectonically stable. There was the marked post-Hercynian relief to start with, and the tectonic phase that led to the unconformity underlying the Bunter Formation. The deposits, moreover, must have accumulated in a region of general, although limited subsidence.

The flat lower boundary of the Peranera Formation cannot be a pediment plane, because it is not erosive (p. 148); the lower boundary plane of the Bunter Formation, however, can be interpreted as a pediment surface (p. 165).

The marked change in climate, which is such an important feature at approximately the Carboniferous-Permian boundary on the northern hemisphere, has been treated by various authors, among them Termier & Termier (1958), Falke (1961), Schwarzbach (1961a, p. 119; 1961b), Feys (1964), and Feofilova (1966). Amongst others, sequences of continental deposits of interest in this context are found in Southern France (Department of Hérault; Stephanian to Lower Triassic; Gèze, 1949; Kruseman, 1966), in the Swiss Alps (the 'Verrucano', same age; von Gümbel, 1888, p. 632; Trümpy, 1966), and in the Cantabrian Mountains (Spain, Polaciones-Comunidad area, same age; Maas, 1968).

CHAPTER IV

PETROGRAPHY AND DIAGENESIS

Mode of presentation

The data pertaining to the petrography and diagenesis of the sandstones are presented in the form of Tables. These Tables are divided into two main sections: (a) a section devoted to the original sediment, subdivided into columns I, II, and III, and (b) a section containing information relating to the diagenetic history of the rock, subdivided into columns IV, V and VI.

(a) Original sediment: columns I, II, and III.

Column I. Gives in succession the composition of the matrix and its percentage in relation to the whole rock (matrix being taken as all the clastic material of less than 30 micron (Dott, 1964), and the average value of the long axes of the 20 largest clastic grains found in the thin section. Together they provide a rough estimate of the grain-size distribution in the rock. Information on the roundness of the grains (scale after

Pettijohn, 1957, p. 59), dimensional orientation of grains, and other primary sedimentary structures follows.

Column II. This column shows the composition of clastic grains larger than 30 micron. The components are arranged in order of frequency. No separate headings are made for monomineralic rock fragments (quartz, feldspar, mica) and polymineralic ones (quartzite, phyllite fragments, etc.).

Column III. This column lists the percentages of the clastic components determined by point counting. An exponent 'e' means that the value is an estimation; a questionmark indicates considerable uncertainty.

(b) Diagenesis: columns IV, V and VI.

Column IV. Gives the authigenic minerals, the term

being taken widely to include all newly formed minerals, overgrowths, and recrystallization products. When numbered 1, 2, etc., a strict order of formation occurs (paragenesis); when not numbered the order of formation is uncertain, or no strict order occurs.

Column V. Gives the percentages of the authigenic minerals.

Column VI. This column gives the textural evidence for authigenesis and for the paragenesis presented in column IV, mentioning crystal habit, pressure-solution, replacement, enclosure, indentation (Glover, 1963), and other textures for each mineral recorded. Bioturbation (in its strict sense an early diagenetic process) and deformation of grains due to compaction are also included.

Diagenesis: the concepts used

The term diagenesis will be used to include all post-depositional changes in sediment prior to metamorphism (Von Gümbel, 1866, p. 28; 1888, p. 334; Walther, 1894, p. 693). The diagenetic environment, thus defined, has its upper boundary at the sedimentary interface; the lower boundary generally is a zone of transition into the lowest grades of regional metamorphism.
In the following the concept of diagenesis as applied in this study is briefly discussed. For a more elaborate treatment of the subject, reference is made to the review of the hand of Dunoyer de Segonzac (1968).

Diagenesis can be studied by two different approaches. In the first, diagenetic changes are related to the depth of burial, the subjects of study being the progress of compaction and concomitant decrease in porosity (Hedberg, 1936; Meade, 1964), the progress of cementation, the evolution of grain contacts (Taylor, 1950), compositional changes in interstitial waters (Von Engelhardt, 1967), mineralogical changes in clay minerals (Scherp, 1963; Stadler, 1963; Dunoyer de Segonzac, 1965; Powers, 1967), and the progressive ranks in coalification correlated with textural and mineralogical changes (Hecht et al., 1962; Kossóvskaya & Shutov, 1964; Esch, 1966; Hedemann & Teichmüller, 1966; Kisch, 1966, 1968, 1969).
The second approach consists of relating diagenetic changes to the sedimentary environment of deposition. It amounts to lateral comparisons, in a time-stratigraphic level, of the characteristics of diagenesis. In a geologic column it means the following, in the vertical sense, of diagenetic changes as the paleoenvironment of deposition, and with it primary lithology, changes. Various Russian authors have studied the diagenetic changes as related to the depositional environment, among them Teodorovich (1946, 1947, 1954; as cited by Larsen & Chillingar, 1967) and Strakhov (1958, 1967).
A pertinent example is given by the relations, carefully worked out by Füchtbauer (1967), between authigenesis and depositional environment in laterally tran-

sitional fluvial and lagoonal Bunter deposits in Germany. Many instances of the close linkage between diagenesis and environment of deposition are treated in the present study.

Sediments accumulate in a wide variety of environments, which can be broadly subdivided into non-marine (eolian, fluvial, lake, glacial), marine (shelf, bathyal and abyssal), and mixed (littoral, deltaic, lagoonal, estuary, and organic reef) (Dunbar & Rodgers, 1958).
In many of these environments the sedimentary components, after their moments of definitive deposition, remain situated in the subaqueous *milieu*. In others, the components accumulate under conditions making long-term or temporary penetration of air, and infiltration of meteoric water into the pore spaces of the sediment possible or even normal. This holds for, amongst others, steppe, savannah and desert environments. Many of the earliest changes then taking place in effect correspond to weathering and soil-forming processes; the reason why they are referred to here as diagenetic lies in the fact that they commonly occurred in generally aggrading sedimentary systems, wherever studied in geologic deposits.
In the processes constituting diagenesis, three main stages are distinguished: (a) early diagenesis, (b) advanced diagenesis, and (c) late diagenesis (epigenesis) (Table III).

TABLE III

DEPTH OF BURIAL			
	Early diagenesis	subaerial	Diagenetic changes directly related to environment of deposition; authigenesis, replacement, bioturbation
		subaqueous	
	Advanced diagenesis		Diagenetic changes leading to lithification; compaction, cementation, replacement
	Late diagenesis (epigenesis)		Diagenetic changes in lithified sediment
	Metamorphism		

Table III. Stages in diagenesis

Early diagenesis refers to processes taking place at and directly below, up to a few tens of metres, the sedimentary interface. Many of these processes result from interaction between the overlying medium (air or water) and the sediment; the products of early diagenesis are therefore closely related to the environment of deposition. Exceptionally, subaerial diagenesis may extend to depths of over 60 m, as in the silcrete-carrying weathering profiles in Australia (Langford-Smith & Dury, 1965). Early diagenesis is used in the same sense, or approximately the same sense, as the terms: 'syngenesis' applied to these processes by some Russian authors (Roukhine, as spelled in French, 1955), 'syndiagenesis' (Fairbridge, 1967), 'early burial stage' (Müller, 1967), and the 'redoxomorphic stage' (Dapples, 1962, 1967).

Early diagenesis applies to both subaqueous and subaerial environments. For cases where the subaerial processes occurred, the term 'subaerial diagenesis' is used. The significance of subaerial diagenesis is similar to that of 'exodiagenesis' (Shvetsov, 1960; cited by: Larsen & Chilingar, 1967), and, in part, to 'hypergenesis' (Dobrovolsky, 1964). Hypergenesis also applies to waterlogged soils. It is doubtful whether the changes taking place in such cases can be easily distinguished, in ancient deposits, from subaqueous early diagenesis when no other evidence is present, e.g. the occurrence of root imprints (seat-earths in coal basins).

Advanced diagenesis refers to the lithification process of the sediment in which compaction and cementation play particularly important roles. Advanced diagenesis starts where the interaction with the overlying medium is reduced, and extends to the depths at which the sediment is largely lithified. In a number of cases advanced diagenesis appears to be ultimately controlled by the primary lithology and environment of deposition (Füchtbauer, 1967; this study). During advanced diagenesis, interstitial waters can still move to a varying degree, enabling reactions between minerals and surrounding pore solutions. With depth the migration of pore solutions is increasingly hampered by the decrease in permeability and porosity resulting from compaction and authigenesis. The pore volume of sandstones rich in easily deformable components such as phyllite, may thus already be strongly reduced at early stages (p. 178). As argued by Von Engelhardt (1959, 1967), the level at which lithification is almost completed determines an important change in the system: the decrease in porosity will halt many reactions between minerals and pore solutions. Between depths of 2000 to 3000 m, clays have practically lost their pore volume (Von Engelhardt, 1959, Esch, 1966). The changes taking place during advanced diagenesis are called 'anadiagenetic' by Fairbridge (1967) and 'locomorphic' by Dapples (1962, 1967). It is to be noted that in particular cases, especially when carbonates are involved, partial lithification may already occur not far below the interface

(early, subaqueous diagenetic calcareous nodules; caliche and silcrete).

The lower reaches of advanced diagenesis are considered by Von Engelhardt (1967) to represent already the transition to metamorphism. Other authors have distinguished a lowermost stage of diagenesis in largely lithified deposits; this stage has been referred to as 'catagenesis' (Strakhov, 1958, 1967), 'epigenesis' (many Russian authors, Packham & Crook, 1960), 'late burial' or 'phyllomorphic stage' (Dapples, 1967), and 'anadiagenesis' (Fairbridge, 1967). In the present study it is referred to as the stage of 'late diagenesis' (Siever, 1959). Late diagenesis begins where lithification is largely completed; it is transitional to low-grade metamorphism characterized by a prevalence of laumontite over heulandite in the zeolite facies, and the critical minerals corresponding to the greenschist facies (Turner & Verhoogen, 1960). According to Turner & Verhoogen (1960), the zone of transition to metamorphism lies at depths of 3000 to 7000 m, the temperature ranging from 100 to 200° C. A temperature of 300° C is considered the value for the on-set of metamorphism by Winkler (1965, p. 136). Schüller (1961) considers diagenesis to end and metamorphism to begin at a depth of about 6000 m, and at a temperature of somewhat more than 200° C. An important criterion for the initiation of metamorphism is the loss of the clastic texture in clastic sedimentary rocks.

AGUIRÓ FORMATION

The petrography and diagenesis of the following rock types will be successively treated:

> sandstones intercalated between conglomerates, (section 1; 35—75 m and 98—130 m intervals)
> mudstones (section 1; 75—98 m interval)
> tuff beds (section 1; 83—88 m interval, sheet I).

Sandstones

Original sediment, provenance. — The data relating to the sandstones are shown in Table IV.
Approximately 60% of the sandstone components con-

TABLE IV

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (phyllite debris) 8% ?	non-calc. phyllite	35			strong moulding of phyllite; closing up of pore spaces
	silty phyllite	26			
Coarse-grained, poorly to moderately sorted	quartz	14	ferric oxides	x	colloform, along fissures
	ser./chlor. quartzite	9			
Quartz, quartzite, chert: angular to subangular	chert	2			
	feldspar	2			
	microsparry limestone	1			
Phyllites: rounded to well-rounded	muscovite	x			
Some quartz milonitic, some lamellar, some showing resorbtion embayments	biotite	x			
	zircon	x			
	tourmaline	x			
Matrix difficult to differentiate from phyllite grains					
Long grains subparallel					

Table IV. Aguiró Formation; sandstones at Aguiró. Mean values after samples 1091, 1092, 868, 1110 (300 points counted in each sample).
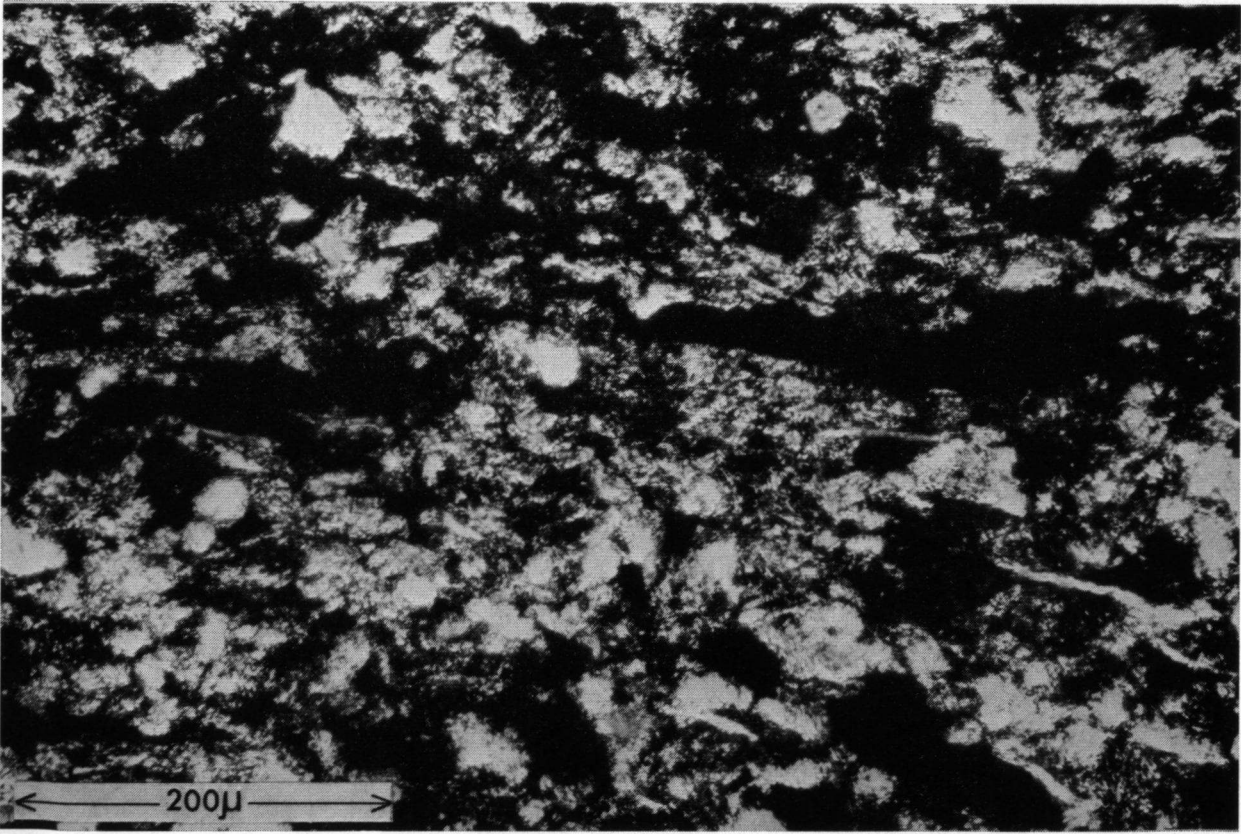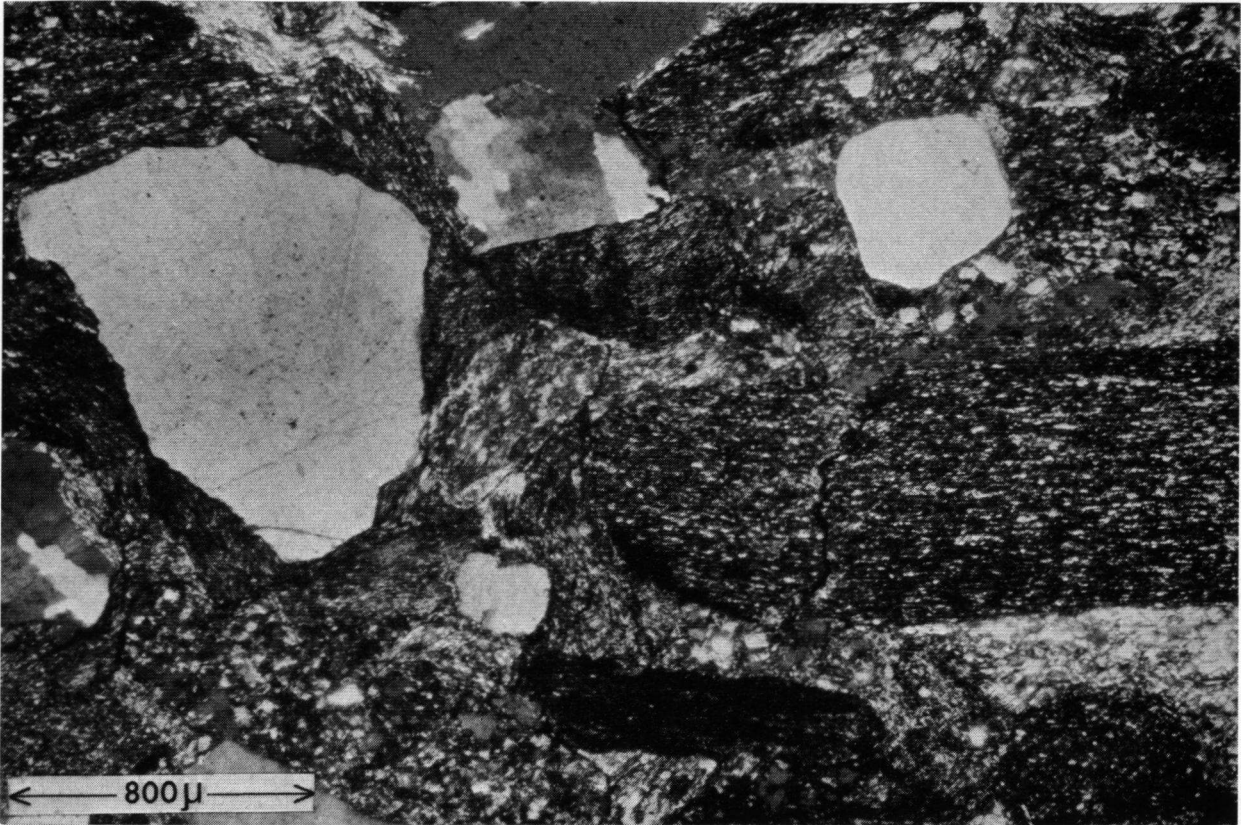
sists of rounded to well-rounded, non-calcareous phyllite and silty phyllite grains having their long axes strictly parallel to their cleavage (Fig. 28). The cleavage in the phyllite grains is a result of the Hercynian tectogenesis (see under Geological Setting, p. 145). Nearly all grains show one direction of cleavage; some show a doubtful second one, and micro-knick zones are present locally.

An investigation of recent river sands from the Mañanet and Flamisell rivers (the occurrence of the Aguiró Formation under discussion lies on the divide of these two rivers (Fig. 1)), shows comparable percentages of fully similar non-calcareous and silty phyllite. Neither in the recent sands nor in the Aguiró sandstones is it possible to differentiate optically between non-calcareous and silty phyllite derived from Devonian and from Ordovician rocks resp. (Appendix, p. 229).

The origin of the sericite/chlorite quartzite grains is as uncertain as that of the non-calcareous and silty phyllite grains, since this type of quartzite occurs in the recent sands stemming from both Devonian and from Ordovician rocks (Appendix, p. 229). Large sericite/chlorite quartzite boulders however, occur in the Aguiró conglomerates at Castelnou de Avellanos (Fig. 3, section 1, sheet I). Quartzite beds of a thickness corresponding to these boulders are not seen in the Devonian at present but they occur frequently in the Ordovician. The argument that the quartzite boulders must have come from the Ordovician is weakened, however, by the possibility that lateral variations in thickness of quartzite beds could have existed in parts of the Devonian now removed by erosion. The quartzite boulders could, theoretically, have been derived from the Devonian, although in the present author's mind the Devonian is a very unlikely source.

The microsparry limestone grains (Table IV) are certainly Devonian components; grains of similar composition occur in the sands of the Peranera River, which has the upper part of its drainage area restricted to the Devonian (Appendix, p. 229). Also, pebbles and boulders of griotte limestone, a type of reddish nodular limestone only found in the Devonian (Nagtegaal, 1969), occur in fairly large numbers near Castelnou de Avellanos (Fig. 3; section 1, sheet I). These observations, added to the fact that the Aguiró Formation unconformably overlies the Devonian, leave little room for doubt that the Devonian acted as a contributor of material to the Aguiró deposits.

Fig. 28. Characteristic coarse-grained Aguiró sandstone, at Aguiró. Rounded, non-undulatory quartz grain at the upper right is probably of volcanic origin. Most of the other quartz grains are undulatory and angular to subangular. Note abundance of phyllite grains and pressure moulding of these grains, making matrix difficult to distinguish from grains. Sample 1110, nicols crossed.

Fig. 29. Aguiró mudstone. Large black shreds are coalified vegetational debris, smaller black spots are ferric oxides after siderite, which widely replaces matrix and quartz grains. Matrix consists largely of phyllite debris and altered fine-grained volcanic material. Sample 1105, plain light.

If one compares the percentages of rock fragments and other constituents in the Aguiró sandstones (Table IV) with those of the sand in the Peranera River (Appendix, p. 229, Table XVI), it is clear that the Aguiró assemblage cannot have been made up by the Devonian alone [1]. Extension of the comparison to include the sands of the Lladorre River (drainage area in the Ordovician) reveals that a combined supply from Devonian and Ordovician cannot be directly visualized either. The significant points of divergence are that the Aguiró sandstones contain too many quartz grains and also contain feldspar and biotite. These three compositional differences may have been determined by the influence of post-orogenic volcanism which was active already in the Westphalian D (see below).

Exclusion of the influence of volcanism does not solve the difficulties, for even if the Aguiró sandstones formed, apart from the volcanic source, from a combination of Devonian and Ordovician supplies they are still too low in such Devonian carbonate-rich components as microsparry limestone, calcareous phyllite, and macrocrystalline calcite fragments (compare Table XVI, Appendix p. 229).

At this point one should realize that there is a limit to the value of comparisons of the kind made here. The composition of recent river sands can be successfully related to the geology of the drainage area, albeit on a large scale (Appendix, p. 229), but too many unknown factors enter in when the relations are applied to an ancient situation.

First, in a fossil sandstone one is looking at fragments of rock masses that in outcrop no longer exist. Lateral lithologic variations, whose character can only be guessed at, may account for otherwise unexplainable compositional differences. In the present case, this influence existed but was probably limited. Lateral lithologic variations are found in the Paleozoic, but in the area under consideration the Paleozoic formations still retain an almost constant gross lithology in the outcrop areas.

A second point to be taken into account is the progression of diagenesis. Although Devonian microsparry limestone fragments occur in the Aguiró sandstones, there is a conspicious absence of calcareous phyllite. As noted in the Appendix (p. 229), this type of phyllite is characterized by varying amounts of replacement calcite; it is otherwise identical to the non-calcareous and silty phyllites. It is conceivable that the replacement by calcite had not yet started when the Hercynian orogenesis came to a close. If the replacement were of a later date, it would follow that, although the Devonian now supplies calcareous phyllite in abundance, it was formerly incapable of doing so. If the replacement really were of a later date, it must have taken place before the Permian, because the Peranera sandstones contain Devonian calcareous phyllite grains (p. 201).

[1] The percentage values are not of equal significance: those of the Aguiró sandstones were arrived at by means of point counting; those of the recent sands, however, were found by means of grain counting.

There seems to be no particular reason, however, why the replacement should have occurred between the Westphalian D and the Permian.

One of the main factors in the type of supply is the paleorelief. Although it must have been pronounced in Westphalian D times (p. 150), it is not known whether it reached the appreciable relief energies of today (Appendix, p. 229). Climate and vegetation are also important factors in this respect. It is possible that these three factors combined in such a way (e.g. moderate relief energies, a climate with sufficient humidity, and the presence of a vegetation cover) as to result in a relatively restricted supply of particulate carbonates as a result of solution during weathering. If all these possibilities are taken into account, we may only conclude, on the basis of the types of rock fragments present, that the Aguiró sandstones probably formed from a combination of Devonian and Ordovician strata, admixed with material of contemporaneous volcanic origin.

Most of the quartz grains in the Aguiró sandstones are polycrystalline and undulatory (Fig. 28). According to Blatt (1967), such grains may derive from a wide range of different rocks; therefore, they have little provenance value. Part of the quartz grain population in the same grain-size range, however, is faintly to non-undulatory, monocrystalline, very clear, and in some cases apparently magmatically resorbed, showing embayments and good rounding (Tröger, 1967; p. 158) (Fig. 28). Such grains occur in minor quantities in the lower part of the section at Aguiró but are more frequent in the upper part. In the upper conglomerate/sandstone unit, rather fresh biotite and plagioclase grains form part of the clastic assemblage. These minerals are also constituents of the tuffs found in the Erill Castell Volcanics and in the Peranera Formation (p. 181, p. 209). It is assumed that in the Aguiró Formation they are likewise derived from tuffs, because *in situ* tuff beds occur also between localities in which sandstone was sampled in the Aguiró section (section 1, sheet I).

These few data suggest that the post-orogenic volcanic activity increased in the course of the Westphalian D and reached a peak with the deposition of the thick sequence of Erill Castell Volcanics (p. 151). Volcanic activity continued, in the form of intermittant deposition, in the Stephanian and in the Permian (p. 153; p. 159).

Biotite and feldspar appear in the sands of the rivers that now drain the granodiorites of the Axial Zone of the Pyrenees (Appendix, p. 229). Their occurrence in the Aguiró sandstones raises the question of whether, in Westphalian D times, the granodiorites could have been exposed in the Aguiró drainage area and have supplied both minerals. As far as the present evidence is concerned, the question may be answered in the negative. The predominant feldspar (25—50% by volume of rock) in the granodiorites is a sodic andesine (30—40% An), followed by microcline (15—20%

by volume) (Mey, 1967, p. 211). No microcline was noted in the Aguiró sandstones, and the plagioclase is more basic than that in the granodiorites (40—50% An).

In average weathering, microcline is more stable than lime-rich plagioclase (Pettijohn, 1956, p. 125). If the granodiorites had been exposed, one would certainly expect some microcline to occur in the sandstones along with the plagioclase. No granodiorite pebbles or boulders were noted in the Aguiró conglomerates, nor do they occur in the Peranera Formation, which overlies the Aguiró Formation and which was probably deposited under conditions of vigorous erosion in a semi-arid climate (p. 163). Only in the still younger Bunter deposits is there direct evidence that the granodiorites were finally exhumed (p. 213).

The content of tuff-derived material varies strongly in the sandstones as well as in the mudstones. One sample collected in the sandstones consists almost solely of tuff material (sample 864). These facts suggest that the tuffs did not form extensive deposits in the drainage basin, which would have led to a more even distribution of tuff components in the sandstones, (compare Malpas sandstones, p. 185), but that tuffs rained down not far from and within the area of accumulation of Aguiró sediment, where they were unevenly dispersed, or were preserved as in the quiet back-swamp environment (see under *Tuff beds*, p. 179).

From the foregoing it follows that the Aguiró sandstones are very immature, both texturally (angular to sub-angular grains of quartz, quartzite, chert) and mineralogically (high content of phyllite). They show, in fact, the same immaturity as the sands now transported in the rivers in the area of study. (Appendix, p. 229).

Diagenesis. — During diagenesis, the Aguiró sandstones remained remarkably inert with respect to authigenesis: no pore-filling authigenic minerals were noted, except for the probably recent introduction of ferric oxides.

These ferric oxides appear in flame-like patterns which locally concentrate along grain boundaries but commonly extend apparently arbitrarily through phyllite grains and phyllite matrix alike. The highest concentration of ferric oxides is found along small fissures and cracks in the rock and along joints. They are therefore assumed to originate from recent weathering processes.

The absence of pore-filling authigenic minerals can be accounted for by:

(1) the particular composition of the sandstones, as expressed in their high content of phyllite, and (2) conditions unfavourable to the generation of early diagenetic products.

The high proportion of phyllite grains present must have hampered the precipitation of cements and other authigenic minerals via purely mechanical processes, i.e. compaction. Probably, moulding of the soft phyllite grains started even at shallow burial stages, which

would have rapidly decreased the pore volume of the sandstones. The passage of pore solutions from which precipitation could have occurred was thus limited at rather early stages, and the sandstones would behave like mudstones and shales with a low porosity and permeability. This may explain the absence of products of advanced diagenesis.

Not all the conditions unfavourable to the formation of products of early diagenesis can be considered here, though one fact may be pointed out. Sandstones deposited in environments sedimentologically comparable to that of the Aguiró sandstones, contain, like those of the Malpas Formation (braided stream facies, p. 157) and those of the Peranera Formation (alluvial fan, p. 163) abundant calcite, presumably formed in response to climatic conditions conducive to the formation of caliche (p. 188, p. 201). If similar climatic conditions had existed during the deposition of the Aguiró sandstones, such calcite could certainly have formed, because the sandstones had a source-area rich in carbonates and the conglomerates carry limestone pebbles and boulders (Devonian, see above).

The absence of the early diagenetic calcite could be an indication, though indirect, that the climate in Westphalian D times was too evenly humid and possibly not sufficiently warm to cause enough evaporation for early diagenetic calcite precipitation.

Among paleoclimatologists and paleobotanists it still is a point of discussion whether the climate of the coal-bearing Westphalian was tropical-humid or temperate-humid (Schwarzbach, 1961, p. 117; Kräusel, 1961, 1964).

The predominance of the highly unstable phyllite grains in the Aguiró sandstones makes it very unlikely that they could have been derived from a region with a tropical-humid climate. In such regions, even with a marked relief, weathering normally extends to depths of 10—20 m over large areas. The presence of the phyllite grains thus speaks for a temperate climate in the Aguiró drainage area (Köppen's Cf climate). Whether the same holds for the accumulation area however, is uncertain, as the distances of transport, nor the elevations of the paleodivides and the accumulation area are fully known.

Mudstones

Some of the mudstones are simply finer grained equivalents of the sandstones just treated. In these types the matrix of the mudstone consists, as far as discernable, mainly of phyllite debris (predominantly sericite, silt-sized quartz, and some chlorite). The debris must have been of sufficiently fine grain-size to permit dimensional reorientation during compaction: the matrix shows good aggregate extinction in thin section (sample 1096).

Most of the mudstones, however, show no aggregate extinction (samples 1098, 1103, 1105). They appear to be largely made up of varying amounts of phyllite debris and, originally, fine-grained volcanic material. The volcanic components may have consisted of glass

dust and fine-grained pumice fragments; now they show complete conversion into a densely intergrown aggregate of kaolinite and microcrystalline quartz. The same alteration products of tuffs are also widely encountered in the Erill Castell Volcanics (p. 181). All mudstone samples investigated contain authigenic siderite; locally, siderite predominates to such an extent (over 90% by volume) that the rock should be termed an ironstone (samples 1093, 1099, 1104). Everywhere, the siderite replaces the mudstone. The crystallization relations of kaolinite and siderite in the Aguiró mudstones are similar to those in the mudstones of the Malpas Formation. A discussion of the latter can be found on p. 190.

A representative sample of Aguiró mudstone is shown in Fig. 29. Vegetational debris, a common component in all mudstones, occurs in the form of irregular coalified shreds. The mudstone proper shows an abundance of quartz of silt-size, and finer-grained phyllite debris admixed with devitrified (kaolinized and silicified) volcanic material. Fine-grained authigenic siderite, making up an estimated 30% (by volume) of the rock, occurs evenly distributed and replaces mudstone matrix and silt-sized quartz grains. The result of the latter process is that the grain boundaries of quartz grains are almost everywhere strongly corroded.

Tuff beds

Only two samples were collected in the tuffs (samples 1097 and 1101). The tuffs are very poorly exposed; the samples in fact had to be dug out, which holds for most of the mudstone samples as well.

The two samples of tuff are similar in appearance. They show many quartz and some andesine/labradorite and biotite crystals 'floating' in an extremely fine-grained groundmass. The crystals are smaller than about 400 micron. The feldspar and biotite are still rather fresh, although some of the biotite is bleached. By volume, the crystals make up an estimated 10%. The groundmass consists of densely intergrown kaolinite and microcrystalline quartz; it probably formed, as suggested in the paragraph on the mudstones, by devitrification of glass dust and pumice fragments. Both samples contain authigenic siderite.

Signs of pedogenesis, which are of frequent occurrence in the Erill Castell tuffs (p. 181), were not noted in the tuffs of the Aguiró Formation.

ERILL CASTELL VOLCANICS

Andesitic tuffs

The Erill Castell tuffs must have accumulated on a hilly land surface, since they overlie fossil slope breccia and contain intercalations of fluvial channel-fills (p. 153).

The composition of the tuffs was investigated by H. de la Roche (Centre de Recherches Pétrographiques et Géochimiques, Nancy, France), who kindly communicated the results of two analyses (Table V). The

TABLE V

constituent	greenish tuff	reddish tuff	average
SiO_2	52.70	58.50	55.60
TiO_2	0.95	0.84	0.90
Al_2O_3	19.75	16.25	18.00
Fe_2O_3	6.94	7.01	6.98
MnO	0.10	0.08	0.09
MgO	1.27	1.29	1.28
CaO	3.99	3.73	3.86
Na_2O	1.86	1.52	1.69
K_2O	2.13	2.07	2.10
Loss on ignition	9.19	7.66	8.43
Total	98.88%	98.95%	

Table V. Composition of Erill Castell tuffs,
after H. de la Roche.

tuffs are apparently of an andesitic composition and may be compared to the overlying sheet of basaltic andesite (Table VI). The MgO, CaO and Na_2O contents are, however, significantly lower; SiO_2, AL_2O_3 and K_2O are somewhat higher, while manganese and iron differ little.

In the average sequence of loss of oxides by weathering, as suggested by Steidtmann (1908), Leith & Mead (1915) and Goldich (1938) (summarized by Pettijohn, 1957, p. 502), CaO, MgO, and Na_2O, taken together, are first carried away, followed by K_2O, SiO_2, Al_2O_3, and iron.

If it is assumed that the analysed tuffs, in their initial composition, differed little from the associated basaltic andesites, their deficiency in certain oxides would agree with the sequence suggested by the authors mentioned. The leaching of MgO, CaO, and Na_2O would have led to a relative increase of the other constituents. However, in order to determine the true nature of the tuffs and the influence of weathering processes, many other analyses will have to be made.

The alteration processes must have been well under way when the entire tuff unit was deposited, since the directly overlying Malpas deposits already contain an abundance of clastic fragments and pebbles of altered tuff (p. 185).

It is a well-known fact that andesitic tuffs alter quite rapidly. An example, mentioned by Hardy as cited by Robinson (1949, p. 415), are the andesitic tuffs of the St. Vincent volcano (eruptions of 1902—03), which within one year were sufficiently altered to support agricultural crops.

An example of the rapid weathering of andesitic tuffs is also found in the North Island of New Zealand. The micromorphologic aspects of the weathering and
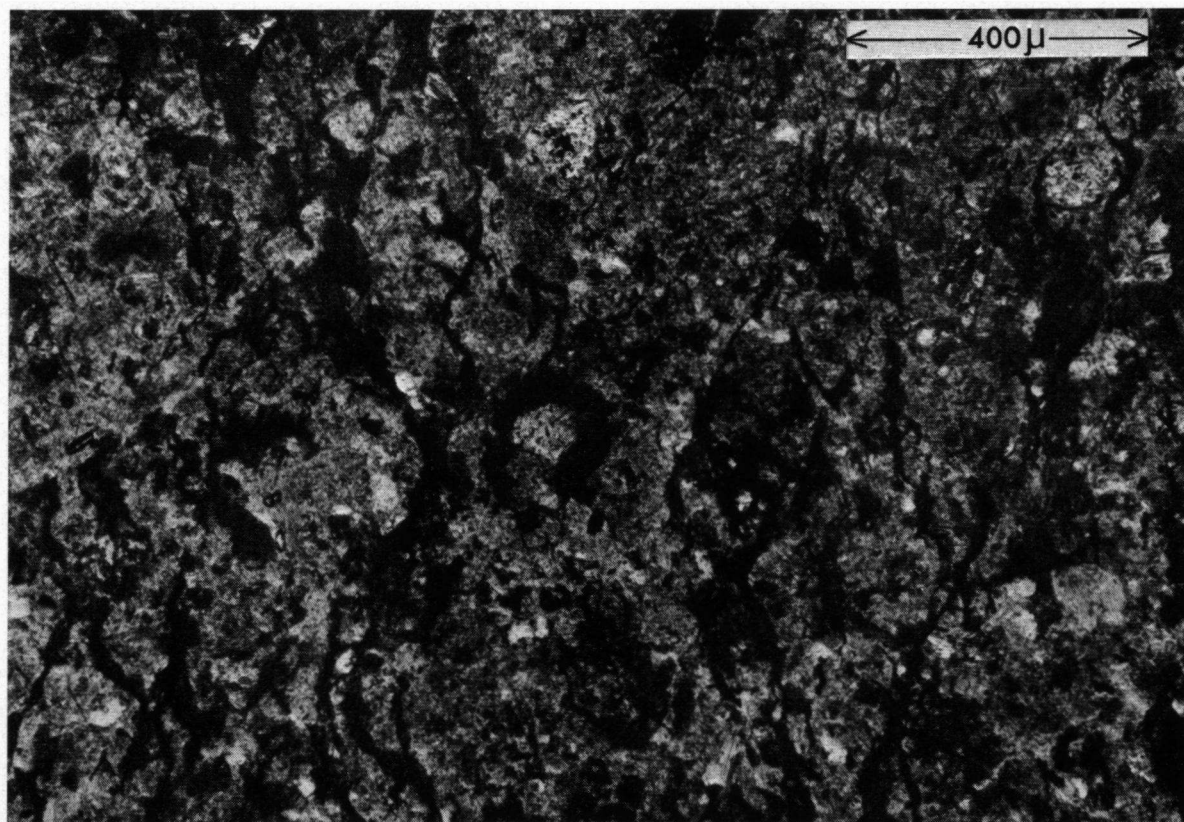
Fig. 30. Medium-grained Erill Castell tuff. The tuff, which originally may have consisted largely of volcanic dust and pumice fragments, is now a dense aggregate of mainly fine-grained kaolinite and microcrystalline quartz. The original clastic nature is vaguely discernable. The network of veins is opaque due to ferric oxides; it probably represents a system of former conducting channels formed in response to pedogenic processes. Stratification is parallel to the long edge of the microphotograph. Sample 1180, plain light.

pedogenetic processes in these tuffs have been studied in detail by Dalrymple (1964), according to whom the alteration of the tuffs appears to have kept pace with deposition. Many of the features noted by Dalrymple, such as local accumulations of colloidal material, the presence of conducting channels running perpendicular to the stratification (Fig. 30), clay coatings, and so forth, can be recognized in the Erill Castell tuffs (very clearly displayed in samples 1170, 1171, 1176, 1177, 1180, 1181). It is noteworthy that these features are far less intense or completely absent in the levels of paroxysmal origin, such as those consisting of graded coarse tuff breccia and those containing many bombs and lappilli (section 2, sheet II).

A detailed discussion of the weathering phenomena and pedogenesis would take us beyond the scope of this study. It will suffice to say that the Erill Castell tuffs show, at well-stratified levels, evidence of rapid weathering (pedogenesis) penecontemporaneous with deposition.

Of the original constituents of the tuffs, only the quartz crystals are clearly recognizable. They are either sharply angular, chip-like, or rounded, commonly showing magmatic resorbtion embayments and microcracks (Fig. 31).

The quartz grains contained in the overlying Malpas deposits exactly fit this description and are of similar size; it is therefore assumed that they derived from the Erill Castell tuffs (p. 185).

The main products of alteration are secondary quartz, kaolinite, and illite. The secondary quartz occurs in two forms, which are the end members of a textural range: (1) microcrystalline textures of varying coarseness, rarely in perfect spherulites as is also the case in the Peranera tuff beds (Fig. 57) or, normally, distributed in vaguely defined patches; and (2) macrocrystalline, mosaic-textured, either vaguely or sharply defined aggregates (Fig. 32). The globular form and microcrystallinity of much of the secondary quartz are suggestive of a colloidal origin (Lebedev, 1967).

Both types of quartz aggregates occur abundantly, as clastic components, in the overlying Malpas deposits (p. 185).

Kaolinite is also found in a microcrystalline form. The microcrystalline kaolinite is commonly intergrown with quartz (macro- and microcrystalline forms); both minerals, together, commonly appear to be pseudomorphs after pumice fragments (Figs. 31, 32).

In sedimentary rocks in which fine-grained crystalline granular quartz or chalcedonic quartz and kaolinite occur in tightly intergrown patterns, it may be difficult to distinguish optically between these two minerals. A convenient method for arriving at a differentiation is a treatment with aniline, in which uncovered thin sections are immersed for 10 minutes in a 0.5% methylene blue solution in water. Quartz and chalcedony do not stain, but the kaolinite is stained blue by absorption of the dye (Mielenz *et al.*, 1950).

After X-ray analyses of a number of representative samples had demonstrated the presence of well-crystallized kaolinite, the aniline treatment was applied to many samples, including

those from the Malpas sandstones and tuffs, Peranera tuffs, and Bunter sandstones.

Microcrystalline kaolinite, interspersed with sericite (illite) also occurs in the form of dense aggregates, large angular fragments measuring up to 2.5 mm (Fig. 31). These aggregates give the impression of being pseudomorphs after feldspars; in outline and size they are similar to the feldspars found in the thick Malpas tuff bed (p. 198) and in the Peranera tuff beds (p. 209). In the Erill Castell tuffs kaolinite also occurs in large crystals (up to 0.5 mm) which, judging again from their habit and shape, are apparently pseudomorphs after mica (Fig. 32, centre). Large crystals of kaolinite are also found in the well-known vermicular habit; these kaolinite crystals may have been fully newly formed.

The strong alteration of the tuffs makes it difficult to define the original mineralogical composition; it seems that the crystal content may have varied from an estimated 3 to 20%, the rest being made up of pumice fragments, glass dust, and accessory glass shreds.

Secondary calcite is rare in levels in the tuffs that are of paroxysmal origin (graded tuff breccia, section 2, sheet II). The well-stratified tuffs, interpreted as having been altered by pedogenetic processes, locally contain secondary calcite. This suggests that the calcite is an illuvial concentrate in paleosol B horizons. There is no evidence of caliche development, however, since textural caliche markers such as encrusting calcite and calcite spherulites (Nagtegaal, 1969) are missing. The replacement calcite present varies from microcrystalline masses to aggregates of anhedral macro-crystals. Staining with alizarin red-S in a cold acid solution (acid concentration between 0.1 and 0.2 N) gives shades of pink. Surface precipitates of Turnbull's blue of varying intensities are produced by treatment with potassium ferricyanide. The results are the same for all samples treated (1172, 1176, 1177, 1178, 1180), from which it follows that the replacement carbonate in the tuffs at Erill Castell is calcite, the greater part being ferroan (Evamy, 1963; Dickson, 1966).

The Erill Castell tuffs show differences in topographic expression. At Erill Castell and along the entire extension of the outcrop area up to Castelnou de Avellanos (Figs. 3, 4 and 5) these tuffs have little resistance to erosion and often show narrow, deep valleys. Just to the West of Aguiró, however, the highest peaks in the local terrain consist of the same Erill Castell tuffs. This surprising phenomenon results from a stronger silicification of the tuffs near Aguiró, as can be established both in the field (the tuffs are chert-like, while at Erill Castell they are friable) and in thin sections. Staining with methylene blue confirms that the tuffs at Aguiró, like those at Erill Castell, contain kaolinite, but have higher silicia contents. To the East of Aguiró, again, the tuffs have little resistance to erosion.

The locally stronger silicification may be related to the paleodrainage pattern. Delvigne (1967) has shown that in the weathering of basic rocks on the Ivory Coast of Africa, silica solutions tend to migrate downslope, leading to silica enrichment in topographic lows and downhill areas. The occurrence of more strongly silicified tuff just at the point where a pronounced paleotopographic low can be indicated (p. 150), suggests that similar processes may have been active in the weathering of the tuffs.

In the two tuff samples collected in the Erill Castell Volcanics near Aguiró (samples 1112 and 1113, section 1, sheet I), the feldspars show much less alteration than at Erill Castell proper, and some are still fresh. The tuffs at Aguiró moreover, show no pedological features.

The tuff samples from Aguiró have a grain-size resembling that of the samples from Erill Castell showing signs of pedogenesis; therefore, they may also represent slowly accumulated deposits. The absence of pedogenesis, and the better preservation of the feldspar in the tuffs at Aguiró agree with the suggested accumulation in a topographic low, the depositional environment possibly temporarily having been that of a shallow lake.

At other paleotopographic lows (at Castelnou de Avellanos and at Erill Castell-Peranera; Figs. 3 and 4), no stronger silification can be inferred from the topographic expression of the tuffs.

Basaltic andesites

Dalloni (1913, 1930) and Misch (1934) already mentioned the occurrence of andesites near the village of Erill Castell (Fig. 4), but as far as the present author is aware, the petrography of the dark greenish-grey lavas has not yet been described.

These lavas are dense aphanitic rocks; only the (altered) mafic phenocrysts can be distinguished with the hand lens. The lava sheet as a whole is strongly jointed, and consequently the rock breaks easily into angular fragments of approximately fist size.

TABLE VI

constituent	L-1186	L-1187	L-1188	average	olivine basalt [x]	basaltic andesite [+]
SiO_2	52.62	55.12	52.22	53.32	51.51	55.83
TiO_2	1.24	1.22	1.35	1.27	1.12	0.84
Al_2O_3	17.22	15.32	17.34	16.63	17.52	18.01
Fe_2O_3	2.54	3.39	1.65	2.53	1.51	2.63
FeO	4.00	6.22	5.58	5.27	6.69	4.07
MnO	0.10	0.06	0.08	0.08	0.15	0.08
MgO	4.78	4.64	4.49	4.64	6.74	5.12
CaO	9.47	7.54	8.59	8.53	8.78	7.40
Na_2O	3.96	2.74	3.54	3.41	3.34	3.64
K_2O	0.44	1.54	0.84	0.94	0.77	1.22
H_2O	2.98	2.22	3.19	2.80	0.64	0.98
P_2O_5	0.23	0.25	0.31	0.26	0.31	0.11
Total	99.58%	100.26%	99.18%		99.08%	99.93%

Table VI. Composition of Erill Castell lavas; samples L-1186 L-1187 L-1188.
   Analysed by Mr. L.F.M.Belfroid, Leiden.
   x after Thayer (1937); + after Williams (1942).

The results of chemical analyses of three samples are shown in Table VI. On the basis of the SiO_2 contents, the rocks can be classed as borderline cases between intermediate and basic rocks (Turner & Verhoogen, 1960, p. 57). Compared to the average compositions of classes of lava computed by Daly (1933), the rocks fall in between the average andesite and basalt, lying somewhat closer to the basalt side.

In composition, the lavas compare well to an olivine basalt (Thayer, 1937) and a basaltic andesite (Williams, 1942) (Table VI). For these reasons, the Erill Castell lava is described here as a basaltic andesite. In thin sections the rocks show a porphyritic, pilotaxitic texture. The groundmass is composed of labradorite microlites, small hypersthene phenocrysts,

Fig. 31. Coarse-grained Erill Castell tuff. Large clast to the left represents a fragment of altered crystal tuff in which the groundmass is converted into fine-grained quartz and kaolinite; quartz crystals showing magmatic resorbtion embayments are preserved, and feldspars are replaced by fine-grained kaolinite, sericite, and microcrystalline quartz (dark, rectangular). Medium-grained tuff to the right contains one phyllite fragment. Sample 1171, nicols crossed.

Fig. 32. Coarse-grained Erill Castell tuff. Light-coloured patches, some of them well-defined but many showing vague boundaries, are probably altered pumice fragments. They now consist of densely intergrown quartz and kaolinite. Many such fragments were supplied to the Malpas sandstones (Comp. Figs. 33, 34, 35, and 36). Just to the right of centre, a mica grain fully altered to kaolinite and darkly stained. Sample 1171, nicols crossed.
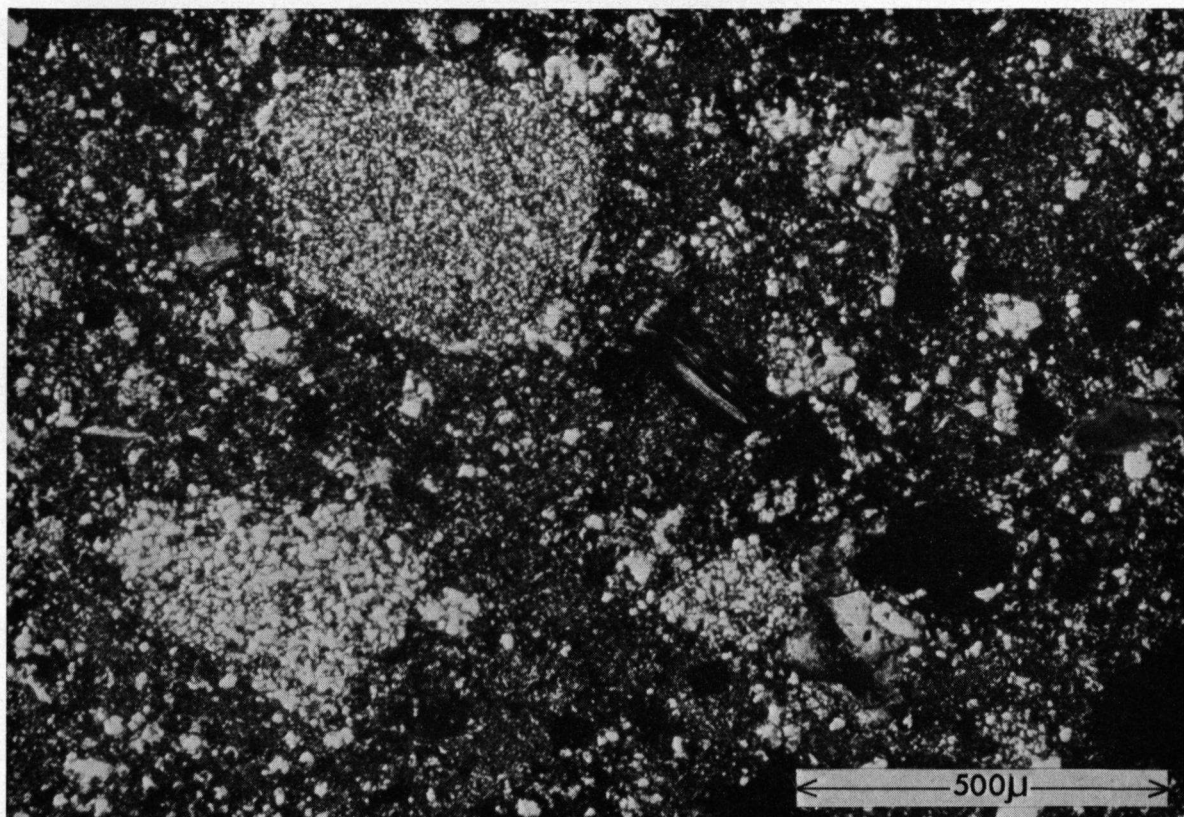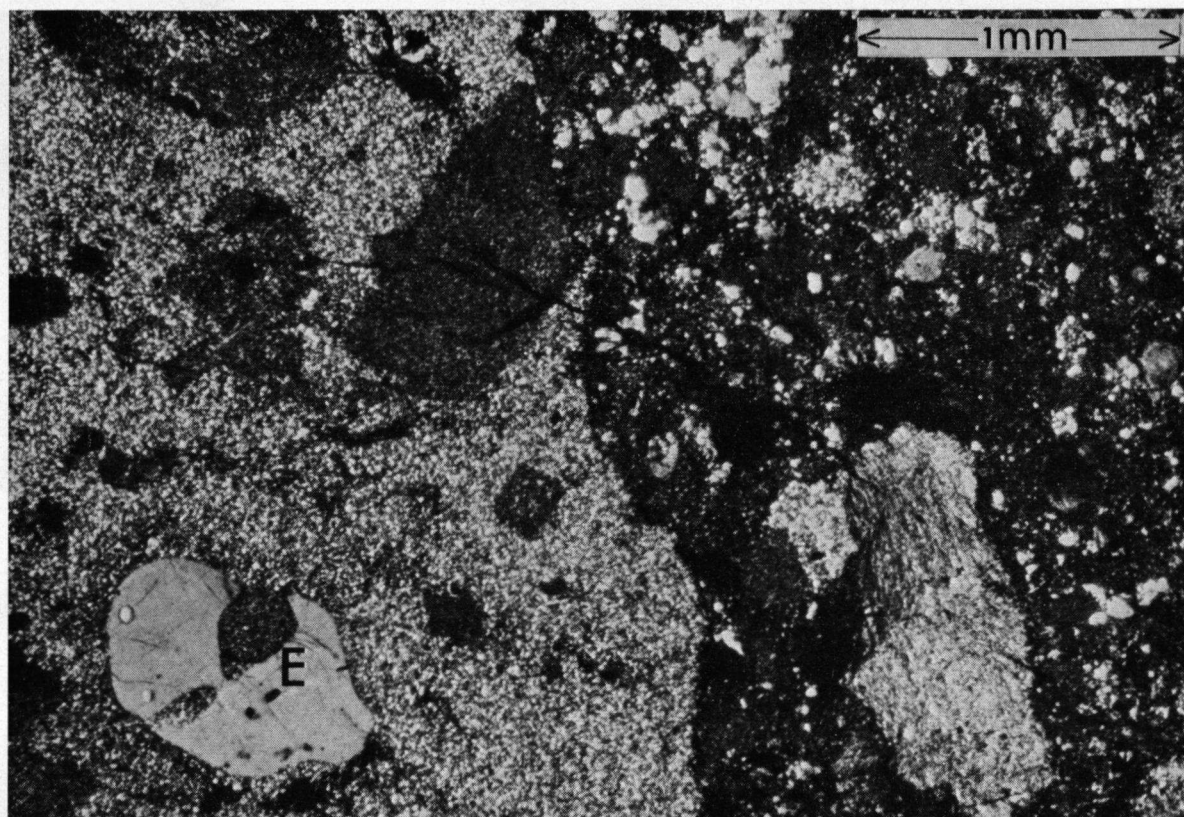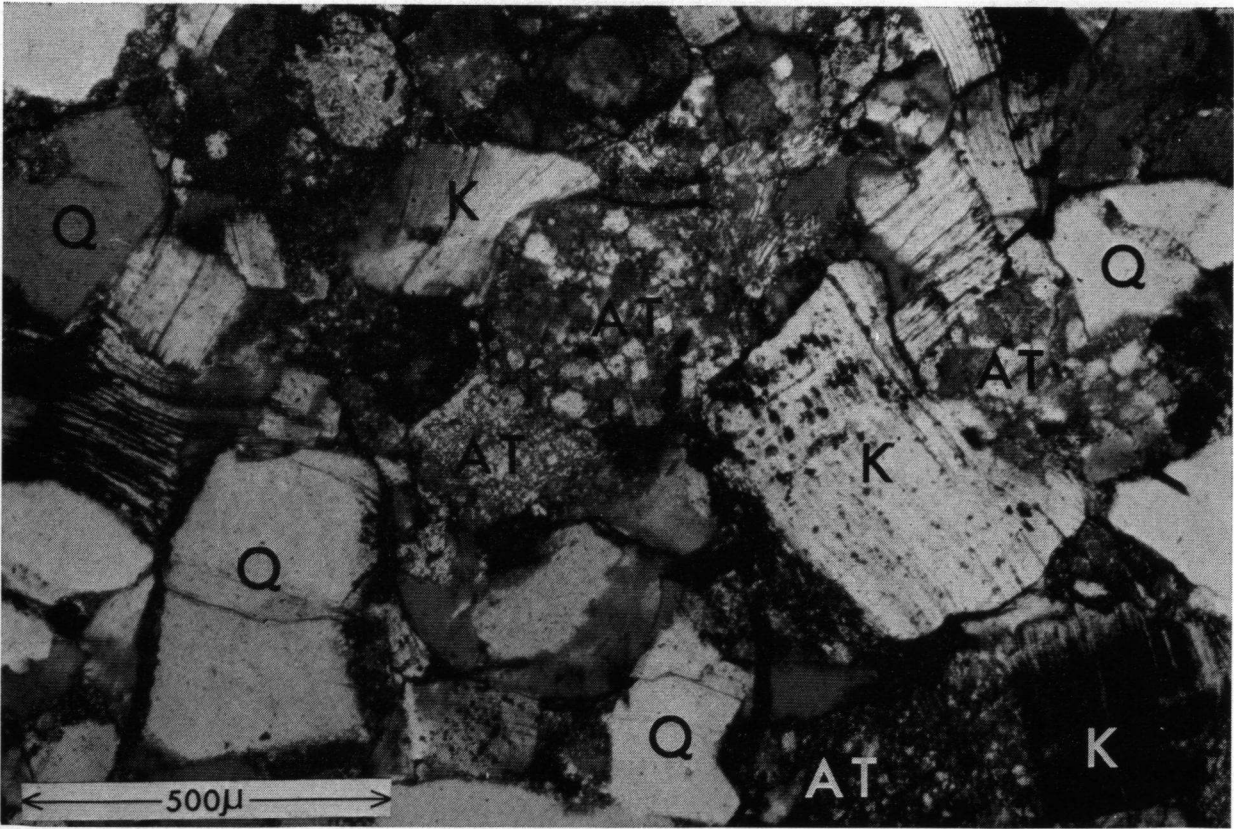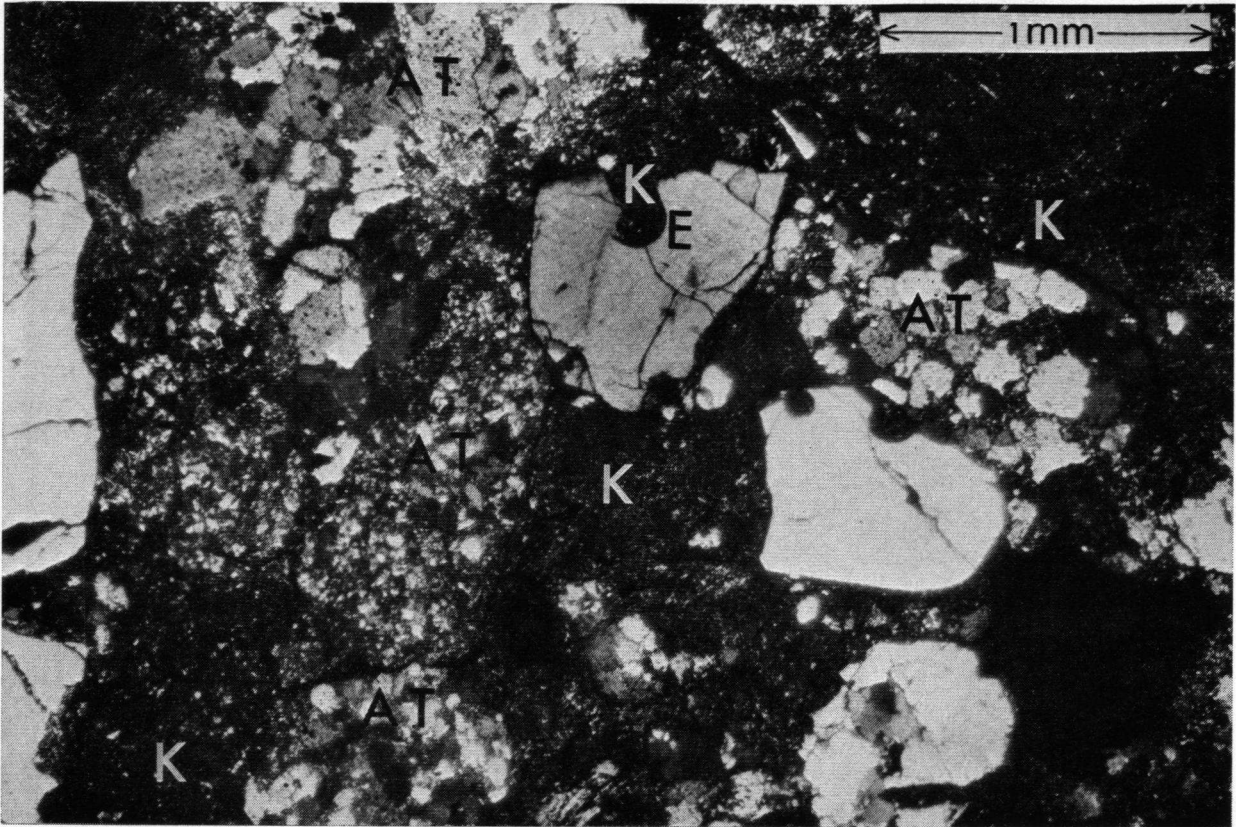
and accessory augite, apatite, biotite, and ore. There are two types of phenocrysts: small labradorites and a fully altered mafic mineral, probably originally olivine.

The labradorite phenocrysts are comparatively rare, making up no more than 1 % of the rock. They are subhedral; some crystals are normally zoned and some show centres occupied by a green, fibrous mineral.

The mafic phenocrysts, which may occupy up to 5 % of the rock, show the characteristic outline of euhedral olivine and the irregular fracture pattern commonly seen in olivine. No olivine is present, however. The phenocrysts now consist largely of antigorite accompanied by magnetite and hematite. In some samples chlorite and a carbonate are also present as alteration products.

MALPAS FORMATION

The petrography and diagenesis of the following rock types will be successively treated:

    sandstones,
    mudstones, sideritic mudstones, and siderite beds
        (ironstones),
    chert (at 89 m, section 3),
    carbonate beds (at 23 m, section 3, and in section 5),
    tuff bed (39—71 m interval, section 3; 30—59 m
        interval, section 4, sheet III).

Fig. 33. Coarse-grained, moderately sorted Malpas sandstone; meandering stream environment. The majority of the grains consist of altered Erill Castell tuff (AT), closely packed. All quartz grains are cracked; two grains show pronounced magmatic resorbtion embayments (E). Fine-grained kaolinite (K) occurs in the embayments and as a matrix (recrystallized) between the grains. (Comp. Figs. 31 and 32.) Sample 1150, nicols crossed.

Fig. 34. Coarse-grained, moderately sorted Malpas sandstone; meandering stream environment. The grains are quartz (Q), kaolinite crystals (K), and altered Erill Castell tuff (AT). The kaolinite crystals have grain-sizes similar to those of the other grains; they could be clastic. Sample 1189, nicols crossed.

Sandstones

The Malpas Formation is everywhere underlain by the Erill Castell Volcanics (Figs. 4 and 5). As will be presently shown, the composition of the detrital components of all sandstones in the Malpas Formation reflects a constant source located in the Erill Castell Volcanics.

The paleocurrent directions of the Malpas Formation point to a source area located in the North (p. 158). Since the fluvial deposits at a lower stratigraphic level than the Malpas Formation (Aguiró Formation) consist largely of pre-Hercynian Paleozoic rock fragments derived from the North (p. 175), the conclusion may be drawn that the Paleozoic to the North was temporarily shielded by a cover of Erill Castell Volcanics, which prevented supply from the underlying rocks. The cover of Erill Volcanics must have been largely removed by the time the deposits belonging to the Peranera Formation were formed, because the Peranera sandstones again contain pre-Hercynian Paleozoic rock fragments in abundance (p. 199).

On the basis of the type of diagenesis in the Malpas sandstones, it is possible to distinguish microscopically between the sands deposited in continuously water-filled, largely meandering streams and those associated with the conglomerates deposited in ephemeral braided streams.

Sandstones deposited in meandering streams. — The composition of these sandstones (mean values of three representative samples) is shown in Table VII. Most of the quartz grains are angular, and some are merely chips; they commonly show micro-cracks and are either undulatory or non-undulatory. Magmatic resorption embayments (Tröger, 1967, p. 158) are common; they are most frequent on the non-undulatory grains (Fig. 33). Some grains show good over-all rounding in addition to embayments.

There can be little doubt that these quartz grains are of volcanic origin. As fully similar grains occur in the underlying Erill Castell Volcanics (p. 181, Fig. 31),

TABLE VII

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (kaolinite mud; recryst.) 13%?	altered Erill Castell tuff	45	kaolinite crystals	3ᵉ	pseudomorphs (?); newly formed
Max. grain-size: 4.5 mm	quartz	27	kaolinite/quartz	13	tightly intergrown, main cement, largely fine-grained mosaic
	kaolinite crystals	7ᵉ			
Quartz: angular; some rounded, cracked, resorbtion embayments	dark volcanic rock	1	siderite	4	subhedral crystals in aggregates
	non-calc. phyllite	x			
Altered tuff fragments: subangular to well-rounded	zircon	x	illite	x	some intergrown with kaolinite crystals
Phyllite: rounded	coalified plant debris	x			
Apparently random orientation			ferric oxides	x	in fissures; impregnating kaolinite (recent weathering)
					compaction moulding of altered Erill Castell tuff

Table VII.   Malpas Formation; sandstones deposited in meandering streams.
        Mean values after samples 1150, 1189, 1179 (300 points counted in each sample; little difference in coarseness).

it is assumed that the majority of the quartz grains in the Malpas Formation derived from the Erill Castell Volcanics, though an unknown amount may have originated from contemporaneous volcanism.

The majority of the clastic grains consist of crystalline granular quartz intergrown with varying amounts of commonly finer grained kaolinite (Table VII). The texture within these grains varies considerably in coarseness (Figs. 33 and 34). Both the texture and the composition of the grains are similar to those of the altered Erill Castell Volcanics; it is therefore assumed that they were, like the quartz grains, derived from this source. Some of the clastic grains in the Malpas sandstones could be simply the altered pumice fragments as found in the Erill Castell Volcanics (p. 181; Figs. 31 and 32).

The grains must have already consisted of interlocking crystals of quartz and kaolinite when they were deposited, as shown by clastic grain boundaries which arbitrarily transect the mosaic pattern. Most pebbles occurring in the braided river deposits near the top of section 4 consist of the same material. These findings show that the Erill Castell tuffs were already altered (silicified, kaolinized) at the time when they contributed material to the Malpas Formation. This agrees with the suggested penecontemporaneous alteration (pedogenesis) of the tuffs (p. 180).

The large, single crystals of kaolinite present a problem, since it is impossible to determine for all the individual crystals whether they were newly formed *in situ,* deposited as clastic monocrystalline kaolinite grains, or were formed, as pseudomorphs, from pre-existing grains (Fig. 34). There is evidence that at least the first two of these possibilities were realized in the Malpas sandstones.

The textural relations as illustrated in Fig. 35 suggest that the greater part of the kaolinite crystal in the centre of the figure developed *in situ.* There are also kaolinite crystals enclosing one or two small, presumably clastic, quartz grains. In relation to the total number of single kaolinite crystals, these cases are not uncommon (an estimated 3%, Table VII).

The deposits that acted as a source for the Malpas sandstones (Erill Castell Volcanics, and probably to some extent Malpas mudstones and shales as well — by 'autodigestion' through the eroding Malpas streams) contain kaolinite crystals of a size similar to those found in the Malpas sandstones. It is therefore possible that the kaolinite crystals were derived, as clasts, from these deposits.

Incidental instances of recent fluvial transport of sand-sized kaolinite crystals are found in both samples of river sand collected in the Peranera River (Appendix, p. 229). Although these samples were expected to consist of a pure Devonian rock fragment assemblage, they proved to be contaminated with material coming from the Erill Castell Volcanics. These Erill Castell components exactly fit the Malpas detritogeneous assemblage; among them are sand-sized kaolinite

crystals originating from the tuffs (Appendix, p. 229; Fig. 71). Some kaolinite crystals which seem to be worn and rounded by transport (not illustrated) are indeed found in the Malpas sandstones. The rounding, however, does not constitute direct evidence of the transport of kaolinite crystals, because it could equally well be an inheritance of the grains after which the kaolinite crystals could be pseudomorphs. Rounding, moreover, is not a neccessary condition for fluvial transport, since the kaolinite crystals transported in recent river sand are angular (Appendix, Fig. 71).

Since fluvial transport of sand-sized kaolinite crystals has been shown to be possible, pseudomorphism of kaolinite after feldspar or mica *in situ,* if it occurred at all, is impossible to demonstrate. No transitional stages from feldspar or mica to kaolinite crystals were observed. If some kaolinite crystals in the Malpas sandstones are pseudomorphs after feldspar, the latter mineral must have come mainly from contemporaneous volcanism, since there is reason to assume that the feldspar in the Erill Castell Volcanics had altered into fine-grained kaolinite before the deposits supplied material to the Malpas Formation (p. 181; Fig. 31).

Various kaolinite crystals show a light brownish tint and are pleochroitic, probably as a result of adsorbtion of humic acids and iron oxides (Kimpe, 1967).

The main cement of the sandstones deposited in meandering streams is kaolinite showing a fine-grained, crystalline granalur texture. It is normally accompanied by small amounts of secondary microcrystalline quartz and chalcedony, both of which are intergrown with the kaolinite (Fig. 35). The growth relations seem to indicate that the minerals formed simultaneously. According to Shelton (1964), quartz is slightly unstable under conditions suitable for kaolinite formation. Indeed, some replacement, albeit limited, of quartz by kaolinite can be observed in the Malpas sandstones (Fig. 35). Still,

Fig. 35. Coarse-grained, moderately sorted Malpas sandstone; meandering stream environment. The relationship of the two quartz grains (Q) and the central kaolinite crystal (K) show that the greater part of the kaolinite crystal must have grown *in situ*. This is also shown by the fact that quartz is slightly replaced by the kaolinite (R). Altered tuff fragment (AT) in the upper left. Fine-grained kaolinite (K), intergrown with quartz (Q) probably represents recrystallized kaolinite mud matrix. Sample 1151, nicols crossed.

Fig. 36. Coarse-grained, moderately sorted Malpas sandstone; braided river environment. The grain at the left has been entirely replaced by sparry ferroan calcite (FC) which is intergrown with ferric oxide (FeOx). Ferric oxide also occurs directly to the right of centre. The ferric oxide probably formed after siderite, which, in a partly oxidized state, occurs in similar grains in other parts of the thin section. The two grains to the right consist of altered Erill Castell tuff (AT). Compare with Figs. 31 and 32. The encrusting rims are made up of fibrous ferroan calcite (FC); sparry ferroan dolomite (D) fills the remaining pore space. Sample 1158, nicols crossed.
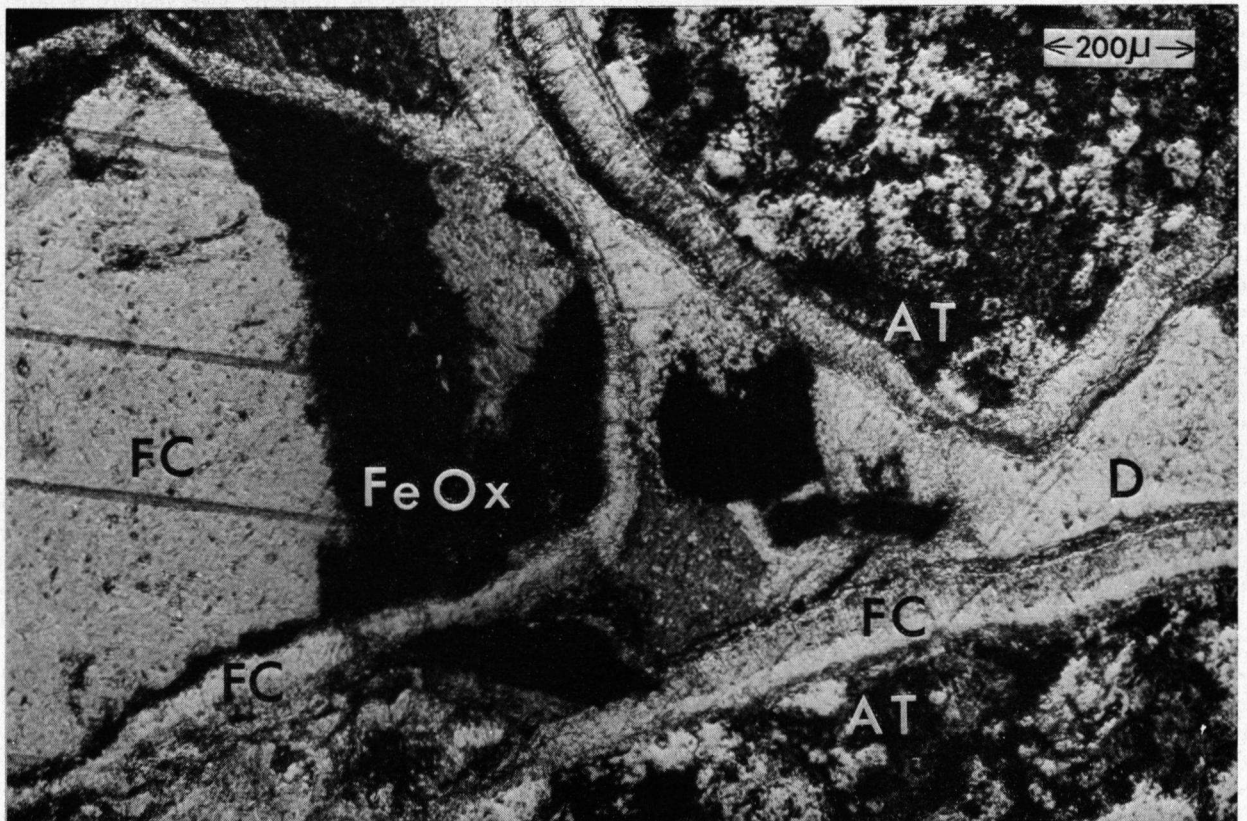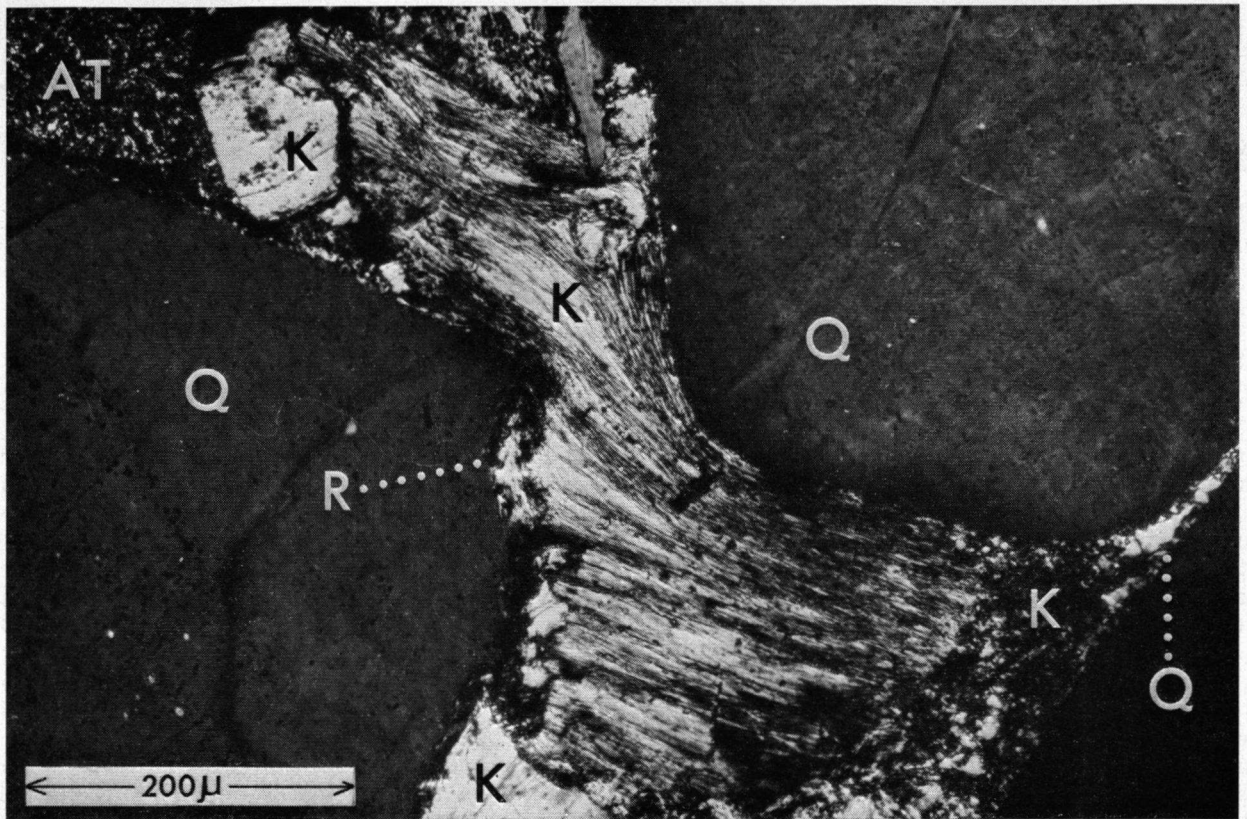
TABLE VIII

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix: kaolin. mud, largely replaced by ferroan calcite	altered Erill Castell tuff	15	ferroan calcite	46	replacing clasts and matrix.
	quartz	3	siderite	1	subhedral
Max. grain-size 2mm	kaolinite crystals	3			
	feldspar	3	ferric oxides	x	after siderite
Other textural properties: see Table VII	biotite	2			
	clay galls	2	ferroan calcite	23	fibrous columnar; encrusting
	dark volc. rock	x			
	phyllite	x	ferroan dolomite	2	sparry pore fills

Table VIII. Malpas Formation; sandstone deposited in braided stream, sample 115s (300 points counted).

the dense intergrowths of the two minerals indicate that the stability ranges should largely overlap.

It is not certain whether the kaolinite cement is a true cement (precipitated from solution or from gels in open pore spaces) or represents a recrystallized kaolinite mud of detrital origin. The latter is probably the case, because no other mud matrix is apparent in the sandstones.

At points where the cement borders clastic grains consisting of altered Erill Castell tuff, it often becomes very difficult to distinguish the detrital grain optically. In these cases, staining with methylene blue is helpful (p. 181); it commonly brings out the outline of the clastic grain quite clearly through differences in kaolinite content.

Subhedral and anhedral siderite crystals occur in small aggregates in nearly all the Malpas sandstones. Growth relations indicate that the major part of the volume now occupied by siderite was acquired by replacement. The same relations indicate that much of the siderite must have formed *after* the apparently authigenic large kaolinite crystals and *after* the larger part of the fine-grained kaolinite/quartz cement.

Accessory iron sulfides, among them pyrite, marcasite, and chalcopyrite, were noted in various sandstone samples. These minerals are far more abundant in the mudstones and shales, however.

Sandstones deposited in braided streams. — The composition of the detrital grains of these sandstones is similar to that of the sandstones just treated; the difference lies, as previously noted, in the type of diagenesis.

Ferroan calcite (lacking in the sandstones deposited in meandering streams) forms the main cement. Three phases of carbonate crystallization can be distinguished (Table VIII).

The first phase is that of replacement. Numerous grains are totally replaced by a single ferroan calcite crystal (pseudomorphs after quartz grains, kaolinite crystals, and grains composed of altered Erill Castell tuff), others partly so. Ferric oxides occur intergrown with the replacement calcite (Fig. 36).

The second phase of carbonate crystallization consists of encrustation by calcite. Numerous individual grains

and aggregates of grains, either previously replaced by calcite (first phase) or still showing their original composition, are entirely surrounded by calcite crusts (Fig. 36). Some of these crusts are composed of columnar and wedge-shaped calcite crystals; others show a dense fibrous structure apparently consisting of calcite whiskers (Nabarro & Jackson, 1958). The crusts are locally dolomitized (third phase, see below). In all crusts the calcite crystals are lengthwise oriented perpendicular to the surface on which growth took place.

The thickness of the calcite crusts in the sandstones varies from a few microns to over 3 mm. Commonly, the thickness of the crust proper also varies; in the majority of cases the thickest part occurs on the upper surface of a grain. This feature is not an infallible criterion for distinguishing top and bottom: the thickest part of a crust has also been found facing in all the other directions.

Many of the crusts have a laminated appearance (Fig. 36). This is brought about by parallel rows of inclusions of tiny aggregates of microcrystalline calcite, or, occasionally, by new generations of calcite crystals. In most cases, however, the calcite crystals extent over almost the entire thickness of a crust.

The third phase of carbonate crystallization is that of sparry dolomite filling remaining open pore spaces (Fig. 36).

The lateral equivalent of the braided stream deposits in section 4, i.e. the coarse sandstones at 221—231 m in section 3, also contain ferroan calcite. In these sandstones, however, the calcite is mainly of the replacement type (first phase).

The deposits directly overlying the calcite-bearing sandstones (samples 1161, 1162, 1218) contain no calcite; they are similar in composition to the mudstones and shales underlying the calcite-bearing sandstones: the calcite-bearing sandstones are thus neatly sandwiched between non-calcitic beds.

These facts suggest that the calcite in the sandstones did not result from some form of carbonate metasomatism during advanced diagenesis, or from a redistribution of calcite from calcareous rock fragments during advanced diagenesis, since the original composition of the sandstone did not include such fragments. It may therefore be concluded that the calcite

was introduced during early stages in diagenesis, which also agrees with the loose packing of the grains and the fact that commonly the crusts are continuous wherever stains are in contact (Fig. 36).

On the basis of this conclusion, the braided stream deposits may be compared with early calcite-cemented sands and gravels in recent stream deposits as described, for example, by Mosely (1965) from wadis in Libya and by Ruhe (1967) from deposits in southern New Mexico. In both cases the deposits are associated with widespread occurrences of caliche. The results of a separate study led the present author to conclude that the microtextures displayed by the calcite in the Malpas braided stream deposits are similar to those of the calcite in recent caliche on the Spanish East Coast (Nagtegaal, 1969).

On these grounds it is suggested that the calcite in the Malpas sandstones formed under climatic conditions conducive to the formation of caliche. As is generally known, caliche forms in areas of low to moderate, seasonal rainfall and high temperatures. The occurrence of calcite in the level of conglomeratic sandstones near the top of the Malpas Formation therefore probably corresponds with a temporary change towards a more seasonal and drier (and warmer?) climate. The very change in sedimentary facies (from largely meandering stream types to braided stream types) may also result from the change in climate.

The calcite-cemented braided stream deposits announce the profound change to the generally semi-arid climate of the overlying Peranera Formation (p. 228).

The origin of the abundant calcite can hardly be
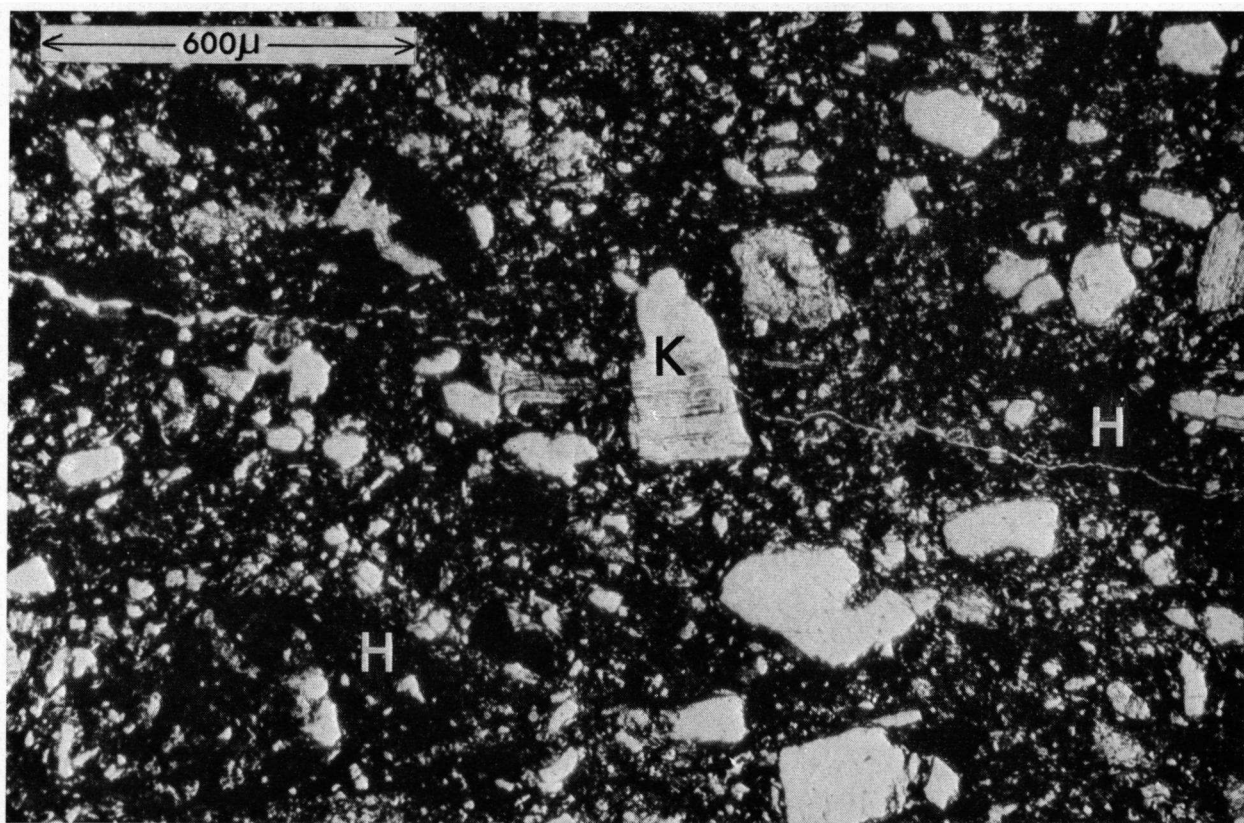
Fig. 37. Tuffaceous mudstone; Malpas Formation. Most of the larger fragments are kaolinite crystals (K). The mudstone matrix is almost completely opaque, due to dense colloform masses of hematite (H). Sample 1119, plain light.

sought, as already mentioned, in the sandstones proper, since these show the same (non-calcareous) composition as all the other Malpas sandstones. Therefore, it may be assumed that the calcite was deposited, by means of evaporation, from solutions transported by the run-off water and possibly also from air-borne calcareous dust.

The evaporation of, and the precipitation of $CaCO_3$ from a carbonate-rich river water may strongly increase the $Mg^{2+} : Ca^{2+}$ ratios in the water (Hut-

chinson, 1957, p. 665). It seems possible that the relative increase of Mg^{2+} after the precipitation of the calcite (and gypsum?) caused the interstitial water to become so enriched in Mg^{2+} that the partial dolomitization of the calcite and the precipitation of dolomite could take place. The sequence in authigenic products ending with the ultimate formation of dolomite would thus find a logical explanation. But Mg^{2+} could also have been derived from the clastic volcanic components contained in the braided stream deposits proper.

Mudstones, sideritic mudstones, and siderite beds

Silt- and sand-sized particles in the mudstones have the same composition as those in the sandstones. The matrix is difficult to determine optically, but probably consists mainly of debris of altered tuffs (from Erill Castell Volcanics).

The mudstones or colluvial tuff (the difference is difficult to distinguish) at the base of the formation are a dark yellowish-brown, in contrast to all the other mudstones in the formation (which are medium grey to black), and they contain a large amount of kaolinite and some quartz grains. The brownish colouring is given by hematite, which occurs in dense masses often rendering the matrix opaque (Fig. 37). The hematite replaces both matrix and larger grains, and must therefore have formed authigenically. The siderite beds in the Malpas Formation all show superficial oxidation to ferric oxides. The hematite in the basal mudstones, however, cannot be the result of present-day weathering of a possible mudstone siderite component, since even the freshest samples show hematite in abundance. The formation of the ferric oxides must therefore have taken place during the Stephanian, which is also indicated by the fact that the brownish mudstones form a distinct and continuous level in the field, extending at least from section 3 to section 4 (Fig. 4). Where the basaltic andesites are overlain by the brownish mudstones, they are strongly altered into a greyish-orange to light brown crumbly rock.

The properties of the dark yellowish-brown mudstones as well as the alteration of the basaltic andesite are interpreted as being the result of a period of deep weathering in which ferric oxides were formed and concentrated in soil profiles (pedalfers).

It is largely a matter of taste whether the level of weathered rock is considered to be the top of the Erill Castell Volcanics or the base of the Malpas Formation. If a sharp erosional break were visible, the former view would certainly deserve preference. No such break occurs, however; sedimentation, though temporarily having been slowed down (*in situ* weathering) or locally temporarily interrupted by erosion (fluvial channels), appears to have been largely continuous. The profound change in sedimentation from pyroclastics to fluvial deposits is shown by field and petrographic evidence to have been gradual, and may be visualized as having taken place in an area where fluvial sedimentation slowly took over from deposition of pyroclastics. Only once again did deposition of pyroclastics become fully dominant, recorded by the thick bed of tuff in the lower part of the Malpas Formation (39—71 interval, section 3; 30—59 m interval, section 4, sheet III).

Kaolinite is the main component of most of the mudstones and shales. In the mudstone matrices it has an extremely fine-grained texture and is intergrown with microcrystalline quartz. The mudstone matrices are identical to the matrices of the sandstones deposited in meandering streams (see above). It is assumed here too that the kaolinite was transported as a kaolinite mud, derived from the already altered Erill Castell Volcanics. Recrystallization and/or further growth of the kaolinite must have resulted in the present dense, fine-grained mosaic texture. Larger kaolinite crystals showing a 'book' and a vermicular habit, occur in most mudstones and shales. True kaolinite tonsteins ('Kristalltonsteine', 'Graupentonsteine'; Schüller, 1951; Guthörl et al., 1956), were not found however.

With the exception of the basal mudstone level, the authigenic products in the mudstones are similar to those in the sandstones. Siderite occurs, however, in far greater amounts and wider textural variety.

All the textural evidence indicates that the siderite formed largely by replacement of other minerals (Fig. 38). Where siderite replaces kaolinite, it sometimes follows the basal cleavage of the mineral (Fig. 39).

The textures displayed by siderite are: (1) anhedral crystals in dense, 10—200-micron mosaics (Fig. 38), (2) evenly distributed, subhedral up to 300-micron crystals, and (3) spherulites with a diameter of up to 1.5 mm, either closely packed or suspended in the mudstone (Fig. 40).

In some siderite bed samples the individual crystals in a mosaic pattern show rows of opaque inclusions aligned parallel to rhombohedral faces. In the centre of such crystals, the crystal faces are flat; going toward the peripheries of the crystals, however, they become more and more curved outward, up to an almost perfect concentric oval structure (Fig. 41). The whole remains one crystal, since it extinguishes simultaneously under crossed nicols. This feature is interpreted as an increasing tendency of crystal growth to take place in all directions; it probably points to a relatively rapid rate of crystallization.

Most of the siderite is stained with dark reddish-brown ferric oxides, probably mixtures of goethite and hematite. In hand-specimens, some siderite samples show the orange colours typical of lepidocrocite.

Because nearly all growth relations of siderite are those of replacement, it is concluded that the beds consisting almost entirely of siderite (ironstones; sections 3, and 4) were formed largely by replacement. It seems very likely, however, that finely disseminated

Fig. 38. Malpas siderite bed (ironstone). The siderite bed was formed by replacement of a sandy mudstone. Crystalline granular siderite (S, larger part of the microphotograph) moderately stained yellowish-brown, entirely replaces the mudstone matrix and partially replaces a detrital quartz grain (Q) and a kaolinite crystal (K). Sample 1207, nicols crossed.

Fig. 39. Malpas siderite concretion (ironstone). The concretion formed by replacement of a medium-grained sandstone. The microphotograph shows a kaolinite crystal (K) partially replaced by siderite, which is stained yellowish-brown (S). Replacement occurred along the basal cleavage of the kaolinite crystal. Sample 1199, nicols crossed.
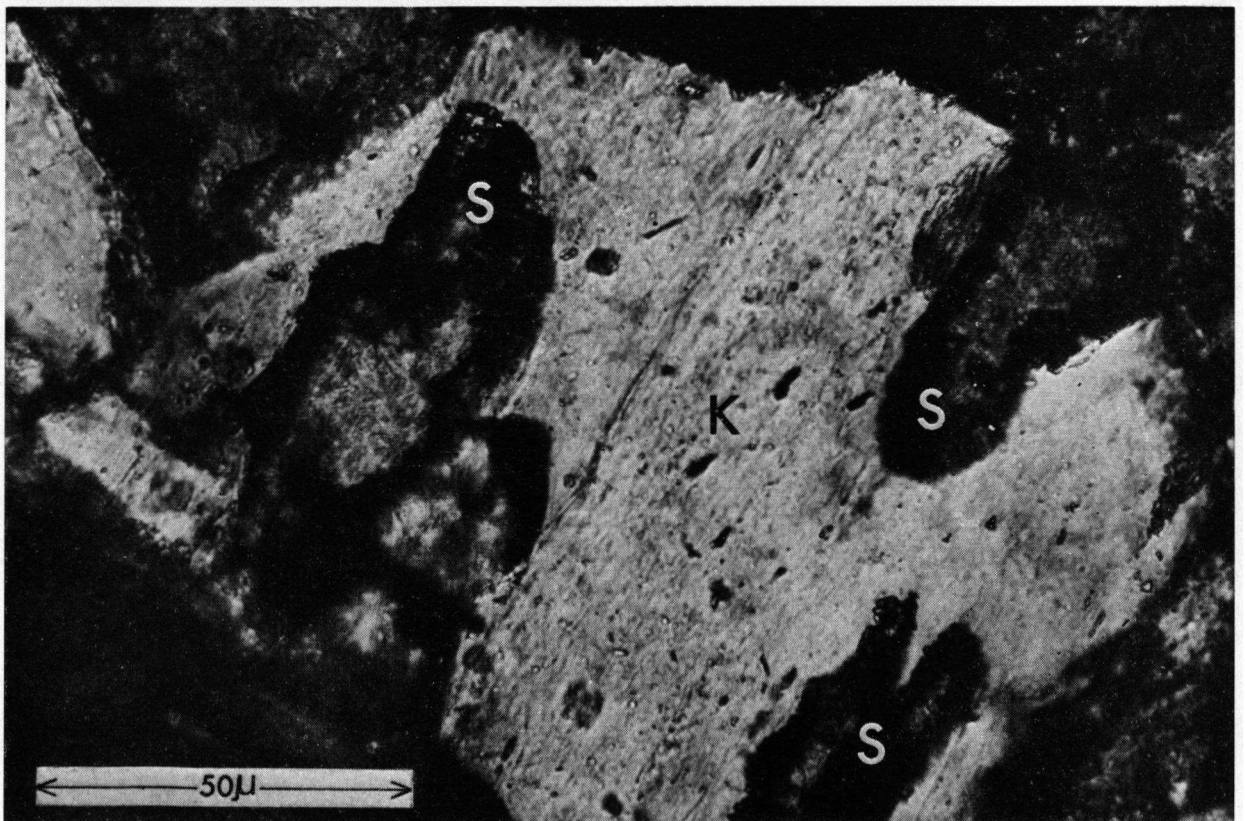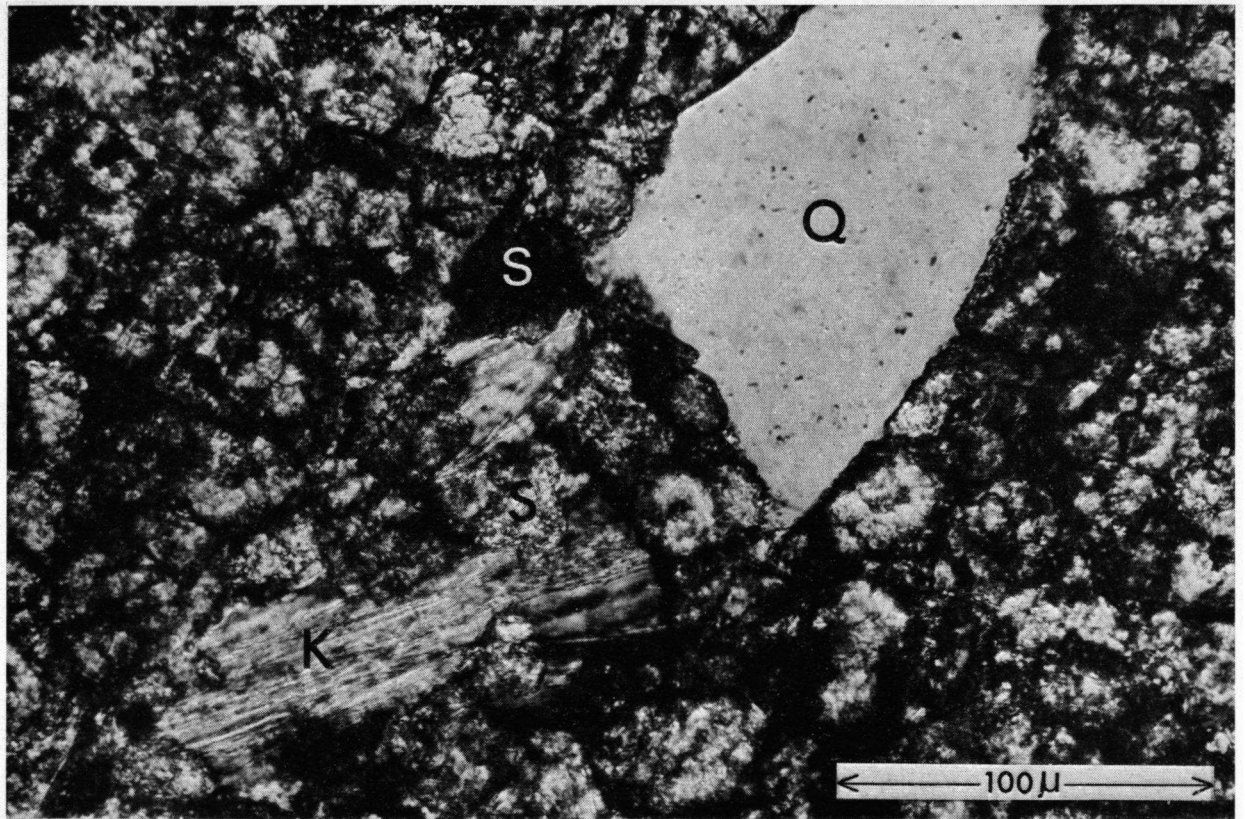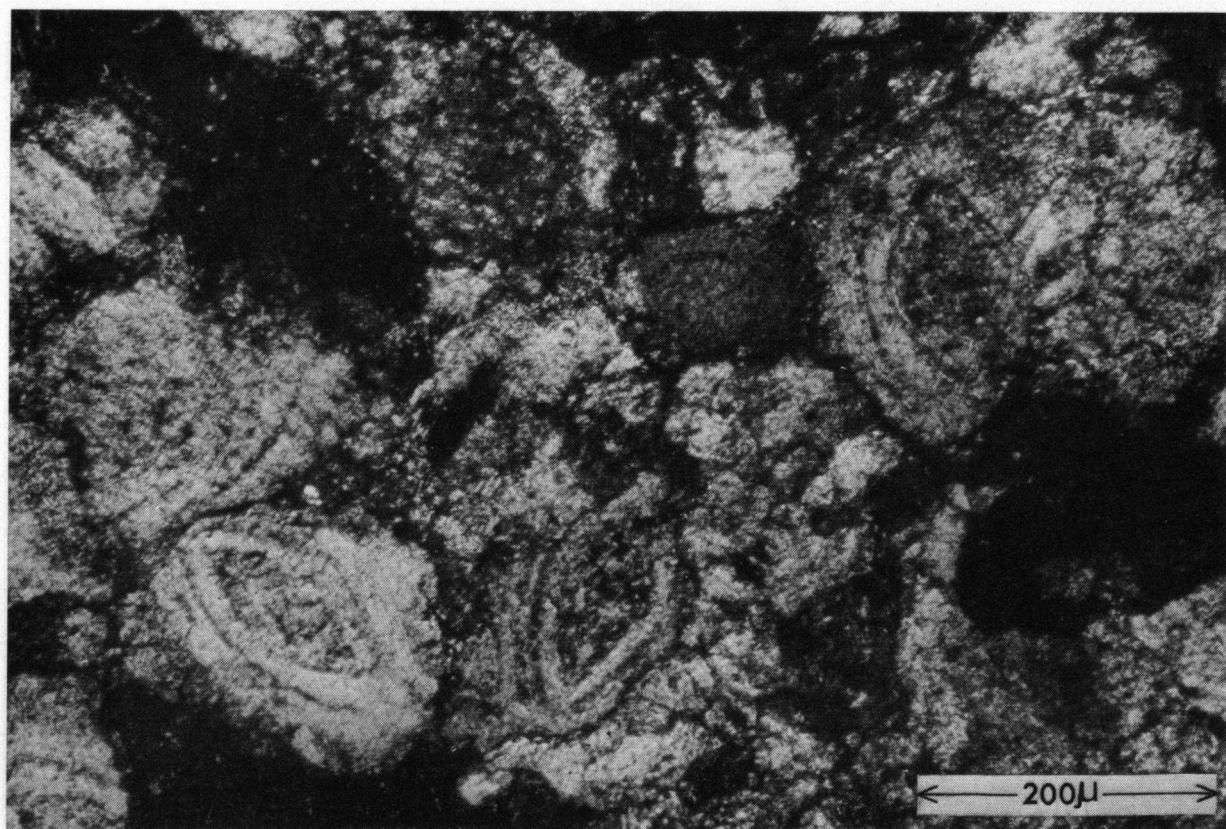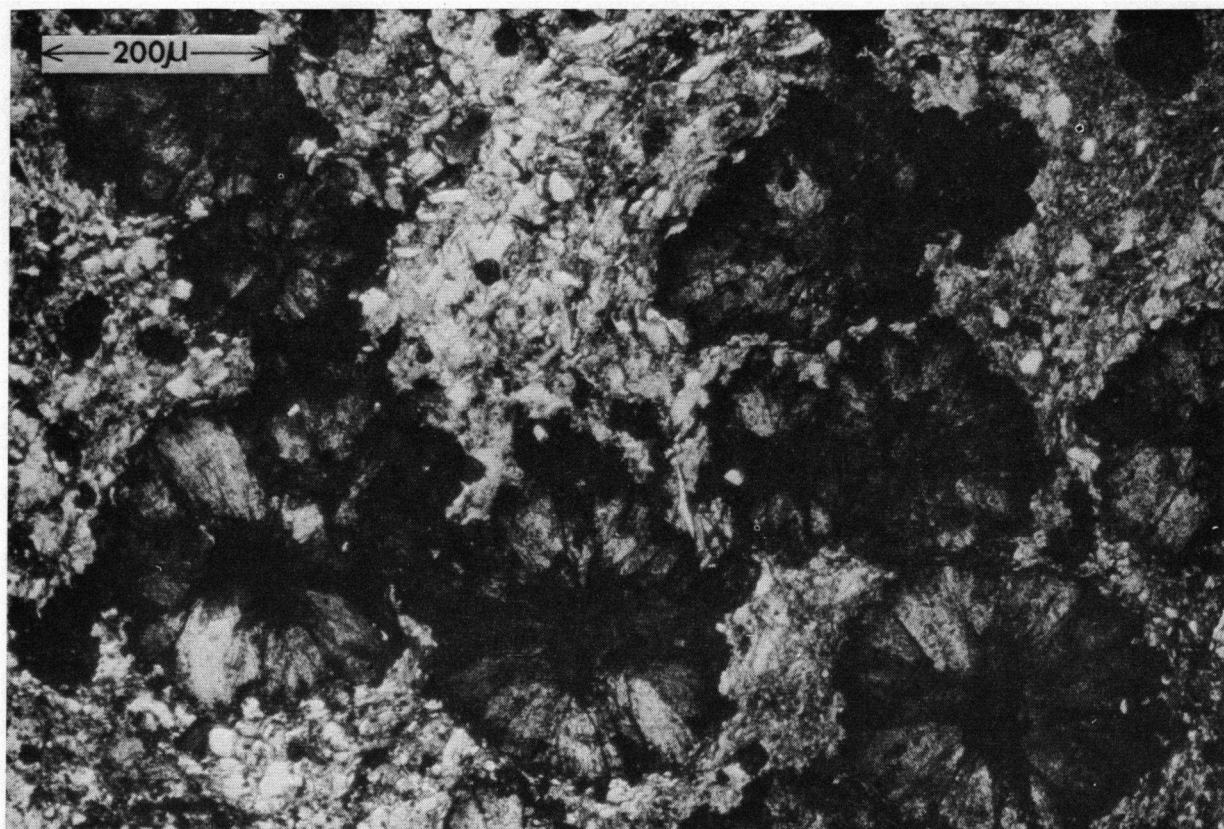
siderite was first deposited *on* the sedimentary interface, because this takes place in recent swamps (Barth et al., 1960, p. 202). The extremely thin, persistent, and fully conformable ironstone beds (thickness 2 to 10 cm) can usually be traced over almost the entire lateral extent of the back-swamp deposits, which suggests that the precipitation of the siderite occurred simultaneously in large parts of the back-swamps.

The formation of the coarser siderite textures (spherulites, rhombs, mosaics of anhedral crystals) and most of the replacement, may have started early, but probably occurred chiefly during advanced diagenesis (Scheere, 1955, 1963).

Mudstones containing sphaerosiderite show not only replacement by siderite, but also alignments of detrital grains parallel to the boundary surfaces of the spherulites (Fig. 41). This texture, which amongst others, has also been described by Gilbert (1958, p. 369), is the sedimentary analogue of what in igneous petrography is called pilotaxitic. In extrusive rocks (e.g. andesites) it is produced by flow. In the Malpas mudstones, however, there is no evidence of flow, because the parallel arrangements of detrital grains occur only in the immediate vicinity of a siderite spherulite and the mudstones containing the spherulites show no field evidence of mudflow deposits.

The occurrence of the texture in the mudstones is therefore better explained as the result of growth pressure of the siderite spherulites (Gilbert, 1958, p. 370) or by compaction. It seems likely that the muds in which the spherulites formed were at least not fully lithified, otherwise such good orientation of the clastic particles (up to 50 micron) could not have been produced.

Some siderite spherulites have nuclei composed of fine-grained aggregates of marcasite measuring up to 200 micron, which indicates that a crystallization phase of marcasite preceded the crystallization of siderite. The combined marcasite/siderite phenocrysts are restricted to mudstones and shales associated with coal stringers.

Coal

Three samples collected from the lowermost levels of the coal in the Malpas Formation (sections 3 and 4; one sample between these two sections, sheet III) were analysed by the *Geologisch Bureau voor het Mijngebied,* Heerlen, The Netherlands, whose cooperation is gratefully acknowledged.

The percentages of volatiles were: 8.2, 7.0, and 7.7% (d.a.f.); and the ash percentages: 33.4, 38.4, and 22.4% respectively.

The coal contained in the Malpas Formation is mined and, after crushing, is used to heat the kilns of the cement factory at Cherallo (Fig. 1). Mining operations are carried out at two levels, the *'primera y segunda capa'* (*primera capa* at approx 211 m in section 3; *segunda capa* at 139 m in section 3, sheet III). The coal used in the cement factory is a homogenized mixture of unknown composition of coal from both these levels. The results of analyses regularly performed on the coal mixtures were kindly communicated to the author. They apply to the years 1966 and 1967, and for each year represent mean values of 1080 analyses. For 1966, volatiles: 8.47% (d.a.f.); ash: 47.94%; and for 1967, volatiles: 8.28% (d.a.f.), and ash: 50.32%. With respect to the volatile content, the coals are anthracites (Patteisky & Teichmüller, 1960). It is of interest to note that the anthracites coexist with and are underlaid by kaolinite-rich rocks. In the drill-hole Münsterland 1 (Hedemann & Teichmüller, 1966; Scherp, 1963; Stadler, 1963) this would correspond to a depth of burial of not more than approximately 3000 m. However, this value has little significance for the present deposits, since anomalously high geothermal gradients probably existed both in the Münsterland deposits (Hedemann & Teichmüller, 1966) aswell as in the present ones (Maladeta granodiorites, post-orogenic (Erill Castell) volcanism. Moreover, in the case of the Malpas deposits, the time of burial at greater depths is unknown.

X-ray analyses

The optical identification of the kaolinite and sidetite was confirmed by X-ray analyses of bulk samples and hand-picked minerals. Illite, chlorite, and illite/chlorite mixed layers are of common occurrence in the mudstones and shales. The origin of these clay minerals was not investigated in detail; they were probably largely clastically supplied, mainly from the Paleozoic basement phyllites, but may in part be authigenic. Anatase occurs in most samples; traces of feldspar and magnetite were noted.

The marcasite, chalcopyrite and pyrite were determined in polished samples.

Chert

Only one level of chert was found in the Malpas Formation; it occurs in the lower part of section 3, and can be laterally traced eastwards up to, but not beyond, the Peranera River (Fig. 4).

The chert is embedded in black shale showing perfect fissility, limited aggregate extinction in thin section, and a high content of organic matter. The mode of

Fig. 40. Spherulites of brown-stained siderite in Malpas mudstone. Clastic particles in the direct vicinity of the spherulites are reoriented to a position parallel to the surface of the spherulites. Some spherulites have a small nucleus composed of marcasite; some enclose clastic quartz particles. Sample 1203, plain light.

Fig. 41. Malpas siderite bed (ironstone). This siderite bed was formed by almost complete replacement of a mudstone. Rows of inclusions in siderite crystals indicate a rhombohedral crystal habit. The successive crystal surfaces are increasingly rounded toward the peripheries. Sample 1194, nicols crossed.

occurrence of the chert is represented by lenses up to 10 cm thick. Each lens can be followed over a distance of several metres and in an occasional case for 20 to 30 metres, before it pinches out and is succeeded by another lens at the same level or at a level some centimetres higher or lower. Commonly, three or four lenses occur in vertical succession over a total thickness not exceeding 2 m (Fig. 11). Rare chunks of silicified wood measuring up to 30 cm protrude from the chert lenses or occur isolated in the black shales. In hand-specimens the rock shows a homogeneous texture, conchoidal fracture, a deep black lustre, and small white specks.

In thin section the chert appears to consist of densely intergrown microcrystalline quartz and chalcedonic quartz. The term microcrystalline quartz is used here for crystalline-granular, commonly undulatory quartz with a fine grain-size (Keller, 1941; Pittman, 1959). In the present samples the grain-sizes of the crystals in the microcrystalline quartz are somewhat coarser than is commonly recorded, since they range from 3 to 300 micron. Chalcedonic quartz (Folk & Weaver, 1952) occurs in up to 1500-micron fibrous masses. These fibrous growths are clear and correspond to the white specks seen in hand-specimens.

The microcrystalline quartz is contaminated with extremely fine-grained, probably colloidal organic matter appearing as a brown stain in thin section, and with plant fragments measuring up to a few cm (Fig. 42).

Scattered through all the samples (1195, 1196, 1219, 1220) are small (up to 800 micron), usually perfect rhombohedres of dolomite. These dolomite crystals show indentation textures with the microcrystalline quartz (Fig. 43). Enclosed in the cherts, moreover, are masses measuring up to a few cm and consisting of very loosely packed aggregates of randomly oriented kaolinite crystals (not illustrated).

The origin of the silica can probably be traced to the continuous leaching of the Erill Castell tuffs, which were exposed in the source area of the Malpas Formation (p. 185).

As shown by Kobayashi (1967), the silica content of rivers draining volcanic areas is relatively high, reaching 40 ppm or more in some Japanese volcanic areas. These values, however, are still well below the equilibrium concentration for amorphous silica, which lies at approximately 120 ppm (Krauskopf, 1967, p.

166). What factor or factors caused the elevated concentration of the silica solutions is not known; but it could have been strong evaporation of the back-swamp surface waters. Indications that the climate during the accumulation of the Malpas Formation was at least at times warm enough to cause strong evaporation are found in the diagenetic history of the braided stream deposits (p. 188) and of the thick tuff bed (p. 198). Furthermore, the presence of limestone beds also points to at least not too cold waters (p. 197).

According to Krauskopf (1959), concentrated silica solutions will ordinarily not precipitate but form colloids. Such silica colloids, which presumably accumulated on the bottom of the reducing Malpas back-swamp environment (association with organic matter), may have given rise to the observed chert lenses.

A possible equivalent present-day occurrence of such inorganic-precipitated silica gels are the black oozes collected from the Missisipi delta. There, they occur on the bottoms of stagnant bays and bayous (Russel, 1936, p. 143).

Millot (1964, p. 325) thinks that the chalcedonic habitus of quartz does not necessarily indicate a transitional gel stage. He argues that chalcedonic quartz is crystalline (Folk & Weaver, 1952), and that the habitus may result from the type of material replaced and the types of ions in solution.

In the present author's opinion, the suspended, completely undeformed, and fragile cell structures of the plant material found in the Malpas cherts (Fig. 42) support the hypothesis of a transitional silica gel stage. Virtually all the mudstones and shales in the Malpas Formation (back-swamp facies) contain some washed-in, fine-grained vegetational debris. This material now has the aspect of coalified films lying parallel to the stratification (similar to those illustrated for the Aguiró Formation in Fig. 29). Where a silica gel occurred on the bottom of the back-swamp area, the sinking vegetational debris would be entrapped somewhere in the gel because a gel has some rigidity. To preserve the delicate cell structure undeformed, the silica gel must have hardened slowly but before even small overburden pressures could develop. The dehydration of the gel must therefore have taken place in the course of early diagenesis. This conclusion is corroborated by the preservation of the enclosed aggregates of randomly oriented, very loosely packed kaolinite crystals. These aggregates likewise must have been protected against compaction pressure by the already hardened silica.

The crystallization history of the dolomite crystals can also be reconciled with the presumed presence of a silica gel, which can be visualized as follows. Crystallization of dolomite could begin in the highest and least concentrated part of the silica gel. Supply of the necessary ions could take place because the gel structure was still sufficiently open. At the same time, the growing crystal could already obtain support from the gel. Once the dolomite crystals came to lie at a somewhat lower level, their growth would be impeded by the denser structure of the gel; many crystals were even

Fig. 42. Malpas Formation; chert bed. Chalcedonic and microcrystalline quartz. Single dolomite crystals (D) are scattered through the rock. Note undeformed delicate cellular structure of plant remains. Sample 1195a, nicols crossed.

Fig. 43. Malpas Formation; chert bed. Dolomite rhomb in microcrystalline quartz. After its formation, the dolomite rhomb probably became corroded and was replaced by quartz. Sample 1195, nicols crossed.
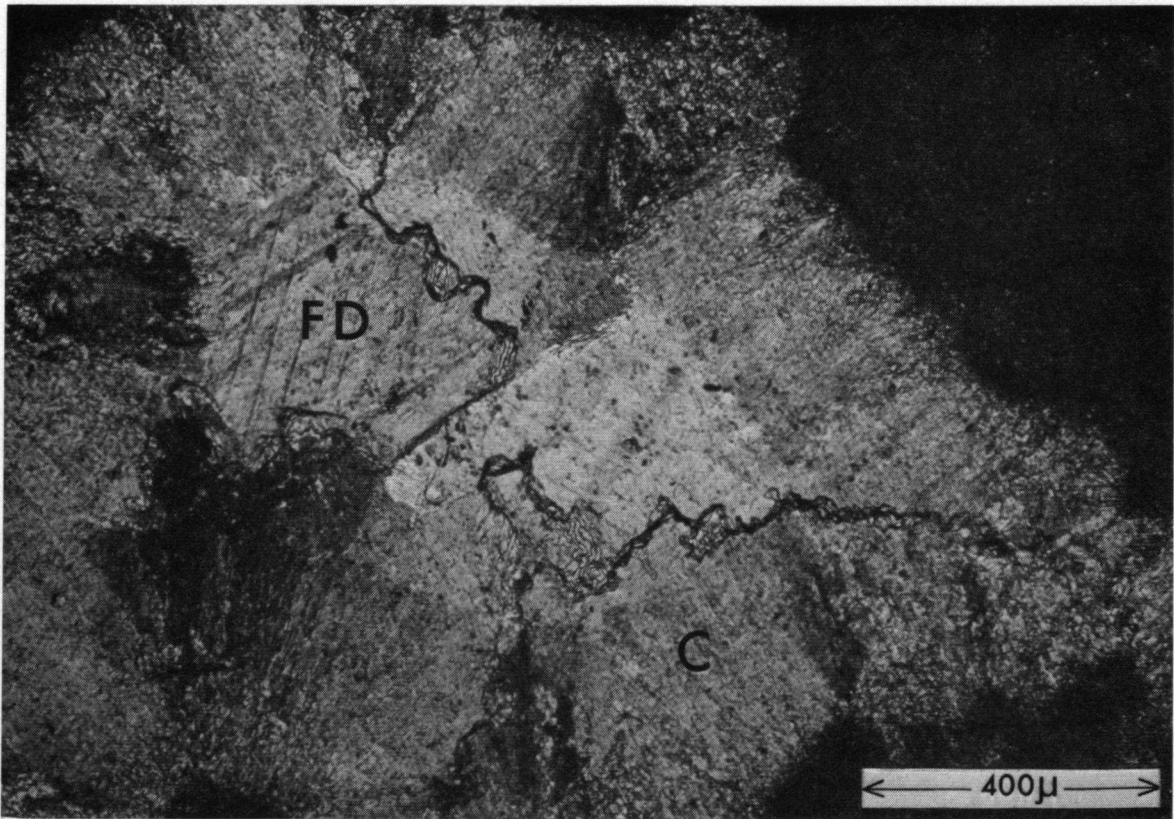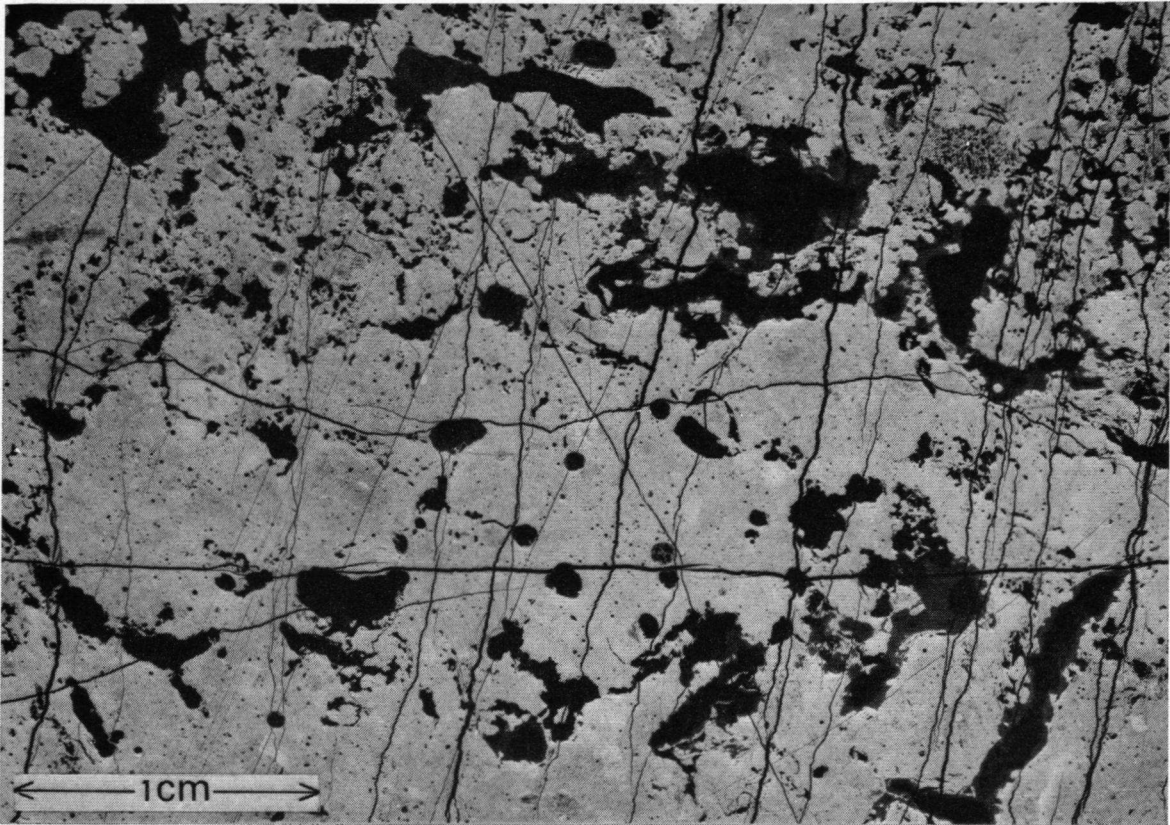
corroded, probably due to an increase of the CO_2 concentrations. The corroded parts of the dolomite crystals were filled by the hardening silica gel, which ultimately must have led to the indentation texture illustrated in Fig. 43.

Carbonate beds

Predominantly dark grey carbonate beds occur at two levels in the sections of the Malpas Formation under study. The lower level is found at the height of 23 m in section 3; here the carbonate is largely ferroan calcite. The upper level is at the very top of the formation, in conformable contact with the overlying red beds of the Peranera Formation (Fig. 4; section 5, sheet III). There, the carbonate is ferroan dolomite in beds intercalated with ferroan dolomite-bearing kaolinitic shales and some mudstones.

Limestone. — The lateral extent of the level of the limestone is not known, due to insufficient exposure; it does not recur in section 4. In the area between sections 3 and 4 (Fig. 4), there is a similar level of limestone, also intercalated between mudstones and shales.

Since the mudstones and shales must have been deposited in fresh-water back-swamp areas belonging to a fluvial environment of predominantly meandering streams, it follows that the intercalated limestones must have been deposited under similar conditions (p. 154). The limestones are extremely fine-grained, the grain-sizes of the calcite being mostly less than 4 micron. They are therefore true micrites (Folk, 1962, 1965). In plain light the micrite is light brown, probably due to dispersed colloidal organic matter; small fragments of vegetational debris also occur scattered through the limestones. Although less well preserved than in the cherts (p. 193), cell structures are visible in these fragments.

Micro-fossils (small gastropods, ostracods(?)) are present in low numbers and poorly preserved condition. The micrite proper characteristically shows indistinct parallel laminations observed as slight variations in the grain-size and colour intensity of the rock. The laminations are curved in various directions; locally, laminated parts are bounded by small fractures. These laminations are considered to be structures formed by carbonate-binding algae; judging from the samples collected (1186, 1187, 1188), the whole limestone unit may have formed in this way.

All six limestone beds typically show numerous small white specks. On examination in thin section, these specks appear to be filled-in voids. Although these former voids are generally somewhat planar and are oriented parallel to the stratification, many are hemispherical or irregular, and some are oriented perpendicular to the stratification. The voids range in size from 20 to 30 micron up to about 4 mm. The general pattern of the voids is illustrated in Fig. 44; a representative void-fill is shown in Fig. 45.

In the void-fills a primary phase of dense, fibrous non-ferroan calcite occurs, followed by a secondary infill of ferroan dolomite. Some voids contain only the fibrous calcite, others have no fibrous calcite but contain anhedral dolomite only. The crystals of the central dolomite fills commonly meet along stylotitic seams; the origin of this is not known (Fig. 45).

In size and distribution over the limestone, the voids resemble the well-known birdseye structure described for many fine-grained limestones; they are therefore considered to be such structures. The reader may recall that in this study birdseye structure was also found in silty and clayey puddle deposits (Fig. 24).

In a recent paper, Shinn (1968) reconsidered the formation of the birdseye structure in fine-grained limestones. After citing various examples of both ancient and recent occurrences, he suggests, also on the basis of laboratory experiments, that the planar type of birdseye is formed by dessication-shrinkage in sub-aerially exposed sediment. This explanation may well apply to the birdseye structure in the Malpas limestones, considering their undoubtedly shallow-water origin.

Limestones in the Malpas Formation are exceptional as compared to most other lithologies. This would be expected also from the fact that the Malpas environment in general must have been a swamp area in which, due to CO_2 formed from abundant decaying vegetational debris, the waters must have been acid. Probably, the limestones could form only under rather special conditions, for instance in extremely shallow parts of the back-swamp areas, with marked depletion of the available CO_2 due to photosynthesis by algae and water that was not too cold. The general presence of birdseye structure in the limestones agrees with this view.

Ferroan dolomite. — At the very top of the Malpas Formation the carbonate beds occur in a facies suggested to be lacustrine (p. 158).

The sequence, which consists of an alternation of ferroan dolomite beds and ferroan dolomite-bearing kaolinitic shales, is more than 20 m thick (only the top part being shown in section 5, sheet III) and can be shown to underlie the wedge of Peranera Formation over its full length (Fig. 4). Fine grain-size, strict, well-expressed parallel bedding, and absence of intercalated fluvial channel-fills, constitute the main evidence that this environment was not a back-swamp area bordering meandering streams as must have been the case in most of the Malpas deposits, but that a true, possibly extensive (though very shallow) lake existed at the close of the Malpas history.

Fig. 44. Malpas Formation; birdseye structure in fine-grained limestone. Sample 1186, plain light, *negative print*, plain light.

Fig. 45. Malpas Formation; birdseye structure in fine-grained limestone. The crystalline fill of one of the voids is shown. It consists of primarily formed, densely fibrous non-ferroan calcite and secondarily formed ferroan dolomite. The stylolites are restricted to the void-fills. Sample 1186, nicols crossed.

The kaolinitic shales and mudstones resemble the back-swamp-deposited shales and mudstones in that they show extremely fine-grained, densely intergrown kaolinite and some microcrystalline quartz; they differ from those deposits in that they contain far less silt and sand-sized grains. Most shales contain some ferroan dolomite, which is apparently secondary since it occurs in irregular aggregates.

Upon staining with a mixture of alizarin red-S and potassium ferricyanide (Evamy, 1963; Dickson, 1966), all samples of the carbonate beds showed Turnbull's blue precipitates. Calcite is apparently absent, and the carbonate should be a ferroan dolomite. This result was checked by X-ray analyses of two samples (1226, 1232). No calcite was detected. The a_0 and c_0 values (in Å) for the ferroan dolomites are, according to calculations by Mr. J. J. M. W. Hubregtse and Mr. C. F. Woensdregt (Leiden), as follows:

$$\begin{aligned}
\text{sample 1226: } a_0 &= 4.817 \pm 0.002 \\
c_0 &= 16.0668 \pm 0.0005 \\
\text{sample 1232: } a_0 &= 4.816 \pm 0.002 \\
c_0 &= 16.0524 \pm 0.0005
\end{aligned}$$

Following Goldsmith et al. (1962), the approximate ratios of iron, magnesium, and calcium were calculated to be: sample 1226: Fe 12, Mg 38, and Ca 50; sample 1232: Fe 5, Mg 44, and Ca 51.

In thin section, the ferroan dolomites are less fine-grained than the limestones treated above, the largest part of the rocks consisting of 10 to 30 micron mosaics of anhedral crystals.

No fossil fragments occur, but some fragmental plant debris was found. Some samples, among them 1226 and 1232, show a structure reminiscent of birdseye. The filled voids, however, are less clearly defined than in the case of the limestone beds; it seems likely that a formerly distinct birdseye structure in a lime-mud was partly obliterated by dolomitization. An extreme shallowness of the lake, as indicated by the presence of the birdseye structure (see discussion under limestones above), would also follow from the presence of mud-cracks (at two levels, section 5; sheet III).

In various samples the presence of algal structures is suspected; for only one sample (1237) is there less doubt, since it shows laminations similar to those described for the limestones.

The generally fine grain-size and the presence of ghosts of structures present in the limestones too, suggest that the ferroan dolomites formed by penecontemporaneous dolomitization of lime-muds. Under precisely what geochemical conditions these lime-muds were converted into ferroan dolomite is not known. However, since at present most dolomitization takes place in environments of strong evaporation, it may be assumed that this was also the case in the Malpas lake deposits. The point is of special significance because the Malpas dolomites occur at the very transition from a sequence of deposits interpreted as having formed under generally humid conditions (Malpas Formation) to a red bed sequence probably formed in a semi-arid climate (Peranera Formation).

The entire transition from the Malpas Formation to the Peranera Formation is covered by section 5 (sheet III). At the height of 4.8 m in the section, the first greyish-red colours appear in the form of flames and mottles; in a 3 m transitional zone the sediment is mottled greenish- and greyish-red, on top of which follow predominantly greyish-red deposits.

The transitional deposits and the predominantly greyish-red deposits consist of very poorly sorted, sandy mudstones. Significant amounts of feldspar (many crystals still fresh), angular quartz, and some mica suggest that these mudstones are mainly reworked tuffs. The distinct crystal tuff bed (at 6.57 m, section 5, sheet III) is similar in composition to most other crystal tuffs found in the Peranera Formation; for a discussion of these tuffs the reader is referred to p. 209. Features typical of the Peranera Formation, such as widespread replacement by non-ferroan calcite, abundant calcite concretions and the presence of encrusting calcite thought to result from soil-forming processes in a seasonally dry and warm climate (p. 207), testify to the definitive drying up of the Malpas lake and the profound change in climate.

Tuff bed

In the lower reaches of the Malpas Formation an approximately 30 m thick, light-coloured tuff bed can be laterally traced over more than 4 km; it is encountered in the traverses of both section 3 and section 4 (sheet III).

Some bombs and lapilli occur in the tuffs, but the high content of coarse pyroclastics found in some levels in the Erill Castell Volcanics is not reached.

The tuff proper is a crystal tuff. Most abundant are plagioclase phenocrysts, many of which are largely replaced by fine-grained kaolinite, sericite, and ferroan calcite. Of secondary importance are quartz crystals, sometimes in a subhedral habit and commonly intensely cracked. Irregular fragments and shards of a greenish, palagonite-like mineral occur in minor quantities. The groundmass of the tuffs consists of fine-grained crystalline granular kaolinite, intergrown with microcrystalline quartz. Secondary calcite, which has replacement relations with all the components of the tuffs, occurs in aggregates of macro-crystals. It reaches an estimated 10 to 20%, by volume, of the rock.

Small spherulites measuring up to 2 mm are a characteristic feature of the tuffs in section 3 (near Erill Castell). Some of the spherulites have a grain (quartz, feldspar) in the centre, but most of them carry no nucleus of this kind. The spherulites show concentrically arranged, alternating laminae of ferroan calcite and colloform hematite. They commonly coalesce, being intergrown into larger aggregates, which shows that the spherulites originated as *in situ* con-
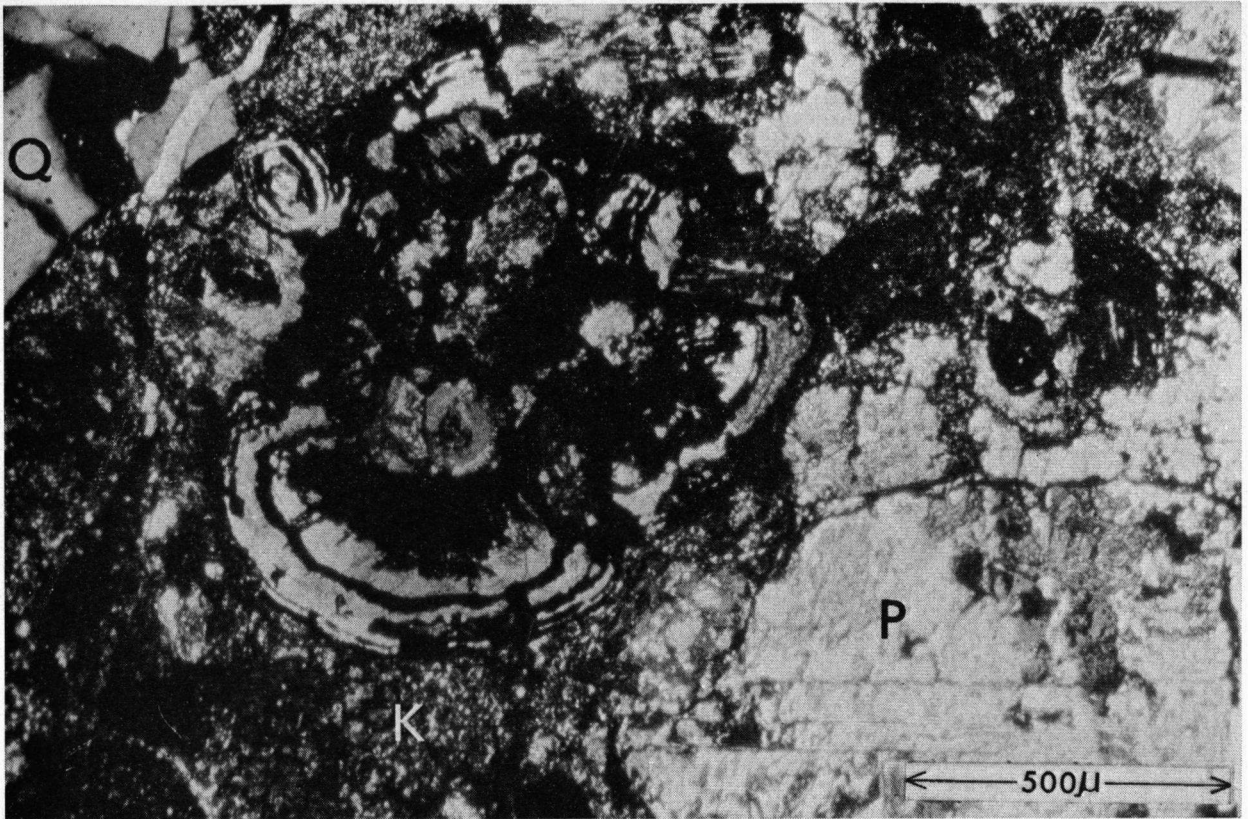
Fig. 46. Malpas tuff bed. Spherulite in a crystal tuff consisting of plagioclase (P), quartz (Q), and an altered ground-mass, mainly fine-grained kaolinite (K). The spherulite shows concentric rings of hematite (black) and ferroan calcite. The plagioclase crystal is partially replaced by ferroan calcite. Sample 1191, nicols crossed.

cretions of ferroan calcite and hematite (the latter originally probably first as hydrous ferric oxides) (Fig. 46).

The number of hematite rings in the spherulites shows little variation; for all spherulites examined in sample 1191 (totalling about 30) it falls between 4 and 6. This suggests a common origin, and that the first ring, the second, etc., developed simultaneously on all spherulites in successive stages. Consequently, the same should hold for the ferroan calcite. Similar calcite spherulites occur densely packed in some Bunter mud-stones (p. 226); in levels of calcrete in the Peranera Formation they are less well developed (p. 207).

Rutte (1958) has described calcite spherulites ('Pseu-dooids') from recent deposits in eastern Spain. According to him, they originate in highly porous superficial deposits in which the temperature, due to insolation, may fluctuate considerably. They are restricted to areas of calcrete formation and consequently must form under climatic conditions similar to those required by calcrete.

In a separate study, the present author compares the spherulites from the Malpas tuffs, the Peranera cal-crete, and the Bunter mudstones with recent ones from eastern Spain.

Apart from differences such as a higher iron content and a coarser microtexture in the ancient ones, the spherulites look very much alike (Nagtegaal, 1969).

Although no calcrete was found in the Malpas tuff bed, it is assumed that the spherulites may have formed under the conditions suggested by Rutte. The water-table would then have been temporarily situated at some distance below the surface of the deposits. Although the water-table in the Malpas environment was normally located at or above the sedimentary interface (p. 171), the situation of a lowered water-table could have existed in the presumably rapidly accumulated, bombs and lapilli-carrying tuffs.

If the formation of the Malpas spherulites is rightly attributed to processes similar to those taking place in the formation of calcrete, their presence forms yet another indication that the climate during the accumulation of the Malpas Formation must have been warm enough to cause strong evaporation.

PERANERA FORMATION

The petrography and diagenesis of the three main rock types of the Peranera Formation, the sandstones (including conglomeratic sandstones), the mudstones, and the tuff beds, were studied.
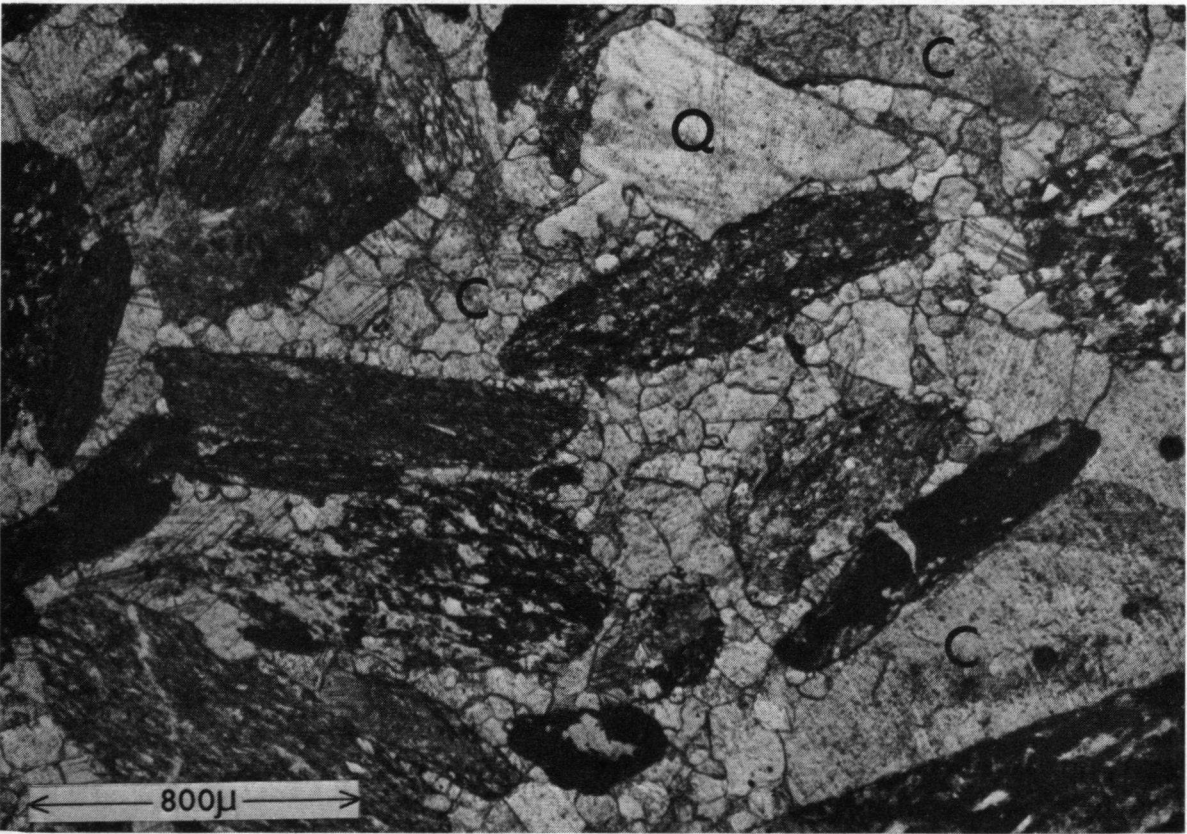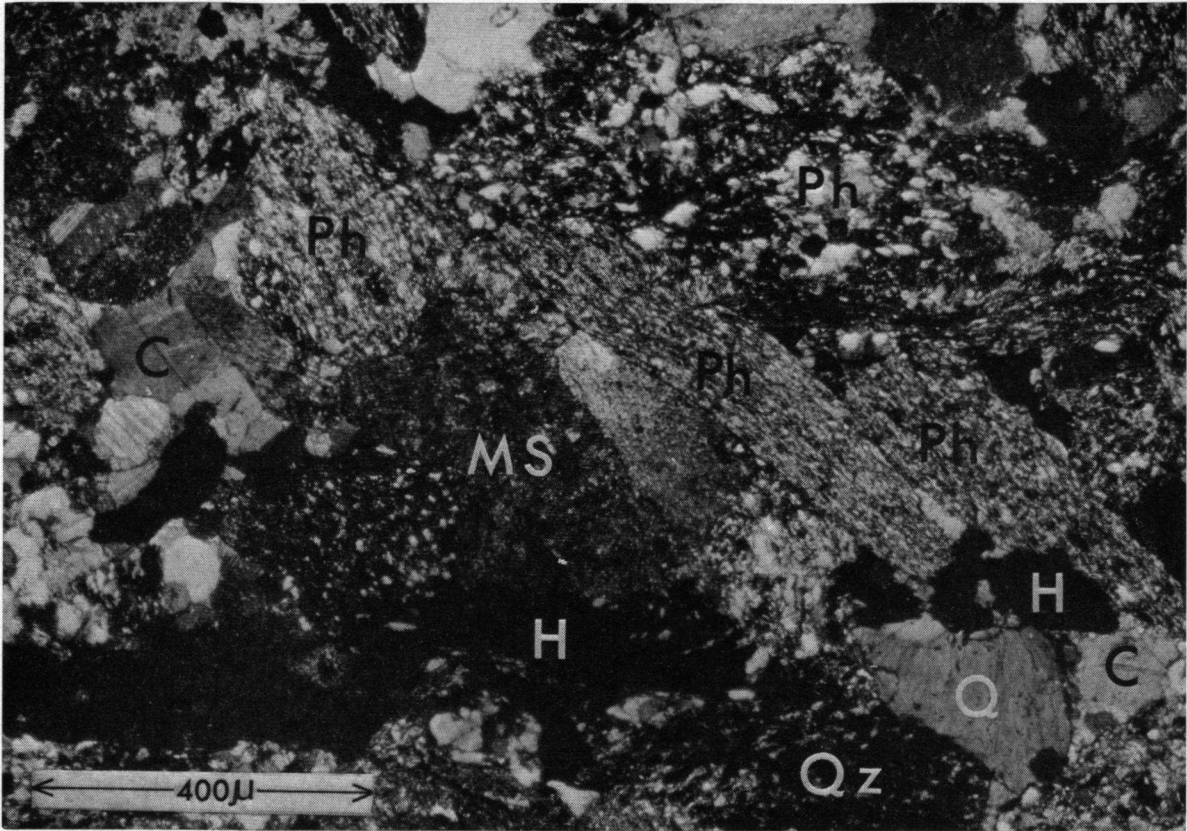
TABLE IX

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (phyllite debris,quartz,feldspar) largely replaced by calcite	ser/chlor. quartzite	20	ferric oxide (largely hematite)	15⊖	colloform, authigenic, and in clastic grains
	non-calc. and silty phyllite	15			
Quartzite, quartz, feldspar. angular to subangular	calc. phyllite	7	feldspar	x	small overgrowths on Na-rich plagioclase
	quartz	4			
Phyllite: rounded	fine-grained limestone	3	calcite	33	sparry cement and replacement, pseudomorphs after clastic grains
	feldspar	2			
Max. grain-size: 1.8mm	bleached biotite	x			
	zircon	x	chalcedony	1	small masses associated with replacement calcite
Phyllite grains in subparallel and imbricate position	tourmaline	x			

Table IX. Peranera Formation, sandstones. Mean values after samples 1305, 1309 (300 points counted in each sample; little difference in coarseness).

Sandstones

Original sediment, provenance. — The composition of the Peranera sandstones is shown in Table IX. The data are based on two samples, nos. 1305 and 1309, taken from the middle reaches of the formation in the Las Iglesias area (section 6, sheet IV).

The bulk of the sandstones is made up of polymineral rock fragments (Table IX; Figs. 47 and 48). Many of these fragments are directly traceable to their source rocks, all of which at present occur to the North of the study area.

Comparing the rock fragments in the Peranera sandstones to those transported by present-day rivers draining the Axial Zone of the Pyrenees (Appendix, p. 229), it is clear that the Devonian must have been present as outcrops in the Peranera drainage area. The components of certain Devonian origin include calcareous phyllite and microsparry limestone, the latter occasionally containing trilobite, echinoderm, and tentaculitid fragments.

It is not certain, however, whether all the microsparry limestone fragments stem from the Devonian. Some are laminated and some have a vaguely globular

internal structure. These types moreover, are devoid of fossil fragments. The limestones from which these components must have derived resemble the calcrete in caliche profiles, and since calcrete with a similar microtexture occurs in the Peranera deposits proper (p. 207) there is the possibility that it may have been available as a source.

As noted in the Appendix (p. 229), it is not possible to classify the non-calcareous and silty phyllite according to whether they came from the Devonian or from the Ordovician.

It seems safe to conclude, that Ordovician rocks occurred in the Peranera drainage area and that they even were an important source of material. The analyses of present-day river sands (Appendix, p. 229) showed that sericite/chlorite quartzite occurs in the sands from both Devonian and Ordovician rocks. The amount of this quartzite rock in the 'Ordovician sands', however, is significantly higher than that in the Devonian ones (29% *vs* 4%). In the Peranera sandstones, the content in sericite/chlorite quartzite fragments locally reaches 20% [1].

The prevailing climate in the Peranera lowlands must have been semi-arid (p. 171). On the basis of the general occurrence in the sandstones of clastic grains rich in ferric oxides and appreciable numbers of grains almost entirely consisting of ferric oxide, it is assumed that ferricretes and soils rich in ferric oxides formed contemporaneously in the uplands, which may have had a somewhat more humid climate (p. 203). For this reason a certain degree of selective weathering may have occurred in the supply areas, leading to a relatively greater supply of non-calcareous (mainly Ordovician) components.

Feldspar and biotite, which are found in the sands of rivers which at present drain the Maladeta granodiorites (Appendix, p. 229), occur in many samples of Peranera sandstone. Both these minerals, however, could come from contemporaneous tuffs, which also are intercalated *in situ* in the entire Peranera sequence. Nowhere in the Peranera deposits have granodiorite pebbles been found. It is therefore unlikely that the granodiorites were exposed in the Peranera drainage areas.

Fig. 47. Coarse-grained, moderately sorted Peranera sandstone. The sandstone is rich in unstable grains such as phyllite (calcareous and non-calcareous) and silty phyllite and microsparry Devonian limestone (MS). The quartz grains are angular (Q). The phyllite grains lie in an imbricate position (current towards the left). Authigenic colloform hematite (H) replaces phyllite grains *in situ*. Quartzite grain (Qz) is stained dark by hematite, presumably before deposition of the grain. Calcite cement (C) only occurs in small interstices between the grains. Compare with Fig. 48. Sample 1304, nicols crossed.

Fig. 48. Coarse-grained, moderately sorted Peranera sandstone. Except for one angular quartz grain (upper centre, Q), the grains consist of various types of phyllite, and unlike those in the sample shown in Fig. 47, are very loosely packed. The calcite cement, which in part has a blocky, encrusting habit and which occupies all the space between the grains, may therefore have been introduced at an early (precompaction) stage. The dark stain, which varies in intensity from grain to grain, consists of ferric oxide included before deposition of the grains. Sample 1302, plain light.

[1] See footnote on p. 177.

In a broad sense, the amount of tuff components in the sandstones diminishes rapidly going upward from the base of the Peranera Formation in the section studied (sheet IV). In the lower reaches of the section, up to the height of about 40 m above the base, components of altered tuff similar to those found in the Malpas sandstones (p. 185) are abundant. They probably derive from the Erill Castell Volcanics, which apparently had not yet been fully exhausted as a source of sediment. Supply of Erill Castell Volcanic material to the Peranera deposits could have been easily realized *via* short routes wherever the Malpas Formation fails and the Peranera deposits directly overlie the Erill Castell Volcanics. (Fig. 5).

The segment from 34 m to 74 m above the base of the section does not provide good information about the type of clastic supply because the deposits are mudstones, strongly replaced by caliche-calcite (p. 207). From 80 to 140 m there are many tuff beds. This period of tuff deposition led to a renewed supply of tuff components to the fluvial sands. Some channel-fill sandstones consist entirely of minimally reworked tuff (samples 1273, 1276, 1277). Above a height of about 140 m, Paleozoic rock fragments dominate in most of the sandstones.

The Peranera sandstones are immature deposits, both mineralogically (high content of phyllite debris) and texturally (angularity of quartz). Although they are richer in sericite/chlorite quartzite grains, their primary lithology is comparable to that of the Aguiró sandstones (p. 175).

Diagenesis. — After deposition, the Peranera sands were modified mainly by three diagenetic processes. These processes are, in chronological order:

(1) authigenesis of ferric oxides [1]
(2) precipitation of locally abundant authigenic calcite
(3) compaction-moulding in zones low in early authigenic calcite.

The Peranera sandstones contain local, colloform, commonly highly irregular aggregates of hematite. The major part of these aggregates must have formed authigenically, since their relations with the adjacent clastic grains are almost invariably those of replacement

[1] Under ferric oxides is understood the (unknown) original iron oxides (hydrous or non-hydrous) formed in the deposits; nearly all the iron oxide now consists of hematite, only traces of goethite being present, according of the results of X-ray analyses.
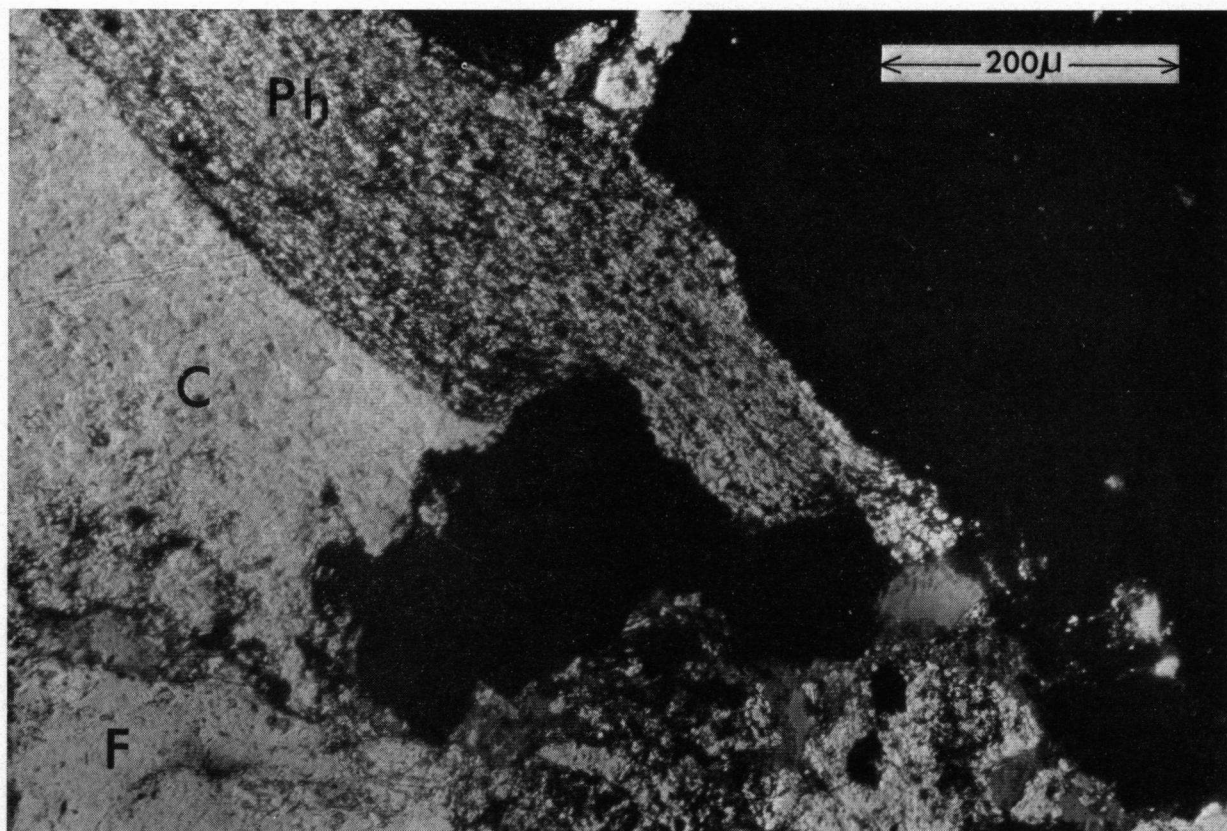
Fig. 49. Coarse-grained, moderately sorted Peranera sandstone. Authigenic colloform hematite (black) partially replacing a phyllite grain (Ph). The local deformation of the cleavage in the phyllite near the point of contact with hematite was probably caused by compaction pressure. The relation indicates that the ferric hydroxide gel from which the hematite must originally have been formed had already hardened at an early stage. Sparry calcite (C) replaces both a feldspar grain (F) and hematite. Sample 1309, nicols crossed.

(Figs. 47 and 49). Quartz, quartzite, and phyllite grains have been attacked with equal facility; plagioclase and limestone grains, however, seem to show less replacement.

Relations like those in Fig. 49, showing a sample in which hematite both replaces and deforms a phyllite grain, indicate that the authigenic ferric oxides had already hardened completely before compaction. On this basis it is assumed that the ferric oxides formed in the sediment at an early stage of diagenesis, through dehydration of a ferric hydroxide gel. This type of hematite is much more abundant in the Bunter sandstones than in the Peranera sandstones. A discussion of its formation is presented on p. 216.

Only a minor part of the total amount of hematite is authigenic, however. Numerous well-rounded phyllite and quartzite grains contain hematite, usually evenly distributed in small patches (Figs. 48 and 50). Such grains sometimes lie in isolation between non-stained grains. Close microscopic observation reveals that the hematite is cut by the rounded grain surfaces. Also, the intensity of 'hematitization' varies from grain to grain (Fig. 48).

These facts indicate that the introduction of ferric oxide occurred before the grains were deposited. The 'hematitization' may either have occurred upstream in the depositional area itself, or in the soils in the drainage basin. The latter possibility seems to be the most likely, as the hematite in the grains is evenly distributed (suggesting formation of ferric oxide in a larger mass of rock), while the *in situ* replacement by authigenic ferric oxide in the Peranera sandstones shows sharp boundaries along irregular fronts of relatively large aggregates (Fig. 49). The non-stained grains were probably derived from levels below the soils bearing ferric oxide or from areas with little ferric oxide in the soils. The introduction of ferric oxide into clastic material prior to its inclusion in the sediment, has recently also been described from Devonian fluvial red beds (Friend *et al.*, 1964; Friend, 1966).

The suggestion that rock fragments may carry considerable amounts of ferric oxides apparently originated by weathering of the source rock, finds support in the presence of ferric oxides in a similar habit in the recent fluvial sands collected in the study area (Appendix, p. 229; Fig. 72).

The fact that ferric oxides in other Permian red beds in part were also clastically supplied has been noted by Feys (1964) and Kruseman (1967). Feys suggested that the clastic ferric oxides were derived from upland remnants of ferricretes formed during the humid Carboniferous. Kruseman thought that the ferricretes could have formed contemporaneously with the deposition of the Permian red beds, due to a more humid climate in high upland areas.

In the present deposits the Stephanian contains direct evidence of the formation of soils rich in ferric oxides (pedalfer paleosol at the base of the Malpas Formation, p. 190). It is questionable, however, whether it would have been volumentrically possible to supply the enormous amounts of Peranera material (up to 700 m thick in our area, over 1000 m thick in other areas; p. 149) without rapidly exhausting the presumed Carboniferous ferricrete profiles in the supply areas. For this reason the explanation of more or less continuous formation of soils rich in ferric oxides in the upland areas, contemporaneous with the deposition of the red beds in lower parts, is preferred.

Differentiation in thin section between hematite that is authigenic *in situ* and hematite contained in clastic grains is often complicated by what are possibly secondary outgrowths of hematite on hematite-carrying grains. For this reason, it is difficult to subdivide the total of 15% given for hematite (Table IX). Probably, however, the authigenic hematite comprises no more than 1 to 3% of the total rock.

The percentage value of 15 for hematite was arrived at, like all the other values shown in Table IX, by point counting under the microscope. It is therefore a volume percentage. In view of the high density of hematite, the weight percentage of Fe_2O_3 could be expected to be still higher, but according to the results of partial chemical analyses, this is not the case, the following values having been found: sample 1305: 6.10% and sample 1309: 5.26% Fe_2O_3. The marked inaccuracy of determinations of ferric oxide percentages by point counting may result from the counting of ferric oxide stains and porous aggregates as hematite.

In summarization of the foregoing the Peranera sandstones may be said to show evidence of:
(1) *in situ* formation of ferric oxides at an early diagenetic stage, and
(2) supply from soils in which ferric oxides had been concentrated.

Both the accumulation area and the drainage basin must therefore have been subject to surface-processes promoting the formation and concentration of ferric oxide. The greyish-red colours of the sandstones are largely due to ferric oxides contained in grains brought in from ferritic soils.

Calcite (33% in Table IX) is, chronologically, the second authigenic mineral formed. In almost all the sandstone samples this is confirmed by calcite replacing authigenic hematite (Fig. 49). Calcite crystals enclosing hematite are also quite common, but in these cases there is not always certainty as to whether the enclosed hematite is authigenic or had once formed part of clastic grains. Almost all calcite in the Peranera Formation is non-ferroan.

In the sandstones, three main types of secondary calcite can be distinguished: (1) calcite in the form of an inert cement, only filling initially open pore spaces, (2) calcite replacing clastic components, and (3) encrusting calcite. The first two types often are impossible to distinguish, because when there is much replacement by calcite, the outlines of former open pores commonly cannot be seen.

Calcite as an inert cement should occur in all Peranera sandstones, because all of them are completely lithified
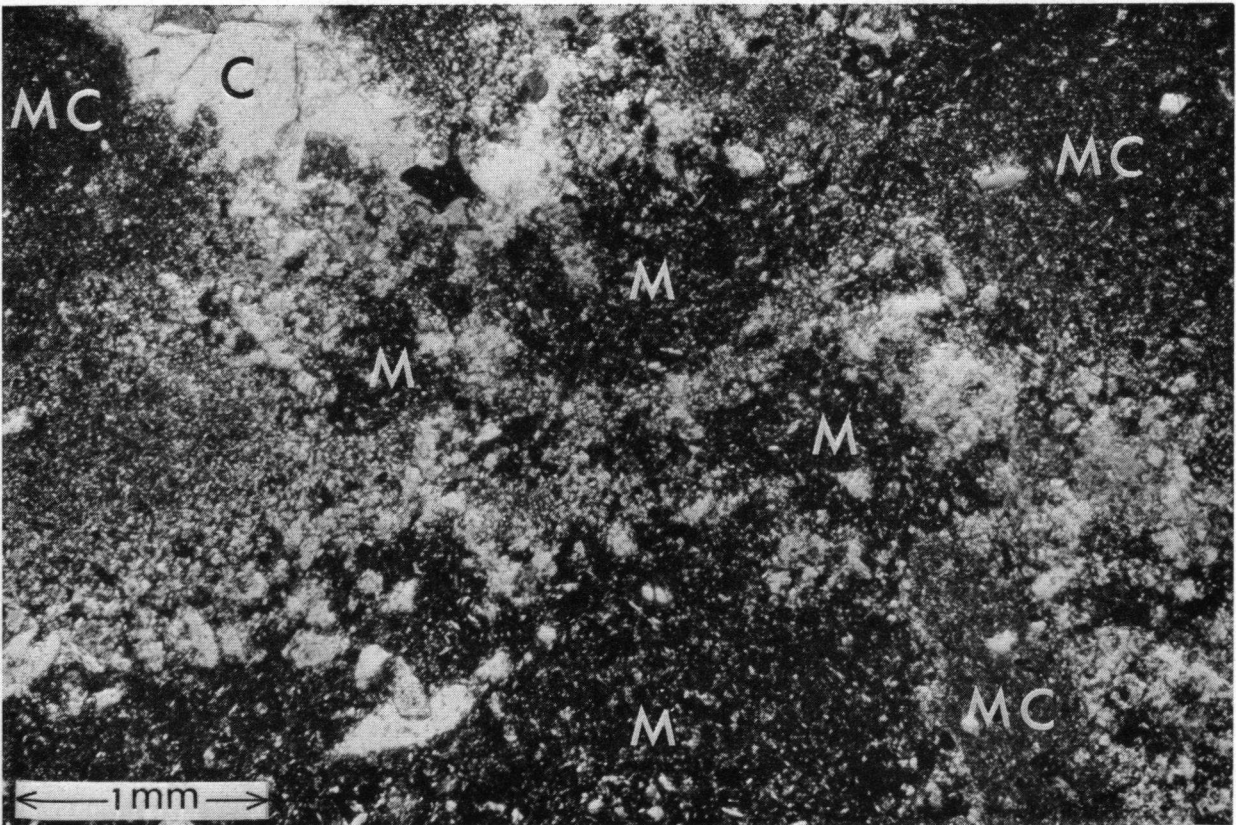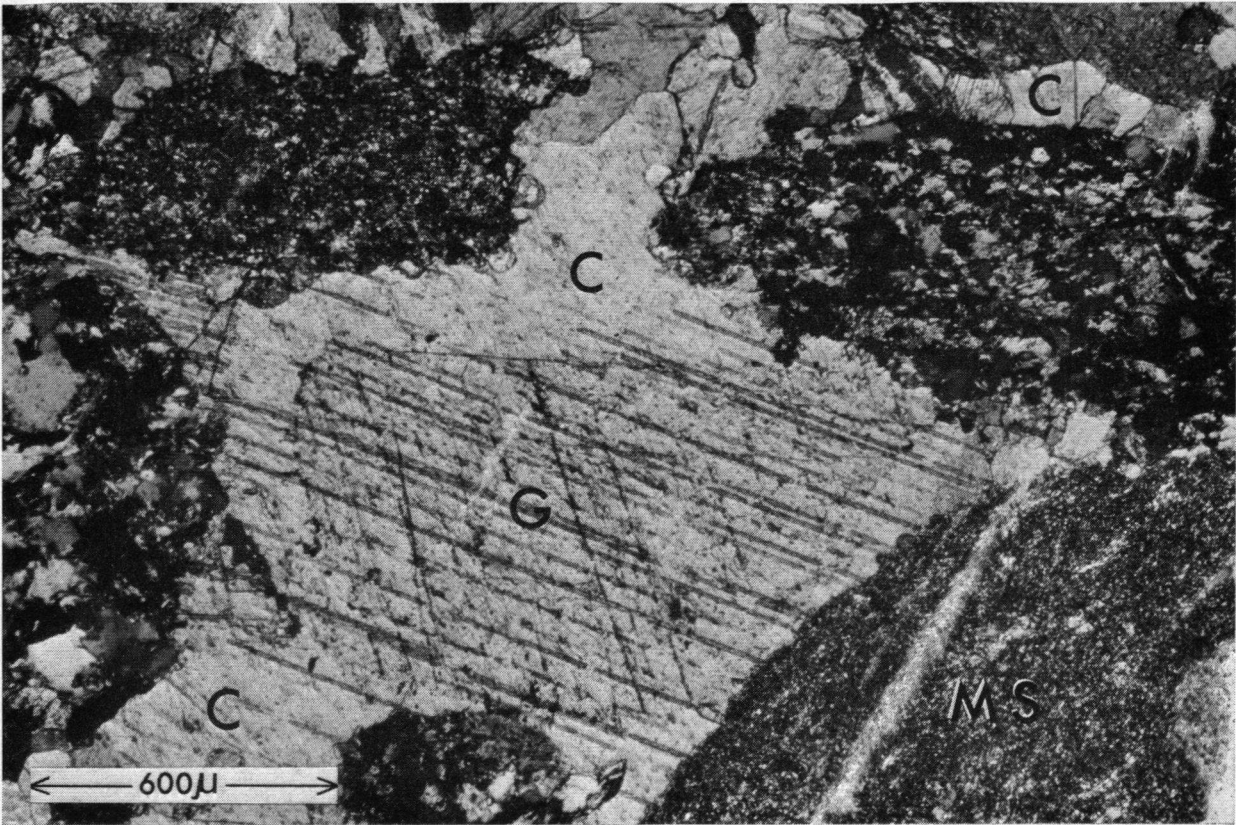
by calcite. The time of introduction of the calcite cement, however, may have differed from case to case. Although the evidence is not entirely convincing, as will be discussed below, a difference in the time of introduction of calcite cement is a reasonable conclusion to draw from a comparison between the amounts of authigenic calcite in the sandstones shown in Figs. 47 and 48. The sandstone shown in Fig. 48 is separated stratigraphically by 12 m from the one of Fig. 47 (section 6, sheet IV). Although the sandstone of Fig.47 possibly had less pore space from the start as the result of a somewhat closer packing (imbrication) and a somewhat higher matrix content, it still seems that the calcite was mainly introduced after compaction (moulding of phyllite grains) had proceeded to a certain extent, whereas in the sandstone of Fig. 48 it may have crystallized almost directly after the sand was deposited, thus preserving the loose packing of the clastic grains.

The loose packing of the sand grains in Fig. 48 could, however, also be the partial result of the driving apart of the grains by growth pressure of the calcite cement (encrusting habit of the calcite). And the possibility also remains that the sandstone in Fig. 48 was not as loosely packed as it now seems, but that the matrix and a number of clastic grains were replaced by calcite. At some points, in the same sample, this is indeed evidenced by ghost structures of clastic grains in single calcite crystals (Fig. 50). In the same microphotograph, replacement by calcite of the ferric oxide-bearing sandy phyllite and quartzite grains clearly results in the impression of a looser packing; in these cases there is no ghost structure to indicate the former grain boundaries.

In some samples (1264, 1274) microcrystalline quartz and chalcedonic quartz are intergrown with the cementing and replacement calcite; it seems likely that these types of quartz represent precipitates of silica mobilized by the widespread replacement by calcite. Only one sample of sandstone (1271) shows cementation with quartz. Volumetrically, however, and judging from the samples collected, these occurrences

of secondary quartz do not seem sufficient to account for all the silica that must have been replaced.

The amount of cementing and replacement calcite in the Peranera sandstones varies from less than 10% to over 80% of the volume of the rock. In the latter cases, only isolated grains and relics of grains float in the calcite (sample 1308). The most common amount of cementing and replacement calcite is from 20 to 40%.

Encrusting calcite occurs in some samples. This type of calcite consists of crusts or rinds composed of blocky or wedge-shaped crystals surrounding grains or aggregates of grains. Various crusts are similar to those found in the mudstones (p. 207) and to those found in the Malpas sandstones (p. 188). These types of secondary calcite in the sandstones closely resemble those found in recent caliche profiles (Nagtegaal, 1969).

Some of the calcite probably accumulated in near-surface horizons as a result of caliche processes or precipitation from run-off water; substantial amounts, however, may have been precipitated at somewhat lower levels from ground-water in the alluvial fans and below the steppe plains. The distinction between the two types is just as difficult in ancient deposits as in young deposits (Ruhe, 1967).

Although not found in the sections studied, barite (field determination) occurs in the Peranera deposits at several localities in the outcrop area shown in Fig. 5. The mode of occurrence is that of veins measuring not more than a few cm in thickness and running oblique to the stratification, and in pockets measuring up to 40 cm. This habit shows that the mineral crystallized during late stages in diagenesis. Barite occurs similarly in the Bunter deposits (p. 224).

Mudstones

Mudstone is used here as a general term for the fine-grained deposits occurring mainly outside the channel-fill deposits. Many of these rocks, however, consist of silt-sized material and should properly be called silt-stones; others contain appreciable amounts of clay-sized material.

Except for a higher content of phyllite debris (up to 60% in some instances), the original sediment of the mudstones is similar to that of the sandstones just treated. Diagenesis, probably in large part of a sub-aerial character, is more pronounced, however.

The mudstones contain more hematite than the sandstones; it may locally render the matrix almost fully opaque. A hematite content of 10 to 30% (volume, P.C.) in mudstones containing little or no replacement calcite is not uncommon.

Because of the fine grain-size, differentiation between authigenic hematite *in situ* and hematite transported in grains or as clastic particles is more difficult than in the sandstones. Nevertheless, on the basis of almost universal replacement relations, most of the hematite seems to be authigenic, which is the reverse of the

Fig. 50. Coarse-grained, moderately sorted Peranera sandstone. In the centre, the ghost structure (G) of a grain entirely replaced by calcite lying in a syntaxial relation to sparry calcite cement (C). The calcite cement has replacement contacts with four quartzite grains, thus producing an apparent loose packing of the grains. The black spots in the quartzite grains are hematite, formed prior to clastic deposition. The microspar limestone grain (MS) at the lower right is of Devonian origin. Sample 1302, nicols crossed.

Fig. 51. Peranera nodular mudstone. Relict shards of mudstone partially replaced by calcite (M, 30—40% calcite) and set in a mass of concretionary calcite representing mudstone almost entirely replaced by calcite (MC, 90—100% calcite). Calcite occurs in the form of microspar (MC) and spar (C); the sparry calcite probably fills former voids. The mudstone consists largely of phyllite debris stained a medium reddish-brown by hematite. Sample 1259, nicols crossed.
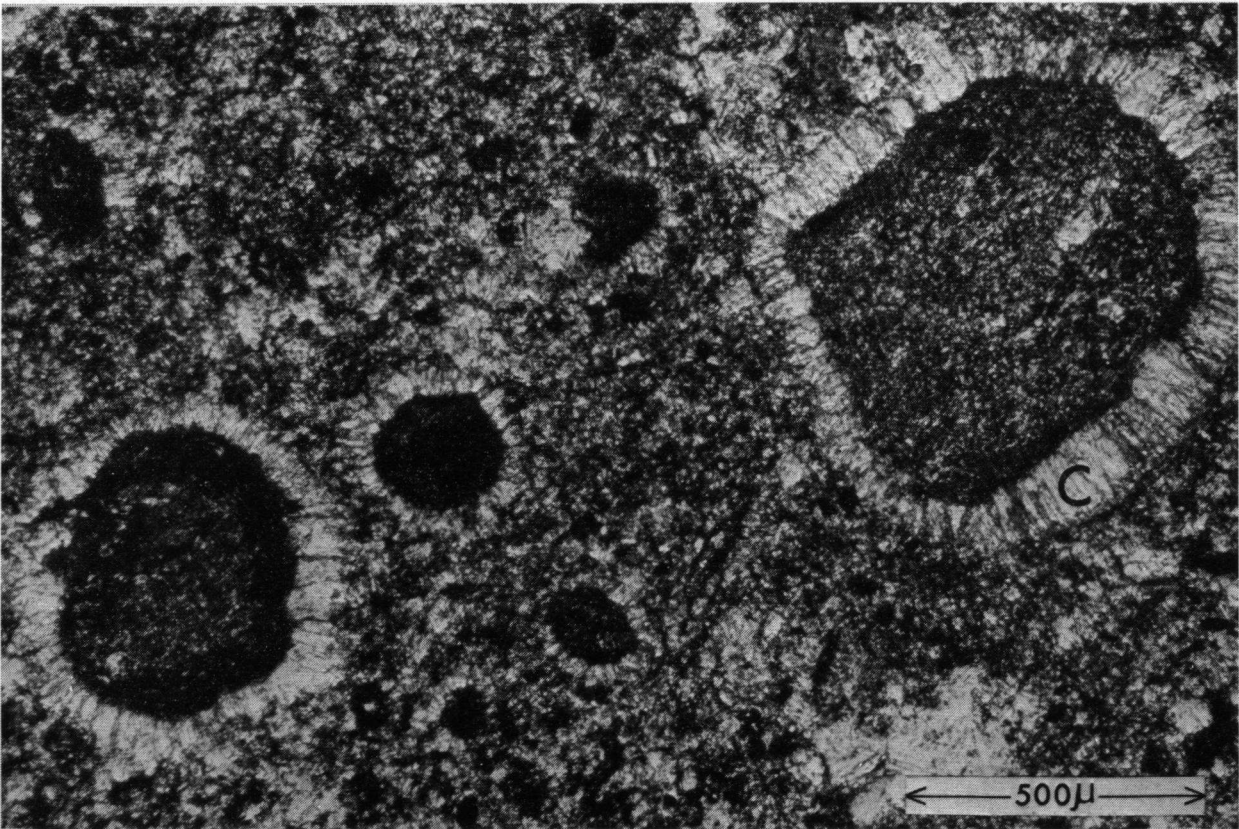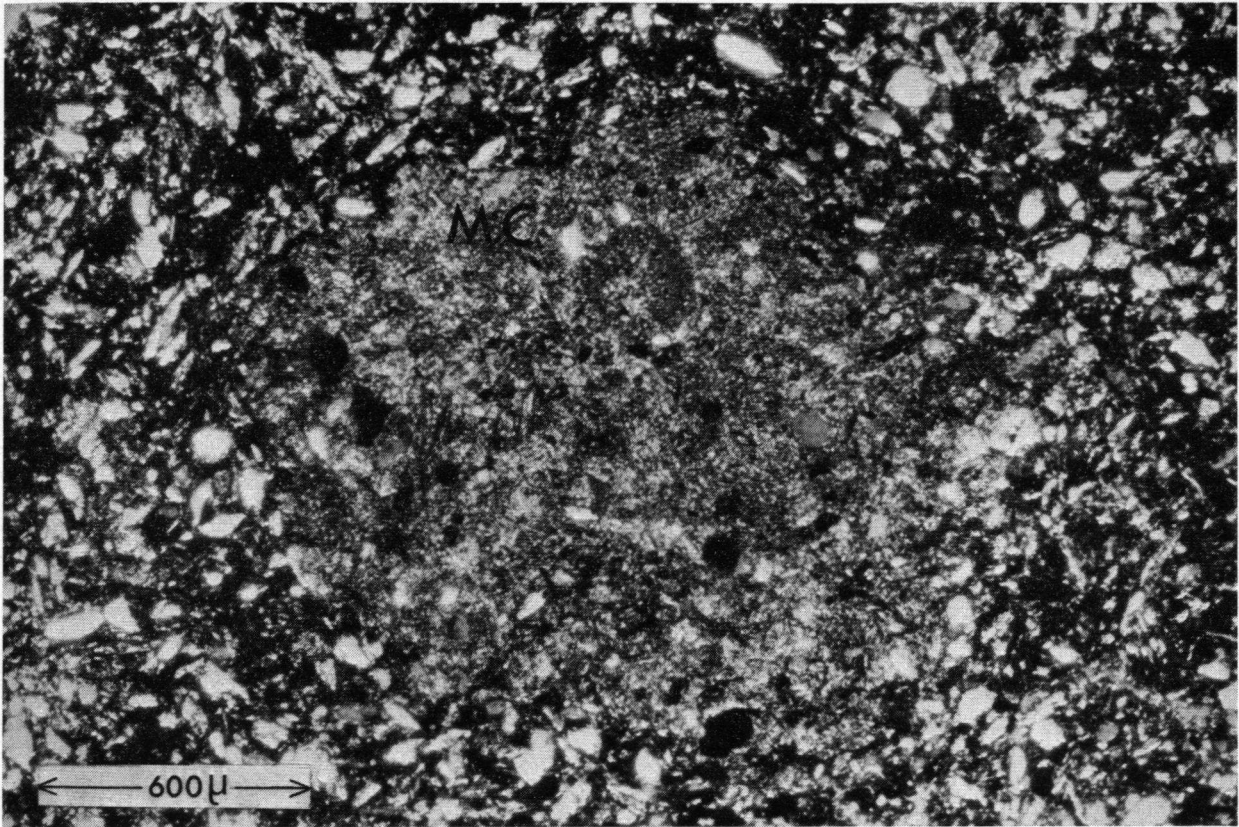
relations in the sandstones, where the authigenic hematite occurs only in relatively small quantities (p. 203).

As in the sandstones, the calcite in the mudstones was mainly introduced after the ferric oxides had precipitated. This is shown by widespread replacement of hematite by calcite and by numerous inclusions of colloform hematite aggregates in calcite concretions. Particularly in mudstones carrying calcite concretions, replacement of mudstone by calcite is volumetrically important. The calcite concretions (densely packed and well-displayed in the 40 to 80 m interval; section 6, sheet IV) must have formed chiefly by bulk replacement of mudstone. This conclusion is reached by the analysis of the relations of calcite and mudstone as shown in Fig. 51, which is a microphotograph of a sample taken from a calcite concretion. This sample shows shards of less completely replaced hematitic mudstone in a rock consisting almost entirely of fine-grained calcite.

As is the case for sphaerosiderite in the Malpas mudstones (Fig. 40) and dolomite phenocrysts in the Bunter mudstones (Fig. 68), small concretions in the Peranera mudstones show a reorientation of clastic particles in their direct vicinities (Fig. 52). The small concretion shown in Fig. 52, which has a diameter of a little more than 1 mm, must have formed largely by replacement, as may be concluded from the numerous included, only partly 'digested', clastic fragments. Whether the reorientation of the clastic fragments bordering the concretion resulted from an outward-directed pressure of the concretion growing in uncompacted mud, or whether it resulted from pressure exerted by compaction, is not known. In either case, however, the nodule should be of a pre-compacition origin. The marked similarity of the numerous Peranera calcite nodules to those in recent caliche (p. 161), of course leaves no doubt as to the early diagenetic origin of the former.

The replacement of the mudstone shown in Fig. 53 was evidently halted at a stage when only some spherical relics of mudstone were still present. The replacement may have been interrupted by the development of calcite crusts which shielded the spherical mudstone relics from pore waters and hence from further replacement. The calcite crusts are of the same type as those in the Peranera sandstones; as in the

Fig. 52. Peranera mudstone. Small microspar calcite concretion in mudstone stained dark by hematite (black). In the interior of the concretion, calcite replaces about 95% of the mudstone. Clastic grains in the direct vicinity of the concretion are oriented parallel to the concretion surface. Sample 1294, plain light.

Fig. 53. Spherical mudstone replacement relics carrying envelopes of encrusting calcite; Peranera Formation. The mudstone relics contain little calcite (5—10 vol. %); outside of the relics, about 95% of mudstone has been replaced by microsparry calcite. Sample 1296, nicols crossed.

sandstones, they occur in only relatively few samples (1296, 1301, 1306).

The calcite in the Peranera mudstones is, with respect to its habit, similar to that described for the sandstones (p. 203); but in the cases of cementing and replacement calcite, it has a much finer grain. The presence of the abundant secondary calcite is attributed chiefly to the formation of caliche, and in part to cementation by ground-water at somewhat deeper levels in the deposits.

Although the mobilization and precipitation of calcite by caliche processes must have taken place almost continuously during the accumulation of the Peranera Formation, the effects are most pronounced in the lowermost 80 m of the formation (section 6, sheet IV). At 112 m above the base of the section there are lenses up to 30 cm thick and composed of an extremely fine-grained, light-coloured limestone. As far as the author is aware, this is the only level in the Peranera Formation containing almost pure limestone. The limestone lenses are underlain by a mudstone with calcite nodules. The limestone shows parallel laminations and contains calcite spherulites; it is therefore interpreted as the calcrete in a caliche profile.

The fact that only one complete caliche profile occurs in a thick sequence of deposits in which caliche processes may have been almost continuously at work, may be partially due to high rates of deposition of clastic material, although the climate proper must have been, of course, of primary importance (Nagtegaal, 1969).

The development of caliche is mainly a pedogenic process (Ruhe, 1967). Many of the levels containing abundant cementing, replacement, and encrusting calcite, in particular the calcite nodules, could be considered to correspond to the B horizons in paleosols. Evidence of the presence of paleosols is also provided by a textural analysis of the mudstones.

Modern pedology makes an increasing use of thin-section studies, the basis of the microscopic approach having been laid in the early thirties by W. L. Kubiena (Pijls, 1964). This type of research, which is generally referred to as 'soil micromorphology', has evolved rapidly and contributes considerably to our understanding of the nature of soils. The wealth of data collected has great value for the sediment petrographer, particularly if he is concerned with continental deposits. Unfortunately, the terminology used by the pedologists is very different from that of sedimentary petrography; most of their terms and concepts have to be converted into geological language.

The microscopical study of the Peranera paleosols was based on the criteria applied by Brewer (1964) for the microscopical analysis of soils. Several of the common elements of soils, such as 'micropeds' (small aggregates of soil), 'closed planes' (compressed planar voids), and 'pedotubules' (tubules filled with soil material) can be observed in the Peranera mudstones.

The pedotubules may have been formed by burrowers and roots; evidence of that both were once present is
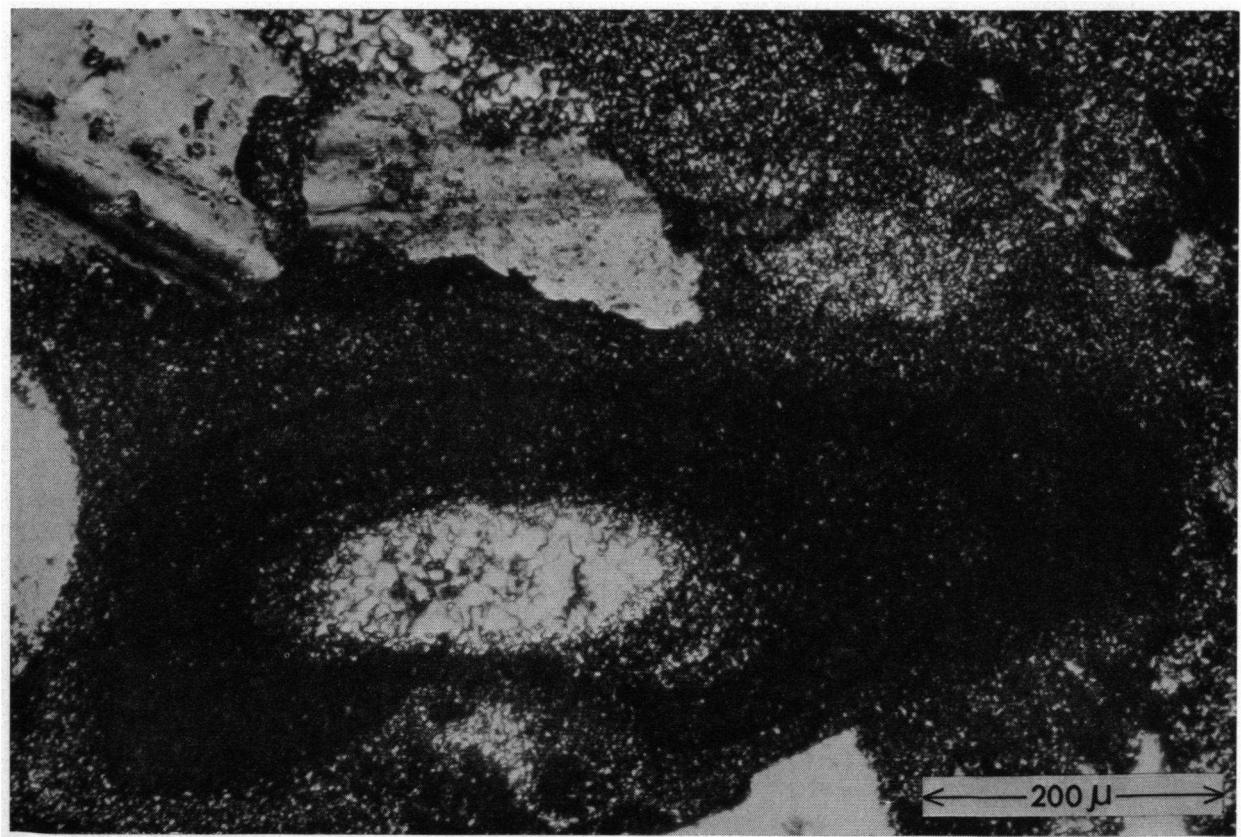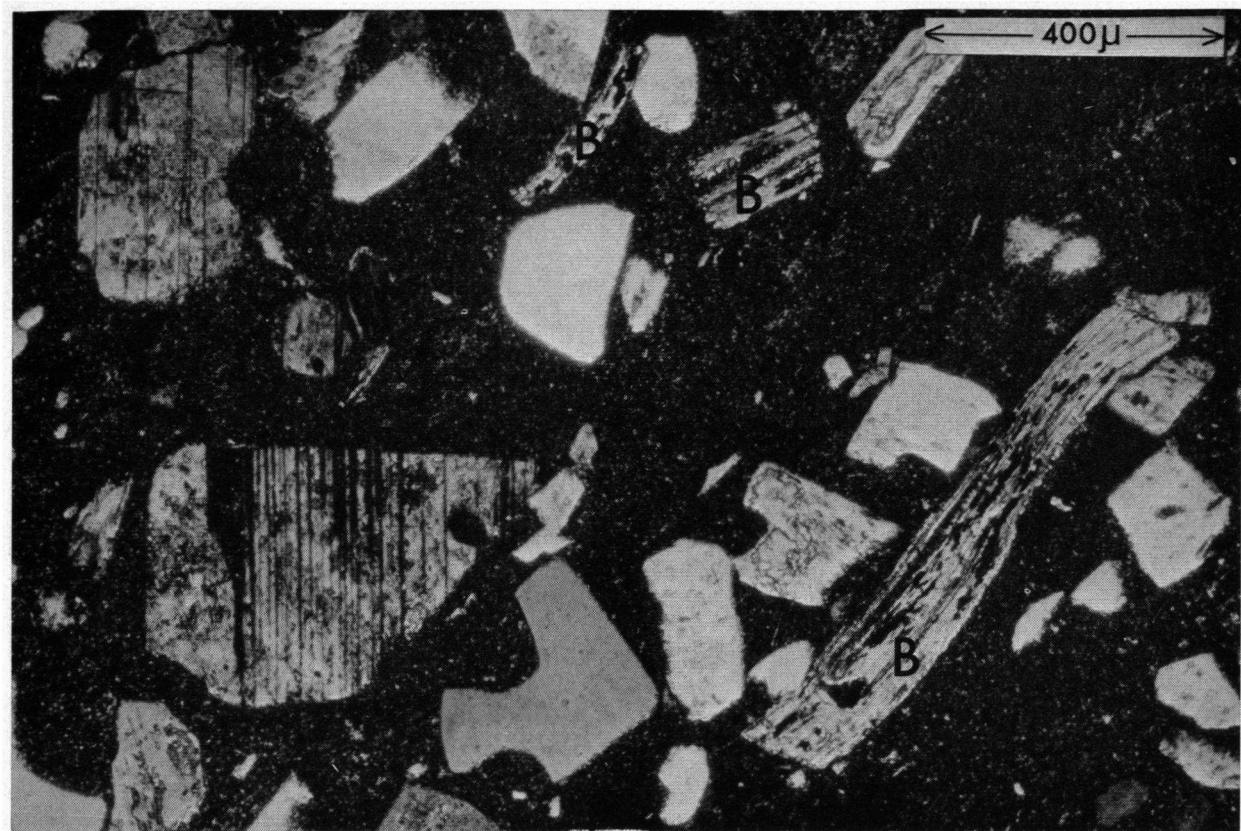
clearly observable in the field (section 6, sheet IV). In some samples of both sandstones and mudstones, the clastic components are arranged in whorl-like patterns thought to result from bioturbation. Some evidence of the destruction of soils by penecontemporaneous erosion is provided by 'sandstones' consisting almost entirely of clasts composed of small aggregates of mudstone, presumably micropeds.

Paleosols such as those occurring in the Peranera deposits were formed in an aggrading system. As previously noted, it follows that the intensity of soil formation, which depends first of all on the climate, must also be related to the rate of deposition. In this respect it is of interest to note that the Bunter paleosols show stronger weathering of components. This would agree with the suggested weaker erosion and presumably lower rates of deposition in the Bunter if compared with Peranera times, but it almost certainly relates to a more humid climate in the time during which the Bunter was deposited (p. 223).

Tuff beds

The tuffs intercalated in the Peranera deposits range in composition between crystal tuffs and vitric tuffs, according to a classification proposed by Pettijohn (1957, p. 332).

Crystal tuffs. — The crystals in the crystal tuffs consist of feldspars, quartz, olive-green (altered) biotite, and accessory zircon and iron ores. The crystals commonly float in an extremely fine-grained groundmass (Fig. 54). The composition of a representative crystal tuff containing an insignificant amount of secondary calcite is as follows: groundmass: 54.8%, feldspars: 34.8%, quartz: 12.6%, biotite: 6.8% (sample 1272; P.C. analysis, 300 points counted).

Among the feldspars, which are only slightly altered, there are a few orthoclase crystals, over 95% consisting of well-twinned, non-zoned oligoclase/andesine. The quartz phenocrysts are commonly non-undulatory

Fig. 54. Peranera crystal tuff bed. Oligoclase/andesine, quartz, and biotite (B) crystals set in a groundmass now composed of chert and minute kaolinite crystals. The groundmass is lightly stained by hematite. The quartz grain just below the centre shows well-developed magmatic resorbtion embayments. The biotite is bleached; the black inclusions are hematite. Sample 1272, nicols crossed.

Fig. 55. Peranera crystal tuff bed. Liesegang's rings composed of hematite concentrically arranged around a former vesicle. The rings invade the groundmass of an oligoclase/andesine-quartz-biotite crystal tuff. The groundmass is now composed of chert containing minute crystals of kaolinite. All the biotite in the thin section is strongly bleached. The hematite rings testify to the mobility of what probably were colloidal ferric hydroxides derived from the early diagenetic oxidation of biotite and the vitric groundmass. The vesicle is filled with chert, the texture of which coarsens inwards. Sample 1268, nicols crossed.

and show pronounced embayments (concave rounding) and occasionally good rounding (convex rounding, Fig. 54). Both types of rounding can be satisfactorily explained by isothermal magmatic resorption due to pressure release during the extrusion of the magma (Tröger, 1967, p. 158).

Most biotite shows signs of alteration. The presence of vermicullite, a probable alteration product after biotite (Walker, 1949), was demonstrated by the X-ray analysis of a Peranera tuff bed containing altered biotite. Oxidation of the iron set free by the alteration of biotite, resulting in the formation of enclosed aggregates of hematite, is clearly displayed in numerous biotite flakes (Fig. 54). The oxidation of the biotite must have occurred *in situ*, since the groundmass of the tuff is usually stained a greyish-red in the immediate vicinity of the grains. The stain commonly becomes lighter going away from a biotite flake (sample 1272). In other samples, complete oxidation of the biotite gives the entire rock a greyish-red colour, while only ghosts of the biotite flakes are preserved (1268, 1267, 1276, 1273). These observations of the *in situ* breakdown of iron-bearing minerals and consequent staining of the surrounding rock, agree with the findings of, among others, Branson (1915), Tomlinson (1916), Robb (1949), Reifenberg (1950), Valeton (1953), Shotton (1956), Razumova (1960), and Walker (1967a).

Only two chemical determinations of the total iron content of the tuffs were carried out: sample 1276: 2.16% and sample 1273: 1.68%. The iron content is lower than that in the Erill Castell tuffs (Table V). The low values of iron in the Peranera tuffs agree with the suggestion that the origin of reddish hues of the tuffs could entirely be a matter of *in situ* mobilization and precipitation of the iron.

There is evidence that after its formation from biotite, and from other sources such as the volcanic glass dust (groundmass of the tuffs), ferric iron was transported over short distances within the tuffs, probably in the form of colloidal ferric hydroxides. Halos of hematite (Liesegang's rings) often invade the groundmass of the tuffs from vesicles which were probably interconnected. The vesicles proper are now filled with chalcedonic quartz, the crystal aggregates of which normally coarsen inwards (Fig. 55).

The greyish-red stain originating from the *in situ* oxidation of biotite is the second demonstrable mode of formation of reddish hues in the Peranera Formation. As already shown, reddish hues also result from hematite contained in clastic grains derived from ferric upland soils (p. 203).

The origin of the authigenic fine-grained hematite aggregates in the sandstones is not known with certainty. They probably also formed from iron liberated by decomposition of iron-bearing minerals, but they may also have derived partially from run-off water (see discussion on ferric oxides in the Bunter Formation, p. 216).

The fine-grained groundmass of the crystal tuffs
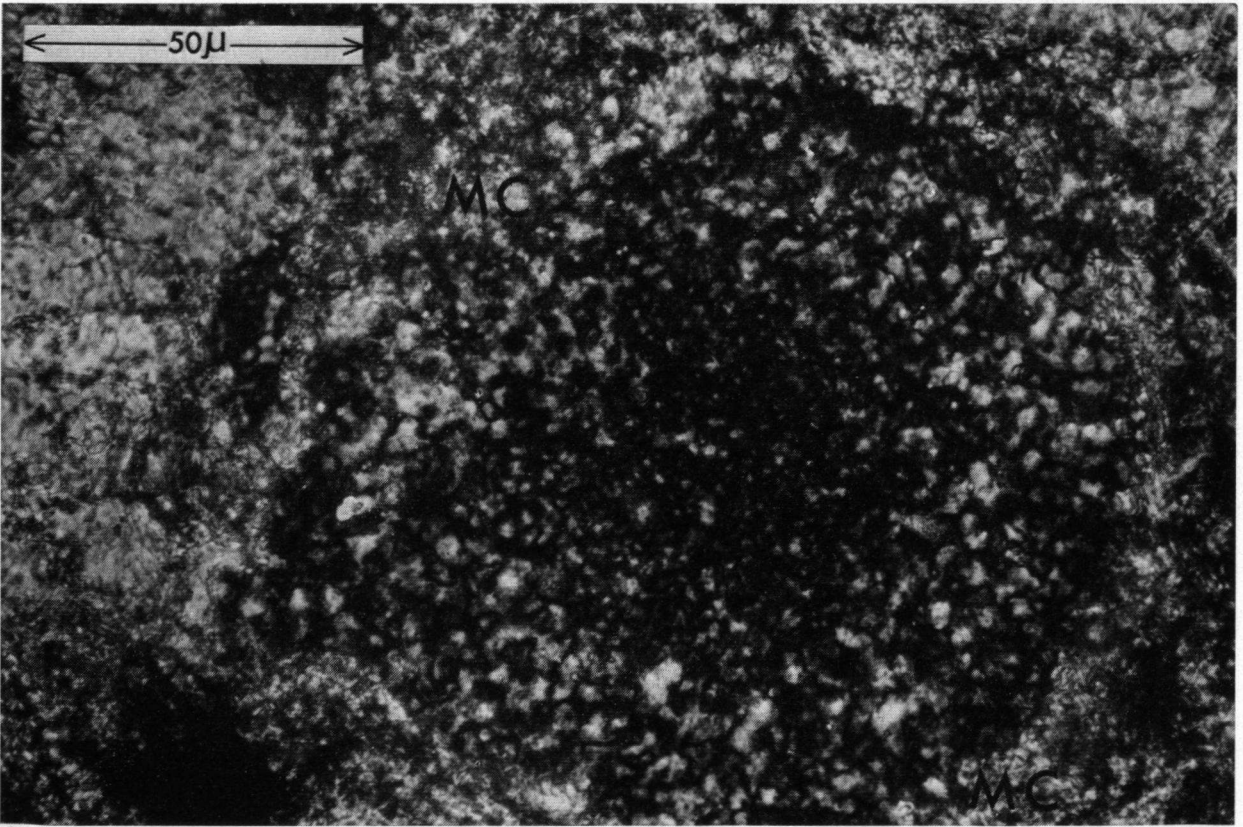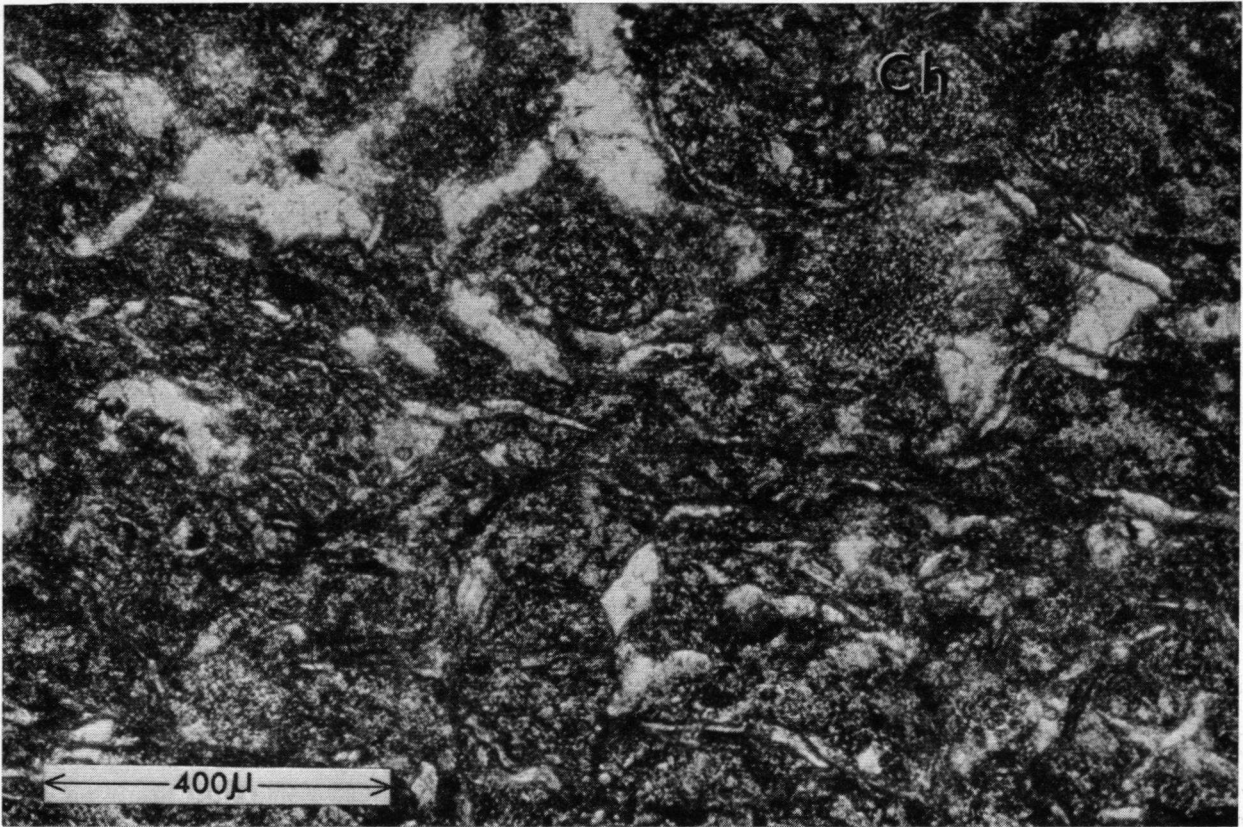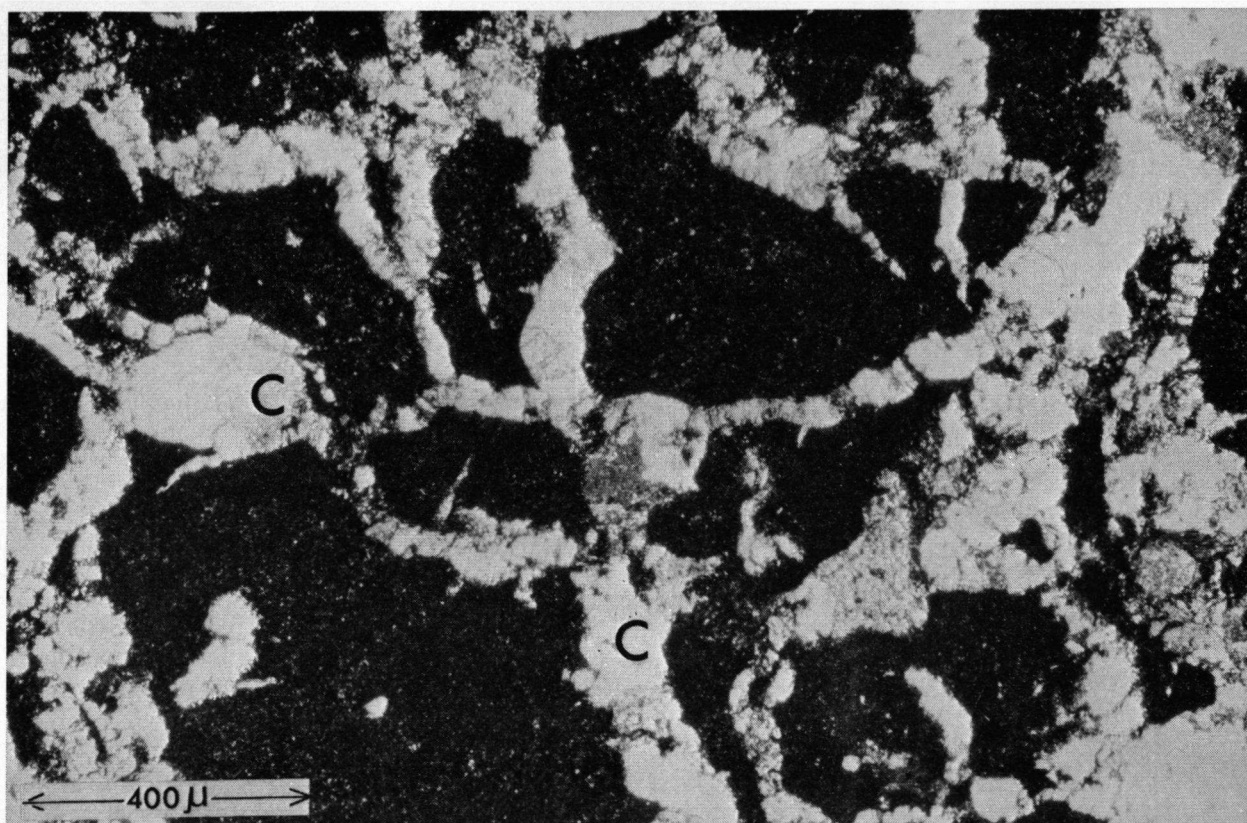
Fig. 58. Peranera fine-grained vitric tuff, septarian nodule. The angular fragments are *in situ* but cannot be exactly matched to each other. Sparry calcite (C) therefore replaces the tuff, but was probably introduced along cracks. Sample 1260, plain light.

consists of tightly intergrown chalcedonic and microcrystalline quartz and kaolinite. These minerals are probably devitrification products after finely divided pumice or glass dust. Because of the fine grain-size, optical differentiation of the chert and kaolinite is difficult, but staining the kaolinite with methylene blue readily brings out the distribution of both minerals (p. 181). The relative amounts of chert and kaolinite vary widely, but chert predominates in all cases.

The intensity of alteration of the tuffs varies from sample to sample, which is probably due to differences in the type of and time of exposure to penecontemporaneous weathering processes. The groundmass is completely altered in all cases, but the amount of secondary calcite and the intensity of the red coloration vary strongly.

Fig. 56. Peranera vitric tuff bed. Vitroclastic texture preserved in a vitric tuff largely replaced by calcite. Chert spherulites (Ch) are common in this sample. Sample 1266, nicols crossed.

Fig. 57. Peranera vitric tuff bed. Chert spherulite, probably formed during devitrification of the vitric groundmass of the tuffs. The microphotograph shows the increase in coarseness of the chert texture towards the periphery of the spherulite, as well as the partial replacement by microsparry calcite (MC). Sample 1266, nicols crossed.

Vitric tuffs. — The majority of the tuffs are vitric tuffs, some of which show a non-welded vitroclastic texture. Nowhere, however, is any glass preserved. Arcuate and linear shreds, which originally must have been composed of glass formed by rapid cooling of lava, now commonly consist of replacement calcite (Fig. 56).

Most of the vitric tuff shows the same composition and texture as the groundmass of the crystal tuffs just described. Most samples of this dense rock contain some small feldspar phenocrysts and somewhat larger biotite flakes and devitrified glass shreds.

Many samples of vitric tuff also contain distinct spherulites composed of microcrystalline quartz. Commonly, the texture becomes coarser towards the peripheries of the spherulites (Fig. 57).

The chert spherulites must have formed authigenically, as a result of devitrification. Their authigenic origin is demonstrated: (1) by the fact that some spherulites were clearly hindered in their growth by nearby phenocrysts or other spherulites, and (2) by instances of incipient spherulitization in the form of interconnected, vaguely concretionary aggregates of chert. The globular shape of the spherulites may result from drop-like concentrations of silica gel formed from supersaturated (with respect to amorphous silica) silica solutions in the tuffs.

The formation of the spherulites, and as a consequence,

the devitrification of the tuffs, must have progressed fairly far when secondary calcite was precipitated in the tuffs. This time-sequence is demonstrated by the frequent replacement of the spherulites by calcite. All degrees of replacement are observed; in one sample (1262) calcite spherulites showing a radial internal structure are simply pseudomorphs after the micro-crystalline quartz spherulites.

The fine-grained vitric tuffs show a special type of concretions. These concretions are oval or almost round, lie parallel to the stratification, and range in largest diameter from 10 to 30 cm. On the basis of these characteristics, they are easily distinguished from the concretions found in the paleosols (p. 161). When broken with the hammer, the concretions invariably show an irregular, dense septaria-like network of calcite veins. In thin section the internal structure is as shown in Fig. 58. Fragments of devitrified fine-grained tuff can more or less still be fitted together, but much 'lost' space is occupied by the sparry calcite. Calcite must therefore have partially replaced the tuffs, but may have initially been introduced along numerous cracks. It is probable, but difficult to demonstrate,

that these cracks formed as a result of the dehydration of a gel (which could have existed in the course of the devitrification process of the tuffs) and that the calcite was introduced at an early stage.

BUNTER FORMATION

The petrography and diagenesis of the two main rock types of the Bunter Formation, the sandstones and mudstones, will be successively treated.

Sandstones

Original sediment, provenance. — The fluvial deposits described so far, i.e. those of the Aguiró, Malpas, and Peranera Formations, contain vast quantities of unstable rock fragments whose sources can be traced to a terrain situated to the North of the study area, as confirmed by the paleocurrent data. The clastic components of the Malpas deposits came from a temporary source, the Erill Castell Volcanics, but the components of the Aguiró and Peranera deposits were largely derived from rocks now making up the Paleozoic core of the Pyrenees.

TABLE X

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (quartz, muscovite, phyllite debris) 11%?	quartz	47			reworked *in situ* (burrowers, plants).
	quartzite	5	ferric oxides (largely hematite)	9*	colloform; some pseudomorphs after
Max. grain-size: 175 μ	muscovite	3			goethite needles; replacing quartz
	chert	1			
Quartz: angular to subrounded	phyllite	x	quartz	20	secondary overgrowths on all grains
	zircon	x			
Many quartz grains elongate (length: width ratios up to 10)	tourmaline	x	kaolinite	3	alteration product from muscovite; replacing secondary quartz
			dolomite	1	anhedral; replacing ferric oxides, quartz overgrowths, and kaolinite
					no interpenetration of clastic quartz grains

Table X. Bunter Formation; fine-grained sandstone. Mean values after samples 1454 and 1455 (300 points counted in each sample; little difference in coarseness).

TABLE XI

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (quartz, muscovite, phyllite debris) 9%?	quartz	49			quartz grains pitted
	quartzite	2	ferric oxides (largely hematite)	3	colloform; replacing quartz grains
Max. grain-size: 360 μ	muscovite	2	quartz	21	secondary overgrowths
	chert	x			
Quartz: angular to rounded, most grains subrounded	phyllite	x	kaolinite	14	aggregates in pore spaces; alteration product from muscovite; replacing secondary quartz
	zircon	x			
Poorly developed dust rings	tourmaline	x			
Parallel laminae (parallel to cross-bedding)			dolomite	x	subhedral crystals
					some muscovite flakes deformed by compaction; no interpenetration of clastic quartz grains

Table XI. Bunter Formation; medium-grained sandstone.
    Mean values after samples 1461 and 1498 (300 points counted in each sample; little difference in coarseness)

The paleocurrent data of the Bunter Formation point to source areas in more or less the same terrain (p. 168). In sharp contrast to the described deposits, however, no unstable rock fragments, except insignificant amounts of phyllite, biotite, and altered feldspar, are found in the Bunter. The bulk of the Bunter sandstones is composed of quartz grains admixed with varying but low quantities of quartzite, chert, and muscovite (Tables X and XI).

The primary composition of the Bunter sandstones is in no way comparable to the composition of the sands supplied from the Axial Zone of the Pyrenees at present (Appendix, p. 229).

The problem of the aberrant composition of the Bunter sandstones therefore concerns two questions: Did the composition of the Bunter source areas differ entirely from that of the Aguiró and Peranera Formations? And, if not, why does the composition of the Bunter sandstones reflect the composition of the source areas so unfaithfully?

There is direct evidence that in Bunter times a new kind of rock previously untapped, had appeared in the supply areas: the granodiorites. This evidence is found at localities where the Bunter deposits directly overlie granodiorite (as at Bielsa, 50 km NW of the section studied).

The presence of granodiorites in the Bunter drainage areas could partially account for the high percentage of quartz grains in the sandstones. A similar influence of the granodiorites is found in the composition of the recent river sands: the sands of the Ribagorzana River, which drains a large area composed of granodiorites, contain 22% quartz grains [1] (Appendix, p. 229).

The granodiorites, however, cannot have been the sole contributors of clastic material, as is clear from the analyses shown in Tables X and XI. Besides quartz there are quartzite and chert grains. Both these types of grains, of course, stem from non-igneous rocks. The quartzite grains are similar to those found in the recent fluvial sands; they are of either Devonian or Ordovician origin (Appendix, p. 229). The chert is similar to the cherts occurring in the Carboniferous (Mey, 1968); this was confirmed by comparison of the relevant thin sections. The majority of the pebbles in the Bunter conglomerates consist of quartzite (most likely from an Ordovician source), a fact also noted by previous authors (Roger, 1965).

It may therefore be concluded that the Paleozoic formations that supplied the bulk of the clastic material in the Aguiró and Peranera deposits, also occurred in the Bunter drainage areas.

This leads us to the question of why so many of the Paleozoic components such as the various types of phyllite and limestone fragments, are absent in the Bunter sandstones.

The two main factors in fluvial supply must be considered in this context: (1) the morphologic setting, and (2) the prevailing climate.

With exclusion of the conditions during deposition of the Iguerri member (see below), the morphologic setting during the accumulation of the Bunter deposits must have been one of extensive lowlands (p. 171). Such a situation is unfavourable for vigorous, deep-cutting erosion, and is therefore unfavourable for the supply of fresh, unstable rock fragments. Under these conditions, it may also be expected that the rate of fluvial deposition in the areas of aggradation was relatively low.

On the basis of the sediment structures and the strong *in situ* weathering (see below), it was concluded that the prevailing climatic conditions during the accumulation of the Bunter deposits were those of the savannah (tropical, seasonal, and moderately humid) (p. 171). To summarize the foregoing, it may be said that both the topographic setting and the climate prevailing during the accumulation of the Bunter deposits may have favoured selective weathering. The result of this process must have been that, in the sand sizes, only those components most stable under weathering conditions, such as quartz, quartzite, chert, and muscovite (Goldich, 1938), were available for transport. It is thought that selective weathering must have been the main factor determining the mineralogical maturity of the Bunter sandstones.

The heavy minerals in the Bunter sandstones (concentrates from crushed samples) consist mainly of tourmaline (rounded and less rounded prismatic forms, brownish and blueish varieties), zircon (rounded and rounded prismatic, colourless and pink varieties), and rutile (rounded). Alterite (heavy minerals altered beyond recognition) occurs with equal frequency. Much less frequent are garnet, andalusite, epidote, hornblendes, corundum, brookite, and anatase. The last two minerals occur in all samples investigated (8 in all); they are probably authigenic. Virgili (1960) found a similar heavy mineral assemblage in the Bunter formation in the Ribagorzana valley.

Among the heavy minerals, tourmaline, zircon, and rutile range among the stablest in average weathering (Pettijohn, 1941; Smithson, 1941; Sindowski, 1949). The stability of the Bunter heavy-mineral assemblage agrees with the suggested penecontemporaneous strong weathering, but it cannot be accepted as evidence, since the stability of the assemblage could in large part be due to recycling from the Paleozoic formations. Some evidence of deep weathering is found in the occurrence of markedly elongate quartz grains in the finest sand-sizes (Tables X, and XI). The size and shape of these grains agree with those of the quartz grains in some phyllite and sericite/chlorite quartzite from the Paleozoic basement. It is probable that they were liberated from these sources by means of weathering.

The results of two chemical analyses of Bunter sandstones collected at Cherallo (Fig. 1) were kindly put at the author's disposal by the management of the El Pirineo cement factory (E.N.H.E.R. Company, Spain), and are shown in Table XIV. The SiO_2 con-
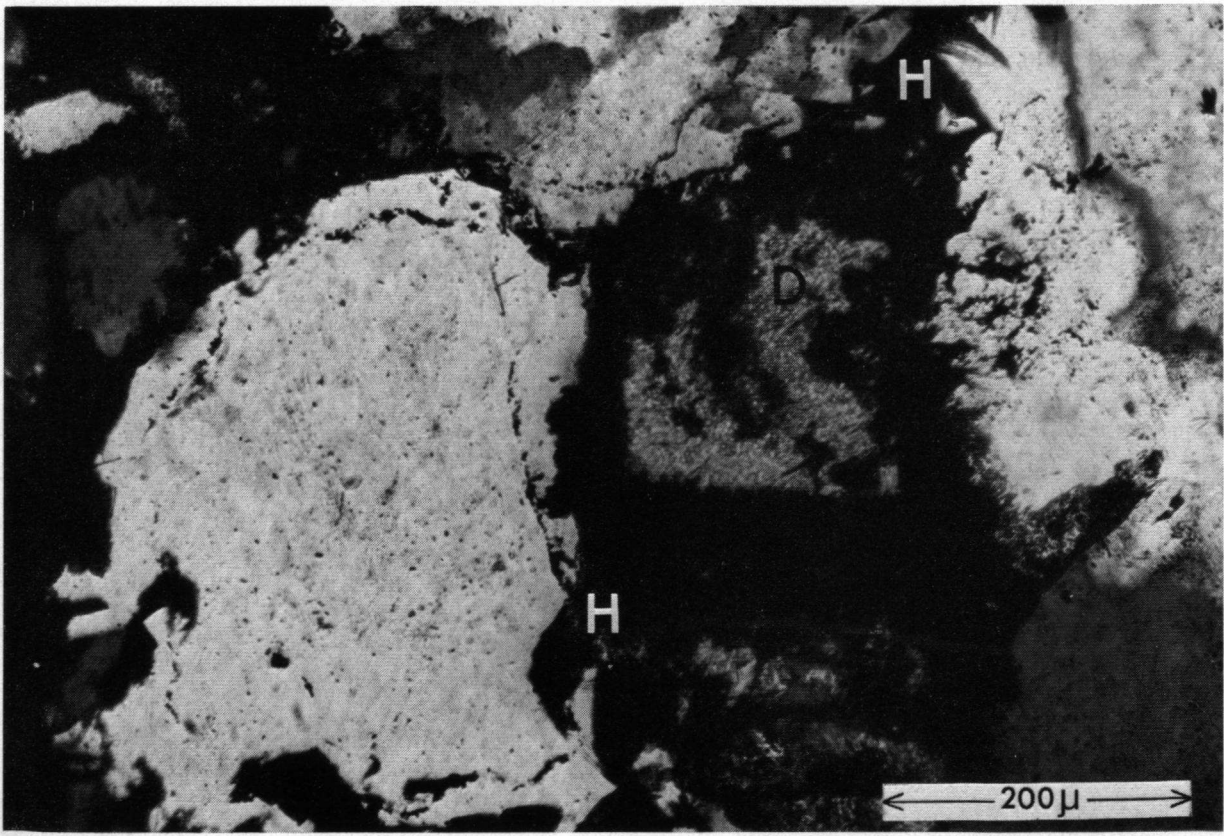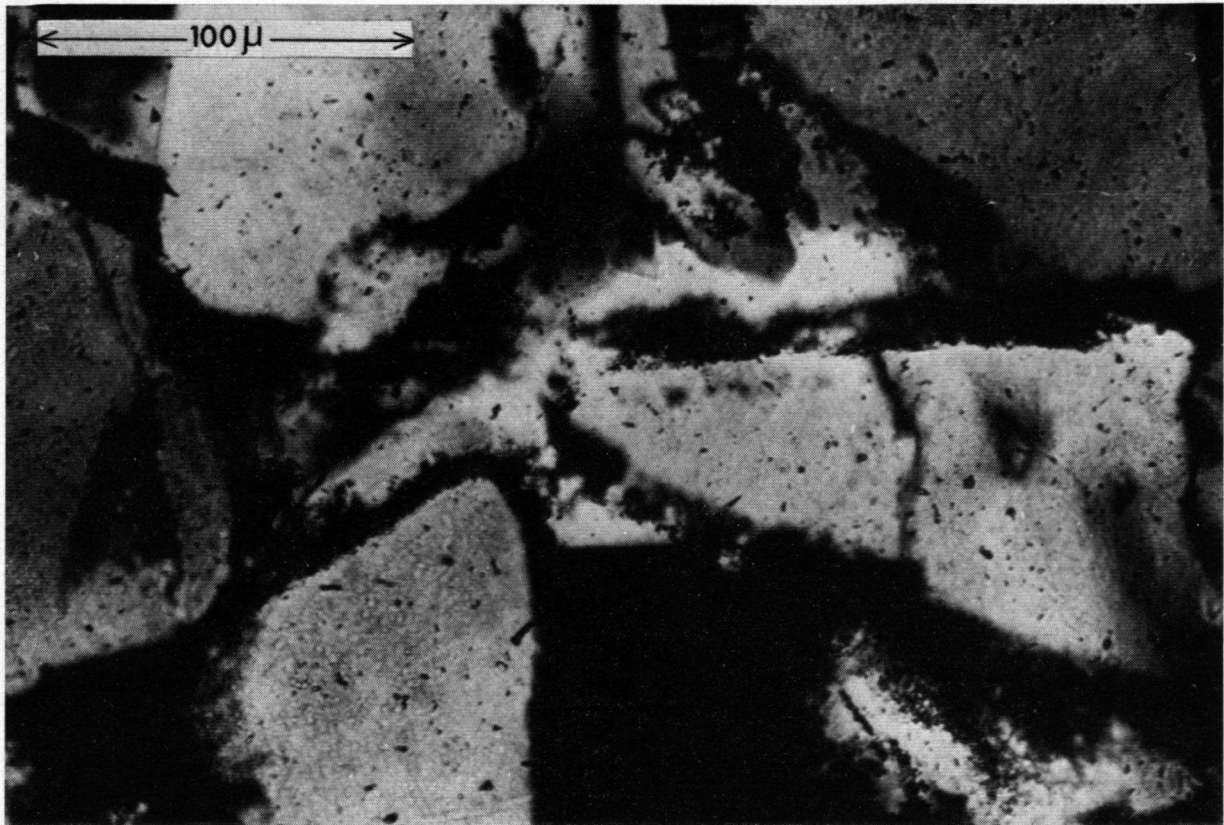
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[1] See footnote on p. 177.

TABLE XII

ORIGINAL SEDIMENT			DIAGENESIS		
Texture	Detrital grains	%	Authigenesis	%	Texture
Matrix (quartz, phyllite) 5%?	quartz	36	ferric oxides (largely hematite)	1	small colloform patches
Max. grain-size 1.2 mm	ser/chlor. quartzite	15	quartz	2	small patches of cement
	fine-grained limestone	9			
Quartz, quartzite: angular to subrounded	non-calc. phyllite	6	kaolinite	2	alteration product from muscovite
Phyllite: rounded	silty phyllite	4			
	feldspar (fully altered)	4	dolomite and calcite	13	main cement; microspar to spar; replacing silicate clasts and matrix
	calc. phyllite	2			
	muscovite	1			phyllite compaction-moulded; lmst grains strongly corroded
	zircon	x			
	tourmaline	x			

Table XII. Bunter Formation: sandstone from Iguerri member.
Mean values after samples 12a and 13b (300 points counted in each sample; little difference in coarseness).

tent is equal to that of the average sandstone, Al_2O_3 and Fe_2O_3 are higher, MgO is lower, and CaO is roughly the same.

These chemical data are given without comment; their significance is not fully clear in view of the still limited amount of chemical data on sandstones in general and in view of the complex diagenesis in the Bunter sandstones (see below).

As has already been pointed out, the Iguerri member of the formation resembles the Aguiró deposits with respect to its geological occurrence (p. 149).

The clastic assemblage and diagenesis of the Iguerri sandstones, however, are very different from those of the Aguiró sandstones (Tables XII and IV), which excludes the possibility that the Iguerri member could be mistaken for another occurrence of the Aguiró Formation.

The clastic assemblage and diagenesis of the Iguerri sandstones can probably best be considered as intermediate between those of the Peranera and Bunter sandstones.

Fig. 59. Fine-grained, moderately sorted Bunter sandstone. Quartz grains are cemented by hematite (black) and quartz. Thick colloform aggregates of hematite envelop the quartz grains and occur also in larger, pore-filling aggregates (on the left and below centre). The hematite (formerly hydrous ferric oxides), which is the first cement formed, corrodes the quartz grains. The second cement is quartz, which completely fills the remaining pore spaces. Sample 1454, nicols crossed.

Fig. 60. Coarse-grained, well-sorted Bunter sandstone. On the left, the outline of a clastic quartz grain indicated by a dust ring composed of hematite. The dust ring probably does not completely coincide with the original grain boundary; its pitted appearance suggests some solution of quartz prior to the syntaxial crystallization of the secondary quartz surrounding it. The deposition of secondary quartz was followed by the formation of ferric oxide, which shows some crystal outlines (hematite). The hematite penetrates the secondary quartz and the clastic quartz grain. The stage at which formation of the dolomite rhomb (D) took place cannot be deduced with certainty from the texture, but in the Bunter sandstones, dolomite generally crystallized during later stages. Sample 785 (Flamisell valley), nicols crossed.

Similarities to the Peranera sandstones are the presence of considerable amounts of unstable components (various types of phyllite and Devonian limestone carrying tentaculitid fragments), and the presence of a mainly calcitic cement.

Unlike the Peranera deposits, however, the clastic components contain no ferric oxides; all the ferric oxide appears to be authigenic, as is also the case for the Bunter sandstones. Another point of similarity to the Bunter sandstones lies in the fact that the Iguerri sandstones carry muscovite and strongly altered feldspar.

In view of the field evidence and petrographic data, it is suggested that the Iguerri deposits are probably an incidental remnant, in the form of a steep-sided valley-fill, of the period of strong erosion which occurred after the deposition of the Peranera Formation.

Diagenesis. — The Bunter sandstones have a highly complex diagenetic history, but the effects of the following processes are nevertheless clearly recognizable:

precipitation of and replacement by ferric oxides [1], dissolution of quartz and precipitation of secondary puartz,
kaolinization of muscovite,
precipitation of and replacement by dolomite.

There is little doubt that the formation of dolomite was among one of the last processes to occur (still after dolomite, accessory barite formed; p. 224), but it is difficult to arrange the other three processes in strict chronological order. As far as the evidence offered by the thin sections is concerned, they could have been going on simultaneously or have overlapped each other in time.

The absence of a strict chronological order applicable to all Bunter sandstones can be seen from Figs. 59 and 60. In the sample shown in Fig. 59, ferric oxide was clearly the first to precipitate. It occurs in colloform,

[1] See footnote on p. 202.

fine-grained aggregates directly on the quartz grain surfaces, and locally invades the quartz, which must be due to replacement of quartz by ferric oxide. Authigenic quartz is the second product; it completely fills the remaining pore space and tightly cements the fine-grained sandstone.

In the sample shown in Fig. 60, a hematitic dust ring indicates the outline of a clastic quartz grain. Secondary quartz appears as the first precipitate; it surrounds the quartz grain almost completely, at least in the two dimensions visible. Ferric oxide may be the second authigenic product; it replaced the secondary quartz and extends even into the clastic quartz grain.

The origin of the dust ring, however, is still uncertain. If it originated from *in situ* precipitation of ferric oxide, the secondary quartz would be a second precipitate as in the sample shown in Fig. 59, except that it was followed by the formation of much ferric oxide. If the dust ring was formed on the grain during some stage in its clastic transport, the secondary quartz would really be the first precipitate, and the relations in Fig. 60 would be the reverse of those shown in Fig. 59.

In the Bunter section studied, almost all the ferric oxide occurs as a first precipitate. As an apperent second precipitate after quartz, ferric oxide dominates locally, as in the Bunter along the Flamisell River, where the sample shown in Fig. 60 was collected.

In such three-component relations in one part of a thin section, between clastic quartz grains, authigenic ferric oxide, and secondary quartz, the chronological sequence of the diagenetic processes may only appear to be simple. The authigenesis of ferric oxides could, at some stratigraphic levels, have postdated the kaolinization to muscovite (p. 222).

The origin of the hematite in the Bunter Formation will be discussed first. Since many points also apply to the authigenic hematite in the Peranera red beds, frequent reference will be made to the latter deposits. The replacement relations of the authigenic hematite with other components, both in the Peranera and Bunter deposits, indicate that at one stage the iron must have occurred in the sediment in a mobile form. This form was most probably a ferric hydroxide sol or gel, as indicated by: (1) the blob-like, colloform habit of most of the hematite aggregates, (2) the extremely fine grain of the hematite aggregates, and (3) the Liesegang's rings of hematite invading the groundmass in the Peranera tuffs (p. 209).

Only a small percentage of the total amount of ferric oxide in the Peranera sandstones probably formed from gels; the equivalent in the Peranera mudstones is higher, whereas in the Peranera tuff beds all the ferric oxide may have formed in this way (p. 209). Petrographic evidence indicates that in the Bunter deposits, too, almost all the hematite may represent authigenic dehydrated ferric hydroxide gels.

The origin of the greyish-red stain is therefore quite different for the Bunter and the Peranera sandstones. In the Bunter sandstones the stain originates from

authigenic hematite, in the Peranera sandstones it originates chiefly from the greyish-red contained in rock fragments (p. 203). The difference is essentially between what Krynine (1949) has termed post-depositional red beds and primary red beds.

The ferric oxide contents of some Bunter sandstones, found by point counting, are included in Tables X and XI. These values were checked by chemical analyses. As was the case for the Peranera sandstones, the P.C. values are too high by a factor of about 3. The weight percentages of Fe_2O_3 found chemically are shown in Tables XIII and XIV.

TABLE XIII

Sample no.		Total Fe_2O_3 %
1455	sandstone	2.29
1463	"	3.03
1474	"	4.61
1496	"	4.22
1498	"	0.51
1470	mudstone	7.39
1489	"	4.37

Table XIII. Total Fe_2O_3 content of Bunter sandstones and mudstones. Analysed by Miss M. van Wijk.

TABLE XIV

constituents	A	B	C
SiO_2	78.90	78.02	78.33
Al_2O_3	7.70	7.89	4.77
Fe_2O_3	2.90	2.16	1.40
MgO	0.46	0.47	1.16
CaO	4.30	3.70	5.50
Loss on ignition	3.68	3.34	–

Table XIV. A and B: Partial chemical analyses of Bunter sandstones. Analysed by chemical laboratory, cement factory at Cherallo. C: Average sandstone, after Leith & Mead (1915).

Because no ferric-oxide-bearing rock fragments are found in the Bunter sandstones, the authigenic ferric oxide cannot be the result of possible mobilization from such a source, but must have formed either from: (1) intrastratal supply of iron from the decomposition of iron-bearing minerals, or (2) introduction of iron by run-off water.

In the Bunter drainage areas, the granodiorites were exposed (p. 213). Present-day fluvial sands derived from the granodiorites carry biotite (Appendix, p. 229). Although strong weathering conditions prevailed in Bunter times, it is likely that some biotite was included in the Bunter deposits, and X-ray analyses of bulk samples (1498, 1495) indeed show the presence of some biotite and of chlorite. The chlorite was probably derived from non-igneous Paleozoic formations (from

phyllites, sericite-/chlorite quartzites). Both biotite and chlorite, but also hornblendes, epidote, magnetite, and various clay minerals, may, as a result of intrastratal decomposition, have been sources of the authigenic ferric oxides. (references to the literature on the subject, p. 209).

In fluvially accumulated deposits, the introduction of some iron via run-off water is to be expected. This would hold especially for the Peranera and Bunter environments, where the alluvial plains would fall dry after each downpour or rainy season. All the iron in the run-off water would then have precipitated.
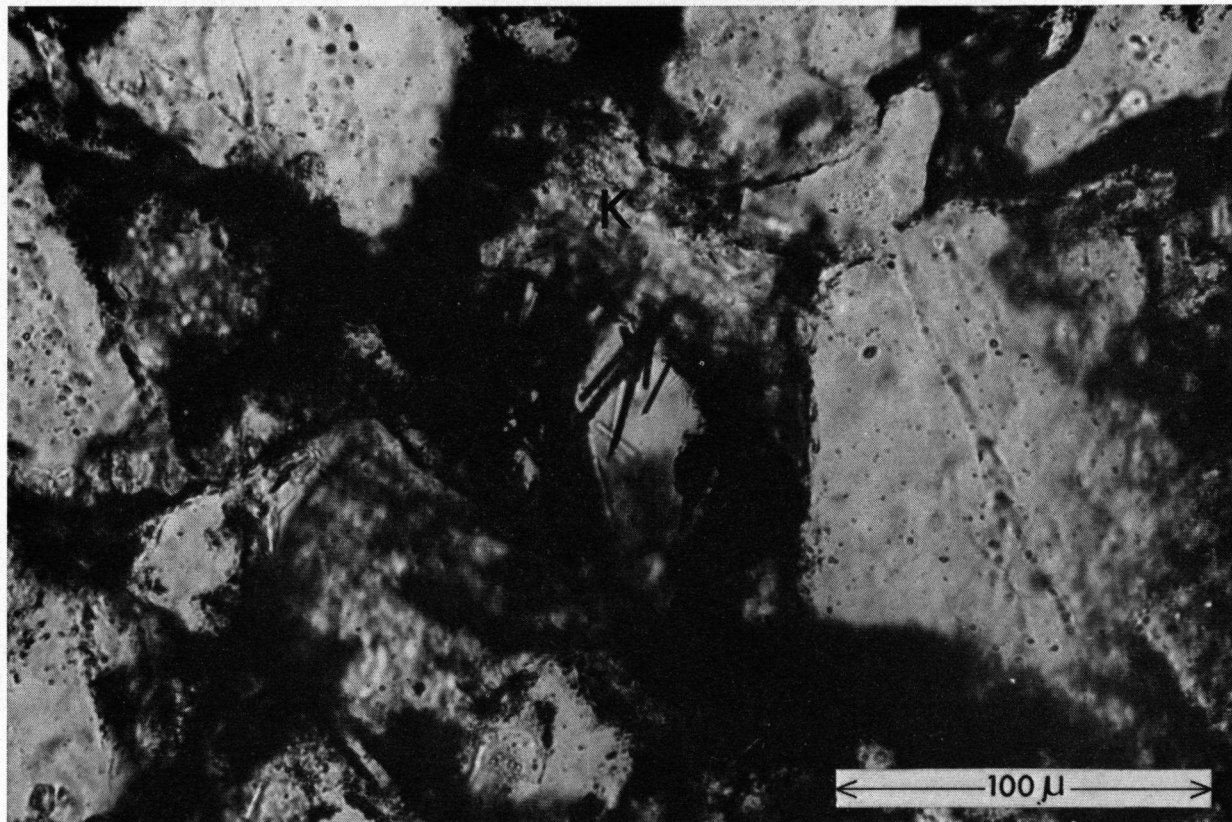

Fig. 61. Fine-grained, moderately sorted Bunter sandstone. (K) is kaolinite formed from mica; other grains are quartz, the black is colloform hematite. Hematite has corroded the quartz grains. The quartz in the centre, which is probably a strongly corroded clastic grain relict, was penetrated by numerous acicular crystals of authigenic goethite (now hematite). Sample 1455, nicols crossed.

Under the conditions of the Bunter and Peranera environments, the iron very probably occurred in its oxidized trivalent state in the soils of the drainage areas and in the run-off water. Since trivalent iron is almost insoluble in most natural waters (Ruttner, 1953, p. 74; Hutchinson, 1957, p. 704;) there can be very little transport of Fe^{3+} in true solution.

Organic complexes and ferric hydroxide sols are compounds that occur widely in soils, from which they can be mobilized and transported. During transport, ferric hydroxide sols may be 'protected' (stabilized) by organic colloids (Barth *et al.*, 1960, p. 202). A supply of these compounds by run-off water infiltrating the sediment during wet periods, could partially account for the authigenic ferric oxides in the Bunter and Peranera deposits.

In view of the low amounts of total Fe_2O_3 in the Bunter sandstones however, the supply of iron by means of run-off water may have been only very small.

The mobilized iron gave rise mainly to the following precipitates in the deposits:

(1) goethite ('*Nadeleisenerz*'), shown in Fig. 61. This form of iron is rare in the Bunter deposits as a whole, but is fairly common in a few samples (1454, 1455). It has not been found in the Peranera deposits. The needles now consist of hematite.

(2) hematite, in many subhedral crystals but also in some euhedral, six-sided flakes. This form of iron, which is as rare as the goethite needles, was found in three samples (781, 785, 787). Well-crystallized, early-formed hematite in red beds has also been recorded by Walker (1967a).

(3) irregular, fine-grained aggregates now consisting largely of hematite. This is the common form of the authigenic iron in the Bunter and Peranera deposits.

Schellman (1959) has shown that both goethite and hematite can crystallize directly from ferric hydroxide gels, albeit under somewhat different conditions. Once

goethite has crystallized, dehydration to hematite is difficult and starts only at temperatures of 150 to 160° C. Van Houten (1961) stated that goethite dehydrates to hematite at a temperature of about 130° C; according to Barth *et al.* (1960, p. 202) dehydration sets in already at 65° C. The influence of aging during a long period of (geologic) time is not known.

According to Schellman, goethite crystallizes from a ferric hydroxide gel at high pH values and, under the influence of adsorbed negative ions (HCO_3^-, SO_4^{2-}), also at neutral pH values. Hematite crystallizes at low pH values, and, under the influence of absorbed positive ions (Mg^{2+}, Ca^{2+}), at neutral pH values. Depending on the types of ions adsorbed, goethite and hematite may form simultaneously.

The significance of these laboratory findings for the red beds under discussion is not entirely clear. Replacement of quartz means that this mineral must have been dissolved at the site at which the new mineral formed. The solubility of quartz is increased significantly only in an alkalic environment at pH values above 9 (Krauskopf, 1959). Replacement of quartz by goethite (needles, Fig. 61) might have occurred in an alkalic environment, and it could also be assumed that the fine-grained aggregates of hematite, which commonly replace quartz, originally consisted largely of goethite. It is noteworthy that in the Peranera deposits the aggregates of hematite rarely replace any plagioclase and limestone fragments (p. 202), which also agrees with alkalic conditions. The replacement of quartz by subhedral hematite crystals (Fig. 60) would be incompatible with the chemical data: at the neutral to low pH values under which hematite forms, quartz is virtually insoluble. It could be argued that the hematite crystals might have formed at greater depths from goethite already present at the same site, but for this there is no direct evidence.

In the Peranera and Bunter deposits, initially present goethite cannot be expected to have been preserved. The increase of temperature with depth of burial, and the permanence at depth such as indicated by the degree of coalification of the Malpas coals (p. 193), probably sufficed to bring about the dehydration of goethite.

Various Bunter sandstone samples provide clear proof that silica fragments (quartz grains, chert, and pebbles) were partly dissolved.

Quartz grains in the Bunter sandstones, as exemplified by the one in Fig. 60, may have pitted surfaces, as indicated by dust rings which follow the irregular grain surfaces either continuously or with interruptions. The pitted surfaces are angular, and were never observed to show even the slightest degree of the rounding that might have resulted from mechanical wear. It is therefore concluded that these surfaces represent corroded surfaces, the corrosion having been brought about by surface dissolution of the quartz grains. Considering the depths in the grains to which corrosion has progressed in various cases, it is reasonable to assume

that small quartz grains in the matrix could have been entirely dissolved (Pye, 1944).

The sample shown in Fig. 60 is of particular interest. Since there is every reason to believe that the authigenesis of the ferric oxides in the Bunter Formation resulted from weathering processes, the ferric oxide, and, according to the growth relations, the quartz overgrowths and the corrosion of the quartz grains, could be early diagenetic.

The results of the dissolution of chert grains and pebbles are much more pronounced. The clastic chert in the Bunter deposits probably derived from pre-Hercynian Upper Carboniferous cherts. The fragments sometimes contain Radiolaria or Hystrichosphaeridae; in plain light in thin section they normally show a light brown stain, probably due to included colloidal organic matter. Dissolution of the grains and pebbles sometimes has progressed so far that only an irregularly shaped relict is left. Such relics are penetrated on all sides by quartz cemented, quartz, quartzite, and other chert grains (Fig. 62). Other chert pebbles are multi-directionally traversed by numerous cracks, now filled with secondary quartz (Fig. 62).

At what stage in diagenesis did the dissolution of the chert occur? To answer this question, the textural relationships in Fig. 63 will be discussed first. The lower part of the microphotograph is occupied by clastic chert, which in this case contains Radiolaria or Hystrichosphaeridae. The dented contact with the clastic quartz grain (on the left in the microphotograph) can only be interpreted as a dissolution surface (dissolution of both clastic quartz and chert), similar to that found along stylolite seams. The same surface continues to the right, to form a dented contact between clastic chert grain and secondary quartz. At this contact, both secondary quartz and chert must have been dissolved. At many points all along the surface, opaque (organic?) matter is concentrated.

To explain these textural relations, two possibilities

Fig. 62. Bunter coarse-grained conglomeratic sandstone. Two chert pebbles (Ch) and one quartzite pebble (Qz) set in a coarse-grained quartz mosaic. The chert pebble to the left is penetrated on all sides by the quartz mosaic (for stylolitic penetration contact, see Fig. 63); the quartzite pebble is penetrated to a lesser degree. Small spherical specks in the chert pebble to the left are Radiolaria or Hystrichosphaeridae The chert pebble to the right is traversed by a dense network of secondary quartz veins, the time of formation of which is not known. The clastic quartz grains (the cores of the mosaic components) do not interpenetrate. Sample 1500, *negative print*, nicols crossed.

Fig. 63. Bunter coarse-grained conglomeratic sandstone. The stylolitic penetration contact of the quartz mosaic into the chert pebble shown in Fig. 62. The outline of two clastic quartz grains is indicated by distinct dust rings; both grains show secondary syntaxial quartz overgrowths. The lower half of the microphotograph is occupied by a part of the chert pebble (Ch). The same stylolitic contact occurs between chert and clastic quartz (to the left) and between chert and secondary quartz (centre). Sample 1500, nicols crossed.
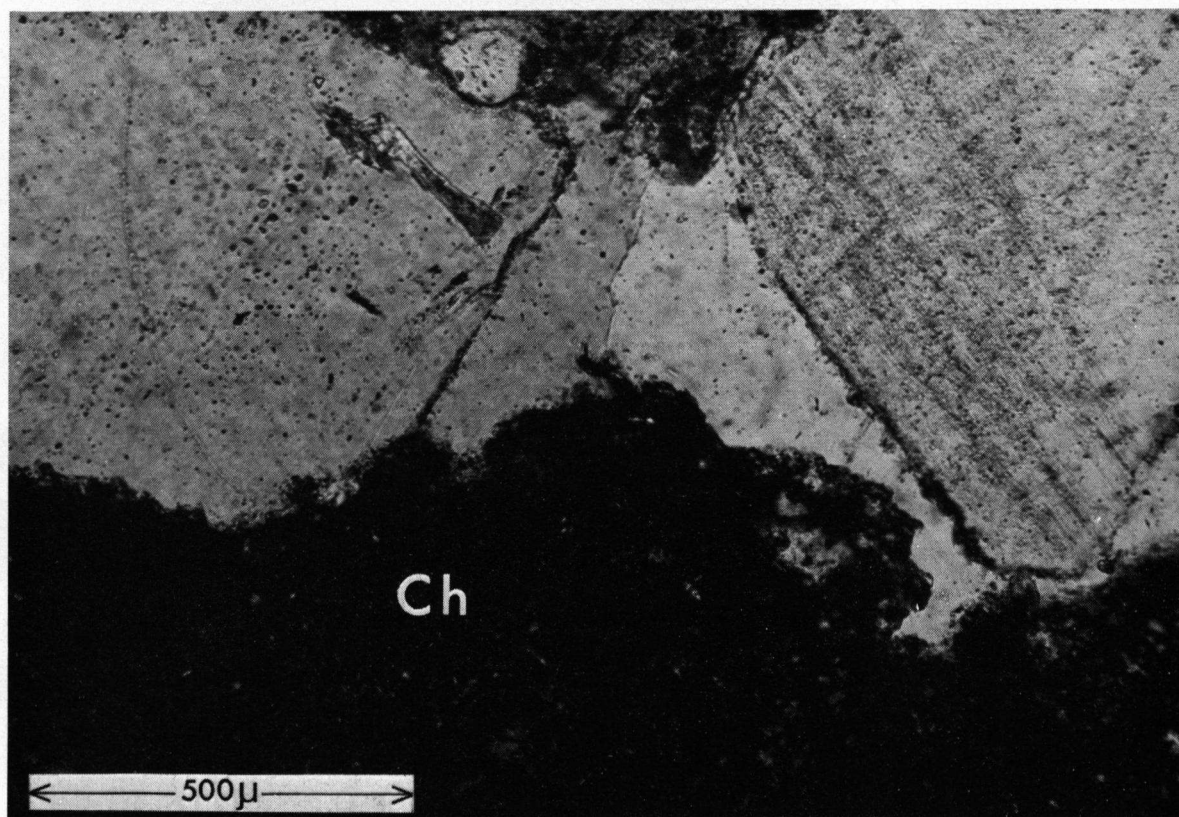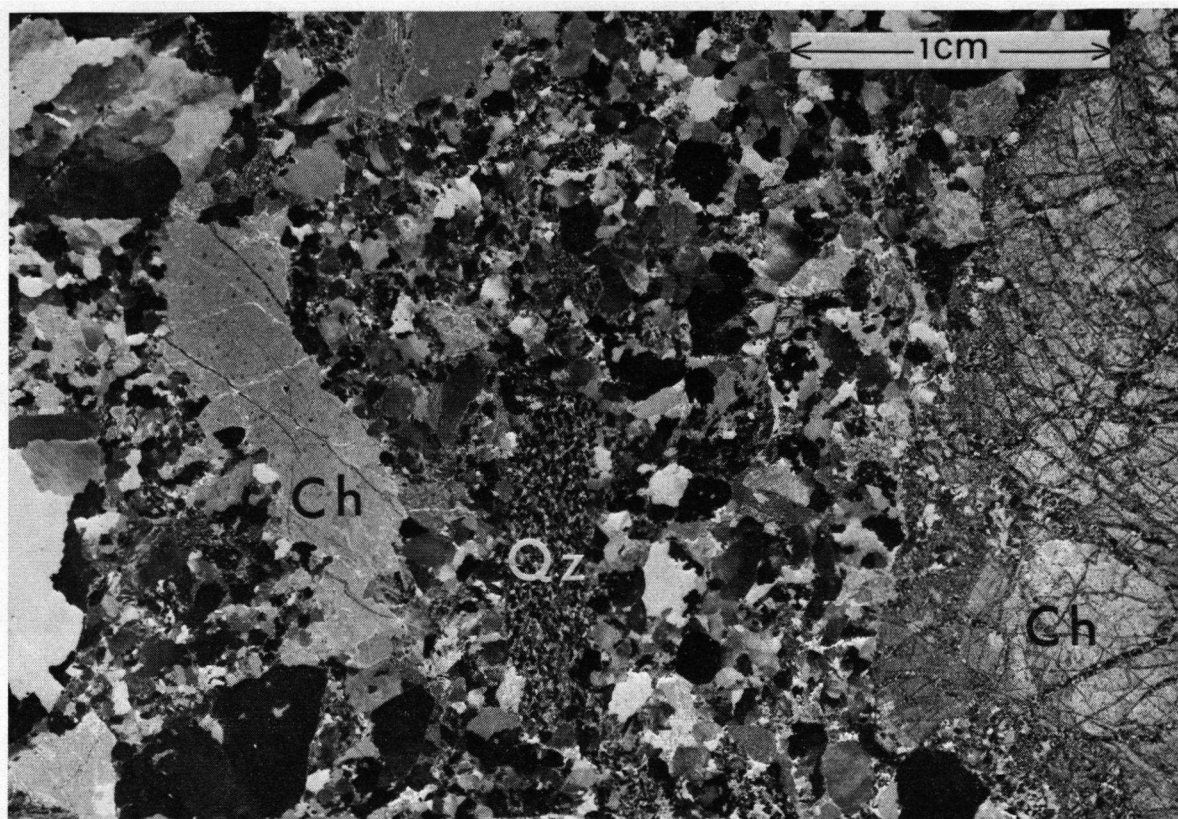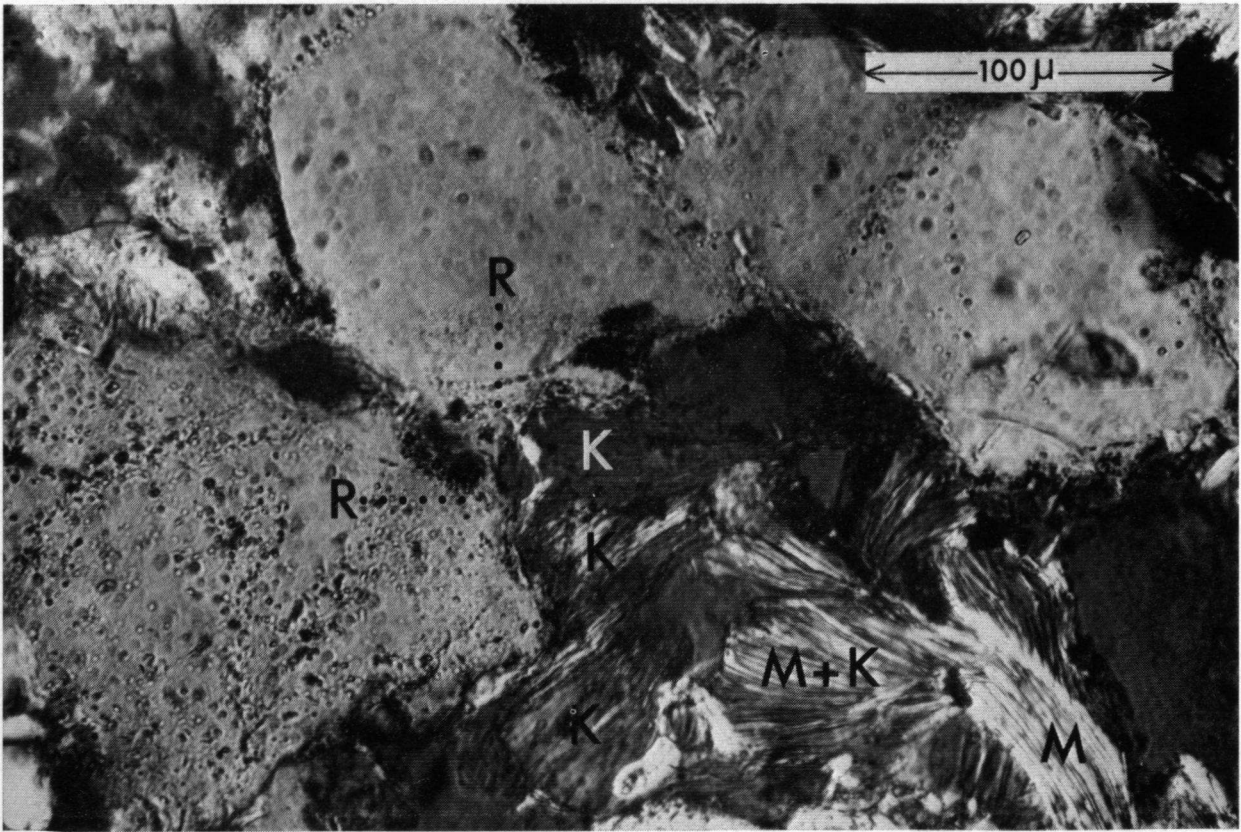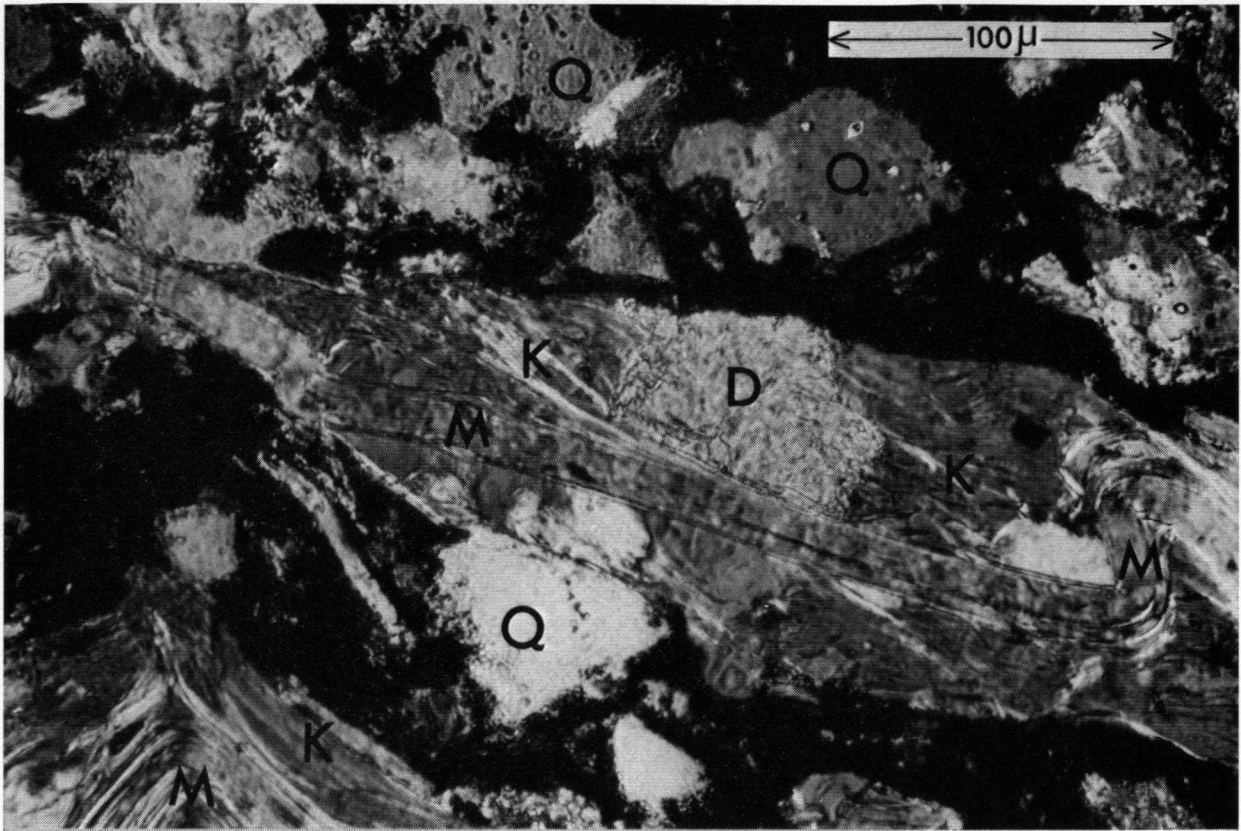
are considered: (1) the dented surface of the chert grain formed, like the pitted quartz-grain surfaces, by dissolution during early diagenesis in response to surface weathering processes. This possibility is discarded, because it fails to explain the deep penetration of clastic quartz grains into the chert; obviously, some overburden pressure would be needed to bring about such penetration. (2) the dented surface of the chert grain is a true stylolite seam, formed by pressure solution (Thomson-Sorby-Riecke principle). This assumption is accepted as valid, because it satisfactorily explains the deep penetration of quartz grains and the local concentration of insoluble residue, which is normal in stylolite seams.

Identical dented surfaces between quartz and chert, also interpreted as stylolite seams, were described by Sloss & Feray (1948). Many pressure-solution phenomena, including those in quartz sandstones, were recently rediscussed by Trumit (1968).

The significance of the relations present in Fig. 63 also lies in the fact that they show that at least some cementation with quartz had been going on *before* the development of the stylolite seam, since the seam cuts both clastic quartz *and* secondary quartz at approximately the same level. Although this does not necessarily date the formation of the secondary quartz during early diagenesis, it testifies to the fact that precipitation of silica had already occurred before overburden pressure could cause pressure-solution of the chert. It also forms an indication that, in the Bunter sandstones, one should take into account the possibility of a cementation with quartz unrelated to that freed by local pressure-solution.

Probable main sources of silica unrelated to pressure-solution in the Bunter sandstones are:

Fig. 64. Fine-grained, poorly sorted Bunter sandstone. The microphotograph illustrates the complexity of the diagenetic processes in the Bunter sandstones. The most likely sequence of diagenesis seems to be: (1) alteration of muscovite (M, still present as an unaltered core) into kaolinite (K) involving a considerable increase in the thickness of the grains, and some replacement of quartz; (2) precipitation of colloform ferric oxides (black, now hematite), involving not only closure of much pore space but also strong corrosion of clastic quartz grains (Q); (3) replacement of the kaolinite by dolomite (D). It is possible, however, that the replacement by dolomite preceded the deposition of ferric oxide. Sample 1491, nicols crossed.

Fig. 65. Coarse-grained, moderately sorted Bunter sandstone. Shown are quartz grains carrying secondary quartz overgrowths, muscovite (M), and kaolinite (K). The clastic muscovite is laterally transitional, *via* interstratified muscovite and kaolinite (M + K), to the authigenic kaolinite. Kaolinite replaces secondary quartz and the clastic quartz cores (R). The 'dust rings' in this thin section are commonly composed of rows of fluid inclusions. Sample 1460, nicols crossed.

(1) silica freed from quartz grains by the replacement with ferric oxides, as already discussed, (2) silica dissolved from clastic quartz grains (pitted quartz grains). Possible other sources of silica lie in the run-off water, concentrated by evaporation, and in the *in situ* hydrolysis of silicate minerals like feldspar; of these latter origins however, there is no direct evidence. Alkalic interstitial waters, and the increase in temperature with increasing depth of burial must also have promoted the mobilization of silica. The normal presence of authigenic dolomite (see below) testifies to elevated pH values of the interstitial waters, since present-day dolomite formation occurs in saline waters having a high pH value (Alderman & Skinner, 1957; Skinner *et al.*, 1962).

The effect of included plant material, release from this source of CO_2, and a consequent lowering of the pH value, probably had an influence only at near-surface levels. The almost general absence of coalified plant material in the Bunter deposits shows that it was rarely included in lower levels; a slow rate of deposition (p. 213) must also have favoured the complete oxidation of plant material at near-surface levels.

The solubility of quartz increases with increasing temperature (Siever, 1962); increased temperatures may therefore have promoted the cementation with quartz, particularly where pressure-solution is involved. Evidence of raised temperatures is provided by the advanced state of coalification of the Malpas coals, which underly the Bunter deposits by about 300 m (p. 193; Fig. 4). The amount of increase of temperature, however, cannot yet be estimated because the permanence of the coal at greater depth is still unknown.

It is extremely difficult to evaluate the extent to which pressure-solution of quartz, quartzite, and chert grains has contributed to the cementation of the sandstones. In quite a number of samples, among them the sample in Figs. 62 and 63, the quartz grains appear to be loosely packed, and hardly any interpenetration of the clastic grains, as indicated by dust rings, can be observed. In other samples dust rings are almost absent and quartz forms an equidimensional mosaic. Still other samples show a vague orientation of the mosaic pattern parallel to the stratification, though nowhere as pronounced as in thoroughly pressure-welded sandstones (Waldschmidt, 1941).

The hazards involved in attributing the cementation of a quartz sandstone to one process or another are very clearly demonstrated by an example from the Woodbine sandstones discussed by Gilbert (1958, p. 318). In these deposits, loosely packed quartz-cemented quartz sandstones and pressure-welded sandstones occur together.

It is perhaps safest to conclude that (1) in the Bunter sandstones studied, dissolution of quartz and precipitation of quartz may have already started during early diagenesis (pitted quartz grains, secondary quartz replaced by ferric oxide), (2) cementation with quartz may have continued prior to the onset of pressure solution (secondary quartz cut by stylolite seams), and

(3) cementation with quartz was completed by the silica freed by pressure-solution. There are, however, no convincing arguments permitting us to distinguish between the first and second phase.

Considering the probability of the early precipitation of quartz, it is instructive to compare the Bunter environment with an environment like that found in Central Australia. Besides other similarities such as widespread pediplanation and authigenesis of ferric oxides (lateritization), the early precipitation of quartz took place there as well, as shown by the occurrence of silcrete (Mabbutt, 1965; Langford-Smith & Dury, 1956). It is, however, doubtful if the Australian silcretes formed in the present-day arid climate. It has recently been argued by Van de Broek & Van der Waals (1967) that the silcrete studied by them in Tertiary deposits in The Netherlands formed in a warm, humid, and seasonal climate. The present Bunter deposits probably formed under similar climatic conditions (p. 171).

The effects of the hydrolysis of muscovite and in all cases the alteration of this mineral to kaolinite, are observable in most Bunter sandstone and mudstone samples from the Tor River valley. Clastic muscovite is a characteristic component of the Bunter deposits in the South-Central Pyrenees; in the Tor valley samples analyzed it occurs in percentages of 2 to 3 (volume) in the sandstones (Tables X, and XI).

Because muscovite is a very stable mineral in weathering, its constant presence in the Bunter deposits, as compared to its very infrequent occurrence in the underlying deposits, is probably partially due to selective weathering in the source area of sediment (see above). The precise source of the muscovite is not known; it may have lain in the pegmatites associated with the granodiorites and also in Paleozoic non-igneous formations. In present outcrop areas in the Axial Zone, muscovite-bearing pegmatites are rare.

In a few Bunter sandstone samples the preserved muscovite is still fresh, but in most samples it has been partially or completely altered into kaolinite. X-ray analyses of the partially altered grains invariably showed the presence of 2M muscovite and kaolinite. The kaolinite occurs interlayered in exfoliated muscovite grains, or a muscovite grain is laterally transitional to kaolinite (Fig. 65). Thick kaolinite crystals can also be observed to carry a muscovite relict in their central parts (Fig. 64). In all the observed cases the transition of muscovite to kaolinite involved an estimated increase in the thickness of the grains of up to 5 times. Due to this process, kaolinite crystals filled in entire pore spaces (Fig. 65). Identical conversions of muscovite into kaolinite have also been described by Scheere (1963) and Jonas (1964).

In a few samples (including nos. 1460, 1499, and 1500) the authigenic kaolinite is reconstituted into illite/muscovite, a transition related to the depth of burial of the deposits and to a supply of potassium (summarized by Kisch, 1969).

It seems reasonable to assume that the kaolinization of the muscovite occurred as a weathering process. No-

where in an accumulating continental sedimentary deposit are the conditions for leaching and subtraction of cations better realized than in the part exposed to weathering, whereas leaching will become more difficult at lower levels because the cation content in the groundwater normally increases with depth. In several instances the authigenic kaolinite was observed to replace both clastic quartz grains and their secondary quartz overgrowths (Fig. 65). If the assumption that kaolinization of the muscovite was a weathering process is justified, we have yet another indication that some of the secondary quartz was precipitated during early diagenesis.

An alternative explanation of the formation of the kaolinite, namely that it formed during later stages, under the influence of slightly acid (CO_2-carrying) waters expelled by the underlying pre- and post-Hercynian Carboniferous can probably be excluded. The Bunter sandstones also contain kaolinite wherever they overly the calcareous Devonian and the Peranera Formation, and 'halted (early) stages' in diagenesis, as found in dolomite crystals, contain kaolinized muscovite as well (p. 226).

In an aggrading system in which weathering (pedogenic) processes of superficial material have been continuously active, it is not surprising that no well-differentiated weathering profiles are found. The uppermost level in the weathering profile, the eluvial or A horizon, will, with continuing deposition of sediment, become the illuvial or B horizon below a new, superimposed eluvial horizon. Continuous repetition of these switches from A to B horizons should result in a uniform sequence of sediment. This scheme, of course, is a strong simplification; the influence of the many factors involved, such as the time of exposure, temporary changes in climate, differences in parent material, etc., should lead to a different result.

What we would expect from applying this principle, in its simplest form, to the Bunter deposits (which could have formed under these conditions) is the following chronological order in diagenesis: (1) eluviation (solution, oxidation, hydrolysis), and (2) deposition of material commonly concentrated in B horizons, such as ferric oxides.

In some Bunter sandstone and mudstone samples, among them sample 1491 (Fig. 64), small muscovite flakes partially altered into kaolinite are fully enclosed in much larger lumpish masses of hematite. This would agree with the sequence suggested above. Although the reverse relation was not noted (kaolinite enclosing hematite), the cases are so scarce that no definite conclusion can yet be drawn from them.

The weathering of muscovite *in situ* also distinguishes the Bunter red beds from the Peranera red beds. In the Peranera red beds, many feldspars are little altered and commonly even strikingly fresh. Biotite, which is less stable than muscovite during weathering, is commonly less altered than the muscovite in the Bunter deposits (p. 209).

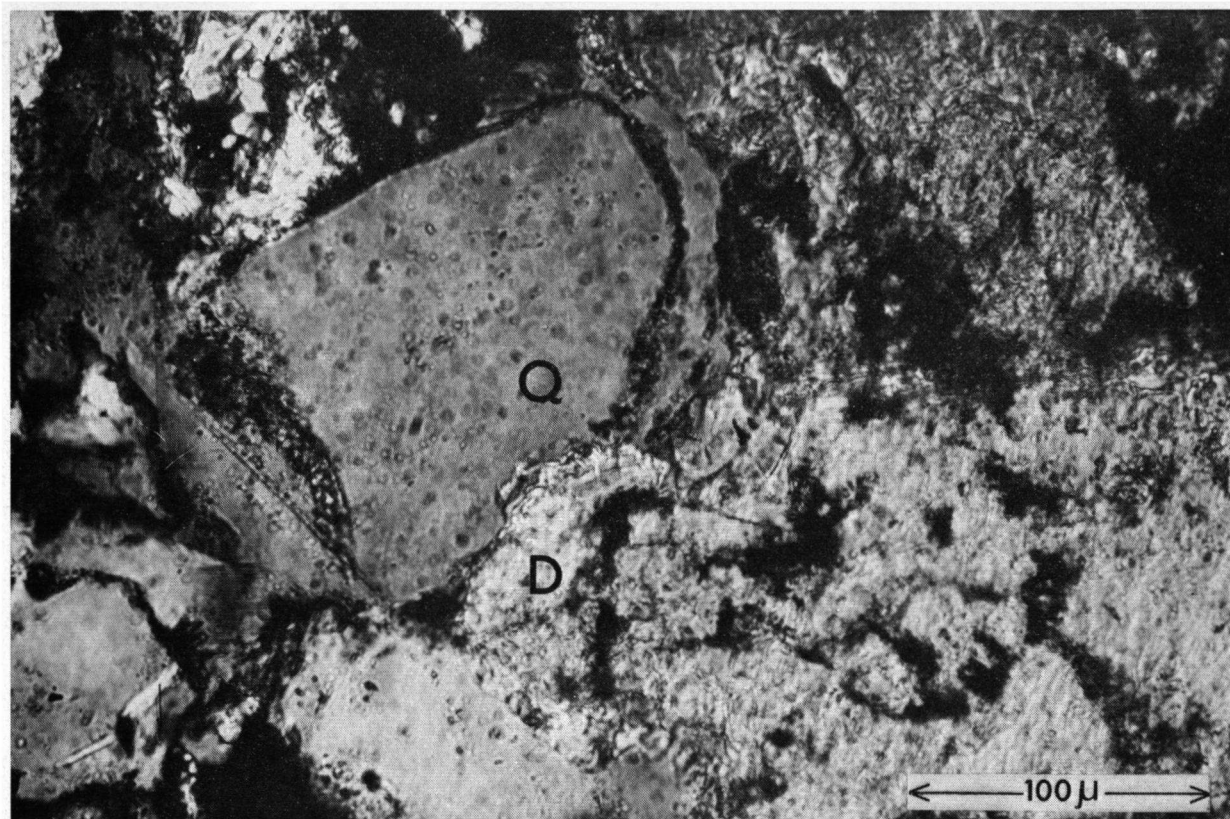The indispensable agent in weathering is water. The

Fig. 66. Medium-grained, moderately sorted Bunter sandstone. Both a clastic quartz core (Q) and its syntaxial quartz overgrowth, separated by a well-developed rim of fine-grained hematite, are partially replaced by a single dolmite crystal (D). The dolomite crystal encloses relics of fine-grained hematite and of quartz. Sample 1495, nicols crossed.

much stronger *in situ* weathering of the Bunter deposits, therefore, point to more humid conditions (savannahs, p. 171) than the prevailing climate in Peranera times (steppes, p. 171). A slower rate of deposition of the Bunter deposits (p. 213) may also have contributed to the stronger weathering of the deposits.

As previously stated, dolomite was among the last authigenic minerals formed in the Bunter sandstones. This relationship to the other authigenic products is shown by (1) dolomite replacing and enclosing colloform hematite aggregates (Fig. 66); (2) dolomite replacing secondary quartz (Fig. 66); and (3) dolomite replacing the kaolinite which had previously formed after muscovite (Fig. 64). The replacements are common, and can be observed in most sandstone samples.

Staining with potassium ferricyanide (reaction on Fe^{2+}: Turnbull's blue) shows the presence of both non-ferroan and ferroan dolomite. The distribution in a thin section of both types of dolomite suggests that the non-ferroan dolomite precipitated first. In aggregates of dolomite crystals the central crystal is commonly non-ferroan; single dolomite crystals often show a non-ferroan centre and a ferroan rim.

The dolomite occurs either in aggregates of anhedral

crystals or in almost perfect rhombs. The crystal sizes range up to about 0.5 mm.

Because no ghost textures were found that could be ascribed to the initial presence of calcite, it is not known whether the dolomite represents dolomitized calcite or formed as a direct precipitate. The Bunter mudstones show evidence of dolomitization of calcite as well as of the direct precipitation of dolomite (see under *Mudstones*, p. 224).

No dolomite concentration was found along faults, fissures, or joints. Although the dolomite formed relatively late in the sandstones, its occurrence is still related to the sedimentary fabric. In the sandstones the dolomite is usually concentrated in laminae lying parallel to the stratification, which is normally the stratification of cross-bedding. Between such laminae with abundant dolomite, the dolomite occurs evenly distributed, which suggests that the mineral was precipitated from percolating groundwaters, preferentially in zones of the greatest permeability.

The possibility that dolomite formation only started after the Bunter Formation had been entirely deposited should be considered. The formation is overlain by the Muschelkalk, Keuper, and Jurassic, all of which are rich in dolomite (Mey *et al.*, 1968). Could the dolomite, carried by downward percolating waters,

have originated from these overlying deposits? Apparently not, because the dolomite concentration is not higher in the top levels of the Bunter Formation, and because some of the mudstone levels are very rich in dolomite while others contain almost none, which suggest a penecontemporaneous origin (p. 224). The formation of dolomite in the Bunter deposits is further discussed in the paragraph on the mudstones.

With the exception of the sericitization of the authigenic kaolinite, accessory barite was probably the last authigenic mineral to crystallize. Barite occurs, as in the Peranera deposits, in the form of thin veins oriented at various angles to the stratification (sample 1474) and in pockets. In one sample (1495) barite occurs as an accessory cement formed after dolomite. This relation is also demonstrated where very large barite crystals in veins entirely enclose much smaller dolomite replacement relics (sample 1474).

Mudstones

Many mudstone samples are nearly opaque in thin section due to evenly distributed, fine-grained hematite. With respect to the total iron content, only two samples were analysed. The content of one of these samples falls in the range of the sandstones (4.37%); the content of the other is almost double that amount (7.39%, Table XIII).

Three chemical analyses of Bunter mudstones made in the El Pirineo cement factory at Cherallo are shown in Table XV. These samples were taken in the quarry near the factory (Fig. 1). The results show, besides a significantly higher content of Al_2O_3 (probably largely from muscovite, kaolinite, and illite), a higher content of total Fe_2O_3 than was found in the sandstones (Tables XIII and XIV). Combination and averaging of all values of total iron shows that the mudstones are richer in Fe_2O_3 than the sandstones by a factor of 2.5.

TABLE XV

constituents	A	B	C	D
SiO₂	58.72	57.40	59.26	58.10
Al₂O₃	20.35	22.75	17.72	15.40
Fe₂O₃ .	7.15	7.70	8.04	6.74
MgO	2.12	1.81	1.77	2.44
CaO	2.30	1.75	5.00	3.11
Loss on ignition	4.90	4.30	5.86	–

Table XV. A,B and C: Partial chemical analyses of Bunter mudstones.
Analysed by chemical laboratory, cement factory at Cherallo.
D: Average shale, after Leith & Mead (1915).

The fact that the finer-grained deposits in red bed sequences have a higher iron content (mainly as hematite) has already been pointed out by Tomlinson (1916). The conclusion that the hematite was clastically supplied as fine-grained material (Dunbar & Rodgers, 1958, p. 210), however, need not be generally valid, nor is the fact that the fine-grained deposits are

higher in total Fe_2O_3 typical of red bed sequences. According to the values computed by Leith & Mead (1915) for the average sandstone and shale, the content of total iron in shale is higher than that in sandstone by a factor of 4.8.

In the Bunter mudstones, as in the sandstones, almost all the hematite shows colloform (non-clastic) textures and generally replaces clastic quartz grains. It is likewise suggested that the ferric iron in the mudstones probably formed from a ferric hydroxide sol or gel (p. 216).
The content of total Fe_2O_3 in the mudstones does not differ essentially from that in the average shale (Table XV). The ferric oxides could therefore have derived chiefly from the intrastratal destruction of iron-bearing minerals. An unknown but presumably small amount of iron may have been introduced by infiltrating runoff water (see discussion under *Diagenesis*, p. 216).

X-ray analyses of bulk samples of mudstone and determinations in thin section show the presence of quartz, muscovite, kaolinite, chlorite and septechlorite, hematite, traces of goethite (probably oxidation products of ferroan dolomite), dolomite, and traces of calcite.

With respect to the course of diagenesis, some similarities to the sandstones can be observed, including replacement of clastic quartz by ferric oxides, kaolinization of muscovite, both to a larger extent than in the sandstones and clearly displayed by almost all mudstone samples. Secondary quartz seems to be absent. The cement relations are difficult to distinguish, however, because of the general hematite stain.
But there are also a number of marked dissimilarities with respect to the sandstones, the most noteworthy being the signs of a strong infuence on the mudstones of pedogenic processes. In virtually none of the mudstone samples is the original texture of the mud preserved[1]; what is observed could be described as: (1)

[1] In the field, ghost structures of current ripples and laminations are seen (p. 167).

Fig. 67. Bunter sandy mudstone. Well-developed authigenic ferroan dolomite rhombs in a mudstone matrix largely composed of authigenic colloform fine-grained hematite (black), kaolinized sericite and muscovite, chlorite, and angular quartz grains (lower right, lower left, top centre). Formation of the dolomite rhombs occurred after the precipitation of ferric oxide, as shown by the included cores of hematite and the hair-thin dolomite parallel to the rhomb surfaces indicating the progressive growth of the crystals. Sample 1481, nicols crossed.

Fig. 68. Bunter sandy mudstone. Imperfectly developed ferroan dolomite rhombs in a fine-grained mudstone matrix. The dolomite crystals enclose small quartz grains, muscovite altered into kaolinite, chlorite, and hematite. Elongate fine-grained clastic components of the mudstone matrix are reoriented in the direct vicinity of the dolomite rhomb due to compaction pressure or growth pressure of the rhomb, or both. Sample 1494, nicols crossed.
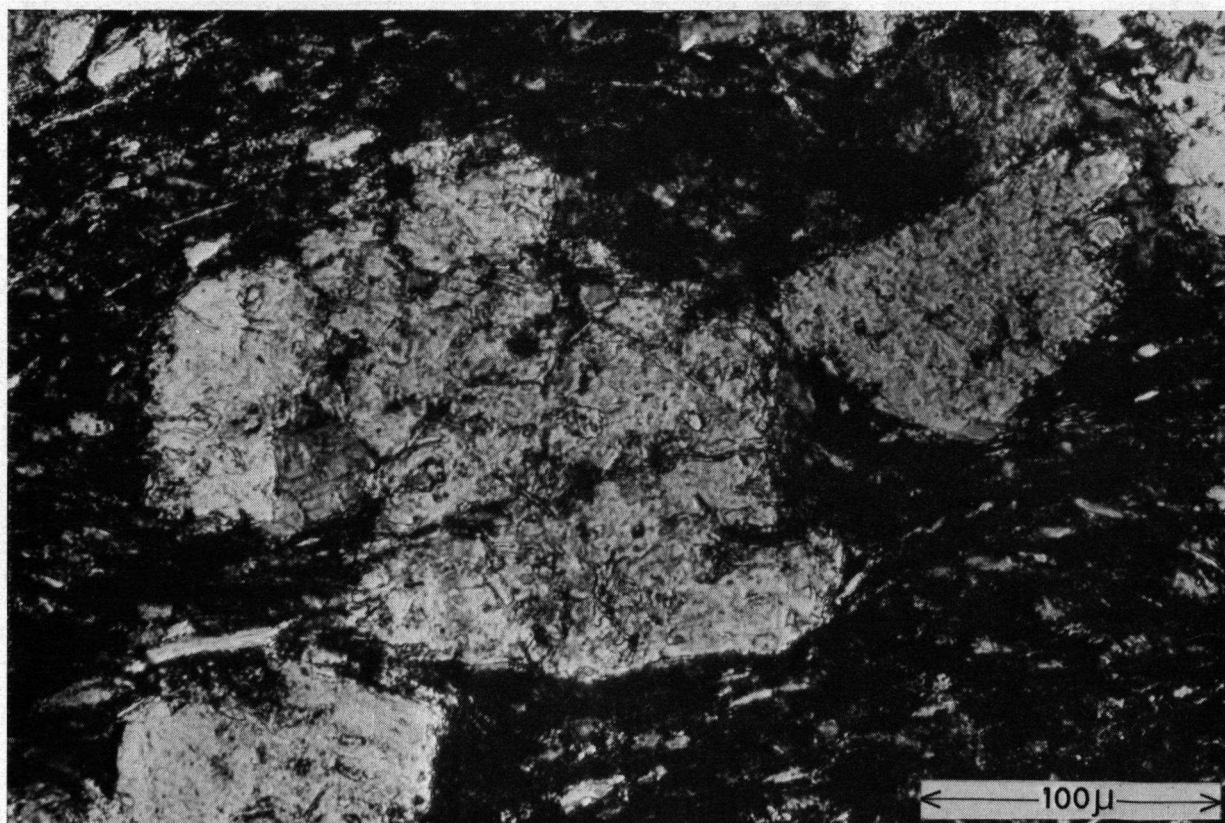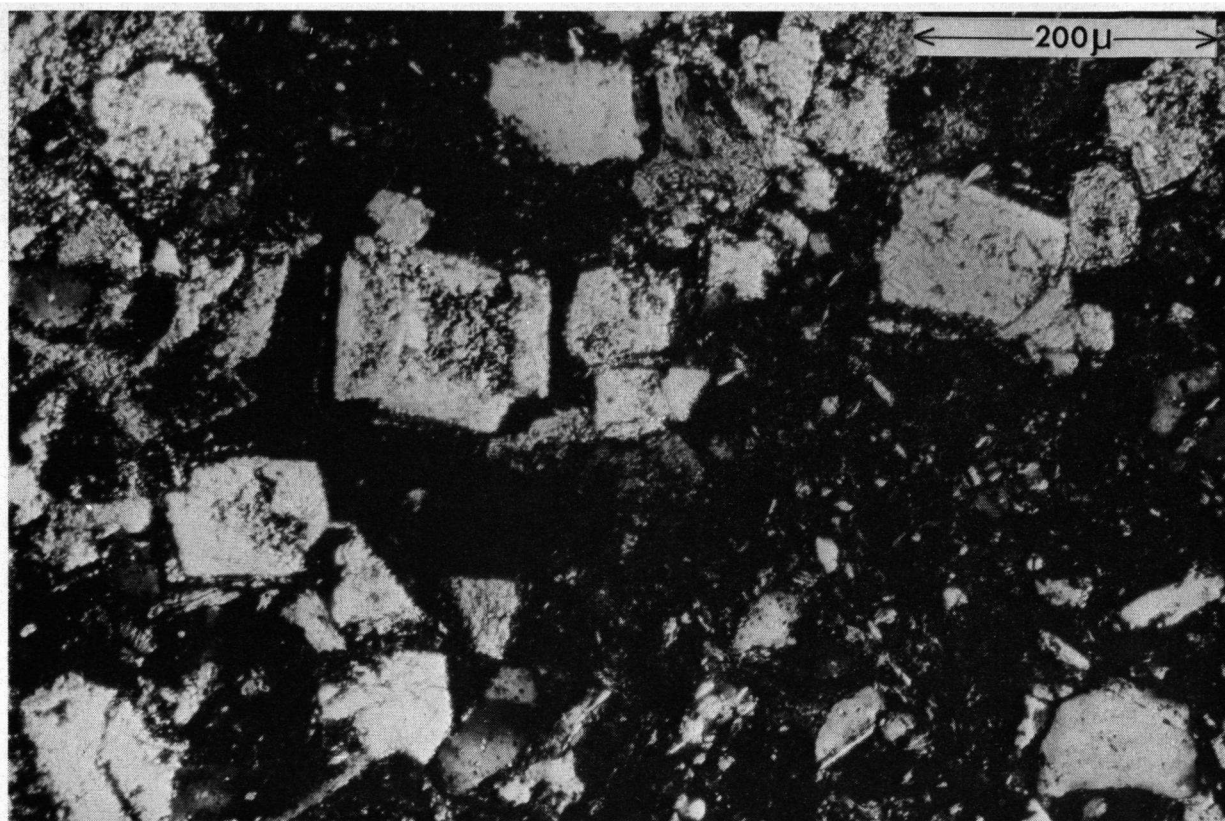
hosts of particles arranged in whorl-like patterns; (2) highly irregular, local concentrations and streaks of the coarsest grains present (silt-size); (3) 'fragments' in equidimensional or oblong shapes of fine-grained material (which a pedologist would probably call 'micropeds'); and (4) rare 'veins' of fine-grained material arranged in parallel aggregates probably representing former conducting channels (comp. Fig. 30). There is therefore no doubt that the muds were

intensely reworked, both by organic activity (burrowers and roots, the signs of which are also visible in the field) and by inorganic processes such as mud cracking, fissure forming, internal transport, etc.

In at least one case, sample 1482, there is evidence that the muds were also reworked by colluvial processes. This sample is largely made up of rounded, sand- and pebble-sized components of mudstone; it must therefore have been deposited as a pebbly sandstone,
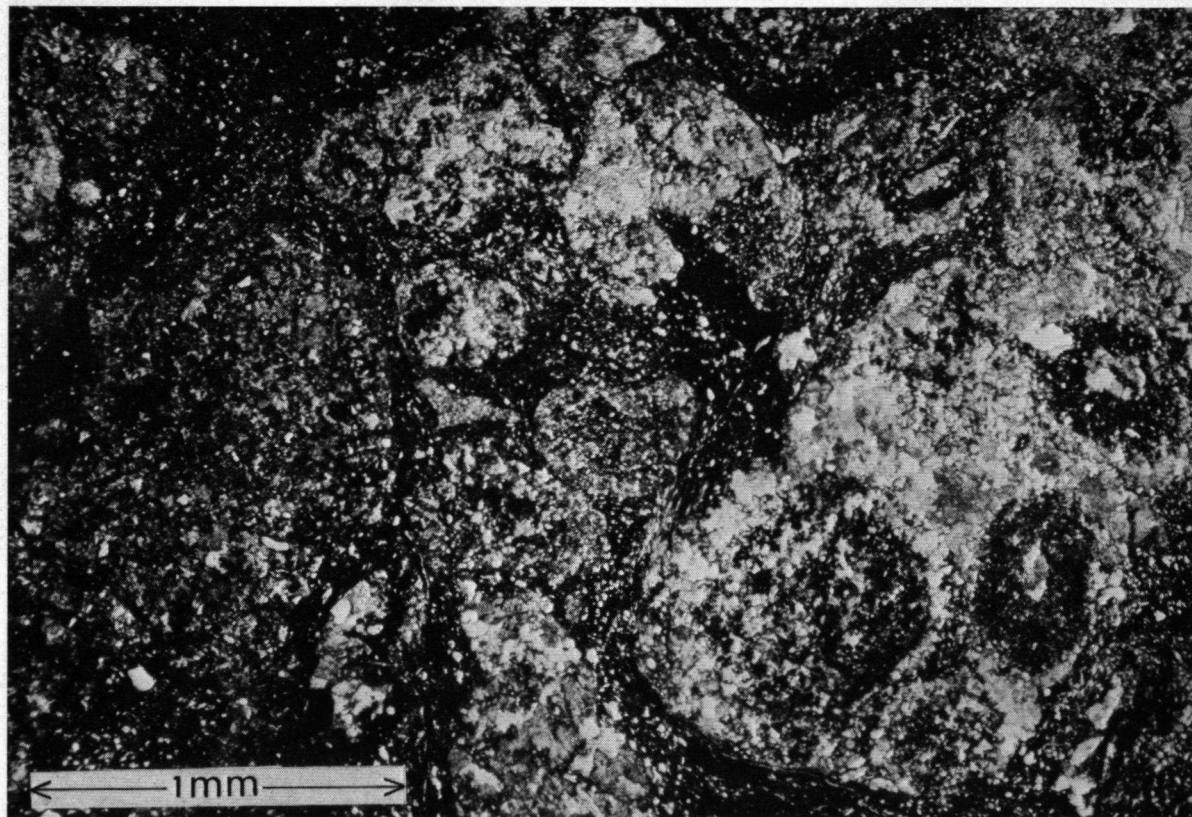

Fig. 69. Bunter silty mudstone. Coalescing spherulites, probably initially composed of calcite, now entirely converted into ferroan dolomite. The same sample shows perfect, isolated spherulites. The origin of the spherulites is thought to be similar to that of those found in recent caliche. Sample 1464, plain light.

but is still largely composed of material of the mudstone grain-size range.

Dolomite (largely ferroan dolomite; some non-ferroan), is present in almost all the samples; this mineral is an equally characteristical component of the Bunter mudstones and the sandstones, but the content in the mudstones varies widely from one stratigraphic level to another. In a rock as little pervious as mudstone this fact excludes the possibility of a late introduction of the dolomite; it is in agreement, instead, with a penecontemporaneous (early diagenetic) origin of the mineral.

The dolomite shows two main textures (1) single dolomite rhombs, distributed in more or less even patterns of varying density, or arranged in curved streaks in the silicate-hematite mudstone matrix (Figs. 67 and 68), and (2) dense aggregates of much finer

grained dolomite (Fig. 69). Besides these two forms, dolomite is sometimes also found in polycrystal units of oblong and occasionally rectangular shapes (sample 1456, 1459a; not illustrated). It is suspected that this latter mode of occurrence represents pseudomorphs after gypsum and anhydrite, but the point cannot be proved, since no partial replacements have been observed.

The single rhombs, which probably represent a direct crystallization of dolomite, commonly contain many fully enclosed minerals, among them small quartz grains, muscovite and muscovite partly or entirely altered into kaolinite, hematite, and chlorite. The types of enclosures show that the dolomite rhombs most probably developed after the precipitation of ferric oxides and after the hydrolysis of muscovite had taken place (Figs. 67 and 68).

On the other hand, there is also evidence that the

dolomite rhombs developed at a stage when the mudstones were not yet fully compacted. Fig. 68 shows three imperfectly developed dolomite rhombs. In the direct vicinity of the large crystal in the centre, the clastic particles of the mudstone (mainly muscovite, some quartz) are oriented parallel to the outer surface of the crystal. This orientation must have been produced either by outward-directed growth pressure of the dolomite crystal or by compaction of the still relatively soft mud around the hard crystal. It may be recalled that the same argument was used to show that the siderite spherulites in the Malpas mudstones and small calcite concretions in the Peranera mudstones had a pre-compaction origin, but it is not known at what advanced stage of compaction such reorientation of clastic components could still have been realized.

The fine-grained dolomite aggregates are probably the result of dolomitization of calcite. In many of the aggregates a distinct, but partly destroyed globular texture produced by the coalescing of spherulites, is preserved (Fig. 69). Isolated, perfectly developed spherulites are also present. These spherulites locally enclose smaller spherulites (Nagtegaal, 1969).

On the basis of the resemblance in shape and size of the spherulites to calcite spherulites in recent caliche, it is assumed that both were formed under similar conditions. The occurrence of vertically oriented nodules resembling those found in the Peranera mudstones and in recent caliche (Rutte, 1958) (p. 161) also indicates that at times caliche did develop in the Bunter mudstones (at 32 and 105 m above the base of section 8, sheet V).

Since the Bunter deposits formed in a fresh-water fluvial environment and since the dolomite in the mudstones appears to be penecontemporaneous, it follows that the Mg^{2+} necessary for dolomite formation must have been largely introduced by run-off water. A direct source of Mg^{2+} may have been Devonian dolomites which probably cropped out in the supply areas of Bunter sediment, but the Mg^{2+} may also have been freed by weathering from many other Paleozoic rocks in the supply areas. No special

conditions had to be fulfilled, however, because Mg^{2+} occurs in appreciable quantities with respect to the other cations in most river waters (Hutchinson, 1957, p. 554; Krauskopf, 1967, p. 320).

Strong evaporation on the Bunter flood plains may have been ultimately responsible for the formation of the dolomite. First, $CaCO_3$ would precipitate (calcite nodules, spherulites), possibly followed, with rising salinity, by precipitation of gypsum (doubtful gypsum and anhydrite pseudomorphs). This is the normal course of precipitation from ordinary river waters (Hutchinson, 1957, p. 566). The Ca^{2+} subtracted in this way may have increased the Mg^{2+}: Ca^{2+} ratio in the uppermost levels in the ground-water to the extent that it could bring about dolomitization of the calcite present (calcite nodules, spherulites) and precipitate dolomite directly (dolomite rhombs in the mudstones). The ground-waters in the Bunter deposits must have been relatively high in Mg^{2+} also at much lower levels, as shown by the late authigenic precipitate of dolomite in the sandstones.

Why is dolomite the characteristic carbonate in the Bunter red beds and why does it occur so rarely in the underlying Peranera red beds? The answer to this question probably lies in the fact that due to the abundant early-precipitated $CaCO_3$, the Peranera deposits were rapidly 'clogged'. The Bunter deposits, to the contrary, remained 'open' much longer and evaporation could proceed furher, thus creating conditions favourable for the formation of dolomite. Because of the more humid climate moreover, the total amount of run-off water (carrying dissolved Mg^2) supplied to the Bunter deposits will have considerably exceeded that supplied to the Peranera deposits.

The high content of authigenic calcite in the Peranera deposits probably resulted mainly from *in situ* mobilization from clastically deposited calcareous fragments (caliche formation). In the Bunter deposits, to the contrary, because of the maturity of the clastic assemblage, such mobilization was possible only to a very limited extent. Here, most of the authigenic carbonates may have been supplied by the run-off water.

PETROGRAPHY AND DIAGENESIS: SUMMARY, COMPARISON, AND CONCLUSIONS

The discussion in the preceding chapters has repeatedly made it clear that close interrelationships exist between the type of diagenesis, the type of depositional environment, and the types of materials available, such as the primary lithology of the deposits and the components supplied by run-off and ground-water.

As is the case in much geological work, we are still far from being able to quantify such relationships. It is even likely that some important factors have so far

escaped attention, and that the relative influences of others are exaggerated or underestimated. Consequently, the following attempt to indicate the relationships between the various factors cannot be more than a crude one. For the continental deposits under study, we find that:

Diagenesis = f [depositional environment + materials available];

in this

depositional environment = f [climate + morphologic setting],

and:

materials available = f [composition source area + climate + morphologic setting].

Furthermore, it holds that:

diagenesis = f [materials available + overburden pressure + geotemperature + permanence at depth].

The significant petrographic and diagenetic characteristics of the deposits in question will be briefly summarized on the basis of the first three functions. The effects of the burial history of the deposits, such as the coalification in the Malpas Formation (p. 193), compaction moulding of phyllite (Aguiró and Peranera Formations, p. 178, p. 202), sericitization of kaolinite in the Bunter Formation (p. 222), dehydration of hydrous ferric oxides (Peranera and Bunter Formations), and pressure-solution (Bunter sandstones) are here left out of consideration.

Aguiró, Erill Castell, and Malpas Formations

The sandstones (braided stream environment) in the Aguiró Formation are very immature, as shown by the predominance of the phyllite components. The morphologic setting must have been one of a pronounced relief (piedmont environment, p. 150). Devonian rocks, which are rich in limestones, must have occurred in the source area of the sediments. Limestone components, however, are almost solely found as pebbles and boulders in the Aguiró conglomerates; they are rare or absent in the sandstones. There is, moreover, no carbonate cement in the sandstones.

The mudstones and shales (back-swamp environment) in the Aguiró Formation contain abundant coalified vegetational debris and coal stringers, and carry authigenic iron sulfides, kaolinite, and siderite. This paragenesis suggests a geochemical environment characterized by neutral to negative Eh values and neutral to low pH values[1]. To permit these conditions to develop, the climate must have been sufficiently humid to keep the back-swamps flooded. A humid climate and low evaporation are also suggested by the absence of carbonate-rich fragments in the sandstones (selective weathering: solution of small-sized, particulate carbonates in the drainage areas) and by the absence of early precipitated calcite in the braided stream deposits.

The Erill Castell tuffs, which were deposited on a hilly land surface, give evidence of climatological conditions comparable to those prevailing during the deposition of the underlying Aguiró Formation and the overlying Malpas Formation. Their present chemi-

cal composition can be attributed to leaching of tuffs with an andesitic composition. Their mineralogy points to intense hydrolysis of silicate components to the extent that hardly any of the original silicate minerals, besides quartz, are preserved. The effects of pedogenic processes are clearly expressed at various levels. Locally, secondary calcite could accumulate, probably in B-horizons of soils.

The Malpas Formation, considered as still representing the fill of a valley, although a much wider one than that of the Aguiró Formation, contains a texturally extremely immature clastic assemblage. This is due to the fact that nearly all clastic material was derived from the directly underlying, easily erodable Erill Castell Volcanics.

Diagenesis in most Malpas mudstones and shales (back-swamp environments) is similar to that of the Aguiró mudstones and shales, and likewise points to flooded back-swamps in a humid climate. Further evidence of a humid climate is provided by the continuous flow in meandering streams, which can be deduced from channel-fill properties (p. 171).

There is good evidence, however, of a stronger influence of evaporation and hence of a warmer climate than that which prevailed during the accumulation of the underlying formations. This evidence consists of: (1) the occurrence of calcite spherulites in a tuff bed, (2) the occurrence of birdseye limestones, (3) the sudden change in fluvial facies from meandering to braided, with a concomitant change in diagenesis to abundant early precipitated calcite, and (4) the occurrence of ferroan dolomite in lake deposits, in the very top part of the formation.

Peranera and Bunter Formations

The Peranera and Bunter Formations are comparable in that they are lowland, mainly fluvial deposits; both of them are red beds, and both contain clastic material derived from the Paleozoic core of the Pyrenees. In many other aspects they differ markedly. There is a pronounced difference in diagenesis. In the Peranera deposits diagenesis is largely restricted to the authigenesis of ferric oxides and the precipitation of calcite, the latter probably extending from the earliest stages (caliche) to later stages in diagenesis. Much of the ferric oxide (now largely hematite) in the Peranera Formation, however, was supplied as parts of clastic particles. The Peranera deposits are, in part, primary red beds; the Bunter deposits, to the contrary, are post-depositional red beds (Krynine, 1949).

Diagenesis in the Bunter deposits is more complex: there is the general authigenesis of ferric oxides, kaolinization of muscovite, probably some early precipitation of quartz (quartz being the main cement), and authigenesis of dolomite. In the Bunter mudstones an onset of caliche formation is found in the form of calcite spherulites and vertically oriented calcite nodules, which were dolomitized at a later stage.

The differences in diagenesis between the Peranera

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[1] The physico-chemical conditions of formation of these minerals are not treated. Reference is made to general texts on the subject, e.g. Tröger (1967).

and Bunter red beds are attributable to differences in the depositional environment and the materials available. The depositional environment of the Peranera Formation was predominantly that of steppes bordering alluvial fans (p. 171); the prevailing climate must have been semi-arid.

The aridity of the climate prevented the alteration of feldspars (feldspar of volcanic origin; still remarkably fresh in many samples), and led to the formation of large amounts of caliche (calcrete, calcite nodules, and zones of early cementation with calcite). Caliche formation was also favoured by included limestone fragments in the generally immature clastic assemblage. The depositional environment of the Bunter Formation consisted largely of savannahs, with a moderately humid but seasonally dry climate (p. 171). The rate of deposition was probably low.

The Bunter clastic assemblage is mature (predominantly quartz, some quartzite, chert, and muscovite), which resulted from selective weathering in the source areas of

sediment (p. 212). The prevailing environmental conditions favoured strong *in situ* weathering of the deposits, which led to the authigenesis of ferric oxides, kaolinization of muscovite, and the early precipitation of quartz mentioned above. Caliche formation was limited by the climate and by a CaO-poor substrate. The Peranera alluvial plains probably fell dry after each downpour; those of the Bunter Formation may have dried up completely only during the dry seasons.

In both types of deposits, however, the penetration of air into the uppermost sediment levels led to the oxidizing conditions characteristic of subaerial diagenesis. The interstitial solutions in the surface levels of both the Peranera and the Bunter Formations were probably neutral to slightly acid due to decomposing organic matter. At lower levels, however, the solutions may have been neutral to alkaline as evidenced by the widespread calcite precipitation in the Peranera deposits, and, in the Bunter, the formation of authigenic dolomite.

APPENDIX

PRESENT-DAY SUPPLY OF FLUVIAL CLASTICS

Evidence from paleocurrent data and the composition of rock fragments suggests that the materials building up the sequence of sedimentary rock under study must have been supplied from northern directions. With the exception of the Malpas Formation, the clastic components of which derived from a temporary cover of volcanic deposits (Erill Castell Volcanics), the material can be assumed to have come from the rocks making up the Axial Zone of the Pyrenees. The main morphologic characteristic, namely southward-flowing streams draining the Paleozoic rocks of the Axial Zone, was therefore probably the same then as it is today.

With this assumption in mind, two samples of recent river sands were collected from each of six rivers draining the Axial Zone of the Pyrenees, with the intention of comparing their composition with that of the fluvial sandstones in the ancient formations. The conditions of formation of the recent river sands are, in brief, as follows: the altitudes of the sample localities vary from 900 m to 1200 m, the heights of the divides between the various drainage areas go up to well over 2500 m. Annual precipitation ranges from about 800 to more than 2000 mm (Lautensach, 1964), air temperatures average 10 to 13° C (Masachs Alavedra, 1954). These values represent gross generalizations, since there are small differences in climate from valley to valley. A detailed treatment of these differences, however, would lead us too far from our subject.

A geological map of the Paleozoic of the Axial Zone, the drainage patterns of the streams, and the sampling localities are shown in Fig. 70. Sands were collected from sand bars in the stream beds. After only drying

and sieving, the 2000 to 53-micron fraction of the sand was mounted in Araldite and thin-sectioned. Following the line-counting method, 200 grains in each thin section were counted and the averages of the countings of the samples for each river were computed. These data, which are shown in Table XVI, and the geological map in Fig. 70, form the subject of the following discussion.

The majority of the sand grains in each river is made up of non-calcareous phyllite and silty phyllite. Unfortunately, no distinction can be made optically between non-calcareous and silty phyllite grains coming from the Devonian (Peranera River; drainage area, upstream from sample locality, entirely in the Devonian) and from the Ordovician (Lladorre River; drainage area almost entirely in Ordovician). In both cases the grains contain abundant sericite and chlorite, and show similar tectonic features such as micro-knick zones and a pronounced first and sometimes a pronounced second cleavage. The silty phyllite is a transitional type in a range running from non-calcareous phyllite up to sericite/chlorite quartzite.

The sericite/chlorite quartzite, which is normally fine-grained, is particularly abundant in the Lladorre River. It is mainly an Ordovician component but also occurs in the Devonian (Peranera River). The other quartzite shown in Table XVI is a quartz-cemented, rather fine-grained, very clean sandstone of uncertain origin. Some phyllite rich in organic matter occurs in all the samples except those from the Lladorre River; these grains probably derive mainly from the Silurian black shales, although some may come from Carboniferous shales.

Calcareous phyllite grains (phyllite rich in replacement calcite), microsparry limestone, and macrocrystalline calcite (occasionally dolomite) grains are components deriving from the Devonian. They occur in all the samples except those from the Lladorre River, and are particularly abundant in the Peranera River. The samples taken in the Peranera River are slightly contaminated: about 5% of the material derives from the Erill Castell Volcanics, including grains composed of basaltic andesite, aggregates of macrocrystalline quartz and fine-grained kaolinite, and sand-sized angular kaolinite crystals (Fig. 71). These components have been subtracted from the counts.

The sands of the rivers whose drainage areas extend into the Maladeta granodiorites clearly show this fact. The influence of the granodiorites is seen in a some-
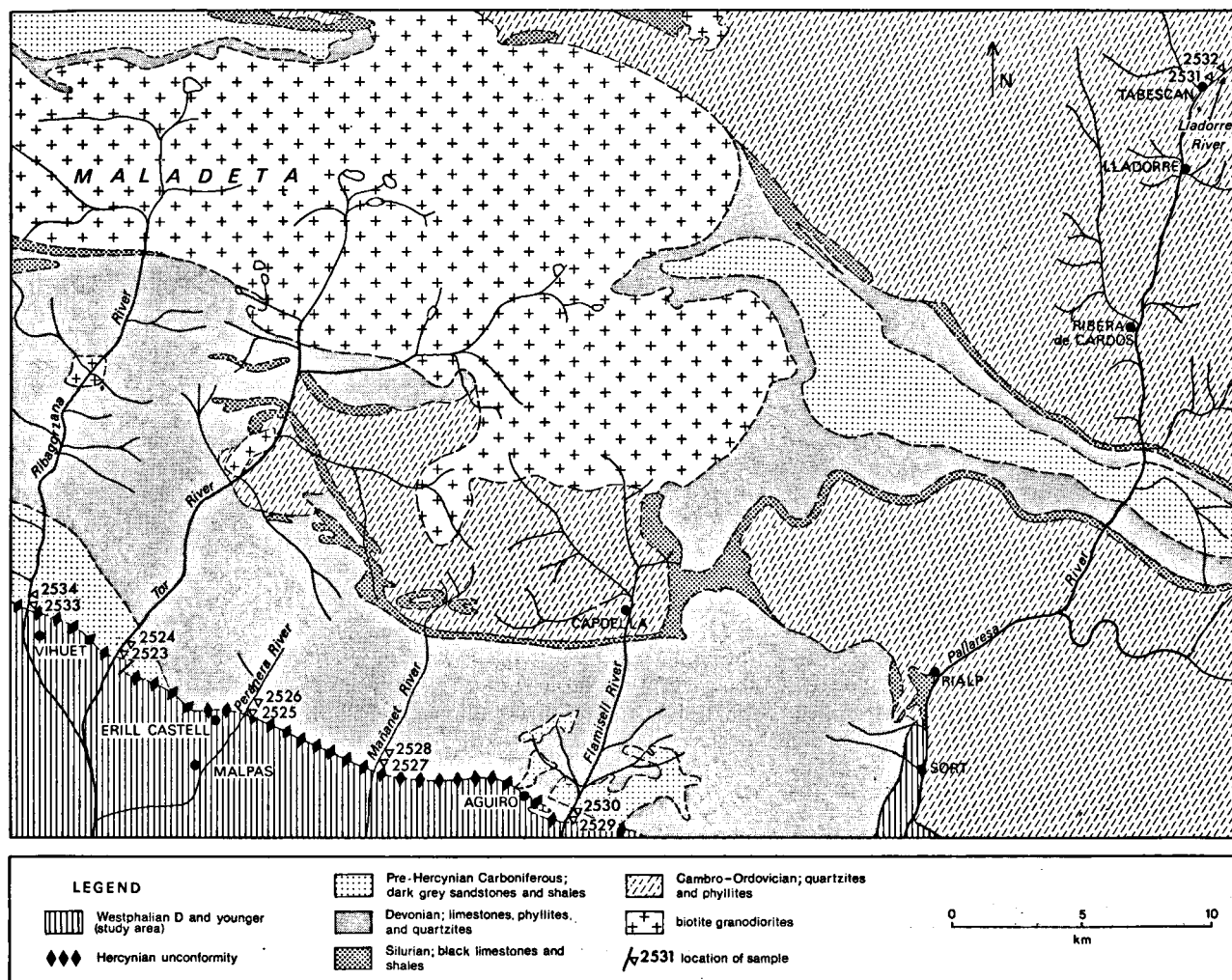

Fig. 70. Slightly simplified copy of part of the provisional Geological Map of the Central Pyrenees (L. U. de Sitter & H. J. Zwart).

what higher quartz content (angular to subangular grains in all observed cases), the presence of feldspars (andesine and occasional microcline), and the presence of biotite (slightly bleached in some cases). The influence of the granodiorites is particularly strong in the Ribagorzana and Tor rivers, both of which drain large areas on granodiorite; it is weaker in the Flamisell River, which also has less granodiorite to draw on. No influence of the granodiorites is seen in the sands of the Lladorre River, since the granodiorites (Auzat-Bassies Massif) occupy only a very small part of the drainage area (not shown in Fig. 70).

The rather high quartz content of the Mañanet River

sands, however, cannot be due to supply from the granodiorites (Fig. 70).

One disturbing factor requires mention: not all the supply comes directly from the outcrop areas as shown in Fig. 70.

Pleistocene glaciers once occupied the upper reaches of all the drainage areas under discussion (Birot, 1937), and locally, considerable amounts of lateral moraine material are still found plastered against the valley walls, fluvio-glacial terraces also being preserved locally. At present, these deposits are being actively eroded.

TABLE XVI

components	sample no	RIBAGORZANA RIVER	TOR RIVER	PERANERA RIVER	MANANET RIVER	FLAMISELL RIVER	LLADORRE RIVER
	locality	2534 / 2533	2524 / 2523	2526 / 2525	2528 / 2527	2530 / 2529	2532 / 2531
Quartz		22	12	3	9	8	3
Quartzite		3	2	1	2	1	1
Ser./chlor. quartzite		–	9	4	17	10	29
Non-calc. phyllite		22	36	37	27	49	22
Silty phyllite		23	25	10	28	26	43
Calc. phyllite		6	3	16	8	5	–
Microsp. limestone		2	3	18	6	5	–
Macrocryst. limestone		3	1	10	2	2	–
Feldspar		8	3	–	–	1	–
Biotite		5	5	–	–	1	–
Not determined		6	1	1	1	2	2
		100%	100%	100%	100%	100%	100%

Table XVI. Composition of recent fluvial sands.

The effect of the presence of glacially transported material favours the occurrence in the samples of material from the upper reaches of the drainage areas. Qualitatively, however, there will be no effect, because no major stream diversions have taken place in the area since the Pleistocene.

This brief survey of the percentages of rock fragments in the recent river sands does not bring out the relationship of the percentages to those of the areas occupied by the various lithologic units in the drainage areas. The conclusion that the sands consist predominantly of non-calcareous and silty phyllite and that the influences of the Devonian and of the granodiorites are clearly discernable suffices for the present purpose. It shows that the supply of clastics in the Westphalian D (Aguiró Formation) and in the Permian (Peranera Formation) did not differ greatly from that of today, although the granodiorites were probably not exposed in the earlier period (p. 178). But this conclusion does pose a problem for the case of the Bunter: there is no point of agreement between the composition of its clastic components and the assemblages found (p. 212).

Two additional observations deserve mention. The first is that a majority (about 70 to 80%) of the non-calcareous and silty phyllite grains and sericite/chlorite
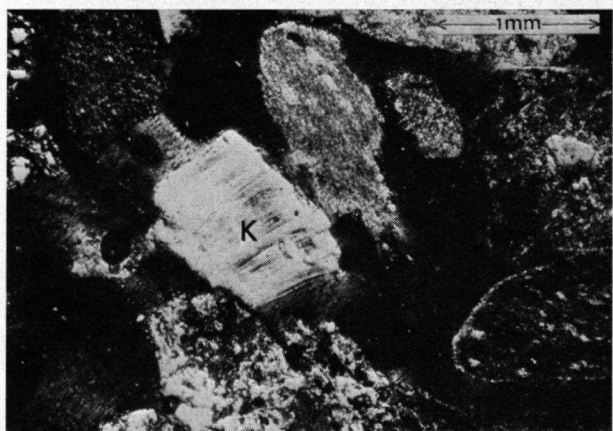
Fig. 71. Thin section of recent river sand mounted in Araldite. Kaolinite crystal from the Erill Castell Volcanics, transported as an angular clastic grain. Lowermost grain consists of altered Erill Castell tuff; the grain directly above the kaolinite grain is fine-grained Devonian limestone. Other grains are various types of phyllite. Sample 2526, nicols crossed (Araldite shows strain-birefringence).

Fig. 72. Thin section of recent river sand mounted in Araldite. Sandy phyllite and non-calcareous phyllite grains, impregnated to varying degrees with ferric oxides. Sample 2530, plain light.

quartzite grains carry varying amounts of ferric oxides, some grains being completely opaque (Fig. 72). In these grains the ferric oxides replace quartz, chlorite, and sericite. An arbitrary and patch-like distribution of the ferric oxides, and an absence of ferric oxides enveloping the grains show that the grains derive from
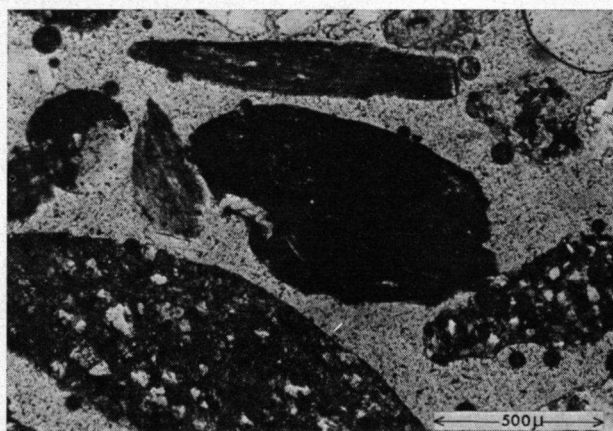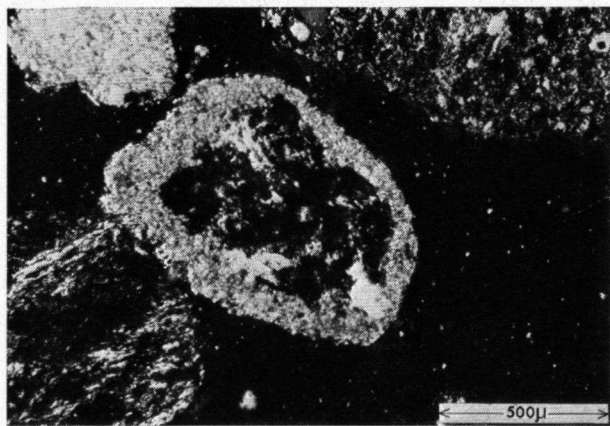

Fig. 73. Thin section of recent river sand mounted in Araldite. Sandy phyllite grain in the centre is stained dark by ferric oxide and carries an envelope of fine-grained calcite. Other grains are non-calcareous phyllite (lower left), silty phyllite (upper right), and limestone (upper left). Sample 2525, nicols crossed.

soil or weathering horizons in which ferric iron is concentrated. Identical grains were described from the Peranera Formation (30 to 40%; p. 201); but they were not found in the Aguiró sandstones. The occurrence in the recent fluvial sands of similar grains carrying ferric oxides lends support to the suggestion that, in the case of the Peranera Formation, they were likewise derived from weathering or soil horizons rich in ferric iron.

The second observation is that various grains in the recent sands are completely or partly surrounded by crusts of calcite (Fig. 73). The thickness of these crusts varies from almost zero to 150 micron. Some are distinctly laminated but others are homogeneous. The calcite occurs in porous or dense masses; it is invariably extremely fine-grained (mosaic textures composed of individual calcite crystals in the order of 5 micron or less).

The calcite crusts occur in all the samples except those from the Lladorre River (Ordovician); they are rare in the larger rivers (on less than 0.5% of the grains), but comparatively abundant (on 4 to 6% of the grains) in the small Peranera River, which drains an area rich in carbonates (Devonian). The crusts occur on grains composed of microsparry limestone, macrocrystalline calcite, and all types of phyllite. They were not noted on quartz and quartzite. Also, in some cases a calcite crust has no nucleus but a central void instead. Locally associated organic fragments in these cases show that the calcite probably formed on small twigs or roots.

In the cases in which the crusts occur on non-calcareous and silty phyllite, there is replacement of the silicate components of the grain by calcite progressing inward from the crust (Fig. 73).

The mode of formation of the calcite crusts is not fully understood. They formed during some stage in transport, because crusts surround rounded grains of varying grain-sizes present in the samples. A possible explanation may be that the calcite precipitated on the grains as travertine or as a result of caliche processes when transport was temporarily interrupted, as in fluvial terraces. An alternative explanation would be that the calcite was bound by algae in locally quiet parts of the fluvial system. Discontinuous crusts present on some grains do not support either of these explanations, since they could have formed due to abrasion in the stream or have resulted from incomplete development of the crusts due to the presence of contiguous grains.

SAMENVATTING

Het eerste hoofdstuk van de post-Hercynische geologische geschiedenis van het centrale deel van de Zuidelijke Pyreneeën ligt besloten in een opeenvolging van fluviatiele en vulkanische afzettingsgesteenten. Deze gesteenten variëren in ouderdom van Westfaal D tot en met Onder-Trias; de som van de maximale diktes van de gesteente-eenheden is meer dan 2300 m.

De huidige studie betreft de oorspronkelijke samenstelling van de gesteenten, de stroomrichtingen, het milieu van afzetting, het vroegere klimaat en de diagenese.

Vijf formaties worden onderscheiden:

    (5) Bontzandsteen-formatie (Onder-Trias)
    (4) Peranera-formatie (Perm)
    (3) Malpas-formatie (Stefaan)
    (2) Erill Castell vulkanieten (Stefaan?)
    (1) Aguiró-formatie (Westfaal D)

De afzettingen van de Aguiró-formatie zijn opvullingen van fossiele dalen die deel uitmaken van een geprononceerd oud reliëf. De formatie bestaat grotendeels uit conglomeraten afgezet door vlechtende rivieren. Eén niveau, bij Aguiró, bevat kool-houdende, tuf-rijke 'mudstone' en tuflagen; het milieu van afzetting komt overeen met dat van het laag gelegen gebied tussen de rivieren. De meeste klastische komponenten, waaronder veel niet-kalkhoudende phyllietkorrels, kunnen herkend worden als afkomstig van het Paleozoische grondgebergte (Axiale Zone van de Pyreneeën; ten Noorden van Aguiró).

De Erill Castell vulkanieten, die bestaan uit gesilificeerde en gekaoliniseerde tuffen en een ingeschakelde laag basaltische andesiet, werden afgezet op een heuvelachtig landoppervlak daar zij lokaal op fossiel hellingpuin rusten en enkele fluviatiele geulopvullingen bevatten. Er zijn aanwijzingen voor penecontemporaine bodemvorming.

De Malpas-formatie bevat koolhoudende schalies en 'mud-stones', zwarte vuursteen, kalklagen en ferro-dolomietlagen, (buiten-stroomgeul en lacustriene milieus), platen van in opwaartse richting fijnkorreliger wordende zandsteen (op-vullingen van voornamelijk meanderende rivieren), en een dik niveau conglomeraat (milieu van vlechtende rivieren). De verdeling van deze afzettingen in het kaartbeeld wijst op de opvulling van een fossiel dal. Bijna alle klastische kom-ponenten in de Malpas formatie zijn afkomstig van de onder-liggende Erill Castell vulkanieten, die reeds omgezet waren. De Peranera-formatie bestaat uit een caliche-bevattend rood-bruin gesteente-pakket. Afzettingsverschijnselen wijzen op het overwegend vlakke gebied benedenstrooms van puin-waaiers. Het oppervlaktewater stroomde intermitterend. Vele van de klastische komponenten zijn dezelfde als die in de Aguiró-formatie, en afkomstig van het Paleozoische grondgebergte. De Bontzandsteen-formatie, die eveneens uit fluviatiele, rood-bruine afzettingsgesteenten bestaat, ligt diskordant op alle oudere afzettingen. Het diskordantievlak wijst op een periode van pedimentvorming. De zandstenen bestaan grotendeels uit kwarts; zij bezitten de grootste mate van rijpheid van de gesteenten uit de gehele opeenvolging.

Het milieu in de Peranera- en Bontzandsteentijd moet veel geleken hebben op dat der huidige steppes en savannes, resp. Al de fluviatiele afzettingen tonen stromingsrichtingen die variëren, wat betreft het totaalbeeld, van West naar Oost, Noord naar Zuid, tot Oost naar West.

Het vroegere klimaat was van het grootste belang voor de afzettingseigenschappen, de oorspronkelijke samenstelling en het soort van diagenese in de afzettingen.

Een humide klimaat was verantwoordelijk voor continu stro-mend oppervlaktewater gedurende de afzetting van de Malpas-formatie en de met water gevulde laag gelegen gedeelten

buiten de stroom-geulen van de Aguiró- en Malpas-formaties. Vroeg-diagenetische parageneses van ijzersulfides, kaoliniet en sideriet en het bewaard blijven van organisch materiaal in deze formaties wijst op zwak tot sterk reducerende om-standigheden en neutrale tot lage pH-waarden.

Een semiaried klimaat (steppe) is in overeenstemming met het intermitterend stromen van oppervlaktewater, afgeleid uit stroomgeul-eigenschappen in de Peranera-formatie. Een meer continu stromen van het water in het Bontzandsteen-milieu wijst op een vochtiger klimaat (savanne). Het niet stabiel zijn van vegetatieresten (wortelgangen komen voor, maar er is nauwelijks enig ingekoold materiaal) wijst op veelal lage grondwaterniveaus en oxyderende omstandigheden. In weer-wil van de overeenkomstige roodbruine kleur, verschillen de Peranera- en Bontzandsteen-formaties sterk in oorspronkelijke samenstelling en diagenese. De klastische komponenten in de Bontzandsteen zijn zeer stabiel. Ferri-oxydes in de Peranera-zandstenen werden veelal klastisch aangevoerd; in de Bont-zandsteen zijn ze authigeen. De diagenese in de Peranera-af-zettingen is hoofdzakelijk beperkt tot vorming van, en ver-vanging door, calciet (caliche); in de Bontzandsteen-afzet-tingen kunnen *in situ* kaolinisatie van muscoviet, mogelijke vroege authigenese van kwarts, en latere authigenese van dolomiet worden waargenomen. Deze verschillen staan ge-deeltelijk in verband met de vroegere morfologie (Peranera-formatie: nog een duidelijk reliëf; Bontzandsteen-formatie: afzettingen op uitgestrekte pedimenten), maar hoofdzakelijk met het paleoklimaat (Peranera-formatie: steppe klimaat; Bontzandsteen-formatie: savanne klimaat).

De samenstelling van recente fluviatiele zanden afgeleid van de Paleozoische kern van de Pyreneeën wordt behandeld in een Appendix. Deze zanden zijn, qua samenstelling, vergelijk-baar met de zandstenen in de Aguiró- en Peranera-formaties.

SUMARIO

El primer capítulo de la historia geológica, post-hercínica, de la parte sur de los Pirineos Centrales está basado en una sucesión de depósitos fluviales y volcánicos, cuya edad varía desde el Westfaliense D hasta el Triásico inferior, inclusive, con una suma de espesores máximos que sobrepasan los 2300 m.

El estudio presente trata la composición original, las direc-ciones de corriente, el medio ambiente de deposición, la paleoclimatología y la diagénesis de estos depósitos.

Podemos distinguir cinco formaciones:

    (5) formación del Bunter (Triássico inferior)
    (4) formación de Peranera (Pérmico)
    (3) formación de Malpas (Estefaniense)
    (2) formación de Erill Castell (Estefaniense?)
    (1) formación de Aguiró (Westfaliense D).

Los depósitos de la formación de Aguiró lo constituyen valles fósiles rellenados que formaban parte de un paleorelieve pronunciado. La formación consiste en su mayoría en con-glomerados depositados por corrientes entrecruzadas. Un nivel, cerca de Aguiró, contiene 'mudstone' rico en tufo y carbón, y capas de tufo; el medio ambiente de deposición corresponde al de una ciénaga. La mayoría de los componentes clásticos, muchos de los cuales son granos de filitas no-calcáreos, pueden ser reconocidos como procedentes del basamento Paleozoico (la Zona Axial de los Pirineos, al norte de Aguiró).

Las volcánicas de Erill Castell, que consisten en tufos silifi-

cados y caolinizados y una capa intercalada de andesita basáltica, fueron depositados en una superficie terrestre on-dulada, ya que descansan localmente sobre escombros fósiles de laderas y contienen algunos rellenos de canales fluviales. Hay indicaciones de pedogénesis penecontemporánea.

La formación de Malpas contiene esquistos arcillosos hullíferos y 'mudstones', capas de caliza y capas de pedernal, ferro-dolomitas (medio ambiente de ciénaga o lacustre), bancos de arena cuyos granos se afinan hacia arriba (rellenos de meandros fluviales) y un nivel grueso de conglomerados (medio ambiente de ríos de corrientes entrecruzadas). La distribución superficial de estos depósitos indica el relleno de un valle fósil. Casi todos los componentes clásticos de la formación de Malpas provienen de las volcánicas subyacentes de Erill Castell, que ya estaban alteradas.

La formación de Peranera es una sucesión de capas rojas que contienen caliche. Los rasgos de deposición indican una zona principalmente llana bordeando conos aluviales, con corrientes intermitentes. Muchos de los componentes dedritogénicos son similares a los de la formación de Aguiró y procedentes del basamento Paleozoico.

La formación del Bunter, compuesta también de capas rojas fluviales, yace discordante sobre todos los depósitos anteriores. La superficie inferior indica un período de formación de pedimentos. Las areniscas del Bunter consisten principalmente en cuarzo y son los de mayor grado de madurez de toda la sucesión.

El medio ambiente de las formaciones de Peranera y Bunter se habrá parecido probablemente mucho al de las actuales estepas y savanas, resp.
Todos los depósitos fluviales muestran direcciones de corriente que varían del oeste al este, del norte al sur y del este al oeste. El clima antiguo ha sido de gran importancia para las características de deposición, la composición original y el tipo de diagénesis de los depósitos.

El clima húmedo fue responsable de corriente contínua de agua superficial durante la deposición de la formación de Malpas y las ciénagas inundadas de agua de las formaciones de Aguiró y Malpas. En estas formaciones, la existencia de paragénesis de marcasita, pirita, caolinita, y siderita, formadas por una diagénesis temprana, y la conservación de material orgánico, apuntan unas condiciones de reducción de débiles a fuertes y un medio ambiente de neutral a ácido.

Un clima semiárido es compatible con la existencia de corrientes intermitentes de agua superficial, que se deducen de las características de los canales fluviales en la formacion de Peranera. En los canales fluviales de la formacion del Bunter el agua corria más continua. La inestabilidad de los restos orgánicos (rastros de raíces presentes pero apenas hay material carbonizado) indica un nivel bajo del agua sub-

terráneo y condiciones oxidantes. A pesar de la semejanza en color rojo, se diferencian fuertemente las formaciones de Peranera y Bunter en su composición original y en su diagénesis. Los componentes clásticos en la formación de Peranera son en su gran mayoría poco estables, en la Formación del Bunter son muy estables. Los óxidos ferrícos en las areniscas de la formación de Peranera han sido traídos en forma clástica, en las areniscas del Bunter son authigenéticos. La diagénesis en los depósitos de la formación de Peranera se limita principalmente a la formación de, y el reemplazamiento por, calcita (caliche); en los los depósitos del Bunter se observan hidrólisis de silicatos, posible autigénesis temprana de quarzo y authigénesis de dolomita. Estas diferencias en parte están relacionadas con la topografiá antigua (formación de Peranera: un relieve aún marcado; formación del Bunter: pedimentos extensos) y en parte con el antiguo clima, entendiendo que el clima fue menos árido durante la deposición del Bunter.

La composición de arenas fluviales recientes, derivadas del núcleo Paleozoico de los Pirineos, es tratada en un apéndice. Estas arenas son comparables, en cuanto a su composición, con las areniscas de las formaciones de Aguiró y Peranera.

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