SEDIMENTARY PETROLOGY OF THE LOWER LIMESTONE GROUP IN THE CARBONIFEROUS CENTRAL BASIN, MIDLAND VALLEY OF SCOTLAND

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A thesis submitted for degree of Doctor of Philosophy

by

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#### ABSTRACT

The Lower Limestone Group was studied in the area of Stirlingshire and Lanarkshire. It consists of alternating limestones, shales, siltstones, sandstones and shales with ironstone nodules and bands. The investigation of the clastic sediments revealed that they were deposited by deltas prograding from the north east. Facies analysis of these deltaic sediments shows the predominance of four main facies, progradational, interdistributary bay, abandonment and channel facies. Six delta advances were interpreted in this study, post-Hurlet, post-Shield' Bed, post-Blackhall, post-Main Hosie, post-Mid Hosie and post Second Hosie Limestone. The study of post-Blackhall delta revealed that it was built by a constructive, fluviallyinfluenced, elongate delta(s) which prograded into a deep basin(s), whereas the other deltas prograded into shallow Changes in the depth of the basin and the basins. sediment load of the rivers, which were probably due to changes in tectonics, climate and eustatic changes in sea level, probably resulted in the differences in thickness of the studied deltas. Petrological study indicated the predominance of stable light and heavy minerals, suggesting the source area consisted of sedimentary rocks, in turn derived mostly from a metamorphic terrain.

The study of the carbonate rocks showed that there are six main facies. Biomicrite and micrite facies (indicating a low energy subtidal environment), dolomitic

biomicrite and micrite (higher salinity subtidal), Ostracodbearing dolomite (hypersaline lacustrine or lagoonal) and calcareous siltstones and sandstones (nearer shore environment than the other facies). Two main penecontemporaneous types of dolomite were deduced; (a) dolomitic rocks that are either unfossiliferous or contain marine fauna and (b) Ostracod-bearing dolomitic rocks. Late diagenetic recrystallisation gives the dolomite the superficial appearance of a late stage replacement while more detailed study shows that it formed early.

Two types of ironstone nodule were recognized, zoned and non-zoned. The mineralogical zoning suggests that there was a change in the chemical environment during the formation of some nodules.

The study of the trace and some major elements indicated that the clay, muscovite and organic material are the location of Ni, Cu, Co, Zn, Pb, Ti and Rb in the carbonates and shales. The Ba, Mn and Fe are located in the carbonate minerals. The high values of Sr in the carbonate rocks suggest an original aragonitic composition for the carbonate.

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CHAPTER 1

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INTRODUCTION

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## 1.1 INTRODUCTION

The Lower Limestone Group has been studied in the Carboniferous Central Basin of the Midland Valley of Scotland from Stirlingshire to Lanarkshire (Fig. 1). The distribution of the Carboniferous sediments in the Midland Valley of Scotland is shown in Figure 2. The age of the Lower Limestone Group is considered to be Brigantian (George <u>et al.</u>, 1977). It is underlain by the Calciferous Sandstone Measures and overlain by the Limestone Coal Group (Craig, 1965).

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Previous studies of the Lower Limestone Group in this area have been mainly stratigraphical and have been carried out mostly by geologists of the Geological Survey. They divided the Central Coalfield into nine different areas in seven of which, described by Carruthers and Dinham (1917); Hinxman et al. (1920); Clough et al. (1920); Hinxman et al. (1921); MacGregor and Anderson (1923); Clough et al. (1925); Dinham and Haldane (1932) and MacGregor and Haldane (1933), the Lower Limestone Group is exposed. Although their stratigraphic work was very detailed and is taken as the basis for this study, they only described the colour. fossil content, and sometimes gave the thicknesses of some of the exposed lithologic units. They reported that the Lower Limestone Group consists of seven carbonate units which alternate with shales, siltstones, sandstones, shales with clay-band ironstones or nodules and occasional thin seams of coal. However, Crampton (in Clough / 1925, p.35) indicated that the shale sediments represented a deltaic



fig. 1 Location map showing the studied area, (modified from Craig, 1965).



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fig. 2 Geological sketch map of the Midland Valley of Scotland showing the distribution of the Carboniferous sediments (after Rayner, 1967) environment and stated that they were deposited "... in or near the sea at the front of the delta of a sluggish river of large volume bringing seaward muds full of organic detritus". He added that the carbonate units were deposited in a comparatively shallow sea at the front of this delta. Read (1959) and Francis et al. (1970) redescribed the stratigraphy of the Lower Limestone Group in the Stirling area. At the Weatherlaw Inlier (section  $28 \triangle$ Fig. 10), a partial section of two carbonate units is exposed separated by shales, siltstones and sandstones. Dinham and Haldane (1932) considered the lower of these to be the Hurlet Limestone and the upper unit to be the Shields Read (1959, p.9) stated that the lower carbonate Bed. unit is not the Hurlet limestone but the Shields Bed while the second represents the base of the Blackhall Limestone. In this study Read's interpretation is followed.

A typical section of the Lower Limestone Group is presented in Figure 3. Different names have been given to the same carbonate unit in different areas by previous workers. In this study, the names shown in Figure 3 are followed.

Goodlet (1957), studied the lithological variations in the Lower Limestone Group by isopach, isolith and ratio maps which show the relative proportions of the shale, limestone and sandstone rock-types and concluded that the sediments of the Lower Limestone Group were laid down by the delta of a large river which debouched from the north. Belt (1975, p.446) suggested a destructive, wave-dominated



Fig.3 Vertical section of the Lower Limestone Group at Hurlet (After Craig, 1965).

Formation names used in this st	udy	Equivalent names
Top Hosie	:	Calderwood Cement, Lingula or Calmy Limestone.
Second Hosie	:	Anvil Limestone, First Kingshaw
Mid Hosie	:	Middle Limestone, Second Kingshaw
Main Hosie	:	Under Limestone, Birkfield, Third Carron
Blackhall	:	Foul Hosie, Fankerton, Wee Limestone
Shield <sup>5</sup>	:	Craignhill Limestone, Limestone C.
Hurlet	:	Murrayshall, Main Limestone.

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delta as the environment of deposition for the sediments of the Lower Limestone Group in the St. Monance area to the NE of the studied area in Fife.

Selim and Duff (1974) studied the carbonate members of the Lower Limestone Group in East Fife. They concluded that the carbonate units were deposited in a marine environment which, on the basis of lithological variations, varied between being weakly to highly agitated. Brown (1977) studied the Hurlet Limestone (the lowest carbonate unit of the Lower Limestone Group) in the north of Ayrshire, to the south of the present studied area. He indicated that it was deposited in a shallow, subtidal, normal marine environment. He added that at the top of the Hurlet Limestone there is evidence of exposure.

The above review shows that there has been no specific and detailed work on the Lower Limestone Group sediments in the studied area. The main conclusion based on previous work is that the non-carbonate sediments were deposited on the delta of a sluggish river and the carbonate units were deposited in a relatively shallow marine environment.

Read (1959) pointed out that the Lower Limestone Group consist of cycles and he proposed the following ideal cycle for the Lower Limestone Group sediments in the Stirling area, although no individual cycle contains every subdivision.

a	Coal	
1	Seat-earth	
k	Fakes	
j	Sandstones	
i	Fakes	
h	Blaes and ironstone concretions, with	th non-marine
	Lamelli_branch5 .	
g	Blaes and ironstone concretions	
f	Calcareous blaes	with
е	Limestone	marine
d	Calcareous blaes	fossils
с	Blaes and ironstone concretions	
b	Carbonaceous blaes	
a	Coal	

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Note:- Blaes is an old mining term for mudstones or shales and fakes are thin-bedded argillaceous sandstones or sandy mudstone.

The shales, siltstones and sandstones together form coarsening-upward sequences interpreted in this study to be the result of delta progradation. Transgression of the sea followed delta abandonment and the carbonate members were deposited.

The aim of the study was to confirm the environment of deposition of the non-carbonate sediments as deltaic, and to classify the deltaic sediments into different environmental facies. The type of the deltas in which the sediments were laid down and the paleogeography are topics given specific attention in this study.

For the carbonate sediments, the study involved the classification of the carbonates into environmental facies, petrology, diagenesis, dolomitisation, trace elements, and some major element chemistry. Specific attention was given to sideritic nodules and bands and to the interpretation of the mineralogical and morphological variation within the sideritic nodules.

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The stratigraphic sections which were studied in the Central (oalfield are mainly stream sections. However, a few partial sections are exposed in old quarries. The grid references of the sections are presented in Table 1 in and their distribution with the studied area is shown in Figure 4. TABLE 1 The Grid References of the Visited Sections

Secti No.	on	Grid Reference	Details of Sections
1	NS	798419	Birkwood Burn.
2	11	804438	Nethan River below Auchenheath House
3		817463	Nethan River below Corramil.
4	11	736454	Crumhaug Burn, near Stonehouse
5	11	740456	Lounsdale Burn, near Nethan River
6		734457	Minholm section, Avon River
7		742466	Avonholm Section, Avon River
8	44	854495	Quarry at Fiddler Burn, near Headsmuir
9	u	873488	Lemuir Quarry, near Headsmuir.
10	L	871516	Thorn Quarry.
11	16	828499	Jock's Gill
12	61	873472	Quarry at Craigen Hill
13	¢.	855480	Nelfield Quarry.
14	ti	836488	Quarry at Oldhill.
15	4	668513	Quarry near Mid-Drumloach
16	1	680788	Corrie Burn
17		657776	2 stream sections between Shield Burn and Spouthead.
18		595757	Blairskaith Quarry.
19	ŧ	661777	Shields Burn.
20	T	640785	Sloughneagh Section.
21	ł	634788	About 500 m N.E. of Balgrochan.
22	U	644786	Baught Glen.
23		615776	Baldow Glen.
25		741877	Bannock Burn
26	1	760891	Sauchie Craig South.

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Section No.	Grid Reference	Details of Sections
27 N	/5 779884	Burn near Old Sauchie
28	"    764875	Weatherlaw Inlier
29	" 798831	River Carrogn
32	" 404656, 412655	River Gryfe, Bridge of Weir
33	" 401605	Howwood
34	· 509597	Quarry at Liverholm.
36	·* 466611	Limeraig Quarry.
37	4 <b>513652</b>	Arkelston Junction
38	· 503599	Hurlet
39	• 657543	Calderwood Glen
39 <b>A</b>	·· 658547	Calderwood Glen
40	·· 662542	Calderwood Glen
41	·· 594547	Quarry at Thorntonhall
42	a <b>621551</b>	Kittoch Water Section
44	··· 587536	Gill Burn
45	<b>992653</b>	River Almond
46	" 9986 <b>28,</b> 986626	Skoli Burn and Breich Water
47	NT 008750	Beecraigs Wood
48	NS 991715	Silvermine Quarry
49	" 98569 <del>4</del>	Quarry at Bathgate Area
50	988693	West Kirkton Quarry
51	·· 607550	Barrhead
52	<b>498592</b>	Quarry near East Kilbride.

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#### 1.2 TERMINOLOGY

The environment of deposition of the non-carbonate sediments of the Lower Limestone Group is considered to be deltaic. It is first necessary to define some of the terms used in the description of delta morphology which will be used in the forthcoming discussion and interpretation of the Lower Limestone Group sediments

Most authors agree that there are three simple main components of a delta; these are:

- 1. Delta plain
- 2. Delta front
- 3. Prodelta.

## 1.2a Delta Plain Environment

This environment includes the deltaic distributary channels with their associated natural lovees and the area between these channels (Fig. 7), which includes, marshes, swamps, lakes and bays (Fisher, 1969a; Donaldson et al., 1970). Coleman and Gagliano (1964) and Allen (1965) pointed out that these delta plains are characterized by shallow water and they are areas of extensive vegetation. Gould (1970, p.5) described the Mississippi delta and noted that the delta plain "... holds many shallow brackish and fresh-water lakes and is indented along its margins by brackish and marine bays". He added also that the surface of the delta plain is zoned into four broad belts grading gulfward along the seaward margin of the plain from the swamp environment through the fresh-water marsh, brackish-

water marsh and saline marsh (Fig. 5).

Elliott (1973, 1974b) summarized the depositional processes, morphological forms and the sedimentary sequences found in the delta plain environemnt. He used the term "interdistributary bay" to include the areas of shallow water (1-4 m deep) between the delta distributaries, which may be open to the sea or partially or entirely closed; and therefore may be fresh, brackish or marine. This usage is adopted in this study. He also added that the bay received sediments either by flood generating processes (the main processes) which include overbank flooding, crevassing and avulsion, or, when the interdistributary bay is open, by longshore currents which produce sand spits and beaches. These processes produce sequences of sediments, the majority of which are small-scale coarsening-upwards sequences.

#### 1.2b Delta Front Environment

This area includes the coarse sediments associated with delta progradation. The nature of the characteristic features of this environment which includes river mouth bars, sand spits, beaches and coastal barriers sands depends on the ratio of the sediment input to the marine reservoir energy (Fisher, 1968; Scott and Fisher, 1969).

1.2c Prodelta Environment

This includes the sediments seawards of the delta front. Although prodelta sediments are largely of clay,



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fig. 5 D

Distribution of swamp and marsh environment on Mississippi deltaic plain. After Gould (1970, fig. 8). they may include a little very fine sand and silt.

1.2d Types of Delta

The classification proposed by Scott and Fisher (1969) is adopted in this study. They classified deltas into four main types; these are (see also Fig. 6).

A - High-constructive deltas - fluvially influenced type.

- 1 Elongate type
- 2 Lobate type.

B - High-destructive deltas

- 1 Wave dominated
- 2 Tide dominated.

#### 1.2d(i) High-constructive deltas

Coleman and Gagiliano (1965) noted that during the progradation of the Mississippi delta, fine sediments, mostly clays), were deposited offshore forming the prodelta facies. Silts and clays represent the distal facies of the mouth bar, whereas coarse sediments (mainly of sands and silts) represent the proximal facies of the mouth bar. Frazier (1967) in his description of the fluvially influenced, constructive type Mississippi delta, pointed out that most of the sands deposited at the distributary mouth are buried by more sands although some is redistributed laterally by waves, while the finer grained sediments are carried further away from the mouth. He indicated also that the delta formation begins with the progradation of a shoreline producing distributary mouth bars the



sediments of which have/coarsening-upward trend.

Fisher (1968) described the delta system of the Gulf Coast Basin (Eocene) and reported that constructive deltas formed at the mouths of a few very large streams in which there was extensive shoreline progradation of elongate to lobate terrigenous delta lobes. He indicated also that the ratio of the progradational to destructional facies is high within this type of delta. Fisher (1969a) pointed out that a delta system containing a large proportion of fluvially influenced facies is considered to be a constructive type and that such deltas are the product of progradation and aggradation. He added that this type of delta is developed under conditions of high sediment input relative to the marine reservoir energy. Scott and Fisher (1969) indicated that delta systems made of large proportion of fluvially influenced facies represent high constructive deltas in which shell concentrations and burrows are restricted to the crest of the distributary mouth bars due to delta abandonment.

### 1. Elongate type:

This type of delta is constructed by a few distributaries (Gould, 1970) and forms under high sediment input containing a high proportion of mud resulting in sand facies prograding over relatively thick mud sequences. Fisk (1961) indicated that the elongate lenticular sand bodies (bar-finger sands) of elongate deltas, consist of well sorted, fine and very fine sands which grade

downwards and laterally into massive delta front clayey He added also that the bar-finger sands are thick silts. directly beneath the distributary channels and show an irregularly decrease in thickness towards their lateral margin (Figs. 7 and 8). Fisk et al. (1954) and Fisk (1961) noted that when the underlying muds of the bar-finger sand are thick, mud lumps or diapirs may develop and their upward movement creates local structures in the overlying sand masses, the recognition of which could be used for the recognition of bar-finger sands in ancient sediments. Fisher (1969b) noted that the vertical variety of facies in the elongate delta sequence contrasts with the prominent lateral persistence of specific facies in lobate high constructive deltas. He added that elongate deltas are developed by progradation over thick mud. Gould (1970) indicated also that the bar-finger sands are distinguished by their geometric form, lithologic features and the relationship to their adjoining fine grained sediments.

## 2. Lobate delta type

This type of delta develops under conditions of high sediment input by numerous streams with relatively little mud, in comparison to the elongate delta type, and produces sand facies which prograde over a thin mud sequence (Fisher, 1969b; Scott and Fisher, 1969). Fisher (1969b) added also that this type of delta is characterised by the lateral persistence of specific facies.



associated facies (after Fisk, 1961).

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## 1.2d(ii) High Destructive Delta

Marine and river processes interact at the delta shoreline and the relative importance of these processes is shown by the delta morphology. When the sediments input is moderate and is slightly exceeded by the energy of coastal processes, a destructive delta is produced in which the ratio of constructional to destructional facies is relatively low (Fisher, 1968). Scott and Fisher (1969) reported that in the destructive delta the fluvially introduced sediments are contemporaneously reworked by marine processes such as waves and tides and they reported two varieties within this delta type (Fig. 6).

#### 1. Tide dominated delta

In this type of delta, the fluvially introduced sediments are redistributed by tidal currents, consequently a series of digitate sand units which normally radiate from the front of the river mouths are produced.

#### 2. Wave dominated delta

This delta is characterised by cuspate to arcuate trend of the sand units which are accumulated in a series of coastal barrier sands flanking the river mouth.

CHAPTER 2

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THE SEDIMENTOLOGY OF THE CLASTIC SEDIMENTS

1.1.1

## 2.1 SEDIMENTARY FACIES

The clastic sediments of the Lower Limestone Group are subdivided into four main facies based on vertical sequences of lithology, sedimentary structures and fossils. The characteristics and the inferred environment of deposition of each facies is presented in Table 2 and Figures 9-14.

## 2.1a Facies A

This facies consists of fossiliferous marine shales which pass gradually upward into laminated non-fossiliferous shales then into siltstones interbedded with shales or silty claystones and finally, in some cases, into sandstones (Fig. 15). The thickness of these coarseningupwards units range from 14.3 metres to 56.7 metres. Mostly, this facies is overlain by thin fossiliferous siltstones or very fine sandstones (facies C). Smallscale coarsening-upward sequences (facies B) sometimes overlie this facies.

The siltstone beds range in thicknesses from 2 cm to 10 cm and their thicknesses increase upward accompanied by the decreases in the thicknesses of the interbedded shales. Internally they have alternating dark (0.1-2 mm) and light (2mm - 1 cm) laminae which exhibit parallel or micro-cross laminations. Sometimes wavy laminae are present but are not abundant. Petrographic study indicates that the dark laminae consist mainly of heavy minerals (the elongated grains lie parallel to the laminae)


















Table 2:

Summary of the recognised facies.

..... continued

<b></b>				>	Facles	
Coarsening-upwards from shale at the base to silt- stones and sandstones. Rootlets and coal are common. The environ- ment of deposition is interdistributary bays				Coarsening upwards, without rootlets, normally grades from A marine shale at the base. The environ- ment of deposition is prograding deltate lobes. # fossiliferous	Characteristic features and environment of facies	
BI	<b>A</b> 4	εv	72	A1	Sub- facles	
l. Parallel Lam.	Same as above	Same as above	Same as sub- factes Al	1. Parallel Lam. 2. Wavy Lam.	Sed Imentary Base	
1. parallej Lam. 2. Wavy Lar	Same as subfacies Al and A2	Parallel and wavy laminations	Same as sub factes Al	1. ParallelLam 2. Wavy Lam 3. Micro-cross	Middle	
<ol> <li>Micro-cross Laminations.</li> <li>Symmetrical ripples when the bedding planes are exposed.</li> <li>Occasionally cross-bedded units.</li> </ol>	1. Micro-cross Lamination.		<ol> <li>Micro-cross</li> <li>Laminations.</li> <li>Apparently</li> <li>Rare cross-</li> <li>Bedding.</li> <li>Symmetrical ripple marks when the bed- ding plane is exposed</li> </ol>	. 1. Cross bedding 2. Apparently massive 3. Micro-cross Laminations	Тор	
<ol> <li>The sandstone units have sharp erosive boundaries.</li> <li>Rootlets and coal are common.</li> <li>Sometimes cut by a channel.</li> </ol>	1. The thickness of shale at the base of the sequence roughly equal to the amount of silt- stones and sandstones at the top.	<ol> <li>No deposition of coarse sediments.</li> <li>Overlain by facies C.</li> </ol>	<ol> <li>Lot of shale interbedded with the top sandstone</li> <li>Normally overlain by facies C and B.</li> <li>Low amount of cross- bedding.</li> <li>Thick shale at the base.</li> </ol>	<ol> <li>The amount of shale is low or negligible within the top sandstone units.</li> <li>Normally cut by channel.</li> <li>Thick shale at the base.</li> </ol>	Characteristic features of the subfacies	
Sudden incursion by crevasse splays	Small scale delta progradation.	Distal position on distributary mouth bar with negligible fluvial influence.	Lateral position on distributary mouth bar with moderate fluvial influence	Axial position on distributary mouth bar with fluvial influence dominant	Environment of the subfacies	
3-7 metres	14.3 m	Thickness r	anges between 20 m to to decrease towards t	be distal facies.	ness	

27 A

Ð	o	B Contd.					Facies	
Distributary channels characterised by 1.The Lower surfaces are scour erosional surfaces. 2. Mostly non- fining upwards	Abandonment facies, two types. a. Fossilifer- ous silt- sandstones b. Coal.							
8 -		B5	B4	B3	В2	facies	Sub-	
<ul> <li>Large scale cross bedding.</li> <li>Horizontal bedding</li> </ul>		Same as above	Same as above	Same as above	Same as above	Base	Sedime	
		Same as above	Same as above	Same as above	Same as above above	Middle	ntary Structures	
		Same as B2 and B3	Same as sub- facies B2	Same as sub- facies B2	Same as above but no cross- bedding units were observed.	Top		
		Marine fossils in the basal shales	Hypersaline fos- sils in the basal shales	<ol> <li>Normally the units have units have sharp erosion- al bases.</li> <li>Sometimes associated with borrows.</li> <li>Large amount of shale inter- bedded with the coarse sedi- ments</li> </ol>	1. Gradational upwards	the subfacles	features of	
Straight distribut- ary channels for the non-fining upwards and meandering for the fining upwards sequences.	<ol> <li>Blogenic rework- ing and periods of non-deposition for the fossili- ferous units.</li> <li>Marshland area for the coal.</li> </ol>	Produced by similar processes of sub- facies B2 and B3 into open bay.	Slow incursions of sediments by crevasse splays into hypersaline (Lagoonal or Lacustrine)bay.	Levee progradation by overbank flooding	Minor mouth bars produced by slow incursions of sediments on crevasse splays.	the subfacles	Environment of	
1-8 metres	5-80 cm.	6-10 m.	8.7 m	10-15 metres	4-8 m	Thic	k.	

27 B



and organic material with a minor amount of light minerals (e.g. quartz). The light laminae consist mainly of quartz with lesser amounts of heavy minerals and organic material (Figure 16).

The sandstones are moderately sorted, of yellowish to brownish colour and range.in thicknesses from 15 cm to 80 cm. Sedimentary structures include large scale trough cross-bedding and micro-cross laminations. In most of the sections, there is no definite vertical sequence of structures and mostly the sandstones appear to be massive. However, these apparently massive sandstone units contain carbonaceous fragments which are inclined to the bedding. Climbing ripples were observed in some sections. Symmetrical wave ripple marks with an average wavelength of 4.5 cm and amplitude of 0.6 cm are common in some sections and restricted to the top of the sequences. A few sandstone units, which range in thickness from 8 cm to 50 cm, were found within the upper part of the shales or within the siltstone sediments. They are apparently massive and have sharp bases.

The coarsening upward nature, the presence of marine fauna in the shales at the base of the sequence, the high proportion of coarse sediments and the presence of thin fossiliferous siltstones and very fine sandstones (facies C) at the top of the facies, indicate that it was deposited by the progradation of constructive, fluvially influenced deltaic lobes into the marine environment. In such a delta progradation the coarse sediments debouched by



streams are dropped at the mouth of the streams forming distributary mouth bars. The interbedded siltstones and shales represent the deposition on the distal parts of these bars, whereas the sandstones form the proximal facies of the mouth bar sediments. The fine material was carried away and deposited below the reach of wave action and represents the prodelta sediments.

Leeder (1972, 1974) interpreted the coarsening-upward sequences, with marine fauna within the basal mudstones, in the Lower Border Group (Tounaisian) of the Northumberland Basin, as having been formed by the progradation of constructive delta complexes into a marine environment. Elliott (1976) also considered that thick coarseningupward sequences sometimes overlain by thin fossiliferous siltstones and sandstones, in the Carboniferous sediments of Bideford Group of North Devon (England), were formed by the progradation of constructive river-dominated elongate delta type.

Coleman and Gagliano (1965) pointed out that the distal sediment (mainly silts with clays) of the mouth-bar of the Mississippi delta show parallel laminations and wave and current produced microcross laminations. They added that the coarse sands and silts of the mouth bar are reworked not by stream currents, but also by waves generated in the open water beyond the channel. They indicated also that parallel laminations could be formed by segregation of particles by differential settling and wavy laminations could be produced by minor irregularities

and obstructions on the deposition surface. Allen (1970) pointed out that ripples and cross laminations are the commonest structures found in fine sediments while dunes and their resulting cross-bedding are typical of coarse sediments.

The parallel laminations which are shown up by the heavy and light minerals in the siltstones of the studied sequence could have been produced by settling from suspension. Due to the difference in the specific gravity of the minerals, alternating parallel laminae of heavy minerals and light minerals could be produced as a result of the sorting action of the random fluctuations of the depositing current. The microcross laminations could be produced by current and wave action. The abrupt alternation of the parallel and micro-cross laminations might be due to fluctuation in sediment supply and current velocity which probably reflect changes in the weather and random fluctuations in river discharge. The wavy laminations might be due to minor irregularities and obstructions on the surface of deposition.

Allen (1970) pointed out that the large scale crossbedding is formed by the migration of subaqueous sand dunes. He added that tabular sets are the product of the migration of straight-crested bed forms, and trough crossbedding the product of linguoid bed forms. Hamblin (1965) reported that sandstones which are apparently massive due to their homogeneity often consist of large scale crossbedding and micro-cross lamination which can only be seen in

x-ray radiographs. Scott and Fisher (1969) indicated that multi-directional trough type cross-bedding produced by oscillation and current ripples are the main sedimentary structures in the distributary mouth-bar sands of the Mississippi delta.

The trough cross-bedded sandstones of this facies indicate bed-load movement represented by migration of subaqueous linguoid sand dunes. The micro-cross laminations, might be due to wave or current action. The apparently massive sandstones might consist of internal stratification since the sandstones are well-sorted and therefore homogeneous. The apparently massive sandstones might also reflect rapidity of deposition. The sands dropped at the mouth of the streams are expected to be buried rapidly by more sands in which there may be no bed load movement and consequently no stratifications are produced.

Four distinct subfacies are recognized within this facies (Table 2). They all have the same general characteristics, outlined above but with certain distinguishing features.

## 2.la(i) Subfacies $A_1$

This is characterised by having less shale between the sandstone beds in the middle and top of the cycle. Two cycles containing this subfacies were observed. One is partially exposed and cut by a channel (section 34, Figure 11), while the sandstone beds of the other are either

highly cross-bedded, apparently massive or micro-cross laminated (Section539,39%; Figure 12) and overlain conformably by marine fossiliferous sandstones and siltstones (Facies C).

In subfacies A<sub>1</sub>, the smaller proportion of shale between the sandstones of the cycle and the presence of a channel indicates that the environment of deposition is the axial or near axial part of a mouth bar. Elliott (1976) org noted that the variations between the sediment/(cycles in the Carboniferous Bideford Group of North Devon (England) are governed by their location with respect to the barfinger sand. He added that axial cycles include welldeveloped mouth bar sediments and/or distributary channel facies and consist solely of progradational facies.

A few sandstone units were found within the upper part of the shales or within the siltstone sediments of this subfacies and also within subfacies  $A_2$  which range in thickness from (8-50 cm). They are apparently massive and have sharp bases. Leeder (1974) interpreted thin, sharply-based, fine grained sandstones in the prodelta sediment of the Lower Border Group (Tournaisian), Northumberland Basin, as due to the sudden incursion of turbidity currents, although he added that storm action, major distributary flooding or delta front slumping might produce such units. Elliott (1975) reported sharp-based sandstone units at the base of progradational deltaic sequences in the Upper Limestone Group (Namurian,  $E_1$ ) northern Pennines. He considered them to be related to

the rapid flood rise with deposition during maximum and waning stage. The apparently massive sandstone units within the prodelta shale or delta front siltstones of this subfacies indicates sudden incursions of sediment. These sudden incursions were possibly produced during storm action or rapid stream flooding episodes.

2.la(ii) Subfacies A,

The sandstones of this subfacies are similar in colour, texture and thickness to the subfacies  $A_1$ , but differ in containing a higher proportion of shales, which range in thicknesses from 40 cm to 2.1 m between the sandstone units of the cycle, and in exhibiting less cross-bedding. Sometimes this subfacies is overlain by facies B and facies C. In Section 40, Figure 12, the sandstones of the cycle pass upward from apparently massive and sometimes cross-bedded in the middle of the cycle into symmetrical wave-rippled (Figure 17) sandstones at the top of the cycle which are in turn overlain by subfacies  $B_3$ .

Subfacies  $A_2$  is broadly similar to subfacies  $A_1$  and therefore formed by the progradation of delta lobes. The high proportion of shales between the sandstone units, the rarity of cross-bedding and the presence of wave ripple marks suggest that this subfacies was deposited in a lateral position on a mouth bar in which there was much less fluvial influence than in the case of subfacies  $A_1$ . Climbing ripples are present within some sandstone units indicating rapid deposition from suspension. The change

in the sedimentary structures (Section 40, Fig.12) within the sandstones of the cycle from apparently massive and trough cross-bedded sandstones at the middle of the sequence to symmetrical wave rippled at the top of the sequence might indicate that the marine process becomes more active towards the top of the cycle.

2.la(iii) Subfacies A<sub>3</sub>

No sandstone units are found within this subfacies, but the marine shales grade upwards into thin dark grey siltstones. These are mainly parallel laminated, although some micro-cross laminations were observed  $(\epsilon \cdot 9 \cdot 5ec_{1} + 2 \cdot 5e_{1} + 2 \cdot 5e_{1$ 

Subfacies  $A_3$  is similar to subfacies  $A_1$  and  $A_2$ , but the absence of sandstone units, the high proportion of shales, and the predominance of parallel laminations indicate the depositional environment to be a distal position, away from the axis, on a distributary mouth-bar with negligible fluviatile influence. In such a site of deposition the sediments are thought to be deposited from suspension as indicated by the predominance of parallel laminations. However, the micro-cross laminations indicate wave or current action, although the wave actions are more Elliott (1976) likely at such a site of deposition. interpreted similar sequences in carboniferous Bideford Group as lateral cycles deposited either, a considerable distance from bar finger axis, or, in a distal position on the mouth bar.

2.1a(iv) Subfacies  $A_4$ 

This subfacies is only exposed in one section (Section 19, Fig.10). It is similar to subfacies A<sub>2</sub>, except that it is only 14.3 metres thick, and shows a normal coarsening upward trend. No fossils were noticed in the basal shale of the cycle but Macnair and Conacher (1914) noted the presence of <u>Posidonomya becheri</u>, <u>Orthoceras</u> and <u>Nautilus</u>, a fauna which indicates a marine environment. The shales pass upward into parallel laminated siltstones in which plant fossils were observed and then sometimes into micro-cross laminated and then micro-cross laminated and apparently massive sandstones.

The discussion of subfacies  $A_1 - A_3$ , indicated that they were formed by the progradation of a constructive delta into a marine environment. Donaldson <u>et al</u>. (1970) described the Gaudalope delta, a constructive type, and reported small scale coarsening upward sequences, on average only about 3 metres thick, produced by progradation of mouth bars into a marine environment.

The coarsening upward trend, the presence of marine fauna at the basal shale and its similarity to subfacies  $A_2$  indicate that subfacies  $A_4$  formed by progradation of small scale mouth bars into a shallow marine environment. However, the water depth was significantly less than in the case of subfacies  $A_1$  to  $A_3$  because the shale at the base of this subfacies is much thinner.

2.1b Facies B

These facies comprise normal coarsening upwards sequences with a range of thickness from 2.5 to 15m i.e.

usually less than in facies A. They either overlie or are laterally equivalent to facies A sediments. Mostly the facies starts with non-fossiliferous highly fissile dark shales and passes gradually upwards into siltstones and sandstones. In some cases these are overlain by more interbedded shales and sandstones. The different examples of this facies are shown in Figure 18.

The siltstones are dark to medium grey and form beds 2-5 cm thick interbedded with the shales or silty claystones. Internally the siltstone beds consist of 0.1-2mm thick dark laminae (mainly heavy minerals, organic materials with little quartz) and 2mm-lcm light laminae (mainly quartz with little heavy minerals and organic materials). The internal sedimentary structures exhibited by these dark and light laminae are parallel, micro-cross and wavy laminations. Rootlets and thin coal seams and sometimes burrows are common (Fig. 19, 20).

The sandstones are light grey to yellowish-brown and range in thickness from 10-40 cm and shales are sometimes found interbedded with the sandstone units. Sedimentary structures are mainly micro-cross lamination; largescale cross-bedding is occasionally present and apparently massive sandstone units are common.

Coleman and Gagliano (1964); Allen (1965) and Kanes (1970) reported that the deltaic plain environment of the Mississippi, Niger and Colorado deltas are areas of shallow water and extensive vegetation. Coleman <u>et al</u>. (1964, p.252) stated that the interdistributary bays of the Mississippi delta are "... areas of shallow, open water which may be



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completely surrounded by marsh or levees, partially open to the sea, or connected to it by tidal channels". Gould (1970) pointed out that the salinity of the Mississippi deltaic plain waries seaward from the swamp environment, through fresh-water marsh, brackish water marsh and saline marsh.

The presence of rootlets and coal, the small thickness of the cycles and their relationship with the facies A, indicates that these facies are interdistributary bay deposits. Greensmith (1965) interpreted an assemblage of sediments consisting mainly of siltstones, sandstones and silty muds with coals in the Carboniferous (alcifereous Sandstone Series of the Midland Valley as on-delta sediments. Leeder (1972) concluded that alternating thin beds of mudstone, siltstone and sandstone with abundant rootlets and plant debris and without fauna represented the on-delta environments of levees and back-swamps in the Lower Border Group. Elliott (1974b) described the genesis of the interdistributary bay sediments and reported that sedimentary facies, the majority of which are coarsening upwards with coal and rootlets, indicate an inter-distributary bay environment.

Several subfacies each with the general characteristics discussed above are recognized within this facies as follows:

2.1b(i) Subfacies B<sub>1</sub>

In this facies the gradual coarsening upwards is interrupted by the presence of sharp, erosive-based sand-

stone units, which sometimes exhibit large scale crossbedding (Fig. 21).

The coarsening upward trend, and the interruption of the basal fine sediments of this subfacies by planar erosive sandstone beds, suggests that these sediments are laid down by crevasse splays. A crevasse splay is formed when the floodwater passes through a distinct breach in the levee of distributary channels into an interdistributary The sudden incursion of such sand-laden highbay. velocity currents into the bay produces sharp, erosivebased coarse beds which interrupted the deposition of the usual bay muds or silts. These muds and silts of this subfacies are thought to have been deposited partly by overbank flooding and partly from crevasse channels in which the fines were carried baywards beyond the coarse sediments of the crevasse splays. The muds and silts settle out from suspension and the segregation of heavy and light minerals produced parallel laminations of the siltstone units. However, some siltstone units show micro-cross laminations. Coleman et al. (1964) reported that wave ripples and sometimes current ripples are the main sedimentary structure of the bay sediments of the Mississippi delta. The cross-laminated siltstones probably indicate wave or current action. The crossbedding and micro-cross laminations of the sandstones indicate the migration of sub-aqueous sand dunes and ripples. However, some of the micro-cross laminated sandstones could reflect wave action. The apparently



because of the second property in the strength of the second property of the strength of the second property of the strength of the second property of the strength of the str

Fig. 21 Crevasse splay sediments subfacies Bl. Note the sharp erosional base of the sandstone cut through the fine sediments (Section 3, Fig.13). Post-Shield' Bed delta.

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massive sandstones might indicate homogeneity or rapidity of deposition.

Coleman and Gagliano (1964) pointed out that subdeltaic lobes formed by crevasses in the shallow bays between or adjacent to major distributaries, form the flesh on the skeleton framework of the major distributaries in the Mississippi delta. Arndorfer (1973) described the discharge pattern in two crevasses of the Mississippi delta and indicated that they built lenticular sedimentary deposits within the interdistributary bay. He added also that maximum discharge was attained in these crevasses during flood tides, when seaward flow is inhibited in the main delta distributaries. Elliott (1974b) in his description of the genesis of interdistributary bay sediments, pointed out that when the bay sediments and sometimes the levee sediments are interrupted by crevasse channel deposits a small scale coarsening upward cycle is produced in which the fine bay sediments are cut by the planar, erosive-based sandstones of the splay lobe. Collinson (1969) interpreted a sharp-based sandstone facies in the Millstone Grit (Namurian), England, as sudden incursions of relatively coarse sediments by crevasse splays into/shallow interdistributary bay environment.

Rootlets are only found in some of the sequences. Leeder (1974) pointed out that a rarity of plant material forming peat beds in this type of environment may possibly be due to rapid on-delta sedimentation and subsidence. Elliott (1974a, 1975) indicated that the absence of rootlet

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suggests a deep interdistributary environment, or central and/or distal site of deposition. The absence of rootlets in some sequences might therefore suggest that they were deposited in a deep central and/or distal bay area of sedimentation rather than a shallow part of the interdistributary bay.

In Silvermine Quarry (Section 48, Fig.14) a sequence considered to have been deposited by crevasse splays was traced laterally for about two miles to the south of the quarry where it is cut by a channel characterised by large scale cross-bedding and interpreted as a crevasse channel (Figs. 22 and 23).

## 2.lb(ii) Subfacies B<sub>2</sub>

This subfacies is similar to the subfacies  $B_1$ , but is characterised by a gradual coarsening-upward trend. Sedimentary structures within the sandstone units are mainly micro-cross lamination although apparently massive units are common. Rootlets are common, although not ubiquitious, either within the siltstones (in the middle of the sequence) or within the top sandstone units. Coal overlies some of the sequences.

Coleman <u>et al</u>. (1964) reported distributary mouth bars formed at the mouth of the crevasse channels in the interdistributary bay of the Mississippi delta. They added that, since the distributaries in the crevasse systems are numerous and often lie close together, the deposits of one bar might merge laterally with the bar deposits of the adjacent channel. They added that micro-cross



Fig. 22 Initiation of channel in the crevasse splay sediments (subfacies Bl) compared with Elliott 1973; figure 28. Silvermine quarry (Section 48, Fig.14), post-Blackhall delta.



Fig. 23 Sketch of channel similar to the Silvermine channel (fig. 22), from Elliott (1973, fig. 28).

laminations of wave and current type are the predominant sedimentary structures and sometimes parallel laminations are present. Coleman and Gagliano (1964), pointed out also that subdeltaic lobes developed in the bays of the Mississippi delta due to advance of closely spaced pairs of crevasse channels. Elliott (1974b) described the genesis of interdistributary bay sediments and indicated that the progradation of permanent crevasse channels could produce minor mouth bars. He added that mouth bar sediments are sites of wave and current processes.

The nature of the vertical sequence and the association of the sediments indicates that these sediments are similar to facies association  $A_1$  but differ in the lack of marine fauna at the base of these units. Therefore the progradation of small mouth bars into an interdistributary bay isolated from the sea or in the landward part of an open bay is most likely to be the origin for this subfacies. If a small scale mouth bar is accepted as the origin of these sediments, their deposition could result from the gradual incursion of crevasse splay(s) into an interdistributary bay similar to the processes explained earlier for the Mississippi delta. The lack of rootlets in some sections might indicate that the bay was too deep for the growth of vegetation, i.e. this subfacies was deposited in the distal and/or central part of the bay.

2.1b(iii) Subfacies B<sub>3</sub>

This subfacies ranges from 10 to 13 m thick. It differs from the above two subfacies in that shales

separate the beds of siltstones and sandstones. The colour of the coarse sediments is grey. Normally, the sandstones have erosional bases which cut through the underlying fine sediments and in some cases the bed thickness increases upwards, whereas sometimes it decreases upwards. In section 40, Figure 12 (Calderwood Glen), the sequence contains horizontal sandstone tubes interpreted as burrows (Fig. 20). Sometimes rootlet-bearing sediments cap these sub-facies. In Blackburn (Section 45, Figure 14) a sequence grades from dark shale to siltstone and very fine sandstone returning back gradually into siltstones and shales (Fig. 24). No rootlets were observed within this section. Parallel, micro-cross and sometimes wavy laminations are the main sedimentary structures observed within the siltstone and sandstone units.

Fisk (1961) indicated that the bar-finger sand (part of prograding elongate delta) of the Mississippi delta is overlain by silty sand and clayey silt levee deposits which grades laterally into the fine grained bay sediments. Coleman <u>et al.</u> (1964) pointed out that the natural levee part of the interdistributary bay sediments of the Mississippi delta is of fine grained sediments deposited from suspension by flood waters. They added that parallel laminations, micro-cross laminations of current type, wavy laminations and climbing ripples are the main sedimentary structures of the levee sediments. Allen (1965) reported that the sediments of the levee which border the stream channels and descend away into the fresh



water back-swamp of the Niger delta, were formed by overbank flooding. He added that the typical sediments of the levee are alternating very fine sands and silts with even and small-scale cross stratification, rootlets and animal burrows. He indicated also that the typical sediments of the fresh-water back swamp are mainly of alternating silty clays or clayey silts with laminae of coarse to very coarse silt. Scott and Fisher (1969) reported that the sediments of the Mississippi delta levees are thickest and coarsest when adjacent to the channel and they grade laterally into interdistributary muds and organic sediments of the marshes. Collinson (1969) noted the presence of burrowing within the interdistributary bay sediments of the Millstone Grit (Namurian), England.

The coarsening upward trends, the presence of large amounts of shale separating the coarse sediments and the presence of rootlets and burrows, probably indicates that these sediments were formed by overbank flooding. This process involves sediment-laden flood water flowing over the banks of the main stream channel as sheet floods without forming /breach or crevasse. The fine sediments mainly settle out from suspension as indicated by the parallel laminations of the siltstones. However, the micro-cross laminations of some siltstones and the sandstone units might indicate current action with the formation of ripples. The sequence at Blackburn (Fig.14, Section 45) is coarsening upward from shales into siltstones and then into thin sandstone units (1-7 cm thick), returning

back gradually into thinner sandstones, siltstones and shales. The sandstone units are probably deposited during the major flood in which the coarse sediments could reach the distal part of the bay and the fine sediments (siltstones and shales) are deposited during minor floods. However, the retreat of the levee, probably during the processes of distributary abandonment or due to a decrease in the magnitude of the flooding, could produce such sequences. At Sauchie Craig South (Section 26, Figure 10) the coarsening-upward trend of the cycle is characterised by an increase in the sandstone thicknesses upward. This is probably due to the encroachment of the levee into interdistributary bay. Elliott (1974b) described the genesis of interdistributary bay sequences and pointed out that the encroachment of levees into the bay produced coarsening upward sequences characterised by interbedded coarse and fine sediments in which the thicknesses of the coarse beds increased upwards.

The discussion outlined above indicates that subfacies  $B_1$ ,  $B_2$  and  $B_3$  represent interdistributary bay sediments. They deposited by flood-generating processes, either by overbank flooding (subfacies  $B_3$ ) or by crevasse channels (subfacies  $B_1$  and  $B_2$ ). No fauna were observed in the basal fine sediments of these subfacies. This probably suggested that the bay in which these sediments were deposited was of fresh water. In other words, they are either deposited in the upper part of open bays (i.e. the inland part) or in closed bays.

2.1b(iv) Subfacies  $B_A$ 

(Section 19, Fig. 9) One example is represented by this subfacies (and has a normal coarsening-upward trend of 8.7 metres thick, starting from shales with clay band ironstones. Ostracods are the main fauna within the ironstone bands and are similar to the fauna of the base of the Blackhall limestone which indicates hypersaline conditions (see Chapter 3). Clough et al. (1925) reported fish remains and entomostraca (ostracods) within the ironstone bands.

Coleman <u>et al</u>. (1964), indicated the existence of fresh to brackish water conditions in most of the interdistributary bays of the Mississippi delta. Gould (1970) reported also a variation in salinity in the interdistributary bay of the Mississippi delta in which the interdistributary becomes more saline seaward. Oomkens (1970) reported small scale fluviolacustrine (non-marine) regressive sequences, in which the brackish water coastal plain basin is infilled by fluviatile sediments producing a coarsening-upward trend in Rhone delta.

The coarsening upward trend and its similarity to the subfacies B<sub>2</sub>, indicates that this subfacies represented minor mouth bar and therefore was formed by slow incursion of sediments from crevasse splay(s) into an interdistributary bay environment. The presence of hypersaline fauna indicated that the bay was of abnormal salinity (lacustrine or lagoon). The absence of rootlets within this subfacies might indicate that the site of deposition was central and/or distal position within the bay. Leeder

(1974) interpreted similar coarsening upward facies as deposited in brackish interdistributary bays in the Lower Border Group (Tournaisian), Northumberland Basin.

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## 2.lb(v) Subfacies B<sub>5</sub>

This subfacies has a coarsening-upward trend with a range of thickness from 6 m to 10 m. Normally the sediments pass upwards from marine fossiliferous shales into non-marine shales, into parallel laminated and sometimes micro-cross laminated siltstones with rootlets and then into micro-cross laminated or apparently massive Symmetrical wave ripples of average wave sandstones. length 7.5cm and amplitude 0.8 cm were observed (Fig. 25, p.49). In the Stirling area, Bannock Burn (Section 25, Fig. 10), when this subfacies overlies the Hurlet Limestone crinoids and bivalves were noted in the basal shale of the cycle. Dinham and Haldane (1932) and Francis/ (1970) indicated that the shale close to the Hurlet Limestone in Bannock Burn carried an abundant marine fauna, including crinoids, bryozoa, productoids, brachiopods, gastropods and bivalves. In Old Sauchie Burn (Section 27, Fig.10) where this subfacies overlies the Shields Bed, the basal shale units have Lingula and other unidentified skeletal fragments. Dinham and Haldane (1932) also reported a marine fauna in the shale at the base of the cycle in this section. In the Campsie area, Corrie Burn section (Section 16, Fig. 9), this subfacies overlies the Hurlet Limestone and is about 3m thick (Fig. 26). Although the exposure is



Fig. 26 Levee sediments prograded into open interdistributary bay environment (subfacies B5). Section 16, Fig. 9 (scale is arrowed). Post Hurlet delta.



Fig. 27 Levee sediments (subfacies B5) section 19, Fig. 9. Post-Main or Mid Hosi@delta.

poor, the Hurlet Limestone passes upward into calcareous shales, then into siltstones with rootlets and finally into sandstones. At Shield' Burn (Section 19, Fig. 9) this subfacies overlies the Main or Mid Hosit Limestone and is represented by a coarsening-upward sequence about 9.5 m thick, in which shales forming the upper part of the cycle are overlain by fossiliferous sandstones (facies C). Although the section is now poorly exposed (Fig. 27) Clough et al.(1925) noted that the basal shales contain ironstone bands with a marine fauna, passing upward into shales with thin seam of coal.

It is indicated previously in the discussion outlined under facies B and also under subfacies  $B_1-B_4$ , that the interdistributary bay is an area of shallow water and of extensive vegetation which might be open and therefore varied in salinity and may be completely surrounded by marshes or levees (cf. Coleman and Gagliano, 1964, 1965; Kanes, 1970; Gould 1970).

The existence of rootlets within the siltstone units and the presence of coal indicates that this subfacies was deposited in interdistributary bays. However, the presence of marine fauna in the basal shales indicates that these bays were open. If this is accepted, the cycles of sections 25 and 27 (Fig.10) were produced by the slow incursion of sediments from crevasse splay(s) in which minor mouth bars were formed in an open interdistributary bay, similar to the processes envisaged for subfacies  $B_2$ . In such a situation, coarsening-upward

sequences would be produced with a marine fauna in the basal shales of the cycle. Rootlets are most common within the siltstones in the middle of the cycle. This is probably due to the infilling and progressive shallowing of the interdistributary bay. The absence of rootlets within the upper-most sandstone units of the cycle and the presence of symmetrical wave ripple marks indicated that wave action become more active towards the top of the cycle. The sequence at Shields Burn (Section 19, Figure 9) which contains coal in the basal mud and passes upward into sandstone and siltstone units and then into shales probably formed by the encroachment of marsh and levee sediments into an open interdistributary bay as indicated The encroachment of levees into an open in Figure 28. interdistributary bay is also suggested for the sequence at Corrie Burn (Section 16, Fig. 9).

Elliott (1975) interpreted small scale coarsening upward sequence with marine fossils at the base and rootlet at the top as the progradational mouth bars of crevasse splays which built into open interdistributary bays in the Upper Limestone Group (Namurian  $E_1$ ) Northern Pennines.

## 2.1c Facies C

Two types of sediments are included in this facies; thin marine siltstones or fine sandstones and coal. The sandstones are grey to yellowish-brown units ranging in thickness from 10 cm to 80 cm which are found overlying subfacies  $A_1$  to  $A_3$  and sometimes subfacies  $B_5$ . When the



sandstones form thick units they consist of thin beds of 2-10 cm thick, interbedded with thin silty claystones.

De Raaf et al. (1965) in their study of the Lower Westphalian sediments of North Devon, England, indicated that during delta progradation, a thick sequence of clastic sediments was deposited representing the constructive facies, whereas thin bioturbated siltstone units represent the destructive facies which formed after delta abandonment. Scott and Fisher (1969) noted that in the Mississippi delta, the upper part of the distributary mouth bar (constructive facies) contains a concentration of shells and burrows formed during delta abandonment. Elliott (1974a) reported thin fossiliferous sandstone units and coal, which overline the constructive distributary mouth bars or interdistributary bay sediments in the Upper Limestone Group, Namurian, E1, North Pennines and considered them as an "abandonment" facies. This term is adopted in this study to represent these marine and coal units.

The marine horizons are considered in this study to represent a limited reworking of the upper part of the mouth bar sediments by marine processes during the abandonment of delta. In other words, they reflect reduction in sediment supply; periods of non-deposition and biogenic reworking.

Within the interdistributary facies (Facies B), thin seams (8-15 cm) of coal were found to overlie or underlie some sequences, sometimes associated with rootlets. Allen
(1965), Kanes (1970) and Gould (1970), reported the development of marshes within the deltaic plains of the recent Niger, Colorado and Mississippi deltas. Donaldson <u>et al.</u> (1970) pointed out that within the deltaic plain of Guadalupe delta, the abundance of thin beds or laminae of peat distinguished the marsh environment from other environments in which silty clays are deposited. The existence of coal within some cycles of facies B is considered to represent an extensive development of marshlands in an area of humid climate. The absence of coal or rootlets in some sequences may indicate deposition in a distal and/or central part of the bay.

## 2.1d Facies D

This comprises sandstones over 1 m thick, normally with erosional bases (Fig. 29). They are well to moderately sorted and of medium grain size. The common sedimentary structures are trough and planar cross bedding (Figs. 30, 31, 32).

The erosional bases of these sandstones correspond to the scoured surfaces of Allen (1964) produced by erosion in the scour pools at the bottom of fluvial channel. The large scale cross-bedding results from the migration of subaqueous dune bed forms (Harms & Fahnestock, 1965; Allen, 1970).

Most of the sections in the area studied indicate the non finfing-upward nature of the channels (Fig. 30) and also the lack of any upward change in sedimentary structures. These features exclude high-sinuousity rivers as a possible origin.





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Fig.31 Large scale trough cross-bedding in the distributary channel of post-Shields Bed delta (Section 12)

Fig. 32 The right side of Fig. 31

Fig.33 Siltstone units as indicated within the crossbedded units in the channel at Liverholm. Post-Blackhall delta (Section 34).

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Moody-Stuart (1966) noted that the absence of levee sediments above channel sediments indicates a low sinuosity origin. The lack of repeated units of pebbles, with large-scale tabular cross-bedding indicates that braided, low-sinuosity channels are unlikely analogous (Allen, 1970; Deegan;1970; Leeder, 1973). Therefore, non-braided, low-sinuosity channels are the most likely origin of this facies.

Siltstone units, ranging in thickness from 8 cm to 30 cm, showing parallel or sometimes faint micro-cross laminations and occasionally interbedded with claystones, are either interbedded with or drape some of the crossbedded units (Figs. 33, 34).

At Liverholm (Section 43, Figure 11) a 30 cm thick siltstone, which is probably discontinuous interrupts a channel. This was probably formed by deposition from suspension in slowly moving water during low river stage on an irregular surface formed by earlier, high stage strong currents, as explained for the River Rio Grande by Harms and Fahnestock (1965). The large thickness of silt, 30 cm (Figures 33, 34) probably indicates the scale of the irregular depressions, which may have been the troughs of large bed forms as explained by Coleman (1969).

Some of the channel sandstones contain sets of tabular cross-beds bounded by apparently massive sandstone units or horizontal beds. Harms and Fahenstock (1965) noted that River Rio Grande contains marginal bars and at low stage the bars are exposed and the talweg flow modifies



their margins. These processes produce tabular sets by bar-front avalanching (see also Leeder, 1973). Alternatively the tabular sets could have resulted from the migration of straight-crested dunes (Allen, 1964; McCabe, 1977).

The channels either cut through the deltaic bar finger sands, subfacies A1 (Figures 35 and 36) or interdistributary bay sediments, facies B (e.g. Figure 12, Section 40, Fig. 10, Section 29). In the Stirling area an 8 m thick channel of sandstone (Section 29, Fig. 10) is considered to have been formed by a major avulsion switching into an interdistributary bay. The sediments of this channel were deposited under the upper flow regime as indicated by massive horizontal bedded sandstones (Fig. 37; compare also with Collinson, 1969, Fig. 12), which grade upwards from the upper part of the lower flow regime as indicated by the cross-bedding at the base of the sequence. The channel at Skoli Burn (Section 46, Fig.14) is unusual in that it has a finging-upward trend, and passes upward from trough cross-bedded sandstones at the base (Fig. 38) into thin units of parallel and sometimes micro-cross laminated siltstones interbedded with shales. This probably represents deposition in a meandering channel.





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# 2.2 DEFORMATIONAL STRUCTURES

These structures were observed mostly in the clean, moderately to well sorted sandstones of facies D and subfacies  $B_1$ . They were also observed within the siltstone laminae of facies A. They include folded laminae considered to be convolute laminations (Figures 39, 40), sandstone collapse structures (Fig. 41) and the disturbance of sandstone units which was probably due to upward movement of mud lumps in the distributary mouth bar (see section 2.3c). The sandstones in which these deformation structures are observed are overlain and underlain by undisturbed sandstone units.

Greensmith (1965) reported deformed laminae in the Calciferous Sandstone Series and considered them to result from earth tremors which repacked the sandstones causing a migrating of pore-water upward with the result that the upper layer became more mobile and susceptible to gravitational slip. Lowe (1975) pointed out that most convolute laminations are formed in coarse, uncohesive and moderately to high permeable beds and develop when the free upward flow of escaping pore fluids is inhibited by cohesive and low permeable units within the accumulated He added that during the waning stage of sediments. current activity the cohesive and less permeable sediments accumulate both because the sediments are finer grained and better packed as a result of the reducing rate of sedimentation. He stated (p.188) also "As pore pressures increase accompanying progressive liquefaction





in the lower part of a sedimentation unit, anticlines might be expected to evolve near the sediment surface by deformation within and above the crests of ripples located immediately beneath accumulating low-permeability layers." Anderton (1976) interpreted convolute foresets in the Jura quartzite as liquefaction structures produced by the upwards migration of pore water causing the arching of stratification around the axis of maximum flow.

The observed deformed laminations are considered to be syn- or post-depositional, not tectonic, and result from a triggering mechanism, such as  $\chi$  earthquake, causing liquefaction. The upward movement of pore water probably caused an arching of laminae around the axis of maximum flow and convolute laminations are produced.

A similar process is thought to be responsible for the collapsed sandstone beds. The upward movement of pore water probably caused a fluidization and a loss of strength in the overlying sediment at the points of maximum flow. However, the sediments in the area between the have been the the sediments of maximum flow need not  $\bigwedge$  fluidized and may/retained their cohesion. The blocks of cohesive sand may then have collapse downwards into the underlying liquefied bed (Fig. 41).

# 2.3 DELTA TYPES OF THE LOWER LIMESTONE GROUP

The Lower Limestone Group consists of seven carbonate units interbedded either with shales, siltstones, sandstones and occasionally thin seams of coal or shales (Fig. 42). Read (1959) pointed out that the Lower Limestone Group consists of cycles in which each cycle passes upward from limestone into shales, siltstones, sandstones and coal. The investigation of these cycles in this study reveals that the non-carbonate sediments of each cycle coarsen upwards and, therefore, delta progradation is suggested as the cause of the cyclicity. Six delta advances are recorded in the area studied. Their recognition is based on facies associations, vertical profiles and on lateral facies variations within the sediments of each delta. Each delta advance was followed by the deposition of a carbonate member formed while the delta lobe was abandoned.

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### 2.3a Post-Hurlet delta

The distribution of the sediments of this delta is presented in Figure 9-14 and Figure 43. In the Stirling area only one partial section (Section 25, Fig. 10) is exposed and is represented by subfacies  $B_5$ , interpreted as the deposit of an open interdistributary bay environment. In the Bathgate area also only one partial section is exposed (Section 45, Fig. 14) and no fossils were observed, either by the present author or by previous workers (e.g. Geike, 1879) in its basal shales. The sediments of this





section are considered to represent subfacies  $B_3$  which indicates levee encroachment into an interdistributary fresh water bay. In the Campsie area, three partial sections are exposed (Sections 16, 20, 22, Fig. 9), one of which is represented by subfacies  $B_5$ , interpreted as due to the encroachment of a levee into an open interdistributary bay environment. The other sections are represented by shales only. The sections exposed in the south of the studied area (Sections 1, 2, 3, 6, 7, Fig. 13, the Section 41, Fig. 12) are considered to represent/post-Shields delta, even though they stratigraphically overlie the Hurlet Limestone as will be explained later.

In conclusion all the studied sediments of the post-Hurlet delta indicate interdistributary bay or shelf prodelta conditions of sedimentation. Although there are no sections representing the progradational (i.e. mouth bar) facies of post-Hurlet delta, from the distribution of coarse sediments the boundary of/delta front can be drawn as indicated in Figure 44. This shows that the dclta front of post-Hurlet delta lies to the north of Milton of Campsie and slightly to the south of the Bathgate The thicknesses between the Hurlet Limestone and area. the Shield Sed (the second carbonate unit in the Lower Limestone Group) is not more than 12 metres. From this it is estimated that the post Hurlet delta was a small scale delta which prograded into a shallow marine environment (slightly more than 12 metres).



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### 2.3b Post-Shields Bed Delta

The Shield Bed represents the second transgression in the Lower Limestone Group. It was only deposited in the Stirling, Bathgate, Campsie and the south west part of the Carluke areas and was not deposited south east of Carluke or in part of the Lanark district (Fig. 45). The detailed study of this carbonate unit will be discussed in the carbonate section.

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Hinxman <u>et al</u>. (1921) noted that in the Birkwood section (Section 1, Fig. 13) and elsewhere in the district west of the Clyde there is no bed equivalent to the Shields Bed (p.45). They added that here the Hurlet Limestone is succeeded by shales and sandstones which are overlain directly by the Blackhall Limestone. They quoted Macnair who thought that, in the Birkwood section, the Shields Bed is probably represented by the highest bed of the Hurlet Limestone.

It is inferred from the distribution of sediments that the post-Hurlet delta, either prograded as one or many lobes. The delta-front and the shelf prodelta sediments of the post-Hurlet delta are located to the north of Sections 1, 2, 3, 5, 7 and 41 (Figure 46). When the Shields Bed transgression started the Shields Bed was deposited directly on the Hurlet Limestone in the area of sections 1, 2, 3, 5, 7, 41 and further south (Figure 47). Here it is suggested that the Hurlet and Shields Bed Limestones form one unit with the Shields Bed forming the upper leaf (Figure 47). This is consistent with the



Diagram showing the lateral variations of the post-Hurlet and post-Shields Bed deltas in the studied area. Notice the Shields Bed and Hurlet Limestone formed one unit at the SW of the studied area.

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fact that, although the Hurlet Limestone (in fact the Hurlet and Shields Bed) is only partially exposed in the area of Sections 1, 2, 3, 5, 7, 41, it is thicker than to the north in the Campsie area (Sections 16, 20, 21 and 22, Figure 9). Consequently the sediments which stratigraphically overlie the Hurlet Limestone in these southern sections (Sections 1, 2, 3, 5, 7 and 41) are thought to represent the post-Shields Bed sediments. Therefore the sediments between the Hurlet and Blackhall Limestone in these sections are considered to represent the post-Shields Bed delta in this study.

The distribution of the post-Shields sediments in the studied area is summarized as follows: In the Stirling area, Old Sauchie Burn (Section 27, Fig.10), two coarsening upward sequences are exposed which represent subfacies  $B_5$  and  $B_2$ , considered to indicate marine and fresh water conditions respectively in an interdistributary bay environment. The other sediments in this area are represented by subfacies  $B_2$  and  $B_3$ , interpreted as fresh water interdistributary bay sediments.

In the Campsie area, Shields Burn (Section 19, Figure 9), the post Shields sediments are represented by two small-scale coarsening upward sequences of subfacies  $A_4$  and  $B_4$ . Subfacies  $A_4$ , a progradational small scale mouth bar, forms the first sequence and is overlain by the hypersaline water interdistributary by sediments of subfacies  $B_4$ . At Corrie Burn (Section 16, Figure 9), a partial section is exposed. The sandstones here are

moderately sorted and have symmetrical wave ripples which indicate a NW-SE orientation of the coastline in the middle of the studied area. The other post-Shield's Bed sediments in the Campsie area are mainly shales. In the Renfrew district in the south of the studied area (Figure 12), the post-Shield's Bed sediments are all shales.

In the SE of the Lanark district (Fig. 13), the smallscale coarsening-upward sequences (Sections 1, 2, 3, 5 and 7) which stratigraphically overlie the Hurlet Limestone are considered to represent post-Shield Bed sediments as explained earlier. No fossils were observed here or mentioned by the previous workers (e.g. Hinxman and MacGregor, 1921). These sediments represented subfacies  $B_1$ ,  $B_2$ , and  $B_3$  which are interpreted as fresh water interdistributary bay deposits. In Section 9 and 12 (Fig.13) where the Shield Bed is separated from the Hurlet Limestone, two partial sections are exposed, one of which is interpreted as a channel (facies D). The distribution of post-Shield sediments is presented in Figures 9-14 and also in Figures 48 and 49.

The previous description reveals that facies B and shales are the main sediments of the post-Shieldsdelta. Only one section is represented by mouth bar sediments, subfacies  $A_4$  (Section 19, Fig. 9). Therefore it is difficult to speculate about the type of delta. However, the thicknesses of this section (14.3 M) indicates that the post-Shields Bed delta prograded into a shallow marine environment; slightly more than 15 m deep.



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At Old Sauchi Burn (Section 27, Figure 10), the two small coarsening upward sequences probably represent the repeated infilling of an interdistributary bay. This interdistributary bay possibly changed in salinity from marine, indicated by the basal subfacies B5, to fresh water represented by subfacies B<sub>2</sub>. At Shields Burn (Section 19, Fig. 10), the two exposed coarsening-upward sequences represented by subfacies  $A_4$  and  $B_4$  are interpreted as formed by the progradation of a mouth bar into shallow marine environment and a hypersaline water interdistributary bay respectively, subfacies  $B_4$ , representing aggradational sediments, either formed at the time of formation of subfacies  $A_4$  or it developed after the progradation of subfacies  $A_4$  when the deltaic plain transgressed over the progradational sediments of the advancing delta (Fig. 50).

#### 2.3c Post-Blackhall Delta

Three types of vertical sequence were recognized in the field within this delta progradation. In two types a fossiliferous marine horizon, the Neilson shell bed (Wilson, 1966), overlies the Blackhall Limestone.

In type 1 which is the thickest, subfacies  $A_1$ ,  $A_2$ and  $A_3$  form the main sediments. Normally these sediments pass upwards from fossiliferous shales into laminated nonfossiliferous shales, siltstones with parallel and microcross laminations, then into microcross laminated, crossbedded, apparently massive and sometimes wave rippled sandstones. Sometimes type 1 sediments are overlain by



type 2 sediments. The type 2 is represented by nonfossiliferous shale which grades into siltstones with rootlets and then into sandstones. Coal is occasionally present at the base. Subfacies  $B_1$  and  $B_2$  form the main sediments of this type. The type 3 is represented by shale only which is fossiliferous at the base.

Fisher (1968) indicated that in a constructive delta where the sediment input exceeds the marine reservoir energy the ratio of progradational to destructional facies is high. Fisher (1969a) noted that in wave and tide-dominated delta, the abandonment facies is not clearly distinguishable from progradational facies. In the post-Blackhall delta, the abandonment facies is easily distinguished from the mouth bar facies (facies  $A_1-A_3$ ). This suggests that the delta was a constructive type.

In an elongate delta with a bar-finger sand, the facies do not persist laterally in comparison with those in lobate deltas (Fisher, 1969b). The thickness of sediments is also variable across a bar-finger sand; it thickens directly beneath the distributary channel and decreases towards its lateral margin (Fisk et al. 1954; Fisk, 1961).

In type 1 sequences of the post-Blackhall delta (facies  $A_1$  to  $A_3$ ) the shale is thick compared with the coarse sediments, and three sections selected perpendicular to the *Palaeocurrent direction* (Calderwood Glen, Section 40; Kitockwater, Section 42; Gill Burn, Section 44, Figures 51, 52) show that there is no lateral persistance of the facies.





In the quarry near Sloughneagh Burn, there is a local disturbance in the strata (Figure 53). The amount of dip in these strata decreases upwards, and the dip directions are opposite to the palaeocurrent direction. It is suggested that this structure was either produced by the upward movement of a mud lump (which would be located to the west of the quarry) or by syn-depositional faulting (in which case the fault would be located to the east of the quarry).

Type 2 sediments are represented by subfacies  $B_1$  and  $B_2$  which are interpreted as fresh water interdistributary bay facies as indicated by the presence of rootlets, coal and the absence of marine fauna. Type 3 sediments represent a shelf-prodelta association (cf. Fisk <u>et al</u>. 1954; Frazier, 1967).

All of these features indicated that the post-Blackhall delta is a highly constructive, fluvially-influenced, elongate type in which the sediment input was high and exceeded the energy of the coastal processes. These sediments are characterised by thick sequences with little shale within the coarse units near the channels (subfacies  $A_1$ ) and thin sequences away from the channel in which the coarse facies have high proportion of shale (subfacies  $A_2$ and  $A_3$ ). These sequences, represent the progradational phase of a mouth-bar, in which the siltstones and sandstones of the mouth-bar represented a delta front sediments and the shales represented a prodelta sediments deposited

Fig. 53a Deformation of the sandstones by mud lump movement or by syn -depositional faulting. Post-Blackhall delta. Quarry 100m west of section 20.



Fig. 53b Sketch of the deformation outlined above. Figures give direction and angle of dip of strata corrected for tectonic tilt.

seaward below the wave action. The depth of the basin in which the post-Blackhall delta was prograded is slightly more than 60 metres. This is estimated from the thickest section in the area studied, 56.7 metres. Type 1 sediments pass laterally Perpendicular to the Palacocurrent direction into an interdistributary bay sequence (Type 2 sediments) in the upstream part of the delta lobes and into shelf-prodelta (Type 3) sediments offshore from the lobes. (Figs.54 & 55).Similar examples, in which the progradational facies passes inland into interdistributary bay facies and seaward into shelf-prodelta facies, are given by Allen (1965) for the Niger delta, Fisk (1961) and Frazer (1967) for the Mississippi delta and by Elliott (1975) from Upper Limestone Group (Namurian E<sub>1</sub>), northern Pennines, England.

#### 2.3d Post-Hosid deltas

These carbonate units are found at the top of the Lower Limestone Group. These are named in this study, as discussed earlier, as the Main Hosie, Mid Hosie, Second Hosie and Top Hosie Limestones. Coarse sediments with a coarsening-upward trend follow each limestone in some sections and are considered to be the interdistributary bay deposits of small-scale deltas. The distribution of sediments within the Hosie units are discussed as follows.

#### 1. Post Main Hosie Delta

The distribution of the sediments within this delta is presented in Figures 56 and 57. In the Stirling area,



. 54 Diagram showing the lateral variations of the post-Blackhall sediments in the studied area. Paleocurrent directions deduced from (R) ripple marks and (C) cross bedding. and a second second

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River Carron section (Section 29, Fig. 10), a small scale coarsening-upward cycle (3-9 metres) is exposed. The cycle represents subfacies  $B_2$  which are interpreted as interdistributary bay sediments (2.1b(ii)). In the Campsie area, Corrie Burn Section (Section 16, Fig. 9), the exposure of the post-Main Hosie sediments is poor and only sandstone beds ranging from 10-30 cm thick forming sequences 1-2 metres thick are exposed. It is difficult to speculate about their environment. In Shields Burn (Section 19, Fig. 9) the Mid-Hosie Limestone, which is stratigraphically above the Main Hosie, is not indicated on the geological map (Sheet XXVIII, Stirlingshire). Clough et al. (1925) reported the existence of the Mid-Hosie Limestone in the quarry about 0.5 mile east of Shields Burn (Section 19, Fig. 9). The section at Shields Burn is now poorly exposed and the thicknesses between the Main Hosie Limestone and the Second Hosic Limestone is about 9.5 m, much less than that exposed at Corrie Burn (Section 16, Therefore the Mid-Fig. 9) to the west of Shields Burn. Hosie Limestone has either coalesced with the Main Hosie Limestone, similar to the situation explained for Shields Bed (2.3b) or it is separated from the Main Hosie Limestone by a thin shale unit. In either case the model presented in Figures 58 and 59 explains the two Therefore, the sediments in this section possibilities. (Section 19, Fig. 9) which are represented by subfacies B<sub>5</sub> and indicate an open interdistributary bay environment, either represent the post-Main Hosic or post-Mid Hosic









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deltas. In the Calderwood Section (Section 40, Fig.12) the Main Hosie Limestone and Mid Hosie Limestone are separated by 1.1 metres of shale. At Kittoch Water, the Main and Mid Hosie Limestones have coalesced into one unit. The model presented in Figure 59 could be applied in this case also. The other cycles of post Main Hosie delta are represented by shales only (Figs. 56, 57 and also Figs.9-14).

# 2. Post-Mid Hosie delta

In the Stirling area, River Carron Section (Section 29, Fig.10), the Mid-Hosit Limestone was not deposited. Dinham and Handle (1932), noted a thin (7.5 cm thick) fossiliferous marine shale about 4 metres above the Main Hosit Limestone. This fossiliferous marine shale is not exposed now, but its stratigraphic position is taken to represent the Mid-Hosit Limestone. The sediments of the post-Mid Hosit Limestone are represented by subfacies  $B_1$  and D which are interpreted as the deposits of interdistributary bay and channel environments. Facies B and D and shales formed the main sediments of this delta in the other sections of the studied area. (Figs.9-14; Figs. 60 and 61).

## 3. Post-Second Hosie delta

Two small scale coarsening upward sequences are exposed at the SE of the studied area (Sections 1 and 11, Fig.13). The sediments of these cycles represented subfacies  $B_1$  and  $B_3$  which are interpreted as fresh water



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19 10 10 10 102 Fig. 61 Diagram showing the boundary of the delta front sediments of the post-Mid Hosi<sup>e</sup> delta Loch 6 Z • 5 10 Km Post Mid Hosie delta • 0m N Section 40 200 1 12-Delta front sediments 10. 1 Section 29. 2 N \* Section 3 Mid Hosie (MDH) MDH Pro delta sediments ..... 200 

interdistributary bay sediments. The other sediments are represented by shales only. The distribution of post Second-Hosie deltaic sediments are presented in Figures 9-14 and Figures 62 and 63.

The above discussion reveals that the sediments of the post Hosiz deltas are represented mainly by facies B and D and shales which indicates interdistributary bay, channel and shelf prodelta environments. The thicknesses of post Hosiz sediments ranges from 50 cm to 12.3 metres. This probably indicates that the post Hosiz deltas prograded into shallow basin(s) of maximum depth slightly more than 12.3 metres.





# 2.4 PALEOGEOGR APHY

Four factors are considered to control the shape and growth of the delta platform in constructive deltas as explained by Fisk et al.(1954). These are:

1, River load

2. Number of main distributaries

3. The depth of water into which the delta front advances4. The amount of subsidence.

In destructive deltas, marine processes such as waves and tides and the sediment input are considered the main factors (Allen, 1965; Scott and Fisher, 1969). However Coleman and Wright (1975, p.109) stated that"whether the sand bodies reflect fluvial or wave dominance depends largely on the ability of the river to supply sediments relative to the ability of the waves to rework and redistribute them."

The depth of the water and river sediment load are considered the most important factors for the Lower Limestone Group deltas. The deltas investigated changed from small deltas (Post-Hurlet and Post-Shields deltas) to a thick elongate type (Post-Blackhall delta) returning back to small deltas (Post-Hosir deltas). The Post-Hurlet, Post-Shields and the Post-Hosir sediments were probably built up by relatively small streams which debouched into a shallow basin(s). It is difficult to speculate about the type of these deltas because mouth bar sections are rare. However, one section representing the distributary

mouth bar of the Post-Shields delta, is characterised by normal coarsening-upward and shows no evidence for wave or tidal reworking such as the presence of tidal channels or shell beds, within the sandstone part of the sequence. This might suggest that these deltas were probably represented by constructive types.

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The post-Blackhall delta is considered to have been formed by a few, large streams which debouched into a deep basin. The sediment input was great relative to the energy of the marine processes, producing distinct and thin abandonment facies relative to thick constructional facies.

Apart from a few sections, no cross-bedded exposures were available to trace the paleocurrent in each delta advances in each section within the studied area. However, the available paleocurrent measurements for each delta are presented in the diagrams showing the distribution of the sediments. For all the deltaic advances the crossbedding measurements represented by the vector mean (Fig. 64) indicate NE derivations for the sediments, whereas, the ripple mark measurements indicate a NW-SE orientation of the coastline. The plotting of the distribution of the coarse sediments (delta front sediments) for each delta (Fig. 64), which probably reflects shoreline progradation, indicates the direction of the delta progradational. The areal extent of coarser delta plain and prodelta sediment is different in each delta advance. Therefore the shape of the shoreline changed from one delta to the other.



Greensmith (1968, p.5) noted that the Central Basin in which the studied area is located is in a NE-SW zone of "... relatively fast downwarp controlled by pre-existing Caledonian structures" during the deposition of the Oil Shale Group (Carboniferous). He added that the local paleogeography and tectonism within this basin and the surroundings controlled the sedimentation pattern. Goodlet (1957) indicated that in the Ayrshire Basin (SW of Central Basin) there are four main northeast trending faults (Fig. 65) causing abrupt changes in the thicknesses of the Lower Limestone Group. However, on the basis of isopach maps he indicated the existence of an areas of non-deposition (land areas) between Ayrshire and the Central Basin and to the Southern Upland Fault (Fig. 66).

The changes in the depth of the basin and of the river load, which resulted in the fluctuation of the thicknesses of the Lower Limestone Group delta and changed the shape and direction of the coarse sediments, are probably due to tectonic, climate and enstatic changes in sea level.

#### Origin of Cyclicity

As indicated on page 72, the Lower Limestone Group Consists of cycles and the investigation of these cycles suggests that they have a deltaic origin. Belt (1975) discussed some Scottish Carboniferous cyclothems and indicated that there are three schools of thought for the origin of cyclicity; the "American School", the "European School" and the "Deltaic School".

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The "American School" places the base of the cyclothem below the sandstones which cut down into subtidal marine shales and relate this downcutting or erosion to eustatic lowering of sea level which in turn is produced by tectonism or climatic change. The "European School" places the cyclothem boundary between the coal or rooty bed and the overlying marine shale or limestone and relates the origin of the cyclicity to tectonism, climatic changes or changes in the pattern of sedimentation. The "Delta School" places the boundary of the cycle between the marine limestone and the overlying shales and considers that the cyclicity results from repeated delta lobe progradation and abandonment. Belt (1975, p.430) concluded that the cycles in the Lower Carboniferous sediments of East Fife were most likely to have resulted from delta progradation and abandonment.

This study of the Lower Limestone Group showed that there is no vertical or lateral persistence of the facies in the investigated delta lobe advances (Figures 9-14). For example, the sediments of two delta lobes (post-Hurlet and post-Shields Bed, Figure 9) are represented by shelf-prodelta facies in section 22 and are succeeded by progradational and aggradational facies (subfacies  $A_2$  and  $B_2$ ) of the post-Blackhall delta. In the south of the studied area (e.g. section 2, figure 13), where the Hurlet and Shields Bed have coalesced, the post-Shields Bed sediments are represented by interdistributory bay facies (Subfacies  $B_2$ ) and succeeded by shelf-prodelta sediments

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of the post-Blackhall delta. In other words the sediments of the post-Hurlet, post-Shields and post-Blackhall delta lobes are quite different in these two sections and thus show lateral and vertical variations. As indicated on page 107, the areal extent of the prodelta and the coarse delta plain sediments is different for each delta lobe advance. The origin of the facies variations discussed above is due to the position of the sediments in each delta lobe with respect to the shoreline (see also figure 55, p.94). Therefore it is suggested that the deltaic model of sedimentation is the origin for the cyclicity in the Lower Limestone Group sediments. If this origin is accepted, therefore the base of the cycle should be taken at the top of the limestone units.

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The question remains as to why each delta lobe was finally abandoned. The delta sedimentation model assumes that the deposition of sediment in distributary channels increases their elevation until they become unstable and switch to a lower area marginal to the original lobe. Eventually, distributaries will switch back into the area of the original lobe after it has subsided in the absence of clastic influx, and a new cycle will form as a result of delta lobe progradation.

Within the Lower Limestone Group in the studied area (and also to the east and northeast in Fife and Midlothian, see George <u>et al.</u>, 1976) the limestone units are persistent in a direction normal to the shoreline. This indicates that either the delta (probably a big delta) periodically

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switched into another area outside the Midland Valley, or there were periodic hiatuses in the influx of clastic sediments. It is not possible to reach a conclusion as to which of the above possibilities is more likely from our knowledge of the Midland Valley alone.

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### 2.5 PETROLOGY OF THE SANDSTONES

#### 2.5a Introduction

From more than sixty studied thin sections of the sandstones, twenty one samples were selected for point counting, 500 grains were counted in each slide. The samples represent the post-Hurlet, post-ShieldS Bed, post-Blackhall and post Hosie deltas. Samples from different environments in each delta were also selected. Besides the environmental considerations the geographic distribution was taken into consideration and the samples were chosen from the north, middle and south of the studied area. The geographic location, the environment and the stratigraphic position of each sample is presented in Table 3.

## 2.5b Light Constituents

Microscopic study of the Lower Limestone Group sandstones, revealed the following main constituents (Table 3).

1. Quartz:

The results show that quartz is the predominant mineral and ranges from 60-88%. Inclusions of brown and green tourmaline, zircon and other unidentified minerals were observed within quartz grains. These inclusions possibly suggest a source area with magmatic (granitic) rocks or gneiss and schist (cf. Lobo and Osborne, 1976). An attempt was made to classify the quartz grains following Blatt and Christie (1963) and Folk (1968) and the following

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Sam- pl No.	Quartz and Chert	Feld- spar	Rock Frag- ments	Silica	Carbo- nates	Iron oxide	Matrix	Other	Total	Quartz	Sect. No.	Facies	Strat- igraphic position	Grid Reference
\$10	71.2	0.0	0.2	3.0	0.0	1.4	18.8	5.4		70.8	25	в	Post-Hu	741876
5538	74.4	0.2	0.0	5.8	4.4	4.0	2.4	8.4		73.4	19	в	Post-SD	661777
3555	67.6	1.6	0.2	3.4	20.2	2.0	2.2	2.8		67.4	22	A	Post-BK	644786
F66	81.4	0.0	0.0	9.2	0.0	3.6	5.2	0.6		81.2	12	D	Post-SD	873472
E2	65.4	3.4	0.6	0.4	28.2	0.2	0.8	1.0		65.4	39A	в	Post-MDH	659541
E12	66.0	0.0	0.2	9.4	0.0	5.8	2.2	3.6		65.4	40	A	Post-BK	662542
E29	46.4	0.8	0.2	0.6	39.6	4.2	2.2	5.8		45.4	42	с	Post-BK	621551
\$33	81.4	0.0	1.2	15.0	0.0	0.8	2.4	0.4		80.2	27	в	Post-SD	779883
N15	69.8	0.0	0.4	1.2	0.0	2.2	22.2	4.2	ples	69.0	48	в	Post-BK	991715
N32	70.2	0.0	0.0	2.8	2.6	2.2	12.2	10.0	BAR	70.2	52	D	Post-BK	498592
26	72.6	1.0	0.0	4.0	0.0	8.8	8.8	4.8	Ξ	71.8	34	D	Post-BK	509597
Z1	75.2	0.0	0.0	0.4	0.0	7.6	14.6	2.2	or	75.0	40.	D	Post-MDH	662 54 3
S6 8	75.2	0.0	0.0	6.0	0.0	15.0	1.6	2.2		75.2	29	D	Post-MDH	798831
F45	77.4	0.0	0.2	9.8	6.8	2.2	2.2	1.4	100	76.2	6	в	Post-SD	734457
F3	83.0	0.0	0.2	12.8	0.4	2.0	0.6	1.0		82.8	1	в	Post-SD	798419
F16	69.6	1.2	0.0	4.6	13.0	4.6	5.6	2.6		69.0	2	в	Post-SD	804432
\$5	67.8	0.4	0.2	2.2	0.0	2.0	21.6	6.0		66.8	25	В	Post-Hu	741876
GS57	76.2	2.6	0.2	2.2	0.0	4.8	3.0	11.0		73.6	20	A	Post-BK	640785
G 520	61.8	3.6	0.4	4.2	0.0	5.0	14.0	11.0		59.6	19	A	Post-BK	661777
E37	47.4	0.6	1.3	0.2	46.9	0.8	1.2	1.4		46.8	44	c	Post-BK	587536
G 558	72.6	0.0	0.2	7.2	0.2	12.4	5.0	2.6		72.0	20	A	Post-BK	640785

Table 3 Modal analysis of the Lower Limestone Group sandstones. Hu = Hurlet; SD = Shield Bed, BK = Blackhall Limestone; MDH = Mid-Hosi@ Limestone. types were recognized:

- A Undulatory (the predominant variety)
- B Non-undulatory
- C Polycrystalline grains in three types
  - (i) composite (fig. 67), (ii) semi-composite (Fig. 68),

(iii) stretched quartz (Fig. 69).

The presence of large composite grains probably indicates that the small undulatory types were derived from them. The undulatory types show a slightly undulatory to strongly undulatory extinction, which might indicate a non-stretched metamorphic origin (Folk, 1968; Deegan, 1970). However, the undulosity sometimes passes into the overgrowth suggesting post-depositional deformation.

#### 2. Feldspar

The feldspars range from 0-4% in the studied sandstones. Microcline and Perthite are the most common types, however undetermined plagioclase and zoned plagioclase (Fig. 70) (zoning indicates an igneous source rock) were observed.

### 3. Other Constituents

Minor amounts of chert and volcanic rock fragments (Fig. 71), were identified but these form a very low percentage. Mica in the form of muscovite was identified, the elongate flakes being bent during compaction.

In conclusion the study of light constituents suggested that the source area of the Lower Limestone Group

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Fig.67 Badly sorted sandstone, showing composite quartz grains. XN (X320). Post-Mid Hosie delta (Section 39A)

Fig.68 Badly sorted sandstone with semi-composite grains showing undulous extinction. XN (X130). Post-Mid Hosie delta (section 39A).

Fig. 69 Photomicrograph showing stretched quartz. XN(X100) Post-Blackhall delta (Section 44).





clastic sediment was mainly metamorphic and acidic (probably granite) indicated by the type of quartz and by the inclusions of brown and green tourmaline and zircon in the quartz grains. However, the presence of volcanic rock fragments suggested a volcanic source area (Andecitic type).

2.5c Classification of the sandstone and the grain size analysis.

Most of the sandstones of the Lower Limestone Group can be classified as quartzarenites according to McBrides classification (McBride, 1963). However, two samples are located in the sublithic arenite field, although they are very near to being quartzarenites (Figure 72).

## Grain size analysis

Fifteen thin sections were chosen for grain size analysis and 500 grains per slide were measured. The measurements were converted to the equivalent sieve size using Friedman's graph (Friedman, 1958). Mean grain size and sorting values (Inman, 1952) were obtained from the frequency cumulative curves (Fig. 73) and are presented in Table 4.

The mean of the grain size ranges from 1.90 to 4.00 (medium sand to very fine sand), although most of the samples had a mean of almost 30 (very fine sand). Sorting values range from 0.60 to 1.330 (moderately well sorted to poorly sorted), but most of the samples are moderately well sorted to moderately sorted. The poorer sorting





The mean grain size and sorting, obtained from frequency cumulative curves Table 4.

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phic position	ids.	lds	lds	 d>	los ie	los 1e	ď	ds.	thall	thall	thall	thall	ds	losie	hall
Stratigra	Post-Shiel	Post-Shie]	Post-Shiel	Post-Shiel	Post-Mid F	Post-Mid F	Post-Shiel	Post-Shiel	Post-Black	Post-Black	Post-Black	Post-Black	Post-Shiel	Post-Mid H	Post-Black
reference	777	883	432	419	831	543	472	883	542	785	550	597	457	543	715
Grid	611	617	804	798	798	657	873	617	662	640	607	509	734	657	166
Facies	£	В	В	В	D	B	D	B	¥	¥	D	B	D	Q	В
Sorting	0, 83	0.79	0.9	0.73	0.9	1.33	0.98	0.8	0.75	0.75	1.25	0.78	0.9	0.65	0.68
Mean grain size (MØ)	3.45	2.8	3.6	3.0	3.2	1.9	2.4	2.7	3.0	3.3	3.2	3.1	3.1	3.9	4.0
Sample No.	GS38	<b>S</b> 33	F16	F3	898	E2	F66	\$30	El2	GS55	N32	26	F45	zı	N15

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values are either due to the accumulation of fine sediment, such as sample in N32, or to the mixing of coarse, medium and fine sediments as represented by sample  $E_2$ , which is a crevasse splay deposits.

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## 2.5d Heavy minerals

Twenty one samples were selected for heavy minerals analysis. These samples represent, post-Shields Bed, post Blackhall and post-Hosic sandstones. Samples representing the different environments of these deltas and different geographic locations were also chosen. These samples were crushed according to Henningsen's (1967) method and 63-250 u size fraction was chosen (see Appendix I ). 300 grains were encountered in each slide and the percentages of heavy minerals species are presented in Table 5. The "ZTR" index (Hubert, 1962) which is the percentage of combined zircon, tourmaline and rutile among the transparent, nonmicace ous detrital heavy minerals was also calculated and is presented in Table 5.

The aim of the study of the heavy minerals was to locate the source area, and also to investigate any cryptic variation in the heavy mineral suite which might be controlled by environment, texture, diagenesis or geographic location.

The following minerals were identified. 1. Zircon.

Zircon makes up 7.0 to 57.4% of the heavy minerals suite. Different shapes and colours of zircon were found (Figures 74, 75 and 76). The main varieties are:

Sam- ple No.	Zircon	Tour- maline	Musco- vite	Rutile	Anatase	Garnet	Opeque Min.	Others	Total	ZTR index	Strati- graphic Position	Sect. No.	Facies	Grid Reference
F66	57.4	10.6	0.66	19.8	6.6	0.66	3.3	0.99		98.14	Post-SD	12	D	873472
\$30	22.26	42.47	6.51	7.88	8.9	3.43	6.43	2.14		92,86	Post-SD	27	В	779884
G \$55	34.09	5.68	11.36	19.32	9.09	5.68	12.5	2.27		87.43	Post-BK	20	A	640785
G <b>S</b> 38	42.0	16.0	24.0	6.0	4.7	4.7	1.3	1.3		91,43	Post-SD	19	в	661777
S52	28.18	12.82	33.64	10.09	3.82	3.73	4.73	3.0	•	88.36	Post-SD	26	B	760891
S6 8	28.5	37.5	1.0	19.5	6.0	4.5	1.5	1.5		93.4	Post-MDH	29	D	798831
F16	56.45	8.06	3.23	19,35	4.84	4,84	1.61	1.61		92.87	Post-SD	2	8	804431
E2	8.7	8.7	2.42	8.7	0.0	65.7	5.9	0.0	es	28.41	Post-MDH	39A	в	6 3 8 5 4 7
533	62.6	14,63	1.22	16.26	3.25	0.41	1.63	6.0	Ide	99.56	Post-SD	27	в	779884
R40	7.0	5.5	1.5	11.5	1.0	68.0	5.0	0. 5	sar	25.94	Post-BK	34	A	509597
F4 5	41,15	25.93	1.23	15.23	12.35	2.06	2.06	0.0	=	97.55	Post-SD	6	B	7344 57
F80	12.79	17.44	2,33	10.47	1.74	52.32	1.74	1.16	æ	43.21	Post-BK	15	A	668513
F3	40,8	37.36	3.49	10.92	2.3	0.0	5.17	0.0	for	100.0	Post-SD	1	в	7984 1 <b>9</b>
G\$58	12.5	42.5	13.5	21.5	8.5	0.0	1.5	0.0	*	100.0	Post-BK	20	A	640785
N13	48.3	16.3	1.3	18.0	9.0	5.3	1.7	0.6	0	93.97	Post-BK	48	B	991715
N15	51.66	8.0	6.0	21.66	7.66	2.66	2,33	0.0		93.97	Post-BK	48	в	991713
N32	12.00	10.0	2.0	7.33	2.33	63.60	3.00	0.33		31.77	Post-BK	52	D	498 592
<b>Z6</b>	28.66	7.66	1.0	12.66	2.33	46.66	0.66	0.33		51.03	Post-BK	34	Ď	509597
<b>Z</b> 1	25.33	8.33	1.33	8.66	1.66	52.0	2.33	0.33		44.72	Post-MDH	40	σ	6 <b>6</b> 1542
GB48	36.36	20.55	7.12	23.72	9.09	1.98	0.79	0.40		97.6	Post-SD	16	в	680788
E12	21.79	6.23	33.46	8.56	8,17	2.14	0.39	0.0		94.45	Post-BK	40.	A	662342

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Table 5 Heavy minerals analysis. SD = Shield Bed, BK = Blackhall Limestone, MDH = Mid Homit Limestone.

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Fig. 74 Photomicrograph showing spherical brown tourmaline (T), and less elongated zircon . (Z) (X320). Post-Shield Bed delta (Section . 12).

Fig. 75 Photomicrograph showing highly rounded zircon (Z), tourmaline (T) and reddish brown rutile (R). (X320). Post-Shield<sup>5</sup> Bed delta (Section 25).

Fig. 76 Photomicrograph showing rounded, slightly elongated and fractured zircon (Z). Notice the zircon inclusion. X320. Post-Blackhall delta (Section 48).


b. Colourless, prismatic with rounded edges.

c. Colourless, euhedral grains.

d. Pinkish, rounded to ellipsoidal.

e. Pinkish, prismatic with rounded edges.

f. Pinkish, euhedral grains.

Inclusions are common within the zircon grains, being most common in the pinkish varieties. Some of the colourless grains are turbid, some are highly fractured and few grains show overgrowths. Although highly elongated grains with euhedral and the rounded prismatic edges are found, these are less than the less elongated or equant grains.

2. Tourmaline

Tourmaline grains range from 5.7 to 42.0% of the heavy mineral suite and were observed in all the specimens. The following types were identified.

a. Green rounded, ranging from highly spherical to ellipsoidal (Figures 74 and 75).

b. Irregular shaped grains.

c. Green prismatic.

d. Brownish to slightly pinkish rounded grains.

e. Brown prismatic.

Some of the grains contain inclusions. Overgrowths were observed but are rare, whereas "Hacksaw" terminations probably caused by instratal solution were also observed.

3. Garnet.

Garnets, of both colourless and pinkish varieties, were observed. These range from 0-68% of the heavy mineral suite. Most of the grains are angular with subconcoidal fractures but some of the grains show rounded edges and inclusions are sometimes present.

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4. Rutile

Rutile ranges from 6-23.7% of the heavy minerals suite and is weakly plleochroic. Two colour varieties were recognized, pale earthy brown and red types. The grains are prismatic with slightly rounded edges.

5. Anatase

This is present as pale yellowish grains with a metallic lustre. They are uniaxial or biaxial with very small 2v. The grains are equidimensional and subangular to subrounded. The identification of anatase was checked using an x-ray diffraction camera.

6. Muscovite

Muscovite, occurring as lath shaped grains, form 0.66 to 33.5% of the heavy mineral suite. It indicates a metamorphic or igneous source (Philip, 1968).

#### 7. Other minerals

Some grains are coated by iron oxides, which makes their identification difficult. However, grass-green, small size grains were observed in a few samples which are tentatively identified as epidote.

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2.5d(i) Relationships between grain size, sorting and the composition of heavy mineral suite

The mean grain size and sorting of the sandstones which were analysed for heavy minerals are plotted against the percentages of each heavy mineral species as indicated in Figures 77 and 78. No relationship exists between the mean grain size and the distribution of the main heavy minerals; zircon, tourmaline, garnet and rutile. However, the plotting of sorting against composition revealed a weak relationship which is best defined with zircon followed by tourmaline and then the other minerals.

2.5d(ii) Relationship between the main heavy minerals

To find if there are any correlations between the concentrations of the various heavy minerals, the frequencies of the main heavy minerals were plotted against each other as indicated in Figures 79, 80 and 81.

The figures show that, apart from inverse relationships between zircon and garnet, zircon and tourmaline and a weak positive relationship between rutile against zircon and tourmaline, there are no other obvious relationships. Since there is no relationship between the main heavy minerals and the grain size and sorting (apart from weak relationship between sorting and zircon frequencies) the above variation is probably inherited from the variation in the source area. Mapstone (1971) related the variation between heavy minerals in the Upper Limestone Group (Carboniferous), Midland Valley of Scotland to the variation within the source area.

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Fig. 78b Relationship between the sorting and the frequency of heavy minerals.







2.5d(iii) Vertical and Lateral Variations in the Heavy

## Minerals

A few specimens were chosen to discover if there is any vertical variation within one deltaic advance or between several advances.

Figure 82b shows the vertical variation in tourmaline, rutile and zircon, for one deltaic advance, the post-Shieldý delta. However, Figure 82e reveals a variation in tourmaline and muscovite for the post-Blackhall delta. There are vertical variations in garnet, rutile, zircon and tourmaline between deltas as shown in Fig. 82.

There is no relationship between the size, sorting and the frequencies of the heavy minerals as discussed above (Chapter 2, Section 2.5d(i)). Therefore, these vertical variations might be related to changes in source area.

To discover any cryptic lateral variations, the values of the zircon, garnet, rutile and tourmaline frequencies were plotted for the post-Shields, post-Blackhall and post-Mid Hosi deltaic sediments in the studied area (Table 6, Figures 83-91).

For the post-Shields delta the map reveals that zircon values are significantly higher in the southern part of the area than the northern part, and the garnet values, in comparison, are low throughout the area. The plotting of rutile and tourmaline does not show any specific trend and the values are variable.

For the post-Blackhall deltaic sediments zircon values are high in the north and northeast which, ranging from













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Fig.89 Map showing the lateral variations of rutile in the post-Blackhall sediments





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Summary of the distribution of the main heavy minerals in the studied area Table 6

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	Post-Shields	Post-Blackhall	Post-Mid Hosie
Zircon	High in the studied area ranging from 22.3% to 63.6% and relatively higher in the south than in the north.	<ul> <li>A. High in the north and north-east ranging from 34.1% to 51.6% with exception of one sample (12.5%).</li> <li>B. Low in the southwest and ranges from 7% to 28.7%.</li> </ul>	The two samples have values of 8.7% and 25.3%.
Garnet	Low values and ranging from 0.0 to 4.8%.	<ul> <li>A. Low in the north and north-east ranging from 0.0% to 5.7%.</li> <li>B. High in the southwest ranging from 46.7% to 52.3% with the exception of one sample (2.1%).</li> </ul>	The two samples have high value; 52.0% and 65.7%.
Rutile	Ranging from 6 to 23.7% without areal variation	Ranging from 7.3 to 21.6 and slightly lower in the south- eastern part than northern and northeastern part.	Constant and of 8.7%
Tourmaline	Ranging from 8.1% to 42.5% without areal variation.	Ranging from 5.7 to 425 without specific trend.	Constant and the two samples have 8.7% and 8.3% values

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34.1% to 51.6% with exception of one low sample (12.5%), but low in the southwest of the studied area where they range from 7% to 28.7%. Garnet is low in the north and northeast (0.0-5.7%) and high at the southwest (46.7% -52.3%) except for one sample (2.1%). No specific trend was found for rutile and tourmaline. Two samples from the post Mid Hosi delta show an inverse relationship between the garnet and zircon whereas the values of rutile and tourmaline are constant.

Muir (1963) in his study of the Carboniferous Millstone Grit from the Midland Valley of Scotland and Mapstone (1971) in the Carboniferous Upper Limestone Group, reported lateral variations of heavy minerals and related these variations to the source area. In the studied Lower Limestone Group sediments, as no relationships were found between grain size, sorting, diagenesis and the frequency of heavy minerals (discussion later on), the lateral variations in heavy minerals may similarly be inherited from the Source area.

2.5d(iv) The relationship between light and heavy minerals

Hubert (1962) noted that heavy mineral stability measured by the "ZTR" index, is usually related to the maturity of the light fractions. He added that these relationships might be due to tectonic quiescence, weathering and mechanical abrasion. In the clastic coarse facies of the Lower Limestone Group the "ZTR" index is more than 87% (Table 5) except for six samples which have a high

concentration of garnet and "ZTR" indices from 26-51%. The heavy minerals stability, coupled with the quartzdomination of the light fraction, indicates a high maturity for the light and heavy mineral fractions. These probably indicate a multicyclicity and/or very prolonged abrasion and weathering as noted for similar stable heavy minerals associations by Jain (1972) in Precambrian Gamri Quartzite, India and Ojakangas(1963) in the Upper Cambrian Lamotte Sandstone, Missouri.

2.5d(v) The effect of sorting, instratal solution, weathering and abrasion on the concentration of heavy minerals.

Van Andel (1959) indicated that four parameters effected the concentration of heavy minerals; selective sorting, instratal solution, weathering and abrasion. A weak relationship exists between the sorting and the frequencies of zircon and, to a lesser extent tourmaline; although this relationship is not clear for the other heavy minerals. Therefore the effect of sorting is very Some grains of tourmaline show "Hacksaw" limited. terminations which indicate solution during diagenesis, but Hubert (1962) considered that instratal solution is unable to produce a high concentration of stable heavy minerals. Weathering and abrasion could produce high association of Walker (1967), on the basis of other stable heavy minerals. workers' data reported that unstable heavy minerals such as hornblende and augite are found in fluvial and marine

sediments derived from igneous and metamorphic rocks, even where a hot, moist climate and deep weathering exists in the source area. However, very prolonged abrasion of the sediments can produce an association of stable light and heavy minerals such as quartz, zircon and tourmaline (Ojakangas, 1963; Jain, 1972).

### 2.5d(vi) Interpretation of the source area

Blatt et al. (1972) reported that euhedral grains of zircon indicate a magmatic (granitic) origin, brown to yellow-brown tourmaline represent metamorphic origin, the other colour varieties of tourmaline indicate granitic or pegmatic sources and most garnets are derived from pelitic schists. They added that sands composed entirely of a stable light fraction and stable heavy minerals (e.g. zircon and tourmaline) might represent recycled sediments, although they did not exclude the effect of abrasion or weathering on the concentration of stable light and heavy minerals. Pettijohn (1975) stated that if heavy minerals are derived from sedimentary rocks the less stable minerals tend to be absent whereas the more stable (e.g. zircon and tourmaline) are predominate and are highly rounded.

Muir (1963) in his study of Millstone Grit stated that the highly elongated zircons were derived from granties or pegmatites, whereas the equant zircons originated from sedimentary rocks or at least from sedimentary rocks derived from a metamorphic terrain. He added that colourless and brown garnets were derived from pelitic schist, rutile from

granulites (quartz mica schist), quartzite, gneiss and phyllite whereas the brown tourmaline originated from slate, phyllite and pelitic schist. Mapstone (1971) in his study of the Upper Limestone Group also reported that highly elongated zircon indicates granitic or pegmatic origin whereas the less elongated grains are from granulites (quartz mica schists) and quartzite. He added that the red and yellow rutile indicates a metamorphic source area, such as granulites (quartz mica schist), quartzite, gneiss and phyllite whilst the colourless and pinkish varieties of garnet indicated pelitic schist. He reported also well rounded tourmaline and considers them to represent poly-cyclic sedimentary origin. Winchester (1974) reported that facies of garnet, silliminite, kyanite, andulsite, biotite and chlorite comprises the Scottish Caledonian Highlands.

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It is suggested that the presence of brown tourmaline, colourless and brown garnet and red and brown rutile indicates a metamorphic origin (e.g. pelitic schist, quartz mica schist, quartzite, gneiss, phyllite and slate). The presence of euhedral zircon and other varieties of tourmaline (e.g. green tourmaline) indicates granite and pegmatite. However, the zircon and tourmaline are highly rounded which might indicate weathering, abrasion or multicyclicity. Weathering and abrasion can be effective in removing unstable heavy and light minerals (Hubert, 1962; Blatt <u>et al.</u>, 1972; Pettijohn, 1975), but it is unlikely that they could produce an association of

stable light and heavy minerals, like that found in the Lower Limestone Group, from a non-sedimentary source without the presence of even a small amount of unstable heavy minerals. Therefore, the predominance of stable heavy minerals, the presence of well rounded zircons and tourmalines, some with overgrowths, probably indicates that the source area of the coarse clastic sediments was sedimentary. However, the presence of a small amount of euhedral zircon and tourmaline probably indicates that these grains either escaped from the effect of weathering or have passed through fewer sedimentary cycles in comparison to the well rounded zircon. The presence of brown tourmaline, colourless and brown garnet and red and brown rutile, possibly indicate that the sedimentary rocks which were the source rocks of the Lower Limestone Group sediments were themselves derived mainly from a metamorphic source. The presence of euhedral zircon grains and other varieties of tourmaline (green type) indicate that these sedimentary rocks were also originated partly from granites and pegmatites.

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If the sedimentary origin is accepted, therefore, the Old Red Sandstone and the ? older Carboniferous sediments and the Old Red Sandstone andesitic lavas (indicated by the presence of volcanic rock fragments) are the main sources for the clastic coarse facies of the Lower Limestone Group.

Greensmith (1965) identified similar heavy minerals to the identified heavy minerals of the

Lower Limestone Group in this study in the Calciferous Gandstone Series and considered the source area for the Calciferous Sandstone Series to be the Old Red Sandstone sediments and igneous rocks (Andesitic Lava). Ali (1976) recognized the same heavy minerals also together with unstable species (e.g. pyroxene, amphibole, kyanite etc.) in the Old Red Sandstone, Midland Valley, Scotland. He indicated that the heavy mineral fraction of the Upper Old Red Sandstone contains 0-26.66% garnet, 0-18.66% zircon, 0-6.6% rutile and 1-11.66% tourmaline whereas the Lower Old Red Sandstone contains 2.66-88.33% garnet, 0-7.33% zircon, 0-2.3% rutile and 0-4.66% tourmaline.

# 2.5e Matrix

The matrix is polymineralic, consisting of very small grains of quartz, sericite, clay and iron oxides, ranging from 0.8-22.2%. X-ray diffraction shows that the kaolinite is the main clay mineral. Although the origin of the matrix is most probably primary, the decomposition of feldspars could have supplied some secondary components to the matrix.

#### 2.5f Cement

Three types of cement are recognized in the studied Sandstones; carbonate, iron oxide and silica. All the studied sandstones contain cementing materials, and either contain all the three or sometimes just two types. In some specimens a high proportion of cement and matrix

exists whereas in others only cementing materials are important and the matrix is insignificant.

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The order of formation of the cement is considered to be as follows.

1. Iron oxides

- 2. Silica
- 3. Carbonates
- 4. Iron oxides.

The features which support the interpretation that iron oxides were the first cementing materials are:

## Petrographic relationship

i) Patches of iron oxide inside the carbonate cement indicating that the carbonate cement was deposited after the iron oxide.

ii) Sometimes the iron oxide defines the overgrowthsof silica (Fig. 92) which indicate that the iron oxideformed before the secondary overgrowths of silica.

## Field relationship

Field observations indicate that the coarseningupwards sequences change from black shale (high organic content) with siderite and/or pyrite to grey siltstones to sandstones which are usually yellowish, suggesting oxidizing conditions during the deposition of the sandstones.

It is indicated in the discussion of ironstone bands (Chapter 4) that calcite formed in weakly alkaline environment and siderite formed in acidic reducing



Fig. 92 Iron oxide cement (arrowed) defining the overgrowth of silica. The overgrowth is better developed on coarse grains than smaller grains. XN(X320). Post Shield Bed delta (Section 12).

Fig. 93 Similar to the above, but with dust defining the overgrowth. XN(X320). Post-Blackhall delta. (Section 22).

Fig. 94 Photomicrograph showing high amount of carbonate (C) replacing the quartz grains (q). XN(X320). Post-Mid Hosi? delta (Section 39A).

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environment near to the sediment water interfaces.

Walker (1967) in his study of the recent Pleistocenepho cene sequence of sediments, in the Sonaran desert. California, and the Late Paleozoic of Colorado indicated that the red pigment in sandstones is due to ferrous iron released by progressive diagenetic alteration of detrital heavy minerals (e.g. hornblende, biotite and iron oxides etc.) and amorphous ferric hydrate (limonite) finally being deposited as hematite from oxygenated pore waters. He added that the climate during deposition of sandstones may be irrelevant to the formation of hematite cement, and the rate of alteration of iron-bearing mineral grains depends on the amount of water that moves through the sediments and the rate of alteration is much slower in arid than moist climate. He stated also that ground water can be oxygenated to a depth of several hundred to thousands of feet below the water table. McBride (1974) indicated that the grey colour of the Difunta Group (Late Cretaceous to Paleocene) of north eastern Mexico deposited in a semi-arid to sub-humid climate, is due to the presence of plant debris and organic materials. He added that most red coloured of deltaic plains are of red hematite coating which formed by early post-depositional alteration of iron-bearing detritus and dehydration of iron hydroxide on the delta plains. He added also that some sandstones are not reddened because not all the organic matter was destroyed.

In the studied Lower Limestone Group, the sequence of

the sediments changes upward from dark shale, into grey siltstones and then into yellowish sandstones. In other words the colour is dark to grey in fine sediments and yellowish-brown in coarse sediments. Heavy minerals study in the sandstones indicates that apart from iron oxides which range from 0.4 to 12.5% no unstable iron-bearing heavy minerals which could produce iron by alteration were identified. The maturity of heavy minerals is interpreted to be inherited from the source area (see heavy minerals section). It is suggested that in the fine sediments the permeability is low and therefore water circulation is limited. The disintegration of organic material probably consumed the oxygen and a reducing environment was produced. This is supported by the presence of siderite or pyrite in the shale. In the coarse sediments and at a shallow depth below the sediment/water interface, there was enough water circulation for oxidizing condition to exist. It is suggested also that the pore water is originally rich in ferrous iron (probably migrated from the underlying shales) or iron released by diagenetic alteration of iron oxides. Therefore, the iron might have been deposited as ferric oxides in the sandstones under oxidizing conditions soon after the deposition of the Sandstones. Some of the sandstones are grey in colour. These mostly contain a lot of organic material, rootlets and are badly sorted. The organic material probably consumed all the oxygen and together with the bad sorting which affected the circulation of pore water reducing

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conditions resulted. Deegan(1970) indicated that the iron was precipitated from solution in oxidizing conditions forming a primary diagenetic iron oxide cement in the Lower Carboniferous rocks, Kirkudbrightshire, Scotland.

The silica cement is represented by quartz overgrowths on quartz grains and is normally defined by dust or iron oxides (Fig. 93). However, in clean quartz grains it is difficult to identify the overgrowths. Quartz overgrowths are best developed in very coarse-grained sandstones and are abundant in mature sandstones. Aalto (1972) indicated that the quartz forming overgrowths in his study of Orthoquartzite sequences (Devonian-Tertiary), Columbia, may have been derived from the solution of fine quartz. Such an origin may be applicable to the Lower Limestone Group sandstones.

Carbonate in the form of calcite and dolomite is sometimes the main cement. The calcite is usually nonferroan, although it can be slightly ferroan, and has locally replaced the silica and iron oxide cements (Figs. 94, 95). The dolomites which are volumetrically less important are microcrystalline ferroan types.

The solubility of minerals, which is a function of physical and chemical factor (e.g. pH, temperature, pressure and permeability) controls the precipitation of diagenetic minerals. At shallow burial depths and when the pore fluids were slightly acidic (due to the presence of  $CO_2$  formed by the bacterial decomposition of organic matter) the silica was precipitated as overgrowths. When



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Fig. 95 Photomicrograph showing the iron oxide cement (I), defining the silica overgrowth(s). Notice the carbonate cement (C) replacing the overgrowth and iron oxide cement. XN(X320). Post-Blackhall delta (Section 22). the pH rose, as  $CO_2$  was depleted at greater depths and high temperatures, the carbonates were precipitated (cf. Sharma, 1965).

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Sometimes the calcite is surrounded by an iron oxide rim. The staining shows that within a single patch of calcite cement surrounded by iron oxide, the part of calcite adjacent to the iron oxide rim is ferroan, although the calcite of the patch is non-ferroan. Greensmith (1965) noted that the iron oxide in the Calciferous Sandstone Series was partially formed by the destruction of ferroan Neal (1969) indicated that the brownish dolomite. weathering surface of the limestones is the result of ironoxide produced by the destruction of ferroan calcite in the Blackjack Greek Formation (Pennsylvanian), Missouri. Al-Hashimi & Hemingway (1973) in ther description of the origin of the recent rusty crusts in the coastal sections of the Middle Limestone Group (Carboniferous), Northumberland, indicated that the existence of iron hydroxide together with rhombohedral calcite in the rusty crusts and the absence both in the unweathered part of the dolostone suggested that the iron hydroxide was formed by the oxidation and hydration of the ferrous iron in the dolomite. They added that this process takes place when the carbonate units are in contact with the present day sea water, as is indicated by the predominance of rusty crusts in the coastal exposures compared with inland exposures. They indicated that the oxidation of ferrous iron and its hydration to form limonite or geothite with the consequent

gradual breakdown of the dolomite crystals can be produced by fresh water (ground water circulation), although the reaction is faster with seawater.

In the Lower Limestone Group sandstones, the ferroan calcite cement probably undergoes a similar type of destruction to the ferroan dolomite and calcite discussed above, the leached ferrous iron being oxidized to iron oxides by the oxygenated ground water. Oxygenated rain water also might help in the oxidation of the leached ferrous iron.

#### 2.5g Diagenesis

The contacts between quartz grains due to diagenesis were classified according to the scheme of Taylor (1950) and three main types recognized

- Simple line to slightly irregular penetration contact (Fig. 96).
- 2. Concavo-convex contact (Fig. 97).
- 3. Micro-stylolite contact (Fig. 98).

The most common types are simple to slightly irregular penetration and concavo-convex contacts, which result from pressure solution due to depth of burial (Taylor, 1950). These types of contacts probably indicate that there was probably sufficient silica released by pressure solution to form the silica cementation. Sibley and Blatt (1976) pointed out that the silica which forms the cement in the Tuscarora Orthoquartzite (Silurian), Virginia, as revealed by cathodoluminescence, originated partly by pressure



Fig. 96 Highly mature sandstones, showing simple straight to slightly irregular contacts (S) and concavoconvex contacts (C). XN (X320) Post-Blackhall delta (Section 20).

Fig. 97 Similar to the above thin section with predominance of concavo-convex contacts in which some of them show faint micro-stylolitic contact. XN (X320). Post-Blackhall delta (Section 40).

Fig. 98 Development of micro-stylolite contact (m). Notice also the predominance of concavo-convex contacts (C) and the low percentage of simple straight to slightly irregular contacts. XN (X320). Post-Shield Bed delta (Section 12).

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solution of quartz grains. However, they noted that part of the authigenic silica could also have been produced by the diagenesis of clay minerals, dissolution of biogenic silica, desilicification of volcanic debris and from normal surface water.

The edges of the quartz grains are often corroded where in contact with the matrix when the latter forms more than 10% of the rock (Fig. 99). A relationship exists between the matrix content, the suturing of the grains and secondary overgrowth of quartz. It was found that as the amount of matrix increases the suturing of the grains decreases (Fig. 99). An increase in the amount of matrix was also observed to accompany a decrease in volume of quartz overgrowths.

It is suggested that the high proportion of matrix decreased the permeability and therefore restricted the movement of pore water and consequently inhibited the formation of quartz overgrowths. The high proportion of matrix also reduced the number of grain contacts, restricted pressure solution and therefore the suturing of the grains is not common. Similar findings and conclusions have been reported by Whisonant (1970) in his study of the basal Cambrian Chilhowee Group in eastern Tennessee and by Aalto (1972) in Orthoquartzite in

## 2.5h Conclusion

The study of light minerals of the clastic coarse sediments of the Lower Limestone Group, which are mainly


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Fig. 99 Slide shows large amount of matrix. Notice that the matrix has corroded the quartz grains at (P) and also notice the small amount of silica overgrowth and lack of suturing of the grains in comparison with the rocks with little matrix. XN(X320). Post-Hurlet delta (Section 16).

of very fine sand and moderately well sorted to moderately sorted, indicates that they consist mainly of stable light minerals; i.e. they are largely of quartz. Minor quantities of chert, feldspar, and volcanic rock fragments are found. Matrix and cement (carbonate, iron oxides and silica) were identified in most of the studied thin sections. Three types of quartz were recognized, undulatory, non-undulatory and polycrystalline quarts. The undulatory type, which is the predominant variety, probably indicates non-stretched metamorphic source rocks. Inclusions of brown and green tourmaline and zircon are common in the quartz grains which might suggest a source area of granite, gneiss and schist.

The heavy minerals study reveals the existence of stable species only. There is no relationship between the different delta facies and the frequencies of heavy minerals. However, lateral variations in the amount of zircon and garnet were observed which might reflect a variation in the source area. The stability of light and heavy minerals probably indicates multicyclicity, weathering and very prolonged abrasion. However, the predominance of well rounded zircons and tourmalines, some of which have overgrowths, probably indicates a sedimentary origin. The brown variety of tourmaline, red and brown rutile and garnet indicate a metamorphic origin, whereas the green varieties of tourmaline indicated granite or pegmatite. Therefore, most likely, the source area of the Lower Limestone Group sediments was sedimentary rocks, derived

from a metamorphic terrain which also include granites or pegmatites, and subordinate volcanic rocks (probably andesite).

Three types of cement were observed; iron oxide, silica and carbonate. The iron oxide cement is probably early diagenetic, formed soon after the deposition, as indicated by the petrographic relationships. The silica, carbonate and latest iron oxides are probably late diagenetic.

The diagenetic features were discussed including the contacts between the grains, the relationship between the matrix and the secondary overgrowths of quartz and the suturing of the grains. Concavo-convex and simple line contacts are the main type of contact between the grains and were probably due to pressure solution at depth. An inverse relationship between the amount of matrix and the amount of secondary quartz and the suturing of the grains were found. This is probably because an increase in the matrix causes a reduction in permeability with a consequent decrease in secondary overgrowth and a reduction in the number of grain to grain contacts which therefore restricts the effects of pressure solution and the suturing of grains. The stages of diagenesis of the Lower Limestone Group sandstones are summarized in Figure 100.

### 2.51 Main Conclusion

The clastic sediments of the Lower Limestone Group are considered in this study to have been deposited by







Formation of iron oxide rim cement (I).





Development of straight to slightly irregular, boundaries with few concavoconvex contacts (c) and formation of silica cement(s)

Development of concavoconvex (c) and microstylolite contacts (m) and formation of carbonate cement (Ca)



Replacement by the carbonate cement of some of the silica and iron oxide cement. The percentage of the concavo-convex and microstylolite increases. The destruction of ferroan calcite cement results in the formation of late iron oxide cement (Li).

Fig.100 Diagram illustrate the diagenetic sequence in the studied sandstone

delta progradation. Four main facies were recognised within these deltaic sediments, progradational, interdistributary bay, abandonment and channel facies. Four subfacies were recognized within the progradational facies. three of them are considered to have been deposited in axial or near axial, lateral and distal positions on a mouth bar. The fourth subfacies is represented by only one section in which it is difficult to speculate about its location on the mouth bar. The interdistributary bay facies are either equivalent horizontally to the progradational facies or overlie them. The interdistributary facies were classified into five subfacies, three of them are interpreted to have been deposited in fresh water bays by overbank flooding or by crevasse The other two subfacies were deposited in open Channels. interdistributary bays indicated by the presence of marine fauna in the basal shales. The abandonment facies are represented by fossiliferous marine siltstones or sandstones, which reflect periods of non-deposition and biogen ) ic reworking, and coal, which reflect an extensive development of marshlands. The channel sediments were mostly non-fining upward and indicate straight-channel types.

Six delta advances, which debouched from the NE, are recorded in the area studied and were each followed by the deposition of a carbonate member after the delta lobes were abandoned. The investigation of these deltas reveals that one of them, the post-Blackhall, represented a

constructive, fluvially influenced elongate delta type prograding into a deep basin. No mouth bar sections were found in the other deltas and mostly their sediments are of interdistributary bay and shelf-prodelta origin. However, their thickness is not more than 15 m which indicates their progradation into shallow basin(s) (slightly more than 15 m). The changes in the depth of the basin and river load, which were probably due to tectonic, climate and eustatic change in sea level, probably caused the fluctuation of the thicknesses of the investigated deltas.

The study of the petrology indicated the presence of stable light and heavy minerals, which probably indicates multicyclicity, weathering and very prolonged abrasion. The predominance of well rounded zircon and tourmaline, some of which have overgrowths, probably indicates a The brown variety of tourmaline, red sedimentary origin. and brown rutile and garnet indicate a metamorphic origin, whereas the green variety of tourmaline indicates granite or pegmatite. It is suggested that the source area of the Lower Limestone Group sediments was mainly of sedimentary rocks derived from metamorphic rocks and granites or pegmatite. Three types of cement were identified and their paragenetic sequence is: iron oxides, silica, carbonates and iron oxides. Three types of contact between the quartz grains due to diagenesis were recognised in which the concavo-convex and the simple line contact are the predominant and probably due to the pressure

solution at depth. Inverse relationship between the amount of the matrix and the amount of secondary quartz and the suturing of the grains were observed. It is suggested that the presence of matrix resulted in the lack in the permeability and therefore a decrease in secondary overgrowths and the number of grain to grain contacts.

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THE SEDIMENTOLOGY OF THE CARBONATES

## 3.1 CARBONATE FACIES

The marine units of the Lower Limestone Group consist of carbonates and shales. The carbonate units were studied in each section and an attempt made to classify them into different facies (Figs.101-105). The term facies refers here to a bed or part of a bed with a particular lithology, fossil content and structure.

An attempt has been made to relate each of these facies to an environment of deposition. The diagenesis and dolomitization of the carbonate facies were also studied in order to construct a paragenetic sequence and to help with the understanding of the processes of dolomitization within the carbonate units. The petrographic terms used here are those of Folk (1959, 1965) and Bathurst (1975).

The following facies were recognized and their distribution is shown in Figures 101-105.

#### 3.1a Biomicrite facies (Facies 1)

The colour of the limestones of this facies ranges from light-medium grey into dark grey with slightly brownish to yellowish-brown weathering surfaces. The thicknesses of the limestone beds of this facies varies from 15 cm to more than 5 metres. When they are thick, they consist of units ranging from 5 to 60 cm usually separated by shale seams (5-25 cm). No structures were observed in the field and all the beds are massive (Fig. 106 ). Most of the units of this facies have sharp



Fig.101Facies distribution of the carbonate rocks in the Campsie Area. (16) number of section, (2) number of facies.

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bases, although some have wavy or rippled bedding. The observed fossils in the field are up to 3 cm long and some of them are concave upward. A small biostrome was observed in the field. This is of micrite composition and the fauna are of crinoids, brachiopods, bivalves and bryozoans which are similar to the surrounding beds. This facies either forms a bed interbedded with noncarbonate sediments (e.g. shales) or forms part of a bed interbedded with the micrite, dolomitic biomicrite and ostracod-bearing dolomite facies.

Microscopic study shows that the biomicrite facies limestones are composed of micrite, allochems, siderite and detrital grains such as quartz and iron oxide. The allochems are mainly skeletal grains (Fig. 107) and make up more than 10% of the volume of the rock. The skeletal fragments include crinoids, brachiopods, bivalves, foraminifera, ostracods, gastropods and bryozoans together with unidentified fragments. Intraclasts and pellets are seldom present and do not form more than 10% of the rock's volume. Some of the fossils, such as crinoid fragments, have straight, unabraded edges (Fig. 108). The insoluble residue, of which quartz and clay minerals are the main constituents, ranges from 3.5% to 37%. The clay is mainly kaolinite. Neomorphism has intensively affected this facies and nearly obliterated the allochems.

Heath et al. (1967) pointed out that crinoids, brachiopods and benthonic foraminifera lived on either





Fig.106 Massive bed of the biomicrite facies (arrowed). The overlying beds are of micritic facies. Hurlet Limestone (Section 16).

Fig.107 Photomicrograph of the biomicrite facies, which consist mainly of crinoids (C) and brachiopods (B). Peel(X100). Hurlet Limestone (Section 41).

Fig. 108 Photomicrograph showing straight edges of the crinoids. Peel (X130). Top of the Blackhall Limestone (Section 16).

172B rite facies (arrowed). | micritic facies. n 16). iomicrite facies, which ds (C) and brachiopods (B). tone (Section 41). straight edges of the Top of the Blackhall . . . . . . . . . . . ..... . .. ----.....

side of wave base where water circulation was good and able to carry food. They added also that bryozoans prefer highly agitated conditions well above wave base, although they have a wide range of tolerance. They interpreted a similar facies to the studied biomicrite facies, from the Bird Spring Group (Pennsylvanian ) of Nevada as the deposit of a weakly to moderately agitated marine environment on the basis of the diversity of fauna and the micritic composition of the matrix. Laporte (1967) also indicated a subtidal environment for a similar facies in Manlius Formation (Lower Devonian) of New York and he considered that the diversity of fauna indicated continuous marine submerge. Braun and Friedman (1969) also noted that an abundant and varied fauna implies a subtidal environment whereas the micrite indicates a low energy environment from Tribes Hill Formation (Lower Ordovician), New York.

Thusu (1972) described a facies from the Rochester Formation (Middle Silurian), Southern Ontario and interpreted the environment to be a calm subtidal environment with open circulation in which he noted that the presence of bryozoans indicate a slow rate of sedimentation. Laporte (1971) pointed out that the good preservation of fossils and a muddy calcareous matrix indicates a deep subtidal facies formed offshore, below mean wave base. However, he interpreted a calcilutite facies in the Paleozoic carbonates of the Central Appalachian, as a shallow subtidal facies formed below mean low water and

above mean wave base and stated (p.731) that "lack of cross-stratification and abundance of mud in the calcilutites does not demand deposition below wave base because not all shallow subtidal deposits need be current agitated".

Purdy (1963) described an identical facies to the biomicrite facies in the Bahama Bank deposited in water less than 6 m. deep. Asquith (1967) indicated that the depth of water under which a similar facies were deposited in the Mifflin member (Ordovician) of SW Wisconsin was less than 10 metres.

The biomicrite facies in the Lower Limestone Group can therefore be interpreted as the deposit of a submerged, marine low energy, outer neritic, subtidal environment as indicated by the nature of the matrix (mainly micrite), The the diversity of fauna and the lack of structures. preservation of fossils such as crinoids which have unbraded straight edges indicates a lack of wave or current action. The depth of water under which the sediments of this facies accumulated was probably of the order of 10 metres. Selim and Duff (1974) described the carbonate units in a section from the Lower Limestone Group at St. Monance, East Fife, and they interpreted the depositional environment as relatively deep water below wave base with weak currents. Some of the skeletal fragments of this facies and also of micrite facies are broken (Figs.134 & 135). Zankel (1969) obtained a relationship between the insoluble residue and lithification and he noted that a high insoluble residue prevents

lithification, thus allowing greater compaction of the unlithified sediment which in turn leads to the destruction of fossils by crushing. The high insoluble residue present in this facies and the destruction of fossils is interpreted as due to compaction during burial.

Intraclasts and peloids (Figs. 109 and 110) were identified in this facies and in the dolomitic biomicrite and micrite facies and in the Ostracods bearing dolomitic facies. Their characteristic features and origin will be discussed as follows:

# Intraclasts

These are angular to slightly rounded or irregular, sharp and poorly sorted. Most of the intraclasts range in size from 0.5 mm to 2.1 mm. However, some units contain large size intraclasts which sometimes exceed 1.5 cm (e.g. Blackhall Limestone, sample no. E27). They consist mainly of micrite and micritic dolomite. Although most of the intraclasts are barren of fossils, some contain ostracods or a marine fauna.

#### Peloids

These are rounded, spherical to slightly elliptical, moderately to well sorted and range in size from 0.2 mm to 0.7 mm. They consist mainly of micrite and micritic dolomite.

Mapstone (1971), Thusu (1972) and Williamson and Picard (1974) noted that the intraclasts and pellets could form by the erosion of incompletely consolidated sediments



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Fig. 109 Intraclast of ferroan dolomite containing skeletal fragment. The neomorphism of the ferroan dolomite has partially destroyed the skeletal fragments (arrowed). Peel (X100), Blackhall Limestone (Section 42).



Fig.110 Peloids and intraclasts of ferroan dolomite. The fossils are ostracods. The groundmass, the intraclasts and peloids are composed of ferroan dolomite. Peel (X100). Blackhall Limestone (Section 23).

#### 1.1.1.1

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by weak current action or by periodic storm activities. However, Williamson and Picard (1974) did not exclude the neomorphic origin for some intraclasts having gradational boundaries with the groundmass. Anderton (1974) proposed a slight drop in sea level by tectonic, climate, or low tide as the mechanism of the formation of intraclasts and pellets by subareal erosion or dessication of the limestone in the Dalaradian sediments, Scotland.

The intraclasts and pellets have sharp boundaries with the groundmass. It is suggested that these intraclasts and pellets are most likely erosional product of incompletely lithified sediments. It is difficult to speculate about the mechanism of erosion whether by weak current, storm activities or subareal erosion or dessication. However, the erosion by weak current is most likely because of the absence of indication of exposure and because of the low energy environment of the Lower Limestone Group sediments.

#### 3.1b Micrite facies (Facies 2)

The limestone beds of this facies range in thickness from 20 cm to 70 cm. Normally the colour of the rocks of this facies is light-medium grey to dark grey with a Slightly brownish weathering surface. No structures were observed in this facies and the beds have sharp, and Sometimes undulatory or wavy, bases (Fig.111). They are interbedded with shales a few millimetres to 17 cm thick and sometimes this facies is associated with



biomicrite facies. In some sections the micrite facies passes upward gradually into calcereous shale, or into calcareous shale than into siltstones and sandstones (Fig. 113a). The insoluble residue of quartz and clay minerals ranges from 22.7% to 49.6%. The clay minerals are mainly kaolinite. This facies is similar to the biomicrite facies in having a micrite matrix, but it differs in containing fewer fossils, normally not more than 4% by volume. The fauna are crinoids, forams. brachiopods and other unidentified shell fragments (Fig.112).

Heath <u>et al</u>. (1967) described similar facies with a micrite matrix in the Bird Spring Group, and considered the environment to be very quiet with poor circulation when the sediments contain less than 3% organic debris and quiet when it contains 3% to 30% organic debris. They related the percentages of fauna to the control of food availability by water circulation. Laporte (1971) suggested a deep offshore environment for identical facies in the Paleozoic carbonate facies and stated (p. 732) "Still farther seaward, or at least toward the center of basin subsidence, the deep subtidal facies passes into less calcareous, more argillaceous sediments, for as the basin deepens carbonate production declines and finer grained clastics become proportionately more significant."

Some of the studied units show rippled or wavy bases. Asquith (1967) described similar features from the Mifflin Member, and he suggested that the wavy character of the bedding indicates continuous movement of the depositional



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interface by wave or current action, although wave or current action was not strong enough to winnow out or prevent the accumulation of micrite. In some sections of the Lower Limestone Group the micrite facies overlies the biomicrite facies and has a higher insoluble residue. In these sections the micrite facies passes upward into shales or into shales then into siltstones and sandstones. (Fig. 113a). This indicates that the lack of fossils probably results from the high influx of clastic sediment and not from an increase in the depth of water. This probably suggests that this facies deposited in a near shore environment presumably nearer shore than the biomicrite facies (Fig. 113b). This interpretation is supported by the wavy bedding which probably indicates continuous movement of the depositional interface by wave or current action. Therefore, the depositional environment of this facies is most likely a subtidal inner neritic environment in which the water depth was shallower than in the biomicrite facies.

In the Stirling area the base of the Blackhall Limestone is represented by 40-70 cm thick beds. These are dark grey in colour with a slightly brownish weathering surface. Mostly they are nodular and irregular or with wavy bedding surfaces. They differ from the above described facies by the absence of fauna and sharp contact with the overlying shale. Microscopic study indicates that these units are mainly of micrite. No fossils were observed, although at the Weatherlaw Inlier section

unidentified skeletal fragment were observed. Some of the units are dolomitized. The dolomitization is thought to be partially or completely related to adjacent igneous intrusions (see dolomitization section).

Belt <u>et al</u>. (1967) interpreted the non-fossiliferous laminated cementstone facies of the Carboniferous Cementstones of the British Isles and Eastern Canada as the deposits of a marginal marine to restricted marine environment in which the high salinity and temperature resulted in a lack of fauna.

It is suggested that these Lower Limestone Group units either represent a deep offshore, subtidal environment (deeper than biomicrite facies, Fig. 113b) or a high salinity environment (e.g. restricted marine, lacustrine etc.). However, as most of these units are completely un-fossiliferous a deep, offshore marine environment is unlikely. The lack of dolomitisation suggests that a high salinity environment (e.g. restricted marine, lacustrine etc.) is also unlikely. Therefore it is difficult to reach a conclusion about the environment of these units and further work is suggested.

3.1c Dolomitic biomicrite facies (Facies 3).

The colour of the rocks of this facies is medium to dark grey with a slightly brownish to yellowish-brown weathering surface. The thicknesses range from 20 cm to more than 4 m. When they are thick, they consist of units ranging from 6 cm to 40 cm interbedded with shale partings 3-10 cm thick. Fossils up to 1 cm in size were

observed in the field. This facies either forms a bed or part of a bed. When it forms part of a bed, it is separated from other facies (e.g. 1 and 5) by a shale seam 5-10 cm thick. Beds of this facies are massive with sharp bases, and are sometimes slightly rippled. It consists mainly of micrite, micritic ferroan dolomite, siderite and allochems (Fig. 114). The allochems are mainly fossil fragments with subordinate intraclasts and pellets. The identified fauna are crinoids, brachiopods, bivalves, foraminifera, bryozoans, and gastropods which together form more than 10% of the volume of the rocks. Intraclasts and pellets were observed but do not exceed 5% of the rock's volume. The neomorphism of calcite, dolomite and siderite has nearly obliterated the original nature of this facies. The insoluble residue, largely quartz and kaolinite ranges from 2.5% to 23%.

Asquith (1967) noted a similar facies containing a marine fauna identical to the fossils identified within this facies in the Mifflin Member and interpreted the dolomitization as an early diagenetic effect produced by the recrystallisation of the metastable fine grained carbonates such as aragonite and high-magnesium calcite in the presence of a  $Mg^{++}$  rich solution. He considered the environment to be shallow marine with a possible range of depth from 6-10 m. Braun and Friedman (1969) described a mottled dolomitic biomicrite and micrite facies from the Tribes Hill Formation and interpreted them as intertidal on the basis of the presence of tidal



channels. However, they noted that the fauna indicates the nearness to subtidal environment. In the Midland Valley in which the Lower Limestone Group is located, Belt et al. (1967), noted that a large body of saline water developed in the Carboniferous rift valley. They indicated that the Midland Valley basin, which was presumably usually connected with the open sea, suffered periodic isolation or semi-isolation from the sea during the thedeposition of/Cementstone Group (Carboniferous).

The presence of the same fauna in biomicrite facies and dolomitic biomicrite facies indicates that the environment of deposition of the two facies is similar. The dolomitic biomicrite facies differs from biomicrite facies in dolomite content only. The presence of the dolomite most probably indicates an environment of higher salinity than biomicrite facies. The dolomite is most likely formed as a replacement product of metastable carbonate grains by  $Mg^{+2}$  and  $Fe^{+2}$  rich solution (see dolomitisation section).

3.1d Dolomitic micrite facies (Facies 4)

### Subfacies 4a

The colour of the rocks of this facies is bluishdark grey with a slightly brownish to yellowish-brown weathering surface. The thickness usually ranges from 30 cm to 40 cm and although one unit is 70 cm thick, some of the units contain a large amount of carbonaceous material. The beds are massive, with sharp bases and

sometimes are slightly undulous. The insoluble residue, largely quartz and kaolinite, ranges from 14.3%-33.3%. This facies is similar to the dolomitic biomicrite facies but it differs in the amount of fauna (Fig. 115). The salinity of its depositional environment was presumably similar to the dolomitic biomicrite facies. The dolomitisation is considered to be early diagenetic as explained in the dolomitisation section. Therefore the environment is considered to be a saline, low energy, shallow subtidal environment (probably shallower than the dolomitic biomicrite facies).

## Subfacies 4b

In the Kittoch Water section (Section 42, Figure 103), a 70 cm thick unit is exposed which represents the base of the Blackhall Limestone. It is nodular and light grey in colour and contains a large amount of carbonaceous material and faint indications of rootlets. Microscopic study reveals that micrite, micritic ferroan dolomite, siderite and allochems are the main components. The allochems form less than 5% by volume and consist of intraclasts only. Some of the intraclasts contain an Ostracod fauna similar to that identified as Cavallina or Parapachites in ostracod -bearing dolomite facies (Robinson, 1977, personal communication) which indicates hypersaline conditions. No fossils were identified in this unit although shell fragments forming less than 1% by volume are probably ostracods.

Belt et al. (1967) pointed out that the change from

fossiliferous lutite (shales or mudstones) into nonfossiliferous laminated cementstones in the Carboniferous Cementstones of the British Isles and Eastern Canada indicate an increase in salinity with an increase in temperature which resulted in a drop in input. They stated (p. 711) that "These facies were deposited in a marginal marine to restricted marine environment...".

This unit is similar to/ostracod-bearing facies, but it differs in the amount of the ostracods. The ostracod bearing dolomite facies is interpreted as the deposit of a lacustrine to lagoonal environment. Therefore, it is most likely that this unit also represents lacustrine or lagoonal conditions but with an even higher salinity than the ostracod -bearing dolomite facies. In such a high salinity environment a lack of fossils is to be expected.

3.le Ostracod -bearing dolomites (Facies 5)

This facies is found at the base of the Blackhall Limestone in the Campsie area and in the Bridge of Weir section. It consists of several thin units which range in thickness from 16 to 43 cm and are separated by thin shale (3-20 cm). The colour of the units is dark-grey with a yellowish-brown weathering surface. Most of the units contain a large amount of carbonaceous materials. The beds of this facies have sharp bases and are sometimes slightly rippled. When the bedding planes are exposed, they are wavy or irregular. This facies passes upward into shales (5-30 cm thick) and then into biomicrite facies or dolomitic biomicrite facies. In two sections

(Baldow Glen and Shields Burn, Fig. 101) this facies passes upward directly into biomicrite facies without a shale separating the two facies.

The insoluble residue, largely quartz and kaolinite, ranges from 3% to 15.3%. The matrix is composed of microcrystalline calcite, micritic dolomite and coarse grained siderite. The allochems are mainly fossils, intraclasts and pellets. The fossil assemblage is dominated by one type of organism, ostracods (Fig. 116). However calcispheres were identified in some units. The intraclasts and pellets, which have sharp boundaries, are composed of micritic ferroan dolomite similar to that in the matrix. When the rocks have undergone intensive recrystallisation, all the fossils, intraclasts and pellets have been nearly obliterated and only their "ghosts" remain.

Williamson and Picard (1974), reported on a lacustrine environment from the Green River Formation (Eocene) in northeastern Utah and northwestern Colorado. They stated (p.752) that "Lagoons and bays that were protected from the full sweep of the wind were common along the irregular shores of the lake". They added that most ostracod bearing rocks (which are dolomitised) are thought to have accumulated in lagoons. They interpreted the dolomitisation as an early diagenetic process formed by replacement unstable carbonate grains.

The Lower Limestone Group ostracods are represented mainly by <u>Cavallina</u> or <u>Parapachites</u> which indicate hyper-

(X130).

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Fig. 116 Photomicrograph of the ostracod-bearing dolomitic facies. The ostracods are arrowed. (X130). Blackhall Limestone (Section 19).

Fig. 117 Photomicrograph showing sandy micrite rock of the calcareous sandy micrite facies with crinoid fragment. Slide (X100). Shields Bed (Section 19).

Fig. 118 Photomicrograph showing sandy sparite rock of the calcareous sandstone facies with brachiopod spine. Slide (X100), Main Hosie Limestone (Section 16).


saline conditions (Robinson, 1977; personal communication). The presence of calcisphere; also indicates a sheltered restricted environment (cf. Marszalek, 1975). These ostracod -bearing units are overlain directly by 20-30 cm thick units (biomicrite, and dolomitic biomicrite facies) containing a marine fauna which represent the top of the Blackhall Limestone. Therefore the base of the Blackhall Limestone represents a different depositional environment from the top. The former was probably deposited in lacustrine or lagoonal condition. This isolation possibly resulted in an increase in salinity due to evaporation to the limit where penecontemporaneous dolomitisation was possible. This is supported petrographically by the selective dolomitisation of the microcrystalline carbonate grains, the sharp boundaries between the dolomitic intraclasts, pellets and the dolomitised groundmass.

3.1f Calcareous siltstones and sandstones facies (Facies 6)

This is characterised by rough undulatory bedding planes. They have wavy or rippled erosional surfaces 2-3 cm in relief. The thickness ranges from 25 cm - 125 cm and the are normally medium grey in colour with slightly brownish weathering surfaces. There are some indications of rootlets. These appear as carbonaceous streaks 1-2mm in width and up to 2 cm in length slightly perpendicular to the bedding. In the field rocks of this facies look more like sandstones than carbonates. The insoluble residue ranges from 68.9% - 87.3%. A marine fauna with

crinoids, brachiopods and bivalves is present and forms less than 5% by volume of the rock (Fig. 117 ). Silty and sandy quartz is the main constituent for this facies forming more than 80% of the rock volume. The other constituents are micrite and micritic dolomite in the sandy or silty micrite rocks (Fig. 117 ) together with calcite spar in the sandy sparite rocks (Fig. 118).

Williamson and Picard (1974) noted identical facies to the calcareous siltstones and sandstones facies in the Green River Formation and considered them to be a shoreline deposits formed on beach, bars and spits. The abundance of silty and sandy quartz and the sparse fauna, probably indicates that this facies was deposited nearer shore than the other carbonate facies and it is interpreted as the deposits of beaches, bars or spits, the currents in these coastal areas were either strong enough to remove the clay and produce a sandy or silty sparite rock of this facies or insufficiently strong to remove all the clay and thus produce a sandy micrite rock. The sandy or silty micrite rocks of this facies are dolomitised. The micrites are more susceptible to dolomitisation than calcite spar. The crinoids fragments are more susceptible to dolomitisation than brachiopods, bivalves and other unidentified skeletal fragments. It is suggested that this facies were deposited in shallow, nearshore marine environment. Possibly the evaporation in this shallow environment and the extensive precipitation of calcium carbonate increased the amount

of  $Mg^{+2}$  in the marine water or in the interstitial water to the point where penecontemporaneous dolomitisation was possible (see dolomitisation section).

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### 3.2 MINERALOGY OF CALCITE AND THE DIAGENESIS

Three types of carbonate minerals were identified, using staining and x-ray diffraction techniques, these are calcite, dolomite and siderite. The dolomite and siderite will be discussed later on (see section 3.3 and Chapter 4) and only the calcite will be discussed in this section. All of the carbonate minerals have suffered diagenesis.

3.2a Calcite

Five types of calcite were identified; these are:

## 3.2a(i) Equant calcite cement

These crystals are mainly pure, equant, ferroan types. Variations in the iron content were observed and the calcite could thus be classified as strongly ferroan, moderately ferroan, weakly ferroan and nonferroan. These minerals occur in fossil cavities, molds and pores as a cement. Calcite crystals increasing in size away from skeletal walls are recognised (Drusy calcite, Figure 119). Bathurst (1975) noted that the cements are characterised by plane crystals boundaries and enfacial junctions. Plane boundaries and enfacial junctions were observed (Fig. 120 ) but most of the crystals have curved boundaries (Fig. 121 ). These curved boundaries and the local replacement by the calcite crystals of the enclosing fossils suggested that these minerals underwent late diagenetic recrystallisation.

Friedman (1968, p. 19) stated that "If a drusy mosaic is found in marine limestones, these limestones must have



Fig. 119 Drusy calcite. Ferroan calcite cement (subsequently slightly recrystallised) increasing in size away from the skeletal wall. Peel (X130). Main-Hosie Limestone (Section 32).

Fig. 120 Ferroan calcite cement (subsequently slightly recrystallised) inside fossil cavity characterised by enfacial junction (arrowed) and plane crystal boundaries lines. Peel (X320). Main-Hosiz Limestone (Section 32).

Fig. 121 Ferroan calcite cement inside fossil cavity characterised by curved boundaries (C.). Notice the growth of calcite cement away from the shell wall (C.). Veinlet of ferroan calcite cuts across the fossil, and the cement. Peel (X320), Hurlet Limestone (Section 16).

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been subjected to subaerial diagenetic change or to fresh waters in subsurface". On the other hand, Bathurst (1975) indicated that the lithification of carbonate sediments can be produced by the simple redistribution of primary carbonate. He reported that this can be achieved mainly by the compaction of carbonate-bearing clays with the formation of stylolites which act as permeability barriers while the CaCO3 released during the stylolite growth is a major source of late diagenetic cement. He added that pressure-solution and dissolution of aragenite can also be the source for late diagenetic cement. The Lower Limestone Group carbonate facies contain a large amount of insoluble residue and mostly contain a considerable proportion of allochems. Therefore, it is suggested that the equant calcite cement might be produced as a result of compaction in these clay-bearing rocks which leads to the solution of CaCO3 and the growth of clay seams which act as permeability barriers and aid the reprecipi-However, tation of the dissolved CaCO3 as cement. pressure-solution, dissolution of aragonite, subaerial diagenetic change and fresh water in the subsurface could supply a subordinate amount of calcite cement.

Oldershaw and Scoffin (1967) noted that in the Halkin imestone (Upper Visean), N. Wales and the Wenlock imestone (Middle Silurian), England, the cement in the argillaceous limestones is ferroan.whereas that in the non-argillaceous limestones is non-ferroan. They pointed out also, that when the non-argillaceous limestones

are adjacent to shale units the cement is ferroan. They deduced that the iron content of the limestones was derived from the clay minerals in the adjacent, interbedded shale units. Carrol (1958) noted that iron can be carried by clays as essential constituents within the crystal lattice. Microscopic observations indicate that whether the limestones contain a little ( 5%) or a lot ( 30%) of insoluble residue, the calcite cement is ferroan. Therefore, iron content in the ferroan calcite cement probably originated in the adjacent shales.

## 3.2a(ii) Needle-like and Micrite Cement

Needle-like and micrite grains are recognised as a cement (Figures 122, 123). The needle-like grains line fossil cavities whereas the micrite are randomly fill fossil cavities.

Shinn (1969) and Taylor and Illing (1969) pointed out that in the Holocene marine sediments of the Arabian Gulf fibrous acicular crystals are an aragonite cement and the micrite is a high-magnesium calcite cement. However, Ginsburg <u>et al</u>. (1967) noted that aragonite forms a micritic matrix in some sediments whereas the highmagnesium calcite forms a fibrous cement as in the Bermuda reef. Friedman (1968) noted that fresh water micrites (low-magnesian calcite) are deposited either biologically by blue-green algae or higher plants or inorganically in lakes such as Green Lake, New York, or in Lake Constance Germany. He noted that only the fossil could indicate



Fig. 122 Needle-like, non-ferroan, marine calcite cement (Nfc) terminated against equant ferroan calcite. Peel (X320). Hurlet Limestone (Section 39).

Fig. 123 Similar to the above. Peel (X320). Hurlet Limestone (Section 6).

Fig. 124 Neomorphic ferroan calcite characterised by curved boundaries and micrite inclusions (M). The dark patches are micrite, brown silica and other insoluble residue. Peel (X320). Hurlet Limestone (Section 25).

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arine calcite cement uant ferroan calcite. one (Section 39).

one (Section 6).

characterised by curved lusions (M). The dark silica and other 320). Hurlet Limestone



whether micrite is of marine or fresh water origin. Folk (1974) pointed out that two factors might produce different shapes of carbonate spary grains; these are Mg<sup>++</sup> and Na<sup>+</sup> in the pore water and the rate of crystallisation. He noted that dissolved Mg<sup>++</sup> ions, particularly in sea water, selectively poison sidewards growth of calcite preventing the formation of large, equant crystals of calcium carbonate and resulting in the growth of fibrous calcite. Accordingly the highly magnesian calcite is micritic or fibrous with very narrow crystals and the aragonite is fibrous.

The environment in which the carbonate units were deposited is considered to be marine on the basis of fossil content. However, lagoonal or lacustrine environments were recognized. It is suggested that the fibrous sparry crystals and micrite (Fig. 127 ) probably represent aragonite and high-magnesian calcite marine cements respectively following Shinn (1969) and Taylor and Illing (1969). These unstable carbonate minerals later changed during diagenesis to low-magnesian calcites as explained by Friedman (1968) and Neal (1969). Similar marine cementation by aragonite and high-magnesium calcite have been recognized in the Carboniferous sediments of Northumberland (Al-Hashimi, 1972; Leeder, 1975).

3.2a(iii) Neomorphic calcite

Calcite crystals with a size ranging from 0.01 mm to 0.12 mm are common in the groundmass of the carbonate rocks and sometimes partially replace the allochems. These

crystals commonly have curved boundaries and contain micrite and clay inclusions (Fig. 124 ). Following the criteria of Folk (1965) and Bathurst (1975), these crystals are considered to be neomorphic calcite formed by the recrystallisation of micrite and into microspar forming the groundmass. The composition of this microspar is mainly ferroan, although non-ferroan types were observed. The analysis of insoluble residue in the studied carbonate units shows that a positive relationship between the insoluble residue and the amount of iron, the insoluble residue in rocks with highly ferroan neomorphic calcite being more than 30% while those with non-ferroan neomorphic calcite contain less than 5%. However, as indicated previously that there is no relationship between the insoluble residue and the amount of iron in the calcite cement. In some samples (Figure 125) which contain a low proportion of insoluble residue (e.g. FlO, Hurlet Limestone, 4.6%).the groundmass is composed mainly of euhedral neomorphic sparry grains. In places when those non-ferroan neomorphic grains have replaced the fossils, only islands (Fig. 126) of ferroan calcite (fossil cavity cement) are preserved surrounded by the non-ferroan neomorphic grains. Therefore the variation in the iron content is probably related to clay content within the insoluble residue. The role of clay minerals as a source of iron is explained by Carrol (1958) as discussed above.



Ilcite (Nfc) without Irk patches are ferroan Froan calcite cement. Insolube residue. Stone (Section 1).

lcite (NFC) replacing ferroan calcite (FC) within fossil cavities nonferroan calcite. stone (Section 1).

te (FC) and micrite (M) ty of brachiopod spine. of neomorphic calcite from 11. Peel (X320), Main-40).



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## 3.2a(iv) Microcrystalline Calcite

These are very small calcite grains and are normally less than 4 micron. They are found mainly in the groundmass, but sometimes fill fossil cavities. Purdy (1963) noted that the lime mud could be produced by four processes: (1) bacterial; (2) derived; (3) physiochemical; (4) skeletal disintegration.

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It is difficult to speculate which one of the above origin is the main contribution to the microcrystalline grains.

### 3.2a(v) Silty Size Calcite

These are non-ferroan clean and elongate grains ranging in size from 0.02 mm to 0.13 mm (Figure 128). Although these grains were observed in most of the rocks they only constitute a very low percentage. Lindholm (1969) reported similar grains in the Onondaga Limestone (Middle Devonian), New York and indicated that they are disintegration products of skeletal material. He noted also that the disintegration resulted either from abrasion and physical breakdown of organic debris in agitated waters or from boring, mastication and ingestion by organisms. Such an interpretation is accepted for the silty calcite in the Lower Limestone Group since it is clear, elongated, badly sorted and Without microcrystalline grain inclusions.



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Fig. 128 Photomicrograph showing the silt-sized calcite (arrowed). Dark patches are micrite, brown silica and other insoluble residue. Peel (X130). Hurlet Limestone (Section 32).

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#### 3.2b Diagenesis

This includes processes which affected the rocks either immediately after the time of deposition or later on. Schmidt (1965, p.128) stated that the term "early diagenesis refers to diagenetic changes which were influenced by the physico-chemical environment and the biologic activity at the site of deposition..." and "Late diagenesis refers to diagenetic changes not influenced by the depositional environment or by the physical-chemical conditions of the supernatant water". An attempt is made here to classify the diagenetic processes in the Lower Limestone Group carbonate units into early or late.

## 3.2b(i) Neomorphism of fossils

Some fossils, like crinoids, have partially disintegrated (Fig. 129), in other words they have undergone the degrading neomorphism of Folk (1965). This process started from scattered points within the fossils and spread until entire fossils were degraded. (Fig. 130).

## 3.2b(ii) Fossil cavity fillings

Ferroan calcite is the dominant cement inside these cavities. Sometimes fibrous, non-ferroan calcite lines the fossil cavities (the first-generation of cement) and is terminated against blocky ferroan calcite considered to be a second generation of cement. However two generations of cement, both of which are non-ferroan are recognised. Micrite, probably from high magnesium calcite, sometimes



exists as a cementing material also. However, some fossil cavities are filled with a large amount of micrite which sometimes has been neomorphosed into spary grains (microspar of Folk, 1965). Sometimes the neomorphic calcite or the precipitated calcite replacing the shell wall of the fossils extends outside into the groundmass.

### 3,2b(iii) Pore and mold fillings

The pores within the rocks are filled mainly by ferroan equant calcite crystals (Figs.131,132). No fibrous non-ferroan calcite was recognized. The absence of the first generation cement may be due to three things (a) the percentage of pores is less than the fossils The percentages of first generation cement content. (originally aragonite) is very low within the fossils cavities compared with the second generation. This is either because any aragonite cement that was precipitated on the outside of shell fragments underwent early diagenetic leaching or recrystallised to coarse equant grains. (b) Some pores resulted from desiccation or deformation of the partially lithified sediments after the initial (c) the microenvironment inside the fossil cementation. cavities was responsible for the first generation of cement.

#### 3.2b(iv) Grumeleuse texture

Grumeleuse texture is common in some rocks (Fig. 133). This texture/characterised by clotted micrite associated with brownish chert, bounded by neomorphic sparry calcite grains.



Fig. 131 Ferroan calcite cement filling mould in Mid-Hosi<sup>e</sup> Limestone (Section 32). Peel (X130).

Fig.132 Ferroan calcite cement filling pore. Peel (X130). Mid Hosic Limestone (Section 32).

Fig.133 Grum@leu% texture. The dark patches are micrite, brown silica and other insoluble residue. The white patches are neomorphic, ferroan calcite. Peel (X130). Hurlet Limestone (Section 25).

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illing mould in Mid-32). Peel (X130).

filling pore. Peel tone (Section 32).

dark patches are micrite, insoluble residue. The orphic, ferroan calcite. mestone (Section 25).



Evamy (1967) noted that grumeleuse texture could be formed as a result of dedolomitisation. He pointed out that late diagenetic dolomite rhombohedra are characterised by central inclusions of mud. On calcitization the dolomite rhombohedra, and in particular the clear rim, alter to a relatively coarsely crystalline mosaic of calcite which resembles cement and leads to the isolation of the central inclusion of the dolomite grains to form a clotted texture known as grumeleuse texture. Baush (1968) noted that clay minerals control the crystal size of neomorphic calcite in limestone; small grain sizes are produced when there is a large amount of insoluble residue and large crystal sizes result when there is only a little. Marschner (1968) described the same relationship and suggested that it may be due to the presence of "protective" envelopes of clay minerals during recrystallisation. However Bathurst (1975, p.512) stated "... that structure grumeleuse evolved by the growth of calcite crystals throughout the mass of an original homogeneous micrite, and the gradual differentiation, thereby, of a more coarsely crystalline, continuous matrix separating discrete residual clotts of microcrystalline (micritic) calcite".

No indication of dedolomitisation was observed within the carbonate of the Lower Limestone Group so such an origin for the grumeleuse texture can be dismissed.

Microscopic observations indicate that in rocks containing little insoluble residue no indication of grumeleuse texture was observed. Instead the rock consists

of mesh-like coarse, neomorphic calcite crystals (Fig. 25 & 126 ), perhaps with a few aggregations of microcrystalline grains but not enough to merit the term grumeleusetexture. In rocks containing a high concentration of insoluble residue, the grumeleuse texture is prominant (Fig.123, p.206) and it is suggested that it probably originated by the recrystallisation of the micritic matrix from scattered points within the groundmass, similar to the processes of "porphyroid" neomorphism described by Folk (1965). When this process is halted by other constituents such as chert and/or clay minerals which probably forms barriers to the growing neomorphic spar, the grumeleuse texture results.

## 3.2b(v) Compaction

Some shells are broken and cemented by ferroan calcite (Figs.134,135). Zankel(1969) considered the crushed shells as a criterion for compaction and he related these features to the effect of insoluble residue preventing the early lithification by cementation or recrystallisation. He noted also that there is no grain compaction when the clay mineral content is less than 2%.

In the carbonate units studied, although the insoluble residue in some units is low (about 2%), most of the units which show compactional effects have more than 30% insoluble residue. The large concentrations of insoluble residue probably prevented early lithification and compaction due to the overburden probably crushed or deformed the organic debris.

of mesh-like coarse, neomorphic calcite crystals (Fig. 125 & 126 ), perhaps with a few aggregations of microcrystalline grains but not enough to merit the term grumeleusetexture. In rocks containing a high concentration of insoluble residue, the grumeleuse texture is prominant (Fig.123, p.206) and it is suggested that it probably originated by the recrystallisation of the micritic matrix from scattered points within the groundmass, similar to the processes of "porphyroid" neomorphism described by Folk (1965). When this process is halted by other constituents such as chert and/or clay minerals which probably forms barriers to the growing neomorphic spar, the grumeleuse texture results.

### 3.2b(v) Compaction

Some shells are broken and cemented by ferroan calcite (Figs.134,135). Zankel(1969) considered the crushed shells as a criterion for compaction and he related these features to the effect of insoluble residue preventing the early lithification by cementation or recrystallisation. He noted also that there is no grain compaction when the clay mineral content is less than 2%.

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Figs. 134 and 135

Photomicrograph showing broken skeletal fragment cemented by ferroan calcite. Peel (X320)

Fig. 134 Shields Bed (Section 27) Fig. 135 Hurlet Limestone (Section 16)

Fig. 136 Veinlet of ferroan calcite cut across brachiopod fragments. Peel (X130) Hurlet Limestone (Section 16).

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### 3.2b(vi) Veinlets

Many carbonate rocks of the Lower Limestone Group contain veinlets of ferroan calcite (Fig.136). These veinlets are due to fracturing either early, during lithification of the rocks, or, later during tectonism. As some of the veinlets cut across the fossils, an origin due to tectonism is more likely.

## 3.2b(vii) Silica diagenesis

Two types of silica were identified within most of the carbonate members of the Lower Limestone Group; brown chert and colourless chert. They are found within nondolomitic, partially and completely dolomitic rocks.

## 1 - Brown chert

This consists of fine, microcrystalline quarts grains identified as chert. The brown chert is found mainly within the groundmass. Some of the fossils (e.g. foramonifera (Fig. 137) and crinoids) and some of the intraclasts and pellets have also been replaced by brown chert.

Silica can have either an organic or inorganic origin (Blatt et al., 1972; Pettijohn, 1975). Organic silica forms when organisms such as radiolarians, diatoms or sponges, extract silica from marine water which is subsequently dissolved and reprecipitated as nodular masses which replace the matrix. Inorganic silica results from direct precipitation of amorphous silica from marine or



Fig.137 Photomicrograph showing foraminifera (F) fragment in which the shell wall is composed of brown chert. Slide (X130) Top of the Blackhall Limestone (Section 19).



Fig. 138 Photomicrograph showing chert (Ch) which has replaced the fossil. Slide (X130). Hurlet Limestone (Section 16).

lake waters, or by devitrification of volcanic glass. Peterson and Borch (1965) concluded that in the lakes associated with the Coorong Lagoon of South Australia, the transported dissolved silica in the lakes originated from detrital quartz and clays. They added that during active photosynthesis by plants and evaporation, the brine approaches its maximum concentration, and may evaporate or sink into the underlying mud. The pH is very low within the mud, because of the available organic material, therefore the silica will be precipitated when it approaches this Eugster (1967) concluded a similar origin for zone. silica in the Lake Magadia, Kenya. This lake is essentially of sodium carbonate brine. Evaporation raises the pH within this lake up to 10, causes dissolution of The dilution of volcanic rocks and clastic fragments. the lake water by fluvial inflow then lowers the pH and hydrated amorphous silicate is precipitated, and is later altered to chert during diagenesis.

Namy (1974) pointed out that organic matter plays an important role in precipitation of silica by reducing the solubility of silica locally and causes its precipitation.

In the studied area, no radiolarians, diatoms or Sponges were identified, so an organic origin for the silica is unlikely. Volcanic activity has been reported in the north of the area studied (Craig, 1965), and the Lower Limestone Basin is considered to consist or connected to semi-connected small basins (Section 3.3). The preliminary interpretation of the origin of the brown

silica is that, evaporation caused an increase in the pH and the silica was released by the dissolution of detrital quartz, clay minerals and/or volcanic glass. The dilution of these small basins by fresh water inflows lowered the pH and the silica was precipitated. Alternatively, the dissolved silica was carried into the Lower Limestone Group basin from the surrounding area and if the pH was low, the silica was deposited as soon as entered the basin.

## 2 - Colourless chert

This is composed of fine microcrystalline grains identified as chert usually filling fossil cavities. Some fossils, such as brachiopods (Fig. 138) and crinoids, are also replaced by chert. In crinoids fragment, it is found that dolomite crystals are concentrated next to those of silica (Fig. 139). Jacka (1974) pointed out that when length-slow chalcedony is replacing high magnesian calcite, such as fusulinds, bryozoans and echinoderms, dolomite will form in the area of silici-He considered these features as indicators of fication. an early replacement of silica before the stabilization of the carbonate minerals. Because of the very small size of the siliceous grains, it is difficult to say whether they are represented a length-slow chalcedony or other types of silica. However, it is most likely that the formation of the dolomite crystals next to the silica suggests their connection with the silica replacement, since the unit in which these dolomite crystals were



found is a non-dolomitic unit. Petrographic observation reveals that patches of ferroan calcite and dolomite are present inside the chert (Fig. 140). This indicates that the silica replacement took place after the formation of ferroan calcite and dolomite.

The colourless silica is probably secondary and either originated from the dissolution of the detrital quartz and clays or precipitated from dissolved transported silica.

3.2b(viii) Diagenetic sequence

Based on the petrographic observations of thin sections and peels, including cross-cutting relationships, the following diagenetic sequence has been constructed (see fig. 141 ).

- Deposition of calcium carbonate mud, allochems, and the influx of terrigeneous materials (Fig.141A).
- Replacement of microcrystalline carbonate grains by microcrystalline dolomite (Fig.141B )
- 3. Formation of the microcrystalline siderite and/or pyrite and brown chert (Fig. 141C). In some rocks (e.g. Blackhall Limestone, Kittock Water section, is Fig. 104 ) in which the groundmass/composed mainly of siderite, there are sharp boundaries with the dolomitic intraclasts and pellets.
- Deposition of sparry calcite cement inside fossil cavities, moulds and fractures (Fig. 141D).



5. Replacement of colourless chert (Fig. 141D).

- 6. Neomorphism of microcrystalline calcite and siderite to large sparry grains (Fig. 141E). This is indicated by the presence of microcrystalline dolomite inside the neomorphic sparry calcite and siderite grains.
- 7. Recrystallisation of microcrystalline dolomite grains and its replacement to sparry neomorphic or directly precipitated ferroan calcite cement (Fig. 141F).

The proposed time relationship of the diagenetic changes in the carbonate units of the Lower Limestone Group are summarised in Figures 142 and 143.



# Figure 143

Diagram summarizing the time relationship of the diagenetic changes in the studied carbonate sediments

	Features	Early diagenetic	Late diagenetic
1.	Microcrystalline carbonate grains		
2.	Allochems		
3.	Microcrystalline dolomite		
4.	Microcrystalline siderite	?	
5.	Pyrite	?	
6.	Equant calcite cement	?	
7.	Needle-like and micrite		
8.	Neomorphism of micro- crystalline calcite and siderite		
9.	Silty calcite		
10	Compaction -		
11	Grumeleous texture		
12	Fossil neomorphism		
13	Veinlet		
14	Recrystallisation of dolomite		
15	Brown Chert		
16	Colourless Chert		
## 3.3 DOLOMITISATION

# 3.3a Early diagenetic dolomitisation

Some of the carbonate units of the Lower Limestone Group have been partially replaced by ferroan dolomite. In most of the units the dolomitisation is not laterally persistent. According to their fossil content the dolomitic bearing rocks can be divided into two types.

- Dolomitic rocks that are either unfossiliferous or contain a marine fauna.
- 2 Ostracod-bearing dolomitic rocks.
- 3.3a(i) Dolomitic rocks that are either unfossiliferous or contain a marine fauna.

This type of rock either contains marine fossils such as crinoids, brachiopods, bivalves, foraminifera or is completely barren. The groundmass is composed mainly of microcrystalline calcite and micritic dolomite. However, some units consist of entirely micritic dolomite.Pellets and intraclasts, of ferroan dolomicrite composition or which have been partially dolomitised and have sharp boundaries with the groundmass, are present within some rocks. Sparry ferroan calcite filling fossil cavities, moulds and pores is also found.

Friedman and Sanders (1967), in reviewing and discussing the dolomitisation problem, pointed out that there are two main processes of dolomitisation; early diagenetic (syngenetic) and late diagenetic (diagenetic). The syngenetic origin is the replacement of unstable carbonate

grains by Mg<sup>++</sup> either by capillary concentration or refluxion processes, penecontemperanous with the deposition of the carbonate, whereas the diagenetic process is the replacement of stable carbonate minerals by subsurface brines, However, the role of insoluble residue (Kahle, 1965; Williamson and Picard, 1974) and the effect of the underlying sediments (Kahle, 1965; Leeder, 1975) are also reported as controls of dolomitisation. The origin of the dolomitisation in the Lower Limestone Group will be discussed in the light of these processes.

# I - Stratigraphic relationship

The character of the underlying sequences probably effects dolomitisation in the following way:

A - The relation of the carbonate units to the mouth bar

At the river section southeast of Calderwood Castle (Section 39 and 39A, Figure 104),the carbonate units which are dolomitised overlie sandstone sequences considered to represent the lateral and axial parts of the mouth bar (See chapter 2). At Kittoch Water (Section 42, Fig.104), and in the Gill Burn Section (Section 44, Figure 104), the Carbonate units are not dolomitised and overlie sequences which represent a distal part of the mouth bar. Even after delta abandonment the depth of the water would have varied across the mouth-bar as shown in Figure 8 (Section 1.2d(i)). Therefore, any evaporation would probably cause a variation in the salinity and consequently a higher concentration of the Mg<sup>++</sup> would have been expected above

the axial and lateral part of the mouth bar than the distal part. However, surface currents would tend to mix in low salinity water from the ocean while the high saline water above the axial or lateral parts of the mouth bar would sink and flow seawards, preventing the formation of a permanent high salinity area above the axial or lateral part of the mouth bar. It is suggested that the brine sinking above the lateral and axial part of the mouth bar probably penetrated the underlying, unlithified carbonate and produced dolomitisation. This idea is not applicable to all the dolomitised units, because not every dolomitised unit is related to the axial and lateral part of the mouth-bar.

# B - The effect of the underlying sandstones

Kahle (1965) pointed out that the presence of porose and permeable sandstones could, if sufficient magnesium could be derived from the shales within the basin, give rise to localized or widespread dolomitisation. The theory of the control of dolomitisation by the underlying permeable sandstone sequences is not applicable to the Lower Limestone Group. This is because many carbonate units overlie sandstone sequences and are not dolomitised while some carbonate units overlying shale units are dolomitised (Figs.101-105).

# C - The effect of the underlying shales Oldershaw and Scoffin (1967) indicated that the limestones adjacent to the shale units in the Halkin

Limestone (N. Wales) and the Wenlock Limestone (England) contain ferroan calcite cement. They considered that the source of the  $Fe^{+2}$  for the calcite cement is the clay minerals within the shale units. Leeder (1975) considered that the origin of the dolomitisation in the Lower Border Group (Touraisian), Northumberland, is due to the released  $Mg^{+2}$  and  $Fe^{+2}$  from the clay minerals in the adjacent shales. This idea is not applicable to the origin of the dolomitisation in the Lower Limestone Group because some carbonate units overlie shales and are not dolomitised.

II - The role of insoluble residue.

The insoluble residue of 63 samples were determined following the method of Bisque and Lemish (1959) (See appendix III). It consists mainly of quartz and clay minerals (mainly kaolinite).

Some authors (e.g. Kahle, 1965; Williamson and Picard, 1974) suggested that at least some or even all the magnesium needed for dolomitisation can be provided by clay minerals. Schmidt(1964) pointed out that dolomitisation affected first the rocks containing a large proportion of clay, even prior to the replacement of the aragonitic fauna. Zen (1959) indicated that clay will enter into chemical reactions which produce dolomite thus: Clay (chlorite or Montmorillonite)+ calcite + carbon

dioxide = dolomite + kaolinite + quartz + water.

Kahle (1965) reported that the clay could contribute to dolomitisation either by (a) ion exchange, in which ions such as  $Na^+$  (in highly mineralized sodium chloride

brines) or Ca<sup>++</sup> (produced by the dissolution of carbonate rocks) replace Mg<sup>++</sup> adsorbed onto clay minerals, liberating it into the solution or by (b) Nucleation and crystal growth processes, in which clay minerals may serve as some sort of catalyst or as centres of nucleation for crystals of dolomite.

There is no relationship between the amount of insoluble residue and dolomite content within the carbonate units of the Lower Limestone Group. The Hurlet Limestone for example has 30-50% insoluble residue in the Campsie area (Section 16, Fig.101) and is not dolomitised, whereas in the south of the area studied it has 2-5% insoluble residue and is dolomitised. However, in some sections it contains a lot of insoluble residue, about 60%, and is dolomitised (Figure 103, Section 36). This lack of relationship between the insoluble residue and dolomitisation also applies to the other studied carbonate units. Kaolinite is present in both dolomitised and non-dolomitised units which also confirms that Zen's (1959) reaction for the formation of kaolinite and dolomite from clay and calcite is not applicable here. Therefore, clay minerals are not likely to be the source of the Mg<sup>++</sup> for the formation of dolomite in the Lower Limestone Group. Lumsden (1974), in his detailed study on the effect of insoluble residue on dolomitisation in (Callville Formation the (Pennsylvanian) and Pakoon Formation (Lower Permain), Southern Nevada, reported that there is neither direct nor inverse relationship between the insoluble residue and the

dolomite content.

The previous discussion indicates that neither the underlying sediments, nor the insoluble residue can help in solving the problem of the origin of the dolomitisation. So it is necessary to look for another origin for the dolomitisation in the Lower Limestone Group.

The current theories for the early diagenetic origin of dolomite involve formation in either supratidal flats (Shinnet al., 1965 and Illing et al., 1965) or hypersaline lagoon (Adams and Rhodes, 1960). In supratidal areas intense evaporation during subareal exposure of the flats may lead to the formation of a brine rich in Mg<sup>++</sup>. The precipitation of gypsum as well as calcium carbonate increases the Mg/Ca ratio in this pore water brine to the point where dolomitisation becomes possible by the replacement of calcium carbonate. The absence of stromatolites and mudcracks and also the diversity of the fauna in the Lower Limestone Group indicates that here a supratidal environment is not likely as the origin of the dolomitisation. The hypersaline lagoon theory indicates that evaporation will produce a dense brine which passes down dip through the underlying sediments resulting in their dolomitisation by the replacement of unstable carbonate grains. The restricted lagoon theory for the origin of the dolomitisation can again be ruled out due to the presence of the diverse marine fauna.

Schmidt (1965) pointed out that the order of decreasing susceptibility of calcium carbonate to alteration into

dolomite (penecontemperaneous dolomitisation), is 1) matrix, 2) aragonite bioclasts, intraclasts, pellets and Ooids, 3) magnesium calcitic bioclasts. Petrographic observations do show that the order of susceptibility to dolomitisation in the Lower Limestone Group is matrix > pellets and intraclast > fossils and sparry calcite grains which corresponds in general outline to Schmidt's (1965) order. In some thin sections (e.g. F52, Figures 144,145) the fossils have suffered selective dolomitisation with some of the fauna being completely dolomitised while the other fossils are not affected. The recrystallisation of the dolomite (a late diagenetic process) destroys the fossil morphology so that their identification becomes impossible. The late diagenetic processes affected all the fossil types also (irrespective of the original composition) which makes the recognition of the susceptibility to dolomitisation of the various fossil types difficult.

Microscopic observations indicate that the main dolomitisation is earlier than late diagenetic recrystallisation and the formation of veinlets as indicated by the following:

- 1 The sharp boundaries between the veinlets of sparry ferroan calcite and the ferroan dolomitic groundmass (Figs.146,147).
- 2 Micritic ferroan dolomite grains identified as inclusions within the ferroan neomorphic sparry calcite grains (Fig. 148), which indicate that the dolomitisation is earlier than neomorphism.



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Fig.144 Dolomitic biomicrite in which some fossils have been selectively dolomitised (arrowed). Peel (X100) Base of the Blackhall Limestone (Section 5).

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Fig. 146 Veinlet of ferroan calcite (V) which cuts across ferroan dolomitic (fd) rock. Notice the sharp boundary between the calcitic veinlet and the dolomitic groundmass. Peel (X130), Main Hosif Limestone (Section 14).

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Fig. 147 Similar to the above, but the micritic ferroan dolomite has replaced the sparry ferroan calcite of the veinlet. Peel (X320), Main Hosit Limestone (Section 14).









on. Photomicrograph tised fossil (probably the adjacent brachiopod . The black patches be brachiopod and its d opaque grains. The oundmass are ferroan Second-Hosie Limestone

lcite (V) which cuts tic (fd) rock. Notice tween the calcitic veinlet undmass. Peel (X130), Main on 14).

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Fig. 148 Photomicrograph showing ferroan dolomite (fd) inside neomorphic ferroan calcite (fc). The dark patches of the groundmass are ferroan dolomite. Peel (X320). Blackhall Limestone (Section 7).

Little is known about the formation of penecontemoraneous dolomite in subtidal environments, which are characterised by a diverse of marine fauna. However, Behrens and Land (1972) noted penecontemporaneous dolomite forming subtidally at a depth of 7-12 metres, in the Holocene Sediments of Baffin Bay, Texas. They noted that the dolomite is the result of precipitation by water having a Mg/Ca ratio near that of normal sea water and stated (p. 160) "... the dolomite was the original precipitate or that it formed by replacement of a texturally equivalent precipitated precursor such as a Mg-calcite mud by reaction with slightly hypersaline interstitial fluids". In ancient sediments Asquith (1967) in/Mifflin Member and Lumsden et al. (1973) in/Spring-Callville Group and Pakoon Formation (Pennsylvanian-Lower Permian), Nevada related the dolomitisation to an evaporation process in shallow water conditions which caused hypersalinity and consequently primary dolomite formed by replacement of original calcium carbonate in the carbonate facies of a subtidal environment which contains a diverse marine fauna.

Goodlet (1957) noted that the southern sector of the studied area is not simple tectonically but was affected by certain faults during the deposition of the Lower Limestone Group. Greensmith (1965,1966) pointed out that the basin of the Calciferous Sandstone Series (directly underlying the Lower Limestone Group) is characterised by barriers, local subaerial volcanoes, submarine lavas and possibly sand bars. He noted also that behind these

barriers, shallow water lagoonal environments developed in which, ostracod -shale, micrite, biomicrite and oomicrite facies were formed. However Belt <u>et al</u>. (1967) pointed out that large bodies of saline water were developed in Carboniferous rift valleys (Scottish Midland Valley and North Ireland) which were presumably connected with the sea although characterised by isolated or semiisolated basinsduring the deposition of the Cementstone Group (Carboniferous).

Badiozamani (1973) interpreted the dolomitisation in the Ordovician Mifflin Member, which was deposited in a broad shallow-marine environment in Wisconsin, as an early diagenetic process due to the mixing of meteoric ground water with sea water. When fresh meteoric ground water is mixed with between 5 to 30% sea water the high Mg/Ca ratio of sea water is maintained but the solution becomes undersaturated with calcite and supersaturated with Calcite can therefore be replaced respect to dolomite. by dolomite without the need for extensive evaporation Meyers and Lohmann (1978) indicated that of sea water. the microdolomite inclusions in the syntaxial cements of the Mississippian Limestones of New Mexico indicated that these cements were originally Mg calcite, which had behaved as closed or semi-closed systems during their transformation into low magnesian calcite. They suggested that these cements were precipitated in the zone of mixing between meteoric and marine phreatic ground waters. They added that the heterogeneous distribution of microdolomite

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reflects the heterogeneous Mg distribution of the precursor Mg calcite and they related the variation of Mg within the Mg calcite as due to fluctuations in the  $Mg^{++}/Ca^{++}$  ratios in the precipitational pore waters at uniform temperatures.

It is suggested that the basin of deposition of the Lower Limestone Group was probably not a simple basin, but characterised by isolated basins (see ostracod-bearing dolomite) or semi-isolated basins in which the dolomitic micrite and biomicrite facies were deposited. Within these sectors of the basin evaporation would have increased the Mg/Ca ratio in the sea water or interstitial water to the point where the dolomitisation became kinetically possible. However, when the Mg/Ca ratio was not high enough, no dolomite would form, and non-dolomitic micrite or biomicrite was deposited. Alternatively the dolomitisation could be induced by mixing of meteoric ground water with sea water (cf. Badiozamani, 1973; and Meyers and Lohmann, 1978). Such mixing is likely in the ground water underneath advancing delta lobe. This is indicated by the lateral and vertical variation of the dolomite within the carbonate units of the Lower Limestone Group which have the same lithology and fossils content (Figs. 101 - 105).

# 3.3a(ii) Ostracod-bearing dolomitic rocks

This type of dolomitisation is found only at the base of the Blackhall Limestone in the Campsie area (Fig. 101) and Bridge of Weir section (Section 32, Fig. 103). The top of the limestone is mostly non-dolomitised and has a

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diverse marine fauna. Clough <u>et al</u>. (1925) noted similar sections of the Blackhall Limestone with the same characteristics around the Campsie area.

The discussion of the Ostracod-bearing dolomitic facies reveals that these rocks are composed mainly of micritic dolomite and allochems represented by fossils, dolomitic intraclasts and peloids. The fossils are mainly ostracods in which the identified genera are Cavellina or Paraparchites which indicates hypersaline condition. Calcispheres were identified but are not abundant (Fig. 149) and consist of a rind composed of sparry calcite and holes filled by micritic ferroan dolomite, and sparry ferroan They indicate sheltered shallow-protected waters calcite. with a restricted or semi-restricted circulation. It is suggested that the base of the Blackhall Limestone was probably formed in a restricted or isolated environment (probably a lagoonal or lacustrine environment) in which penecontemporaneous dolomitisation took place due to This is indicated by the fauna and is evaporation. supported petrographically by the fact that the micrite is more dolomitised than the sparry calcite cement, the fauna and the similarity in composition in spite of the sharp boundaries between the pellets, intraclasts and the groundmass (Fig. 150). The presence of microcrystalline calcite in the groundmass and within the fossil cavities, either randomly distributed or as laminae alternating with ferroan dolomite (Fig. 151), indicates that part of the microcrystalline calcium carbonate changed to lowmagnesian calcite before the dolomitisation was completed.



Fig.149 Calcisphere in which the rind is composed of calcite, and the cavity filled by ferroan calcite (fc) and micritic ferroan dolomite (fd). The dark patches in the groundmass are ferroan dolomite. Peel (X320). Base of the Blackhall Limestone (Section 23).

Fig.150 Intraclasts and peloid composed of ferroan dolomite similar to the groundmass ans with sharp boundaries. Fossils are ostracods. Peel (X100), Base of the Blackhall Limestone (Section 19).

Fig. 151 Laminae of nonferroan calcite (nfc) and ferroan dolomite (fd) inside the ostracod cavity. The dolomite inside the cavity and the groundmass has undergone recrystallisation. Peel, (X320). Base of the Blackhall Limestone (Section 23). the rind is composed of ty filled by ferroan calcite roan dolomite (fd). The roundmass are ferroan O). Base of the Blackhall ).

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n calcite (nfc) and ferroan the ostracod cavity. The cavity and the groundmass has isation. Peel, (X320). Base sestone (Section 23).

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#### 3.3b Late diagenetic

Petrographic evidence indicates that late diagenetic processes are extensive and affect all the dolomite facies in the Lower Limestone Group as explained by the following points

- 1 Dolomite crystals in the completely dolomitised groundmass replace the sparry ferroan calcite filling some veinlets (the latest calcite) (Fig.147, p.228).
- 2 Sparry and micritic crystals of dolomite replace both sparry calcite crystals in the fossil cavities, mould and pores and neomorphic calcite (Figs. 152, 153).
- 3 Dolomite replaces all the fossil types, irrespective of their original composition.
- 4 The random distribution of iron within the sparry dolomitic grains (Fig.154 ) probably indicates that when the micritic dolomite grains recrystallised into coarse sparry grains the original patchy distribution of the iron was still retained even within a single crystal. However, all the allochems were destroyed and only "ghosts" of fossils or pellets and intraclasts are preserved.

The above points indicate that the late diagenetic processes include the recrystallisation of the original dolomitic mud which is considered to have had an early diagenetic origin. Such a late recrystallisation makes the dolomite appear, at first sight, as if it formed by a late stage replacement rather than during early diagenesis, as more detailed work shows.



Fig. 153 Sparry ferroan dolomite (fd) replaced sparry ferroan calc ite (fc) inside the pores. The dark patches of the groundmass are ferroan dolomite. Peel (X130). Hurlet Limestone (Section 2).

Fig. 154 Random distribution of the iron (I) within the sparry dolomitic grains as indicated by staining. Peel (X320), Hurlet Limestone (Section 38).



hedral sparry ferroan ng neomorphic ferroan e bivalve cavity. The groundmass are ferroan 30), Hurlet Limestone

ite (fd) replaced sparry c) inside the pores. the groundmass are ferroan 0). Hurlet Limestone

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3.3c The effect of igneous intrusion on dolomitisation

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All the carbonate units that are associated with igneous intrusions are dolomitised. Most of the dolomite is a ferroan type, but sometimes it is nonferroan (e.g. the top of the Blackhall Limestones in Sections 26, 27, Figure 102). Both types of dolomite are replacive as indicated by the replacement by dolomite crystals of skeletal fragments, sparry calcite cement etc.

Field observation indicated that the sediment associated with igneous intrusions underwent the following types of alteration:

- The shales adjacent to intrusions are lighter in colour (Sauchi Craig South Section, Figure 102) and are harder than the shales away from the intrusion.
  The coals adjacent to intrusions are hard and coked.
  Pyrite is common with the sediments associated with igneous intrusions.
  - 4 Breccias composed of igneous rocks formed during intrusion were found associated with the carbonate units (e.g. Sauchi Craig South Section, Fig. 102).

Dunham (1959) indicated that Fe, Mg and SiO<sub>2</sub> were transported from igneous rocks such as dolerite into some of the adjacent Carboniferous sediments in Yorkshire. He noted also that the veins of ankerite in the sediments associated with the igneous intrusions were probably formed by the Fe and Mg leached from the intrusions. There is no doubt that the dolomite in the carbonate units

associated with the igneous intrusions in the Lower Limestone Group partly or completely originated from igneous intrusions. The iron and/or magnesium could be removed directly from intrusions as indicated by Dunham (1959) or liberated by the reaction of clay minerals, siderite and original dolomite in the sediments with the heated solutions derived from the intrusions.

#### 3.3d Origin of Iron

As discussed earlier, the dolomite which is a ferroan type, is thought to be early diagenetic in origin. Apart from the dolomite associated with igneous intrusions, little is known about the early diagenetic origin of ferroan dolomite. Hough (1958), in his description of the Precambrian banded iron formations in the Lake Superior district, indicated that rapid decay of source rocks in  $\alpha$ humid climate, could supply adequate quantities of ferrous iron to the basin of deposition. Braun and Friedman (1969) noted a ferroan dolomite within a supratidal dolomitic facies, in the Tribes Hill Formation. They pointed out that the iron entered late in the depositional or early in the diagenetic environment. Greensmith (1965) suggested a primary or very early diagenetic origin for the ferroan dolomitisation in the Calciferous Sandstone Series (Carboniferous) of the Midland Valley of Scotland. He noted also that the abnormal proportion of the iron was related to the erosion of an iron-rich provence or to pene-contemporaneous volcanicity. Mapstone (1971) considered that the ferroan dolomitisation in the Upper

Limestone Group was an early diagenetic process which took place soon after deposition and before compaction as a result of reaction with the sea water trapped at the time of deposition. He pointed out that the postdepositional changes of the trapped sea waters caused the introduction of  $Fe^{++}$  which entered into the dolomitisation before the sediments were compacted.

Apart from minor volcanicity in the north of the area studied (Craig, 1965, p.360), no pene-contemporaneous volcanism has been reported during the deposition of the Lower Limestone Group. However, the presence of coal seams in the Lower Limestone Group sediments indicates that the climate was humid. Therefore, the high proportion of iron was probably due to the erosion of an iron-rich provenance in humid climate and its incorporation by diagenetic processes within the sediments before the compaction and stabilisation of the carbonate grains.

# 3.4 CLAY MINERALS

Clay minerals were studied in the carbonate and shale units, in an attempt to find if there is any environmental and/or diagenetic relationship between them and the different lithological units of the Lower Limestone Group. The methods used for clay preparation and the obtaining of oriented clay samples are described in Appendix IV.

The result revealed that kaolinite is the main type of clay mineral in marine units (carbonates and shales) and non-marine units (sandstones and shales). However, peaks corresponding to illite and mixed-layer clays were identified in some marine and non-marine units, but they are very small and rare compared to the kaolinite peaks.

Keller (1970) reported that, clay minerals are generated in large part by weathering of alumins silicate He added that clay could be produced by direct rocks. crystallisation from solution, crystallisation from a colloidal gel and by diagenesis from other clay minerals. Zen (1959) pointed out that kaolinite could be produced together with dolomite by the reaction of montmorillonite and chlorite with calcite. Parham and Austin (1967, p. 868) stated that "... the presence of abundant kaolinite suggests that it formed in a warm moist climate under acid condition". Grim (1953) noted that the high pH value necessary for calcite to form causes kaolinite to be unstable and montmorillonite, illite and chlorite formed instead. Keller (1970) thought that kaolinite is unstable in the marine environment and changes to illite

and mica although Grim and Loughnan (1962) state that "the kaolinite appears to be unaffected by the marine environment". Keller added that the kaolinite dissolution is retarded if it is accompanied by other clays minerals which give silica rapidly than kaolinite and therefore approach equilibrium conditions in the marine environment.

Kaolinite is present in the dolomitic and nondolomitic units of the Lower Limestone Group which indicates that it is unlikely to have originated by the reaction of montmorillonite and chlorite with calcite. The occurrence of kaolinite in marine and non marine units indicates a common origin. Most probably the kaolinite originated as a weathering product in the source area under humid weathering conditions. The small concentrations of illite and mixed-layer clays probably originated from the diagenetic alteration of the detrital kaolinite and/or marine montmorillinite. However, as the illite and mixedlayer clays occur in both marine and non-marine units suggests that they are probably entirely the result of the alteration of kaolinite.

# 3.5 DESCRIPTION OF THE CARBONATE UNITS

In this section, the lithology, texture and the vertical and lateral variation within each carbonate unit will be discussed.

# 3.5a Hurlet Limestone

This unit marks the base of the Lower Limestone Group. Its total thickness varies between 1.5 m to more than 5 metres. Internally, it consists of thin limestone units 5-60 cm thick, sometimes separated by calcareous shales ranging in thickness from 5 cm to 25 cm.

The allochems consist of fossils, pellets and intraclasts. The identified fauna are crinoids, brachiopods, bivalves, gasteropods, bryozoans and foraminifra. A lateral and vertical variation in the percentage of fauna and, to a lesser extent, the type of fossils is recognised. The pellets and intraclasts are found in one section only (Section 6, Figure 105). The pellets are spherical to rounded and range in size from 0.6 mm to 0.9 mm whereas the intraclasts are angular and range in size from 1.1 mm to 1.6 mm. The groundmass is composed mainly of micrite, sparry calcite cement fills the fossil cavities, pores and moulds. Siderite is common in some sections whereas pyrite was noticed in one section. Brownish and colourless chert is common in the groundmass and in the fossil cavities. The percentage of insoluble residue varies and consists of clay minerals, mostly kaolinite and quartz.

In Corrieburn (Fig. 101, Section 16) a vertical section

was taken within the Hurlet Limestone (Fig.113a) to show if there is any vertical variation. This shows that the base is represented by the biomicrite facies and the top is represented by the micrite facies. The insoluble residue changed from about 30% at the base to about 50% at the top. These variations probably indicate that the depositional environment of the Hurlet Limestone changed from a subtidal, outer neritic environment to a subtidal inner neritic with a greater terragineous influx. The change from subtidal outer neritic to inner neritic, probably indicates the beginning of the Hurlet sea regression, accompanied by the beginning of delta progradation with its resulting increase in terrigeneous sediment input and adverse effect on carbonate secreting This is confirmed in the field in that the organisms. biomicrite facies passes upwards into a micrite facies, then into calcareous shales and finally into siltstones and sandstones.

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In the Campsie area, the Hurlet Limestone, changes from biomicrite or micrite at the east and west of the area to dolomitic calcareous siltstones facies in the middle of the area (Fig. 101). This variation probably indicates a change in the environment of the Hurlet Limestone from subtidal inner and outer neritic to nearer shore represented by coastal environments such as beach, bars or spits in which the calcareous siltstone facies was deposited.

The insoluble residue varies from 2.5% to 69.7% (not including the calcareous siltstone facies). The low values were obtained in the south of the studied area (Fig. 155) where four analysed samples have insoluble residues of 2.5% (Sample F47) 7.8%, 4.6% (F.46), (F10) and 15.4% (F15). These values are much lower than those obtained in the north and middle of the area (Fig. 155) which probably indicates that these samples were deposited in an offshore environment away from any terrigenous influx, in contrast to those from the middle or north of the area which contain a high value indicating nearness to the shore.

Some units are dolomitised. The micritic matrix is more susceptible to dolomitisation than the other constituents. The dolomitisation is considered to have been controlled by the depositional environment (see Section 3.3). Therefore these dolomitised units were probably formed in highly saline semi-isolated basins of deposition.

In conclusion the Hurlet Limestone shows a range of variation in lithology, as indicated by the presence of different facies. The recognised facies are: biomicrite (low energy, outer neritic subtidal); micrite (subtidal, low energy, inner neritic); dolomitic biomicrite (highly saline, low energy) and dolomitic calcareous siltstones (coastal deposits). The distribution of facies in the Hurlet Limestone is presented in Figure 156.









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3.5b Shields Bed

This is the second marine horizon in the sequence. Its thickness ranges from 50 cm to 80 cm. It is exposed at three localities only; Shields Burn (Fig. 101, Section 19), Old Sauchie Burn (Fig. 102, Section 27) and the Weatherlaw Inlier (Fig. 102, Section 28). In Shields Burn (Campsie area) it is represented by the dolomitic sandy micrite facies with a 70.7% insoluble residue. The organic constituents, forming less than 4% of the rock volume, include crinoids, brachiopods and unidentified shell fragments. In Old Sauchie Burn (Stirling area) this unit is represented by a biomicrite facies with 4.9% insoluble residue. The skeletal fragments forming about 25% of the rock, consists mainly of crinoids, bivalves, brachiopods, foraminifera, gastropods and other unidentified fragments. In the Weatherlaw Inlier, the Shields Bed is represented by the micrite facies with a marine fauna.

Although the Shield Bed is only exposed in three localities, there is a lithological difference between these three exposures. The three localities (Fig. 157 ) are represented by calcareous sandstone, biomicrite and micrite facies which probably represent a variation in the environment of the Shield Bed from coastal deposits to a subtidal, low energy environment.

3.5c Blackhall Limestone

This unit ranges in thickness from 1.3 m to 4.5 m. Internally, it consists of thin beds (16 cm to 80 cm),









sometimes separated by shales ranging in thickness from 3 cm to 80 cm. It differs from the other units in having a fauna which is different at the base of the unit from the top. This subdivision was noted by Clough <u>et al</u>. (1925) around the Campsie area. However, it is not persistent throughout the studied area and has only been found in the Campsie area and the Bridge of Weir sections.

In the Campsie area, the Blackhall Limestone can be divided into two subunits. The basal one consists of yellowish-brown, ostracod-bearing dolomite which forms several bands ranging in thicknesses from 16 cm to 43 cm separated by thin shale units (3-20 cm). The groundmass is composed mainly of microcrystalline dolomite, calcite (micritic) and siderite. The dolomite which is ferroan is sometimes neomorphosed into sparry grains. Sparry ferroan calcite is also found in the fossil cavities, pores and the moulds. Brownish and colourless chert is also found. The insoluble residue ranges from 3% - 15.3% (Fig. 158) and the clay consists mainly of kaolinite.

The allochems consist of fossils, pellets and intraclasts. The fossils are mainly ostracods, however, calcispheres were identified in some sections. The ostracods are represented by <u>Cavellina</u> or <u>Paraparchites</u> which indicate hypersaline conditions (Robinson, personal communications, 1977). The pellets are spherical to rounded and range in size from 0.4 mm to 1.0 mm. The intraclasts are large, badly sorted and range in size from 0.8 mm to 2.4 mm.

The top unit ranges in thickness from 20 cm to 50 cm and consists mainly of micrite and siderite. Ferroan dolomite was identified in one section but is not abundant. Ferroan sparry calcite fills the fossil cavities, pores and moulds. The identified fossils are crinoids, brachiopods, foraminifera and ostracods which together form 7-15% of the rock. Intraclasts (2.2 mm to 2.8 mm in diameter) and pellets (0.7 mm to 0.8 mm) were recognised also. Biomicrite and dolomitic biomicrite form the main facies of these units.

The faunal differences between the two subdivisions are not found to the northeast and south of the Campsie area. In the Stirling area the Blackhall Limestone is represented by two subunits, separated by shale. The basal subunit is not fossiliferous whereas the top is nodular and contains marine fossils forming from 5-20% of To the south of the Campsie area (Fig.104). the rock. two sections are exposed in which the Blackhall is nodular and forms two subdivisions also. In one section (Section No. 41) both of the subunits contain marine fossils, whereas in the other (Section 42) the top contains marine fossils whereas the base is unfossiliferous except for the presence of ostracod fragments which form less than 1% In the south of the area studied (Fig.105), of the rock. the Blackhall Limestone occurs either in one, two or several bands all containing marine fossils, separated by thin units of shale. The insoluble residue is not usually more than 11%, but one unit contains 31.9%. Most

of the bands are dolomitised. The origin of the dolomitisation is considered to be controlled by the depositional environment (see Section 3.3).

The above discussion shows that there is a lithological variation which probably reflects a change in depositional environment during the deposition of the Blackhall Lime-The facies recognised in this unit within the stone. studied area are: ostracod bearing facies (lacustrine or lagoonal), biomicrite (low energy, outer neritic, subtidal), micrite (low energy, inner neritic subtidal), dolomitic biomicrite or micrite (highly saline, low energy, subtidal environment). The distribution of facies and the insoluble residue values in the Blackhall Limestone is shown in Figures 159-161. It is suggested that the base of the Blackhall Limestone was deposited in lacustrine or lagoonal environment in the Campsie area, south of Campsie (Section 42) and around the Bridge of Weir section. In the south of the studied area (Fig. 105) it was deposited in a normal marine environment. Transgression of the Blackhall sea then followed in the north and marine units represent the top of Blackhall Limestone were deposited above the lacustrine or lagoonal units.

# 3.5d Hosie Limestone units

There are between three and four of these units in the studied area. The facies distribution and the environment are discussed below:

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Fig. 159 Map showing the distribution of the insoluble residue in the top of the Blackhall Limestone





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## A - Main Hosie Limestone

It ranges in thickness from 30 cm to 1.7 m and the colour is dark-grey with a yellowish-brown weathering surface. It consists mainly of micrite, micritic dolomite and allochems. The allochems are represented mainly by fossils, intraclasts and pellets. The identified fauna are brachiopods, crinoids, forams, gastropods and other shell debris which make up to 40% of the rock volume. The intraclasts and pellets are subordinate and not more than 5% of the rock volume. The insoluble residue, mainly quartz and kaolinite, ranges from 6-20.5%.

Dolomitic biomicrite, biomicrite and calcareous sandstones are the main facies within this unit. The environment probably represent, shallow, low energy subtidal (biomicrite), highly saline (dolomitic biomicrite) and coastal deposits (calcareous sandstones). The distribution of the facies within the Main Hosit Limestone is presented in Figure 162.

#### B - Mid-Hosie Limestone

The thickness of this unit ranges from 45 cm to 50 cm. It is similar to the main Hosie Limestone in colour, composition and fossil content. The environment of deposition was probably shallow, low energy, subtidal (biomicrite facies) and sometimes highly saline (dolomitic biomicrite). The distribution of facies in the Mid-Hosie Limestone is presented in Figure 163.

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# C - Second Hosie Limestone

It is dark grey in colour with a yellowish-brown weathering surface and the thickness ranges from 25 cm to The groundmass consists of micrite and micritic 50 cm. Quartz, chert and siderite are found in minor dolomite. quantities and sparry ferroan calcite fills the fossil cavities, pores and moulds. The identified fauna are brachiopods, crinoids, forams, gastropods and other shell debris which ranges from 14-22%. Angular large intraclasts and rounded pellets were recognised in some sections. Only the dolomitic biomicrite facies was recognised within this unit which probably indicates a highly saline, subtidal, low energy, inner neritic environment. The distribution of facies in the second Hosie Limestone is presented in Figure 164.

# D - Top Hosie Limestone

This marks the top of the Lower Limestone Group. It is bluish-dark grey in colour with a brownish weathering surface and it ranges in thickness from 30 cm to 40 cm.

The matrix consists of micrite, micritic dolomite and siderite. The insoluble residue ranges from 17.2% to 33.3%. The fauna is mainly of brachiopods, and bivalves and other shell fragments which do not make up more than 5% of the rock. However, sometimes when the unit has undergone intensive diagenetic dolomitization, only the "ghost" of the fossil is present. This unit is represented by one type of facies; dolomitic micrite



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## 4.1 INTRODUCTION

Ironstone bands and nodules, with a brownish weathering surface, are common within the shales of the Lower Limestone Group sediments. The shales in which these bands and nodules are found are of dark colour and highly fissile. The bands and nodules are distributed in the shelf-prodelta sediments of progradational deltas and in the marine and lacustrine sediments of interdistributary bay environment. However, some of the fresh water interdistributary sediments also contain bands and nodules. The distribution of bands and nodules in the Lower Limestone Group sediments are presented in Figures 9-14.

#### 4.2 IRONSTONE NODULES

Ironstone nodules are common within the Lower Limestone Group, and range in size from 1.5 cm to 50 cm. Most of the nodules are ellipsoidal with long axes parallel to the bedding. However, spherical, irregular and triangular nodules were also observed.

The ellipsoidal nodules mostly range in size (represented by the long diameter) from 7 cm to 25 cm and sometimes reach up to 50 cm. The spherical nodules are smaller and mostly range in size from 1.5 cm to 3 cm, but sometimes reach 8 cm. The triangular and irregular nodules range in size from 5 cm to 18 cm. In some of the nodules crinoids and brachiopods together with other skeletal fragments were identified, although others were barren. Many of the nodules show septarian structures.

Most of the nodules have a "swelling" on the upper and lower surfaces. Bowes (1973), described some of the nodules in Corrie Burn (Section 16, Fig. 9) as having a local concavity or "dimple". The "dimples" were observed in the swollen part of most of the nodules together with other irregularities. However, some of the swollen parts are smooth. All the swollen nodules have a brecciated centre.

Two types of nodules were observed; these are:

# 1 - Non-zoned nodules

These nodules are ellipsoidal, triangular or irregular (Figs. 166 and 167). No "swelling" was observed on the upper surface. The nodules are composed mainly of siderite and sometimes micritic calcite was identified. The identified fossils are crinoids, brachiopods and skeletal fragments. In some sections the non-ellipsoidal nodules coalesced into bands.

## 2 - Zoned nodules

The zoning (Figure 168) takes the form of variation in mineralogy between the centre, middle and the rind of the nodules. Samples were taken from the centre, middle and the rind of many nodules and examined by x-ray diffraction and stained thin sections. This indicated that the centres of the nodules are mainly of micritic calcite with subordinate siderite and micritic ferroan dolomite; the middle is composed mainly of micritic calcite and siderite and the rind consists of siderite



Fig. 166 Ellipsoidal, non-zoned nodule. The white patches are marine fossils, mainly crinoids.

Fig. 167 Triangular nodule. These nodules are mostly coalesced into bands.

Fig. 168 Zoned nodule in which the centre (C) is composed mainly of calcite, the middle (M) of calcite and siderite and the rind (R) of siderite.

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only. Quartz and kaolinite were identified by x-ray diffraction in all the zones.

Girin (1970) described the same features from Middle Jurrassic sediments in the High Caucasus and noted that organic and carbonic acids are released by the destruction of organic matter in sediments. These acids leach out cations from the clay and dissolve dispersed carbonates. As a result the pH of the pore solution increases and a weakly alkaline reducing environment is produced in which  $Ca^{+2}$ , Mg<sup>+2</sup> and Mn<sup>+2</sup> can be transported by diffusion (so calcite will be precipitated) but resulting in the very limited mobility of  $Fe^{+2}$  (because  $Fe^{2+}$  is not mobile in alkaline solution). When the supply of readily leached cations is exhausted the pH will fall due to the continuous production of acid and under such conditions Fe<sup>+2</sup> will then be leached and siderite formed. Walton (1972) noted that siderite could precipitate in more acidic solutions if the concentration of the total iron increases. However, Curtis (1967) indicated that the siderite formed in a neutral to alkaline environment of pH 7-8.

The siderite nodules in the Lower Limestone Group probably started to grow in an alkaline reducing environment where  $Ca^{2+}$  and  $Mg^{2+}$  were available. These elements could then have been leached out and precipitated as calcite and dolomite, while iron underwent reduction from the ferric to ferrous state. However, if only some of the iron was transported at this time, a little siderite would have been precipitated with the calcite, a stage

represented by the formation of the centre of the nodules. The concentration of reduced ferrous iron would then increase and be leached out at the same time as  $Ca^{2+}$ ,  $Mg^{2+}$ and  $Mn^{2+}$  with the precipitation of siderite at the same time as calcite leading to the formation of the middle zone. The leaching of ferrous iron would continue, while the other cations ( $Ca^{2+}$  and  $Mg^{2+}$ ) became exhausted until only the siderite was formed, a stage represented the formation of the rind, the outer zone.

The cores of the nodules are usually spherical or only slightly ellipsoidal, whereas the outer surfaces are usually ellipsoidal. Spherical siderite nodules are also found, but these are usually small compared with the ellipsoidal types. The spherical nodules and the spherical to slightly ellipsoidal central parts of the nodules probably formed early in porous uncompacted sediments. Raiswell (1971, p.165) described the Cambrian and Liassic concretions of South Wales and Dorset and stated that "within the same lithology early concretions that formed in porous, uncompacted sediments are more spherical than the later concretions which developed in compacted anisotropic sediments ... ". The ellipsoidal shape of the rind, indicates its formation in compacted anisotropic sediments in which the solutions needed for the rind formation flow along, rather than normal to the bedding planes. This is supported by the lack of septarian structures within the rinds, as discussed later on.

## Brecciation of the nodules

Some of the cores in zoned and non-zoned nodules are brecciated (Figs.169-171). The brecciated fragments are composed mainly of micritic calcite as revealed by staining and x-ray diffraction. However, sideritic breccias were observed in some nodules. When calcitic and sideritic breccia occurs in the same nodule, the calcitic breccia tends to occupy the centre, whereas the sideritic fragments are located near the rind. All the brecciated nodules are accompanied by septarian structures which are filled by ferroan calcite. Ferroan calcite with pure, equant crystals also fills fossil cavities in the nodules and bands. The origin of this ferroan calcite is discussed in the carbonate section (Chapter 3).

Zanger et al. (1969) considered that the brecciation within concretions in the Fayetteville Black Shale (Mississippi) of Arkansas was largely due to gas pressure produced by the bacterial reduction of coprolites which form the core. Raisewell (1971) pointed out that septarian structures are produced by the dehydration of the initially plastic concretion centres and are limited to concretions forming early in porous, water-laden sediments. This interpretation is applicable to the Lower Limestone Group nodules described here.

If the cores are fragmented by contraction cracking and the rinds are slightly coherent, the rind will subsequently collapse under the increasing weight of the overlying sediments. The formation of contraction cracks



Fig. 169 Nodule showing that the core was brecciated into a few uniform large pieces and on irregular upper surface was produced. Notice also the septarian structures.

Fig. 170 Nodule in which the core was fragmented into large and small pieces and a swollen upper surface was produced.

Fig. 171 Same nodule as above showing the "dimple" (d) and "swollen" (s) features.

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is thought to be the main reason for brecciation coupled with the reduction of volume due to losses of water and the crushing of the rind by subsequent pressure. If the cracks extended more toward the rind (which probably indicates that part of the rind was formed in slightly uncompacted sediments), the rind will also be brecciated (Fig. 169). If the core was fragmented into a few relatively large pieces or into both large and small pieces (Fig. 170), a swollen or irregular surface, possibly with a dimple, would be produced during the subsequent collapse under pressure. On the contrary, if the core was brecciated into small and uniform pieces (Fig. 172), or if fragmentation occurred in concretions of a completely sideritic composition which grew in relatively compacted sediments no "swellings" would be produced (Fig. 173) during collapses, although irregular rough surfaces could Some of the brecciated nodules have a smooth be formed. swollen surface without irregularities (Fig. 174). These nodules are spherical to slightly ellipsoidal and were most likely formed in a porous, isotropic medium, probably at shallow depth. Therefore, the rind would not collapse when the core was brecciated because of the insignificant load of the overlying sediments. The stages of brecciation of the nodules are summarized in Figure 175.

In the Corrie Burn Section (Section 16, Figure 9) the ironstone nodules within the shale of the post-Blackhall deltaic sediments decrease in size upward. The sediments of this section represent a deltaic progradation in which the shale of the prodelta coarsens in grain size upward



Fig. 172 Nodule showing the core was brecciated into relatively uniform pieces, consequently no "swellings" were produced, but rough irregular surface was formed instead.

Fig. 173 Nodule of uniform, sideritic brecciated core in which no "swellings" were produced.

Fig. 174 Slightly spherical nodule of brecciated core and smooth swollen surface.

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core was brecciated into pieces, consequently no oduced, but rough irregular instead.

sideritic brecciated core .ngs" were produced.

nodule of brecciated core surface.



266 "dimple" swelling Brecciation into different sizes core Redistribution of the fragments after compression ind Septarian Breciation into equal structures large pieces A. Formation of the spherical to slightly ellipsoidal nodules in uncompacted sediments. "Dimples" and nodules in uncompacted sediments. "swellings" are produced. Rough, irregular surface Rind Core Redistribution of the fragments Septarian structures after compression B. Formation of the cores of the nodules in relatively compacted sediment in which ellipsoidal cores are produced. Brecciation of the cores and then readjust-ment of the fragments. No "swelling" formed but instead rough irregular surface. - pressure of the overlying sediments Fig. 175 Diagrams summarising the stages of brecciation of the nodules. . .. in.

and changes gradually into interbedded siltstones and shales of the delta front and then into sandstones of the distributary mouth-bar. The ironstones are large when enclosed in the fine grained prodelta clayey shale and become smaller upward as the shale becomes coarser. Moore (1966) noted that the prodelta sediments contain more clay than the delta front sediments. As the clay is considered to be the source of iron for formation of the siderite, the size of the ironstone nodules may be controlled by the proportion of clay in the sediments.

## 4.3 IRONSTONE BANDS

The ironstone bands range in thickness from 3 cm to 25 cm. The composition of these bands, as revealed by staining and x-ray diffraction, is mainly siderite. However, micritic calcite was identified in some bands but is not abundant. Some of the bands are fossiliferous and include crinoids, brachiopods, ostracods, bivalves and unidentified skeletal fragments. However, some of the bands contain mainly an ostracod fauna of <u>Cavellina</u> or <u>Paraparchites</u> which indicates hypersaline conditions (see Section 3.1e). Microscopic observations indicate that the siderite crystals range from 4 to 80  $\mu$  in size. In some bands siderite has replaced the fossils (Figure 176).

Taylor and Spears (1967) considered that the siderite bands of the Yorkshire Coal Measures (Carboniferous) England, were originally limestone bands. They pointed out that when the rate of sedimentation is slow in a reducing environment, the sulphate reducing bacteria cause



Fig. 176 Photomicrograph showing a crinoid fragment, within an ironstone band, that has been partially replaced by siderite. Peel (X130) Ironstone band (section 1).

the formation of pyrite and later ankerite. On the contrary, a high rate of sedimentation results in a lower concentration of sulphur species in the pore solution and siderite is formed by the replacement of calcite grains. Curtis and Spears (1968, p.264) stated that "it is doubtful, however, if siderite need always be a replacement of a primary carbonate". They noted that the ankeritesiderite bands discussed by Taylor and Spears (1967) are perhaps the only examples of siderite bands replacing original limestone in the Yorkshire Coal Measures (Carboniferous) England. Curtis and Spears (1968) went on to suggest that the siderite of Carboniferous and Jurassic Ores of Yorkshire, England could form as a primary diagenetic precipitate formed under the same physicochemical conditions below the sediments/water interface as those described above. Diagenetic precipitation is considered by many authors to be the origin of siderite bands (Hough, 1958; Hallam, 1966; Frank, 1969; and Sellwood, 1971). Curtis (1967) noted that siderite forms in a reducing environment with Eh ranges from -0.2 to -0.3 V. However, Curtis and Spears (1968) stated that very low Eh is not reached in the bottom waters of marine or estuarine environment although it can be reached in the pore water of sediments, although, Skopintsev et al. (1966) reported a low value of Eh (-0.172 ) in the stagnant bottom waters of the Black Sea. Curtis (1967) showed that the siderite will only grow where there is a high partial pressure of  $CO_2$ , higher than that found in normal sea water saturated

with CaCO<sub>3</sub>. He concluded that siderite concretions must therefore develop within the sediments.

The marine fossils content (e.g. crinoids, brachiopods ...) of some of the ironstone bands in the Lower Limestone Group indicates that the water was not stagnant. The absence of benthonic fauna in most of the bands could be related to the existence of a muddy substrate and not to the lowering of Eh (cf. Curtis and Spears, 1968). Therefore sideritic bands with marine and hypersaline fossils and the non-fossiliferous siderite bands were probably formed as early diagenetic precipitates in a reducing environment below the sediment/water interface, where there was a high rate of sedimentation and in which a very low Eh and a high partial pressure of  $CO_2$  could be The physicochemical conditions could not be reached. reached on the sea floor. The replacement features probably indicate that the siderite underwent subsequent neomorphism (see section 3.2b).

Shrinkage cracks were observed within the ironstone bands in one section (Kittoch Water section). Frank (1969) suggested a dewatering of the diagenetically precipitated colloidal iron carbonate as an origin for similar shrinkage crack in Kiowa Formation (Early Cretaceous), North-Central Kansas. Shrinkages represented by septarian structures are found in the nodules of the Lower Limestone Group and interpreted as dehydration of initially plastic concretions as discussed earlier. It is difficult to speculate about the origin of these cracks

in the ironstone bands, whether they were formed by subaerial exposure or by a dewatering process. However, no indication of exposure was observed within the sediments of the Lower Limestone Group, which suggests that dewatering is the more likely process.

The iron which formed the siderite could have been released into solution from the lattices of clay minerals, from ferric oxides or from hydrated ferric oxide coating detrital clay minerals (Curtis, 1967; Frank, 1969). The iron underwent redistribution in the ferrous state beneath the sediment/water interface and was precipitated as siderite under moderate to strongly reducing conditions.

## 4.4 FORMATION OF NODULES AND BANDS

Whether the siderite forms nodules or bands probably depends on the depth in which the nodules or bands were formed and on the rate of deposition. Raiswell (1971) described the Cambrian concretions of South Wales and pointed out that at a small depth and with continuous deposition the sediments compacted rapidly and as the concretion grew, and the laminae, which are initially parallel in the loose uncompacted host sediments were preserved by cementation and developed first at the central zone of the concretion (Fig. 177). He stated (p.154) "As compaction progressed laminae in the sediment were deformed around the relatively rigid, cemented concretion and further concretionary growth preserved these deformed laminae in subsequent layers of the concretion". He concluded that these concretions are characterised by



septarian structures and of deformed laminations and suggested a 10 m maximum depth for their formation. Raiswell (1971) also indicated that at greater depth, the concretions develop parallel laminae because of the slow rate of compaction and suggested a 100 metres maximum depth for these concretions which are characterised by the lack of septarian structures. He added that the rate of deposition could control the type of laminae (whether parallel or deformed) and the formation of bands and stated that the Liassic concretions in Dorset, formed at small depth (about 5 m) with a low rate of deposition and therefore that these concretions developed parallel laminae because of the absence of compaction and are characterised by septarian structures. They may finally coalesce into bands.

The discussion of Raiswell (1971) reveals that concretions with deformed laminae and septarian structures can form at small depths (10 m) and with rapid compaction, whereas concretions with parallel laminae can form at greater depth (100 m). Septarian structures are found in nodules formed at small depth (5 m) in sediments which had a slow rate of deposition. The discussion of the zoned nodules in the Lower Limestone Group (Section 4.1) reveals that the centre, which is spherical, formed in a porous, isotropic medium whereas the rind is ellipsoidal and formed in less porous and slightly compacted sediments. It is suggested that at a small depth with continuous deposition, the Lower Limestone Group sediments compacted rapidly and

concretions formed instead of bands. This was probably because progressive rapid compaction reduced the porosity of the sediments which in turn affected the movement of the pore water (which carried the necessary elements for the formation of sideritic nodules) eventually halting the growth of the nodules and preventing their coalition into bands. At a greater depth the compaction was slow, whether the deposition was continuous or not, and bands formed because of the continuous and uniformly flow of pore water along the bedding planes. However, some of the bands contain shrinkage cracks and some of the nodules which coalesced into bands are slightly ellipsoidal. The rate of deposition is considered to control the formation of bands (cf. Raiswell, 1971). At a small depth and with a slow rate of deposition the sediments are porous and isotropic. In such conditions the growing concretions probably coalesced into bands. This is unlikely because these concretions should be spherical in such conditions. It is suggested that some of the bands which are formed by the coalition of slightly ellipsoidal nodules and the shrinkage cracks bearing nodules were formed in slightly compacted sediments, a stage corresponding to the formation of the rind in the zoned nodules. Therefore the concretions are probably early diagenetic and formed within the top of 10 m of continuous sediments deposition and rapid compaction, whereas the bands are either early diagenetic and formed at the same depth of the nodules in sediments of slight rate of deposition or late diagenetic and formed at greater depth.

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CHAPTER 5

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GEOCHEMISTRY

# 5.1 INTRODUCTION

Nine trace elements were analysed by XRF Spectrometry in 59 carbonate rocks and 21 shales. Three major elements were analysed by Atomic Absorption Spectrometry, two of which were also analysed by XRF. The carbonate samples were grouped into three petrographic types, limestones (non-dolomitic), partially dolomitic limestones and dolostones, to find if there are any corresponding variation in the trace and major elements. However, the lateral and vertical variations within the carbonate units in the studied area, were considered in the selection of the samples. For shales, the selection of the samples was random and includes two main groups, marine and non-marine.

The trace elements analysed by XRF Spectrometry in the carbonates and shales were; Sr, Rb, Ba, Ti, Pb, Zn, Ni, Cu and Co. The major elements analysed by Atomic Absorption Spectrometry were Ca, Mg and Mn in the carbonate rocks. Fe and Mn were analysed in carbonates and shales by XRF Spectrometry.

#### 5.2 TRACE ELEMENTS

#### Strontium

The data reveals that the strontium concentration in the limestones ranges from 211 to 1907 with an average of 638 ppm. In partially dolomitic rocks the concentration of this element ranges from 203-1614 ppm with an average of 631 ppm. The dolostones have a strontium content much lower than the limestones and dolomitic limestones ranging

from 53 to 1254 ppm with an average of 225 ppm (Table 7).

Veizer and Demovic (1973) related the concentration of Sr to the original mineralogy; aragonite contains much more strontium than high or low magnesium calcite. They pointed out that early diagenetic dolomite contains a higher concentration of Sr than late diagenetic dolomite, although Weber (1964) concluded that the concentration of Strontium is higher in secondary than in the primary dolostones. He considered that the secondary dolostones formed by the replacement of aragonite or calcite and primary dolostones by the direct precipitation of dolomite or penecontemporaneous replacement. However, he did not exclude the effect of adsorption on clay minerals and added that much Sr is lost during the conversion of aragonite into calcite or dolomite.

The plotting of insoluble residue against the Sr content in the three carbonate types (Fig. 178), indicates that there is no positive relationship, which suggests that the Sr is mainly concentrated within the carbonate fractions.

The average Sr value within the analysed limestones and dolomitic limestones is higher than in Weber's (1964) (Table 8) secondary and primary dolostones and is within the range of the highest average values of Veizer and Demovic (1973) (600-700 ppm) which are found in sediments of an originally aragonitic composition. Therefore the high values of Sr in the Lower Limestone Group carbonates could reflect an original aragonite composition.

The dolqstone rocks have the lowest Sr content within the carbonates of the Lower Limestone Group, although one

No. of Section Sr ppm Mn ppm -Mn ppm Co ppm Ba ppm Rb ppm mdd mdd hpm bba I.R. Ca % T102 -Unit Samp Cu P.e Mg 2 uz NI (XRF) (Ab) TABLE 7a 23.70 25.40 56.50 31.30 195 734 6772 3681 0.22 0.2 0.46 0.093 5159 5890 2907 1449 4420 6340 2370 1450 1.99 1.09 21.7 1.75 0.66 24.10 1.36 0.6 10.4 0.58 0.53 27.35 12 50 59 40 31 44 59 43 24 30 48 47 20 13 31 71 35 27 45 26 329 517 41 41 42 42 61 99 E20 E22 E25 E30 BK (B) BK(T) BK(T) Hu MH & MDH MH 116 657 894 2754 1259 F51 GB2 GB4 GB6 GB7 GE28 GS61 R4 R9 S26 S35 F42 GS63 R12 S35 F42 GS63 R12 GS65  $\begin{array}{c} 6.5\\ 2.67\\ 30.50\\ 40.86\\ 49.56\\ 28.21\\ 68.70\\ 39.98\\ 28.10\\ 3.45\\ 20.51\\ 4.92\\ 33.78\\ 31.90\\ 7.78\\ 48.01\\ 48.90\\ 5.88\\ 48.42 \end{array}$ 22 0 103 91 18 48 220 69 34 22 7 14 2 16 102 137 98 56 9 62 1907 82 1242 1020 559 562 535 541 779 443 329 807 387 484 437 323 727 211 295 501 1907 648  $\begin{array}{r} 24\,90\\ 1220\\ 1230\\ 810\\ 470\\ 440\\ 16\,80\\ 270\\ 470\\ 14\,70\\ 14\,70\\ 14\,70\\ 1550\\ 2560\\ 33\,50\\ 61\,50 \end{array}$ 7 39 16 16 16 16 21 32 32 32 32 27 5 6 21 32 39 16 20 225 222 30 28 33 56 222 28 14 28 18 37 16 35 43 20 25 53 0.018 0.095 0.2 0.11 0.17 0.27 0.30 0.02 0.11 0.30 0.02 0.12 0.28 0.36 0.36 0.36 0.32 46 12 38 43 27 31 85 44 16 31 10 30 24 9 40 31 61 69 0 38 6 7 15 6 12 13 8 7 9 9 5 4 9 16 33 12 17 2 4 MH Hu Hu Hu Hu BK(T) Hu BK(T) 2849 1129 139 195 293 208 476 193 92 163 150 109 375 697 288 121 238 14 19 9 14 29 35 16 11 0 18 2 1 42 37 53 2 28 1222 897 352 517 2040 458 593 1465 8955 4702 BK(T) MDH MH SD BK(B) BK(T) Hu Hu BK(T) MDH MH 1620 3151 3580 9968 1008 1463 1173 945 970 1330 690 2278 2685 2606 15.2 0.88 23.2 12 19 638 0.16 57 29 1005 Average 36 51 TABLE 7b 3180 9070  $\begin{array}{c} 0.27\\ 0.23\\ 0.02\\ 0.02\\ 0.04\\ 0.07\\ 0.05\\ 0.05\\ 0.02\\ 0.03\\ 0.04\\ 0.11\\ 0.06\\ 0.11\\ 0.02\\ 0.09\\ 0.59\\ 0.03\\ 0.21\\ 0.02\\ 0.02\\ 0.03\\ 0.21\\ 0.04\\ 0.05\\ 0.05\\ 0.16\\ \end{array}$ 4172 9687 7898 450 920 3343 1787 4150 2366 1633 2150 2366 1633 1332 5575 978 2224 5301 1487 656 1704 7068 1469 4825 5126 1980 E5 E27 F10 F15 F15 F15 F15 F15 F15 F15 F17 F17 F17 F52 F53 GB40 GS40 GS40 GS40 GS50 GS50 GS50 GS50 GS50 GS28 GB26 GB27 GS30 GS28 R41 TH BK(B) BK(T) Hu Hu BK(B) BK(B) BK(B) BK(B) BK(C) BK( 

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 TABLE 7c

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Table 7. Chemical data for limestones (7a); dolomitic limestones (7b); and dolostones (7c). The abbreviations of the carbonate units are indicated in Figure 3. I.R. = insoluble residue, B = Base, T = Top.

L.G.G. Carbonate Units 1964 L.L.G. Shales Dolost. (Primary Wedepohl et al. (1970, 1972, 1974) Weber Wedepohl et al. (1970, 1972, 1974) Limestones Dolomitic Dolostones Limestones (Secondary 3182 2684 Mn (XRF) 5618 606 245 237 631 637 ppm 174 225 325 187 Sr ppm +45 164 109 Rb 33.9 12 20 19 39.8 1005 Ba 668 485 487 546 85.8 51.4 90 Cu 95\* 24 34 29 65 6.74 41 5.72 126 ppm 50 57 Ni 36 108 6 Co 12 14 13 28 0.13 0.16 TiO2 0.05 1.07 187 202 202 1100 ppm 95 200\* 550 Zn 59 51 69 14 20 **P**b 36 20 38 51 CT

21.6 68.2 18.2

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TABLE 8

The Comparison of the Lower Limestone Group (LLG) data with the data of other workers

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+ Dolostones \* Bituminous shale

Average	GS94	GS29	S34	S31	R15	GS70	GS41	S37	GS25	<b>GB45</b>	F78	S27	GS23	GB25	GB39	GB29	GB3	F48	F19	E23	E4	No.	Sample
	P?	જ	N	N	NS	N	ΝP	N	SN	NP	NS	M	MP	M	MP	M	M	M	MP	M	MP	ment	Environ-
108	219	72	73	131	113	67	95	127	118	118	60	121	101	50	167	142	55	79	112	124	132	ppm	1N
65	85	65	40	57	94	76	79	45	58	84	36	53	93	40	92	67	44	58	73	58	72	ppm	Cu
28	71	31	19	38	39	20	30	16	27	25	23	29	20	18	28	24	38	16	44	20	24	ppm	Co
325	7	133	192	160	112	1162	81	1463	134	311	6	163	280	451	247	258	409	422	128	397	301	ppm	Sr
109	75	115	171	160	65	198	133	158	159	159	53	180	202	21	108	101	31	31	54	50	70	ppm	Rb
69	123	84	23	73	134	140	58	26	70	52	34	55	66	41	95	69	36	56	115	48	54	ppm	Zn
51	53	74	28	27	7	31	47	18	70	23	27	51	35	24	67	127	37	126	31	57	118	ppm	Pb
484	464	1.80	531	463	269	234	420	1143	278	351	307	676	474	411	274	201	1499	227	187	1363	225	ppm	Ba
1.07	1.42	1.21	1.03	1.12	3.07	1.7	1.07	0.68	86.0	1.05	0.8	1.01	1.1	0.26	0.7	0.7	0.57	0.56	1.3	0.77	0.61	8	T102
606	1640	904	559	423	1523	388	222	67	439	387	2645	549	69	688	621	577	403	1647	1349	1781	2009	ppm	Mn

TABLE 9 Chemical Data for Shales

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NNGS .... Marine Non marine Pyrite Siderite

Average

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sample contains 1254 ppm. Petrographic study indicates that these rocks underwent late diagenetic changes, including recrystallisation and the Sr was most probably leached out during these late diagenetic processes. Thompson (1972) reported that Sr concentration is low in recrystallised carbonate facies due to the mobility of this element during diagenesis.

In shales the average Sr content ranges from 6 to 1463 ppm with an average of 325 ppm. The marine shales range in Sr content from 258-451 ppm. The non-marine shales fluctuate in their Sr content as is indicated by the wide concentration range of 6-1463 ppm (Table 9).

In marine shales (calcite bearing) the Sr is possibly located in the calcite structure as described in the carbonate section. The Sr in the non-marine shales (not containing calcite) is probably in the detrital minerals (e.g. feldspar) or adsorped on clay minerals. Sample S37 contains the highest value of the analysed shales, 1463 ppm, and x-ray diffraction reveals that this sample consists mainly of orthoclase. Deer et al. (1966) reported that  $Sr^{2+}$  could replace Ca<sup>2+</sup> in feldspar.

#### Rubidium

Average Rb values are, for limestones and dolomitic limestones 19 ppm and for dolostones 12 ppm (Table 7 ). Weber (1964) and Wedepohl <u>et al</u>. (1972) concluded that rubidium is concentrated in the clay fractions of the insoluble residue of the carbonate rocks. Weber (1964) added also that there is no significant difference
between the rubidium content of primary and secondary dolostones. Degens <u>et al</u>. (1957) pointed out that there is no structural position for rubidium in kaolinite but rubidium can substitute for potassium in illite, montmorillonite and micas.

The plot of Rb against the insoluble residue (Fig. 179) shows a positive relationship, which suggests that the rubidium is concentrated in the clay minerals. The study of the clay minerals reveals that kaolinite and muscovite are the main minerals within the clay fraction. Therefore, the rubidium is most likely concentrated in the muscovite structure.

The rubidium content in the Lower Limestone Group shales ranges from 31 to 202 ppm, with an average of 109 ppm.

Degens <u>et al</u>. (1957) concluded that Rb is a marine environmental indicator, on the basis of its concentration in illite rather than kaolinite. In the analysed samples the rubidium distribution within the marine shales is lower than the non marine shales. This suggests that the rubidium is concentrated within the detrital muscovite minerals similar to the carbonate facies.

## Barium

Table 7shows that the average barium content ofthe limestones is 1005 ppm of the dolomitic limestones is668 and of the dolostone is 487 ppm.

Wedepohl <u>et al</u>. (1972), although indicating a low average value in carbonates of 90 ppm, reported high values



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of 900-1800 ppm in (retaceous limestones from the USA. He stated also that the location of the barium is either detrital clay minerals, redistributed, during diagenetic processes which can lead to the formation of  $BaSO_A$ , or carbonate minerals. However, he related the high Ba content of black shales to their organic material. Veevers (1969), related the barium content of limestones to their insoluble residue, whereas, Thompson (1972) noted a higher barium content in recrystallised limestones. Weber (1964), pointed out that barium is somewhat greater in abundance in primary dolostones (86 ppm), than in secondary dolostones (51 ppm). He also added that barium is strongly extracted from seawater by adsorption onto clays and is largely removed in near shore environments, only a negligible amount being carried to the open sea to be precipitated in deep sea sediments.

The plotting of insoluble residue against the barium content (Fig. 180) indicates a weak positive relationship up to 250 ppm barium, although there is no relationship above this value.

The environment of deposition for the carbonate rocks of the Lower Limestone Group is a near shore environment (see Chapter 3) which suggests that high Ba values are to be expected, the Ba being adsorped onto clay minerals (cf. Weber, 1964). The recrystallisation of these carbonates may have allowed more Ba to be incorporated within the carbonate minerals. Therefore, most likely the high amount of barium within the limestones and dolomitic



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limestones of the Lower Limestone Group is due to the recrystallisation and to the organic materials (plant types). However, barite of biochemical and/or hydrothermal origin could give rise to high barium values.

In the dolostone rocks the average value of barium is less than in the limestone and dolomitic limestone rocks. However, it is more than Weber's (1964) average value for primary and secondary dolostones. The petrography indicates that the dolostone rocks are strongly neomorphosed in comparison to the limestone and dolomitic limestone rocks and low values probably result from the strong diagenetic processes that have affected these rocks. Wedepohl <u>et al</u>. (1972) pointed out that during diagenesis barium is either gained or lost.

In shale the average value of the Ba content is 485 ppm. Degens <u>et al</u>. (1957) concluded that barium is not an environmental indicator. They added also that Ba is associated with oxides, hydroxides or carbonates. Wedepohl <u>et al</u>. (1972) pointed out that biotite and Kfeldspar are the most important carriers for barium. However, he reported the high value in black shales is connected with the organic materials.

Table 9 indicates that there is no difference in barium content in fresh water shales or marine shales. The high barium content within the Lower Limestone Group shales is probably related to the large amount of organic material and is probably present adsorped on clays, since no biotite was identified within the associated sandstones

and shales of the Lower Limestone Group. However, local concentration of organic materials, the presence of oxides or hydroxides might affect the concentration of barium within the analysed shales.

## Nickel

The average content of nickel in limestones and dolomitic limestones is very similar, 56 and 50 ppm, whereas the dolostones contain much less at 36 ppm.

Degens et al. (1957) and Wachs and Hein (1974) pointed out that nickel is concentrated in marine organic matter. Thompson (1972) reported higher concentrations of Ni in recrystallised limestones; 75-90 ppm. Weber (1964) found a difference in Ni content between primary and secondary dolostones, although, because of the dispersion of the data and the small number of samples, the difference in Ni content is not significant. However, he concluded that the source of Ni is the organic material and the clay minerals.

The plotting of the insoluble residue against Ni content (Fig. 181 ) reveals a positive relationship. Therefore the Ni content within the carbonate rocks of the Lower Limestone Group is most likely concentrated in the clay minerals.

In shales the average value of nickel is 108 ppm. Degens <u>et al.(1957)</u> concluded that marine shale is more concentrated in Ni than fresh water shales. Wedepohl <u>et al.</u> (1970) reported a high concentration of nickel in sulphides. The data shows that there is no difference between

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Ni content in the Lower Limestone Group marine and fresh water shales. The data also shows that pyritic shales contain a large amount of Ni. However, some of these shales are marine. Therefore it is difficult to reach a conclusion about the source of the anomalous value of Ni, but the origin of Ni as a whole could be related to clay minerals, organic materials and pyrite.

#### Copper

The average Cu content of the limestones is 29 ppm, of the dolomitic Limestones is 34 ppm and of the dolostone is 24 ppm.

Wedepohl et al. (1970) concluded that the accumulation of Cu in the sedimentary environment is probably connected with detrital matter; iron minerals. However, Wachs and Hein (1974) pointed out that Cu could be concentrated by organisms. Weber (1964) noted an average Cu content of 5.72 ppm.for primary dolostones and 6.74 ppm for secondary dolostones, although he reported a high average value for silty dolostones (160 ppm) and argillaceous dolostones (68.5 ppm). He added that the source of Cu is mainly clay minerals and organic material.

The plotting of Cu content against the insoluble residue (Fig. 182) shows a positive relationship which suggests that the clay minerals as the location of this element. However, the higher average of Cu within the carbonate rocks of the Lower Limestone Group compared with the average value given by Wedepohl <u>et al</u>. (1970) might

Cu ppm 30 10. 60-40 50-Fig. 182Relationship between Cu and the insoluble residue (I.R.) \*\* 10 120 20 30 40 50 60 . 70 x Limestone ▲ Dolomitic Limestone o Dolostones s Siderite •98 80 I.R.%

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suggest other source(s) of Cu such as organic material and/or iron oxides.

In shales the Cu content ranges from 36 ppm to 94 ppm with an average of 65 ppm. The pyritic shales have the highest Cu content; 73-93 ppm. However, one non-pyritic shale sample contains 94 ppm.

Wedepohl <u>et al</u>. (1970) concluded that the bituminous shale contains higher Cu concentration (average 95 ppm) than the non-bituminous shale (average 35 ppm) and that the source of Cu is either organic matter, pyrite or clay minerals.

It seems that clay and organic material are the main locations of copper in the analysed samples, whereas, the highest values (73-93 ppm) are due to pyrite content.

#### Cobalt

There is no significant difference in cobalt content between the analysed limestones, dolomitic limestones and dolostones (Table 7 ) and their average value is 13 ppm.

Wedepohl <u>et al</u>. (1970) reported that cobalt may replace Fe<sup>2+</sup> and Mg<sup>2+</sup> cations as the ionic radius of cobalt is intermediate between the ionic radii of these ions. The insignificant difference in cobalt content in the different carbonate rocks of the Lower Limestone Group indicates that cobalt is related neither to the type of the carbonate fraction or to diagenetic processes. However, the plotting of cobalt against the insoluble residue (Fig. 183 ) shows a positive relationship suggesting that it is concentrated within the clay fractions.



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Co ppm 20 24 28 16 12 a Fig. 183 Relationship between Co and the insoluble residue 1 10 20 30 40 • 50 .. 60 70 80

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(I.R.)

x Limestone • Dolomitic • Dolostone s Siderite limestone

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In shales the Co content ranges from 16 to 71 ppm with an average of 28 ppm. No significant variation exists between the marine and non-marine group. The origin of Co is most likely the clay minerals similar to the carbonate rocks.

### Zinc

The limestone and the dolomitic limestone rocks of the Lower Limestone Group have nearly similar zinc contents, 51 and 59 ppm respectively, whereas the dolostones have the lowest value, 14 ppm (Table 7 ).

Veevers (1969) pointed out that there is no relationship between Zn and insoluble residue and added that Zn is positively related to dolomite and Mg;  $Zn^{2+}$  substituting for  $Mg^{2+}$ . Wedepohl et al (1970) related zinc content to the carbonate fraction and noted that Zn might enter the carbonate fraction during diagenesis. However, he did not exclude the effect of organic material in concentrating this element and added that sulphate reducing bacteria could concentrate Zn from natural waters, probably by sulphide precipitation. Veevers (1969) and Wedepohl et al. (1970) thought that zinc might be adsorped on the surfaces of clays during the dissolution of carbonates. Weber (1964) reported higher zinc values in primary dolostone (1100 ppm) than in secondary dolostone (550 ppm) and also found high values in argillaceous dolostone (5600 ppm). He related the zinc to the clay and organic materials.

The plotting of zinc against magnesium content (Fig.

184 ) reveals no clear relationship between the two elements. However, the plot of zinc against insoluble residue (Fig.185)shows a strong relationship which indicates that the zinc content is related to the clay minerals in the studied carbonate facies. The low values of zinc within the dolostone rocks are, most probably, due to their low concentrations of insoluble residue. However, the abnormally high values might be due to the activity of the sulphate reducing bacteria.

In shales the average content of zinc is 69 ppm. Most of the samples containing more than 70 ppm are pyritic, although some pyritic shales contain less than 60 ppm and some non pyritic shales have more than 70 ppm zinc (Table 9 ).

Degens <u>et al</u>. (1957) stated that zinc is concentrated in the organic fraction of fresh water shales. Wedepohl <u>et al</u>. (1970) reported an average of 95 ppm in nonbituminous shales, whereas those having higher concentrations of organic material contained up to 200 ppm. He considered the iron minerals and clay minerals to be the main locations of zinc in non-bituminous shales.

It seems to be that the clay minerals are the main location of the zinc in the Lower Limestone Group shales. However, the highest values ( >70 ppm) might be related to the pyrite content and/or organic materials.

#### Lead

Average Pb values are, for limestones 36 ppm, for dolomitic limestones 38 ppm and for dolostones 20 ppm.

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Veevers (1969) considered that Pb is concentrated in the soluble fraction of the carbonate rocks. Wedepohl <u>et al</u>. (1974) stated that calcite and dolomite cannot incorporate appreciable concentration of lead, because sea and interstitial waters usually contain very little lead. He added that plants can concentrate large amounts of Pb and he quoted Köster (1969) that the kaolinites can have higher than 40 ppm lead.

The plot of Pb against insoluble residue in the carbonate rocks of the Lower Limestone Group reveals a positive relationship (Fig. 186 ). Kaolinite was identified as the main clay mineral of the insoluble residue. Therefore, the Pb concentration in the carbonate rocks is most likely related to the kaolinite. However, the contribution of lead from the organic materials should be taken into consideration also.

In shales the average lead content is 51 ppm. Six samples contain lead from 57 to 118 ppm; some of them are pyritic.

Degens et al. (1957) and Wedepohl <u>et al</u>. (1974) reported a high concentration of lead in shales rich in organic materials. Wedepohl <u>et al</u>. (1974) added also that lead could be concentrated in pyrite.

The main location of lead in the Lower Limestone Group shales is considered in this study to be clay minerals (kaolinite), similar to the carbonate rocks. The anomalous value ( >57 ppm) is probably related to pyrite content, high concentration of organic material and/or clay minerals.



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#### Titanium

The analysed samples of the carbonate and shale sediments of the Lower Limestone Group, indicate that the average distribution of titanium (calculated as  $TiO_2$ ) is 0.16% ppm in Limestones; 0.13% in dolomitic limestone; 0.05% in dolostones and 1.04% in shales.

Wedepohl <u>et al</u>. (1970) pointed out that titanium is found as a major constituent of oxide, titanate and silicate minerals. He added that titanium usually substitutes for Fe or Al. Veizer and Demovic (1973), related the titanium concentration to insoluble fractions. They reported that Ti can be used as an indirect measure of the amount of  $\leq 2$  u clay fraction. Weber (1964) indicated an average of 202 ppm in primary dolostones, and 187 ppm in secondary dolostones. Although he referred the titanium concentration mainly to the clay fractions, he added the possibility that organic matter may concentrate this element.

Plotting TiO<sub>2</sub> against the insoluble residue shows a positive relationship (Fig. 187). Titanium bearing minerals (e.g. Rutile and Anatase) were identified in the sandstones associated with the studied carbonate and shale sediments. Therefore, most of the titanium is probably concentrated in clay minerals (kaolinites) and/or other detrital insoluble fractions such as titanate or oxide minerals.

In shales, the data reveals that non-marine shales contain more titanium than marine shales. This is most



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likely due to the higher percentages of detrital minerals (e.g. kaolinite, titanates and oxides) in non-marine than marine shales.

#### 5.3 MAJOR ELEMENTS

#### Manganese

The Mn distribution in the soluble fractions and the whole rocks of the carbonate of the Lower Limestone Group is presented in Table 7. The analysed samples (XRF results) reveal that the average Mn content in limestones is 2685 ppm, in dolomitic limestones is 3182 ppm and in dolostones is 5618 ppm.

Veevers (1969) concluded that the concentration of Mn is usually proportional to Fe and both of them are concentrated in the insoluble residue. Bencini and Turi (1974) reported that Mn can be transported as adsorped on clay minerals and could be substituted for  $Ca^{2+}$  in calcite structure rather than aragonite structure during diagenesis. They pointed out that dolomitisation did not produce a significant change in Mn content. However, Rao and Naqvi (1977) reported a more than three fold increase in Mn content in dolostones compared with their limestones in the Lower Ordovician sequence of Tasmania, Australia. Weber (1964) noted that no significant difference in Mn content between primary and secondary dolostones and he related the Mn content to the dolomite structure. However, he added also that argillaceous dolostones contained a higher Mn content (640 ppm) than primary and secondary

dolostone (245, 237 ppm respectively).

The plot of Mn against the insoluble residue (Fig. 188) in non-sideritic rocks indicates that there is no However, the plotting of Mn against Fe and relationship. Mg reveals a positive relationship (Figs. 189, 190). The petrography of the carbonate indicated that the samples in which the groundmass is composed mainly of highly ferroan calcite have more Mn than the samples in which the groundmass is composed mainly of non-ferroan calcite. The ferroan calcite is interpreted as the result of late diagenetic processes (see Chapter 3). Samples F10 and F47, taken as examples, have a groundmass which consists mainly of non-ferroan calcite, although they contain a very small amount of ferroan dolomite (the presence of dolomite is interpreted to increase the Mn content). The Mn and Fe contents of these two samples are lower than the non-dolomitic rocks (e.g. GBr, GB6, GB7, GS61) in which the groundmass is composed mainly of ferroan calcite. High value of Mn and Fe were obtained in the dolostone rocks which are mainly of ferroan dolomite type (Table 7).

The discussion of the carbonates (Chapter 3) suggested that the carbonate rocks were deposited as aragonite which later changed by diagenesis into lowmagnesian calcite. The analysed values for Mn indicated unusually high values which are not expected in aragonite minerals. Therefore the high amount of Mn in the studied non-sideritic carbonate rocks probably is due to substitution of  $Mn^{2+}$ , which was probably adsorped on the



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clay minerals, for  $Ca^{2+}$  and  $Mg^{2+}$  in calcite and dolomite structures during late diagenetic processes. The high values of Mn in the siderite bearing carbonate facies are probably related to the high concentration of Mn within the siderite structure. Deer <u>et al</u>. (1966) reported that both  $Mn^{2+}$  and  $Mg^{2+}$  could be substituted for the  $Fe^{2+}$  in siderite.

In shales the manganese ranges from 222 to 2645 ppm with an average of 909 ppm. Bencini and Turi (1974) explained the Mn enrichment in the Posidonia Marls of the Tuscan sequence (Mesozoic), Lima Valley, Northern Apennines, as due to adsorption on clay minerals. They added that reducing conditions after deposition allowed a large amount of  $Mn^{2+}$  to substitute for  $Ca^{2+}$  in the calcite structure.

The highest value of Mn was recognized in calcareous, sideritic and some pyritic shales. The shales of the Lower Limestone Group contain a lot of organic material. The discussion of the ironstones (Chapter 4 ) revealed that the decomposition of the organic materials consumed the oxygen and reducing environment were produced and sideritic and/or pyrite were produced depend on the rate of sedimentation, and sulphide and carbonate activities. It is suggested that most of the Mn within the analysed shales is adsorped on clay minerals. However, the highest values are most likely due to the available  $Mn^{2+}$  being substituted for Ca<sup>2+</sup> in calcite, Fe<sup>2+</sup> in siderite and, to less extent, Fe<sup>2+</sup> in pyrite. Deer et al. (1966) pointed

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out that the replacement of  $Fe^{2+}$  by  $Mn^{2+}$  in pyrite is very limited.

#### Iron

The average values of the iron are, for limestone 1.52%, for dolomitic limestone 1.72% and for dolostones 2.67%.

Veevers (1969) indicated that iron might be located in the insoluble residue whereas Wedepohl <u>et al</u>. (1970) noted that  $Fe^{+2}$  could replace Mg<sup>+2</sup> in dolomite structures because of the close similarity between the radii of the two ions.

The plot of Fe and I.R. (Fig. 191) shows a weak positive relationship which is better shown in the region of 30% insoluble residue and 2% Fe. The plot of Fe against Mn suggests a positive relationship (Fig.192). The discussion of the Mn revealed that Mn entered into carbonate minerals during late diagenetic processes. However, the discussion of iron in the dolomite section (Chapter 3) revealed that iron entered early into carbonate minerals.

It is suggested that the location of the iron in the studied carbonate rocks is the late diagenetic ferroan calcite, iron oxides, pyrite, siderite and the clay minerals. For the dolomite bearing rocks the iron probably replaced  $Mg^{2+}$  in the dolomitic structure earlier, before the stabilisation of the carbonate minerals, and during late diagenetic processes.

In shales the iron ranges from 0.55% to 13.16% with an average of 3.5%. The highest values are present in



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the siderite and pyritic bearing shales. The location of the iron in shale is probably similar to the carbonate rocks i.e. iron oxides, pyrite, siderite and the clay minerals. However, in marine shales Fe is probably located in calcite by substitution for Ca<sup>2+</sup>.

# 5.4 HORIZONTAL AND VERTICAL VARIATIONS

Ba, Sr and Mn, which are not related to the insoluble residue, and Rb and Zn which were randomly selected from the trace elements having a positive relationship to the insoluble residue, were chosen to see if there are any horizontal variations within two selected carbonate units; the Hurlet and Blackhall Limestones.

The plotting of the data (Figs. 193 - 201) reveals no specific trends . The distribution of these data also indicate that Zn, Ba, Rb and Mn varied randomly within the Hurlet Limestone and Blackhall Limestone. The random variations in Zn and Rb are controlled by the insoluble residue; in Ba are controlled by the degree of diagenesis and in Mn are controlled by siderisation, dolomitisation and other diagenetic processes. The plotting also reveals that Sr values are almost constant within the Hurlet Limestone and to a lesser extent within the top of the Blackhall Limestone, and are more variable within the base of the Blackhall Limestone. The constant values of Sr within the Hurlet Limestone probably indicates that Sr is related to the carbonate fraction. However, the variations within the Blackhall

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Limestone might be related to diagenesis. It was found petrographically that diagenesis affected the base of the Blackhall more than the top of the Blackhall which, in turn, was affected more than the Hurlet Limestone.

Four stratigraphic sections were chosen to show if there is any vertical variation within the different carbonate units of the Lower Limestone Group (Figs. 202-205). For this purpose, Sr, Mn and Ba. which are not related to insoluble residue, and Rb, Pb and Ti, which have a positive relationship to the insoluble residue, were chosen. The same conclusion concerning the variation of these elements horizontally could be applied to the vertical variations. The trend of variation of Pb, Rb and Ti follows the trend of the insoluble residue, whereas Sr, Mn and Ba do not.

#### 5.5 CONCLUSION

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The analysed trace elements Ni, Cu, Zn, Pb, Co. Ti and Rb are concentrated in the insoluble residue of the carbonate rocks. This suggests the possibility that clay minerals are the main source for these elements. However, rubidium is considered to be contributed from detrital muscovite. Organic material may also concentrate these elements and possibly be responsible for the anomalous values of these elements in comparison to average value given by Wedepohl <u>et al</u>. (1970, 1972, 1974) and Weber (1964).

No relationship exists between the insoluble residue and Ba, Sr and Mn, which suggests that they are concentrated

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Fig. 202 Vertical variatio, of some of the analysed elements,  $TiO_2$  and insoluble residue (I.R.). Scale; l cm = l m for carbonates; l cm = 20 m for non carbonates.

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Fig. 203 Vertical variation of some of the analysed elements, T10, and insoluble residue (I.R.). Scale: 1 cm = 1 m for carbonate; 1 cm = 20 m for non-carbonates.

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within the carbonate fraction. Barium which is thought to be adsorped on clay minerals seems to enter the carbonate fraction during recrystallisation. Iron is probably located in the late diagenetic ferroan calcite, dolomite (early and late diagenetic), iron oxides, pyrite, siderite and clay minerals.

Sr is reported to be high in aragonite and low in calcite minerals (cf. Veizer and Demovic, 1973), whereas Mn is high in calcite and low in aragonite (cf. Bencini and Turi, 1974; Rao and Naqvi, 1977). In the carbonate rocks of the Lower Limestone Group, both Sr and Mn are high. However, the dolostones have low values of Sr (Table 7).

Rao and Naqvi (1977) reported a positive relationship between Sr and Mn in the Lower Ordovician sequence of Tasmania, Australia. They related this relationship to the mixing of continental saline brine and marine brine which induced dolomitisation. They stated that (p.1047) "High Mn content in the dolomites is probably due to higher salinity of the continental brines which were capable of leaching Mn from the adjacent terrigenous clastics under reducing conditions". They added that Sr is less concentrated in the continental brines than in the marine brines.

In the Lower Limestone Group, Sr has a positive relationship with Mn in the limestones and dolomitic limestones, whereas no relationship exists in the dolostones (Fig. 206). The possibility of losing Sr during

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diagenesis is reported by many authors (e.g. Weber, 1964; Thompson, 1972) whereas no such loss was reported with respect to Mn during diagenesis. On the contrary Mn could increase during diagenesis, since Mn, together with Fe, substitutes for Ca and Mg in the formation of ferroan carbonates.

The rocks in which the groundmass is composed mainly of neomorphic ferroan calcite contain more Mn than those in which the groundmass is composed mainly of non-ferroan calcite. This suggests that Mn enters the calcite structure together with Fe (and also the dolomite structure) during late diagenetic recrystallisation. This probably explains the unusual high amount of Mn and the positive relationship between the Sr and Mn in the Lower Limestone Group sediments.

No difference in trace elements has been found between the marine and non-marine shales. The clay minerals are thought to contain Ba, Ni, Cu, Zn, Pb, Ti and Mn in this study. The anomalously high values for Ni, Cu, Zn and Pb are probably due to incorporation in organic matter and/or pyrite and for Mn and Fe is calcite, siderite and pyrite. Muscovite is considered to be the main location of the rubidium and kaolinite of the Pb. The carbonate fractions are considered as the main sites of the Sr in the marine shales together with muscovite and K-feldspars. In the non-marine fractions muscovite and/ or K-feldspars are considered to be the main Sr bearing minerals.

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# APPENDIX I

## HEAVY MINERALS STUDY

Henningsen (1967) indicated that the crushing of indurated rocks does not affect the heavy mineral frequency and he recommended crushing in an electrically powdered Jaw Crusher with a minimum opening of 5 mm. Carver (1971) recommended the heavy mineral size fraction between 20 to 30 as being most representative of the source area.

The sandstone studied for heavy minerals are highly indurated. They were crushed in a Jaw Crusher with a 5mm opening. The disintegrated samples were crushed carefully with rubber rolling pin to disintegrate the aggregations or uncrushed fragments. The samples were washed with water and the clay fractions and impurities were siphoned The powdered, dried samples were then seived and the off. 20 to 30 size fractions were chosen for heavy mineral study. Bromoform of specific gravity 2.89 was used to separate the light fractions from heavy minerals. Microscopic observations indicate that all the heavy minerals were stained with iron oxide which made their identification Jain (1972), in his study of the Precambrian difficult. Gamri Quartzite, boiled the heavy mineral fractions with HCl to remove the iron oxide pigments. Carver (1971) indicated that the treatment of the heavy minerals with HCl could effect the apatite and some opaque minerals.

The studied heavy minerals were investigated for apatite and no indication of it was observed. Therefore, part of the heavy mineral fractions were treated with 10%

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HCl and heated carefully until the iron oxides were removed. The heavy minerals were mounted in oil (the same refractive index as Canada balsam) and studied under the microscope.

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# APPENDIX II

# STAINING

Four methods of staining were used to select the one applicable to the study of these carbonate rocks. These are:

1 - Evamy (1963) method

The solution of this method is prepared by mixing 0.2% HCl with 0.2% alizarin red S and 0.5-1.0% potassium ferricyanide. With this method the non-ferroan calcite gives a red colour and Fe-poor to Fe-rich calcite gives mauve to purple. It was found very difficult to differentiate between non-ferroan and ferroan calcite when this method was used.

# 2 - Dickson (1965) method

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The solution of this method is prepared by mixing 0.2 gm alizarin red S per 100 cc and 1.5% HCl and 2.0 gm potassium ferricyanide per 100 cc and 1.5% HCl, in which 3 parts of alizarin red S was mixed with 2 parts of potassium ferricyanide. The application of this method results in part of the shell fragments and some of the calcite grains remaining colourless, as if they were nonferroan dolomite. This is probably due to the use of relatively strong HCl (1.5%) which reacts strongly with the rocks and removes the pigments produced by staining and consequently the calcite does not stain.

3 - Davies and Till (1968) method This method follows Dickson's (1965) for preparing

the solution and is recommended for pells. The application of this method gives the same result which is obtained by Dickson's (1965) method.

4 - Katz and Friedman (1965) method

This method mainly followed Evamy's (1963) method and the staining solution was prepared by dissolving 5 gm of potassium ferricyanide and 1 gm of alizarin red S in a distilled water containing 2 ml concentrated HCl. The solution was warmed to  $40^{\circ}$ C. Whether polished rocks or slides were stained the time used for staining is 4 minutes.

This method was used and it was found applicable to differentiate the carbonate minerals in the Lower Limestone Group. This is probably because of the longer time of staining (4 minutes) and the low concentration of HCl (0.2%). The etching solution used is 10% HCl for the polished rocks as recommended by Katz and Friedman (1965) and 1.5% for slides as recommended by Dickson (1965).

### APPENDIX III

# INSOLUBLE RESIDUE

The amount of insoluble residue was determined according to the method of Bisque and Lemish (1959) as follows:

l0 gm of the powdered sample was dissolved in 3 NHCl for half an hour at  $60^{\circ}-80^{\circ}$ C. Some of the samples, especially the dolomitic rocks, were analysed by x-ray diffraction to check that all the carbonate had dissolved. The insoluble residue was then weighed and recalculated to a percentage.

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#### APPENDIX IV

# CLAY MINERALS

The Ostrom (1961) method was followed for the separation of clay from the carbonate in this study. He recommended 0.3M acetic acid to dissolve the limestones and 0.11 M HCl to dissolve the dolostones. The steps for the dissolving of carbonates and calcareous shales are as follows:

1 - 20 g of the powdered sample were placed in 500 ml beaker and 200 ml of 0.3M acetic acid, if the rock was limestone, or 0.11 M HCl, if the rock was dolostone, were added.

2 - The mixture was stirred until the reaction ceases. The liquid decanted and the processes repeated until the reaction had completely ceased. This process usually takes more than a day.

3 - The liquid was separated from the solid by filtering and dried at room temperature and the powder was examined by the XRD to ensure the dissolving of the carbonate.

For preparation of oriented slides, Gipson's (1966) method was used as follows:

1 - The powder was placed in 1000 ml. graduated cylinder and distilled water was added.

2 - The mixture was stirred and then allowed to settle for approximately six hours. All the materials greater than 0.002 mm in diameter would have settled below a depth of 10 cm at the end of six hours.

3 - Glass slides were mounted on inverted petri dishes

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and placed on the bottoms of one litre beakers.

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4 - The suspension was allowed to settle overnight or for about 12 hours.

5 - The remaining suspension was then siphoned from the beaker carefully (not disturbing the slides). The slides were removed from the inverted petri-dishes, dried at room temperature and analysed by x-ray diffraction.

6 - The same slides were heated for 550 c for one hour after they had been analysed by x-ray diffraction and then reanalysed by the XDR. This is to differentiate  $7^{\circ}A$ chlorite peak from  $7^{\circ}A$  kaolinite, the heating destroyed the kaolinite.

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