

- Title Magnetic and Sedimentological Analyses of Quaternary Lake Sediments from the English Lake District
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MAGNETIC AND SEDIMENTOLOGICAL ANALYSES OF QUATERNARY LAKE SEDIMENTS FROM THE ENGLISH LAKE DISTRICT.

DONALD C.H. McLEAN

A thesis submitted in partial fulfilment of the requirements of the Council for National Academic Awards for the degree of Doctor of Philosophy.

March 1991

Luton College of Higher Education

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MAGNETIC AND SEDIMENTOLOGICAL ANALYSES OF QUATERNARY LAKE SEDIMENTS FROM THE ENGLISH LAKE DISTRICT. ABSTRACT

Results of mineral magnetic, mobile element, and granulometric analyses of Holocene sediments from Buttermere and Crummock Water (two closely-linked lakes in the north-west of the English Lake District) are presented. These are used to: (1) identify effects of internal (lacustrine) and external (catchment) controls on sedimentation; (2) establish catchment source-lake sediment linkages and assess the value of mineral magnetic techniques in palaeolimnological studies; (3) identify major catchment environmental changes.

Analyses of lake sediment fabrics (using sediment thin sections, SEM clay flake analysis, standard granulometric analysis, and mineral magnetic indicators of grain size change) indicate that river plume sedimentation is the normal sediment dispersal mechanism in these lakes. Thin (≤ 3.0 mm) chlorite-rich laminae, found at intervals in the otherwise homogeneous Holocene sediment sequence, are probably formed by trapping and concentration of fine, platy particles within lake waters. They are subsequently deposited during lake overturn. This represents an "internal" control on sedimentation. A model of sedimentation processes operating in these lakes is developed, incorporating river plume sedimentation, episodic density surges, and lake thermal structure.

Mineral magnetic measurements allow the objective subdivision of the lacustrine lithostratigraphy, identifying broad changes in lake sediment characteristics. Samples from both lake catchments are clustered into six magnetically distinct groups - despite the lithological complexity of the catchment. Comparison of these with the lake sediments has enabled identification of major sources during the Holocene. Following deposition of relatively unaltered bedrock-derived material during the Late-glacial ("primary" sources), secondary sources (which may include glacial diamicts, soils and stream sediments) dominate the lake sediments. Direct input of topsoil-derived sediment from *circa* 1000 A.D. onwards (during and following the main period of Norse settlement of the Lake District) is identified by its distinctive mineral magnetic characteristics, (high Xfd% values, $>\sim4\%$). Industrially-derived magnetic spherules contribute significantly to the mineral magnetic characteristics of the more recent sediments, (mainly those post-dating *circa* 1900 A.D.). These are used to construct a proxy chronology for recent sediments.

Catchment environmental changes are mainly related to stabilisation of vegetation following deglaciation and, from *circa* 2 000 B.P., anthropogenic effects of deforestation and land disturbance, thus increasing lake sediment accumulation rates. These findings are broadly consistent with the interpretation of the Lake District Post-glacial sediment sequence presented in studies by Mackereth, (1966a), and Pennington, (1981), demonstrating a uniformity of lake and catchment development within the Lake District. A prominent minerogenic layer present in the Buttermere and Crummock Water sediment sequence however broadly correlates with similar horizons deposited in other Lake District lakes from *circa* 7 400 - 5 000 B.P. These have been previously interpreted as composed of topsoil-derived material derivd from human actions, (Pennington 1973, 1981). In the Buttermere and Crummock Water sediments reworked from <u>within</u> the lake basins, probably following lowered lake water levels during the period *circa* 7 300 - 5 300 B.P. Thus it is suggested that a re-interpretation of similar Lake District lacustrine sediments using the methods employed in this study would be appropriate.

Mud, mud, glorious mud, Nothing quite like it for cooling the blood, So follow me, follow, Down to the hollow, And there let us wallow, In glorious mud.

> Donald Swann, "The Hippopotamus Song" (Personal communication, December 1990)

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DECLARATION

The material of this thesis is based on the author's previously unpublished independent research. Material used from other sources is acknowledged within the text where applicable.

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ABREVIATIONS.

ARM Anhysteretic Remanent Magnetisation;

- κ volume or whole core susceptibility;
- IRM Isothermal Remanent Magnetisation;
- IRM_n the isothermal remanent magnetisation after subjecting

a sample to a specified field (n) measured in milliTesla;

- LOI loss-on-ignition;
- MD multidomain (re: magnetic grain size);

mT milliTesla;

- NRM natural remanent magnetism
- PEG polyethylene glycol
- PSD psuedo single domain (re: magnetic grain size);
- SEM scanning electron microscope;

SD single domain (re: magnetic grain size);

- SIRM Saturation Isothermal Remanent Magnetisation;
- SP superparamagnetic (re: magnetic grain size);
- T Tesla;
- χ mass specific susceptibility;
- χ lf low frequency mass specific susceptibility;
- χhf high frequency mass specific susceptibility.

(n.b. appendix 1 provides an explanation of mineral magnetic theory and the mineral magnetic parameters used in this study - which should be consulted by those unfamiliar with this methodology)

1. INTRODUCTION, AIMS, AND REVIEWS.

"lake and their drainage basins....provide a unifying framework within which to study environmental processes and ecological change on a wide variety of timescales" (Oldfield, 1977, page 461)

1.1 Introduction.

Oldfield (1977) and Haworth and Lund (1984), for example, have demonstrated the value of lake sediments for investigating environmental change over a wide variety of timescales. Timescales of events observed recorded in lake sediments range from diurnal, subseasonal, annual, and historic to "ancient"; environmental factors recorded include external processes, such as catchment erosion (and hence soil stability and formation), vegetation change, and, latterly, anthropogenic effects both inside and outside of a given catchment. There are also internal factors such as changing lake water quality, thermal structure, and internal sediment distribution processes. That so many factors (traditionally divided into individual studies within discreet scientific disciplines) may affect limnological processes has for some years been recognised as providing a unique opportunity for the study of a wide range of environmental variables, principally through the investigation of accumulated lake sediments (Frey, 1969). This thesis utilises such lake sediment records from part of the English Lake District to investigate the lake/catchment relationship and environmental changes during the Holocene using a range of techniques.

Because of the wealth of information potentially available from such investigations there is a large body of literature detailing studies both of ancient and of contemporary lake systems. The bulk of these have employed analysis of the biogenic components of the sediment to investigate both external changes such as landscape evolution, and internal lake processes. Lake District examples including Haworth, 1985, Pennington, 1943, 1947, 1970). In these studies, biogenic material is used as,

> "an amalgam....representing time (seasonal changes), space (habitats in and around the lake) and environment (the range of

conditions pertaining),".

(Haworth, 1985, page 64)

This approach offers a powerful methodology, but is an examination of only part of the properties of a lake sediment. Despite often comprising the bulk of the material deposited (70% or more in oligotrophic lakes, Mackereth 1965), the non-biogenic, clastic, fractions of lake sediments have in some respects received more limited study. As in the case of studies of te biogenic fraction, analysis of the non-biogenic components of lake sediments can also provide information of past soil states, and hence of erosional processes and of climate, but in addition yield direct information on the provenance of the lake sediments.

The generally rather uniform appearance of many Post-glacial lake sediments, often described simply as "brown mud" in the Lake District (Pennington, 1981), has probably not encouraged extensive sedimentological investigation of the clastic component. In some of the few studies of inorganic fractions of lake sediments from the Lake District, both Holmes (1968) and Chambers (1978) tried to relate their composition to catchment sources using grain size distribution and clay mineralogy. In comparison, the more structured, glacial lake sediments observed in other countries have been and still are the subject of extensive theoretical and field investigation (for example, Kuenen, 1951, Catto, 1987). These have attracted some attention in the Lake District both individually (Smith, 1959a) and in conjunction with the studies of biogenic components mentioned above.

Mackereth (1965, 1966a) supplied a conceptual and practical advance for palaeolimnology with the application of methods of chemical analysis, demonstrating that these, as well as visual stratigraphy could supply information on both "external" as well as "internal" processes (as defined above). In particular, sediments incorporate information on the weathering conditions experienced by catchment substrates prior to their incorporation in lake deposits, and on the palaeo-redox states both of catchment soils and of lake sediments. Subsequent chemical investigations of a wide variety of lake sediments have confirmed the value of this approach (for example, Engstrom and Wright, 1984; Huttunen, Meriläinen, and Tolonen, 1978; Pennington, 1981), but have also highlighted the difficulty of interpreting chemical stratigraphies in view of potential changes experienced in lake waters by both dissolved and particulate matter during and after deposition. The development and application of magnetic techniques of investigation to lake sediments have provided similar practical and conceptual advances, particularly in the study of the lake-catchment <u>system</u>. As with Mackereth's use of element chemistry, it is not so much that the techniques of what is now termed the "mineral magnetic" approach were new (although developments have taken place), since there was already an established body of physical theory and practical application in relation to the study of "rock magnetism". Rather, the <u>application</u> of the technique to this novel material (lake sediments) represented an advance in palaeolimnology through the analysis of a particular clastic component of the sediments, (Thompson *et al*, 1975).

The major advantages for palaeolimnological investigations of the mineral magnetic approach are: (i) it permits rapid whole core correlation of sediment (a great advantage for visually homogeneous horizons, Dearing *et al*, 1981); (ii) that it also provides a means of correlation between soil, substrate, and sediments allowing the more widespread investigation of source-sediment linkages, (Oldfield *et al*, 1985a). In both cases this analysis is relatively rapid, safe, may use samples of small size, and is non-destructive. Hence additional analysis is not precluded. Published studies which have applied this methodology to the detailed investigation of source-sediment linkages have concentrated on catchments of relatively simple geology (for example, Stott, 1986, 1987, Yu, 1989) which have shown marked soil-bedrock distinctions in terms of their magnetic properties. Thus, the use of this mineral magnetic methodology for source-sediment linkage investigation remains to be demonstrated in rather more complex catchments (for example, in upland, glaciated areas, where less well-developed soils, and the widespread distribution of glacial diamicts may considerably complicate the identification and clear distinction of catchment sources).

Lake sediments provide a record of environmental change, unusual in comparison to most terrestrial deposits for their continuity and potential level of detail. Whilst the biogenic components of such sediments have frequently been investigated, relatively few detailed studies have been made of the non-biogenic sediment components, particularly in the visually rather homogeneous Holocene lake sediment sequences from the Lake District.

The non-biogenic component can provide more direct information than the biogenic of on the catchment landsurface conditions (for example, weathering, and erosion), and may also allow identification of sediment sources and variations in their relative importance over time. Advances in analytical procedures, particularly of "mineral magnetic" methods, allow the use of the clastic components in relatively rapid investigation of both internal and external processes through core correlation and sediment source identification.

1.2. Aims.

As discussed in the previous section, many investigators have studied landscape development but often through less "direct" methods, (for example using pollen and diatom analysis), rather than via the characteristics of the primary sediment (which should show the most obvious provence relationships). Analytical developments, particularly in the field of mineral magnetism, now facilitate the further examination of the non-biogenic fractions of the sediments of oligotrophic lakes, in order to examine <u>directly</u> the relationship between sediments and sources, (see review of the applications of mineral magnetics, section 1.4). This study therefore aims to use analysis of the non-biogenic fraction of lake sediments to study the physical processes of landscape evolution and of catchment source-lake sediment linkages during the Holocene.

The Buttermere and Crummock Water catchment in the English Lake District provides a system, as yet largely unexplored in any detail, but one for which there is a framework of landscape and vegetation evolution available, (discussed here in chapter 2, section 2.2). This has been developed largely through the investigation of sediments from Windermere, and sites in its catchment, including Blea Tarn and Blelham Bog, by workers such as Mortimer (1949), Mackereth (1965), Pennington (1947), Haworth (1969), Oldfield, (1970), Macan, (1970). More comprehensive bibliographies are provided by R.A.Smith (1974, 1990) and Haworth and Long (1989). The geology of the catchment, and the morphological characteristics and trophic status of these two lakes, (discussed in sections 2.1 and 2.3) indicate that methods mineral magnetism would be suitable for the study of source-sediment relationships in this catchment, in terms both of the potential supply of

magnetic minerals, and of their stability following deposition, (see also discussion in section 1.4). Studies of landscape evolution and vegetation history from adjacent lakes and cacthments provide a framework within which environmental change can be considered, (section 2.2).

Buttermere and Crummock Water share the same glacial trough, and are linked by the Buttermere Dubbs river, which is approximately 1 km in length. Investigation of such closely-linked water bodies provides an opportunity to assess the influence of an upstream lake (Buttermere) acting as a sediment trap on sedimentation in the lower, (Crummock Water).

This study therefore aims to assess major Holocene environmental changes in the Buttermere and Crummock Water catchment, to establish source-sediment linkages between lake sediments and catchment substrates, and to identify the effects of "external" and "internal" factors on sedimentation in the two lakes via a study of the non-biogenic component of the sediments.

1.3. Methodology.

A summary of the approach used to achieve these aims (section 1.2) would be to:

i) to review the characteristics of the two lake basins and their catchment using published sources;

ii) to collect lake sediment cores and collect catchment soil and substrate samples representative of possible sediment sources;

iii) to construct a sediment chronology which provides a framework within which to evaluate observed changes;

iv) to use standard geochemical techniques to analyse the lake sediments, allowing assessment of changes in catchment erosion style;

v) to characterise samples of lake sediments and catchment material on the basis of their mineral magnetic properties allowing investigation of source-sediment linkages;

vi) to assess patterns and processes of lake sedimentation via variation in sedimentation patterns and grain sizes between cores, and using sediment micro-fabrics as a guide to

depositional mechanisms.

Post-glacial sediment sequences from the lakes in the English Lake District are largely uniform in appearance (Mackereth 1965, 1966a, Pennington, 1981). The visual methods of analysis of clastic sediments, used to interpret sediment supply and depositional processes in many glacial lake sediment sequences (N.D.Smith, 1978, 1981, Ashley, 1975, Ashley, Shaw, and Smith, 1985), are therefore generally inappropriate. However, microscopic examination of even visually homogeneous sediments may still be of value in determining depositional processes through recognition of sediment structures too small to be seen in hand specimen, (Stürm, 1979, O'Brien *et al*, 1980). Such analysis is therefore used as a supplementary method in this investigation, (chapter 5).

Techniques of mineral magnetic analysis of sediments are particularly appropriate in the study of visually homogeneous sediment cores, permitting core correlation, and also correlation between lake and source sediments (Thompson et al, 1975; Bradshaw and Thompson, 1985; Oldfield et al, 1985a). Such techniques are therefore used as the primary investigative tool in this study, as they facillitate both the investigation of external processes of sediment supply (via identification of source-sediment linkages), and internal lake processes such as sediment focusing (Dearing, 1983). They also enable the study of within-lake variations in grain size through examination of the appropriate magnetic parameters. Such methods have been applied to the analysis of lake sediments since the early 1970's (Thompson, 1973, Thompson et al, 1975), and have proved a rapidly developing methodology, but are not yet widely used or understood. Therefore, a brief discussion of the basis of the methodology of mineral magnetism and its application is conducted in the following section. (A more complete discussion of the physical basis, techniques of mineral magnetic measurement, of the instrumentation employed, and the definition and interpretation of mineral magnetic parameters used in this study, is provided in appendix 1).

Magnetic analyses are supplemented in this investigation by a number of chemical and

physical techniques in order to provide complementary and corroborative evidence of source, and catchment weathering and erosion processes. These additional techniques include chemical analysis of mobile elements, and mineralogical analysis using X-ray diffraction (XRD) analysis. Such a combined approach allows identification of relative weathering states of source material (for example, freshly eroded bedrock or mature, leached, soil). Such techniques and their interpretation have a demonstrated application to the study of lake sediments (Engstrom and Wright, 1984; Dean and Gorham, 1976). Their application to this study is discussed with the results presented in chapters 3 and 4.

Many factors act in combination to influence the type and distribution of sediments within a lake. Mostly these might be considered as "external", including the processes actually influencing the types and rates of sediment supplied. This may include the types of material available (dependent initially upon catchment geology and geomorphology) and the types of weathering and erosion processes operational. Internal factors, may also exert important controls on the final characteristics of the sediment though. For any such investigation it is therefore important to posses an understanding of these controls and how they interact. The final section of this chapter (section 1.5) therefore outlines the principal controls on lake sediments, and sedimentation processes operating in catchments broadly comparable to the Buttermere and Crummock Water system.

1.4. Application of mineral magnetic techniques to lake sediment studies.

Detailed explanations of the causes and physical basis of rock magnetism, from which the techniques of mineral magnetics are derived, are provided by Stacey and Banerjee (1974) and Tarling (1983). Thompson and Oldfield (1986) give explanations of both the physical basis, and the applications of mineral magnetism in a wide range of environmental studies. Such descriptions will therefore not be giev in detail in this study, although a summary of the physical basis of mineral magnetism and the terms and magnetic parameters used in this study is provided in appendix 1.

The basis of the mineral magnetic approach is that, by measurement of the response of a sample to various controlled magnetic fields, the concentration, granulometry, and mineralogy of its magnetic assemblage can be characterised (see appendix 1). Iron is an element widespread in many natural systems, and although it is usually of low absolute concentration, it often dominates the properties of natural magnetic assemblages. The magnetic properties of iron compounds are sensitive both to chemical and to thermal transformation, such as may be experienced in "normal" pedogenic fermentation processes, and in forest and heath fires (Mullins, 1977; Maher and Taylor, 1988). In a variety of previous studies it has been found both that specific iron compounds can be indicative of particular environments of formation (Maher, 1986), and that these characteristics frequently remain conservative during transport within geomorphic systems, (J.P.Smith, 1986). In essence, the approach therefore utilises a naturally "labelled" material and provides a means of tracing its movement through a system in order to allow the idebtification of source-sediment linkages, and hence the processes and controls responsible for these transfers.

Two major applications of mineral magnetism are observed in published limnological studies. These are: i) core correlation; ii) characterisation and correlation of sediment and source material.

Successful core correlation requires areal continuity and synchroneity of deposition. Whilst

it is well known that sedimentation in lakes is not uniform over an entire basin (Lehman, 1975), changes in accumulation rates are often gradual. Core sampling of sufficient density allows correlation of sedimentary features across basins. The first published studies incorporating mineral magnetism (Thompson, 1973, Thompson *et al*, 1975) utilised whole core susceptibility profiles for core correlation. Cores from Lough Neagh collected 8 km apart were successfully correlated across the lake. Correlation based on magnetic susceptibility profiles was confirmed by comparison with diatom stratigraphy. This method has been extended by multi-core studies such as those of Bloemendal *et al*, (1979), Dearing *et al*, (1981), and Dearing (1983), where the speed of measurements of whole core susceptibility on unextruded cores permitted detailed analysis of closely-spaced data, and hence the fine resolution evaluation of changes in patterns of sediment accumulation within lakes. Dated features from a "master" core have been transferred to others on the basis of similarity of their magnetic profiles. Combined with the greater sampling the speed of the method allows, this is useful in the calculation of more representative sediment influxes for lakes, (Dearing, 1986).

ii) Characterisation of, and correlation between lake sediments and their sources, is based upon a variety of infield and remanent responses of samples, allowing calculation of mass specific magnetic parameters, (as opposed to volume susceptibility measurements frequently used for initial core correlation). These results may then be compared, preferably on a multivariate basis, between samples of lake sediments and catchment material. The data derived from these measurements is dependent upon magnetic mineralogy, concentration, and grain size, and so provide a means of characterising magnetic assemblages. Precise evaluations of magnetic mineralogy and grain size within natural samples are, as yet, largely not possible using these techniques. This is due to the highly variable grain sizes, shapes, compositions, the variety of impurities, and the range of weathering products often encountered in natural sediment assemblages. These provide potentially very complex magnetic mineralogies, (Bradshaw and Thompson, 1985). Application of mineral magnetism in this area therefore relies principally upon sufficient differences between the characteristics of sources to allow differentiation between them. These characteristics must also remain conservative during the movement of the material

Chapter 1, introduction, aims, and reveiws

through a system and its deposition on the lake bed.

The first condition depends on the initial characteristics of the material, such as the bedrock geology, and the subsequent processes, such as weathering history, soil-formation, and, in relation to more recent sediments, the nature of anthropogenic activity particularly via industrial emissions. The magnetic characteristics of many minerals, and of "typical" rocks are known (for example Tables 3.4 and 4.3 in Thompson and Oldfield, 1986). Characteristics of soils have been investigated and frequently exhibit enhancement in the upper horizon through specific soil-forming processes (for example Maher and Taylor, 1988) as well as by the action of fire (for example, Longworth *et al*, 1979). These also often reflect a strong dependence on the underlying geology (for example, Dearing and Flower, 1982). However, the range of possible influences of these variables within a catchment is large. Although a general assessment of the rock types present and their likely contribution to the magnetic characteristics of soils and sediments in a catchment can sometimes be made using published sources, it is often necessary to evaluate empirically the magnetic characteristics of potential sediment sources within particular catchments.

The second condition (that magnetic characteristics acquired in particular environments remain conservative in transport and after their deposition as lake sediment) is dependent on several other factors and remains a subject of research (J.P.Smith, 1986). During transport, material may encounter a variety of chemical gradients and mechanical wear. These are likely to be of limited impact in the study of Holocene sediments, but ultimately the effects of diagenesis would place a time limit on the application of the method.

Simple diminution of grain size during movement within a sedimentary system may be accommodated for in interpretation of differences between sediment and source characteristics. Previous studies have established empirically that important minerals (in terms of their control of the magnetic characteristics of a bulk sediment sample) remain identifiable during their transit through lake-catchment systems (for example, Oldfield *et al*, 1978a, Bradshaw and Thompson, 1985.

Previous investigations utilising mineral magnetism in this fashion include: Oldfield *et al* (1983a), Oldfield *et al* (1985a). Thompson and Oldfield (1986, chapter 16) discuss details of a study based upon the Rhode River catchment in the United States; Bradshaw and

Thompson (1985) and Hirons and Thompson (1986) have investigated the role of particle size in relation to the study of sediment-source linkages in Icelandic and Irish lake sediments; and J.P.Smith (1986) studied source-sediment linkages using the combined chemical and magnetic characteristics of sediments from Shropshire/Cheshire meres.

Whilst the studies discussed above have demonstrated that the magnetic properties of materials can be retained in lacustrine sediments, such that changes in sources over time can be identified, the quantitative expression of proportions of source materials has presented a problem. Stott (1986, 1987) described magnetic mixing models for sediments deposited in a reservoir (the Macclesfield forest area, England). In this study, the mixing models was simple in that only two major sediment sources were operational in the system. Yu and Oldfield (1989) described a method where quantitative expressions of the proportions of source material in cores from the Rhode River (USA), were obtained for six major source categories. This represents a progressive use of mineral magnetism in the study of source-sediment relationships which has yet to be more widely and more routinely incorporated into the field as a whole.

The direct comparison of the mineral magnetic characteristics of lake sediments and source material is based on a detrital model of the accumulation of magnetic minerals. Diagenetic alteration or authigenic growth of magnetic minerals subsequent to sediment deposition would severely complicate the interpretation of lake-catchment sediment linkages, and several studies have identified the presence of authigenic magnetic minerals in certain lake sediments. Hilton and Lishman (1985) and Hilton *et al*, (1986a) found magnetic iron sulphide in surface sediments from Esthwaite Water, Blelham and Grasmere in the English Lake District. Authigenic greigite (Fe₃S₄) and rare pyrite (FeS₂) particles were observed in older Holocene sediments from Loch Lomond. The presence of authigenic greigite was inferred throughout much of the <u>overlying</u> Holocene material (Snowball and Thompson, 1988). Snowball and Thompson (1990) have observed greigite formed in cores from Lough Catherine, (Co. Tyrone, Northern Ireland). In their studies of authigenic magnetic

minerals in lake sediments, Snowball and Thompson (1988) also indirectly infer the presence of greigite which they think provided a significant contribution to the total magnetic assemblage in sediments from Lyn Geirionydd (North Wales), and much of the Holocene sequence of Lake Windermere. In the latter, the presence of greigite was inferred from small decreases in volume susceptibility of sediment cores over periods of four years or longer. These were thought to be due to the oxidation of greigite and thereby to the decrease in magnetic susceptibility over time.

Snowball and Thompson failed to identify authigenic magnetic minerals in the sediments of the oligotrophic Lake Svinavatn however, either by variation of volume magnetic susceptibility over time, or by thermomagnetic analysis of the lake sediments. The lowermost, least organic, of the Windermere sediments similarly exhibit no evidence of the formation of authigenic magnetic minerals. The sediments of oligotrophic lakes therefore appear unlikely to undergo authigenesis of this kind.

From the work of Snowball and Thompson (1988, 1990) and Hilton *et al* (1986a) it appears that the sediments of biologically <u>productive</u> lakes are most susceptible to the formation of magnetic iron sulphides - due to the activity of sulphate-reducing bacteria during the decomposition of organic matter. Hilton *et al* (1986a) have demonstrated a minor reduction in wet sediment volume susceptibility over time of recent sediments from Buttermere, suggesting the possible presence of an authigenic magnetic fraction. Changes in volume susceptibility over time in most cores may be expected owing to changes in sediment sample geometry, particularly if any variation in sediment moisture content occur. These may contribute to the changes in susceptibility measured by Hilton *et al*. Authigenic magnetic minerals therefore probably do not <u>dominate</u> the magnetic characteristics of the complete magnetic assemblage in oligotrophic Buttermere however, and Hilton *et al* (1986a) suggest that,

> "it is in any productive lake with anaerobic sediments that diagenetic magnetic minerals will contribute a major amount towards the magnetic records of a core".

> > (Hilton et al, 1986a, page 331)

Both Buttermere and Crummock Water are oligotrophic lakes (Luther and Rzöska, 1971), and available evidence suggests that their sediments are unlikely to be significantly affected by the formation of authigenic magnetic minerals, at least in significant quantities. In the following analysis, a model of detrital accumulation of magnetic minerals has therefore been used in the interpretation of source-sediment linkages based on their magnetic characteristics.

1.5. Lake sediments and sedimentation processes.

Sediment grain size distributions in lakes have been described as being ideally typified by a regular, concentric, "belt-like" pattern, (Figure 1.1). Coarser grains around a lake margin grade into finer towards the centre of the lake. This is a result of the normally zoned distribution of hydraulic energy within a lake. The breaker zone (the zone of highest energy) is located on the outer margins, and are followed by a zone above, and a zone below, wave base (the area of lowest hydraulic energy). In the absence of other influences, such energy zonation produces an areal gradation, where the coarsest beach lag around the lake margins grades into finer sediments below wave base in the deeper areas of the lake (Frey, 1969). Examples of lake sediments exhibiting approximate belt-like distributions are presented by Thomas and Jaquet (1975) for Lake Superior, and Jaquet *et al* (1975) for the Lake of Geneva, (Figure 1.1).

External and internal processes distort this "ideal" pattern, and an appreciation of the controls on actual sedimentation is necessary if sediments are themselves to be used as an investigative medium. Figure 1.2 illustrates the relationships between these controls. It can be seen that many of the factors are interrelated, although for the purposes of the following discussion, the major variables necessarily have to be considered seperately. For brevity, discussion is also directed towards those controls and processes which operate in lake-catchment systems with characteristics similar to the Buttermere and Crummock Water system.

The controls are:

i) Climate.

Climate indirectly affects lakes and their sediments by influencing many <u>external</u> processes, which include variable water supply, and weathering and rates of erosion in the catchment. Climatic effects on <u>internal</u> lake processes may be direct, through their influence on lake water thermal characteristics, or less direct, such as through their influence of biological characteristics of the lake, as illustrated in Figure 1.2.

Climate directly affects lakes internally by heating or cooling of lake water thereby



Figure 1.1. Grain size distribution in the Lake of Geneva, showing approximate concentric "belt-like" distribution of decreasing mean grain sizes towards the deep lake centre, (see text for discussion). After Jaquet *et al*, (1975).



Figure 1.2. Schematic representation of the major controls on lacustrine sedimentation and their interrelationships. (Controlling factors are discussed individually in the text).

influencing the development of the thermal characteristics of lake water. In conjunction with characteristics such as basin morphology and climatically-driven variables such as wind, this may lead to the formation of thermal stratification in freshwater lakes of low sediment input which exerts a profound influence upon both the distribution and the form of sediments.

Through its influence on catchment geomorphic processes, seasonality of climate promotes both marked differences in types and rates of sediment input). It also affects the formation of thermal stratification in lakes which may enhance sediment sorting by periodically trapping finer particles in discreet water layers. Both factors encourage the development of rhythmic variations of sediment texture, and hence the development of laminae. The Lake District is presently subjected to a North Temperate maritime climate which is not strongly seasonal, hence the seasonality of sediment input is not marked. Sediment sorting may occur during lake stratification although the uniformity of other Lake District Holocene sediment sequences (Pennington. 1981, and discussed in section 1.1) suggests that the effects of water stratification are not strongly developed in the lakes of this region.

The effects of wind in a lake are dominantly in the form of surface waves, which can drive currents extending throughout the water column, and in the production of seiches, which are oscillatory changes of water level within a lake. In general, wind influence becomes of greater significance for lakes of increasing size. The relatively small size of most lakes inhibits formation of long period waves characteristic, for example, of the marine environment (Sly 1978).

The effects of surface waves depends greatly on the morphological characteristics of the lake. Direct influence on bottom deposits is limited to broad shallow lakes, where waves of longer wave length can develop, and where effective wave depth may approache the sediment/water interface. Examples of such lakes include Lake Balaton (an extreme example from Hungary), and Bassenthwaite and Derwentwater in the English Lake District (mean depths 5.3 m and 5.5 m, and surface areas of 5 km² and 5.5 km²

respectively, lake locations shown on Figure 2.1). All exhibit considerable disturbance of the bottom deposits as a result of wave action, (Sly 1978, Pennington 1981). The depth of the Buttermere and Crummock Water lake basins will inhibit any direct disturbance of the bottom sediments from wind-induced waves.

Production of seiches is caused by the effective "piling up" of water at one end of a lake owing to wind action. When the wind ceases or slackens, the lake surface oscillates along the long axis in a "seesaw-fashion" (Lindell 1980) as the water returns to the horizontal. If the lake is stratified, internal seiches may also occur, typically with an amplitude between one and three orders of magnitude greater than the surface form (Lindell 1980, Lemmin 1987). Attempts have been made to model such seiches (for example, Horn *et al* ,1986) who based their analysis upon data obtained from measurement of internal waves in the Zürichsee. From this they suggest that, whilst surface seiches are not of great significance, internal seiches may form along the thermocline and so promote vertical mixing between layers. Vertical mixing of particulates originally trapped within the discreet water bodies allows the lake-wide dispersal of this fine sediment (by horizontal water movements). This may thus form an important process for the distribution of fine sediment over an entire lake.

ii) Geology.

The principal effects of geology in this context are in influencing erosion and catchment geomorphology and hence sediment type and supply. Through its control on erosional and geomorphological processes, catchment geology also influences the development of the lake basins, and thereby both the efficiency of sediment retention (primarily dependent on the morphometry of the lake basin) and the effects of wind.

Geology also influences chemical coagulation of sediments (greatest in hard waters with low dissolved organic carbon contents, Weilenmann *et al*, 1989) and sediment pelletisation by small organisms (Smith and Syvitski, 1981) via the control of bedrock on water chemistry and nutrient supply. Water chemistry and nutrient supply are also influenced by many other factors, notably, the climate, floral and faunal development. These factors are

all important in detrmining sedimentation rates and lake trapping efficiency, hence the distribution and form of lake sediments.

iii) Geographical location and aspect.

Geographical location, in conjunction with lake basin size and morphometry, is of importance in its effect on the magnitude of the Coriolis force; and in determining the climate experienced by the area. The Coriolis effect is a product of latitude and the earth's angular velocity, (in the northern hemisphere) deflecting aqueous inflows to the right-hand side of a lake. The magnitude of this force which increases with increasing latitude. The response seen in lake sediments are increases in sedimentation rates and mean grain size along right-hand shores (in the northern hemisphere), as, for example, observed in the recent sediments of Bow Lake, Alberta, (N.D.Smith, 1981).

The Coriolis effect, in conjunction with large inflowing rivers, may exert sufficient force to promote an horizontal river-plume driven lake-wide circulation. Such effects have been recorded in the Swiss lakes Brienz and Briel (Wright and Nydegger, 1980) and Kamloops Lake, British Columbia (Hamblin and Carmack, 1978), where the sedimentary effects where seen in the deposition of coarser sediments along the northern shore, (the "right-hand-side" relative to the inflowing river plume).

Aspect is of particular importance in relation to microclimate, such as the effect of wind surface stress, and also for the development of geomorphic processes within the catchment - with consequent implications for rates and types of sediment supplied. For example, the long axes of Buttermere and Crummock Water, are orientated north-west and north-north-west in an area of predominantly westerly winds. They are therefore potentially subject to significant influence from wind surface stress. The basin profiles however (chapter 2, section 2.3), place much of the lake volume below wave base. Hence wind effects will be restricted to the formation of seiches - which may in turn promote mixing within the epilimnion and between the epilimnion and deeper waters, (see i, climate, above).

The north-easterly aspect of the southern side of the Buttermere trough also ensures that a considerable area of the valley sides are well-placed for the development of local periglacial and glacial features (Clough, 1977). For example, well-developed deposits from corrie glaciers originating from the Loch Lomond Readvance are evident on certain north-east facing slopes (see chapter 2). Corrie development was generally restricted to micro-climatically "optimal" locations (Sissons, 1980; Clough, 1977). Development of such features within a catchment is most significant in terms of supply, which, characteristically in active glacial areas, is highly seasonal and therefore likely to promote the formation of distinct sedimentary laminae.

iv) Basin morphometry.

Basin morphometry affects sediment type and accumulation through its interaction with the other major variables shown in Figure 1.2. Via depth, morphometry determines the susceptibility of any deposit to disturbance through wind and wave action, and is of critical importance in the accumulation and preservation of stratified sediments.

O'Sullivan (1983) identified the specific morphometric conditions of a flat lake bottom (preventing slumping and lateral creep of sediments) and sufficient basin depth (avoiding mixing by storm waves or lake circulation) as among the most important criteria for the production and preservation of lacustrine laminae. These are fulfilled in both Buttermere and Crummock Water (chapter 2, section 2.3). Depth is also a major constraint in the development both of stratification and of circulation patterns, which in turn affect the distribution and form of sediments, (discussed below).

v) Thermal characteristics.

Insolation warms surface waters which, when heated above 3.86°C (the temperature of maximum density), become relatively less dense than the cooler water beneath, and forms a more buoyant, relatively stable warmer surface layer called the epilimnion. This overlies the cooler hypolimnion, (Hutchinson, 1957). Other factors, such as basin characteristics

and wind stress, permitting, continued warming of more buoyant surface waters leads to a more clearly defined distinction between epilimnion and hypolimnion, and the development of stable thermal stratification. This usually persists until seasonal changes in insolation and air temperature induce cooling of the upper layers which, as they near the temperature of maximum density sink and may mix with the hypolimnitic waters below.

Stratification of lake waters is a major influence on the form of inflowing plumes take (namely, underflows, interflows, or overflows). Plumes flow to, and subsequently along, layers of water of the same density. Underflows move downwards until equilibrium density is achieved, and may thus travel along shallow slopes for relatively long distances. They cannot climb mid-lake barriers however, or lateral slopes owing to their reliance upon their relatively greater density to promote downward movement (Matthews, 1956, Stürm and Matter, 1978).

Where sediment plumes develop as underflows, linear increases of mean grain size away from the point of plume entry will be most marked and will extend furthest into lake basins. Mixing with lake water is minimised. Hence linear "fingers" of sediment of relatively coarser mean grain size will be superimposed upon a general decrease in mean grain sizes from the lake margins inwards. For example, in the Lake of Geneva, a combination of relatively high sediment load and strong temperature difference habitually promotes the development of the inflow of the River Rhóne as an underflow. Consequently, at the eastern end of the Lake of Geneva, there is an ingress of sediment of coarser mean grain size than that found in the surrounding lake, (Houbolt and Jonker, 1968). This distorts the concentric pattern of decreasing sediment grain sizes displayed elsewhere in the lake.

The development of long-term underflows may also generate lake-wide circulation patterns, but in the vertical plane, denser inflowing water plunging to the lake bottom promotes a higher level return current (Carmack *et al*, 1979). Laboratory experiments have shown that the water surface in the plunging area is depressed, necessitating the return flow (Friedman and Sanders, 1978, p.511). This is a process also observed in nature by

Gould (1960) at the inflow of the Colorado River into the Hoover Dam. Development of such a current system will encourage greater mixing and the subsequent wider distribution of nutrients and finer sediments entrained in the return flow in the upper lake waters (Carmack, *et al*, 1979).

Formation of overflows and interflows promote either rapid transit of fine material out of a lake, or a more complete mixing of sediment over the entire lake water surface. In the former case, sediment trapped in the upper layers may be expelled as water is discharged. This material is thereby lost to the immediate system (Weirich, 1985). In the latter case, there is a greater opportunity for complete mixing across the lake surface by wind and wave action while the sediment remains trapped in the epilimnion, with subsequent wider distribution on the breakdown of water stratification.

N.D.Smith (1981) observed changes in sedimentation patterns in Bow Lake, Alberta, where interception of sediment-rich water by a newly-formed upstream lake caused a reduction in density of the inflowing tributary. This in turn promoted the development of interflows moving above the hypolimnion, an effect expressed in the sediments by a change from thicker laminations preferentially formed in the lowest parts of the lake floor to thinner layers with a wider distribution generally unaffected by bottom topography.

Similar processes may have operated in Buttermere and Crummock Water, during the change from denser, glacier-derived inflows to river inflows at the Pleistocen/Holocene boundary, with Buttermere periodically acting as an upstream sediment trap for Crummock Water. Variations in relative densities of inflows and lake thermal structure would result in changing sedimentation patterns in each case (Pharo and Carmack, 1979).

Hilton (1985) developed predictive models of expected sediment redistribution processes according to lake basin morphometry. These suggest that sediment redistribution processes in lakes with basin characteristics similar to those of Buttermere and Crummock Water, are dominated by intermittent complete mixing. This occurs during periods of overturn, when the fine material trapped in the upper layers becomes available for redistribution over the entire lake bed. The process provides pulses of finer sediment for deposition after each overturn, and results in a vertical sediment sequence characterised by sediment couplets of coarser material overlain by finer sediment derived from overturning lake water.

The effects of variable sediment input on sediment distribution were not considered in Hilton's models, but have been discussed by Stürm (1979). He demonstrated that the interaction of cycles of sediment input (for example, during either continuous or discontinuous input during different seasons) with changing patterns of lake stratification may result in characteristic grain size variations in the resulting sediment. Such "micro-structures" may be recognised in thin sections, and therefore potentially afford the opportunity of reconstruction of past lake/catchment characteristics via detailed (microscopic) examination.

Stürm neglects, however the possible influence of underflows produced by sediment-rich (and therefore relatively dense) inflows, and slope (particularly delta-slope) failure within the lake body. Such episodic density currents have been termed "sediment density surges" by Pharo and Carmack (1979), in order to distinguish them from the more continuous currents arising from river inflow. Sediment density surges produce thicker beds of coarser mean grain size than would normally occur at a particular depositional site. As with density underflows, movement, and hence sediment deposition, will be restricted to the deeper lake bed areas.

vi) Biological factors.

Biological processes are dependent on a number of other controls (for example, climate, nutrient supply, and water stratification) and are often expressed directly in lake sediments by the deposition and preservation of pollen and algae, which may contribute to the formations of distinct seasonal laminae (O'Sullivan, 1983). Organisms may also exert important effects on sedimentation processes through pelletisation of fine suspended sediment. Such material, temporally suspended in the epilimnion, may be lost to the system as water is flushed from a lake, or be transferred to profundal waters during phases
of lake overturn. The "premature" deposition of such fine sediment has been cited as an important process in certain glacial lakes (Smith and Syvitski, 1981), and its effects in other lake systems requires further investigation.

The significance of bioturbation of sediments depends on other lake characteristics, particularly the availability of oxygen in profundal waters. Such activity seems unlikely to cause widespread redistribution of sediment, but may be of great importance in terms of the low preservation potential of delicate sedimentary structures such as thin laminae. Biological activity will have less impact where sedimentation rates are high, and will be discouraged by the formation of stable stratification and incomplete lake circulation (meromixis), when the oxygen recharge potential in profundal waters is reduced.

The trophic status of a lake is a general measure of its nutrient load, may therefore reflect the level of biological activity, and hence is of significance for the potential for preservation of sedimentary structures (with respect to bioturbation). Oligotrophic lakes, such as Buttermere and Crummock Water, should posses a high preservation potential, low trophic status suggesting low levels of bioturbation. Eutrophic lakes however, frequently suffer oxygen depletion in the deep parts of the water column, discouraging the bioturbating effects of macro-organisms. Certain types of biological activity may be widespread in oligotrophic lakes. For example, Ashley (1975) has observed trace fossils in supposedly barren ancient glacial lake sediments, and Duck and McManus (1987), and Morrison (1987), have reported chironomid larva trails in shallow, oligotrophic, proglacial lake sediments. Significantly, Duck and McManus (1984) observe that chironomids may prefer the fine-grained substrate characteristic of the <u>deeper</u>, quieter water of lake basins. Contemporary studies are less likely to directly observe such trails in these more inaccessable deeper waters.

In summary, whilst a number of controls may interact to determine the final form and distribution of lake sediments in a complex way (Figure 1.2), many of their influences on the form of the deposited sediment can be predicted. It is therefore practicable to achieve

the aims of this study, (section 1.2), to determine the state and influence of both internal and external controls by working back from examination of lake sediment type, form, and distribution.

2. LAKE AND CATCHMENT CHARACTERISTICS.

Introduction.

This chapter describes the characteristics of Buttermere, Crummock Water and their catchment, and what is known of recent (Holocene) developments within it. Reference is made to controlling factors discussed in Chapter 1 (section 1.5).

2.1. Catchment characteristics.

2.1.1. Geology.

The Buttermere/Crummock Water catchment is situated in the north-west of the English Lake District, (Figure 2.1). Its solid geology based on the published geological map for the area (Arthurton *et al*, 1980) is shown on Figure 2.2. The geology of the Lake District is discussed by Mosely (1978). The Buttermere and Crummock Water catchment contains rocks from three main geological formations:

- (1) Skiddaw Slates from the Skiddaw Group;
- (2) Borrowdale Volcanic Group;
- (3) the Ennerdale Granophyre.

(1) Skiddaw Slates.

This geological Group comprise the oldest rocks exposed in the Cumbrian Mountains, which extend from Cleator Moor on the west of the Lake District to Troutbeck in the east. They form the greater part of the bedrock in this catchment, namely the area to the north and north-east of the two lakes, and immediately to the south-west of Buttermere (Figure 2.2).

The Skiddaw Group comprises a range of mudstones, siltstones, sandstones, and conglomerates lying beneath the Borrowdale Volcanics, and are considered to represent a deep-water turbidite suite from the south-east margin of the proto-Atlantic (Jackson 1978, p.81). Locally, spillitic lavas and tuffs are found, and in the upper part of the sequence, intercalations of andesite lavas and tuffs. However, mudstones and siltstones are the major



Figure 2.1. Location of the English Lake District, and the lakes.



Figure 2.2. Solid geology of the Buttermere and Crummock Water catchment. (Redrawn from Arthurton *et al*, 1980).

units present in the Buttermere/Crummock Water catchment, (Arthurton et al, 1980).

(2) Borrowdale Volcanic Group.

The Borrowdale Volcanic Group were previously known as the Borrowdale Volcanic <u>Series</u>, (Millward *et al*, 1978). Following the revision of terminology presented by Millward *et al*, (1978) they are now termed Borrowdale Volcanic <u>Group</u>. Rocks of this group outcrop in the extreme east and south-eastern parts of the Buttermere and Crummock Water catchment (Figure 2.2).

The lithologies comprising the group are varied. Formations are characterised by a common association of lithologies rather than a particular rock group. This is a product of complexities resulting from the operation of several volcanic centres at any one time, with later Caledonian folding and faulting, (Millward *et al*, 1978). Rocks in this group characteristically weather to produce a "craggy" landscape with development of a thin soil cover.

The two formations of the Borrowdale Volcanic Group present in the Buttermere and Crummock Water catchments consist of undivided intermediate tuffs and andesite lavas (Arthurton *et al*, 1980). The latter tend to be massive, and the tuffs are mostly strongly cleaved lithic tuffs with a high chlorite content, (Millwood *et al*, 1984). These form the "green slates" which have been widely quarried in the Lake District, and which until recently (1986) were quarried at Honister in the south east of the Buttermere catchment. These rocks therefore provide a large reservoir of chlorite-rich material which has contributed greatly to the sediments in the two lakes studies here.

(3) The Ennerdale Granophyre.

The Ennerdale granophyre is emplaced immediately to the south-west of Buttermere, contributing to the high topography on the south-western shores of the two lakes. The bulk of the formation is a fine-grained, pink, quartz-alkali, feldspar rock, common accessory minerals being chlorite (replacing biotite) and epidote, with traces of magnetite, apatite, sphene, and zircon (Firman 1978, p.146).

The age of the Ennerdale granophyre is not certain. K-Ar dates of 334 ± 18 Ma (Brown et

al, 1964) have been questioned by Firman (1978, p.146), who grouped this deposit with the "older" Lake District intrusions - of possible Ordovician age, pending further isotopic dating studies.

The sediment derived from all these rocks and supplied to the lakes may therefore be of varied composition. The major components of the lake sediments formed from them should consist of quartz, together with feldspars and clay minerals. Of the latter, chlorite should be present in high proportions, reflecting its high concentration in the Borrowdale Volcanic Group (Millward *et al*, 1978, p.115). Afurther source of replacive chlorite is the Ennerdale granophyre (Firman 1978, p.149). XRD results from bulk analyses of samples throughout the lake sediment sequence (Chapter 4) broadly confirm this sediment composition.

Sediments from catchments of slate bedrock (*sensu stricto*) have been shown to carry low magnetic susceptibility, a product of the small input of primary ferromagnetic mineral (Figure 2.3, Thompson and Oldfield, 1986, p.103, their Figure 3.2, Stott, 1987). In the Buttermere and Crummock Water catchment area, sediment is derived from the Ennerdale granophyre and Borrowdale Volcanic Group andesites, as well as the Skiddaw Slates, so that the amount of primary ferromagnetic material likely to be present in the lake sediment cores is somewhat greater, with a corresponding increase in the susceptibility of bulk sediment samples. Secondary soil-derived minerals may further contribute to the magnetic component of the sediments, as well as atmospheric produced during the Industrial Revolution, (Oldfield *et al*, 1978b, Oldfield *et al*, 1981, Goldberg *et al*, 1981).

Expected susceptibility values for sediment from this catchment may therefore lie in the range $1.2 - 22 \ 10^{-8} \text{m}^3 \text{kg}^{-1}$, (Figure 2.3, and Thompson and Oldfield 1986, their table 10.2). Samples collected for mineral magnetic analysis are therefore likely to fall well within the sensitivity range of the sensing equipment used (Appendix 1).

Remanence-carrying minerals Specific susceptibility 10 ⁻⁸ m ³ kg ⁻¹					
iron	2 x 10 ⁷				
goethite (aFeOOH)	70				
haematite (αFe_2O_3)	60				
maghematite (γFe_2O_3)	4 x 10 ⁴				
magnetite (Fe ₃ O ₄)	5 x 10 ⁴				

Other iron-bearing	minerals Specif	fic susceptibility	$10^{-8} \text{m}^{3} \text{kg}^{-1}$
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 biotites (Mg, Fe, Al) silicates	5> 95
olivines (Mg, Fe)SiO ₂	1> 130
pyroxenes (Mg, Fe) ₂ Si ₂ O ₆	5> 100

Other rocks, minerals, and materials Specific susceptibility 10⁻⁸m³kg⁻¹

feldspars (Ca, Na, K, Al) silicates- 0.5"clay"10illite~ 15kaolinite- 2quartz (SiO2)- 0.6water (H2O)- 0.9granite~ 300*Ennerdale Granophyre (microgranite)~ 260siltstone20*Skiddaw slate~ 13*Borrwdale Volcanic Group slate~ 20		
"clay"10illite~15kaolinite- 2quartz (SiO2)- 0.6water (H2O)- 0.9granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	feldspars (Ca, Na, K, Al) silicates	- 0.5
illite~ 15kaolinite- 2quartz (SiO2)- 0.6water (H2O)- 0.9granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	"clay"	10
kaolinite- 2quartz (SiO2)- 0.6water (H2O)- 0.9granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	illite	~ 15
quartz (SiO2)- 0.6water (H2O)- 0.9granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	kaolinite	- 2
water (H2O)- 0.9granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	quartz (SiO ₂)	- 0.6
granite~300*Ennerdale Granophyre (microgranite)~260siltstone20*Skiddaw slate~13*Borrwdale Volcanic Group slate~20	water (H ₂ O)	- 0.9
*Ennerdale Granophyre (microgranite) ~260 siltstone 20 *Skiddaw slate ~13 *Borrwdale Volcanic ~20 Group slate ~20	granite	~300
siltstone 20 *Skiddaw slate ~13 *Borrwdale Volcanic ~20 Group slate	*Ennerdale Granophyre (microgranite)	~260
*Skiddaw slate ~13 *Borrwdale Volcanic ~20 Group slate	siltstone	20
*Borrwdale Volcanic ~20 Group slate	*Skiddaw slate	~13
	*Borrwdale Volcanic Group slate	~20

Figure 2.3. Typical mass specific susceptibility values for common matierials and some rocks and minerals found within the Buttermere and Crummock Water catchment.

(* Data from this project - see appendix 6 and discussion in section 4.4.3, other data from Thompson and Oldfield (1986) their tables 3.4, 4.3, and 4.6).

A comparison of typical specific susceptibilities for various lithologies including those found within the Buttermere/Crummock Water catchment area is provided in Figure 2.3. From this, it is suggested that differences in mineral magnetic characteristics between the major geological groups present in the Buttermere and Crummock Water catchment will allow major changes in sediment sources for the Buttermere and Crummock Water sediments to be identified using the methodology discussed in Chapter 1 (section 1.4).

2.1.2. Geomorphology.

Topography and place names of the Buttermere and Crummock Water catchment area are shown on Figure 2.4. The catchment covers a total of 43.6 km². There are considerable variations in relief within this area, varying from 852 m on Grasmoor (NY 174 204) to 100 m around the lake shores. The Buttermere trough, which contains both lakes, is orientated north-west/south-east, turning north-north-west/south-south-east approximately 1.5 km from the southern end of Crummock Water. Processes of glacial erosion and deposition have been instrumental in the formation of this landscape, and are discussed below.

i) Glacial modification.

The valleys of the major lakes in the Lake District are all considered to be a product of glacial erosion, ice having radiated from the central mountain massif (cf. Marr 1916, Pennington 1978b). The Buttermere trough exhibits some well-developed characteristics of glacial modification, the valley morphology suggesting a major source for the valley glacier from an ice dome located towards the east.

Glacial overdeepening has been of great significance in the upper reaches of the Buttermere trough, and valley profiles, characteristic of erosion by glacial processes are illustrated in Figure 2.5. Form ratios on selected profiles (Figure 2.5) are typical of glaciated valleys





Figure 2.5. Valley cross-sections along the Buttermere trough, selected sections shown with calculated form ratios and parabolic curve functions,

(form ratios >~0.2, Graf, 1970).

The general valley profile, with a well-developed head (exhibiting the highest value form ratios of the trough), is typical of Icelandic-type glacial troughs associated with erosion beneath ice caps. Ice spilling over the trough end causes the greatest erosion in this part of the valley (Linton, 1963, Sugden and John 1976, p.178). This is in agreement with the ice dome mechanism generally proposed for the substantial formation of the Lake District glacial valleys, (Pennington, 1978b).

Corrie formation has also occurred on north-east facing slopes above the southern shore of Buttermere and Crummock Water (Sissons, 1980). These were last active during the Loch Lomond Stadial (*op cit*).

Depositional features associated with this phase of glacial activity are still identifiable in the Buttermere and Crummock Water catchment. Remnants of a cross-valley (terminal) moraine are present along the wester end of Crummock Water, and of a lateral moraine along the north-west margin of the lake. Where sections are exposed, such as at the lake shores, moraines are seen to be composed of clay diamicts which are usually oxidised in their upper, more weathered, zones.

Trial pits dug during catchment substrate sampling revealed the presense of similar clay-diamicts throughout much of the catchment, for example, at localities in the valley head area, and underlying the high peat area of Buttermere Moss to the north of Buttermere, (Chapter 5, Figure 5.1). Glacial diamicts therefore prabably cover much of the catchment and form the immediate parent material for many of the contemporary soils. They are likely to have formed a significant source for the lake sediments, particularly during the Loch Lomond Stadial (when glacial activity was generally restricted to development of corrie glaciers (Sissons, 1980)) and immediately following de-glaciation when regional pollen analytical evidence has indicated that vegetation cover was initially sparse and land surfaces therefore relatively unstable (see section 2.2).

Further modification of the valley occurred during the Loch Lomond Stadial with the

development of corrie glaciers, generally restricted to the north-east facing slopes optimal for development of climatically sensitive corries (Clough 1977). Three corries in the Buttermere and Crummock Water catchment, Ling Comb (OS reference NY 175 145); Chapel Crags Cove (NY 166 134); and Burtness Comb (NY 159 159) (Sissons, 1980), have been identified as active during this period. All three exhibit well-developed steep head- and side-walls, with hummocky moraine and linear till ridges preserved in their mouths. Only Chapel Crags Cove retains a corrie lake (Bleaberry Tarn). Ling Comb presently drains into Crummock Water, and Chapel Crags Cove and Burtness Comb directly into Buttermere.

Such corries, and the sediments eroded by the glaciers within them, must have been significant sources for lake sediments deposited during the Loch Lomond Stadial. In Windermere for example, the Late-glacial sequence of laminated clay is attributed to material supplied by active corrie glaciers (Pennington, 1981).

ii) Post-glacial modification.

The most dramatic post-glacial modification of the Buttermere/Crummock Water valley was the development of the two lakes themselves, and of the large alluvial fans at Gatesgarth (NY 194 144), Hassness (NY 187 157), Rannerdale (NY 164 186).and Buttermere Dubbs south-west of Buttermere village (NY 170 168). (Buttermere Dubbs divides the Buttermere trough, allowing the development of the two separate lakes). The forms are of low relief, typical gradients in the main plain areas of 1:150, 1:380, 1:90, and 1:300 respectively. At present they appear stable, and bear little relation to the contemporary streams flowing across them. The fans are therefore relict features, probably formed in the early Holocene after the retreat of the main valley glacier. It has been suggested that they consist of torrential meltwater and solifluction debris formed during the climatic amelioration following the Late-glacial (Dodd 1982, p.76).

The result of all of these processes of erosion and deposition has been to provide a wide,

deep, and flat-floored valley, sub-divided by the Buttermere Dubbs alluvial fan, within which Buttermere and Crummock Water have developed. The specific morphometric characteristics of the two lakes are discussed in section 2.3, "Lake characteristics", following.

2.2. Regional vegetation history and contemporary land use.

Aspects of the vegetational and climatic history of the Buttermere and Crummock Water catchment can be reconstructed with reference to investigations from other Lake District catchments, notably the extensively-studied and independently-dated sediment sequences of Lake Windermere and the tarns and bogs within its basin (Walker and West, 1970; Pennington, 1981; Haworth, 1985). These studies employ a range of biological variables to determine the nature of ancient floristic communities and, thereby, the influence of regional factors such as palaeoclimate and local effects such as the nature and stability of soils.

Changes in these factors may cause significant variations in the characteristics of lake sediments (discussed in section 1.5). Major changes identified in previous studies, mainly from within the Lake District, are summarised below.

i) The Late-glacial, (circa 11 000 - 10 000 B.P.).

Diatom assemblages in sediments of this period are of low diversity or absent (Haworth, 1985). Pollen and insect assemblages provide evidence of cold climatic conditions, with grass heath vegetation containing ground herbaceous taxa such as *Rumex* and *Artemesia* (Coope and Pennington, 1977, Pennington, 1981). Arboreal pollen (*Pinus*) has been recorded in sediments of this age, but lack of corresponding macroscopic remains suggests that this pollen represents distant rather than local sources, and thus that Pine woodland was not present in the region at this time (Pennington, 1970). The existence of such sparse, heath vegetation suggests the presence of a relatively unstable and easily eroded land surface, particularly following probable disturbance by frost.

On the basis of these characteristics alone, lake sedimentation rates would be expected to be high, and the resulting sediments highly minerogenic. Lack of abundant diatoms suggests stressful lake water conditions, which, in view of evidence from the pollen record, would be turbid, as well as cold.

ii) The early Post-glacial, (circa 9 700 - 5 000 B.P.).

Changing environmental conditions in the early Post-glacial (*circa* 9 700 - 5 000 B.P.) are recorded by increases in numbers and diversity both of diatom and pollen assemblages. Diatoms directly record changes in lake water quality, with the increased Post-glacial diversities attributed particularly to enhanced nutrient loads within lake waters and a reduction in turbidity following reduced sediment loading (Haworth, 1985). Changes in the water quality reflect external factors, namely the development of organic soil profiles, increased delivery of organic material to lakes, and the stabilisation of land surfaces. The latter was initially brought about by the development of pioneer vegetation including herbs like grasses, sorrels, and shrubs such as Juniper, and subsequently by a complete forest cover (up to altitudinal limits of *circa* 850 m). Changes in forest composition from birch and locally pine, to elm and oak, and alder in particular are indicative of regional climatic warming and increases in precipitation (Pennington, 1970, 1981).

These climatic and vegetational changes resulted in the stabilisation of land surfaces and the increased delivery of organic matter to Lake District lakes. The resulting more organic sediments accumulated less rapidly than the underlying glaciogenic material (Pennington, 1981). The rapid change from the clay- to mud-texture seen in the sediments at this (Late-glacial/Post-glacial) transition has been attributed to the input of terrestrial organic matter containing humic compounds which caused flocculation of clay particles (Holmes, 1968).

From *circa* 7 500 B.P., diatom assemblages record a change from alkalinic to acidic or indifferent taxa which flourished in less nutrient-rich waters, and whose presence implies the prevalence of more oligotrophic lake conditions. These changes are attributed to (a), the reduced inputs of base-rich freshly-eroded bedrock material and vegetative stabilisation of glacial deposits, and (b), reduced input of leached bases from soils following their gradual impoverishment during leaching in the previous *circa* 2 000 years (Haworth, 1985, p.69). Such assemblages, with abundant *Cyclotella*, still characterises the contemporary sediments of Crummock Water.

Evidence exists for climatic variation within this period, following initial increases in temperature and moisture, although there is variation between sites, reflecting in part the difficulty in distinguishing in the pollen record between successional and climatically induced vegetation change. Changes in both sedimentary iodine and in pollen stratigraphy were tentatively attributed to decreased precipitation by Mackereth (1966b) and Pennington (1970). Similar reductions in iodine are also associated with a minerogenic layer in four tarns in the south and south-west of the Lake District and dated at 7560 ± 160 B.P. in one of them, Burnmoor Tarn (Pennington, 1970). These layers were therefore interpreted as reflecting lowered lake water levels during a period of decreased precipitation (*op cit*). Lake sediments from this period therefore successively reflect (a) the major (Late-glacial to Post-glacial) change in climate and catchment erosion, (b) the gradual stabilisation of land surfaces resulting in a reduction in sediment input, as well as (c) less well-understood variations in climatic conditions which, at some sites, have resulted in lowered lake water levels with consequent changes in type and style of sediment deposition.

iii) Mid Post-glacial to present, (from circa 5 000 B.P.).

Towards the end of the mid Post-glacial (*circa* 5 000 B.P.) variations in regional pollen assemblage provide evidence of a number of general environmental changes. These are characterised by (a) a reduction in elm pollen (the widespread *circa* 5 000B.P. Elm Decline (Pennington, 1981), and (b) an increase in the pollen of grasses and a general decrease of tree pollen, particularly of oak (Oldfield, 1963; Pennington, 1970). The causes of the Elm Decline have not been clearly identified. It may reflect a combination of subtle climatic change, the selective destruction of the tree by Early Neolithic peoples, or the occurrence of a pathogen such as Dutch Elm Disease (Pennington, 1981).

From *circa* 5 000 B.P. therefore (this date representing a mean of radiocarbon dates for the Elm Decline in the Lake District (Pennington, 1970)), reduction in forest cover occurs in the Lake District, with cultivation of cereals and establishment of grazing. Vegetational change of this type is recorded by increasing rates of sedimentation, and, generally by

reductions in the organic content of limnic sediments. In extreme cases, increased input of mineral material in this horizon has led to lithological changes (from organic mud to clay-mud) as recorded at Barfield Tarn (Pennington, 1981). This has been interpreted as due to ploughing of slopes during prehistoric and early historic agriculture (Pennington, 1981).

Following the Elm Decline, the initial deforestation, and early farming, studies of sediments from the Lake District emphasise the importance of local rather than regional effects, as lakes responded to more localised anthropogenic activity within particular catchments. In the north-east of the Lake District, Haworth and Allen (1984) for example, observe an oscillation of diatom assemblages above the Elm Decline, which they interprete as the biological response to temporary lake enrichment from the introduction of unweathered sediments after a series of erosional events (probably associated with clearances within the Ullswater catchment).

Specific studies of prehistoric land use and vegetation changes in the Buttermere and Crummock Water catchment have not been published. A possible Stone Age burial mound, and the remains of Romano-British settlements located on Grasmoor and in the area at the western end of Crummock Water, are identified by Collingwood (1928). These indicate local occupation and therefore some degree of change within the Buttermere and Crummock Water catchment certainly from around 2 500 B.P. Evidence of further influence is provided by records of the use of forests around Crummock Water for swine from 1100 A.D. (*op cit*, p.77), and by a licence for fortifications to a manor-house at the western end of Crummock Water during the 14th century (*op cit*, p.105). Pennington (1981, p.214) reports a "thick deposit of mineral sediment associated with...mediaeval cultivation" in Loweswater (to the west of the Buttermere and Crummock Water. There appears to have been a much larger human population in these valleys during the Medieval period than is presently found (Pennington, 1981).

These intermittent references demonstrate that there have been several intermittent periods (if not continuous) human occupation within the Buttermere and Crummock Water

catchment since the Elm Decline. Sediments post-dating this event are therefore likely to record changes similar to those observed in other Lake District lakes. These exhibit increased rates of sediment accumulation, and deposition of more highly minerogenic sediments following the first phase of deforestation and ploughing by Early Neolithic farmers, such activity being concentrated in the western Lake District (Pennington, 1981). The shallower lakes responded with more dramatic changes in lake biota during this period, owing to a reduction in the hypolimnion volume and the consequent development of reducing conditions at the mud-water interface (increasing seasonal releases of nutrients from the mud surface, Mortimer (1941)).

More profound ecological changes were produced following the settlement of Norse migrants (*circa* 900 - 1000 A.D.). More extensive forest clearance led to enhancement of sediment accumulation rates (by a factor of ca. four). Anomalous ¹⁴C dates are associated with the inwash of "old" soil-derived organic material in Windermere, Ennerdale Water, and Loweswater (Pennington, 1981). Higher sedimentation rates were maintained with the establishment of mediaeval settlements and agriculture in many areas, including the Buttermere/Crummock Water catchment. By the 1600's, effects of human activity were enhanced by the establishment of quarrying and mining activities in the valley head area. Ore from the Borrowdale Volcanic Group and Skiddaw Slates was worked (Rollinson, 1967, p.104). During the 1960's and 1970's, prior to installation of settling ponds, this resulted in the formation of sediment-rich plumes in Buttermere (Pennington, 1987, pers. comm.). Quarrying ceased in the Buttermere and Crummock Water catchment in 1986 when the Honister Green Slate quarry closed.

After about 1900 A.D., changes in certain of the Lake District lake sediments have been observed, with the development of "black ooze" overlying the organic Post-glacial sediments (Pennington, 1943). This is only seen in sediments of lakes located in the more intensively-settled catchments and is related to input of anthropogenic nutrient-rich effluent (Pennington, 1981).

The present land use in the Buttermere/Crummock Water catchment is devoted mostly to sheep on the mountain and hill slopes, with hay meadows and limited cattle grazing on the more fertile alluvial plains surrounding the two lakes. These areas cover *circa* 1.6 km², approximately 3.7% of the catchment area. Foster *et al*, (1989) have demonstrated that "poaching" (that is, the trampling and disruption of soil by animals) around lake margins by cattle can result in significantly enhanced sediment inputs. The effects in this catchment are likely to be restricted however, becuase of the limited grazing around the lake shores. Areas of peat such as Buttermere Moss (NY 194 169) and over Melbreak (NY 147 190) have developed on the high hill tops and also in the large low-lying area to the east of

Buttermere (NY 193 146, see Figure 2.4). Buttermere Moss covers nearly 1 km^2 and the peatland over Melbreak about 0.75 km². Visual inspection of these deposits indicates that they are not presently actively eroding, and that they are therefore unlikely to be of significance for present sediment inputs to Buttermere and Crummock Water.

Extensive woodlands no longer exist in the Buttermere and Crummock Water catchment, although there are several remnants. The largest, Burtness Wood (NY 182 153 - NY 172 163) is a coniferous plantation lying along the western shores of Buttermere. Extending north-west along the western side of Buttermere Dubs are mixed deciduous woods, together covering *circa* 1.1 km². Another relatively large area of mixed woodland, Laithwaite Wood, which extends out of the catchment, is on the extreme north-east shore of Crummock Water. This is about 0.4 km² in area, of which about 0.2 km² is within the catchment. There are a number of smaller stands, mainly on the upper slopes of the fans on the western shore of Buttermere and Crummock Water.

As discussed in the preceding sections, the catchment ecology and sediments of other lakes in the Lake District have been significantly altered particularly by deforestation. Development of woodland within a catchment enhances land stability, and hence generally moderates sediment input to catchment lakes. Both planting and then intermittent felling, here principally concentrated within the coniferous plantation located alongside Crummock Water, will lead to localised increases in sediment erosion (Likens *et al*, 1969) and therefore to variations in sediment type and accumulation rates. Distinguishing minor woodland clearance events in the sediment record of Buttermere and Crummock Water would require a fine-resolution chronology not available in this study (Chapter 3).

To conclude, using published work, five main phases of environmental change within the Lake District during the last 11 000 years have been highlighted, and their general effects on lake sediments and sedimentation discussed. These are summarised in Figure 2.6, which provides a framework against which results from Buttermere and Crummock Water can be tested.

2.3 Lake characteristics.

Morphometric details for the two lakes are summarised on Figure 2.7. This data is based upon original soundings recorded by Mill (1895), which are reproduced in Ramsbottom (1976). Figure 2.8 illustrates the bathymetry of the two the lakes.

2.3.1. Buttermere.

Buttermere which is relatively uniform in outline, and appears approximately oblong, with a maximum length of 2 km and surface area of 0.94 km² (Figure 2.7). The bathymetric map (Figure 2.8) and the longitudinal and cross-sections derived from these, (Figures 2.9 a and 2.9 b) also illustrate the lake form. This is a simple trough, with steeply-sloping walls and a nearly flat floor (cf. Mill, 1895, p.69). The trough has a mean depth of 16.6 m, with a maximum depth of 28.6 m at the eastern, valley head end. The main tributaries of Buttermere are Warnscale Beck (entering at the southern corner); Gatesgarth Beck (discharging midway along the south-east end of the lake); the Hassness How Beck on the north-eastern shore, Comb Beck almost opposite it on the southern shoreline; and Sourmilk Gill, which flows in at the north-western corner of Buttermere (Figure 2.4). The relative hypsographic curve for Buttermere (Figure 2.10 a) also illustrates the lake form and is the basis for the definition of the lake form concept (Håkanson 1981). This curve approximates closely to the boundary between linear and concave, to give a form

Recent circa 1900 - 1987 A.D.

More intensive farming, commercial forest plantations, limited urbanisation in some parts of the Lake District; increased recreational pressure increases surface erosion.

Increased sediment input, increased proportion of minerogenic sediment; increased anthropogenic organic effluents into some lakes; more widespread deposition of novel contaminants (including magnetic spherules) from outside of catchments via atmospheric deposition.

Late Post-glacial circa 950 A.D. - 1900 A.D.

Effects of man now dominate climatic effects on lake sedimentation.

Waves of settlers and deforestation; communities well-established by medieval period, developing more intensive agriculture including deeper ploughing, leads to disturbance of upper and <u>lower</u> soil horizons. Increased sediment input, increased proportion of minerogenic sediment; anthropogenic organic effluents into some lakes.

Mid Post-glacial circa 5 000 - 1 000 B.P. (950 A.D.)

Anthropogenic effects significant in addition to climatic effects on lake sedimentation.

Subtle change in climate/pathogen/anthropogenic effects leads to Elm Decline.

First indicators of anthropogenic disturbance; initial forest clearance; primitive cultivation and ploughing causes soils disturbance.

Increased sediment input, possible pulses of more minerogenic sediment from anthropogenic clearance events.

Early Post-glacial circa 10 000 - 5 000 B.P.

Climatic amelioration; pioneer vegetation followed by development of mature forests.

Less intense physical erosion; gradual stabilisation of landsurface; development of stable soils.

n.b. climatic variation leads to variation in erosion intensity, and, at some sites, lowered lake water levels.

Reduced sediment input, with increased percenatge of organic material; any changes in lake level imply changes in sediment type and distribution.

Late Glacial circa 10 000 B.P.

Glacial conditions and sparse vegetation.

Unstable and actively eroded landsurface.

High input of minerogenic sediments; rythmic sedimentation from corrie glaciers.

Figure 2.6. Summary of Holocene environmental change, major erosion processes and their possible effects on lake sediments in the Lake District (based on literature reviwed in the text).

A) Buttermere.

Contour (m)	Area enck km ²	osed %	Maximum length	Mean depth (m)	Lake area (km ²)	Volume $(m^3 \times 10^6)$	Drainage basin' area (km ²)
0	0.94	100	20	16.6	0.04	15.2	16.9
7.62	0.75	79.4			0.94	13.2	10.9
15.24	0.58	61.8					
22.86	0.28	29.4					
28.65 (maximum depth)							

B) Crummock Water.

Contour (m)	Area enclo km ²	osed %	Maximum length	Mean depth (m)	Lake area (km ²)	Volume (m ³ x 10 ⁶)	Drainage basin area (km ²)
0	2.52	100					
7.26	2.17	86.2	4.0	26.7	2.52	66.4	43.6
15.24	1.85	73.3					
22.86	1.58	62.8					
31.48	1.35	53.5					
38.10	0.85	33.7					
43.89 (maximum depth)							

Figure 2.7. Morphometric details of Buttermere and Crummock Water. Based on bathymetric data from Mill (1895) and Ramsbottom (1976).



Figure 2.8. Bathymety of Buttermere and Crummock Water (metres), and lines of cross-sections shown on figure 2.9.



Figure 2.9. Longitudinal- and cross-sections from Buttermere and Crummock Water, (after Mill, 1895).

The lines of section are shown on figure 2.8. The black shaded parts use the same vertical and horizontal scales, the unshaded parts show the sections with depths exagerated 15 times.

A) Relative hypsographic curve for Buttermere.



-80

-100

ρ

20

40

1

C) Terminology and class limits for the classification of lake forms, (Håkanson 1981, figure 48).

80

100

60

Cum Area %



Figure 2.10. Relative hypsographic curves for Buttermere and Crummock Water, and class limits for the classification of lake forms.

classification of Cma. The application of the lake form concept is in optimising lake volume determinations and planning hydrographic surveys, however, it also provides a useful classification system, allowing rapid comparison of form between lakes (*op cit*).

The mean depth ratio (that is, the ratio of mean depth to maximum depth, $z:z_m$) has been used by Lehman (1975) to define lake basin morphology. Models of lake sedimentation proposed by Lehman assume an elliptical lake bed area, but differ with respect to the basin shape. Mean depth ratios are limited to a certain range of values depending on the shape of the geometric solid to which a basin approximates. Mean depth ratios of 0.333 - 0.5, 0.297 - 0.5, and 0.5 - 0.667, apply to hyperboloid, sinusoidal, and ellipsoid-shaped basins respectively. As the ranges for the hyperboloid and sinusoidal basins overlap, further examination of volume distributions is necessary in order to determine which model a lake basin conforms to. Visual inspection of basin profiles (Figure 2.9) suggests that Buttermere is an ellipsoid. This is confirmed by its mean depth ratio of 0.58, which falls within the depth ratio range for ellipsoid-shaped basins (Lehman, *ibid*).

2.3.2. Crummock Water.

Crummock Water has a maximum length of 4 km, and a surface area of 2.52 km^2 (Figure 2.7). Its outline is less regular than that of Buttermere, and the lake axis is slightly curved, changing from north-north-west to approximately north-west nearly 1.5 km from its southern end where the lake is constricted by the rocky prominentaries Hause Point (NY 1615 1820) and Low Ling Crag (NY 1570 1830, Figure 2.4).

The largest tributary entering Crummock Water is Buttermere Dubbs (the outflow from Buttermere) which has developed a large delta in the south-west corner of the lake. This can be identified in the bathymetric map west of "X" (a cross-section reference point, Figure 2.8). Smaller tributaries, mainly draining from the high valley sides surrounding the lake include Mill Beck (which enters at its south-eastern corner), Rannerdale Beck, located approximately midway along the lake on the northern shore, Scale Force, which enters in two arms near the head of the lake on the southern shore, and Park Beck, (which discharges close to the lake outflow), and which flows directly from Loweswater, (a small, shallow lake, mean depth 8.4 m (Ramsbottom, 1975), situated in the extreme west of the catchment of Crummock Water, see Figure 2.4). Both Park, Rannerdale, and Mill Becks enter over large plains of low relief (Figure 2.4), developed over the relict alluvial fans discussed in section 2.1 ("Post-glacial modification"). The presence of the Buttermere Dubbs delta at the head of Crummock Water suggests that this tributary is the most significant source of incoming sediment, although analyses of stream sediment loads to test this have not been undertaken.

The relative hypsographic curve (Figure 2.10 b) for Crummock Water, unlike that for Buttermere, does not posess a simple, single, point of inflection. It still lies predominately between the linear and concave shape class limits (Figure 2.8 c), and thus the lake may be classified as concave (form Cmi).

Profiles for Crummock Water (Figure 2.9 a, c, d) illustrate the general form of the lake as a flat-bottomed trough with steep sides. Sections of these lateral slopes are formed by steep outcrops of the adjacent bedrock, which are among the steepest lateral slopes in the Lake District (Mill, 1895). The concave hypsographic classification (Figure 2.10 b), the regular basin profiles (Figure 2.9), and the mean depth ratio of 0.61 all demonstrate that Crummock Water, like Buttermere, conforms to the ellipsoidal basin sedimentation model of Lehman (1975). Like Buttermere, the lake floor is described as nearly flat (Mill, 1895).

Both Buttermere and Crummock Water are presently highly unproductive lakes (Luther and Rzöska, 1971). Buttermere is classed as extremely oligotrophic, particularly with respect to its size, and has therefore been notified as a Site of Special Scientific Interest forming one of the Lake District lakes least affected by human modification of the environment (Ratcliffe, 1977).

The deep Lake District lakes are all generally considered to be warm monomictic, and are therefore characterised by one complete overturn of the water column each year in autumn (Hilton, 1985). The break down of lake stratification allowis the redistribution over the

whole lake basin of very fine sediment previously trapped in the epilimnion (further implications of lake thermal structure on sedimentation are discussed in Chapter 1, section 1.5).

Both Buttermere and Crummock Water are therefore very similar in terms of "external" characteristics, such as climate, geology, and geomorphology, and in terms of their morphometry and trophic status. They differ markedly in terms of their basin volume however, with Buttermere occupying less than half the area of Crummock Water. While sediment sources for Buttermere are derived directly from the catchment, the major tributary for Crummock Water, Buttermere Dubbs, is the outflow from Buttermere. Buttermere may therefore act as an upstream sediment trap for Crummock Water.

Crummock Water also receives the outflow (Park Beck) from Loweswater located in the extreme west of its catchment (Figure 2.4). This enters Crummock Water on its south-west shore, close to the main outflow. Loweswater is distinctly different in both morphometry (area, 0.64 km2, mean depth 8.4 m, maximum depth, 16.0 m, Ramsbottom, 1975) and productivity, with development of periodic oxygen deficits (Pennington, 1981). Direct comparison of lake sediments, as is undertaken here between oligotrophic Buttermere and Crummock Water (detailed in the following chapters) is therefore not facilitated, and, with its increased productivity, sediments from Loweswater do not conform to the oligotrophic lake conditions preferable for mineral magnetic studies (see discussion, section 1.4). Sediments from <u>within</u> Loweswater are therefore not included in this study, although, as the lake is situated within the Crummock Water catchment, samples from around Loweswater are used in the investigation of source - sediment linkages undertaken in this study (chapter 4).

In summary, with flat bottoms located well below storm wave base, Buttermere and Crummock Water are morphologically suited to the preservation of undisturbed, continuous sediment sequences. They are presently oligotrophic, and have remained so throughout the Post-glacial (section 2.2, Haworth, 1985). They therefore provide good sites at which to examine Post-glacial sediments, and in particular to study processes of landscape evolution and sediment transfer by direct analysis of their clastic components. Results of these analyses are presented and discussed in the following chapters. Initial discussions of results in each chapter is made, allowing integrated thematic discussion in the concluding chapters.

3. LAKE SEDIMENT STRATIGRAPHY AND CHRONOLOGY.

3.1. Introduction.

Visual, physical and magnetic analyses are used to characterise various lithostratigraphic units identified in the Buttermere and Crummock Water sediment sequences, and also form the basis of thematic discussion in subsequent chapters. A sediment chronology is constructed using several complementary dating techniques, and this provides a framework within which environmental change is subsequently considered.

Palaeolimnological studies have frequently assumed that the most representative record of accumulation is obtained from sediment cores collected from the central and/or the deepest area of a lake, with subsequent study based on analyses solely of cores from these locations (for example, Mackereth 1965, 1966a; Haworth and Allen, 1985; Sandgren and Risberg, 1990). Whilst assumptions of <u>continuous</u> sediment accumulation at the centre of a lake may be justified, Davis and Ford (1982), and Dearing (1983) have shown that <u>patterns</u> and <u>types</u> of sediment accumulation may change in a complex fashion through time and across the lakebed.

The lake in Dearing's study is relatively small (an area of 55 ha), shallow (maximum depth 5 m), and is surrounded by a flat, treeless landscape. It is consequently more easily influenced by changes in tributary pattern and wind direction. Both studies demonstrate however, that it is prudent to demonstrate conformity ofaccumulation, particularly in studies of lake/catchment interaction, where consideration is given to thicknesses of sediment units. This is achieved through analysis of a suite of cores, collected from across a lake basin is preferable for studies utilising lake sediment records (*op cit*). A multiple coring approach has therefore been adopted in this study providing low density, but lake-wide, sampling of the sediment sequence.

Eleven sediment cores 0.5 to 6 m in length were collected from Buttermere and Crummock Water between June and September 1987. A 6 m Mackereth corer (Mackereth, 1958) was used to obtain all the long cores and a mini-Mackereth corer (Mackereth, 1969) to provide undisturbed samples up to the sediment/water interface. Mackereth cores exhibit minimal sediment disruption (Mackereth, 1958, 1969, Smith, 1959b). Compaction is minimised by incorporation of a Kullenberg piston, although smearing of the outer surface of the core does sometimes occur. In order to prevent contamination, the outer rind of sediment is thus usually removed when detailed stratigraphic studies are undertaken.

Non-vertical sediment cores may be recovered if either of the corers used does not seat correctly on the lake floor. Identification of horizontal laminae in all cores employed in this study (see sediment descriptions, section 3.2) confirm that the cores were all collected in an approximately vertical position.

As is usual in lake sediment studies, cores were mainly collected from the flat, deeper areas of the two basins (Figure 3.1), aiming to avoid sediment slumping and hence stratigraphic disturbance from lateral slopes. The cores were collected on a transect along both lakes thus providing samples most sensitive to down-lake changes in sediment quality.

3.2. Methods.

3.2.1. Stratigraphic division.

Stratigraphic division of the Buttermere and Crummock Water cores is based on their visual characteristics, and is supplemented by a number of analyses which individually have further application in investigation of catchment environmental history (see subsequent chapters). Following collection, long cores were extruded in the field into lined core trays and wrapped in polythene sheeting for transport back to the laboratory. Short cores were held vertically during transport and then extruded in the laboratory. Prior to extrusion of the short cores, whole core (volume) susceptibility measurements of all cores were made at 2 cm intervals.



Figure 3.1. Bathymetry of Buttermere and Crummock Water (after Ramsbottom, 1976) and core locations. (Suffix "S" indicates, short, mini-Mackereth cores, "L" indicates, long, six-metre Mackereth cores, bathymetrical depths in metres).

Using clean plastic blades in order to avoid contamination, particularly from metallic particles which would affect subsequent magnetic analyses, cores were sliced into 2 cm thick sections and dried at 40°C. Each slice was weighed before and after drying allowing determination of moisture content, and stored in clean, labelled plastic bags.

Selected long cores (C2L, B2L) were additionally sub-sampled when fresh, using a cut plastic syringe to take known volumes of fresh sediment. These were weighed wet, after drying at 40°C, and again after ignition at 550°C for two hours, and were subsequently used for geochemical analysis (Chapter 4). These measurements allow determination of moisture content, bulk density values, and loss-on-ignition (LOI), and follow the physical analysis regime of sediments discussed by Bengtsson and Enell (1986).

Grain size distributions were determined by standard pipette analysis after the method in BS 1377, with analyses based on sub-samples from either single dry slices or from the bulked remains of up to three slices (equivalent to up to 6 cm of wet sediment) in the less dense upper sediments. Data up to the sediment/water interface for the Crummock Water analyses were obtained by combining analyses of core C2L and the adjacent short core C3S. The two cores were correlated on the basis of their whole core susceptibility.

Investigation of the elemental composition of laminae within the sediment sequence was carried out by EDAX analysis of small samples taken from well-developed laminae. A standard mineralogical sample was used for comparison of these results.

For mass specific mineral magnetic measurements, dry sediment samples were gently crushed by hand in a pestle and mortar and tightly packed into preweighed 10 ml plastic pots which, prior to packing had been placed in a magnetising field of 300 mT. Those pots with measured remanences of >1 mT were rejected in order to avoid significantly influencing low remanence samples.

Magnetic measurements were carried out in the order:

(1) low and high frequency susceptibility (χ lf, and χ hf respectively);

(2) anhysteretic remanent magnetisation (ARM);

(3) isothermal remanent magnetisations (IRM) after placing samples in forward fields of 20 mT, 300 mT and 1 000 mT (IRM₊₂₀, IRM₊₃₀₀, SIRM);

(4) isothermal remanent magnetisations (IRM) after placing samples in reverse fields of 20

mT, 40 mT, 100 mT, and 300 mT (IRM₋₂₀, IRM₋₄₀, IRM₋₁₀₀, IRM₋₃₀₀).

The instruments employed are described in appendix 1.

Mass specific magnetic results are obtained from the determination of these parameters by normalising with respect to the sample mass. This eliminates variations caused by sample size and thus the amount of sediment actually present in a given sample.

Organic matter effectively "dilutes" magnetic assemblages, and, with variation in organic content between samples, potentially introduces an additional variability in concentration of magnetic material. In order to avoid this, and to allow direct comparison between samples of differing organic content, the weight contributed by organic material (as determined by loss-on-ignition) was subtracted from each 10 ml sample weight and the resultant "effective" weight used to calculate all mass specific magnetic results.

Mass specific mineral magnetic variables respond to the combined effects of mineralogy, concentration, and grain size within a sample. Ratio parameters, derived from mass specific measurements, may be calculated in order to reduce the effects either of mineralogy, concentration, or grain size. For example, susceptibility is highly influenced by the total ferrimagnetic content of a sample. Normalising a variable with respect to susceptibility therefore reduces the effect of changing concentrations of ferrimagnetic material between samples.

Ratios used in this study include:

(1) frequency dependent susceptibility ($\chi f d\%$), which is mainly dependent on the proportion of fine grained (superparamagnetic, SP) material in a sample (*circa* 0.02µm in isodiametric magnetite, Zhou *et al*, 1990);

(2) ARM/ χ , high values indicating the presence of single-domain (SD) ferromagnetic grains;

(3) IRM₊₂₀/ARM, high values indicating the presence of coarser ferrimagnetic grains at the SD/MD boundary;

(4) "soft", soft %, "hard", and hard % (ratios and percentage ratios derived from the forward fields IRM_{+20} , IRM_{+300} , and SIRM). These parameters reflect the proportion of "soft" and "hard" magnetic minerals in a sample.

More detailed explanations of the calculation and interpretation of the magnetic parameters used are given in appendix 1.

Mineral magnetic measurements are used in this chapter as an additional method of stratigraphic subdivision, with cluster analysis of the lake sediments based on their mineral magnetic characteristics providing an objective grouping of the sediment samples. Ten mineral magnetic variables, five mass specific (χ , ARM, SIRM, soft, and hard) and five ratio parameters (χ fd%, IRM+20/ARM, ARM/ χ , soft %, and hard %), characterising the sediments in terms of magnetic mineralogy, concentration and grain size (see appendix 1).

3.2.2. Chronological methods.

Sediment chronology has been constructed using three complementary methods. These are (1) comparison with independently-dated lake sediment sequences from other Lake District sites, (2) analysis of an atmospheric contaminant with a known emission history (magnetic spherules), (3) and Natural Remanent Magnetism (NRM).

Analysis of the sedimentary record of atmospheric contaminants with a known emission history provides a basis for dating recent sediments, and is the principle of dating utilised in 137 Cs dating (Pennington *et al*, 1973). Other contaminants such as carbonaceous
spheres, produced during the combustion of fossil fuels, have also been used to date lake sediments (Renberg and Wik, 1984). The sedimentary record of magnetic spherules observed in sediments from Crummock Water has been used in a similar way. This provides a dating technique for the more recent of these sediments (McLean, 1991). The extraction and counting method used here is described in appendix 2, and, briefly, consists of passing a sediment slurry over a magnet and removing that portion attracted to it. Care was taken to avoid: (a), contaminated sediment from the outer surface of the core - "smeared" during the coring process, and (b), contamination from dust and dirty glassware. An adjacent long and short core (C2L and C3S) were combined on the basis of the similarity of their magnetic susceptibility profiles (Figure 3.2) in order to provide a complete sediment sequence up to the sediment/water interface. Sampling was directed by comparison with the magnetic susceptibility profile for the combined core. Susceptibility responds to changes in the total concentration of magnetic minerals in the sediment, and so provides a guide for optimising sampling to locate major changes in numbers of magnetic spherules.

The earth's magnetic field is sufficiently strong to influence the direction of magnetisation and orientation of iron oxide crystals, and, under suitable sedimentary conditions, these signals may become locked and preserved in fine-grained sediments (Thompson, 1984). These <u>direction-dependent</u> magnetic properties are termed natural remanent magnetism (NRM), as opposed to the <u>non-directional</u> magnetic properties of materials which are their mineral magnetic properties.

Changes over time in direction and strength of the geomagnetic field, which may vary from a few degrees to a complete reversal (although such a "polarity reversal" has not occurred in the last 100 000 years), may thus be preserved in the sediment record. Being of regional extent (that is, comparable over areas roughly of continental size, see Thompson, 1984, his Figure 5.3), variations in NRM offer a means of correlating sediment cores between sites. Regional "master" curves illustrating variations in declination (direction) and intensity (strength) have been published (Thompson and Turner, 1979, Thompson 1984), and, with independently dated horizons, form a comparative method for dating sediments

from other locations. NRM measurements were thus carries out on cores from Buttermere and Crummock Water in order to provide a declination magnetostratigraphy and hence a comparative dating method.

For NRM measurements, two long cores from each lake were selected, in order to allow corroboration of results. These were processed whilst fresh, preceding slicing, drying, and any other analysis. Lengths of core, supported in non-magnetic plastic tubing, were measured on a slow-speed spinner magnetometer (Molyneux *et al*, 1972). Measurements were taken at 5 cm intervals, but were more closely-spaced towards ends of cores (see below). The magnetometer was calibrated at the beginning and the end of each run using a rock standard of intensity 938 x 10^{-8} . Am². The noise level of the system (discussed by Molyneux *et al*, 1972) is such that declination directions for these sediments were typically repeatable to within 1°.

Absolute orientations of the whole cores were not known. Therefore all declinations are relative. Declinations across core breaks were made equal in order to fit corresponding sections against one another, following Denham (1981). Whilst this procedure is simple, for example, if a curve is changing angle rapidly with respect to changing depth across a core break, a slight break in slope on the adjusted plot may result, this problem only arises where declination changes are of large magnitude and changes very rapidly. Any possible effects on plotted measurements were reduced here by using more closely-spaced sampling intervals towards the ends of the core sections.

The resolution of the whole core magnetometer (the minimum distance at which a section of core will have no detectable effect upon the fluxgate detector) used in this study has been estimated as 10 cm (Richards, 1987). Material above and below the fluxgate detector may thus influence readings, and the effects on declination measurements will be to smooth values obtained, as several thin slices are effectively being measured together. This is not a problem in this study as, although individual peaks and troughs may be slightly smoothed, the general shape of the declination curve is retained, which enables correlation with "master" curves. The effects on intensity measurements is for values to fall off towards the ends of core sections where the volume of magnetic material measured is effectively reduced.

3.3 Results.

3.3.1. Stratigraphic analyses.

The stratigraphy of the cores is shown diagrammatically in Figure 3.2 together with their whole core susceptibility measurements. It should be noted that near surface features of long cores may exhibit slight depth variation between cores owing to loss of material inherent in the 6 metre Mackereth coring procedure (Mackereth, 1969). Four distinctive sedimentary units may be identified, giving the general sequence:

Unit 1) microlaminated clay; Unit 2) clay/gyttja (detritus mud/clay transition); Unit 3) uniform grey silty-clay; Unit 4) gyttja (brown detritus mud).

Visual characteristics of these are described below. (Colour notations follow the Munsell system and are from dried sediment unless stated otherwise). Together with results of other physical and magnetic analyses (Figures 3.3 and 3.4), they exhibit distinctive variations in characteristics with the changing lithology. Results of the magnetic analyses record similar patterns of change between cores (illustrated by the whole core susceptibility logs in Figure 3.2). Therefore results of one core from each lake (B2L and the composite core C2L/C3S) are presented in this chapter as being representative of the lake-wide magnetic stratigraphy (Figure 3.4).

Clustering of sediment samples from these two central cores, based on a comprehensive range of mineral magnetic parameters with the same level of similarity (0.15) used for both B2L and C2L/C3S, has allowed "objective" division of the sediment sequence (Figures 3.5 and 3.6). Clustering is performed in the first stage of the PATN computer programme (Pattern Analysis Package, Belbin, 1987), used subsequently for sediment source comparison and identification (chapter 4). The clustering procedure used is the unweighted pair group centroid (following the method of Yu and Oldfield, 1989). These exhibit





Figure 3.2. Lithology and whole core susceptibility (κ , non-specific SI units) for all Buttermere and Crummock Water cores.

No short cores penetrate below unit 4 and are therefore composed entirely of gyttja. Inset indicates core sites. Core logs shown progressing westerly from the head of Buttermere.



Figure 3.3. Moisture content (as percentage wet weight) for Buttermere and Crummock Water cores. Adjacent long and short cores C2L and C3S are combined, giving a complete sequence to the sediment/water interface. Loss-on-ignition profiles are shown for cores B2L and C2L/C3S.

No short cores penetrate below unit 4 and are therefore composed entirely of gyttja. Inset indicates core sites. Core logs shown progressing westerly from the head of Buttermere.





Figure 3.4. Mass specific and ratio parameters for: A), Buttermere core B2L; B), Crummock Water, composite core C2L/C3S, (adjacent cores combined on the basis of similar susceptibility profiles).

Units are: susceptibility 10^{-8} m³kg⁻¹; ARM, SIRM, Soft, and Hard, 10^{-6} Am²kg⁻¹; ARM/X, 10^{2} Am⁻¹; Xfd%, soft%, hard%, IRM₊₂₀/ARM, dimensionless percentage ratios.



Figure 3.5. Dendrograph showing clustering of 89 downcore sediment samples from Buttermere, core B2L. (Average mineral magnetic characteristics of cluster groups and their depth ranges shown on figure 3.7).



Figure 3.6. Dendrograph showing clustering of 102 downcore sediment samples from Crummock Water, core C2L. (Average mineral magnetic characteristics of cluster groups and their depth ranges shown on figure 3.7). ς, ε

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ordered changes with depth. At the level of similarity used, the magnetic sub-groupings are used to subdivide the cores. Average parameter values fro the magnetic sub-groups identified are shown in Figure 3.7.

Each unit is now described in turn.

Unit 1) Microlaminated clay.

This is a stiff silt-clay with rare sub-rounded to angular pebbles, and more commonly, angular coarse sand-size grains of grey slate. Its recovered thicknesses of 0.8 m in Buttermere and and 1.1 - 1.4 m in Crummock Water. As no lower bounding horizons were recovered, these are minimum thicknesses. Moisture content and LOI values are the lowest of the whole sequence, similar at both sites (3.9% and 3.5% respectively). At such low values, contributions from interstitial water (lost on heating to 550°C) become significant. These figures therefore indicate the presence of an essentially inorganic sediment. Microlaminae, similar to those observed at the base of Unit 2, are present throughout this unit. In core C3L "fanning" of microlaminae in Unit 1 indicate that this sediment is completely overturned, the fold centred at 4.6 m. Such major disturbance is not present in other cores from Buttermere or Crummock Water, but is typical of the slumping which may occur in these fine grained sediments. This is thought to result from gravitational sliding of a sediment mass, following over-loading in the up-slope area (Smith, 1959a).

Cluster analysis divides this unit into two magnetic subgroups (I and II, Figure 3.7). These exhibit low total ferromagnetic concentrations and are characterised by a hard magnetic response. The minimal ferrimagnetic component is characterised by the presence of coarser (SD/MD) grains.

Unit 2) Clay/gyttja.

Where undisturbed, this unit maintains a relatively consistent thickness within each lake, approximately 0.75 and 1.1 m in Buttermere and Crummock Water respectively. At its upper boundary, it is similar in appearance to Unit 1, a brown detritus mud, and both units posess similar LOI values. Narrow bands of grey clay are more common in the lower part

A) Buttermere (core B2L)

<> mass specific parameters>						< ratio parameters>					
Depth (m) X	f	ARM	SIRM	Soft	Hard	Xfd %	IRM ₊₂₀ /ARM	ARM/Xlf	Soft%	Hard%	
10 ⁻⁸ m ³ kg ⁻¹	<		10 ⁻⁶ A.m ⁻²	kg ⁻¹	>1		timensionless ratio	10 ² Am ⁻¹			
Subgroup V	TT		*****								
0.0 - 0.53 78.	6	203.5	5420.1	965.9	149 3	6.8	4.8	27	179	28	
[Lithological uni	ι4,	gyttja; n	= 25)	,,	1 19.2	0.0	4.0	2.1	11.0	20	
Subaraun V	т				*****		******		*****		
Subgroup V	1 0	1366	3304 7	650 6	60 7	67	4.2	26	170	216	
[Lithological uni	ο ι4.	gyttia; n	= 5)	0.9.0	09.7	0.7	4.2	3.0	17.2	210	
Subgroup V	~										
0.67 - 1.81 21.	2 • 1	121.0	2955.1	629.7	95.8	3.9	3.5	5.8	15.0	3.2	
		. gyuja, n	- 23)	wir Printing of service state ser							
Subgroup IV	V										
1.91 - 4.21 20.	9	170.1	4261.9	488.8	63.9	3.3	2.9	7.9	11.4	1.6	
[Lithological groups 4 and 2, gyttja and clay/gyttja transition; $n = 23$)											
Subgroup II	I										
4.31 - 4.41 19.	2	57.7	2288.4	214.2	142.7	1.4	3.7	3.0	9.4	6.5	
[Lithological gro	up	2, clay/gy	yttja transit	ion; (n = 2	2)						
Subgroup I	[****	
4.51 - 5.01 23.	1	22.4	1598.6	162.2	214.4	1.6	7.5	1.0	10.2	13.5	
[Lithological gro	up	s 2 and 1,	clay/gyttja	transition	and micr	olaminat	ted clay; n = 6)				
Subgroup T										6854 ar 4 6 4	
3 57 &											
5.11 - 5.41 18	.77	12.4	655.4	73.3	80.1	1.0	6.0	0.7	11.3	11.8	
[Lithological gro	up	s 3 and 1,	grey clay a	nd microl	aminated	clay; n =	= 4)				

B) Crummock Water, (core C2L/C3S)

<	<	mass	specific p	arameters	>	<	ratio par	ameters		>
Depth (m)) 10 ⁻⁸ m ³ kg	(lf -1<	ARM	SIRM -10 ⁻⁶ A.m	Soft -2 _{kg} -1	Hard	Xfd % di	IRM ₊₂₀ /ARM	ARM/Xlf 10 ² Am ⁻¹	Soft%	Hard%
Subgroup 0.01 - 0.11 83 [Lithological gr	VIII 3.0 roup 4	197.4 4. gyuja;	9545.7 (n = 7)	1221.6	536.7	1.8	6.2	2.4	12.8	6.0
Subgroup 0.13 - 0.19 49 0.23 & 0.65 - 0 [Lithological gr	VII 9.2).67 roup 4	156.2 4, gyttja; 1	5165.6 n = 5)	73 9.1	470.0	5.8	4.8	3.2	14.4	9.0
Subgroup 0.21 & 0.25 - 0.63 34 [Lithological gr	VI 4.3 roup	136.1 4, gyītja; 1	4162.1 n = 21)	437.8	775.1	4.7	3.3	4.0	10.5	19.4
Subgroup 0.69 - 2.85 2: & 3.45 - 3.55 [Lithological g	V 5.7 roups	210.3 4 and 2, ;	5397.2 gyttja and	556.8 clay/gyuja	261.3 transitio	3.1 on; n = 41	2.7	8.3	10.7	5.1
Subgroup 1 2.95 - 3.35 20 [Lithological gr	IV 0.5 roup	67.7 3, grey cla	2539.6 ay; n = 5)	260.5	236.4	2.1	3.8	3.3	10.1	7.3
Subgroup 1 3.65 - 3.95 14 [Lithological g	111 4.2 roup	82.2 2, clay/gy	3035.6 tija transi	211.4 lion; n = 4)	126.0	2.0	2.6	5.8	7.0	4.1
Subgroup 1 4.05 - 4.45 1 & 5.55 - 5.65 [Lithological g	II 2.8 roups	24.8 2 and 1, 0	1281.6 clay/gyttja	87.8 transition	194.2 and micr	2.5 Tolaminat	4.1 cd clay; n = 5)	2.1	6.8	15.9
Subgroup 4.55 - 5.45 1 [Lithological g	I 8.4 100p	9.9 1. microla	1179.5 Iminated c	92.3 lay; n = 10	331.9)	1.4	9.8	0.5	8.0	27.8

Figure 3.7. Average values of all magnetic parameters used for cluster analysis for lake sediment magnetic subgroups, (figures 3.5 and 3.6), A) Buttermere, B) Crummock Water.

of the unit, passing into a silty clay at the junction with Unit 1 below. Here LOI values average 14.2% and 11.6% in Buttermere and Crummock Water respectively. Dark green horizontal laminae, as in Unit 4, and sand-size vivianite grains are found irregularly throughout the unit. Numerous sub-horizontal leaf, and more rarely, woody fragments are found in the upper part, becoming rarer below and absent at the base. In addition, towards the base of the unit, sub-horizontal microlaminae are seen, which become more common downwards. The micro-laminae are each less than 0.5 mm thick, distinguished as subtle lineations of varying shade within the generally grey silty clay.

The magnetic characteristics for this unit are similar to those of the lower part of unit 4, although concentration-dependent parameters are lower, and most values, particularly in the Crummock Water sequence are generally more variable.

The magnetic characteristics of this lithological group are transitional between those of units 4 and 1. Hence, in the cluster analysis, samples from the upper part of this group are located with samples from the base of the gyttja (magnetic subgroups IV and V, B2L and C2L/C3S respectively) whilst those from the base of this group are correlated with those from the top of the microlaminated clay (magnetic subgroup II in both B2L and C2L/C3S). Samples from the middle section of the clay/gyttja transition are clustered as separate magnetic subgroups in both sediment sequences (magnetic subgroup III in both B2L and C2L/C3S).

Unit 3) Uniform grey silty-clay.

This is a distinctive silt-clay horizon with sharp upper and lower contacts, consistently narrow where penetrated in the Buttermere cores (*circa* 1 cm in thickness), and varying from 5 - 46 cm in thickness in the Crummock Water cores (thickest in the central core, C2L). An apparent thinning of this unit downlake in Crummock Water (the unit is *circa* 1 cm thick in C3L) can only be tentative as C3L shows obvious evidence of sediment disturbance immediately below this unit (see discussion of unit 1, following). Where this unit is thickest an horizontal fabric is defined by occasional leaf fragments, with rare sand-size vivianite grains found throughout. Both LOI and moisture content fall sharply in comparison to the sediments above and below, as seen in C2L where this unit is thickest,

with an average LOI of 7.8%.

Also where thickest, the magnetic characteristics of this unit consist of a sharp reduction in concentration-dependent variables (for example, SIRM), and of the grain size-dependent parameter ARM/ χ , with a corresponding increase in hard and grain size-dependent parameters (hard, hard%, IRM₊₂₀/ARM). Whilst this unit forms a separate magnetic subgroup in C2L/C3S (subgroup IV), in core B2L, where it is represented by a single sediment sample, it is clustered with sediments from the base of unit 1, the microlaminated clay, B2L magnetic subgroup I.

Unit 4) Gyttja.

This is a greyish yellow (2.5Y 7/2) detritus mud, which forms the uppermost unit of the Buttermere and Crummock Water sequence, varying in thickness from 3.5 - 5.3 m and 2.1 - 2.6 m in Buttermere and Crummock Water respectively. Distinct horizontal laminae, dark green when wet and fresh, drying to olive brown (2.5Y 4/5) and 0.5 - 3 mm thick are spaced irregularly but frequently throughout the unit. Numerous horizontal or sub-horizontal leaf fragments and occasional sand-size grains of vivianite are also found throughout the unit. Both LOI and moisture content values are generally constant in the lower part of the unit, but reach a marked peak and trough in the upper 0.8 - 0.5 m (Figure 3.3). Average LOI values are 22.3% (Buttermere) and 16.8% (Crummock Water), falling to minima of 11.7% and 9.0% respectively in the uppermost sediment.

Magnetic analyses allow further subdivision of this unit, with four subgroups identified in both cores. The topmost unit (VIII, 1 - 11 cm in C2L/C3S) is absent in B2L owing to sediment loss in the coring procedure. This subgroup is characterised by the magnetic properties of atmospherically-derived magnetic spherules (identified by microscopic analysis and used in a proxy chronology, section 3.3.2). Magnetic parameters indicate that this is the most concentrated magnetic assemblage of the all the units, with a mainly ferrimagnetic mineralogy, and the largest proportion of coarse (SD/MD) ferrimagnetic grains.

The remaining magnetic subgroups within unit 4 (VII, VI, V in C2L/C3S, and VII, VI, V,

and IV, in B2L) exhibit initially high but rapidly decreasing concentrations of the whole ferromagnetic assemblage and of the fine (SP) magnetic grain size component (decreasing

average values of χ , SIRM, and χ fd%). The deepest subgroup (V, C2L/C3S, and IV, B2L) both include sediment from the upper part of unit 2, the clay/gyttja transition, as well as from the lower part of the gyttja, unit 4.

Whole core (volume) susceptibility (κ) logs (Figure 3.2) exhibit a consistent pattern between cores as do selected LOI profiles and the moisture content logs (Figure 3.3). The latter is the inverse if the whgole core susceptibility logs (Figure 3.2). The main features of whole core susceptibility profiles are:

i) a prominent double peak in the upper part of unit 4;

ii) consistently low values for the lower portion of unit 4;

iii) high values for unit 3, where this unit is sufficiently thick for 2

cm spaced magnetic measurements to register it;

iv) high and variable values in unit 1.

The laminae found irregularly throughout Units 2 and 4 are dark green when fresh, drying to an olive brown (2.5Y 4/5) with a flaky texture, and vary in thickness from approximately 0.5 to 3 mm. They are most easily separated from the surrounding material in the Crummock Water sediments, dried core slices splitting preferentially to expose the desiccation-cracked surfaces of individual laminae. In Buttermere, similar laminae are present, but contrast less distinctly with the surrounding sediment. Bulked material from laminae found throughout the Crummock Water Unit 4 gives average LOI values of $10\% \pm 1.2\%$, contrasting with 22.3% $\pm 4.5\%$ and $17.3\% \pm 3.3\%$ for the surrounding sediment of Buttermere and Crummock Water respectively. Separate determinations of LOI for laminae from the surrounding sediment.

Between 45 - 53 distinct laminae were counted in the cores from Buttermere and 98 - 103 in those from Crummock Water respectively. These were not correlated between cores.

EDAX (energy dispersive X-ray) analysis of laminae from each lake recorded an elemental composition dominated by Si, Al, with K and Fe present, which is consistent with the presence of chlorite or illite (Figure 3.8) although it is noted that chlorite is also present in the surrounding sediments (Chapter 4, XRD analyses).

This composition and the colour of hand specimens, suggest that chlorite is indeed the main mineral present. The most significant differences between these laminae and their surrounding sediments are therefore their relatively lower organic content and the marked colour differences.

Granulometric data from central cores from Buttermere and Crummock Water are shown on Figure 3.9. The Crummock Water data is more variable throughout the sequence, which may partly reflect a denser sampling regime used for these cores. Several changes in grain size composition are shown in both sequences however. These are:

 a generally increasing proportion of finer, clay-size, material upwards from Unit 1 to reach maximum values in Unit 2;

2) a decreasing proportion of clay above Unit 2 towards the sediment surface. In addition, the Buttermere profile exhibits a marked decrease in the proportion of clay from 0.75 m upwards, with minimum clay content values of 20% by weight found at the core top (N.B., the Buttermere profile does not include sediment from the sediment/water interface as no adjacent short core is available);

3) a marked increase in the proportion of sand above Unit 3 in the basal part of Unit 4, declining rapidly to values of 1 - 3% of total sediment weight, with a second, smaller increase in sand in the upper part of this unit.

Granulometric data for bulked samples of green laminae are also shown on Figure 3.9 b. Only data from the Crummock Water sequence are available, as laminae from the Buttermere sediments could not be reliably separated from the surrounding sediment.



Figure 3.8. EDAX spectra for a) "standard" chlorite sample;

b) green lamina material from core C2L, 1.73 m depth, (unit 2);

c) green lamina material from core C1S, 0.63 m depth, (unit 4).

Au present on all spectra derives from the coating procedure for the SEM. Spectra from the core samples (b and c) are consistent with a dominantly chlorite/illite clay composition, compared with the standard shown in a). Differentiation of laminae from the surrounding sediment is mainly by their distinct green colour in wet and dry samples, (grain size and LOI differences are also marked - see text for discussion).



by standard pipette analysis

Replicate analyses were carried out, each using material picked from up to ten laminae in order to collect sufficient material for each analysis (*circa* 10 g per pipette analysis). With a clay content of 39%, silt - 59.1%, and sand - 1.9%, the laminae exhibit a similar coarse (sand) component, but an increased proportion of finer (clay-size) material compared to the surrounding sediments.

3.3.2. Chronological analyses.

For lithostratigraphic comparison, reference is made to published descriptions of the deep water sediment sequence from Windermere. This is compared to that of Buttermere and Crummock Water in order to allow correlation with dated sediment boundaries representing regional events.

The Windermere sequence consists of a thin "surface ooze" (representing recent organic deposits derived mostly from 20th century anthropogenic effluents) overlying a thick "brown mud", which represents most of the post-glacial Holocene sediments. This is followed by an "upper laminated clay" which is a glaciolacustrine deposit from the Loch Lomond Stadial, the lower boundary of which is dated at *circa* 11,000 B.P. Below this is an organic silt from the Windermere Interstadial, 11,000 - 14, 623 \pm 360 B.P.), and a laminated clay, representing Devensian glaciolacustrine deposits (Pennington, 1943, 1981; Coope and Pennington, 1977).

A deposit similar to the Windermere "surface ooze" is not present in the Buttermere and Crummock Water sequence, probably reflecting the much lower human population in these catchments than found in Windermere. The Buttermere and Crummock Water "gyttja" is therefore chronologically equivalent to the Windermere "brown mud" and "surface ooze" together.

The lowest section of the Windermere "brown mud" immediately above the "upper laminated clay" was originally termed "a transition zone of narrow clays",

"...a zone of 1.5 m thickness in which narrow bands of pink unlaminated clay are separated by brownish grey organic deposits. All these narrow clays fade out laterally and do not appear in shallow water (less than 25 - 30 m).." (Pennington, 1943, page 4)

This description, replaced in later publications by a more generalised stratigraphy, for example Mackereth (1971) and Pennington (1981), describes accurately the Buttermere and Crummock Water unit 2, the transitional clay/gyttja.

The "upper laminated clay" of the Windermere sequence, a product of corrie glacier activity during the Loch Lomond Stadial, lies directly beneath the "zone of narrow clays" (equivalent to the Buttermere and Crummock Water unit 2). This suggests that the "upper laminated clay" is equivalent to the Buttermere and Crummock Water Unit 1, also a microlaminated clay. In the Buttermere and Crummock Water catchment, evidence for three corrie glaciers all situated in the high ground immediately to the south of the two lakes (Chapter 2, section 2.1), and active during the Loch Lomond Stadial, which would have fed directly into the two lakes, has been presented by Sissons (1980). The upper boundary of the Buttermere and Crummock Water microlaminated clay is therefore correlated with the Windermere late glacial/post-glacial boundary and is considered synchronous with that feature.

Results of magnetic spherule counts are expressed as numbers of spherules per gram of dry sediment, and are shown in Figure 3.10. Low values of < 40 spherules.gram⁻¹ are found throughout most of the core where examined, but rise gradually at 19 cm below the sediment/water interface to 50 - 100 spherules.gram⁻¹, with a dramatic increase above 11 cm, rising to maximum values approaching 2,000 spherules.gram⁻¹ in the uppermost sediments.

Associated with the changing spherule concentrations are marked changes in size



Figure 3.10.Magnetic spherule concentrations, expressed as numbers of spherules per gram of dry sediment, for nine selected samples, and mass specific susceptibility for composite core C2L/C3S, $10^{-9}m^3kg^{-1}$.



Figure 3.11. Size distribution of magnetic spherules in samples from core C2L/C3S. Distribution of cosmic ("natural") spherules shown by superimposed line (data from Puffer *et al*, 1980). Dates shown for each sediment sample are linear interpolations between the sediment/water interface (1987 A.D.) and NRM-dated horizons, hence are estimates as no account changing sediment accumulation rates between dates is made. With this error source, the similar calculated age for the major increase in spherule numbers (11 cm, 1905 A.D.) and suggested age, 1930 A.D., is reasonable.

distribution characteristics (Figure 3.11). Size distributions in the upper sediments become increasingly characterised by peak values of the finest fraction counted (1 - 10 μ m). The superimposed line on each spherule size distribution plot of Figure 3.11 represents the size distribution of cosmic ("natural") spherules presented by Puffer *et al* (1980). In Crummock Water, the size distributions of "Pre-Industrial" spherules are similar to the "natural" distributions, whilst those from "Late Industrial" distributions are not similar.

Plots of intensity and declination are shown in Figure 3.12 A and B. Declinations are relative, joined at core breaks by equalising declinations on either side of the break (as discussed above). Intensities are expressed in values relative to the magnetic rock standard used (938 x 10^{-8} .Am²).

Core B3L exhibits a very low intensity over its entire lower part, but a sharp increase above 0.8 m. This is similar to the whole core susceptibility pattern (Figure 3.2) where values of ca. 3 (non-specific SI units) increase to between 12 and 30 at around 0.9 m. Whilst the other long cores all exhibit increases in whole core susceptibility of similar magnitude, none contain such a low values as B3L, suggesting that either no magnetic material was deposited in this part of Buttermere (the lake head) over this period, or that these minerals have subsequently been removed.

The intensity profile of core B2L exhibits a generally higher and more variable pattern than that of B3L. Reference to the whole core susceptibility log (Figure 3.2), suggests that changes in the actual concentration of magnetic grains present is unlikely. Whole core susceptibility values remain broadly consistent for much of the total core length (between 0.75 - 4.4 m). Thus variations in intensity must be due to changes in the degree of alignment of the magnetic grain assemblage in the sediment.

The relative declination plots (Figure 3.12 B) exhibit close similarity <u>within</u> each lake where the sediment is undisturbed, but no correlation <u>between</u> Buttermere and Crummock Water. The declination record from B2L is scattered, which is a reflection of the low



Figure 3.12.A) intensity curves (x10⁻⁸Am⁻²) for long cores B3L, B2L, C2L, C3L. B) relative declination curves for long cores B3L, B2L, C2L, C3L.

intensities recorded for this core. Typically these result in poorer declination reproducibility (Barton and McElhinny, 1981).

Several centimetres more of the uppermost sediment were retained in core C2L than in core C3L. For comparative purposes, discussion of core C3L is limited to a section above*circa* 2.5 m. Sediment below this contains evidence of severe disruption (Figure 3.2) but where undisturbed, C3L and C2L both exhibit very similar patterns of intensity and relative declination.

3.4. Discussion.

3.4.1. Stratigraphy.

Four visually-distinctive sediment units are present in both the Buttermere and Crummock Water cores. Mineral magnetic data, moisture content, and LOI for selected cores provide criteria, in addition to the lithology, for correlation between cores.

Mineral magnetic characteristics reflect changes in mineralogy, grain size, and total concentration of magnetic material within the lithological units. These results indicate that the most concentrated magnetic assemblage is located in the upper part of unit 4 (the upper one metre). Initially the magnetic assemblage in this part of the sediment sequence is dominated by a hard, antiferromagnetic, component, with sharp increases in the proportion of magnetically soft minerals in the upper 0.2 m. The proportion of coarser magnetic grains present also increases markedly in the upper 0.2 m of the Crummock Water sequence, corresponding with a decrease in the proportion of very fine (SP) grains which are abundant in the lower part of this section. These characteristics are consistent with the observed increase of magnetic spherules found at this level (section 3.3.2), which are typically enhanced in these coarser magnetic grain sizes (Hunt *et al*, 1984).

In the lower part of unit 4, and throughout unit 2, increases in the ratio ARM/ χ indicate increasing proportions of fine (SD) magnetic grains. Concentrations of very fine (SP) grains are also reduced (cf. lower χ fd% values), as is the total concentration of ferrimagnetic minerals (indicated by decreasing values of χ and SIRM). Both the hard and the soft variables indicate generally unchanging proportions of ferrimagnetic and antiferromagnetic mineralogies in these sections, although the total concentration of ferromagnetic material decreases at the base of unit 2.

The clustering of lower samples from unit 2 with unit 1 (the microlaminated clay), illustrates the similarity of sediment characteristics between the two groups. At the upper

boundary of unit 2, magnetic characteristics of the sediments are also similar to those of the lowest sediments from unit 4, although this gradual transition is "interrupted" by unit 3. In Unit 3, sudden increases in the proportion of coarser (MD) magnetic grains and of magnetically hard mineralogies, with reductions in finer (SD) grains and the total concentration of the magnetic assemblage are recorded. These characteristics are similar to those of the microlaminated clay, unit 1.

Variations in the thicknesses of lithological units, and in the depths of certain features of the sediments are indicative of differential sedimentation rates across the two lake basins. Sedimentation rates have therefore varied through time. This is most marked in Buttermere in unit 4, with thicknesses varying from ≥ 5.3 m in B3L to 3.6 and 3.4 m in B2L and B1L respectively. In Crummock Water differential sedimentation rates are most marked in unit 3, thicknesses varying from 0.46 m in C2L, 0.05 and 0.02 m in C1L and C3L respectively. Further discussion of sediment focusing and sedimentation styles based on data presented in this chapter is given in chapter 5.

Distinct laminae are observed throughout Units 4 and 2 in both lakes. These are less organic than the surrounding sediment, and produce EDAX spectra consistent with a clay composition rich in chlorite. They contain similar proportions of coarser, sand-size, material, but greater proportions of the finer, clay-size, fraction.

Granulometric data from the central area of each lake records concentration of the finest sediment (with respect to total sediment weight) in Unit 2, with a gradual and slight coarsening-up of material in all cores exaimed in both lakes. This trend is more marked in Buttermere. No great similarities between bulk sediment grain size distributions, and lithological unit can be identified <u>between</u> Buttermere and Crummock Water however.

3.4.2. Chronology of the lake sediments.

i) Lithostratigraphic correlation.

In section 3.3, the similarities between the sediments of Windermere, and those of Buttermere and Crummock Water were demonstrated. The waning of the corrie glaciers of the Loch Lomond Stadial is represented in Windermere by a change from rhythmically laminated inorganic to more homogeneous and organic sediments. This is correlated with an identical lithostratigraphic change in the Buttermere and Crummock Water sediments. The change has been dated in the Windermere sediments using the ¹⁴C method to 10 013 \pm 350 B.P. (Mackereth, 1971). This provides a maximum age for the sediments near the base of the Buttermere and Crummock Water cores.

ii) The record of magnetic spherules.

Magnetic spherules are produced in only two natural environments (volcanic plumes and cosmic dusts). During prolonged high temperature combustion of fossil fuel they may be released into the atmosphere along with waste gases (Fisher *et al*, 1978).

Previous studies have shown that spherule concentrations have increased in a variety of sediments (for example, marine, estuarine, lacustrine, and bog deposits). The first increase coincides with the onset of the Industrial Revolution but latterly, much larger increases have been recorded, thought to reflect the development of large coal-burning stations and smelting plants (Locke and Bertine, 1986, Oldfield *et al*, 1981). Such studies have also demonstrated that spherules of industrial origin are widespread, having been identified in marine sediments at least 3,000 miles from major likely sources (Parkin *et al*, 1980).

Magnetic spherules therefore provide widespread cryptic marker horizons. Low numbers of spherules of cosmic origin provide a background level, and the development of modern industry and particularly of coal-fired power generation produces spherules in numbers at least two orders of magnitude greater than background (Locke and Bertine, 1986; Oldfield *et al*, 1978b; Oldfield *et al*, 1981). Volcanic eruptions possibly introduce erratic increases

in spherule numbers, but their effect on sediments is not discussed in the published literature.

Identification of changes in spherule concentration within a sediment sequence therefore provides a method of identifying the onset of the Industrial Revolution and of later industrial developments within a large region. Whilst these effects are widespread, absolute accuracy in dating using this technique is not possible.

The increase in numbers of spherules observed in the Crummock Water sediments is consistent with the predicted pattern of increase discussed above. The marked change in the size distribution of spherules is also consistent with this interpretation. Previous workers have observed that assemblages of industrially-derived spherules are characteristically enriched in the finer ($<20\mu$ m) fraction (McElroy *et al*, 1982). These finer fractions are increasingly dominant in the more recent sediment of Crummock Water (see Figure 3.11, samples A, B, and C - 3 cm, 7 cm, and 11 cm depth, which show size distributions increasingly dominated by the < 10 µm size fraction). Their size distributions become increasingly dissimilar to that of a sample of cosmic spherules, shown as a superimposed line of each graph on Figure 3.11.

Changing size distributions cannot be used as conclusive evidence for the origin of spherules, but do provide corroboration in this case. They suggest that the observed changes in spherule concentration and in size distribution reflect dilution of low numbers of cosmic spherules by high numbers of industrially-derived spherules, originally from the onset of the Industrial Revolution, but most markedly from the early 20th century (see following).

Oldfield *et al* (1978b) have dated the initial increase in spherule concentrations as observed in several U.K. ombrotrophic peat bogs to *circa* 1800 A.D. More closely-dated profiles from several Swedish bogs have been presented by Oldfield *et al* (1981), where, by the 1930's, spherule deposition rates increased by 20 - 50 times the values recorded for the 1800's. This coincides with the period of the development of iron smelting industries in Sweden.

Profiles observed in sediments from North America are generally similar to those from western Europe, although initial increases in spherule concentrations have been dated to the mid-1800's, with the dramatic increases from 1900 - 1910 A.D. owing largely to the commissioning of coal-fired power stations (Locke and Bertine, 1986, Goldberg *et al*, 1981).

Ochsenbein *et al* (1984) dated a marked change in concentration of trace metals in sediments from Blelham Tarn (in the Windermere catchment), to 1900 A.D. This they considered as reflecting post-Industrial Revolution increases in airborne trace metal pollution, which includes magnetic spherules. Crummock Water is situated approximately 20 km north-east of Blelham Tarn, so that the marked increase in magnetic spherule concentrations present in its sediments is probably derived from similar sources, and hence probably also dates from this period (*circa* 1900 A.D.).

Oldfield and Richardson (1990) report a generally similar pattern of deposition of atmospheric magnetic particles of industrial origin from thirty two lake sediment profiles mainly collected from upland Wales and the Scottish Highlands. They (*op cit*) report,

"...an increase in deposition from the early-mid 19th century onwards followed by a steep rise in the last 3 - 5 decades".

(Oldfield and Richardson, 1990, p.330)

The record of magnetic spherules found in Crummock Water is very similar to that of other independently-dated British and West European profiles, exhibiting low background values increasing by orders of magnitude in more recent sediments. Similarity of the records allows transfer of dates between profiles. These suggest dates of 1800 - 1850 A.D. for the initial increase in spherules above "background" levels (about 20 cm in the Crummock Water profiles) and *circa* 1930 for the marked increases caused by to the development of modern industry (about 11 cm). The present data therefore provide a general chronological framework for the upper sediments of Crummock Water which is absent in the long cores used fro NRM determinations.

A further test of this theory is provided by consideration of a contemporary sedimentation rate for Buttermere calculated by Pennington (1981) at 0.2 cm.yr⁻¹. This suggests an approximate <u>maximum</u> depth of 11.4 cm for sediments dated at 1930 A.D. in this lake. The upper susceptibility peak, which spherule counting has identified as the onset of the major increase in spherule numbers, occurs at 8 - 10 cm on the Buttermere mid-lake susceptibility profiles (Figure 3.2). Thus, dating by identification of increasing spherule numbers and consideration of contemporary sedimentation rates, locate the 1930 A.D. sediment horizon at 8 - 10 cm and 11.4 cm respectively.

Linear interpolation between the sediment/water interface and the most recent NRM-derived dated horizon (1800 A.D., see Figure 3.14 following) provides an approximate date of 1905 A.D. for the major increase in spherule numbers (Figure 3.11, "Late Industrial"-type magnetic spherule distributions, 11 cm depth). Acknowledging that this interpolated date/depth is very approximate, as linear interpolation takes no account of changing sediment accumulation rates, the correlation between the date of *circa* 1930 A.D. for the onset of the recent dramatic increase in spherule numbers (preceding discussion) and inferred dates from local chronologies is reasonable.

Two independent lines of evidence (a contemporary accumulation rate for Buttermere based on 137 Cs dating by Pennington (1981), and onterpolation between an NRM-derived dated horizon) therefore show a general agreement with dating based on changing magnetic spherule concentrations. This suggests that the magnetic spherule record provides a proxy chronology in sediments post-dating the Industrial Revolution. As doubt exists, principally over the exact date of increases in the combustion processes from which the spherules derive, as well as the influence of emisions from more local industry, this date is valuable in the context of this study, but is used for guidance.

In the same way that changes in pollen concentration should be considered within the context of varying lake sediment accumulation rates (Davis *et al*, 1984), changing lake sediment accumulation rates will, of course, cause apparent changes in spherule concentration. Sediment accumulation rates <u>have</u> varied over time in both Buttermere and

Crummock Water (Chapter 4, section 4.2) particularly in more recent times. Human influence has caused increases in sediment inputs in lakes throughout the Lake District (Pennington, 1978a). As spherules are ultimately derived from the atmosphere, increases in total sediment input would tend to dilute the spherule record. Observed values for the recent increases of spherule concentrations in Crummock Water therefore represent minimum values, and as such do not alter the interpretation of the spherule record and its use as an approximate dating method.

iii) Natural Remanent Magnetostratigraphy.

Intensity is not an ideal between-lake correlation parameter as it is dependent on intrinsic sediment characteristics - the quantity and type of magnetic material available for orientation, as well as how this material has actually been orientated by the strength of the earth's magnetic field. Declination measurements alone provide an adequate basis for comparison with published sequences, although intensity measurements can be of value in the interpretation of these, hence their measurement here.

Despite the general lithological similarities, the correlation of NRM characteristics between Buttermere and Crummock Water is poor. Thompson (1984) reported a wide variability in the quality of lake sediment magnetic records, which may be due to a variety of factors, including:

- (a) lack of deposition of sufficient iron oxide;
- (b) inappropriate sizes of magnetic crystals;
- (c) imposition of secondary magnetisation on the primary magnetic component.

Subtle variables are therefore important for the retention of high quality NRM records, where the quality of the preserved record may differ even in lakes located close to each other. Hence the value of NRM for comparison between sites must be evaluated empirically, as here, after core measurements have been taken and compared with "master" records.

Evidence suggests that it is the Buttermere NRM record that is anomalous and does not reflect declination variations at the time of sediment deposition. All cores (except core B3L) exhibit a similar lithostratigraphy. The declination record for cores B2L and B3L is identical, but their lithostratigraphy is different however. This suggests that the declination record in the Buttermere sediments is not related to the time of sediment deposition, and that adequate "locking" of magnetic grain orientations did not occur at the time of deposition. This suggests that the Buttermere sediments do not hold a stable remanence, and that the pattern measured from these cores has been imposed later. Variations caused by later changes in the earth's magnetic field result if the sediment fails to "lock" the grains in their original alignment, a process normally achieved soon after sediment deposition by the growth and subsequent dewatering of gels within the sediment (Stober and Thompson, 1979).

The exact reason for the poor record of remanence in the Buttermere sediments may be a product of:

i) deposition of magnetic particles of inappropriate size;

ii) disruption during dewatering, (Stober and Thompson, 1979);

iii) realignment of particles by active bottom currents prior to their burial and the stabilisation of remanent palaeomagnetic directions;

iv) insufficient growth of stabilising gels owing to intrinsic properties of the sediment.

The most appropriate grains for the preservation of high quality NRM records must be sufficiently small and magnetic for reorientation by the earth's magnetic field. Stober and Thompson (1979) identified SD ferrimagnetic material as being the most favourable for preservation of high quality detrital NRM. Smaller (SP) particles are unable to retain a remanent magnetisation, whilst larger (MD) particles naturally contain several magnetic

domains, apart from being physically larger and therefore less easy for the earth's magnetic field to reorientate (Turner and Thompson, 1979).

Mineral magnetic measurements of the Buttermere and Crummock Water sediments have been made in order to characterise their magnetic mineralogy and granulometry. These are used elsewhere in this study to aid source identification (chapter 4) and to assess sedimentation mechanisms (chapter 5). They are briefly referred to here in order to assess the quality of the sediments as recorders of NRM.

The ratio ARM/ χ provides a measure of the concentration of SD ferrimagnetic material in a sample. Figure 3.13 illustrates logs for all of the Buttermere and Crummock Water long cores. High values imply a greater concentration of SD ferrimagnetic particles, and lower values a reduction in their concentration and an increase in concentration of MD grains.

Whilst very low ARM/ χ values are typical for much of the length of core B3L, core B2L shows values comparable to both cores C2L and C3L.

Therefore, although appropriate size (SD) grains <u>are</u> of very low concentration in core B3L, SD ferrimagnetic grains are present in B2L, C2L, and C3L in approximately similar concentrations (as well as in the other Buttermere and Crummock Water long cores). Mineral magnetic measurements (Figure 3.13) suggest that an absence of appropriate magnetic grains is not responsible for anomalous lake-wide <u>declination</u> readings from the Buttermere cores, although this may be the reason for the very low <u>intensity</u> values measured in B3L. This is related to lake hydrodynamic conditions, where "inappropriately"-sized, coarser MD, magnetic grains are preferentially deposited in the lake head area.

Severe disruption during dewatering would disturb grain orientations, and so might be responsible for poor NRM record. Operation of such a process is unlikely as horizontal laminae are preserved throughout the sediment column (section 3.2). The Buttermere sediments do display higher water contents at equivalent horizons throughout the sediment column than the Crummock Water sediments (Figure 3.3), so realignment of particles at a



Figure 3.13. Downcore logs of the ferrimagnetic grain size dependent parameter ARM/X (units = 10^2 .Am⁻¹). Higher values indicate higher proportions of the finer (SD) ferrimagnetic component, lower values = lower proportions of the SD component and a higher proportion of the coarser MD component in a sample. (n.b., SD magnetic size range corresponds to grain diameters of approximately 0.02 - 0.04 µm in isodiamteric magnetic; the MD size range is > approximately 0.05 µm; further discussion of magnetic parameters is provided in appendix 1).

microscopic level, sufficient to disturb grain orientation but not to destroy features such as laminae, might have occurred. The similarity of the declination record for both B2L and B3L suggests that the process is not localised or random however, which would be expected during dewatering.

The action of bottom currents in realigning particles, prior to their stabilisation by growth of gels, would also be expected to operate on a lake-wide basis, and so would be expected to promote the similarity of NRM between cores. Given the preservation of fine laminae within the sediment sequence however (section 3.2), this explanation of anomalous NRM declinations seems less likely than the influence of intrinsic sediment characteristics, such as sediment porosity, favouring particle realignment.

The process of acquisition of remanence described by Stober and Thompson (1979) invokes realignment of grains in the uppermost wet sediment. These are stabilised by the growth of gels within the sediment, rather than by immediate fixing by dewatering of the sediment to below a critical level (contrary to the hypothesis of Verosub, 1977).

The Buttermere sediment is generally more organic than that of Crummock Water (Figure 3.3). It also exhibits greater moisture content and porosity values at equivalent sediment horizons than sediments from Crummock Water. (Porosity values for equivalent near-surface sediment horizons (correlated on the basis of similar whole core susceptibility profiles (Figure 3.2) are 86% and 77% for Buttermere and Crummock Water respectively (calculation method and data in appendix 3)).

Higher porosity and water content provide greater space and lubrication for particle movement, therefore favour interstitial particle realignment, (Verosub, 1977). Such sediment characteristics may therefore explain the apparently anomalous declination record from Buttermere.

The preceding discussion has largely discounted the use of the Buttermere declination curves for comparison with other declination records. Identification of other dated horizons in these cores, such as distinctive pollen horizons, would provide more conclusive



Figure 3.14. Comparison of NRM master curves from Windermere and Loch Lomond (transformed declination, redrawn from Turner and Thompson, 1979, Thompson and Turner, 1979), and Crummock Water relative declination curves. Symbols refer to correlated features which may be sharp (for example, "d") or drawn out, (for example, "g"). Dates are in calibrated years B.P.

evidence for these sediments. This could be undertaken in further studies of these sediments. The Crummock Water cores do not show any major anomalies however, and are consistent between themselves where sediment disruption (such as below 2.5 m in core C3L) has not occurred. They are therefore acceptable for comparison to published data.

Figure 3.14 shows the Crummock Water curves alongside published, independently dated "master" curves from Windermere and Loch Lomond (Turner and Thompson, 1979). Correlation of features between the two sets of curves has been made by eye. Such a process is subjective to some degree, but the features correlated are prominent and may be sharp (for example, "d" and "f") or drawn out (for example, "g"). It can be seen that correlation is generally good, with closely-spaced features allowing more precise correlation in the upper 2 m, and less distinctive and more widely-spaced features below. Correlation of these profiles therefore allows transfer of a series of absolute dates to the Crummock Water sequence. These are illustrated on Figure 3.14. The use of these for calculation of accumulation rates and subsequently catchment erosion rates is discussed in Chapter 4.

3.5. Summary.

The lithology of the Buttermere and Crummock Water sediment sequence has been described. It is consistent within and between lakes, and is similar to the deep water sediment sequence of other lakes in the region, notably Windermere. Other physical and magnetic characteristics of the Buttermere and Crummock Water sediments record ordered changes with depth, and indicate distinct variations in lake sediment type, and hence of material received from the catchment. Clustering of lake sediment samples based on their mineral magnetic characteristics generally supports division of the sequence based on gross visual characteristics, "objectively" identifying magnetic subgroups within the broad lithological divisions. These provide the basis for comparisons of lake sediments with catchment source samples undertaken in the following chapters (chapter 4).

A chronology of the Buttermere and Crummock Water sediments has been established
Core	Depth (m)	Feature	Inferred date		
C2L/C3S	0.00	sediment/water interface	1987 A.D. (n.b. absolute date)		
C2L/C3S	0.11	start of major spherule increase (= base of upper increase in susceptibility, present in all short cores)	<i>circa</i> 1930 A.D.		
C2L/C3S C3L	0.20 0.00	NRM declination feature a	150 B.P. (1800 A.D.)		
C2L/C3S C3L	0.41 0.22	NRM declination feature b	450 B.P.		
C2L/C3S C3L	0.47 0.29	NRM declination feature c	600 B.P.		
C2L/C3S C3L	0.81 0.61	NRM declination feature d	1 000 B.P.		
C2L/C3S C3L	1.42 1.33	NRM declination feature e	2 000 B.P.		
C2L/C3S C3L	1.63 1.51	NRM declination feature f	2 600 B.P.		
C2L/C3S C3L	2.79 2.49	NRM declination feature g	4 900 B.P.		
C2L/C3S	3.25	NRM declination feature h	7 100 B.P.		
C2L/C3S	3.58	NRM declination feature i	8 300 B.P.		
C2L/C3S	4.07	NRM declination feature j	9 100 B.P.		
B3L B2L B1L C1L C2L/C3S C3L	n/a 4.43 4.21 3.25 4.52 n/a	lithostratigraphic correlation (unit 2/unit 1 boundary)	<i>circa</i> 10 000 B.P.		

Figure 3.15. Summary of dated horizons and dated cross correlation features available from the Buttermere and Crummock Water sediments using NRM declination curve matching, magnetic spherule concentration curves, and lithostratigraphic correlation.

using three complementary methods. These provide a series of dates mostly for the Crummock Water sequence. Using the established lithostratigraphy, and features from both Buttermere and Crummock Water derived from physical and mineral magnetic analyses, transfer of some of these dates between the two sequences can be made, allowing calculation of sedimentation rates for each lake (chapter 4).

The Buttermere NRM record appears to be anomalous. This may be related to sediment characteristics. Systematic physical differences between sediments from both Buttermere and Crummock Water have been identified and their probable effects on the NRM recording capabilities of the sediments discussed.

The poor quality of the NRM record from a core from the lake head of Buttermere (B3L), which is also the deepest part of the lake, is partly a reflection of the inappropriate magnetic grain sizes present in this sediment, in which the coarser, heavier, grains have been preferentially deposited. This result is significant for the core sampling regime for other studies intending to use NRM measurements, implying that areas likely to receive input of coarser grain sizes should be avoided as lower quality NRM records will be obtained from sediment dominated by coarser MD magnetic grains.

Local lithological and chronological frameworks have therefore been established, Figure 3.15 summarises the dated sediment horizons obtained from all of the three chronstratographical methods used. Together with the framework of regional environmental change discussed in chapter 2 and further sediment analyses, the following chapters therefore aim to establish the environmental conditions and the sources within the catchment responsible for the observed changes in sediment type (chapter 4) and in lake sedimentation style (chapter 5) recorded over time.

4. CATCHMENT WEATHERING AND SOURCE-SEDIMENT LINKAGES.

4.1. Introduction

In chapter 3 the changing sediment sequence of Buttermere and Crummock Water were identified and described. As illustrated on Figure 1.2, these may reflect a combination of changing conditions within the catchment ("external" factors) and within the lakes basins ("internal" factors).

In this chapter, analyses examining external factors contributing to the observed sediment sequence are presented. The aim is to establish rates of sediment accumulation and delivery, the conditions of weathering and erosion operating in the catchment, and the catchment source-lake sediment relationships. Major conclusions from each analytical method are discussed in order to provide the basis for an integrated discussion (chapter 6) within the chronological framework established in chapter 3 and regional vegetation and climatic developments discussed in chapter 2.

4.2. Methods.

4.2.1. Sediment accumulation and delivery.

Sediment accumulation rates in Buttermere and Crummock Water have been calculated in the manner of Davis and Ford (1982). The methods used are described in detail in appendix 4. Dry density values are based on the mass of volumetric samples of fresh sediment collected from the core at 10 cm intervals (section 3.1). Deposition rates have been calculated from the chronology of sedimentation developed in chapter 3. Sediment inputs and catchment net sediment supply rates are derived from dry weight accumulation, and lake and catchment area data (appendix 4).

Accumulation rates for core C2L were calculated up to the present (the sediment/water interface at 1987 A.D.) using the adjacent core C3S. The two cores were correlated on the basis of similar overlapping whole core susceptibility profiles. Rates for the remaining Buttermere and Crummock Water cores were calculated using dates transferred from core C2L on the basis of well-correlated prominent features of susceptibility or lithological boundaries. As such features are few, rates are necessarily averaged over much wider time intervals than for C2L and C3L.

The effective lake area used for calculation of sediment input rates is less than the total lake surface area. Shallow marginal zones and steep basin sides are excluded, as they probably do not contribute significantly to the total sediment accumulation area. This is an assumption of the ellipsoidal sedimentation model (Lehman, 1975) discussed in section 2.3, which is supported by practical investigations of various lakes by Davis and Ford (1982), and Davis *et al* (1984). The effective lake basin area used for calculation of accumulation rates in Buttermere and Crummock Water is shown in Figure 4.1. This area, corresponds to the flat, basal region of the lake floor, avoiding both steep lateral slopes and the deltaic regions, where sedimentation patterns may be highly variable. In this study, these parts of the lakes have not been investigated in sufficient detail.

Terminology used here follows the definitions of Davis and Ford (1982) such that *accumulation rate* refers to net sedimentary material accumulated (mg.cm⁻².yr⁻¹); *input* equates to total sedimentary material delivered to the lake sediment (kg.yr⁻¹). *Catchment net sediment supply rate* describes the total export of material from the watershed (kg.ha⁻¹.yr⁻¹). The last is a guide to catchment erosion rates, which cannot be directly assessed, principally because of lack of quantitative data on sediment storage characteristics within the Buttermere and Crummock Water catchments (Oldfield *et al*, 1985b). This and other assumptions used for these calculations are discussed in detail in section 4.4.1, the discussion. (Expression of rates as dry weight values avoids "artificial" variations produced by differential sediment compaction, a problem inherent in accumulation rates expressed in units of sediment thickness per year).

4.2.2. Catchment weathering and erosion regimes.

Catchment weathering and erosion processes are examined using several complementary techniques. XRD has been used to provide weathering indices, which are based on relative concentrations of the resistant and less resistant minerals identified, especially quartz and feldspar respectively (Dean and Gorham, 1976; Chambers, 1978). Analysis of concentrations of mobile elements, and variations in mineral magnetism are used as additional indicators of changing erosion regime, and their application is examined in the discussion in this chapter.



Figure 4.1. "Effective" lake basin areas (shaded) used for calculation of lake sediment accumulation rates, (method shown in appendix 4). Effective lake basin areas are those areas below storm wave base, avoiding lateral slopes (hence slope creep/failure), and avoiding delta areas (which have very variable accumulation patterns), where sedimentation may be considered as continuous and generally conformable lake-wide. Delineation of effective lake basin area relies mainly on morphometric characteristics and therefore can only be approximate, (further discussion of assumptions in the calculation of these rates is discussed in section 4.4.1). Bathymetrical contours in metres.

i) XRD Analysis.

Sub-sampling for XRD analyses was based on the whole core susceptibility profile, which recorded the most marked mineralogical changes at the boundary of Units 1 and 2, and the upper part of the gyttja Unit 4. Core C1L was sampled most closely around these boundaries. Widely-spaced sub-samples for the Buttermere sequence (core B1L) were used to provide a very general comparison with the Crummock Water results.

Sediment sub-samples were taken from the dried 2 cm slices. The outer sediment from each slice was removed and discarded in order to avoid possible contamination by smearing during coring. Sub-samples crushed to a fine, uniform powder in an agate pestle and mortar and back-packed into aluminium sample holders for analysis. Three analyses were repeated after igniting the sediment at 550°C for three hours in order to aid clay identification. Analyses were performed on a Siemens x-ray diffractometer D500 with

DG1 x-ray generator, using Cuka radiation at 20 kV, 6 mA, with a ratemeter setting of 2 x

 10^2 cps, a scan rate of $1^{\circ}20$.min⁻¹, and a chart speed of 1 cm.min⁻¹. Quantitative expressions for the proportions of the most abundant minerals (quartz, feldspar, chlorite and illite) are obtained by calculating the integrated line intensity values (the product of the maximum peak height and peak width at half peak height), after the method of Norrish and Taylor (1962).

ii) Chemical analysis.

The reason for analysing the chemical properties of the Buttermere and Crummock Water sediments is to investigate the sedimentary geochemical evidence for changing catchment erosion processes. The hypothesis originally proposed by Mackereth (1966a) and discussed more recently, for example, by Engstrom and Wright (1984) suggests that the relatively straightforward determination of mobile element concentrations along a sediment profile can provide evidence of the intensity of erosion within a catchment.

Samples for chemical analysis from Buttermere and Crummock Water were collected at 10 cm intervals from the central cores B2L and C2L respectively. Material up to the

sediment/water interface was obtained for Crummock Water using the adjacent short core C3S. The two were correlated on the basis of their whole core susceptibility profiles (Figure 3.2). Samples had previously been ignited for two hours at 550°C for LOI determination (section 3.2). Finely crushed sub-samples from each of these residues were digested using the solution method 2 of Bengtsson and Ennell (1986). A 1:4 concentrated nitric acid:perchloric acid solution, which liberates bound elements from particulate matter into solution. Replicate digestions were made at intervals in order to ensure reproducibility. Quantitatively diluted, filtered digests were analysed in an EEL flame photometer for K and Na. Readings were converted to ppm using calibration curves determined by standard aqueous solutions.

4.2.3. Sediment source linkages.

i) Mineral magnetic comparison.

Lake sediment sources are investigated primarily through a comparison of mineral magnetic characteristics of lake sediment and catchment source. Such a methodology has been successfully used in other studies of source-sediment linkages (for example, Oldfield *et al*, 1985a, Oldfield *et al*, 1989, Sandgren and Risberg, 1990, Stott, 1986, 1987). The value of this methodology, and the fulfilment of criteria for its successful application to lake sediments are discussed in section 1.4.

Samples collected from the catchment reflect the major lithologies, glacial diamicts, and the soils developed on these in a variety of micro-environments, whether or not these appeared as currently active sediment sources. During collection, the use of iron tools was avoided where possible (in case of contamination), and, when pits were dug, samples were extracted using a plastic trowel.

Magnetic characteristics are strongly influenced by particle size, and J.P.Smith (1986) has shown that analysis of the <4 \emptyset fraction of catchment sediment sources provides a more direct comparison with mid-lake sediments. Magnetic domain differentiation is mostly based on effective grain sizes of 1 μ m and below. Sieving to this level separates not so much the magnetic grain sizes identified by the methods employed here, but removes variable proportions of the sand size range generally composed of quartz, and which would otherwise "dilute" the magnetic characteristics of samples. This has been observed, for example, in the magnetic characteristics of natural sediment samples by Oldfield *et al* (1985).

Catchment samples were therefore sieved and the $<4\emptyset$ fraction used for all subsequent analyses. Subsequent measurement and clustering of results were used to identify magnetically distinctive sediment source types. The determinations were performed in the same way as for the lake sediment samples (chapter 3).

Comparison of clustered lake and catchment source samples are made using the multi-parameter pattern analysis package PATN (Belbin, 1987). PATN allows the succinct presentation of a large volume of multi-parameter data on a single two-dimensional plot, so that general trends and interrelationships of sample characteristics can be more easily identified than by the use of a series of more conventional bivariate plots.

Analysis by PATN is based on a principal co-ordination analysis (PCoA) which calculates and plots eigenvalues and eigenvectors for all sample attributes. Parameter reference points are located around the plot boundaries and labelled with the parameter they represent. In the resulting two-dimensional ordination plot, the proximity of a sample to a parameter reference point, as measured along the straight line passing through the origin and the parameter reference point, represents the relative magnitude of that parameter value.

Stress, that is, distortion of original information, may occur during the transformation and plotting of data points. Compromises have to be made between samples that are similar with respect to certain parameters, but dissimilar with respect to others. A measure of the stress is provided by correlation coefficients positioned next to each parameter reference point, which indicate the degree of correlation between the original and the transformed data.

Thus, while the stress introduced into the data by the manipulation and plotting procedure remains low, objects plotted close together are similar. Those more widely separated are dissimilar with respect to all the parameters used to form the data set.

Volume susceptibility (κ) profiles of lake sediment cores alone have been used as a guide to the degree of bedrock weathering and hence of the type of dominant weathering processes in lake catchments (Zielinski, 1989). The basis for this hypothesis is that, if

catchment bedrock is the dominant source of magnetite for a lake sediment, variations of κ will reflect the degree of erosion and supply of bedrock-derived material.

Potentially, these measurements may thus serve as a guide to sediment source (that is, bedrock or not) and hence of catchment weathering regime. The techniques employed and the results of mineral magnetic studies of representative lake sediment cores from Buttermere and Crummock Water were described and presented in chapter 3. The utility of this simple approach assessed in section 4.4.3.

ii) Clay composition.

Identification of changing sediment source types is supplemented by the use of changes in the chlorite:illite ratio. In these catchnments, such minerals are respectively derived from primary (that is rock, and in this area particularly the Borrowdale Volcanic Group) and secondary sources (soils, mainly a product of feldspar degradation). Chlorite and illite contents are determined using semiquantitative data obtained from the XRD analysis of sub-samples from cores C1L and B1L, as discussed above.

4.3. Results.

4.3.1. Sediment accumulation and delivery.

Lake sediment accumulation, sediment input, and catchment net sediment supply rates are shown on Figures 4.2 and 4.3. Detailed accumulation and catchment net sediment supply rates for core C2L and widely-averaged rates for all cores are illustrated graphically in Figure 4.4 a and b. Data for core C3L are only available for the upper 2.5m, as below this

Date of horizon, [and NRM	Sedi thicl (cm)	iment kness	Time interval (years)	Deposition time (yr/cm)		Dry density (mg/cm ³)		Accumulation rate, (mg.cm ⁻² .vr ⁻¹)		Lake sediment input 4	Catchment net supply rate
feature]	C2L/ C3S	C3L		C2L	ĊĴĹ	CZL	C3L	C2L	GL	(x 1C kg.yr ⁻¹)	(kg.ha ⁻¹ .y1 ⁻¹ ,
1987 A.D	11	[N/A no adj-	57	5.2	[N/A no adj-	≈300	[N/A no adj-	57.7	[N/A no adj-	94.6	217
1800 A.D 150 B.P. [a]	9	short core]	130	13.0	cent short core]	410	cent short core]	32.2	cent short core]	53.0	122
	21	22	300	14.3	13.6	431	≈ 500	30.2	36.8	54.9	126
(00 D.D	6	7	150	25.0	21.4	576	496	23.0	23.2	37.9	87
600 B.P. [c]	34	32	400	11.8	12.5	283	283	24.0	22.6	38.2	88
[d]	61	72	1000	16.4	13.9	310	310	18.9	22.3	33.8	78
[e] 2600 B.P.	21	18	600	28.6	33.3	271	386	9.5	11.6	17.3	40
[f]	116	98	2300	19.8	23.5	315	353	15.9	15.0	25.3	58
4900 B.P. [g]	46		2200	47.8	DV/A	800	DV/A	16.7	DIA	27.4	63
[h]	33	core dist-	1200	36.4	core dist-	1022	core dist-	28.1	core dist-	46.1	106
[i]	49	urbed]	800	16.3	urbed]	1197	urbed]	73.5	urbed]	121.5	277
[j] 10,000B.E	54		900	16.7		1428		85.5		140.2	321

Figure 4.2. Lake sediment accumulation rates, inputs, and catchment net sediment supply rates for cores C3S/C2L and C3L, (see text for discussion).

Date of	Correlated feature	Accumulation rates (mg.cm ⁻² .yr ⁻¹)sediment input as kg/yr, catchment net sediment supply rates as kg/ha/yr							
feature		B1S/B3L	B2L	B1L	Lake-wide average	C4S/C1L	C3S/C2L	C3L	Lake-wide average
1987 A.D. 150 B.P.	sediment/water interface upper susceptibility	88.2	≈ 61.7	[N/A no adjacent short core]	accumulation rate	48.1	43.3	[N/A no adjacent short core]	accumulation rate45.7 lake sediment input75.0 catchment net supply rate172
860 B P	trough	41.5	28.3	26.8	accumulation rate32.2 lake sediment input19.6 catchment net supply rate116	37.4	26.5	30.8	accumulation rate
5270 B.P.	lithological boundary	>31.6*	20.4	19.3	accumulation rate23.7 lake sediment input14.5 catchment net supply rate86	11.4	15.0	17.9	accumulation rate14.8 lake sediment input24.2 catchment net supply rate56
7330 B.P.	unit 4/unit 3 lithological boundary	[N/A - no	0.8	0.8	accumulation rate	1.9	17.9	0.4	accumulation rate
≈10,000B.P.	unit 3/unit 2 lithological boundary unit 2/unit 1	sediment recovered]	13.7	13.1	accumulation rate13.4 lake sediment input8.2 catchment net supply rate48	47.0	48.8	[N/A core disturbed]	accumulation rate47.9 lake sediment input78.6 catchment net supply rate180

Figure 4.3. Wide interval lake sediment accumulation, input, and catchment net sediment supply rates for all Buttermere and Crummock Water long cores, (* Calculation assumes lower boundary of this unit is immediately below the base of the recovered core, hence this must be a minimum rate).



Figure 4.4. Lake sediment accumulation rates and catchment net sediment supply rates for Buttermere and Crummock water.

a), widely-spaced lake sediment accumulation rates (mg.cm⁻².yr⁻¹, light shading) for Buttermere and Crummock Water long cores. More closely-spaced dated intervals available for cores C2L/C3S and C3L allow calculation of more detailed sediment accumulation rates, (shown by superimposed thick black lines).

b), lake-wide average sediment accumulation rates (mg.cm⁻².yr⁻¹, light shading) and catchment net sediment supply rates (kg.ha⁻¹.yr⁻¹, dark shading) for Buttermere and Crummock water, with detailed catchment net sediment supply rates for the Crummock Water catchment determined using the more closely-dated rate data available for composite core C2L/C3S.

Catchment net sediment supply rates may be equivalent to catchment erosion rates, but do not take into account catchment storage mechanisms, and loss due to lake outflow for example, and are therefore be subject to errors, (see discussion in section 4.4.1).

the sediments are disturbed.

Accumulation rates for the more closely-spaced horizons of C2L and C3L (Figure 4.4 a) are very similar. High rates recorded by unit 3, corresponding to the early post-glacial (*circa* 7 000 - 10 000 B.P.), decrease steadily to reach minimum values of approximately 10 mg.cm⁻².yr⁻¹ in both cores during the period 2 000 - 2 600 B.P. Thereafter, sedimentation rates increase gradually, with a more rapid increase to over 57 mg.cm⁻².yr⁻¹ in the most recent 500 years.

The general trend of sediment input, and hence, catchment net sediment supply rates, follows the pattern of accumulation rates. The most detailed data available is for Crummock Water (Figures 4.2 and 4.4 b). Here, the highest rates are found in the early post-glacial (a peak catchment net sediment supply rate of 321 kg.ha⁻¹.yr⁻¹) falling to a minimum from 5 000 - 2 000 B.P. (40 - 60 kg.ha⁻¹.yr⁻¹) and increasing towards the present, where the most recent net sediment supply rate is in excess of 200 kg.ha⁻¹.yr⁻¹. Figure 4.3, which shows both detailed and widely averaged rates follows this general pattern, although significantly higher rates occur at certain horizons. For example, much lower rates are recorded in Buttermere than in Crummock Water in the early post-glacial (7 300 - 10 000 B.P.), and much higher rates in the most recent time interval (1987 A.D. - *circa* 1800 A.D.).

4.3.2. Catchment weathering and erosion.

i) XRD results.

Locations of sediment sub-samples used for XRD analysis, and the whole core susceptibility profiles employed to direct the XRD sampling regime are shown on Figure 4.5. Typical XRD traces are presented on Figure 4.6. All traces exhibit close similarity, with peaks indicating the presence of:

> quartz, (3.34 Å; 2.46 Å; 2.28 Å; 2.24 Å; 2.13 Å; 1.98 Å); chlorite, (14.7 Å; 4.69 Å; 3.52 Å; 2.82 Å; 2.42 Å; - selective changes after sediment ignition at 550°C);



Figure 4.5. XRD sediment sampling positions (**n**) for cores B1L and C1L, shown with volume susceptibility logs used to direct the XRD sampling regime. Depths in metres, volume susceptibility in non-specific SI units.



Figure 4.6. XRD traces for C1L and B1L sediment samples. Depth on each core trace refers to sample depth (m) down core. Abbreviations used are: Al = aluminium; Qz = quartz; Ill = illite; Chl = chlorite; Kaol = kaolinite.

Instrument operated using Cuca radiation with the following set-up: 20 kV, 6 mA,

1°20.min⁻¹, cps = 2 x 10², original chart speed = 1 cm.min⁻¹.

illite, (9.90 Å; 4.46 Å; 2.60 Å; 2.42 Å; no change in peaks after sediment ignition at 550°C); kaolinite, (7.00 Å; 4.48 Å; 3.52 Å; peaks lost after sediment ignition at 550°C); micas, (4.97 Å; 4.09 Å; 3.83 Å; 3.73 Å; 3.26 Å; 2.96 Å; all peaks very low indicating low concentrations near the resolution limit); feldspars, (3.19 - 3.21 Å)

aluminium, (2.34 Å; 2.03 Å; - a contaminant from the aluminium sample holders used).

The XRD traces are very consistent between cores. Changes in peak height and minor reductions in background "noise" levels (from 5 - 6% to approximately 3% at the top and bottom of the cores respectively) may both be accounted for by error due to differences in packing (up to 15% in extreme cases). A reduction in background "noise" is also caused by a decrease in the organic content of the samples (cf. LOI values, Figure 3.3, page 64). The results therefore suggest that there are no major differences in bulk mineralogy between the samples.

Figure 4.7 shows the integrated line intensity profiles from core C1L for the four most significant minerals identified (quartz, plagioclase feldspar, chlorite, and illite), calculated as detailed in the method section (page 101). The quartz:feldspar and chlorite:illite ratios are derived from these calculated intensities.

In order to allow direct comparison of samples of differing total mineral content, integrated line intensities are expressed as percentages of the total line intensity of the four minerals from each sample. Large variations are seen in the quartz:plagioclase ratio, high ratios in the upper *circa* 0.25 m and below 3 m depth. These changes are due to lower proportions of feldspar in the microlaminated clay (Unit 1) and the upper 0.5 m (Unit 4).

ii) Chemical analysis

Results of analyses of the mobile elements K and Na, expressed as $mg.g^{-1}$ of dry



Figure 4.7. Integrated line intensity profiles for core C1L. Values, expressed as percentages of total line intensity for each sample, are calculated from observed XRD line intensities for illite, chlorite, plagioclase feldspar, and quartz. Ratios for chlorite:illite and quartz:feldspar are also shown, and are used as indices of changing catchment sediment source and erosion style, (see text for discussion)



Figure 4.8. K and Na profiles for A), core B2L and B), composite core C2L/C3S. Labels a - g mark peak values, with dates extrapolated from an NRM-derived time/depth curve. Silt proportion and LOI curves are shown for comparison, (see text for discussion).

sediment weight, are shown in Figure 4.8. Prominent peaks in parameter profiles are labelled ("a" to "g") in order to facilitate identification and discussion. The main features of the profiles are:

1) There is a close resemblance between the K and Na profiles within each lake;

2) Maximum values for each element ("a") are located at the base of the sediment profile in each lake. These fall sharply above Unit 1. High values are reached again in the upper part of Unit 4 ("f" and "g");

3) Between the upper and lower peak Na and K values (a, f, and g) are a series of peaks of smaller amplitude (labelled "b", "c", "d", e"). These are present in both profiles but which exhibit greater amplitude variation in Buttermere.

4.3.3. Sediment source linkages.

i) Mineral magnetic-based comparisons.

The PATN analysis of mineral magnetic characteristics of catchment and lake sediment samples i snow discussed. Results are considered separately to allow identification of groupings within each catchment. Results of mineral magnetic analyses of all catchment samples are listed in appendix 6. Locations of sampling sites are shown in Figure 4.9, and results of a cluster analysis of catchment samples on Figure 4.10. Ordination plots for the Buttermere and Crummock Water catchments and lake sediment samples are illustrated in Figures 4.11 and 4.12 respectively.

Cluster analyses, using the unweighted group centroid method (after Yu (1989); Yu and Oldfield (1989); and as used for clustering the lake sediment samples - section 3.2.1), identifies six magnetically distinct source groups from each catchment. The three main lithologies each form one group (1 - 3). Substrates largely derived from the Ennerdale Granophyre account for a separate group in each catchment (group 4). Upper, weathered zones of glacial diamicts form a separate group in each catchment (group 5). Most soil A-



Figure 4.9. Location of catchment sampling sites in the Buttermere and Crummock Water catchments (Descriptions of mineral magnetic characteristics of samples shown in appendix 6).



Suffix letter describes type of deposit:

a = soil A-horizon; b = soil B-horizon; p = peat ss = stream sediment u = diamict, upper

(weathered) horizon 1 = diamict, lower (unweathered) horizon.

Figure 4.10. Results of cluster analyses of catchment samples from a) the Buttermere catchment, and b), the Crummock Water catchment. Using the same level of similarity for both catchment sample sets, six distinct sample groupings are identified. These correspond to the three lithologies present in each catchment, (Skiddaw Slate, Borrowdale Vocanic Group, Ennerdale Granophyre, groups 1, 2, and 3, respectively), substrates derived principally from the Ennerdale Granophyre (group 4), upper, weathered, diamicts (group 5), and undivided soils, stream sediments, and unweathered diamicts (group 6).



Figure 4.11. PATN ordination plot (first two dimensions of rescaled configuration and direction cosines of fitted property vectors) for Buttermere catchment and lake sediment samples. Distances between samples indicates degree of similarity or difference in terms of their magnetic properties. Ten marked points around the plot correspond to the ten magnetic parameters used in this study. Catchment samples, which fall into six distinct groups (see cluster analysis, figure 4.10) form a "reference" frame from which the lake sediments are mostly derived. Plotted positions of lake sediment magnetic subgroups (Roman numerals, I - VII) are mean values of clustered groups from 82 downcore sediment samples from Buttermere central core B2L, (figures 3.7 and 3.5 respectively).



Figure 4.12. PATN ordination plot (first two dimensions of rescaled configuration and direction cosines of fitted property vectors) for Crummock Water catchment and lake sediment samples. Distances between samples indicates degree of similarity or difference in terms of their magnetic properties.

Ten marked points around the plot correspond to the ten magnetic parameters used in this study. Catchment samples, which fall into six distinct groups (see cluster analysis, figure 4.10) form a "reference" frame from which the lake sediments are mostly derived. Plotted positions of lake sediment magnetic subgroups (Roman numerals, I - VIII) are mean values of clustered groups from 102 downcore sediment samples from Crummock Water central core C2L, (figures 3.7 and 3.6).

and B-horizons and stream sediments comprise a further group which displays rather variable mineral magnetic characteristics (group 6). Summary data of the mineral magnetic characteristics of all these source groupings are shown on Figure 4.13.

Ordination plots (Figures 4.11 and 4.12) illustrate the relative magnitude of mineral magnetic characteristics for source samples for the two catchments, and the relation of these to lake sediment magnetic groupings. Correlation coefficients (r-values) for original and transformed data are shown next to parameter reference points on the circumference of the ordination plots. With r-values greater than 0.68, they are all statistically significant or highly significant. The ordination plots therefore generally represent a true summary of sample characteristics. However, as the values of correlation coefficients decrease, the possibility of individual plotted points deviating from their optimum position with respect to particular parameters increases, and is of significance for particular samples (which are noted in the following section).

The main features of the source-sediment relationships in the Buttermere catchment (Figure 4.11) are as follows. The microlaminated clay (lake sediment magnetic groups I and II) clusters close to the Borrowdale Volcanic Group. The lower section of the Clay/Gyttja transition (lake sediment magnetic group III) plots in the upper left quadrant, and in terms of possible source, is therefore transitional between the Borrowdale Volcanic Group, the Skiddaw Slates and Ennerdale Granophyre (samples from locations 8 and 10). It is closest in terms of magnetic characteristics to the unweathered diamict (sample 281).

The remaining lake sediment magnetic groups (IV, V, VI, and VII) are located close to the undivided group of catchment soils and stream sediments. As indicated by larger values of

the parameters ARM/ χ , χ fd%, and soft %, they contain greater proportions both of very fine (SP) and fine (SD) magnetic grains, and a greater total proportion of soft, ferrimagnetic, material in the magnetic assemblage.

Trends of source-sediment relationships (Figure 4.12) are similar in both catchments. Lake

Group	Source type	Summary mineral magnetic characteristics
1	Skiddaw Slate and directly-	minimum total ferromagnetic concentration, low X, SIRM;
	derived substrates	antiferromagnetic mineralogy dominant, hard, hard% values
		relatively high.
2	Borrowdale Volcanic Group	low total ferromangnetic concentration, (higher than Skiddaw
	and directly-derived substrates	Slate);
		dominantly ferrimagnetic, antiferromagnet component
		significant.
3	Ennerdale Granophyre	maximum total ferromagnetic concentration, very high X,
		SIRM;
		ferrimagnetic component dominant, antiferromagnetic
		component very low;
		coarse (MD) grain sizes dominate - high ARM,
		IRM ₊₂₀ /ARM.
4	Ennerdale Granophyre-	similar to Group 3 but slightly less concentrated ferrimagnetic
	derived substrates	assemblage, antiferromagnetic component significant;
		coarse (MD) component dominant, fine (SP) component
		minimal.
5	Upper (weathered) diamicts	high total ferromagnetic concentration;
		ferrimagnetic component dominant;
		significant fine (SP) fraction - high Xfd% values.
6	Undivided soils, lower diamicts,	characteristics intermediate to preceding groups;
	stream sediments	ferrimagnetic component usually dominant, antiferromagnetic
		component significant.

Figure 4.13. Catchment sediment source mineral magnetic groups and summary mineral magnetic characteristics. Groups are clustered from samples within each catchment, (figure 4.10), and magnetic characteristics are summarised using data shown in appendix 6, (displayed graphically on ordination plots on figures 4.11 and 4.12).

sediment magnetic groups I, II, III, and IV (the microlaminated clay, the lower clay/gyttja transition, and the grey clay) are most similar to the Skiddaw Slates and Borrowdale Volcanics in terms of magnetic mineralogy (cf. soft% and hard% values), but are enhanced in the fine (SD) grain size range. The increasing contribution of secondary (soil) sources is demonstrated by the progression of groups I - III towards the group of undivided catchment soils (catchment group 6) near the centre of the ordination plot. Lake sediment magnetic group IV represents a reversal of this trend, with characteristics similar to group II. It therefore posses a greater similarity to the Skiddaw Slates and the Borrowdale Volcanics.

The magnetic groups from the upper lake sediments (V - VIII) plot close to group 6 (undivided soils), although the lake sediment groups are located in the upper half of the ordination plot, actually <u>above</u> catchment group 6. They are therefore similar in terms of magnetic mineralogy and concentration to source group 6, but are enhanced in the <u>finer</u> (SD) grain size range. Thus, whilst the proportions of minerals and their total concentration has remained similar between source and lake, there has been a relative increase in the proportion of finer grain sizes in the magnetic assemlage. This may be due to actual increases in the amount of the finer material, or to decreases in the amount of coarser (MD) material.

Figure 4.14 shows an ordination plot of the lake sediment magnetic groups from both lakes. This clearly illustrates a very similar trend of changing mineral magnetic characteristics between lakes, but a relatively consistent "offset" in the plotted positions of equivalent lake sediment magnetic groups. This reflects slight differences in the composition of the magnetic assemblages from each lake. Wth the exception of lake sediment magnetic groups II and III, equivalent groups from Buttermere are enriched in the finer (SP) and coarser (MD) ferrimagnetic grain sizes, as indicated respectively by higher

 χ fd% and IRM₊₂₀/ARM values.



Figure 4.14. PATN ordination plot for Buttermere (\bullet) and Crummock Water (X) lake sediment magnetic subgroups. Plotted positions of Buttermere sediment subgroups marked with circles (O), Crummock Water subgroups with crosses (X), and distances between plotted points indicate degrees of similarity or difference in terms of their magnetic properties. Tie lines link lithostraigraphically equivalent lake sediment magnetic groups, (depth ranges of groups shown on figure 3.7). Ten marked points around edge of plot (\bullet) show reference positions of the ten magnetic parameters used in this study. Plotted positions of sediment groups are mean values from the clustered sediment samples from Buttermere and Crummock Water respectively, (figures 3.5 and 3.6).

Volume susceptibility (κ) profiles of all sediment cores are shown in Figure 3.2 (p.63), and are used here in conjunction mainly with mass specific magnetic measurements (Figure

3.4, page 65) to test the hypothesis of Zielinski (1989) that κ profiles alone may be used to define the degree of relative bedrock weathering in certain catchments. A consistent pattern of variation between cores is seen, namely, high values at the base (unit 1), near the middle (unit 3) where this unit is well-developed, and towards the top (upper *circa* 1 m of unit 4) with low values in most of the remaining sediment. Of these, the peaks at the base and in the middle of the cores are exactly correlated with relative increases in IRM₊₂₀/ARM. These indicate the presence of increased proportions of coarse (MD) ferrimagnetic material. Neither of these zones exhibit any significant increase in χ fd%, an unambiguous indicator of the presence of very fine (SP) magnetic material, naturally formed in a soil environment (Maher, 1986, Maher and Taylor, 1988). A small increase in IRM₊₂₀/ARM at the very top of the sediment profile (retained only in core C2L/C3S) corresponds with a sharp fall in χ fd%.

Mineral magnetic measurements, the magnetic characterisation of catchment source samples and clustering of lake sediment samples therefore provides a sound basis for the identification of catchments source-lake sediment linkages, and changes in these over time. Detailed discussion of these linkages is found in section 4.4.3, (p.133).

ii) XRD-derived source indices.

The chlorite:illite ratio for the Crummock Water sediments (Figure 4.7) exhibits small variations across the boundary of Units 1 and 2, and in the upper 0.5 m (Unit 4). The lower feature is due to a reduction in the proportion of chlorite, and the upper to an increase in the proportion of illite.

4.4. Discussion.

4.4.1. Lake sediment accumulation/catchment erosion rates.

Care is needed in the interpretation of processes such as accumulation, input, and subsequently, erosion rates, from a single or even a few cores owing to possible effects of sediment focusing, changing patterns of sediment accumulation with time, and the influence of lake basin shape on apparent accumulation rates (Davis *et al*, 1984, Dearing, 1983, Lehman, 1975).

In this study core sampling density is low. However, the regular basin shape, longitudinal and cross-sectional profiles, and flat basin bottoms (section 2.3, p.44) ensure that limited sampling density is still likely to identify major changes in sedimentation patterns. Furthermore, both visual inspection of basin profiles (Figure 2.9) and comparison of mean depth:maximum depth ratios (0.58 for Buttermere and and 0.61 for Crummock Water) with classes presented by Lehman (1975, discussed in section 2.3) indicate that both lakes approximate to simple ellipsoidal sedimentation basins. For such lakes, sediment is most likely to be distributed over the entire basin floor with little occurring on the steep side walls. In the absence of strong current-induced focusing, models of sedimentation in elliptical basins also predict insignificant differences between measured and actual sediment input to the basin (*op cit*).

Calculation of catchment erosion rates however, requires detailed knowledge of sediment transport and storage mechanisms within both the aquatic and terrestrial systems. These have been discussed by Oldfield *et al* (1985), who identified ten major criteria for the calculation of <u>meaningful</u> catchment erosion rates. These are:

i) a well defined catchment;

ii) a low autochthonous organic sediment component;

iii) stable lake levels;

iv) detailed chronology of sedimentation;

v) detailed core correlation;

vi) no major development of reed-beds;

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vii) surveyed grid of cores;

viii) no major deltaic deposition;

ix) minimum outfall loss;

x) no long term sediment storage in catchment.

Criteria i - vi are generally fulfilled in this study. The core sampling density <u>is</u> low however, although the relatively regular shapes of the Buttermere and Crummock Water basins (as discussed above), and the uniformity of the stratigraphy, suggest a), that uniform sedimentation takes place across the basins is encouraged, and b), that major changes in the sediment accumulation pattern should be easily identified. Apart from unit 3, which, with large variation in unit thickness between cores, implies the operation of strong sediment focusing, sedimentation does appear to have been relatively uniform within each lake (see also discussion in chapter 5, section 5.4).

Criteria vii - x are unknown in detail in this study, and therefore impose further constraints and unsubstantiated assumptions on the calculation of any catchment erosion rates. In particular, because sediment storage mechanisms in this catchment have not been investigated, there can be little confidence that the calculated rates reflect <u>catchment-wide</u> changes in erosion intensity as opposed to the transfer of material from sediment <u>stores</u>. Additionally, as a necessary simplification, localised deltaic sedimentation has been ignored. Detailed core sampling of these areas would otherwise be required.

Because of these additional possible sources of error, <u>instead</u> of catchment erosion rates, catchment net sediment <u>supply</u> rates are presented here. These are calculated in the same way as catchment erosion rates (namely, lake sediment input ÷ catchment area, see appendix 4), but the use of a different name acknowledges the greater potential errors which may be present. However, net sediment supply rate is be related in some way to "true" catchment erosion rates, and does therefore provide a basis for comparison with catchment erosion rates presented in other published studies.

The widely averaged accumulation rates presented in Figure 4.4 a record a similar general

trend for the Crummock Water cores. Initially high early post-glacial rates decline in the mid Post-glacial and then increase again markedly in the most recent sediments. Widely averaged early post-glacial accumulation rates do not reach the same (high) peak values as for the more detailed rates because the wide time interval forced by scarcity of correlation features therefore incorporates both very high and relatively low accumulation rates. Between core variations in accumulation rates in Unit 2 (*circa* 5 270 - 7 330 B.P.) suggest marked sediment focusing toward the lake centre (core C2L). Sediment accumulation rates at the lake centre are an order of magnitude greater than for other cores. This makes calculation of lake-wide accumulation rates for both Buttermere and Crummock Water (Figure 4.4 b) for this period speculative. Recent accumulation rates are indicative of a more uniform sediment accumulation across the main lake basin. In the last 1 000 years increased accumulation rates across the whole basin and a shift to relatively greater accumulation at the lake head relative to other areas are recorded.

Lake-wide accumulation rates (Figure 4.4 b) for Buttermere contain certain differences compared to those for Crummock Water, although the general pattern of change (of a decrease from the early post-glacial rate, and marked increase in the upper sediments corresponding to the last 1 000 years of deposition) is similar. For example, the early post-glacial rates in Buttermere of *circa* 13 mg.cm⁻².yr⁻¹ are only 30% of those of Crummock Water fro the same period. This may reflect a loss of detail forced by wide averaging, but the higher organic contents of the Buttermere cores produces a lower dry density values, and a comparatively smaller unit thickness. This suggests the existence of "real" variation in dry sediment accumulation rates between the two lakes.

Uniformly low rates for Unit 2 in cores B2L and B1L indicate that sediment accumulation during this period of the early Post-glacial was minimal. Whether this is due to sediment focusing, as in Crummock Water, cannot be determined because this unit is not penetrated in core B3L, from the deepest part of the lake. Strong focusing shown by the Crummock Water sediments suggests that lake-wide sediment accumulation rates from this period must be regarded as extremely tentative. Accumulation rates in the sediments above Unit 2 are comparable to those in Crummock Water, although maintaining consistently slightly higher values. A similar pattern of increasingly high rates in the lake head area, where the major river inflows taker place, is seen. The only figure available for the most recent period (*circa* 1800 A.D. - 1987 A.D.) is at 88 mg.cm⁻².yr⁻¹, nearly double that of the rate at the lake head in Crummock Water .

Data from published studies of catchment erosion, while not providing exact analogues, do afford a basis for <u>general</u> comparison of the accumulation and net sediment supply rates calculated in this study. The Hubbard Brook catchment experiment remains one of the most closely-monitored watersheds. Studies there have yielded catchment erosion values in the range 7 - 79 kg.ha⁻¹.yr⁻¹ (average 25 kg.ha⁻¹.yr⁻¹) for a mature forested, and 16 - 380 kg.ha⁻¹.yr⁻¹ (average 156 kg.ha⁻¹.yr⁻¹), for subsequently "experimentally" deforested catchment respectively (Borman *et al*, 1974). These data suggest that, with no other disturbance of the land surface, an average 6.5-fold increase in catchment erosion rates takes place following deforestation.

Changes of a similar magnitude have been observed by Davis (1976), where deforestation and development of agriculture resulted in a calculated approximate 10-fold increase in catchment erosion rates in a small, low-lying drainage basin. Catchment erosion rates rose from *circa* 90 to 900 kg.ha⁻¹.yr⁻¹ on the establishment of European agriculture.

Approximate catchment erosion rates of 10 kg.ha⁻¹.yr⁻¹ have been calculated by Pennington (1978a), suggesting rates for the Ennerdale catchment (in the English Lake District) prior to human settlement, and a similar value for the "near natural" catchment of Loch Clair (N.W.Scotland).

In Crummock Water, the highest net sediment supply rates are found in the early post-glacial, where an 8-fold increase over minimum rates is observed. The total catchment deforestation observed in the Hubbard Brook studies (discussed above) provides probably the most direct analogue to the recently deglaciated landsurface. Minimum net sediment supply rates of around 40 - 60 kg.ha⁻¹.yr⁻¹ in the period 7 100 - 2 000 B.P., are

comparable to tentative rates for undisturbed upland catchments proposed by Pennington (1978a). These rates increase by a factor of 5.5 in recent times (to 217 kg.ha⁻¹.yr⁻¹). The Buttermere and Crummock Water results are therefore similar to those discussed above in terms of absolute values and the magnitude of changes, and the consistently low sediment accumulation rates up to *circa* 2 000 years B.P. This suggests that any anthropogenic modification of the catchment until then was of limited extent.

Comparison of sediment accumulation rates between Buttermere and Crummock Water shows that prior to *circa* 5 000 B.P., accumulation and catchment net sediment supply rates in the Crummock Water catchment (which includes that of Buttermere) were between two or three times those of the Buttermere catchment. After *circa* 5 000 B.P., all rates increase in the Buttermere catchment, equalling or exceeding those in Crummock Water. In the most recent of the widely spaced time intervals (1987 A.D. - *circa* 1800 A.D., Figures 4.3 and 4.4), rates in Buttermere approach twice those of Crummock Water.

Variations in sediment accumulation and catchment net sediment supply rates for Buttermere and Crummock Water are therefore consistent with the known record of environmental change in the Lake District, and the predicted events shown in Figure 2.6. They are produced by changes in climate (and thereby in geomorphic processes) and thereafter (that is, post *circa* 5 000 B.P.) by the combined effects of climate and anthropogenic influence (section 2.2, p.38). The variations in rates <u>between</u> Buttermere and Crummock Water suggest a marked change in sedimentation pattern, initially with Crummock Water as the major sediment sink. After 5 000 B.P., Buttermere displays the higher sediment accumulation rates. This reflects changes in sediment source and/or lake hydrodynamics. The first are investigated in this chapter, and the latter in chapter 5).

4.4.2. Catchment weathering and erosion regimes.

i) XRD analysis.

In addition to the mineral types present, their crystallinity and concentration, XRD peak

intensities are strongly influenced by particle size and by preferential particle orientation within a sample. The sample preparation method used here minimises particle size and orientation effects by a) thorough grinding to a talc-like consistency (Till and Spears, 1969), and b) back-packing into sample holders rather than using strong sample compression or slurry evaporation methods (Norrish and Taylor, 1962).

Calculation of the integrated line intensity values can be significantly affected by the estimation of the position of the baseline within the "noise" at the base of a trace. The method of calculation used here is robust, as peak width is measured at half peak height, minimising errors in baseline positioning ($op \ cit$). Variations in mineral proportions and their ratios for all Crummock Water samples are each expressed as a percentage of the total line intensity for the sample. This allows direct comparison of variations in mineral proportions between samples (Figure 4.7).

Artificial variation in percentage mineral proportions, due to a large variation of one mineral causing a percentage change in the remainder, must be considered by examining raw, as well as transformed, data. Comparison with the raw data (that is, intensity profiles not expressed as a percentage of a sample's total intensity, appendix 5) confirms the existence of similar profiles for both data sets. Thus no artificial variation is imposed by mathematical transformation as percentages of a closed number system.

The ratios obtained from these data (Figure 4.7) can provide a useful index of the degree of weathering of the supplied sediment (Dean and Goreham, 1976), and hence of the types of erosional regime operating within a catchment (Chambers, 1978). As all feldspars are less durable both physically and chemically than quartz, then other factors remaining constant, with prolonged weathering, the proportion of quartz relative to feldspar will increase.

Within the limitations of the wide sampling intervals employed, variation in the quartz:feldspar ratio from the Crummock Water sediments suggests the existence of two periods of increased erosion, one at the base of the core, coinciding with the deposition of Unit 1, the second in the upper 0.5 m of the core (corresponding to the upper part of Unit

4). The intervening period, although represented by fewer data points, contains much lower quartz:plagioclase ratios, suggesting a long period of relatively less intense catchment erosion, with greater *in situ* weathering of source sediments.

ii) Chemical indicators of erosion regime.

The mobile elements K and Na in lake sediments have been successfully used as one line of evidence for soil erosion, for example by Pennington (1981) for Blelham Tarn in the English Lake District, and by Huttunen, Meriläinen, and Tolonen (1978) for Hakojärvi in southern Finland. The underlying hypothesis, originated by Mackereth (1965, 1966a), is that periods of active erosion do not permit the development of deep, weathered soil profiles. Thus mass transport of relatively unleached soils should increase the mineral fraction of lake sediments as a whole and enhance their K and Na content. Less active catchment erosion allows development of more stable soil profiles, deep weathering diminishing the base content of mineral material prior to its removal and subsequent deposition as lake sediment. Hence high K and Na values in a lake sediment profile suggest relatively active catchment erosion processes and low K and Na values relatively less erosion. The lower values are a product of greater leaching of physically stable soil profiles.

Some later writers have questioned the direct application of Mackereth's hypothesis. Chambers (1978) observed a positive correlation between K and illite (reflecting K in the illite crystal lattice), and the association of illite with the finer (silt and clay) component of sediments from Brotherswater (the English Lake District). He concluded that mobile elements such as K may not be direct indicators of soil erosion, but are related to the proportion of sedimentary illite and particular particle size fractions in lake sediments. However, a major source of sedimentary illite is the degradation of feldspars in weathered soil profiles. Hence, measurement of K may record the input of soil-derived material, although not in the direct manner that Mackereth originally proposed. Particle size characteristics of lake sediments are in turn influenced by several factors including available source material, intensity of erosion and transport mechanisms, and lake hydrodynamics.

Engstrom and Wright (1984) report a further variable for consideration in the interpretation of lake sediment geochemical records, namely the influence of authigenic minerals and biogenic mineral Si which can contribute significantly to the total dry weight of freshwater sediments. These will tend to dilute the mineral chemical profile, and the effects are,

> "greatest in highly organic deposits and least in minerogenic sediments", (Engstrom and Wright, 1984, p.31)

Engstrom and Wright contend that such a process of dilution by biogenic material may be responsible for the trend in the Na and K profiles observed by Mackereth from the English Lakes.

Comparison of chemical profiles and silt fraction (which mirrors clay proportion) together with LOI values for cores B2L and C2L are shown on Figure 4.8 (p.113). Chemical and silt proportion profiles are initially similar in Unit 1, becoming less so above. Some similarity in shape but not magnitude is identifiable in Unit 3, where small increases in both silt fraction and of K and Na are observed.

Biogenic fractions have not been determined in this study, and therefore their possible diluting effects cannot be directly evaluated. However, the organic content of the Buttermere and Crummock Water sediments is approximately indicated by LOI, and usually is related to lake productivity. LOI may therefore be used as an approximate indicator of productivity, hence as a guide to likely changes in the proportion of sedimentary biogenic Si.

The weak inverse relationship between LOI and the Na/K profiles (Figure 4.8) therefore supports the idea of dilution of allogenic material by input of biogenic matter. However, this effect is not at all marked.

This discussion has identified complications in the straightforward interpretation of chemical profiles as proposed by Mackereth (1965, 1966). Evidence from the Buttermere
and Crummock Water sediments shows that variation in both grain size and in the proportion of inorganic biogenic material may influence the chemical profiles determined. These influences are not straightforward however, and the successful application of Mackereth's hypothesis in other studies (for example, Pennington, 1981, Huttunen *et al*, 1978, discussed above, also of Heathwaite, O'Sullivan, and Appleby, 1991) suggest that it is not invalidated by these latter criticisms, but rather that chemical profiles respond to several influences. These include:

- i) the intensity of catchment weathering and erosion;
- ii) variations in grain size distribution;
- iii) the proportion of biogenic mineral material present;

These provide further complicating factors which should be considered in the application of this method.

The Buttermere and Crummock Water K and Na profiles therefore suggest that the sediments laid down in the upper 0.5 m, and at the base of the profiles were laid down during periods of intense erosion. These correspond to the periods of the Loch Lomond Stadial (ending *circa* 10 000 B.P.), and that from *circa* 1 000 B.P. to the present.

From *circa* 10 000 to about 1 000 B.P., lower mean K and Na values indicate a generally less intense rate of physical erosion occurring in the catchment. Although these <u>may</u> have been altered by changes in-lake production of biogenic Si (as discussed above) this effect is unlikely to be significant in an oligotrophic system such as this. Also within this "middle" period, four less distinct increases in K and Na values indicate increased supply of less-weathered sediment to the lakes, and hence denote intervals of enhanced mass transport of sediment. The positions of these features and the more recent K and Na peaks, with their approximate ages (calculated from the chronology established in section 3.3) are included on Figure 4.8 (p.113).

4.4.3. Source-sediment linkages.

i) Mineral magnetic indicators.

Before analysing the source-sediment relationships indicated by comparison of catchment and lake samples, some discussion to explain the observed magnetic characteristics of the catchment sources is needed. These are summarised in Figure 4.13 (page 120).

The Skiddaw Slates, group 1, exhibit minimum values for the concentration-dependent magnetic parameters and maximum values for the hard % parameter. These suggest the existence of very low concentrations for the total magnetic assemblage dominated by "hard", that is antiferrimagnetic, mineralogies. Such characteristics are a feature of slate substrates, similar magnetic properties having been observed, for example, from North Wales, Exmoor, and the Macclesfield Forest (Maher, 1986, Stott, 1987).

The Ennerdale Granophyre (group 3) shows maximum concentration-dependent

parameters (χ lf and SIRM), of ARM (indicating a dominant proportion of coarser-grained, SD/MD material), and of the soft parameter (indicating a high proportion of soft, ferrimagnetic mineralogies). The plotted position of the Ennerdale Granophyre (Figure 4.11, page 117), where a very high ARM value has distorted the plotted position to suggest a higher χ fd% value than actually measured shows the presence of stress within the plot for this sample.

The Borrowdale Volcanic Group (group 2) are transitional in their plotted position, and therefore in their magnetic characteristics, between the Ennerdale Granophyre and the Skiddaw Slates. This is due to a low absolute concentration of ferromagnetic material, where the hard, antiferrimagnetic mineralogies tend to dominate the measured magnetic characteristics.

Group 4, sediment derived from the Ennerdale Granophyre, is similar in terms of total

ferrimagnetic concentration (χ lf and SIRM) to the Ennerdale Granophyre, the underlying bedrock, but is disassociated from its parent material particularly by its lower ARM (a measure of the total concentration of the coarser (MD size) component). The IRM₊₂₀/ARM parameter, a measure of the coarser (MD) fraction of the ferrimagnetic component only, remains similar for the two groups. Such a pattern suggests the preferential diminution of coarse grains of the antiferromagnetic component during weathering. This group is represented in the Crummock Water catchment by a sediment from location 19 (sample 19ss), collected from a stream whose major tributaries drain the Ennerdale Granophyre, with other tributaries from the area underlain by the Skiddaw Slates (see location map, Figure 4.9, page 115). This group displays characteristics intermediate between the two contributing bedrock sources (the Ennerdale Granophyre and the Skiddaw Slates) and the soils (group 6).

Group 5 consists of samples from the uppermost, weathered, horizon of clay-diamicts. The upper parts of these are weathered in comparison to the underlying horizons, with oxidation characteristically expressed as a deeper red than that of the underlying material (10 YR 6/4 compared to 10 YR 6/3). Their positions on the ordination plot indicate the presence of a higher total ferromagnetic concentration than, for example, that found in the Skiddaw Slates or Borrowdale Volcanic Group.

Higher $\chi fd\%$ and ARM/ χ values than most other groups reflect greater proportions of the fine, SP, and SD grains. This may be a product of physical size reduction during the genesis of the diamicts or secondary mineral growth during weathering. That values of these variables are greater than those of the underlying unweathered diamict (samples 281 and 211) would tend to support the second hypothesis.

Maher (1986) and Maher and Taylor (1988) have shown that that such development, leading to <u>enhancement</u> of finer ferrimagnetic oxides mainly in the SP size range, is a characteristic process in many soil-forming environments. Mullins (1977), has reported such magnetic enhancement occurring preferentially within the clay fraction (<2 μ m) of

soils. This fraction constitutes a major component of this material.

Group 6, the remaining soils and stream sediments sampled in each catchment, are clustered as a relatively diffuse group towards the central area of the respective ordination plots (Figures 4.11 and 4.12). The group lies between the extremes provided by the bedrock samples, but is more closely associated both with the unweathered diamict (for example, samples 281 and 211 in the Buttermere and Crummock Water catchments respectively). This suggests either that the glacial clay-diamict present throughout the catchment forms a major source for the group 6 sediments, and/or that the two groups share common secondary magnetic minerals arising from weathering in the subaerial environment.

Both of these relationships are in fact likely: diamicts are widespread throughout the catchment, and most soils are developed directly on these rather than on the bedrock. The upper horizons of the diamicts will also have been subjected to soil-forming processes during initial plant colonisation and pedogenesis. Thus, similarity between the magnetic characteristics of diamicts and soils would be expected (soils having "inherited" magnetic characteristics from their parent material). Also, similar evolution between diamict and the contemporary organic soils of group 6 would be expected. This has resulted in enhancement of certain magnetic properties within the upper diamict horizons (namely, increases in the concentration of the soft, ferrimagnetic, magnetic component in the finer (SP and SD) grain size ranges). That the clay-diamicts generally exhibit a greater enhancement is a function of the predominance of the clay (< 2 μ m) fraction in this substrate, more susceptible (as discussed above) to the development of secondary "pedogenic" ferrimagnetic minerals.

No consistent differentiation between soil horizons is displayed in these results. This contrasts with the findings of previous studies such as those by Stott (1986, 1987). In these however, the soils under investigation were underlain by more homogeneous lithologies (gritstones and slates), characterised by consistently low susceptibilities and an antiferrimagnetic (hard) mineralogy. These characteristics observed in this catchment in the

magnetic behaviour of the Skiddaw Slates only). In the studies undertaken by Stott (*op cit*), these characteristics facilitated differentiation between soil horizons: the A-horizons being characterised by high concentrations of *in situ* secondary ferrimagnetic minerals and atmospherically-derived magnetic spherules; the B-horizons by values intermediate between A-horizons and the slate bedrock (Stott, 1987). Developed on steep and/or exposed hillsides, many of the soils in the Buttermere and Crummock Water catchment are also relatively immature, leading to the reduced input of a uniform soil magnetic signature.

In summary, the wide distinction between bedrock types in this study has effectively obscured any differentiation of catchment soil material with respect to their vertical position within soil profiles. However, the results potentially provide a method of differentiating catchment sources with respect to geographical position. The widespread distribution of glacial diamicts, which form the parent material for many of the catchment soils, "blurs" such geographical distinction however, as unweathered diamict samples from widely separated parts of the catchment (samples 281 and 211, Figures 4.11 and 4.12 from the extreme east and west ends of the catchment respectively) exhibit magnetic characteristics similar to those of the Borrowdale Volcanic Group. Hence soils from different locations share a rather similar parent material.

Although these results preclude detailed identification of sources with respect to their position in a soil horizon, they do provide a good basis for comparison of the lake sediment samples with primary (bedrock) and secondary (diamict, soil, or stream sediment) sources, and variations in these with time. This is considered in the discussion below.

There are, however, a number of methodological compromises to be considered. First, quantitative estimates of proportions of source groups present in the lake sediments have not been attempted in this study. This reflects the complexity of potential sediment sources. Although quantitative estimates of such potential sediment sources based on mineral magnetic analyses of lake and catchment sediments <u>have</u> been made (Stott, 1986, Yu and Oldfield, 1989), these studies were based on catchments substantially simpler than that of

Buttermere and Crummock Water. In terms of variety of bedrock lithologies, secondary sediment sources (where, for example, in the Buttermere and Crummock Water catchment, frequently less well-developed soils have resulted in a less significant soil magnetic signature), and a widespread drift mantle (as present over much of the Buttermere and Crummock Water catchment). This study area therefore represent a rather more complex catchment.

Secondly, Hirons and Thompson (1985) and Yu (1989), amongst others, have demonstrated that mineral magnetic properties of natural sediment samples are influenced

by the size fraction measured, even in the range below 63 μ m (4 ϕ). For example, Yu

(1989) separated sediment samples into 1 ϕ intervals between 0 and 10 ϕ , and observed variations in mineral magnetic properties between these classes, <u>including</u> those classes within the silt and clay size intervals (4 - 8 ϕ , and 8 - 10 ϕ respectively). Samples of catchment material from Buttermere and Crummock Water were therefore sieved to < 63

 μ m (4 ϕ) in order to reduce an additional source of between sample variation.

Separation to the level of <u>single</u> phi size classes is impractical in this study however, owing to the relatively large number of samples (*circa* 230) used for source - sediment comparisons. J.P.Smith (1986) reported that comparison of the < 63 μ m fraction of catchment samples to lake sediment samples provided an improvement on comparisons based simply on bulk sediment samples. Therefore, as an approximation, this compromise is considered acceptable. The size fraction used for analyses in this study (< 63 μ m) is therefore a compromise. This is an improvement on bulk sediment comparisons, but less precise that use of more specific size fractions. Some variation between samples may therefore be explained by between sample variation in the < 63 μ m classes, in addition to variation in changing source components.

This source of variation, additional to that caused by actual changes in sediment sources, is illustrated by the direct comparison of lake sediment magnetic groups, as shown on the ordination plot, Figure 4.14. This shows a very similar pattern of change of the magnetic characteristics of sediments between lakes, and reflects relative enrichment in most of the

Buttermere sediments of both very fine (SP) and coarser (MD) ferrimagnetic grains (as observed in the results, section 4.3.3). This <u>could</u> be a product of changing sediment sources however, <u>systematic</u> differences between equivalent pairs of groups from each lake, despite significant changes in over all magnetic characteristics of each pair, suggest that they originate from common source sediments which have changed over time. Such systematic changes between equivalent sediment groups from each lake are most easily explained by the depletion of the Crummock Water sediments of particular sediment fractions, with Buttermere acting an an upstream sediment trap.

Relative to equivalent Crummock Water groups, the Buttermere sediments are generally enriched in the coarser (MD) ferrimagnetic fraction. Such coarser, denser, material will naturally settle out preferentially in the headwater areas of a lake-sediment system. This would not be expected for the much finer SP fraction, which is also enriched in the Buttermere sediments, although examples of very fine material closely associated with grains approximately an order of magnitude larger (the finer adhering to the coarser) have been observed during the investigation of other lake sediments. For example: Bradshaw and Thompson (1985) deduced the presence of coarse (MD) and possibly finer (SD) grains coating non-magnetic particles "like currants on a bun"; and Morrison (1981) observed small clay grains ($\leq 3 \mu m$) adhering to larger particles ($\geq 20 \mu m$) during SEM examination of a variety of glacial lake sediments. A similar process, would explain the simultaneous deposition of both coarse and very fine components of the magnetic assemblage, allowing the transit of a more restricted rage of fine grains through Buttermere, to be subsequently deposited in Crummock Water. Pipette analysis of bulk sediment samples (section 3.3.1, Figure 3.9) indicates a consistently greater proportion of fine material (clay range) in the Crummock Water sediments relative to those of Buttermere. This is consistent with the hypothesis of Buttermere acting as an upstream sediment trap, filtering a proportion of coarser sediment before it reaches Crummock Water.

Thus, whilst the exact mechanism of enrichment of the Buttermere sediments in particular size fractions remains unidentified, magnetic and bulk granulometric analyses do point to an hydrodynamic interpretation (very fine grains adhering to much larger grains settling out preferentially nearer the point of sediment input to the lake) and highlight a natural

modification of the sub 63 μ m sediment fraction. However, the observed offset of Crummock Water lake sediment magnetic groups on ordination plots might have been reduced or removed by the analysis of much more specific size fractions.

The particle size dependence of mineral magnetic measurements is, in this respect, something of a problem. Although more specific particle size separation, and consequently particle-size-related magnetic measurements, may be undertaken, such separations are time consuming and become somewhat impractical in studies involving large numbers of samples. Indeed, it is the relative speed of the mineral magnetic approach that originally stood out as a major advantage of the method (Thompson and Oldfield, 1986).

Despite these inherent problems, the value of the mineral magnetic approach has been demonstrated by its successful application in numerous investigations (section 1.4). In this study, even with the additional variability resulting from changes in the < 63 μ m fraction, the mineral magnetic results combine to provide a convincing argument for source-sediment relationships. These are also generally corroborated by the other techniques employed (see discussion, chapter 6). The major points to note here are:

i) catchment samples, are easily objectively clustered into lithologically distinguishable groups (Figure 4.10, and illustrated on ordination plots, Figures 4.11 and 4.12);

ii) sediments from both Buttermere and Crummock Water, are easily objectively clustered into groups, which are not only stratigraphically coherent, but enable differentiation of equivalent sediment groups <u>between</u> the two lakes (despite "artificial" variation between sediments from the two lakes, Figure 4.14, page 122);

iii) correlation coefficients between actual and plotted sample values on all ordination plots are statistically significant, suggesting little loss of data during mathematical transformation;

iv) from i, ii, and iii, above, ordination plots of catchment and lake sediments therefore

reflect <u>actual</u> changes in sample mineral magnetic characteristics. These plots also illustrate distinct similarities between source and lake sediment (Figures 4.11, 4.12), for example between the lower lake sediment magnetic groups (I, II, III) and the Skiddaw Slates and Borrowdale Volcanic Group, and between the upper lake sediment groups from unit 4 (groups V - VII and V - VIII, Buttermere and Crummock Water respectively) and the undifferentiated soils of catchment group 6.

The patterns are clear, even with the additional variability between samples potentially arising from grain size variation in the sub 63 μ m fraction. The basal groups (I - III, Buttermere, and I - IV, Crummock Water) exhibit greatest similarity to the Skiddaw Slates and Borrowdale Volcanic Group, with a progressively increasing contribution of secondary sources (represented by catchment group 6). The overlying lake sediment groups are similar to the secondary sources (catchment group 6), indicating an increasingly concentrated magnetic assemblage, with increasing proportions of the fine, SP, component

(cf increasing χfd% values) a clear indication of derivation from (upper) soil horizons (Maher and Taylor, 1988).

Zielinski (1989) proposed that in catchments of magnetite-bearing bedrock, the input of unaltered bedrock-derived magnetite is the principal control on magnetic susceptibility.

Hence it was suggested that variations in κ along a sediment profile may be used as a guide to the efficiency of physical erosion processes which lead to deposition of unaltered bedrock material.

This argument is simplistic, in that no account is taken of secondary sources of magnetite. This may be formed in developing topsoils in areas with or without primary (bedrock) magnetite sources (Maher and Taylor, 1988) or of deposition of atmospherically-derived magnetic particles which significantly affect the magnetic records of recent sediments from many western European and North American lakes (Oldfield and Richardson, 1990; Locke and Bertine, 1986; Goldberg *et al*, 1981).

The characteristic magnetic properties of topsoil derived magnetite however (notably, high

 χ fd% values) reflect the small (SP) size of secondary magnetite grains formed in the topsoil environment (Maher, 1986; Maher and Taylor, 1988). The deposition of magnetic atmospheric particles in large numbers is a geologically recent phenomenon, post-dating the Industrial Revolution (Clymos *et al*, 1990, see also chapter 3). Thus, in catchments containing primary magnetite sources, which are likely to dominate the magnetic susceptibility of a lake sediment record, susceptibility measurements <u>may</u> be used as a rough guide to efficiency of catchment physical erosion processes. This should preferably

be considered in conjunction with mass specific variables, such as $\chi fd\%$, which can identify the presence of "soil" magnetite (Maher and Taylor, 1988) and hence differentiate between erosion of primary and secondary sources. In addition, microscopic examination of sediment magnetic extracts may identify the presence and likely contribution of industrially-derived magnetic spherules (McLean, 1990).

Of the lithologies present in the Buttermere and Crummock Water catchment, the Ennerdale Granophyre is a primary source of magnetite, typically of coarser (MD) grain sizes. The patterns of changing volume susceptibility (κ) and χ fd% outlined in the results (section 4.3.2) may therefore indicate increased erosion of primary sources leading to the observed high values at the base of the sediment cores (unit 1).

Increased values of κ towards the top of the sediment cores are mostly associated with increases in the χ fd% variable however. These identify the presence of a very fine (SP) magnetic component, suggesting supply of secondary "soil" magnetic material.

High κ values towards the sediment/water interface (from 11 cm to surface in core

C2L/C3S), associated with decreased $\chi fd\%$ and increased IRM₊₂₀/ARM, imply a relative reduction in the proportion of SP magnetic grains and an increase in coarser (SD/MD) grains. These <u>could</u> be derived from the Ennerdale Granophyre (cf. mineral magnetic

characteristics of catchment samples, appendix 6), but after the Industrial Revolution, inputs of atmospheric magnetic spherules, typically enriched in the coarser magnetic grain sizes (Hunt *et al*, 1984) are another potential source. In this case, they have been visually identified as contributing to this most recent increase in susceptibility (section 3.4.2, part ii).

Variations in κ in the Buttermere and Crummock Water profiles therefore identify the periods of most intense erosion as having been during the deposition of the Late Glacial minerogenic material (unit 1), and the uppermost sediment (*circa* 0.5 m) of unit 4. However, reference to mass specific parameters identifies the upper increase in κ as derived from secondary, soil, rather than primary, rock, sources. Near the sediment/water interface, enhanced κ values derive not from changes in erosion intensity, but from the input of magnetite-rich atmospheric spherules of industrial origin. With the possibility of such misinterpretation using κ profiles alone, particularly when applied to recent sediments. Zielingkile (1080) have the input of the sediments.

sediments, Zielinski's (1989) hypothesis is considered as extremely unreliable. It is important to corroborate volume susceptibility results at least with reference to mass specific magnetic parameters.

ii) XRD-derived indicators.

Changes in the ratio of chlorite to illite may also be used as a guide to the proportion of soil- and rock-derived material incorporated into lake sediments. Chambers (1978) in a study of contemporary sediments from Brotherswater (also in the English Lake District), found that this ratio varied with distinct changes in types of source material. In the Lake District, sources of chlorite are primary, mainly from erosion of the chlorite-rich slates within the Borrowdale Volcanic Group, whereas sources of illite are secondary. These are mainly soils, where the mineral is derived from the weathering of feldspars.

Changes in the chlorite:illite ratio therefore record variations in the proportion of soil-derived and rock-derived material, allowing use of the ratio as an indicator of changing

sediment source type. Changes from primary, rock, to secondary, soil, sources may also reflect varying catchment erosion processes. In this study, the chlorite:illite ratio derived from semiquantitative XRD data from core C1L is used to supplement mineral magnetic investigation of changing sediment sources and types.

As discussed in section 2.1, the chlorite-rich tuffs from the Borrowdale Volcanic Group, form the major source of the mineral within the Buttermere and Crummock Water catchments. The first variation in the chlorite:illite ratio in the Crummock Water profile, at the base of core C1L is associated with an high proportion of chlorite. It therefore indicates greater physical weathering of primary (that is, rock) sources, and this material is derived from the Borrowdale green slates.

The second variation, near the top of core C1L, is associated with a higher proportion of illite, suggesting increased delivery of a more weathered secondary source material. The synchronous rise in illite and fall in feldspar for this second period (Figure 4.7) suggests that the latter is being degraded to its weathering product, illite. The most likely source for this material is increased erosion of weathered soils in the catchment, supplying some feldspar together with increased proportions of its weathering product.

4.5. Summary

Sediment accumulation rates, sediment input, and catchment erosion rates have been calculated for Buttermere and Crummock Water (Figure 4.4). The most detailed data is available for the more closely-dated Crummock Water sequence, but correlation between Buttermere and Crummock Water has allowed calculation of more widely-averaged rates for Buttermere.

The pattern of sediment accumulation rate change is:

i) peak rates near the case of the sediment sequence (early Post-glacial);

ii) steadily declining rates to circa 2 000 B.P.;

iii) increasing rates post circa 2 000 B.P. to the present.

The areal pattern of sediment accumulation within the lakes has also changed over time. At the base of the sequence, rates were initially relatively consistent across the lake basins (units 1 and 2). Strong sediment focusing occurred during the deposition of unit 3. Increases in sediment rates become proportionately greater in the lake head (delta) areas towards the top of unit 4.

Catchment net sediment supply rates are presented as a guide to catchment erosion rates. Comparison with erosion rates from published studies indicates that the magnitude of change between the highest and lowest net sediment supply rates in Buttermere and Crummock Water are similar to those observed in the experimental deforestation of Hubbard Brook (Bormann *et al*, 1974). In Buttermere and Crummock Water, this peak in sediment supply corresponds to the change from a sparsely vegetated landsurface of the Late-glacial and early Post-glacial, to a more densely vegetated landsurface thereafter.

The lowest net supply rates (*circa* 40 kg.ha⁻¹.yr⁻¹) are of a similar, in order of magnitude, to approximate catchment erosion rates of 10 kg.ha⁻¹.yr⁻¹ calculated for Ennerdale (western Lake District) and Loch Clair (N.W. Scotland) by Pennington (1978a). This is considered reasonable in view of the potential errors associated with the calculation of both figures, (section 4.4.1, page 125).

XRD and mobile element (K and Na) analyses provide an indication of catchment weathering and erosion regimes (Figures 4.7 and 4.8 respectively). These analyses identify two major periods of more intense physical erosion within the catchment. The first in the early Post-glacial, and the second dating from *circa* 1 000 B.P. (reaching maximum intensity at *circa* 150 B.P.). The more detailed mobile element analyses also suggest the existence of several less marked periods of active physical erosion between these two periods (Figure 4.8).

Despite methodological compromises, the mineral magnetic approach provides clear

indications of changes in source types (Figures 4.11 and 4.12). Early Post-glacial sediments at the base of the sediment sequence exhibit most similarity to unweathered bedrock sources. Thereafter, secondary sources (including catchment soils, stream sediments, and glacial clay diamicts) increasingly contribute to the lake sediments. Towards the top of the sequence (from *circa* 1 000 B.P.) a marked increase in the proportion of fine (SP) magnetic grains indicates increasing input of topsoil-derived material.

As a guide to the relative proportions of primary (bedrock) and secondary (soil) sources, the chlorite:illite ratio (determined from semiquantitative XRD data) broadly corroborates the relationships recorded by the mineral magnetic comparisons (Figure 4.7). Glacial and early Post-glacial sediments contain high chlorite:illite ratios, indicating the relative dominance of primary sources. Towards the top of the sequence, in unit 4, increases in primary sources are indicated, but rising proportions of illite suggest the enhanced input of soil-derived material, the illite most probably derived from degradation of feldspars within the soil environment.

5. LAKE DEPOSITIONAL MECHANISMS.

5.1 Introduction.

As well as acting simply as a sink within which catchment-derived sediments are deposited, "internal" lake processes may be of importance in further modifying the sediments supplied. In oligotrophic lakes with clastic input, these transformations are restricted mainly to changes in physical sorting rather than the authigenic processes such as the growth of secondary sulphide minerals and dissolution which may occur in eutrophic lakes (see discussion in section 1.4, also Hilton *et al*, 1986, Snowball and Thompson, 1990). Therefore in this chapter, physical variations in sediment properties, that is, sediment distribution, texture, and grain size variation are examined in order to provide information on the lake sediment depositional mechanisms and thereby the influence of the lake water body on the form and distribution of sediments. The type of analyses used and reasons for their use are discussed briefly below.

5.2 Methods.

On the micro-scale (*circa* 0.1 - 1 mm), vertical variation in grain size and sediment texture can be used to determine the influence particularly of sediment influx and lake water stratification and their interrelation (Figure 5.1, and Stürm, 1979). From Figure 5.1, it may be seen that different combinations of sediment input mechanism and lake water stratification produce characteristic vertical variations in grain size distribution. This figure illustrates, for example, how continuous sediment texture. Similar influx conditions during stratification which subsequently breaks down (box 5) produces a "chaotic" sediment with bands enriched in fine clays following the lake overturn. Identification of vertical variations in sediment textures, requiring thin section examination of undisturbed sediments, may therefore be used to interpret ancient lake water stratification and sediment influx characteristics.

Granulometric analyses (see chapter 3) indicated that, on a very broad scale, vertical grain



Figure 5.1. Idealised sedimentary structures formed by interaction of two hydrological parameters (water stratification and suspended matter influx) in an oligotrophic lake dominated by clastic deposition. (After Stürm, 1979).

size variation within the sediment column is not pronounced. Visually, the sediments are homogeneous apart from the green (chlorite) laminae commonly present, at irregular intervals through the sediment column. Sampling for thin sections was therefore directed towards those containing green laminae, with surrounding homogeneous material retained above and below laminae in the prepared material. Moreover, these laminae, although present in Buttermere, Crummock Water, and the Windermere sediments (author's personal observations of all sequences) have been described only rarely and briefly, for example by Pennington (1943) and by Mackereth (1965, 1966a) in a passing reference, "visible stratifications in the [Post-glacial] organic matter, often in the form of ... dark green laminations" (Mackereth, 1966a, page 170).

The samples from Buttermere and Crummock Water were selected mainly from the long cores B2L and C2L during initial slicing, which allowed close inspection of the fresh sediment. No thin sections of the clay rich units (3, 1, and the lower part of unit 2) were studied, as impregnation of these was never satisfactory.

Sediment thin sections were mostly prepared by impregnation of fresh undisturbed sediment blocks with G4 (a commercial damp sealing compound). Undisturbed blocks of sediment were extracted from fresh cores using small Corbienier tins and successively dowsed in G4, which replaces water within the sediment matrix. Blocks were finally cured at around 60°C. Comparison with thin sections prepared using the more standard method of PEG (polyethylene glycol, molecular weight 6000) impregnation indicates that the only discrepancy between the two methods is a different shade to the background colour of the sections.

Impregnation using G4 ideally provides a more durable sediment block than PEG, which has been used for the impregnation of unconsolidated sediments in other studies (O'Brien, 1971). PEG solutions did not soak satisfactorily into the small sediment blocks used and do not provide a hard enough block for grinding. PEG is a waxy and ductile compound whilst most component grains are hard and non-ductile. They are therefore frequently ripped from the block rather than being ground with it.

As PEG is water soluble, after impregnation, thin sections must be ground individually

using a paraffin based paste, which is also time consuming. The G4 solution therefore provides a harder impregnated sediment block, although holes within thin sections were frequently encountered where material was not completely impregnated and bonded.

On a still smaller scale, of the order of microns, the orientation of clay flakes within a sediment may be used as an indication of either a turbiditic or hemipelagic mode of deposition. O'Brien *et al* (1980) demonstrated by SEM analysis of fabrics that a random clay flake orientation is associated with deposition from a turbidity flow, and a preferred orientation is associated with hemipelagic sedimentation. These differing fabrics are produced by contrasting sedimentation styles. Turbiditic clay deposited more rapidly in the flocculated state produces unsorted, random fabrics. Hemipelagic clay deposited more slowly from dilute suspensions allows the attainment of preferential flake orientation hence formation of sediment fabric showing an ordered fabric (*op cit*).

Small samples (up to 2 cm in length) for SEM analysis were extracted from fresh and undisturbed sediment cores and impregnated by immersion in a warm liquid PEG solution (*circa* 60°C) care being taken to retain sample orientation. PEG is water soluble, and gradually replaces water in the sediment matrix so that subsequent drying does not cause disruption of the sediment fabric. Samples were sheared along the faces subsequently examined in order to ensure exposure of a "natural" fabric (slicing a sediment block causes orientation of particles on a small scale) using the method of Tovey (1971). V-shaped notches are scored around the sample face and fracturing achieved by gently bending and pulling the sample. These samples were mounted and coated for SEM examination, and examined on an ISA model 100-A scanning electron microscope.

Variation in sediment unit thicknesses provides an indication of the action of sediment focusing. In turn, this is related to the prevalence of deposition from under-, inter-, or overflows in a lake. Areal grain size variation similarly reflects depositional mechanisms as discussed in section 1.5. Deposition of particles from inter- or overflows distributes grains more evenly over the entire lake and results in uniform rates of deposition over the basin,

whilst underflows concentrate relatively coarser material into areas of low topography (Matthews, 1956; Smith, 1981). Stratigraphic variation is examined directly by changes in thickness between correlated features of two or more cores, and is further quantified through the calculation of sediment accumulation rates.

Sediment grain size distributions are traditionally determined by pipette analysis of sediment samples (as detailed in BS 1377). This method is not particularly sensitive to small variations in the finest, clay size, fractions which analyses of the central cores from each lake have shown form the bulk of the sediment (section 3.2 Figure 3.9). Variations in grain size identified using mineral magnetic parameters are located in predominately in this size range, providing a more rapid and very sensitive measure of size variation within the fine fraction. Furthermore, pipette analyses established that the gross grain size variation along the sediment column is not particularly marked. As this analysis is a time consuming method, particularly if applied in a multi-core study, further analyses were not undertaken. Comparison of grain size variations between the Buttermere and Crummock Water cores is therefore based on comparison of logs of certain mineral magnetic parameters, namely those primarily influenced by grain sizes of the magnetic assemblage. These are the

ARM-derived parameters ARM/ χ and IRM₊₂₀/ARM for the SD and coarser MD size ranges respectively, which are used as surrogate measures of changing grain size trends of the complete sediment.

Application of all these techniques to the Buttermere and Crummock Water sediments allows the investigation of the sedimentation processes (hemipelagic or turbiditic) operating at certain horizons, and of vertical and areal grain size distribution. These offer an assessment of the role of internal lake processes in influencing sediment distribution and the possible modification of catchment-derived sediment assemblages.

5.3. Results.

5.3.1. Sediment fabrics.

Figure 5.2 shows the locations of samples taken for thin section and for SEM examination. Representative photomicrographs of thin sections and SEM samples are shown on Figures 5.3 - 5.7.

i) thin section examination.

The main features apparent in thin section are:

i) The sediment groundmass is generally homogeneous (Figure 5.3 a and b), with a range of grain sizes from fine clay to maximum diameters of *circa* 0.4 mm, 1.5 \emptyset (that is coarse/medium sand) although grains of this size are rare (N.B., most of the light patches looking like large quartz grains are holes in the thin section);

ii) Green laminae are darker than the groundmass (Figures 5.4 and 5.5) which may be due to a higher proportion of organic or finer (silt/clay) material. LOI values for the green laminae are lower than those of the surrounding sediments however, indicating that their darker colour is due to a greater proportion of fines;

iii) Because of the very fine nature of all the sediment, standard optical mineralogical identification of grains is not possible;

iv) As observed in hand specimens (section 3.2), laminae in the Buttermere sediments are often less distinct, with less well-defined upper and lower boundaries than those from Crummock Water (Figures 5.4 and 5.5 respectively);

v) Basal contacts are always sharp. Upper contacts are usually sharp, but may rarely be gradational or "diffuse", particularly in the less well-developed Buttermere laminae (Figure 5.4);

vi) Although usually sharp, many laminae/groundmass boundaries are "wavy". Rare, poorly-developed flame structures indicate slight sediment disturbance, probably the result of compaction (Figures 5.4 a and c).



Figure 5.2. Sampling positions along Buttermere and Crummock Water long cores B2L and C2L for thin section (thick line) and SEM (thin line) specimens.



Л

1 mm



b) C2L 198 cm. Homogeneous groundmass. Sub-horizontal layer

across centre of picture is a leaf, edge-on. Scale bar = 1 mm.

Figure 5.3. Examples of the homogeneous and unsorted sediment groundmass from Buttermere and Crummock Water. Changing shade across the field of view are due to variation in the thickness of the section. The distinctive bright patches are small holes in the thin sections. All photomicrographs taken in plane polarised light (PPL).



a) B2L, 15 cm. Darker band in centre of picture is a poorly-developed green (chlorite) lamina. This section was made by PEG impregnation. Note the resultant lighter shade background. Scale bar = 1 mm, PPL.



b) B2L 82 cm. Darker band in centre of picture is a poorly-developed green (chlorite) lamina. Both upper and lower contacts are gradational. PPL



c) B2L 189 cm. Darker band in centre of photo is a green (chlorite) lamina. Note sharp upper and lower contacts. Yellow tint is due to a slightly thicker thin section. Scale bar = 1 mm, PPL.

1 mm

Figure 5.4. Examples of green (chlorite) mud laminae from the Buttermere sediments. Small undulations in all these laminae may reflect side-pressures due to the coring process in addition to compaction during burial. All pictures taken in plane polarised light (PPL).



 $\begin{bmatrix} a \\ a \end{bmatrix}$ C2L 198 cm. Darker band is a green (chlorite) lamina. Note sharp upper and lower contacts, also undulations of lamina and poorly-developed flame structures (lower left boundary), indicative of sediment compaction. Scale bar = 1 mm, PPL.

b) C2L 205 cm. Dark band in lower section of picture is a green (chlorite) lamina. Note sharp upper boundary, (there is a gap in the thin section over the lower boundary of the lamina). Scale bar = 1 mm, PPL.

1 mm



 \hat{a} c) C2L 25 cm. Two fine green laminae (centre of photomicrograph). Note their undulations and sharp boundaries. Scale bar = 1 mm, PPL.

1 mm

1 mm

ii) SEM examination.

The locations and frequency of core samples taken for SEM examination are shown on Figure 5.2. Photomicrographs of clay fabrics (Figure 5.6, and 5.7) are here marred by charging on parts of the specimens, causing loss of fine detail at the high magnifications used. However, the following distinct features may be seen:

i) "Open" horizontal clay flake fabrics. These characterise the gyttja, unit 4 (Figure 5.6);

ii) Closely-packed but still horizontal fabrics in the grey clay (Figure 5.7 a);

iii) In most samples, including i) and ii) above, areas with less well-defined horizontal fabrics were also observed, ranging from slight disruption of clearly horizontal fabrics (Figure 5.6 b), to what may be a primary random clay flake orientation (Figure 5.7 b).

5.3.2. Between-core grain size variation.

Comparison of grain size changes between cores is achieved through examination of the

 IRM_{+20}/ARM and ARM/χ (Figure 5.8 a and b) parameters which are predominantly dependent on magnetic grain size. It should be remembered that, as with all other Buttermere and Crummock Water long core diagrams, near surface depth values of long cores may be influenced by core loss.

IRM₊₂₀/ARM provides a measure of the proportions of the soft, ferrimagnetic, component, high values implying a greater proportion of the coarser (MD) component and low values a greater proportion of finer (SD) grains. ARM/ χ provides a guide to the proportion of SD size magnetic grains in an assemblage, high values implying a greater proportion of this component.



 $10 \,\mu\text{m}$ a) C2L 0.12 m, (unit 4, gyttja). Horizontal clay flake fabric. Clay flakes long axes are orientated approximately across the picture width, and face into the centre of the picture. Scale bar = $10 \,\mu\text{m}$.



10 μ m b) C2L 0.53 m, (unit 4, gyttja). Horizontal clay flake fabric, (long axes are orientated to the long and short axes of the photomicrograph). Note slight disturbance of fabric, (particularly on left hand side of picture), which is a secondary sediment fabric alteration, and the relatively "open" texture to the fabric, adjacent flakes showing mostly point contacts with one another. Scale bar = 10 μ m.



10 μm c) B2L 4.30 m, (unit 2, clay/gyttja transition). Horizontal clay
 flake fabric. Note the more compacted nature of the fabric, which may reflect greater compaction and dewatering at depth, but also the greater clay content of this unit. Scale bar = 10 μm.

Figure 5.6. Horizontal clay flake fabrics from units 4 and 2.



^{10 μ m a) C2L 2.66 m. Horizontal clay flake fabric. Note the compactness of the fabric, particles staked on top of one another. This reflects the platiness of the particles. Scale bar = 10 μ m.}



10 µm

b) C2L 2.66 m. Severely disturbed or possibly primary random
clay flake fabric. Fabric is still relatively compact in comparison to those from more organic-rich units. Scale bar = 10 μm.

Figure 5.7. Clay flake fabrics from unit 3, the grey clay.





b) ARM/X parameter (10^2 Am^{-1}) , most sensitive to magnetic grains in the SD size range, (approximately 0.02 - 0.04 μ m for isodiametric magnetite).

Figure 5.8. Size dependent magnetic parameters for all long cores, (see text for discussion and appendix 1 for explanation of parameters).

Logs for these two variables (Figure 5.8 a and b) record changes in proportions of particular grain sizes from the Buttermere lake head, east towards the Crummock Water outflow, all of which are progressive. In both lake sediment sequences there is a gradual proximal-distal <u>reduction</u> of the coarse (MD) component at equivalent stratigraphic horizons (Figure 5.8 a), most marked in the more widely-spaced Crummock Water cores, whilst there is a progressive <u>increase</u> in the proportion of finer (SD) magnetic grains.

5.3.3. Sediment focusing.

Lithological and whole core susceptibility (κ) logs are presented in chapter 3 (Figure 3.2) are reproduced here as Figure 5.9 with correlated features marked with letters. These provide comparisons of differential sedimentation within the two lake basins, the logs of κ allowing more detailed comparison between cores than from comparisons of gross lithology alone.

In the most recent sediments (from the short cores and the upper 1 m of the long cores)

comparison of κ peaks indicates that the highest sedimentation rates are found in the lake head areas. For example, in core B3L both of the peaks labelled as "a" and "b" in Figure 5.9 are expanded, recording greater detail compared to cores B2L and B1L (N.B., different short and long core sediment diameters account for the variation in absolute

magnitude of respective κ profiles). Similarly, in core C1L peak b covers a greater depth of sediment indicating an increased accumulation rate at this horizon than seen in either core C2L or C3L.

Differences in calculated sediment accumulation rates between cores are quantified and summarised in Figure 5.10 (over-leaf), which shows percentage changes of rates between successive pairs of down-lake long cores. Significantly higher sediment accumulation rates are found in cores from the Buttermere lake head area than in distal cores. For example, 88 mg.cm⁻².yr⁻¹ for B1S/B3L may be compared to 61 mg.cm⁻².yr⁻¹ for core B2L for the





Figure 5.9. Lithology and volume susceptibility, all cores. Correlated features illustrate changing unit thickness, hence variation of sediment accumulation rate with location. No short cores penetrate below unit 4 and are therefore composed entirely of gyttja. Inset indicates core sites. Core logs shown progressing westerly from the head of Buttermere.

period 150 B.P. to present (Figure 4.2). This represents an increase in the lake head core of approximately 40 %. This pattern is maintained throughout the Buttermere sequence, with the greatest enhancement (54.9 %) observed in the mid-Postglacial (the period 5270 -860 B.P.). In older sediments, rates between cores are approximately the same, although no data is available for the lake head area.

Approximate date of sediment horizon	B1S/B3L vs. B2L (Lake Head area)	B2L vs. B1L (Lake Head area)	C4S/C1L vs. C3S/C2L	C3S/C2L vs. C3L
 1987 A.D	+43.0 %	(not available)	+11.1 %	(not available)
150 B.P.				
150 B.P.				
860 B.P.	+46.6 %	+5.6 %	+41.1 %	-16.2 %
860 B.P.				
5270 B.P.	>+54.9 %	+5.6 %	-31.6 %	-19.3%
5270 B.P.	(not available)	0.0 %	-842.1 %	(not available)
7330 B.P.				
7330 B.P.	(not available)	+4.6 %	-3.8 %	(not available)
10 000 B.P.				

Figure 5.10. Percentage enhancement of sediment accumulation rates between adjacent long cores from Buttermere and Crummock Water (based on sediment accumulation data shown in Figure 4.2). Positive numbers indicate enhanced sedimentation at the site of the first named core; negative numbers refer to the second named core.

In Crummock Water, the pattern of sediment accumulation is more variable through time. In the more recent sediments (post-dating *circa* 860 B.P.) accumulation is greatest in the lake head area (core C1L). In the mid- to late-Postglacial (5270 - 860 B.P.), it is concentrated mid-lake, and also towards the western end of Crummock Water, where the tributary from Loweswater enters. The grey clay (unit 3) exhibits pronounced sediment focusing towards the lake centre, whilst below this, rates between cores are broadly similar, although no data is available for this period from the western end of the lake (core C3L).

5.4. Discussion.

In hand and thin section, the sediments examined are generally structureless and unsorted, with a range of grain sizes $\leq 1.5 \text{ } \emptyset$ (although the larger sizes are rare). This corresponds, in Stürm's (1979) model, to the "chaotic" sediment (Figure 5.1, Boxes 1 or 2), indicating hydrological parameters of continuous sediment influx with either a stratified or an unstratified water column.

Through examination of sediment structure at a very detailed level (of the order of microns, where individual clay flakes are identifiable), clay flake fabric analysis provides an additional indicator of sedimentation processes, at a level of detail not considered by Stürm in his (1979) sedimentation model. Under SEM examination, horizontal fabrics, indicative of hemipelagic sedimentation processes (O'Brien *et al*, 1980), are common, with what may be primary random flake fabrics only rarely identified.

Thus, both the very fine scale examination of clay flake orientation, and the somewhat coarser examination of sediment structures on the scale of millimetres in thin and hand section suggest that sedimentation is essentially continuous, with deposition from dilute sediment plumes.

The green laminae spread throughout the cores indicate distinct, episodic, changes in sedimentation processes, and imply the input of discreet pulses of finer sediment. This might be achieved through discontinuous catchment-based erosion events, and Pennington (1943), who described similar laminae present in the Windermere sediments, suggested that the chlorite-rich rocks from the Borrowdale Volcanic Group formed the source for this material. However, she was unable to propose an mechanism for the process concentrating chlorite into discreet layers on the lake bed. Given the well-sorted and very fine-grained nature of these deposits, intermittent catchment erosion events such as "catastrophic" slope failure, introducing large volumes of chlorite-rich sediment into the lakes seems unlikely. Such inputs, highly sediment laden, might be expected to contain a variety of different sized material, rather than the uniform deposit observed.

These laminae therefore represent pulsed deposition of fine clay, more probably initially trapped in epilimnic waters, and then deposited following breakdown of lake stratification.

This would lead to the deposition of a,

"distinct non-gradational clay layer on top of the earlier deposited coarser layer" (Stürm, 1979, page 283)

Morrison (1981) demonstrated that the <u>shape</u> of particles was of far greater importance in the production of lacustrine laminae than is generally acknowledged in the literature. He observed that the trapping of platy minerals, such a chlorite, may be achieved through the action of wind alone, causing what might be termed "turbulent resuspension" of fine material in the upper waters, with no requirement for well-developed aqueous thermal stratification.

However, Morrison's study was conducted on a small proglacial lake experiencing strong daily katabatic winds, which kept the upper water layers well, and regularly mixed. Buttermere and Crummock Water do not experience winds of such strength and regularity, and thus probably require some development of thermal aqueous stratification to provide a sediment trap. Morrison's observations highlight the susceptibility of particular sediment fractions to redistribution within the lake environment, and the predominantly chlorite composition of the laminae in the Buttermere and Crummock Water sediments suggests that grain shape, as well as size, is an important factor in the production of these laminae.

O'Sullivan (1983) cites the mechanism of winter freezing causing deposition of "an amorphous layer formed in the still conditions under ice".

This actually represents similar hydrodynamic conditions to the possible mechanism of lake overturn described above. Surface water cooling from above 4 °C to freezing passes the temperature of maximum density, so sinks beneath the epilimnion. Fine material trapped in the previously buoyant and more turbulent upper layers thereby becomes liable to deposition at the lake bed.

If so, this mechanism provides the potential of a further proxy dating method for the more recent of these sediments via a comparison with recorded weather conditions. If the green laminae are formed when the lakes are very occasionally ice-covered, it may be possible to correlate laminae produced under these conditions with weather records. Unfortunately, although historical meteorological records from Honister Pass (the extreme east of the

Chapter 5, lake depositional mechanisms

Buttermere catchment) have been consulted, records of prolonged winter freezing are not maintained. This hypothesis would therefore be better tested in more populated catchments, for example, Windermere, where local records of weather and lake conditions may be more readily available.

Hilton (1985), on the basis of climatic and morphological characteristics alone, suggested that the deep English Lakes, (which include Buttermere and Crummock Water) experience intermittent complete mixing. This promotes distribution sediment throughout the whole water column which may then settle out of suspension to the lake bed.

The physical examination of the sediments undertaken here supports this hypothesis. The green laminae are formed following intermittent complete mixing of the water column and the deposition of fine and platy particles previously trapped in the epilimnion. The breakdown of stratification and subsequent development of still water conditions promoting deposition of fine suspended material <u>may</u> be initiated by the infrequent formation of winter ice on the lakes, according to the mechanism cited by O'Sullivan (1983).

Intermittent complete mixing, which evidence so far presented implies is the primary mechanism in the production of distinct clay laminae, is imposed upon a regime of generally continuous hemipelagic sedimentation of a broader range of grain sizes. These also include a proportion of fine material, suggesting either deposition from interflows (below the potential trap of the epilimnion) or from overflows if lake thermal structure and/or turbulent resuspension processes are essentially inactive. This suggests that the development of marked aqueous stratification (causing trapping of sediment and its subsequent lake-wide distribution) is not a regular occurrence in Buttermere and Crummock Water.

Therefore, except for the sediment comprising the green laminae (see discussion in the following paragraph), sediment supplied to Buttermere and Crummock Water may not become trapped within discreet water layers. It is unclear whether this reflects an absence

of distinct stratification, or input of sediment plumes below the epilimnion as interflows. Underflows are unlikely however, given the characteristic hemipelagic fabrics observed under SEM.

The green laminae <u>may</u> represent short-lived, infrequent sediment inputs from rocks of the Borrowdale Volcanic Group (the principal source of chlorite). In particular, their good sorting makes this less unlikely though. More probably, the green laminae represent deposits of suspended material trapped and concentrated in the epilimnion, and deposited after the breakdown of periodic water stratification.

SEM examination of samples from the grey clay (unit 3) demonstrated the presence of a strongly orientated, horizontal clay flake fabric, indicative of hemipelagic sedimentation, where flakes attain a preferred orientation whilst settling from suspension (O'Brien *et al*, 1980). Within the same unit however, are less strongly, or possibly randomly orientated clay flake fabrics (Figure 5.7 b), indicating rapid deposition from more concentrated sediment plumes, that is, turbiditic sedimentation (*op cit*). Thus sedimentation styles within this unit are more variable than in the overlying gyttja, unit 4.

The sediment accumulation data (Figure 5.10) indicate that throughout the period of deposition of unit 4, sedimentation in Buttermere was concentrated in the deep lake head area, declining sharply mid-lake, with a further small decline towards the distal end. From Figure 5.8 a and b progressive proximal-distal changes in magnetic grain size characteristics are also apparent. In both Buttermere and Crummock Water, generally higher IRM₊₂₀/ARM values in cores from the lake head contain the highest proportion of coarser, MD, ferrimagnetic grains. These proportions decrease east, towards the outflow.

The opposite pattern is shown in Figure 5.8 b, where an <u>increase</u> in ARM/ χ implies an increasing proportion of finer, SD, magnetic grains.

Such a sedimentation pattern is consistent with deltaic deposition concentrated in the lake head area where the largets grains, including the coarsest magnetic grain sizes, occurs. This is called river plume sedimentation (Hilton *et al*, 1986b; Pharo and Carmack, 1979). The relative similarity of sediment thicknesses and hence of accumulation rates in the cores from the rest of the lake suggest more uniform deposition in this area. This would result from more widely-mixed and evenly distributed sediment, possibly derived from inter- or over-flows (Stürm and Matter, 1978).

The pattern in Crummock Water is more variable. From the base of the sequence upwards, the main features of sedimentation in Crummock Water are:

i) To the base of unit 3, sedimentation rates are nearly equal between the mid-lake and lake head areas;

ii) Strong sediment focusing operated during the deposition of unit 3;

iii) In the mid- to late-Postglacial period (5270 - 860 B.P.) greatest sediment accumulation occurred at the distal end of the lake, followed by the central area, with lowest rates at the lake head (see Figure 5.10). Total sediment accumulation rates were generally low during this period;

iv) After *circa* 860 B.P., minimum sediment accumulation occurs in the lake centre, with the greatest accumulation rates in the lake head area (860 - 150 B.P.);

v) In the most recent period (150 B.P. to the present) sediment accumulation is marginally greater at the lake head, although there is no data available to confirm the contemporary rate at the distal end.

This pattern of sediment accumulation in Crummock Water suggests that two major sediment tributaries have been of importance, one at each end of the lake. During the period of deposition of the gyttja (unit 4) most sedimentation has occurred near these source inputs by river plume sedimentation. In the mid-Postglacial, Park Beck (the outflow from Loweswater), was of relatively greater significance although, notably, total
sedimentation rates are very low during this period (averaging 14.8 mg.cm⁻¹.yr⁻¹). The influence of the western sediment source is thus only distinguishable when lake-wide sediment accumulation rates are relatively very low.

Changes in sedimentation rate in more recent periods demonstrate the increased importance of the source at the eastern (Buttermere) end of Crummock Water. As this becomes dominant, absolute lake-wide sedimentation rates initially double, and then triple in the most recent period (150 B.P. to present). Grain size differences, as illustrated in Figure 5.8, remain fairly consistent however, with a well-defined progressive decrease from the lake head to the distal end in the proportion of coarser, MD, ferrimagnetic grains (decreasing IRM_{+20}/ARM values, Figure 5.8 a), and an increase in the proportion of finer, SD, magnetic grains (Figure 5.8 b).

Unit 3, the grey clay, exhibits evidence in Crummock Water of strong sediment focusing towards the lake centre. This unit reaches a maximum thickness of 0.46 m in core C2L, but elsewhere is less than 0.05 m thick. In Crummock Water, the unit exhibits a pronounced coarsening of grain sizes in the mid-lake core C2L, with the proportion of coarser, MD, ferrimagnetic grains increasing, and that of finer, SD, grains decreasing (Figure 5.8).

Grain size analyses of bulk samples (Figure 3.9) record a relative increase and decrease in the proportions of silt and clay respectively, corroborating the evidence of coarsening as suggested by mineral magnetic analyses. Such concentration of relatively coarse sediment towards the deep areas is typical of sediment deposition from density underflows (as discussed in section 1.5). These are restricted to the deeper areas of a basin, and may originate from processes such as delta/lateral slope failure, or from input of, for example, sediment-rich and therefore relatively dense, fluvial inflows. In Buttermere, the grey clay exchibits a uniform thickness of 0.02 m in the central and distal cores (B2L and B3L respectively) and is not penetrated in the core from the deepest area of the lake (B3L). It is therefore not possible to test whether strong sediment focusing was operational in Buttermere during the deposition of this unit.

No lower boundary surface for the micro-laminated clay, unit 1, was recovered. Therefore it is difficult to assess the complete sedimentation characteristics of the unit. However, similar peaks in κ in cores B2L and B1L (labelled "d", Figure 5.9) suggest little or no sediment focusing between these two sites. As this unit is not penetrated in the deepest area

of the lake, sediment focusing there, with a more even distribution of sediments over the remaining lake basin, as in unit 4, cannot be ruled out.

In Crummock Water, correlation of the κ profiles of cores C1L and C2L is tentative, but that of C1L may be expanded relative to C2L such that the two maximum values are correlated (peak "d", Figure 5.9), indicating an increased sediment accumulation rate in the lake head area. Concentration of sedimentation there is consistent with the rapid deposition of material supplied from a point source in the lake head area during typical deltaic sedimentation (Fulton and Pullen, 1969), finer material being distributed more evenly over the lake basin, possibly as inter- or overflows.

5.5. Summary.

Evidence from sediment fabric, sediment unit thickness, and grain size changes have been used to interpret depositional mechanisms operational in Buttermere and Crummock Water. Sedimentation is mostly hemipelagic, and presently concentrated in the lake head areas, with deposition of the coarser grain sizes also in these areas - a "river plume sedimentation" regime. Crummock Water posses sediment sources at both ends of the lake, supplied in each case from upstream lakes, although that from Loweswater (the western input of Crummock Water) is only distinguishable during periods of generally low sedimentation rates (the mid-Postglacial).

Strong sediment focusing was operational during the time of deposition of unit 3 (the grey clay) although SEM examination of fabrics indicates that deposition included hemipelagic settling as well as turbidity flows. The cause of this distinct change in sedimentation style has not been determined, although the similarity of these sediments to underlying minerogenic glacial material in terms of their mineral magnetic characteristics (chapter 4)

supports the suggestion (section 2.2) that this unit may record a period of lowered lake water levels (as seen in several small tarns in the Lake District at this time, Pennington, 1970). Slumping of lateral slopes owing to water draw down effect, with the transfer of somewhat coarser sediment to the central lake areas, is a typical phenomenon seen in sediments following a reduction in lake water level (Gilbert and Desloges, 1987).

Therefore, internal lacustrine processes of Buttermere and Crummock Water do not strongly modify the sediment already delivered. Coarser material is dumped in deltas, and deposited by hemipelagic settling through the water column at open water locations typical river-plume sedimentation.

The presence of the green laminae demonstrates intermittent action of internal lake processes, where a particular sediment fraction (finer, platy clay particles) has been concentrated within the water body and then deposited probably after the breakdown of stratification. This process may be enhanced, or may be dependent on the development of winter freezing, promoting prolonged still lake conditions, and thereby the deposition of much larger amounts of fine material than under unfrozen conditions. This mechanism is therefore superimposed on the "normal" hemipelagic/deltaic sedimentation regime.

In the more closely documented of the Lake District lakes, such as Windermere, it may be possible to correlate such laminae with the occasional freezing of the lake. This would test whether this process is important in their formation and enable construction of a proxy chronology.

Chapter 6. DISCUSSION AND CONCLUSIONS.

6.1. Mineral magnetism and the study of lake sediments.

Previous studies have demonstrated the value of mineral magnetic measurements, particularly

 κ , whole core or volume susceptibility, as a rapid means of core correlation, both within (Appleby *et al*, 1985, Dearing *et al*, 1981, Dearing, 1983), and, in certain circumstances, between lakes (Oldfield *et al*, 1978a). In this study, mineral magnetic measurements have allowed both cross-correlation of dated horizons, and more detailed examination of the effects of differential sedimentation within and between Buttermere and Crummock Water than is possible using lithology alone.

Mineral magnetic analyses are both relatively rapid and non-destructive, and hence, for sedimentological studies, may be preferable to correlations based on more time consuming identification and analysis of biogenic sediment components. The ability to measure variation

of κ from unextruded cores is, in this respect, unrivalled.

Where little variation in the characteristics of sediments occurs, the value of magnetic measurements as a correlation tool is reduced. This is the case in much of the Holocene sediments of Buttermere and Crummock Water <u>and</u> in the Windermere Holocene sediment sequence (Stober and Thompson, 1979). In such circumstances few alternative methods capable of allowing rapid <u>and</u> detailed core correlation exist.

In the Buttermere and Crummock Water catchments, determinations of whole core NRM measurements of declination might provide a better method, allowing correlation between cores and <u>between</u> lakes, in addition to the construction of a proxy chronology. Measurement of whole core declination variations can be as rapid as those of whole core susceptibility, and may also, as in this study, be performed on fresh unextruded cores (Section 3.3.2). As seen in the Buttermere sediments, retention of accurate declination records is by no means assured as these are dependent on specific sediment characteristics. These characteristics have

not as yet been clearly defined. Fine-grained relatively homogeneous sediments are a prerequisite though (Thompson, 1984).

In addition, the equipment used for whole core declination measurements is neither portable nor capable of field operation, requires a dedicated personal computer, and is more expensive. Thus, while determination of NRM declination provides a potential means of core

correlation comparable to measurements of whole core susceptibility (κ) in terms of speed, and is more valuable in terms of its potential as a dating tool and for core correlation <u>between</u> lakes, it is unlikely to be as widely suitable, nor so convenient to measure logistically.

Mineral magnetic measurements have also been successfully used in this study as a means of identifying changing grain size distributions in bulk sediments. Changing values of specific susceptibility (χ or χ lf) have previously been observed in lake sediments and were seen to have been produced by changes in sediment source.

Dearing *et al* (1981) for example, observed changing values of χ in cores of recent sediments from Llyn Peris (north Wales), where higher values were found to correspond to periods of increased channel scour, which supplied coarser-grained ferrimagnetic material(of higher specific susceptibility. Similarly, Björck *et al* (1982) observed that in addition to a dependency on the type of bedrock from which they were derived, χ was dependent on the particle size distribution of sediment samples. This observation, if repeated elsewhere, could

also allow the use of χ as a guide to changing bulk sediment grain size distributions.

In this study mineral magnetism is used in a rather different way to indicate bulk sediment grain size trends. This represents a rather different use of the techniques than performed in the studies discussed above.

Pipette analyses of bulk sediment samples indicate that, in general, variations in grain size are not marked in the Buttermere or the Crummock Water sediments, which contain abundant

silt and clay (Figure 3.9, page 75). The use of ratios (such as ARM/ χ and IRM₊₂₀/ARM) reduces the influence of changing ferromagnetic concentrations, and consequently mainly reflects changes in the proportion of particular magnetic grain sizes. They have therefore been used to identify changes in the major components of the magnetic grain assemblages, and, by inference, of the complete sediment assemblage.

This approach has a number of advantages over conventional techniques of analysis of sediment grain size. These are that it:

i) allows identification of very subtle changes in grain size;

ii) is particularly suited for the analysis of fine-grained sediments - such as those from Buttermere and Crummock Water;

iii) is much faster than most conventional techniques of grain size analysis;

iv) is non-destructive and may be performed on sediment samples of small volume.

This technique, suitably cross-correlated with measurements of actual bulk sediment grain size distributions, has much potential for more rapid mapping of variation of approximate grain size distribution over entire lake beds.

The recent sediments of many North American and European lakes, including Buttermere and Crummock Water, have been shown to contain increasing numbers of magnetic spherules. These are derived mainly from industrial discharges, mostly from increases in atmospheric discharges from coal-fired power generating stations. The sediments exhibit marked increases in spherule concentrations at *circa* 1900 - 1930 A.D. (Goldberg *et al*, 1981;

Oldfield and Richardson, 1990; McLean, 1991).

This is easily identified in sediments by determination of a concentration dependent parameter

such as κ , or, on a mass specific basis, χ , or SIRM. Thus magnetic measurements may be used for the rapid identification of a widespread, approximately-dated, cryptic marker horizon in lake sediments.

Lakes may receive inputs of ferrimagnetic minerals from a variety of sources, for example, from primary, bedrock, and secondary, soil, <u>catchment</u> sources, as well as magnetic spherules from the atmosphere. Identification of the horizon at which the major increase in <u>spherule</u> concentration occurs should therefore be corroborated, as here, by microscopic identification of magnetic spherules.

Atmospheric magnetic spherules are typically enriched in the coarser, MD grain sizes (this study, and Hunt *et al*, 1984). Measurements from this study suggest that use of the ratio IRM_{+20}/ARM , which primarily reflects the concentration of such coarse, MD ferrimagnetic material in a sample (see appendix 1), may, in conjunction with a concentration-dependent magnetic parameter, often be sufficient to identify the recent increase in spherule concentration, without the need for visual confirmation. This was not possible Buttermere and Crummock Water sediments however, because one of the catchment sources (the Ennerdale Granophyre, see discussion in section 4.4.3) supplies minerals with similar magnetic characteristics to those of the spherules.

Mineral magnetic measurements are readily analysed using numerical techniques, as it is a data-based analytical technique. Here, multivariate analysis facilitate the direct comparison of sediment magnetic groups from Buttermere and Crummock Water (as shown in Figure 4.14, page 122). This demonstrated that the similarity of sediment characteristics in both lakes reflects the similar evolution of their catchments. Relatively consistent variations in grain size between equivalent sediment groups from each lake, are interpreted as being due to the action of Buttermere as an upstream sediment trap for Crummock Water. This demonstrates the value of mineral magnetic analyses both as a sedimentological tool and in the interpretation

of environmental change.

Results of mineral magnetic analyses presented in chapter 4 (Figures 4.11 and 4.12, pages 117 and 118 respectively) also demonstrate that, based on their respective mineral magnetic characteristics, clear associations between lake sediments and catchment sources may be made. Data analysis, again using multivariate statistical techniques in order to produce ordination plots of lake and catchment samples, facilitate the representation and interpretation of data from the magnetic analyses.

These findings illustrate the utility of the mineral magnetic approach to the identification of source - sediment linkages in this catchment, despite its greater complexity (in terms of differing lithologies, widespread cover of glacial drift over much of the catchment, and poor differentiation of soil horizons) compared to catchments previously investigated using a similar methodology (Oldfield *et al*, 1985a; Smith 1986; Stott 1986, 1987; Yu 1989). This work suggests that this technique would be appropriate for investigations of sediment - source linkages in many more lake catchments in the U.K.

Several methodological problems in the mineral magnetic approach adopted here are identified however.

i) Measurement of the mineral magnetic characteristics of more specific particle size ranges would reduce the effect of between sample variations in grain size on magnetic characteristics. However, separation of specific particle size ranges in the sediment fraction

below 4 ϕ (63 μ m) is time consuming, and, for studies involving large numbers of samples, may become impractical.

ii) The present identification of source-sediment linkages is qualitative. This approach has

allowed identification of the major sources, and the likely contribution of additional sediment supplies.

Quantitative estimates of the relative proportions contributed by catchment sources, would represent a further refinement of the method. It would allow more precise identification of the relative contribution of <u>subsidiary</u> sediment sources, as well as the quantification of separate erosion rates for each catchment source, thus offering significant refinement of environmental interpretations.

Yu (1989) and Yu and Oldfield (1989) have discussed a method of obtaining quantitative estimates of sediment sources using magnetic measurements. This is based on laboratory mixing of source samples, which are then subjected to analysis by mineral magentic techniques.

Multiple regression using samples' magnetic characteristics allow calculation of multiple regression equations, defining the proportion of source materials in a sediment of given mineral magnetic characteristics. Linear programming is used so that a series of different magnetic characteristics can be considered together, and to introduce constraints to the calculated source proportions.

This approach has the advantage of incorporating additional constraining conditions. For example, these include not accepting source ratios considered unlikely to occur naturally, and introducing wider error margins forced by within-lake modification of the size assemblage of the sediment (as has happened in Buttermere and Crummock Water).

Further research using these techniques in the Buttermere and Crummock Water catchments might therefore allow the assignment of figures for the relative contribution of sources of the lake sediments. This would therefore further refine the interpretation of changing sediment source, and help in the construction of sediment budgets for both lakes.

Exact determinations and identification of magnetic mineral components, and hence their

proportions in a natural sediment sample, is not possible using the mineral magnetic approach. This is due to the very wide range of grain sizes and shapes, and chemical composition of iron oxides found (Bradshaw and Thompson, 1985). Thus, although the mineral magnetic approach is unlikely to offer a means of precise and absolute quantitative expressions of magnetic mineral proportions in natural samples, it does provide a rapid method of identifying the broad magnetic characteristics of materials, which are frequently specific to particular sedimentary environments. Such relative speed, ease and non-destructiveness facilitate comparison and matching, which is of great value in environmental investigations, where analysis of numerous samples is frequently desirable.

6.2. Catchment evolution in the Holocene.

6.2.1. The Buttermere and Crummock Water catchments.

No detailed studies of environmental change in the Buttermere and Crummock Water catchments have been published, although Mackereth (1965, 1966a) presented chemical analyses for a 5.5 m sediment core from Buttermere. Similarities in variations of the concentration of sodium, potassium, nitrogen and carbon content of the sediments from Buttermere, Ennerdale, Windermere, and Esthwaite suggested broadly similar environmental histories for all these catchments.

The present study is therefore the first to investigate the Holocene record of environmental change in the Buttermere and Crummock Water catchments in detail, based upon a <u>range</u> of geochemical and other sedimentological data. Correlations between changing lake sediment accumulation rates, quartz:feldspar ratios, and changing potassium concentrations are shown on Figure 6.1 a, with source indicators summarised on Figure 6.1 b. All lines of evidence, each based on particular sediment characteristics, indicate similar, synchronous, changes within the Buttermere and Crummock Water catchment, and provide evidence for three distinct periods of environmental change. These are:

1) A phase of active erosion in the Late-glacial and land in the recently de-glaciated landscape (pre-10 000 - *circa* 8 300 B.P.); the pre-10 000 B.P. Sediments dating from before 10 000 B.P.are derived from relatively fresh, mechanically eroded bedrock. Catchment net sediment supply rates exceed 250 kg.ha⁻¹.yr⁻¹.

2) This was followed by a period of less active erosion from *circa* 8 300 - 1 800 B.P. Sediments here are derived from mainly weathered sources, as indicated by the low quartz:plagioclase ratio, and low K and Na values for much of this period. Variation in concentrations of K and Na at times during this period indicate pulses of more intense physical erosion. Catchment net sediment supply rates remain consistently low (40 - 60 kg.ha⁻¹.yr⁻¹).



Figure 6.1. Summary diagram showing correlations between A), sediment accumulation rates, erosion intensity, and B), sediment source indicators for Buttermere and Crummock Water. Dating based on NRM and spherule proxy chronologies, correlations between cores based on prominent susceptibility features, lithological changes, and/or correlations between chemical mobile element peaks. Note that, apart from composite core C2L/C3S, remaining long cores are missing material at the sediment/water interface. Lithological sysmbols are the same as used throughout, (keys on figures 3.2 and 3.3).

3) An interval of renewed erosion from *circa* 1 800 B.P. to the present. The high quartz:plagioclase ratio suggests transport of chemically weathered material, and the low chlorite:illite ratio indicate input of sediment derived from secondary sources. The high K and Na values also suggest the input of less weathered material, derived from more intense physical erosion. Sediments here are therefore derived both from chemically weathered material and from less weathered sources. Catchment net sediment supply rates are high, ranging from 80 to 200^{+} kg.ha⁻¹.yr⁻¹ in the most recent sediments.

The change from active to less active catchment erosion in the first two periods reflects changes in erosional regime responding initially to "natural" - non-anthropogenically initiated - change. Climatic amelioration resulted in: a) de-glaciation, and hence the removal of a major agent of physical erosion; and b) the development of environmental conditions suitable for the growth of pioneer vegetation and ultimately of forests, thus allowing the eventual development of mature, more stable soils.

The persistence of high rates of catchment erosion until *circa* 8 300 B.P. (within unit 2, the clay/gyttja, cf Figure 6.1 a), illustrates that the stabilisation of soils was a rather gradual process. A small peak in K and Na values at *circa* 9 000 B.P. (K peak b, Figure 6.1 a) indicates increased mass transport of sediment, hence variations in landscape stability during this period due to natural catchment perturbations.

During the second period (*circa* 8 300 - 1 800 B.P.), chemical analyses (in the form of peaks in the concentration of K and Na) indicate three phases of increased mass transport of relatively unleached material both to Buttermere and Crummock Water. These suggest a decrease in landscape stability at these times. The Crummock Water profiles which record this are rather smoother than those from Buttermere, demonstrating more marked response of Buttermere to these changes, probably because of the concentration of erosion within its catchment. Averaged sediment accumulation rates exhibit generally higher values for the

Post-glacial in Buttermere compared to Crummock Water, supporting the idea of the existence of more intense catchment erosion in the Buttermere catchment.

The earliest of these minor episodes is dated by the NRM declination method at *circa* 6 900 B.P., and coincides with the change to a minerogenic sediment (that is, to unit 3, the grey clay). Interpolation between horizons dated by NRM suggests that this layer was deposited between *circa* 7 330 and 5 200 B.P. Several lines of evidence suggest that this unit records not so much a change in catchment environmental conditions, but rather in <u>internal</u> lake characteristics. The of which it is comprised sonsists mainly of reworked minerogenic sediments of glacial origin from <u>within</u> the lakes, rather than from outside. Lines of evidence used to substantiate this hypothesis are:

i) the clustering, based on magnetic characteristics, of unit 3 with stratigraphically lower minerogenic glacial sediments (magnetic subgroups I and II);

ii) the uniform sediment accumulation rates in surrounding sediments;

iii) the strong sediment focusing operating at this time, which suggests the existence of significant changes in lake hydrodynamics;

iv) subdued increases in mobile element concentrations;

v) constant χ fd% values.

In terms of magnetic properties, unit 3 is more like the underlying glaciogenic sediments than any other sediment source. Additionally, clustering of the lower sediments of unit 4 and the upper sediments of unit 2 (cf. Figures 3.6 and 3.7) suggest that generally constant catchment environmental conditions were "interrupted" during the deposition of unit 3 (section 3.4.1, page 85).

Sedimentation rates are uniformly low both above and below this unit (Figure 6.1 a). Calculated accumulation rates for unit 3 itself can only be speculative, owing to the complications caused by sediment focusing. Focusing towards the lake centre would be expected to cause an apparent <u>increase</u> in the calculated rates in the central core (C2L). In fact, detailed sediment accumulation rates calculated for core C2L for this period (Figure 6.1 a) are the lowest for the whole Crummock Water sequence, implying even lower general sediment accumulation rates calculated on a lake-wide basis. Increased erosion of catchment sources is likely to cause faster lake sediment accumulation. In fact, in Crummock Water unit 3, the opposite is seen.

Sedimentological evidence of strong sediment focusing towards the deeper areas of the lake, suggests the incidence of a rather dramatic change in lake hydrodynamic conditions. The generation of underflows by input of sediment-laden stream water (should this unit be derived from <u>catchment</u> sediment sources) <u>could</u> be invoked as an explanation for this sediment focusing however - as opposed to any change in <u>internal</u> lake characteristics.

If topsoil-derived material were a significant source for the sediments of unit 3, values of

 χ fd% would be enhanced. Mineral magnetic analyses show that they are not. Unit 3 is probably not associated with increased sediment supply from catchment streams either. Contemporary samples of stream sediment also cluster with catchment soils. Similarly, although values of K and Na do increase within unit 3, these are small in comparison to those arising from periods of known soil instability and soil input (compare peaks "c", unit 3, to "f" and "g", unit 4, Figure 6.1 a). This suggests that the unit was not derived from the sudden influx of either soil or stream sources.

Thus, various lines of evidence indicate that the sediment forming unit 3 is not derived from topsoils. In terms of their mineral magnetic characteristics, samples from this unit do not exhibit similarities to contemporary stream sediments either. Increased downcutting by streams probably therefore does not represent a source for these sediments. The most likely source of minerogenic sediment at such low accumulation rates, is glacial minerogenic material <u>reworked</u> from within the lake basins themselves.

Lowering of lake water levels promotes destabilisation and reworking of marginal sediments (see discussion, section 1.5, also Pharo and Carmack, 1979, Shaw *et al*, 1978). This could be caused by an effective decrease in catchment precipitation. This would promote reduced transfer of sediment from the catchment, and hence lower sediment accumulation rates.

The remaining K and Na peaks of this period (d and e, Figure 6.1 a) are not associated with such dramatic lithological changes and <u>may</u> represent periodic anthropogenic disturbance of the catchment. From examination of the diatom record, Haworth and Long (1984) inferred small fluctuations in water quality in the sediments of Ullswater. These temporary changes in the dominant diatom taxa occurred after forest clearances and were interpreted as following anthropogenic modification of the catchment (discussed in section 2.2). "Slash and burn" is the usual way Neolithic peoples modified natural forest vegetation. Small areas of forest were cleared for cereal and other crops, and domesticated animals browsed around these areas (Turner, 1970). Such periodic occupation is consistent with the observed "pulsed" nature of the K and Na profiles from Buttermere and Crummock Water. However, "natural" perturbations, such as at *circa* 9 000 B.P. (K peak "b", Figure 6.1 a), which pre-date evidence for anthropogenic activity in the area, cannot be ruled out.

The onset of increased net sediment supply rates has here been taken as the beginning of the third period of catchment history. This begins at *circa* 1 800 B.P. Then, until *circa* 450 B.P., net sediment supply rates are modest compared to subsequent increases. Hwoever, values still reach nearly double those of the minimum rates observed in the mid-Post-glacial (78 and 40 kg.ha⁻¹.yr⁻¹ respectively, Figures 4.3 and 4.4).

The whole of this period is marked by consistently increasing catchment net sediment supply and lake sediment accumulation rates. K and Na values rise to their Post-glacial maxima at *circa* 450 B.P. (Figure 6.1 a) and then do not fall again to the low "background" values seen in the mid-Post-glacial. Such protracted disturbance of catchment soils is

consistent with the presence of more permanent settlement and farming within the catchment, as identified in other parts of the Lake District by Pennington (1981).

The lower of the K and Na peaks within this period (f, Figure 6.1 a) is dated to *circa* 860 B.P. (1100 A.D.). This coincides with the widespread Viking immigration of the Lake District dated by Pennington (1981) at around 1 000 A.D. This phase of migration prodiced extensive local forest clearance, recorded in the Lake District sediments studied by Pennington by increases in accumulation of all variables except carbon. Evidence for disruption of catchment soils during this period in the form of ¹⁴C anomalies, has been found in sediments from Loweswater .

It is highly likely that the increases in sediment accumulation, catchment net supply rates, and other variables in the Buttermere and Crummock Water sediments at this time also reflect this wave of Norse settlement and consequent catchment modification. In fact, mineral magnetic data positively identify <u>topsoils</u> as a main source for these sediments.

This is illustrated in Figure 6.1 b by the marked increase of $\chi fd\%$ values (exceeding 6 % at *circa* 1.5 m depth), which "can ... be used specifically to infer soil erosional inputs" (Maher and Taylor, 1988).

Whilst data from Buttermere and Crummock Water indicates consistently increasing rates of accumulation and supply, the slightly finer resolution data from the chemical analyses suggests that after these initial clearances there was a decline in input of more freshly eroded material, and thus a relative stabilisation of the landsurface. This is followed by further increases in K and Na values, in the quartz:feldspar ratio, and a decrease in the chlorite:illite ratio, with peak values dated at *circa* 450 B.P (1500 A.D., Figure 6.1 a). Evidence of increases in lake accumulation rates, and deposition of increasingly minerogenic sediments at around this time has been found by Pennington (1981) in Blelham Tarn and Loweswater. These changes were ascribed by that auther to the effects particularly of deeper ploughing during the cultivation of cereals and crops which included hemp and flax. Such deeper ploughing affected more minerogenic sub-soils as well as

topsoil horizons (op cit).

Once again therefore, it is highly likely that the more recent increases in sediment accumulation rates, mobile element concentrations, and the quartz:feldspar ratio in the Buttermere and Crummock Water sediments, reflect human activities. Sediment layers recording medieval events are found in nearby Loweswater, and there is documentary evidence for medieval building activity and land disturbance both at the foot of Crummock Water, and in the Honister quarries above the head of Buttermere (Collingwood, 1928, pp. 77 and 105, Rollinson, 1967, p.104, see also discussion in Chapter 2, section 2.2). The input both of weathered soil mineral material and of less weathered substrate from the increasingly deforested catchment is recorded by the simultaneous increase in the quartz:feldspar ratio and decrease in the chlorite:illite ratio respectively (Figure 6.1 a and b). This again is consistent with the mobilisation of both top- and sub-soils by this deeper ploughing.

Consistently high lake accumulation and catchment net sediment supply rates following this period point to the continued human occupation of this catchment and of erosion up to the present. Decreases of, for example, sediment LOI (Figure 3.3), indicate the continued erosion of less weathered sources in the catchment and, as suggested by Pennington (1981), that following the extensive Viking forest clearances of 900 - 1000 A.D., there is,

"...a permanent change in the ecosystem..., formation of organic soils on the catchment no longer kept pace with the rate at which they were transferred to the lakes."

(Pennington, 1981, p.214)

6.2.2. Regional (Lake District) Holocene catchment evolution.

The preceding discussion demonstrates that the pattern of environmental change identified in the Buttermere and Crummock Water catchments is broadly similar to trends observed from other Lake District studies. These changes are summarised on Figure 2.6. This pattern, as indicated by results from Buttermere and Crummock Water, is outlined as follows.

Following deglaciation, a long period of relative catchment stability which lasted for much of the Holocene period was characterised by relatively low and consistent values for lake sediment accumulation rates and other variables. This ended at *circa* 1 800 B.P. Relatively modest increases in lake sediment accumulation rates (from 10 to about 20 mg.cm⁻².yr⁻¹, Figure 6.1 a) are seen at this initial level. Changes, mainly in pollen assemblages of cultivated plant seen in sediments from other lakes (Pennington, 1970, 1981), suggest anthropogenic modification of the catchments during this period, and therefore that the changes in lake sediment characteristics at sites across the Lake District, including at Buttermere and Crummock Water, posses an anthropogenic cause.

Increased sediment accumulation rates at around 1 000 B.P (to 24 mg.cm⁻².yr⁻¹, Figure 6.1a) coincide with a period of widespread Norse settlement in the Lake District (Pennington, 1981). This evidence points to more intense human activity in the Buttermere and Crummock Water catchments, and probably to selective deforestation. Chemical evidence indicates this is followed by a period of relative stability (from *circa* 700 B.P., Figure 6.1 a), during which lake sediment accumulation rates remained approximately constant (*circa* 23 mg.cm⁻².yr⁻¹).

Mineral magnetic analyses of the Buttermere and Crummock Water sediments provides positive evidence, for the first time in Lake District studies, of the <u>direct</u> input of topsoil-derived material from *circa* 1 000 B.P. onwards (indicated by high χ fd% values in the upper 1.5 m, Figure 6.1 b). Previously, sources of this material were <u>assumed</u> to be catchment soils, assumptions being based on chemical (mobile element) data (Mackereth, 1965, 1966a), and on the correlation of synchronous changes of pollen assemblages and sediment characteristics,

"The invariable correlation found between changes in pollen and changes in sediment composition can only be explained by the relationship of both variables to catchment soils."

(Pennington, 1981, p.199)

Further increases in lake sediment accumulation rates indicate continued human impact during the medieval period, with ploughing probably being responsible for the introduction of both top- and sub-soil derived material to the lakes. Continued settlement and farming in the catchment are most likely to be responsible for the further increases in net sediment supply and in accumulation rates. Lake sediment accumulation rates in Crummock Water, for example, presently at nearly 60 mg.cm⁻².yr⁻¹, Figure 6.1 a).

The interpretation placed on the Buttermere and Crummock Water grey clay (unit 3) presented in the preceding section differs from that produced by observations of similar layers noted in other Lake District sites. Available evidence from this study demonstrates that this unit in Buttermere and Crummock Water is most likely derived from reworking of material from within the lakes, probably following lowering of the lake water levels.

In the usually rather homogeneous Post-glacial sediments of Lake District lakes and tarns, two, apparently distinct, minerogenic horizons have been observed which predate more recent anthropogenically-induced changes. Minerogenic layers identified in several Lakeland tarns (Burnmoor Tarn, Blea Tarn, Low Tarn, and Brant Rake Moss, all in the upland southern Lake District), dated to 7560 ± 160 B.P., were, on the evidence of changing iodine concentrations (Mackereth, 1966a, 1966b), interpreted as reflecting lowered water levels following a period of decreased precipitation (Pennington, 1970). These deposits therefore probably represent sediment reworked from shallower, marginal areas of the lake during lowered lake water levels. Thus, lower precipitation levels around 7 560 B.P. may have caused a reduction in water levels in these small Lake District tarns, with consequent changes in sedimentation regime.

At other Lake District sites (including Barfield Tarn, lowland south-west Lake District, and Blelham Tarn, central Lake District, and within the Windermere catchment), Pennington (1981) has reported the interuption of an organic sediment sequence by a lithologically similar layer,

> "... a lithological change from mud to clay-mud which represents the extreme manifestation of the increased input of mineral soils".

> > (Pennington 1981, p.211)

In these sites, this change coincides with pollen analytical evidence of the Elm Decline and the appearance of pollen from cultivated cereals. It is radiocarbon dated at *circa* 5 000 B.P. $(3370 \pm 120 \text{ B.C.}, \text{ and } 3285 \pm 55 \text{ B.C.}, \text{ Blea Tarn}, \text{Pennington}, 1973)$, and is attributed to increased erosion of "mineral soils" following an early, Pre-Neolithic, phase of Lake District deforestation.

Pennington (1981) postulated that, following deforestation, an increase in run-off led to "downcutting of streams into the mineral soils of catchments" (page 209) to form the more minerogenic layers observed. Further increases in soil erosion were provided by ploughing of slopes during prehistoric and early historic agriculture. It is interesting that this layer in Blelham Tarn, dated at *circa* 5 000 B.P. and ascribed mainly to effects of deforestation and prehistoric agriculture, <u>also</u> exhibits evidence of sediment focusing. Thus,

"the rate of sediment accumulation increased more at sites near the deepest part of the tarn than at more peripheral sites."

(Pennington, 1981, p.211)

The dates for these minerogenic layers are shown in Figure 6.2 (overleaf). The ¹⁴C dates are mean dates for each deposit, based on bulked sediment samples (of 10 cm thickness for Blea Tarn, Pennington, 1970), and can therefore only be approximate, particularly if, as in Crummock Water, sediment deposition rates were very low during this period. The suggested dates from Buttermere and Crummock Water (based on NRM declination curve comparison) thus form an approximate "envelope" encompassing the published ¹⁴C dates.

Thus, apart from their lithological similarities, various datings suggest that, whilst individual horizons are not synchronous, a minerogenic layer present in each sequence represents the same general period of similar environmental conditions experienced by all these lakes.

LAKE/TARN	DATE/DATING TECHNIQUE	REFERENCE
Buttermere and Crummock Water (unit 3)	7 330 - 5 200 B.P. (NRM declination curve comparison)	this study
Burnmoor Tam	7 560 ± 160 B.P.	Pennington, 1970
Blea Tam	5 323 ± 120 B.P. 5 238 ± 55 B.P.	Pennington, 1973 - " -
Blelham Tarn	5 538 B.P.	Pennington, 1981

Figure 6.2. Comparison of published ¹⁴C dates for selected minerogenic layers in Lake District lacustrine sediment sequences, and the suggested age of the grey clay layer in Buttermere and Crummock Water.

This conclusion contradicts Pennington's (1973, 1981) interpretations of increased catchment erosion, specifically including topsoils, as the source for these sediments, but supports her (1970) environmental interpretation of the <u>within-tarn</u> source for the minerogenic sediments from Burnmoor Tarn. Interpretations, substantially based on mineral magnetic analyses of sources of the Buttermere and Crummock Water sediments, suggest that unit 3 is derived from within-lake reworking of sediments (section 6.2.1), and definitely not from catchment topsoils. Pennington's (1970) environmental interpretation therefore should have more widespread application throughout the Lake District.

The "environmental" interpretation of this clay layer is of particular interest, as controversy

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surrounds the cause(s) of the Elm Decline of *circa* 5 000 B.P. Pennington (1973, 1981) noted that whilst the decrease in elm pollen (both in percentage and absolute terms) observed in a range of lake sediment profiles may be accompanied in some areas by human impacts (elm could be used as animal fodder, as well as for fuel and a building material), its widespread and apparently synchronous occurrence throughout the British Isles suggests other causal factors - such as a subtle climatic change and/or tha incidence of a pathogen such as Dutch Elm Disease.

In the light of the results from Buttermere and Crummock Water, and the environmental interpretation of unit 3 proposed here, a reexamination of the sediments of some of the sites studied by Pennington (for example, from Blea Tarn, Barfield Tarn, and Blelham Tarn) using a mineral magnetic approach is recommended. This would establish whether the minerogenic layer present in a number of Lake District sequences, and frequently interpreted as reflecting increased <u>catchment</u> erosion, is actually derived from such sources (as suggested by Pennington, 1973, 1981) or is really reworked lacustrine sediment (as in Buttermere and Crummock Water). Broadly synchronous reworking of marginal lacustrine sediments in lakes across the region would be indicative of a climatic change (a decrease in net precipitation) - identification of which affect interpretations of causes of the Elm Decline.

6.3. Sedimentation mechanisms in oligotrophic lakes.

The model of the major controls on lake sedimentation presented in this study (Figure 1.2, page 16) illustrates the potential complexity of such a system, but also how, by working back from a study of sediment characteristics, details of controlling factors, and hence aspects of the environmental history of a catchment can be deduced. This is the basis of the "lake-watershed" approach (Oldfield, 1977), which has been used throughout this project. In this study, specific investigation of sedimentation processes within Buttermere and Crummock Water has also been undertaken (chapter 5). This provides greater detail for processes operating in what is shown as the end point on Figure 1.2, and labelled simply as "sedimentation".

First, Buttermere and Crummock Water, being closely linked, are likely to suffer effects of sediment trapping by the upstream lake. N.D.Smith (1981) noted effects of trapping on the characteristics of the sediments of a downstream lake (Bow Lake). There, the effects of formation of an upstream lake (following recent glacial retreat) were mainly the trapping of a proportion of the coarser sediment discharged from the glacier. This was expressed in Bow Lake by:

i) decreased sedimentation rates;

ii) decreased thicknesses of rhythmites (which thinned towards a visually homogeneous sediment in the distal zones of the lake);iii) the formation of sediment overflows and interflows rather than underflows, owing to the decreased turbidity, and hence density, of the waters of the principal tributary.

As discussed in section 4.4 (p.135), relatively consistent differences in the magnetic characteristics of lake sediment groups between Buttermere and Crummock Water, demonstrate the systematic capture of particular grain size ranges in the former. Thus, in comparison to those of Buttermere, the sediments of Crummock Water are relatively fine grained, as illustrated in Figure 3.9 (page 75). This observation leads to a further complication in the interpretation of the mineral magnetic analyses from this study, as trapping has introduced additional sorting of the fine fractions of catchment sediments. The

lake sediments therefore do not provide an <u>exact</u> analogue to sieved catchment samples (see discussion, section 6.1).

More unexpected was the finding that not only is the coarser sediment fraction preferentially deposited in Buttermere, but that the finest (SP) size was also relatively enriched in Buttermere and depleted in Crummock Water. Such an association of grain sizes, with diameters approximately an order of magnitude different, has been noted in other studies (Bradshaw and Thompson, 1985, Morrison, 1981). This effect is most probably explained by the large surface attraction of very fine (SP) magnetic grains. Morrison's results demonstrate that this phenomena in lake sediments is not restricted to ferrimagnetic material however, and hence may therefore be widespread.

Deposition of part of the sediment component must reduce the turbidity, hence the density, of the outflow (Buttermere Dubbs) from Buttermere. A reduction in inflow denisty will discourage the formation of underflows in favour of interflows or overflows. Hence, a further effect of Buttermere on sedimentation in Crummock Water will probably have been in encouraging the formation of interflows or overflows in favour of underflows.

Pharo and Carmack (1979) presented a model of lake sedimentation based on three mechanisms, namely:

i) delta progradation;

ii) formation of episodic sediment density surges (mainly by delta face failure);

iii) river plume sedimentation.

Despite its simplicity, sedimentological and geochemical observations in Kamloops Lake could be interpreted on the basis of this model (*op cit*).

Buttermere and Crummock Water represent a similar system (deep, oligotrophic, and with allochthonous clastic sediment sources), although they are much smaller in terms both of lake and catchment dimensions, and of the magnitude of sediment inputs. (Kamloops Lake is 25 km long, with contemporary sedimentation rates > 1 cm.yr⁻¹ in the delta front area,

compared to rates of 1 - 2 mm in Buttermere and Crummock Water, calculated using data from this study and from Pennington, 1981). Sedimentation in Buttermere and Crummock Water can therefore only be <u>partially</u> interpreted on the basis of Pharo and Carmack's sedimentation model.

As discussed in chapter 5, sedimentation in Buttermere and Crummock Water is dominated by river plume sedimentation and delta progradation. Thus, deposition is concentrated in the deltaic areas at the lake heads, with decreases in both sediment accumulation <u>rates</u> and of dominant <u>grain sizes</u> away from these areas. This reflects the gradual settling of suspended material as river plume energy, and hence competence, is lost by mixing with the lake water.

No evidence is seen in either the Buttermere or the Crummock Water sediments of the frequent sediment density surge activity ("the episodic formation of strong downslope movements of sediment-laden water", Pharo and Carmack, 1979, page 528) inferred from the sediment characteristics of Kamloops Lake (*op cit*). As discussed in section 1.5, this promotes the deposition of "linear fingers" of coarser sediment in the deeper areas of a lake. Their formation in Kamloops Lake was attributed mainly to delta slope failure, promoted by,

i) high rates of delta progradation (about 14 m.yr⁻¹), leading to formation of steep and unstable delta fore-slopes;

ii) development of relatively large amplitude waves over the 25 kmlength of Kamloops Lake able to attack and destabilise shorelines,particularly at the delta front;

iii) large annual variations in lake level (about 7 m), promoting destabilisation and allowing attack of parts of the delta face, encouraging periodic collapse.

Sedimentation rates are relatively low in Buttermere and Crummock Water. Hence deltas are less likely to build up large "metastable" slopes. Neither are there such large variations in inflow characteristics, owing to the relatively moderate maritime climate of the region. Both of these factors discourage such regular slope failure as inferred by Pharo and Carmack (1979) in Kamloops Lake. In fact, the only feature in the entire Holocene

sequence that may be attributable to this type of process is (our old friend) unit 3, the grey clay. This exhibits a slightly coarser sediment assemblage than the surrounding material (Figure 3.9), and is concentrated in the deeper parts of the lake basin. Consequently (in conjunction with other evidence, such as mineral magnetic indicators of sediment source) it is interpreted as representing a within-lake, reworked, deposit, probably produced by a fall in lake water level and the consequent exposure of marginal sediments (discussed in Section 6.2.1). The absence of <u>regular</u> density surge deposits in the Buttermere and Crummock Water sequence is thus indicative of the generally very stable and consistent catchment, input, and lake characteristics of this system. Incorporation of this sedimentation mechanism (the formation of episodic density surges) in a model applicable to Buttermere and Crummock Water is still necessary, as it has been an active process during the lakes' history.

Despite acknowledging the role of thermal stratification in influencing levels at which river inflows enter and travel along a lake, the sedimentation model presented by Pharo and Carmack (*op cit*) does not acknowledge the influence of stratification in the production of discreet sediment structures. This, as discussed in chapter 5, is the major mechanism involved in the production of the green chlorite laminae in Buttermere and Crummock Water. (Other factors, such as the development of a winter ice cover, may also be of importance for the formation of these laminae - as discussed in section 5.4).

The effect of stratification is to trap and concentrate a particular sediment fraction, deposited as a visually distinguishable layer following the lake overturn. The operation of this mechanism, and its effect on the vertical variation of sediment characteristics is illustrated in Stürm's (1979) sedimentation model (Figure 5.1, page 147, this report), where boxes 5, 7, and 8, depict the production of laminae following trapping of fine particulates during lake stratification.

The irregular occurrence of these layers in the Buttermere and Crummock Water sediments suggests the rather sporadic <u>effective</u> operation of this mechanism. This is interpreted in this study as mainly being due to the injection of river plumes below the epilimnion and/or insufficient development of water stratification, and hence ineffective trapping of fine



Figure 6.3. Principal sedimentation processes and effects on sediment distribution and texture which may occur in an oligotrophic lake. Model based on identification (chapter 5) of sedimentation processes operating in Buttermere and Crummock Water, and on sedimentation models presented by Pharo and Carmack (1979), and by Stürm (1979).

particulates (see section 5.4). Both processes being, to a degree, interdependent.

Undoubtedly, the chlorite-rich Borrowdale Volcanic Group bedrock within this catchment form a ready supply of chlorite, which provides the most obvious characteristic of the green laminae. Chlorite grains posses markedly different hydrodynamic properties to the bulk of the sediment (Morrison, 1981), and thus the laminae provide a prominent "marker" facilitating the identification in the sediment record of the operation of the above process. The process is in fact likely to be widespread in deep, oligotrophic lakes, as illustrated in Stürm's (1979) model ("core boxes" 5, 7, and 8, page 147). It may thus operate in lakes, including Kamloops Lake, where thermal stratification occurs. Easy identification of the operation of this process, however, relies on a supply of hydrodynamically and <u>visually</u> distinctive sediment, such as platy chlorite. It is thus possible that this process has been overlooked in the description and interpretation of many lake sediment sequences - even Windermere, where similar chloritic laminae are easily distinguished from the surrounding sediment.

Identification of the sedimentation processes operating in Buttermere and Crummock Water has therefore both confirmed aspects of Pharo and Carmack's (1979) model of sedimentation, and highlighted some of its deficiencies in terms of its wider application. A more widely applicable model must incorporate the potential effects of lake stratification as a discreet sedimentation process (as illustrated in Stürm's (1979) model of idealised sedimentary structures) as well as its potential influence on other factors. The model shown in Figure 6.3 integrates the principal mechanisms of both sedimentation models, illustrating those mechanisms potentially operating in a clastic, oligotrophic lake system. This is an improvement both on Pharo and Carmack (1979), and Stürm (1979), although the latter illustrates in greater detail the various vertical variations in sediment textures resulting from the interaction of just two hydrological parameters (suspended sediment influx and water stratification). The processes shown in the model in Figure 6.3 can explain the sedimentological characteristics identified in Buttermere and Crummock Water, the inferred sedimentation processes involved, and also provides a framework against which results from future lake sediment studies may be compared.

6.4. Conclusions.

1) Mineral magnetic analyses are of particular value in limnological studies, not only because they can help elucidate particular problems, such as correlation of visually homogeneous cores and investigation of source - sediment linkages, but because this type of analysis may also be applied to a wide <u>range</u> of different areas of a study.

2) As in a number of other lake sediment-based studies (for example, Appleby *et al*, 1985, Dearing *et al*, 1981; Dearing, 1983) core correlations using mineral magnetic measurements have proved valuable for cross-correlation of dated features and identification of changing lake sediment accumulation patterns in Buttermere and Crummock Water. Where sediments change relatively little over time - as for periods during the Holocene as recorded in the Buttermere and Crummock Water lake sediments - and consequently no magnetic marker horizons are produced, the value of this technique for core correlation is necessarily reduced. Few other correlation techniques are appropriate under such circumstances.

3) Mineral magnetic measurements provide a convenient and relatively rapid method of characterising and thereby tracing movements of natural sediments within a defined sedimentary system. These techniques do not allow the precise identification of magnetic mineral compositions. This reflects the very wide range of minerals, concentrations, and grain size ranges encountered in natural samples.

4) This study has extended the range of applications that may confidently be explored using the mineral magnetic methodology. Magnetic measurements used here have, for the first time:

a) formed the basis for identification of subtle magnetic grain size changes which occur within a lake;

b) have been used to identify a widespread cryptic and approximately-dated marker horizon;

c) have demonstrated that mineral magnetic measurements may be used for the identification of sediment-source linkages in lithologically complex catchments, when previously these studies had been restricted to rather simpler catchments,.

Recent development of methods allowing quantitative estimates of source components in derived sediments (Yu, 1989, Yu and Oldfield, 1989) represent a further refinement of this approach, which increases its value for environmental investigations.

5) This study has broadly confirmed interpretations of Lake District catchment changes proposed by Mackereth (1965, 1966a) and Pennington (1981). The data used for these published studies was obtained from other Lake District sites (including those of Windermere, Ennerdale, and Esthwaite). This demonstrates a uniformity of catchment and lake development throughout the region.

6) Evidence from this study, principally based on mineral magnetic analyses, has provided the first positive evidence for sudden inwash of topsoil in historical times. This inwash correlates with marked increases in catchment net sediment supply rates around the time of the wave of Norse immigration to the region (*circa* 900 - 1 000 A.D.) and is therefore interpreted as reflecting their modification of the region.

7) Unit 3, the "grey clay" is best interpreted as recording a lowering of lake water levels and the subsequent focusing of marginal glaciogenic mineral sediments towards the deep lake centre. This conflicts with previous interpretations of broadly comparable minerogenic horizons observed in other lakes (cf Pennington, 1973, 1981). Further research is required to confirm the age and areal distribution of this unit, and pollen analysis of sediment cores could provide a direct indication of vegetation change and the position of unit 3 with respect to the Elm Decline of *circa* 5 000 B.P.

8) Sedimentation is carried out mainly by the process of river plume dispersal, and hence is concentrated in delta areas, with decreases in both sedimentation rates and dominant grain

sizes in distal areas of the lake.

9) The presence of chloritic laminae indicates that the lake water body may directly influence sedimentation under certain conditions. Similar laminae are present in the Windermere sediments (author's own observations), but have only rarely and briefly been described (Pennington, 1943; Mackereth, 1965). Their more widespread occurrence is predicted for similar periodically stratified, oligotrophic lakes, but identification is only made easy when, as in Buttermere and Crummock Water, the laminae are composed of visually distinctive forms of sediment.

10) A model of lake sedimentation mechanisms has been presented, based on the integration of those developed by Stürm (1979) and Pharo and Carmack (1979). This explains sedimentation processes occurring in Buttermere and Crummock Water, and is potentially more widely applicable. It provides a comparative framework for identification of sedimentation processes probably operational in similar lake systems.

This study has therefore demonstrated the value of the mineral magnetic methodology in lake sediment studies, has allowed construction of a more detailed environmental history of the Buttermere and Crummock Water catchment than previously available, in the process highlighting a major anomaly in the regional interpretation of *circa* 5 000 B.P. minerogenic layers observed in a number of other lakes in the Lake District (Pennington, 1970, 1973, 1981). It has also allowed some general interpretations of sedimentation processes in clastic, oligotrophic lakes.

As one of the aims of the project, these conclusions also demonstrate the value and the uses to which analyses of <u>clastic</u>, non-biogenic, components of lake sediments can be put. Their value in limnological studies has been enhanced by analytical and methodological developments in the field of mineral magnetism. Analyses of clastic sediment components can provide more direct evidence, for example, for the provenance of sediments, and allow substantial interpretation of environmental conditions operational through history.

Throughout, reference has been made to results from published studies based largely on analyses of biogenic components of lake sediments, as well as to those obtained directly from analyses of the sediments of Buttermere and Crummock Water. These two sources of information are not exclusive however, but, ideally, should be complementary. Such an integrative approach, where a more complete understanding of an entire lake watershed, a complex biogeochemical system, is obtained through use of a variety of complimentary techniques - an interdisciplinary approach - has been emphasised by, for example, Frey (1969), as well as Oldfield (1977) and O'Sullivan (1979).

Nevertheless, this study has demonstrated the great value of analysing one component of a lake sediment (the clastic, non-biogenic sediment fraction) which has traditionally received somewhat less direct attention than biogenic material. In this study, use of the clastic, inorganic sediment fraction has allowed investigation of a range of areas of environmental interest, specifically: investigation of lake sediment distribution and depositional processes; identification of source - sediment linkages; and interpretation of environmental change during the Holocene.

APPENDIX 1.

MINERAL MAGNETISM, INSTRUMENTATION, PARAMETERS, AND INTERPRETATION.

Physical basis of mineral magnetism.

Detailed explanations of the physical basis of palaeomagnetism are provided by Stacey and Banerjee (1974), Tarling (1983), and Thompson and Oldfield (1986). Magnetism, and the response to applied magnetic fields, is produced at the atomic scale by the spin and/or orbital motion of electrons around the nucleus of an atom. Different electron pairing arrangements and spin directions within particular lattice arrangements result in contrasting magnetic characteristics. These characteristics remain constant for similar crystal lattices and mineral compositions, and therefore allow identification of particular minerals or groups of minerals according to a magnetic "signature". All substances thus posses magnetic characteristics, although for many (diamagnetic and paramagnetic materials for example) these will be of relatively very low magnitude. The magnetic signature of particles is influenced by three major factors: composition (mineralogy); grain size; and grain shape. Theoretical implications of these are briefly discussed below.

Compositional effects.

All materials exhibit some type of magnetic response, and have been classified accordingly as either paramagnetic, diamagnetic, ferromagnetic, ferrimagnetic, or antiferromagnetic. Iron exhibits very strong magnetic responses, and, as the fourth most abundant element in the earth's crust, its minerals (mainly the iron oxides) account for most of the magnetic response of a rock, with iron sulphides or manganese oxides becoming relatively more important with very low concentrations of iron oxides (Thompson and Oldfield 1986, p.13). The iron oxides are classed as ferromagnets, with variant subgroups (ferrimagnets and antiferromagnets) being more commonly found in natural materials (Thompson and Oldfield 1986, p.14).

Ferrimagnetism.

Ferrimagnetic minerals crystallise with a spinnel structure, that is, a face-centred cube - which is very flexible in terms of the cations it can accept. Examples include magnetite

(Fe₃O₄) and maghemite (γ Fe₂O₃), these possessing a spinnel structure, with two types of magnetic sites containing antiparallel moments of different magnitudes (Thompson and Oldfield 1986). The unequal magnitude of the moments results in a net magnetism which is parallel to and in the same direction as, an applied field. Some of this is retained on removal from the field as a remanent magnetism.

Antiferromagnetism.

Antiferromagnetic minerals crystallise with a rhombohedral unit cell (the corundum structure) and possess two antiparallel magnetic sublattices, which, ideally, are equally opposed and which would thus exhibit zero net magnetisation. Imperfections of the lattice structure, spin canting (a slight modification of true antiferromagnetic antiparallelism), and compositional impurities all result in the production of imperfect antiferromagnets, which exhibit net magnetisation in an applied field and a remanent magnetisation on removal from

it. Haematite (\approx Fe₂O₃) is a common natural imperfect antiferromagnetic mineral, the imperfections being a result of spin canting (*op cit*).

Diamagnetism.

Diamagnets are materials which exhibit weak negative magnetisation (that is, of opposite direction to the applied field). This is due to field interaction with the orbital motion of electrons (all electron spin motions are paired and hence balance one another), which is lost on removal from the field. Common diamagnetic minerals include quartz, the feldspars, calcite, and water (*op cit*).

Paramagnetism.

Paramagnetic materials exhibit a weak, positive magnetisation, which is a result of partial reorientation of the electron spin magnetic moments when placed in a field. These are easily rearranged by thermal agitation, and so the spontaneous magnetisation of paramagnetic materials is temperature dependent. Such magnetisation is also lost on removal from the applied field. Common paramagnetic materials include olivine, pyroxene, garnet, biotite, iron and manganese carbonates, and amorphous iron oxide precipitates (Foster *et al*, 1985).

In samples containing a mixture of all of these groups, effects caused by diamagnetism, and by paramagnetism, tend to be swamped by those resulting from ferrimagnetic and antiferromagnetic material. Applications of mineral magnetism have therefore usually involved distinguishing the relative abundances of "hard" (that is antiferromagnetic or "haematite-type") and "soft" (ferrimagnetic or "magnetite-type") mineralogies, (Thompson and Oldfield, 1986).

Paramagnetic and diamagnetic effects are generally small in comparison to the magnetic effects produced by ferrimagnetic and antiferromagnetic material. The diamagnetic effects of water can frequently be seen in the volume susceptibility measurements of fresh sediment cores. The wettest sediments at or just below the sediment/water interface may exhibit lower volume susceptibility values owing to the weak negative diamagnetism of water.

Grain size effects.

The magnetic characteristics of a grain are strongly influenced by its size (Day *et al*, 1977). This is due to the fact that within grains exceeding a certain size, it is more efficient for the magnetisation to break down into individual volume elements (domains), within which electron spins are parallel. These are known as multidomain (MD) grains, and the domains are orientated so that the increased magnetic forces in such a grain are reduced by mutual interaction.
As grain size is reduced, there is eventually only sufficient volume for a single domain - it is now energetically advantageous for each single grain to form its own, single, magnetic domain. Such grains are known as single domain (SD) grains.

For very small grains, a threshold is reached where the energy of thermal vibration at "room temperature" is of equal or greater magnitude to their magnetic energy. Approximate energy equivalence allows continual thermal reorientation, so that these grains cannot retain a stable remanent magnetism. They do however exhibit a very strong spontaneous magnetisation under an applied field, similar to, but of much greater magnitude than, paramagnetic material. Hence they are called "superparamagnetic" (SP) grains.

The presence of superparamagnetic material is thus characterised by high susceptibility, low remanent magnetism, and high frequency dependent susceptibility (Xfd - a derived value from susceptibilities measured at different frequencies, see section "Parameters...", following).

A further domain state is termed pseudo-single domain or two-domain (PSD grains). These are intermediate in size between single domain and multidomain grains, and are considered to be multidomain-type ("size") grains that possess lattice imperfections which effectively "lock" domain walls. Grains of this type therefore fail to exhibit remanence changes in steady proportion to an applied field (Tarling, 1983), and have relatively poorly understood characteristics. They exhibit aspects of both single domain, and multidomain behaviour (Dunlop, 1981).

The critical sizes for a change from MD to SD grains for magnetite is in the region of a grain length of $0.1 - 1 \mu m$, but this varies with grain composition and shape (Figure A1, following page, illustrates shape effects on magnetite grains). In contrast, the multidomain/single-domain transition size of haematite is much larger than that of magnetite. This is largely a product of the lower saturation magnetisation of haematite. The critical size is greater than 0.1 cm - implying that most natural haematite grains will be of the single domain size range (Thompson and Oldfield, 1986).

Identification of the domain state of a grain thus allows inference of its possible size. Current understanding of grain domain theory is incomplete however, particularly for the PSD domain. Empirical data supplements the theoretical through the investigation of pure, synthetic, sized ferrimagnets, and magnetic parameters have been shown to display continuous variation across the entire grain size range, with X, Xfd, and X/ARM being particularly responsive to size changes at the fine to ultrafine end of the magnetic grain size spectrum (Maher 1988).



Data from "Untitled Data"

Figure A1. Relation of domain state to grain dimensions for magnetite, grain length in μ m. (After Thompson and Oldfield 1986, their Figure 2.9).

Grain shape effects.

Effects of shape in modifying domain state are illustrated in Figure A1. Magnetic anisotropy also results from shape effects, where magnetic properties vary with the direction of the grain axes. There are three forms of magnetic anisotropy: magnetocrystalline anisotropy; strain anisotropy; and shape anisotropy.

Magnetocrystalline anisotropy describes the phenomenon whereby various axes of a crystal possess different magnetic properties. This means that it may be harder to magnetise a crystal along certain crystallographic axes. This is important for mineral magnetic measurements since magnetocrystalline effects depend solely upon crystal structure and composition, and in natural imperfect antiferromagnets (haematite, for example) this causes their characteristic low spontaneous magnetisations and very high coercivity (that is, the ability to retain an imparted remanence whilst under a strong opposite negative field).

In haematite, the basal plane of crystals is about 100 times more readily magnetised than any other, whilst the internal anisotropic forces in magnetite, for example, are relatively very weak (Tarling, 1983). This implies that ferrimagnetic grains are readily magnetised and demagnetised, while antiferromagnetic grains exhibit a much greater resistance to magnetisation, termed magnetically "hard" characteristics. Once magnetised, hard, antiferromagnetic, grains will then show considerable resistance to demagnetisation. Such characteristics allow the use of remanence measurements and ratios derived from these, to distinguish between mainly ferrimagnetic and mainly antiferromagnetic magnetic assemblages (as discussed below under "Isothermal Remanent Magnetisation").

Strain anisotropy describes the effect whereby a grain changes size when subjected to a magnetic field (that is the piezo-electric effect), or conversely, whereby its magnetic properties change on application of stress. This effect is sometimes considered to be a modification of crystalline anisotropy (Thompson and Oldfield 1986, p.8).

Like crystalline anisotropy, shape anisotropy means that magnetisation of a sample is easier in certain directions than in others. This is caused by the external magnetostatic forces from the magnetic poles of a body being produced by the interaction of all domains. In spherical grains, since the separation of the surface poles is constant in all directions, this results in zero shape anisotropy, whilst, conversely, in a rod there is high shape anisotropy. This effect may also be of significance when considering the magnet characteristics of a whole sample. Samples are therefore normally kept in the same relati orientation for all measurements, in order to maintain this as constant within sample

Instrumentation.

The magnetic measurements used in this study were all collected using a combination of a Bartington M.S.1 susceptibility meter with a core scanning sensor (M.S.1C) for whole core measurement, and a dual frequency single sample sensor (M.S.1B) for dried sediment samples at low and high frequency (0.47 and 4.7 kHz respectively). A Molspin pulse magnetiser (producing applied fields of up to 1 T), and Molspin flux magnetometer were used for the determ, ination of isothermal remanent magnetisation measurements); and a Molspin anhysteretic magnetiser, for measurements of anhysteretic remanent magnetisation. Anhysteretic magnetisations were acquired in an a.c. field smoothly decreasing from 100 mT to zero with a superimposed field of 0.05 mT are grown and then measured using the Molspin flux magnetometer.

Parameters and their interpretation.

Individual parameters and their relation to grain characteristics are discussed below. For determination of specific grain characteristics, parameters are normally used in conjunction with each other, in order to account for the maximum number of variables.

Parameters other than volume susceptibility are expressed on a mass specific basis, that is, after normalisation by the sample weight. Further, varying organic contents between samples are accounted for by subtracting the approximate weight due to organic content prior to normalising (here termed "effective weight"). Organic contents were estimated by determination of loss-on-ignition. The effective weight is therefore calculated as: We = Wt - (Wt x LOI)

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^{-100}_{-100}
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We = effective weight Wt = total weight, (including organic fraction) LOI = loss-on-ignition (percent value) Normalising on this basis therefore allows comparison between samples of different weights and different organic contents.

Susceptibility.

Susceptibility is defined as "the ratio of induced magnetisation to applied field", that is, a measure of the degree to which a sample is attracted to a magnet (Oldfield *et al*, 1978a). It is therefore influenced by the concentration of ferrimagnetic minerals in a given sample, but is also affected by their size, shape, and internal stress of these minerals (Thompson *et al*, 1975, pp.687-688). If ferrimagnets are of low concentration in a sample, antiferromagnets (haematite for example), and, at still lower concentrations, paramagnetic and diamagnetic material may contribute to the susceptibility. Additionally, the presence of even a small amount of superparamagnetic material results in a disproportionately high susceptibility reading. The Xfd parameter identifies the presence of superparamagnetic material in a sample (see the discussion of Xfd following).

Susceptibility measurements performed in a weak alternating field are the most reliable (Mullins, 1977). The sensors used in this study operate using this principle, switching between two different measuring frequencies for Xfd measurements, namely, 0.47 and 4.7 kHz. In mineral magnetic studies three susceptibility measurements are commonly used. These are: volume susceptibility (k); low frequency mass specific susceptibility (Xlf); and high frequency mass specific susceptibility (Xhf).

Measurements of whole core susceptibility (k) are routinely made on wet sediment, including whole sediment cores, and may therefore involve inaccuracy in terms of the exact sample size, and will be affected by the water content of the sediment. Preservation of the sediment/water interface (as with Mackereth minicores) will additionally affect susceptibility readings, owing to the diamagnetic properties of water. Whole core susceptibility measurements therefore record <u>relative</u> changes of susceptibility, and readings in SI units are dimensionless.

Mass specific susceptibility measurements allow normalised readings to be calculated (SI

units $m^{3}.kg^{-1}$), which can thus be compared directly between different samples. Mullins (1977) discusses a range of uses for susceptibility measurements in soil science, some of which have been under experimental investigation since the 1950's. However, since susceptibility is dependent upon several variables (the types of mineral present, their relative concentrations and grain size distribution, for example), the use of susceptibility alone may be extremely ambiguous. In contrast, Maher (1986) and Özdemir and Banerjee (1982) for example, were able to attempt more detailed identification of the magnetic minerals present in a number of soils, and to differentiate them on the basis of the combined use of susceptibility and a <u>range</u> of other mineral magnetic parameters.

Measurements at different frequencies (Xlf and Xhf) are made in order to calculate the frequency dependent susceptibility (Xfd) for a sample. Multidomain grains have been shown to exhibit very small reductions in susceptibility with increased frequency (of the order of 0.3% per ten-fold increase in frequency). Smaller grains (around the single domain/superparamagnetic grain size boundary, namely, diameters of ~ 0.02 μ m in isodiametric magnetite, Zhou *et al*, 1990) show a marked reduction in susceptibility with increasing frequency of measuring field. This is due to the short delay in response to changing magnetic field conditions by the finer grains (magnetic viscosity), which becomes more significant at higher frequencies of measurement. This difference is expressed as a percentage of Xlf (see equation below), and the Bartington dual frequency susceptibility sensor, which was used in this study, measures at frequencies of 0.47 kHz and 4.7 kHz for Xlf and Xhf respectively. Dunlop (1981) has suggested that susceptibility differences produced by such changes in the frequency of measurement are the best test of superparamagnetic behaviour in a sample. The range of percentages so far encountered in natural samples is 0-24% (Thompson and Oldfield 1986, p.56).

The formula used for the calculation of frequency dependent susceptibility (Xfd%) is,

$$Xfd\% = \frac{Xlf - Xhf}{Xlf}$$
. 100

Isothermal Remanent Magnetism (IRM).

Isothermal remanent magnetism (IRM) is "the magnetic moment induced in and retained by a sample after it has been placed at room temperature in a magnetic field" (Oldfield *et al*, 1978a). IRM is usually measured for a number of forward magnetic fields of different magnitude, up to the saturating value, and then a series of "back fields" (that is of opposite polarity to the applied saturating field).

Remanence results from the alignment of magnetic domains of the component grains in a sample. Beginning from a natural, randomised, state, an applied field causes domain walls to "jump" past energy barriers within a crystal. On removal of this field, certain of these will be unable to roll back through the energy barriers to their original alignment, and are said to have undergone Barkhausen jumps. These cause the observed remanence. When all domains are aligned, the theoretical saturation level has been reached. When the sample is removed from this field there is a certain loss of magnetism (relaxation), to give the saturation isothermal remanent magnetism (SIRM).

SIRM is mainly affected by mineralogy, ferrimagnetic mineralogies resulting in a relatively low, and antiferromagnetic mineralogies in a much higher SIRM. The grain size of the soft component also produces a significant effect. In magnetite it causes a rapid and large increase in SIRM over the change from SP to the SP/SD boundary, followed by a less rapid, eventually gradual, decline through to MD grains (Thompson and Oldfield 1986, their Figure 4.7). SI units for SIRM measurements are in m.Am².kg⁻¹.

Measurement of SIRM is in fact limited by the size of the maximum field available in a laboratory. Samples in this study were "saturated" under a field of 1 000 mT (1 T) which is sufficient to saturate soft, ferrimagnetic grains. Hard, antiferromagnetic, components of a sample are unlikely to be fully saturated in a field of this magnitude (Robinson 1986).

The various backfields determined for a particular sample can be plotted together as a curve of remanence hysteresis (or coercivity plot). The shape of these curves depicts mineralogy and grain size, hard assemblages producing relatively flatter curves (owing to the relatively very high fields required for saturation), and soft assemblages steeper curves. Changes in

grain size tend to move the curve along the axis depicting backfield magnitude(usually plotted as the horizontal, "x"-axis) while still retaining the general curve shape (Thompson and Oldfield, 1986, their Figure 4.10). Saturation remanences greater than 0.2 T are typical for hard mineralogies, and values in the range 0.02 T (grain sizes of approximately 1 μ m) to 0.05 T (greater than 100 μ m) for soft mineralogies (Thompson *et al*, 1980). Certain backfield ratios have been shown to discriminate effectively between particular magnetic properties. The S-ratio (Hirons and Thompson, 1986), or simply "S" (Maher, 1986) is the value obtained from the 0.1 T backfield (IRM_{-100mT}) reading divided by SIRM. The basis of this ratio is that ferrimagnetic material will be fully saturated in a field

of 1 T and be demagnetised by a reverse field of 0.1 T, antiferromagnetic material exhibiting a greater resistance to demagnetisation. Thus, soft assemblages typically produce strongly negative "S" values, and harder assemblages show positive "S" values Maher (1986).

Variations in S primarily reflect changing proportions of ferrimagnetic (magnetite-type) and antiferromagnetic (haematite-type) mineralogies, and is concentration independent. Grain size variations of the magnetic components will affect S values, but this is only a "secondary" effect (Robinson 1986). Such ratios are dimensionless, and may be expressed as a ratio of SIRM (between -1 and +1), or as a percentage of SIRM.

HIRM (the ratio: IRM₋₃₀₀ - SIRM, Robinson, 1986) records relative changes in the total concentration of the antiferromagnetic components. This is based upon the observation that IRM acquisition in fields greater than 300 mT is effectively due to antiferromagnetic minerals only (Collinson, 1975). Therefore the difference between SIRM and IRM₋₃₀₀ is concentration dependent, but responds only to changes in mineralogy. High HIRM values will indicate increasing proportions of antiferromagnetic ("hard") components.

Remanence ratios used in this study are: soft (IRM₊₂₀), hard (SIRM - IRM₊₃₀₀); soft % (that is, soft normalised with respect to SIRM, IRM₊₂₀/SIRM); and hard % (that is, hard normalised with respect to SIRM, [SIRM - IRM₊₃₀₀]/SIRM). The use of these ratios is based on the discussion preceding, where it was stated that IRM acquisition in fields

greater than 300 mT is effectively due to the presence of antiferromagnetic minerals alone (Collinson, 1975). Only minerals of very soft magnetic characteristics will retain remanences after being subjected to a low field of 20 mT, and will be fully saturated after subjection to a field of 300 mT. Any increase in IRM on application of the "saturating" 1 000 mT field must be due to the presence of minerals of much harder magnetic characteristics. These "hard" parameters can therefore be used as a guide to the relative importance of haematite a common antiferromagnetic mineral), whilst the "soft" parameters provide the best approximate indication of the relative importance of magnetite in a sample (Oldfield and Richardson, 1990).

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The remanence ratios used in this study therefore discriminate between the minerals displaying extremely soft, ferrimagnetic, characteristics, and those with much harder, antiferromagnetic, characteristics. High values of the soft and soft % parameters thus indicate a magnetic assemblage rich in the ferrimagnetic component, and *vice versa*. High values of the hard and hard % parameters imply an abundance of the antiferromagnetic component, and *vice versa*. Expression of the soft and hard parameters as a percentage of SIRM (soft % and hard %) provides an indication of major mineralogy independent of the total concentration of remanence carrying magnetic minerals in a sample.

Anhysteretic Remanent Magnetism.

Anhysteretic Remanent Magnetism (ARM) represents the field retained after subjecting a sample to a strong alternating field which is smoothly reduced to zero in the presence of a small steady field (Thompson and Oldfield, 1986). The fields used for samples in this study were a maximum a.c. field of 100 mT, and a small d.c. field of 0.04 mT (following Maher 1986, and Robinson 1986). SI units are Am².kg⁻¹.

ARM is affected both by mineralogy and grain size. However, it has been shown that grain size variations are the more significant factor (for example, Johnson *et al*, 1975). In particular, ARM discriminates between changes in domain state at the SD/MD boundary, SD grains (diameters $0.02 - 0.04 \mu$ m, Maher, 1988) exhibiting markedly higher values than MD grains (although the changes are in fact progressive, Gillingham and Stacey, 1971). The relatively high ARM values of the finer SD grains results in a disproportionate

contribution of these values on the ARM measurements of mixed grain size (that is, mixed domain state) assemblages. Large changes in ARM values might therefore reflect relatively subtle changes in grain size. Interpretation of records of ARM may become complicated however by variations caused by grain interaction and its relatively complicated dependence on changing grain size (Thompson and Oldfield 1986, p.29).

Interparametric Ratios.

All the parameters discussed in the preceding sections ultimately reflect changes in more than one characteristic of a particular assemblage, noticeably concentration, mineralogy, and grain size/domain state. Whilst certain of these are relatively specific and are of value when used independently, combinations, as ratios, may be used to form interparametric ratios eliminating dependence upon certain variables.

ARM ratios.

Banerjee *et al* (1981) and King *et al* (1982) discuss the theory and use plots of ARM vs. X to distinguish magnetite domain state, and hence grain size, in natural samples. King *et al* (1982) additionally provide a quantitative model, based upon data from prepared magnetite grains, for predicting the grain size of magnetite grains from specific X and ARM values. Unfortunately, complicating factors such as grain size variation within assemblages (particularly if these include SP-size magnetite), shape effects, and possible influences of the non-magnetic matrix on X values, preclude the quantitative use of this for natural samples. King *et al* (*op cit*) therefore suggest that, at present, the best use of the parameter is probably for identification of distinct groups of samples containing magnetite of the same predominant grain size. The method has been used by Robinson (1986) though for example, who, by comparing ARM/X with changes in other down core parameters, was able to relate it to changes in dominant domain state (and hence grain size) in natural marine mixed sediment assemblages.

ARM/X is therefore of use in identifying grain size fluctuations of the ferromagnetic components of a sample and eliminates variation due to variable sample concentration. In

general, high values of ARM/X imply a large proportion of fine SD ferrimagnetic particles (diameters $0.02 - 0.04 \mu m$, Maher, 1988). Finer particles will tend to dominate the ARM response of a mixed assemblage even when volumetrically unrepresentative relative to other magnetic fractions (Robinson 1986). Assemblages dominated by coarser particles (PSD or MD grains) exhibit lower ARM-ratios than SD-dominated assemblages, although where absolute values of both SIRM and X are low, low ARM/X ratios are indicative of the presence of paramagnetic iron compounds (Williams 1987).

As X may be influenced by both ferrimagnetic and antiferromagnetic material, if present in sufficient concentrations, the ratio IRM_{+20}/ARM has been used here to identify the domain state of the soft magnetic component in isolation. This follows from the discussion above, where it was shown that only soft, ferrimagnetic grains will be identified by the IRM_{+20} parameter ("soft"), and so the IRM_{+20}/ARM parameter allows identification of the dominant domain state of a particular fraction of the ferromagnetic assemblage to provide further discrimination between sediment samples.

р 14 14

SIRM/X ratio.

This parameter (expressed in SI units in kA.m⁻¹) is less sensitive to variations in the total concentration of magnetic minerals in a sample. It is still influenced by the type, size, and shape of magnetic particles, so that where, for example, samples incorporate similar mineral types, grain morphometry will be of greater significance. Also, a combination of fine and SP-size magnetite will result in a similar SIRM/X ratio to that of a sample dominated by coarse magnetite. The presence of SP grains in a sample is readily established by analysis of the Xfd response however, so comparative inspection of this parameter should allow a more accurate identification of the true grain characteristics.

Typically, SIRM/X values greater than 200 kA.m⁻¹ signify a dominantly hard mineralogy ("haematite-type"), and values in the range 1.5 to 50 kA.m⁻¹ indicate a dominantly soft ("magnetite-type") mineralogy (increasing with size) (Thompson *et al*, 1980). Snowball and Thompson (1990) have used the SIRM/X ratio to identify samples where the authigenic sulphide greigite is the main magnetic mineral present. Empirically, SIRM/X

ratio values above 40 kAm⁻¹ identified those samples with abundant authigenic greigite and values < 40 kAm⁻¹ indicated the presence of detrital magnetic assemblages (*op cit*). In the same study, analysis of pure greigite extracted from Loch Lomond sediments exhibited an SIRM/X ratio of 70 kAm⁻¹.

APPENDIX 2.

PREPARATION OF MAGNETIC SPHERULE EXTRACTS.

A) Extraction procedure.

1) Sediment samples were dried at $\leq 40^{\circ}$ C in order to avoid hard baking of clays.

2) All glassware was thoroughly washed to ensure it was dust-free.

3) Acetone was used to make up a sediment "slurry" in order to speed evaporation and to prevent accumulation of large volumes of liquid during the extraction. A face mask was worn to prevent inhalation of the fumes.

4) A large permanent magnet with a glass petri dish, here temporally attached by Blu Tak over one pole of the magnet, was assembled in advance. The magnetic gradient was increased by attaching iron bolts, or other similar ferrous objects, to the pole - effectively reducing the pole surface areas, and keeping the magnetic field constant.

5) A small sample of known weight (≤ 1.000 g)) was gently crushed, mixed in a test tube with acetone to form a slurry and further disaggregated by placing the mixture in an ultrasonic bath for 10 - 15 minutes.

6) Some slurry was pipetted from the test tube into the petri dish. More acetone was added if necessary in order to thin the sediment slurry.

7) Gentle agitation of the magnet allowed the whole sediment slurry to pass above the magnetic pole situated beneath part of the petri dish. Opaque magnetic particles congregated above the magnetic pole directly above the petri dish.

8) When all magnetic particles appeared to have been withdrawn from the slurry, the remaining liquid (the "non-magnetic" slurry) was drawn off with a pipette and retained.

The magnetic fraction, still grouped above the magnetic pole, was brushed off the petri dish into a small beaker on removal of the magnet.

9) Steps 6 - 8 were repeated until all the original slurry had been processed.

10) The "non-magnetic" slurry was rewashed (steps 6 - 8) in order to ensure complete retrieval of magnetic particles.

11) The magnetic slurry was finally washed repeatedly (steps 6 - 8) until clay adhering to the magnetic particles had been removed, in order to give a clear liquid in which opaque black, magnetic particles rapidly settled out.

B) Mounting and Spherule Counting.

1) The acetone above the magnetic material in the final slurry was gently pipetted off until a small amount remained which was left to evaporate away.

2) The now dry magnetic material in the beaker was transferred to a glass well slide. A moistened fine paint brush, viewed through a supported magnifying glass, was used to pick up material.

3) Cover slips, each with a 10×10 mm grid drawn on (to enable complete coverage of the slide with no repetition) were glued over the slide well.

4) Slides were viewed on a binocular microscope at up to 100 x magnification, with an eyepiece graticule for measurement of spherule diameters. Slides were placed on a white slip of paper with strong side-lighting as this was found to give the best contrast and illumination for easy identification of spherules, which are distinguished by their regular shape and metallic lustre.

APPENDIX 3.

POROSITY CALCULATION METHOD AND DATA USED.

(Method after Briggs, 1981)

Here, "porosity" is defined as a measure of the total volume of pore spaces in a sample. This is generally less useful than values for average pore space diameter, but pore-space volume is much more readily measured than average pore diameters.

Porosity, P, as a percentage of the total sample weight:

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= (1 - Db/Gs).100
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where: Db = dry bulk density Gs = specific gravity

Dry Bulk Density, Db, (g.cm⁻³):

dry weight of known volume of sample (g) volume of fresh sample (cm³)

Specific Gravity, Gs, (g.cm⁻³): M2 - M1

(M4 - M1) - (M3 - M2)

M1 = weight (g) empty density bottle.

M2 = weight (g) density bottle + oven dried (105°C), crushed sample.

M3 = weight (g) density bottle + oven dried sample, bottle filled with distilled water.

M4 = weight (g) density bottle + distilled water only.

Data from equivalent horizons from C3L and B2L:

Measurement	Core, and depth (cm)								
	C3L, 7	C3L, 9	B2L, 17	B2L, 19					
m1	33.2014	34.2251	34.2251	33.2014					
m2	38.0770	39.1249	41.0029	41.4041					
m3	85.3422	86.2049	87.8464	86.9512					
m4	83.0931	84.1864	84.1864	83.0931					
Specific gravity (g.cm ⁻³)	1.86	1.70	2.17	1.89					
Dry bulk density (g.cm ⁻³)	0.4062	0.4062	0.3183	0.2630					
Porosity %	78.2	76.1	85.3	86.1					

APPENDIX 4.

CALCULATION OF LAKE SEDIMENT ACCUMULATION, INPUT, AND CATCHMENT NET SEDIMENT SUPPLY RATES.

(Method after Davis and Ford, 1982)

1. accumulation rate, (mg.cm⁻².yr⁻¹),

= sediment dry bulk density (mg.ml⁻³) deposition time (yr.cm⁻¹)

where:

bulk dry density, (g.cm⁻³),

= dry weight of known volume of sample (g)
volume of fresh wet sample (ml³)

and:

deposition time, (yr.cm⁻¹), is the number of years taken for a 1 cm thickness of sediment to accumulate, calculated using thicknesses of sediment between (mainly NRM) dated sediment horizons.

2. Lake sediment input, (kg.yr⁻¹),

(from (1), accumulation rates are shown as mg.cm⁻².yr⁻¹ = 10^{-2} kg.ha⁻¹.yr⁻¹)

= accumulation rate (kg.ha⁻¹.yr⁻¹) x basin area (ha)

(n.b. effective basin areas are: Buttermere 0.61 x 10^{2} ha; Crummock Water 1.64 x 10^{2} ha, illustrated in figure 4.1)

3. Catchment net sediment supply rate, (analogous to catchment erosion rate - see discussion, section 4.4.1 - expressed in kg.ha⁻¹.yr⁻¹),

= lake sediment input (kg.yr⁻¹) catchment area (ha)

(n.b. catchment areas are: Buttermere 16.9 x 10^{2} ha; Crummock Water 43.6 x 10^{2} ha).



Appendix 5. "Raw" XRD integrated line intensity profiles (that is, profiles not calculated as closed number system percentages) from C1L for illite, chlorite, plagioclase feldspar, and quartz, based on measured XRD line intensity for each mineral system. (compare with figure 3.8 for results expressed as percentages, which allow direct comparison between different mineral groups).

Sample name	Location	Desciption	LOI (%)	XII	ARM	SIRM	Solt	Hard	Xid%	ARM/XII	IRM(+20)/arm	Solt %	Hard %
Sarpenane	Loounon			$10^{-8} \text{ m}^{3} \text{kg}^{-1}$	10 ⁻⁶ Am ² kg ⁻¹		10 ⁻⁶ Am ² kg ⁻¹			10 ² Am ⁻¹	ratio		
			·	13.1	0.560	89.97	2,631	37,167	0.1	0.043	4.698	2.9	30.0
Skiddaw Slate	4	Slate bedrock	1	13.1	0.000	00.07	2.001						
Borrowdale Volcanic Group	28	green slatey tuff (chlorite- rich) bedrock	1	20.5	20.480	470.30	102.363	41.617	3.4	0.999	4.998	21.8	8.8
Ennerdale Granophyre	10	micro-granite bedrock	/	260.6	2963.910	92476.3	28505.4	169.1	0.6	11.370	9.620	30.8	0.2
50	5	upper clay-diamict	26.7	152.9	479.136	9521.20	4635,669	88.021	11.4	3.133	9.675	48.7	0.9
5р	5	(weathered) A-horizon (peaty)	76.7	7.6	5.347	358,13	66.364	Ð.824	1.8	0 .700	12.411	18.5	2.5
65\$	6	stream- sediment	14.6	14.6	6.909	975.19	131.871	43.549	0.1	0.472	19.087	13.5	4.5
7a 7ss	777	A-horizon stream- sediment	14.2 13.8	53.3 48.3	138,492 136,184	4637.174 4449.001	1262.657 1245.691	152.316 155.048	7.4 7.7	2.600 2.822	9.117 9,147	27.2 28.0	3.3 3.5
8a	8	A-horizon woodland soil	65.2	288.9	286.541	47151.408	6269.027	2263.315	2.5	0.992	21.878	13.3	4.8
9a 955	9 9	A-horizon stream- sediment	14.6 15.0	58.2 63.297	85.593 98.416	9084,959 9304.841	1137.066 1310.752	353.088 633.629	3.6 4.1	1,471 1.555	13.285 13.318	12.5 14.1	5.9 6.8
10a 10b	10 10	A-horizon B-horizon	52.6 31.1	162.4 95.4	149.371 91.937	46294.502 31799.817	3732.156 1638.023	2202.667 2422.032	0.674 1.2	0.920 0.964	24.986 17.817	8.1 5.2	4.8 7.6
10u	10	upper clay-diamict (weathered)	27.3	179.9	162.396	61722.290	2099.005	3246.785	0.397	0.903	12.925	3.4	5.3
13a 13b	13 13	A-horizon B∙horizon	36.8 27.6	27.5 34.255	51.427 73.553	3641.615 3583.900	534.850 628.381	329.533 396.198	4.0 4.8	1.868 2.147	10.400 8.543	14.7 17.5	9.0 11.1
14b	14	B-horizon	21.8	47.828	125.262	4194.143	833,123	210.284	6.1	2.619	6.651	19.9	5.0
155	15	B-horizon	17.0	30.122	49.677	2516.744	426.996	194.698	4.0	1.649	8,595	17.0	7.7
16a	16	A-horizon	12.2	30.980	49.533	3214.080	704.231	304.268	1.8	1.599	14.217	21.9	9.5
28u	28	upper clay-diamict	20.7	242.0	493.141	15030.493	2264.530	623.665	8.3	2.038	4.592	15.1	4.1
281	28	(weathered) lower clay-diamict (unweathered)	16.4	76.8	60.530	5453.524	366.441	668.339	4.9	0.129	6.054	6.7	12.3

Appendix 6. Buttermere catchment sediment source sample mineral magnetic characteristics. Key to sample names: prefixed numbers = site number; suffixed letters: a - soil A-horizon; b - soil B-horizon; p - peat; ss - stream sediment; u - diamict, upper (weathered horizon); u - diamict, lower (unweathered horizon).

T		Description	LOI (%)	хн	ARM	SIRM	Soft	Hard	Xid%	ARM/XII	IRM(+20)/arm	Soli %	Hard 🛠
Sample name	Location	Description		$10^{-8} \text{ m}^{3} \text{kg}$	1 10-6	Am ² kg ⁻¹	10 ⁻⁶ A	m ² kg ⁻¹		10 ² Am ⁻¹	ratio		
					0.560	89.97	2 6 3 1	37 167	0.1	0.043	4 698	29	30.0
Skiddaw Slate	3	Ordovician slate bedrock	(13.7	0.300	03.87	2.031	37.167	0.1	0.043	4.000	2.5	50.0
Borrowdale Volcanic Group	28	green slatey (utf. (chlorite- rich) bedrock	,	20.5	20.480	470.30	102.363	41,617	3.4	0.999	4.998	21.8	8.8
Ennerdale Granophyre	10	micro-granite bedrock	1	260.6	2963.910	92476.3	28505.4	169.100	0.6	11.370	9.620	30.8	0.2
10	1	upper ctay-diamict (weathered)	4.5	210.1	581.721	14054.683	5470.3B6	547.080	9.8	2.769	9.404	38.9	3.9
2a	2	A-horizon	32.1	255.0	424.897	30463.656	5444.795	1312.103	5.7	1.667	12.814	17.9	4.3
11a	11	A-horizon	15.5	20.6	40.104	2584.158	457.123	340.974	2.3	1.948	11.398	177	13.2
1155	11	stream-	10.8	40.7	62.196	5134.429	997.299	377.246	2,8	1.528	16.035	19.4	1.3
12u	12	upper clay-diamict (weathered)	6.9	62.7	237.695	6511.990	234.809	318.490	7.9	3.789	0.988	3.6	4.9
17a	17	A-horizon	12.6	79.9	206.603	3561.192	1785.908	64.081	11.0	2.587	8.644	50.1	1.8
184	18	A-horizon	26.0	76.2	156.446	6176.613	1908.110	161.708	8.2	2.052	12.197	30.9	2.6
185	18	B-horizon	16.5	70.1	141.433	3671.557	1812.656	81.235	9.9	2.019	12.816	19,4	2.2
1955	19	stream- sediment	19.6	49.3	45.039	9529.219	772.739	1410.346	1.3	0.914	17.157	8.1	14.8
206	20	B-horizon	10.5	7.8	3.173	194.839	19.249	41.514	1.4	0.406	6.066	99	21.3
210	21	upper clay-diamict	16.8	114.4	224.462	5900.939	1828.028	252.706	9.2	1.962	8.144	31.0	4.6
211	21	(weathered) lower clay-diamict (unweathered)	3.8	21.4	13.111	2698.458	108.205	219.327	0.971	0.614	8.253	4.0	8.1
2235	22	stream- sediment	13.1	55.2	97.133	6642.326	887.662	456.363	2.3	1.759	9.139	13.4	6.9
23a	23	A-horizon	18.4	44.2	144.054	4422.466	689.249	361.007	5.1	3.264	4.785	15.6	8.2
242	24	A-horizon	13.8	34.9	126.115	5336.625	634.210	457.973	3.7	3.612	5.029	11.9	8.6
25a	25	A-horizon	16.1	29.3	83.805	2301.217	361 192	97.114	7.3	2.858	4.310	15 7	4.2
265	26	B-horizon	15.0	40.2	57.054	3096.815	371.778	156.715	5.8	1.418	6.516	12.0	5.1
27a	27	A-horizon	16.5	89.821	213.451	6907.385	1281.218	257.286	8.0	2.376	8.002	18.5	3.7
Fly ash	Fiddler's Ferry coal power station	Fly ash	4.3	2585.5	114.321	465173.757	17497.693	12666.267	2.0	0.431	15.703	3.8	2.7

Appendix 6. Crummock Water catchment sediment source sample (and fly ash sample) mineral magnetic characteristics. Key to sample names: prefixed numbers = site number; suffixed letters: a - soil A-horizon; b - soil B-horizon; p - peat; ss - stream sediment; u - diamict, upper (weathered horizon); u - diamict, lower (unweathered horizon).

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