THE IDENTIFICATION OF EARLY LEAD MINING: ENVIRONMENTAL, ARCHAEOLOGICAL AND HISTORICAL PERSPECTIVES FROM ISLAY, INNER HEBRIDES, SCOTLAND.

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ABSTRACT

This thesis investigates whether lead mining can be detected using palaeoenvironmental data recovered from freshwater loch and marsh sediment. Using radiometric time-frames and geochemical analyses the environmental impact of 18th and 19th century mining on Islay, Inner Hebrides, Scotland, has been investigated. The model of known mining events thus produced has been used to assess previously unrecorded (early) lead mining activity. Previous mining in the area is suggested by 18th century accounts that record the presence of 1,000 "early" workings scattered over the north-east limestone region. While there is little to support the often repeated assertion that lead mining dates back to the Norse Period (*circa* 10-11th centuries) it is clear that it may well have been an established industry prior to the time of the first historical records in the 16th century. In order to use a palaeoenvironmental approach to the question of mining history and its impact, the strategy has been to use integrated loch and catchment units of study. The areas considered are; Loch Finlaggan, Loch Lossit, Loch Bharradail and a control site at Loch Leathann. Soil and sediment geochemical mapping has been used to assess the distribution of lead, zinc and copper within the catchments. Environmental pathways have been identified and influx of lead, zinc and copper to the loch sediment has been determined through the analyses of cores from each loch basin. Archaeological fieldsurvey and the re-examination of the results from mineral prospecting data across the study region provides new evidence on the geographical extent and contaminatory effects of leadmining in this area. This study shows how the effect of lead mining can be identified in the palaeoenvironmental record from *circa* 1367 AD onwards, so mining in Islay does indeed predate the earliest known archaeological and historical records.

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TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

APPENDICES

Appendix 1 Plates

Appendix 2 : Loch Finlaggan and Finlaggan Marsh

Appendix 3: Loch Bharradail

Appendix 4: Loch Lossit

Appendix 5: Loch Leathann

Appendix 6: XRD data

CHAPTER 1: INTRODUCTION

Mining is one of the earliest industries of great cultural importance as the supplier of metal and stone. Mining changed dramatically as craft production gave way to later industrial development. From the earliest times to the present, base metals have been exploited resulting in a long but often fragmentary archaeological record. As well as the immediate workings, mining in general and base-metal extraction in particular has left a legacy within the environmental record.

Great Britain has a long history of base-metal extraction beginning in the prehistoric period. The archaeology of early historic and prehistoric mining is constrained by the problem of dating the field evidence. This evidence is often partly destroyed or obscured by later phases of mineral extraction, while even where evidence survives on the ground, typologies of mine types are notoriously unreliable indicators of their true age. For example, bell-pits are often assigned to the medieval period whilst these and other features such as shallow workings or deeper trials may well date to the Dark Age or prehistoric period. If we are to build up an effective long-term picture of mining in general and lead mining in particular, it is considered necessary to look away from the zone of mineral extraction to nearby depositional environments where sedimentary or geochemical evidence survives. For those reasons this thesis has concentrated on the environmental impact of mining as a proxy record.

Metal mining causes varying levels of environmental pollution, the extent, concentration and scale of which will differ according to the type of metals being extracted. The processes employed in the refinement of ores and the type and duration of mining operations can also play a major part in governing the extent of local or regional contamination. Geochemical investigations around former mining areas provide useful qualitative and quantitative data on the concentration, distribution and the routing of heavy metals along environmental gradients (Davies, 1983; Thornton and Abrahams, 1984 and Alloway, 1990). Such data form the basis of modern impact assessment studies that investigate important environmental issues ranging from past to present pollution trends and modern day environmental hazards.

The calculation of heavy metal flux in lacustrine sediment enables researchers to monitor pollution inventories that may be attributed to one or more point-sources.

As a base metal, lead (Pb) has been widely exploited as a secondary source of extractable silver and is usually found in association with other metals such as copper (Cu) and zinc (Zn). Extensive lead mining has occurred on the island of Islay, where lead mineralization is recorded over 40 km^2 and deposits were accessible either at the near surface or in deep mine workings. The lead field is at the centre of a historically important region once under the control of the Lords of the Isles, maritime rulers of the Western Isles between the 13-15th centuries (Caldwell and Ewart 1993). Archaeologists working at the site of *Eilean Mor* (Castle Finlaggan) at the head of Loch Finlaggan have raised the question of the economic importance of lead following the recovery of lead artefacts from securely dated 13-15th century contexts. If these lead artefacts are manufactured from local lead ores then this indicates lead mining several centuries prior to the earliest currently known documentary evidence. Recognising the significance of galena (lead ore) as a secondary source of extractable silver in one of the most politically important regions in the Western Isles, Islay offers an ideal opportunity to extend a study from known mining activity to earlier periods when mining is probable but yet unknown.

Extending the record of mining activity would be a simple matter if we were able to obtain a sample of lead from a securely dated archaeological context and source its provenance within the landscape. Unfortunately at the present time this is not possible. Recent studies aimed at determining the geographical source of ores used in the manufacture of ancient metal artefacts have relied extensively on Pb isotopic analyses. The technique involves the use of thermal ionization mass spectroscopy (TIMS) to examine the abundance ratios of the isotopes $^{206}Pb/^{204}Pb$, $^{207}Pb/^{206}Pb$,$^{208}Pb/^{206}Pb$ within ancient metal artefacts (Budd *et al* 1995). The use of Pb isotopic data for archaeological provenancing rests on the assumption that Pb isotopes do not fractionate to a significant extent in geological, extraction and manufacturing processes (Gale and Stoss-Gale 1992). Given the assumed stability it was possible that isotopic ratios within lead objects would remain characteristic of ore bodies from which they were originally derived. Unfortunately experimental work and kinetic theory modelling by Budd and co-workers (1995, 145) has shown that metallurgical process involving the removal of lead by evaporation or thermal alteration during the smelting process do seriously alter the isotopic composition of the lead. This being the case, an alternative line of enquiry is adopted using a palaeoecological approach to determine when mining became important in a given region.

Within the study area there are nine freshwater lochs. Six of the lochs have evidence of lead mining within their respective catchments. This geomorphological setting offers the chance to extend the record of mining through the inferred impact on the environmental record.

In this thesis, geochemical inventories will be constructed from sediment cores recovered from four lochs that will be directly related to mining activities within three catchments and provide a control site at a fourth.

1.1 Approach and aims

The aim of this study was to establish criteria for the recognition of early mining from the palaeoenvironmental record by focusing on a case study on Islay. To do this I have adopted an analogue approach; with the key steps being:

- Firstly to determine the evidence for the scale of historically recorded lead mining; its duration and distribution
- Secondly to assess the other sources of heavy metal abundance and their relative distribution within the study area
- Thirdly to identify the possible routes of heavy metals from natural and anthropogenic point-sources into loch sediment
- Further, to review the mechanisms by which heavy metals are retained in near-surface sediment and the degree of post-depositional movement
- Lastly to construct a chronology of heavy metal deposition and assess the reliability of the chosen radiometric dating methods. This is essential to establish the link between the environmental, historical and archaeological record and to identify periods of earlier (undocumented) mining activity.

As a result the aims of this thesis are:

- To consult documentary records and maps in order to obtain an historical perspective of 18th and 19th lead mining activity
- To expand the archaeological record with a detailed archaeological survey in former lead mining areas within the study area
- To establish the heavy metal distribution in catchment soils
- To develop a conceptual model of the environmental pathways to identify the likely sources and routes of lead contamination into soils, freshwater and alluvial sediment
- To identify three loch-catchments affected by mining and one control loch-catchment
- To determine heavy metal inventories and other physical properties in loch and marsh sediment using atomic absorption spectrophotometry and other analytical techniques
- To construct an environmental history using ^{14}C ^{210}Pb ^{137}Cs and other relative dating methods to link mining events with the environmental record.

1.2 The structure of this thesis

Chapter 2 reviews the history of non-ferrous metal mining. Here the discussion develops chronologically drawing on evidence for lead exploitation from the earliest times through to the more industrialised Roman Period and into the Early and Post Medieval Periods. Industrial archaeology has played a central role in developing our understanding of the scale and geography of non-ferrous metal mining in Great Britain. The principal metals extracted were lead, zinc, copper, tin (Sn) and iron (Fe). Gold (Au), silver (Ag) and arsenic (As) were of considerable importance in some areas (Thornton and Webb 1979). Today there is little left of this extractive industry but its legacy is still apparent within the landscape.

The second part of Chapter 2 reviews the implications of mining in relation to environmental gradients and the mechanisms that lead to heavy metal contamination in the terrestrial and lacustrine environment. The section develops thematically with a review of how researchers have developed a historical perspective in order to monitor the rise of pollution from both base-metal mining and from atmospheric sources. In order to place these events within a time-frame, the construction of long or short-term chronologies are assessed.

A fundamental part of any thesis has to be its research design, supported by methods which are selected to test various hypotheses. A systems approach is here adopted and the methods used in this thesis integrate a wide range of laboratory-based and field-survey methods. These are explained in Chapter 3.

Chapter 4 examines the topographical and environmental background of the study area. This chapter reviews the development of lead mining and its archaeological remains in north-east Islay. The latter have been identified to assess what survives in the 'cultural landscape' today. Over the centuries, much of this archaeological evidence has been lost due to 'masking' or deliberate clean-up operations. Clearance of tailings heaps for track building has denuded many features associated with the industry. A brief account is given of other on-going archaeological research and previous geochemical survey in the study area.

Lochs Finlaggan, Bharradail, Lossit and Leathann are discussed in Chapters 5, 6, 7 and 8 respectively. The loch-catchment unit of study is maintained throughout each of the chapters and the results of the findings of both soil and sediment geochemistry are examined accordingly. Chapter 9 will bring together the results obtained from the geochemical analyses of loch sediment and addresses a series of important issues. Firstly we need to establish whether the data obtained from chemical analyses is the result of mining, or of another natural process.

These might include, diagenitic (*in situ*) alteration, chemical remobilisation, bioturbation as a result of sediment mixing by benthic fauna and increased chemical influx from non-mining events occurring in the catchment. The latter might include natural erosion processes, historical land-use, afforestation and deforestation and/or any combination of these factors. Secondly, time frames constructed to support the historical and environmental record will be critically assessed. Thirdly, we have to be sure that the integrity of the cores is sufficient in terms of their 'steady state' formation. We are fortunate that near-surface sediment disturbance can be assessed using the inventories of the radionuclides ^{210}Pb and ^{137}Cs. Finally, Chapter 10 will conclude with a general review of the overall achievements and the wider implications of this thesis.

CHAPTER 2: MINING, METALS AND THE ENVIRONMENT

2.1 Introduction

Since the prehistoric period the search for stone and metals has left its mark in some way or other within the landscape and one could argue that apart from agriculture, our earliest industry was extractive. The physical evidence for early mining is very rare but from the Roman Period we have a slightly clearer picture of the scale of base-metal extraction. The environmental impact from 'ancient' mining and smelting processes has had far-reaching consequences at local, regional and global scales. Base metal mining can have serious implications for the terrestrial and aquatic environment and will often lead to the formation of highly contaminated soils and sediment. In order to assess the scale of contamination in soils and sediment, the heavy metal inventory obtained has to be compared to soils and sediments obtained from areas with little or no contamination. This will then provide a 'benchmark' or 'baseline' level which will help assess whether or not contamination is indeed present either at a low, moderate or critical level. Ecosystems contain many complex environmental pathways in which heavy metals can be routed and these are best understood using descriptive 'flow' diagrams.

This chapter is divided into three sections. Section 2.2 will examine the development of non-ferrous mining in Great Britain. The latter part of this section will discuss the development of the lead industry in Scotland. Although the industry was not geographically as extensive as in England it was nevertheless intensive in at least two regions. Moreover, in Scotland, we have some excellent historical documentation which relates to the earliest phases of metal mining that commenced on a commercial basis in the Early Medieval Period. Historical records allow us to glean information on the mining technology that developed in the Later and Post-Medieval Period. Some of these important technological achievements are placed alongside lead output statistics which show the production trends within the Scottish industry.

Section 2.2 will discuss the presence of trace metals in the terrestrial environment and their routing along different environmental gradients. The lake water-shed ecosystem (Oldfield 1983 citing Likens 1972) provides a spatially bound framework within which aspects of material flux may be isolated and quantified, and their interactions through time studied as they are reflected in the environmental record.

The long history of metal mining in Britain has led to high levels of heavy metal contamination in soils at long-abandoned sites. Often metal-rich solutes from mine tips (commonly termed tailings) will invariably find their way along fluvial pathways resulting in high metal concentrations in both stream, lake and estuarine sediment. High concentrations of trace metals which are above their 'normal' background concentration in soils or sediment may also arise as a result of natural agencies and this phenomenon is discussed in the light of recent research.

The sediments formed in freshwater lochs and in ombrotrophic peat bogs provide a valuable and complementary situation for historical monitoring. Section 2.4 will examine the presence of trace metals in the lacustrine environment. This section will also concentrate on the theme of environmental routing which will explain how pollutants may be incorporated into freshwater sediment. The environmental impact from base metal mining and smelting can often be monitored several kilometres from its origin or 'point source'. The rapid increase of industrialisation since the mid-nineteenth century has been coincident with the rise in European fossil fuel burning (Renburg 1985). Moreover, the increase in atmospheric pollution from leaded petrol has had a profound effect on the global environment. Some aspects of this research are explored briefly in conjunction with the mechanisms that may be attributable to the fixation of heavy metals into freshwater sediment (these subjects are dealt with in greater detail in Chapter 9).

In the final section of Section 2.4, a simplified model is presented to bring together the important environmental issues that are raised in this chapter. Such models are important if we are to understand the principal sources and routes of heavy metal contamination and their influx into soil and sediment in the study area. Starting from a lead ore deposit, environmental gradients are shown passing through several stages (both natural and industrial) to eventually enter into a freshwater loch and marsh sediment.

2.2: Base-metal mining

2.2.1 A brief history of lead mining

The history of lead mining and the development of the industry within the British Isles is fragmentary from the Roman through to the Post Medieval period. Metal mining (mainly copper) in Wales dates back to the Bronze Age, with artefacts having been found at Parys Mountain and the Llandudno Mine on the Great Orme. The earliest evidence so far for the use of metallic lead in Britain and Ireland has come from a Late Bronze Age (LBA) cist cemetery at West Water Reservoir, Peebleshire, (Hunter 1994). Lead has often been described as a 'mystery metal' in British prehistory (Hunter and Davis 1994). Although it was smelted and used for alloying bronzes from the LBA, actual lead artefacts are few and far between. Its presence in the archaeological record in the prehistoric period is restricted to a few lead/tin alloyed objects from the LBA (Needham and Hook 1988) but becomes more common in the Late Iron Age and Roman Period.

Until the Roman era the quantity of lead exploited was on a small scale in comparison to the usage of other metals. However with the annexation of southern Britain into the Roman empire the mineral wealth within the regions bearing galena were rapidly exploited (Figure 2.1). It is probable that lead was mined on a small scale in Britain before the Roman conquest. We can be fairly certain that Caesar's expedition to Britain was prompted by the knowledge of the existence of lead and silver. Strabo writing at this time describes the major exports of Britain as grain, cattle, gold, silver, iron, slaves and hunting dogs. Later the writer Tacitus was to mention gold, silver, and other metals as the 'wages of victory' (*pretum victoriae)* a statement that Salway (Salway 1981, 72) interprets as a form of economic propaganda giving Rome ample justification for the Claudian invasion of Britain.

The exploitation of lead ore on an industrial basis began very soon after the Romans arrived, in AD 43. Two pigs or ingots of lead were found in the Mendips with inscriptions dating them to AD 49. Derbyshire pigs are distinguished by the letters *EX. ARG.*, *LTV* and *LVTVD* which is believed to refer to *Lutudarum* which according to Ford and Rieuwerts (1981, 7) may have been the site of Matlock, or Chesterfield. Equally, the letters could refer to a whole area of a mineral field. Pigs containing the letters *BRIT. EX. ARG.* have fuelled much debate as to their origin. Ford and Rieuwerts (1981, 7) cite several possible translations which include '*ex argento*' meaning "made from silver"; '*ex argenteriis*' "from the silver mines"; or *'ex argentiia officina'* "from the silver refinery".

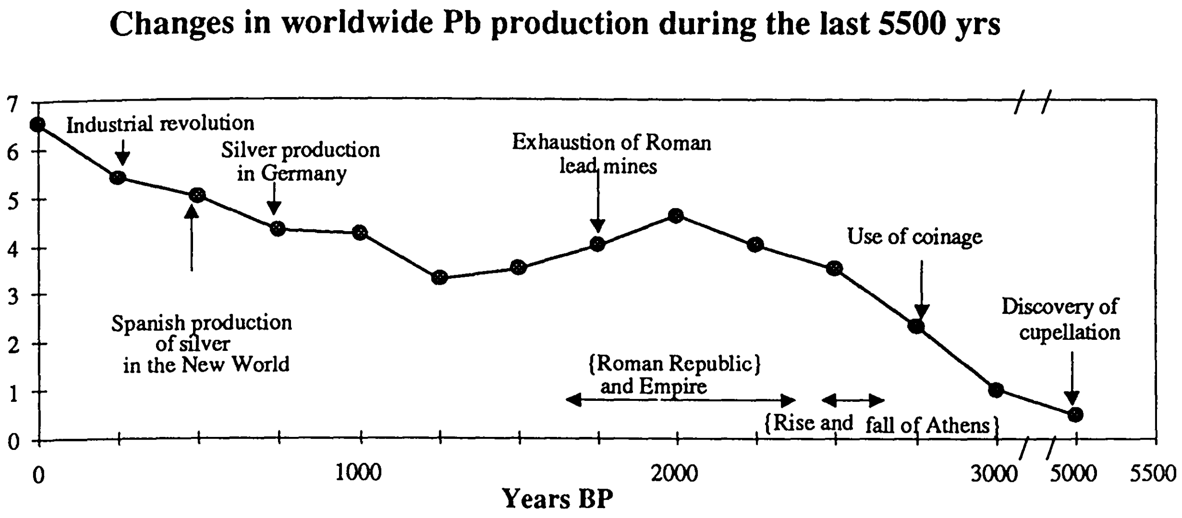

Figure 2.1 Changes in global lead production (after Hong *et al* 1994) Note: 0-7 scale is lead production in K tons yr^{-1}.

The lead mines in England developed rapidly under private contractors (*conductores*) and from the inscriptions on the pigs, it has been possible to identify most of these with specific mining areas and ports associated with their export (Salway 1984). With the establishment of permanent forts and the later development of towns, vast quantities of sheet lead were required for water supply. These included elaborate systems of piping and cisterns which supplied water to the towns and houses throughout the Roman Empire. Domestically, lead was used in a variety of pewter utensils and tableware, ornaments, jewellery, cosmetics and medicinal remedies.

Silver extracted from lead using a technique known as cupellation (Hodges 1989, 94) was vitally important to the imperial government both for coinage and under the Late Empire for official payment in plate and ingots (Salway 1981, 633). The scale of silver production in Britain at this time is not clear. Inscribed silver ingots have been found but Tylecote (1986,70) argues that in view of the low average silver content of British ores compared to the higher silver content of the lead ores of Iberia, Greece and Asia Minor, Britain was probably an importer of silver in the Roman Period.

According to Ford and Rieuwerts (1981, 8) between the end of the Roman Period and the Norman Conquest both the Saxons and the Danes continued mining in the Derbyshire region but it is thought on a much smaller scale than previously. In the 9th century, mines at Wirksworth in Derbyshire were attached to the Abbey of Repton and were evidently of considerable importance because lead worth 300 shillings had to be paid as annual rent to

Christ Church, Canterbury. After the destruction of Repton Abbey in 874 the Manor of Wirksworth passed into the hands of the Danish King Ceolwulf. Danish influence in the region is to be found mainly in place-name evidence. The Alport-by-Youlgreave mining field in Derbyshire is said to be derived from the Saxon *Aldgroove* or *Auldgroove* meaning Old Mine (Ford and Rieuwerts 1981, 81). The Odin Mine near Castleton, Derbyshire, traditionally derives its name from being worked at the time of the Danes. Traditional uses of lead at this period included the manufacture of pewter. Pewter was used widely in the Scandinavian world as a metal for the production of brooches (Graham-Campbell 1980, 106).

Evidence for mining in the Early Medieval Period is to a large extent derived from documentary sources. The Domesday survey of England in 1086 records the existence of at least seven lead works (*plumbaria*) in the Derbyshire area but few elsewhere (Tylecote 1986). It is not clear if the term refers to smelters or mines. By the twelfth century mines were worked extensively in the Bakewell region (Ford and Rieuwerts 1981, 9). In 1288 a set of mining laws were set down and overseen by a Barmaster[1] who presided over the Barmoote or leadminers' court. Such courts were held regularly well into the early 18th century in Yorkshire and Derbyshire (Gill 1993). Before this period unwritten codes of mining practice or customary laws regulating mining practices may have been a tradition that originated in the Anglo-Saxon Period.

With the rise of monasticism across the British Isles, the demand for lead stimulated further development within the industry. Mining resumed in Wales during the 12th and 13th centuries, after its conquest by Edward I, when lead was needed for roofing castles and churches. Silver was mined for coin manufacture during the reign of Elizabeth I, from argentiferous lead ores in Cardiganshire (Pearce 1993). The 16th century marks the introduction from Germany of water powered air-blast smelting furnaces. During the 17th century, lead mining became widespread and mines in north east Wales rose in importance. Naturally, more evidence survives from the Industrial Period of the 18th and 19th centuries where mining statistics and the physical remains of lead and copper mining operations are usually more extensive. In 1768, the 'Great Lode' was discovered at Parys Mountain, on Anglesey and produced 3,000 tons of copper. Together with the adjacent Mona Mine, Anglesey, this region dominated world copper production from 1770-1790 (Pearce 1993). The growth of mining during the 18th Century made Britain the world's major lead and copper exporter. After the Napoleonic Wars, mining in Europe developed rapidly but the British industry was slow to recover. Lead prices fell sharply in the 1870s as cheaper lead entered the

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[1] Barmaster was a Crown official who dealt with all lead mining enquiries and customs.

market from overseas. The discovery of large ore deposits in Australia and the USA lowered the international price of lead still further. Cheap imports flooded the British market and the home market declined rapidly. However the silver that could be extracted from lead ore kept many mining concerns operational even though the price of lead had by this time fallen (Davies, pers.comm.).

An extensive corpus of historical data exists on the major lead producing regions of England and Wales during the 17th and 18th centuries. The main concentrations of mines in England were to be found in North Yorkshire, centred around the towns of Alston (Raistrick & Jennings 1965) and Grassington (Gill 1993); in the Derbyshire Peak District mines were centred on Matlock and Castleton (Ford and Rieuwerts 1981); and in the Mendips around Charterhouse (Gough 1967). In Wales the industry was widely dispersed throughout Flintshire and Cardiganshire (Morrison 1971) and in the Clwyd[2] region (Frost 1994).

2.2.2 The history of lead mining in Scotland

The earliest use of lead in the archaeological record is from a recent Early Bronze Age (EBA) cist burial at West Water Reservoir near West Linton in West Lothian. The recovery of a unique two-strand necklace comprising a string of cannel coal and a strand of lead beads provides the earliest evidence to date for the use of metallic lead in Britain and Ireland (Hunter 1993; Hunter and Davis 1994). While the West Water necklace clearly shows at least sporadic use of lead in EBA, nothing is known of the methods of ore extraction or processing during this period. Indeed there is currently no archaeological evidence for the organised mining of metalliferous ores in scottish prehistory. It can only be assumed that much of the galena exploited in this period was recovered from alluvial deposits rather than from mined sources. The archaeological record remains mute regarding evidence of mining in Scotland until the medieval period.

A synthesis of mining records from Scotland entitled *'Early Records relating to Mining in Scotland'* was published by R.W. Cochran-Patrick in 1878. The author has brought together many of the early historical accounts of mining housed in the archive at West Register House, Edinburgh. Part of the following discussion is based on extracts from this important publication. For more detailed statistical accounts of production and ownership of Scottish metalliferous mines in the 19th century, the reader is referred to a synthesis of the *Mineral Statistics 1845-1913*, and the *Annual Reports of the Inspectors of Mines* by Burt *et al* (1981).

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[2] Clwyd incorporates the old counties of Denbighshire, Flintshire and parts of Merioneth.

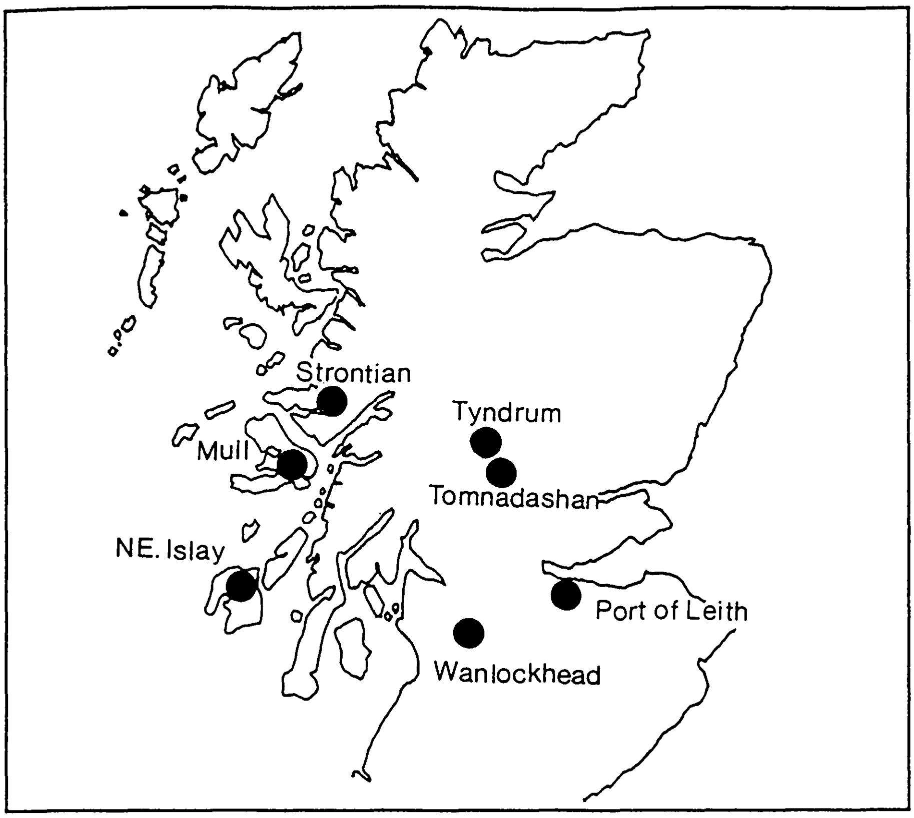

Figure 2.2 Location map of the Scottish lead fields discussed in the text

Mining has been recorded in Scotland since the Middle Ages. The first historical notice relates to gold and is mentioned in a grant by David I to the Abbey of Dunfermline in 1153. Monastic involvement with gold mining was well known in this period and developed alongside the procurement of other metals and minerals such as lead ore and coal. In 1239 the monks of Newbattle Abbey were given a grant to mine lead on Crawford Moor and in 1264 the Exchequer Rolls show a payment by the Sheriff of Lanarkshire for the carriage of lead from Crawford Moor to Rutherglen. Towards the post-medieval period the exploitation of base-metals was rapid and developed with the advent of new technologies. R.W. Cochran-Patrick cites from a letter that is dated from 1604 to the Earl of Suffolk, Lord Chamberlain on the progress of mining in the Wanlockhead region:

> " I began my works with dames whereby I meane to teare ye earth and discover such vains of mettall"

This account refers to the practice known as hushing, whereby water was allowed to wash away surface deposits in an attempt to reveal sub-surface mineral deposits. This may well be one of the earliest accounts of hydraulic mining in Scotland. In certain parts of the Yorkshire Lead field this practice that was later to bring the mine owners into disrepute for damage to properties caused by flooding and the alteration of water courses. For the same period, 1604, the writer also refers to draining mines with a system of adits which again appears to be a technique employed at an early period in mining history in Scotland.

2.2.3 The Post Medieval Period

Scotland's trading links with overseas ports is reflected in a reference from the document *Analecta Scotia* that records that between 1585 and 1590 15,717 stone weight of lead was shipped for foreign use from the Port of Leith. The Port of Leith appears to have been the main route to the principal markets in the Low Countries, especially as early as 1719. An interesting account from *Henry Kalmeter's Travels In Scotland, 1719-1720* (Smout 1978, 29) mentions:

> In Rotterdam there is a company of merchants who, by a contract with My Lord Hopetoun ship out all this lead ore at their own risk and expense from Lith (Port of Leith) where it is ground up very fine, and used in the manufacture of all sorts of porcelain vessels and for that reason it is called 'Potter's Ore'

In industrial terms, lead mining in Scotland became established around 1690 and continued, sporadically, until the early 19th century The villages of Leadhills and Wanlockhead[3] (Figure 2.2) in Lanarkshire are important in the history of lead mining in Scotland (Downs-Rose and Harvey 1979a & 1979b and Smout, 1979). From 1845-1913 a total of 211,849 tons of lead ore were sold, yielding over 800,000 ozs of silver (Burt *et al* 1981, 9). Nearly all of this was derived from the Lowther Hills area of Lanarkshire/Dumfries border. The Leadhills and Wanlockhead mines (total production 190, 333 tons of ore) accounted for 90% of Scotland's total lead output. In the second half of the 19th century the Scottish lead industry accounted for 9-10% of the UK total lead production. With the exception of the lead mines of Leadhills, Wanlockhead and Islay (total lead production between 1862-1896 for Mulreesh Mine was 1,513 tons; see Figure 4.3) the lead workings at other locations in Scotland (Figure 2.2) were relatively small and short lived, such as the mines at Tyndrum and at Tomnadashan, alongside the shore of Loch Tay Perthshire (Bainbridge 1970). Strontium, in Argyllshire, gave its name to the element Strontium, first discovered here in 1764. There are many small lead mine sites scattered across Galloway and Argyll especially around Loch Fyne and on the island of Mull, Inner Hebrides. A gazetteer listing most of the Post Medieval metal mines of Scotland has been produced by Landless (1993).

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[3] The open-air museum at Wanlockhead includes a unique bellows and smelter house that has been investigated by archaeologists and is now reconstructed in parts. Nearby a beam engine from the Straight Steps Mine has been preserved and is the only complete water-powered beam engine left in Scotland.

Although relative to the rest of the UK, lead output was small (never rising above 10% of the total UK production) nevertheless its value to the Scottish economy was significant, firstly owing to improved methods in the refinement of silver and the demand for 'potters ore' and sheet lead, in the Low Countries and home market.

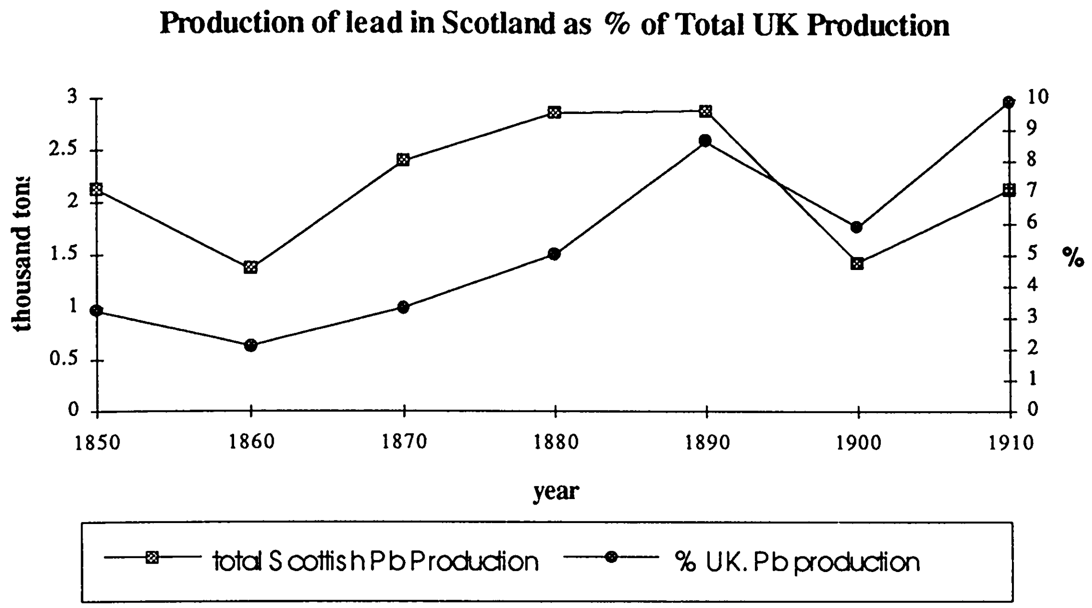

Figure 2.3 (after Burt *et al* 1981) showing the total lead produced and its relation to the net UK net total production.

2.2.4 A brief review of lead mining, processing and smelting techniques

In the Roman Period excavations were carried out with picks, hammers and wedges and when the rocks proved too hard, by 'fire-setting'. This involved lighting fires against the face, and while still hot, dousing it with cold water, the rapid temperature change causing fracturing of the rock. As mentioned previously, a common method of prospecting for lead ore was the removal of surface soil and peat by the method that later became known as 'hushing' (Ford and Rieuwerts 1981). The method involved the damming of rivers or burns and flooding the land to remove topsoil above potential ore-bearing deposits. In the 18th centuries this brought the mine owners into conflict with adjacent land owners due to the rearrangement of water courses and flood damage in the lower valley bottoms.

By the 18th century land for mining generally was offered by landowners to leaseholders on a 36 year tenure. The immediate ore bearing strata was termed a "bargain", and was usually worked on a piece-work basis according to the amount of lead ore smelted. A standard ingot weight was called a bing.

Where the lead ore outcropped close to the surface (hence less labour input) the price per bing was taken into account, thus lowering the amount that could be earned. Surface deposits in most cases were worked first, simply due to the ease of ore extraction. Shallow twisting workings termed "trials" or "rakes" are typical of landscapes that have been mined for lead and resulted from the miners following the strike of the near-surface ore deposits. In many cases, large areas of non-ore producing strata had to be cleared, particularly where drainage shafts (adits) were required. In Derbyshire this work was termed "dead-work": for this, the miners received no or limited pay. Hunt (1970) claims that, in Yorkshire, this work was often sub-contracted to other workers whose speciality was solely the removal of non-ore bearing deposits and the construction of drainage adits.

Until recent times, lead was mined using hand pick and shovel. Lead has rarely been extracted from depths comparable with those of coal seams. However as with the early coal industry, drainage problems were encountered, usually calling for elaborate systems of water management that commonly diverting streams to power water wheels, which in turn could lift water from the mines. A later and significant 19th century development in drainage was the use of steam driven beam engines to pump water from the mines. The upstanding beam engine house at Mulreesh (Plate 6, Appendix 1) and the previously mentioned water-powered beam engine at Wanlockhead are unique survivors of the technology employed in leadmine drainage.

2.2.5 Draining and ventilating the mines

Drainage and mine ventilation were considerable obstacles to the driving of long levels and the sinking of deep workings. Before the 17th century various types of pump were in use. One of the earliest methods employed was that of winding water in leather buckets by means of a windlass. An improvement on this method was the so called 'chain pump' (Ford and Rieuwerts 1981,14). This method involved a long endless chain containing a series of leather buckets which ran through a long barrel of wood and iron hoops. A later development, but on the same design, was the 'Rag and Chain' pump. In this pump a series of rags and leather plugs drew water upwards as the chain ascended through the barrel. Around the 17th century drainage adits or 'soughs' were usually driven from a lower contour or from the base of a lower valley. These effectively lowered the water table to new workable limits. In many cases, the drainage water was channelled down-slope from the mine shafts (termed shaft and adit mining) via drainage leat systems (Plate 12, Appendix 1) or commonly into wooden troughs (termed over-shots) that often powered water wheels, providing power for ancillary devices connected with ore processing.

2.2.6 Ore Preparation

Lead ore was usually crushed by hand ("dressing") or mechanically by crushing mills. The ore had to be graded, a process which required the galena to be separated from its host rock ("tails"). The oldest separation technology is gravity separation in water. In the traditional process the fine-ground ore/rock mixture was placed in a compartmented box called the "jig". The compartments were separated by wire mesh and the vertical movement of water caused the heavy ore minerals to travel to the bottom leaving the waste behind. The middle compartment contained the "middlings", a mix of ore and rock which was processed by "buddling" in which water flowing down a gently inclining surface swept away the lighter rock leaving behind the ore. At the final stage the ore termed "bouse" was removed for smelting.

2.2.7 The lead smelting process

With a melting point of 327° C lead could easily be smelted in a domestic fire. The survival of Roman and Medieval lead smelting hearths in the archaeological record is limited due to their poor chance of survival. A lead smelting hearth dated to the first century (*c.* AD.76) has been recovered from Pentre Ffwrndan near Flint (Tylecote 1986). Lead smelting in its earliest form involved the cutting of a shallow pit commonly termed a 'Bole' (Ford and Rieuwerts 1982, 19). In its simplest form a pit was dug and surrounded by a low wall. A void allowed prevailing winds or bellows to maintain a blast. Galena was placed in the bottom of the pit intermixed with peat or wood as a fuel. When a sufficient temperature had been achieved the lead would melt out through a channel cut in the base of the pit and residual slag could be remelted for further refinement using charcoal.

Technological achievements in the smelting process developed rapidly with the introduction of water-blown blast furnaces, a technology that developed in Germany[4]. Many of the ensuing refinements came from the iron industry that developed in tandem with the lead industry. The technologies employed would vary in different regions according to local situations and the level of economic investment. For example the Pates Knowes smelt mill is a rare survivor from the 18th century at the Wanlockhead Mining Museum. A set of bellows driven by a water wheel supplied sufficient blast to two hearths (known as Scotch ore hearths) and produced on average 1,000 tons of smelted lead per year (Harvey *et al* 1980). The conventional Scotch ore hearth was a critical component of the lead industry and a description by Downs-Rose *et al* (1979a) is worthy of mention:

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[4] The reader is refered to Agricola's, *'De Re Metallica'* 1556 which is an encyclopedia of mining technology in 16th century Germany. The significance of this work lies in the detailed engravings showing the variety of mining equipment and processing techniques used in German mines in the 16th century.

> "The conventional Scotch ore hearth had a bottom which consisted of a heavy cast iron sump from six to twelve inches in depth which was kept full of molten lead during the smelting process, on top of which floated the fire of coal and peat. The fire had to be 'watched', that is, supplied with fuel, ore and lime, and the mixture stirred from time to time with the gavelock, or poker, and the partly smelted ore raked forward onto the work stone where it could absorb oxygen before being returned to the fire. Molten metal overflowed from the sump, trickled down the sloping work stone and was collected in the sumpter pot whence it was ladled into moulds and cast into bars."

A contemporary account of this operation is mentioned in *Henry Kalmeter's Travels In Scotland, 1719-1720* (Smout 1978, 30-32) and describes in great detail the primary smelting process and the subsequent re-heating of waste slag in the 'slag hearth' using coke to obtain the necessary heat required for further refinement.

The 19th century saw the development of the so called 'Cupola' or reverberation furnace that was commonly in use in the larger lead fields of Yorkshire and Derbyshire. In this type of furnace the fuel and coal was burnt in a grate that was separated by a small wall or bridge from a saucer-shaped hearth (termed sumpter-pot) in which the lead ore or 'charge' was placed, so as to avoid contamination. Flames from the fire 'reverberated' from the low arched roof of the furnace, causing the lead to separate from the waste or slag, and then passed through a flue to a tall chimney that provided the draught, (Ford and Rieuwerts 1981,118 and Gill, 1993, 126). Such furnaces demanded large stocks of coal and were established near good supply routes and away from population as their hazards to human health became more widely known. Not all the lead in the concentrate was recovered in the furnace. Some of it became mixed with the slags, and some went up the chimney through volatilisation.

Lead smelting mills were bad neighbours and the mills often caused serious contamination to soils and vegetation (commonly giving rise to the term of "belland" or poisoned land) In 1781 certain smelt mills at Grassington in N.Yorkshire were charged with being responsible for poisoning cattle and mitigation procedures were common (Gill 1993, 129). The Middleton Dale smelt mill in Derbyshire (1770s) circumvented this problem by constructing a horizontal flue to a remote chimney on the side of a hill. The owners achieved good success as far as avoiding the pollution of the immediate locality and were also able reclaim lead-dust that had settled out along the flue (Gill 1993,130).

2.2.8 Summary

This section has outlined the development of the lead mining industry in Great Britain with particular reference to Scotland. It has been argued that base metal mining was second to agriculture in terms of environmental impact amongst our earliest industries. The prehistoric evidence for mining tends to be artefactual and rather indirect. For the Roman period tools and ingots survive in greater numbers and the regional ore fields that were exploited in England are widely known. The Dark Age period is rather mute on mining evidence but the amount of metal work from the material culture spanning this period attests to the exploitation of a variety of metal ores. For the Early Middle Ages we have the written record, mostly originating from monastic and fiscal accounts. By the Industrial Period there is a wealth of documentary and archaeological evidence still surviving at some of the major ore fields within England and Scotland. The long history of base metal mining in Great Britain has led to large tracts of contaminated land, and many long-abandoned mines are still a major source of pollution. The processes and route ways by which heavy metals enter the wider environment from both natural and man made sources and the consequence of environmental impact is now considered.

2.3: Heavy metals in the terrestrial environment

2.3.1 Heavy Metals in soils

Over the last 200 years following the start of industrialisation huge changes in the global budget of 'critical elements' at the earth's surface have occurred (Figure 2.4 and Table 2.1 below). These have begun to modify the regulatory systems which have evolved over millions of years (Wood and Wang 1983). It is generally conceded that mining and metallurgy can cause soil pollution, but it is not always easy to define a pollution level and the cut-off grade at which metal concentrations constitute a man-made contamination of the soils. Thornton (1979, 159) maintains that the total trace element content of soil reflects to varying degrees that of the parent material. Both bedrock or transported overburden can be modified by soil-forming processes such as podsolization, gleying, leaching and organic matter accumulation. Other important factors such as soil pH, drainage regime and redox potential can influence the formation and relative mobility of trace elements in soils. According to the parent materials' geochemical history, the soil will have inherited a stock of elements which are then redistributed as a result of pedogenesis.

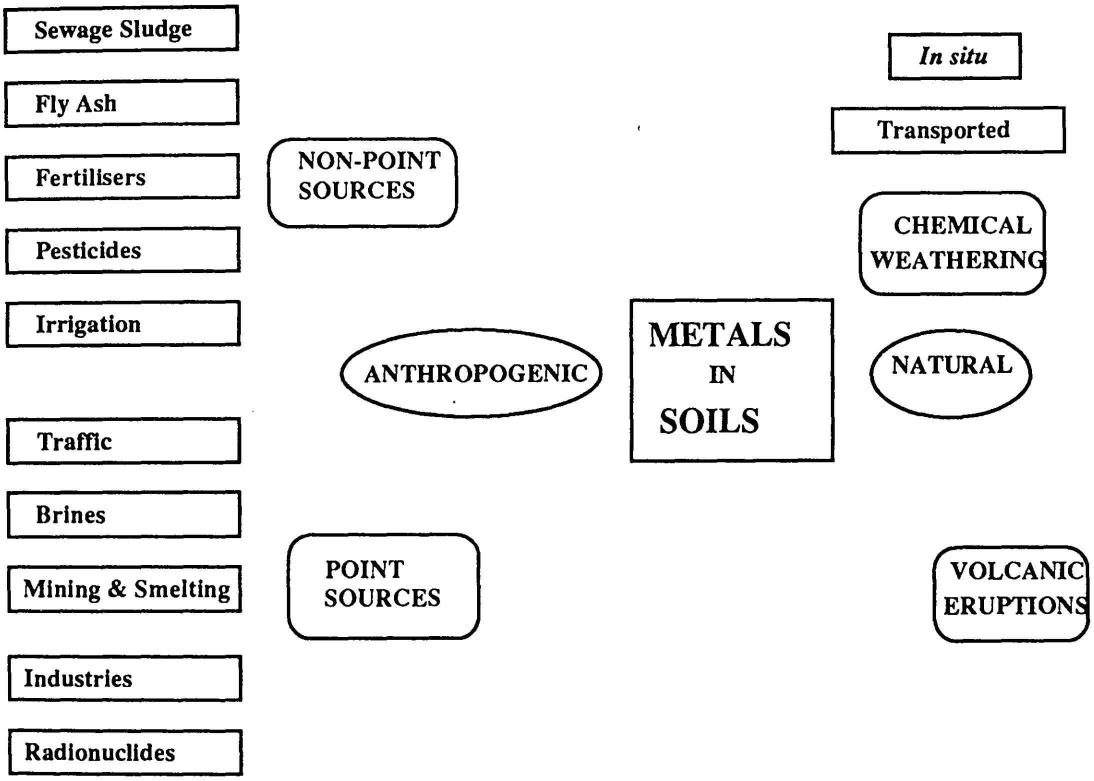

Figure 2.4. Diverse sources of anthropogenic and natural inputs of metals into soils, after Plant and Raiswell, 1983.

Ten major elements, (O, Si, Al, Fe, Ca, Na, K, Mg, Ti and P) account for over 99% of the total element content of the earth's crust (Alloway 1990). The remaining elements from the periodic table are referred to as 'trace elements'. In current literature metals such as lead, zinc and copper are commonly referred to as trace metals. The concentrations of individual trace metals are normally less than 1000 mg kg^{-1} (0.1%), but average content is rarely above 100 mg kg^{-1} (0.01%) (Davies 1983).

Metallic elements having densities exceeding 5kg dm^3 are called heavy metals (Cottenie and Verloo 1984). A limited number are essential for plants and animals (e.g. Fe, Mn, Zn, Mo and Co). If they are not present and available in the soil at a minimal level, deficiency will occur, but excessive amounts could be considered toxic. Lead and mercury are examples of nonessential trace metals. Wood (1974) has classified elements into three groups: (1) non critical, (2) toxic but very insoluble or very rare, and (3) very toxic and relatively accessible (Table 2.1 below).

(1) Non critical			**(2) Toxic but very insoluble or very rare**		**(3) Very toxic and relatively accessible[2]**		
Na	C	F	Ti	Ga	**Be**	**As**	Au
K	P	Li	Hf	La	Co	**Se**	**Hg**
Mg	Fe	Rb	Zr	Os	**Ni**	Te	Tl
Ca	S	Sr	W	Rh	Cu	Pd	**Pb**
H	(Al)[1]	Cl	Nb	Ir	**Zn**	**Ag**	**Sb**
O	Br	Si	Ta	Ru	Sn	**Cd**	Bi
	N		Re	Ba		**Cr**	Pt

Table 2.1 Classification of elements according to toxicity and availability (Wood 1974).

(1) Aluminium is toxic for aquatic and terrestrial biota when mobilised at low pH values.

(2) Elements in bold type are United States Environmental Protection Agency priority pollutants.

The cycling and movement of metals through the environment can best be understood by using a conceptual model of a simple biogeochemical cycle. A general review of the biogeochemical cycle has been produced by Ross (1994) and King (1995) and more detailed studies of heavy metals in the environment are provided by Salomons and Forstner (1984), Pacyna (1986, 1987); Nriagu *et al* (1988) and Nriagu (1988, 1990).

A biogeochemical cycle is the way in which all elements are naturally recycled through the environment. The principal components of the cycle are:

- *Source of chemical elements*:

The source determines how elements are physically and chemically distributed and to what extent they are available within the cycle

- *Transport mechanisms*:

Commonly these mechanisms include water, wind, gravity and living organisms and are heavily influenced by climate

- *Site and fate of deposition*:

The elements may be deposited as part of the sediment in a reservoir or they may be taken up by a plant and perhaps enter the food chain. Not all deposition sites are permanent, and chemical elements can be remobilized if physical or chemical conditions change.

It was mentioned earlier that all soils contain varying degrees of trace metals. Most metals are released by the breakdown of primary ore and rock-forming minerals that occur in the soil either as secondary minerals or as firmly bonded components in clays and hydrous oxides (Bridges 1970). A relatively small part of the total amount of metal derived from the weathering of parent rock or ore may be held as readily extractable components of organic matter or as ions adsorbed on clay particles (Mitchell and Burridge 1979,18). Firmly held metals in clay minerals and hydrous oxides account for most residual metal in areas where unstable ore minerals are oxidizing in freely drained leached environments. Organic matter may contain appreciable amounts of metal and the greater part of the metals will have usually been introduced by ground and stream water and decaying vegetation. Bonding mechanisms are extremely variable and complex. These may range from simple adsorbed ions to metal-organic complexes with metal incorporated into the structure of living organisms.

2.3.2 Naturally occurring geochemical anomalies

Even before any mining activities are undertaken, high metal concentrations in relation to mineralisation exist in soils. Laville-Timsit (1991) stated that:

> "Pollution can only be defined in relation to its natural environment, and this requires, at the very least, a good knowledge of regional geochemical backgrounds"

Unmined Pb-Zn ore bodies occurring near the soil surface will invariably lead to naturally high surface concentrations of these metals as a result of soil weathering processes. Exceedingly high concentrations of a given metal above average concentration in uncontaminated soils are termed anomalous. A 'geochemical anomaly' has been defined as a "deviation from the norm" Hawkes and Webb (1962). More specifically, a geochemical anomaly is a departure from the 'background' value that has been established in the region of study. This is usually a mean or 'control' value taken to reflect no contamination from ore deposits (King 1995). As an ore is weathered, the ore minerals will tend to be dispersed leading to a geochemical pattern at the surface of the soil.

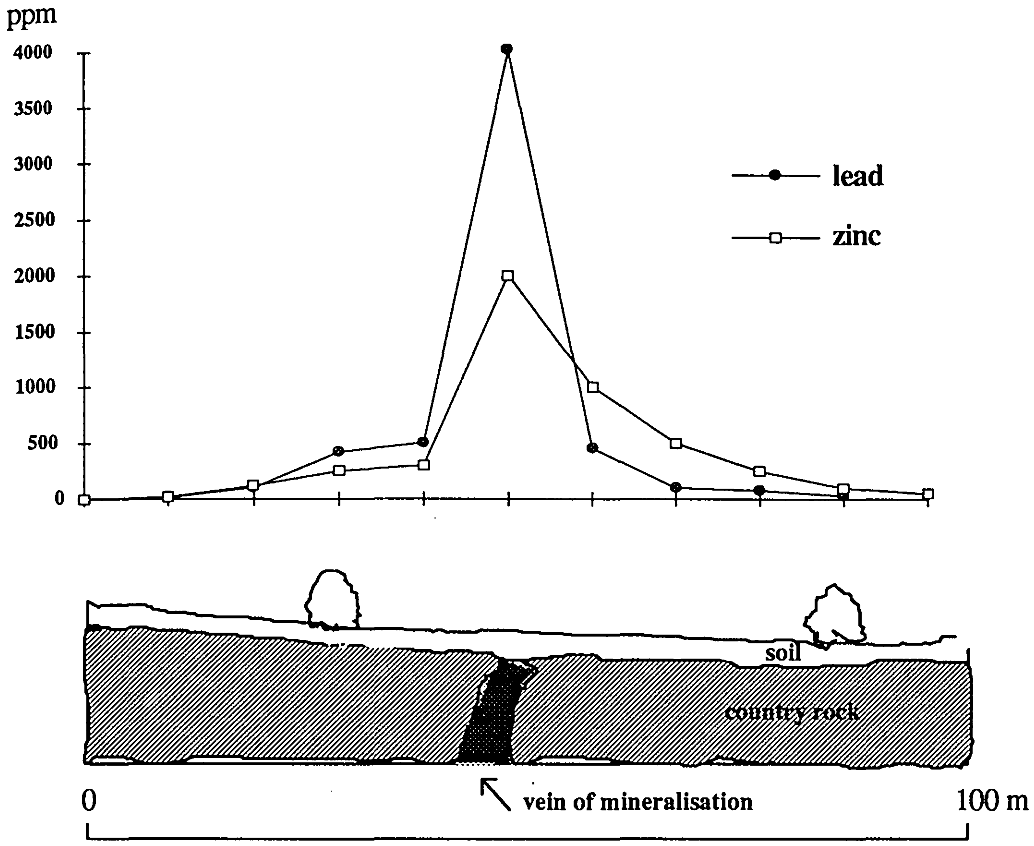

Figure 2.5 Relationship between residual soil anomaly and Pb-Zn vein at Porter's Grove, Wisconsin after Hawkes (1962).

The anomaly is often substantially wider than the ore bearing strata in the underlying bedrock. In general, immobile elements such as Pb tend to give less dispersed anomalies than do the more mobile elements such as Zn. Super adjacent patterns more or less symmetrically dispersed about the source are termed halos (Hawkes and Webb 1962). On sloping ground, however, anomalies generally tend to fan out asymmetrically in the downslope direction.

Downslope distortion is usually the effect of physical movement of metal-bearing particles by soil creep aided by solution and precipitation from laterally moving subsurface waters (Hawkes and Webb 1962). Figure 2.5 above shows the typical concentration/dispersal patterns in the soil derived above a Pb-Zn ore body. Lead tends to be reasonably immobile and will be concentrated in a halo whereas the more soluble Zn has usually dispersed further from the ore body leading to asymmetrical patterns in the surface soils (see Chapter 5, Figures 5.10 and 5.11 as an example).

The formation of a geochemical anomaly within soils and sediments is dependent upon the retention of heavy metals by inorganic and organic components or both. Several factors, including pH, redox potential, inorganic mineralogy and humus content will effect the retention mechanism. The role of humic and fulvic acids and their effect in attracting trace metals has been the subject of analyses, mainly in laboratory based experiments. Ferguson and Bubela (1974) conducted experiments designed to test the hypothesis that humic matter was a regular source of metal-fixation. Using aqueous solutions of Cu (II) Pb (II) and Zn (II) interacting with organic matter recovered from algal specimens, they were able to show that under suitable conditions, a significant proportion of the metal is removed from solution by sorption on to the particulate organic matter within the algal suspension. The work undertaken by Ulrich Forstner (1977 and 1989) on the study of metal remobilisation in waters and sediment from rivers in Germany illustrates the complexities of diagenesis within this material. Forstner's work showed that humic and fulvic acids played a significant role in trace metal uptake. Calcareous and hydrolytic precipitates of iron and manganese oxides were an additional source for trace-metal fixation. Aspects such as the parent geology and ground water retention, solubility, slope and general weathering environment will have a bearing on the rates of metal dispersal. Chapter 4 further explores some of these points in relation to metal dispersal in old mining areas within NE Islay.

2.3.3 Heavy metal concentrations in 'old mining' areas

Human activity has influenced soil development since earliest times and the identification of a 'natural' soil is probably impossible. The global contaminatory effects from prehistoric and Roman metal processing are now seen to be much greater than was previously thought (Hong *et al* 1994). Throughout the Medieval Period and later during the Industrial Revolution, trace metal contamination in soils increased in conjunction with increased output from mining and smelting operations (Martin *et al* 1979; Davies *et al* 1974 and Davies 1983). In more recent times the dispersal of inorganic contaminants is an inevitable consequence of industrial and urban development. Such contaminants travel the same environmental route-ways and suffer the same environmental fate as similar trace materials released by weathering and other natural processes.

Principal base metal	**Guest constituent > %**	**Guest constituent < %**
Galena (PbS)	Ag As Bi Sb Se	Zn Cu Cd Fe Hg Mn Ni Sn Ti
Sphallerite (ZnS)	Ag As Cd Cu Fe Hg Mn Sb Sn	Ba Co Cr Ca Ge Mo Ni Tl V
Chalcopyrite (CuFeS2)	Mn	Ag As Bi Cd Co Cr In Mo Pb Se Sn Ti

Table 2.2 Common trace constituents of sulphide ore minerals. Some of these constituents occur up to several percent of the host mineral where others are only present in smaller concentrations. (after Davies 1983,429).

As discussed earlier in section 2.1.2, mining and metal processing are one of the primary anthropogenic sources of trace metals in the environment. Mining is second to agriculture as the world's oldest and most important industry (Down and Stock 1977). The mining and processing of base metals is one of the oldest ways that humans have increased the concentration of trace metals in the environment. Very high levels of potentially toxic elements such as As, Cd, Cu, F, Pb and Zn are frequently associated with areas of metalliferous mineralisation and old mining activity. Table 2.2 lists the relative order in concentration of the guest constituents that are commonly found in ore deposits (termed gangue) from which a metal or metals have not been extracted.

2.3.4 Regional trends in soil contamination

The problems of long abandoned mines as a source of pollution continue to pose significant environmental risks to their immediate surroundings (Younger 1994). In the case of lead mining, long after mines have ceased to be commercially productive, continual erosion and infiltration of acid mine leachates from mine spoil leads to phenomenally high concentrations of Pb, Zn and Cu.

The publication of the Wolfson Geochemical Atlas in 1978 highlighted the localised environmental impact of old mining activity throughout England and Wales (Webb *et al* 1978). The atlas was produced by the Imperial College Geochemistry Research Group who analysed about 50,000 stream sediment samples from within England and Wales.

The results showed several areas to contain potentially toxic concentrations of trace metals and these are summarised in Table 2.3 below.

Region	**Mineral type**
South West England	Cu-Sn-As-V
Mendips	Pb-Zn
Central and north Wales	Cu -Pb-Zn
Lake District	Pb-Zn- F
Pennines	Pb-Zn-F
Peak District	Pb-Zn-F

Table 2.3 Areas of mineralisation associated with potentially toxic trace element levels in Britain after Howarth and Thornton (1983).

The results of chemical analyses from mine spoil around the United Kingdom are summarised in Table 2.4 and show the relative geographical trends across the country. In Britain, the past disposal of metal rich waste minerals has been shown by previous studies to be one of the primary sources of present day trace metal pollution of terrestrial and aquatic ecosystems (Davies and Roberts 1975, 1978, Wolfenden and Lewin 1978, Davies and White 1981, Davies, 1983, Lewin and Macklin 1987).

Mining region	**County**	**Sites**	**Lead**	**Zinc**	**Copper**
S.W. England	Devon & Cornwall	16	48-2,070	26-1090	65-6,140
W.& N.W. England	Shropshire & Cheshire	12	840-26,000	980-21,000	15-7,260
N. Pennines	N.Yorkshire & Cheshire	8	605-13,000	470-28,000	29-140
S. Pennines	Derbyshire	17	10,800-76,500	12,700-28,000	23-97
Lake District	Cumbria	7	2,070-7,630	4,690-7,370	77-3,800
Mid Wales	Powys & Dyfed	10	1,670-54,000	475-8,000	67-195
N. Wales	Clwyd & Gwynedd	19	6,400-76,000	113,000-127,00	30-5,750
S. Scotland	Dumfries & Galloway	6	4,730-28,300	16,00-31,400	125-657
N.E. Islay*	**Argyll**	**5**	**15-5,000**	**12-2,280**	**2-142**
Agricultural soil	General	n/a	2-200	10-300	2-100

Table 2.4 Copper, lead and zinc in spoil from abandoned mines in the United Kingdom after (Johnson *et al* 1994) and (*) after (Cazalet 1974). All values are in mg kg^{-1} air dried substrate.

In contrast with modern mining technology, the methods used for ore recovery by Britain's historical metal mines were a great deal more inefficient (Davies 1983). It is estimated for 19th century mining in Wales between 15 to 38 tonnes of waste rock was produced during the concentration of one tonne of lead ore (Jones 1922, in Davies 1983). For example, in the Peak District the method of milling was so inefficient that miners would rework the waste material to recover ore that had been missed (Ford and Rieuwerts 1981).

Davies and Lewin (1974), Lewin *et al* (1977), Wolfenden and Lewin (1977) and Macklin (1985, 1989, 1992 and 1994) have studied heavy metals in soils and sediments both in river valley cross-sections and in dated sediments in developing meandering loops. The degree of slope was seen as a major factor in the dispersal of heavy metals in the vicinity of old mining sites. Exposed fluvial sediments weather to form soils and invariably contain significant levels of heavy metals that are widely dispersed across valley floors.

Location	**Lead**	**Zinc**	**Copper**	**Cadmium**
Slope soils				
Mine area	33-1680 (270)	20-1900 (193)	5-25 (16)	0.6-2.1 (1.2)
Non mining	17-57(30)	35-157 (82)	3-5 (10)	0.6-20 (1.5)
Alluvial soils				
Mine area	90-2900 (1420)	95-800 (455)	17-42 (30)	______
Non-mining	24-56 (42)	88-171 (129)	4-31 (19)	______

Table 2.5 Relative values in mg kg^{-1} for soils in Cardigion, West Wales recorded from slope and alluvial soils. The ores that were mined in this region were predominantly Pb and Zn (after Davies 1983). (values in parenthesis = the mean in mg kg^{-1}).

The data in Table 2.5 illustrates how soils in West Wales differ in metal content according to topography and liability to contamination. In contrast to present day mining operations there were no regulations governing the disposal of solid and liquid wastes. The mine operators were free to dispose of the waste in whatever way they wanted. As a result of mining and milling the waste water was highly charged with sand, silt-sized ore particles and metal solutes. After the removal of the valuable ore, the most convenient way of disposing of the tailings (solid waste) was to either back fill the old mine site with waste rock or to dump the tailings on nearby land. According to Davies (1983,434) after the solid and liquid wastes had been disposed of "the metals and their compounds may be released as solutions, colloids or suspended particles in aqueous solution, or as gases, aerosols, dusts from smelter stacks, ore stock piles and tailings." In areas of historic mining activity the local environmental pollution problems arise because once the metals have been released into the environment they are free to follow natural geochemical and ecological pathways. The three major pathways for solid waste are by atmospheric, fluvial and gravitational transport. Figure 2.6 shows the interrelated movement of solid and liquid wastes within the environment. For example solid waste can be moved through the surrounding environment in a number of different physical and chemical forms.

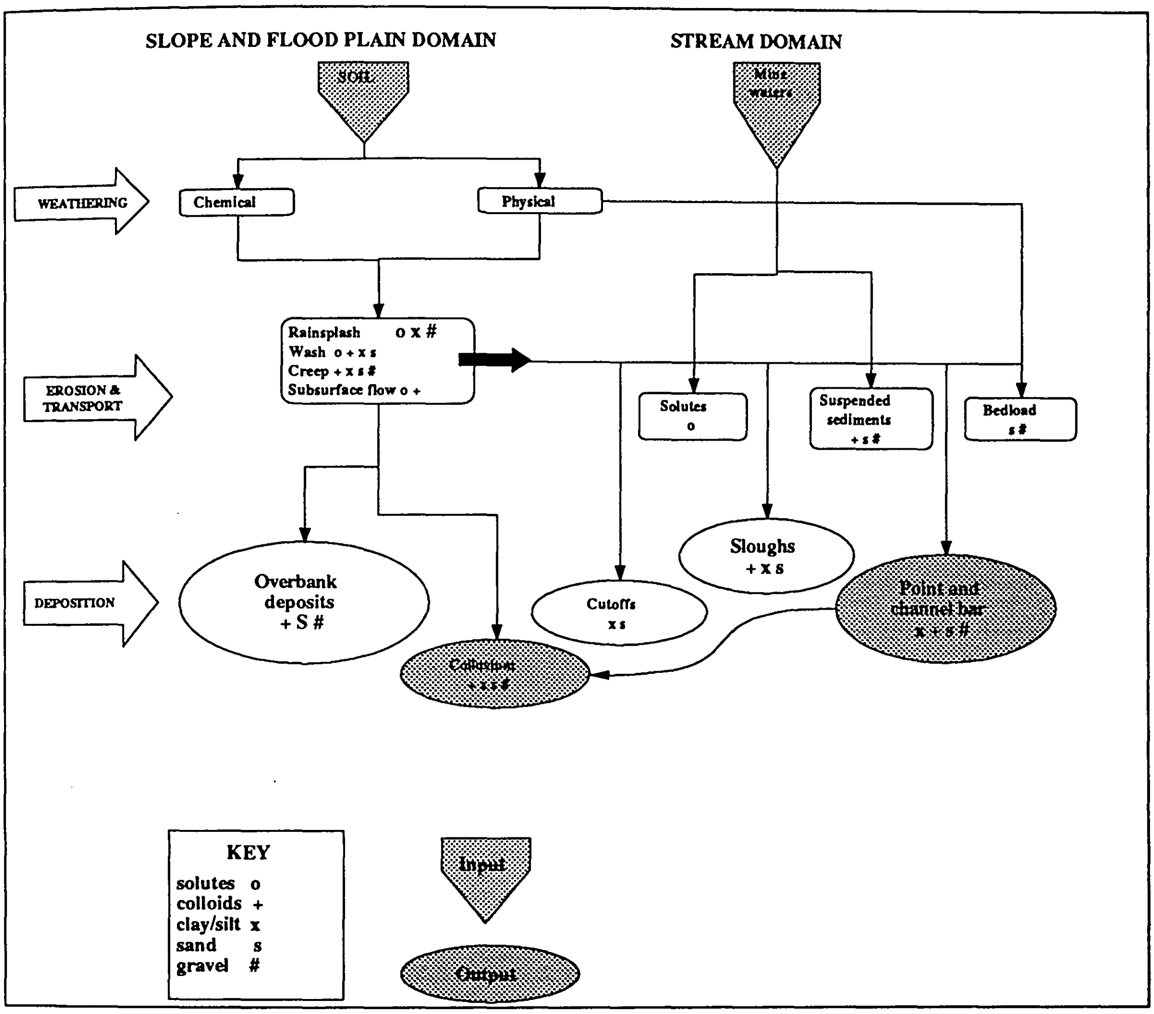

Figure 2.6 Conceptual model showing the dispersal of mine wastes by fluvial activity in west Wales (after Davies 1983, 436).

There are a considerable number of previous studies showing the problems of trace metal pollution associated with the historical mines of England and Wales. The most recent is the series of regional geochemical atlases constructed by The British Geological Survey which cover most of the British Isles. Using geochemical dispersal plots, the atlases depict the concentrations of the major and minor elements found in stream sediment based on a sample interval of 1 km^2. This interval of sampling scale has to be adopted in regional studies to facilitate the vast areas covered within the survey.

In contrast to the amount of research undertaken in England and Wales, there have been few attempts to determine the local environmental impact of historical metal mining in Scotland. This could be due to the fact that mining has tended to be on a much smaller scale than in England and Wales although arguably the lead mines centred in the Wanlockhead region are an exception. However to date only limited research has been undertaken on monitoring the environmental impact of lead mining in this region. In 1982 the Clyde River Purification Board (CRPB) conducted geotechnical surveys on 12 historic mine drainage levels and two culverts down slope from two lead smelter sites. As expected the survey showed extremely high concentrations of Pb, Zn, Cu, Cr, Fe and Mn in the sediment from the levels. The maximum values obtained in the survey for Pb, Zn, Cu and Cd were 16,343; 1,614; 281; 44 and 4.2 ($\mu g\ g^{-1}$ dry weight) respectively, (CRPB Report TO1-C 1982).

2.3.5 Summary and implications

To summarise, base-metal mining accentuates and accelerates natural processes. The development of underground workings, open pits, mill tailings, and spoil heaps and the extractive processing of ores enhances the likelihood of releasing chemical elements to the surrounding area in large amounts and at increased rates relative to unmined areas (King *et al* 1995). The major trace metal pollution problems associated with historical mines are contaminated land (Mattigod and Page 1983), surface water pollution (Forstner and Wittmann 1979) and stream sediment enrichment (Abrahams and Thornton 1987; Bradley 1982 and Wolfenden and Lewin 1978).

This section has explored how heavy metal contamination persists in surface soils as a direct results of base metal mining. The principal mechanisms governing the environmental route-ways of these potentially harmful heavy metals have also been examined. There is a large amount of data in the environmental record on the impact of historic base metal mining within England and Wales. In Scotland there appears to have been considerably less research undertaken on this area of study. The implications for this study are that mining can lead to substantial contamination over a wide area, especially downstream in fluvial systems. In considering attempts to identify metal-extraction in the absence of archaeological data it appears possible that freshwater sediment provides an appropriate medium for recording and measuring mining activity. Part III of this chapter will now discuss the implications of heavy metals in the aquatic environment.

2.4: Heavy metals in the lacustrine environment

2.4.1 Introduction

Section 2.3.4 reviewed the implications of heavy metals in the terrestrial environment and showed that abandoned mines produce high levels of potentially toxic materials that enter the aquatic environment along fluvial pathways as a result of weathering processes. In this section the lacustrine environment is discussed in relation to research on lake sediment. Lake sediments have the ability to contain and preserve a record of pollution inventories that can be reliably linked to historical events and changes that have occurred in the lake's catchment. The scientific approaches commonly applied to lake sediment have been wide ranging and can vary considerably according to the questions being raised.

Lakes (lochs) are depositional sedimentary environments that have been shown to be suitable for the incorporation of permanent and often short-term fixation of metals and organic pollutants. Lake sediments record information about erosion events that occurred in pre-cultural times within the lake's catchment (Mackereth 1966; Stumm *et al* 1978; Milcher 1983). Where man has interfered primarily with the terrestrial ecosystem, such as by metalliferous mining, cultivation, forest clearance and afforestation, the varying degrees of this interference can be established within the geochemical record of the lake. The occurrence of unexploited mineralisation within the catchment may also lead to changes in the chemical record within the sediment (Hilton *et al* 1985; Laville-Timsit 1991 and Erel *et al* 1991).

The cultural impact on the geochemistry of sediments has been an important criterion for the investigation of lake sediments, particularly in palaeoenvironmental reconstruction and in studies that have monitored the advent of industrialisation (Kemp 1976; Bertine & Mendeck 1978; Hamilton-Taylor 1979 and Rippey *et al* 1982). This is usually inferred from a general knowledge of local point sources (e.g. sewage discharges and metal smelters) and more regional diffuse sources (e.g. vehicle emissions and coal burning). Recent studies have measured the regional and global impact of pollution sources as a result of the growth of contemporary industries such as mining, metal smelters and power stations (Thornton & Abrahams 1984; Hermanson 1991 and Hallberg 1991). Elsewhere, researchers have used palaeolimnological techniques to investigate the effects of eutrophication and acidification. The reconstruction of changes in lake acidity and the direct link with 'acid-rain' is now widely understood (Nriagu and Coker 1983; Maitland *et al* 1987 and Battarbee *et al* 1988). These processes are discussed in greater detail in Chapter 9 Section 9.4.1.

2.4.2 Trace metal concentrations in lake sediment

The following review of previous investigations summarises the approaches and applications that have been applied to the study of pollution history from soils and lake-sediment with an emphasis on production-related activities such as mining and smelting processes. The historical perspective is maintained throughout these studies. For reasons of brevity the following is a summary, and a discussion on specific dating methods applied to pollution monitoring is postponed until Chapter 3 Section 3.8.1.

Because of their impact on human health the last two decades have seen the development of a large corpus of literature concerning the distribution of heavy metals within the environment, with particular attention being given to the fate of lead within a variety of biological and non-biological media. The use of lake-sediment, ombrotrophic peats and alluvial soils for palaeocological reconstructions using the onset of industrial development both at a local and regional level has been widespread (Farmer *et al* 1983; Lee and Tallis 1979; Bertine and Mendeck 1978; Forstner 1976, 1977; Lewin and Macklin 1987; Hermanson 1991; Hallberg 1991 and Entwhistle *et al* 1995).

Commonly, researchers have measured down-core heavy metal concentrations and attributed increases in heavy metals to particular events that have been linked to local or regional industrial processes. A historical perspective has been developed to monitor such changes that have arisen due to the rise and decline of a given industry, usually supported by historical documentation and often supported by using rigorous radiometric time-frames. It has been argued (Alderton 1985) that if analytical results from such studies are to be anything more than qualitative, an accurate time-frame for the sediments needs to be established (see Chapter 3 Section 3.8).

Lake sediments provide an extensive store of biological data on postglacial environmental chang and have been the focus of research on short-term environmental changes. In particular, the contribution to studies on the problems of eutrophication[5] and acidification is evident by the wealth of literature on the subject (for an overview see Mannion 1989 and Oldfield 1977 and in detail Battarbee 1977, *et al* 1978, 1980, 1984, 1986 and 1988). Among the range of biological micro-fauna that can reflect such changes, the use of diatoms, chrysophyte (*Mallomonodaceae)*[6] chironomid[7] (*Diptera*) and cladoceran[8] assemblages

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[5] The term eutrophication describes the biological reaction of aquatic systems to nutrient enrichment, the eventual consequence of which is the development of primary production to nuisance proportions (Marsden 1989)

[6] Chrysphaceae are a form of phytoplankton (Munch 1980)

[7] Chironomidae are midge larvae identified by the morphology of their head capsules (Hoffman 1986)

examined in conjunction with trace metal inventories forms a powerful means of understanding the pollution history of a lake. Diatoms are single-celled microscopic plants belonging to the algal class *Bacillariophyceae*, (Dixit *et al* 1992), the common feature of which is a distinctive, highly ornamented cell wall composed primarily of silica oxide (SiO_2). Each cell wall is made up of two main pieces called 'frustules' or valves. Diatoms are key components of nearly all fresh and saline environments and are quick to respond to environmental change owing to their narrow environmental optima and tolerances.

Using diatoms as a means of understanding the changes in water quality, Guppy and Happey-Wood (1978), and Elner and Happey-Wood (1980) examined the history of two linked but contrasting lakes in Snowdonia, Wales. The studies combined a chemical and biological approach using a chronology based on ^{210}Pb and radiocarbon dating. Their work showed that 19th century copper and slate mining within the catchment of the lakes resulted in an increase in the influx of copper into sediment. This resulted in the suppression of the diatom community *Cyclotella* that was abundant prior to the onset of mining in the 19th century. Another efect on diatoms has been seen elsewhere, namely in the Lake District where Hamilton-Taylor (1979) recorded a close association with a high Pb, Zn and Cu influx (1180 ppm, 589 ppm and 140 ppm respectively) and the diatom group *Asterionella*. This high metal loading compared to baseline concentrations has been attributed to the general rise in local urbanisation and use of pleasure craft on the lake.

Using chemical analyses on radiometrically dated cores with supporting pollen analyses, Davison *et al* (1985) were able to show that copper mining (*c*1850) in the catchment of Coniston Water in the English Lake District provided a clear sedimentary record of this local mining activity. Although mining effluent entered the lake at one location, elevated concentrations of Cu, Fe, Mg, and Ca were observed at five sites away from the main point source of pollution. Moreover, their work showed that random transport processes were involved in the distribution of sediment material contrary to the assumptions that maximum metal accumulation will take place at the deepest point in the lake and that the rate of accumulation will decrease in shallower water.

Similar trends have been observed in the Great Lakes of North America where later in post-1935 sediment, Pb values are in the order of 100-200 $\mu g\ g^{-1}$ compared to background concentrations of $< 50\ \mu g\ g^{-1}$ which are typical of Pb profiles. Chromium and Hg are also seen to peak towards the surface at around this depth in sediment profiles. In other studies

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[8] Cladoceran are arthropods (segmented invertabrates) see review by Frey (1986)

these metals are seen to be the result of chlor-alkali plants that developed along the shores of the Great Lakes since *c*1900 (cf. Thomas 1972; Kemp *et al* 1974; Kemp and Thomas 1976a; 1976b and Kemp and Dell 1976). These authors examined the effects of pollution from one location or point source. Elsewhere the effect and implications of air-borne long-range pollution transport has been established (Rippey *et al* 1982).

The concentration-depth behaviour of Mg, Cr, Ni, Cu, Zn, Hg, Pb and P in three sediment cores from a central site in Lough Neagh, Northern Ireland was examined for changes in sedimentary flux of these elements. Two main periods of change were found. A change in the catchment erosion-leaching regime in the 17th century caused by documented widespread woodland clearance which produced increased sedimentary Mg, Cu, and Pb concentrations. Based on a ^{210}Pb chronology, a second and larger change occurred after about 1880 A D with Cr, Cu, Zn, Hg and P concentrations increasing toward the sediment surface. Differing P and trace-metal concentrations in conjunction with estimated anthropogenic sedimentary flux and background atmospheric contributions suggest that atmospheric sources made a substantial contribution to more recent Cu, Zn, Hg, and Pb sedimentary contamination. This led the author to conclude that the trace-metal contamination of Lough Neagh is part of a global pattern.

Hong and co-workers (1994) have examined sections of a core 3000 m long which was drilled at Summit, Greenland. Examinations of ice-core sections covering the period 3000-500 years ago shows that lead is present at concentrations four times as great as their natural values from about 2500-1700 years ago (500 BC-300 AD). These results show that Greek and Roman lead and silver mining and smelting activities polluted the middle trophosphere of the Northern Hemisphere on a hemispheric scale two millennia ago, long before the industrial revolution. It is proposed that 80,000 metric tons of lead were produced around 2000 years ago and it is estimated that 5% of the mined and smelted lead was emitted to the atmosphere producing a peak value of *c*.4000 metric tons lost to the atmosphere (Hong *et al* 1984, 1842).

Many trace elements are emitted to the atmosphere because of their volatility in high-temperature combustion and metal extraction processes (Peirson *et al* 1973; and Pacyna 1984). The implications for long-range transportation of heavy-metals derived from industrial sources and from the urban sector has given rise to a series of studies that have applied chemical analyses to a range of biological materials ranging from terrestrial peats to riverine sediment (Livett 1979; Gorham *et al* 1984 and Gorham *et al* 1990).

In the Pre-Cambrian Shield Region of Canada, mineral exploration companies make explicit use of the upper levels of lake sediment for geochemical prospecting. This approach is based on the variability of geochemical suites of economic importance within the lake sediment which is taken to reflect regions with increased mineralisation within the catchment (Hood 1979). Attention to chronologies is not a general requirement as the presence or absence of the chemical suites are all that is required. Qvarfort (1977) working in the Bergslagen region of central Sweden, examined the contamination problems within different types of material (till, water, stream and lake sediment) to arrive at an understanding of the effects of vertical and lateral contamination of Quaternary deposits due to wind-borne, metal-rich dust and smoke from different types of mining and manufacturing activities. The study area was mined intensely from the Middle Ages for copper and lead and by the early part of the 18th century ten smelters were scattered throughout the region. Although the research was aimed specifically at geochemical prospecting in a highly contaminated area, the study achieved its principal aim of determining the potential use of different types of sampling material and the determination of natural influences on mineralisation in a region highly polluted by four centuries of mining activity.

The stratigraphic implications of heavy-metals in alluvial sediments has received much attention in the last decade (Macklin 1985; Macklin and Dowsett 1989; Macklin *et al* 1992) with the investigation of alluvial deposits from river terraces in the Tyne basin. The South Tyne river and its upland tributaries drain substantial areas of limestone and the Northern Pennine Orefield. The influence of base-metal mining in the Northern Pennines has been seen to have an influence on the metal budget of the South Tyne river with the increased quantity of fine and coarse sediment delivered to river channels in the Tyne catchment. Significant quantities of sediment-borne metals enter local river channels through the erosion of mining-age alluvium during periods of flooding. This has led to the dispersal of fine-grained sediment high in Pb, Zn and Cd values and has arguably resulted in poor performance in crop production (Macklin *et al* 1992).

2.4.3 Summary

Lake sediments provide a useful medium with which to observe long and short-term geochemical trends as a result of changes within the catchment. The routing of metals and their eventual fixation into sediment is complex and often poorly understood. Diagenetic changes brought about by co-precipitation mechanisms form one major aspect of sediment chemistry and speciation studies are at the forefront in understanding the role and diffusion of the metals commonly recorded in lacustrine sediments. There is a wealth of scientific data to show that the anthropogenic loading of lead in the upper surface of lake sediment can be several orders of magnitude higher than in the deeper regions of the sediment. If diagentic and other post-depositional changes can be identified then often the chemical and physical properties within the sediment can be directly related to a number of natural and anthropogenic agencies. Short-term changes within the sedimentary record spanning the last 500 years form a fundamental aspect of historical monitoring. A critical aspect in the development of a historical perspective is the construction of rigid time-scales with support from historical and other environmental indices. Such time-scales permit hypotheses of cause and effect to be rigorously evaluated.

2.4.4 Implications for this study

It was stated in Chapter One that there is a need to examine the principal sources of contamination within the study area. This is important in order to understand the routing of natural and anthropogenic derived contamination into soils, particularly when assessing the scale of soil contamination away from the areas of mineralisation (i.e. in the control catchment of Loch Leathann). Of equal importance is the need to gain an understanding of the principal environmental pathways of heavy metals into the sedimentary environment. In order to achieve this objective an examination of the 'pollution record' from other mining districts has provided comparative data and other information relevant to the cycling of metals through the terrestrial and aquatic environment.

The issues that have been raised above are now brought together in a simplified conceptual model showing the major gradients that lead from a hypothetical lead ore deposit through its various production processes and the likely pathways that can result in the contamination of the loch and alluvial sediment. The model (Figure 2.7) shows three principal pathways relative to the study area and these are now summarised:

(1) Lead mining and contamination of the fluvial environment ('the mining route')

Old open-cast workings and shaft and adit mines contain extensive spoil heaps that weather over time leading to local and often regional pollution. Reclamation and subsequent clearance of metal-rich spoil along with wind blown metal-rich dust can also lead to increased contamination. Metal-rich seepage from spoil heaps will enter into ground waters and streams where they can become incorporated into stream sediment.

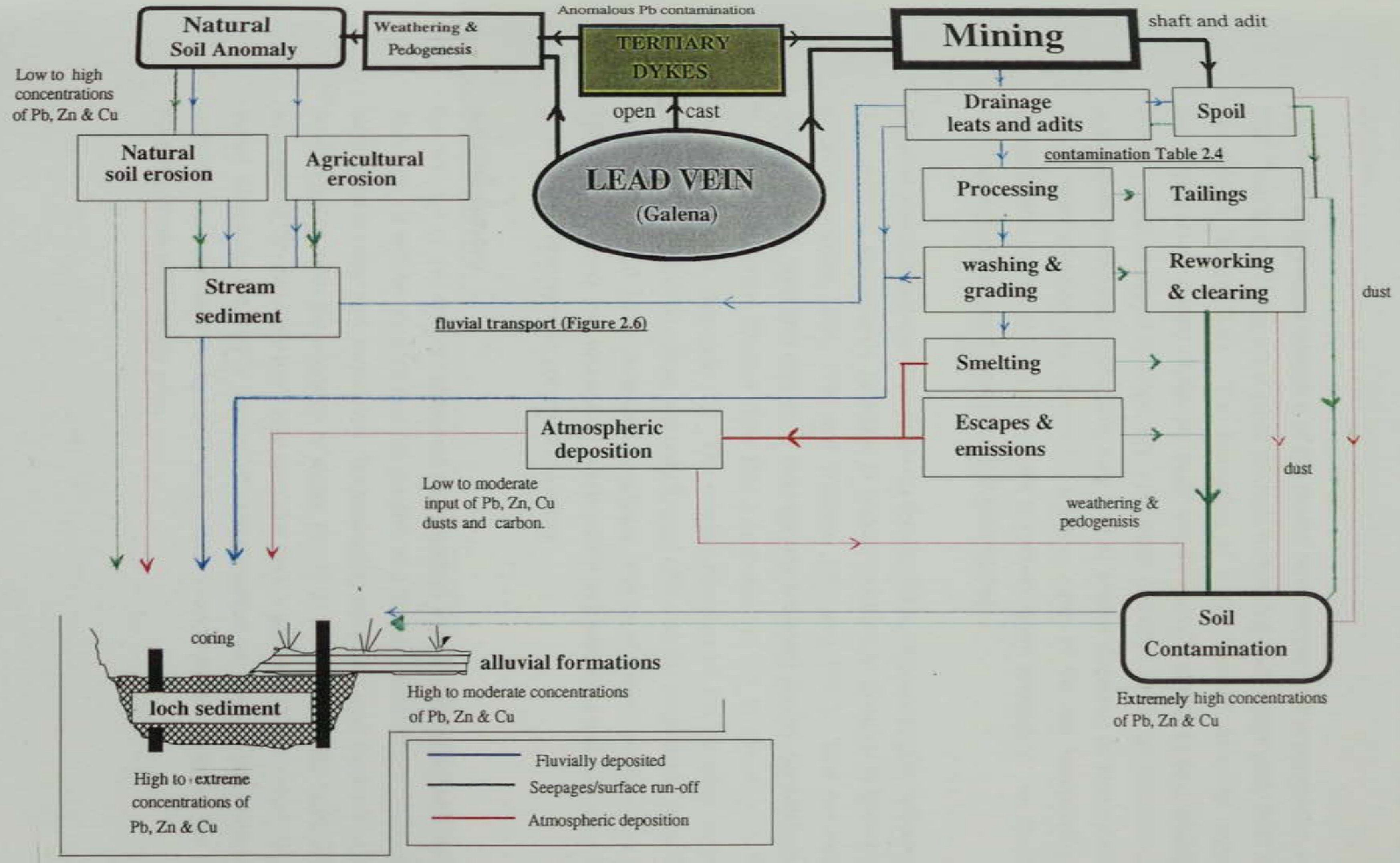

Figure 2.7 Conceptual model of the major environmental pathways of heavy metal contamination into soil, freshwater loch sediment and alluvial sediment in the study area.

The porosity of the local limestone will warrant elaborate methods of mine drainage using adits and leats. Drainage waters will be charged with heavy metals, particularly whilst the mine is in operation. Ore washing and grading leads to highly contaminated waters that can easily enter the fluvial environment as accidental or deliberate run-off into nearby water courses.

(2) Natural soil contamination of the fluvial environment ('the natural route')

Natural soil anomalies are derived through weathering and pedogenesis over local unexploited ore bodies (Figure 2.4). The formation of a geochemical anomaly within soils will be dependent on the nature of the ore body and the metal's ability to be retained or remobilised. Factors such as aspect of slope, eH, pH, redox potential, drainage patterns and the effects of agriculture particularly ploughing may lead to greater dispersal of heavy metals across areas where mineralisation has occurred. Streams close to the ore bodies may receive waters charged with metals to form significant stream-sediment anomalies. As we have said earlier, this forms the rationale for geochemical prospecting.

(3) Atmospheric fall-out deposition on the fluvial environment (the 'atmospheric route')

The onset of atmospheric pollution is widely recorded in Scottish freshwater sediment from *c.*1850 (Battarbee 1984; 1995 and Williams 1991 and 1992). This has resulted in a major influx of lead, zinc and copper in near-surface sediment across Scotland due to the rise in industry. Historical sources show that a lead smelter was in operation in Islay in the 18th century (Plate 5, Appendix 1). The coastal situation of the Freeport smelter (Figure 4.2) would in all probability have had only limited effect on the study area. Windblown dust from spoil tips would have presented a greater source of heavy metal input into local soils particularly where ore processing (particularly mechanised crushing) was undertaken. This issue will be discussed in greater detail below.

2.5 Conclusions

Section 2.2 of this chapter discussed the historical and technological achievements in the lead industry. It will be recalled that the English and Scottish lead industries developed rapidly in tandem with other new industries. Technological advances were made alongside the iron and coal industry, with the adoption of steam power to drain mines and work ancillary crushing and hauling devices. We have also seen that poor legislation in the control of potentially toxic waste materials has left a legacy of contamination at many abandoned sites. This has provided the necessary background with which to address the consequences of environmental contamination in the study area.

The routing of heavy metals and their dispersal in the terrestrial environment has been examined in Section 2.3 which has demonstrated that the long-term storage of metalliferous-rich particulate matter can lead to extremely high heavy metal concentrations. The implications of this have been shown to be serious, often resulting in long-range pollution away from its original origin or point source. Aspects of metal concentration and the definition of the 'geochemical anomaly' have also been considered. This subject has a direct bearing on the interpretation of the heavy metal distribution maps that show the level of soil contamination over land effected by metal mining.

The legacy of metal pollution at sites formerly mined for lead and other metals is well recorded in a variety of environmental indices. Historical landscapes and the effect of centuries of mineral extraction are now slowly being recorded and are now recognised as a threatened archaeological resource. Wales in particular is a country ideally suited to this type of study owing to a protracted history of lead and copper mining. Prior to this study, little work on this topic has been undertaken in Scotland.

Section 2.4 has dealt with cultural impact on the sedimentary environment of lochs. It was demonstrated that loch sediments can provide an extensive archive of biological and chemical data with which to monitor environmental changes. Aspects of this study are wide ranging and some of these are developed further in Chapter 9. The concluding section of Section 2.4 has put forward a model to construct three primary environmental pathways. These were simplified under the general heading of ‘mining’ ‘natural’ and ‘atmospheric’. Where streams flow through regions that are locally contaminated as a result of near-surface mineralisation, then one might expect an increase in the heavy metal burden along this route. Where mining has occurred over such regions, the burden is likely to be greatly increased, particularly if adits or leats drain from the mine in the direction of, or directly into a given loch.

The significance of the atmospheric route cannot be understated. As we shall see below, there is no freshwater loch in Scotland that has not undergone some form of near-surface enrichment from this route.

To conclude, the model provides us with the necessary background information to proceed with the implementation of a series of analyses in which to evaluate the relative scale of environmental impact in loch sediment and soils as a result of 18th and 19th century lead mining.

An integrated approach has been adopted using methods from a variety of disciplines which include Chemistry, Limnology, Geology, Geochemistry, Palaeoecology and Field Archaeology. The implementation of the various methods provide a holistic approach to link the archaeological remains of the abandoned leadmines with the environmental record contained within the loch and marsh sediment. The following chapter will now describe the methods that have been adopted within this thesis.

CHAPTER 3: METHODS AND MATERIALS

3.1 Introduction

(a) Analytical

This chapter describes the methods and materials that have been used in this thesis. Different sample preparation were used according to the requirements of methodologies. For instance, oven or air dried sediment was required for radiometric dating and chemical analyses, whilst X-radiography, pollen analyses, pore-water content along with pH required the use of wet sediment. A summary flow diagram (Figure 3.1) outlines the order in which these procedures were undertaken. A multidisciplinary approach has been applied and many of these methods have been adapted and modified where necessary to suit the needs of analyses which together combine into a holistic study of the loch-catchment unit. This approach has allowed an estimation of the environmental impact of lead mining activity in both local soils and on the chemical influx in loch sediment.

(b) Field Survey

It was mentioned in Chapter 1 that the archaeology of mining is notoriously imprecise owing to the lack of a relative dating evidence. Often the features we are trying to date and interpret have been partly destroyed or 'masked' by later workings. The true scale and geographical extent of the abandoned mines are rarely shown on modern maps. It was for this reason that detailed archaeological fieldsurvey was undertaken over the study area.

Firstly, in order to establish the distribution of land affected by mineral extraction a pilot survey was undertaken across the study area to identify the extent of abandoned mine workings. The survey then recorded and mapped the lead mining sites at both a local and regional level. The relationship between local geology and the presence of lead mineralisation was assessed from previous geochemical survey data. This largely governed which lochs would be suitable for investigation. The results of field survey are summarised in a general distribution map of lead mining sites and ancillary features concerned with this industry (see Chapter 4 Figure 4.2).

Secondly, when the distribution of lead mines had been established, four lochs were then chosen for further investigation. Lochs Finlaggan, Bharradail, Lossit and Leathann (control loch) were chosen for geochemical study and a detailed account of the criterian for their selection is provided below (Chapters 5, 6, 7 and 8 respectively). Bathymetric survey and core recovery was undertaken, which was then followed up by sub-sampling to identify the physical properties of loch and marsh sediment. The identification of the physical properties of lead ore, loch and marsh sediment employed X-ray diffraction and X-radiographic techniques to provide visual and other useful semi-qualitative data. After further sub-sampling and acid digestion of sediment samples, routine chemical analyses were undertaken to determine the concentration of Pb, Zn, Cu, Ca, Mn and Fe. The results of these analyses have been computed to produce depth versus concentration profiles to show the relative changes in chemical influx. Chronological control has been established based on the sediment inventory of the radionuclide ^{210}Pb. Additional chronological support has in some cases included ^{14}C, ^{137}Cs, pollen analyses and reference to historical sources.

(c) Archival

Thirdly, a desk-based study was designed to include the analysis of cartographic sources in the National Map Library of Scotland and a search for documents and plans housed in the archives of the Islay Estate. Additional geotechnical data has been acquired from mineral prospecting companies who conducted large scale soil geochemical surveys across the study area in 1973 and 1983. A re-evaluation of parts of this data has provided new estimates on the degree of surface-soil contamination by Pb, Zn and Cu in the catchment of the lochs chosen for investigation. The chemical data were also useful in supporting the hypothesis that no mining had been undertaken within the Loch Leathann catchment. At a local level this information is significant in terms of our understanding of the extent and scale of heavy metal pollution resulting from mine drainage-waters and tailings heaps.

(d) Detailed study

Further research was undertaken to determine the impact of local lead mining in the catchment of Loch Finlaggan based on biostratigraphical and geochemical analyses of cores from Finlaggan Marsh. This work was designed to monitor the changes in sediment development and chemical stratigraphy produced by natural alluvial processes, historical lead mining, agriculture and more recent afforestation in the Finlaggan catchment.

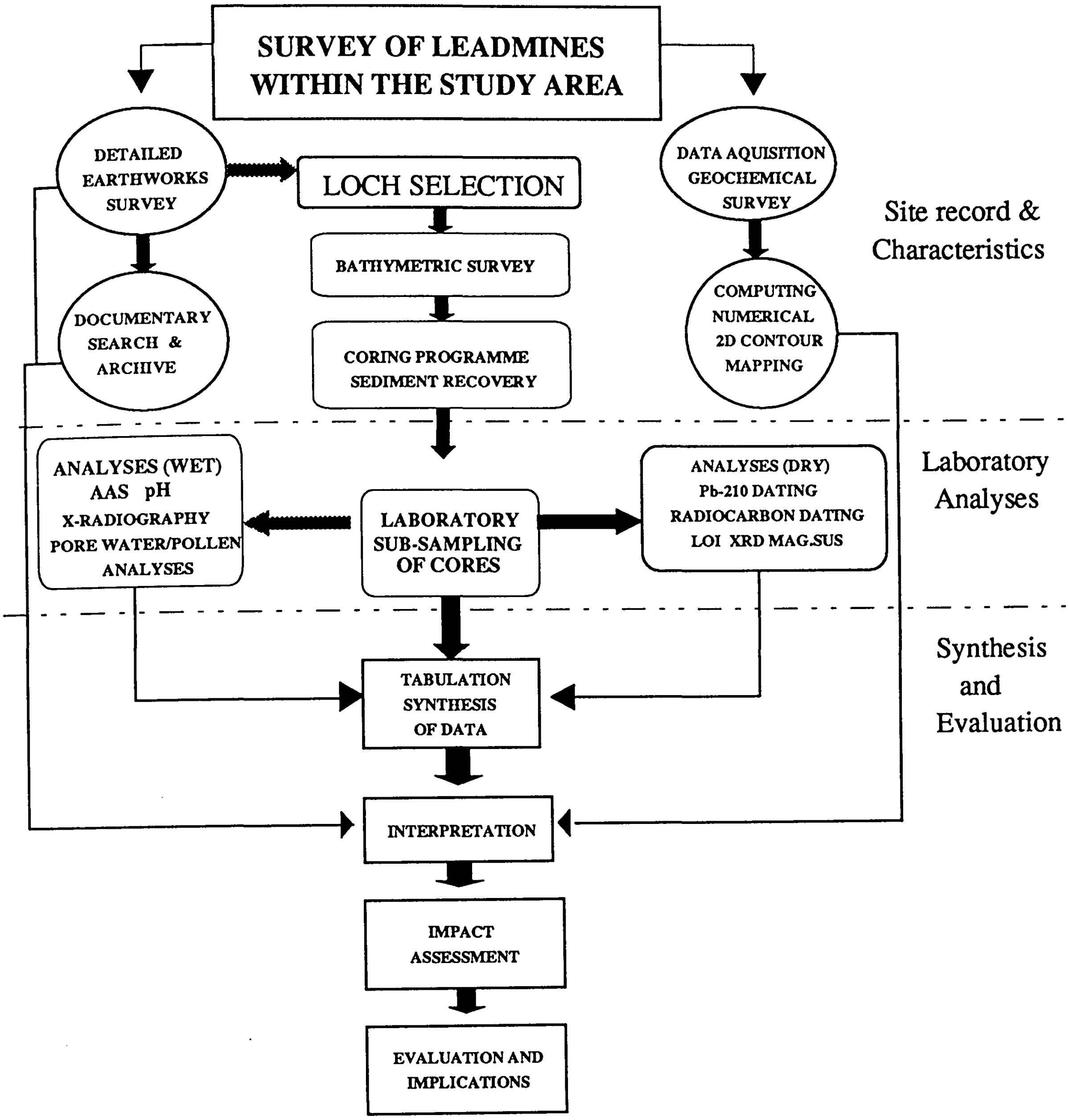

Figure 3.1 Flow diagram of the methods employed in this study.

3.2 Field Survey Methods

3.2.1 Archaelogical survey at selected mine sites

The survey comprised two stages, of which the first was a desk-based assessment of cartographic, bibliographic and aerial photographic sources. The second stage involved survey using a plane-table, prismatic compass, 360 degree protractor, 100 and 30m tapes. This is a very efficient method of survey, allowing one person to traverse quickly over large areas of the landscape. A plan is drawn directly on the plane-table which is annotated as the survey progresses. The method employed in this survey produced an accuracy of ± 50 cm. The appropriate scale for a given set of features was determined by the size of upstanding mine remains and their geographical extent. Parallel base lines aligned to Magnetic North were placed onto a perma-trace covered plane-table. Compass bearings were made against sighted ranging-poles and the distance noted along a 100 m tape and where appropriate, right angle offsets were taken across specific features. Aspects such as steepness of slope were annotated by using a system of hachures used to indicate relief features. In the field, notes were taken of the character and state of preservation of mining features together with identification of the extent of mine tailings. A large proportion of the time taken on the survey was devoted to field assessment of identifiable mines and examination of features such as trials and other open-cast workings This allowed areas of mining activity to be demarcated onto a sketch plan and annotations made directly onto Ordnance Survey maps. These were supported by a 35 mm black and white and colour slide photographic record of all the main features of each mine alongside the mining landscape in general. Examination of the Sites and Monuments Record (SMR) along with aerial surveys housed in the RCAHMS was also carried out.

3.2.2 Summary

The results from this programme of research have provided new information on the geographical extent and state of preservation of the mine workings. Previously unrecorded information regarding the different phases in their development has been recognised. Certain features observed in this study, including shafts and adits, are being continually back-filled by farm refuse and will consequently be lost. Some of the upstanding features such as walls and earthworks are being eroded or removed for track repair. Some of the upstanding remains at the Mulreesh Leadmine (see Figure 5.4) are identified as unique in terms of Scotland's industrial heritage. The mine surveys undertaken in this study make a contribution to the industrial archaeological record of Islay. The survey results confirm leadmining activity within an area between *c* $8k^2$. The results may also prove useful in any future mitigation programme to safeguard these important archaeological remains.

3.3 Re-interpretation of Previous Geochemical Exploration data from NE Islay.

3.3.1 Summary

An assessment of the environmental impacts of mining should be compared with areas that are undisturbed. Ideally, information about an area prior to human interference would serve this purpose; however, this type of information is rarely available (King *et al* 1995). In most cases, comparison areas are chosen that are geologically similar but have not experienced 'adverse' human interference. The effect of human interference is commonly evaluated by simple statistical techniques or by using the comparison, or a 'background site' to provide a baseline. Loch Leathann and its surrounding soil chosen as a control as the loch shared the same geological characteristics as the other lochs chosen for investigation and there was no visible evidence of mineral extraction within its catchment.

3.3.2 Analysis of previous geochemical data

We needed to be certain at the outset that there were no naturally occurring geochemical anomalies within the catchment soils, which might have led to significant increases in the heavy metal burden within the loch sediment. Moreover, it was important to be able to assess the present day concentrations of Pb, Zn, and Cu across the areas previously mined. This would then provide information on the geographical extent of contamination and its relationship to soils and stream sediment within the study area.

Original soil geochemical data of Pb, Zn and Cu (in ppm) from the 1983 Domego Resources Limited (see Chapter 4 Section 4.4) survey has been re-interpreted using Unimap, part of the Uniras series of mapping and graphical software in the University of Edinburgh. Soil samples were originally analysed using conventional AAS and concentration values were expressed in parts per million (ppm). Metal concentrations were plotted directly on to base maps and contours depicting increases in Pb, Zn and Cu were placed to demarcate highly anomalous areas. This author re-plotted the data according to eastings and northings co-ordinates (designated X and Y) with the Pb, Zn and Cu values placed on its respective Z co-ordinate. This provided three columns suitable for computation by the Unimap software into a two dimensional contour map. The 2D contour maps are powerful in their ability to display both the variance in metal concentration and their close conformity to local geological boundaries and other areas contaminated by mine waste. The complete data set used in the formation of the geochemical distribution maps are shown in Appendix 2-5.

3.3.3 Summary of results

The results from this work have shown that there is significant variance between the catchment of lochs Finlaggan, Lossit, Bharradail and the control catchment, loch Leathann. The results also show that the criteria in the selection of this loch and surrounding soils as a background is perfectly valid (see Chapter 9 Figure 9.1). The catchment geochemical distribution maps show that the soils adjacent and over old mine workings are extremely contaminated. Surface enhancement of Pb is in excess of 5000 ppm at the old workings at Portnealon, in the catchment of Loch Finlaggan. Assumptions regarding the influence of Tertiary activity, natural unmined mineral enhancement and geological boundaries provide important information with which to estimate background Pb, Zn and Cu concentrations within a given catchment.

3.4 Bathymetric Survey of Selected Lochs

3.4.1 Introduction

Bathymetric surveys have provided an insight into the formation and physical characteristics of lochs, including profile shape and depth. Other important hydrological parameters, such as seasonal changes to loch levels, flushing and net water retention can be acquired from bathymetric survey (Murray and Puller 1910, and Lyle 1987). The only work of this nature on Islay was a partial bathymetric survey of Loch Finlaggan undertaken by the British Geological Survey in liaison with the National Museums of Scotland (Ruckley and Chesher 1990).

Bathymetric Surveys were undertaken over four days in May 1992 on Lochs Lossit, Leathann and Bharradail by this author. The conditions were favourable with little or no wind. Echo sounding surveys were made from small fibreglass rowing boats with a speed in the region of 2 knots. The survey apparatus was a Seascribe Mark 2 echo sounder operating at a chart speed of 120 cm/hr and calibrated in feet. The transducer was mounted at the stem of the boat. Power was supplied by a 12 volt portable battery. The equipment proved ideal when working in locations that were shallow but free of pond weed. This proved to be the major drawback in shallow locations, here a 2m ranging rod being used to take depth readings. The need for a constant speed and straight trajectories were the most demanding requirement of the survey. At each site surveyed, echo sounding transects were run between identifiable points on the shore and occasionally offshore features such as islands were used. These provided useful intermediate reference points along a transect. Transect lines and end points were marked on to a base-map at 1-5000 scale. The number of transects taken varied according to the size of the loch. A sounding line was used to calibrate echo soundings taken at the coring stations and at random intervals during the survey. Allowing for the soft nature of the loch sediment the error between echo readings and line soundings was in the order of plus or minus 1 ft (30.05 cm). The chart plot was interpolated to an appropriate scale and plotted on to the base maps. An attempt was made to survey Loch Fada and Loch Bharadail but this was abandoned due to the mass of vegetation and the constant fouling of the transducer.

3.4.2 Summary of Results

Most importantly, the survey helped to locate the optimum coring position within the loch basin. Loch Bharradail proved impossible to survey owing to the density of surface vegetation which impeded the transducer. The results of bathymetric survey from lochs Leathann and Lossit and only partial survey of Loch Bharradail (Figures 6.6, 7.5 & 8.5) have provided new data concerning their geomorphology. The development of the loch basins clearly show regions of increased sedimentation which can be attributed to infiltration by major inlets. Additional information has been gained on the position and depths of the loch between the islands and shoreline in Loch Ballygrant and Lossit.

3.5 Gravity Coring, Design and Operation

3.5.1 Introduction

In very simple terms a gravity corer is a hollow tube of a given length that is heavy enough to penetrate soft lake sediment. The classic 'Mackereth Corer' (Mackereth 1958, 1969) has a core tube that is pushed into the sediment by pneumatic pressure provided by compressed air. For the purpose of this study, a corer was developed to obtain larger amounts of sediment in a core tube of greater diameter than the standard 5 cm diameter used in the Mackereth corer. The coring device (Plate 9, Appendix 1) was purpose-built in collaboration with the departments of Geology and Geography and subsequently modified by the writer in the field. The principal for this design was originated by the Loch Ness Research Centre and has the advantage of obtaining cores with a greater quantity of sediment in cores up to 2m long (A Shine pers.comm.).

3.5.2 Design and operation

The coring device consists of three components which include a 1m galvanised steel tube (scaffold pole) welded to a stainless steel base-plate. A plastic bin bolted to the base-plate allows weights to be added according to operating depth and sediment type encountered. The base-plate houses the core tubing (PVC drainage tube with 9.5 cm diameter) by two retaining bolts. At the lower end of the core tube an expansion ring-seal allows a vacuum to be maintained. A trigger device (Stenhouse-slip lifeboat quick-release bolt) is activated by the 'pendulum' weight when contact is made with the sediment, thus allowing the corer to sink into sediment. After approximately 30 minutes the corer is relieved of its weights by way of a separate hauling line. Buoys (net-floats) attached to the corer maintain buoyancy and allow the corer and trapped sediment to rise by hauling on the main lowering line. Once the corer is on the surface the device is lifted into the boat and the core tube is dismantled on the shore.

Before transportation back to the laboratory the floculant sediment-water interface has to be removed to prevent sediment mixing in the upper region of the core.

3.5.3 Summary of results

In total eleven cores were obtained over a period of two weeks in September 1992. The coring device operated well in the shallower lochs (Lochs Bharradail and Leathann) but often proved problematic in retaining sediment which had been recovered from the deeper lochs such as Loch Lossit and Loch Finlaggan. In all probability, this was due to the expansion of gas bubbles within the sediment which was affected by pressure differences as the core tube came to the loch surface. Greater success was achieved when shorter core tubes were fitted which was probably due to a reduction in sediment weight which minimised sediment loss from the core tube as the device was lifted into the boat. As previously mentioned an important objective for this author was to locate the deepest region of the this position minimises the risk of sediment slumping which is a strong possibility in steep sided U-shaped basins. Multiple coring allows much greater control over this problem (Dearing 1986). However it is well known from pollen recruitment studies (Bonny and Allen 1984 and Davis *et al* 1984) that sediment densities and formation rates can be radically different between one location and the next. In this study, two replicate cores were taken within 1-2 m apart from the deepest part of the loch. Once the first coring station had been established and marked with a buoy its position and known depth were recorded. Subsequent visits to the coring stations were made easier by locating the maximum depth and aligning the boat within the convergence lines of three azimuth bearings that were taken on near shore features.

3.5.4 Russian and Dutch coring at Finlaggan Marsh

In Quaternary studies macro-fossil analyses are usually part of wider palaeoecological research, and the selection of a sampling site depends on the general aim of the investigation. Since peat is deposited in a stratified manner one can trace the development of a marsh or mire by studying the succession of peat types in its profile, (Aaby 1986, and Aaby and Digerfeldt 1986). Once the peat types have been identified and the boundaries between one type and its successor have been established it is then possible to draw up a longitudinal or transverse profile (Moore and Bellamy 1974, Moore 1986, and Dearing 1986). It is usual to exaggerate the vertical axis (Chapter 5 Figure 5.29) in order to demonstrate more effectively the topographic features of the marsh. Regularity between the profile positioned is assumed particularly with close-interval coring. In some cases this may not be justified, but generally the close relationship between the stratigraphic sequence observed in each core allows the positioning of dotted correlation lines to link horizons. observed within the profile.

Biostratigraphic sampling at Finlaggan Marsh were undertaken using a Dutch and Russian corer with test-pitting in the shallow regions of the marsh. An X-Y transect was established along the length of the marsh and approximately over its centre. Figure 5.28 shows the X axis which is 54 m in length. Cores were taken at 2 m intervals and levels were taken at the surface of each coring station using a theodolite. The transverse axis (profile not shown) intersected the X axis at 32 m and was 22 m in length. Munsell colour codes and detailed notes on the characteristics of each core were established. A research design (see Figure 3.2 below) was established which utilised a modification of Digefeldt's (1986, 132) approach in the study of past lake-level fluctuations which included the construction and interpretation of paleo-shoreline stratigraphies based on multiple core analyses. The results from this work show that the marsh consists of lake gyttja[1] and fen-type and detrital marsh peats. A sand horizon intersects the marsh sediment at 150 cm depth and this is ascribed to a major flood event. This sequence is supported by palynological evidence. The results are provided in more detail in Chapter 5 Section 5.9.1.

3.6 Laboratory Methods

3.6.1 Sub-sampling and preparation

3.6.2 Introduction

A sample strategy was adopted as in Bengtsson and Enell (1986) whereby one half of a core would be used for wet and the other for dry analyses. Sample storage and the need to maintain sample stability were carefully considered. The risk of microbiological contamination was kept to a minimum by storage in a dark cold store. Core sediment was periodically monitored for signs of degradation. Before sampling careful scrutiny of the cores showed that four cores had undergone serious mechanical disturbance during recovery or whilst being transported to the laboratory. Theses were discarded and only those whose sediment integrity was sound were sampled.

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[1] Gyttja is a term for fine muds and silts in lacustrine deposits.

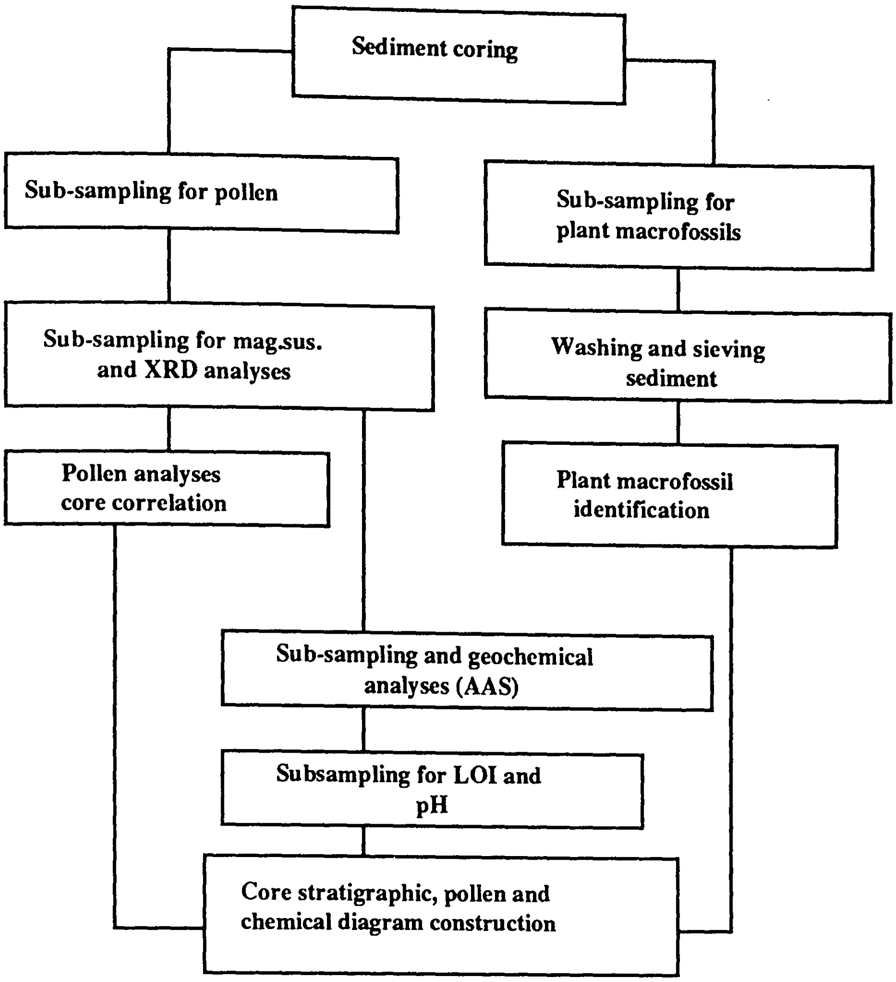

Figure 3.2 Flow diagram of the principal methods used in the biostratigraphical investigation at Finlaggan Marsh.

3.6.3 Core splitting and sampling strategy

A great deal of consideration was given to the method of splitting the core tubes. The input of any extraneous matter such as small fragments of plastic and fine metal particles from cutting tools would have been a serious source of contamination and affected the final results of chemical analyses. To avoid this problem, the core tubes were mounted on a traversing stage and placed beneath a bench mounted drill and drilled longitudinally using a bit from a centre lathe borer. This tool was very effective in allowing corresponding grooves to be cut to a depth of *c* 3 mm along the length of the core tube. Following this procedure, the core tubes were manually cut into two sections with an acid-washed Stanley knife. Acid washed (1M HCL) 'Former' PVC plastic sheets (2 mm thick) corresponding to the tube length and width were carefully inserted through the groove and into the sediment allowing the division of the sediment core. No plastic shavings from either cutting tool entered the sediment, thus minimising the risk of contamination. Finally prior to storage, the cores were wrapped tightly in cling film, capped with sterilised dividing 'Former' plastic sheeting, and labelled.

The cores were stored at 4°C in darkness to minimise micro-biological activity until required for sampling.

3.6.4 Uniform sediment sampling and equipment used

Before sampling the surface sediment was cleaned transversely with a stainless steel spatula. Biostratigraphic and colour changes were recorded using a Munsell chart and other lithostratigraphic information was placed onto record sheets. A stainless steel cutting tool with the same profile as the core tube allowed contiguous samples to be extracted at 1cm intervals. Each sample was placed in a labelled polythene bag and stored at 4° C until required. All tools were cleaned in 1M HCL between each sampling interval to minimise the risk of cross contamination.

Chemical and radiometric analyses required air dried sediment. The sample weight was recorded to 3 decimal places prior to air drying in an electric oven for a period of 48 h at *c* 50° C. The dried samples were re-weighed and recorded to determine the dry/wet sediment ratio. This allows an approximate estimation of pore-water loss on drying. The water content by weight of the sediment was obtained from $Mw/(Mw+Ms)x100\%$ where Mw and Ms represent the weight of water and dry sediment in each sediment section respectively (Farmer 1977). The sample was homogenised with an agate pestle and mortar which was wiped with 1M HCL between each sampling interval to avoid cross contamination. Dry samples were stored away from the light in 22 ml plastic universal containers.

3.6.5 Magnetic susceptibility

Magnetic susceptibility (χ) is an easily measured component within soils and sediment and is expressed as the ratio of the magnetism produced in a substance to the intensity of the magnetic field in which it is produced (Oldfield 1977, Thompson and Oldfield 1986). The measurement of 'magnetizability' is often approximately proportional to the volume of ferromagnetic oxides within a sample. Altered lithologies resulting from mining, historic burning regimes, acidification and podsolisation in surrounding iron-rich soil can in some cases lead to enhanced peaks in magnetic susceptibility within lake sediment profiles (for a detailed review see Dearing *et al* 1981; Dearing and Flower 1982; Dearing 1986 and Lagerås and Sandgren 1994).

Sediment re-adjustment resulting from a rapid influx of mineralogenic matter can easily be monitored which can allow additional chronological support for ^{210}Pb derived time-frames and multiple-core correlation (Oldfield *et al* 1978, 1983 and Dearing 1992).

Several authors have shown that industrial processes involving fossil fuel combustion can often lead to high magnetic enhancement at the surface of ombrotrophic peats and lake sediments (Tolonen and Oldfield 1986, Thompson and Oldfield 1986). The extent of the relationship between sedimentary magnetic enhancement from atmospheric particulate fallout is often seen to be in accord with the deposition chronology of pollutant heavy metals from specific point sources (Williams 1991).

Dried samples (40° for 24h) were placed into 10 ml plastic tubs and measured for magnetic properties using a Bartington Instruments MS2 meter. Maximum efficiency was obtained using the high frequency range under the cgs (counts per gram per second) and later converted to SI units using $\chi_{cgs} = \chi_{SI}/4Pi$. In the case of the Finlaggan Marsh cores, the core tubes were placed through a Bartington loop sensor on the high frequency range.

3.6.6 Summary of results

The routine analyses for magnetic susceptibility were achieved with good success. The loop sensor proved to be the most efficient method of quantifying internal magnetic properties whilst the wet sediment was examined *in situ* in the plastic guttering. For the loch sediment the process took longer with drying and single measurements using the 10 ml pot sensor. The maximum susceptibility value was attained in Lochs Bharradail and Finlaggan, both sediments achieving 3 $\mu m^3 \, kg^{-1}$ respectively. Loch Lossit recorded 2.8 $\mu m^3 \, kg^{-1}$ whilst Loch Leathann had the lowest magnetic susceptibility with 1.8 $\mu m^3 \, kg^{-1}$. The replicated Finlaggan Marsh showed excellent correlation across the biostratigraphical boundaries and both had a maximum concentration of $1.1 \mu m^3 \, kg^{-1}$.

3.6.7 Loss on Ignition

The method followed that outlined by Bengtsson and Enell (1986) and provides a rough indication of the amount of organic matter in a sample. The inverse of the % organic matter lost to ignition provides an estimate of % mineral matter remaining in the residue. Two grams of dried sediment were weighed in a numbered porcelain crucible and placed in an electric muffle furnace at 350° C for eight hours. After removal and cooling in a desiccator the sample was re-weighed and the result was calculated as:

$$\% \text{ Loss on ignition} = \frac{(\text{wt of oven dry soil used- wt ignited residue}) \times 100}{\text{wt of dry soil used}}$$

The residue remaining after loss on ignition was retained for examination of tephra shards. Owing to the suspected presence of tephra shards in the Finlaggan Marsh sediment, 350° C was chosen as the optimum temperature for the non-volatilization of shards. (V. Hall pers. comm).

3.6.8 Summary of results

Loss on ignition was routinely undertaken and the greatest LOI was recorded in the sediment from Loch Bharradail at 40% which in comparison to Lochs Lossit (35%), Finlaggan (20-25%) and Leathann (30%) shows a greater percentage of organic matter.

3.6.9 Determination of pH within sediment sections

The analysis of pH was undertaken to determine the scale of alkalinity and acidity in specific sediment sections. The pH potential within sediment sections may allude to processes that have occurred within the sediment caused by natural chemical changes or external conditions such as acid-mine drainage. It is beneficial to obtain pH measurements on fresh sediment from newly extruded cores, usually by inserting the electrode of the pH-meter into the sediment. However due to the nature of the corer this method was not possible. Measurements of pH were undertaken only on dry sediment (10g dw.). In a comparative examination between wet and dry sediment, measurements taken on wet sediment were seen to be in the order of 0.80 pH units higher. The values were determined on a Pye Unicam digital pH meter and represent dry sediment values ranging between pH 3.5-5.5 within the four lochs examined. Although informative, single pH readings cannot reflect the past trophic conditions of a lake. For this, diatoms are a more sensitive indicator of changes in acidity (and alkalinity). Diatom analysis is a highly specialised subject and whilst recognising their potential for this study, owing to the constraints of time and expertise, they could not be examined.

3.6.10 Summary of results

The pH values obtained within this study are partially dependent on sediment type. For example the Finlaggan Marsh sediment is by its nature physically different in parts with a high mineral content. This combined with a high sulphide component has promoted a very low pH (2.5) in the basal region of Finlaggan Marsh Core 2. Lochs Lossit, Bharradail and Finlaggan have values ranging between 4.4-5 pH units. The maximum pH was recorded in Loch Leathann (range between pH 5.2-5.6) which has been attributed to the buffering effects of neutral spring waters flowing from the nearby Dalradian Limestone.

3.7 Chemical Analyses

3.7.1 Introduction

The 'total' content of polluting elements in soils and sediments can be a useful parameter in characterizing the contamination intensity. The diagnosis of contamination levels essentially depends on the adoption of a suitable procedure for sample pre-treatment rather than the choice of analytical technique. The choice of pre-treatments is varied and essentially forms two approaches; these are either 'total' or 'sequential'. There is no method that systematically yields higher results for all metals although methods incorporating HF in combination with aqua regia (see below) result in the greater liberation of heavy metals into solution (Cottenie and Verloo 1984). Commonly, such methods are more appropriate for partitioning different phases with mineral and organic fractions. Since this study is not concerned directly with biological effects on living biota (e.g. plants and animals) a 'total' extractive approach has been adopted, but it is recognised that sequential extraction techniques are superior in estimating the concentration of partitioning extractable elements.

3.7.2 Atomic Absorption Spectrophotometry

Atomic absorption spectrophotometry (AAS) involves monitoring the absorption of particular light frequencies by dissociated atoms. The light energy spectra which any element can absorb, will vary according to the energy potential of the atoms within the sample. Atomic characterisation is achieved when the atoms are reduced to a "ground state" whereby the atoms are reduced by dissociation to a minimal energy level. The ground state is achieved by excitation in response to thermal energy, causing collision of atoms and energy loss. The main components of the apparatus are shown in Figure 3.3.

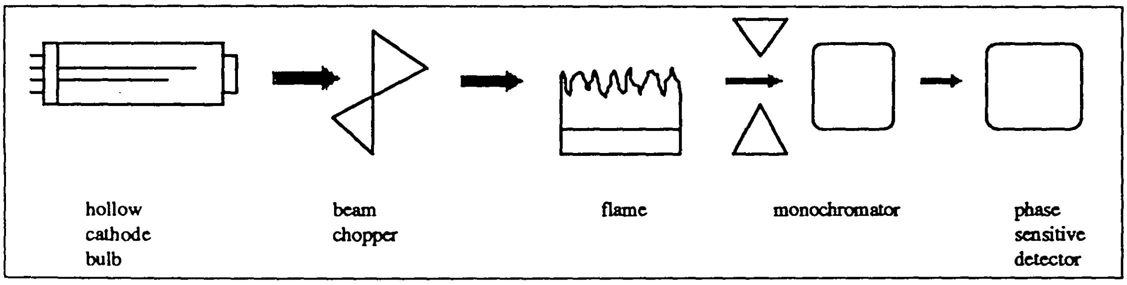

Figure 3.3 Schematic diagram of a single beam atomic absorption spectrophotometer, after Harrison *et al* (1991).

3.7.3 AAS Operating procedure

The instrument used was a Pye Unicam SP9 with on-line computer. Operation procedures and instrument calibration followed the specifications outlined in the instrument operation manual (Whiteside 1979). Optimal wave-lengths and lamp currents varied according to those specified under normal operating conditions. Wave-length, lamp current and band-pass frequency were standardised to ensure precision between replicate measurements across the different sample batches. Table 3.1 summarises the concentration of standard solutions, wavelengths, and band-pass widths used in the analyses.

Element	Wavelength (nm)	Band pass	Standard Conc. (ppm).	Sample dilution
Pb	217.6	0.5	1-5-10-25-50	none
Zn	213.9	0.5	1-5-10-25-50	none
Cu	324.9	0.2	1-5-10-15-20	none
Ca	422.7	0.5	5-10-25-50-100	none
Mn	279.5	0.2	10-25-50-100-250	none
Fe	248.3	0.2	10-25-50-100-250	1-10

Table 3.1 Standard solution concentrations, sample dilution and wavelengths for elements.

The volume of the analyite was standardised to 25 ml. The Fe concentration in the Loch Bharradail sediment was extremely high and beyond the range of the detection limit of the apparatus, therefore the analyite was diluted by a factor of 1 part analyite to 9 parts de-ionised H_2O to bring the Fe concentration down to within a range of optimum absorbence frequency. Five calibration standards were used using *Spectrosol* (BDH) standards, which were made up daily for each sample run. Final absorbancy readings (ppm) were converted using the formula described below, allowing the metal concentration in 1g of dried sediment to be determined:

$$\frac{\text{Absorbance (ppm) x volume of extractant (ml)}}{\text{dry weight of sample digested (g)}}$$

$$= \quad \text{Cu } \frac{(0.169 \text{ ppm x } 25 \text{ ml})}{0.56 \text{ g.}}$$

$$= \quad \text{Cu } \underline{7.54 \text{ mg kg}^{-1}}$$

mg kg^{-1} (equivalent to 1 g of dry sediment)

The trace-metals Pb, Zn, Cu, (total) and the cations Ca, Fe and Mn (total) were measured and are expressed as mg kg^{-1} based on 0.5g of acid digested sediment. Cadmium could not be measured owing to its extremely low concentration. The *'standard addition methods'* were applied (Whiteside 1979) to attempt a measurement but this was abandoned owing to the inherently low concentration (estimate <001-005 ppm) which appeared to be below the detection limit of the apparatus. A recent hydrochemical study of stream sediment taken from a small burn that runs through the Portnealon mine into Loch Finlaggan failed to detect Cd on identical AAS equipment (Short 1994). This lends additional support to the theory that if Cd was in extremely low concentration within catchment soils it is therefore likely to be low in the loch sediment. The results of the chemostratigraphic profiles are expressed in depth versus concentration graphs along with a general summary chart and are shown in the appropriate pages within Chapters 5,6,7 and 8.

3.7.4 Pre-treatment and decontamination of glassware and equipment

To minimise the risk of spurious results all glassware was thoroughly acid-washed to minimise sample contamination using the following procedure, (Bryant 1993 pers comm). All glassware was heated in 8M HNO_3 in a 5L Pyrex beaker for 2-3h at 90° C. After cooling, the glassware was transferred to a 5L beaker and reheated to 90° C in de-ionised water for 2-3h. After this period the glassware was rinsed in de-ionised water and placed in a drying oven. Plastic-ware was allowed to soak overnight in cold 8M nitric acid and rinsed thoroughly in de-ionised water before use.

3.7.5 Acid digestion procedure prior to analyses by AAS

The method followed that used by the University of Ulster Freshwater Laboratory, (Rippey 1992 pers comm). A hot extraction method was used to digest the sediment using $HNO_3/H_2SO_4/HCLO_4$ acids in the ratio 5:1:1 (vol/vol/vol). This is a pseudo-total digestion method, removing metals from all but the most resistant fractions of sediment (these fractions being the mineral lattices, which can only be brought into solution using HF).

Using acid washed glassware throughout, dry sediment was weighed to 0.5 grams in a 10 ml Teflon beaker. Five ml of concentrated nitric HNO_3 (AnalaR grade, BDH Chemicals) was added to the sample and allowed to stand at room temperature overnight. The sample was then heated and the acid evaporated to near dryness. The sample was further treated with a mixture of five ml of HNO_3, 1.0 ml of 60% $HCLO_4$ and 1.0 ml of concentrated H_2SO_4.

The mixture was digested at moderate heat until the liberation of brown NO_2 fumes had ceased. The solution was further digested for ten minutes after the appearance of white perchloric fumes. The digest was diluted to about ten ml of deionised water and brought to boiling point for the inclusion of Fe and Mn. The digestate was filtered via Whatman No.41 filter paper. Following this, the solutions were made up to volume using 25 ml of 1M hydrochloric acid.

Duplicate samples were digested for each sediment section. Two blanks were obtained with each sample batch, by using the reagents as in the digestion procedure, without the sediment. Samples and blanks were placed in a dark cold store to minimise the risk of alteration. The mineral residue from the digestate was retained for the examination of tephra shards.

3.7.6 Quality control assessment procedure: a comparative test

In order to establish the reliability of the analytical procedures employed in this study, comparison between two extractive techniques was undertaken. The hot acid extraction technique described above ($HNO_3/H_2SO_4/HCLO_4$ acids in the ratio 5:1:1) was compared against an 'in-house' reference soil routinely used by the Department of Environmental Science, University of Bradford. The reference soil (Tadcaster Reference Soil, hereafter TRS) has a certified value of 312.36 ppm total Pb obtained by AAS using acid digestion with Aqua Regia (HCL/HNO_3 in the ratio of 3:1). The object of this exercise was to (a) establish the overall difference between the two digestive reagents used to digest the TRS. This would allow comparisons of the efficiency of the oxidising reagents between the two reagents and (b) reaffirm that operating techniques were of the same standard as the Bradford laboratory using the same AAS instrumentation, calibration and calculation of the results. The results of a comparative exercise are outlined below.

The digestion procedure using Aqua Regia was as follows. Using acid washed glassware, 5 duplicate samples were produced in a batch of five using 1.0 g of the TRS and allowed to stand with 10 ml of the Aqua Regia on the hot plate. The heat was increased allowing the solution to boil to near dryness. The samples were made up to volume with the addition of 100 ml of de-ionised water and allowed to cool. The digestate was then filtered through an ashless filter (Whatman 41 grade) into a 25 ml plastic 'Universal' container.

3.7.7 Summary of the results of the recovery test

The results expressed in parts per million (ppm) show that there is a significant difference between the two extraction methods and variation within the sample batches themselves. The Coefficient of Variation (CV) expressed as a percentage of the standard deviation divided by the mean takes account of the variation of values within the sample batch. The CV value for the nitric/perchloric digestate has been established with the omission of the 620 ppm value that is an outlier.

TRS (Aqua Regia) ppm	TRS (Nitric/Perchloric) ppm
360	260
350	260
410	220
320	**620**
450	300
370	160
410	170
400	200
440	240
310	330
mean **382**	mean **237**
Sd **45.3**	Sd **53.4**
CV **12%**	CV **22.4 %**

Table 3.2 Summary results of recovery test using Aqua Regia and Nitric/Perchloric digestion techniques.

The Department of Environmental Science, University of Bradford, recovered 312.36 ppm Pb_{total} (SD 5.21%) based on 20 replicates supported by British[2] and European[3] soil reference standards and their total is based on the mean of five batches. The mean value in this experiment using Aqua Regia on the TRS was 382 ppm, (column 1 in Table 3.2) showing that there has been good recovery of Pb using the Aqua Regia digestion technique. The mean value resulting from the nitric/perchloric method is lower and there is greater within-sample variation as observed by the greater CV value.

In general two conclusions can be made: (1) the nitric/perchloric method is susceptible to greater within-sample variation owing to the 'matrices' or chemical effects that have a tendency to suppress or promote the liberation of a given ion into solution. (2) The AAS apparatus was running to its maximum efficiency, calibration to known standards and computation of data is to a reliable standard shown by the values achieved in this experiment. It is well known that the Aqua Regia method is favoured as a common technique due to the

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[2] British Standard Reference Soil No. 2704

[3] European Standard Reference Soil No. 1646

reagent's ability to be a strong oxidising agent, (J Farmer 1994 pers comm). The nitric/perchloric method is a tried and tested alternative digestion method on lake sediment and is used routinely by the University of Ulster Freshwater Laboratory. Comparative tests by this laboratory using this digestion method against a stronger HF digestion technique were found to give comparative results of recovery in total metal concentrations (B Rippey 1992 pers comm).

3.8 Dating the Environmental Record

3.8.1 Introduction

This section summarises the methods commonly applied to dating sediments. The majority of earlier published work concerning metals in sediments interpreted the changes in the palaeoenvironmetal environmental record only qualitatively (e.g. Mackereth 1966). More recently researchers have relied on radiometric dating and in some cases varve and tephra identification as a means of establishing reliable time-frames. For sediments to exhibit a reliable record of historically derived pollution, several prerequisites have to be met in order to establish meaningful chronologies, the absence of which will only allow qualitative assessment. Radiometric dating methods using ^{14}C, ^{210}Pb and ^{137}Cs represents an important, if not the most important technique for establishing chronologies based on annual rates of sedimentation.

3.8.2 ^{210}Pb radiometric dating method

The ^{210}Pb dating technique is commonly used in palaeolimnology and in some circumstances can provide an accurate chronology spanning the last 150 years. The determination of an annual rate of sedimentation allows a date to be placed with relative confidence providing disturbance to the upper portion of the core has been minimal. Usually proposed 'dates' can be correlated with known events or changes within the landscape or lake catchment. Palaeomagnetic, stratigraphic, macro fossil and pollen analyses have been used to support ^{210}Pb time scales. Further support in some cases can be gained using the radio isotope ^{137}Cs, a product of recent nuclear fission.

There have been numerous applications of the ^{210}Pb dating technique to the determination of sedimentation rates in off-shore, coastal marine, lake and marsh environments (Krishnaswamy 1971; Pennington *et al* 1976; Farmer 1977, Bryant *et al* 1993; Thompson *et al* 1993; Varvas and Punning 1993). For a general review of the methodology and models applied in determining sediment rates see Wise (1980); Olsson (1986) and Oldfield and Appleby (1984).

^{210}Pb is a natural isotope [half life (t½) =22.3 yr^{-1}] and forms part of the ^{238}U decay series (Krishnaswamy 1971). Disintegration of the intermediate isotope ^{226}Ra [half life (t½) = 1622 yr^{-1}] yields the inert gas ^{226}Rn. This in turn decays [half life (t½) = 3.38 days] through a series of short-lived isotopes to ^{210}Pb and is supplied to lake sediments as part of the particulate erosive input. The ^{210}Pb formed by *in situ* decay of this radium is termed the 'supported' ^{210}Pb and is normally assumed to be in radioactive equilibrium with the radium. There are other routes of ^{210}Pb into sediment and these are shown in the ^{210}Pb pathway diagram below.

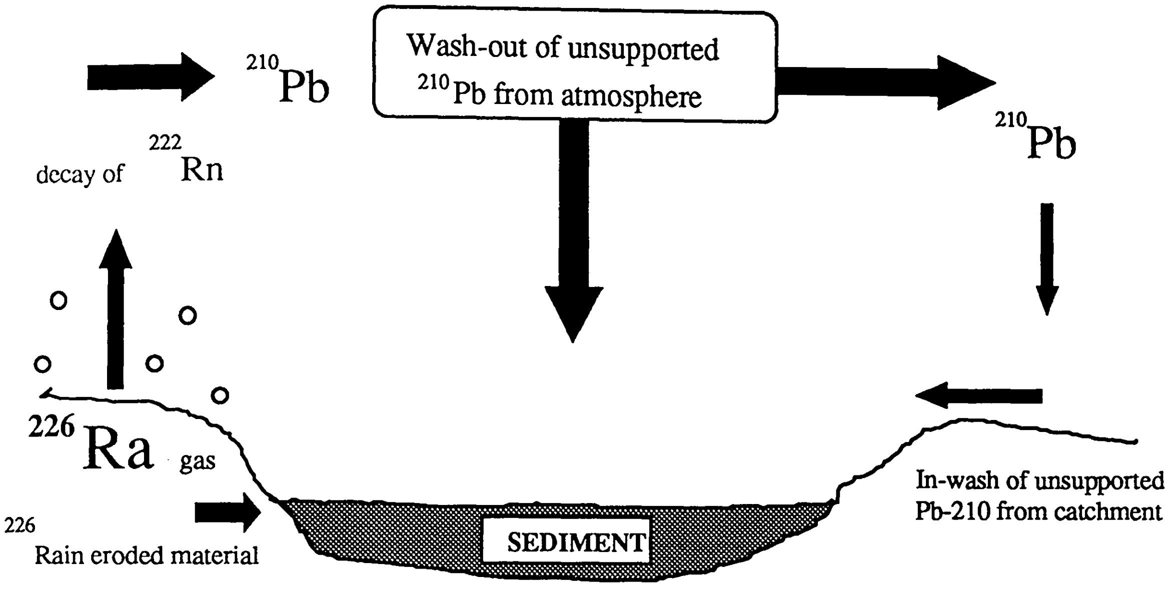

Figure 3.4 Simplified pathway by which ^{210}Pb reaches lake sediments after (Oldfield *et al* 1984).

^{210}Pb activity in excess of the supported activity is called the 'excess' or 'unsupported' ^{210}Pb. In dating by ^{210}Pb it is the unsupported component only which is used, since once incorporated into the sediment it decays exponentially with time in accordance with its half-life. The supported ^{210}Pb activity is estimated by assay of the ^{226}Ra. Once the supported ^{210}Pb activity has been estimated, the unsupported ^{210}Pb can be determined by subtraction from the ^{210}Pb 'total' activity. There are a number of models that have been developed and these are reviewed in detail by Appleby and Oldfield (1978, 1984). The method employed in this study is outlined in detail below.

3.8.3 ^{137}Cs as a tool for chronological support

Research beginning in the late 1960s has shown that ^{137}Cs deposited across the landscape by fallout from nuclear tests is a unique and useful tool for studying recent erosion and aspects of sedimentation and intra-site core correlation (Pennington 1976; Davis 1984 and Appleby, *et al* 1990). Radiocaesium [half life (t½) = 30 yr^{-1}] emits a strong gamma-ray (662keV) making its measurement in environmental samples relatively easy and accurate without special chemical preparation or separation. ^{137}Cs has been used as a tracer to measure and delineate patterns of erosion and deposition within the landscape to provide data on erosion processes (cf. extensive bibliography in Ritchie *et al* 1990). Radiocaesium has also been used to determine the sediment rates in a wide variety of depositional environments for sediment deposited since 1954.

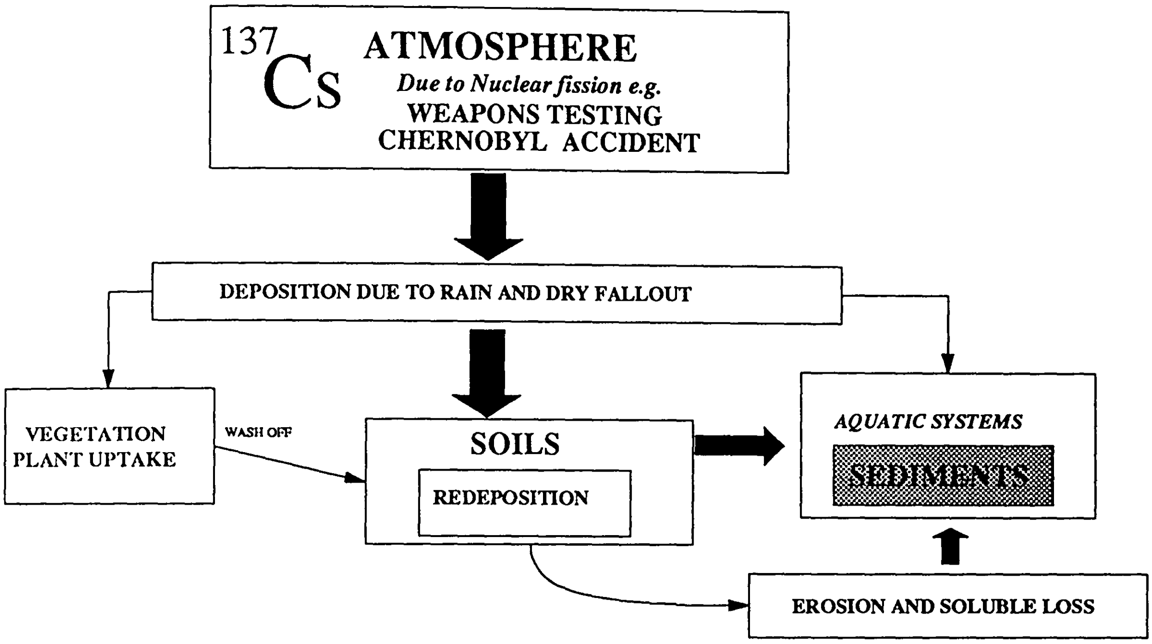

Figure 3.5 showing the routing of ^{137}Cs into sedimentary environments.

Providing data on erosion rates and on sediment accumulation rates, ^{137}Cs is a unique tool for studying the complete erosion and sedimentation cycle across the landscape. Recent research on Chernobyl derived ^{137}Cs distribution and mixing in Scottish freshwater loch sediment has highlighted the problems with this isotope's mobility in organic and clay-rich sediment (Bryant *et al* 1993).

Global dispersal of ^{137}Cs commenced in 1952 with high-yield thermonuclear testing. Periods of lower fall-out have been recorded and related to moratoria on nuclear weapons testing between the years 1958-1961. The Chernobyl accident in 1986 resulted in extremely high levels of ^{137}Cs fall-out across many parts of NW Europe (Clarke 1988).

3.8.4 Pollen analysis as a dating support for ^{210}Pb chronologies

Often pollen analysis has been used as a time horizon in lake sediments to support ^{210}Pb chronologies where the pollen type is derived from well established events within the lake's catchment.

In the Great Lakes region, North America, changes in pollen history have been recognised as a useful regional isochrone. Forest clearance by early 19th-century settlers resulted in the rapid spread of ragweed (*Ambrosia*). Marked increases in Ambrosia pollen values within the sediment profiles have been related to this period and used widely as a corollary to support other data such as increases in trace metal influx where no radio-metric dating was applied. Another much used synchronous event dated to the early 1930s is the decline in chestnut pollen. Chestnut (*Castenea*) pollen production declined as a direct result of Chestnut blight that was endemic throughout North America and into Canada (Anderson 1974).

Pennington (1985) working on lake sediment from Coniston Water, Lake District, was able to show consistency with a pollen chronology and a radiometric time-scale in a study aimed at determining the impact of copper mining. An increase of oak pollen resulting from the maturation of former oak coppice (used for local charcoal production) and the rise in pine pollen as a result of local 20th century plantations, supported ^{137}Cs and ^{210}Pb chronologies.

3.8.5 X-radiography and tephra reconnaissance

X-radiography has been found to provide a rapid and economical method of locating very thin fine grained tephra of Icelandic origin in Scottish peats (Dugmore *et al* 1992a and 1992b). The method is also useful for assessing bioturbation, differing grain size distribution, amount of organic material and other biostratigraphical aspects of the sediment. Other stratified materials such as fine 'inwash' laminations have been seen to be useful for identifying marker horizons and other isochrones in archaeological and palaeoenvironmental studies (Buckland *et al* 1981, Dörfler 1992). Tephra from the Hekla 4 (H4) eruption (c. 2310 ± 20 BC) has been seen to be incorporated into peats from Caithness in the north of Scotland and Ireland (Dugmore *et al* 1992b and Hall *et al* 1994a, 1994b). Owing to the close proximity of Islay to the coastline of Antrim (c. 25 km) it seemed feasible that tephra might be located within Islay and if found, would then provide a very useful isochrone with the County Antrim sites.

An acid digestion procedure described by Pilcher (1992) was adopted. Optical examination using a geological microscope was undertaken on 20 sub-samples. The Finlaggan Core 1 and the Loch Leathann Core 2 were examined for tephra horizons using a SCANRAY 120L machine at British Geological Survey, Edinburgh. Maximum resolution was obtained using various radiation exposure times according to the relative density of the sediment matrices within the cores. Exposure times varied between 50-90s using a range of between 25-35 Kv. The optimum current range was between 2-6nA according to sediment densities.

3.8.6 Results of X-radiography and tephra reconnaissance

The X-radiographic technique provided a 1:1 reproduction and showing horizons and the grain-size distribution of mineral phases within the lithostratigraphic unit of the cores. In the Loch Leathann Core, the X-radiographs showed a fine lamination at 23 cm which is attributed to fluctuations in water level due to the insertion of a small dam on the eastern shore of the loch at the turn of the century. With regard to tephra identification, the sheer volume of diatoms present within the samples presented a problem in the recognition of tephra shards. To overcome this problem attempts were made to filter out any tephra shards using a 75μ mesh sieve in an ultrasonic bath. Repeated washings down to 10μ mesh sieve failed to improve sample clarity. The sheer number of fragmented diatoms and other fine minerogenic particles made the task of identifying tephra so difficult the procedure had to be abandoned owing to the constraints of time.

3.8.7 The calculation of annual rates of sedimentation in this study

This procedure was undertaken at the Scottish Universities Research and Radiocarbon Dating Laboratory at East Kilbride. The samples for analysis were oven dried at 40°C and homogenised by grinding and prepared into pellets for analyses by instrumental γ-spectroscopy. The sediment was pelletised under 10 tonnes psi to produce a pellet with a diameter of 4.8 cm. Maximum counting efficiency is normally achieved on a standard pellet weight of 20g, (B. Anderson 1993 pers comm.) However, there was insufficient sediment within the sediment sections to achieve this weight. The pellets ranged from 8-15g. ^{210}Pb and ^{137}Cs activities were determined using a Low Energy Germanium Detector (LEGe.) which is a low energy photon spectrometer for the energy range of 3-500 ke V. The detection efficiency was determined for each of the nuclides by recording the gamma spectra of standards consisting of sediment 'spiked' with ^{210}Pb and ^{137}Cs. The bulk sediment was prepared from dried homogeneous loch sediment taken from the scrapped Ballygrant core at below 1m depth. Sediment at or below this depth will be depleted of Pb_{excess} and its homogeneity ensured comparable composition and density with the other samples.

Counting efficiencies covering the range of pellet weights are shown in Table 3.4 and the final results with their errors are listed in Appendix 2-5.

Pellet weight (gm)	% counting efficiency
5	9.28
10	7.42
12.5	7.22
15	3.73
20	3.37

Table 3.3 Counting efficiency of the individual pellet weights in the range of 8-15 grams.

The counting efficiency of the LEGe detector was established by counting individual pellet weights over a 48 h period. The counting statistics were then decay corrected to 1:9:92 and computed to establish the overall percent counting efficiency for each pellet weight and show that the larger the pellet's asymmetry the greater the effect of self absorbency of the γ nuclide resulting in a reduction in counting efficiency.

The data from γ counting was manipulated using the 'constant initial concentration' linear regression model (Appleby and Oldfield 1978) requiring log normalisation of the 'unsupported' ^{210}Pb activity which is expressed as Bq kg^{-1}. The log normalised data is then linearly regressed to determine the slope of the gradient line or best fit between the data points. The construction of the annual sediment rate for a given core was calculated according to:

= D/t, i.e. Sedimentation rate = Depth/time
$t = 1/S \times D$
$A_t = A_0 e^{-\lambda/s \times D}$
where A_t = activity at a given depth
and where A_0= activity at the surface
$\ln A_t = \ln A_0 - \lambda/S \times D$
$D = -S/\lambda \ln A_t + S/\lambda \ln A_0$
therefore, the gradient (m) = $-S/\lambda$
so, $S = -\text{gradient} \times \lambda$
where $\lambda = 0.693/t_{1/2}$ and $t_{1/2} = 22$ years

The total inventory of $^{210}Pb_{excess}$ and ^{137}Cs was established in order to estimate the amount of radionuclide flux to the surface of a given loch and its catchment surface, this being expressed as Bq m^{-2} yr^{-1}.

The total inventory has been established from the following:

Inventory(total) = Activity per gram of ^{210}Pb x the total number of grams in_each depth increment. The values are summed down the core till zero $^{210}\text{Pb}_{\text{excess}}$ activity is attained. The total sum is then divided by the area of the core (core area = 70.89 sq m).

The Flux was established from:
Flux = ^{210}Pb inventory x $\lambda = (0.693) \div t½ \, (22.3 \text{ yr}^{-1})$.

3.8.8 Summary of the results of ^{210}Pb and ^{137}Cs determination

The results from ^{210}Pb have allowed the construction of an annual sedimentation rate over the upper portions of the loch sediment (Table 9.4 and 9.5). Loch Bharradail had the greatest surface (0-1 cm) $^{210}\text{Pb}_{\text{excess}}$ maximum of 338 ± 28.4 Bq kg^{-1} at 1 sigma error. Loch Finlaggan had the lowest surface $^{210}\text{Pb}_{\text{excess}}$ concentration with 61 Bq kg^{-1} at 1 sigma error. No annual rate of sedimentation could be formulated for this core owing to the inverted nature of the $^{210}\text{Pb}_{\text{excess}}$ which has resulted from mechanical mixing of the upper portion of the core. The results from ^{137}Cs have been fairly successful in lending additional support to the ^{210}Pb-based chronology (Figures 9.7).

3.9 Pollen Analyses: Methods and Preparation

Detailed pollen analysis was undertaken on the Finlaggan Marsh core (Core 1) and on the 1-30 cm sections of Loch Bharradail, Leathann and Lossit sediment. The objective of this method was primarily to develop supporting palynological time sequences by relating the Finlaggan pollen spectra to the regional pollen record in the historical period. The upper portion of the cores were examined for ***Pinus sp.*** pollen content to estimate the relative percentage frequency in relation to early 19th century pine plantations. It was hoped that this might provide a 'proxy' dating method to support the ^{210}Pb time-frame. Identification followed the keys of Moore and Webb (1978) and Moore *et al* (1993) with additional use of type slides of fossil and modern reference material. Nomenclature followed Clapham *et al* 1962. Relative pollen values were obtained by counting 300 pollen grains per slide. To aid the description and interpretation of the pollen stratigraphy at Finlaggan Marsh the diagram has been divided into a series of local pollen assemblage zones on the basis of changes in the proportions of arboreal to non-arboreal pollen (AP:NAP ratio) and of variations in the composition of these components (see Section 5.9.1 and Figure 5.43) The principal method used consists of a seven-step procedure incorporating a modification of that described by

Moore and Webb (1993) and the swirling technique devised by Hunt (1985). The method employed was as follows:

(1) Sample preparation

Four ml of distilled water was placed in a graduated nalgene centrifuge tube and displaced to 5ml by adding the sample. At this point 40 µl of 0.2g/50 ml of exotic marker spores (*Lycopodium clavatum*) were added from a previously prepared aliquot. The sample was centrifuged at 3,000 rpm for c.5 minutes and the supernatant was decanted.

(2) HCL Treatment

In order to remove free calcium carbonates approximately 6 ml HCL (10% w/v) was added to the sample and left until effervescence ceased. 6 ml of distilled water was added to the sample and centrifuged for five minutes and the supernatant poured off.

(3) KOH Treatment

KOH (Potassium Hydroxide) allows the break down of lignin and other vegetation matter within the sample. 6 ml of 10% KOH was added to the centrifuge tube and placed in a boiling water bath for 20 minutes. The sample was centrifuged for 5 minutes at 3000 rpm and the supernatant was poured off. Six ml of distilled water was added, stirred and centrifuged at 3000 rpm for 5 minutes.

(4) Sieving and swirling

The sample was poured onto a 150u nylon sieve with a 10u sieve placed below. The sample was gently washed with filtered tap water. The vegetation flot was retained for later analyses. The residue contained on the 10u sieve was poured onto a large clock glass. The swirling action allows any inorganic particles to remain in the centre of the glass whilst the supernatant can be gently poured back into the 10u sieve. This process was repeated four or five times according to the amount of inorganic material present in a given sample. Residues were further centrifuged and a pellet formed in the base of the centrifuge tube.

(5) Hydrofluoric acid (HF) treatment

This method was only used on samples that exhibited a high degree of inorganic material such as clay or silica particles. The supernatant was decanted and 6 ml HF was added to the sample. The pellet was stirred using a wooden stick and placed in a boiling water bath for 10 minutes or up to 15 minutes if the sample displayed a gritty texture. The sample was centrifuged at 3,000 rpm for five minutes and the supernatant neutralised with NaOH (Sodium hydroxide) in a plastic beaker. The pellet was resuspended in 6 ml 10% HCL and placed in the water bath to warm. The supernatant was poured off after centrifuging at 3,000 rpm.

(6) Acetolysis

The sample was dehydrated by adding 6 ml glacial acetic acid to the pellet. After spinning and decanting 6 ml of acetolysis mixture (9:1 ratio of acetic acid and concentrated sulphuric acid) was added to the sample tube and placed in a water bath for 1 minute. After spinning and decanting, the pellet was resuspended in 6 ml of glacial acetic acid and centrifuged. The supernatant was poured off and washed by centrifuging in distilled water.

(7) Staining and mounting

The water was decanted off and 6 ml of warmed Methylpropan-2-ol was added with one drop of safranin stain. The pellet was mixed well with a stirring stick and allowed to stand and then centrifuged at 2,000 rpm and decanted. A further wash with 6 ml of Methylpropan-2-ol was carried out before decanting into 5 ml plastic tubes. Several drops of silicone oil were added to the tubes. The tubes were placed in a previously warmed drying cupboard and allowed to stand for 24h before the pollen samples could be mounted.

3.9.1 Summary of results

The results from pollen analyses have provided a detailed pollen diagram from Finlaggan Marsh (see Figure 5.41). Biostratigraphic changes within the profile can be positively linked to events in the pollen spectra. This evidence has allowed assumptions to be put forward regarding the development of the marsh and subsequent hydrological and geomorphological changes brought about by mining, farming and more recent afforestation within the catchment of the loch. The use of pollen as a relative dating method to support the ^{210}Pb-derived annual sedimentation rates proved to be of limited use owing to taphonomic problems with *Pinus* sp. pollen grains and their low abundance over the upper portion of the sediment sections in the loch cores.

3.10 X-ray Diffraction Analyses

3.10.1 Introduction

This method can identify specific crystalline chemical compounds with a high degree of certainty and is commonly used to identify minerals in aquatic sediments, (Davidson *et al* 1985 and Harrison *et al* 1991). It is particularly well suited for use with clay minerals, some of which are very similar in composition, whilst other minerals have discrete structures that provide characteristic diffraction patterns. These patterns are obtained from the fact that the atoms within a crystal are within a regular arrangement of planes which act as a diffraction grating for X-rays (Bragg equation). When the conditions of the Bragg equation are met, X-rays may be 'reflected' from planes of atoms. Up to 12 samples (3-4 g dw.) were placed in stainless steel containers housed in a carousel. Each sample forms a mass of finely divided crystals in a random orientation which is rotated to maximise the number of orientations. Each set of mineral crystals has its own set of d-spacings that provide a unique diffraction pattern which is detected and matched statistically to an on-line record of diffraction data. Graphical representation of match-score statistics are then produced to show the mineral composition of a given mineral (Harirson *et al* 1991, 246).

3.10.2 XRD on selected mineral and sediment sections

In this study, X-ray diffraction was undertaken using a Philips PW 1800 on three mineral fractions surrounding a piece of galena recovered from a spoil tip at the South Ardachie mine close to Loch Bharadail. The main objective was to understand the composition of these secondary minerals (gangue materials) associated with the lead ore. An understanding of their mineral composition would allow an insight into the weathering potential of the mine waste. The resulting contamination around the site will ultimately have a direct bearing on the elemental concentration within the sediment profile and in theory be greatly influenced by lead-rich leachates entering the loch via fluvial pathways. Three separate mineral fractions were identified in association with the galena. The sample of Pb ore was cut on a petrology rotary cutter in preparation for mineral separation. These were separated from the ore and ground to a fine powder (<63u) with a pestle and mortar and at this juncture were ready for insertion into the apparatus.

A pilot study was also undertaken to identify the mineral phases within specific sediment sections of the Loch Bharradail and Loch Leathann cores. Both lochs share similar geologies but the land-use histories have differed to the extent that mining has occurred only in the Bharradail catchment so in theory X-ray diffraction ought to provide an index of minerals associated with lead mine derived solutes. The method was less successful in identifying the mineral phases within the sediment sections compared to the lead ore. Twelve samples from specific sediment sections based on the results from AAS were processed and the results have identified quartz and muscovite as the major mineral constituents in the loch sediments. The procedure failed to identify minerals that could be associated with the suite of minerals recorded in the grab sample from the South Ardachie spoil tip. For the XRD technique to work with any success, minerals have to be present in greater than 1% concentration (J.Angel 1994 pers comm.). Researchers elsewhere have also had varying success using lake sediments to identify mine wastes (Kemp *et al* 1976 and Davison *et al* 1975).

3.10.2 Summary of results

The results of the X-ray diffraction analysis provide useful information regarding the composition of a grab-sample of lead ore taken from mine tailings at the South Ardachie leadmine. These results (see Appendix 6) show the primary and secondary constituents within the sample. Some of these constituents are easily oxidised (e.g. pyrite) to form extremely local acidic conditions leading to increases in heavy metal contamination as these products weather. Additional analyses on specific sediment sections within the cores have provided qualitative information on principal mineral suites. In the case of the Finlaggan Marsh Core 2, a semi-quantitative approach was adopted to reflect the mineral abundance in the minerogenic sections of this core (see Table 5.5).

3.11 Conclusions

This chapter has described the methods and techniques that have been employed in this thesis. The structure of the research design has been outlined in Figure 3.1. A more detailed discussion of the results of each method is described in the appropriate sections of Chapters 5, 6, 7 and 8. The adoption of an integrated approach combining archaeological field survey, analytical chemistry, palynology, radiometric dating procedures, geochemical and geological analyses was essential to assess the input of contamination from mine workings into the loch sediment. When required the techniques used and the sampling procedures have been modified according to the requirements of the analytical investigation. Routine sediment pre-treatments and chemical handling have adhered to a strict regime of de-contamination of all glassware to minimise technical error. The study has benefited from the ample volume of sediment available within each sediment section using the coring device described. Had a conventional corer (i.e. Mackereth piston type) been used then in all probability there would not have been sufficient material with which to undertake such a detailed programme of analyses.

The topographical surveys have been useful in providing the scale and extent of mine tailings in the vicinity of the lochs and have made a useful contribution to the data base of historical mining in a lesser known lead mining region of Scotland. The next chapter will investigate the history of lead mining in Scotland and place the sites within their environmental setting and in a wider context of lead mining in Great Britain.

CHAPTER 4: ISLAY, LOCATION AND PREVIOUS WORK

4.1 Introduction

For two centuries (*c.*1300-1500) Islay was an important maritime power base for the Lords of the Isles. The social and economic lifestyle of the Lordship is now better understood mainly as a result of six years of archaeological research by the National Museums of Scotland (NMS) on two fortified islands at the northern end of Loch Finlaggan (Caldwell and Ewart 1993). The economic and archaeological potential of the region is rich. Prehistoric archaeology is represented by numerous crannogs, with hill forts and duns located in prominent places (RCHAMS). Elsewhere there is a wealth of monumental archaeology dating to the Early Medieval period but in the preceding Dark Age the archaeology is poorly understood. There is a noticeable dearth of Norse archaeology, although the island abounds with Nordic place-name evidence. Visiting geologists interested in the complex structure of Islay have often repeated the statement that lead was mined in the Norse Period, but archaeological evidence has yet to support these claims.

Section 4.2 of this chapter will examine the structure, geomorphology and vegetational history of the region to place the study area in its environmental setting. A review of recent palaeoenvironmental work is given. This sheds new light on the palaeoecology of the island and forms the backdrop for palynological work undertaken at Loch Finlaggan Marsh. Recent paleoenvironmental reconstructions in the western part of Islay have been undertaken using pollen analyses. This data, combined with environmental archaeological research from the north-east region has shown that the island had a diverse woodland cover up until the Bronze Age period. The considerable differences in vegetation communities on the west and east of the island appears to be largely the result of contrasting soils that have formed as a result of different bedrocks. In the east the Dalradian complex consists of mainly fertile limestones whilst the west is made up of the acidic Lewisian Gneiss. Geological deposits of economic importance are to be found associated mainly with the Dalradian Limestone groups.

Section 4.3 will discuss the implications of previous geochemical survey across the study region. It has been mentioned above that data from this research has been re-examined in order to estimate the geochemical distribution of lead, zinc and copper in those areas previously mined and within parts of the catchment chosen as a control. This is followed by a brief review of current archaeological research.

Section 4.4 will examine the implications of geochemical exploration in NE Islay. This was undertaken between 1973 and 1983. More recent work undertaken by the British Geological Survey and others will aslo be discussed. Section 4.5 will briefly review recent archaeological work. Section 4.6 will assess and examine the historical significance of lead mining in north-east Islay. It was stated in the opening chapter that an analogue approach to the identification of early mining needed to pay close attention to the more recent periods of documented lead extraction. This objective has led to the examination of historical accounts and other statistical data in order to glean information on the timing, scale and extent of mining operations. This information provides much of the historical background and is a fundamental component which lends additional support to the interpretation of the archaeological record which is at best ill defined and at worst notoriously fragmentary.

4.2 Environmental Setting

4.2.1 Location

Islay rests between the coast of Antrim, Northern Ireland and the Argyll peninsula of western Scotland (Figure 4.1below). Islay is the southern most island of the Inner Hebrides and is situated 13 miles (20.8 km) from the mainland and about 23 miles (36.8 km) from the north east coast of Ireland. Islay lies to the west of the island of Jura from whic it is separated by a one mile wide channel called the Sound of Islay. The island is 19 miles (30.4) wide and 25 miles (40km) from north to south and is almost divided in two by Loch Gruinart to the north and Loch Indaal to the south. The climate is maritime and influenced by predominantly westerly winds. The average rainfall tends to be high with between 1300 and 1600 mm per year.

4.2.2 Geological setting

The structure of Islay has been the focus of much attention by geologists in recent years owing to the sheer complexity of different lithologies that make up this region. Three principal components form the structure of Islay.

The eastern side of the island comprises mainly Dalradian metamorphic rocks such as quartzites, limestones and phyllites (Figure 4.2). The blue-grey limestones are generally to be found within the Lossit Limestone (formerly Islay limestone) and Ballygrant Formations that lie within the Blair Atholl sub-group of the Dalradian sequence. The major slates and phyllite sequences of Islay form part of the Bharradail Member of the above mentioned Ballygrant Formation.

The western region of the island is known as the Rhinns. This gives rise to the so-called Rhinns Geological Complex. Two principal lithologies form the structure in this region. The first is the Lewisian gneiss with the intrusive epidiorite and hornblende schists (Muir *et al* 1994). The complex geology of Islay has led to a landscape that is not dissimilar to the borders region of southern Scotland. Soils on Islay are young, mainly being formed over the last 10,000 years, providing fertile arable and improved pasture on the limestone with rough grazing in the more acidic regions. Extensive tracts of peat cover 25 % of the island

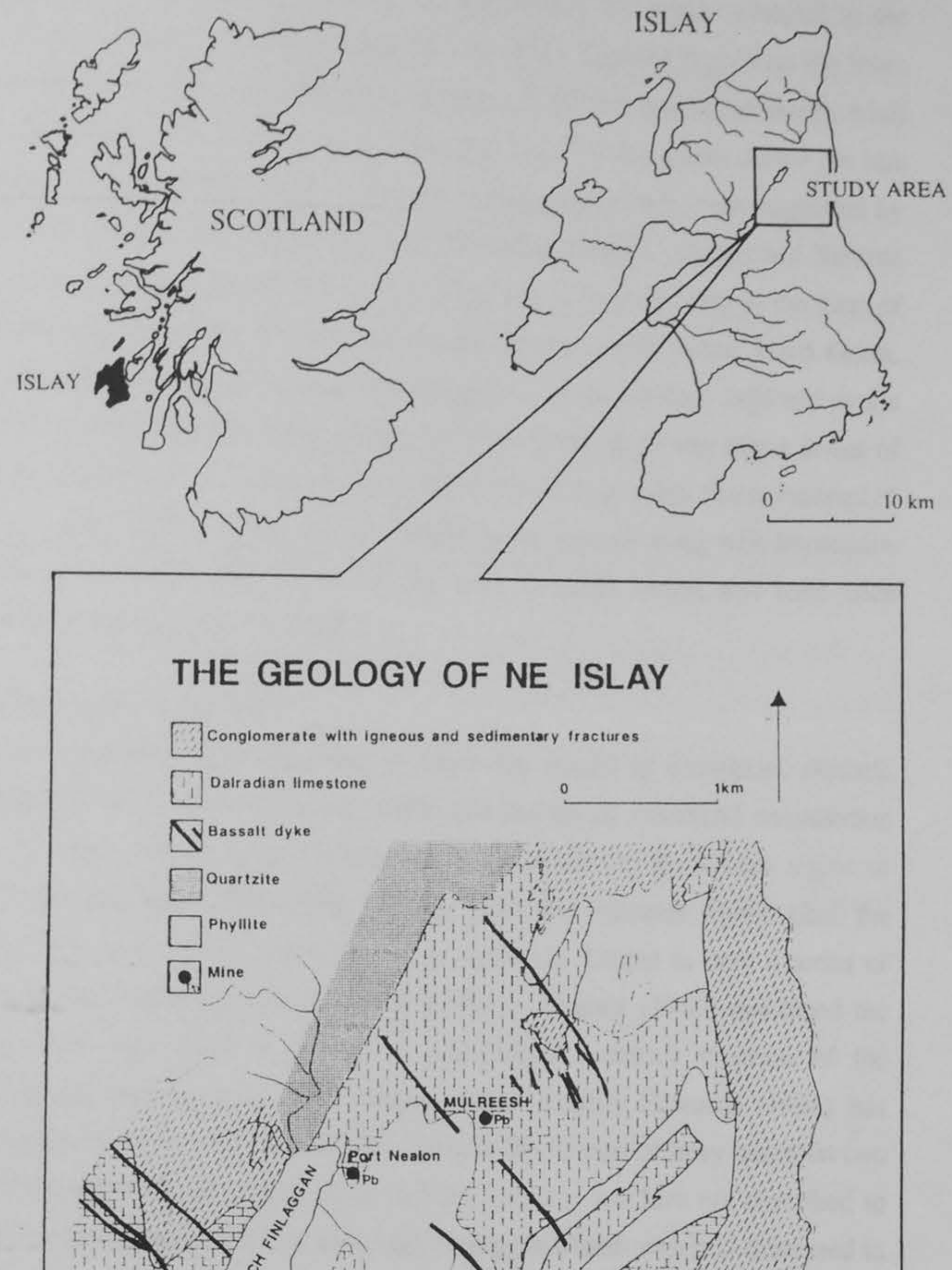

Figure 4.1 Location of Islay, study area and the local geology of NE. Islay.

4.2.3 Quaternary geomorphology and sea-level change

The cumulative results of glacial erosion during the Quaternary are widely recorded in the Inner Hebrides. Owing to their location immediately west of the Scottish Highlands the Inner Hebrides were mostly glaciated by the Highlands ice-sheet (Late Devensian ice-sheet). Mull and Skye are the exception, being covered only by locally nourished ice sheets during the last glacial period (Sissons 1974, 1977, 1983). The Loch Lomond Readvance was suggested by Sissons (1981) to have terminated amidst the Inner Hebridean islands. Synge and Stevens (1966) claimed to have identified the western extent of ice-limit in western Islay in the form of an end moraine complex that transects the middle of the Rhinns region below Loch Gorm. Dawson (1982) refuted their proposition with the recognition of an ice-limit adjacent to the head of Loch Indaal where an end moraine extends for some 8 km. It is here that a series of deposits occur as an assemblage of ridges and depressions that include many linear features of fluvio-glacial origin. Islay hosts a series of Late Glacial raised beaches along with impressive Post Glacial shorelines. These can be seen at the head of Loch Indaal and have been extensively surveyed by Dawson (1979, 1982).

4.2.4 Palaeoenvironmental background

Palaeoenvironmental research is now beginning to assess the impact of ecological, climatic and cultural factors that have effected the expansion and decline of woodland colonisation within the island. To date, palynological research has been confined to the Rhinns region of Islay on *Loch a' Conailbhe* and Cultoon Bog (Bennett 1989 and Edwards 1994). Until the work undertaken by Edwards (1994), palynological research was limited to only a series of summary pollen diagrams undertaken on truncated profiles. Bennett (1989) combined the palynological data from two cores to provide a post-glacial summary diagram of the vegetation history of the western side of the island. More recently Edwards (1994) has reconstructed the vegetation history of the southern region of the Rhinns of Islay based on two complete profiles. The vegetational history of the Rhinns, Oronsay and Jura are described to present a backdrop for recent palynological data from Finlaggan Marsh which is discussed in detail in the next chapter.

In Islay, the long history of peat extraction for fuel has invariably resulted in the loss of the upper sections of peat profiles resulting in incomplete cores and biostratigraphic sequences. To some extent this hampered the completion of a complete pollen sequence that represented the Holocene. This problem has been circumvented using cores from lochs or infilled lacustrine basins. A tentative picture of the local environment of the Rhinns of Islay was first constructed by Bennett (1989) who combined the pollen assemblages from Loch Conailbhe (3 m core) and from Coultoon Bog (*c*.5 m of ombrotrophic peat).

In the former, the pollen was degraded and in the latter the upper portions were truncated by peat cutting. Subsequently, Bennett was able to conclude that during the Postglacial Period, Islay was probably well forested with the exception of regions of higher altitude. *Ulmus* and *Quercus* became established along with the shrubs Guelder rose and buckthorn which are now extinct on the island. By about 7000 BP *Alnus* colonised the shores of mires in the region of the loch. Soil deterioration fostered the onset of heathland communities with the expansion of *Calluna vulgaris* which spread rapidly by 5000 BP. Contributing factors leading to the treeless landscape would have been climate, anthropogenic impact and a trend towards increasingly acid soil conditions.

A dated absolute (concentration) pollen diagram covering the period *c*.7500-4000 BP has been published from the Sorn Valley, Central Islay (McCullagh 1991). The work aimed to provide a back-drop to a Neolithic site at Newton and was also limited to the mid-Holocene owing to the truncation of overlying peat. The work showed that disturbance of woodland, possibly of anthropogenic nature, occurred at around 5000 BC followed by an increase in Coryloid pollen which may be the result of clearance by fire. These events are followed by a period of open woodland. The contribution of *Ulmus* pollen decreases at about 3150 BC and local woodland is soon cleared. A pit from the nearby archaeological site was dated by charcoal to 2930 ± 60 (GU-1951) and was sealed by two horizons both of which contained significant amounts of *Ericacea* pollen which was taken to suggest the spread of heathland associated with the onset of acidification possibly caused by overgrazing.

Following pilot studies by Agnew *et al* (1988) more detailed palynological work has been recently published by Edwards (1994) from the site of *Loch a'Bhogaidh*, Rhinns of Islay (National Grid Reference NR 225576). Six radiocarbon dates are available from specific events within local pollen assemblage zones (see Table 4.1 below). Two pollen diagrams (percentage and concentration) represent a limited Late Glacial and a fuller Holocene record. The earliest stratigraphic and palynological evidence for the site comes from assemblage zones LABI and LABII.

From these sections an open tundra-like environment is proposed with open-habitat taxa such as *Cyperaceae*, *Poaceae* undiff. *Lactuceae*, *Artemisia*-type, *Caryophyllaceae* undiff., *Rumex acetosa* and *Selaginella selaginoides*. Zone LABIIa records high values of *Betula*, *Pinus*,*Corylus avellana*-type, *Empetrum* and *Filupendula*. Open water is assumed and ascribed to the species *Myriophyllum alterniflorum* which suggests that the thermal environment was not too extreme.

Local pollen assemblage zone	Depth (cm)	Estimated date (^{14}C yr. BP)	Subzone	Depth (cm)	Estimated date (^{14}C yr. BP)
LABIg~*Betula-Corylus*	70-28	270-90	——	——	——
LABIf~*Poaceae-Cyperaceae*	262-70	1860-270	——	——	——
LABIe~*Betula-Poaceae-Cyperaceae*	366-262	3610-1860	——	——	——
LABId~*Betula-Quercus-Alnus*	462-366	5470-3610	LABId(ii)	418-366	4610-3610
			LABIc(i)	850-586	5470-4610
LABIc~*Betula-Corylus-Isoetes Lacustris*	850-462	9780-5470	LABc(ii)	586-462	6940-5470
LABIb~*Betula-Poaceae-Cyperaceae*	964-850	11070-9780	LABIb(ii)	910-850 964-910	10460-9780 11070-10460
LABIa~*Cyperaceae-Lactuceae*	184-964	12430-11070	LABIa(ii)	1024-964	11750-11070
			LABI(ii)	1084-1024	12430-11750

Table 4.1. Summary table of Local pollen zones and subzones at *Loch a Bhogaidh* (after Edwards 1994).

At this position Edwards records a high concentration of microscopic charcoal. He proposes that natural fires by lightning in a dry tundra-like environment seem most likely as an explanation for the increase. A basal date of 10910 ± 450 BP (I-15254) from the lowermost organic gyttja provides the onset of more temperate conditions. Hereafter, there is a rise in *Betula* and *Corylus avellana* together with *Juniperus communis* along with *Myriophyllium alterniflorum*, *Potamogeton natans*-type and *Isoetes lacustris* indicating lacustrine conditions at the site leading to the formation of gyttja. A terrestrial ground flora includes such taxa as *Poaceae*, *Cyperaceae*, *Empetrum*, *Rumex acetosa*, *Lactuceae*, *Filupendula* and *Plantago maritima*. There is also a sharp rise in *Betula* frequencies which date by extrapolation to *c.*10320 BP which corresponds well with mainland dates for *Betula* expansion between *c.*10400 BP and 9800 BP (Edwards citing Lowe and Walker (1977) See also Vasari (1977) and Birks (1977, 1980)). *Corylus avellana*-type expands at the expense of *Betula* (at *c.*10460 BP and *c.*9910 BP) which is comparable with a mainland date of 9230 ± 120 BP (Q-1416) at *Loch Cill an Aonghais*, Argyll (Edwards (1994) citing Peglar (1977) in Birks (1980)).

At the beginning of subzones LABI c(ii) and LABII d (i) *Betula-Corylus* woodland and *Salix* carr-type woodland appear to be the main arboreal element along with *Ulmus*, *Pinus* and *Quercus*. The expansion of *Ulmus* (5%) is dated to *c.*8760 BP, with *Quercus* (5%) appearing

earlier at *c.* 8620 BP. An *Alnus* rise is estimated at *c.*6570 BP which accords well with a date of 6250 BP for the rational alder limit at Newton in the Sorn Valley (McCullagh 1991).
For the period *c.*5470-3610 BP at *Loch a' Bhogaidh*, there is an increase in percentage and concentration of *Alnus* and a reduction of *Corylus avellana*. From *c.*5230 BP, *Ulmus*, *Pinus* and *Quercus* are reduced and an increase in *Fraxinus*, *Poaceae*, *Rumex acetosa* and *Ranunculaceae* occurs.

Charcoal frequencies are taken to represent Neolithic and early Bronze Age clearances which are coincident with open land herbaceous taxa. Edwards noted good synchroneity with a reduction in *Pinus* frequencies by *c.*3920 (382 cm) and the widespread British Isles decline of *c.*4000 BP. Between the period *c.*3610-90BP there is an extensive decline in woodland pollen alongside a non-arboreal pollen increase from 11.6% to 45.6%. Charcoal and cereal-type pollen underlines the role of Bronze Age clearances alongside the rise in mineral content caused by terrestrial run-off which is attributed to mixed arable and pastoral farming close to the pollen site. Alongside these events is a shift towards a deteriorating climate. The expansion of heathland (*c.*3610 BP) is attested by a reduction in woodland and shrub pollen (to around 5% TLP). The presence of *Sphagnum* may be taken to indicate that soils were becoming impoverished and blanket peat was spreading within the catchment. Edwards suggests that this was either a result of land-use pressure or perhaps as a result of progressive natural deterioration brought about by climatic change (Edwards 1994, 768).

Slightly further afield from the Rhinns of Islay, archaeological investigations in 1968 (Mercer 1968) from a washing-limit deposit of the highest post-glacial transgression at Lealt Bay, Isle of Jura (approximately 25 km NNE) incorporated limited palynological study. The 4 m undated profile showed that *Pinus* and *Betula* were decreasing from 10% and 85% respectively with a sustained increase in alder at 7500 BP (Boreal zone VI- Atlantic zone VIIa transition sensu Godwin). *Alnus* became locally dominant with *Ulmus* and *Quercus* present below 10%. At around 2500BP (Sub-Boreal/Sub-Atlantic transition zones VIIb/VIII *sensu* Godwin) *Ericacea*, *Gramineae*, *Cyperaceae* and *Sphagnum* all rise exponentially towards the surface. The significance of this work was that oak and elm never became locally dominant probably as a result of climatic gradients. This seems to have been the case in the Finlaggan catchment (Figure 5.43).

Jura's present lack of trees is a near-ultimate stage in a long process; the rate of deforestation has, however, increased markedly since the opening of the Sub-Atlantic period. On the basis of the then pollen-stratigraphical data, Birks (1977) proposed that within western Scotland three broad areas could be delimited on the basis of similarities in vegetational histories and mid-postglacial pollen assemblages. These are: a region of predominant oak forest with birch centred on the mainland of Argyll and south-west Scotland, including Jura, Arran, and eastern Mull and extending northwards to southern Skye; (b) a region of predominant birch forest with some and oak confined to Wester Ross; (c) a region of predominant birch forest characteristic of northern Scotland and the more exposed regions on the islands of the Inner Hebrides. Further work by Williams (1977) and Birks (1980) and Birks and Williams (1983) did not radically alter this model. Birks (1987) suggests that these mid post glacial patterns were almost certainly determined by broad scale natural climatic gradients of winter and summer temperatures, length of growing season and precipitation. Exposure to westerly gales is a further limiting factor for the suppression of arboreal communities.

Age (Years BP)	**Local pollen Zone**	**Predominant Vegetation**
9500-9800	LCq	Herbaceous communities.
6200-9500	LCr, LCs, AAe	Birch-hazel scrub with oak and elm: willow alder on damp soils.
4000-6200	LCt p.p. Strand	Birch-hazel scrub with oak and elm with willow and alder on damp soils.
3500-4000	LCt p.p.	Scrub clearances and pasture.
2600-3500	LCu AAg p.p.	Grassland and pasture-arable ?
1500-2600	LCv AAg AAh	Secondary birch scrub (Colonsay)
		Grassland and pasture (Oronsay)
500-1500	LCw AAj	Grassland, pasture and heath.

Table 4.2. Inferred Holocene vegetation history of Oronsay and Colonsay (after Birks 1987).

The vegetation history of Colonsay and Oronsay has been reconstructed from two radiocarbon dated pollen profiles by Birks (1987). These islands are only 12 km to the NW and it would not be surprising if their vegetation history in the early Holocene was similar to Jura and Islay. Birks summarises the results of two pollen profiles from a peat site of *An t-Aoradh,* (AA) Oronsay and *Loch Cholla* (LC) on Colonsay, these data are shown in Table 5.7. and shows that by 4000 BP, Birch-hazel scrub was predominant on Oronsay and Colonsay.

4.2.5 Conclusions

The contribution made by palynological work has increased our understanding of the vegetation history over the last two millenia. Although only limited work has been carried out, the results show the gradual trend towards open-heath which was brought about by three principal agents (a) human interference, (b) soil deterioration with a trend towards podsolisation and (c) suppression of tree colonisation due to strong westerly winds. More work of this nature needs to be undertaken on the limestone region in the north-east of Islay. Pollen analyses at Finlaggan Marsh (Figure 5.43) shows that this region is more floristically diverse than the more exposed western region of the Island. The north-east region is certainly more fertile (being predominantly calcareous) and sheltered in many parts. This would in all probability have promoted a richer vegetation pattern than we have seen in the previous studies. The relevance of other pollen studies to this thesis is that the pollen diagrams provide a vegetation history which is essential in order to compare the results of palynological investigation at Finlaggan Marsh. The following section will now review the economic geology and the importance of mining within the NE region of Islay.

4.3: Economic geology and previous work

4.3.1 Introduction

This section will now examine the different types of materials that have been exploited in the study region. The region has a rich mineral resource that has been periodically exploited in the past. Today only limestone is quarried on a commercial basis. The mineral wealth in the region has been recently explored by geochemical prospecting companies and an account of their findings is given. A brief survey of recent archaeological research in the region reviews the work undertaken at the historically important site of Castle Finlaggan which is a fortified island at the head of Loch Finlaggan and forms the subject of the next chapter.

4.3.2 Lead (Figure 4.2)

Lead has been mined from twelve narrow veins that each trend 035-135° over an area of 40 km^2 (Gallagher 1991). The lead mineralisation is associated with the Dalradian Limestone, dolerites and phyllites, known collectively as the Lossit Limestone[1], which outcrop around the village of Ballygrant (N. Ruckley pers. comm). Within the study region, three sets of veins, trending north-north-west and north-east have been worked, with ore being recovered from replacement bodies in several areas, but generally in close proximity to the tertiary dykes (Wilson and Flett 1921). The dominant sulphide mineral is galena, with lesser chalcopyrite and sphalerite. The principal gangue minerals in association with the galena are breccias of calcite and dolomite with subordinate quartz (Plates 1, 2, 3 and 4 Appendix 1).

4.3.2 Copper

Copper was mined at Kilsleven, (Mine 9 in Table 4.3 below) situated some 1.5km north-east of *Loch a' Chuirn*. Wilson and Flett (1921) state that a vein with a north-west trend was seen to be 2-3 ft wide and covered a distance of about a quarter of a mile. The mine was reputed to have been discovered in 1760 and was first mined for copper. At some later period, lead ore was extracted.

4.3.4 Manganese

Manganese was mined in the 19th century on a small scale on the Dun Athad peninsula on the Mull of Oa, south west Islay. Here, former workings have been identified but the period of mining is uncertain. The ore occurred as Pyrolussite (MnO_2) forming metal veins which were seen in association with replacement bodies of quartzite (Wilson and Flett 1921).

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[1] The Lossit Limestone is a new classification of the former Islay Limestone (Barnett 1959)

Distribution of lead mines and related features in the study area.

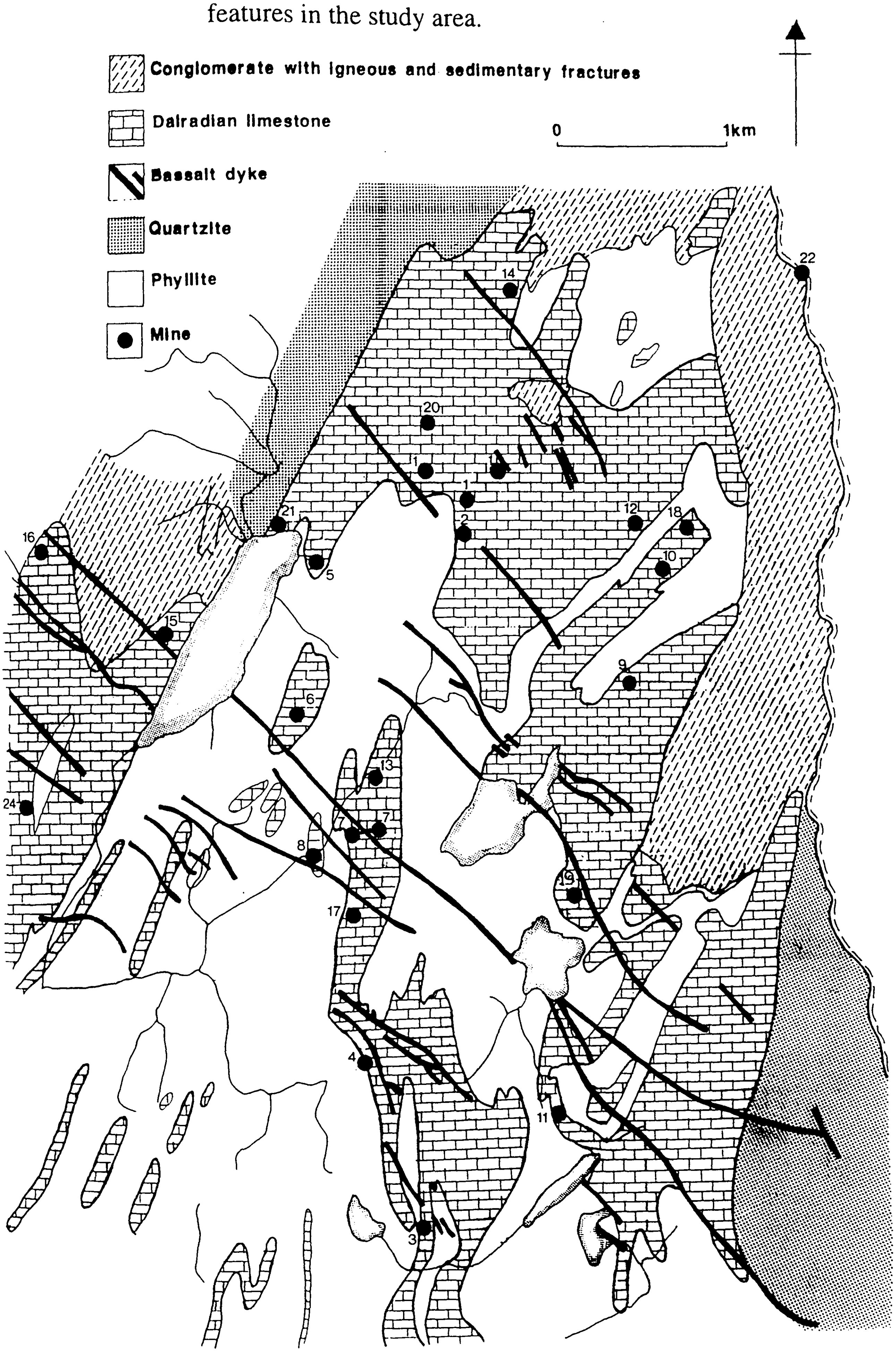

Figure 4.2 Distribution of leadmines in relation to the limestone in NE. Islay (see Table 4.3 overleaf for site names).

Summary table of the site names, location and bibliography

No	Site Name	Grid. Ref.	Mineralogy	Reference
1 2	Mulreesh and Washings site	NR 401601	Gl Sp Cp	Wilkinson, 1907; Wilson and Flett, 1921; Barnet, 1959; Tindale, 1983; Burt, et al 1981; Callender and Macaulay, 1984; Cressey, 1993 and 1994.
3 4	South Ardachie North Ardachie	NR 399635 NR 395643	Gl Gl	Smith, 1895; Wilson and Flett, 1921; Callender and Macaulay 1984; Cressey, 1993. 1:25000 OS. Sheet NR 46/56
5	Portnealon	NR 394683	Gl	Smith, 1895; Cressey, 1993.
6	Robolls Mine	NR 338671	Gl	Wilson and Flett, 1921; Cressey, 1993.
7	Ballygrant	NR 398664	Gl	Wilson and Flett, 1921; Callender and Macaulay, 1984.
8	Gartness	NR 385695 NR 397659 NR 397658	Gl (Ag?)	Wilson and Flett, 1921; Cazelet, 1974; Callender and Macaulay, 1984.
9	Kilsleven	NR 415671	Gl Cp	Wilson and Flett, 1921; Callender and Macaulay, 1984.
10	Carnbeg	NR 414674	Gl	Callender and Macaulay, 1984.
11	Glasgo Beag	NR 409643	Gl	Smith, 1895; Calender and Macaulay, 1984.
12	Keills	NR 417683	Gl	Callender and Macaulay, 1984.
13	Woodend	NR 400667	Gl	Smith, 1895; Callender and Macaulay 1984.
14	Balulive	NR 406609	Gl	Callender and Macaulay, 1984.
15	Sean-ghairt	NR 382667	Gl	Smith, 1895; Callender and Macaulay, 1984, OS. 2nd ed. 1878. (NMLS)
16	Ballytarsin	NR 357612	Gl	Smith, 1895; Callender and Macaulay, 1984.
17	Cnoc Dhiarmaid	NR 373684	Gl	1:25000 OS. NR46/56
18	Knocklearoch	NR 397644	Gl	Wilson and Flett, 1926.
19	Loch a Chuirin Bhig	NR 417676	Gl?	1:10560 Argyll Sheet 189 NW.
20	Cul Dreasach	NR 421684	Gl	Barnett, 1959.
21	Finlaggan West Shore site	NR 391684	Gl	Cressey 1994 (unpublished)
22	Smelt Mill	NR 429700		Miller, 1772 (Brit. LIb. Ms15.509) Langlands, Argyllshire, 1801. (NMLS)

Table 4.3 Table of site names and location as shown in Figure 4.2 on the previous page. (Gl = galena, Sp = zinc, Cp = copper, Ag = silver and NMLS is National Map Library of Scotland).

4.3.5 Bog Iron

In 1772 Pennant described a *"species of iron called bog-ore"* in a stratified deposit not far from the lead mines. The feasibility of extracting iron ore from bogs was later considered by Frew in 1871 who mentions that bog iron occurred *"in many parts of the island immediately below the peat moss and without any seeming connection with the geological character of the rocks near it"*. Around 60 tons of iron ore was excavated from the bogs to the north of Port Ellen in 1867 and assayed at 50% iron. However, the reserves appear to have been sporadic and were not considered worthy of exploitation (Callender and Macaulay 1984, 41).

4.3.6 Limestone

Dalradian limestone is presently being mined at a quarry in Ballygrant (Grid ref. NR 398663). The limestone is of high quality ($CaCO_3$ 95·17 to 97·30 %) and is crushed locally for road surfacing and for agricultural improvement.

4.4. Geochemical Exploration in NE Islay

Geochemical and geophysical surveys have been extensive across the NE region of Islay. From 1970-1974 grants were made available by the Highlands and Islands Development Board to investigate the potential for reworking regions of mineralisation and to locate previously unknown mineral deposits of economic importance. More recently the British Geological Survey (BGS) has undertaken geochemical analyses on stream sediment as a contribution to their recent South of Scotland Geochemical Atlas (SSGA). Two separate studies have been undertaken by undergraduate students to investigate the influence of contamination from lead workings using stream water and sediment analyses. The results from these surveys clearly show that old mine workings are still a source of heavy metal contamination in this region (Short 1994). In 1970 the Dunlossit Estate, NE Islay, entered into agreement with Rio Tinto Exploration Ltd (hereafter RTZ) to prospect for minerals over NE Islay. The initial work programme was over four years, comprising reconnaissance geochemical survey using surface soil and stream sediment. This resulted in the compilation of a series of geochemical distribution maps across the limestone belt in the study region. This work formed the basis of a follow-up survey using geophysical and test-drilling. The stream sediment surveys identified three regions as having high anomalous values in Pb, Zn and Cu. Anomalies to the east of the village of Ballygrant and in the head waters of the Kilennan and Barr rivers identified the need for further geochemical and geophysical survey (Cazalet 1974).

A new programme of geochemical survey was undertaken. Phase 1 investigated the three anomalous regions using soil samples (228 samples), sampled along transects 1000ft (304.8

m) apart. Strongly anomalous values were found in the Ballygrant and Kiells region, however the high anomalous values recorded in the Barr River region were not substantiated by the soil geochemical survey and tended to confirm a secondary environmental origin (Cazalet 1974). Phase 2 was initiated to in-fill the regions with high anomalous values identified in the Phase 1 survey. Soil-sampling was extended to cover the whole of the limestone outcrop and focused on the region where anomalies were incompletely defined by the earlier survey.

A follow up geophysical survey was undertaken to explore the depth and extent of ore bearing strata in the Kiells and Lossit region. Seven and a half linear miles of detailed induced polarisation resistivity survey were conducted on lines 600 ft (220 m) apart. Over the limestone and shales background resolution was good with a high response to depths of up to 50-60 m which only fell where the lines passed on to softer conglomerates and sandstone. Interference by graphite partings (inclusions in the Ballygrant Limestone) resulted in high background values masking the presence of mineralisation (Cazelet 1974). In these areas the survey was abandoned in favour of the results obtained in the earlier geochemical (phase 3) survey. Strong geophysical anomalies were identified to the east of Ballygrant and south-east of Loch Lossit. These were supplemented by additional soil transects to in-fill between the transects of the previous survey.

The result of extensive geotechnical survey on the limestone belt in NE Islay confirmed strong Pb anomalies with weaker Zn and Cu values occurring to the east of Kiells, Ballygrant and to the south of Loch Lossit. In these areas, the anomalies could be explained in part by contamination from old workings. Higher anomalous values identified away from the mined areas were accounted for by pockets of mineralisation. This led RTZ to conclude that the region possessed more extensive mineralisation than that worked in the last century.

In 1982, following recommendations made as a result of the three phase survey by RTZ, detailed geological mapping was carried out in the Kiells-Killsleven and Loch Lossit regions at 1:10,500 scale. The objective was to investigate in greater detail the anomalous regions identified earlier and provide stratigraphic control over the mineralised regions (Tindale 1983). Three diamond drill hole surveys were also carried out to check stratigraphic relationships and determine the source of the Pb, Zn and Cu soil anomalies. Base metal mineralisation was found to coincide along fault zones and the mineralisation fluids were extremely weak. Overall, the drilling program could not account for any obvious source for the soil anomalies. To the south of Loch Lossit during a local geological survey, what appeared to be an old trench and shaft were found directly above the anomaly. This led the team to conclude that the remains of old workings were the source of contamination resulting in the significantly high anomalous values in the Lossit region. The previous work undertaken in the early 1970s and 1980s and more recently in 1991 by the British Geological Survey have made NE Islay unique in having the most comprehensive geochemical survey undertaken in any 40 km^2 mineralised region in the United Kingdom.

4.5. Archaeological Research

The archaeological record of Islay is broad, with evidence for human activity from the Mesolithic through to the Post Medieval. In the interest of brevity this section will discuss only two current research projects. For a detailed review of the artefactual and monumental record of Islay the reader is referred to the archaeological inventory compiled by The Royal Commission on The Ancient and Historical Monuments of Scotland, (RCAHMS) Argyll Inventory Volume 5.

4.5.1 Southern Hebrides Mesolithic Project: University of Reading

Work is currently being undertaken in the Rhinns of Islay by the Southern Hebrides Mesolithic Project (SHMP) in the University of Reading. The SHMP was initiated in 1988 as a three year research project to study Mesolithic settlement in the southern Hebrides. The aims of the project were to address Mesolithic settlement patterns from a regional perspective with survey and excavation of unexplored regions of Islay and the island of Colonsay (Mithen 1992, 1993). Two seasons of field work were conducted during 1989. More recently a program of test pitting at the site of *Coulererach* in the Loch Gorm region has led to the recognition of a dense artefact scatter of Mesolothic character, largely buried below 1.75m of peat. To obtain absolute dates and aid palaeoenvironmental reconstruction, work began in 1994 to recover peat samples above and below the layers at which the artefacts were recovered (Mithen pers comm.).

4.5.2 Finlaggan Project: National Museums of Scotland

A six year programme by the National Museums of Scotland to investigate the historically important site of 'Castle Finlaggan' is nearing completion. The island-site (Figure 5.14 and 5.19) once enclosed by a timber palisade lies close to the northern shore at the head of Loch Finlaggan. The main island of *Eilean Mor* is 180m in length and about 75m transversely. A smaller island *Eilean na Comhairle* has a diameter of about 30m and lies *c.*35m to the south of the main island. (Chapter 5 Figure 5.3). *Eilean Mor* is now separated from the mainland by a narrow reed-grown channel of shallow water.

Formally, access was gained onto both islands via stone (and probably timber) causeways. The site in its natural setting has evoked a passionate response from one researcher documenting the history of the occupants of the site:

> "Finlaggan is a dreary place, for more than anywhere else does it seem to symbolise the ruins of the ancient Lordship not as a fighting and political force, but as the great centre in Alba for the ancient civilisation of the Gael" (Grant 1982)

More recently the director of excavations stated:

> "Finlaggan, is the so-called 'Centre of the Lordship of the Isles', the place of lordly inauguration ceremonies and meetings on the Council of the Isles; a site of national importance" (Caldwell 1993).

The Finlaggan Project stands unique among the limited archaeological work so far undertaken anywhere in the Western Isles (Caldwell and Ewart 1993). A holistic approach has been adopted within the excavation strategy that has included underwater reconnaissance survey, (Ruckley and Cheshire, 1990) to a more recent topographical survey of the landscape surrounding the loch by RCAHMS in 1993. A programme of environmental and archaeobotanical processing is another facet of the excavation strategy and is now providing an insight into the range of crops being brought on to the site. The geomorphology of the site's formation is now understood (Chapter 5 Figure 5.30).

Excavation between 1990-92 on the island of *Eilean Mor* has revealed evidence of a rampart fortification and the layout of several dwellings. A paved road has been discovered between two main areas of occupation. A major phase of occupation appears to have ended at the end of the 15th or the beginning of the 16th century with the replacement of substantial medieval buildings by houses and ancillary buildings leading to the development of a township in the 16th century. During the course of excavations, the presence of several hearths and large spreads of rakings from peat and wood ash were interpreted as part of a cooking area. A series of hearths were archaeomagnetically dated and provide a date range of 1425-1475 (GeoQuest Associates 1994). Other areas of carbonaceous-rich material within *Eilean Mor* have been interpreted as evidence of smithing. Iron slag has been recovered elsewhere on the site and is possibly associated with phases of rebuilding. Scraps of lead in the form of droplets have also been recovered from several 14-15th century contexts and from along the shoreline. The lead droplets ('chaplets') may be the waste products from mould-casting which leaves a characteristic piece of waste (cf. Hodges 1989, 71).

Progress has since developed with the discovery of a large 'castle' on the island of *Eilean na Comhairle* the so called Council Island. Robbed of its upper courses, it appears that the castle or hall-house had been deliberately dismantled perhaps at the behest of James IV in the 1490s to serve as warning to all who might assume control over the lands formerly held by the Lordship of the Isles. Excavation in 1993 revealed the extent of the burial ground adjacent to the chapel. Extra-mural graves were seen to be of an early date, perhaps even pre-dating the earliest phase of main occupation at the site. Whether this evidence points to a monastic cell predating the earliest secular occupation on the island is a question to be addressed by more work at a later date. Work will be undertaken in the autumn of 1995 to assess the archaeological implications of various possible prehistoric features within the landscape adjacent to *Eilean Mor*.

4.6: The importance of Lead Mining in Islay

4.6.1 Introduction

This section will discuss the historical evidence for lead mining in the study area. It will be recalled that the first aim in this study is the identification of the geographical extent and duration of 18th and 19th century lead mining. In order to achieve this aim, an examination of the historical record has been undertaken. This has provided useful historical information to support the results from the archaeological survey of the mining remains and these will are reviewed in Chapters 5, 6 & 7 respectively.

The discussion focuses on the wider implications of the economic importance of lead within the region. Documentary sources are reviewed alongside other statistical records to show that lead was exploited far more intensely than previous literature would suggest. It has been argued by Barnett (1959, 65) that Islay was probably the second most important region of lead mining in Scotland. It is no surprise that in the history of lead mining, Islay remains one of the lesser-known regions of lead production in the UK. However, the area exploited for lead was geographically smaller in comparison to the major lead fields on the Scottish mainland.

Statistical information concerning the lead industry on Islay has been collated from the records of the Islay Estate by Wilson and Flett (1921, 67-68), and Burt *et al* (1981, 18). Callender and Macaulay (1984) have also produced a monograph and visitor's guide that contains historical and technological information relating how the mines functioned on a commercial basis. These authors also sketch-mapped features such as shafts, trials and adits along with standing buildings associated with the abandoned mines. However, an important issue not addressed was the possibility of their being more than one, or possibly even multiple phases of mining activity at a given site. Moreover the writers failed to recognise the true extent of the mining remains. Credit is however given to this work as it provided the author with primary location data and a useful bibliography on Islay's later mining history.

4.6.2 Historical Perspectives

There have been claims originated by Wilson (1921) and subsequently quoted by others since e.g. Barnet (1959), Edmond (1983) and Callender and Macaulay (1984) that "lead was mined on Islay in the Norse Period" (8th and early 11th centuries in the Western Isles). Yet this claim is unsupported by any real material or documentary evidence. However, place name evidence and funerary remains do reflect Norse influence on Islay during this period, but evidence of Nordic settlement is lacking. A much quoted documentary account of the presence of lead on Islay comes from Donald Monro, Dean of the Isles, who in 1549 wrote:

"In Ila is meikle lead ure" [In Islay is much lead ore]

However the earliest reference for lead mining is dated 1511-1512 in the accounts of the Lord High Treasurer of Scotland which mention:

> "Item, the viij day of Januare, compt maid with William Striveling for expenses maid be him apone the sayng of the led myned of Ilay, for the careage tharof, for the biggein of ane hous to the said mynd, for charcoill and other expensis" (Paul 1902, 274).

The recognition of 'earlier' or 'old workings' in contemporary accounts of 1770 attest the scale and geographical extent of the many trials and pits within north-east Islay. Charles Freeburn writing in October that year describes the presence of these features and his description is worth quoting in full:

> " This island contains of 300 square miles, the extent quhairof [whereof] is too much for any person to examine with accuracy in a mineral way. But there is upwards of 40 square miles, contiguous, to where I have bestowed a good dale of trouble and expense, that can ever be truly and wholly mineral. As over the whole extent last mentioned there has been 1000^{d} tryals made in different parts, which have all produced lead ore, and when cleared up still shew from one to six inches thick. But, by their manner of working, quhilk [which] was running along the course of the veins as deep as the soft soile went, whenever hardness or water met them they [the miners] left of working. I have opened above 150 of these old workings and found them as above described, and have prosicuted sundry [others] of them to a considerable depth, and always found the ore continuing, widening sometimes to drift wide, quite pure at 14 fathom deep (c 31 m) and at one place 24 fathom deep" (c 54 m), (Smith 1896, 462).

Barnett (1959, 66) in a geological survey of the north-east of Islay was the first to recognise that the remains of lead mines found in at least fourteen locations could be assigned to more than one phase, showing immense awareness when he wrote:

> "An inspection of the workings gives the impression that they may be divided roughly into two groups-'former' and 'old' and that many of the old workings are very old, possibly medieval, while some, only doubtfully recognisable as workings, may date back to an even earlier period."

Whether Freeburn's estimate of 1,000 "old workings" is possibly an overestimate cannot be determined, but we can be certain that most of what he observed has been lost to later 18th and 19th century mining. It is noteworthy that Barnett possibly recognised what remained of some of these features and quite rightly assigned their origin to a much earlier period.

On Islay in the late 17th century a dual economy existed whereby tenant farmers were granted leases from the principal land owner to mine the lead ore. Lead extraction appears to have been sporadic until the mid-18th century, probably as a result of financial constraints on the land owners and the rise and falls in lead prices.

Thomas Pennant, in his *Tour of Scotland and Voyage to the Hebrides 1772*, wrote that the lead industry was "working under the control of a Mr Freeburn of Freeport near Port Asgaig" (Plate 5, Appendix 1). The site of the 'air furnace' at Freeport as sketched by John Frederick Miller, 1772 is now occupied by the distillery at Caol Isla near Port Askaig, Islay. The furnace would have been suitably positioned to receive coal as ballast cargo from the mainland. In the picture, a large stack of peat can be seen in the foreground and was certainly being used alongside wood and coal in the smelting process. Callender and Macaulay (1986, 45) cite a clause mentioned in a mineral lease dated to 1836 which mentions:

> 'liberty and free leave [is given] to raise and carry of peats and turf from the farm of Persibols for the purpose of smelting and refining ores and metals'.

It is noteworthy to mention that there appears to be two chimney stacks associated with the smelter, which possibly suggests the presence of two sets of hearths. Were this to be the case, then one set would have been the primary smelter (first smelt) and the second for refining the slags of the first smelt. Interestingly there also appears to be some form of chute leading from the top of the hill down into the smelter building. If this indeed was the case then it saved carting heavy loads of dressed ore down the steep incline to the foreshore.

Silver was a valuable by-product that was extracted in this period by cupellation. We know that the Islay lead ore was particularly silver-rich (see Chapter 7 Table 7.3). Pennant recognised this when he wrote:

> *"lead veins were extensive and provided forty ounces of silver from a tun of metal."*

This point emphasises the importance of lead ore as the major source of silver at this period. Useful statistics concerning lead and its production on Islay between the years 1769 and 1774 are recorded in the Kildalton Charter chest and are as follows:

Item	Tons	Cwt
Bar lead	260	1
Ore	72	6
Slag of Lead	90	

Table 4.4. Lead output statistics between 1769-1774 (after Wilson *et al* 1921)

The scale of the lead working in north east Islay can be envisaged in a report entitled '*State of the Mines in Islay*'. The report was compiled by Alexander Shirriff in 1770, a visiting agent for the Leadhills mining company of Lanarkshire. He compared the shipment expenses between the Islay and the Leadhills works as follows:

For Leadhills	£	s	d
Suppose lead at £14 15s per ton			
Royalty to Lord Hopetoun	16	13	4
Cartage to Leith at 30s per ton	10	0	0
Commission and shipping charges	3	0	0
total cost =	**30**	3	4
For Islay			
Suppose lead at £14 15s per ton	£	s	d
Tack duty	12	10	0
Cartage to sea at 2s per ton	1	5	0
total cost =	**13**	**15**	**0**
Balance: Islay below Leadhills	£16	8	4

Table 4.5. Transportation costs between Islay and Leadhills (Smith 1896).

These statistics show that lead then was some £16 18s. 4d per ton cheaper in transportation cost from Islay to the port of Leith. We know that Leith was an important trade-route for lead and that dressed ore (in powder form as against smelted lead) was exported from Islay to the Scottish mainland in 1776 (Section 6.3.1). For the next fifty years or so it is highly probable that there was sufficient demand from the Scottish mainland for Islay lead.

In the early 19th century production appears to have slumped and declined until 1836. At this time new leases were granted to a prospecting company to explore the island's potential for other minerals that included gold, silver, zinc, copper, tin, cobalt and manganese. The result of these agreements in physical and economic terms are not clear. Shortly after 1836, interest developed in Islay's potential for increased lead production and in 1862 leases were granted to allow further investment in the industry. Output records based on Home Office Tax Returns (Figs 4.3 and 4.4) provide records of ore extracted and lead produced at Mulreesh. The output data reflects a steady rise in lead production with a maximum peak recorded in 1867. Figure 4.4 shows that silver production follows the same trend as lead output. The silver was probably extracted as a litharge from the lead, a by-product of the primary smelting operation. No records survive detailing where this operation took place but it is possible that secondary processing may have been undertaken elsewhere on the Scottish mainland. This appears to be the case, as dressed ore was going directly to the Scottish mainland.

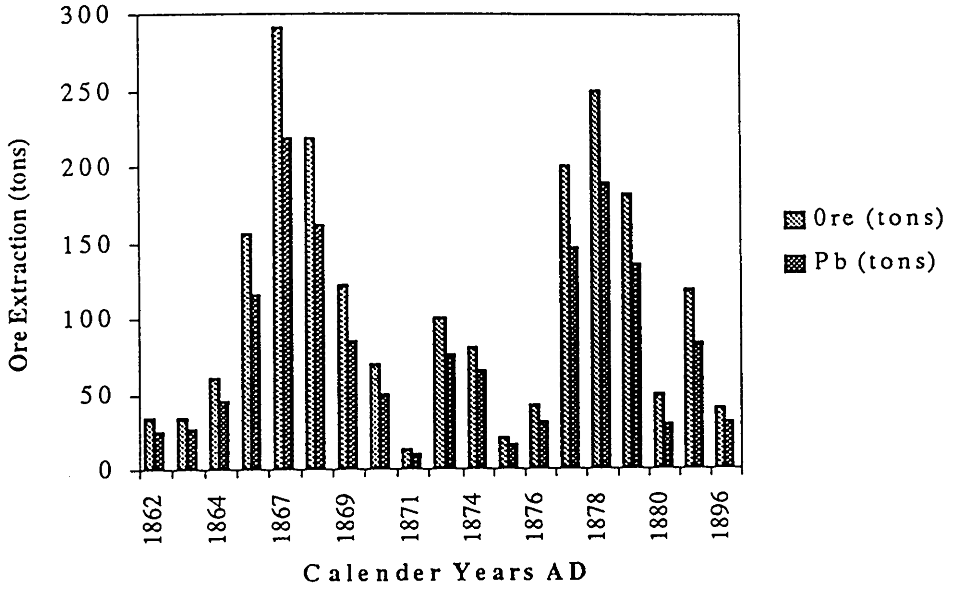

Figure 4.3 Ore extracted and Pb produced between 1862-1896 (after Burt *et al* 1981)

The breaks in yearly production may reflect the vagaries of the lead market or troubles with the share holding Islay Lead Mining Company Limited. A report from their first meeting in London in 1863 recounts the clearing of water courses and the installation of a beam engine and two boilers. This represents a considerable investment in the industry when faced with falling world prices and increasing competition from the mainland. The argentiferous[2] quality of the Islay ore may well account for such investment when faced with increasing competition from the mainland. In 1896 production was limited to 40 tons of ore by eight underground workers that was processed by eight surface workers (Burt *et al* 1981).

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[2] Meaning the quality and quantity of silver.

Lead production finally ceased in 1898 and the Cornish beam engine at Mulreesh was salvaged by an English salvage company.

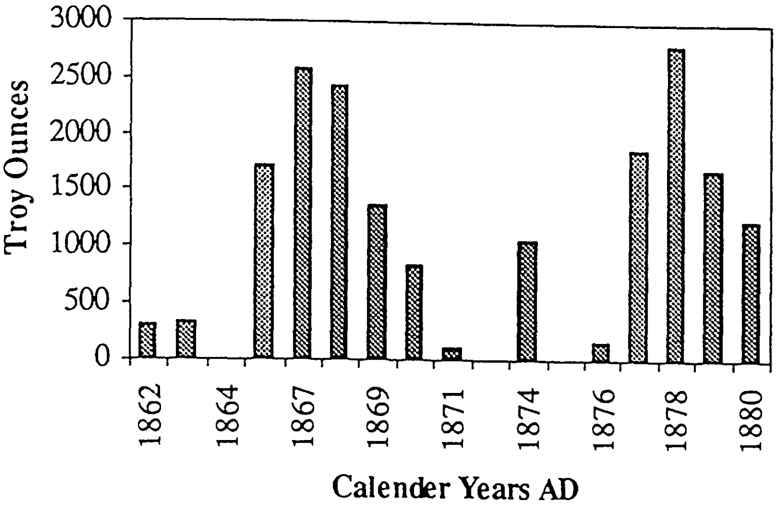

Figure 4.4 Silver produced between 1862-1800 (after Burt 1981).

4.7 Conclusions

This chapter has reviewed the environmental setting of the island and the geographical position of lead mines within the study area has been discussed. The 1973 and 1983 geochemical and geophysical survey across parts of north-east Islay area provide a clear understanding of the extent of mineralisation and its contaminatory effect over land previously mined.

Collectively, these projects form a significant corpus of geochemical data and probably represents one of the most intensively surveyed mineral regions of the British Isles, especially taking into consideration the recent BGS geochemical survey and other collaborative geochemical surveys by this author and two undergraduate students. The palaeoenvironmental record for Islay is now better understood in the light of recent palynological research (see Chapter 5 Section 5.9.1).

The recognition of 'Norse' mining activity based on surface evidence alone is probably impossible. However we can be fairly certain that there has been lead mining in north-east Islay, occurring as early as 1511-12 based on historical sources. This account is given additional significance by being the earliest historical account to mention lead in the region. Moreover the account goes some way to explaining the numerous "old workings" recognised as early as 1770. In all probability throughout the 17th and to the early-18th century, lead mining occurred intermittently. This appears to have been due to contractual and economic reasons rather than the lack of exploitable lead deposits. Soon after this period the industry developed which intensified into a fully mechanised concern at the long-standing Mulreesh mine by the early 19th century. A full account of the archaeology of these sites is given in the next chapter.

Now that the history of mining has been discussed, the scale of what remains of 18th and 19th century mining evidence can be examined. It is at this juncture that we also move to the examination of freshwater loch sediment that forms the central theme of the next four chapters. The following chapter will examine work undertaken in and around Loch Finlaggan.

CHAPTER 5: LOCH FINLAGGAN AND FINLAGGAN MARSH

5.1 Introduction

Chapter 1 stated that four lochs lay at the centre of this research. The catchment of each was to be investigated for archaeological remains and the geochemical environment examined to assess the scale of environmental impact resulting from mine-derived soil contamination. The link between this and the sediment formed within the lochs could then be examined to identify earlier mining events that may have been occurring within the catchment. Loch Finlaggan was the first of the four lochs to be investigated. This chapter will describe the work undertaken in and around Loch Finlaggan and for ease of description this chapter is arranged into five parts.

Section 5.2 places the loch in its environmental setting and describes the geology, soils and vegetation surrounding the loch. Section 5.3 discusses the results of archaeological field survey that was undertaken to identify lead mines within the catchment. Section 5.4 deals with the results of earlier and more recent geochemical survey that was carried out at the Portnealon mine which lies near the loch's eastern shore. In Section 5.5 the results of chemical analyses from a sediment core recovered from the loch basin are described and assessed. Section 5.6 will discuss the results of biostratigraphical and geochemical investigations undertaken at Finlaggan Marsh, an alluvial formation on the northern side of the archaeological site of *Eilean Mor*. Finally, Section 5.7 will examine the results of pollen analyses undertaken to provide a 'proxy' time-frame to date specific events occurring within the sedimentary profiles at the marsh.

Loch Finlaggan forms the largest loch of the four examined in this study. The catchment has a wealth of evidence for lead mining. The loch was selected for study because of the apparent close relationship between the loch and these archaeological remains. Some of these sites are recorded in historical accounts and date from the 18th and 19th century. Archaeological fieldwork has shown that many of the features date to an earlier period and may be indirectly related to the historically important island site of *Eilean Mor* positioned at the head of the loch. It will be recalled that the excavators of the site were keen to determine the economic importance of lead at the time of the Lordship of the Isles (13-15th centuries). To what extent local lead ore was exploited remains an underlying question at the centre of this research. It has been mentioned previously that the environmental record contained within loch sediments presents a means of identifying early lead mining.

In order to identify the evidence for the scale of 18th and 19th century lead mining a detailed archaeological survey was undertaken to map its general distribution around the loch. An important aspect of this work has been the identification of drainage routes leading from the mining areas. These form an important environmental pathway identified in the conceptual model (Figure 2.7).

This model identified the main environmental gradients from a lead mine into loch sediment. By identifying the extent of mining in the catchment, the principal sources of heavy metal contamination in the catchment could be better understood. Previous and recent results of geochemical survey at the site of Portnealon close to the eastern shore has provided useful data on the present-day contamination of lead, zinc and copper in soils over the Portnealon mining region. Collaborative work undertaken on a core recovered from a drainage adit at the Portnealon site shows provides new information on the degree of seepage emanating from the site into the loch.

It was mentioned above that sediments obtained from lochs and other depositional environments (peat deposits and riverine silts) can provide a useful historical pollution inventory with which to assess the effects of contamination from a number or indeed single point sources. With this in mind it was decided to obtain a series of cores from the deepest part of Loch Finlaggan. The chronology of heavy metal deposition within the sediment would then allow the establishment of links between the environmental, historical and archaeological record and possibly help identify periods of earlier (unknown) lead mining activity.

On the northern side of *Eilean Mor*, an extensive tract of alluvial peat has formed as a result of a continual influx of sediments down the Finlaggan Burn that forms the main inlet to the loch. The marsh contains both a lacustrine and marsh peat sequence to a depth of 4 m. The lower sections contain loch sediment which gives way to marsh peat and is estimated to have formed rapidly over the past 500 years. It was predicted that deeper sections of the sequence ought to contain low 'background' lead concentrations, but as a result of lead mining within the catchment we should see a noticeable enhancement in the Pb, Zn and Cu inventories, particularly within the uppermost sediment sections (*c*.0-50 cm depth). A detailed coring programme was undertaken at the site in order to identify the remains of likely inwash material that could be associated with nearby mining activity. The results of these investigations will be described below. The discussion will first focus on the characteristics of the loch to place it in its environmental setting.

5.2: Environmental Setting

5.2.1 Location

Loch Finlaggan (Plate 11, Appendix 1) is situated in the north-east of Islay and is the longest loch on Islay with a length of approximately 1 km and a catchment of approximately 610 ha (Table 5.1). The loch basin is a glacially-deepened valley running south-west by north-east along a major fault line (The Finlaggan Fault). The west side of the loch rises from 60 m to a summit of 175 m above sea level. The eastern shore rises from 60 m to a summit of 120 m. In general, the east and western shores are stony with little in the way of established marsh.

Grid reference	NR 385675
Core date	September 1992
Catchment geology	limestone - phyllites
Catchment type	grazing-plantation
Loch altitude (m)	60
pH (September 1992)	7.24
Conductivity (μ mhos)	280
Max. depth (m)	12.8
Loch area (ha)	63.4
Catchment area (ha)*	609.6
Catchment:loch ratio	9.6
Afforestation (%)	45.7
Net relief (m)	175 (m)

Table 5.1 Loch Finlaggan ~ site characteristics. (note * excluding the loch).

There are numerous springs and small burns feeding into the loch from both sides of the valley. The major inlet of water into the loch is via the Finlaggan Burn that flows along the aforementioned fault line. The river is 2.5 m wide where it enters the loch but in adverse conditions the discharge can be extreme when the burn bursts its banks and becomes a torrent over 10 m wide. The difference in winter and summer water level on Loch Finlaggan is estimated to be in excess of 1m above the summer mean of 60 m above sea level. Three islands are situated within the loch and each has evidence of occupation. At the northern end of the loch there are two islands, *Eilean Mor* [1] and *Eilean na Comhairle*[2].

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[1] *Eilean Mor* = Main Island
[2] *Eilean na Comhairle* = Council Island

The island of *Eilean Mor* is formed from a Dalradian limestone outcrop and has been enlarged through human occupation. The island *Eilean na Comhairle* (Council Island) is a man-made island consisting of several phases of occupation resting on a natural limestone outcrop. On the eastern shore of Loch Finlaggan (Grid Reference NR387674) there is another small island called *Eilean Mhuireill*[3] which has yet to be investigated archaeologically.

5 2.2 Geology

The geology to the north-west of Loch Finlaggan comprises phyllites and shales overlain by acidic glacial till (Figure 5.1). Limestone bounds the south-western and part of the north-eastern shore of loch Finlaggan. The so called 'Robolls Hill' on the eastern side of the loch is formed by phyllites and acidic shales overlain by glacial till that encircles an outcrop of Dalradian limestone. The region at the north-west side of the loch consists mainly of glacial till that in places has formed extensive drumlin fields. To the north of the loch, the fault line runs north by north-west forming a clear division between on one side, the Dalradian limestone and quartzite on the other. Two north-west trending tertiary olivine basalt dykes transect the loch from east to west (Figure 5.1).

5.2.3 Soils, vegetation and catchment land use

Within the Finlaggan catchment, four soil-types are classified by the Macaulay Institute for Soil Research (MISR). The predominant soil type is assigned to the Deecastle Association providing soils for grazing and arable. This type borders the north-east and western regions of the catchment. The soil is derived from Dalradian Limestone with occasional phyllites. Two other soil types have been identified as the Organic and the Durnhill Series. The former is derived from blanket peat, whilst the latter is formed over glacial drift. The region to the south west of Loch Finlaggan is of the Foundland Series, a soil derived from slates, phyllites and other metamorphic argillaceous rocks. These are poorly drained acid soils with a vegetation cover of heather-moorland species that is suitable for only rough grazing. The predominant herbaceous plants are the blanket bog flying-bent communities with a mixture of herb-rich *fescue* grassland. *Calluna vulgaris* is established over much of the acid soils. Marshland communities thrive in the regions susceptible to waterlogging with *Carex sp.* and *Juncus conglomeratus*. The shrubs *Salix sp.* and *Myrica gale* are well established along the sides of small burns that run down to the shoreline.

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[3] *Eilean Mhuireill* = Muriel's island

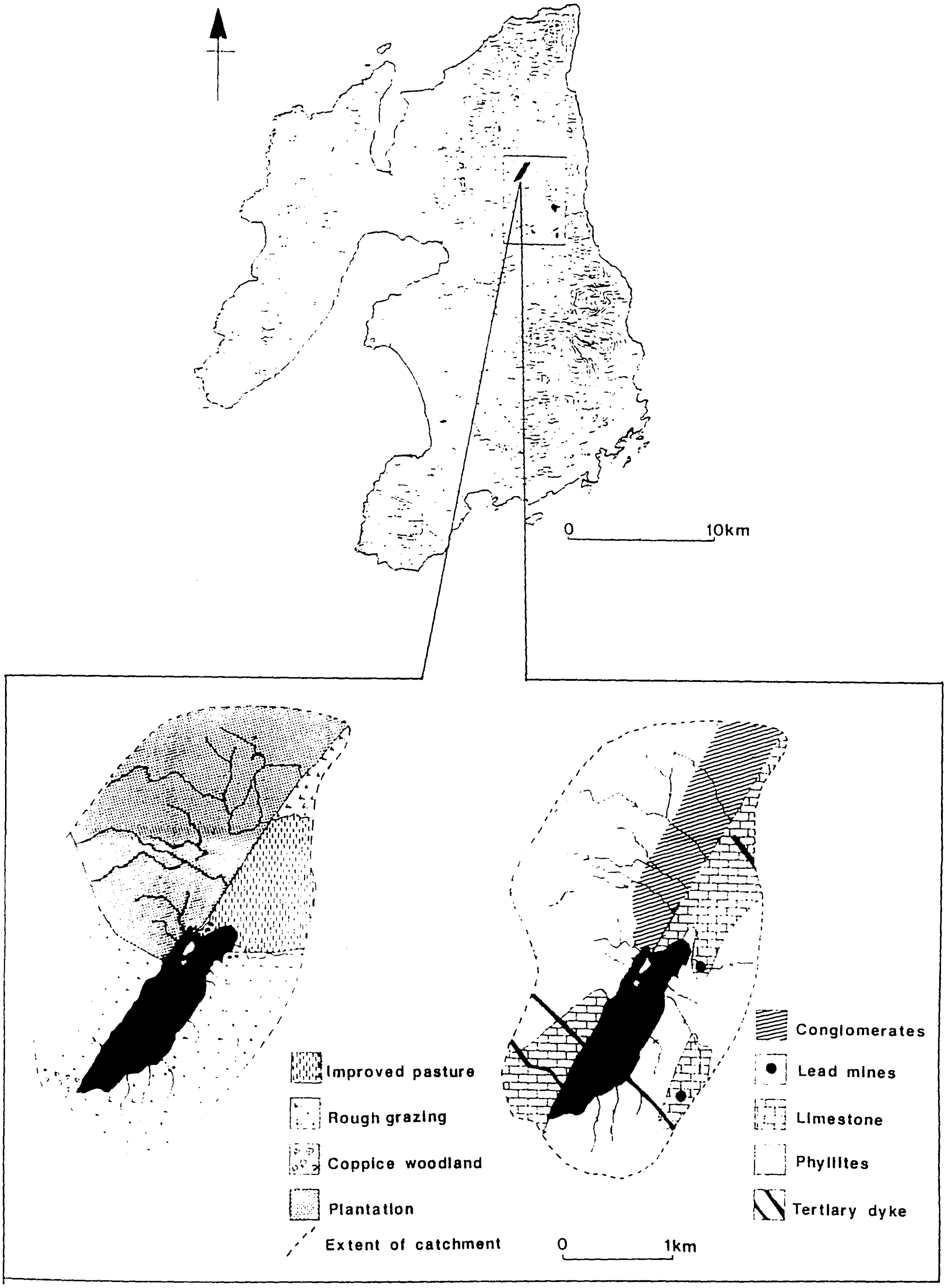

Figure 5.1 Location of Loch Finlaggan, present day land use and local geology.

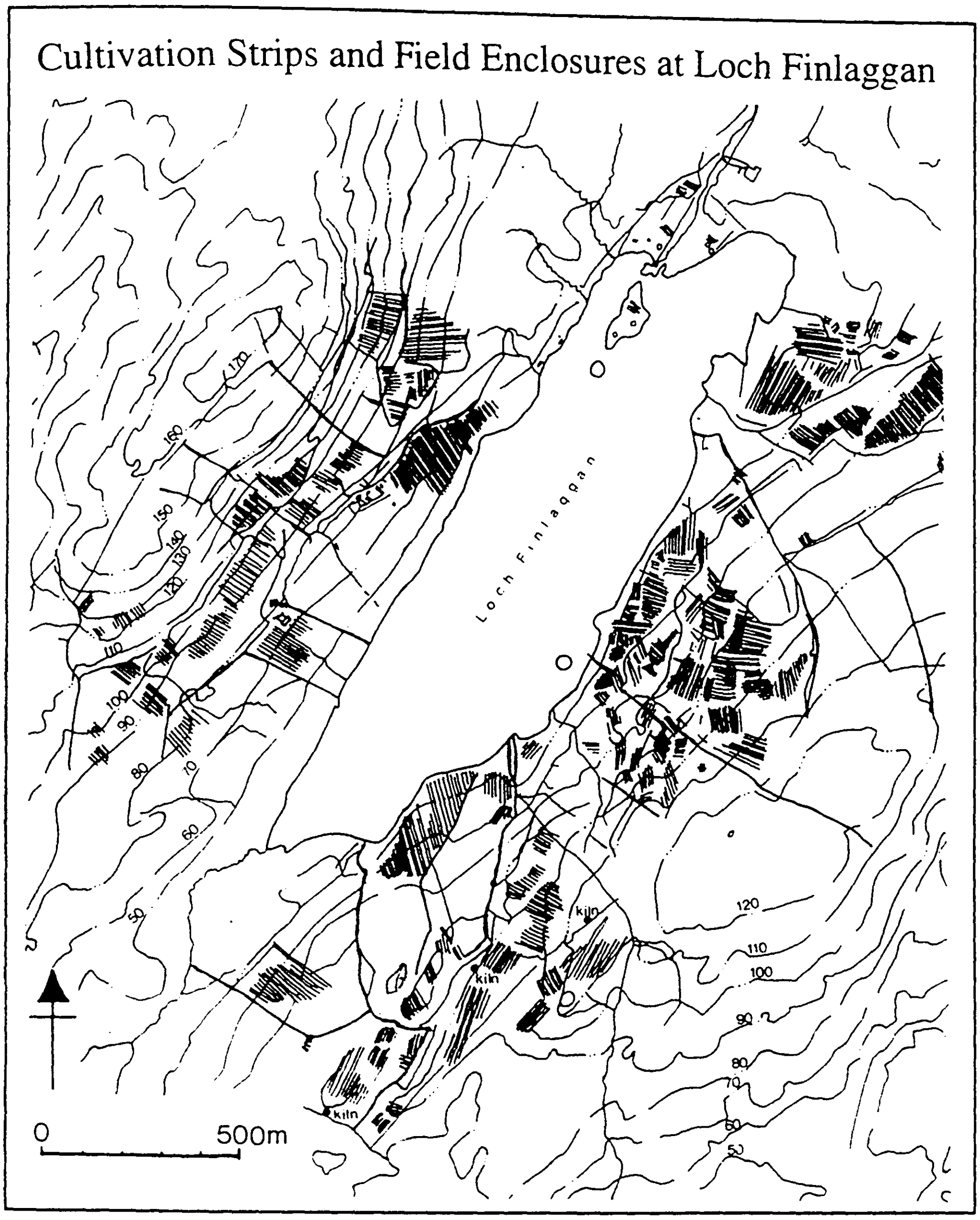

Figure 5.2 Cultivation strips and field enclosures surrounding Loch Finlaggan (after RCHMS survey 1993).

Meadow communities on the improved pasture include *Ranunculus repens*, *Circium arvense*, *Vicia lutea* and *Rumex acetosa*. To the north west of Loch Finlaggan an area of approximately 45.5% of the catchment was afforested with sitka spruce and lodge-pole Pine by the Forestry Commission in 1982 (Table 5.1). At the southern end of the loch a tract of abandoned coppice woodland survives, with stands of *Corylus avellana* and wind-stunted standards of *Quercus sp.* In all probability, the coppice may have been formed from secondary woodland (D. Boyd[4] pers.comm.). No records survive of when this woodland was managed. Solitary wind-stunted birch and oak trees survive on the eastern side of the loch. At the northern end of the loch a well established tract of *Phragmites australis* forms a belt around the northern shore of *Eilean Mor*. At the southern end of the loch a mixture of *Phragmites australis* and *Nymphaea alba* give way to an extensive tract of marshland through which the outflow meanders eventually to form the River Sorn. This river drains most of the Limestone Belt and eventually joins Loch Indall to the south of the island.

The former potential for arable land use has been investigated in a recent survey undertaken by The Royal Commission for Historic Monuments of Scotland (RCAHMS 1993). This work has identified several phases of rig and furrow enclosed by a series of field systems (Figure 5.2). The field systems themselves lie within a much older framework of earlier land allotment. The earthwork boundaries run at right angles to the shore forming large compartments of field enclosure. Archaeobotanical remains from 13-14th century contexts from *Eilean Mor* confirms the local cultivation of barley and oats (S.Boardman pers comm. 1993). The presence of so much rig and furrow supports the notion that much of the landscape adjacent to Loch Finlaggan was under intense arable throughout the medieval period and increasingly so in the post medieval period when population was at its highest (Storrie 1981, 1983). Later in the mid-late 19th century population declined leading to large scale abandonment of remote settlements, such as the township of *Sean-ghairt* on the south-west shore of Loch Finlaggan.

The previous discussion has placed the loch in its environmental setting. The following section will now examine the results of archaeological field survey.

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[4] Mr Boyd is Factor for the Islay Estates.

5.3: Archaeological Survey of lead mines in the Finlaggan Catchment

5.3.1 Introduction

This section concentrates on the results of archaeological field survey of abandoned mine sites in and on the edge of the Finlaggan catchment. It was mentioned above that the archaeology of mining is constrained by the problem of dating the field evidence. In most cases, the evidence that survives on the ground is notoriously difficult to date. Dating the remains from many different periods is even more problematic due to the fact that many of the features are identical and hence, any typology of site forms can rarely provide clues as to the age of the site concerned. While recognising the problems involved with dating and interpretation, much can be gained from earth-work surveys providing there is some supporting evidence from the historical and cartographic record.

It was important at the outset to determine the extent and physical remains of the latest phases of lead mining in the catchment. A second objective was to establish the main routes of mine drainage into the lochs as these form the principal source of metal influx into sediment, forming a direct physical and environmental link between the lead mine and loch basin. This important environmental route is one of several that are summarised in the conceptual environmental pathway model (Figure 2.7). Three mines and an area containing evidence of early prospecting have been identified within the catchment and their location is shown in Figure 5.3 and in Figures 5.4-5.9 4. Portnealon, Robolls and the West Shore shafts all have drainage adits leading into the loch. The Sean-ghairt Mine is related to the deserted township after the same name (Callender and Macaulay 1984) but no surface remains of the mine can be seen in the landscape today. Mulreesh mine rests on the fringe of the Finlaggan catchment on a 100m contour overlooking the Finlaggan Burn. This site has the most impressive 'early' and 18-19th century upstanding archaeological remains in the study area. As a result of archaeological field survey, the location and routing of adits at the sites are now known and several phases of mining activity has been established. Importantly, the geographical extent of mining activity within the locality is now better understood.

5.3.2 The Mulreesh Mine Archaeological Survey (Site 1&2 in Figure 4.2 & Table 4.3)

Given the position of the Mulreesh mine and its close relationship with Loch Finlaggan it was estimated that mine drainage may have entered into the loch. As a result of archaeological fieldsurvey, it is now clear that the two drainage adits identified at the mine

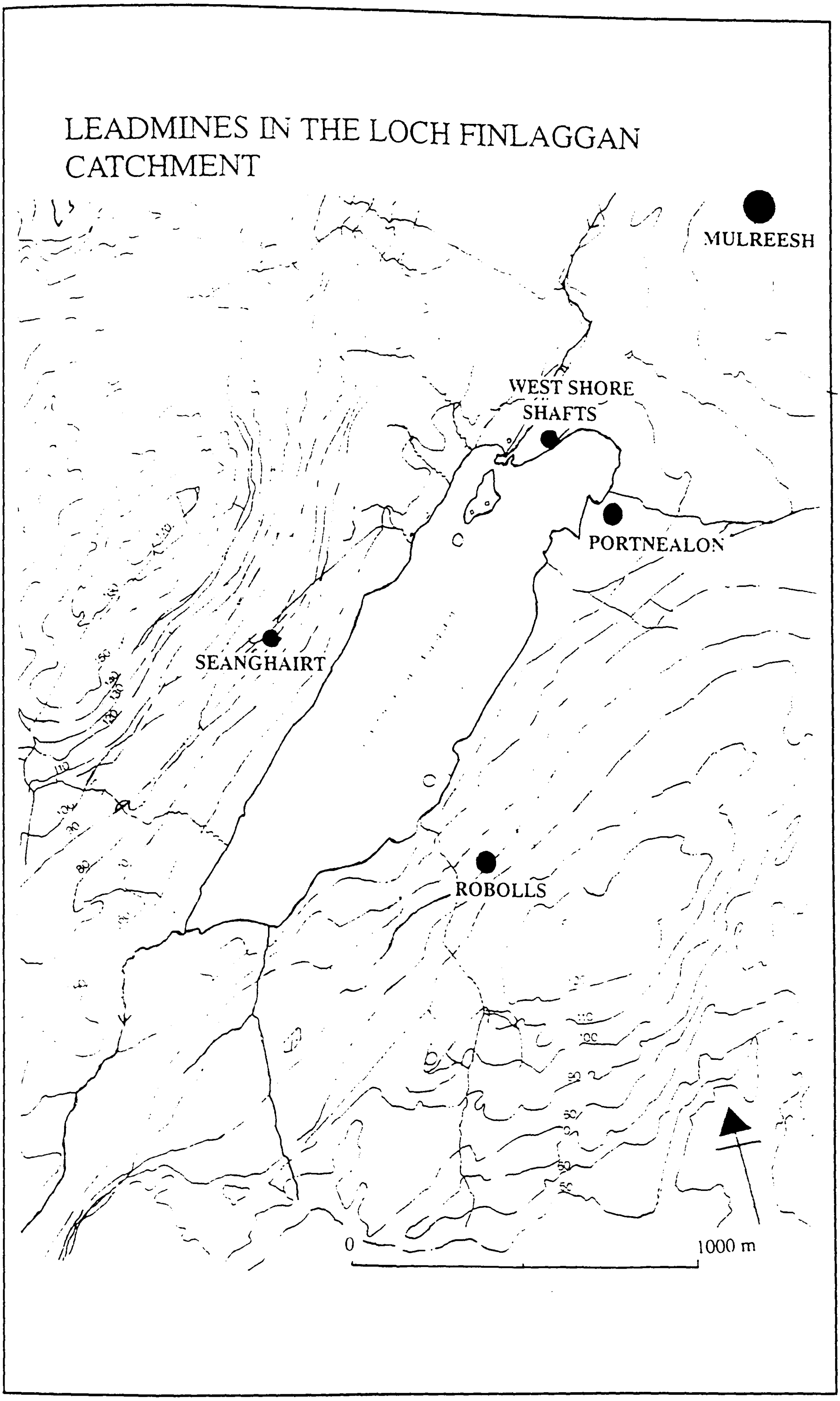

Figure 5.3 Location map of leadmines in the catchment of Loch Finlaggan.

did not drain towards the loch. It is also established that waste water from the site known as the 'Washings' (see below) did not drain towards the loch but drained into a small burn that eventually forms the main inlet into Loch Allan approximately 3 km to the east (NR 425679).

5.3.3 Historical note

As previously mentioned Mulreesh was the principal works of the Islay Lead Mining Company and was the most mechanised, and certainly the last to be worked in the study area. A north-west trending vein of calcite and dolomite with galena, sphallerite, pyrite and chalcopyrite was worked in the 18th and 19th centuries. In 1770 a 64ft (24 m) long shoot had been worked to a depth of 132 feet (50 m). It ranged up to 4ft (1.52 m) wide and the sole of the level was 10 inches to 2ft wide. The mine was later sunk to a depth of 300ft (114 m) and worked by four levels which were 60ft (22.8 m) apart (Smith 1896). By the mid 19th century, the Mulreesh mine had a steam driven Cornish beam engine for pumping water from the mine shafts and for raising ore. Later workings followed cross-course mineral veins to form extensive connecting galleries. A schematic section drawing of the mine shafts and galleries was produced in 1874 by the mine manager H.M.Vercoe. A section drawing of the mine is shown in Figure 5.5 and shows the areas of worked-out ('stoped') ground and provides the names and function of the shafts (e.g., Ladder, Engine and Old Drawing Shaft). These shafts and the two drainage adits have been identified in the Mulreesh earthworks survey which is now described.

5.3.4 Survey description and interpretation

The survey (Phase 2) was undertaken in April 1994 using the method outlined in Chapter 3 Section 3.2.1, and is a follow-up study to previous work undertaken in 1993. Fieldwork identified the presence of buildings and their property boundaries to the south east of the main 19th century complex of Mulreesh. A striking feature brought to light during the survey was the large tracts of hummocky ground broken by only denuded tailings heaps between shafts and the ruined mine buildings.

The Mulreesh earthworks are now described in anti-clockwise direction and individual features have been numbered (in parentheses) to assist the reader in their identification and interpretation (Figure 5.4). Phase 1 represents the earliest phase and comprises four dwellings and a series of shafts situated below the 80m contour in an area to the south of the main 19th workings. Three adjoining rubbish-filled shafts are flanked by a track-way leading down to a hollow way that runs between buildings A, B, C and D. A water-filled adit drains southwards eventually to join a nearby burn (1).

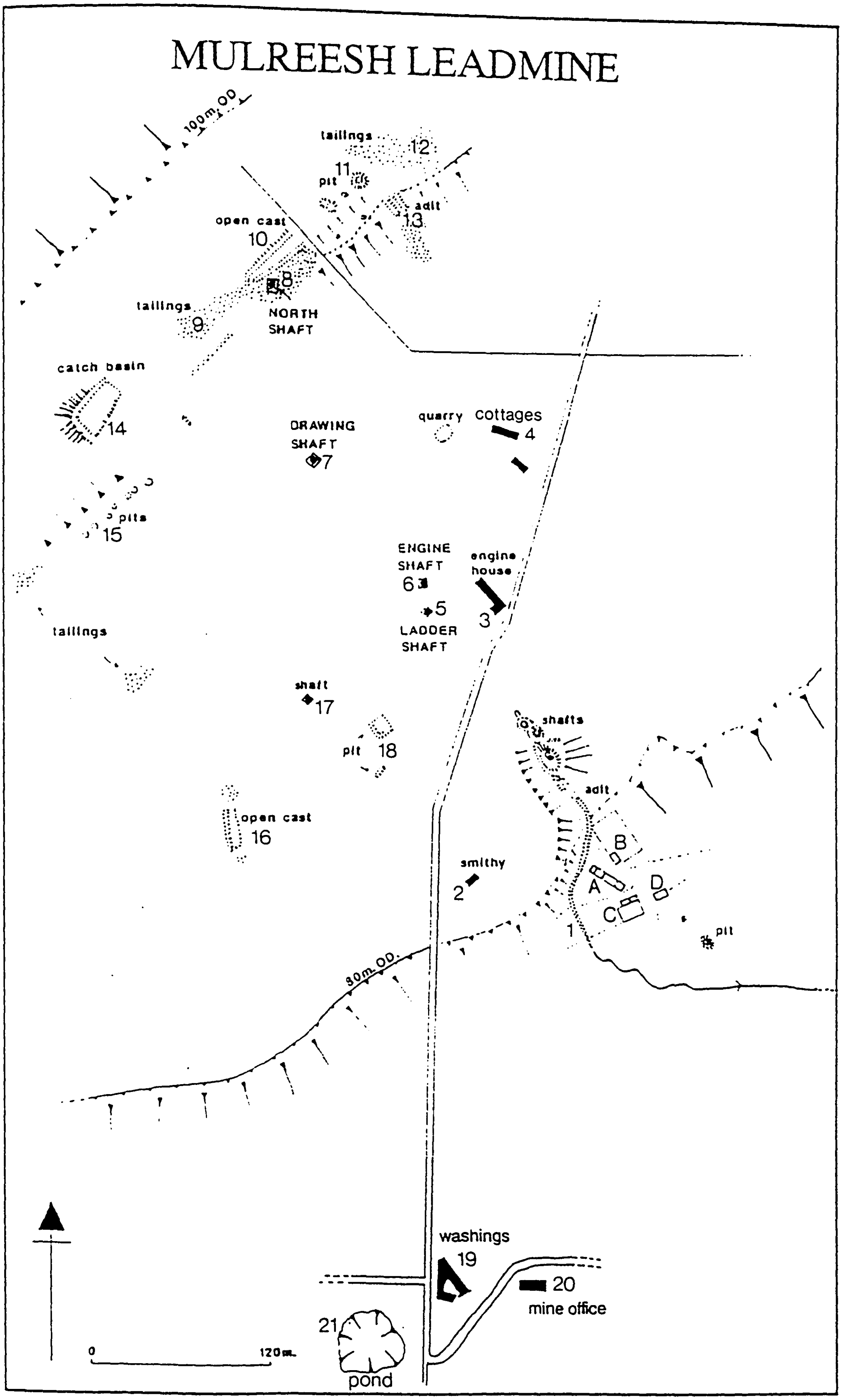

Figure 5.4 Mulreesh leadmine survey (Site 1&2 in Figure 4.2 and Table 4.3).

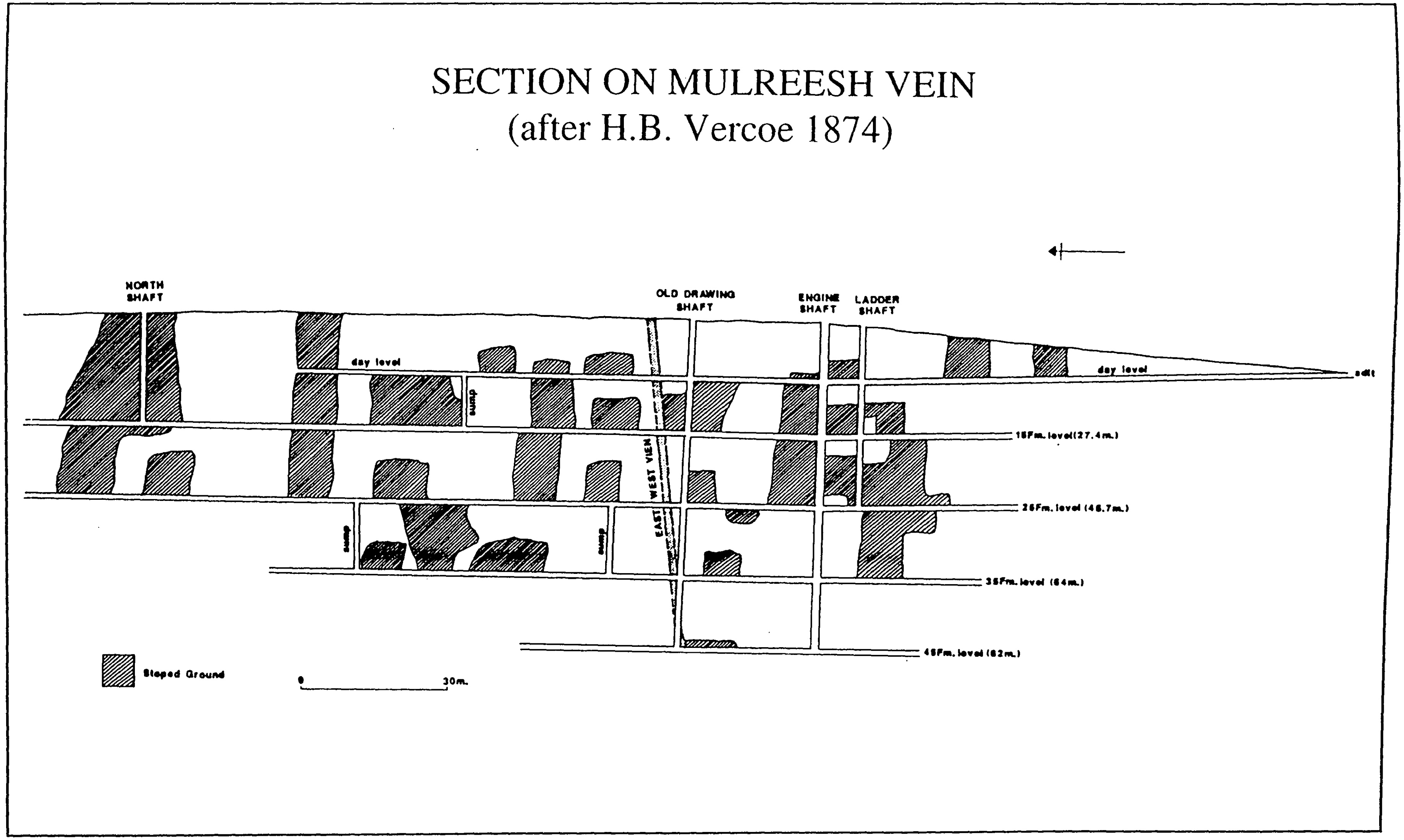

Figure 5.5 Section drawing of Mulreesh leadmine (after Vercoe 1874).

At the time of the survey the adit contained water to a depth of about 1m and was free-flowing. The north bank of the adit overlies the boundary of building B and is later in age than the close. This assumption is supported by the fact that a box-drain in the centre of the metalled track is overlain by the adit bank. Building A incorporates large blocks of roughly dressed Dalradian limestone, most probably quarried from the nearby workings. A large stone at the base of the southern end of building A has within it a sectioned shot-hole (3 cm in diameter). This shows that this building is either contemporary or slightly later than the nearby workings and the fabric of the building is made from mine rubble. Building C has a rectangular enclosure with very pronounced upstanding earth banks. Building D adjoins the parallel banks and flanks a small track that runs west. To the south of this track an area rig and furrow survives along with a small trial pit and a shaft. Moving east from this region some 60m to the east stands the ruins of what has been described as the smithy (2) or small workshop (Callender and Macaulay 1984).

Across the track to the north of the phase 1 earthworks is the rubble-built engine house (3) that has been built on to the gable end of a single storey building. This building was last occupied as recently as the early 1970s. North of this building are the ruins of two cottages (4) that are associated with the mine. Beyond the engine house there is a large tract of flat featureless land with only the remains of fenced-off shafts. The shaft (5) close to the engine house is lined with dressed stone and has a depth of about 40 m. The Engine Shaft (6) and Drawing Shaft (7) are also fenced off and are too dangerous to investigate. The North Shaft (8) is now flooded completely but the remains of what appears to be winding gear and timbers survive at its head. This region contains the largest tailings dumps in the study area ((9) and Plate 7). Some of the tailings are grassed over but where they are exposed, an assortment of large irregular shaped stones of limestone and quartz can be seen. Immediately north of the North Shaft a large open cast (10) runs northwards up to a drystone boundary wall constructed of rubble from the spoil heaps.

Over the wall and running in a north-westerly direction is an escarpment with five small pits and a back-filled shaft (11). Another large tailings dump can be seen to the north of the pits (12). Cut into the escarpment is an adit with a rubbish-filled entrance. Two large banks approximately 1.5 m high lead away from the adit to an area of large boulders (13). The course of this adit was established as north by north east along gently sloping ground littered with mine spoil.

Situated 60 m south-west of the North Shaft is a large ovoid feature with banks that are in places up to 2 m high containing standing water at its northern end (14). The feature is tentatively described as a catch basin for holding water and may have been supplied by a series of wooden troughs from the North Shaft (8). It is noteworthy that Callender and Macaulay (1984) assume that the remains of a small wall adjacent to the North Shaft is all that remains of a plinth to support a pump for lifting water from the shaft. Were this to be the case, then it would be possible that the waste water was contained within the catch basin. South of the basin on a north by north-westerly alignment are seven pits or shafts (15). They are all filled to the surface with farm refuse but their origional diameter was established at 3 m. More mine tailings can be observed to the south-east of the pits along with an open cast (16) that is 30 m in length and 5 m wide. The open cast follows the alignment of a tertiary dyke. The significance of this feature may lay in the fact that lead ore has been exploited from a near surface ore outcrop that was in contact with the dolerite. A fenced off shaft (17) of unknown date lies to the north east of the open-cast and does not relate to those shown on Vercoe's plan and may be somewhat earlier than 1874. East of this shaft is a rectangular feature comprising a shallow scoop with denuded banks (18). To the south is a pit filled with rubbish associated with the last period of occupation of the cottage with adjoining engine house.

Contemporary with the engine house and approximately 500 m to the south is an ore dressing plant with reservoir for powering a stamp (19) and mine office (20) adjacent. The stamp's function was to crush the lead ore prior to washing. A series of troughs for washing and grading the ore would have been nearby. Water was supplied to the reservoir by a leat that traversed the hillside for a distance of approximately 3 km from a small loch that was artificially dammed to control a regular supply of water to the reservoir. It is also likely that water was pumped the short distance from the mine using overshot troughs to supply a catch basin near the site. The Ordnance Survey (First Edition 1856) map show the presence of a pond (21) close to the washings and this probably acted as a catch basin to store water from or to the reservoir. It is a great shame that the remains of the hydraulically driven stamp have been lost as this site is unique to Scotland.

5.3.5 The 'Washings', description and interpretation (Site 2 in Figure 4.2 & Table 4.3)

Figure 5.6 shows a more detailed plan of the reservoir and associated features. Two rubble built walls some 1.5m thick form the remains of the reservoir. This feature was built into a terrace by the removal of slate. The inner face of the wall shows evidence of a mortar lining. The walls have been breached quite recently and substantial amounts of stone have been robbed for track repair. An iron pipe (C) is still *in situ* and is marked on the plan as a dotted line.

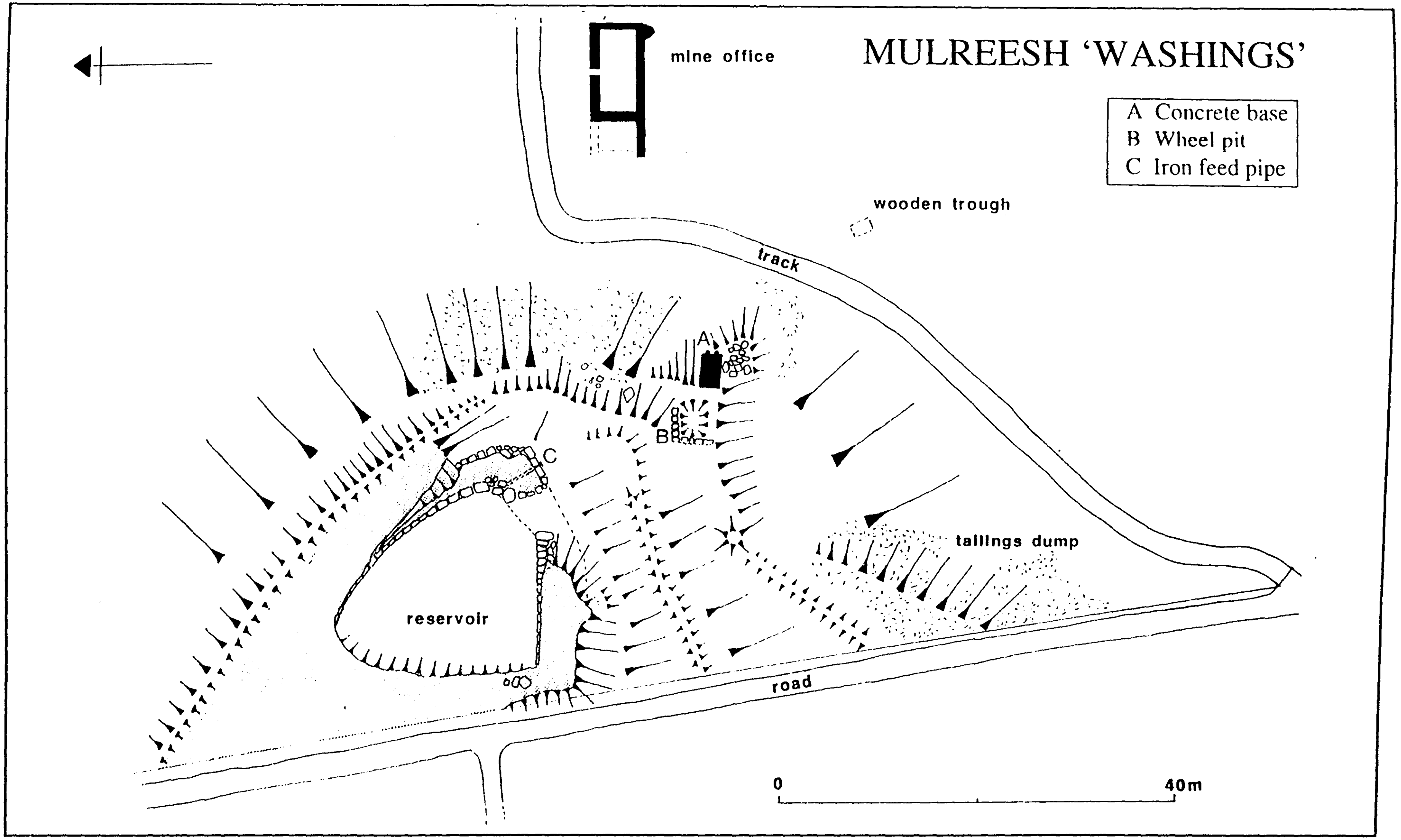

Figure 5.6 Detailed survey of the 'Washings' part of Mulreesh leadmine
(Site 2 in Figure 4.2 and Table 4.3)

This pipe connected to an over-shot wheel that gravity-fed water to a trip hammer or stamp. A wheel-pit (B) of unknown depth is situated to the west of a concrete base or plinth (A) which is approximately 2 m long and 1 m wide. The interior of this feature is lined with roughly dressed rubble. A single concrete block forms what would have been a pair of wheel plinths (A). Two large iron bars, presumably bolts, protrude in front of the blocks. Large pieces of timber work have been dislodged from the site and these lie alongside the second dislodged concrete plinth. Fifteen metres from the stamp-base are the remains of a wooden trough buried by fine tailings. This is probably the remains of a washing trough. A building to the east of the stamp-base is described as the mine-office, (Callender and Macaulay 1984). The building incorporates rough dressed stone and an unusually large buttress on its gable end.

5.3.6 Portnealon Mine Survey (Site No.5 in Figure 4.2 & Table 4.3)

Historical note:

The mine workings (Figure 5.7) lie on a gentle slope adjacent to the north-east shore of Loch Finlaggan at Grid reference NR391678. The works are named after the 18th century 'tack'[5] called '*Portineilan*' which refers to "the port serving the islands of loch Finlaggan" (Callender and Macaulay 1984). The *Book of Islay* records (Smith 1895, 458) that "Portnelan" was a vein discovered in *c.*1745 along with the vein at Balgrand (Ballygrant, Site 7 Figure 4.2). When reporting on the state of the mines in 1770, Mr Alexander Shirriff considering the possibility of linking the mine with nearby Mulreesh, wrote:

> " Portnealon vein is discovered at the surface in different places with some ore in it for trying of the vain. A cross cut is driving from Portnealon Loch, [Loch Finlaggan] advanced in open cast 24 fathoms [*c.*480 m] and close drift 22 fathoms will cutt the vain in drawing 5 fathoms... If on driving north on this vain it be found to carry ore, it will serve as a level to Mulrees works to which it points. The distance may be 400 fathoms and will, when at the Mulrees works, be 12 fathoms under the old soles" [old worked-out areas] (Smith 1895, 459).

The description accords well with the surface evidence obtained from earthworks survey and at this conjecture, given the distance between the mines, attempts to link the two at that period would been very ambitious indeed.

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[5] Tack is a term used for a parcel of rented land

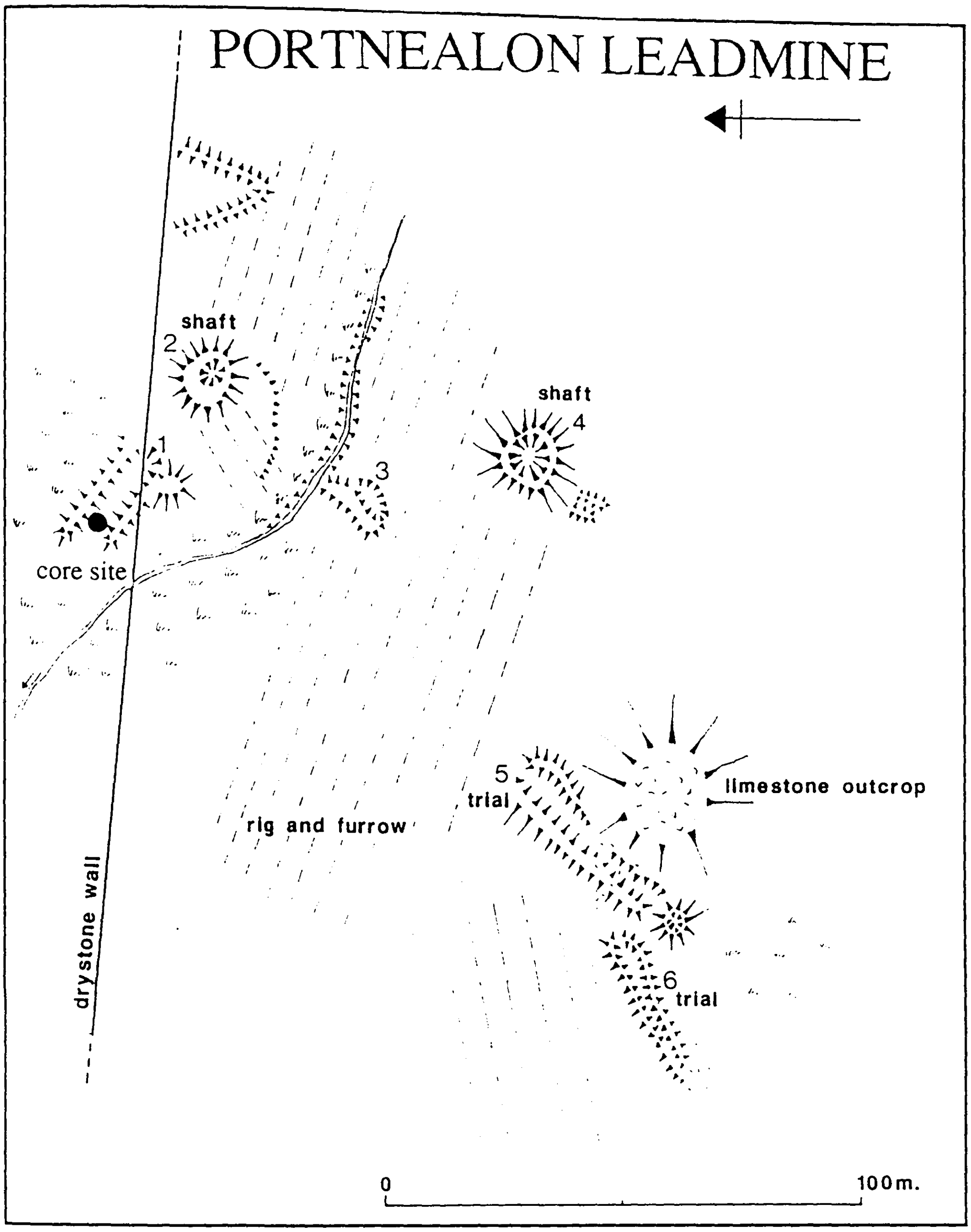

Figure 5.7 Portnealon leadmine survey (Site 5 in Figure 4.2 and Table 4.3).

The earthworks survey was undertaken in bright conditions using the methods described in Chapter 3 Section 3.2.1. The following text has numbers placed in parentheses which are also located in Figure 5.7 to aid identification. A water-filled adit (1) with denuded banks up to 1m high is transected by a drystone wall that forms a boundary between the Finlaggan and Keppols Farms. A shaft (2) with surrounding spoil up to 2.5m is situated 20 m to the north. A series of trials (3) (open cast workings) have a depth of about 1m and each is surrounded by spoil which is 1-2m high in places. On roughly the same alignment as shaft (2), there is a second shaft which has internal banks 2m deep with surrounding spoil to a height of 2 m.. To the south of these workings a natural limestone outcrop is flanked by two deep trials (5-6) that traverse upslope for about 47 m. All the features mentioned lie over the remains of rig and furrow which are clearly earlier than the lead mining period.

5.3.7 Robolls Mine Survey (Site 6 in Figure 4.2 & Table 4.3)

These workings are to the south east of loch Finlaggan on sloping ground about 60 m from the eastern shoreline. An account of Robolls mine does not appear in the Book of Islay and it has been suggested by Wilson (1921) that the mine dates to a later period than the 18th century. He also claims that the site was one of the last to be worked but provides no evidence to support this theory. Alternatively, these workings may not have been visited by Alexander Shirriff in 1770 and this may account for their absence in contemporary records.

A north by north-west trending trial (1) has within its interior four shafts now full of refuse that mask their true depth. Mine spoil is evident both outside and within these features and a large spoil tip (2) is situated 5 m north of these shafts. North of these features is a north-south trending trial that is joined at right angles by a tertiary dyke (3). Spoil tips are evident along the length of the open-cast feature (4). Immediately south of this area a building (5) has been interpreted as a smithy (Callender and Macaulay 1984) The building is rectangular and comprises rough dressed stones one to two courses high. Distinction between room divisions and entrances can be made.

Directly south of the building a farm track runs in a southerly direction towards the village of Ballygrant. At right-angles to the track runs a smaller track that is flanked on both sides by two drainage adits (6) and (7). At the time of the survey the interior of the adit (7) to the south of the track was refuse-filled and flooded to a depth of about 2 m.

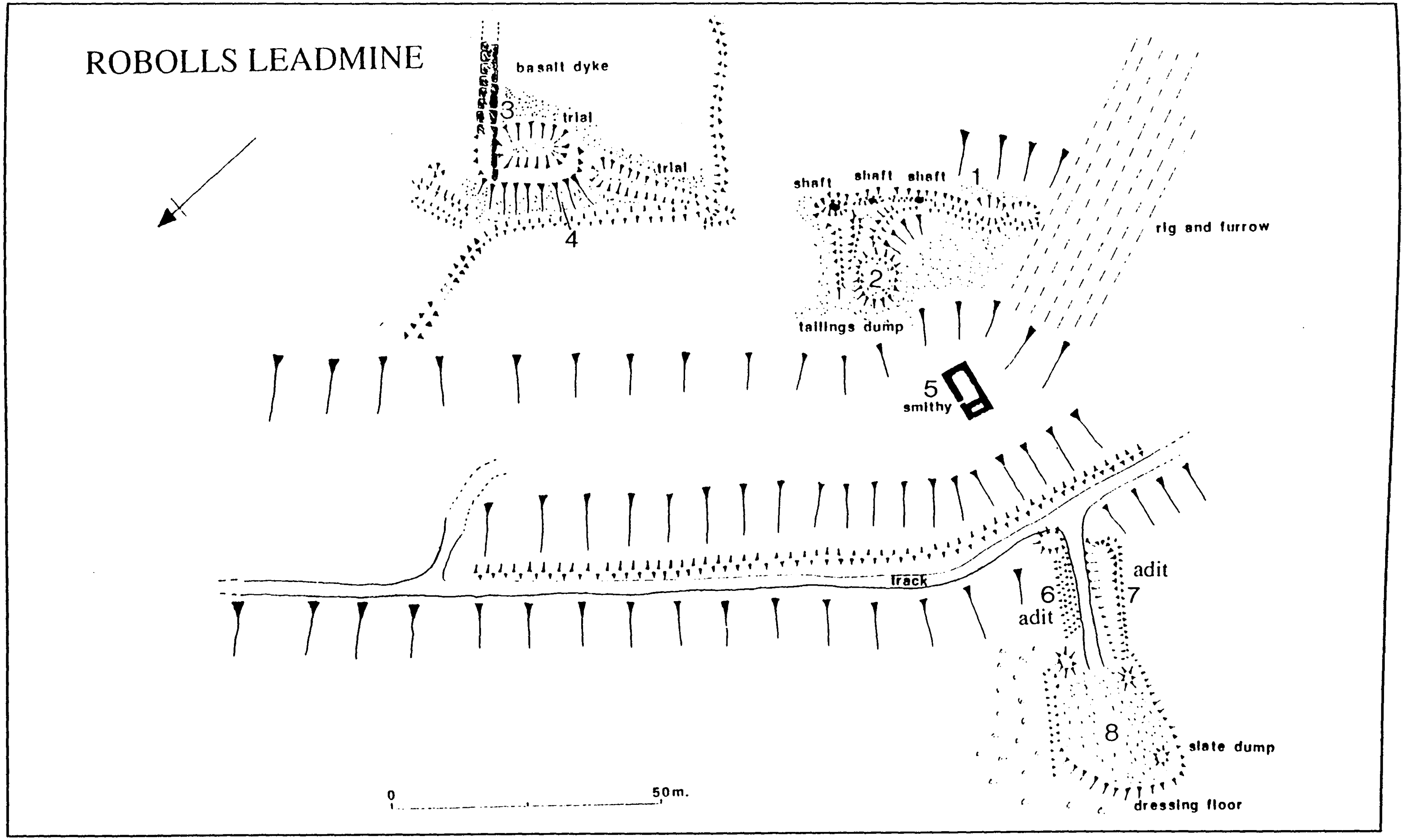

Figure 5.8 Robolls leadmine survey (Site 6 in Figure 4.2 and Table 4.3).

A large ditch and bank led from this feature for a distance of about 20 m, the interior of which contained seepage water draining in the general direction of the loch. Close by there is a flat area that is poor in surface vegetation with mine spoil covering an area of 30 square metres. A denuded spoil tip consisting of slates and phyllites suggests deliberate sorting of mine waste and perhaps points to the area's use as a dressing floor (8). It is suggested that the open cast feature (1) containing the three mine shafts might date to an earlier episode of lead mining. The near-surface lead has been exploited to an unworkable depth and then later re-worked by the more technically advanced shaft and adit mining method. Placing a date on the earlier open-cast is impossible in the absence of some relative dating material but it is most probably much earlier than the 18th century and most likely 17th century in origin.

5.3.8 West Shore Site Survey (Site 21 in Figure 4.2 & Table 4.3)

At the head of Loch Finlaggan and situated on a geological boundary between phyllite and limestone are two shafts (1) and (2) (bell-pits) with surrounding spoil. The banks of spoil surrounding the shafts stand approximately 1.5 m high. The northern shaft rests on a plateau that resembles a lynchet[6] (3). This terrace has formed as a result of continued cultivation which encloses a patch of rig and furrow that is only visible in suitable lighting conditions. The southern shaft is close to the loch and has slightly more elongated sides than the northern shaft. A trial (4) has been cut into the side of the shore forming a small basin with quite pronounced internal slopes. One can only speculate if the two shafts were ever joined by a gallery. In all probability they were as this would allow an ingress of air making it possible for the miners to exploit a pipe-vein of galena. In 1993 the writer recovered an iron kibble-hoop[7] from the loch adjacent to the trial. The site does not appear to have developed beyond two shafts and a trial. There is however possible evidence of quarrying close to the Finlaggan Burn where a small rectangular face has been cut away from the outcropping limestone.

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[6] Lynchet- a term used to denote a terrace formed by cultivation.

[7] A Kibble hoop is part of a heavy iron-clad barrel that was winched up and down the mine shaft.

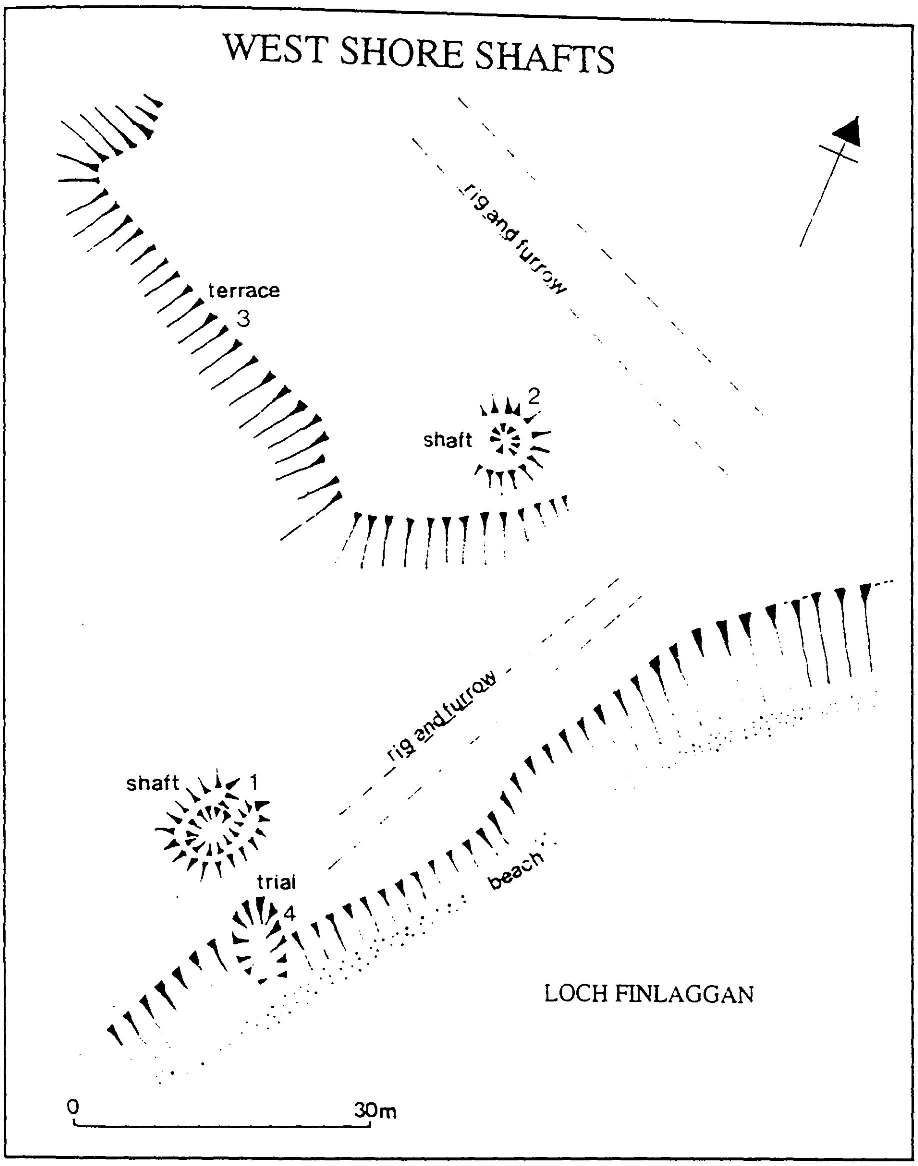


Figure 5.9 West Shore Shafts survey (Site 21 in Figure 4.2 and Table 4.3).

5.3.9 Implications of the archaeological surveys in the Finlaggan catchment

With regards to the Mulreesh mine it is concluded that the archaeological remains of the mine are more extensive than previously thought. Several tons of tailings were taken for use as hard-core in road improvement schemes in the early 1970's (D. Boyd pers comm). It would seem then, that what survives today is only a fragment of what must have been an extensive mining operation. One can only envisage the scale of the drainage and hauling systems employed at the site. A series of over-shot troughs probably supplied waste water from the engine house to the washing plant. Hauling by cable-winch would have required a system of hawser and pulleys to lift the ore in heavy iron-clad kibbles. All this can only be estimated from the position of the shafts depicted on Vercoe's gallery section drawing and the fragmentary remains of hawser foundation blocks adjacent to the engine house.

Two phases of mining activity at the site are established with some degree of certainty. The first is by tenant farmer-miners who occupied the dwellings (ABCD). A tentative date of the mid to the late-18th century is ascribed to this part of the site based on the survey evidence and contemporary accounts. The large open-cast feature following the Tertiary dyke and the line of refuse filled pits (Features 15 & 16 in Figure 5.4) may well date from the 18th century or earlier.

The rest of the Mulreesh complex owes its development to the early and later parts of the 19th century. This is indicated by the upstanding archaeological remains, historic accounts and 19th century output returns listing annual lead and silver production. Importantly, the survey has shown quite conclusively that the adits at this mine do not drain towards Loch Finlaggan. It will be recalled from Chapter 1 that such information was significant in our understanding of the routing of heavy metal contamination into loch sediment.

The results of field survey closer to Loch Finlaggan have shown that the mines are not as extensive as at Mulreesh. However all the adits identified do enter into or flow in the direction of the loch. At the Portnealon site it is concluded that only one phase of mining activity can be established and this phase dates to the late 18th century according to contemporary accounts. The most impressive remains at this site are the bell-pits and the open cast feature. Robolls mine to the south of Portnealon has extensive remains and here two phases are indicated by the archaeological remains and only limited references in the historical accounts. The first phase of activity was a small-scale attempt to extract near-surface outcrops of lead from a north by north-west trending open cast. This feature was then worked to an unknown depth by the insertion of four mine shafts at a later date.

The adits are flooded and seepage flows in the direction of the loch aided by the steepness of a slope, at the base of which the adits emerge.

The survey has shown that the remains of the mines are in parts extensive and 18-19th century phases of activity can be identified. However there are several features that in all probability are much earlier than the two phases recognised, especially at Mulreesh and Robolls mines. Importantly, the survey has identified the scale of the 18th and 19th century mining phases and their distribution even though many features have been backfilled and cleared for track repair. The survey has also shed new light on the drainage routes from the sites. From the stand point of archaeology the fieldwork makes an important contribution to the industrial record which hitherto has been lacking in this region of Scotland.

5.4: Geochemical Survey in the Finlaggan Catchment

5.4.1 The Catchment Distribution of Pb, Zn and Cu based on the DRL Geochemical Survey

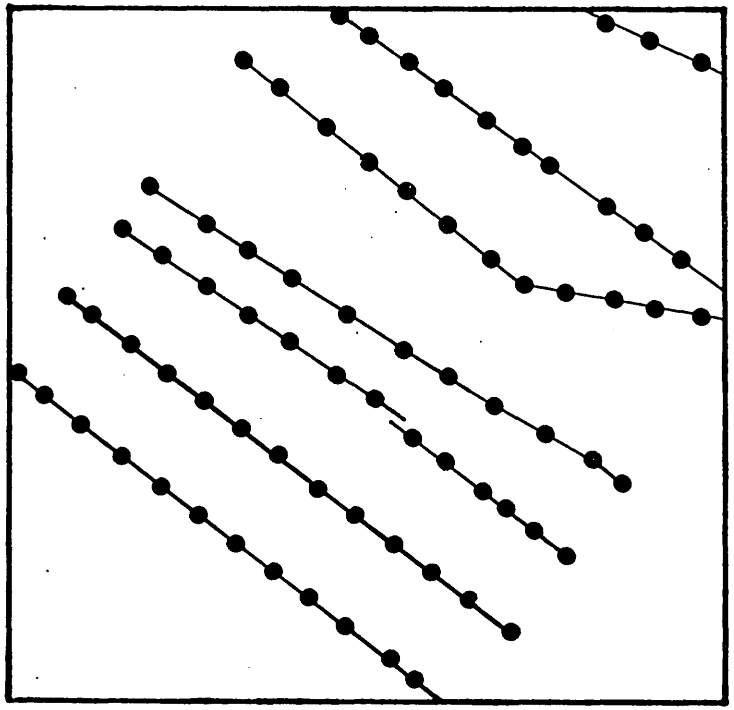
The geochemical survey undertaken by Domego Resources Ltd (DRL) in 1983 at the site of Portnealon determined the concentration of Pb, Zn and Cu in surface soils. The aim of the survey (see Chapter 4 section 4.4) was to assess the extent of mineralisation in order to establish the economic potential for reworking existing ore bodies at the mine. The highest values obtained within the samples was 5000 ppm Pb with a secondary anomaly of 2280 ppm (Table 5.2). These two values reflect the highest levels of soil contamination within this study area. Using the methods described in Chapter 3 Section 3.3 this author has re-plotted the data to show the relative distribution of these metals across the site. Table 5.2 below summarises the statistics of the data set used in the contour plots.

STATISTICS	Pb	Zn	Cu
Observations	79	79	79
Maximum value	5000	2280	142
Minimum value	15	12	2
Mean value	139	105	22
Median value	129	108	21
Standard deviation	96	66	20

Table 5.2 Statistical characteristics of Pb, Zn and Cu distributions based on DRL geochemical data from Portnealon Mine, Finlaggan catchment (all values in ppm).

5.4.2 The catchment distribution of Pb (Figure 5.10)

The distribution pattern for Pb shows an outlying value of 5000 ppm with a minimum value of 15 and a mean of 139 ppm respectively. The standard deviation values suggest high between-sample variation. The geochemical distribution map for Pb shows two halos that are related to the position of mining activity. The 5000 ppm halo is related to Feature 3 in Figure 5.3 and the 2280 ppm halo is related to Feature 4 in Figure 5.7 respectively. There is a sharp division between the blue (below 10 ppm) category and the green (between 133 ppm and 224 ppm) category. This division appears to be entirely dependent on the underlying solid geology. The green region demarcates the calcareous Dalradian Limestone and the blue area defines the acid phyllites and shales. There is a clear pattern of dispersal away from the open cast feature that corresponds very well with the demarcated region proposed by the author of the DRL survey.



DRL Geochemical Survey
Pb, Zn and Cu soil sample
points

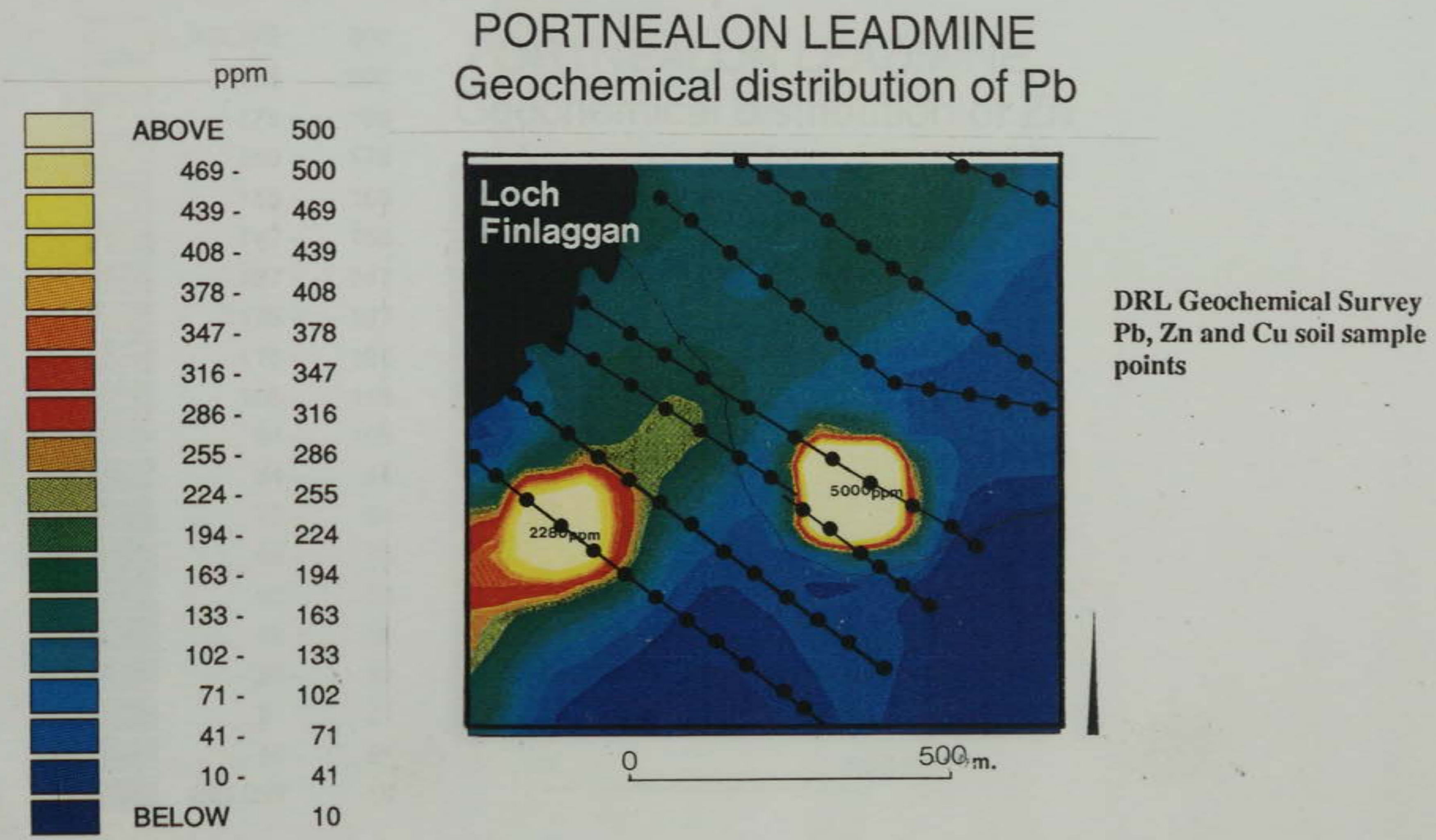


Figure 5.10 Distribution of lead in surface soils across Portnealon leadmine (after Domego Resources Ltd 1983).

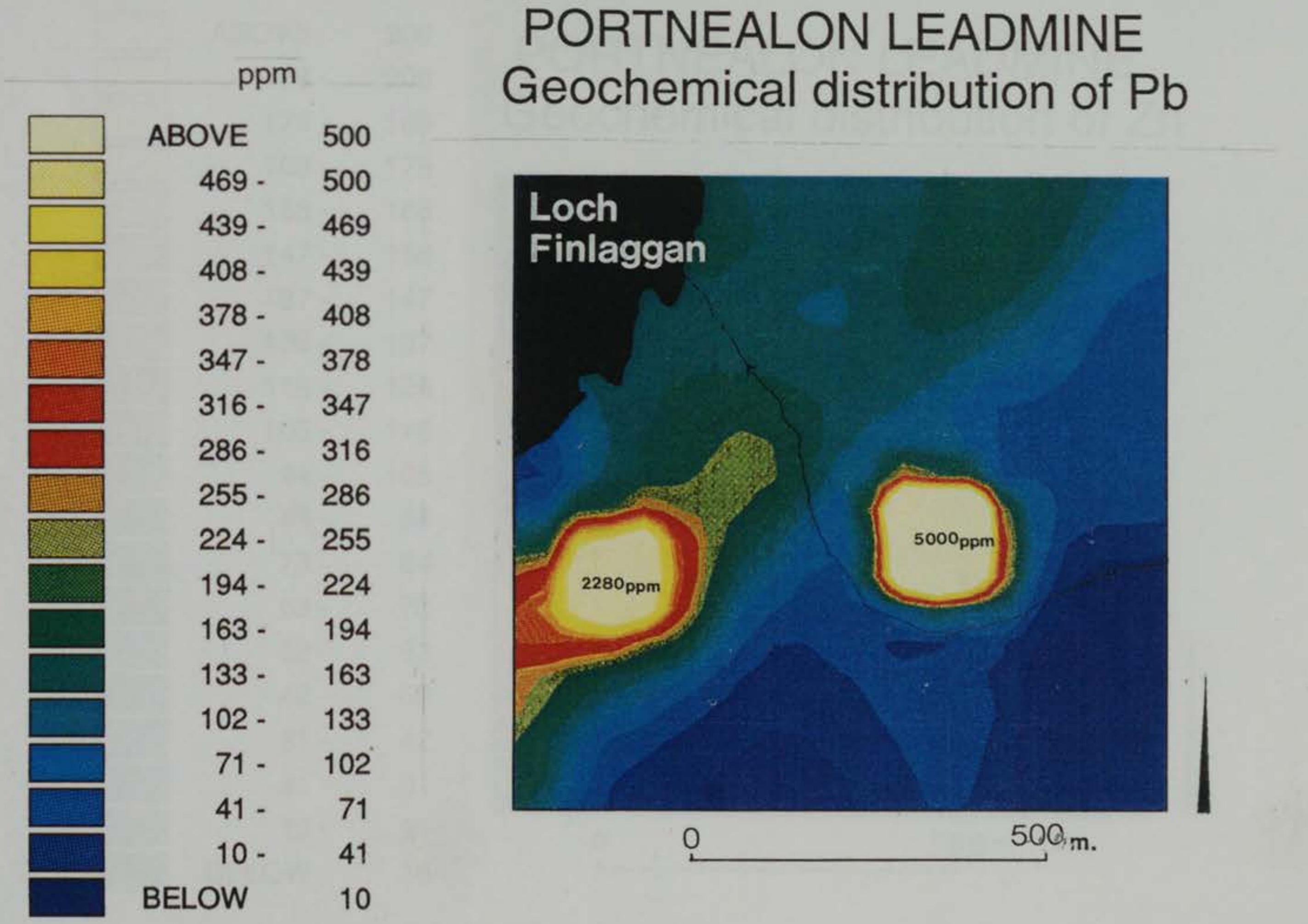


Figure 5.10 Distribution of lead in surface soils across Portnealon leadmine (after Domego Resources Ltd 1983).

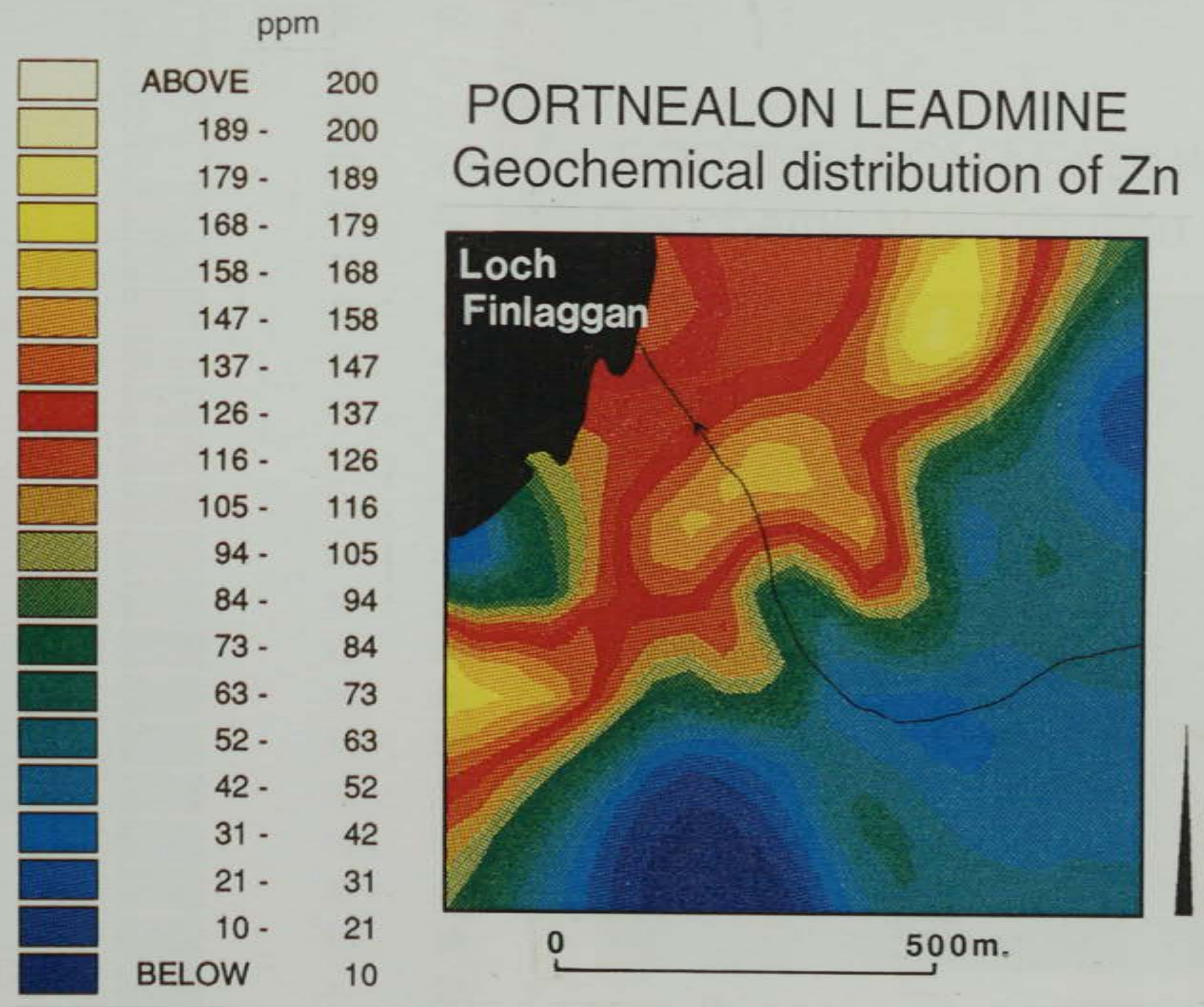


Figure 5.11 Distribution of zinc in surface soils across Portnealon leadmine (after Domego Resources Ltd 1983).

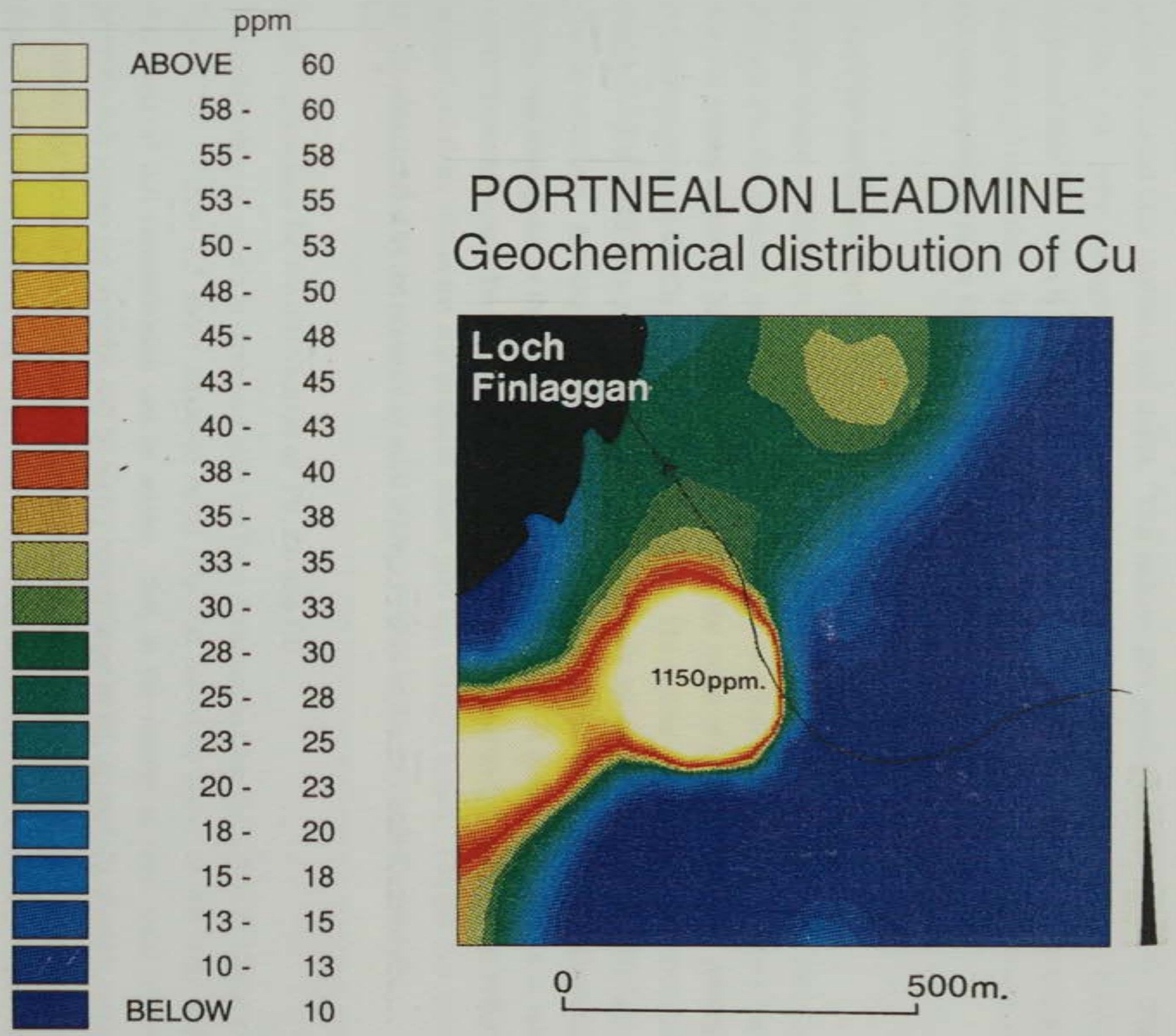


Figure 5.12 Distribution of copper in surface soils across Portnealon leadmine (after Domego Resources Ltd 1983).

The regions shown in red have Pb values of between 286-347 ppm and these values are closely confined to the spoil tips and show that Pb is not widely dispersed as is the case for Zn and Cu. A value of 150 ppm Pb is within the range of 'normal' soils but is well above the 'background' established at 24 mg kg^{-1} (see Chapter 9, Figure 9.2).

5.4.3 The catchment distribution of Zn (Figure 5.11)

Zinc has a recorded maximum of 2280 ppm with a minimum of 12 ppm. The mean value was 105 ppm with a lower standard deviation that reflects moderately high within-sample variance. The geochemical distribution of Zn shows a clear demarcation between the two geological types. The plot shows a wide dispersal pattern for Zn across the limestone, with lower Zn values recorded over phyllites and shales. This reflects Zn's greater mobility over Pb. The presence of mine tailings has led to isolated concentrations of Zn that is more widely dispersed than Pb. This is to some extent predictable given this metals tendency to be mobile in more acid conditions. In general we are observing a more natural dispersal pattern due to the geomorphology of the site.

5.4.4 The catchment distribution of Cu (Figure 5.12)

Copper attains a maximum of 142 and a minimum of 2 ppm respectively. The mean value is 22 ppm and the standard deviation is low in comparison to either Pb and Zn and shows that there is greater similarity in Cu concentration across the range of values. There are striking similarities between the Cu and Pb dispersal patterns. The highest value attained is shown on Figure 5.12 at 1150 ppm and forms a concentric anomaly set on the edge of the limestone-phyllite boundary. A background value of below 10 ppm is shown as the blue area becoming lighter and then green as the transects traversed the limestone. It is apparent that Cu is not widely dispersed across the Limestone as is Zn. This is due in part to Cu's complexion within organic fractions within the soil matrices where they are strongly bound. Copper has been locally concentrated by the presence of mine tailings evident by the very high Cu anomaly.

5.4.5 Implications for the distribution of Pb, Zn and Cu

The distribution of metals in terrestrial soils has been discussed in Chapter 2, Part II. (see also Figure 2.7). Figure 2.5 above modelled metal dispersal behaviour above an ore body and demonstrated that contamination can be severe. Soil in the vicinity of mine spoil where sulphide-rich wastes are common will be highly concentrated in Pb, Zn and Cu, in most cases well above the 'normal' background concentration established in 'unpolluted' soils. As we have seen, the relative mobility of a given metal is a key factor in its ability to be laterally dispersed within the soil. Downslope distortion in the metal distribution patterns (i.e. the move away from symmetrical patterns) is usually the effect of physical movement of metal-bearing particles by soil creep aided by solution and precipitation from laterally moving subsurface waters (Hawkes and Webb 1962, 173). This latter point is of significance at Portnealon where the substrate in the vicinity of the mined area is free draining Dalradian Limestone. The groundwater emanating from the more acid phyllites and shales upslope will be acidic and charged with precipitates high in Fe and Mn oxides that will promote the release of Zn and Cu ions (by dissolution) from the lattices within the gangue minerals in the mine spoil (see below, Section 6.8.7). Furthermore the fact that Portnealon is on a gentle incline, has certainly been a contributory factor in the greater dispersal of Zn. At the site, geological boundaries are sharply defined as a result of the retention or relative mobility of these three metals. In the case of Pb, Zn and Cu the concentration is significantly lower on the acidic phyllites and shales. The permeability of the limestone region is clearly leading to the dispersal of metal-rich groundwaters which are leaching from the site.

This theory was tested in a collaborative project between the author and an undergraduate of ecology (Short 1994 unpublished). The project has investigated the relative Pb, Zn and Cu concentration of leachates within a sediment-filled adit leading from the Portnealon mine (see Feature 1 in Figure 5.7 above). A 120 cm deep core was obtained from the adit using a Russian corer and analysed using the same acid digestion procedure described in Chapter 3 section 3.7.4.

The results of this study are summarised in Figure 5.13 and show that the lower sediment sections contain relatively high concentrations of Pb and Zn. This indicates sub-surface movement of water from the old mine workings. The concentration of Cu is greatest near the surface and has probably been enhanced near the sediment-water interface region (*c*.25-30 cm depth) as a result of redox control. In contrast to Cu, the concentrations of Pb and Zn share a similar trend throughout the core. In the case of Pb, the concentration is lower nearer the surface and increases to a maximum of 400 mg kg^{-1} at 110 cm in the brown silt layer. Lead may have been diffused from the upper region of the core and concentrated in the more minerogenic sections of the profile. ANOVA statistics (analyses of variance) for each metal showed there was significant variation between the samples and the relative order of concentration established as Pb>Zn>Cu. The results confirm that the adit is still receiving moderately high levels of Pb and Zn from mine drainage almost two centuries after the mine ceased operations.

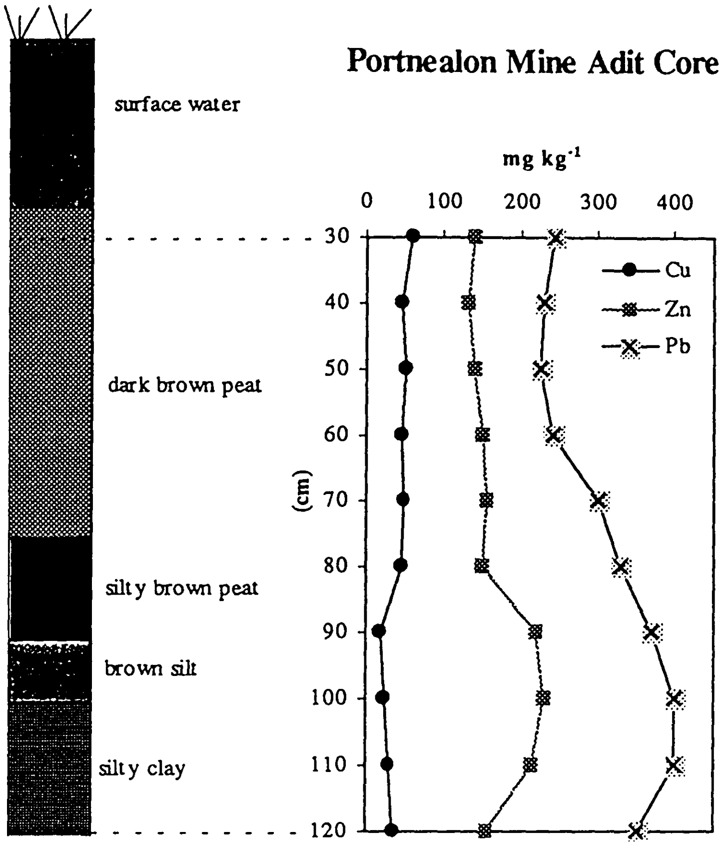


Figure 5.13 Total Cu, Pb and Zn concentration of the Portneaolon adit core based on AAS analyses (after Short 1994). See Feature 1 in Figure 5.7 for the location of this core.

5.4.6 Conclusions

The re-interpretation of the DRL geochemical survey shows conclusively that Pb, Zn and Cu have been significantly enhanced in the soils across the former site of Portnealon. The two dimensional plots relate the close conformity between the two highly anomalous zones and the two extant mine features Feature 3 and 4 in Figure 5.7. In the case of Portnealon the distribution of heavy metals in soils is tightly controlled by the underlying bedrock geology. The degree of slope and the presence of the burn and adit leading from the site are the principal vectors in moving heavy metals into loch sediment. More recent geochemical analyses of sediments obtained from the adit (Feature 1 in Figure 5.7) have shown that seepage waters flowing through the adit have moderately high concentrations of Pb, Zn and Cu (Short 1994).

The results of work undertaken on sediment recovered from the loch will be discussed in the following section.

5.5 Geochemical Investigations in Loch Finlaggan

5.5.1 Bathymetry Survey and Core Recovery

As part of investigations for the survival of archaeological remains around the island of *Eilean Mor* a bathymetric survey was undertaken by the British Geological Survey in liaison with the National Museums of Scotland (NMS) (Ruckley and Chesher 1990). This survey was undertaken using echo soundings and a chart record interpolated to the appropriate scale. The results of this survey are produced as Figure 5.14. The plan records depth in feet with associated conversion scale in meters. The deepest part of the loch is approximately 40ft (12.19m) with the shallowest regions being the southern portion of the loch where the mean depth is in the region of 10 feet (3.04m). The data from this survey was available to the writer and used as a guide to locate four optimum coring stations and these are shown in Figure 5.14.

Core 2 was recovered in September 1992 and was 82 cm in length. Examination of the other cores back in the laboratory showed that Cores 1, 3 and 4 were severely degraded due to sediment cracking probably due to a rapid loss of pore-water in the upper sections of the core. Core 2 was seen to have some small fissures running laterally across the sediment. These were probably due to desiccation and/or sediment mixing due to water turbulence at the sediment water interface. It was predicted at this stage that the results from ^{210}Pb determination would reveal the degree of sediment mixing at the surface. Sediment mixing is one of the major constraints in palaeolimnology (Crusius and Anderson 1991, Cumming 1993). Mixing may be due to: (1) bioturbation near the sediment water interface, (2) gas movement from the underlying sediment, (3) turbulent mixing (4) mechanical disturbance (i.e. lifting the core/core transportation). Any of these parameters will undermine the integrity of the sediment's preservation and limit the establishment of a reliable time-frame. It is therefore unfortunate that the results of ^{210}Pb analyses show that there has been mixing in the upper section of the core.

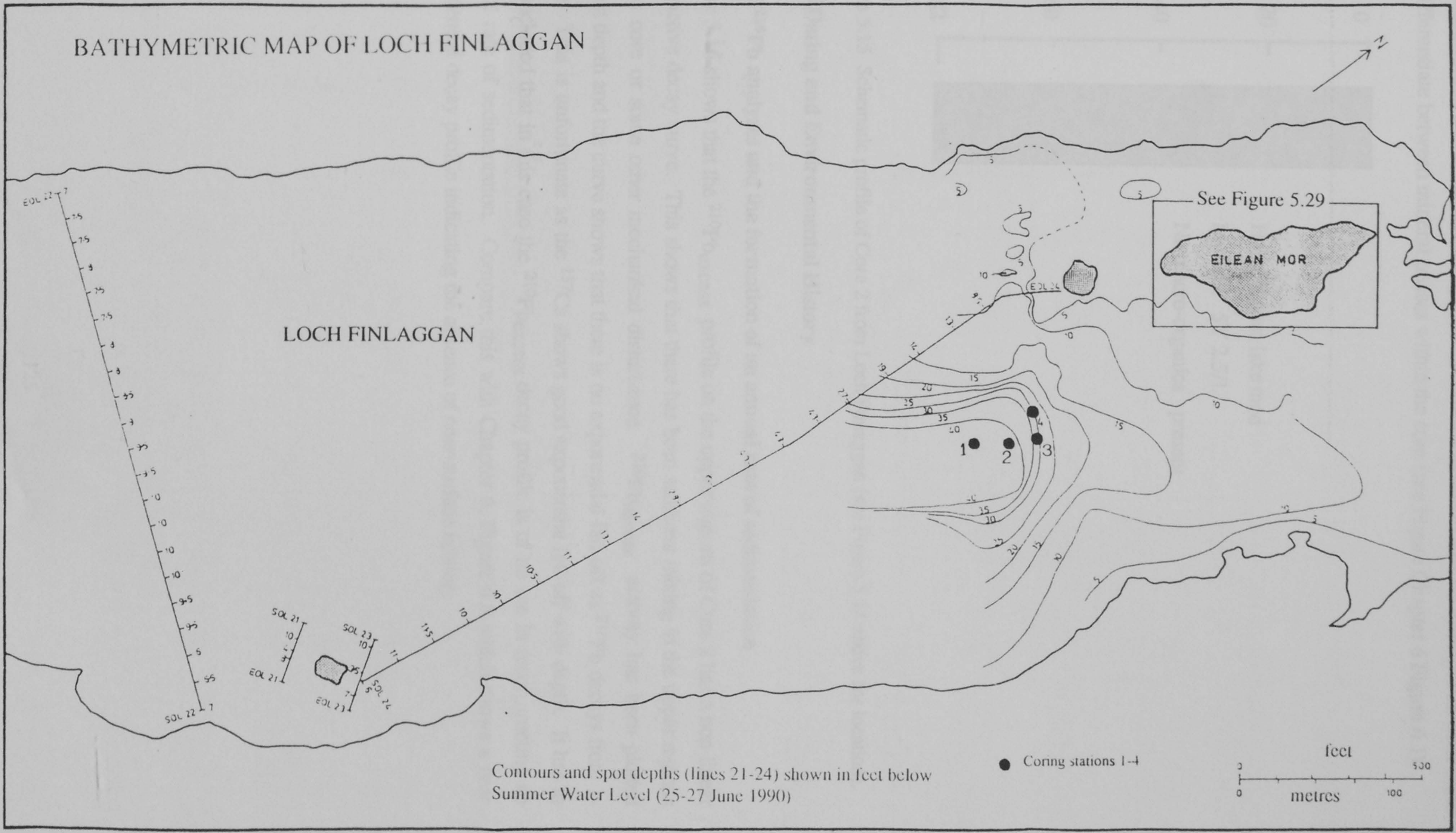


Figure 5.14 Bathymetric survey map of Loch Finlaggan (after Ruckley and Cheshire 1990) inset is *Eilean Mor* or 'Finlaggan' archaeological site.

The sediment consisted of a very flocculant lacustrine mud with no variation in texture or colour. No plant-macro material was observed within the core. No laminations or changes in the composition of the sediment could be discerned and due to this no attempt has been made to differentiate between mineral phases within the core (see Figure Chapter 6 Figure 6.1).

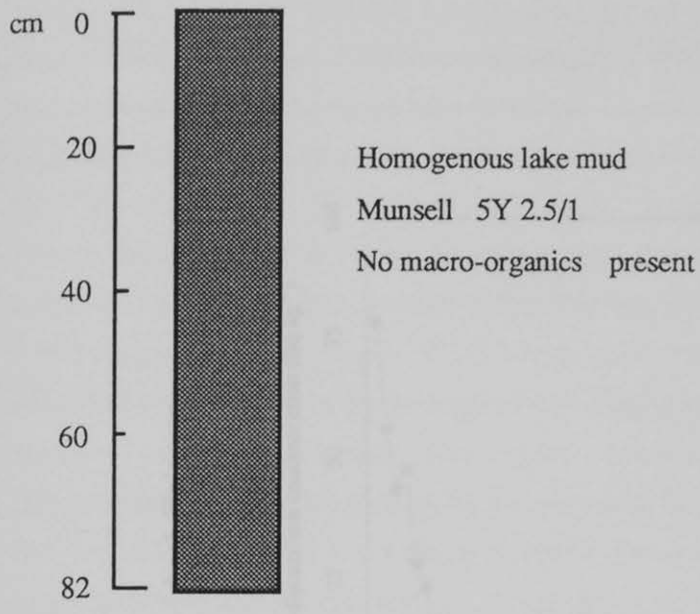


Figure 5.15 Schematic profile of Core 2 from Loch Finlaggan (see Figure 5.14 above for location).

5.5.3 Dating and Environmental History

5.5.4 ^{210}Pb analyses and the formation of an annual rate of sedimentation

Figure 5.16 shows that the $^{210}Pb_{excess}$ profile on the upper regions of Core 2 has a non-linear radioactive decay curve. This shows that there has been sediment mixing in the upper regions of the core or some other mechanical disturbance. $^{210}Pb_{excess}$ activity has been plotted against depth and the curve shows that there is no exponential fall-off as ^{210}Pb decays from 1-11cm. This is unfortunate as the ^{137}Cs shows good exponential fall-off with depth. It has to be concluded that in this case the $^{210}Pb_{excess}$ decay profile is of no use in reconstructing an annual rate of sedimentation. Compare this with Chapter 6, Figure 6.8 which shows a near exponential decay profile indicating the absence of near-surface mixing.

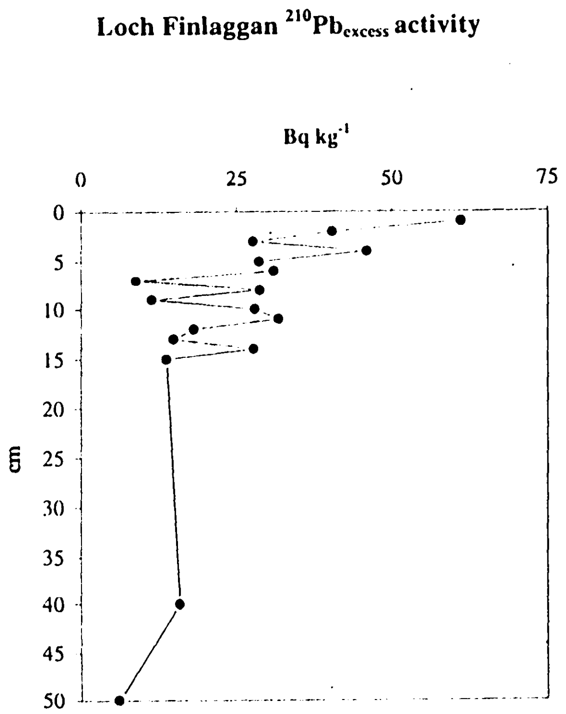


Figure 5.16 Depth versus $^{210}Pb_{excess}$ activity.

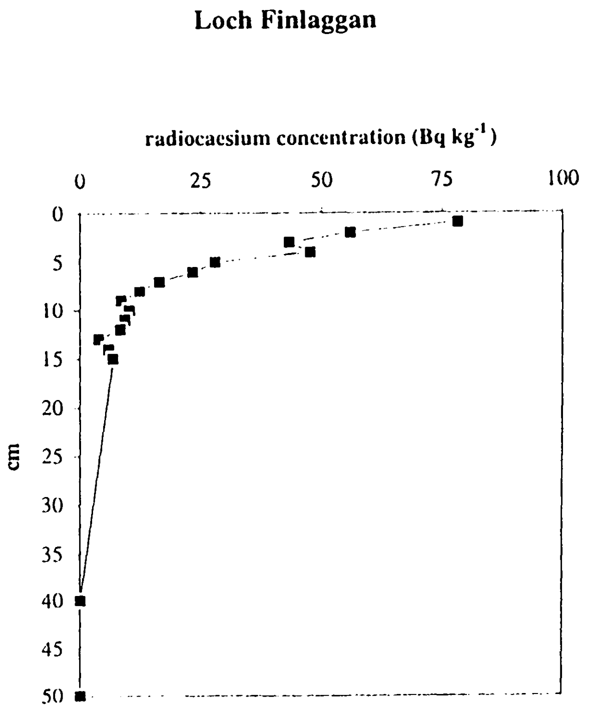


Figure 5.17 Depth versus ^{137}Cs activity.

5.5.5 Chronological support using ^{137}Cs

Radiocaesium activity has been plotted against a depth of 1-50 cm (Figure 5.17). The depth v concentration curve shows that there is near exponential decay from 1 cm with 78.2 ± 1.7 Bq kg^{-1} at 1 sigma down to 17 cm with 5.9 ± 1.3 Bq kg^{-1}. There is a subsidiary peak at 4 cm with 47.6 ± 1.3 Bq kg^{-1} which is accounted for by the effects of the 1963 maximum weapons testing fallout deposition. Without a supporting ^{210}Pb chronology the relative precision of this event cannot be established with certainty. Overall this radionuclide appears not to have been affected by the mixing effects in the upper region of the core which have seriously altered the ^{210}Pb decay profile. Several reasons to account for the apparent mixing towards the upper surface can be advanced. It has been mentioned that Loch Finlaggan is one of the longest lochs in Islay. In adverse weather conditions near-sediment turbulence can be very high. Wave height can be in excess of 1m which combined with strong currents could lead to continued resuspension of the sediment-water interface. Alternately, there are a number of biological benthic fauna such as *chirinomidaea* which burrow into sediment and cause mixing (Chapter 2 Section 2.4.2). Finally, as we have seen, the rapid loss of pore-water in cores 1, 3 and 4 could account for their damaged state. Paradoxically, although the integrity of the upper section of Core 2 appeared good it is now clear from the ^{210}Pb record that the upper section of the core has been disturbed.

5.5.6 Chronological support using pollen analyses

Pollen analysis was undertaken using the procedures outlined in Chapter 2 Section 3.9. Limited work was undertaken on 1-16 cm to establish the maximum concentration of the species *Pinus sylvestris* which is widespread among the estate plantations and gardens in the study area and date to the late 19th Century. No near surface peak in the concentration of *Pinus* pollen could be discerned.

5.5.7 Geochemical Inventory and Physical Properties of Loch Finlaggan Core 2

The chemical flux of Pb, Zn, Cu, Ca, Fe and Mn and physical properties are shown as heavy metal concentration plotted against depth and are expressed as mg kg^{-1} per dry weight of sediment (dw.0.5g). The heavy metal profiles are shown individually (Figures 5.18-5.27) and collectively to allow comparisons to be made. The heavy metal concentrations were established before the results of ^{210}Pb were known. The analytical procedure is described in Section 3.7.4. Summary statistics of these data are shown in Table 5.3.

Statistics	Pb	Zn	Cu	Ca	Fe	Mn
Observations	42	42	42	42	42	42
Maximum value	236	219	43	54	1725	4950
Minimum value	83	96	22	5	763	2800
Median value	124	156	28	25	1144	3500
Mean value	125.5	154.9	29	26.3	1167	3573
Standard Dev.	23.6	3.8	4.5	13	164.4	82.4

Table 5.3 Statistical characteristics of the chemical influx data obtained from Loch Finlaggan Core (all values in mg kg^{-1}).

Lead (Figure 5.18)

The depth versus concentration profile of Pb shows limited variance from the base of the profile at 82 cm to 26 cm where values range between 140-130 mg kg^{-1}. At the 26 cm sediment section there is a noticeable rise in Pb concentration of 236 mg kg^{-1}. There is a decline concentration to 12 cm where 100 mg kg^{-1} Pb is attained. from this position to the surface there is an increase towards the near-surface with 140 mg kg^{-1} decreasing at 2 cm with 135 mg kg^{-1}.

Zinc (Figure 5.19)

Zinc shows a similar trend to Pb concentration profile. From the base of the profile there is a slight rise to 68 cm with 170 mg kg^{-1}. A subsidiary peak can be observed towards the centre of the profile and at between 18-22 cm where 160 mg kg^{-1} is attained.

Copper(Figure 5.20)

From the base of the core the Cu concentration rises intermittently with 25 mg kg^{-1} to 40 cm. There is little variance in concentration until 18-20 cm where 38 mg kg^{-1} are attained. From this position there is a decrease in concentration to 10 cm where values rise at 6 cm with 36 mg kg^{-1}. There is a decline in concentration to 27 at 1-2 cm.

Calcium (Figure 5.21)

Calcium shows wide variability throughout the sedimentary profile. From the base of the core there is a protracted increase from 19 mg kg^{-1} to 66 cm with 49 mg kg^{-1}. From this position there is a sharp decrease in concentration to 56 cm where 17 mg kg^{-1} are attained. Upwards from this sediment section values fluctuate between 10-15 mg kg^{-1} until 26 cm where Ca increases to a sub-surface maximum of 44 mg kg^{-1} at 2-6 cm.

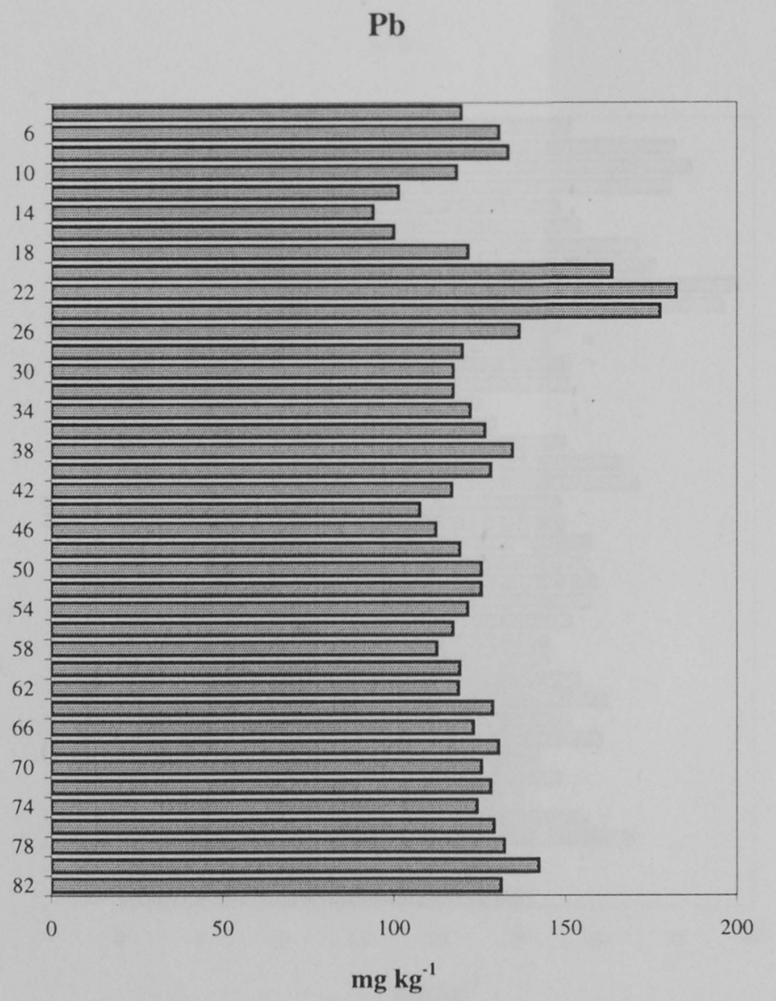

Pb

Figure 5.18 Depth versus lead concentration

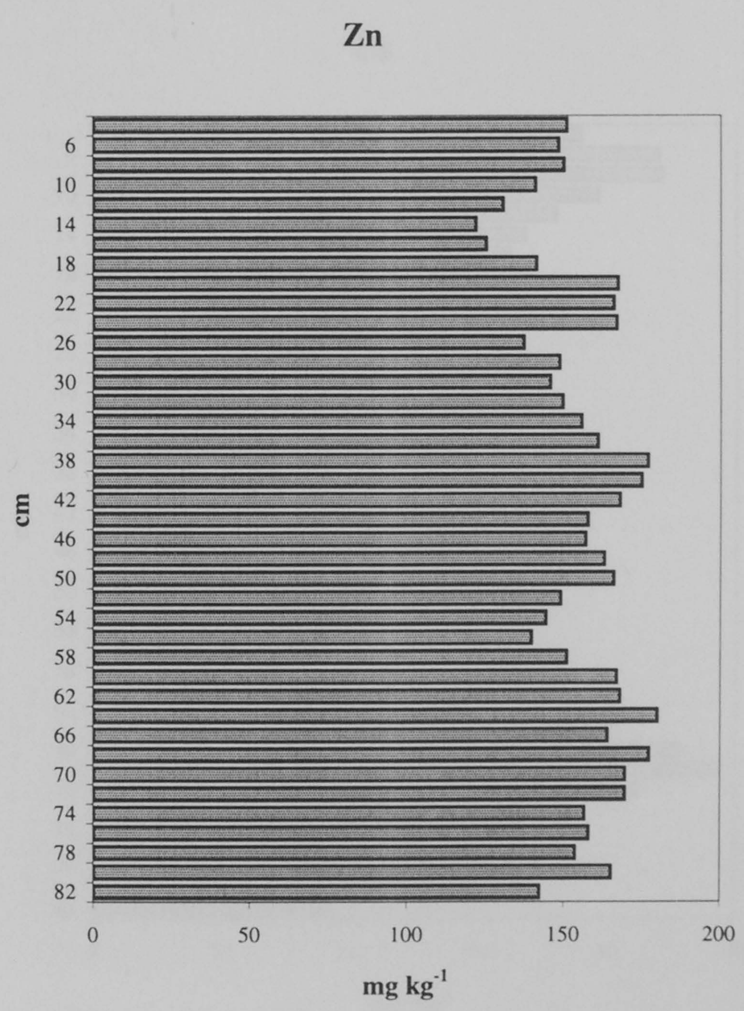

Zn

Figure 5.19 Depth versus zinc concentration.

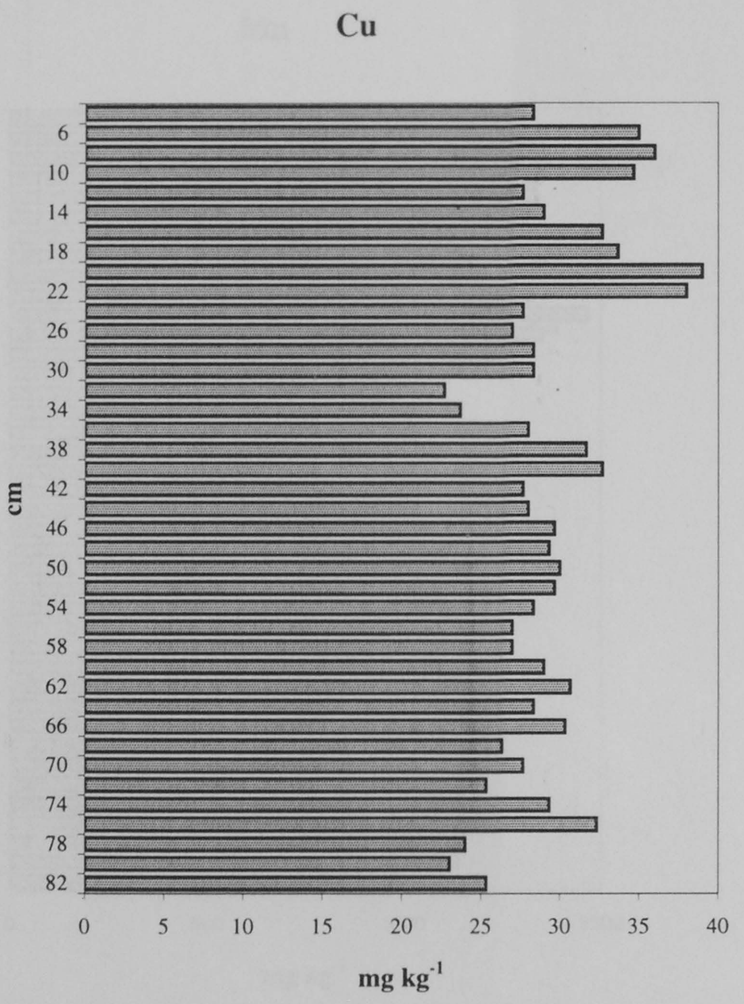


Figure 5.20 Depth versus copper concentration.

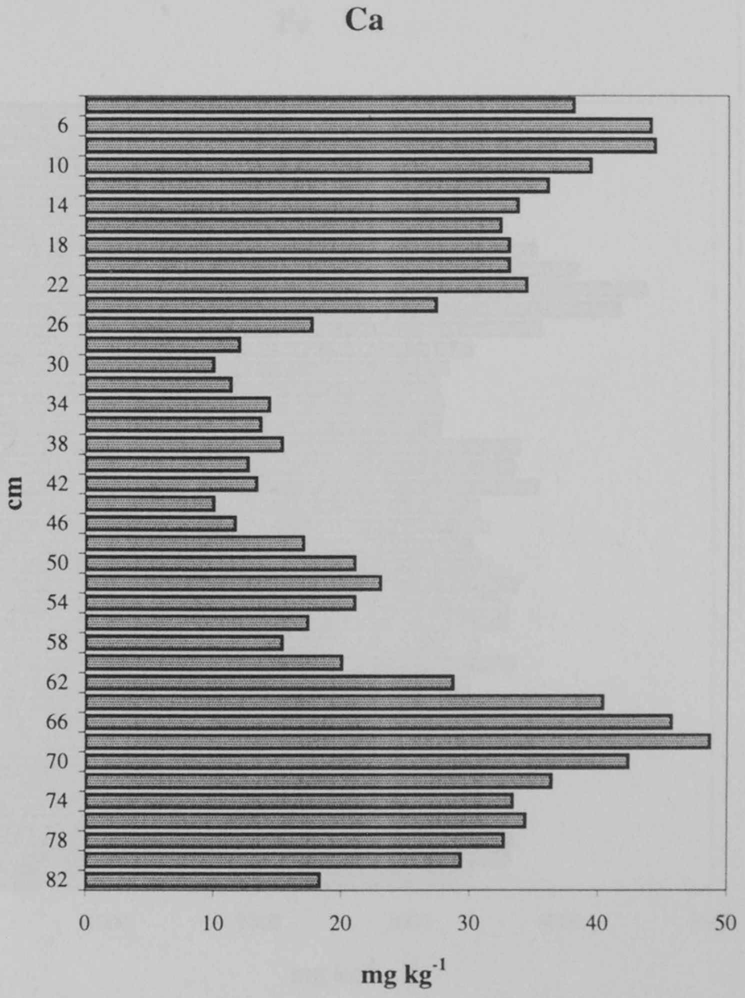


Figure 5.21 Depth versus calcium concentration.

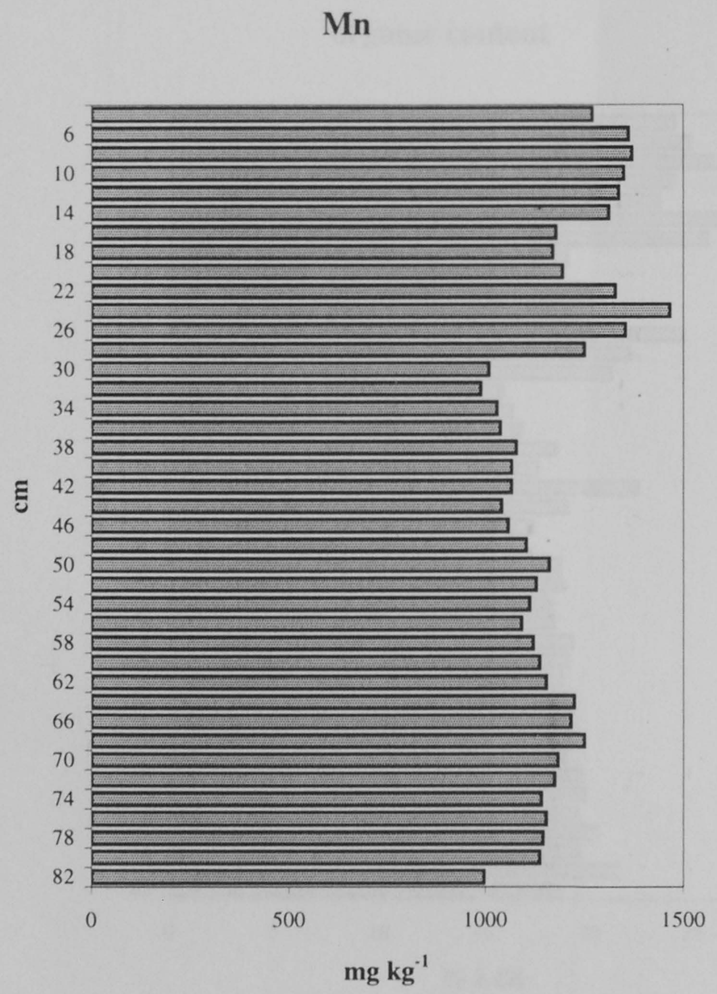


Figure 5.22 Depth versus manganese concentration.

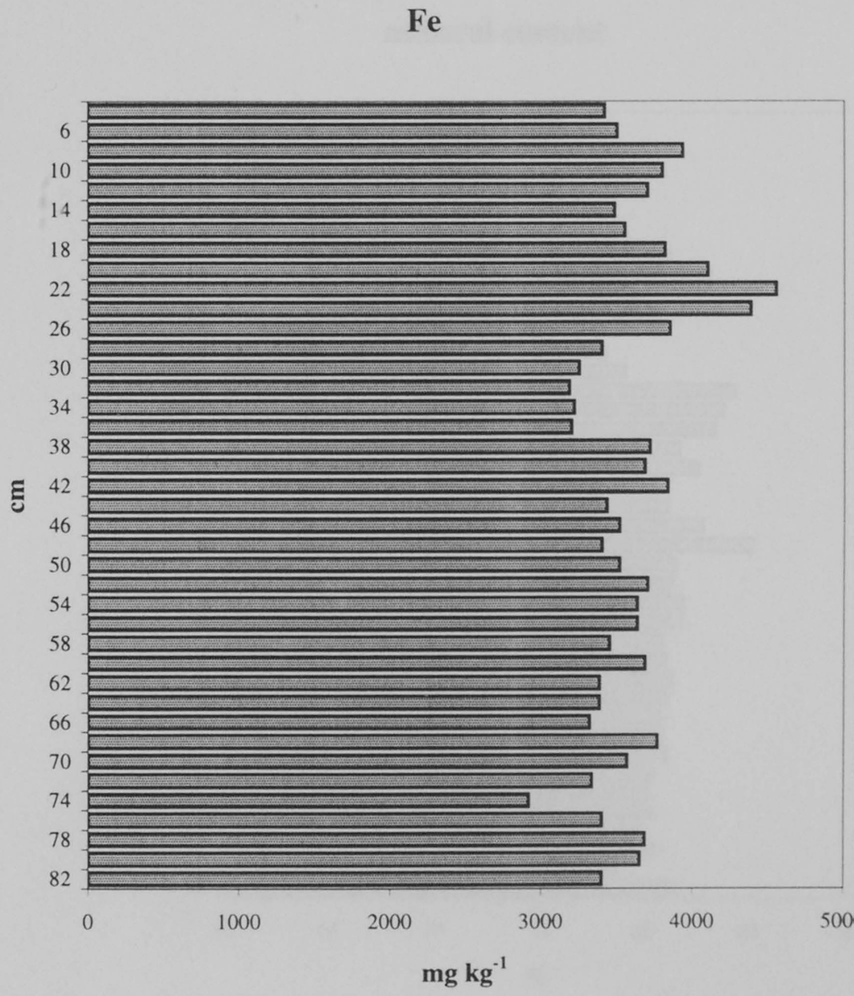


Figure 5.23 Depth versus iron concentration.

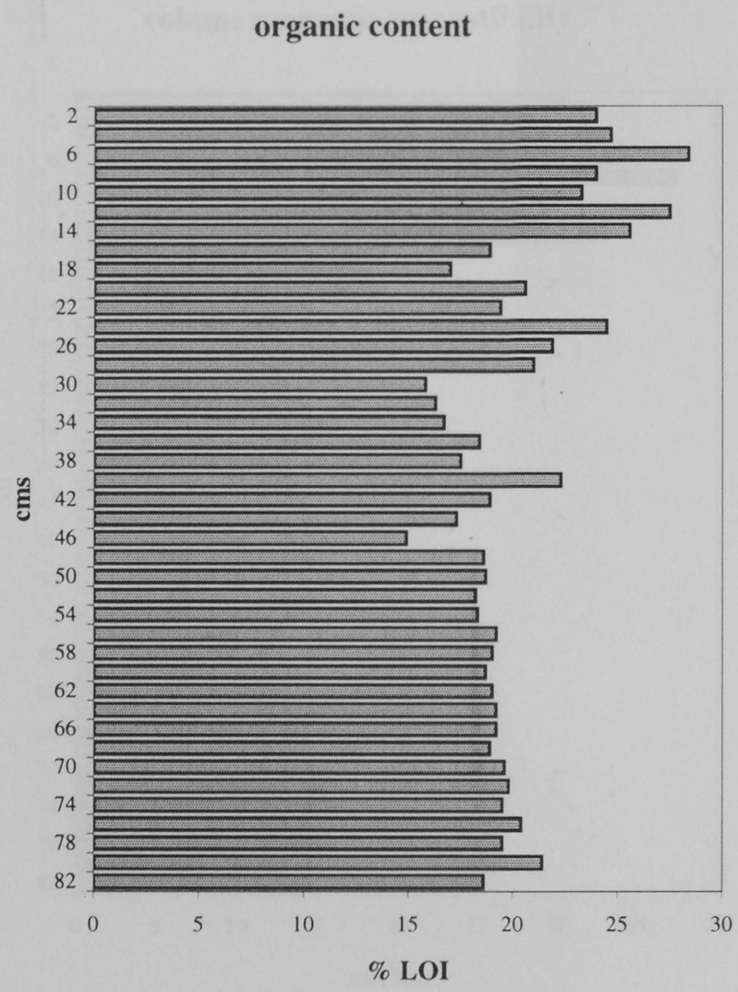


Figure 5.24 Depth versus organic content (350 C).

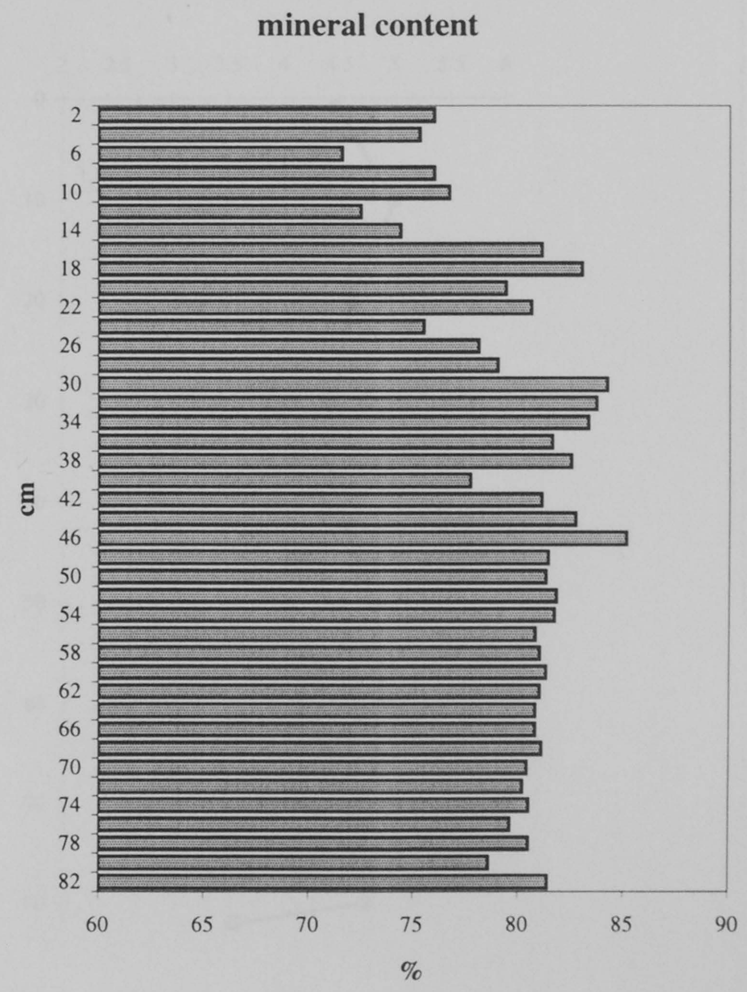


Figure 5.25 Depth versus mineral content.

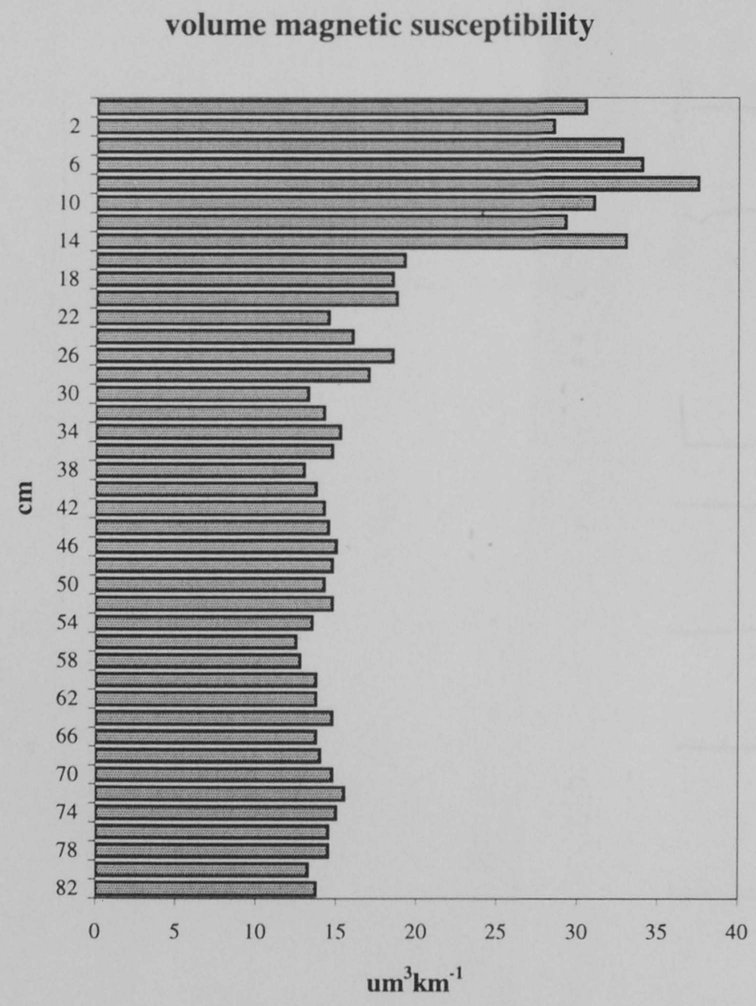


Figure 5.26 Depth versus volume magnetic susceptibility.

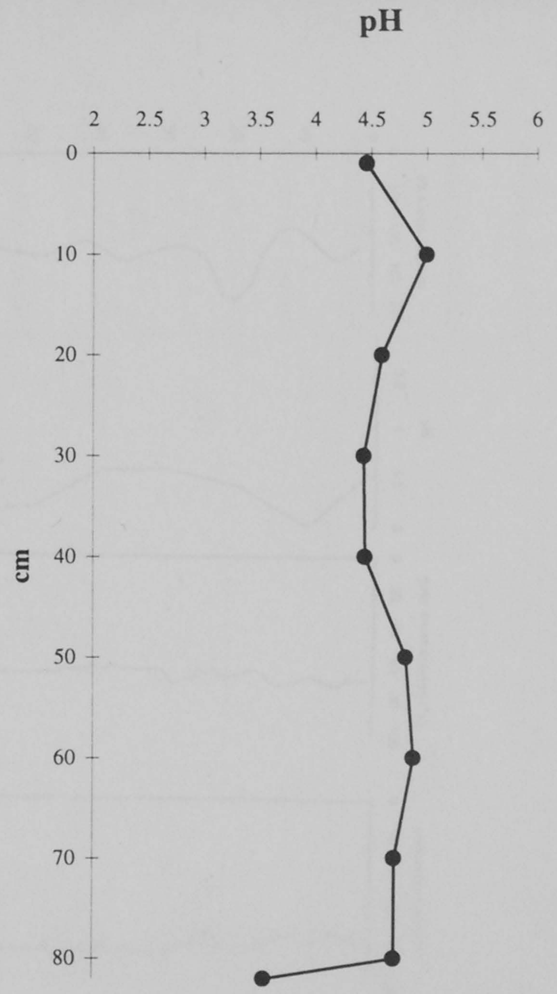


Figure 5.27 Depth versus pH concentration.

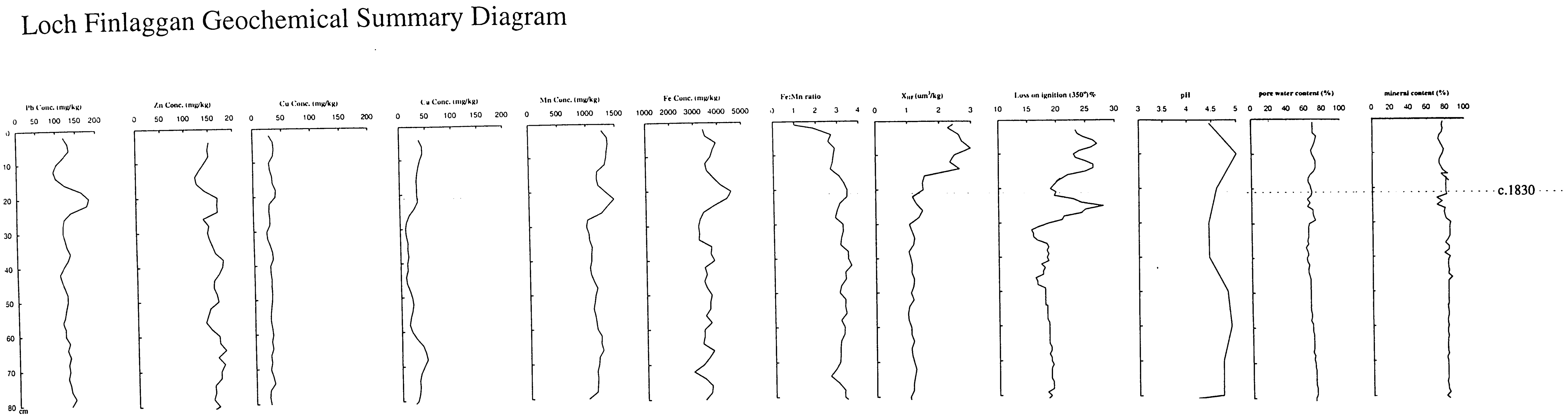


Figure 5.28 Loch Finlaggan geochemical summary diagram.

Iron (Figure 5.22)

The concentration profile for Fe follows the same trend as Pb and Mn where variance is limited until the rise in concentration at between 20-22 cm. At the base of the profile the Fe concentration is established at 3500 mg kg^{-1}. This remains constant until 26 cm where there is a rise to 4500 mg kg^{-1}at 20 cm. A noticeable decrease occurs at 12 cm with 3500 mg kg^{-1} with a slight rise thereafter before a near surface value of 3500 mg kg^{-1} is attained.

Manganese (Figure 5.23)

Manganese shows limited variability from the base (1000 mg kg^{-1}) until 22 cm where an increase occurs with 1480 mg kg^{-1}. From this position there is a decrease then a slight rise with a surface concentration of 1400 mg kg^{-1}.

Iron:Manganese ratio (Figure 5.28)

The Fe:Mn ratio curve shows limited variability from the base of the profile at 80 cm to 5cm depth, where the ratio ranges around 3. At 5 cm there is a noticeable decrease to 1.

Organic matter based on loss on ignition (LOI) (Figure 5.24)

Percentage organic matter based on LOI shows values in the order of 20% with a slight increase to 27% at 6 cm. These values are more or less in keeping with Loch Leathann (mean organic content = *c*.25%) and reflect a low input of allogenic organic matter entering the sediment.

Mineral content based on the inverse of the LOI profile (Figure 5.25)

The inverse of the data obtained from LOI provides a rough estimate of the percentage mineral content within the sedimentary profile. The results show that sediment includes a *c*.80-84 % mineral component which has intermittently decreasing values towards the surface of the core. At this position values range between 74- 82 %.

Volume magnetic susceptibility (Figure 5.26)

From the base of the core to 20 cm there is limited variability in volume susceptibility with values ranging from 1.04 -1.2 μm^3 kg^{-1}. However, from 18 cm there is a sharp increase at 10 cm where volume susceptibility increases to a maximum of 3.04 μm^3 kg^{-1}. From 10 cm there is a decline in values to 2.24 μm^3 kg^{-1} at the surface.

pH (Figure 5.27)

At 82 cm there is a sharp increase in pH from 3.5 to 4.5 pH units. From 80 cm upwards to 10 cm where a maximum of 5 pH units are attained, the values range from between 4.3- 4.4 pH units. From 10 to the surface there is a decrease to 4.4 pH units.

5.5.8 Implications of the geochemical inventory

The geochemical summary diagram (Figure 2.8) shows that there is a correlation between the peak in Pb and the curves of Zn, Cu, Ca, Mn and Fe at *c.*20 cm depth. These peaks are approximately coincident with the start of the increase in both magnetic susceptibility and pH. Prior to this concerted peak, the metal curves for Pb, Zn, Mn and magnetic susceptibility show limited variability with slight fluctuations until *c.*20 cm. Organic and mineral content are similar to the data obtained for Loch Leathann (see Chapter 8 Figure 8.20) where the organic content is similar (mean 25%) with *c.*75% mineral component. The proportionally higher mineral content is perhaps due to erosion input from farming practices within the near vicinity of the loch (see Section 5.2.3 and Figure 5.2 above).

The volume magnetic susceptibility profile is very interesting as it shows a rapid increase in values towards the upper-region of the core. The straightness of the curve before this increase would suggest relatively stable inputs in magnetic mineral constituents prior to a large input of sediment with a higher concentration of magnetic material. The rise in magnetic mineral is most probably the combined result of lead mining and the intensification of agriculture within the catchment. It has already been mentioned that recent field survey has shown the extent of the remains of rig and furrow. It is interesting that there is a sub-surface (2cm) decline in heavy metal concentration and in the physical properties. This is noticeable in the Cu, Ca Fe:Mn ratio and magnetic susceptibility alongside the decrease in pH. There may have been a slight trend towards acidification from *c.*10cm towards the surface but this cannot be supported without examination of the diatom record between these sediment sections. The bimodal nature of the Ca profile does not conform with the other metal profiles. The input of allochthonous material could account for this but arguably were this to be the case one might have expected an additional rises in Fe, Mn and possibly magnetic susceptibility.

Partial sediment disturbance between 1-15 cm has seriously constrained any attempt to establish an annual rate of sedimentation based on the ^{210}Pb inventory. Paradoxically the ^{137}Cs decay profile appears not to have been affected by sediment mixing or by biological disturbance. The usual convention in lake-sediment investigations is to use the ^{137}Cs weapons testing maximum deposition of 1963 as a supporting chronozone for ^{210}Pb derived annual rates of sedimentation. In the absence of a reliable ^{210}Pb decay profile the ^{137}Cs profile is used here to establish a tentative sedimentation rate of 1.3 mm yr^{-1}. This is based on the subsidiary peak at 4 cm which is suggested as being the weapons testing maximum of 1963 (compare this with the ^{137}Cs decay profile for Loch Bharradail Figure 6.10). Four cm of sediment has been deposited from 1963-1992. An approximate sediment rate of 1.3 mm yr^{1} has been calculated using 40 mm/29 yr^{-1} =1.3 mm yr^{-1}. This provides an approximate date for the Pb and Cu maximum at 20 cm as *c.*1835. This date is later than the historical accounts of 1770 for the Portnealon leadmine. It is therefore concluded that the concerted rise in metals could be attributed to the period of maximum intensification in the agricultural base surrounding the loch which is coincident with the maximum peak in population in 1830 based on census records (Storie 1983, 510). The Pb, Zn and Cu profiles from Finlaggan Marsh Core 2 show a similar rise towards the surface of the core. It must be emphasised that the suggested date of *c.*1835 can only be tentative owing to the lack of primary ^{210}Pb data.

Sediment samples were not submitted for radiocarbon determination owing to the presence of limestone within the catchment (see discussion in Chapter 6 section 6.10). Additional chronological support by pollen analyses was also undertaken to assess the influx of *Pinus* pollen resulting from estate plantations established in the early 19th century. Unfortunately the results from this aspect of investigation were negative and therefore of limited use as the % frequency is low (below 2%) in the uppermost sediment sections. Accepting that the integrity of the chemical influx may to some extent have been affected by partial near-surface mixing and in the absence of a rigid chronology then only qualitative assumptions can be made regarding the timing of events that have occurred in the geochemical profile. Firstly, there is close conformity between the peaks in Pb, Zn Cu, Ca, Mn and Fe at between *c.*20-24 cm depth. This concerted rise occurs well below the redox boundary at the sediment water interface (*c.*0-1 cm) and therefore the scavenging effects of Mn can be ruled out. Upward migration of soluble Mn under reducing conditions in sediment pore waters leading to surface precipitation by oxidation is well known and has been described in detail by Mackereth (1966); Weiler (1973) and Forstner (1977).

We know that lead mining has been extensive in the mid 18th century close to the shores of Loch Finlaggan. Three drainage adits have been identified and have been shown to be the principal route of lead-rich solutes into the loch. The rise in Pb, Zn and Cu may well be due to the mining events alongside the intensification of land-use within the catchment. Intensification in the latter half of the 18th to mid-19th century has promoted a rise in Fe and Mn due to an increase in soil-surface run-off. Similarly there has been a response in organic matter which may also corroborate with this to some extent. Deeper fluctuations in elemental concentration may represent variable sediment influx. High sedimentation rates and small elemental concentrations can produce an irregular profile due to non-uniform deposition of anthropogenic and natural materials (Kemp *et al* 1975, 449). The Fe:Mn is in the order of 3 mg kg^{-1} which is higher than the Fe:Mn ratio recorded in Lochs Leathann (1.6 max.) and Lossit (1.3 max.).

Mackereth (1966) recognised that the supply of these elements to the lake sediments could be controlled by changes in soil composition, and he hypothesised that the differential mobility of Fe and Mn could be attributed to alteration of the lithology within the catchment. This would include natural weathering processes and leaching caused by acidification in near surface horizons.

5.5.9 Conclusions

Owing to sediment mixing in the upper region of the core a reliable sedimentation rate based on the ^{210}Pb excess inventory could not be established. However a tentative date of *c.*1835 based on linear extrapolation of 1.3 mm yr^{1} calculated from the ^{137}Cs profile might be credible to explain the concerted peaks in Pb, Zn and Fe and magnetic susceptibility at 20 cm depth. It is at around this period (1831 based on census records) that Islay's population was at its highest and the agricultural base had to intensify to cope with this. There is a wealth of documentary evidence describing new improvements such as lime burning, enclosure, reclamation of land from the sea and more importantly the cultivation of land previously considered marginal, the so-called moss-lands (Storie 1983, 560). It is argued that the density of run-rig remains in the catchment of Loch Finlaggan must surely be attributed to this period. They are not earlier than the mining remains. However this can only be said for rig and furrow in the areas that have been mined. It is probably impossible to attribute dates to rig and furrow away from the mines without other archaeological evidence.

We have seen that the remains at the former lead workings are extensive. At Portnealon this has resulted in anomalous surface-soil enrichment in Pb, Zn and Cu. Paradoxically, the late 18th century period of lead mining appears not to have led to a major increase in either Pb, Zn or Cu, within the loch sediment. The peak in Pb, Zn and Cu at 20 cm could be the result of both mining and farming. It will be recalled that lead mining and farming were carried out alongside each other in the form of a dual economy and indeed we may well be observing the combined effect of these activities.

5.6 Geochemical Investigations at Finlaggan Marsh

5.6.1 Introduction

Given the close proximity of lead mining evidence at Portnealon, Robolls mines and the West Shore site, it was assumed that a 'chemical response' from early mining activity might be recorded in biostratigraphy within the marsh. Moreover, the National Museums of Scotland excavating on the island of *Eilean Mor* were also keen to link the vegetation history of the marsh with on-site environmental strategies. From a geomorphological standpoint, the marsh would provided useful information for understanding the wider aspects of palaeoenvironmental change close to the island. This has been investigated using stratigraphical analyses of 32 cores taken from two transects across the marsh (Figure 5.29). The methods used in this procedure are described above in Section 3.5.4.

Previous exploratory work (G. Coles pers comm.) showed that at the 'nose' of the delta, sediment extended down to a depth of 4 m. A pilot survey using Russian and Dutch corers showed the presence of fen-peat and minerogenic bands. The construction of a relative timeframe based on biostratigraphy was possible owing to the anaerobic nature of the sediment. A palynological time-frame could be used as a relative dating method for linking lithological, chemical and palynological changes within the profile to the local and regional palaeoecological record. Events recorded in profiles could also be related to human activities closer to the site, such as deforestation, agricultural intensification (*c.*1830s), recent afforestation (1982) and more importantly 18th century leadmining. It was also estimated that the deeper sediment sections ought to provide a lower 'background' heavy metal inventory in contrast to the near-surface sediment where the heavy metal burden might be expected to be high owing to the rise in pollution over the last 150 years (Lee and Tallis 1973, Livett, *et al* 1979, Van Geel 1989).

5.6.2 Location of Finlaggan Marsh

The marsh is approximately triangular in shape and 65 m in length and over 45 m at its widest point and is situated on the north-east side of *Eilean Mor* (Plate 10, Appendix 1 and Figure 2.59). The marsh has developed on a slate and phyllite spur between *Eilean Mor* and the shore of the loch. The site has formed as a result of hydroseral succession in the sheltered lee of *Eilean Mor* in conjunction with increased sediment loading resulting from high drainage basin relief. The surface vegetation of the marsh comprises a mosaic of emergent aquatic species including *Nymphaea alba* and *Equisetum fluvitile* forming an outer fringe whilst stands of *Phragmites communis* dominate the shallower interior.

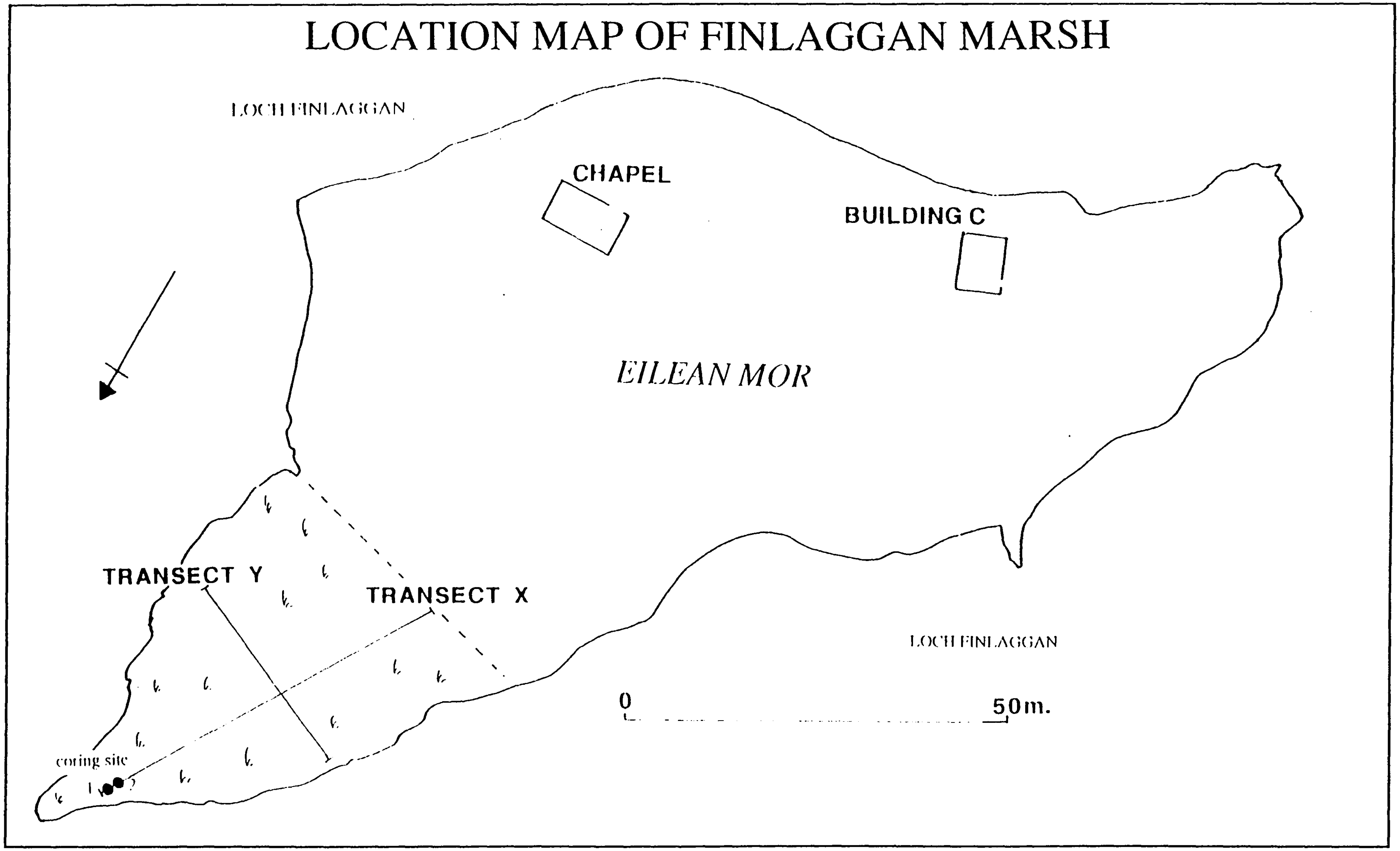


Figure 5.29 Location plan of Finlaggan Marsh and the X and Y coring transects.

STRATIGRAPHICAL PROFILE OF FINLAGGAN MARSH (TRANSECT X)

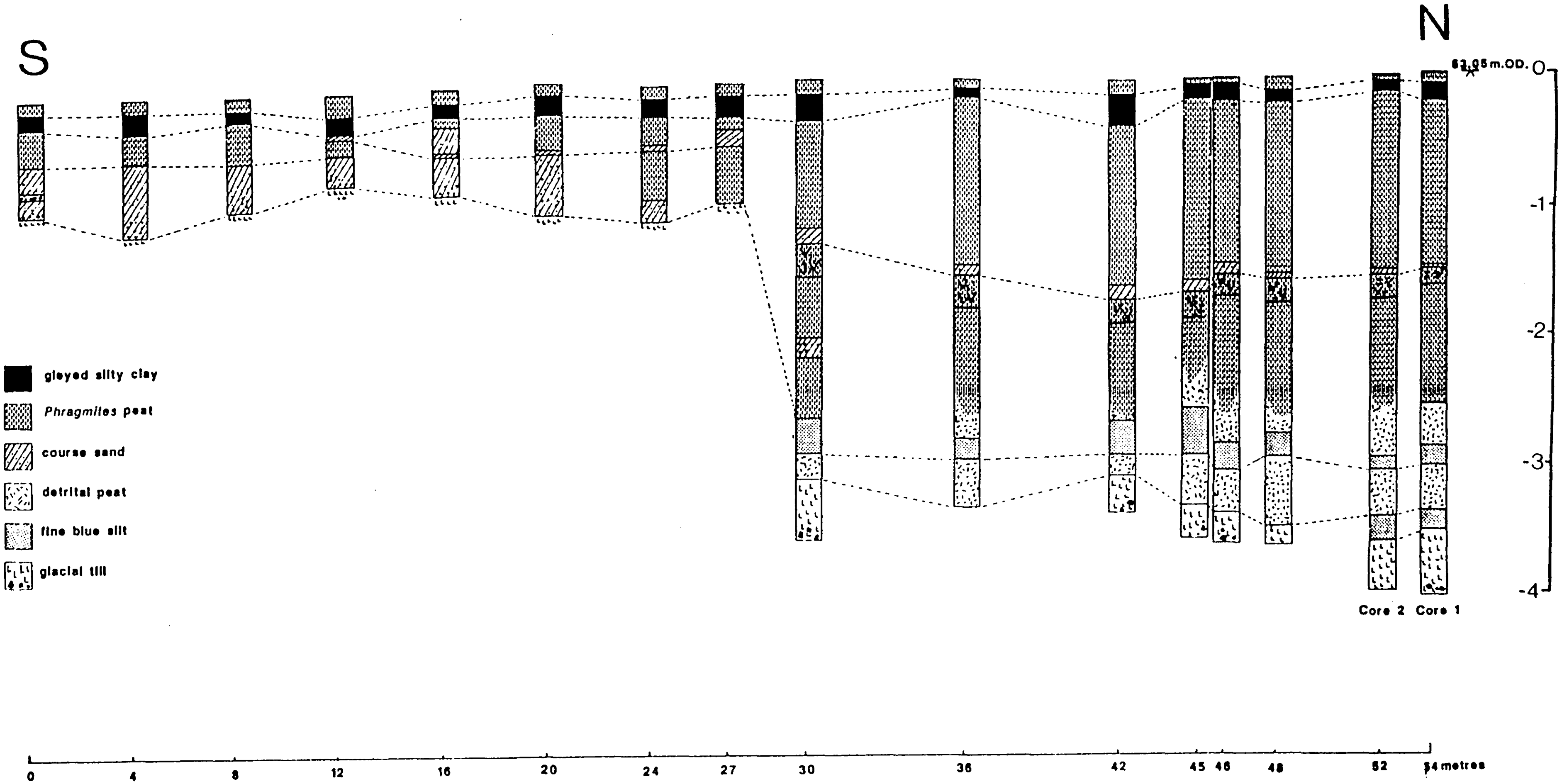


Figure 5.30 Biostratigraphic profile of Finlaggan Marsh (Transect X) and the position of Cores 1 and 2.

Towards the centre of the marsh the vegetation increases in density with *Galium palustre*, *Sagittaria sagittifolia* and *Valeriana dioica*. The marsh is seasonally inundated as the loch reaches its mean winter water-level which varies considerably according to winter rainfall patterns. In 1982 a large proportion of the loch's catchment was afforested (45% Table 5.1). Single throw downhill tine-ploughing (Binns 1978) at right-angles to the Finlaggan burn led to a short-lived but relatively high input of mineral matter in the form of fine silts. These are evident within the upper part of the stratigraphical sequence. (see Unit 2 in Table 5.4 below).

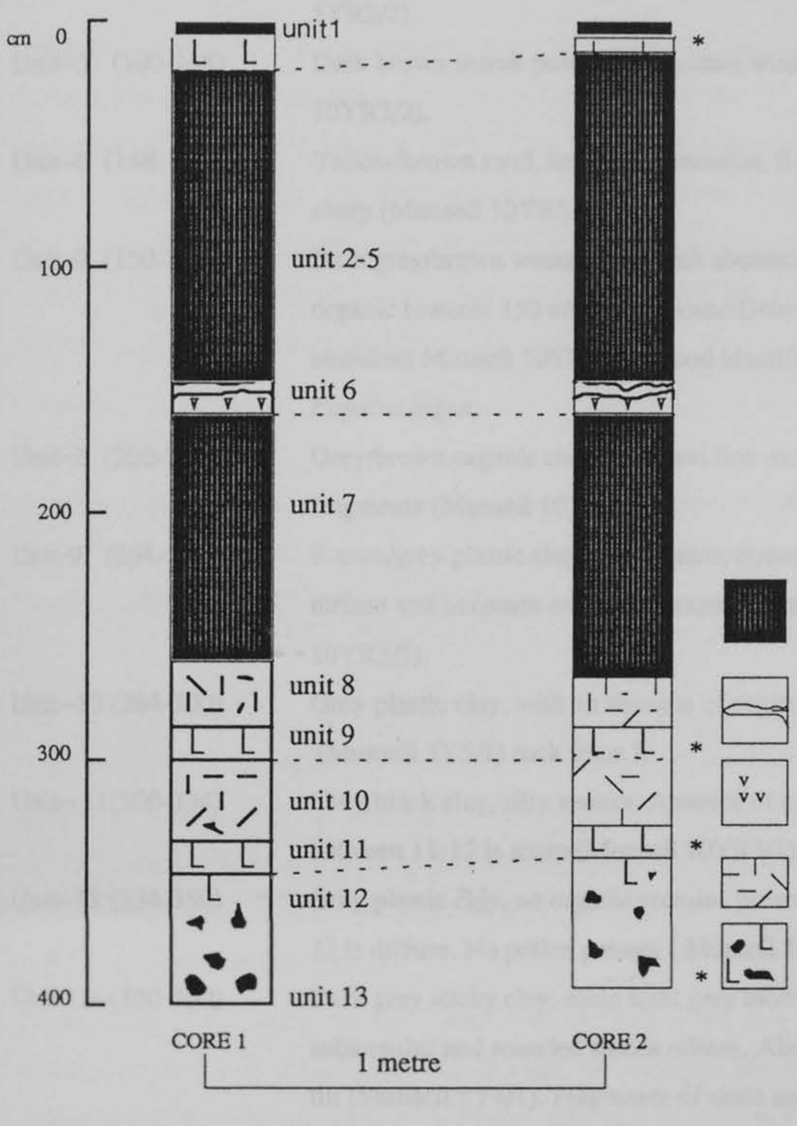


Figure 5.31 Biostratigraphic characteristics of Core 1 and 2 (*) denotes XRD sample position.

Unit	Depth	Description of Core 1
Unit~1	(0-9)	Brown/olive marsh peat, roots abundant with woody fragments, becoming less humified towards the surface (Munsell 10YR4/2).
Unit~2	(9-24)	Grey/brown silty clay, absence of organic remains. Boundary at 24 cm is very sharp. Inwash material (Munsell 5YR5/2).
Unit~3	(24-38)	Grey/brown marsh peat, rich in leaf and stem fragments, roots abundant (Munsell 7YR3/2).
Unit~4	(38-100)	Olive grey marsh peat with leaf and stem fragments <3 mm (Munsell 5YR3/2).
Unit~5	(100-148)	Dark brown marsh peat with abundant woody fragments (Munsell 10YR3/2).
Unit~6	(148-150)	Yellow/brown sand, no organic remains. Boundary at 150 cm is very sharp (Munsell 10YR5/4).
Unit~7	(150-254)	Dark grey/brown woody-peat, with abundant roots, becoming noticeably organic towards 150 cm. *Cyperaceae/Gramineae* stems and rootlets abundant Munsell 10YR3/2). Wood identified as *Corylus avellana* and *Populus nigra.*
Unit~8	(200-254)	Grey/brown organic clay, abundant fine roots with occasional leaf fragments (Munsell 10YR3/3).
Unit-9	(254-284)	Brown/grey plastic clay, few rootlets, boundary between units 9-10 is diffuse and becomes coarser in texture towards 148 cm (Munsell 10YR3/3).
Unit~10	(284-300)	Grey plastic clay, with an absence of organic remains and pollen (Munsell 5Y5/1) rock flour ?.
Unit~ 11	(300-334)	Grey/black clay, silty texture. Absence of organic remains. Boundary between 11-12 is sharp (Munsell 10YR3/1).
Unit~12	(334-350)	Grey plastic clay, no organic remains present. The boundary between 12-13 is diffuse. No pollen present (Munsell 10YR5/1) rock flour ?.
Unit 13~	(350-400)	Dark grey sticky clay, some light grey mottles, occasional gravels of subangular and rounded stones <4mm. Absence of organic matter. Glacial till (Munsell 5Y4/1). Fragments of shale and phyllite (200-300 mm).

Table 5.6 Lithostratigraphic descriptions of the Core 1 profile from Finlaggan Marsh

5.6.3 Stratigraphic correlation of Cores 1 and 2

The biostratigraphic characteristics of Cores 1 and 2 are described in Figure 5.31 above. The lihostratigraphic units 1-13 are described in Table 5.4 above. Stratigraphic changes within both cores allow corresponding horizons to be perfectly matched. Down-core changes within cores 1 and 2 were correlated by visibly matching the corresponding horizons and by using the relative changes in volume magnetic susceptibility. The results of this particular analyses are now described.

5.6.4 Summary description of the volume magnetic susceptibility within Cores 1 & 2

Magnetic susceptibility has allowed a higher degree of objectivity and supports the visual correlation using the stratigraphic units within Cores 1 and 2. The equipment used has been described in Chapter 3 Section 3.6.4. The results of the core correlation using down-core concentration in volume magnetic susceptibility (hereafter VMS) are shown in Figures 5.40 and 5.41. The minerogenic sections provide a higher concentration in magnetic susceptibility which is in sharp contrast to the organic sections within the profile of the peat sections that show a background susceptibility of 0.1 $\mu m^3 kg^{-1}$.

Core 1 (Figure 5.40)

Against a background of 0.1 $\mu m^3 kg^{-1}$ Core 1 has a basal magnetic susceptibility of 1.2 $\mu m^3 kg^{-1}$. Two smaller peaks are coincident with the two minerogenic bands at 300 cm and 350 cm depth (Units 10 and 12 respectively). A subsurface peak of 0.5 $\mu m^3 kg^{-1}$ is related to Unit 2.

Core 2 (Figure 5.41)

The background susceptibility is the same as Core 1. Susceptibility at the base of this core is similar to Core 1 with 1.1 $\mu m^3 kg^{-1}$. Two peaks occur before the 350 cm depth peak of 0.5 $\mu m^3 kg^{-1}$. Again there is a peak at 300 cm depth and at the sub-surface with 0.4$\mu m^3 kg^1$.

5.6.5 Summary of the results

Down-core VMS within Cores 1 and 2 shows that there is a good correlation between the mineral horizons to support the visual links between the lower lithostratigraphc units. The minerogenic horizons appear to be relatively rich in ferromagnetic minerals. It has been recognised elsewhere that Late-glacial sediments are high in magnetic minerals (Thompson and Oldfield 1986) which reflect and integrate the primary magnetic mineralogy of the freshly exposed parent material surrounding the lake basin at the close of the last glaciation.

The horizons consisting of mainly organic detritus and woody fragments contain virtually no ferro-magnetic particulate material. In the lower mineral regions at between 200-400 cm depth it is suggested that the VMS has been 'enhanced' by sulphide rich minerals (Figure 5.42). Here there is a rapid decline from 4.5 to below 2.5 pH units. There is an isolated rise in pH (3.50) which is coincident with 350 cm depth and forms the base of Unit 11 that comprises a very fine blue silt (see Table 5.4 and Figure 5.31).

Sulphide producing mechanisms such as the anaerobic decomposer bacteria *Desulphovibrio* promote reducing conditions which utilise oxygen in a combined form with sulphur and sulphate ions. This leads to the generation H_2S which can dramatically reduce the pH (Moore *et al* 1974, 99-100). Under the prevailing reducing conditions within the upper sections of the sediment, both the secondary and primary ferromagnetic minerals (i.e. pyrites, iron oxides and detrital magnetite formed either *in-situ* or derived from surrounding catchment soils) are readily mobilised at the redox interface and can remain in solution as ferrous hydroxide in the form of geothite. Although potentially low in magnetic susceptibility (Thompson and Oldfield 1986,17) hydroxide complexion on to minerals as surface coatings is highly likely in oxygen deprived environments. The density of these mineral components in Units 12-13 clearly provides a 'magnetically enhanced' environment in contrast to the overlying paramagnetic marsh peat.

5.6.6 X-ray diffraction analyses on Lithostratigraphic Units 2, 9, 12 and 13 (Core 2)

X-ray diffraction was undertaken to determine the major mineral phases within samples taken from Units 2, 9, 12 and 13 respectively. The relative abundance of the major mineral phases are established as (*trace* 1-5%), (✓ 5-25%), (✓✓ 25-50%) and (✓✓✓ >50%).

Depth	Unit	Muscovite	Na/Feldspar	Chl.	Pyr.	Gyps.	Quartz	Kaol.
14 cm	2	✓	✓✓	*trace*	—	—	✓✓✓	*trace*
280 cm	9	✓✓	✓✓	*trace*	✓✓	✓	✓✓✓	*trace*
350 cm	12	✓✓✓	✓✓	✓	—	—	✓✓✓	*trace*
400 cm	13	✓✓✓	✓✓	✓	✓	✓	✓✓✓	*trace*

Table 5.5 Relative abundance of the major mineral phases within selected samples. (note: Chl./Chlorite, Pyr./Pyrite, Gyps./Gypsum,Quartz and Kaol./Kaolinite). 1% is equivalent to 1000mg kg^{-1}.

5.6.7 Implications of XRD analyses

Muscovite is more abundant within the minerogenic sediment towards the base of the profile. Muscovite (KAl_2 ($AlSi_3O_{10}$)) is a detrital mineral formed from weathered granite and schists and possibly reflects the presence of local weathered epidiorite. The sodium rich Na/feldspars are present within all the samples and a common constituent of silicate minerals. Chlorite is in trace amounts within Units 2 and 9 and slightly more abundant in Unit 12 and 13. Chlorite occurs as an alteration product of ferromagnesian minerals and can also occur in low grade regionally metamorphosed rocks, (Whitten and Brooks 1978, 368). Pyrite (FeS_2) is more abundant in Unit 9 and a lesser component of Unit 13. This is a sulphide mineral occurring as an accessory in igneous rocks and in ore veins. It is also a major component within sulphide-rich anaerobic sediment. Gypsum ($CaSO_42H_20$) is recorded in low amounts in Units 12 and 13. This mineral is termed an 'evaporite' derived as a result of the evaporation of saline water, (Whitten and Brooks 1978, 219). Quartz (SiO_2) is the predominant mineral component present in all four samples and is strongly linked to the quartzite geology adjacent to the site. This material occurs as a common vein-stone in sedimentary rocks and igneous metamorphic rocks and is easily weathered out of cleavage zones (Whitton and Brooks 1978, 375).

The presence of chlorite and pyrite in the lower section of the core is attributed to the base geology (phyllite and shale) and is the causative factor in the generation of the extremely low pH. Pyrite is unlikely to be responsible for the high volume magnetic susceptibility at this level. Thompson (1986, 3-17) notes that pyrite is paramagnetic (i.e. of low magnetic retention) and is a typical authigenic iron sulphide formed in freshwater sediment in the absence of oxygen. In conclusion the minerogenic layers examined by XRD reflect a local catchment-derived source of metamorphic mineralogy. Erosion of limestone facies is likely to be absent owing the highly acidic nature of the basal regions which would have rapidly dissolved any inwashing calcareous mater. Alternatively, this material was never transported to the coring site.

5.6.8 The Geochemical Record at Finlaggan Marsh

5.6.9 Introduction

The physical properties and heavy metal concentrations have been plotted against depth. Figures 5.33-5.41 show the plots of the individual chemical profiles. These are brought together collectively in the geochemical summary diagram Figure 5.42 which enables comparisons across the range of heavy metals and other properties.

The methods and reagents used in this analyses have been described in Chapter 3 Section 3.7.4. The metal concentration values are expressed as mg kg^{-1} dry weight of sample (dw 0.5g^{-1}). Sediment from the surface of the core (1-2 cm) was not sampled for chemical analyses due to the presence of root activity which may have invalidated the results. Close interval sampling was carried out over the top metre of the core (2 cm intervals) and below 1m samples were taken at 10 cm intervals. This procedure concentrates the bulk of the analyses over 2-100 cm depth which will incorporate sediment formed throughout the historic period. Calcium was not analysed owing to the extremely acidic nature of the sediment that would have rendered any results from this element meaningless.

Lead (Figure 5.33)
The Pb concentrations are described from the base upwards. Basal concentration in Pb is 58 mg kg^{-1} rising steadily to 100 mg kg^{-1} at *c*.340 cm. An intermittent peak of 100 mg kg^{-1} is seen at 280 cm. After this depth the values decline erratically to 170 cm with 9 mg kg^{-1}. The values increase to 73 mg kg^{-1} at 96 cm. From this region the values fall sharply to 40 cm were the Pb value is only 5 mg kg^{-1}. From this depth the values rise sharply to a sub surface maximum of 128 mg kg^{-1}.

Zinc (Figure 5.34)
The Zn concentrations are described from the base upwards. At 370 cm a maximum value of 260 mg kg^{-1} is recorded and is well within the minerogenic region of the core. Values decline and rise intermittently to 65 mg kg^{-1} at 290 cm. From this point there is a gradual increase at 240 cm with a Zn concentration of 215 mg kg^{-1}. There is a sharp decline at 190 cm with 30 mg kg^{-1}. A peak is noted higher in the profile at 120 cm with 150 mg kg^{-1}. From this region in the core the values decrease quite sharply to *c*.50 cm where the values remain low until 18 cm. From here the values rise sharply to a sub-surface concentration of 238 mg kg^{-1}.

Copper (Figure 5.35)
The Cu concentrations are described from the base upwards. The concentration values range from 16 to 40 mg kg^{-1} until a maximum peak is recorded at 290 cm where a value of 65 mg kg^{-1} is reached. From this location in the core the values decrease intermittently to 170 m where values remain constant until 120 m with a smaller peak of 35 mg kg^{-1}. The values remain erratic towards the surface where the value remains lower than the base with only 11 mg kg^{-1}.

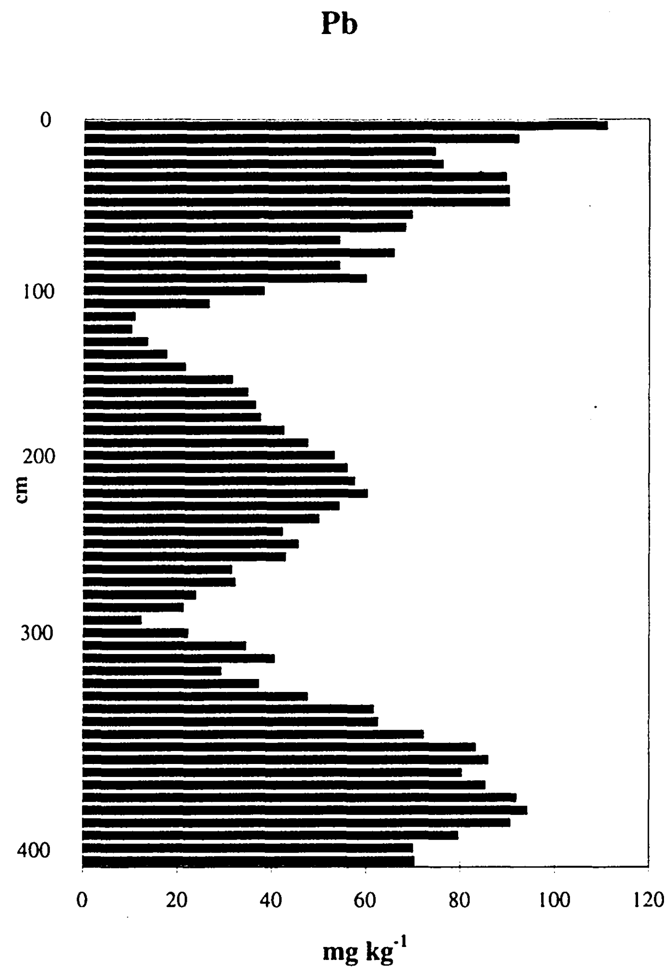


Figure 5.32 Depth versus lead concentration.

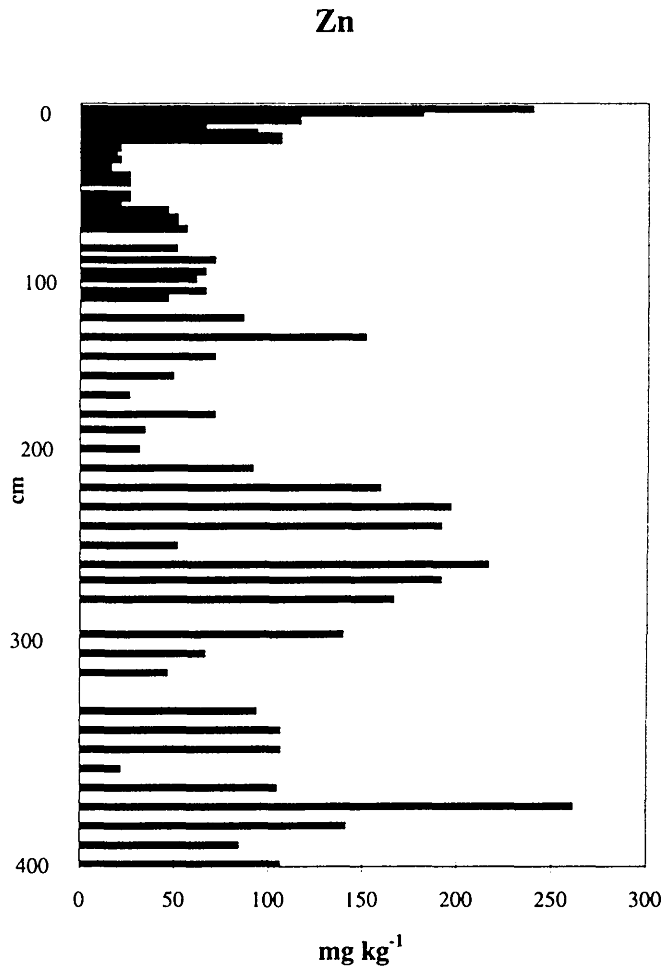


Figure 5.33 Depth versus zinc concentration.

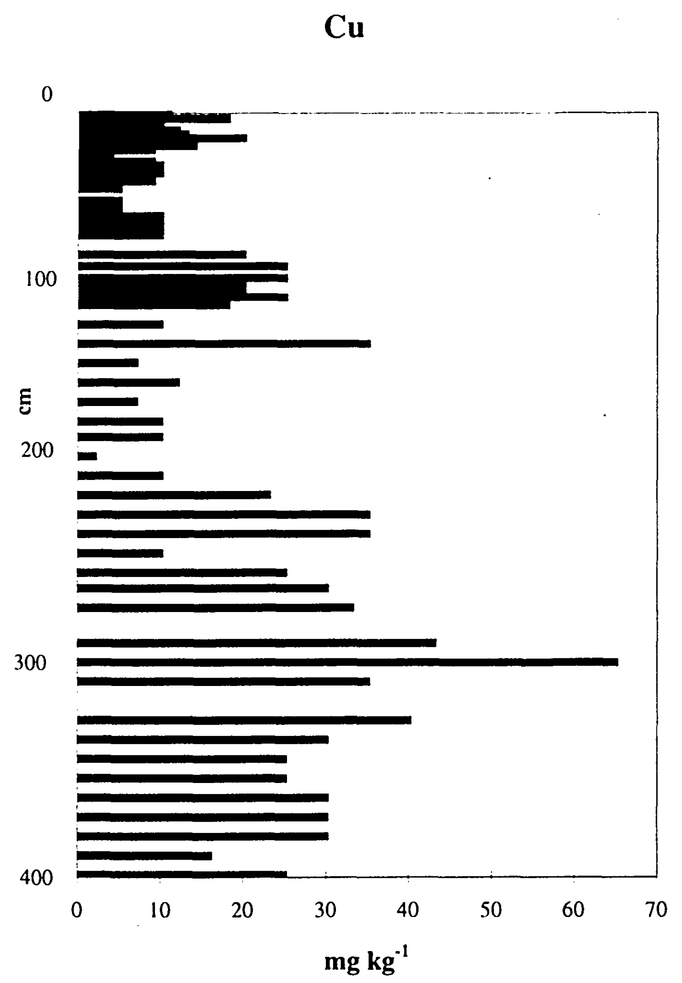


Figure 5.34 Depth versus copper concentration.

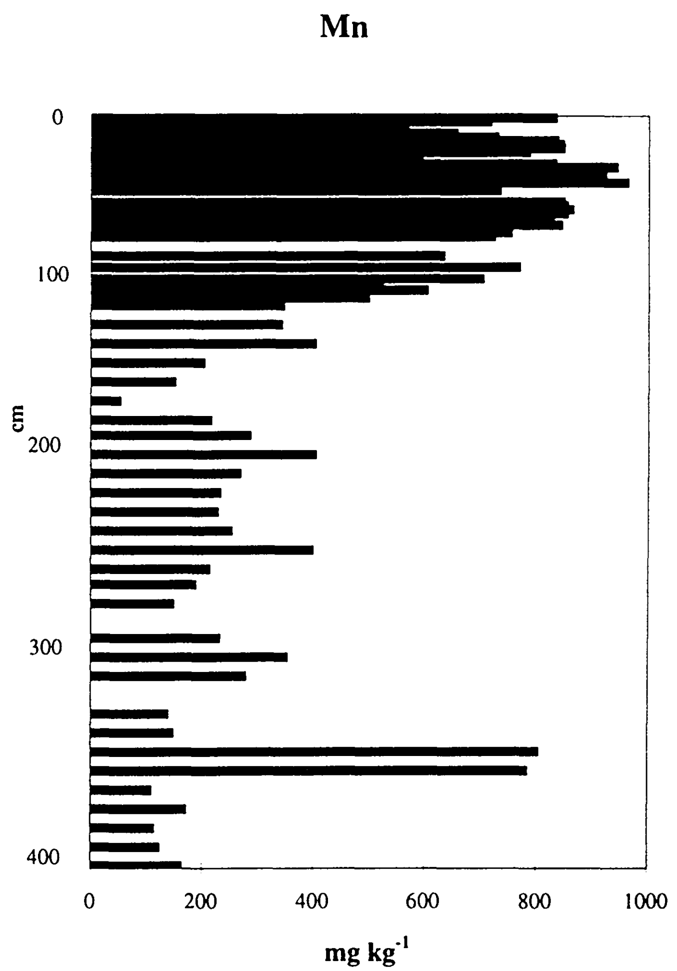


Figure 5.35 Depth versus manganese concentration.

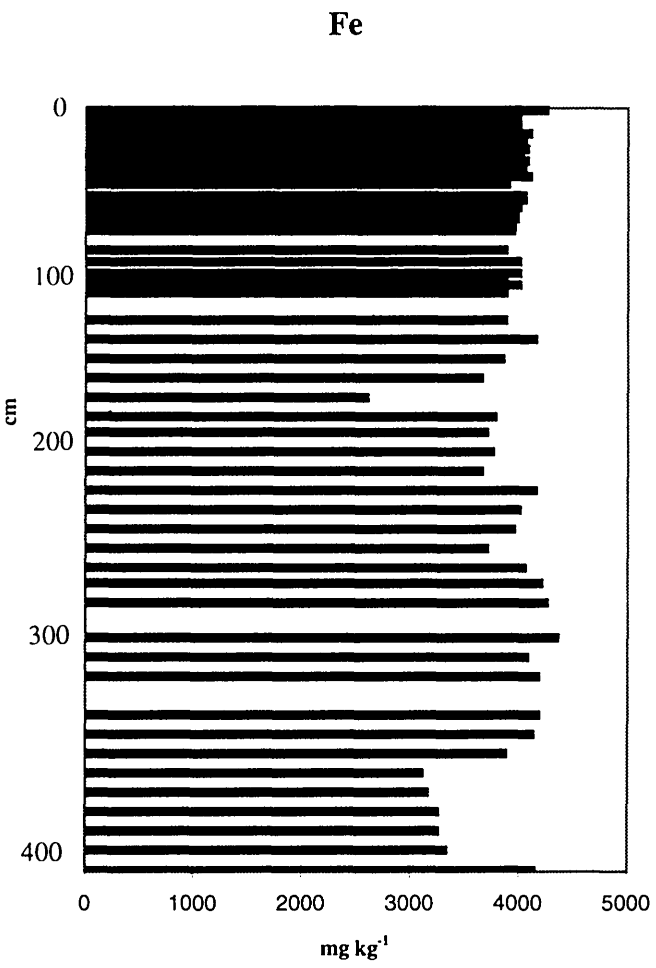


Figure 5.36 Depth versus iron concentration.

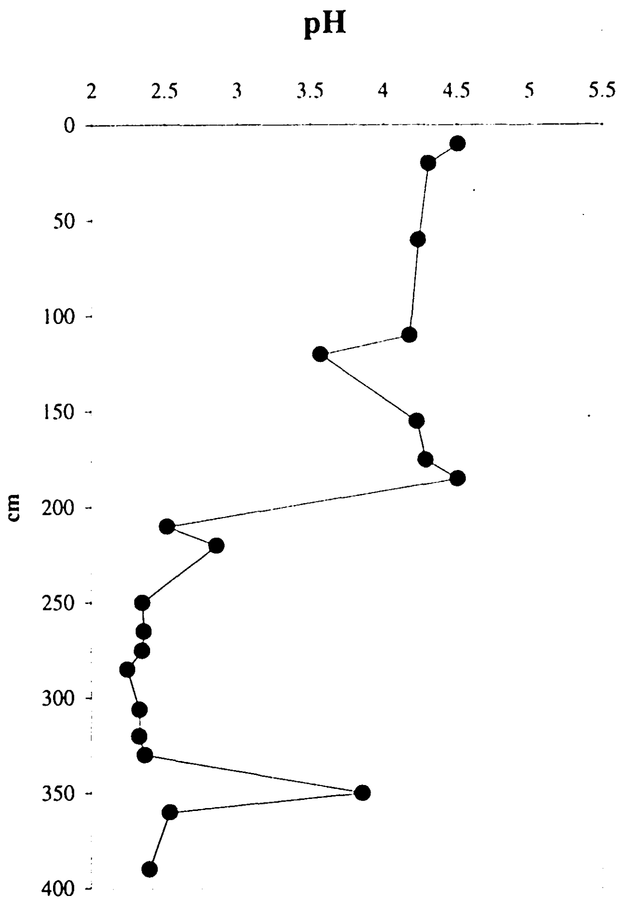


Figure 5.37 Depth versus pH content

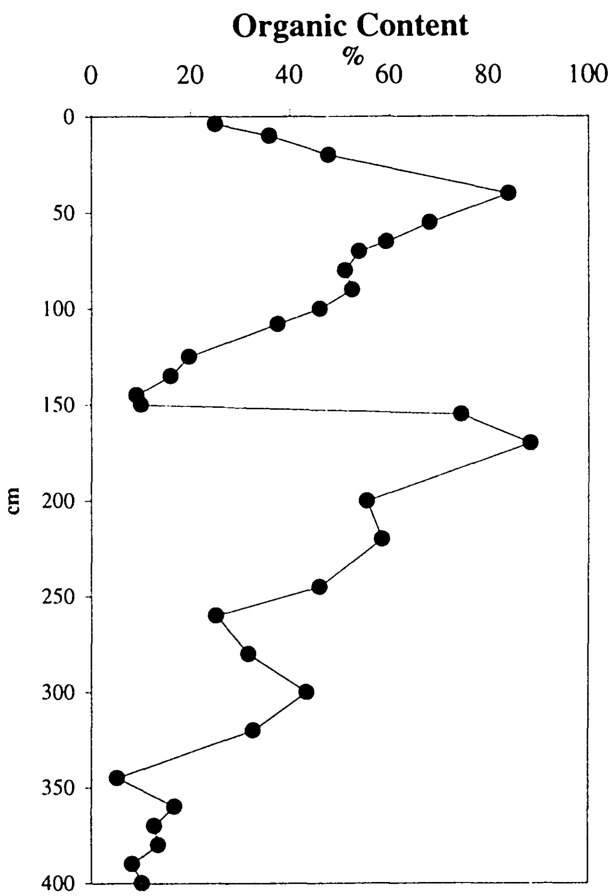


Figure 5.38 Depth versus organic content (350 C)

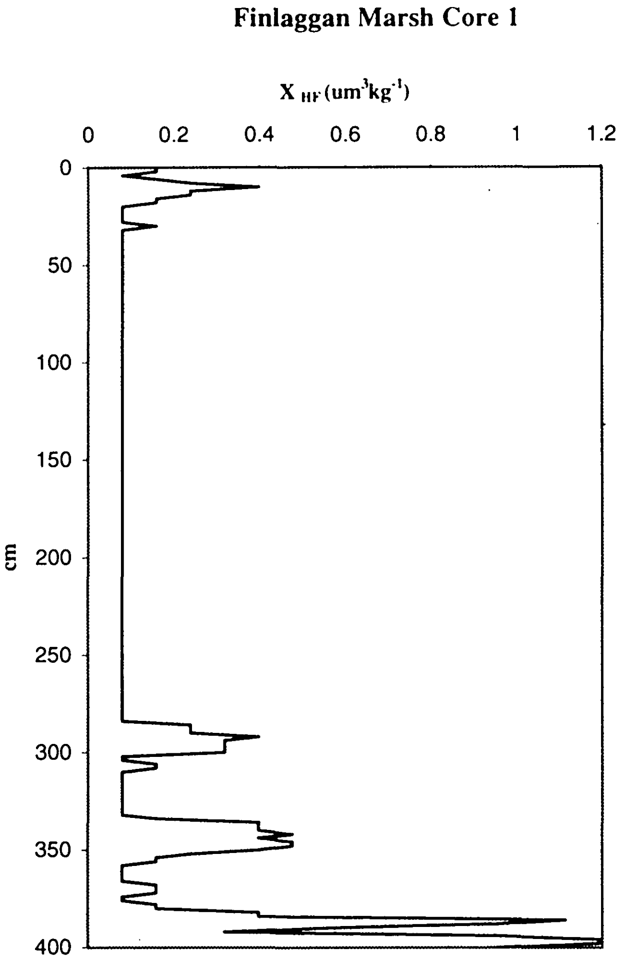


Figure 5.39Depth versus volume magnetic susceptibility.

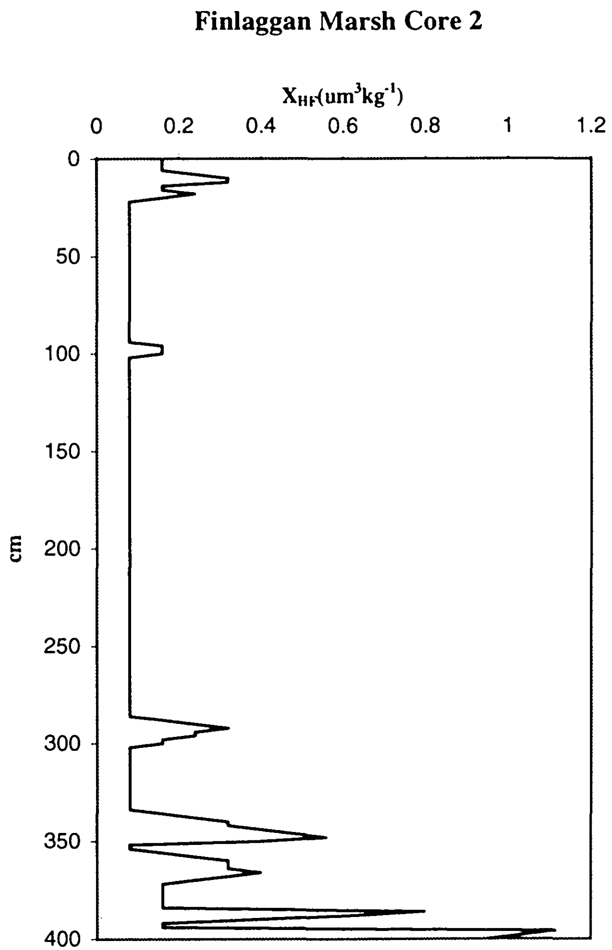
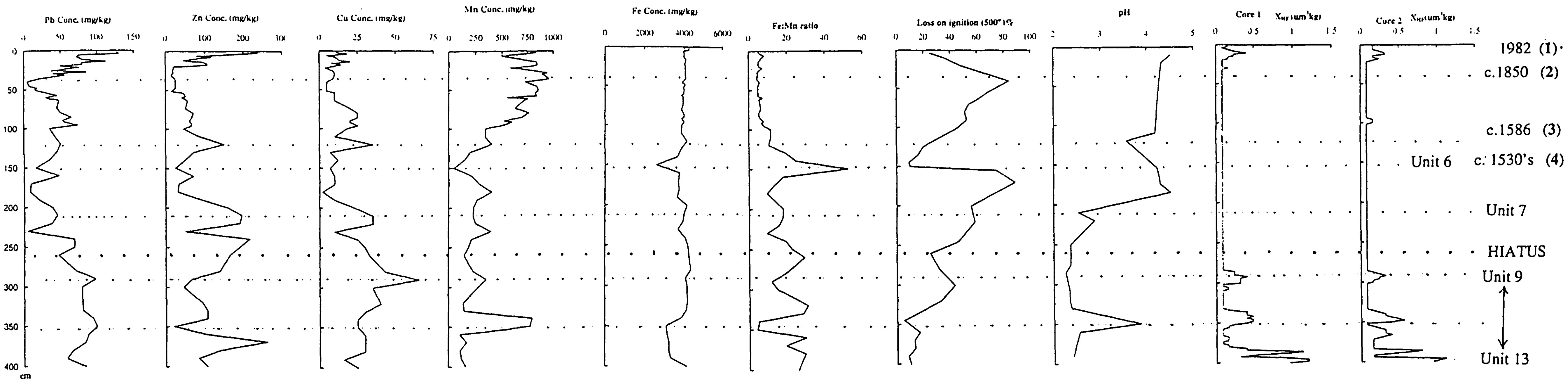


Figure 5.40 Depth versus volume magnetic sisceptibility.

Finlaggan Marsh Geochemical Summary Diagram



Approximate date	Event in the Profile	Interpretation
(1) 1982 (recorded)	Increase in Pb, Zn and Cu at 6 cm	Afforestation of 45% of the catchment
(2) c.1850	Onset of the rise in Pb, Zn and Cu	Atmospheric fall-out deposition
(3) c.1586*	Rise in Pb, Zn Cu Ca Mn	Possible chemical response to lead mining
(4) c.1530*	Decrease in all metals and LOI	Major flood event at Unit 6 (Little Ice Age ?)
Unit 7	Rise in Pb Zn and Cu. Rapid rise in pH	Development of the marsh less acid conditions
Unit 9-13	Peaks in Zn and Mn. High mag/sus. Low pH	Reducing-acid conditions mainly minerogenic inwash

Summary of possible dates and events within the Finlaggan Marsh geochemical diagram. 1850 date based on estimated onset of atmospheric fall-out deposition. (*) based on an extrapolated annual rate of sedimentation rate of 142 mm yr^{-1} using the above atmospheric pollution as a benchmark

Figure 5.41 Finlaggan Marsh geochemical summary diagram.

Manganese (Figure 5.36)

The Mn concentrations are described from the base upwards. Basal values are low with a mean concentration of 132 mg kg^{-1} between 400-360 cm. From this region in the core the values rise sharply to 800 mg kg^{-1} at 340 m. After this rise the values increase and fall intermittently until at about 140 cm where there is an exponential rise to 883 mg kg^{-1} at 14 cm. From here the values rise and fall to a sub-surface peak at 830 mg kg^{-1}.

Iron (Figure 5.37)

The Fe concentration curve shows limited variability. The basal Fe concentration is just over 4000 mg kg^{-1} and falls slightly at 360 cm depth with *c.*3000 mg kg^{-1}. From this position there is a rise to 4000 mg kg^{-1} where this value is maintained throughout the profile.

pH (Figure 5.38)

The pH concentration is lowest at the base of the core (2.5) until 210 cm where there is a rapid increase to 4.2. This remains more or less constant to the surface of the core.

Loss on ignition (Figure 5.39)

The organic content based on LOI shows that between the base of the core to *c.*250 cm the organic content is low (<40%) then rises from this point to over 80% as the marsh develops. There is a rapid decline in organic matter coincident with the sand horizon (Unit 6). The decline in organic material is associated with the minerogenic Unit 1 at the surface of the core.

5.6.10 Implications of the Finlaggan Marsh geochemical inventory

Dealing first with Pb, Zn and Cu, there appears to be some conformity between these three metals as all are seen to peak towards the surface. The concerted rise in these metals occurs from 10-6 cm and may be attributed to afforestation of the catchment (Table 5.1). Deep-rill ploughing preceding afforestation (approximately 45% of the catchment) in 1982 has led to a massive influx of mineral matter across the marsh. At 40 cm there is a protracted increase in Pb, Zn and Cu and this is tentatively attributed to the onset of atmospheric pollution at around 1850 (Chapter 9 Section 9.5.4). At 125 cm there is a concerted peak in Pb, Zn, Cu which may be the result of metalliferous-rich solutes entering the loch. This event is dated to approximately 1586. At 150 cm there is a decline in all metal concentrations due the deposition of white sand. This material (below 10% organic matter based on LOI) has low metal retention capacities hence the extremely low metal influx at this point. The sand horizon is attributed to a major flood event and dated approximately to 1531. At *c.*225-230 cm depth there is peak in Pb, Zn, Cu which may be attributable to redox alterations caused by a rise in pH. The pH curve shows a dramatic rise from 2.6-4.5. The intermittent peaks in Pb, Zn and Cu concentrations towards the base of the core (below 260 cm) are attributed to the influence of minerogenic sediment. The distribution of Zn shows some up-core variation, particularly towards the middle and basal regions. The highest Zn value is attained in the mineral horizon at 360 cm depth which is attributed to the controlling sulphide-rich conditions within this section of the core. As the core becomes more organic towards the surface so the Zn concentration decreases until the surface maximum of 250 mg kg^{-1} is attained.

Copper values throughout the core can be considered low with the peak at 65 mg kg^{-1} (290 m depth) perhaps being slightly anomalous. Copper has a strong affinity with organic matter with which it is strongly bound. The apparent peak at 290 cm is well within the minerogenic section so it seems likely that another form of metal complexion is leading to enrichment at this position. The Fe concentration is almost linear and shows no variability. This is surprising in the light of the fluctuating pH and magnetic susceptibility profiles. The down-core concentrations of Mn show that values are concentrated towards the surface. The peak at 350 cm is attributed to diagenetic alteration within this part of the profile. The gradual rise towards the surface is interesting and perhaps points to a removal of this element upwards towards the redox zone. This phenomenon is well known in lake sediments where controlling redox conditions usually prevail (Kemp *et al* 1976, Davidson *et al* 1985 and Williams 1992).

5.6.11 Summary

Limited geochemical investigations on sediment sections from Finlaggan Marsh establish that Pb, Zn and Cu concentrations can be attributed to specific events occurring in the catchment. Diagenetic control can be observed within the minerogenic sections towards the base of the core. This has given rise to a sustained rise in Pb (400-*c*.290 cm) and a Zn maximum at 365 cm. The exponential rise in Pb within the near surface sediment sections can be related to the onset of atmospheric pollution and recent afforestation (note the spike in VMS at 6-8 cm depth). It has been mentioned that arable farming rose in intensity alongside a rapid population increase in the early 19th century (Storie 1983). The need for increased productivity at this period saw new and marginal areas brought into cultivation. This has undoubtedly led to increased run-off of surface soils resulting in the rise in Pb, Zn and Cu and VMS towards the surface of the profile.

5.7 Palynological Investigations on Finlaggan Marsh Core 1

5.7.1 Introduction

Pollen analysis was undertaken to establish a relative chronology for the chemical profiles using the methods described in Chapter 3 Section 3.9.1. The percentage pollen diagram (Figure 5.43) shows the general trends in the pollen values as a percentage of the sum of the total land pollen (TLP). The diagram has been ordered into a series of five local pollen assemblage zones (LPAZ Fin A-E) based on the criteria of relative changes in pollen values and in the changes in the variations in the pollen spectrum. The local pollen assemblage zones are described from the base to the surface of the profile.

5.7.2 Description of the Local pollen assemblage zones

Lpaz. Fin A

This zone rests below the lowest band of non-polleniferous silt and is characterised by maximum *Pinus* values (42%). *Betula* increases proportionately towards the upper region of the zone. Some *Alnus* grains were present and could be due to contamination at this lower level. *Juniperus* and *Salix* are low in frequency as are the Coryloid group (combined counts of *Corylus avellana* and *Myrica gale*). *Empetrum* is predominant with over 65% showing this plant was established on the nearby porous acidic soils. Open ground indicators are represented by *Ericaceae*, *Gramineae*, *Cyperaceae* and *Rumex*. Disturbed ground indicators are represented by *Plantago lanceolata* with lower frequencies of *Artemisia*.

Lpaz. Fin B

This zone rests between the upper and lower boundary of the non-polleniferous zones and is characterised by decreasing numbers in *Pinus* pollen. A slight increase in *Betula* occurs in this zone and *Quercus* appears for the first time. *Salix* is present but low in frequency. The *Coryloid* group rises substantially to 43% TLP. *Plantago* rises to 10%. *Rumex* is also present along with *Filupendula*. *Cyperaceae* is dominant with submerged aquatics *Isoetes*, *Equisetum* and *Typhacea* which are lower in frequency.

Lpaz. Fin C

This zone is situated between the non-polleniferous zone upper boundary and 2 m depth. The zone is characterised by an increase in arboreal pollen. *Pinus* fluctuates alongside *Betula* which rises proportionately towards the upper middle region of the zone with over 40% to form the maximum concentration throughout the core. Other arboreal groups appear for the first time with *Quercus* and *Ulmus* which remain low (<1%) towards the upper region of this zone and probably represent transportation from further afield. *Salix* is more frequent suggesting colonisation on the margins of the mire. Coryloid types rise sharply at the expense of *Betula* which declines at this point in the profile. The *Gramineae* component rises to 60% but decreases as *Cyperaceae* rises towards the upper region of the zone reflecting mire conditions at the site. As with the previous zone *Phragmites* would have been developing along the flanks of the mire. *Typhacea* was locally present but low in frequency at this time. This zone also sees increasing numbers of herbaceous pollen. *Filupendula*, *Rumex*, *Plantago*, *Artemisia* and *Ericaceae* rise proportionately in this zone.

Lpaz Fin D

This zone extends upwards from 200 cm to 100 cm and is characterised by a significant rise in aquatic species. *Pinus* is present but remains low as *Betula* rises then decreases towards the upper region of the zone. Alder frequencies are gradually sustained but again the numbers are not significant. *Ulmus* is present but very low (<1%) throughout the zone. *Fraxinus* appears for the first time but is low in number. *Salix* is present and *Coryloid*-types make a proportional increase towards the upper region of this zone with over 50%. *Ericaceae* declines dramatically whilst the *Gramineae* and *Cyperaceae* types remain consistent throughout this zone. There is a substantial increase in the number of herbaceous species among which *Rumex* rises to over 20%. This disturbed ground indicator is supported by lesser numbers of *Chenopodiaceae* and *Caryophyllaceae*. *Plantago* and *Artemisia* are also present. *Filicales* and *Pteridium* increase in number alongside *Ranunculus* types that emerge and reflect open areas. Of significance is the exponential increase in the aquatics *Eriocolon* and *Typhaceae*.

Lpaz Fin E

Arboreal pollen remains low with *Pinus* and *Betula* decreasing in number. *Alnus* is present but low in frequency. *Quercus* rises at the base of the zone to just over 10% and declines soon after. A rise occurs near the surface with *Casteneae sativa* (Sweet chestnut) and probably represents pollen transport from one of the estate plantations as does the species *Picea*. *Ericaceae* rises sharply reflecting the acidic conditions we see in the catchment today. Herbaceous flora rise throughout this zone reflecting the predominance of both cultivated apastoral land, with the increase in *Ranunculus*, *Rumex* and *Compositae liguliflorae*.

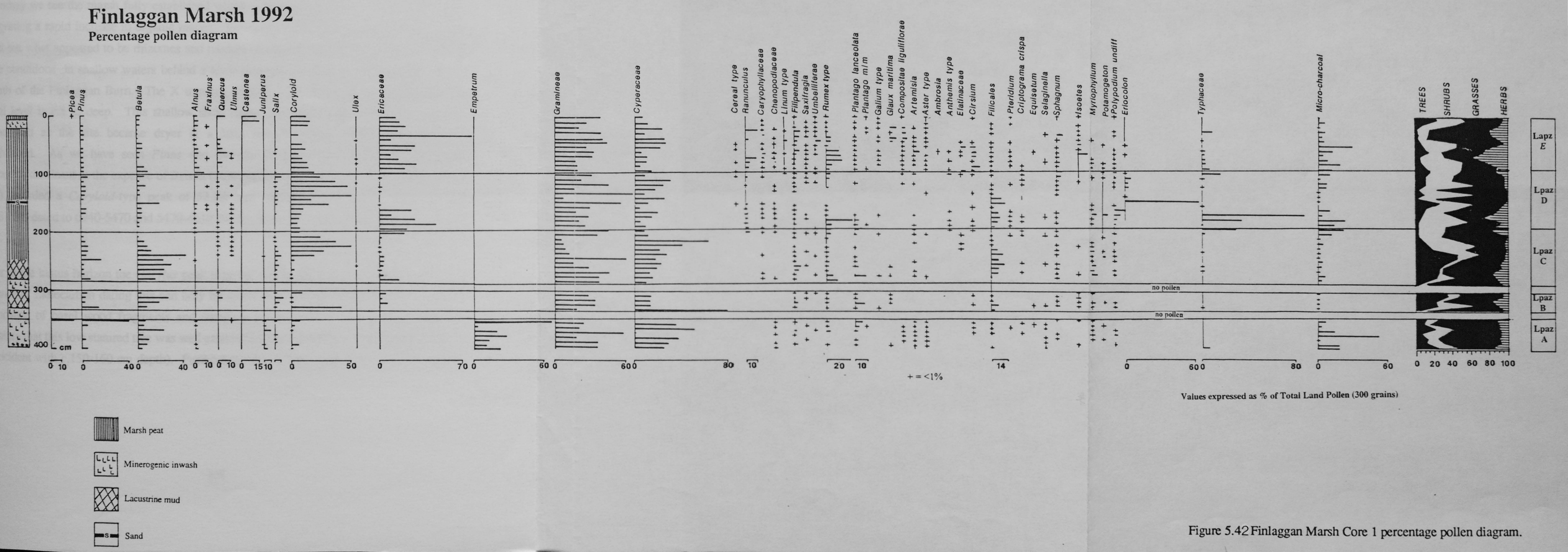


Figure 5.42 Finlaggan Marsh Core 1 percentage pollen diagram.

5.7.3 Dating the Finlaggan Marsh cores using the pollen and chemical record

In the absence of radiocarbon dating only a qualitative assessment can be made regarding the timing of events at Finlaggan Marsh. Table 5.8 below is a summary of the marsh sequence with proposed time-frame based on the pollen, chemical and sedimentary record. Owing to the abundance of open-heath type taxa in the basal minerogenic zones (Lpaz a-b) it is proposed that this section of the core may represents a pollen assemblage that has formed within the Late Glacial period. The basal sediments contain angular fractured stones of various sizes and are most likely to be of fluvio-glacial origin. At the Lpaz a-b boundary there is a rise in *Pinus* frequency to 40% which suggests it was present locally. The presence of *Empetrum* (60%) and *Juniperus* in lesser amounts (10%) suggests a Lateglacial date for this section of the pollen profile. Edwards (1994) recorded a lack of Juniper in the Lateglacial pollen assemblage from Loch a' *Bhogaidh*, Rinns of Islay.

The sediment described between Units 8-11 are akin to lake gyjtta and contains increasing amounts of organic matter content derived from lacustrine plants growing when the marsh was in the early stages of formation.

The upper boundary of Unit 8 to the base consists of a fully lacustrine formed deposit seperated by a distinct hiatus between the marsh peat above. Directly above the hiatus boundary we see the marsh fully established which is reflected by the rise in the LOI curve, suggesting a rapid increase in organic matter. No macro-fossils could be identified to genus level but what appeared to be rhizomes and rootlets of aquatic vegetation reflect the onset of mire conditions in shallow waters behind a levee between the shore of *Eilean Mor* and the mouth of the Finlaggan Burn. The X transect (Figure 5.29) shows that from 0-28 cm the basal level is *c.*1 m deep. This shallow shelf region has been rapidly colonised by marsh plants and as the site became dryer at a later period *Corylus avellana* became well established. As we have seen *Pinus* and *Corylus avellana* was locally present (50%) becoming dominant at the expense of *Betula* (10%) in the middle of Lpaz *C*. Edwards (1994, 759) recorded a *Coryloid*-type peak of 52.2% and 66.2% TLP in zones (LABIc(i) and LABIId(i) dated to 6940-5470 and 5470-4610 BP respectively at *Loch a Bhogaidh* (see Table 4.1).

Prior to the hiatus horizon the *Corylus* peak may fall within this broad date range but in the absence of radiocarbon dating this can only be speculation. Higher in the profile, the sheer abundance of hazel wood fragments and complete hazel nuts show with some degree of certainty that this low statured tree was well established prior to the onset of the Little Ice Age (coincident with *c.*150-160 cm depth). Furthermore the fact that hazel nuts are from mature trees perhaps suggests that these were at this time, not being managed for roundwood in this particular location (cf. Rackham 1980).

The sand horizon is immediately above the woody layer and suggests a major flood event and soon after there must have been a dramatic increase in water level. This remained high enough for submerged aquatics to become established (cf. *Ericolon* peak in zone). This flood event may have led to sand bars as a result of levee formation and might be taken to be the result of increasing climatic deterioration at around the early 1500s (the Little Ice Age?).

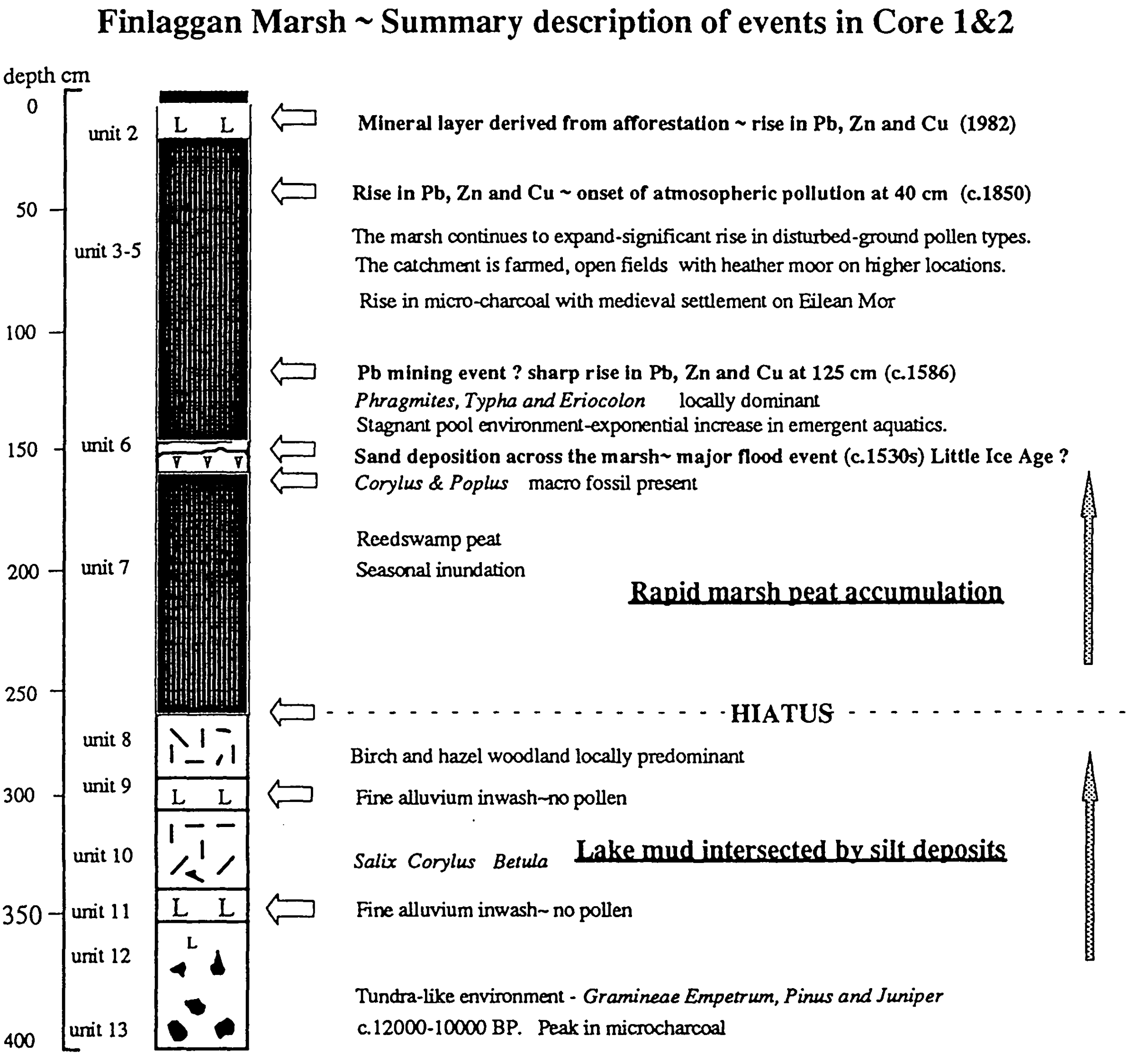


Table 5.8 Description of events in the local environment inferred from Core 1 and 2.

Charcoal in pollen studies has traditionally been associated with woodland clearance (Clark 1982; Patterson, *et al* 1987; Edwards 1990 and Bennett *et al* 1990). Edwards (1990) has identified a positive relationship between woodland decline and the coincident increases in microscopic charcoal. Accepting the arguments for and against the fire retarding quality of Britain's native species (Rackham 1980), the anthropogenic role in woodland clearance by fire, particularly in the Mesolithic and the Mesolithic-Neolithic Transition (Edwards 1990,71) seems conclusive. The link between deliberate burning and its effect on depositional environments has been further strengthened using a combination of palaeocological analyses in conjunction with fire-history records. These have allowed the construction of transportational models and importantly the role of post-fire vegetation succession and its effect on local pollen assemblages (McDonald *et al* 1991).

It is interesting to note in this study that the microscopic charcoal curve exhibits three interesting peaks (Figure 5.43). It is suggested that the charcoal rise in Lpaz A is either an allogenic component of glacially derived material or residual material from wildfires in a very dry Lateglacial environment.. High frequencies of charcoal in Lateglacial minerogenic deposits are not rare (Whittington *et al* 1990, Edwards 1994, 757). At the C/D Lpaz boundary there is another sharp rise in microscopic charcoal that is coincident with a rapid decline in *Coryloid*-type pollen and it is tempting to interpret this with deliberate woodland removal. Higher in the profile there is a rise and fall-off in charcoal (*c*.40cm depth) which could be ascribed to medieval settlement and post-abandonment cultivation on *Eilean Mor*. There is no archaeological evidence to suggest that occupation occurred later than the early 17th century (Caldwell *pers comm.*) and this may account for the decline in local airborne charcoal influx into the marsh sediment. If we take into consideration that the core site is 60 m down-wind from a large centre of occupation then this gives this hypothesis increased plausibility.

It is clear from the Finlaggan pollen diagram that the landscape around the site at one time (*c*.1200-1000 BP) was predominantly tundra-like with grasses and *Empetrum* alongside stands of mixed Pine and Birch woodland on the more acidic soils. Later Hazel colonised the more fertile limestone regions and in particular around the margins of *Eilean Mor*. Here at least some of the trees reached maturity before being inundated by rising loch levels. Whether the increase in water level was due to a deteriorating climate combined with an ever decreasing woodland cover we cannot be sure.

Towards the upper sediment sections (2 cm depth) *Picea* is recorded (<1%) along with *Castenea sativa* (Sweet Chestnut) which were both introduced in the 19th century in gardens on the nearby Lossit and Islay estates. The rise in Pb, Zn and Cu at 40 cm has been attributed to atmospheric deposition (Chapter 9 Section 9.5.1). This event is approximately dated to *c.*1850. The mineral deposit at 6-10 cm depth has been formed as a direct result of afforestation in 1982. A calculated sediment rate is established from the influx of Pb, Zn and Cu at 40 cm (i.e. the onset of industrial derived pollution *c.*1850).

This allows an approximate annual rate of sedimentation of 142 mm yr^{1} which is in broad agreement with the sediment that has formed above the 1982 afforestation event. When this value is extrapolated linearly down the profile the hiatus event (i.e. upper boundary of Unit 8) dates approximately to 1200 AD which is taken as the onset of marsh formation. As farming practices intensified through the Prehistoric Period so we can invisage a gradual shallowing at the mouth of the burn due to sediment loading. The gradual erosion of the shoreline surrounding *Eilean Mor* has led to additional increase in sediment to form the 'tail' of the marsh. The marsh developed as the region became more sheltered and shallow enough to allow the onset of hydroseral succession. According to the pollen record *Phragmites australis* appears to have been the most important colonizer in the history of its development.

5.8 Conclusions

Section 5.2 of this chapter placed the loch in its environmental setting and showed that there is great diversity in the geologies, soil types and vegetation around the loch. The largest and most significant modern land use change has been afforestation of 45% of the catchment in 1982. This resulted in the inwash of fine silts across the near surface of Finlaggan Marsh due to the effects of deep ploughing. There is substantial evidence for intense land use in the early part of the 19th century with extensive tracts of runrig cultivation following early 19th century land-use pressure. In parts these are seen to be integrated into an organised system of land allotment dating to the Medieval and possibly the Prehistoric Period.

In Section 5.3, the discussion concentrated on the identification of lead mines within the catchment. The mines were surveyed in order to determine the extent of the physical remains of the latest phases of mining activity. It was stated above that it was important to establish the principal route of mine drainage as leats and adits form a direct environmental gradient for the influx of heavy metals into the loch basin This work also has shown that two phases of mining can be identified at the Mulreesh and Robolls mines dating to the 18th and 19th centuries. At Portnealon the mining remains relate only to a late 18th century phase. Both the Mulreesh and Robolls mines contain evidence to suggest that some of the remains do appear to be much earlier based on the evidence of open-cast trenches adjacent to Tertiary basalt dykes. The significance of these features is that near-surface lead deposits have been easily exploited from contact zones close to the dykes. Section 5.4 formed a re-appraisal of previous geochemical survey at the Portnealon mine and the results show that Pb, Zn and to a lesser extent Cu are highly concentrated within surface soils owing to the presence of contamination from mine spoil. Similar results were obtained from a sediment core taken from an adit close to the mine. This highlighted the amount of heavy metal contamination formed as a direct result of seepage flowing from the site.

Section 5.5 discussed the results of chemical analyses from a single core taken from the deepest part of Loch Finlaggan. The results from the calculated ^{210}Pb inventory at the upper most region of Loch Finlaggan Core 2 shows this section of the core to be affected by sediment disturbance. This has constrained the estimation of an annual rate of sedimentation using the ^{210}Pb record. The ^{137}Cs record allows only a tentative chronology for part of the core (0-20cm) and therefore only a qualitative assessment can be made regarding the geochemical inventory. A tentative date of *c.*1835 based on linear extrapolation of 1.3 mm yr^{-1} calculated from the ^{137}Cs profile is proposed to date the concerted peaks in Pb, Zn, Fe and magnetic susceptibility rise at 20 cm depth. At around this period (based on the census records of 1831) Islay's population was at its highest and the agricultural base had to intensify to cope with this. This resulted in a massive intake of new and marginal land for agricultural purposes. This may have given rise to higher concentrations of Pb, Zn, Fe and magnetic susceptibility.

Section 5.6 discussed the results of geochemical and stratigraphical investigation at Finlaggan Marsh. The biostratigraphy suggests that at its base, the sediment was formed initially by lacustrine sediment with overlying marsh sediment being derived mainly from the remains of *Phragmites australis* (common reed) and mineral inwash derived from the Finlaggan Burn.

It will be recalled that Core 2 was recovered to provide a 'background' geochemical inventory with an estimated low Pb, Zn and Cu content. An increase towards the near surface of the core was estimated to contain a chemical response to 18th and 19th century atmospheric fall-out deposition and possibly contamination from lead mining. The calculated near-surface increase in Pb and Zn (estimated to about *c.*1850) is not a result of mining but most probably atmospheric fall-out deposition (Chapter 9 Section 9.5.1). The upper-most sediment sections have also received secondary enrichment of Pb and Zn in the inwash silt band (Unit 2) formed as a result of large-scale afforestation in 1982.

Given the distance between the marsh and the 18th century mines of Portnealon and Robolls it is concluded that these sites have had no direct environmental impact within the marsh sediment. However, at 125 cm depth there is a small concerted rise in Pb, Zn and Cu which could be taken to suggest mining activity, and is estimated to have occurred around *c.*1586. The two bell-pits at the West Shore site may have been prospected around this period. Related ore washing and processing in the Finlaggan Burn might be attributed to the rise in Pb, Zn and Cu at this position.

Future radiocarbon analyses on sediment sections within Finlaggan Marsh Core 1 will allow a more rigid time frame to be established and link specific events in the geochemical and pollen record to other palaeoecological events recorded in the regional pollen record previously described above in Section 4.2.4. Until then, only a tenuous link can be proposed for the palynological record obtained from Finlaggan Core 1 and the above mentioned studies. The palynological and biostratigraphical record decribed in Section 5.7 suggests several major shifts due to the action of continued point-bar abrasion, an event also marked by the sand horizon at 150 cm depth. It can only be speculated whether this was the effect of a single or multiple erosion events. What is certain is that this event marks a severe change in the palynological record. Soon after the deposition of the sand the hazel declined rapidly, possibly as a result of flooding. The rise at *c.*1m depth in Pb, Zn and micro-charcoal may also be taken to reflect medieval occupation on *Eilean Mor* sometime in the 13-14th century with aerial fall-out deposition of micro-charcoal originating from domestic fires and industrial hearths. It is also tempting to assign the decrease in micro-charcoal at 12 cm depth to the general decline and abandonment of the site sometime in the early 17th Century.

The palaeoecological methods employed at the marsh establish that there is a close relationship between the palaeoenvironmetal record and historical environmental changes and human activities at *Eilean Mor*. Only tentative evidence survives with which to suggest an influence from local lead mining. In general, the chemical stratigraphy is mainly being controlled by five principal agencies: (1) natural erosion events, (2) hydrological changes in water levels, (3) land use intensification, (4) atmospheric fall-out deposition and (5) recent afforestation.

Finaly, this chapter has highlighted some of the problems of identifying and dating the chemical response of lead mining in loch and marsh sediment. The following chapter will now examine the results from work carried out in and around the catchment of Loch Bharradail.

CHAPTER 6: LOCH BHARRADAIL

6.1 Introduction

Loch Bharradail was selected for study on the grounds of their being extensive remains of lead mining within its catchment. It will be recalled that a main objective at the outset of the study was to assess the scale of 18th and 19th century lead mining. A pilot survey in 1992 revealed the geographical extent of lead mining remains and further topographical surveys were undertaken in September 1993. This work identified large earthworks associated with open casts, shafts and mine drainage features. The results of archaeological survey are reviewed in the light of contemporary historical accounts that document various aspects of the workings.

Firstly, Loch Bharradail and the mine are linked by a leat. This has been identified as a significant environmental pathway by which heavy metals will enter into the loch sediment (see Chapter 2 Figure 2.7). The proximity between mine and loch was suspected to have severely influenced the chemical and physical record in the loch sediment. Such alterations ought then to provide an inventory of environmental changes in the short term, such as the commencement and cessation of mining operations at the mine.

Secondly, the soil surface environment for Pb, Zn and Cu is evaluated in relation to spoil tips and local geology to determine the scale of heavy metal contamination resulting from the mine. Soil geochemical investigations by a mineral prospecting company around Loch Bharradail have provided numerical data on the concentration of Pb, Zn and Cu in the local soils. A re-evaluation of the data from this survey points to natural geological and mine derived influences to account for anomalous soil values of Pb, Zn and Cu at the site.

Thirdly, the sedimentary flux of Pb, Zn, Cu, Ca, Fe and Mn is examined from Loch Bharradail Core 2. Observations based on depth versus concentration profiles are supported by a ^{210}Pb and ^{137}Cs chronology. Additional limited chronological data has been inferred from pollen analyses with supporting historical accounts of the scale of mining operations. Analyses of ^{14}C could not provide a longer supporting time scale due to the influx of old carbon, a result of soil disturbance from mining or cultivation in the catchment. The calculated dates from ^{14}C dating are all within the same age range thus limiting their potential to support a ^{210}Pb-derived time-frame.

The environmental record from Loch Bharradail is assessed in order to identify early (unrecorded) periods of mining. The problems and overall success of this particular investigation is discussed alongside the environmental record obtained from Loch Leathann which was chosen as a control.

6.2 Location

6.2.1 Topographical setting

Loch Bharradail (Plate 13, Appendix 1) lies at an altitude of 100 m OD (Table 6.1). The loch is roughly triangular in shape with bays at its south western and northern end. A small island is situated within the loch towards the northern end with evidence of occupation of an unknown date. The western shoreline has a substrate of small stones and silts, whilst the eastern side of the loch has a shear face of out-cropping phyllites that is up to 3m high in places. There are two major inlets into the loch. The primary inlet is a small burn that originates as the outflow stream from Loch Fada (Grid reference NR 636408). The second major inlet is an artificial watercourse or leat (Plate 12 and Feature 20 in Figure 6.2) that drains an adit. There are also many smaller seepage burns leading into the loch.

Grid reference	NR 636408
Core date	September 1992
Catchment geology	limestone-pyhllites
Catchment: land-use type	rough grazing
Loch altitude (m)	100
pH	7.4
Conductivity (μmhos)	120
Max. depth (m)	6.1
Loch area (ha)	3.74
Catchment area* (ha)	149.2
Catchment:loch ratio	39.8
Afforestation	0
Net relief (m)	178 (m)

Table 6.1 Loch Bharradail ~ site characteristics (note* excluding the loch).

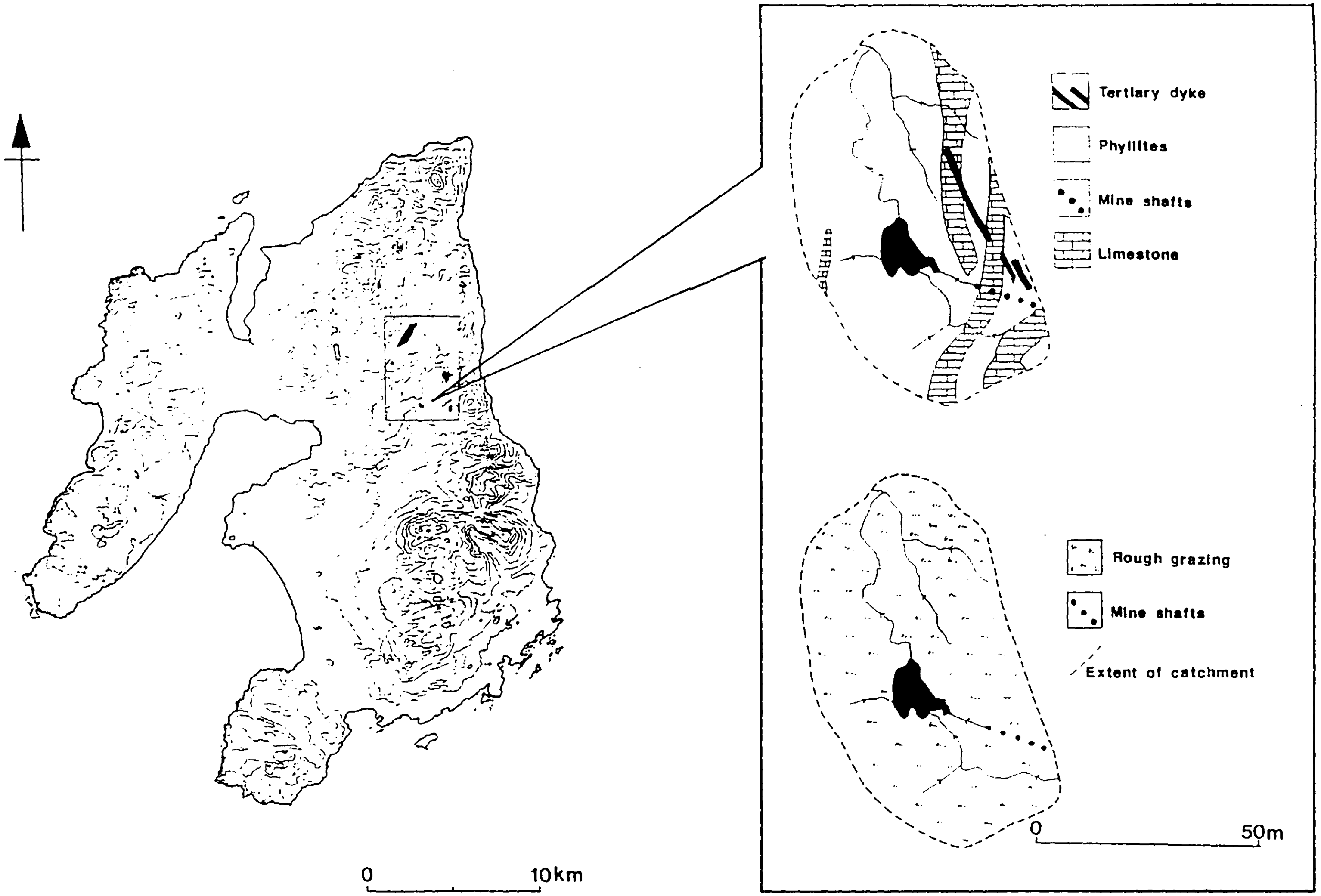


Figure 6.1 Location of Loch Bharradail with local geology and present day land use.

6.2.2 Geological setting (Figure 6.1)

Loch Bharradail is surrounded on all sides by acidic slates and phyllites. A Dalradian limestone (Appin Group) escarpment is situated to the north-east of the loch and trends in a north by north-westerly direction. A Tertiary dyke runs approximately parallel with the escarpment. Mineralisation has occurred extensively in this region and Pb was exploited both within the catchment and to the north at the site known as North Ardachie. In the 19th century limestone was quarried along the escarpment for agricultural improvement.

6.2.3 Soils, vegetation and catchment land use (Figure 6.1)

The topography surrounding the loch is contrasting, with a slope to the west that rises to an altitude of 178 m. On the eastern side of the loch, the land is undulating for about *c* 400 m then rises sharply to a limestone escarpment at 190 m OD. At the north end of the loch there is an outflow burn (*Allt Loch Bhara-dail*) that flows northwards eventually to join the River Sorn. The soil types within the Loch Bharradail catchment are the Foundland Series of drift deposits that are derived mainly from slates and phyllites. Peaty glays and podsols are predominant in the region and support blanket bog plants such as *Festuca* grassland and *Calluna vulgaris* along with heath-rush type vegetation. On the freely drained limestone escarpment the shallow Deecastle Association comprise brown rankers and light loams.

The limestone escarpment and the lower region of phyllite and shales supports rough grazing. Extensive peat cuttings can be seen to the south-west of the loch and are related to the occupation of two small steadings that were abandoned in the late 19th century (D.Bell pers comm.). Flying-bent grassland communities are widespread and alkaline loving species such as the *Campanula rotundifolia* and *Orchis mascula* are frequent. The vegetation in and around the loch margins is very dense with marshland vegetation. The predominant marsh types are *Phragmites australis* with interspersed *Cladium* sp. On the edge of the loch *Potamageton natans* is well established and towards the centre of the loch *Nymphaea alba* is predominant. On the spoil heaps, vegetation is noticeably sparse owing to enhanced heavy metal concentrations. Where flora is established, the predominant types are *Plantago lanceolata L*, and *Rumex obtusifolius*.

6.3 Historical and Archaeological context

6.3.1 Historical Context

The *Book of Islay* (Smith 1895, 456-467) has been the primary source of information concerning the history of lead mining on Islay. Unfortunately, no maps or plans accompany the texts which show the extent of operations at South Ardachie but we are fortunate in having detailed descriptions relating the scale of operations at the mine. The mine of South Ardachie is situated on the eighteenth century land holding of 'Ardochy' and two accounts survive recording the extent and the limit of the mineralisation. In 1770 the lead mine was visited by Alexander Shiriff who described the workings in great detail. He mentions that the site was the principal mine on Islay in 1770 and records the presence of several open cast features that were worked *'in former times'*. In his report, *State of the Mines in Islay* he also mentions:

> "An east-west vein is presently being worked for a length of 30 fathoms" (54.6m) the vein is crossed by two whindykes (Tertiary dykes) and ore is up to 2½ feet wide along one of their sides"

Shortly before 1770 Charles Freeburn the then manager of the mining operations on Islay, referred to several cross-cut veins that were being worked together. These veins were beneath areas worked at an earlier period. He mentions the presence of :

> "two lead veins, one producing near one hundred tons of ore and 'more for the taking'"

Between these two veins more ore was available, Freeburn noted:

> "All the intermediate ground betwixt these two veins are closely chequered with other veins that have been opened formerly, all holding ore, but none of them prosecuted to any depth" (Quoted Smith 1895, 463).

It appears that the mine was particularly rich in ore and again we see reference to previously worked-out areas, a footnote by the same writer dating to 1769 mentions the richness of the new seams:

> "This last mentioned vein is now nine feet wide, and has well mixed ore almost from side to side. The last five days' workings took out 4 and ½ tons of ore from one set of four miners" (Quoted in Smith, 1895, 464).

The documentary sources make numerous references to Tertiary dykes. There can be little doubt that the lead ore is most certainly associated with metamorphic contact zones between the Olivine basalt and the Dalradian limestone. Alexander Shiriff mentions 'brangled' ore which refers to breciated lead ore, a mixture of the primary ore, dolerite and limestone. An example of contact metamorphism is shown in Plate 2 Appendix 1 which was obtained from the limestone quarry in Ballygrant. We know from the Daybook of Daniel Campbell of Shawfield (1767) that two cargoes of rough and dressed ore were shipped to the mainland in 1776 at a cost of £22.1s and £24.10s Sterling respectively, to a merchant named Ronald Crawford of Glasgow (Ramsay 1991, 193). This account is interesting as it shows that the lead was leaving Islay in both a rough and dressed state and not only in ingot form. This probably relates to the requirements of the merchant who was possibly involved in or running a pottery industry and required powdered lead for lead-glaze manufacture.

6.3.2 Archaeological field survey at South Ardachie (Site No 3 in Figure 4.2 and Table 4.3)

6.3.3 Introduction

The archaeological survey of the South Ardachie mine was undertaken using the methods described in Chapter 3, Section 3.2.1. The principal aim of the survey was to gain an understanding of the extent of the remains of the mine and in particular to identify the presence of drainage features leading into Loch Bharradail. One of the problems with the archaeology of mining is the difficulty in relating earthworks and other surface features to descriptions provided in historical documents. Mid-18th century accounts for mining on Islay mention only the depth the miners worked to and to some extent, the quality of the ore deposits being mined. Most often these statements are poor in describing technological information and the geographical extent of the surface activities of the industry. Today, the major characteristics of the earthworks comprise surface workings and a series of spoil tips with related features such as open-cast workings and mine shafts. The spoil tips vary in composition from local country rock (5-150 mm) to fine secondary tailings (<2 mm). The latter are confined to the valley slopes and in the region of the line of mine shafts. Several small dumps of limestone, and pyrite-rich slate are dispersed around the entrances of the shafts. The region described as Area B is enclosed in parts by field boundaries forming an enclosure of arable land.

6.3.4 The earthworks survey

For ease of description the survey has been divided into two areas (A and B) as the workings are extensive covering an area of about 12 ha and individual features are numbered (1-20) respectively.

Area A

Area A is set in a valley bottom surrounded by land now used as rough grazing. The Mulindry road crosses a former east-west oriented quarry feature for a distance of *c.*45 m and is in places *c.*6-8 m in width (1). A number of smaller workings on the same alignment can be seen close by. Several terraces made up of mine spoil descend to a north-south trending adit with a farm refuse-filled interior (2). The trial is intersected by a shaft (3). Next to the shaft is an adit (4). The banks of this feature are well preserved with water draining along the base of the valley floor towards the small burn leading from Loch Fada and eventually into Loch Bharradail (5). Leading away from the adit, the land rises steeply with a large rectangular bank enclosing rig and furrow (6). At the crest of the slope a series of features includes a small test pit cut into the limestone outcrop (7). Rock drill holes and evidence of fire scaring suggest this feature is an exploration test-pit caused by blasting. A plateau of rough pasture is bounded by a region of large tailings dumps that lead to the crest of the slope that falls steeply down to Area B.

Area B

A series of roughly east-west trending trials are enclosed by a sharply defined enclosure bank (8). The most northerly feature is the largest with an unknown depth (9). Commonly, most of these large features have been backfilled with farm rubbish masking their true depth, and in this case, their possible function. It is suggested that this feature may have been an adit as seepage runs downslope into a leat that has been cut through an enclosure bank and a trackway (10). A small rectangular building (11) has been reverted into the bank alongside the track (Plate 8 Appendix 1). The age and function of this building are unknown as is that of the building immediately to the west (12). To the south of building (12) there is a large opencast with extensive tailings aligned east-west (13).

To the south of this feature, a large ovoid tailings dump is present (14). The tailings within the tip are exclusively iron pyrite-rich phyllites and slates. To the west of this feature a series of five rubbish-filled shafts have on average an internal diameter of 6-8 m (15-19). Each shaft is surrounded by a ring of waste that makes them resemble bomb-craters. The shaft farthest from Area B adjoins a large leat 3 m deep between large banks (20). This feature traverses an area of poor grazing for some 170 metres (Plate 12, Appendix 1).

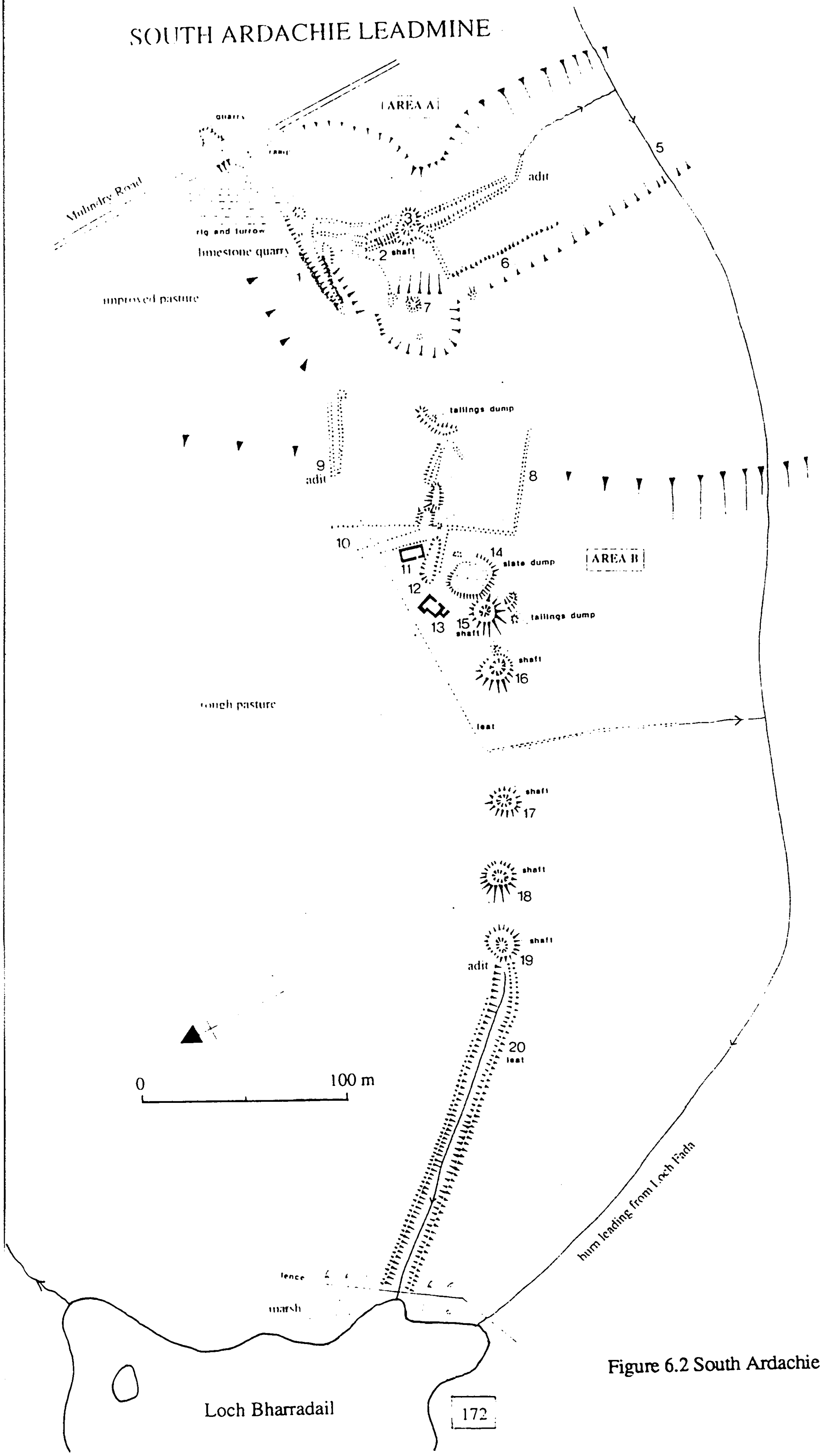


Figure 6.2 South Ardachie leadmine survey.

A point worthy of note is that the material incorporated in these well-pronounced banks is mine tailings. This is evident by their verdant fertile slopes and the presence of *Campanula rotundifolia* (Common Harebell) a plant that prefers alkaline soil.

6.3.5 Interpretation of the results and phase reconstruction

Some doubt is cast on the antiquity of the large open cast quarry on the east side of the Mulindry road. This feature was suggested to have followed the strike of a Tertiary dyke, (Callender and Macaulay 1986, 19). However, I would argue that evidence for mining alongside this feature is lacking as there appears to be no basalt waste. Olivine basalt is, and was then, of no economic importance to the miners and the lack of tailings from this material supports the argument that no dyke was present in the near vicinity of this feature. Secondly, the alignment of this feature does not conform with the general trend of north by north-west as is common with Tertiary activity in the area. A nineteenth century lime kiln situated at Grid reference NR.404648 may well have burnt limestone worked from this quarry. A ramp-like feature oblique to the alignment of the road may have been used as a ramp to lift limestone from the valley floor. Substantial pieces of iron work and timber are still *in situ* and point to this feature being much later than the last episode of lead mining.

Four phases of activity may be identified

(1) *Enclosure banks and early field systems* (Features 6 and 8 in Figure 6.2). The survey has shown that some of the mine workings are superimposed over an earlier field system made up of two distinct compartments. Collectively, these components formed an earlier land holding (commonly termed a tack) with rights to farm the land and extract minerals where they occurred within the tenancy. It may be possible that the field systems with enclosing earthen banks and buildings relate to the 16-17th century. By the mid to late 18th century the South Ardachie lead mine was in full-scale operation. Whether the buildings are contemporary with the last phase of mining is a question that cannot be determined without further archaeological investigation. In all probability the buildings were used for storing tools and other mining equipment.

(2) *Open cast features* (Features 8-9). These are the earliest features at the site and it is proposed that they date to the early 16th century or possibly earlier. Such features were recognised in the 18th century accounts as being 'of earlier times'. The numerous open-cast features are probably the opportunistic workings by tenant farmer-miners who exploited near-surface lead deposits.

(3) *Mine shafts and the leat extending into Loch Bharradail* (Features 15-20) The contemporary accounts for South Ardachie mention *"shafts being sunk"*. It is suggested that these features date to the 18th century based on the technology employed in lead mining at this period (Raistrick 1965).

(4) *Limestone quarry in area A* (Feature 1) The early 19th Century saw a period of intense agricultural improvement both locally and on a national scale. Vast quantities of limestone were required for lime burning to improve the quality of farmland. The presence of a nearby 19th century lime kiln (less than 1km from South Ardachie) suggests that the large quarry in Area A was a local source of limestone for lime production.

6.3.6 Implications for the study area

The survey has shed new light on the scale and extent of the mining remains at South Ardachie. Features (8-9) provide evidence of lead extraction dating to a period earlier than the mid-18th century. This theory is supported by contemporary sources which repeat that 'old workings' were continually encountered as the South Ardachie mine developed. Perhaps these old workings were used as a 'prospecting guide' to deeper ore deposits which were more easily exploited using the shaft and adit method of mining. However, we do have clear evidence of ore prospecting as feature (7) incorporates rock-drill holes and evidence of powder blasting.

Moving to the problems of mine drainage, the earthwork banks that form the leat (20) represents an inexpensive means of mine-drainage control. The leat has been dug to a depth of about 2 m and the up-cast has been increased to a greater height using mine tailings. An adit has been placed to exploit the 60 m contour from which the landscape slopes gently down towards Loch Bharradail. Without this form of drainage-water management, ore deposits in Area B which forms the slope of the limestone escarpment, could not have been exploited due to gallery flooding. Contemporary accounts shed no light on the types of pump used at the mine but at this period in mining history, the 'chain' pump (Ford and Rieuwerts 1981,14-15) would have been the type most commonly in use. Moreover, the historical record is noticeably silent as to why South Ardachie did not continue into the 19th century as was the case for the Mulreesh complex. At the former, could problems with flooding have outweighed the expense and effort getting to deeper ore-bodies ? Given the position of the free-draining limestone and the high seasonal rain-fall patterns for the region this seems a likely possibility.

6.4 Geochemical Environment

6.4.1 The catchment distribution of Pb, Zn and Cu based on the DRL geochemical survey

The data obtained from the DRL survey has been re-plotted as discussed in Chapter 3 Section 3.3.2. Table 6.2 provides descriptive statistics of the data used to produce the contour plots. The highest concentration values were omitted from the input data and fall within the 'above' category in metal concentration (ppm). If the largest outlying value (650 ppm) is included in the plot there is a tendency for the software (Unimap) to exaggerate the upper range of the data leading to the suppression of the lower size class interval. For ease of description, the plot title 'catchment distribution' is used in this case to mean land adjacent to or surrounding the loch. In real terms 'catchment' would include a much wider area than shown in the two dimensional contour plots.

The plots should be seen as the optimum representation of the data and display the total concentration for Pb, Zn and Cu the values of which have been increased by the presence of mine waste and mineralisation within the vicinity of Loch Bharradail. (Compare these with the Pb distribution surrounding Loch Leathann (Table 8.2) where no mineralisation has been observed.)

STATISTICS	Pb	Zn	Cu
Observations	51	51	51
Maximum value	650	380	58
Minimum value	10	5	2
Mean value	92	94	10
Median value	60	70	8
Standard deviation	104	81	11

Table 6.2 Statistical characteristics of Pb, Zn and Cu concentration based on DRL geochemical data from South Ardachie mine, Loch Bharradail catchment (values in ppm).

6.4.2 Catchment distribution of Pb (Figure 6.3)

On the basis of the data in Table 6.2 the background (average) concentration for Pb is established at between 40-80 ppm. The maximum concentration for Pb was 650 ppm with a minimum value of 10 ppm. The high standard deviation value of 104 ppm reflects high within-sample variation across the range of Pb values. Lead concentrations are highest within the limestone. Around the mine shafts, the concentration of Pb is moderately high with 120-140 ppm. A linear distribution ranging between 180-200 ppm is attained from the position of the shafts that trend towards the south. East of the loch and still within the edge of the limestone belt, an anomalous value of 650 ppm Pb demonstrates relatively high contamination.

6.4.3 The catchment distribution of Zn (Figure 6.4)

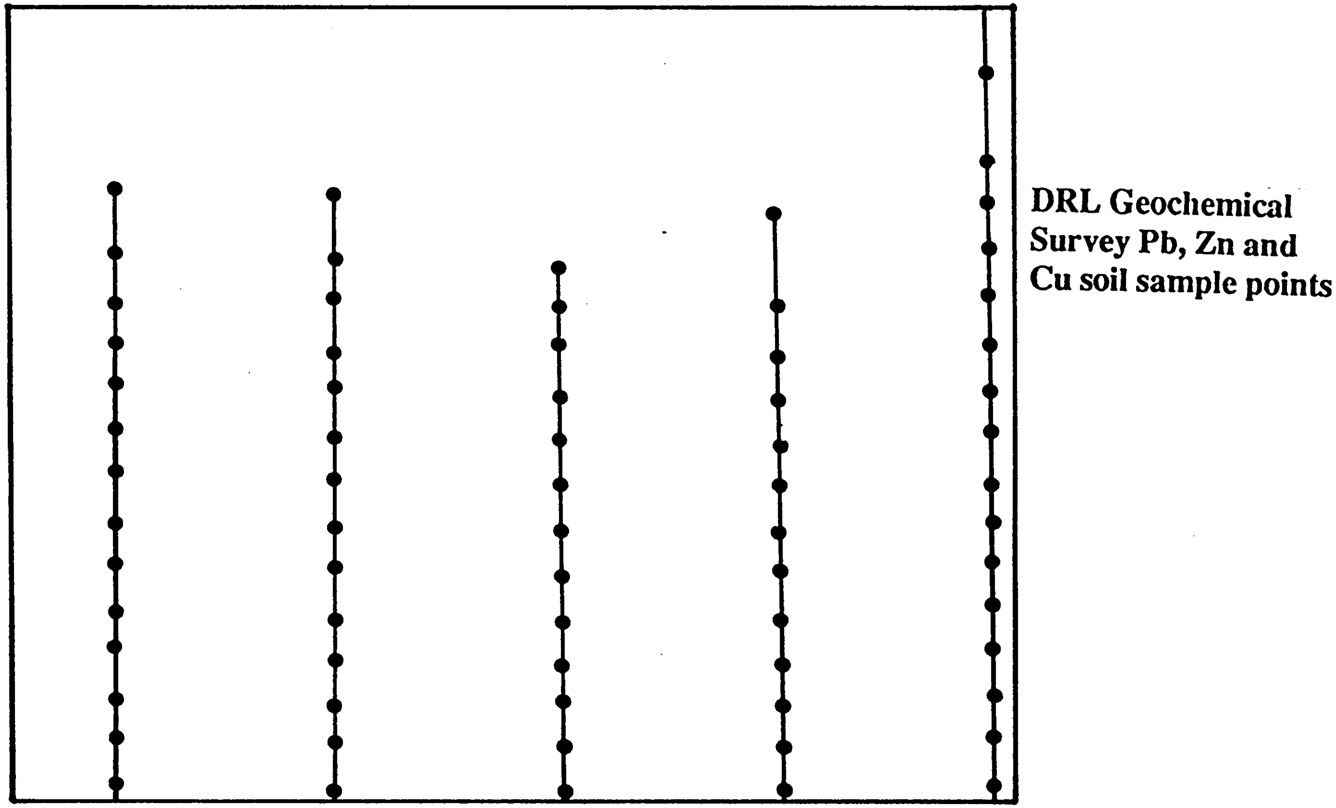
Zinc has a maximum concentration of 380 ppm and a minimum of 5 ppm. With a mean of 94 and a standard deviation of 81 respectively, there is high within-sample variance across the range for this metal. Background concentrations of between 80-100 ppm can be observed in the area to the east of the loch. The area to the west of the loch has lower Zn concentrations with 20-40 ppm. A linear distribution of 100-120 ppm Zn demarcates the edge of the limestone belt. Two anomalies are present within this region. The southern anomaly is concentric with between 120-140 ppm and rising to between 200-220 ppm. To the north a similar anomaly can be seen and is concentric with values ranging from 160-180 ppm, peaking at above 220 ppm.

6.4.4 The catchment distribution of Cu (Figure 6.5)

Copper attains a maximum concentration of 58 ppm against a minimum of 2 ppm. The mean value is 10 ppm with a standard deviation of 11 ppm showing negligible variance across the range of Cu values. Soils adjacent to the loch are low in copper (<10 ppm). The values increase towards the region of the limestone escarpment where a moderately high value of 58 ppm was recorded in the geochemical survey. This anomaly is surrounded by a halo surrounded by Cu values in the range of 20-30 ppm.

6.4.5 Assessment of the observations of Pb, Zn and Cu

The distribution of Pb in the catchment of Loch Bharradail is primarily the result of enrichment from mine spoil and to some lesser extent from naturally derived Pb due to mineralisation within the Dalradian Limestone. The BGS geochemical survey records low or moderate Pb values of 15-70 ppm in the non-mineralised regions of the Dalradian Limestone. Low levels (20-40 ppm) are recorded on the Islay Subgroup of tillites and quartzites and both these geological types occur adjacent to the loch. Zinc appears to be confined in a linear pattern along the Dalradian limestone as a result of the influence of mining spoil and in all probability demarcated according to the distinct geochemical boundary between the limestone and phyllites.


DRL Geochemical
Survey Pb, Zn and
Cu soil sample points

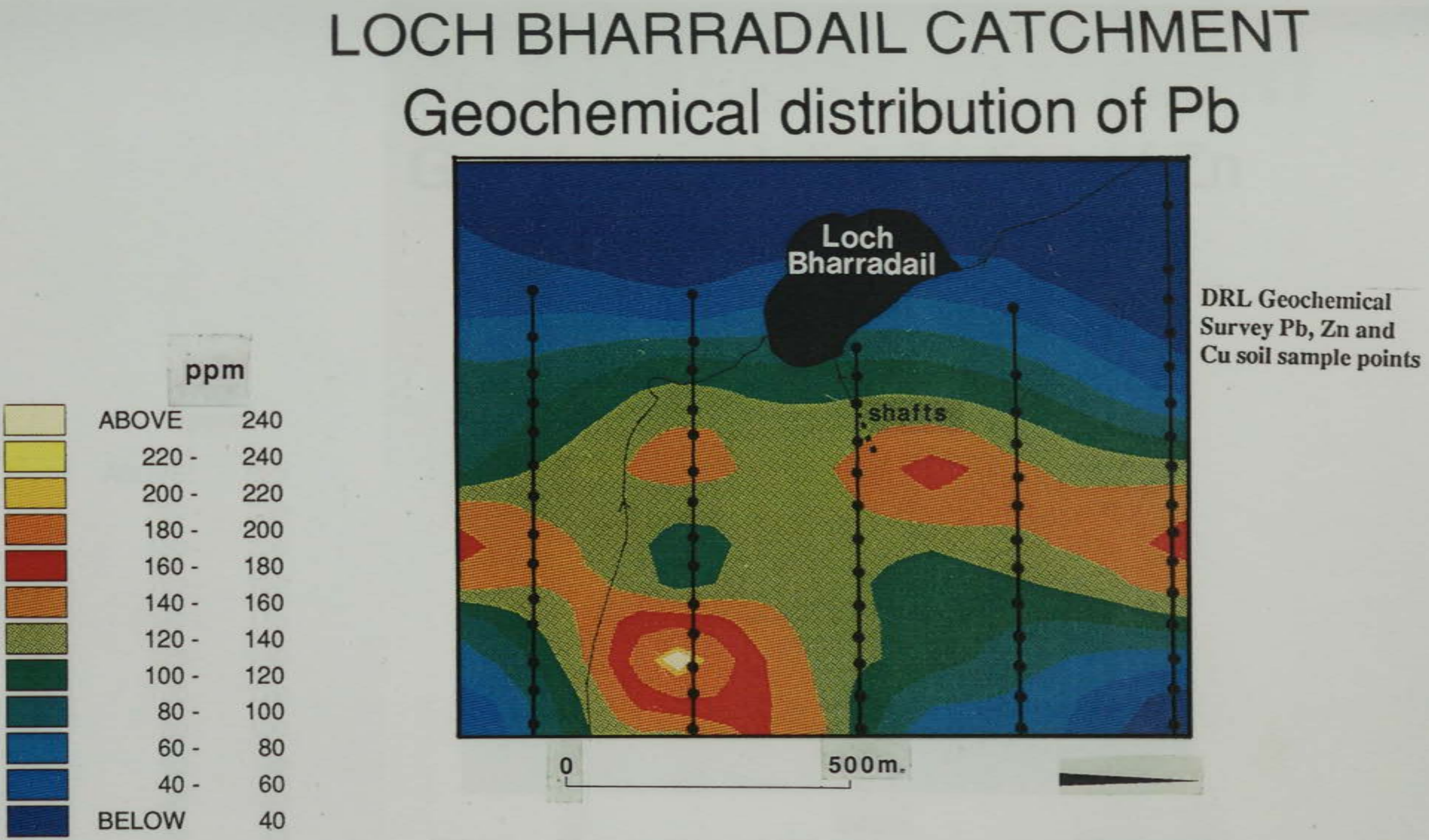


Figure 6.3 Distribution of lead in surface soils across South Ardachie leadmine (after Domego Resources Ltd 1983).

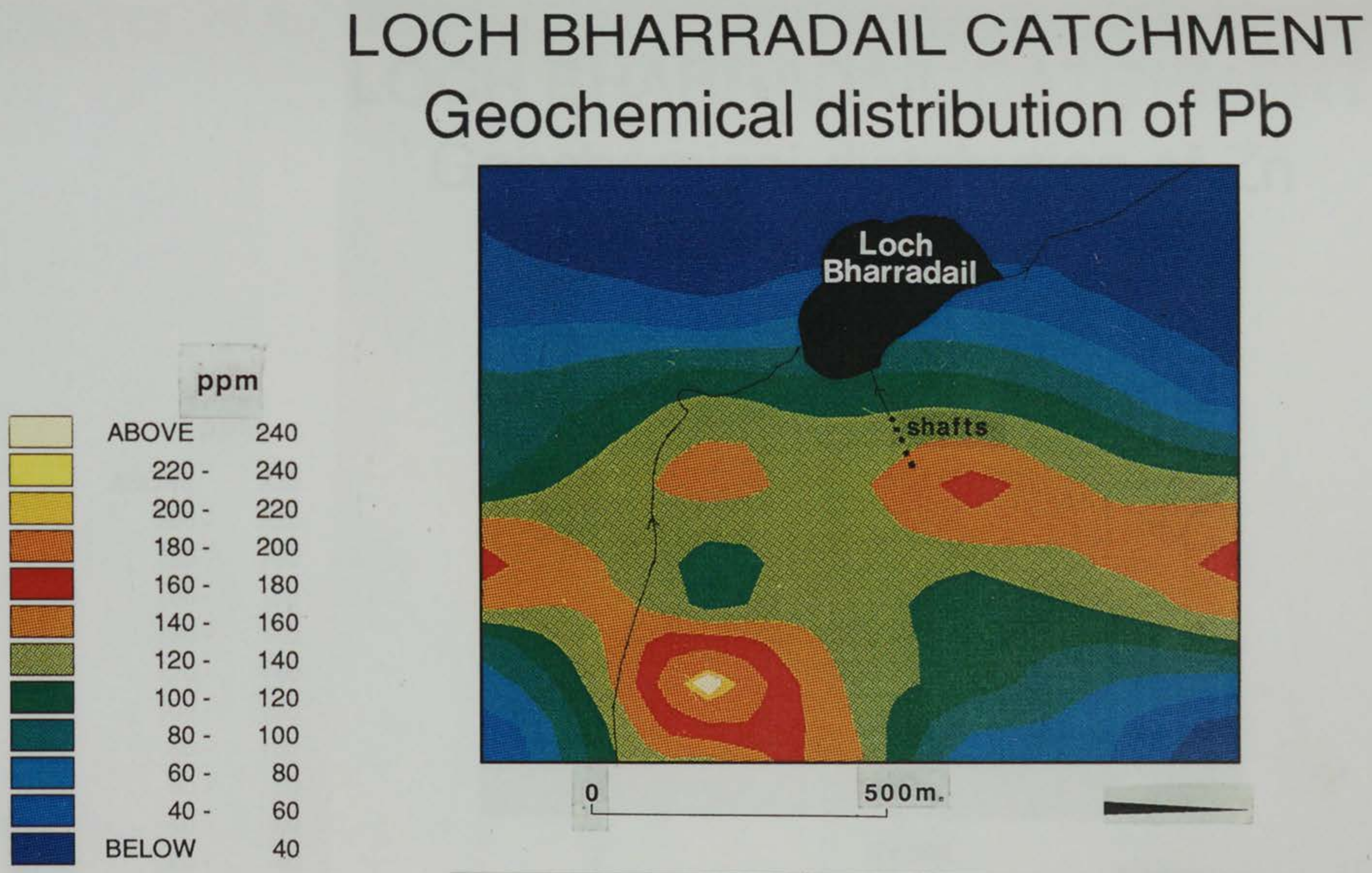


Figure 6.3 Distribution of lead in surface soils across South Ardachie leadmine (after Domego Resources Ltd 1983).

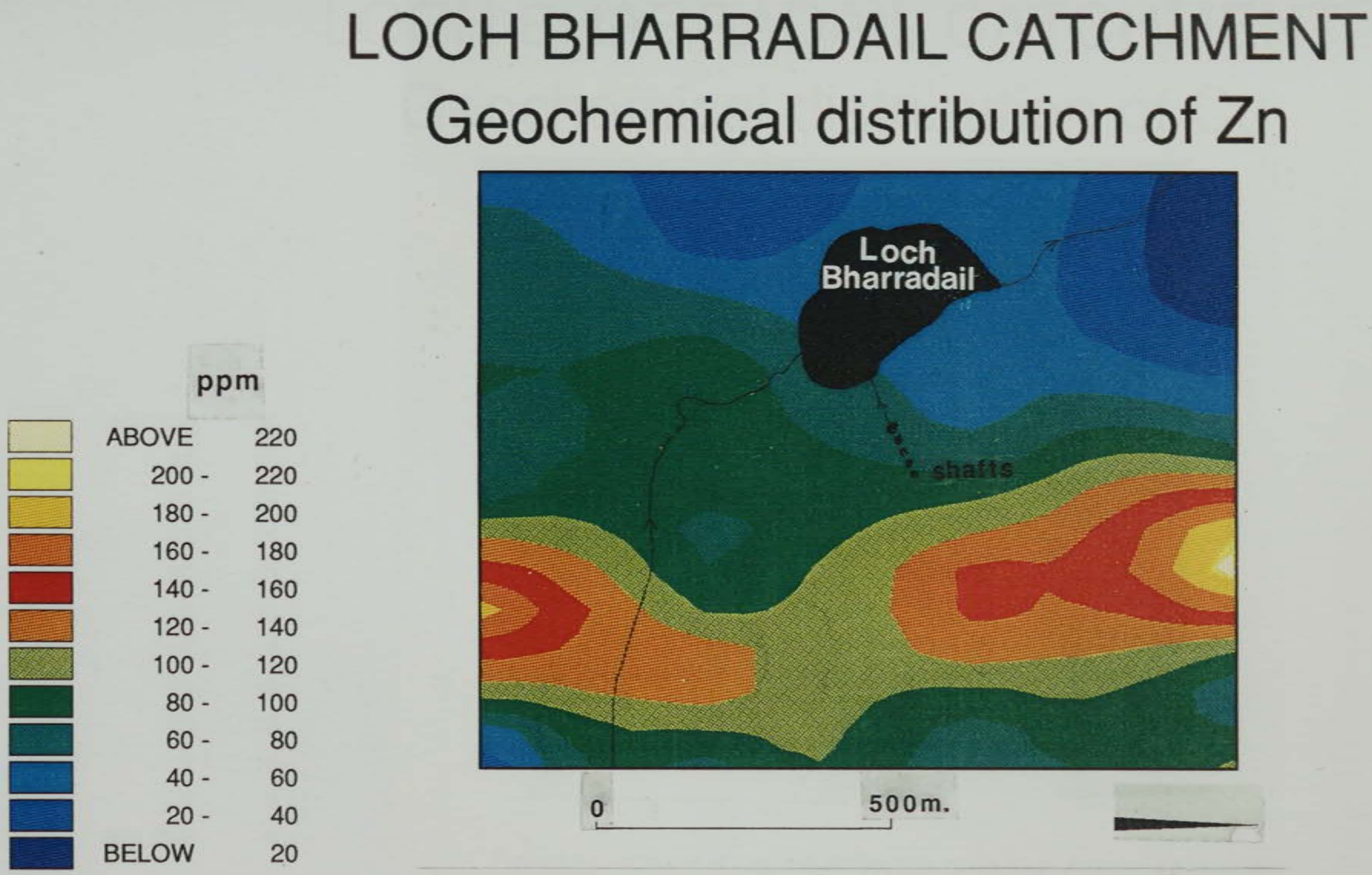


Figure 6.4 Distribution of zinc in surface soils across South Ardachie leadmine (after Domego Resources Ltd 1983).

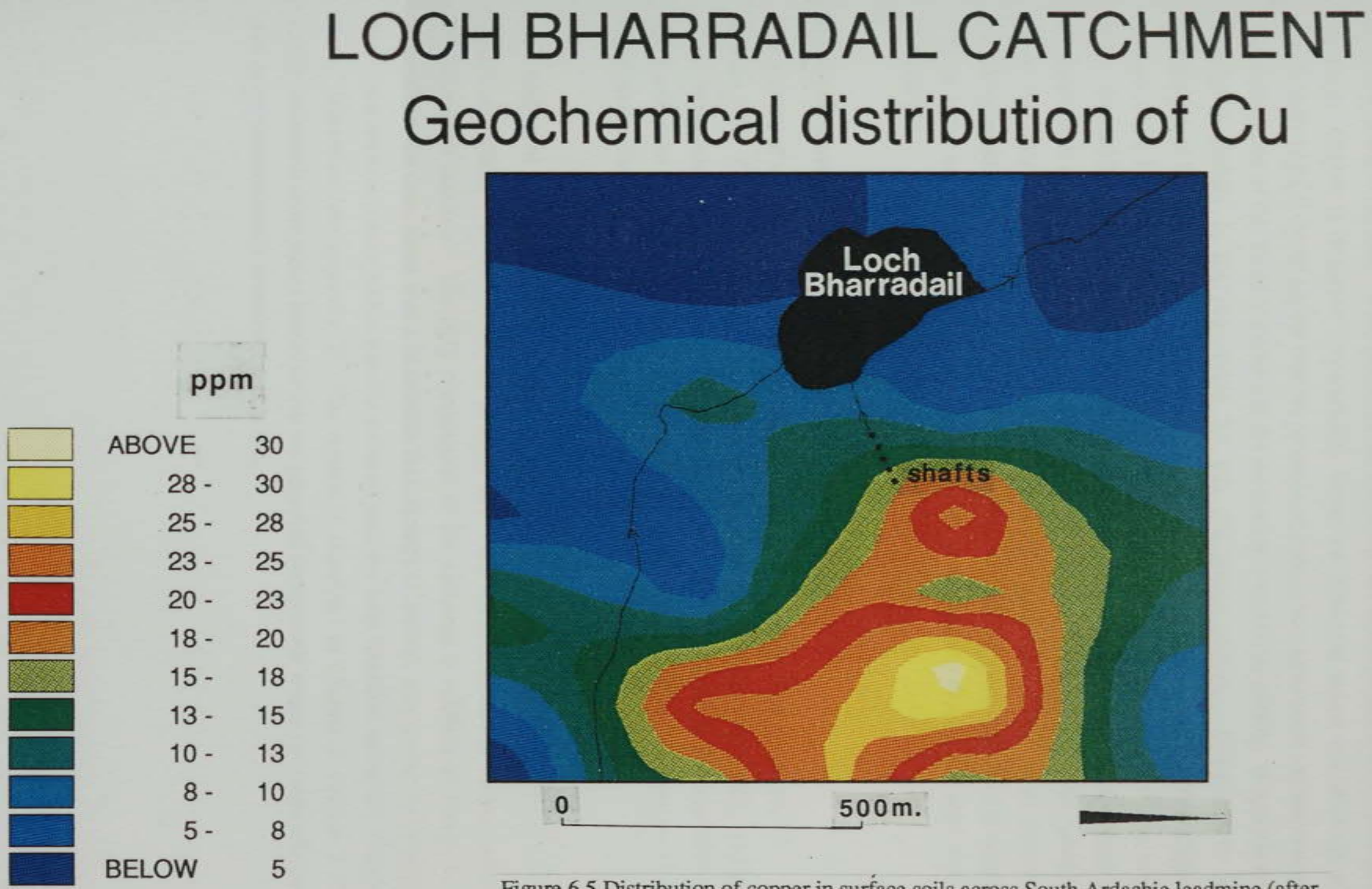


Figure 6.5 Distribution of copper in surface soils across South Ardachie leadmine (after Domego Resources Ltd 1983).

The slate and phyllites region surrounding the loch has concentrations in the range of 60-80 ppm Zn. Zinc appears not to be influenced by the gradient of the slope leading from the limestone down to the loch. Were this to be the case then one would expect a wider pattern of dispersal. Copper is dispersed concentrically within the limestone region and although not significantly high, it would appear that the presence of mining has led to some soil enrichment. The distribution of this metal conforms to the areas most disturbed by mining. It appears that underlying geology is not responsible for this distribution pattern. Compared to the distribution of Pb and Zn which is closely related to the presence of the limestone. Copper appears to be less influenced by geology and more likely reflects contamination from mine spoil. The BGS survey records low (5-20 ppm) values overlying most of the Appin Group of limestone with some enrichment where mineral veins have cut the limestone. The quartzite facies are poor in Cu (<20 ppm). Sporadic anomalies (>50 ppm) have been observed overlying basaltic intrusions. The presence of the Tertiary activity within the local limestone may to some extent be enhancing the Cu values but contamination from mine spoil seems more likely.

6.5 Bathymetric Survey of the loch and core recovery

The method of survey and core recovery and sampling is described in detail in Chapter 3, Section 3.5.2. Owing to the density of submerged vegetation, a complete bathymetric survey of the loch could not be carried out. The central region of the loch was devoid of vegetation and found to be the deepest part and coring was undertaken at this position. The positions of the replicate coring stations are shown on Figure 6.6.

6.5.1 Sediment characteristics of Core 2

Core 2 was selected for analyses on the grounds that it showed no evidence of damage within the uppermost sections. The bulk composition of the sediment is outlined in Figure 6.7. Throughout the core, there was a noticeable homogeneity in texture, colour and consistency. The major mineral phases within specific sediment sections were identified using Dry Powder X-ray diffraction (see appendix 2) The method is described in Chapter 3 Section 3.10. Computed match score tables establish that the principal components within the major mineral phases are predominantly muscovite and quartz.

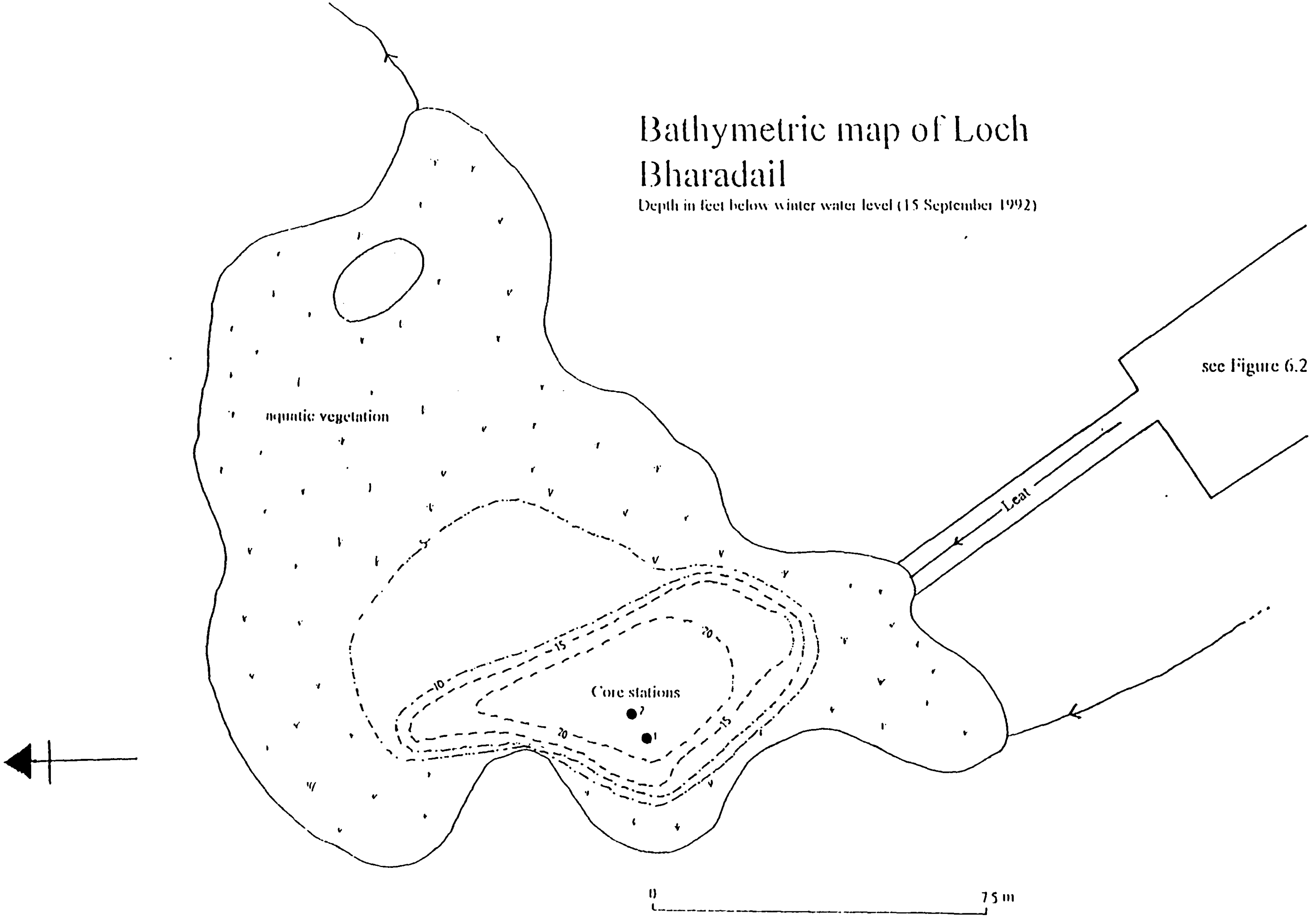


Figure 6.6 Bathymetric map of Loch Bharradail.

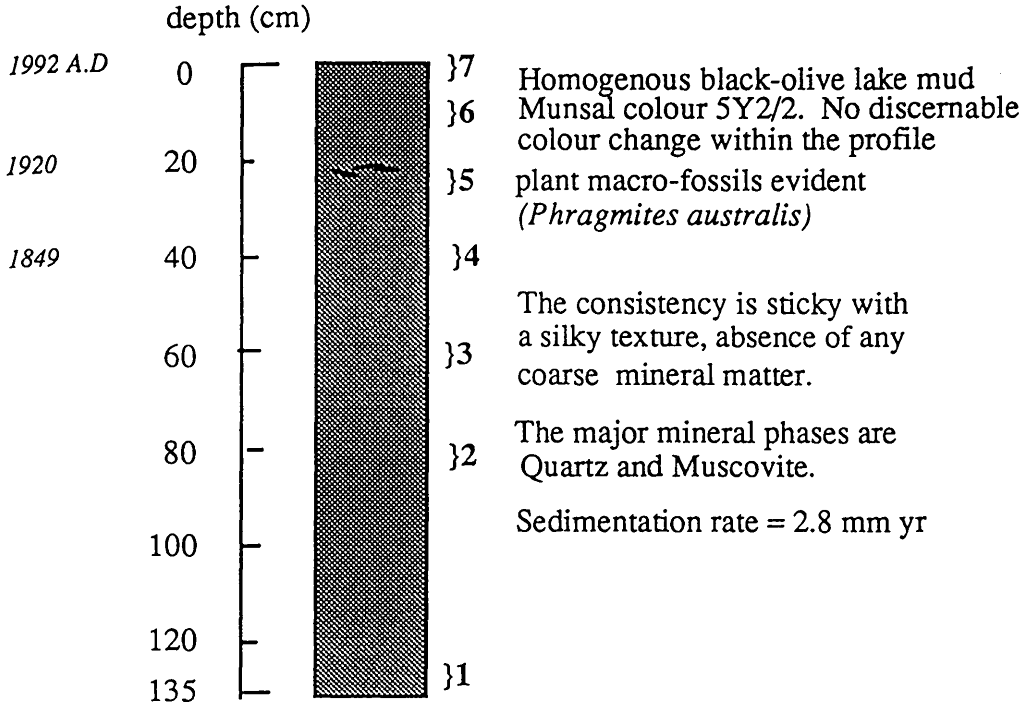


Figure 6.7 Summary description of Core 2 with radiocarbon dating sampling positions (7-1) and linearly extrapolated ^{210}Pb-dates (1992-1850*)* based on an annual sedimentation rate of 2.8 mm yr^{-1}.

6.6 Dating and Environmental History

6.6.1 ^{210}Pb analyses and the formation of an annual rate of sedimentation

The chronology for Core 2 has been established using the ^{210}Pb method outlined in Chapter 3. section 3.8.7. The extrapolated dates on the left of the profile in Figure 6.7 are based on an annual rate of sedimentation of 2.8 mm yr^{-1}. $^{210}Pb_{excess}$ activity was detectable to a depth of 20 cm with a surface maximum (0-1cm) of 338 ± 28.4 Bq kg^{-1} at 1σ. These values have been decay corrected to September 1992 and are shown in Figure 6.11 with error bars placed at 2σ. The results allow a ^{210}Pb-derived sedimentation rate of 2.8 mm yr^{-1} allowing a chronology that has been plotted (in italics) and shown alongside the core in Figure 6.8. The chronology has been linearly extrapolated down the core, but beyond around the 1840's these dates must be viewed as only tentative in the absence of corroborative chronological support. Radiocarbon dating does not support the linearly extrapolated ^{210}Pb-derived annual rate of sedimentation. However on the upper sections of the core (0-20 cm) it is proposed that the established ^{210}Pb chronology is more reliable.

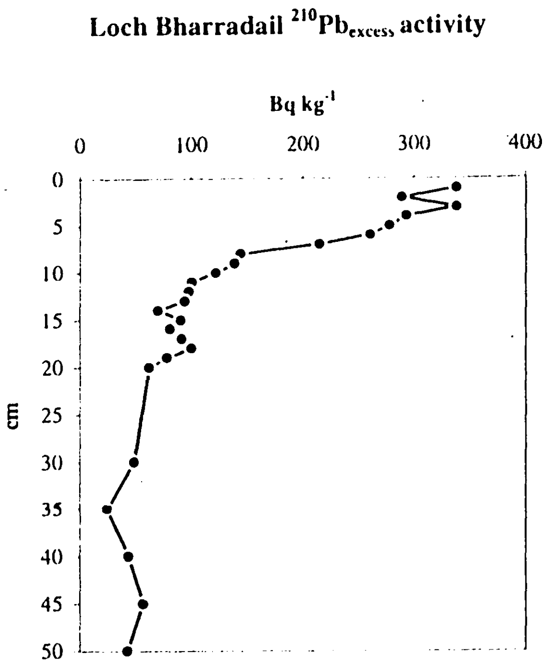


Figure 6.8 Depth versus $^{210}Pb_{excess}$ activity.

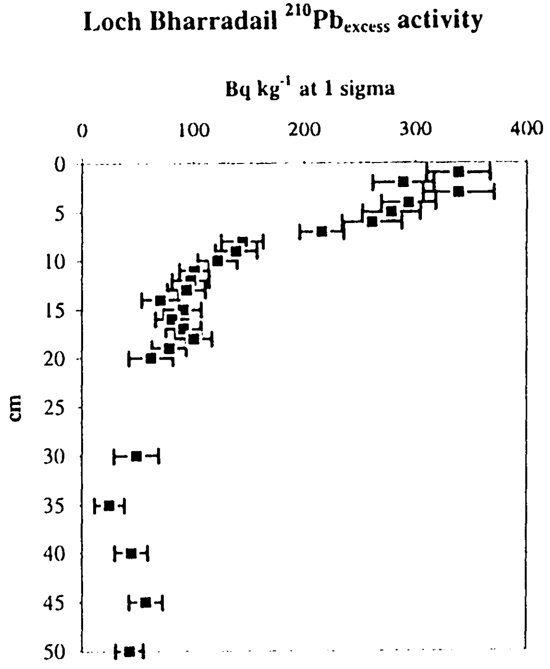


Figure 6.8 Depth versus $^{210}Pb_{excess}$ with error bars at 1 sigma.

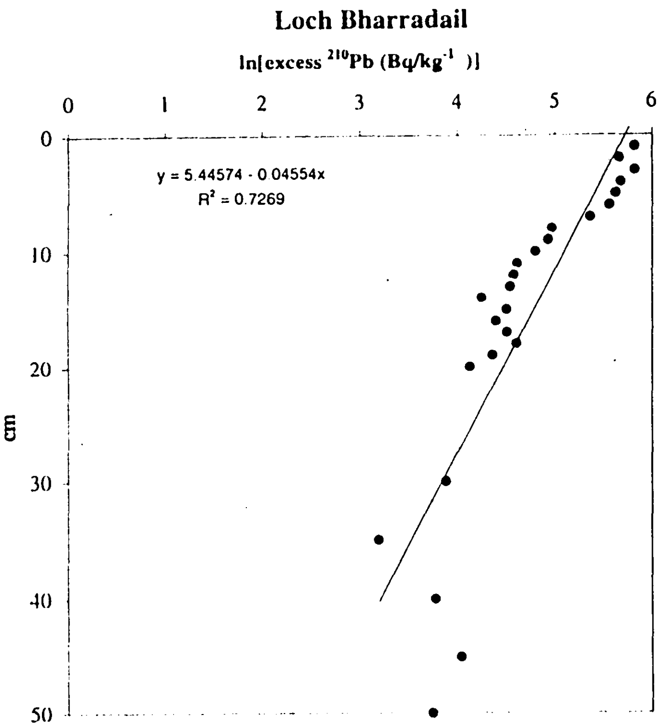


Figure 6.9 Linear regression plot of log normal $^{210}Pb_{excess}$ activity.

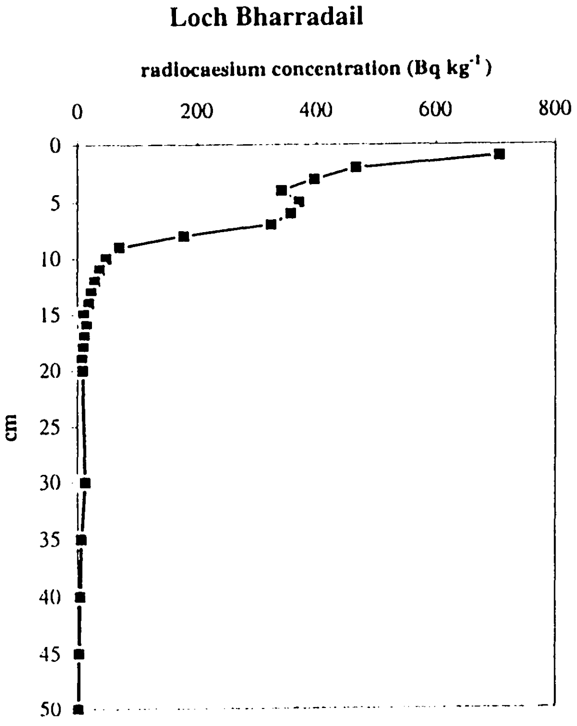


Figure 6.10 Depth versus ^{137}Cs activity.

6.6.2 Chronological support using ^{137}Cs

Radiocaesium in the Loch Bharradail sediment is detectable to 40 cm with a surface (0-1) ^{137}Cs maximum of 706.5 ± 9.2 Bq kg^{-1}. The ^{137}Cs concentration exhibits an exponential decrease from the surface with a subsidiary subsurface maximum in the 4-7 cm section. This may be attributable to the 1963 maximum in weapons testing fallout deposition, with higher overlying concentrations as a result of Chernobyl nuclear accident fallout deposition in 1986. Using the ^{210}Pb derived sedimentation rate, the sediment sections 3-5 cm fall between a date range of 1981-1974 which is 11 years later than the weapons testing maximum taken between the mid-fifties to 1963. The subsurface concentration (proposed weapons testing maximum) is 370 ± 5.8 Bq kg^{-1} and has undergone some diffusion down the profile. Recent research on Chernobyl derived ^{137}Cs distribution and mixing in Scottish freshwater loch sediment has highlighted the problems of its mobility in organic and clay-rich sediment (Bryant *et al* 1993). Diffusion could further be exacerbated by burrowing benthic animals such as *Chironomidae* (Bryant *et al* 1993). Elsewhere, Comans (1989, 367) reported that post-depositional processes in radiocaesium were probably caused by ion exchange with NH_4^+ which reaches high concentrations in anoxic porewaters.

6.6.3 Chronological support using pollen analyses

It was mentioned above that pollen analysis has proved useful as a means of chronological support in ^{210}Pb dated sediment profiles (see Chapter 3 Section 3.8.4). Limited pollen analyses were undertaken between sections 1-30 cm to establish the maximum concentration of the species *Pinus sylvestris* which is well established on the island both in estate plantations dating to the late 19th century and in more recent Forestry Commission plantations. Figure 6.2.3 shows a subsurface pine maximum of 8% at 4 cm. This position is dated to 1975 by the ^{210}Pb inventory. It is most probable that the pine maximum represents a regional origin as there is no pine growing within the catchment of the loch. It is therefore concluded that the pine curve is of little use in supporting the Loch Bharradail chronology. Based on the ^{210}Pb-derived sedimentation rate one would have expected the Pine pollen to begin to be concentrated at around between 30-26 cm equivalent to the years 1884-1900 when estate plantation pine was locally established. The apparent low frequency at this depth is probably due to a combination of taphonomic mechanisms (flotation to the loch margins) and climatic trends being the major influence in its dispersal. It may well be the case that pine pollen was extremely low in frequency after the plantations had become established and failed to register in the Bharradail sediment at this period in time due to the reasons given above.

6.6.4 Chronological support using radiocarbon

Four bulk samples from specific points within the Pb concentration profile were radiocarbon dated by the NERC Radiocarbon Dating Laboratory. Three samples were analysed by the Scottish Universities Research and Radiocarbon Dating Laboratory, East Kilbride, Scotland. The results of radiocarbon analyses are summarised in Table 6.3. and in Figure 6.11. Unfortunately both the radiocarbon dated sections and pollen data are of limited use as an independent support to the ^{210}Pb-derived chronology.

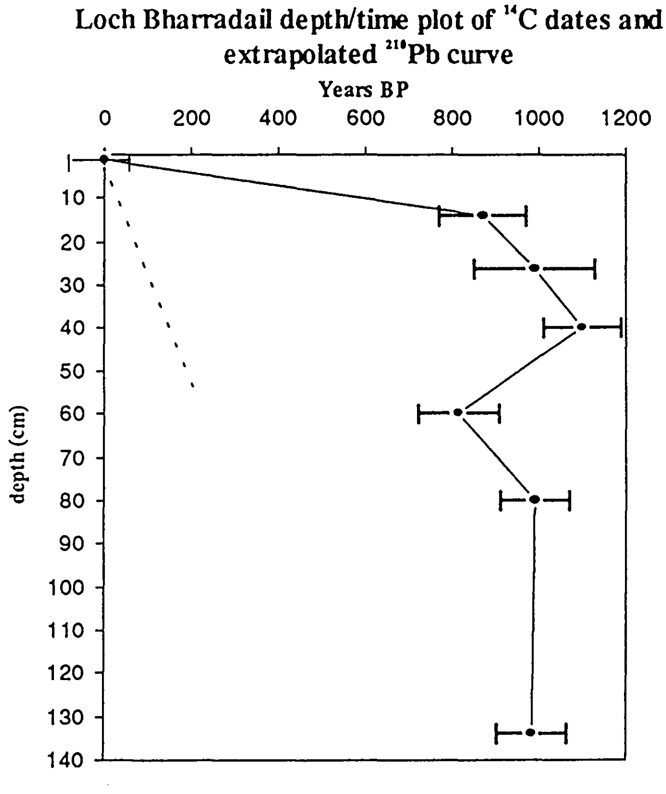


Figure 6.11. Depth/time plot of the seven radiocarbon dates on Bharradail Core 2. Error bars are placed at 2σ. The dotted line is the degree of slope based on ^{210}Pb-derived annual rate of sedimentation of 2.8 mm yr^{-1})

Figure 6.11 shows a depth/time plot of the radiocarbon dated bulk samples. These data show that there is no linear trend of increasing age with depth as one might have expected. Sample 4 (38-43) cm is significantly older at 1100 ± 45 BP than sample 3 positioned at 59-62 cm and dated to 805 ± 45 BP.

Sample	Lab. Code	Depth of sample	Age (years BP)	± 1 sigma	$\delta\,^{13}C$ (± 0.1%)
}7	GU-4219	1-5 cm	Post 1950	0.6%	-29.4
}6	GU-4209	12-16 cm	870	50	-28.6
}5	GU-4208	24-28 cm	990	70	-28.6
}4	SRR-5119	38-43 cm	1100	45	-28.5
}3	SRR-5120	59-62 cm	805	45	-29.2
}2	SRR-5121	79-82 cm	990	40	-29.0
}1	SRR-5122	132-135 cm	985	40	-28.2

Table 6.3 Summary of ^{14}C- results: Sample }5-7 were positioned to support the ^{210}Pb chronology. Sample }4 was positioned to date the maximum Pb concentration. Sample }3 was positioned to date the upper position of the minimum Pb concentration. Sample }2 was positioned to date the lower point of the Pb minimum concentration and }1 was positioned to date the base of the core.

6.6.5 Implications and discussion of the ^{14}C results

Dating lacustrine deposits by ^{14}C has been seen to be problematic (Jones *et al* 1985, Pennington 1991 and Entwistle *et al* 1995). The problem lies in the fact that sediments receive erosional inwash of catchment-derived material which may include old carbon. This invariably results in anomalously old ^{14}C dates. This has certainly been the case at Loch Bharradail. The depth/time curve has established that the ^{14}C dates are all within the same age range and suggest that there has been a major influx of material rich in old carbon. It can only be assumed that peat digging within the catchment has produced a high influx of mineral matter of the same age. Alternatively, the disturbance of the ground surface due to lead mining may have resulted in a greater influx of detrital material of the same relative age. The degree of slope established from the ^{210}Pb curve shows the disparity between these two dating methods. Although the ^{14}C dates to not conform to a trend of increasing age with depth they do suggest a return to steady state conditions possibly as a result of the cessation of mining in the early 19th century. This theory is based on the slope of the curve from sample 4 (38-43 cm depth) to near-surface sample 1 (1-5 cm).

6.7 Geochemical inventory and physical properties

The chemical influx data for Pb, Zn, Cu, Ca, Fe and Mn is shown in Figures 6.12-6.22. Descriptive statistics from the data set are shown in Table 6.4. The trace metal concentrations were determined using the procedure outlined in Chapter 3, Section 3.7.3. The following text summarises the metal concentrations and physical properties from the depth versus concentration profiles. Organic and mineral content, magnetic susceptibility and pH are shown in Figures 6.18-6.22. Elemental concentrations are expressed in mg kg^{-1} dry weight.

Iron has been converted to percentage concentration due to its high concentration within the replicate samples (e.g. 1% = 1000 mg kg^{-1}).

Statistics	Pb	Zn	Cu	Ca	Fe	Mn
Observations.	69	69	69	69	69	69
Maximum value*	925	530	31	175	26%	2563
Minimum value*	275	195	13	40	7%	538
Median value*	645	425	23	88	18%	888
Mean value*	621	404	22.1	92	18%	955
Standard Dev.	149	85.1	4.4	31.1	3791	313

Table 6.4 Summary statistics of the Core 2 data set (* = mg kg^{-1}).

Lead (Figure 6.12)

Starting from the base of the core at 134 cm, Pb is concentrated at 790 mg kg^{-1} and declines to 600 mg kg^{-1} at 120 cm. From this depth there is a rise in Pb values to 780 mg kg^{-1} at 108 cm. From this position the Pb values fall sharply to 440 mg kg^{-1} at a depth of 96 cm. From here Pb rises sharply to 700 mg kg^{-1} at 88 cm. It is from this point that another decline occurs with 275 mg kg^{-1} at 68 cm, forming the lowest concentration in the profile. From 66 cm to 48 cm the Pb concentration rises to a maximum of 925 mg kg^{-1}. The values fall slightly but remain at 640 mg kg^{-1} at the surface of the core.

Zinc (Figure 6.13)

The base of the core has a Zn concentration of 500 mg kg^{-1}. From here there is an intermittent decline in concentration to 66 cm where 240 mg kg^{-1} is attained. From this position there is an erratic rise to 520 mg kg^{-1} at 16 cm. From this here there is a slight reduction to 430 mg kg^{-1} at 1-2 cm.

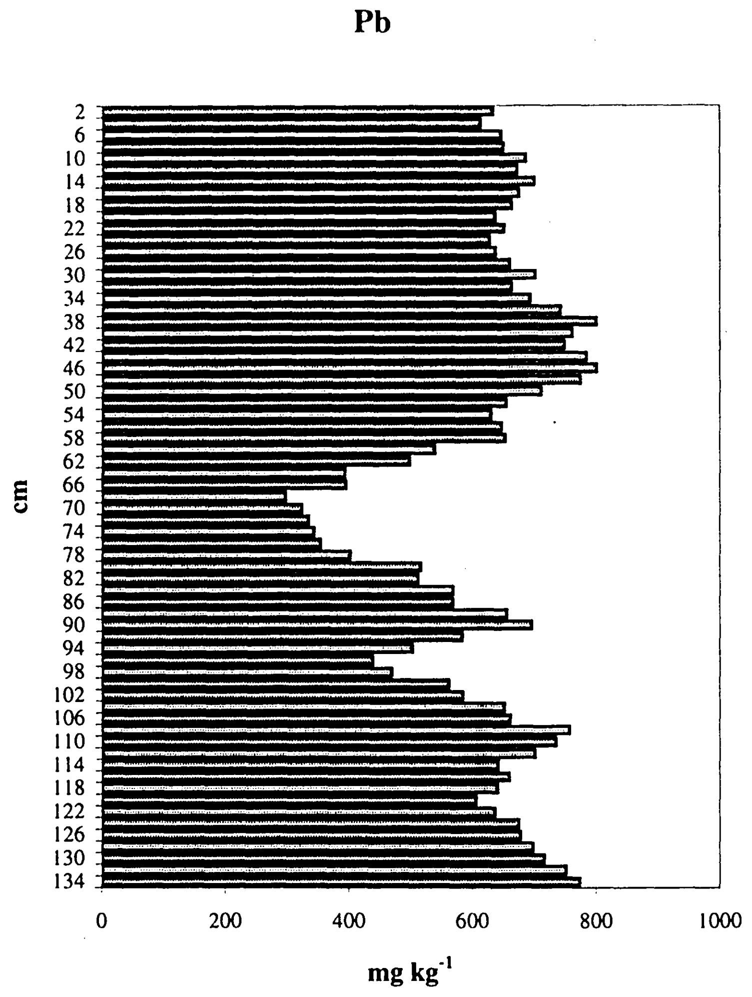


Figure 6.12 Depth versus lead concentration.

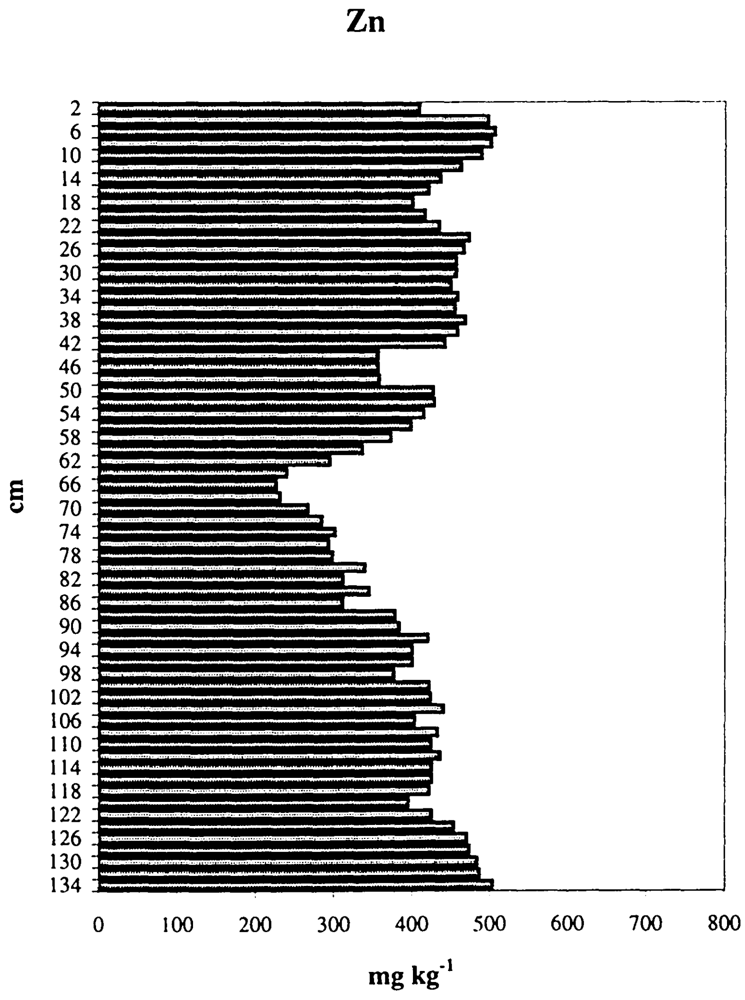
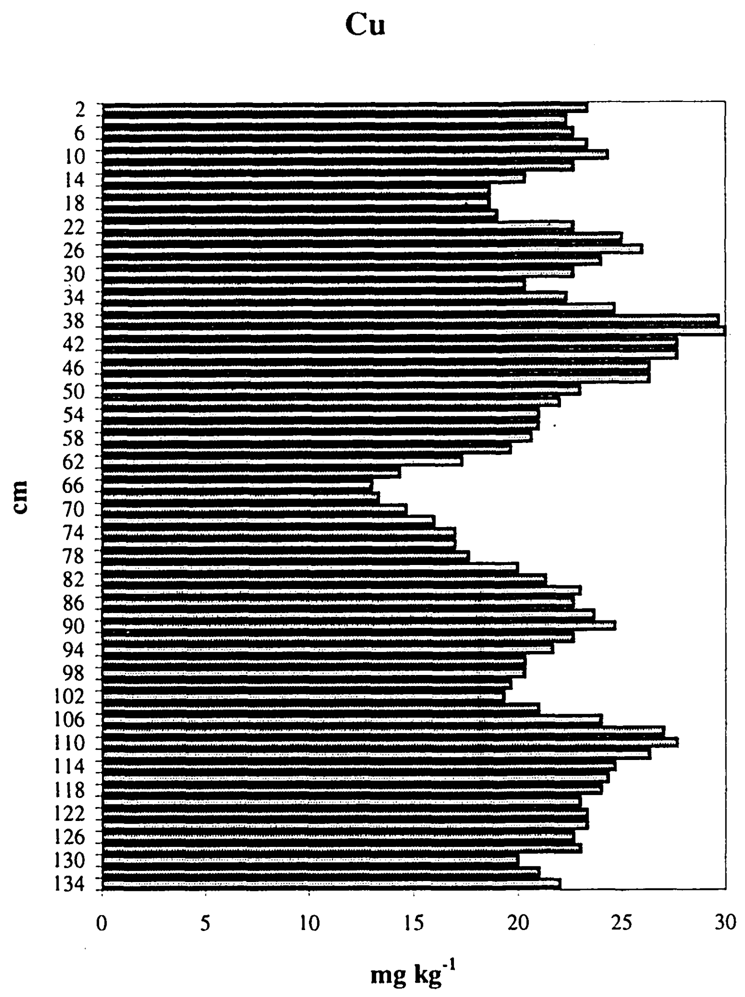
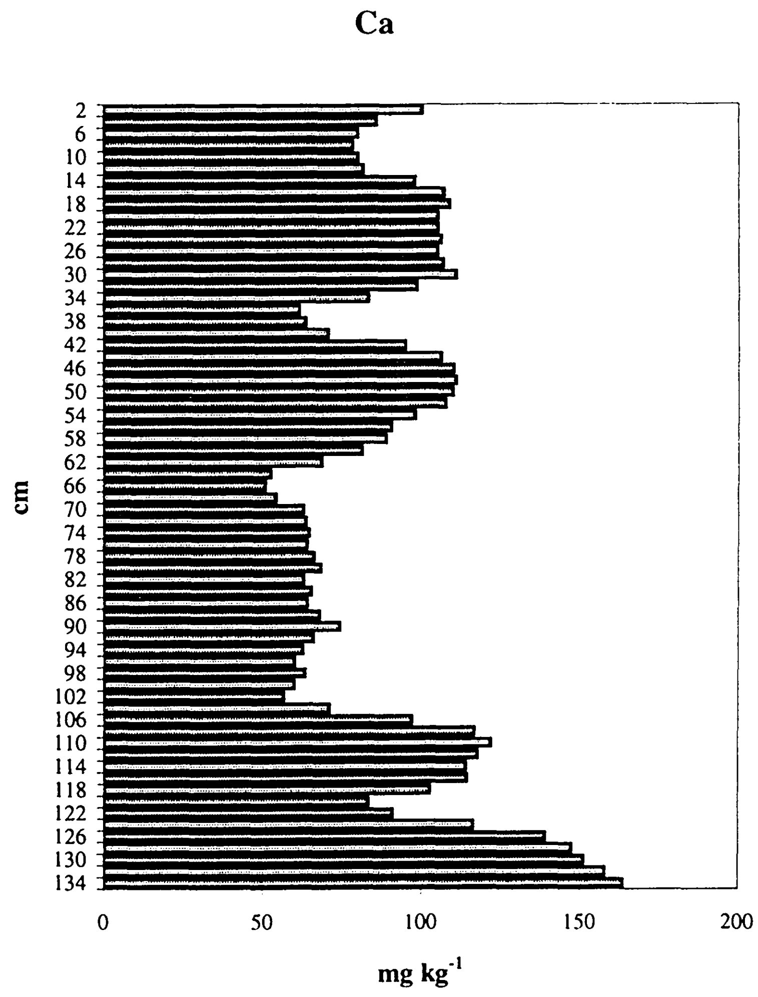


Figure 6.13 Depth versus zinc concentration



Cu

Figure 6.14 Depth versus copper concentration.



Ca

Figure 6.15 Depth versus calcium concentration.

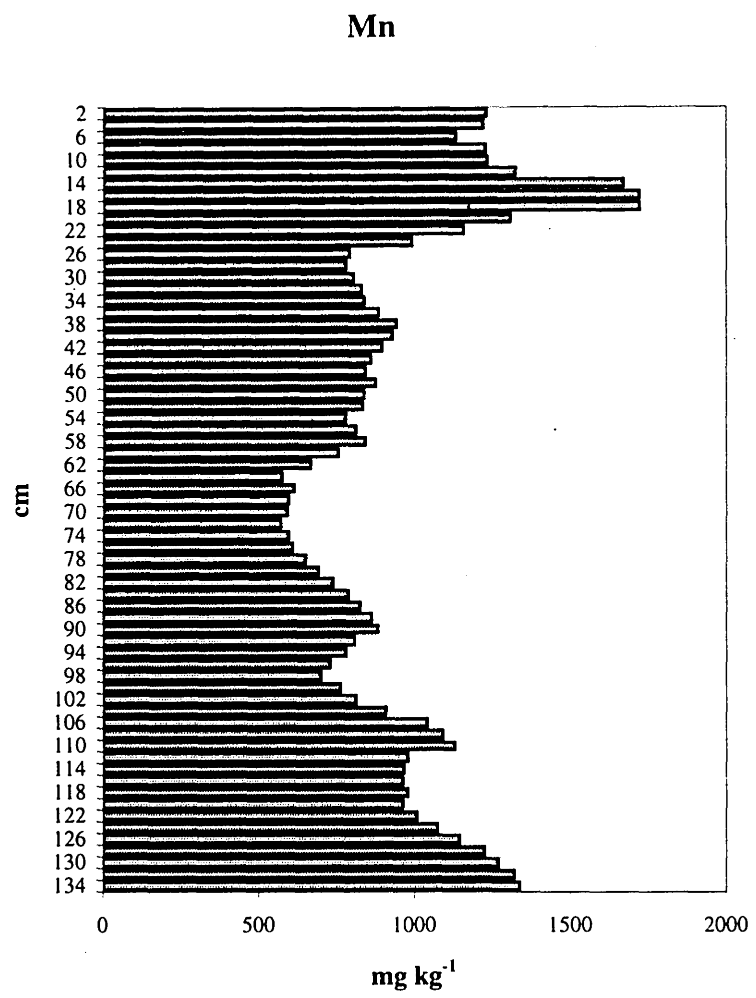


Figure 6.16 Depth versus manganese concentration

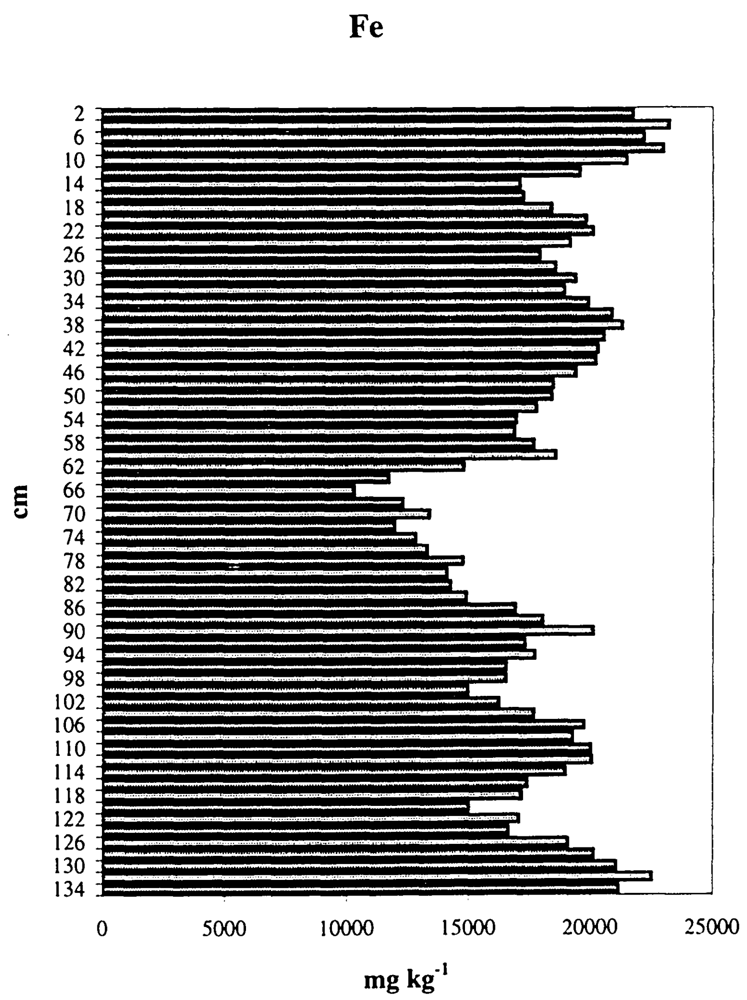


Figure 6.17 Depth versus iron concentration

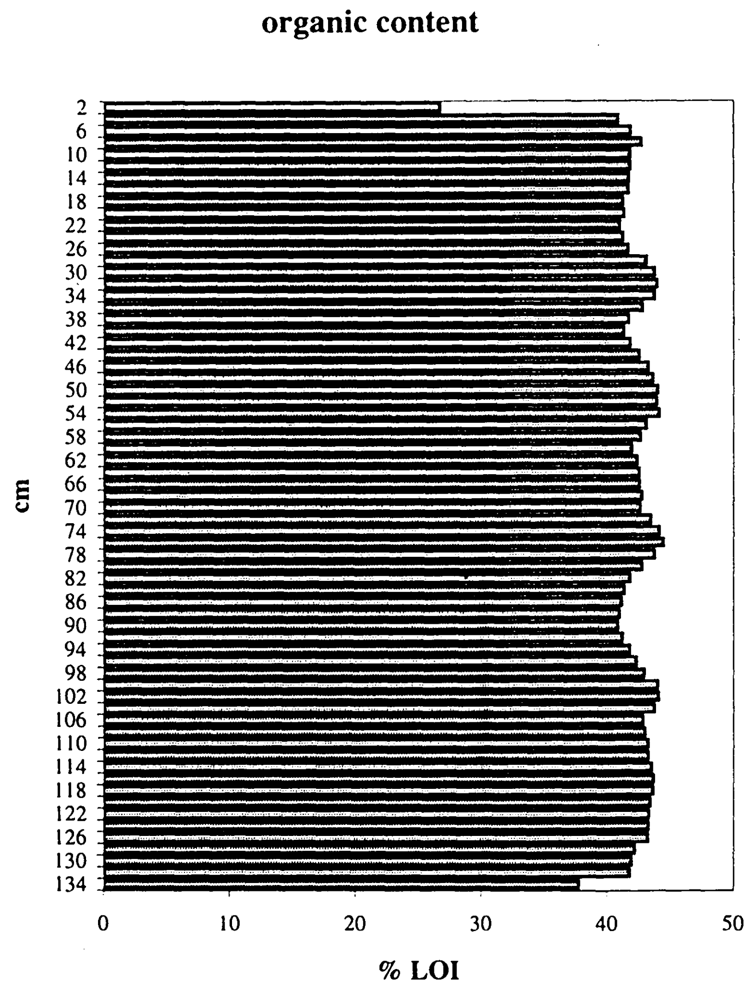


Figure 6.18 Depth versus % organic content based on loss on ignition (350 C)

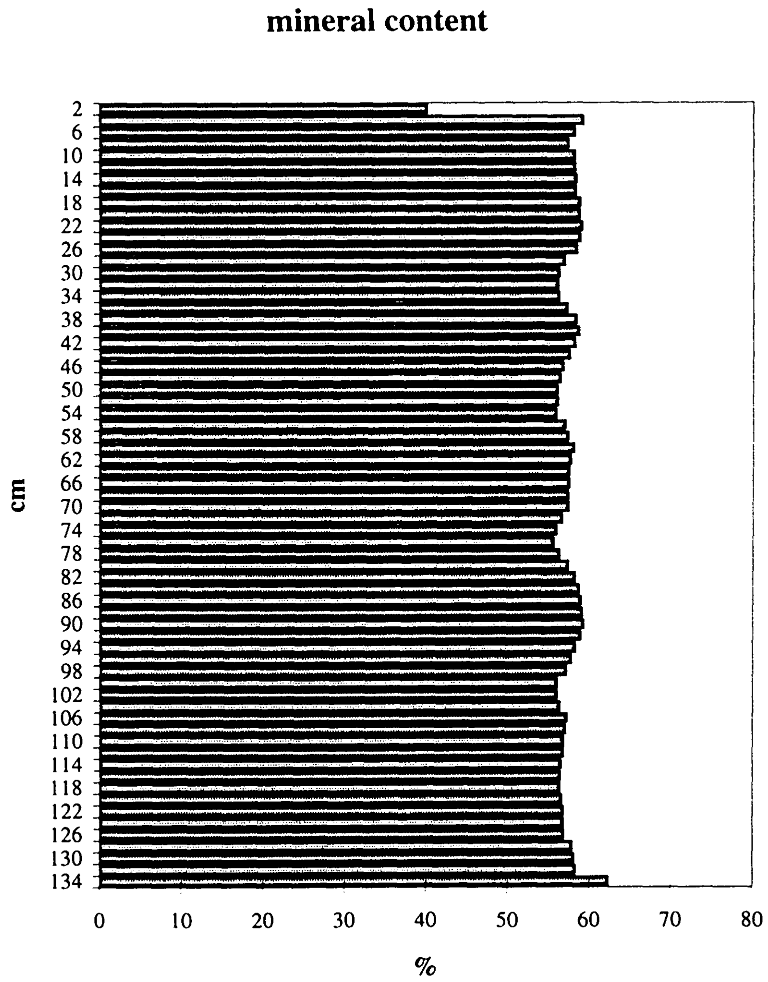


Figure 6.19 Depth versus % mineral content.

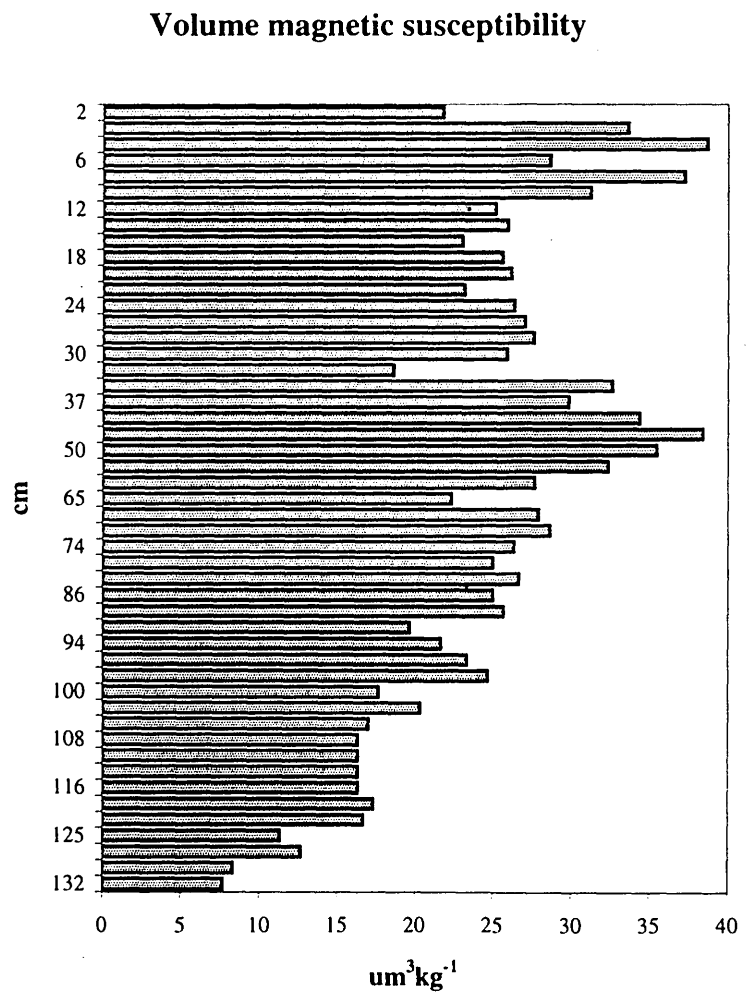


Figure 6.20 Depth versus volume magnetic susceptibility

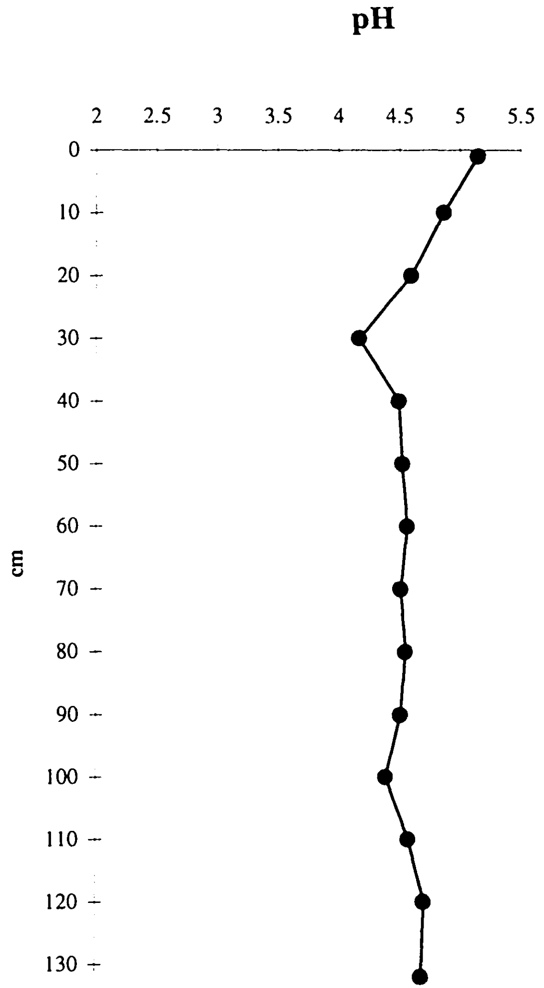
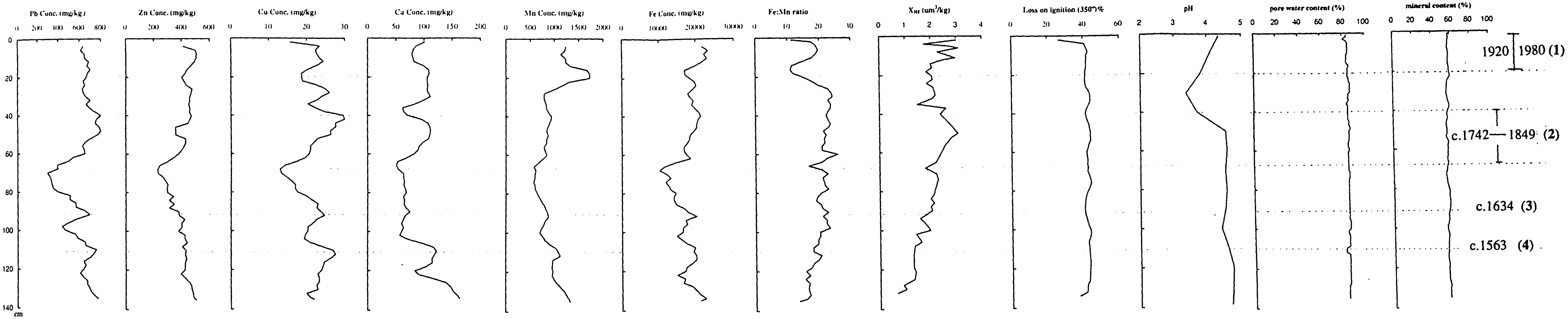


Figure 6.21 Depth versus pH.

Loch Bharradail Geochemical Summary Diagram



Extrapolated ^{210}Pb Date	Event in the profile	Interpretation
(1) c.1920-c.1980	Rise in near surface concentration of Zn Cu, Fe and magnetic susceptibility.	Effect of atmospheric input of Zn and Cu, both decline c.1980. Rise in susceptibility and Fe result of clearance of tailings for track repair.
(2) c.1742-1849	Increase in Pb, Zn, Cu, Ca, Mn, Fe, Fe:Mn ratio and magnetic susceptibility between c 70-40 cm.	Influence of mine-drainage waters. Spans the latest phase of operations at the mine. The leat constructed about c.1742. Peak period of lead production, high discharge of mine drainage into the loch.
(3) c.1634	Rise in Pb, Zn, Cu Fe and Ca.	Input of metals as a result of lead mining.
(4) c.1563	Rise in Pb, Cu, Ca, Fe.	Pb and Zn levels are high in concentration derived as a result of small scale lead extraction or other ground disturbance (ploughing ?)

Summary of possible dates and events within the Loch Bharradial geochemical diagram based on linear extrapolation of ^{210}Pb-derived sediment rate of 2.8 mm yr^{-1}.

Figure 6.22 Loch Bharradail geochemical summary diagram.

Copper (Figure 6.14)

At the base of the core the Cu concentration is 23 mg kg^{-1} rising slightly to 27 mg kg^{-1} at 110 cm. From here there is a sharp decline to 19 mg kg^{-1} at 102 cm rising to 25 mg kg^{-1} at 90 cm, soon after there is a rapid decline in Cu concentration to 14 mg kg^{-1} at 66 cm. From here, there is an exponential rise to 30 mg kg^{-1} at 38-40 cm. From this position the trend is erratic to a sub-surface value of 24 mg kg^{-1}.

Calcium (Figure 6.15)

From the base at 164 mg kg^{-1} there is a decline in Ca concentration to 84 mg kg^{-1} at 120 cm. From this point there is a small rise to 125 mg kg^{-1} at 110 cm. The values remain constant, ranging from 58-68 mg kg^{-1} between 104 and 66 cm. From this last point there is a substantial peak in concentration to 118 mg kg^{-1} at 50 cm falling soon after to 60 mg kg^{-1} at 36 cm. A rise to 115 at 32 cm is sustained to 18 cm where the values fall to 78 mg kg^{-1} at 12 cm, rising to a sub-surface peak at 116 mg kg^{-1} at 1-2 cm.

Manganese (Figure 6.17)

The base of the core has 1300 mg kg^{-1} Mn. From here an intermittent decline in values is seen to 112 cm with 1100 mg kg^{-1}. From 100 cm at 700 mg kg^{-1} there is a rise in values to 90 mg kg^{-1} at 90 cm. From here there is a decline to 64 cm with 600 mg kg^{-1}. From this position in the core there is a rise to 30 cm where the Mn concentration peaks to a maximum of 1700 mg kg^{-1} falling to a sub-surface value of 1200 mg kg^{-1}.

Iron (Figure 6.18)

At the base of the core the Fe concentration is 2.3% and falls to 1.5% at 120 cm. At 112 cm the Fe values rises to 2%. There is a decline in concentration to 1.3% at 98 cm with an increase to 2% at 90 cm. From this position there is a decline to 1% at 64 cm. From here there is a sharp but erratic rise to 2.3% at 36 cm. Values fall to 1.56% at 16 cm with a subsurface increase of 2.4%.

Iron:Manganese ratio (Figure 6.22)

The Fe:Mn ratio shows an intermittent increase from the base where 15 is attained. A maximum value of 26 is attained at 60 cm depth. From this position this value is constant until 40 cm where it declines to 12 at 20 cm depth. The values increase with a subsurface peak of 20.

Water Content (Figure 6.22)

Water content is estimated from the wet/dry ratio after oven drying providing an estimate of the pore water content within each sediment section. From the base of the core to 58 cm the water content ranges from 84-86%. There is a peak of 86% at 48 cm. A maximum of 87% occurs at 26 cm with an erratic decline thereafter to a sub-surface value of 83%.

Organic matter based on loss on ignition (Figure 6.18)

The organic matter is fairly constant throughout the profile with values ranging between 41-44%.

Mineral content (Figure 6.19)

The mineral content remains constant throughout the profile at around 58%.

Volume magnetic susceptibility (Figure 6.20)

Volume susceptibility shows a steady increase in concentration from the base of the core with 1.2 $\mu m^3\ kg^{-1}$ units to 42 cm where a subsidiary maximum of 3 $\mu m^3\ kg^{-1}$ units is attained. Values decrease intermittently to rise at a near-surface peak of 3 units $\mu m^3\ kg^{-1}$.

pH (Figure 6.21)

pH values are established on 10 gr of dry sediment at 10 cm intervals down the core. The method gives only a crude estimate of variance down the profile. From the base of the core to 15 cm there is limited variability. From this position there is a decline from 4.5 to 3.2 pH units. From 15 cm there is a steady increase towards the surface of the core to 4.3.

6.7.1 Implications of the geochemical inventory

A number of patterns emerge on examination of the chemical profiles. The first point to mention is that none of the metals are near the 'baseline' or X axis in the concentration versus depth profiles. This shows that between 0-135 cm there has been chemical enrichment in all six metals throughout the profile. Secondly, there are remarkable similarities in the concentration patterns of Pb, Zn Cu and Mn. The summary diagram of all the metals examined shows that between *c.*64 -70 cm depth there is synchroneity across the Pb, Zn, Cu, Ca, Mn, Fe, Fe:Mn ratio and magnetic susceptibility. At this position Pb declines to 290 mg kg^{-1} and this value can be assumed to be the nearest one can achieve to a 'near-baseline' value in the absence of deeper sediment. Before the decrease in Pb concentration there are two smaller peaks occurring before 64-66 cm and noticeably at between 86-90 and 108-112 cm.

At this last position this trend is also shared in the Cu, Ca, Mn, and Fe concentration profiles. Manganese appears to have undergone some near-surface enrichment. Water and organic content are relatively uniform in distribution confirming the homogenous consistency observed throughout the profile. There appears to be some conformity between the peak in the volume magnetic susceptibility profile and the total Pb profile at *c*.50 cm. In comparison to the susceptibility profile from Loch Leathann, the Bharradail values are higher than one might have expected. This is due to the higher input of ferrogenic magnetic material entering the loch. This is accounted for by generally high Fe values (compared to Loch Leathann) and Loch Bharradail's significantly higher loch:catchment ratio. (see Tables 6.1 and 8.1). The Fe:Mn ratio mirrors the Fe profile from *c*.70 cm upwards and shows good synchroneity with the Zn and Cu profile at 20 cm.

The pH values obtained for Loch Bharradail show that from the base to 40 cm depth, conditions have been relatively acidic which is not surprising given the catchment soils and the influx of acidic-mine waters. However the change to a higher pH towards the surface of the profile is very interesting. We might be observing a cessation or at least a decline in acid-mine input which is being off-set by less acidic drainage waters into the loch. Today the loch is highly eutrophic and it is assumed from the pH trends that this condition has prevailed in the past and will to a large extent have been exacerbated by mine-water drainage. This water may well have been of a higher pH than surface water run-off and certainly higher than peat-bed drainage waters. Time did not permit the examination of the diatom spectrum which is more susceptible to internal changes and is a more reliable indicator of nutrient and pH fluctuations over time. Finally, the integrity of the core appears to be sound in terms of bioturbation or mechanical mixing based on the excess ^{210}Pb activity profile which shows a good linear exponential decay pattern.

The Loch Bharradail core has a maximum concentration of 925 mg kg^{-1} Pb at 48 cm. This is considerably higher (720 mg kg^{-1}) than the surface maximum of 205 mg kg^{-1} Pb (1-2 cm) attained in the control loch (Chapter 8 Figure 8.10). In the absence of mineralisation in the catchment of Loch Leathann it is suggested that the observed near-surface Pb maximum is attributed to atmospheric fall-out deposition (see Chapter 9 section 9.4.4).

In sediment profiles, Zn is commonly associated with Pb and Cu as major contaminants from industrial pollution and base metal mining (See section 9.4.4 below). In the aquatic environment Zn primarily resides in detrital oxides and feromagnesesian silicates, which weather slowly in most environments (Jones and Bowser 1978). In the immediate vicinity of sulphide deposits, oxidation weathering may promote high concentrations (>100 ug/l) of dissolved Zn sulphates and carbonates (BGS citing Ure and Berrow, 1982). Acid mine water can locally accumulate Zn up to the percent range of concentration. The depth v concentration profile for Zn in the Bharradail core shows a decline from the base up towards 64 cm. This is in keeping with Pb, Cu, Ca, Fe and Mn. In the absence of deeper sediment the Zn background is established as 230 mg kg^{-1}. This value is obtained from the lowest range in concentration between 64-70 cm. The British Geological survey established moderately high values (300-800 ppm) over the Dalradian Limestone region of north-east Islay. Several anomalous Zn values (>1000 ppm) at Mulreesh were attributed to enrichment from waters emanating from the mine.

Interestingly, high Zn levels were recorded within the Colonsay Group Formation around *Cnoc Dubh* (Grid Reference NR445570). There is no mineralisation within this region and the high Zn levels attained were attributed to the numerous north-west trending dolerite dykes in this area. In the case of the Bharradail core, the combined effects of mining and the presence of the Dalradian limestone has led to relatively high concentrations of this metal. Sphallerite is associated as a secondary mineral in association with galena. The XRD data (Appendix 3) shows that this mineral is present within the mine tailings at the South Ardachie mine.

With respect to Cu, relatively high Cu concentrations in mine and natural waters arise in areas where sulphide deposits are exposed at the soil surface (Chapter 5 Figure 5.12). Abnormally high concentrations may occur as aureoles in country rocks and organic matter. Collins (1973) found that gradients in declining concentration occurred with distance downstream from a copper mine tailings pond. Dilution of the metal was ruled out in favour of sorption by iron hydroxides present as sediment coatings and complexed with organic matter. The profile for Cu follows the trend of Pb with two peaks in concentration rising from the base of the core then falling to a 'nearest to baseline' value of about 15 mg kg^{-1}. It is surprising given the presence of so much mine waste within the catchment that these values are not significantly higher. Copper is usually a guest mineral with galena in the form of the sulphide copper mineral, chalcopyrite. The British Geological Survey recorded low levels of Cu in the Dalradian limestone of north-east Islay (5-20 ppm). Average enrichment was noted as 30 mg kg^{-1} Cu. Copper has a strong affinity with organic matter and is readily absorbed (Rippey, 1982; Williams 1992). According to Robb (1994, 35) the weathering of pyrite and the production of acid mine waters may readily dissolve Cu and Al.

The following equation relates the process of dissolution in an oxidising environment,

$$CuFeS_2 + 2H_2SO_4 \Leftrightarrow CuSO_4 + FeSO_4 + 2H_2S$$

and $H_2S + 4O_2 \Leftrightarrow 2H_2SO_4$

to give

$$FeCuS_2 + 4O_2 \Leftrightarrow FeSO_4 + CuSO_2$$

It is the generation of sulphuric acid $2H_2SO_4$ that readily attacks the more alkaline secondary 'guest' minerals within the waste ore. In the case of Loch Bharradail, the sedimentary concentration of copper is likely to form a gradient, becoming weaker away from the primary input (e.g. the mouth of the leat) towards the centre of the loch. A similar situation has been recorded elsewhere. Davison *et al* (1985) examined the sedimentary records of copper mining activity in Coniston Water in the English Lake District. Here Davison and co-workers recorded high concentrations of copper in the solid form (max. concentration 10 mg g^{-1}) of calcopyrite which was derived from a single point source, namely the ore washing processes within the catchment.

Moving on to the Ca profile the distribution of this element has a maximum concentration at the base of the core. The values fall sharply with a slight fluctuation before falling to between 60-56 mg kg^{-1}. There is an erratic increase towards the upper regions of the profile. X-ray diffraction identified the presence of calcite ($CaCO_3$) and dolomite $CaMg\ (CO_3)_2$ as the major gangue minerals associated with Galena in the grab sample from the South Ardachie mine tailings (Table 3 Appendix 6). Given the relative ease which these gangue minerals weather in acid environments in theory one would have expected a higher concentration of Ca within the sediment profile. Kemp *et al* (1976) noted that low carbonate concentrations in cores from Lake Superior, USA were indicative of a slow rate of sedimentation and *in situ* dissolution of detrital carbonates. In the case of the Bharradail sediment the presence of Dalradian limestone within the catchment is the primary source of this carbonate element. Calcite could not be identified using X-ray diffraction analyses within the Bharradail sediment. Concentrations need to be in the 1% (e.g. equivalent to 1,000 ppm) range to be detected by this technique.

Manganese is concentrated at the base and towards the near-surface of the Loch Bharradail core. Upward migration of soluble Mn under reducing conditions in sediment pore waters leading to surface precipitation by oxidation is well known (e.g. Mackereth 1966 and Weiler 1973). This major element is commonly seen to be another relatively mobile metal in freshwater sediment. In sedimentary rocks the Mn content is controlled by the geochemistry of the source rock and the redox conditions of the local depositional environment. Manganese tends to be enriched in shales and greywackes (ca 700 ppm) relative to coarser quartzitic sandstones and grits (ca 170 ppm) In the fluvial environment, Mn enters stream sediments in a variety of solid phases that include ferromagnesian silicates, Fe-Ti oxides and adsorbed coatings on clay minerals (Kelts and Hsü 1979). Under acid conditions the dissolution of Mn is rapid relative to alkaline conditions. In solution Mn is governed by prevailing redox conditions. Most of the Mn which reaches an anoxic sediment is rapidly reduced and released to the overlying water, notably by the replacement from insoluble Mn^{4+} to the soluble Mn^{2+} (Davison 1982).

Mine sites and peaty areas with impeded drainage provide stream waters with high levels of manganese (>100 mg L^{-1}) and rapid re-precipitation occurs in oxygenated waters. This mechanism is seen to be instrumental in scavenging trace elements such as As, Ba, Pb, Co, Cu and Zn, (Williams 1992). In the case of the Bharradail core the highest concentration can be observed towards the upper-most region of the core and is assumed to happen under the control of near surface redox conditions.

Moving now to factors controlling Fe, the concentration profile in the Bharradail core exhibits large concentrations at the base (22%) falling erratically to the 70-64 cm region (12%) then rises to the surface (23%). The processes that have led to high concentrations within the profile may be due in part to the high input of ferric hydroxide resulting from the weathering of pyrite, FeS_2. This mineral is widespread in the spoil dumps (Plate 4, Appendix 1) and occurs as rhomboid inclusions in the local slate. Pyrite has been seen to be one of the most easily weathered products on mine spoil. Davies (1983, 434) mentions that pyrite is a common, but valueless component occurring in sulphide ores which is often highly concentrated in mine tailings. In a wet environment pyrite oxidizes in the reaction that can be summarised:

$$S_2 + 14H_2O + 15O_2 = 8H_2SO_4 + 4Fe(OH)_3$$

As a result of this, very acidic conditions occur, notably with the generation of sulphuric acid which in oxidising conditions, readily breaks down silicates and other secondary materials leading to the evolution of extremely acidic leachates (<2 pH) that can readily enter the fluvial environment. The Fe^{2+} ion is highly soluble resulting in high dissolved Fe loading in the streams draining peat lands or other acid and reducing terrain. The latter component of the equation (ferric hydroxide) is often observed as forming a red-brown 'ochre' that stains stones and vegetation within drainage channels leading from abandoned mining areas.

It is debatable to what extent the influence of 19th century peat extraction has had concerning the Fe levels within the Bharradail sediment. It is likely that much of what is observed within the sediment will be a result of increased loading in ferrous-rich materials emanating from the poorly drained, podsol regions in the catchment. The acidification of soil enhances the rate of podsolization and consequently the release of many metals which are otherwise rather less immobile. Low pH alters soil microbial ecology, which in turn will influence biogeochemical processes in soils, included related metal cycles (Salomons and Forstner 1984).

6.8 Conclusions

In the absence of documentary evidence before the late 18th century the palaeoenvironmental record contained within the loch basin provides the only available avenue for understanding longer-term evidence of lead mining at South Ardachie. Assuming 'steady-state' conditions and supposing that sediment compaction has been negligible then on the basis of the ^{210}Pb-derived chronology one can infer that the loch has received a significant input of Pb and Zn for at least the last 500 years. This raises the question as to whether lead was being extracted at this early period from within the near vicinity of the loch. It was mentioned earlier that the *Accounts of the Lord High Treasurer of Scotland* described lead being transported from Islay between 1511-12 A.D. Using the sedimentation rate of 2.8 mm yr^{-1} this period falls between 67-68 cm depth in the profile. Paradoxically, this period falls within the sediment sections at 64-70 cm depth that display the lowest concentration of Pb within the core.

We cannot be sure which mine was producing lead at this period (1511-12) but the mine at South Ardachie has by far the most extensive evidence of 'early' mining based on field survey. Within 70-135 cm of the core Pb concentrations range between 300-750 mg kg^{-1} in comparison to the Loch Leathann control core that has a background value of 30 mg kg^{-1} Pb. In all probability it would appear that 'small-scale' lead extraction was being undertaken at least before the beginning of the 16th century as lead mining must account for the high values in Pb and Zn. We know from the historical accounts summarised in Section 6.3.1 above, that the mine was active in 1770 which based on ^{210}Pb-derived annual rate of sedimentation correlates with the 60 cm sample within the sediment. At this position the Pb values rise dramatically towards the 40 cm depth position (*c.*1849). We can not be certain when this mine ceased production but it is estimated to be at or slightly later than the turn of the 18-19th century. It is at this period that deeper lead deposits were beginning to be exploited at Mulreesh. In summary it can be seen that there is good conformity with the Pb, Zn and Cu profiles covering the peak period of lead production at South Ardachie (*c.*1750-1840). Two subsidiary peaks are also attributed to earlier phases of mining dating to *c.*1634 and *c.*1563 respectively.

Extrapolated ^{210}Pb Date	Event in the profile	Interpretation
(1) *c.*1920-*c.*1980	Rise in near surface concentration of Zn Cu, Fe and magnetic susceptibility	Effect of atmospheric input of Zn and Cu, both decline *c.*1980. Rise in susceptibility and Fe result of clearance of tailings for track repair
(2) *c.*1742-1849 AD	Increase in Pb, Zn, Cu, Ca, Mn, Fe, Fe:Mn ratio and magnetic susceptibility between c 70-40 cm.	Influence of mine-drainage waters. Spans the latest phase of operations at the mine. Alexander Shiriff's (1770) recognition of 'earlier workings. Leat constructed about 1742?.
(3) *c.*1634 AD	Rise in Pb, Zn, Cu Fe and Ca	Input of metals as a result of lead mining .
(4) *c.*1563 AD	Rise in Pb, Cu, Ca, Fe	Pb and Zn levels are high in concentration derived as a result of small scale lead extraction or other ground disturbance (ploughing ?).

Table 6.5 Summary of possible dates and events within the Loch Bharradail sediment profile based on linear extrapolation of ^{210}Pb-derived sediment rate of 2.8 mm yr^{-1}. Note events (1-4) are shown on the summary diagram, Figure 6.22.

Chemical analyses of the Loch Bharradail core allows assumptions to be made on the origin of metal-rich solutes observed within the sedimentary profile. The principal routes of metal-rich material entering the loch have been established from evidence obtained from archaeological field survey. The principal route for the main input of solutes containing high Pb-rich material would have been along the 'leat' (Plate 12 Appendix 1) and along other small burns that drain into the loch from within the mining region. Included will be acidic leachates seeping from the spoil heaps as surface run-off.

There are other minor routes, including seepage-anomalies that flow from the limestone escarpment and out of the old peat cuttings. Constant erosion of the loch shoreline is another route of metal influx into sediment. After the abandonment of the mine, the subsequent clearance of old spoil tips over the last century may well have increased the heavy metal burden in the loch sediment. Importantly atmospheric fall-out deposition of Pb is an additional component within the near-surface of the core.

The ^{210}Pb-derived sediment rate of 2.8 mm yr^{-1} and the supporting ^{137}Cs has allowed a fairly accurate time frame to be established for the upper region of the core. It is unfortunate that the results of the radiocarbon analyses do not provide meaningful results that lend additional support to the ^{210}Pb-derived chronology. The wider implications of this problem are discussed in detail in Chapter 9 Section 9.6. Investigation and analyses of sediment recovered from Loch Lossit form the basis of the following chapter.

CHAPTER 7: LOCH LOSSIT

7.1 Introduction

Loch Lossit was chosen for investigation on the grounds of there being good evidence for lead mining within the catchment although this is on a much smaller scale compared to the mines in the catchment of Loch Finlaggan and Loch Bharradail. The close proximity of the mining remains with Loch Lossit (located 500 m from the loch) appears to have increased the sedimentary flux of Pb, Zn and Cu. Documentary sources show the mine to have been in operation no later than the mid 18th century. A drainage adit at the mine has possibly been the principal source of contamination into the loch at this period. Secondary contamination from natural sources and recent land use changes in the catchment have also been recorded. In keeping with other regions of known mining activity in NE Islay, the surface evidence resulting from early mining episodes within the limestone region immediately south of the loch have been lost due to the clearance of tailings, back-filling and land improvement practices over the centuries. However, subsequent 'clean-up' operations over the years by farmers may have led to a wider distribution of contaminants in surface soils about these former sites.

In order to distinguish between natural and anthropogenic origins chemical data from the sediment sections within the core a good understanding of the local environment within the catchment needs to be obtained. Consultation of soil and solid geology sheets of the area have provided an understanding of soil types and geological formations that are likely to influence the soil geochemistry and lead to naturally occurring soil contamination. As a means of additional support and towards a better understanding the local geochemical environment within the Lossit catchment, the results of DRL survey have been computed to show how mine-derived Pb, Zn and Cu are distributed in nearby surface soils. These data also show that there is also natural contamination within the immediate area of the loch providing a data set to complement the chemical inventories from the loch sediment.

Chronological control has been established using only the ^{210}Pb and ^{137}Cs inventories. Supporting radiocarbon dating could not be undertaken owing to the close proximity of the Dalradian Limestone. This could have effected the outcome in radiocarbon determination. Older or in some cases younger dates are derived as a result of 'hard-water error' or as we have seen, by the input of 'old' carbon. Only limited pollen analyses was undertaken in order to allow additional support to the ^{210}Pb-derived chronology but the results from this programe of analyses appears to be of little value as local *Pinus* sp. appears to have entered the sediment in only small amounts due to its flotation to the loch margins. The results of the bathymetric survey have been rewarding and the data obtained from this survey provide an insight into the previously unrecorded geomorphology of the loch. It is concluded that there may be a direct response to Pb mining observed by an elevation in Pb concentration. A near-surface concentration in all metals can be observed and this is attributed to an increase in sedimentary flux resulting from recent land-use changes and atmospheric pollution.

7.2 Location (Figure 7.1)

Loch Lossit, is situated 1 km to the south-east of the village of Ballygrant and is one of the larger freshwater lochs on the Dunlossit Estate (Plate 14, Appendix 1). The loch is irregular in shape with three small bays situated at the east, west and southern shore. The maximum depth of the loch was recorded at 50 feet (15.24 m). The pH of the water was established as 7.5 with a conductivity of 180 μ holms. The loch lies at an altitude of 110 m (Table 7.1) surrounded by steep hill-slopes that rise sharply on its south-west and south-eastern sides rising to a height of 146 m OD. on *Beannan Dubh*. The loch is fed by five small burns and the main outlet is a man-made leat situated to the north of the loch which eventually flows into Loch Ballygrant. The substrate of the shorelines is predominantly strewn with large boulders and elsewhere with fine shingle. Stretches of silt are located at the southern end of the loch giving way to marsh.

Grid reference	NR 408654
Core date	September 1992
Catchment geology	limestone-phyllites-shales
Catchment type	pasture-woodland-forest
Loch altitude (m)	110
Max. depth (m)	15.24
Loch area (ha)	18
Catchment area*(ha)	136
Catchment:loch ratio	7.1
Afforestation (%)	28
Net relief (m)	146

Table 7.1 Loch Lossit site characteristics (note* excluding loch)

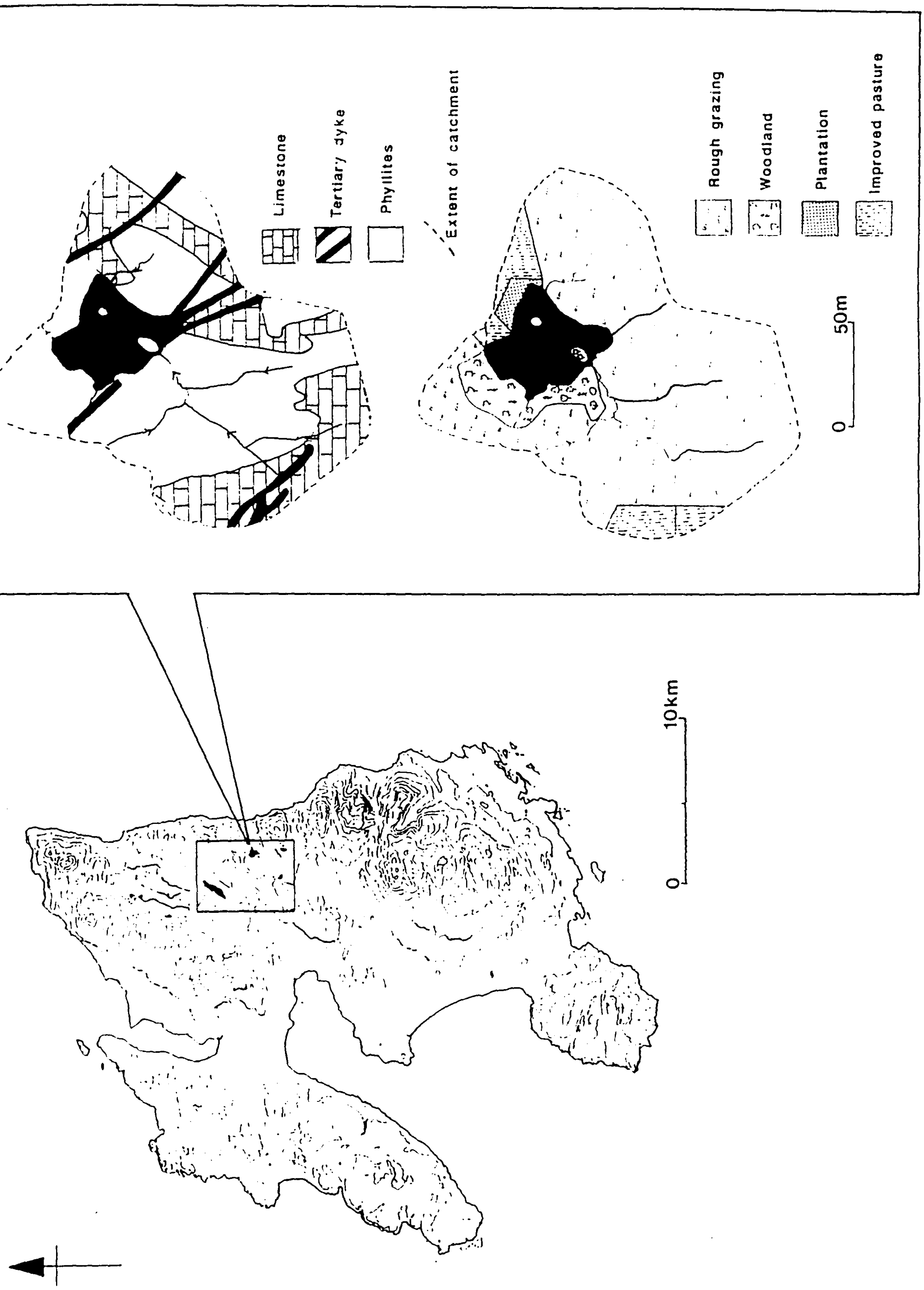


Figure 7.1 Location of Loch Lossit with geology and present day land use.

7.2.1 Geology (Figure 7.1)

Loch Lossit is bounded on its north-western side by phyllites and acidic shales. Dalradian Limestone outcrops on its southern and north-eastern shore. Tertiary activity is represented by three north-west trending olivine-dolerite dykes that terminate in the south-east corner of the loch. One of the dykes outcrops on the north-east shore. In general the topography surrounding the loch is hummocky with fluvio-glacial drift deposits that have given rise to impeded drainage and consequent podsol formation.

7.2.2 Soils, vegetation and catchment landuse (Figure 7.1)

The soil types surrounding the loch arise as a result of underlying parent geology. Two principal soil types are present, namely the Deecastle and Foundland Series. The former comprises drifts derived from Dalradian limestone forming brown-forest soils and rankers. Rich bent-*fescue* grassland provides good grazing, most of which is improved pasture with mainly sheep and cattle. The Foundland Series is predominantly acidic, derived from the weathering of slate, phyllites and other weakly metamorphosed argillaceous rocks. The Foundland soil supports only the moist Atlantic heather moor communities, and hence this land is given over to rough grazing. Podsol formations are common on the limestone regions where there is a tendency towards waterlogging resulting from poor drainage.

Close to the eastern shoreline the loch has been planted with mixed deciduous woodland species including *Pinus* sp., *Larix* sp., *Fraxinus* sp. and understory *Salix* sp. with *Alnus* sp. On the western side of the loch there is a small Sitka Spruce plantation. The islands in the loch have been planted in parts with *Larix* sp. but pioneering *Fraxinus* sp. are well established. The emergent vegetation around the north-eastern part of loch consists of large tracts of *Phragmites australis*, *Scirpus lacustris*, and *Carex rostrata*. The loch supports a healthy population of brown trout and forms the main water supply for the nearby village of Ballygrant. No out-board engines are permitted o the loch, a tradition maintained within the Islay and Dunlossit Estates.

7.2.3 Lead mining history

Lead mining occurred in the 18th century at the mine known in this period as Glasco Beag (derived from *Glasgow Beg* or Little Glasgow) now within the land holding of Knocklaroch. Glasgow Beg lay on the southern part of the Ardochy land holding (NR409643) and was also probably part of the holding of the North Ardachie lead mine.

A furnace and tool shed are attributed to Glasco Beag (Ramsay 1991, 123) but no physical evidence survives of these features in the landscape today. The mining region is shown on the 1882 1st Edition Ordnance Survey map. The *Book of Islay* (Smith 1895,458) relates how in 1745 owing to disputes over debts between various financiers of the lead mining industry ,the smelt mill (precise location not known) was impounded and the tools stored at the mine of Little Glasgo. We have no indication how long this concern was in operation as no records of output survive. Today the surface evidence of mining is very scant indeed and where fragments of the former works survive, these are confined to a limestone spur approximately 500 m to the south of Loch Lossit. Here a small drift cuts through the edge of a ridge to form a series of open-cast cuts. Two hundred metres north of the small drift there is a small rubbish filled shaft with tailings in the near vicinity. There are what appear to be small open casts nearby. The site has been extensively cleared of mine tailings for track building and repair and it is difficult to determine the extent of former mining evidence from the natural surface outcrops of limestone that undulate across this area. Numerous small burns traverse the mining region and lead in the general direction of the loch. In view of the poor nature of the evidence and the sparse distribution of mine tailings no topographical survey was undertaken in this region.

7.3 Geochemical Environment

7.3.1 The catchment distribution of Pb, Zn and Cu based on the DRL geochemical survey

The chemical data obtained from the DRL survey has been computed into two-dimensional contour maps as discussed in Chapter 3 Section 3.3.2. Table 7.2 provides descriptive statistics of the data used to produce the contour plots. The highest concentration values observed in the DRL survey were omitted from the primary input data that produced the distribution maps as there is a tendency by the software to suppress the lower class intervals when the data is interpolated.

Data smoothing parameters have not been applied as these tend to soften the contours leading to loss of definition between the mid-range class intervals. The highest values (outlying anomalies) from the DRL soil transects fall within the 'above' category in metal concentration (parts per million) and the maximum recorded value for either Pb, Zn or Cu has been placed manually on the diagram. The plot title 'catchment distribution' is used in this case to mean land adjacent to or surrounding the loch. In real terms 'catchment' would include a much wider area than that shown in the isopleth plots. The plots show the range and distribution of Pb, Zn and Cu in the vicinity of Loch Lossit and provide an objective representation of the previously hand-plotted DRL data. Additional reference has also been made to the recently published geochemical survey by the British Geological Survey's *Regional Geochemistry of Southern Scotland and part of Northern England 1993*, hereafter referred to as GSS (Geochemistry of Southern Scotland). A national hydrogeochemical survey was undertaken on a 1 sq km basis and the GSS geochemical data has been supplied under licence to the writer. The geochemical results from the survey covering the study area provide additional information on the geochemistry of the fluvial environment.

STATISTICS	Pb	Zn	Cu
Observations	97	99	94
Maximum value	900	1900	64
Minimum value	5	5	1
Median value	35	50	7
Mean value	101	126	12
Standard deviation	182	237	13

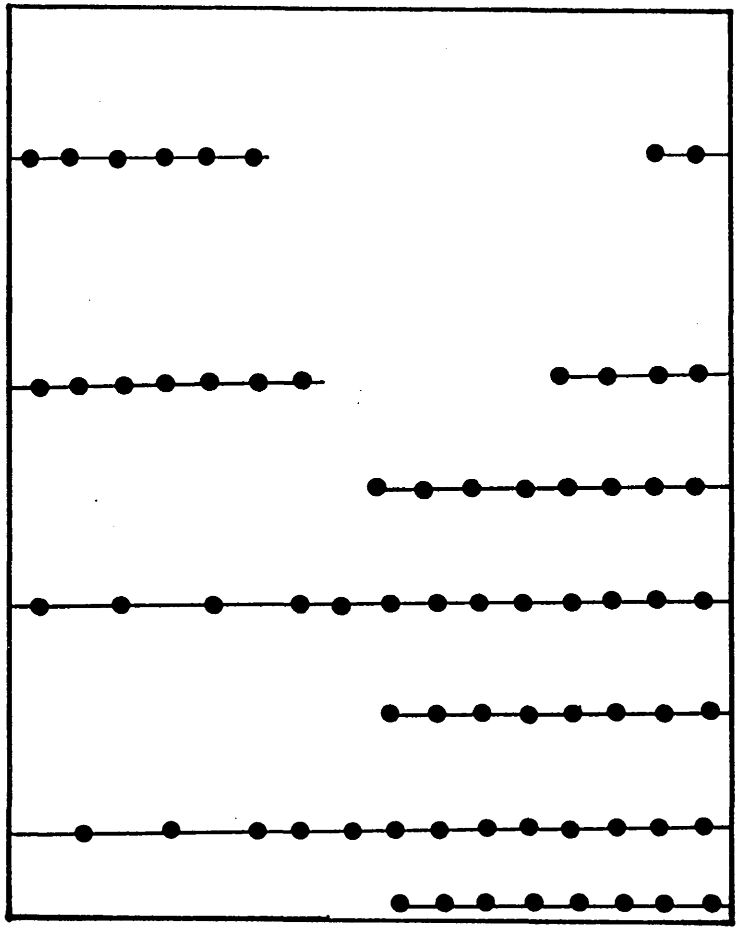
Table 7.2 Statistical characteristics of Pb, Zn and Cu distributions based on DRL geochemical data over soils near the former lead mine of Glasgow Beag, Loch Lossit (values in ppm).

7.3.2 Catchment distribution of Pb (Figure 7.2)

Lead shows a maximum of 900 and a minimum of 5 ppm respectively. With a mean of 101ppm and a standard deviation of 182 ppm respectively this shows that there is extremely high within-sample variation. The distribution of Pb forms a linear anomaly on a north-south alignment, terminating at the southern shore of Loch Lossit. The minimum value attained in the DRL survey was 5 ppm with a maximum of 900 ppm. Background concentrations are in the region of <25 ppm. A greater proportion of the Pb values fall between 169-350 ppm.

7.3.3 Catchment distribution of Zn (Figure 7.3)

With a mean value of 126 ppm and a high standard deviation of 237 ppm the values show very high variation across the range of samples. Zinc is concentrated at its greatest to the south-east of the loch. The highest value is in excess of 1900 ppm which is extremely anomalous. The background concentration for Zn is in the region of <100 ppm. Zinc concentrations in soils close to the loch are in the range of 25-144 ppm.



DRL Geochemical Survey
Pb, Zn and Cu soil sample
points

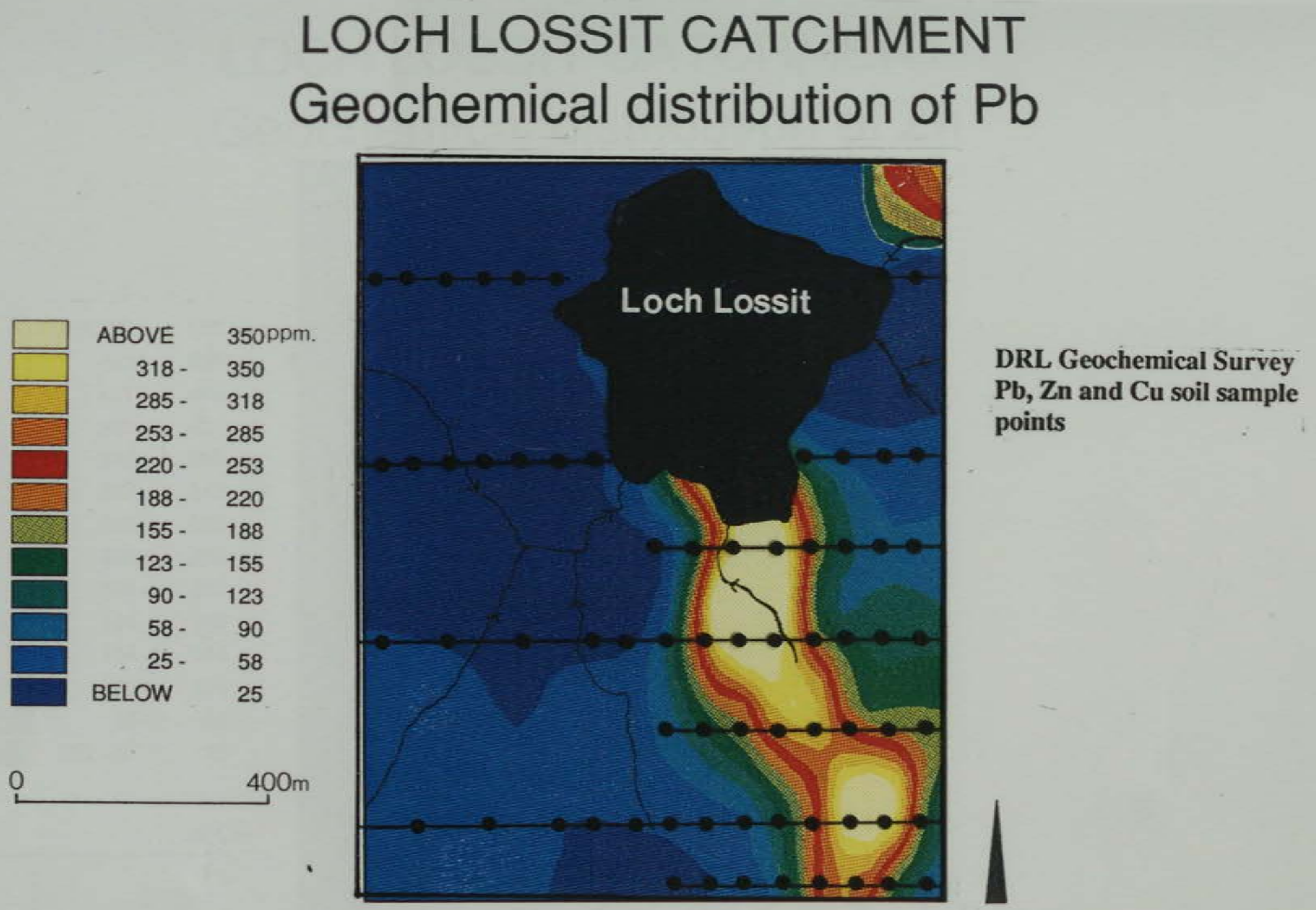


Figure 7.2 Distribution of lead in surface soils in the Loch Lossit catchment (after Domego Resources Ltd 1983).

LOCH LOSSIT CATCHMENT
Geochemical distribution of Pb

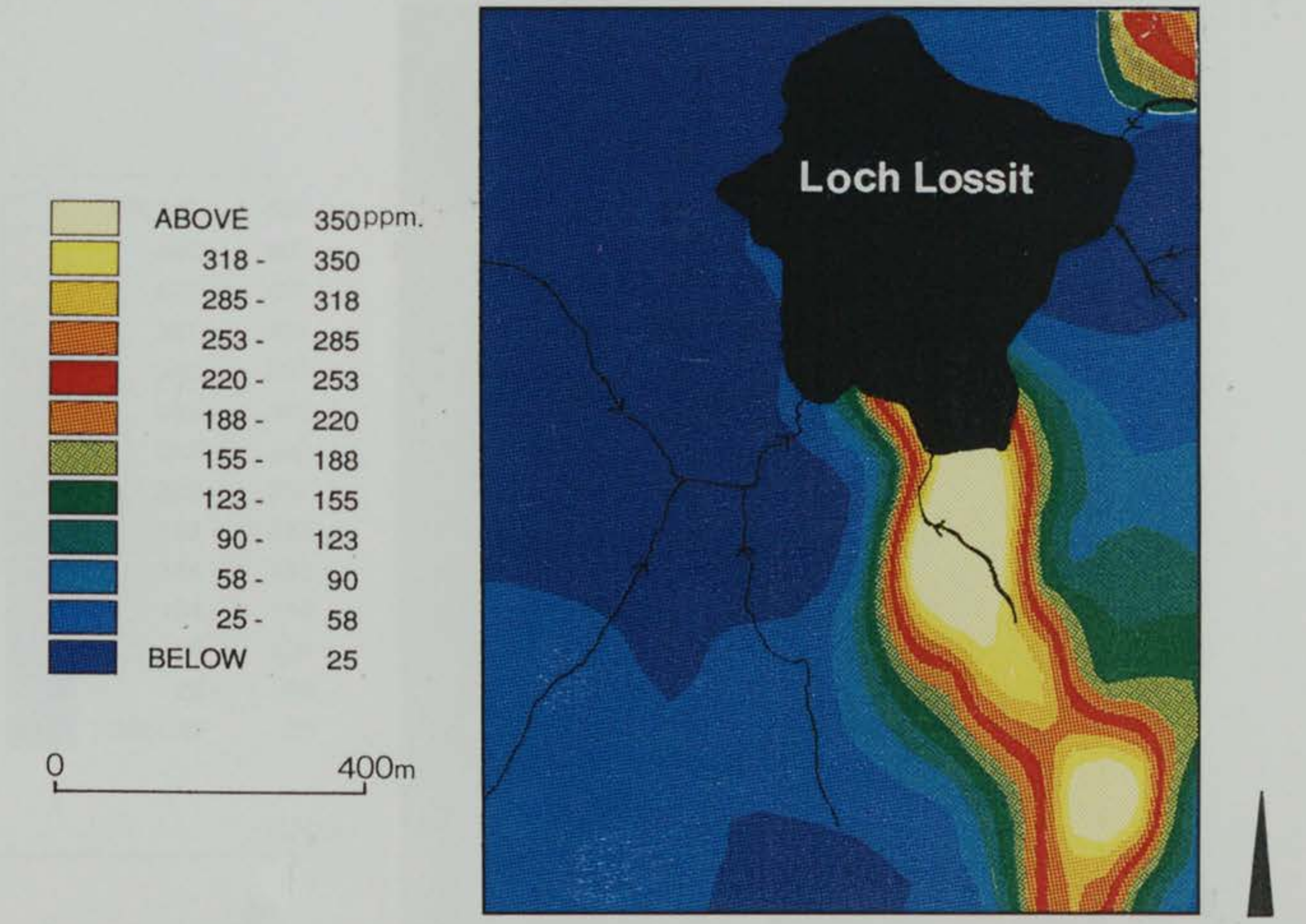


Figure 7.2 Distribution of lead in surface soils in the Loch Lossit catchment (after Domego Resources Ltd 1983).

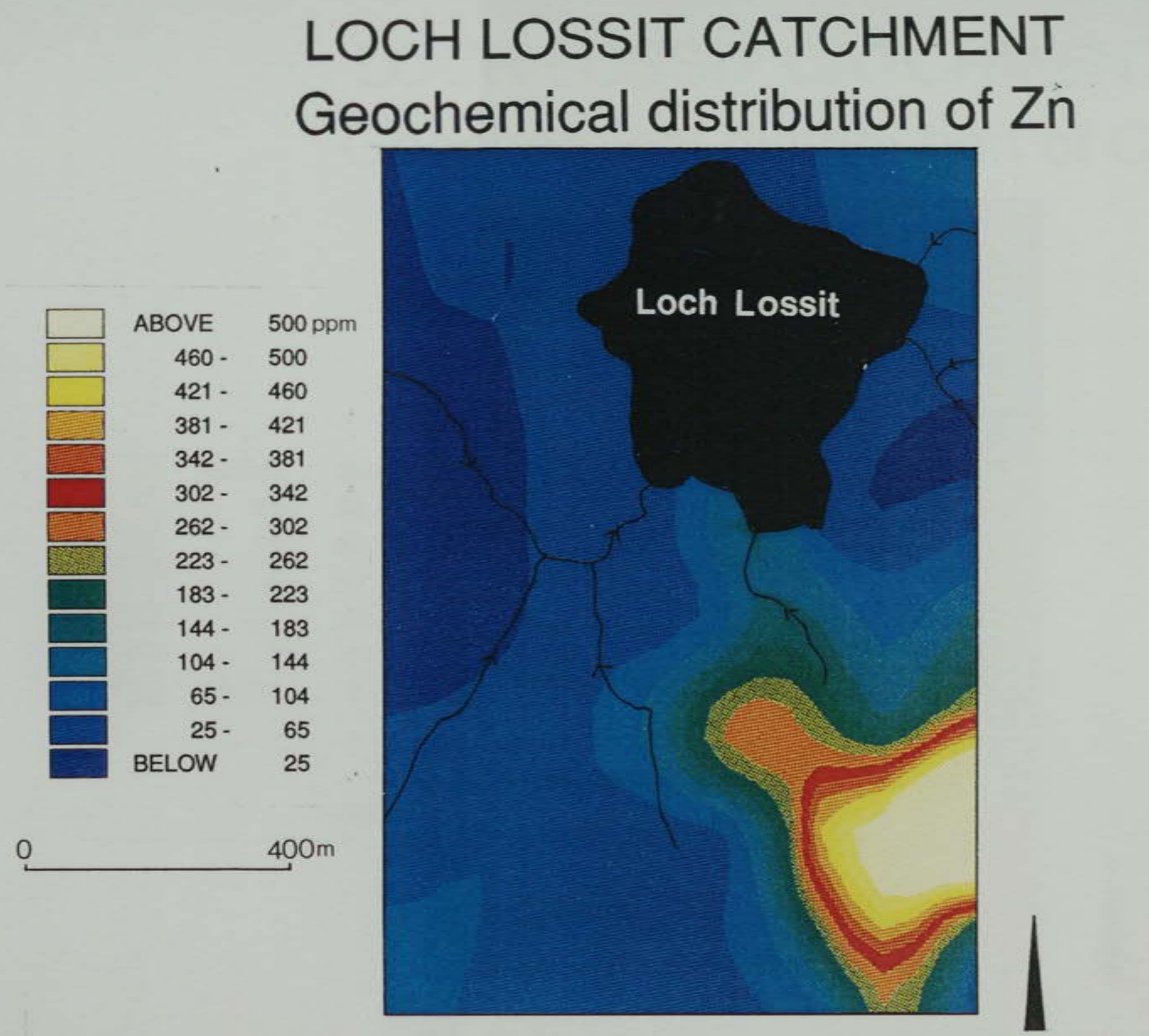


Figure 7.3 Distribution of zinc in surface soils in the Loch Lossit catchment (after Domego

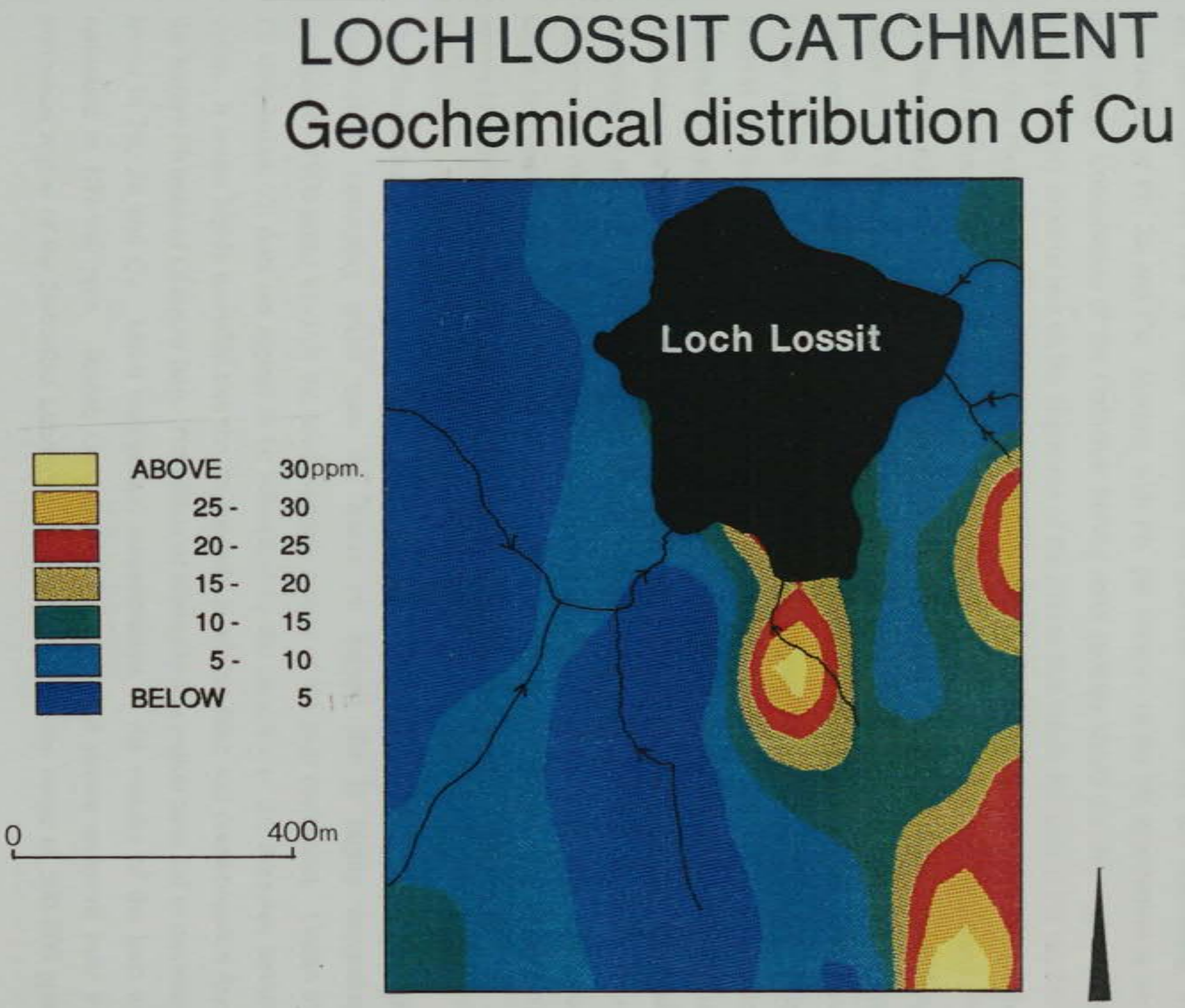


Figure 7.4 Distribution of copper in surface in the Loch Lossit catchment (after Domego Resources Ltd 1983).

7.3.4 Catchment distribution of Cu (Figure 7.4)

Copper has a maximum of 64 ppm and a minimum of 1 ppm, alongside a mean of 12 and low standard deviation of 13 ppm respectively. The low standard deviation shows that the within-sample variation is low and the values across the range of data are of a similar order of magnitude. Copper dispersal about the vicinity of the loch is confined to a series of haloes near the southern shore. The Cu values range from between 25-30 ppm in soils adjacent to the loch. Background concentrations for Cu are <15 ppm.

7.3.5 Assessment on the observations of Pb, Zn and Cu

An assessment can now be made regarding the possible factors that are controlling the distribution of Pb, Zn and Cu. Starting with Pb, the linearity of the Pb distribution is very interesting. Consultation of the Ordnance Survey solid geology sheets for this region show tertiary activity more or less on the alignment of the stream that enters the loch at the southern shore (see Figure 7.1) At this point three tertiary dykes can be seen to converge across the Dalradian limestone and terminate on the southern shore. The linearly dispersed Pb distribution follows the alignment of the dykes and it is suggested that these are a controlling factor in Pb dispersal in soils overlying these features. It is interesting that there is no corresponding alignment in elevated Pb values where a dyke re-emerges on the north-west corner of the loch. The soil sampling transects in this region were taken on a much wider interval unlike the region to the south of the loch where samples were taken at a closer interval. In other areas (denoted by the blue areas in the plot) the background concentration (Pb background < 25 ppm) is in keeping with values for Pb recorded on the Dalradian limestone by the GSS. The stream-sediment survey recorded a background concentration of between 15-70 ppm. Low Pb levels (20-40 ppm) were attained over the Islay Subgroup tillites and quartzites of central and southern Islay. The 900 ppm value recorded in the DRL survey is highly anomalous and may be due to mineralisation or secondary enrichment.

The distribution pattern of Zn is much broader than the Pb dispersal. Although the pattern of dispersal is noticeably wider than the linear Pb haloes, Zn is highly anomalous (maximum~1900 ppm) towards the south-western corner of the loch catchment. Unlike the Pb distribution Zn does not appear to be influenced by the presence of the intrusive tertiary dykes. It seems highly probable that the Zn anomaly is arising from soil contamination from the former Pb mine of Glasgow Beg. The effects of mining in this region have led to increased levels in Pb, Zn and Cu. Mean background concentrations in the vicinity of the loch are estimated at 100-140 ppm. British Geochemical Survey found stream-sediment over the north-east region of the Dalradian Limestone to vary with Zn in the range of 300-800 ppm.

This metal displays a great deal of variance owing to the complicated geological structure of the region. Both the GSS and the DRL surveys have identified anomalous Zn values over the Dalradian limestone and these are often found to delineate historical sources of mineralisation. This has been the case in the region of Mulreesh (former lead mining region) where values >1000 ppm were recorded in stream sediment receiving drainage waters from the mine. In other regions where no mineralisation is recorded high Mn levels (ca. 5%) have been associated with anomalous Zn (>1000) levels and attributed to secondary enrichment (GSS,.87).

In the Lossit catchment Cu follows the same trend in both Pb and Zn but is particularly less concentrated (maximum~64 ppm) in the limestone belt to the south of the loch. Contamination from mining is controlling the Cu levels and to some extent tertiary activity may also be exerting an influence where Cu is dispersed across the region where the dykes converge. Elsewhere, the distribution of Cu in the catchment soils is low (Cu background < 20 ppm) and is within the range of values (< 20 ppm) observed by the GSS over the differing geologies of the Appin Group of central Islay. These include the Islay Limestone, Mulloch Dubh phyllite, Ballygrant Limestone and the Bharradail Phyllite, (Stephenson and Gould 1994). Low values (5-20 ppm) were recorded over the Islay limestone except where mineralisation is known (e.g. Mulreesh with >100 ppm Cu). The quartzite facies of north-east Islay were low in Cu with <20 ppm. In reducing environments (argillaceous peaty-glays) not known to contain mineralisation, detrital matter and secondary Fe and Mn oxides can exert control over Cu leading to enhanced concentrations (BGS/GSS, 47).

Some interesting trends emerge in the soil-surface geochemistry. Near-surface chemical influences by the tertiary dykes and Pb-rich detrital material resulting from former mine-tailings dumps have clearly led to the increased soil surface Pb values observed to the south-west region of the loch. The distribution of Pb in sedimentary rocks is controlled by primary carriers such as feldspars, micas and sulphides. Clay minerals and organic matter are also important in the retention or promotion of Pb in surface soils (Huff 1952).

From the distribution plots it can be observed that Zn is dispersed more widely than either Pb and Cu in the region of the loch. This is probably due to the presence of iron-rich sandy soils derived as drift deposits between the limestone outcrops. The former soil types promote acidic conditions combined with a tendency for increased podsolization due to poor drainage conditions in what is essentially a region of high precipitation. The predominance of pyritic slate and phyllite within the Lossit catchment must be also be a contributing factor in Zn's wide distribution that is augmented by the acidic and highly ferrous nature of these geological types. Dissemination of zinc may be brought about by lower pH levels in these soils. Wedepohl (1978) observed that the distribution of Zn in sedimentary rocks is primarily controlled by the abundance of detrital ferromagnesian silicates, magnetite and clay minerals. Elsewhere, Wedepohl (1978) has observed generally poor Zn values (30-50 ppm) in feldspathic sands compared with greywackes (70-100) and shales (50-190 ppm). Research has shown that the composition of mine-tailings from the former mine tailings in NE Islay are particularly rich in Zn so it is not surprising that this metal will be high in concentration in surface soils around the sites formerly mined for Pb. Edmond (1982, 13) in a spectrographic evaluation of dump material and samples of ore from specific sections in deep diamond-tip drill cores at the Mulreesh lead mine recorded the following levels of Pb, Zn, Cd and Ag. The values shown in Table 7.3 suggest that the Pb ore in NE Islay is zinc-dominant and silver enriched.

Source of sample	Pb (mg kg^{-1})	Zn (mg kg $^{-1}$)	Cd (mg kg^{-1})	Silver (g/ton)
Spoil tip (grab sample)	3,430	11,100	2,000	1.58
Drill hole (D83-1)	unkown	21,000	unknown	14
Drill hole (D83-3)	59,000	70,000	unknown	10

Table 7.3 showing the elemental concentration of Pb, Zn, Cd and Ag after Edmond (1982).

The distribution of copper around the loch is moderately low in keeping with the results obtained by the GSS. Their findings indicated low Cu values over the Dalradian Limestone group (< 5-20 ppm). Similarly, the quartzite and slate facies of the Islay Subgroup was equally poor in Cu (< 20 ppm). Baker (1990) stated that over peat (histosols) Cu was moderately low in concentration with between 15-40 mg kg^{-1} and on sandy soils (drift deposits) 2-10 mg kg^{-1} was the normal concentration. The abundance of Cu is much higher in igneous rock types (basalt 90-100 mg kg^{-1}) and the presence of dolerite is likely to be controlling some local surface contamination. Tailings that are rich in sulphide minerals, particularly the Cu mineral chalcopyrite will promote increased contamination of local soils. The acidic nature of soils derived over phyllites and shales will have a tendency to promote dissolution of Cu at the soil surface. It has been found that in general, peaty soils high in organic matter are usually deficient in available Cu (Baker 1990, 163). Including Cu dispersal through non-ferrous mining, the controlling factor in soil-surface Cu will be the humic acid, organic ligand and clay mineral content in the local soils, all of which play a major role in the adsorption and complexion of Cu (Robinson 1981, Baker 1990).

7.3.6 Conclusions

Contamination from historic Pb mining in the vicinity of the former mine of Glasgow Beg has led to highly anomalous values in Zn in surface soils around the site. Copper is less dispersed and low in concentration across the region effected by mining. Towards the shores of the loch there are highly anomalous values in Pb arising from the effects of the near-surface influences of dollerite tertiary dykes. South from the loch the influence of historic mining is the controlling factor leading to increased concentrations in Pb.

7.4 Bathymetry Survey and core recovery

The preceding discussion has examined in detail the characteristics of physical and chemical environment adjacent to Loch Lossit. The results of bathymetric survey will now be discuseed.. A Bathymetric survey was undertaken on the loch in May 1992 to locate the maximum depth of the profundal region in order to obtain the optimum position for coring sediment. The methods and apparatus used in the bathymetric and coring programmes are described in Chapter 3 Section 3.4. The results of the bathymetric survey revealed relatively shallow loch margins ranging in depth from between 5-10 ft (1.52 - 3.05 m) and shelving gradually to a central basin at just over 45 ft (13.8 m). The deepest part of the loch was located immediately north-east of the small island situated at the south of the loch. Here the maximum depth was recorded at 50 ft (15.24 m). Repeated bathymetric sweeps over this position confirmed an east-west trending trough and this position was deemed to be the optimum coring position. Two cores were obtained from this position and are shown as coring stations C1 and C2 on the bathymetric map (Figure 7.5). The sediment-water interface was removed from the core tubes prior to transportation to the laboratory to limit the effects of sediment mixing in the upper section of the cores. Later examination of the cores showed the integrity of Core 1 was doubtful as a series of small fissures had appeared in the upper region of the core. This was probably due to mechanical disturbance precipitated by a rapid loss of moisture content during transit back to the laboratory. Core 2 (hereafter Loss 2) appeared to be in a good state of preservation with no evidence of disturbance and was therefore chosen for analyses.

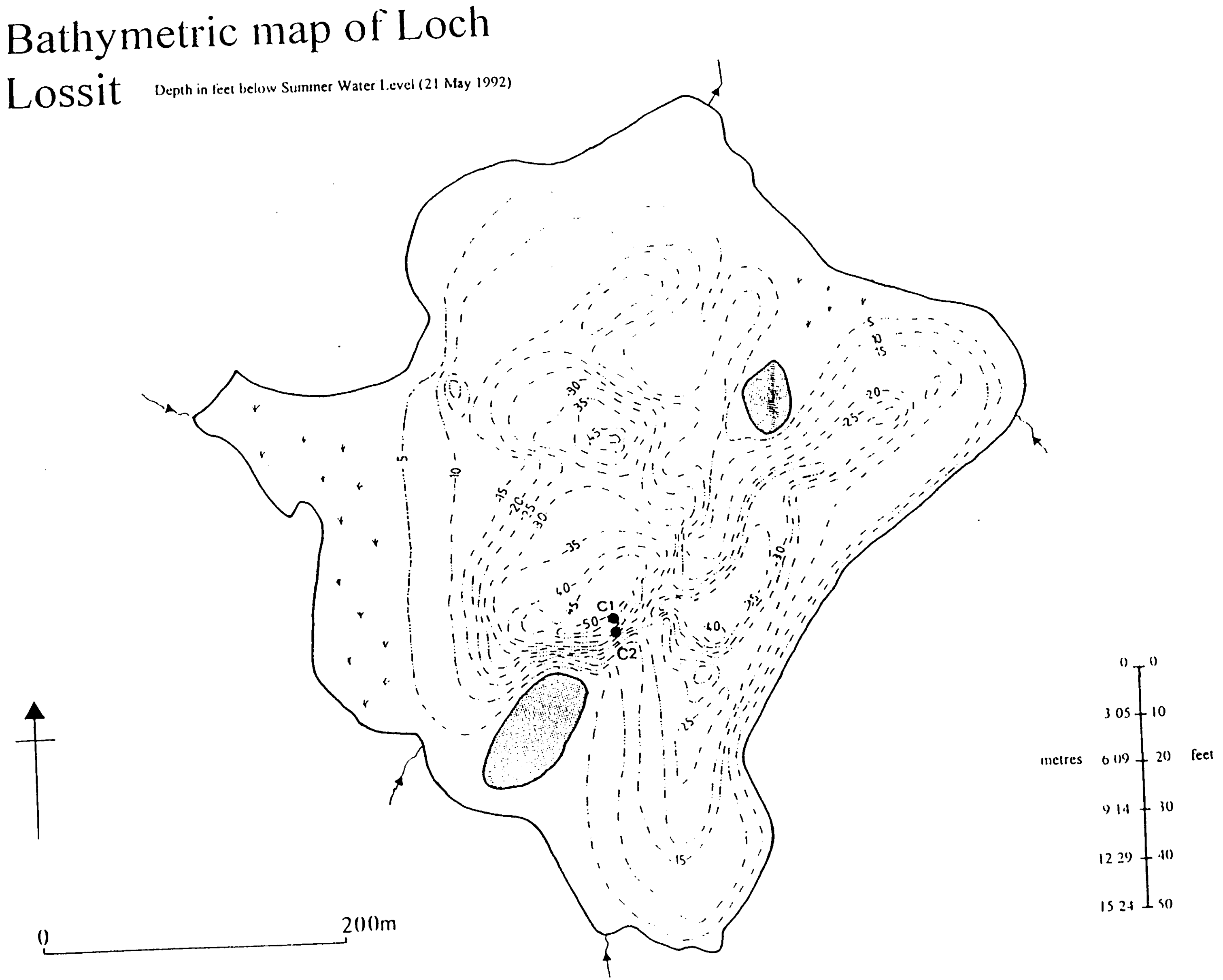


Figure 7.5 Bathymetric survey of Loch Lossit and coring stations.

7.4.1 The physical description of Core 2

In common with the other loch cores in this study, the Lossit Core 2 was seen to be homogenous and uniform in consistency, colour and texture. There was an absence of plant-macro material within the sediment. This is usually the case when cores are recovered from the profundal[1] zones of lochs which tend to be less prone to the incorporation of organic detritul material commonly recovered from the littoral[2] regions.

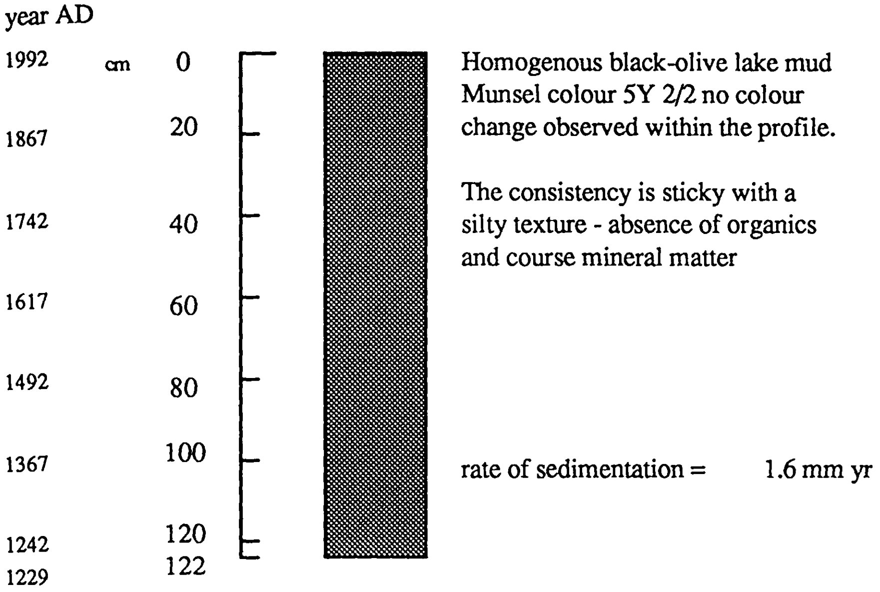


Figure 7.6 Summary description of Core 2 with extrapolated ^{210}Pb dates (1992-1229 AD) based on a ^{210}Pb-derived annual sediment rate of 1.6 mm.

7.5 Dating and Environmental History

7.5.1 ^{210}Pb analyses and the formation of an annual rate of sedimentation

The chronology for Core 2 has been established using the ^{210}Pb method outlined in Chapter 3 Section 3.8.2 ^{137}Cs has provided additional chronological support in the upper portion of the loch sediment. Linear Extrapolation is based on an annual rate of sedimentation of 1.6 mm yr^{-1} derived from the 'Constant Initial Concentration' linear regression model of log normal 'unsupported ^{210}Pb activity versus depth. The procedure is described in detail in Chapter 3 Section 3.8.2. No radiocarbon analyses were undertaken on this core owing to the close proximity of limestone to the loch. It was considered that this would have seriously effected the outcome of any radiocarbon analyses leading to 'hard-water error' (Chapter 6 section 6.6.2).

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[1] The profundal region of a lake refers to its central or deepest basin.

[2] The littoral region of a lake refers to its marginal or near-shore locations.

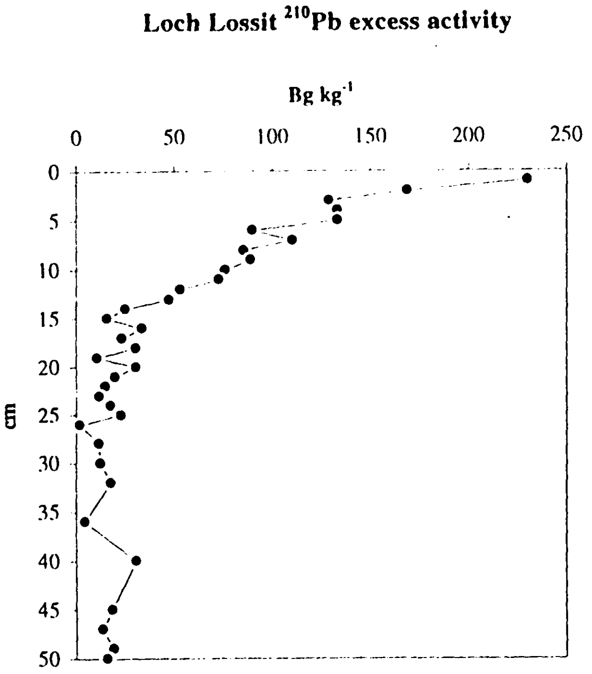


Figure 7.7 Depth versus $^{210}Pb_{excess}$ activity.

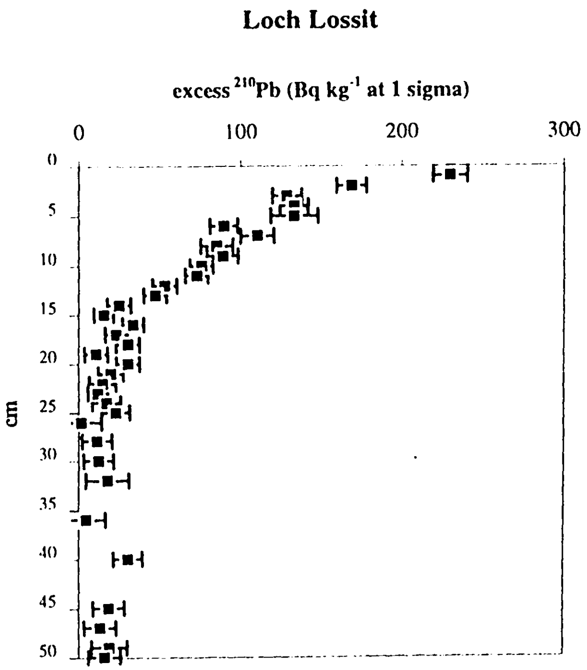


Figure 7.7 Depth versus $^{210}Pb_{excess}$ with error bars at 1 sigma.

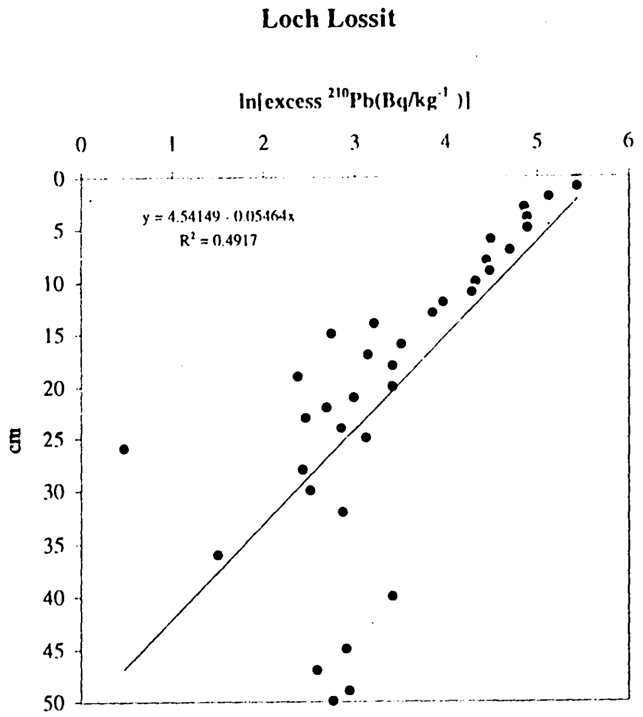


Figure 7.8 Linear regression plot of log normal $^{210}Pb_{excess}$ activity.

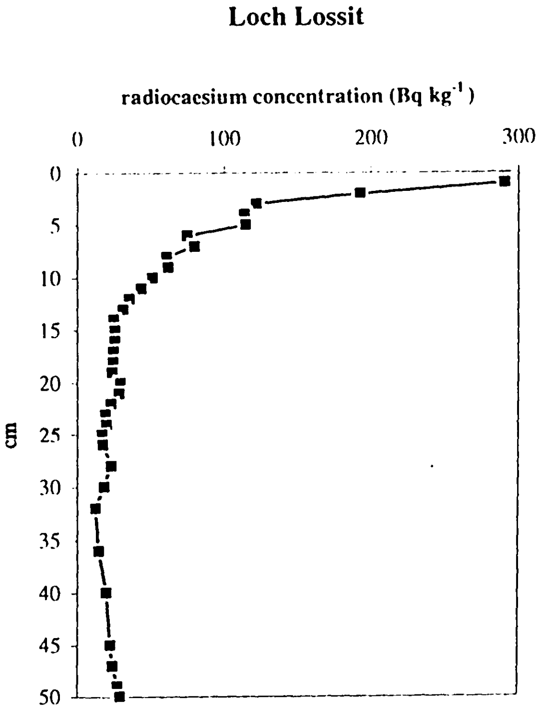


Figure 7.9 Depth versus ^{137}Cs activity.

Dates obtained from radiocarbon determination are likely to have been significantly older and in error of their true age range limiting the use of radiocarbon analyses on sediment containing allogenic mineral matter derived from the nearby limestone.

Most studies of ^{210}Pb-dating accept that the supply of unsupported ^{210}Pb to lake waters is governed primarily by local or regional meteorological factors and hence is reasonably constant on time scales of the order of 100-200 years, (Oldfield and Appleby 1984). Pb-210_{excess} activity has been detectable to a depth of 50 cm with a surface maximum (0-1 cm) of 230 Bq kg^{-1} ± 10.6 at 1 sigma. These values have been decay corrected to September 1992 and are shown in Figure 7.7 with 2 sigma error bars. The results allow a ^{210}Pb-derived sedimentation rate of 1.6 mm yr^{-1} providing a chronology that has been plotted alongside the core in Figure 7.6. The chronology has been extrapolated down the core, but beyond around the 1840's extrapolated dates must be viewed as tentative without a supporting radiocarbon chronology (cf. 6.6.5 above). For the upper region of the core (0-20 cm) it is argued that the proposed ^{210}Pb chronology may be more reliable. The upper region of sediment (0-50 cm) has a reliable ^{137}Cs inventory which provides supporting chronological control. The results of the ^{137}Cs chronology will now be discussed.

7.5.2 Chronological support using ^{137}Cs

Radiocaesium in the Loch Lossit sediment is detectable to 50 cm with a surface (0-1 cm) ^{137}Cs maximum of 290.5 ± 4.0 Bq kg^{-1}. The ^{137}Cs concentration exhibits an exponential decrease from the surface with a subsidiary subsurface maximum in the 4-5 cm section. Using the ^{210}Pb derived sediment rate, the sediment sections 4-5 cm fall between a date range of 1967-1960. This corresponds to the 1963 maximum in weapons testing fallout deposition, with higher overlying concentrations as a result of the Chernobyl nuclear accident fallout deposition in 1986. This subsurface concentration (1963 maximum) at 4-5 cm is 113.9 ± 1.6 and 114 ± 1.7 Bq kg^{-1} respectively. In all probability there has been some diffusion down the core thus lowering the down-core concentration. ^{137}Cs is mobile within core surface pore-waters and this diffusion could be further exacerbated by burrowing benthic animals such as *Chironomidae* (Bryant *et al* 1993). The radiocaesium concentration profile (Figure 7.9) shows that caesium is still active at a depth of 50 cm. Baseline values have not been reached and this is either a result of 'noise' within the counting apparatus or inherent radioactivity activity still present at this depth, a result of the mobility in sediment of this radio nuclide.

7.6 Geochemical Inventory and Physical Properties

The sedimentary fluxes for Pb, Zn, Cu, Ca, Fe and Mn are shown in the summary diagram (Figure 7.20). Descriptive statistics from the data set are shown in Table 7.4 The following descriptions summarise the concentration of the individual metals and are displayed in the depth versus concentration profiles (Figures 7.10-7.15). The relative order in maximum chemical concentration is Mn >Fe > Zn > Pb > Ca > Cu. Metal concentrations are expressed in mg kg^{-1} per dry weight of sediment (sample dry weight = 0.5 g). Organic and mineral content, magnetic susceptibility and pH content are shown in Figures 7.16-7.19.

Statistics	Pb	Zn	Cu	Ca	Fe	Mn
Observations	62	62	62	62	62	62
Maximum value	135	415	38	75	2490	3033
Minimum value	35	123	10	8	1230	1348
Median value	78	173	17	50	1625	2174
Mean value	85	187	18	48	1693	2116
Standard deviation	27	57	6	13	273	352

Figure 7.4 statistical data used in the depth versus concentration profiles.

7.6.1 Observations

Lead (Figure 7.10)

From the basal concentration of 124 mg kg^{-1} there is a decrease at 116 cm with 98 mg kg^{-1}. From this position there is a sustained rise in concentration to 135 mg kg^{-1} at 108 cm. Here the curve remains constant until values decrease gradually to 58 mg kg^{-1} at 78 cm depth.. There are three intermittent smaller peaks (maximum 75 mg kg^{-1}) towards the surface. A rise in concentration occurs between 6-2 cm with 96 mg kg^{-1}. The concentration of Pb in the surface section of sediment at 2-1 cm depth is 78 mg kg^{-1}.

Zinc (Figure 7.11)

Zinc is concentrated at the base at 187 mg kg^{-1}. The concentration remains fairly constant until 58 cm with 235 mg kg^{-1}. After a small fluctuation, there is a noticeable peak towards the surface of the core with 366 mg kg^{-1} at 8 cm depth.. At 2-1 cm the surface concentration is 249 mg kg^{-1}.

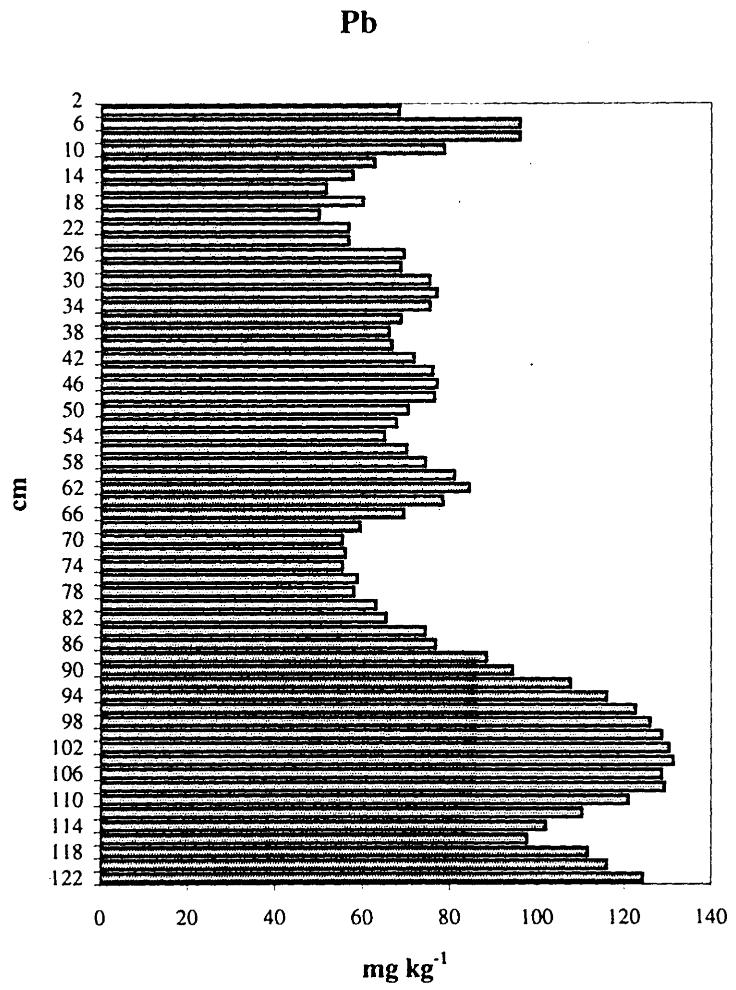


Figure 7.10 Depth versus lead concentration.

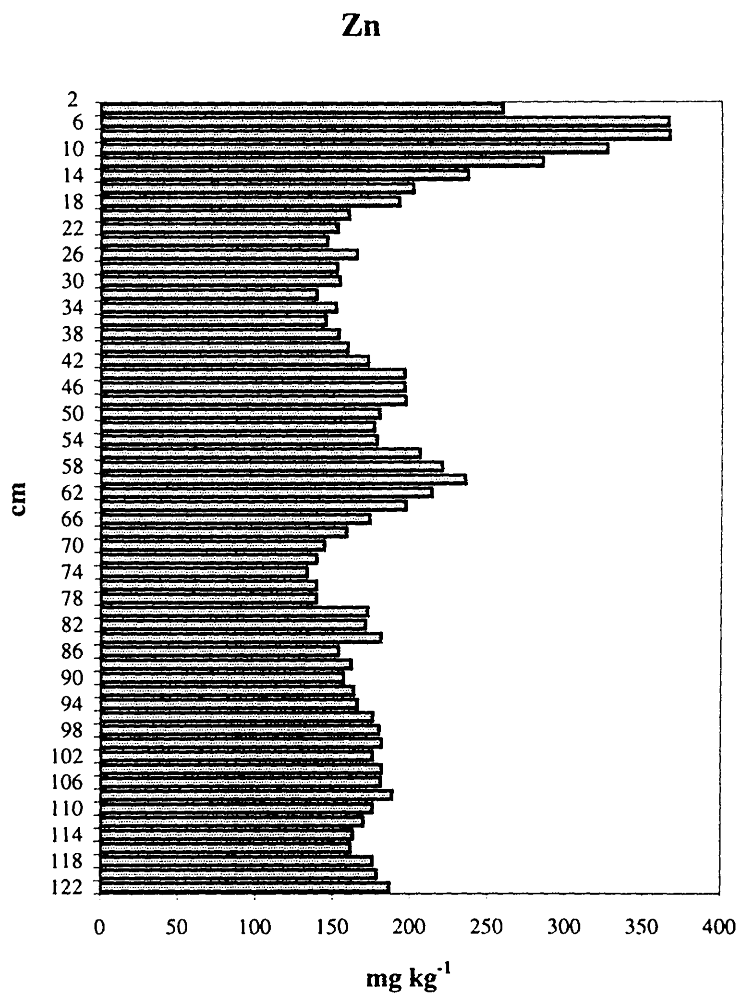


Figure 7.11 Depth versus zinc concentration.

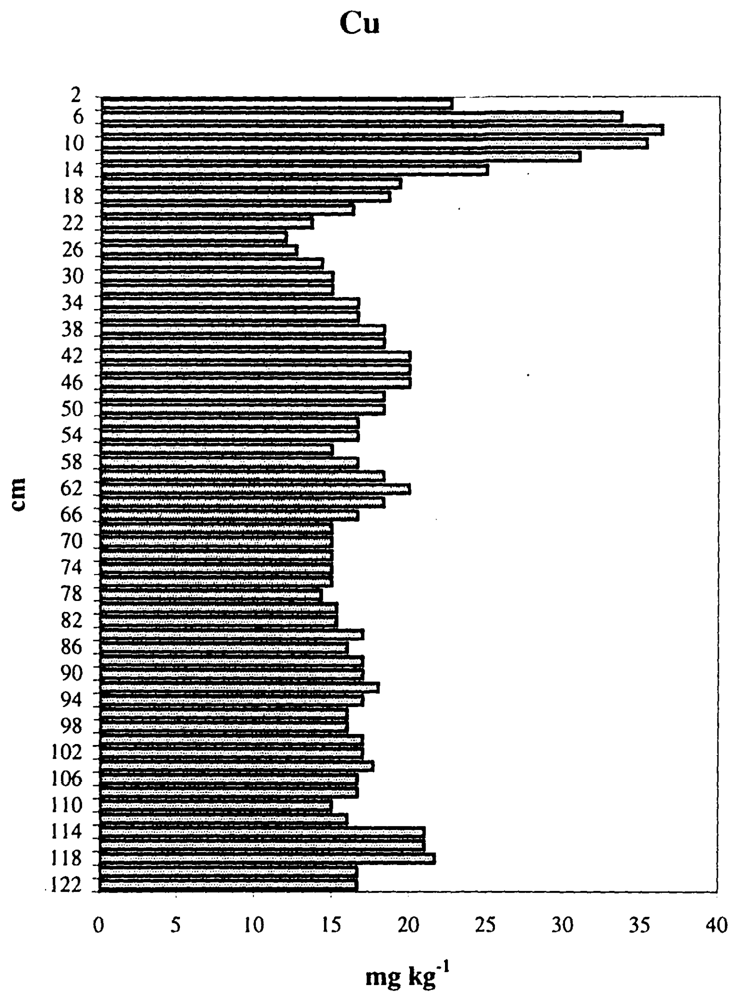


Figure 7.12 Depth versus copper concentration.

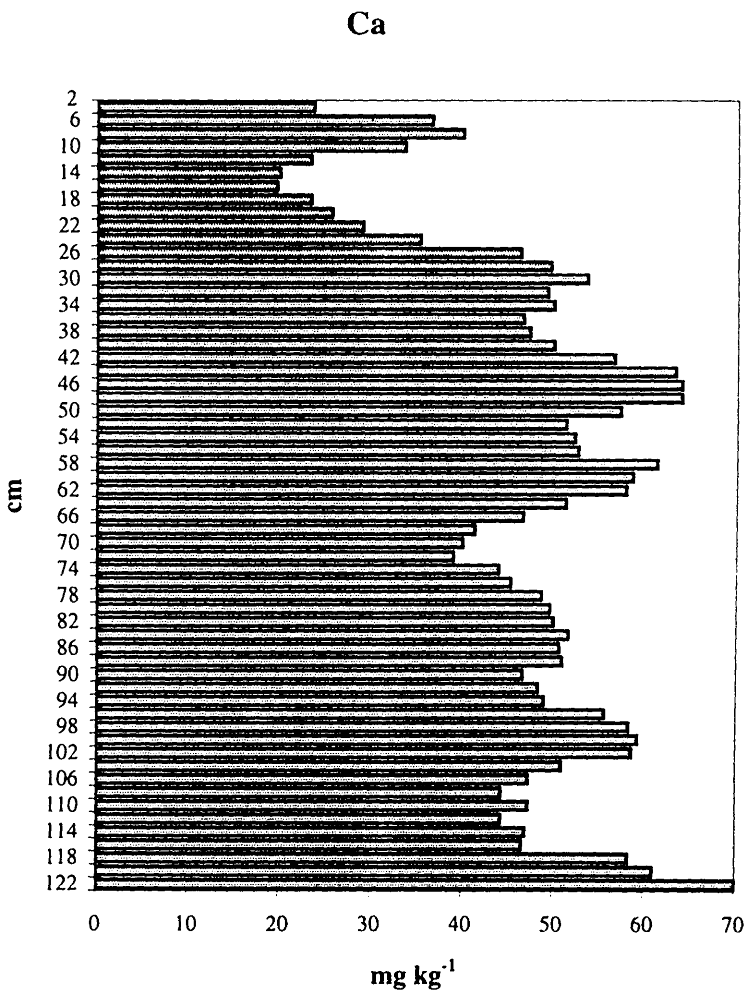


Figure 7.13 Depth versus calcium concentration.

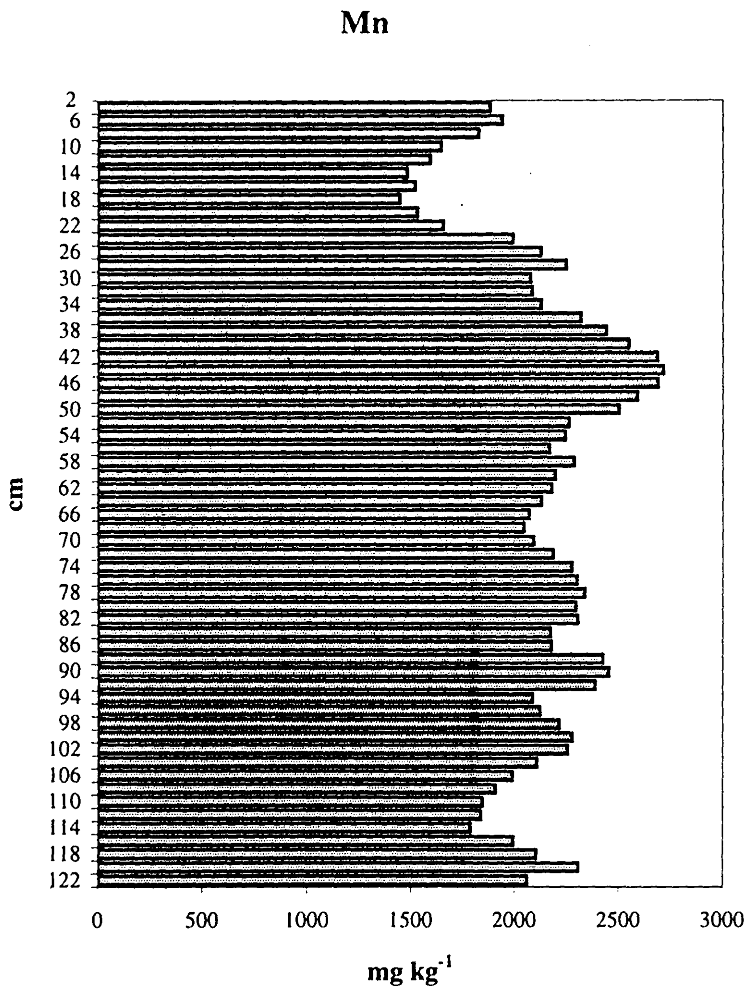


Figure 7.14 Depth versus manganese concentration.

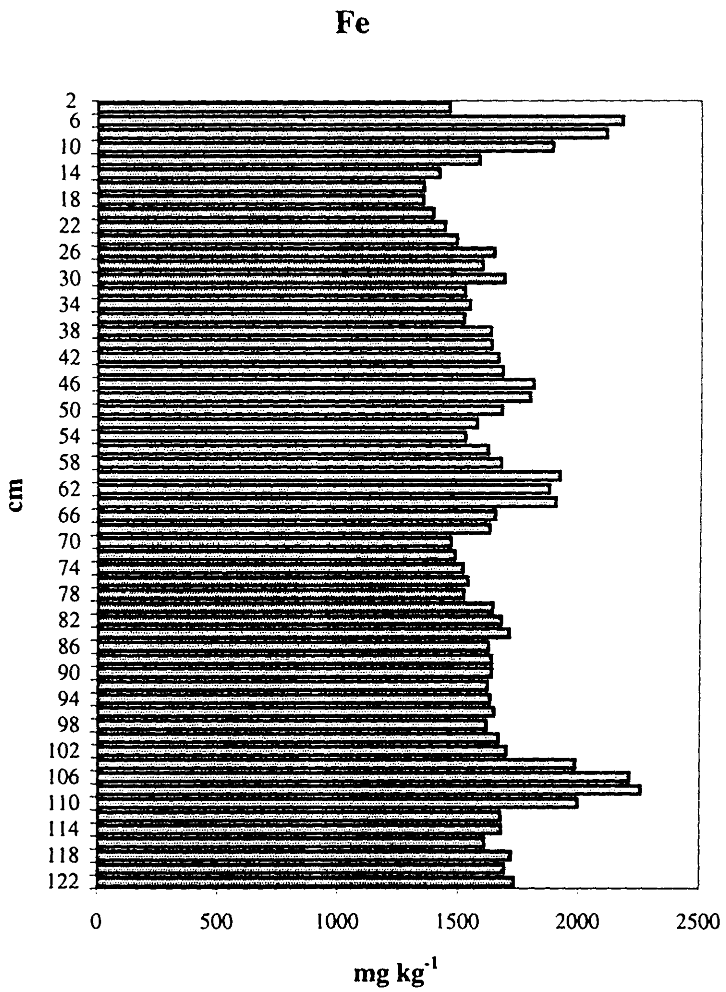


Figure 7.15 Depth versus iron concentration.

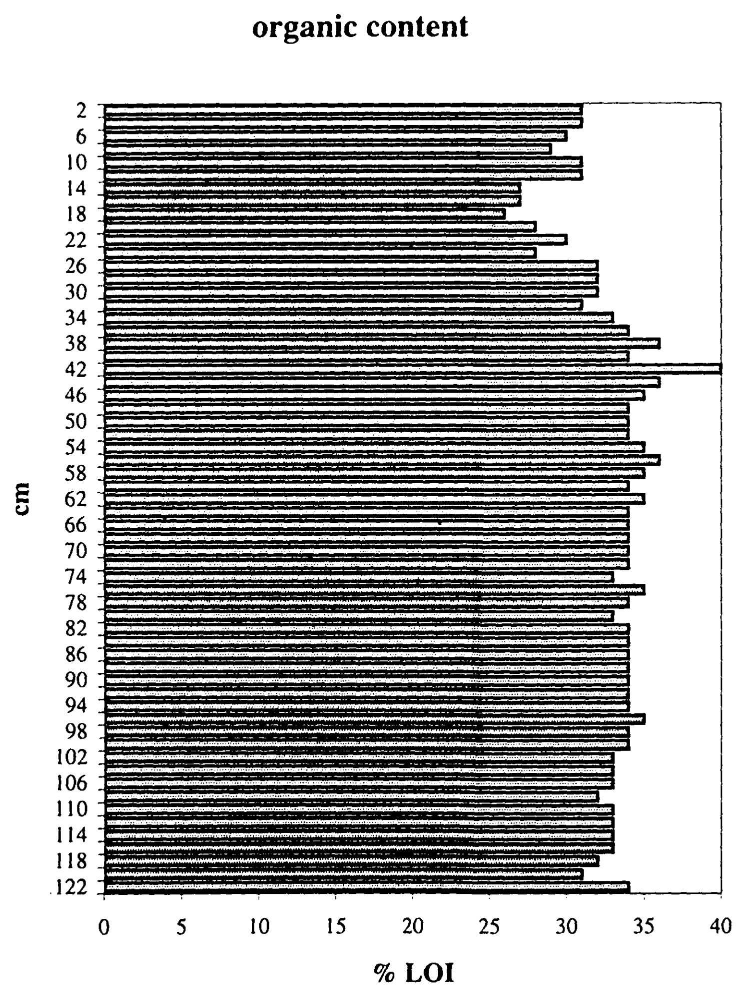


Figure 7.16 Depth versus organic content.

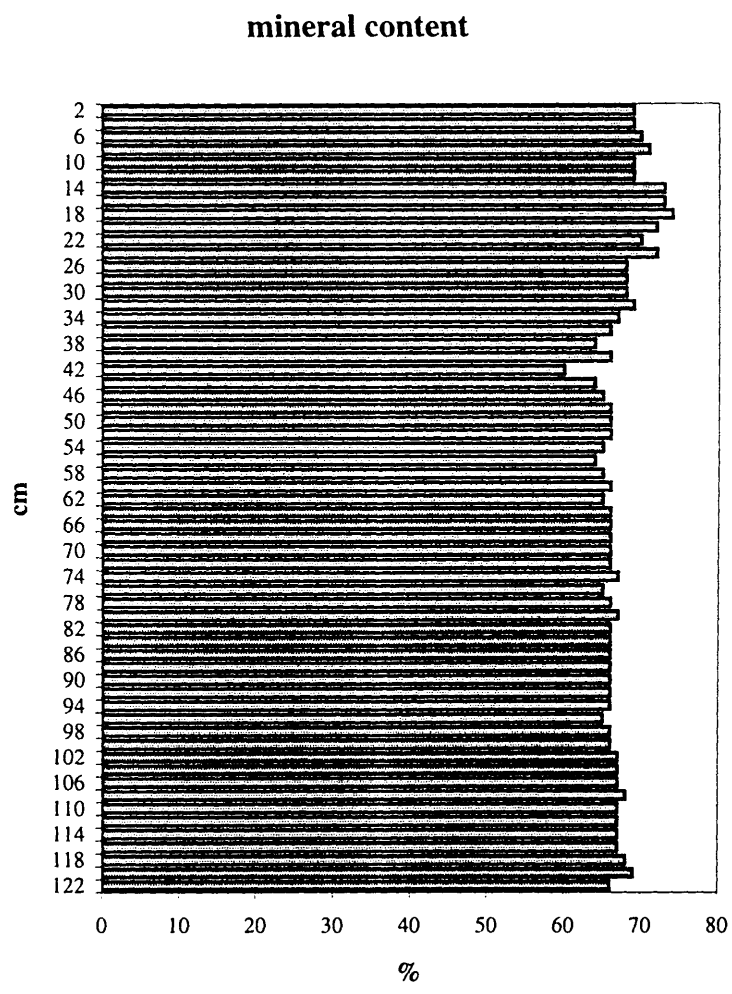


Figure 7.17 Depth versus mineral content.

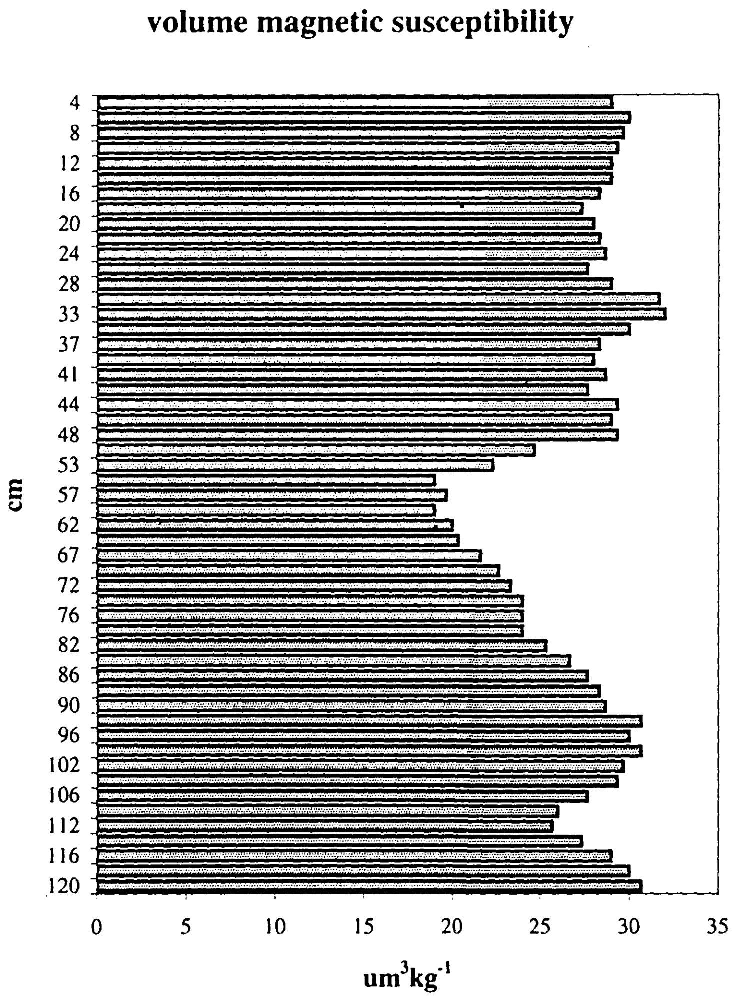


Figure 7.18 Depth versus volume magnetic susceptibility.

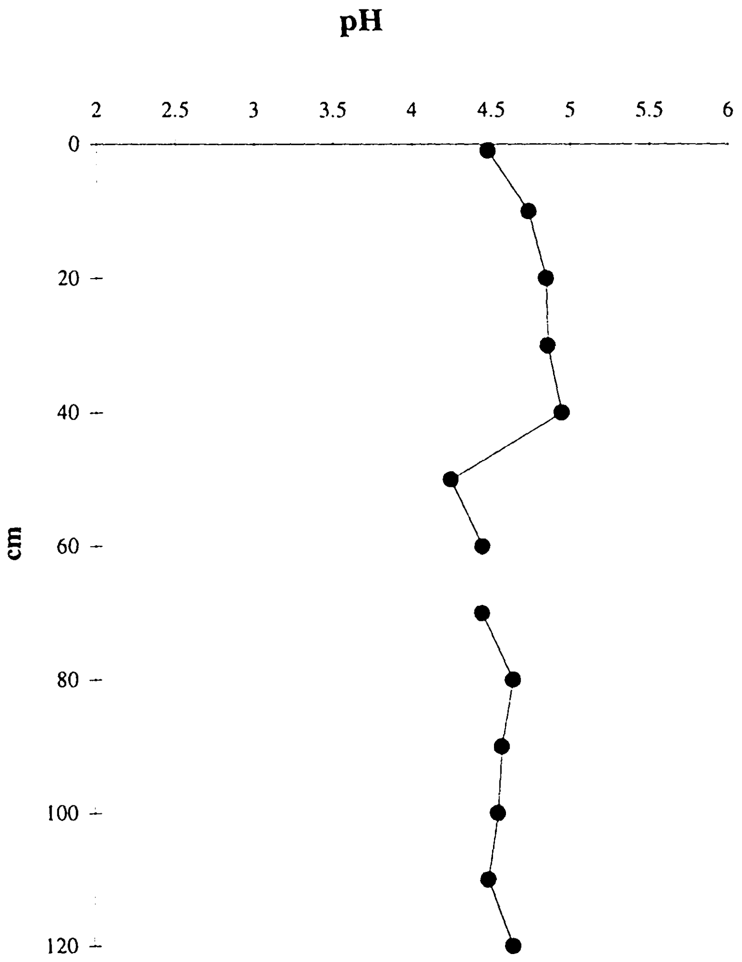
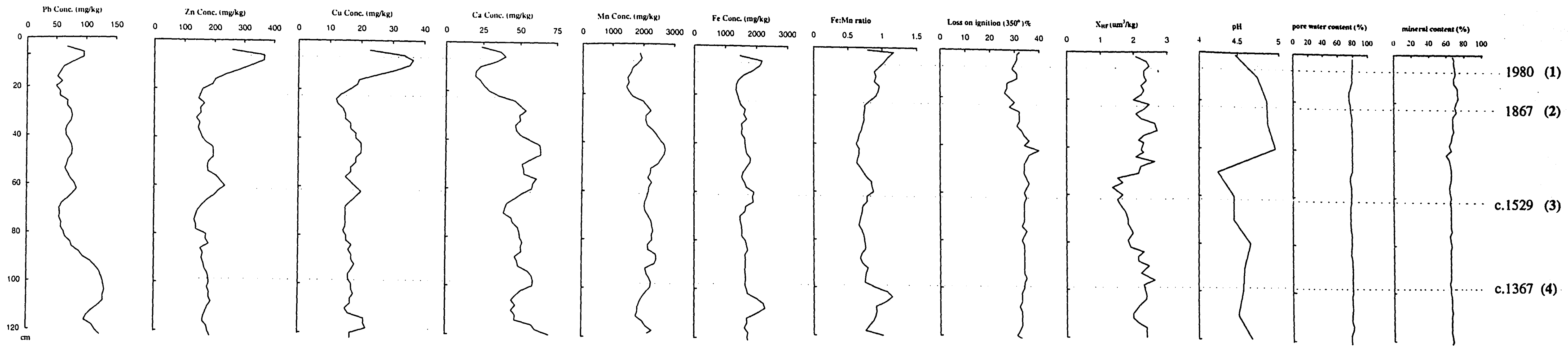


Figure 7.19 Depth versus pH concentration.

Loch Lossit Geochemical Summary Diagram



Extrapolated ^{210}Pb Date		Event in the profile	Interpretation
(1)	c.1955	Near-surface maximum of Pb, Zn, Cu and Ca	Peak in atmospheric fall-out deposition
(2)	c.1867	Rise in Pb, Zn and Cu	Onset of atmospheric fall-out deposition
(3)	c.1529	Small increase in Pb, Zn, Cu Ca and Fe	Lead mining? Influx of lead-rich solutes into the loch.
(4)	c.1367	Maximum concentration in Pb and Ca	Early lead mining ?

Summary of possible dates and events within the Loch Lossit geochemical diagram based on an extrapolated annual sedimentation rate of 1.6 mm yr^{-1}.

Figure 7.20 Loch Lossit geochemical summary diagram.

Copper (Figure 7.12)

The base of the core has 16 mg kg^{-1} Cu with a small rise (22 mg kg^{-1}) occurring at between 116-112 cm. The concentration remains constant until 60 cm with a rise at 20 mg kg^{-1} and 40-44 respectively. A sharp peak occurs towards the upper surface of the core starting at 24 cm to 10 cm where a maximum concentration of 36 mg kg^{-1} is attained. There is a decline to 22 mg kg^{-1} at 2-1 cm.

Calcium (Figure 7.13)

The base of the core has 70 mg kg^{-1} Ca falling to 46 mg kg^{-1} at 116 cm. From here the values remain constant until 106 cm. Between this point and 98 cm there is a rise to 60 mg kg^{-1}. At 90 cm there is a decline to 46 mg kg^{-1}. An erratic rise occurs until 68 cm from this point a rise occurs to 64 mg kg^{-1}. At 42 cm a second peak occurs with 65 mg kg^{-1}. At 34 cm there is a smaller peak at 55 mg kg^{-1} and decline to 14 cm with 20 mg kg^{-1}. From here to 6 cm a rise to 40 mg kg^{-1} is noted. At the surface 2-1 cm Ca is at 23 mg kg^{-1}.

Manganese (Figure 7.14)

The concentration of Mn at the base of the profile is at 2000 mg kg^{-1}. There is a rise to 2400 mg kg^{-1} at 120 cm. An intermittent rise occurs from here to 44 cm where a maximum of 2700 mg kg^{-1} Mn occurs. A reduction in concentration occurs with 1400 mg kg^{-1} at 18 cm. A rise occurs towards the surface from this point to 4-2 cm with 1900 mg kg^{-1}.

Iron (Figure 7.15)

The basal value for Fe is at 1600 mg kg^{-1} and remains constant until 114 cm. From this point to 106 cm it rises to 2300 which forms the maximum value in this profile. There is a reduction in concentration at 102 cm with 1600 mg kg^{-1}. Two smaller peaks occur towards the surface of the profile. At 60 cm Fe is at 1900 and at 44 cm at 1700 mg kg^{-1}. There is a reduction from this point to 12 cm and there is a rise to 4 cm with 2300 mg kg^{-1}. Sub-surface concentration (2-1 cm) is at 1600 mg kg^{-1}.

Iron:Manganese ratio (Figure 7.20)

The Fe:Mn ratio curve has its maximum concentration towards the base of the core with 1.3. After a small decrease there appears to be little variability, including only a notable decrease at 4 cm from 1.2 to 0.6.

Mineral content (Figure 7.17)

The concentration of mineral matter is constant from the base of the core up to 36 cm with 66 % mineral content. From 36 cm there is a slight rise towards 18 cm with 75 %. Values decline slightly to 14 cm with 68 % whereafter this value remains constant towards the surface.

Water Content (Figure 7.20)

Water content is expressed as percentage concentration. Fluctuations in concentration occur up the core, starting from the base where values range from 78-81%. From *c*.35cm there is a sharp decline in water content to 76% where there is an increase towards the surface at 80%.

Organic Matter (Figure 7.16)

Organic matter is expressed as percentage concentration. The organic content of the core remains relatively constant from the base to 48cm with 32-36%. From here there is a decline in concentration at *c*.20 cm were 25 % organic matter is attained followed by a slightly higher rise towards the surface with 32 %.

Magnetic susceptibility (Figure 7.18)

The results show that there may have been some enhancement towards the lower and upper portion of the core. This is due possibly to the influx of metals to sediment as a result of mining within the catchment and possibly in the upper profile as a result of farming practices. Starting from the base of the profile 0.19 $\mu m^3 kg^{-1}$ at 122 cm have been attained. After an intermittent rise at *c*.100 cm there is a gradual decrease in values with 0.09 $\mu m^3 kg^{-1}$ at 52 cm. From this position there is a sharp rise to 0.2 units at 48 cm. The curve from this point is erratic with numerous small peaks towards the surface of the core.

pH concentration (Figure 7.19)

The pH values are fairly constant from the base of the core with pH 4.7 to pH 4.2 at 50 cm. There is a rise to pH 5 at 40 cm which tails off to pH 4.5 at the surface of the core.

7.6.2 Implications of the geochemical inventory of Core 2

The relative order in maximum chemical concentration is Mn >Fe > Zn > Pb > Ca > Cu. The depth versus concentration profile for Pb is markedly different from the other metal profiles observed in this core. Table 2.6 above summarises the likely events that have given rise to the Pb concentrations within the profile. Lead sees its maximum concentration near to the base of the core and falls intermittently up the core before a rise at the sub-surface. No other metal behaves in this manner and there appears to be no inherent relationship with the other profiles with the exception of Fe.

Zinc on the other hand, shares similar trend of distribution with Cu. Both metals have two fairly synchronous peaks at around 50-58 cm and both decline in concentration before reaching a sub-surface maximum. Calcium is difficult to interpret as this ion is unevenly distributed throughout the sediment profile as well as being an autocthonous component of the loch system. The profile for Fe sees its maximum nearer the base than the surface. There may therefore be a diagenetic relationship between Fe and Pb. Manganese declines away from its maximum concentration in the 45 cm region of the core. This metal seems to exert no influence on the other metals (see Section 9.5.3 below). The distribution in organic matter determined from loss on ignition shows that organic content was low throughout the core with lesser amounts from *c*.45 cm a further decrease can be observed around 25 cm. It would appear that there has been a slight increase in mineral influx, perhaps due to erosion processes occurring in the catchment. To some extent the water content may be correlated to this event as there appears to be a decline in water content at more or less the same region thus reflecting a more minorgenic consistency within this section of the core. The mechanisms that control the chemical processes commonly observed in sediment profiles will be discussed in Chapter 9 Section 9.5.

In the Lossit core the Pb profile attains a maximum concentration at 102 cm with 135 mg kg-^{1}. This value is not significantly high in comparison with the maximum concentration in the Loch Leathann profile (205 mg kg^{-1}) or the minimum Pb value of 275 mg kg^{-1} observed in the Bharradail core described in the previous chapter. Using the ^{210}Pb-derived sediment rate of 1.6 mm yr.$^{-1}$ the maximum Pb concentration falls at 1367 AD. As we have seen in Chapter 4 Section 4.6.2 this date would predate the earliest historical accounts of lead mining on Islay by some 144 years. This length of time is not that considerable and what we are seeing is possibly a response from small scale lead mining activity within the vicinity of the loch. It was mentioned above that the physical extent of mining at the 18th century mine of Glasgo Beg appears to have been on a much smaller scale in comparison to mining operations that occurred at the contemporary site of South Ardachie.

There appears to be some correlation between magnetic susceptibility and the increase in Fe and Pb. To what extent these metals are leading to increased magnetic enhancement cannot be ascertained. If this was a result of an increase in sediment influx to the loch then one might have expected an increase in allogenic mineral matter and this does not appear to have been the case if we consider the linear trend observed in the mineral matter profile. It is suggested that increases in Mn and more probably Fe are a controlling factor resulting in slight enhancement within the magnetic susceptibility profile. It has been mentioned previously that increases in Fe and Mn can lead to increases in magnetic susceptibility. Following Mortimer (1942) and Mackereth (1966) these high concentrations in Fe and to a lesser extent Mn are interpreted as a reflection of leaching from surrounding iron-rich soils under increasingly reducing conditions. Manganese does not show an increase in concentration at 102 cm therefore diagenetic control by this metal over Pb can be ruled out. To some extent there may be some influence in the Fe concentration at this position but it appears to be very slight. The concentration of Fe at this position is 2200 mg kg^{-1} and is not sustained as is the case for Pb. Higher in the Pb profile there are three subsidiary peaks between 76-24 cm. The lowest peak at 62 cm (80 mg kg^{-1}) is matched by peaks in Zn and Cu at the same position. On the basis of a 1.6 mm yr^{-1} ^{210}Pb-derived annual rate of sedimentation, this position would date to 1529 which is well within the range of earliest dates mentioned in the historical accounts for lead extraction on Islay.

In all the depth versus metal concentration profiles there is an increase in metals at around 26 cm. This position falls at the date 1842 which may relate to land use changes within the catchment. It is at around this period that major improvements in drainage and formal planting in the catchment occurred (*New Statistical Account* 1845). Furrow and tile drainage were being brought into use to drain wetter regions, along with an intensification in peat cutting for fuel to meet the needs of an increasing population and the newly emerging distilleries. Part of the sub-surface metal concentrations include an atmospherically derived Pb component. Copper and Zn show a marked increase towards the surface and atmospheric fall-out deposition again is likely to account for this. Aspects relating to this topic are developed in more detail in Chapter 9.

With the onset of agricultural improvement in the near vicinity of the loch and a shift towards greater land improvement, we are likely to see an intrinsic mineral component entering the loch resulting in a chemical response towards the near-surface of the core. Deeper in the profile it is argued that mining has promoted the input of a higher Pb burden into the loch basin. A major erosion event unrelated to mining could be responsible for the sustained Pb increase. However this seems unlikely, given the fact that there is no corresponding increase within the other metal profiles.

Extrapolated Pb-210 Date	Event in the profile	Interpretation
c.1842 AD	Increase in metal influx Pb, Zn and Cu (26 cm).	Mid-19th century land-land use changes (New Statistical Account 1845).
c.1529 AD	Intermittent peak in Pb, Zn and Cu.	Possible response of small-scale Pb lead extraction in the Lossit catchment.
c.1367 AD	Protracted rise on Pb at 102 cm with 135 mg kg^{-1}.	Possible Pb extraction but predates earliest historical account for Pb extraction by *c*.144 years.

Table 2.6 Summary of possible dates and events within the Loch Lossit profile based on linear extrapolation of ^{210}Pb-derived sediment rate of 1.6 mm yr^{-1}.

7.7 Conclusions

The surface geochemical environment has been explained in relation to the geological structure of the immediate area around to Loch Lossit. It is clear that there has been enhancement to the soil-surface concentration of Pb (900 ppm) resulting from the contaminatory effects of mine tailings around the former mine of Glasgow Beag. The complicated structure of the geology surrounding the loch has given rise to a wider dispersal pattern in Zn in comparison to either Pb and Cu. It is not surprising that the Tertiary activity is leading to enhancement in Pb. However, Zn appears to be less influenced by this activity which can be observed by lower values over the dyke convergence-point immediately adjacent to the loch. It can therefore be assumed that Zn is a better indicator of anthropogenic contamination over this type of terrain (i.e. where the dykes tend to mask the effects of lead mining). The relative mobility of Zn in soils is controlled chiefly by pH and therefore in increasingly acid soils (peaty-podsols) we can expect greater mobilisation. Where acid mine spoil has been dispersed in clean-up operations then we are likely to see larger dispersal patterns due to wider surface contamination. Copper values are low with background values in the order of < 20 ppm and are not significantly high in the surface of the Loch Lossit core where a Cu maximum 38 mg kg^{-1} was recorded. In the absence of radiocarbon dates within the profile the established time frame within the core is based upon linear extrapolation of the ^{210}Pb-derived sediment rate of 1.6 mm yr^{-1}.

Pollen analyses failed to recognise a rise in Pinus pollen coincident with large scale estate planting in the late 19th and early 20th centuries. The low Pine content within the upper portion of the core was due to the fact that the pollen has failed to enter the sediment possibly owing to its concentration by flotation at the loch margins. The results from the calculated ^{210}Pb inventory show that in the Loch Lossit sediment this radio nuclide was in much lower concentration (230 Bq kg^{-1} ± 10.6 at 1 sigma) at the sediment surface in comparison to Loch Bharradail (338 Bq kg^{-1} ± 28.4 at 1 sigma.). This is possibly due to the size of the Lossit catchment (Catchment:loch ratio =7.1) in comparison to that surrounding Loch Bharradail (Catchment:loch ration = 39.8). The lower volume of drainage into Lossit may account for the observed ^{210}Pb deposition trend. The results of chemical analyses of the sediment from Loch Lossit has provided evidence of mining within the catchment. The sustained peak in Pb at 102 cm is attributed to major influx of Pb and exploitation of this metal appears to be the likely cause. The next chapter will discuss the implications of Loch Leathann were no mining occurred within its catchment.

CHAPTER 8: LOCH LEATHANN

8.1 Introduction

The identification of a suitable loch as a control site has been an integral part of this study. As with the reagent blank, used to measure the bias and precision of the chemical analyses (Chapter 3, Section 3.7) another important aspect is the control population or relevant control site (Black 1988). If the results of the study in a given area are to be judged as high, low, or insignificant, the results must be compared to the results obtained from another area (i.e. the control site). Initially, Loch Fada (Grid Reference NR 408638) was predicted as being a suitable loch for a control but owing to its shallowness and emergent aquatic vegetation this loch was rejected in favour of Loch Leathann. It was assumed that the chemical record from the loch would provide an inventory reflective of a catchment not affected by lead mining. The results from sedimentary chemical analyses would then provide a set of 'benchmark' values to compare against the heavy metal inventories from Lochs Lossit, Finlaggan and Bharradail. In theory sediment from these lochs should have increased Pb, Zn and Cu concentrations resulting from contamination derived from local lead mining. At the outset the following criteria were addressed:

(1) Is there evidence of lead extraction within the Leathann catchment?

There had to be no evidence of lead mining within the Leathann catchment. Examination of geological maps, documentary evidence, aerial photographs and field-walking on the limestone region adjacent to the loch showed that there was no surface evidence for lead mining.

(2) Is the geology similar to the other lochs under investigation?

In order to maintain a degree of geological conformity between the control loch and the others previously mentioned, a loch was needed that shared the same geological characteristics as the lochs Finlaggan, Bharradail and Lossit. The loch did indeed share the same geological characteristics as those above.

(3) Has the influence of (2) led to surface-soil contamination within the Leathann catchment, and could this be observed within the loch's sedimentary chemical inventory?.

Soil metal values needed to be at normal background level in order to support the fact that catchment derived metals would have been in order of magnitude lower than in the lochs associated with nearby mine sites. The Domego Resources Limited (DRL) geochemical survey covered the catchment of the loch and the values recorded were within normal 'background' levels observed in uncontaminated soils.

8.2. Location

8.2.1 Topographical setting

Loch Leathann (Plate 15, Appendix 1) is a remote spring-fed loch at an altitude of 140 m OD (Table 8.1). The loch is irregular in shape with a narrow bay at its northern end. A wider bay is situated on its western shore. The shoreline has a substrate of small rocks and stones with sand and silt being more predominant in the north bay. The main inlet is via a small spring situated on the northern shore. The spring water had a conductivity of 440μmhos with a pH of 7. The main outlet is via a small sluice in the western bay. The pH at the southern end of the loch was recorded as 7.1 with a conductivity at 84 μmhos. The loch forms the head water of the Barr River that eventually joins the River Laggan. The topography adjacent to the loch is undulating with sloping ground towards the Barr River. At the turn of this century the loch was periodically dammed and allowed to bring the Barr River into spate promoting optimum conditions for game fishing. No dates for the end of this practice are available in the estate records and the sluice has not been maintained for many years (D.J. Mcphee & D. Bell pers comm.)[1] Today the loch still supports a healthy population of wild brown trout.

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[1] Donald James MacPhee is the Headkeeper for the Dunlossit Estate, Islay.
Donald Bell is the tennant farmer of the land surrounding Loch Leathann.

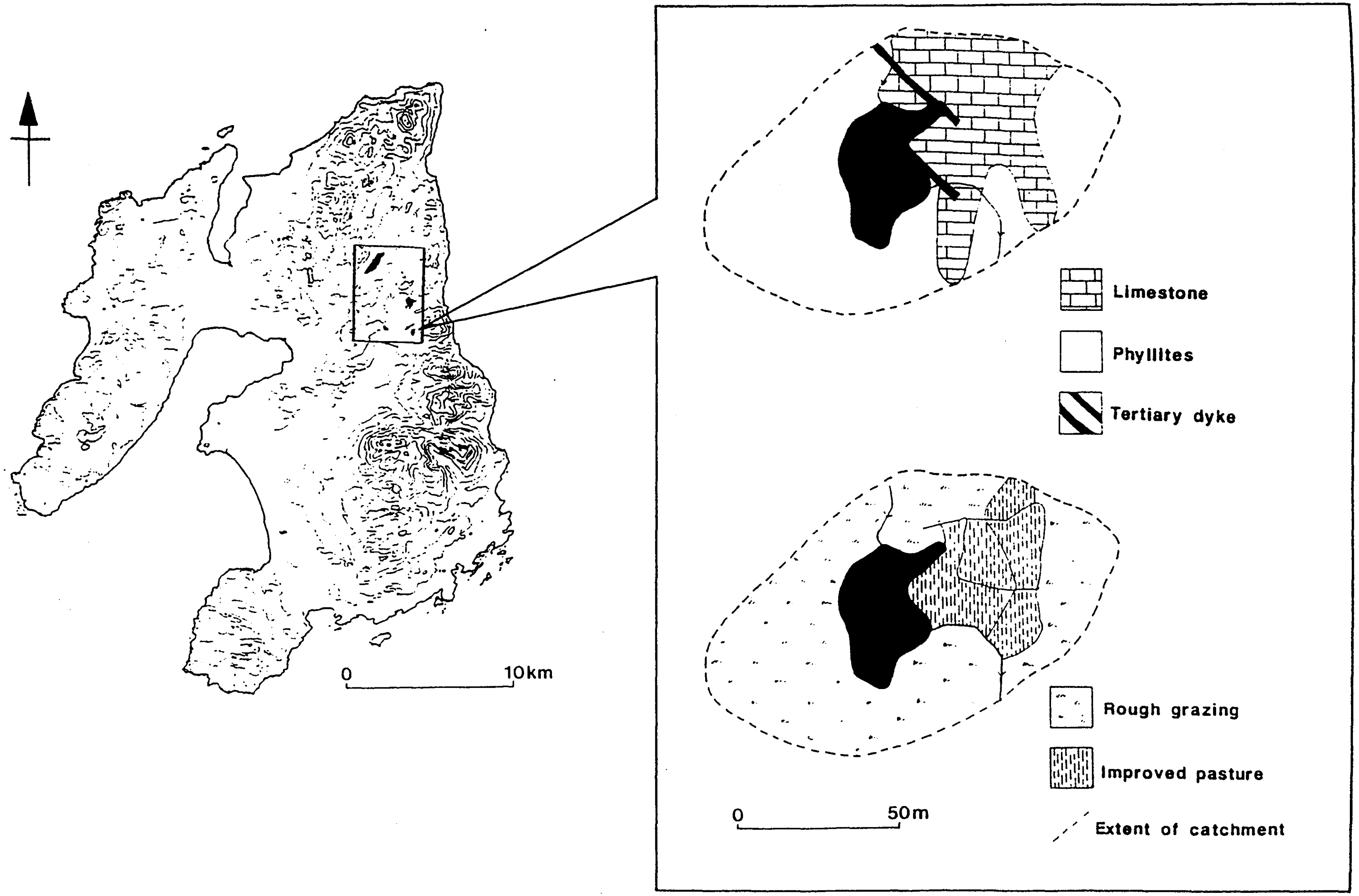


Figure 8.1 Location of Loch Leathann with geology and present day land use.

Grid reference	NR 633412
Core date	September 1992
Catchment geology	limestone-Phyllites-shales
Catchment type	improved & rough grazing
Loch altitude (m)	120
pH	7.1
Conductivity (μmhos)	440 (spring) 84 (loch)
Max. depth (m)	12.19
Loch area (ha)	5.9
Catchment area (ha)*	27.8
Catchment:loch ratio	4.7
Afforestation (%)	0
Net relief (m)	140

Table 8.1 Loch Leathann ~ site characteristics. (note* excluding the loch).

8.2.2 Geology (Figure 8.1)

The geology surrounding the loch is formed from two distinct and starkly contrasting geological types. To the south and west of the loch the geology consists of acidic Phyllites and shales. Dalradian Limestone is present on the north and eastern side of the loch. Here the limestone forms a spur that is intersected by a small outlet stream. Tertiary activity is represented by two basalt olivine dykes that trend in a north by north-westerly direction.

8.2.3 Soils, vegetation and catchment land use (Figure 8.1)

The Dalradian Limestone supports the Deecastle Association soil type (MCURI). The acidic Phyllites and shales have promoted both the Foundland and Durnhill soil series. Both soils are relatively shallow and acidic, derived from fluvio-glacial drift deposits. The soils above the eastern shoreline are exposed in section to a height of 1.5 m and contain glacial drift with well defined podsols immediately above. In general, the vegetation is sparse along the shore margins except in the north bay where the vegetation includes the species *Equisitum fluvitile, Iris pseudacorus, Juncus bulbosus, J.affusus, Litterella uniflora, Myriophyllum alterniflorum, Carex demissa* and *Myrica gale.*

Away from the loch the vegetation is dominated by flying-bent moorland interspersed with *Calluna vulgaris* and stunted *Myrica Gale* in the wetter hollows. There has been some land management within the catchment over the last century mainly with improved pasture on the limestone belt that nowadays supports grazing by sheep and cattle. The predominant vegetation on the acidic Phyllites and shales are the heath-moor land types *Calluna vulgaris* and *Molinia* grassland that provides rough pasture. About 1km downslope from the loch there is a large tract of former peat beds that were dug extensively in the 19th century for commercial and domestic fuel.

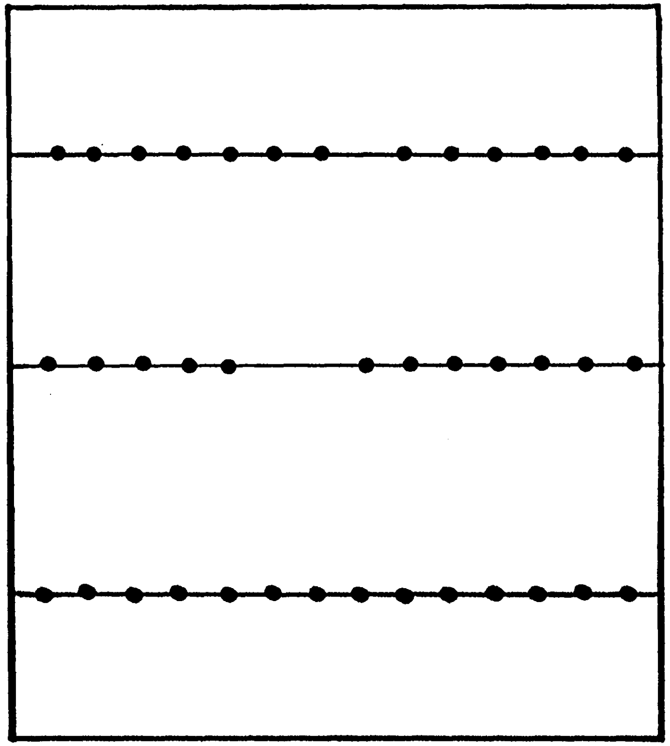
8.3. Geochemical Environment

8.3.1 The catchment distribution of Pb, Zn and Cu based on the DRL geochemical survey

Soil geochemical survey was undertaken around Loch Leathann in 1983. The data produced from this survey show extremely low concentrations for Pb, Zn and Cu within the immediate vicinity of the loch. This evidence supports the theory that no Pb mineral veins outcrop near the soil surface. The data (Table 8.2) complements the results from the analyses of the loch sediment metal concentrations and show that metal contamination has been the result of natural agencies except for the near-surface sections of the sediment that have received an atmospherically derived 'pollution' component. The DRL data has been interpolated into two dimensional contour plots to show the distribution of these metals within the loch catchment. The values for Pb, Zn and Cu are in sharp contrast to the surface metal concentrations in the other loch catchments described previously. As has been shown, the values for Pb, Zn and Cu in the other loch catchments can be seen to be much higher, owing to either natural mineralization, land-use practices and lead mining activity. Metal input of Pb, Zn and Cu will be seen to be lower in the Leathann loch sediment. In all cases metal concentrations will be controlled by the catchment geochemistry, past land-use practices and importantly recently atmospherically derived pollution.

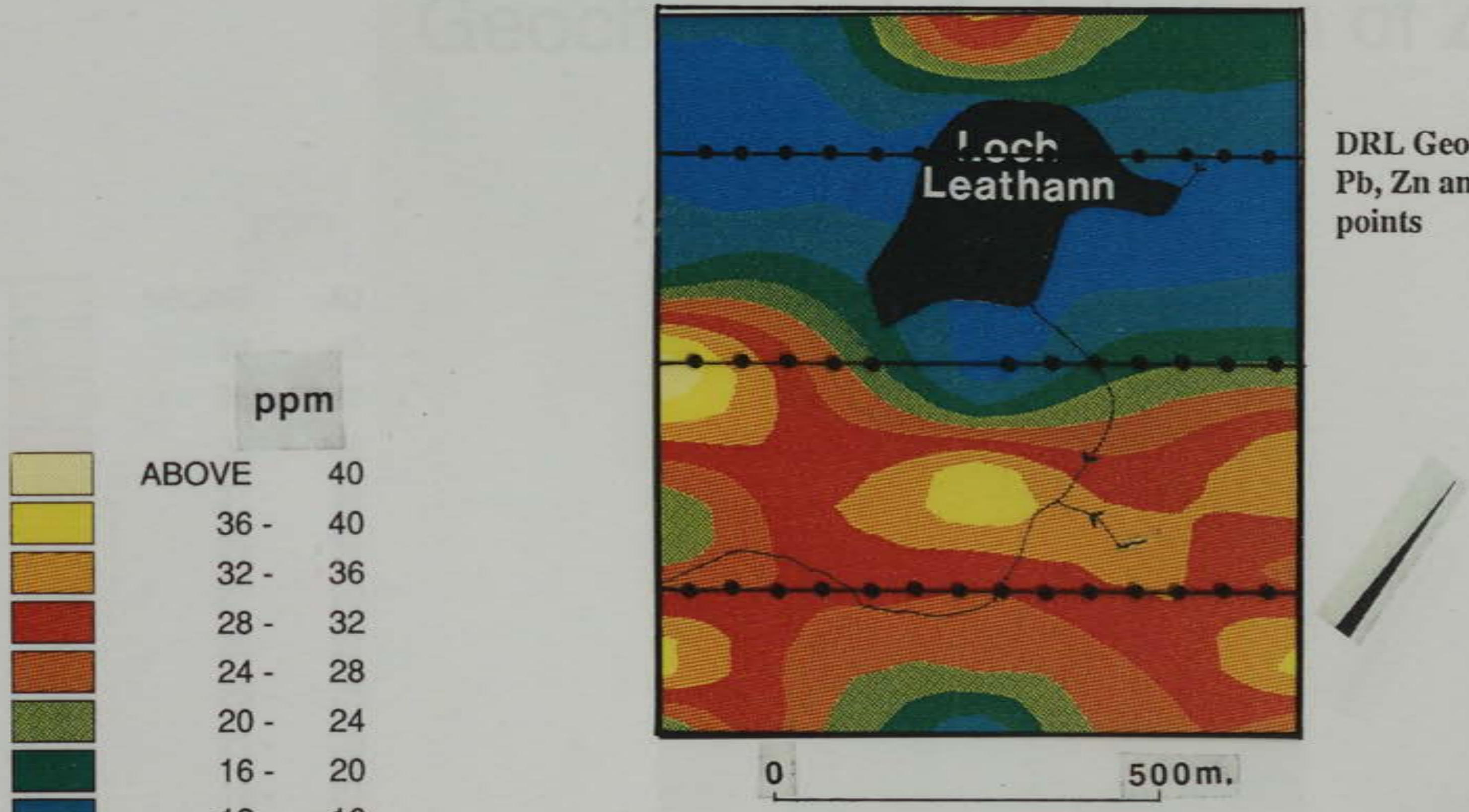
STATISTICS	Pb	Zn	Cu
Observations	33	33	33
Maximum value	55	75	62
Minimum value	5	5	2
Mean value	22	24	5
Median value	20	15	3
Standard deviation	15	19	10

Table 8.2 Statistical characteristics for Pb, Zn and Cu concentration based on the DRL geochemical data (values in ppm).



**DRL Geochemical Survey
Pb, Zn and Cu soil sample
points**

LOCH LEATHANN CATCHMENT
Geochemical distribution of Pb



DRL Geochemical Survey
Pb, Zn and Cu soil sample
points

Figure 8.2 Distribution of lead in surface soils in the Loch Leathann catchment (after Domego Resources Ltd 1983).

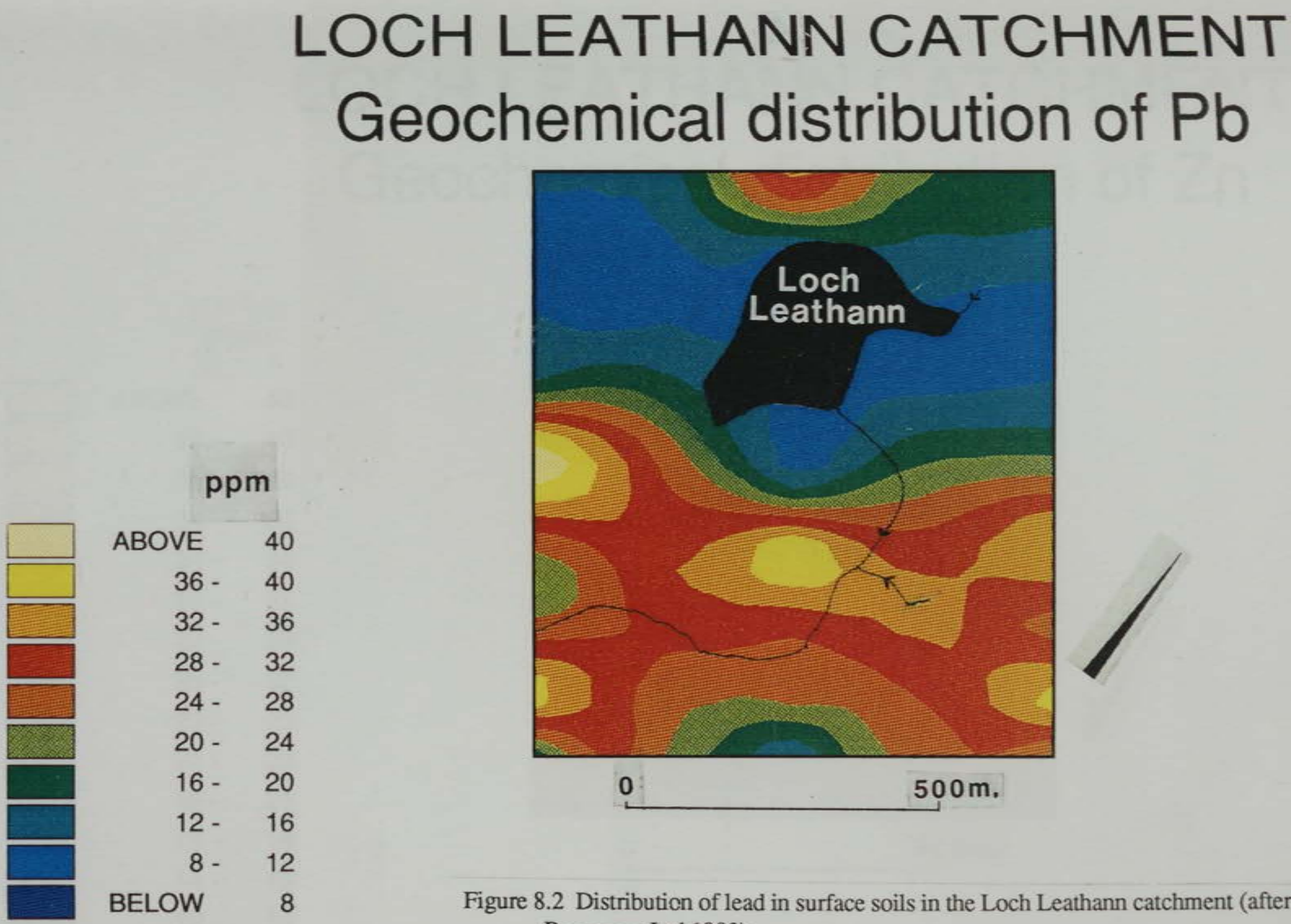


Figure 8.2 Distribution of lead in surface soils in the Loch Leathann catchment (after Domego Resources Ltd 1983).

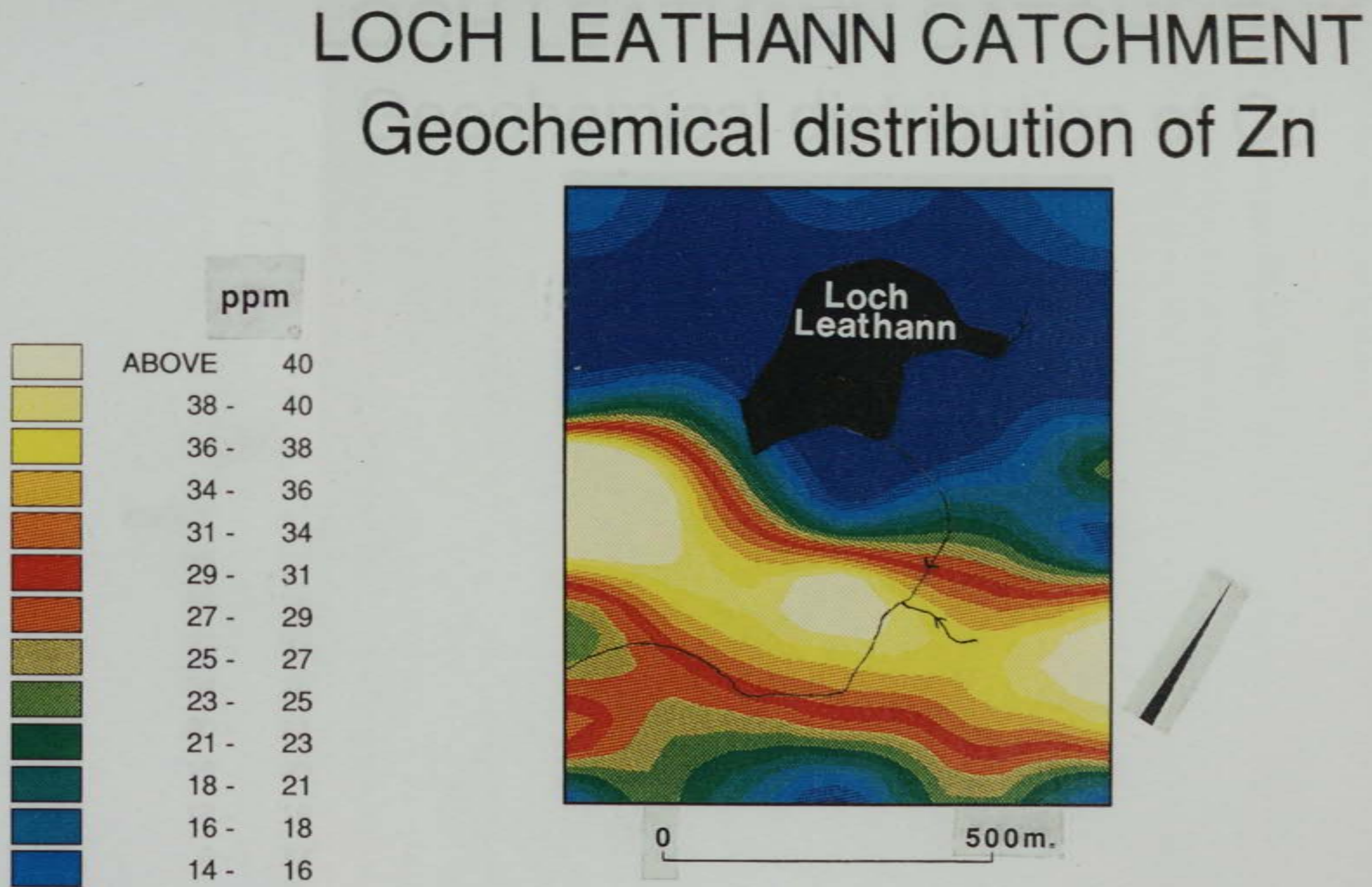
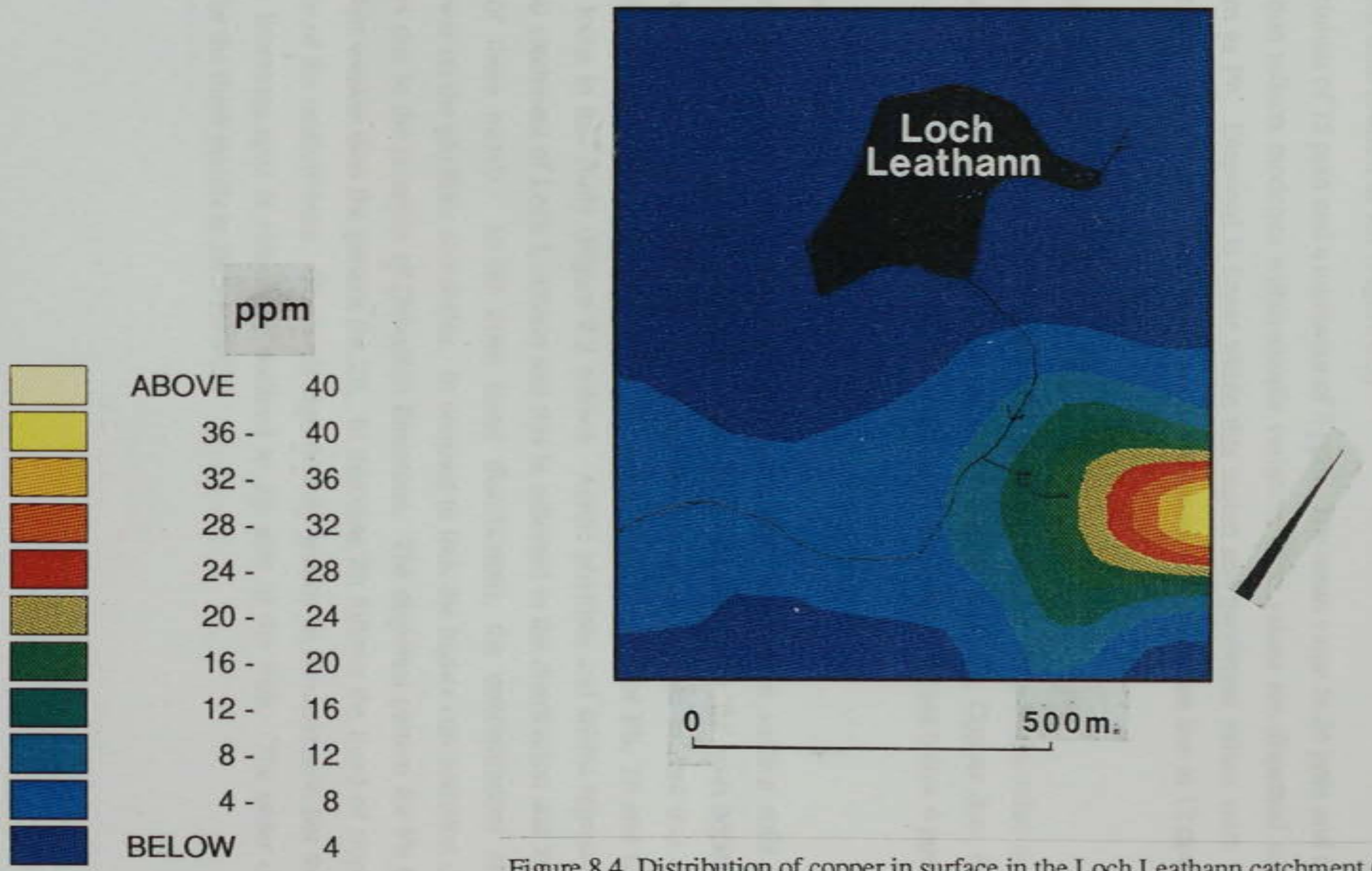


Figure 8.3 Distribution of zinc in surface soils in the Loch Leathann catchment (after Domego Resources Ltd 1983).



LOCH LEATHANN CATCHMENT
Geochemical distribution of Cu

Figure 8.4 Distribution of copper in surface in the Loch Leathann catchment (after Domego Resources Ltd 1983).

8.3.2 Catchment distribution of Pb (Figure 8.2)

Lead has a maximum value of 33 and a minimum of 5 ppm respectively. The mean is 22 ppm and the standard deviation of 15 ppm shows low within sample variation for this set of values. Lead is dispersed assymmetricaly to the east of the loch where a maximum of 50 ppm is attained. The lowest values for Pb are within the near shore regions of the loch where values are below 12 ppm. The overall mean value established from the total surface result divided by the number of samples is 10 ppm.

8.3.3 Catchment distribution of Zn (Figure 8.3)

Zinc has a maximum of 75 ppm and a minimum of 5 ppm. The mean value is 24 ppm and the standard deviation reflects moderate within-sample variation. Zinc values are dispersed with the same pattern as Pb. Dispersal is linear within this region and maximum values were 75 ppm. Again as with Pb the values for Zn adjacent to the shores of the loch are low at 12 ppm.

8.3.4 Catchment distribution of Cu (Figure 8.4)

Copper has a maximum of 62 and a minimum of 2 ppm respectively. The mean value is 5 ppm with a standard deviation reflecting moderate within-sample variance. Copper does not follow the trend of distribution as Pb and Zn. The vicinity of the loch has values below 4 ppm.

8.3.5 Assessment on the observations of Pb, Zn and Cu

It has been mentioned above and in Section 2.3.2, that metal dispersal in surface soils is directly related to parent geology, and degree of slope. A metals affinity to 'fix' with organic matter and its solubility resulting from pH is also of importance. It is quite evident that all three metals are very low when compared to the catchment distributions for Pb, Zn and Cu from the other lochs in this study (Figure 9.2 below). Acidic phyllites and shales represent 85% of the total catchment of Loch Leathann and this is reflected in the distribution and low concentration of these metals. In all three metal distributions, the concentrations are significantly lower on the phyllites and shales. In contrast to this, the higher concentration of Zn to Pb occurs due to the presence of Dalradian limestone. The dispersal pattern for Pb is much broader but weaker than the pattern for Zn. In this case Zn follows the trend of slope along the course of the outflow burn. Copper is significantly weaker in concentration but the position of the limestone spur is tentatively outlined to the east of the loch. The order of concentration for the three metals is Zn > Pb >Cu.

Bathymetric map of Loch Leathann

Depth in feet below Summer Water Level (21 May 1992)

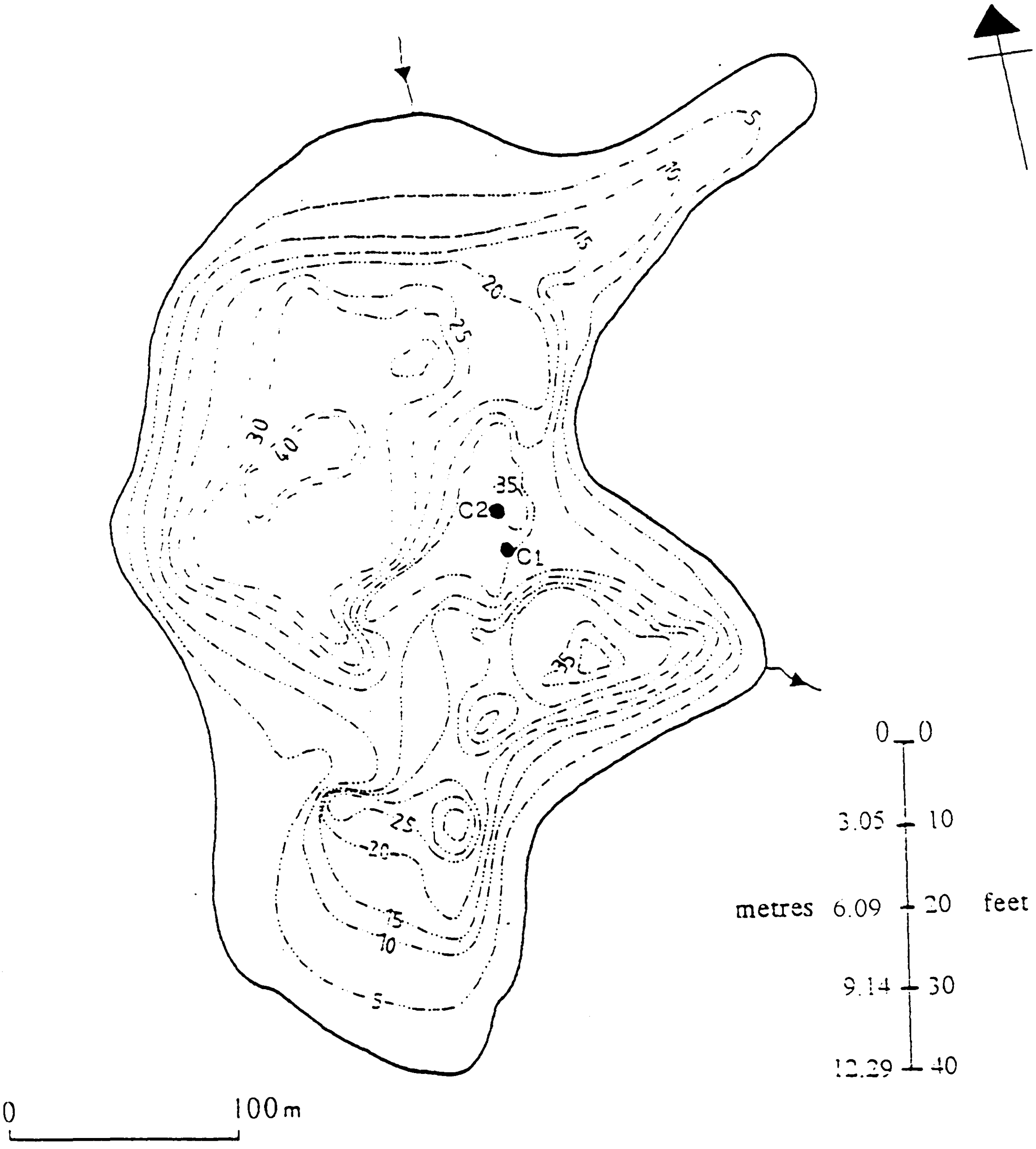


Figure 8.5 Bathymetric survey of Loch Leathann and coring stations.

8.4 Bathymetric Survey of the loch and core recovery

The principal aim of the survey was to establish the morphometry of the loch and define areas suitable for obtaining cores. The survey was carried out in September 1992 using the method outlined in Chapter 3 Section 3.4. The contour survey shows the depth in feet below summer water level on the 21st May 1992. The survey established that the loch had a maximum depth of 40 ft. (*c.*12.29 m) and was probably fomed by scouring in the glacial period. The western shore was seen to shelve rapidly to the area of greatest depth. In contrast, the northern, eastern and southern bays shelve only gradually to form the central basin of the loch. Two cores were obtained from the positions marked on the bathymetric survey map (Figure 8.5) using the device in Plate 9, Appendix 1 under operating procedures defined in Chapter 3 Section 3.5. When both core tubes were split for sediment sampling, Core 1 was seen to be very disturbed in the upper 20 cm section and it was considered that the sediment would be of little use in this study. For the establishment of chronologies based on ^{210}Pb it is of critical importance that the upper portion of the core shows no evidence of mixing or slumping. The sediment in Core 2 was seen to be in a good state and showed no evidence of disturbance.

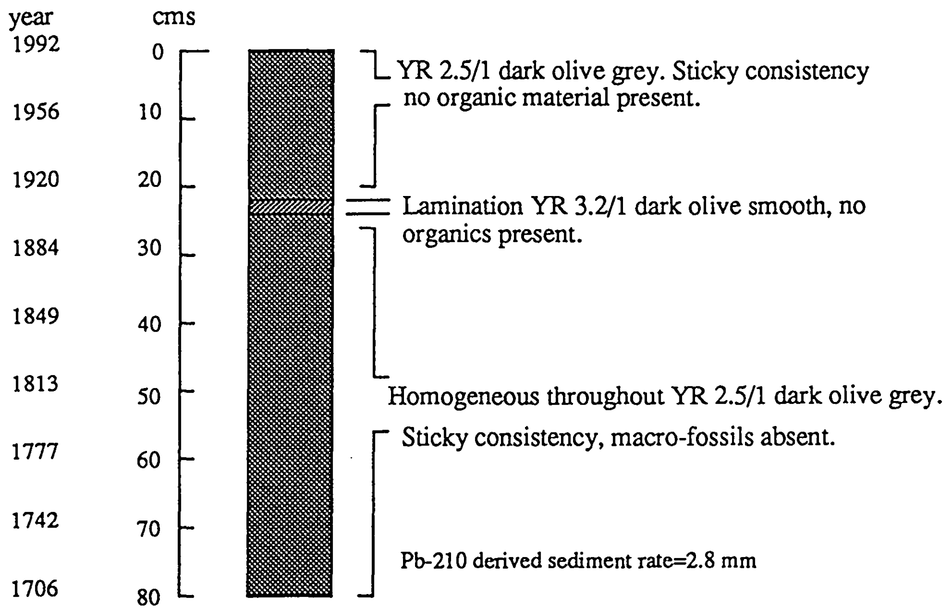


Figure 8.6 Descriptive summary of Core 2 sediment characteristics.

8.4.1 Sediment characteristics of Core 2

The sediment of Core 2 is similar to the loch sediment recovered from Lochs Bharradail, Lossit and Finlaggan. As with these lochs, the sediment shares the same homogeneity in colour, texture and consistency. The Leathann sediment comprised a very dark olive-black lake mud. The sediment was anoxic and very odorous giving off a strong smell of sulphur dioxide.

X-ray diffraction analyses on selected sediment samples revealed that the dominant mineral phases within the sections (see Appendix 2) where muscovite and quartz. These are the main constituents of weathered mineral particles and form a common detrital component in freshwater lake sediment (Jones and Bowser 1978). X-ray diffraction failed to identify any significant differences in mineral composition between the lamination found at 23 cm and the sediment above and below this horizon.

8.5 Dating and Environmental History

8.5.1 ^{210}Pb analyses and the formation of an annual rate of sedimentation

The methodology and limitations of this dating method have been described in Chapter 3 Section 3.8.2) The results of ^{210}Pb analyses and data manipulation are shown in Figure 8.7 and 8.8). The results show that $^{210}Pb_{excess}$ activity has been detectable to a depth of 50 cm. A sub-surface maximum (2-3 cm) of 234.4 ± 24.3 Bq kg^{-1} at 1 sigma has been attained. The data has been decay corrected for September 1992 and is shown linearly regressed in Figure 8.8 and also plotted with 2 sigma error bars.

Using the formula already described, the data allow a ^{210}Pb-derived sedimentation rate of 2.8 mm yr^{-1}. Linear extrapolation based on the explicit assumption of steady state conditions throughout the loch's sediment formation provide the dates that are positioned alongside the core in Figure 8.6 above. The decay profile (Figure 8.7) indicates that some slight mixing may have occurred at 1-2 cm. The trend from 2-3 is one of exponential decay to 7-8 cm. After this position there is a slightly erratic decay curve but the results are good when compared to the Finlaggan excess ^{210}Pb which shows severe sediment mixing due to reasons already explained in Chapter 5 Section 5.5.4. In comparison to the other lochs in the study the annual rate of sedimentation in Loch Leathann is the highest. The ^{210}Pb derived sediment rate of 2.8 mm yr^{-1} is relatively rapid and this may be accounted for by the erosion and general instability of exposed soils, water residence time and sediment distribution within the loch. The loch:catchment ratio of 4.7 was the lowest among the four lochs examined in this study. The loch's catchment is high (>140 OD.) and the input of sediment via seepage is relatively high, particularly in adverse weather conditions. Further sediment influx may be derived from a spring that flows into the loch from the limestone spur outcropping on the northern shore.

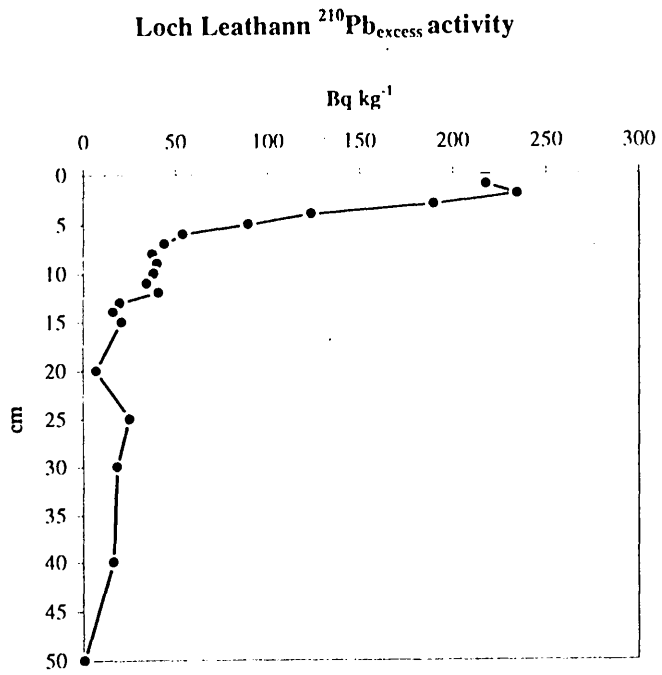


Figure 8.7 Depth versus $^{210}Pb_{excess}$ activity

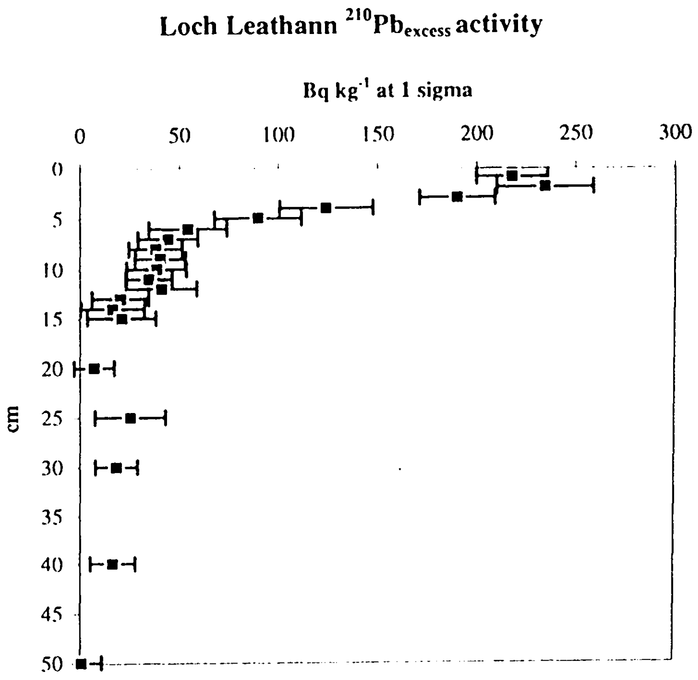


Figure 8.7 Depth versus $^{210}Pb_{excess}$ with error bars at 1 sigma.

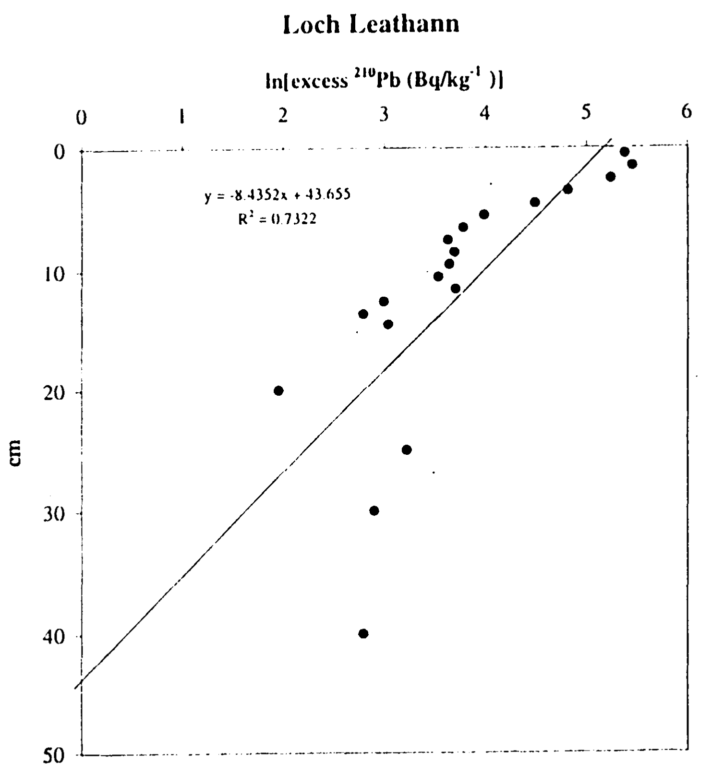


Figure 8.8 Linear regression plot of depth versus log normal $^{210}Pb_{excess}$ activity.

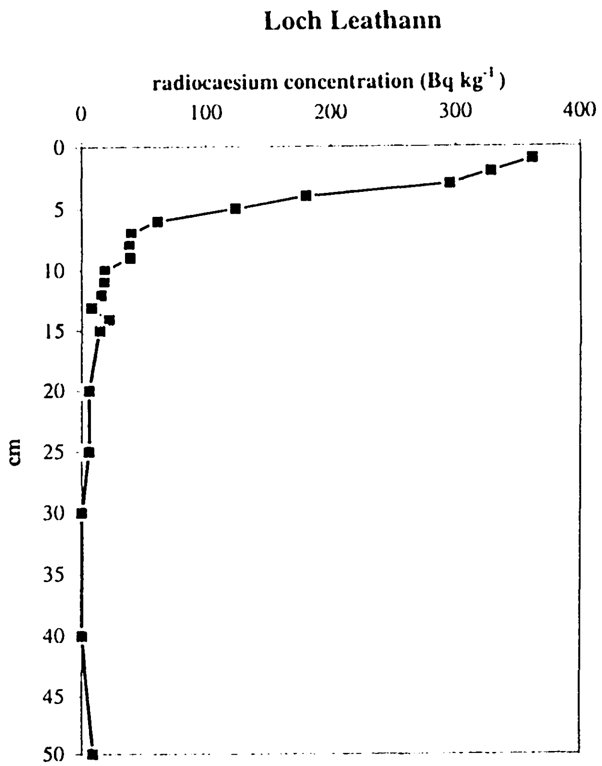


Figure 8.9 Depth versus ^{137}Cs activity.

Sediment sections were not submitted for radiocarbon determination owing to the presence of limestone within the catchment and the short time span that was estimated to be present across the length of the core (0-80 cm). The ^{210}Pb-derived time-frame already described has a supporting ^{137}Cs decay inventory which is discussed below. It has already been mentioned that the loch was artificially dammed to promote spate conditions in its outflow river, the river Barr. No documents survive recording the building of a small sluice on the eastern side of the loch. However, following discussions with the present Gamekeeper (Mr D.J.McPhee) it seems that the sluice was built at the turn of the century. It is argued that the lamination can be attributed to sediment focusing brought about by the lowering of the loch. This lamination is shown in Figure 8.6 at 23-24 cm which according to the annual sediment rate of 2.8 mm yr^{-1} dates to 1903, which would be within agreement with the introduction of a sluice to flood the Barr river.

8.5.2 Chronological support using ^{137}Cs

Radiocaesium has been detectable from 1-30 cm. Figure 8.9 shows the near surface ^{137}Cs concentration (0-1cm) is at 361.4 ± 4.9 Bq kg^{-1} at 1 sigma. The decay curve shows exponential decay from this position to 7 cm where the radioactive decay profile is vertical to 9 cm. Further decay from this point is steady to 13 cm with 7.6 ± 1.5 Bq kg^{-1} at 1 sigma. There is a rise to 22 ± 1.4 Bq kg^{-1} at 14 cm showing that there may have been some re-suspension of material with higher radio caesium content. This may be due to the smearing effects by the coring device. From 14 cm the decay profile reaches the baseline at 30 cm where ^{137}Cs activity is depleted. The upper surface of the profile has received a large proportion of the Chernobyl derived ^{137}Cs that has decayed exponentially from the upper region of the sediment down to 7 cm. The position where the activity becomes vertical is assumed to be what remains of the nuclear weapons testing maximum of 1963. It has been mentioned previously that ^{137}Cs is prone to vertical and lateral diffusion in the pore-waters of freshwater lake sediment (see Chapter 3 Section 5.5.5 and 9.6.3). Taking into consideration the general limitations of ^{137}Cs due to its tendency to be relatively mobile it can be concluded that the ^{137}Cs profile is in broad agreement with the proposed ^{210}Pb derived sediment rate. This aspect is devloped in more detail below.

8.6 Geochemical Inventory and Physical Properties

Analytical procedures undertaken are the same as have been described in Chapter 3, Section 3.7.2. Volume magnetic susceptibility was undertaken on dry sediment using the procedure described in Section 3.6.5. Pollen analyses for the species *Pinus sp.* was carried out as described under section 3.9.

Statistics	Pb	Zn	Cu	Ca	Fe	Mn
Observations	56	56	56	56	56	56
Maximum value	205	103	15	185	1870	2000
Minimum value	10	35	3	25	970	625
Median value	18	48	7	110	1280	950
Mean value	30	53.2	7.2	110.7	1303	1047
Standard deviation	38.4	14.9	2.6	29.1	187	316

Table 8.3 Summary statistics of the data set for Loch Leathann chemical analyses, values in mg kg^{-1}.

Lead (Figure 8.10)

Starting from the base of the core Pb concentrations are at *c.*30 mg kg^{-1} and fluctuate up to 52 cm where the concentration is 40 mg kg^{-1}. From 22 cm the Pb values rise exponentially from 22 mg kg^{-1} to a sub-surface peak of 205 mg kg^{-1}.

Zinc (Figure 8.11)

Basal concentrations of Zn are at 49 mg kg^{-1} and rise only gradually at 38 cm were 62 mg kg^{-1} are attained. A decline in concentration can be observed at 28 cm (*c.*45 mg kg^{-1}) and from this position a sharp rise at the sub-surface give values at 88 mg kg^{-1}.

Copper (Figure 8.12)

Copper values from the base upwards are low and below 8 mg kg^{-1} until a slight rise at 54 cm with 11 mg kg^{-1}. From this position the values decline to 4 mg kg^{-1}. An exponential rise occurs from this position to the surface at 12 mg kg^{-1}.

Calcium (Figure 8.13)

Basal concentrations of 120 mg kg^{-1} are attained for this element. From the base to *c.*24 cm the values rise and fall intermittently. A significant decline in concentration can be observed at 24 cm at 30 mg kg^{-1}. From this depth the concentration of Ca rises very sharply to a maximum of 190 mg kg^{-1}. From this depth the values decline towards the sub-surface at 90 mg kg^{-1}.

Manganese (Figure 8.14)

The concentration of Manganese at the base of the core is at 1300 mg kg^{-1} and declines erratically to 74 cm at 600 mg kg^{-1}. From this depth the concentration rises slightly to 1000 mg kg^{-1} at 44 cm. From this depth the values decline slightly then peak at 22-24 cm where

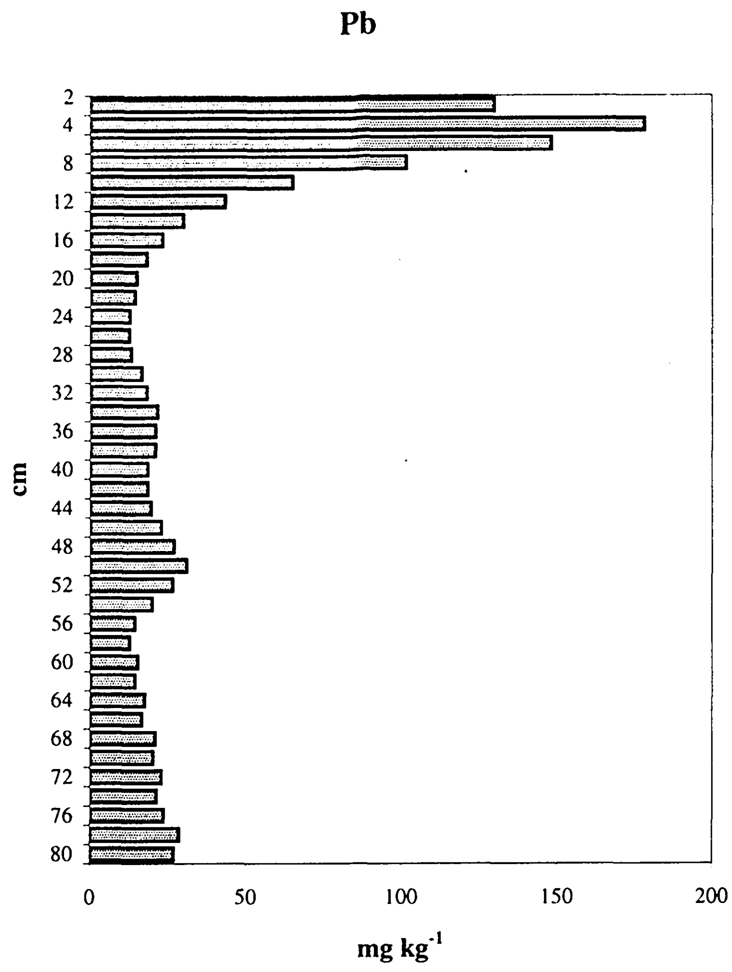


Figure 8.10 Depth versus lead concentration.

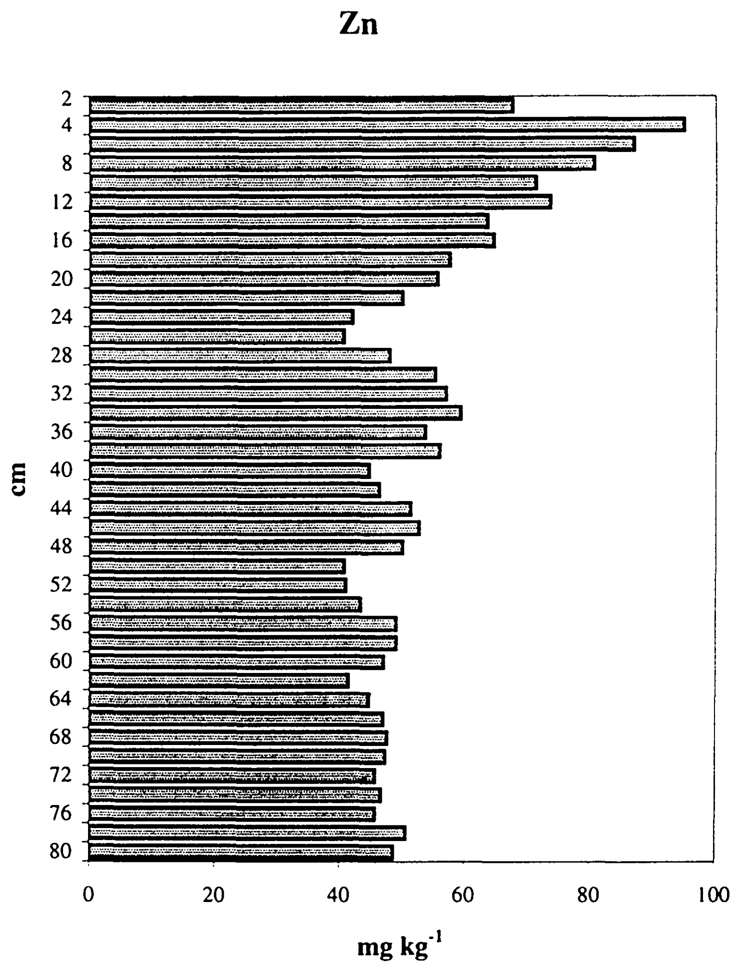


Figure 8.11 Depth versus zinc concentration.

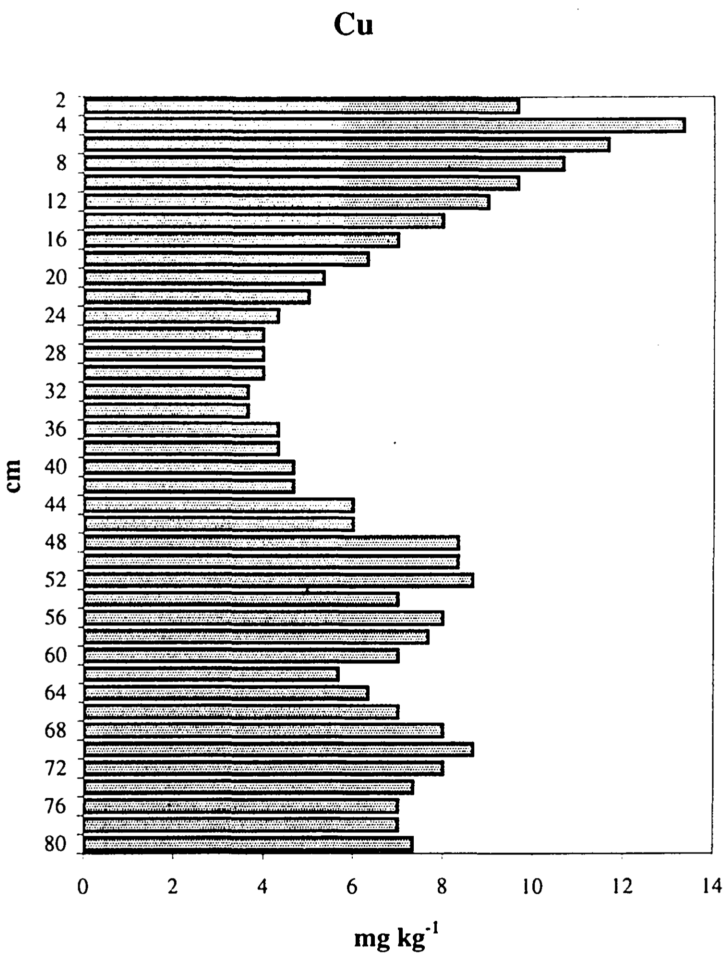


Figure 8.12 Depth versus copper concentration.

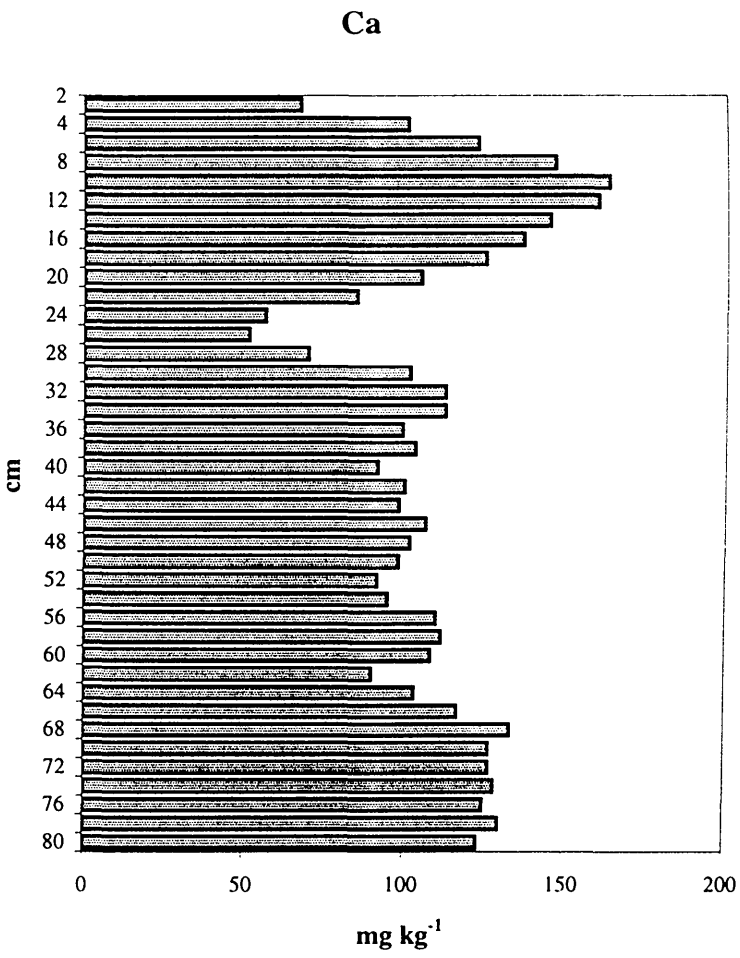


Figure 8.13 Depth versus calcium concentration.

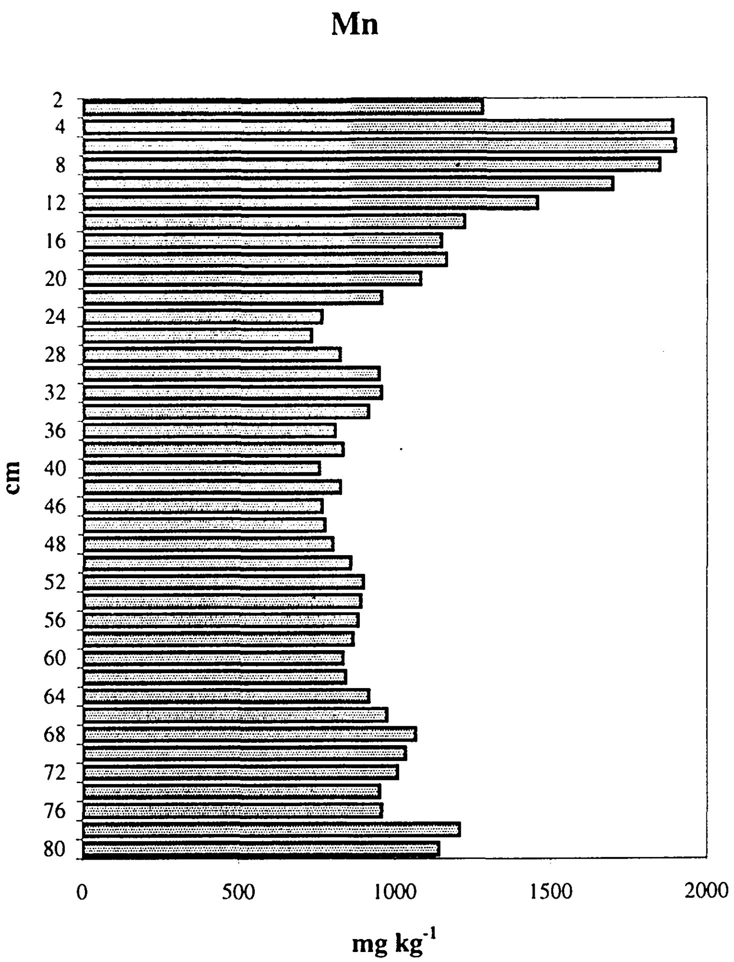


Figure 8.14 Depth manganese iron concentration.

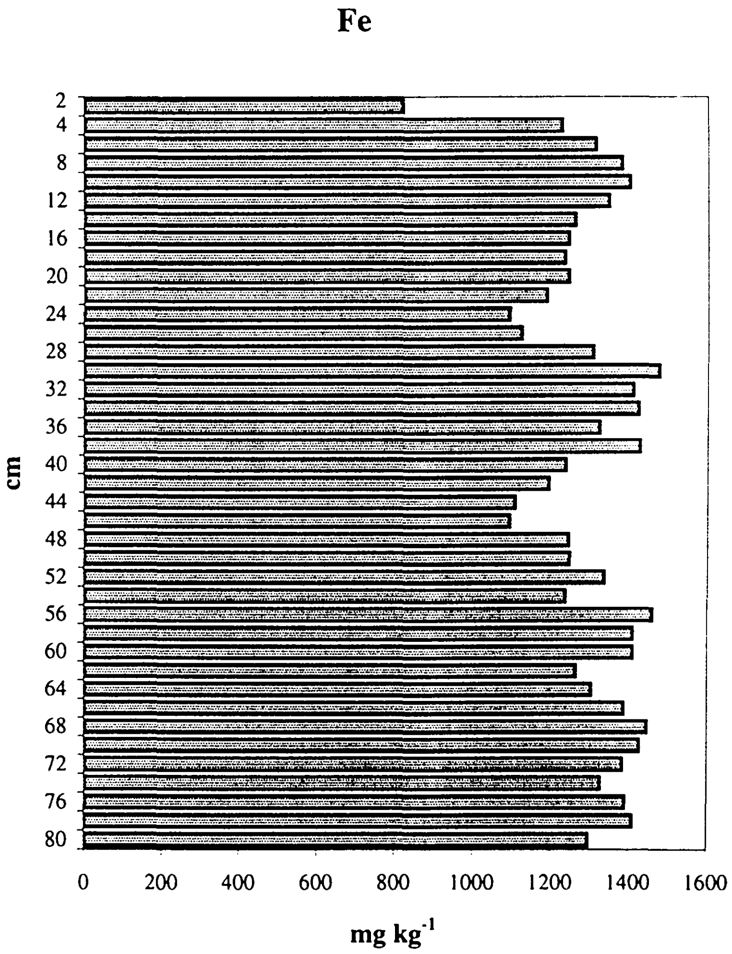


Figure 8.15 Depth versus iron concentration.

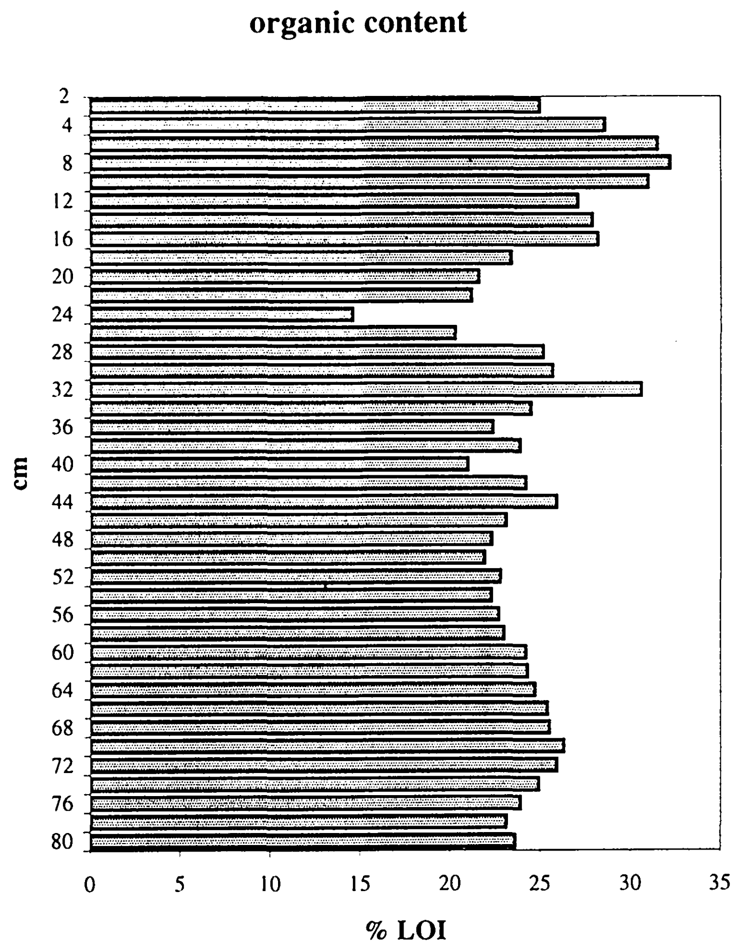


Figure 8.16 Depth versus organic content (350 C).

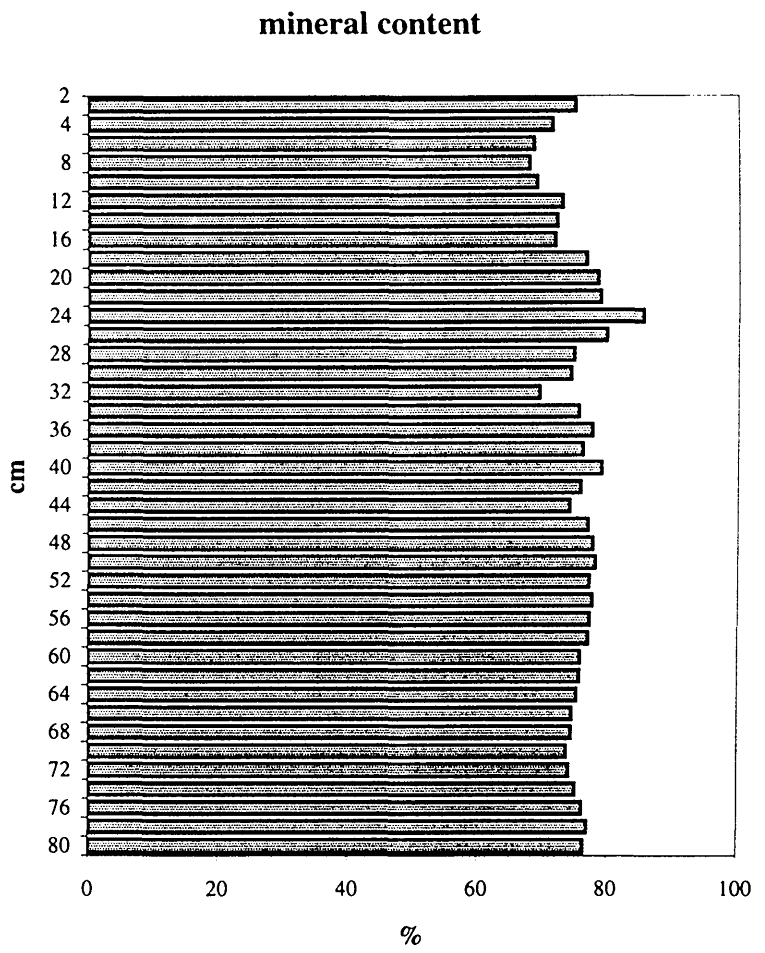


Figure 8.17 Depth versus mineral concentration.

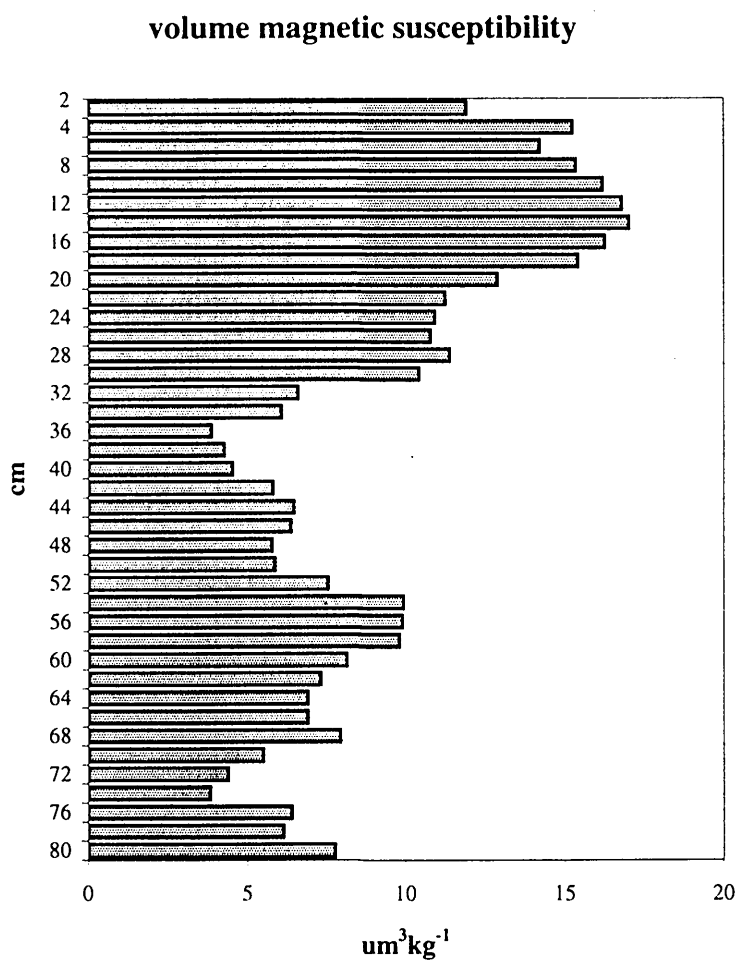


Figure 8.18 Depth versus volume magnetic susceptibility.

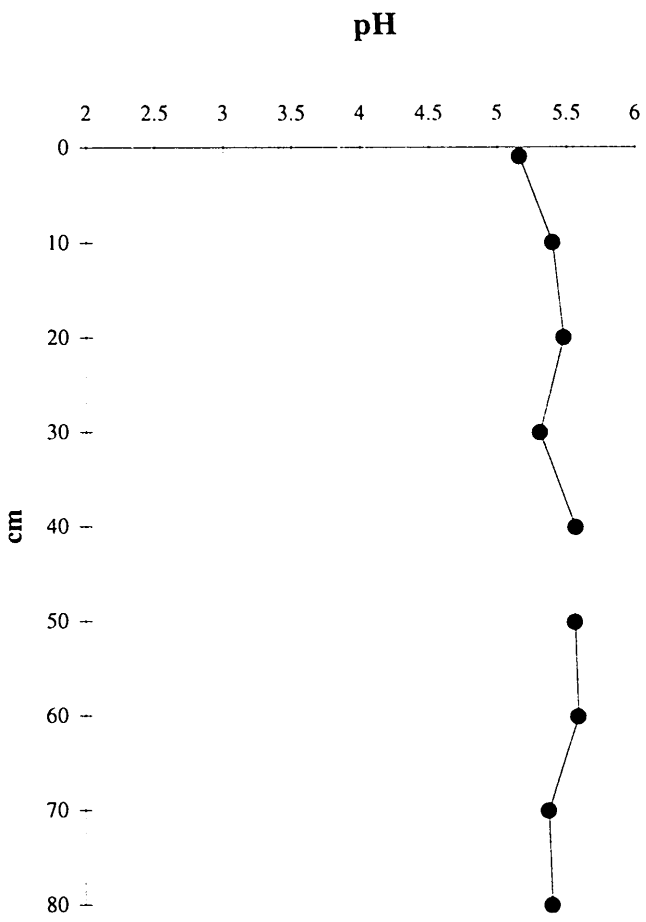
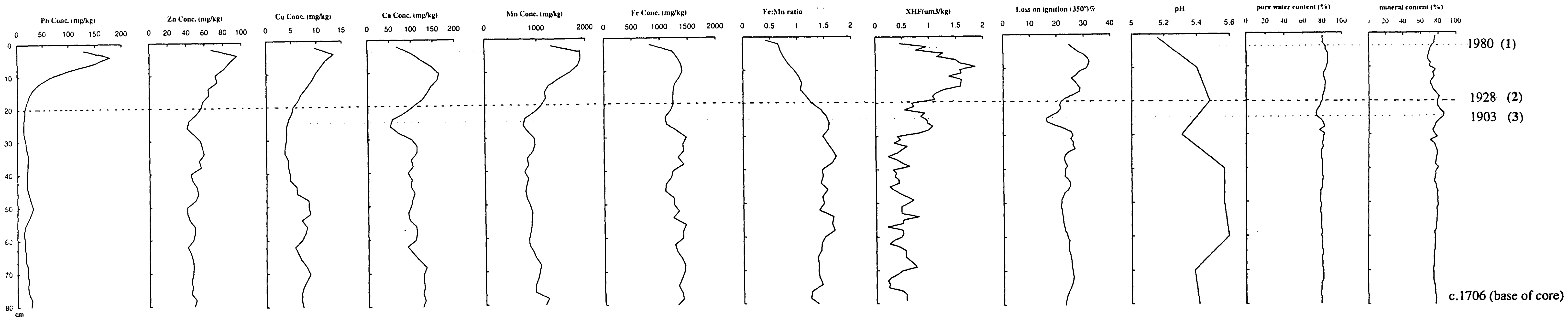


Figure 8.19 Depth versus pH concentration.

Loch Leathann Geochemical Summary Diagram



Extrapolated ^{210}Pb Date	Event in the profile	Interpretation
(1) c.1980	Maximum in Pb, Zn and Cu	Atmospheric fall-out deposition
(2) c.1928	Exponential rise in Pb, Zn and Cu. Sharp rise in mag sus.	Onset of atmospheric fall-out deposition
(3) c.1903	Decrease in LOI. Slight rise in mineral content.	Lamination horizon at 23cm. Dam built to flood the outlet burn, resulting in sediment focusing.

Summary of possible dates and events within the Loch Leathann geochemical diagram based on an extrapolated annual sedimentation rate of 1.8 mm yr^{-1}.

Figure 8.20 Loch Leathann geochemical summary diagram.

Manganese cont....

the maximum value of 1,750 mg kg^{-1} are attained. From this region the concentration falls to the sub-surface of 1300 mg kg^{-1}.

Iron (Figure 8.15)

Iron concentrations from the base towards the surface are erratic and range from 1100 mg kg^{-1} to 1400 mg kg^{-1}. There appears to be little variation throughout the profile.

Pore-water content (Figure 8.20)

Water content was established by taking the value from the wet/dry ratio after drying at 50° C for 48h. The values obtained give a rough estimate pore water content and can be related to the relative amount of mineral and organic matter present within the sediment. From the base upwards the water content is 79%. This value continues somewhat erratically to *c.*26 cm. A sharp decline can be observed after this point where the water content is lower at 73% at 26 cm. From this point the values rise exponentially to the subsurface value of 85%. A slight decrease to 80% can be observed at the surface.

Organic content by loss on ignition (Figure 8.16)

At the base of the core the organic content is 23% and continues intermittently up the core to *c.*30 cm and increases slightly to 25%. From here the organic content declines to 16% at 25 cm. A gradual rise occurs with a peak at 7 cm where the organic content increases to 31%. From here the values decline to the near surface at 24%.

Mineral content (Figure 8.17)

The mineral content was obtained by subtracting the organic values from 100% to provide an approximation of mineral content within each sediment section. Although rather crude, it does provide an assessment of relative increases in mineral matter that may relate to an increase of catchment derived material into the sediment. The average mineral content is 75% with a rise at 24 cm to 85 %.

Magnetic Susceptibility (Figure 8.18)

Magnetic susceptibility units fluctuate between 7-11 μm^3 kg^{-1} from the base up to 27 cm. From this point there is a marked increase with a maximum 25 μm^3 kg^{-1} at 10 cm from the surface of the core. The units decline to a sub-surface value of 17 μm^3 kg^{-1}.

pH (Figure 8.19)

The pH concentration has been established at 10 cm intervals down the core. Values in pH units range from between 5.42 at the base to 5.16. The stability observed in the pH profile may also be attributed to a high conductivity (440μmhos) in spring water that enters the loch. The pH values are noticeably higher than the other pH values recorded in lochs Finlaggan, Lossit and Bharradail. The input of spring water from the limestone at the northern end of the loch may have acted as a buffer against the onset of more acidic conditions.

8.6.1 Implications of the geochemical inventory

The general trend of the chemical flux within the Leathann profile is, with the exception of Fe, an increase in metals at or near the surface. The heavy metals Pb, Zn and Cu appear to be enriched in proportion to metal values in the surrounding soils. The Pb profile is in keeping with many other investigations where a greater proportion of the lead has been attributed to atmospheric sources (see below). The Pb maximum at 1-2 cm in the sediment (150 mg kg^{-1}) is enriched by 14% in relation to a mean Pb soil background value of 22 ppm. Zinc has a surface sediment enrichment of 95 mg kg^{-1} and follows the same trend as Pb. The observed maximum for Zn in the surrounding soil is 75 ppm with a mean of 5 ppm. In the sediment, the background concentration is considerably higher at ca. 50 mg kg^{-1} showing a 10% increase in concentration. Copper has an interesting profile with values that are relatively constant at around 8-10 mg kg^{-1} from the base of the core falling at 46 cm to 5 mg kg^{-1}. A sharp rise occurs towards the surface of the core surface with 15 mg kg^{-1}. Against a background concentration of 5 ppm in the surrounding soil, it appears that copper has undergone some enrichment towards the upper region of the core.

The calcium profile also shows some near-surface enrichment. There is a noticeable decline in concentration at 24 cm (25 mg kg^{-1}) which is synchronous with the decline in organic and water content in this sediment section. Iron remains constant throughout the profile ranging from 1300-1500 mg kg^{-1}. This may reflect steady-state conditions within the sediment's formation. Manganese is interesting as there appears to be a near two-fold increase from *c.*900-1000 mg kg^{-1} to 2000 mg kg^{-1} at 1-2 cm. This increase is attributed to diagenetic factors; these are attributed to this increase and are probably due to prevailing redox conditions within the sediment and overlying water (Chapter 9 Section 9.5.4). The organic content based on LOI shows relative consistency until a decline at 24 cm. At the same position there is a slight rise at the near surface and from here the organic content falls at 1-2 cm.

The inverse relationship of the organic matter profile provides an approximation of the mineral content of the core. At 24 cm there is a slight rise to 85% which is coincident with the lamination shown in the stratigraphic descriptions. The pore water content in the sediment shows an inverse relationship with the mineral component. The average pore water content is 78% and declines to 72% at 24 cm. This must be the result of increased sediment input or focusing at 24 cm. There is a rise to 84% pore water content at the near-surface. In general, the percentage pore water content increases towards the surface and this is to be expected owing to the flocculant and less compacted nature of sub-surface sediment within the core.

It is now possible to review the possible origins, both natural and artificial, of the metals examined in the down-core enrichment patterns in the Leathann sediment. The hypotheses that the loch would be suitable as a control appears to be correct. This is based on the very low baseline Pb, Zn and Cu concentrations observed within the depth versus concentration profiles. Two chemical groups can be distinguished within the metal concentration patterns. The first group includes Pb, Zn, Cu all of which which concentrate towards the near-surface of the core. These are a result of atmospheric fall-out deposition. Mn and Fe shows limited down-core variability and alongside Ca forms a predominant soil erosion. The latter has been observed as also being and autochthonous component (Engstrom and Wright 1984). The limited variability of the curve shows that the influx of these metals has been fairly constant over time and perhaps reflects that land-use and ultimately sediment loading, has remained relatively stable, at least over the length of time represented by the sediment examined.

Cartographic sources show that there have been no major alterations to the landscape within the immediate vicinity of Loch Leathann, at least since 1854 (Ordnance Survey 1st Edition). Volume magnetic susceptibility and Mn share a similar trend with a gradual rise towards the surface. It has been mentioned previously that Mn can control and scavenge metals towards the surface of sedimentary profiles. In this case there is indeed a rise in Mn towards the surface of the core but to what extent Mn is controlling these metals cannot be ascertained.

In contrast to Loch Lossit, the Loch Leathann annual rate of sedimentation is established at 2.8 mm yr^{-1} which is the same as Loch Bharradail. The loch:catchment ratio for Loch Leathann is established at 4.7 which is the lowest ratio within the group, receiving drainage water from a catchment established at 27.8 ha. It is somewhat surprising to observe such a high rate of sedimentation when the catchment and overall conditions (i.e. size of the loch and relatively unaltered surrounding landscape) are proportionately smaller than the rest of the loch catchments investigated in this study. The sedimentary flux for Ca in Loch Leathann is the highest recorded within this study and not surprisingly so too is the pH concentration with values ranging between pH 5.42 at the base to pH 5.16 at the sediment surface. It may well be that the higher rate of sedimentation may be accounted for by internal factors such as those described by Battarbee (1986), which include water retention time, sediment focusing due to the morphology of the loch and increases and fluctuations in mineral/organic material from land-management practices.

It is proposed that there has been significant atmoshpheric enhancement of Pb, Zn and Cu starting from 20cm, 24cm and 26 cm respectively. Based on the ^{210}Pb-derived sediment rate of 2.8 mm yr^{-1} the rise in Pb, Zn and Cu fall between a date range of 1900-1920 (22-26 cm depth). Fossil fuel combustion and later Pb additives in petrol are thought to account for the near-surface increase in these three metals. The implications for these trends are dealt with in Chapter 9 Section 9.5.1 and are reviewed against the geographical distribution of atmospheric fall-out deposition observed at other locations in Scotland.

Extrapolated Pb-210 Date	Event in the profile	Interpretation
1980 AD *c.*1900-1920 AD.	Maximum Pb, Zn and Cu Onset of the rise in Pb and Zn at 20-24 cm.	Atmospheric fall-out. Possible overlap with the introduction of Pb in petrol and an atmospheric fossil-fuel component.
c. 1903 AD.	Lamination of finer sediment at between 23-24 cm. Reduction in the LOI profile	Effect of sediment focusing due to the introduction of a leat to periodically raise the water level in the out-flow river.
*c.*1927-1706 AD.	Background concentrations in Pb, Zn and Cu.	Background levels reflect absence of mineralisation within the catchment.

Table 8.4 Summary of the possible dates and events within the Loch Leathann profile based on linear extrapolation of the ^{210}Pb-derived annual rate of sedimentation rate of 2.8 mm yr^{-1}.

8.7 Conclusions

In the absence of mineralisation adjacent to Loch Leathann, the depth-concentration profiles for Pb, Zn and Cu can only be explained by increased atmospheric sources. Surface-soil Pb, Zn and Cu concentrations are lower in comparison to the enrichment of these metals recorded in the loch sediment. It is proposed that the lamination horizon (23-24 cm depth) may have been the result of fluctuations in the mean water level while the sluice was in use. Alterations to the hydrological regime with the introduction of the sluice at the turn of the century may have had an effect on the metal budget within the sediment profile, but this is as yet unknown.

Table 8.4 above summarises the major events observed within the profile. Owing to the absence of mineralisation the soil geochemical background concentrations for Pb, Zn and Cu are appreciably low in comparison with the other catchments. Importantly the sediment has received a significant increase in atmospherically derived Pb, Zn and Cu which contrasts with the low background values deeper in the sediment profile. This shows that although the loch is remote and some distance from any source of industrialisation the loch has not remained unaffected by atmospheric pollution. The integrity of the core is supported by a reliable ^{210}Pb and supporting ^{137}Cs inventory in the upper sediment sections. These data show that no physical alteration due to biological or mechanical mixing has occurred to the near surface sediment. On the basis of the results from sediment chemistry, the use of Loch Leathann as a control loch has been justified beyond any reasonable doubt. The next chapter will now examine some of the wider issues that have been raised in Chapters 5, 6, 7 and 8. The results of geochemical analyses at Lochs Finlaggan, Bharradail, Lossit and Leathann will be synthesised to establish the overall success of the approach taken for predicting early mining in the study region.

CHAPTER 9: DISCUSSION

9.1 Introduction

One of the principal questions raised in this study has been "how can we identify and date episodes of small scale lead mining in the absence of documentary records"?. This study set out to examine the environmental impact of 18th and 19th century lead mining within the limestone belt of NE Islay with the objective of establishing criteria for the identification of earlier unrecorded mining episodes from the palaeoenvironmental record. It will be recalled that an analogue approach has had to be adopted in order to identify early mining. Five key steps were taken and each posed a series of questions:

(Step 1) To first examine the evidence for the scale of historically recorded lead mining; its duration, and its manifestation in the palaeoenvironmental record.
How does the archaeological record relate to contemporary late-18th century accounts that describe the extent and scale of mining operations. What information can the historical record provide to establish the criteria for the environmental impact of lead mining?

(Step 2) To identify the sources of heavy metal abundance and their relative distribution within the study area.
Do soil contamination levels differ between the former mining regions and the area selected as a control?

(Step 3) To identify the possible routes of heavy metals from natural and anthropogenic point-sources into loch sediment
To what extent has atmospheric fall-out deposition affected the near-surface regions of the sediment. Are there local similarities and how do they relate to pollution trends seen elsewhere?

(Step 4) To evaluate the mechanisms by which heavy metals are retained in surficial sediment and the degree of post-deposition movement
Can diagenetic alteration and the enhancement of certain heavy metals be identified in any of the Islay cores?

(Step 5) To assess the reliability of the chosen radiometric dating methods
What are the limitations of these dating methods ? How can they be integrated with other lines of 'proxy' dating evidence?

9.2 THE EVIDENCE FOR THE SCALE OF HISTORICALLY RECORDED LEAD MINING.

9.2.1 Introduction

It will be recalled that in order to establish the present day sources of contamination from the abandoned mine workings, the remains had first to be mapped in order to define their true extent. It was also stated that it might be possible to determine different phases of mining activity. To this end we could then establish if there was evidence to suggest earlier periods of activity pre-dating the historical accounts. The following discussion will now assess the results of archaeological field survey.

9.2.2 Problems of period recognition in mining archaeology

One of the most comprehensive lead mine surveys undertaken to-date has been carried out by the Clwyd-Powys Archaeological Trust (CPAT) of Wales in 1993. In recognition of the vulnerability of what remained of its industrial heritage, the Clwyd Metal Mines Survey (CPAT Report No 88), conducted rapid fieldwork and recording to identify the remains of the industry. Prior to this work, the archaeological potential of mining sites in Clwyd was poorly understood resulting in poor consultation and liaison between planners and archaeologists. In some cases this led to the total destruction of many mine sites with little or no archaeological recording. The survey identified the greatest modern-day threat to be shaft-capping and large scale quarrying, leading to the total destruction and masking of earlier features. The masking of surface features is all too common where spoil tips from later mining episodes bury earlier features. The 18th century method of shaft and adit mining allowed miners to venture much deeper for ore deposits, which often cut through the earlier shallow mine workings and invariably led to their loss at the surface.

On Islay the situation has to some extent been similar to the Clwyd region although masking has been less extensive, arguably the main loss of features has occurred directly as a result of tailings removal for road building and track repair. Further loss has occurred as a result of the reclamation of former mined areas back into farm land. More recently, back-filling of open-casts features, shafts and shallower trials with farm-refuse, is now making it difficult to identify the true depth and extent of many of these features.

A major constraint in mining archaeology is the problem of period recognition and the assignment of dates to often nothing more than a series of ill-defined shallow workings. A general rule of thumb in this study has been an estimation of the relative age of many of the features based simply on the idea of superimposition, whereby if a given feature can be identified as lying above another feature, then the one lying below is clearly earlier. This has been useful in establishing different phases of mining activity at a given site. Often the historical records have been useful in estimating the age of the earthworks or other extant features such as shafts and adits. A good example of this is shown by the adit depicted as a day level on the mine plan at Mulreesh, drawn by Vercoe in 1874 (Figure 5.5). Today this feature flows through a large leat with banks that overlie a metalled trackway leading down to the buildings labelled A, B, C and D in the Mulreesh Survey (Figure 5.4). These buildings[1] (which have incorporated mine-waste in their construction) are assumed to have been abandoned earlier than *c.*1870-74, when the adit and banked ditch was constructed. This allows a date of the late 18th century period to be placed on these features which in all probability represent an early phase of farmer-miner occupation.

As in Clwyd, there is good documentary evidence for 18th mining but unfortunately a lack of supporting drawings and plans. As was often the case, many of these old accounts relating to the mines were aimed at a readership which was only concerned with the quality and amount of ore being exploited at that time. Technological detail can only be estimated by reference to other mining regions where the industrial archaeology has been better preserved. However, in reality there will remain many 'grey areas' where the historic and archaeological record is often fragmentary.

Another important aspect to come to light from this work has been the identification of mine workings that have exploited near surface metamorphic[2] ore deposits formed in association with Tertiary dykes (see mines 1, 3, 4, 7, 8 and 23 in Figure 4.2). It is suggested that many of the shallow trials associated with these features probably represent the earliest workings across the study area. This assumption is based on the simple fact that the ore found alongside the dykes was easily accessible with the minimum effort. Once the ore became difficult to extract due to depth or flooding the early miners moved on to exploit new near-surface deposits. This must in all probability, have led to the early recognition and exploitation of shallow brecciated ores alongside the dykes.

It will be recalled that Charles Freeburn in 1770 observed and certainly reworked many of what can be taken to be shallow workings, that may have been excavated as early as the Middle Ages. (Section 4.6.1 above). Freeburn's reference to a thousand trials could well be an exaggeration, but nevertheless, the description does point to *ad hoc* exploitation by the

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[1] The buildings are not depicted in any of the cartographic sources.

Metamorphic contact[2]-large scale action of both heat and pressure producing a wide range of new minerals associated with igneous intrusions (Whitten and Brooks 1978).

farmer-miners who mined the shallow ore deposits wherever they occurred. Fieldwork in the limestone quarry at Ballygrant (within the region of Mine No. 7 in Figure 4.2) recorded a mineral vein of galena and chalcopyrite in a breccia of limestone and dollerite (see Plate 2 in Appendix 1). The distribution map of the lead mines (Figure 4.2) in the study area shows that in some cases the mines are situated well away from Tertiary activity, but in all cases the mines are situated on Dalradian limestone. Typically, the mines not relating to Tertiary activity have exploited discrete breccias along major fault zones (Gallhaggar, pers comm.).

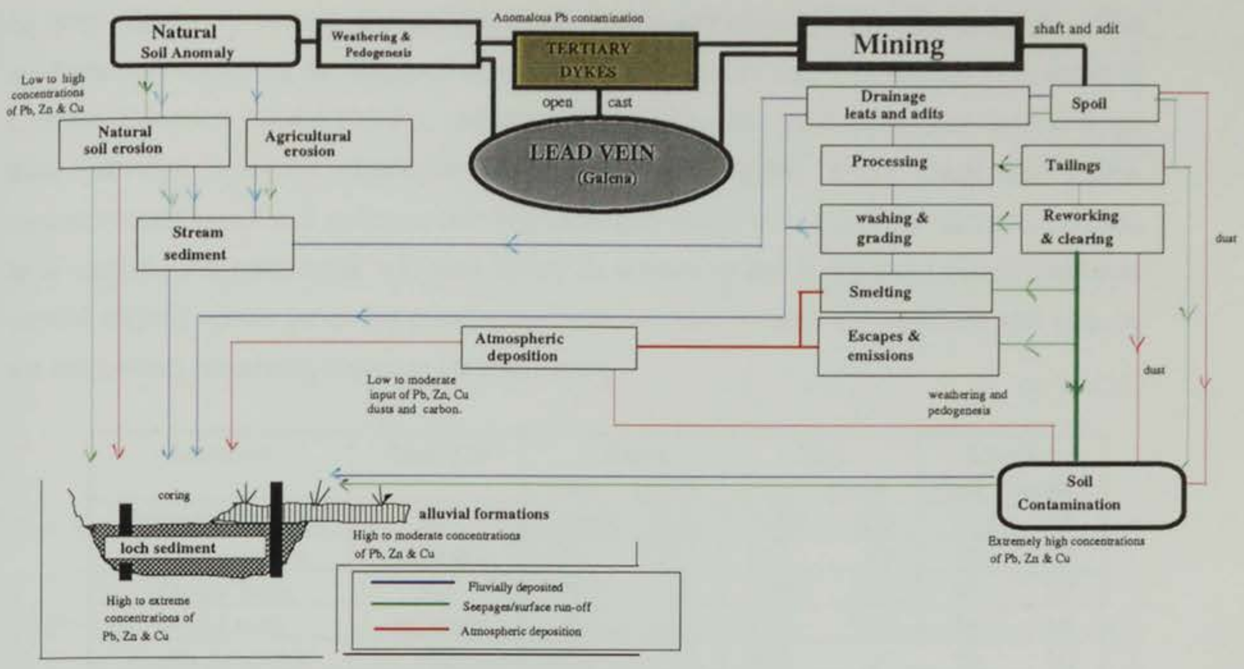
9.2.3 Implications for the study area

In summing-up the success of the archaeological field survey, it is concluded that there has been significant reclamation at many of the sites examined. This has resulted in the loss of many early mining features. Furthermore, it is proposed that what survives within the landscape today is essentially a small fragment of what was formerly a structured industry integrated into an agrarian economy. It has been mentioned that contemporary sources from the mid-late 18th century describe the presence of numerous 'earlier' workings. Some of these are visible today at South Ardachie and Mulreesh Mine. As we have seen elsewhere, the study area is not untypical of other lead fields in Great Britain where many mining features have been lost due to more modern mining practices and recent clearance and repatriation into farmland. Archaeological fieldwork now provides a detailed archaeological record of some of these important remains and this data certainly provides a useful archive as well as producing new information on a lesser-known mining region of Scotland.

9.3 THE IDENTIFICATION OF THE SOURCES OF HEAVY METAL CONTAMINATION IN SOILS AND SEDIMENT IN THE STUDY REGION

9.3.1 Introduction

Chapter 1 mentioned the importance of identifying the source and distribution of heavy metal contamination in the study area. This has been achieved using the loch-catchment unit as a basis of investigation. Chapter 2 Section 2.3 examined in detail the environmental issues arising from base metal mining. A conceptual environmental pathway model (Figure 2.7) has been constructed in order to determine the principal environmental gradients by which heavy metals can enter into loch sediment in the study area. Geochemical data from the DRL mineral reconnaissance survey was critically re-evaluated using modern mapping software.



The conceptual model of the main environmental gradients of contamination in soil, freshwater and alluvial sediment in the study area. This is produced in a larger format as Figure 2.7, page 35.

The results show the distribution of Pb, Zn and Cu soil contamination and its close conformity with both areas previously mined and other natural occurrences, such as Tertiary activity and the possible effects of commercially unexploited mineralisation. Given the scale and geographical extent of the former mining areas (see Figure 4.2) it is not surprising that significant soil contamination has occurred.

9.3.2 Mining versus naturally derived heavy metal contamination

Three important issues have arisen from the re-evaluation of the DRL data. Firstly, it is clear that there are highly anomalous increases in Pb, Zn and Cu which are formed over regions where no evidence of mining has been recorded. This is particularly noticeable in the southern shore region of the Loch Lossit catchment where three Tertiary dykes are seen to converge. (Chapter 7 Figure 7.1). At this junction, the concentrations of Pb, Zn and Cu are enhanced by chemical alterations in the local regolith possibly due to metamorphic ore formation associated with the dyke swarm. It has been mentioned above that Tertiary activity has been an important factor in the early exploitation of local lead deposits.

Secondly the catchment distribution of Pb, Zn and Cu shows very clearly the presence of geological boundaries which are sharply defined where the Dalradian limestone meets acidic phyllite and outcropping shales. This is demonstrated at the site of Portnealon Mine

(Figure 5.12) where Dalradian limestone is bounded by phyllites. These geological types can be seen tightly to control the concentration and distribution of Pb, Zn and Cu. The mechanisms controlling the dispersal of metals in surface soils has been discussed in detail in Chapter 2 Section 2.3.4. Thirdly, and most importantly, the geochemical distribution maps have provided scientific objectivity in assessing the relative heavy metal and cation concentrations in soil and sediment between areas previously mined and the catchment chosen as a control. An assessment will now follow in respect to the background (Loch Leathann control region) versus increased metal concentrations found within the sediment and soils in the catchments containing evidence for lead mining.

Location	**Medium**	**Lead**		**Zinc**		**Copper**	
		max	**mean**	**max**	**mean**	**max**	**mean**
Portnealon mine	soil	5000	236	2280	105	142	22
Loch Finlaggan	sediment	139	125.5	219	96	43	29
Glasgo Beag	soil	900	135	1900	128	64	12
Loch Lossit	sediment	101	85	415	187	38	18
South Ardachie	soil	650	92	380	94	58	10
Loch Bharradail	sediment	925	621	530	425	31	22
No mining	soil*	55	22	75	24	62	5
Loch Leathann	sediment*	205	30	103	53.2	15	7.2

Table 9.1 Summary table of the maximum and mean values taken from the 1983 Domego Resources Geochemical Survey of NE Islay compared to the mean and maximum Pb, Zn and Cu in loch sediment. (*) denotes control soil and sediment, note: all values are in mg kg^{-1}).

Table 9.1 summarises the catchment distribution of these heavy metals in the near vicinity of the lochs Bharradail, Lossit, Finlaggan and Leathann. The highest concentration in Pb, Zn and Cu was found adjacent to Loch Finlaggan at Portnealon Mine in the Finlaggan catchment. In comparison, note the very low concentration observed in the Loch Leathann catchment which shows that the soils adjacent to the loch are below the average proposed for uncontaminated soils (Table 9.1). These values show that there is no mineralisation or other forms of serious contamination in the catchment. Examination of the calculated soil geochemical concentration values within the catchment soils show that there are high isolated anomalous values at Portnealon, Glasgo Beag and South Ardachie mines. At all three sites, the mean concentration of Pb and Zn is significantly higher than the control area and also above the average soil concentrations listed for uncontaminated soils (Table 9.2, see also Figure 9.1.) Copper is low in all cases, with the highest enrichment at Portnealon Mine.

Element	**Average in lake sediment***	**Average in soils**	**Normal range**	**Conc. in soil considered toxic**
Pb	31	30 (24)	2-200 (2.5-85)	100-400
Zn	123	50	10-300	70-400
Cu	46	20	2-100	60-125

Table 9.2. Typical (total) concentrations of Pb, Zn and Cu in 'normal' soils adapted from Ross (1994, 9) and Alloway (1990, 31) (*) denotes Forstner's (1977) 'world' average from 74 pelitic lake sediments. Note: values in mg kg^{-1} . The values in parenthesis are for Scottish upland soils established by Berrow *et al* (1987).

The soil data and results for the mean metal influx to sediment (Table 9.1, Figure 9.2) demonstrate that the mean concentration for Pb, Zn and Cu in the Loch Bharradail sediment is significantly higher than the mean content observed in soils across the South Ardachie mine. Furthermore, the baseline value for Pb at Loch Bharradail was established as 346 mg kg^{-1} which is 11 times higher in order of magnitude than the average of 10-30 mg kg^{-1} recorded for uncontaminated soils cited in the general literature.

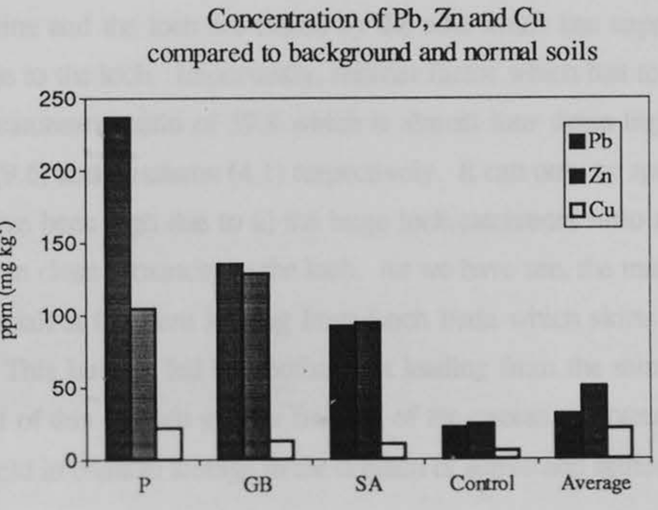


Figure 9.1 Mean concentrations of Pb, Zn and Cu at the Portnealon (P), Glasgo Beag (GB) and South Ardachie (SA) leadmines compared to the control soil and the concentration in average soils as defined in Table 9.2. (note: Background soils are the soils surrounding Loch Leathann)

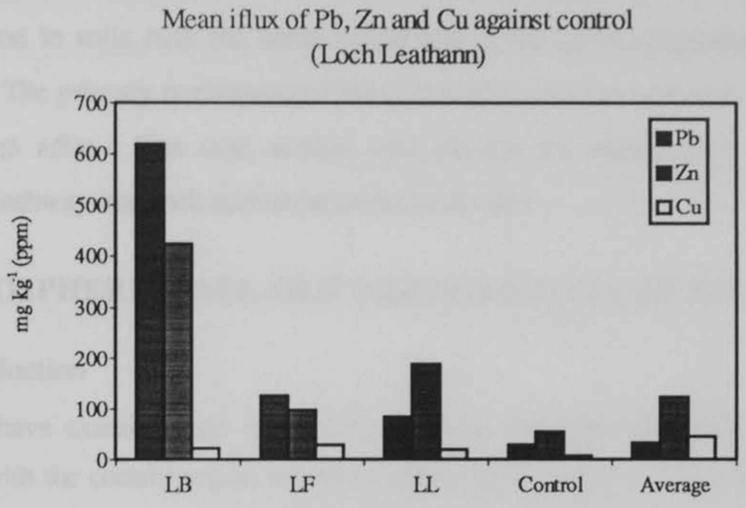


Figure 9.2 Mean sedimentary flux of Pb, Zn and Cu from Loch Finlaggan (LF), Loch Bharradail (LB), and Loch Lossit (LL) in comparison to Loch Leathann (control) and Forstner's (1977) world average from 74 pelitic lakes.

The results confirm conclusively that Loch Bharradail has a higher heavy metal influx into sediment than Lochs Lossit, Finlaggan and Leathann. The reason for this is explained by the fact that the South Ardachie mine is the most extensive of the mines examined in the survey. Moreover, the mine and the loch are linked by the adit which has supplied a high burden of lead-rich drainage to the loch. Importantly, another factor which has to be taken into account is the high loch-catchment ratio of 39.8 which is almost four times higher than Lochs Lossit (7.1) Finlaggan (9.6) and Leathann (4.1) respectively. It can only be speculated that the metal burden would have been high due to a) the large loch:catchment ratio and b) the presence of lead ore deposits in close proximity to the loch. As we have see, the main natural inlet leading into Loch Bharradail is the burn leading from Loch Fada which skirts the edge of the South Ardachie mine. This burn is fed by another leat leading from the mine (Feature 4 in Figure 6.2). As a result of this a much greater fraction of the excess or potentially available Pb, Zn and Cu will be held in channel storage in the domain of active and settled stream sediment.

Metal redistribution mechanisms have been studied by Bradley (1982) using source-sediment and discharge in a mineralised region of Wales. It was demonstrated that surface coatings of Pb, Zn and Cu vary according to discharge. A dilution effect commonly observed in soluble load investigations was also seen to apply to metal coatings on various particle-class sizes using X-ray fluorescence spectrometry (Crecellious and Piper 1973; Hamilton-Taylor and Willis 1990).

This section has demonstrated that there is a large contrast in the amount of heavy metal contamination in soils over the areas mined and much lower concentrations in the control catchment. The primary environmental route of metals into the sediment has been via streams and drainage adits. The next section will address the significance of the atmospheric deposition pathway into loch sediments in the study area.

9.4 ATMOSPHERIC FALL-OUT DEPOSITION IN THE STUDY REGION

9.4.1 Introduction

So far we have examined the effects of mining in the catchment soils. These have been compared with the control region where no mining has occurred. It has been demonstrated in the conceptual pathway model that there are three principal routes leading to high heavy metal flux into the lochs in the study area. The first was identified as natural contamination due to the natural weathering processes of unexploited ores. The second pathway was by direct drainage from mining and processing. The third route was via atmospheric deposition. The significance of atmospheric deposition cannot be overlooked, particularly in explaining the near-surface Pb and Zn maxima in the Loch Leathann and Lossit cores. The implications of long distance pollution transport and its influx into freshwater sediment will first be reviewed drawing on the results of similar findings in Scandinavia and in Scotland. The purpose of this is to show that the results obtained from Lochs Lossit and Leathann are far from unique and that they conform to trends elsewhere in upland Scotland. It will also be shown that Islay's atmospheric record is part of a wider European and global problem.

9.4.2 The wider implications of atmospheric deposition

Large-scale pollution mapping has been undertaken in Sweden by Renberg *et al* (1994, 324) who carried out an extensive geochemical survey of 131 Swedish lakes. This figure comprised two sets of sediment cores. The first set consisting of long-cores between 1.5 and 4 m in length from 19 lakes for the investigation of pre-industrial pollution trends. The second set consisted of 0-35 cm depth near-surface sediment cores from 112 lakes. These were used to monitor trends in atmospheric pollution in Sweden from about 1600 to the 1980's. Chronological control was based on AMS-dated plant macro-fossils and ^{210}Pb dating methods. Land use histories, pH and diatom inventories were also included in the study. The results showed convincingly that atmospheric lead deposition increased above background levels more than 2,600 years ago. A small, but marked lead increase was observed about 2,000 years ago, preceded by a significant increase at around 1,000 AD. This increase accelerated during the nineteenth and particularly into the 20th century, with a deposition maximum occurring at about 1970. This led Renburg (1994, 323) to conclude that before the onset of nineteenth century industrialisation, lead concentrations had already reached 10-30 times previous background levels as a result of atmospheric deposition.

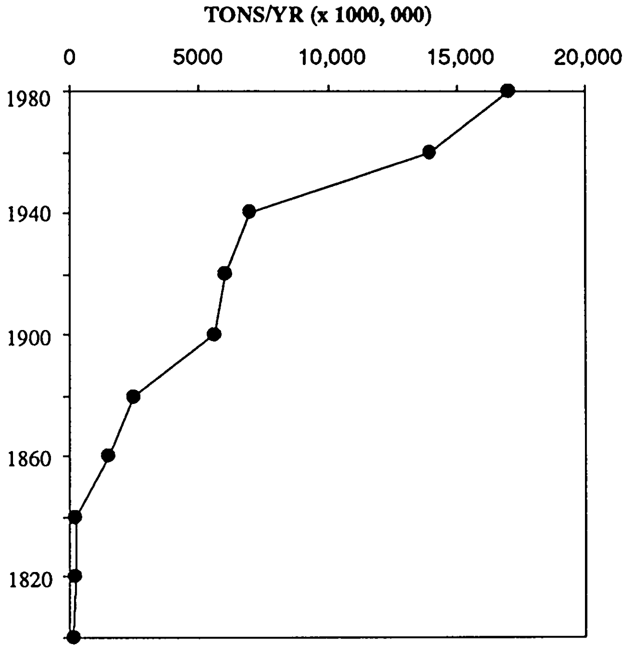


Figure 9.5 European coal combustion trends during the post-industrial period (after Renburg 1985a).

It was further suggested that this pre-industrial airborne pollution was derived from extensive production of lead smelting in Europe that was started by Greek and Roman cultures. According to Renburg and co-workers, the cumulative deposition from anthropogenic sources in pre-industrial times (~600 BC to 1800 AD) was at least as large as the cumulative deposition during the industrial period (1800 AD to the present). These results demonstrate large-scale pre-industrial lead contamination that is closely in accord with deep-ice core analyses from Greenland.

Analyses of the Greenland ice core covering the period from 3000 to 500 years ago, shows that lead is present at concentrations four times as great as natural values from about 2500 to 1700 years ago (500 BC. to 300 AD.) (Hong *et al.* 1994). Results show that Greek and Roman lead and silver production polluted the middle trophosphere on a hemispheric scale two millennia ago, long before the Industrial Revolution. Cumulative lead fallout to the Greenland Ice Sheet during these eight centuries is estimated to be as high as 15 percent of that caused by the massive use of Pb alkyl additives in petrol since the 1930s. Boutron *et al* (1994) have observed a similar trend in ancient Antarctic ice. Human activity has led to an increase in Pb concentrations during recent centuries and this is observed as being in the order of a tenfold increase, whereas in Greenland it is estimated that there has been a two hundred fold increase. Significantly, this increase is followed by a 7.5-fold decrease during the past two decades (1980-1990) mainly as a consequence of the rapid fall in the use of Pb alkyl additives in the USA. This trend is supported by isotopic data based on source apportionment of ^{206}Pb/^{207}Pb (Boutron *et al* 1994).

9.4.3 The acidification problem in upland Scottish lochs

A great stimulus to this science has been the ‘acidification syndrome’ brought about by the fact that the aquatic life of many of our lakes was being affected by acidified waters with increasing trends towards eutrophication and a reason was sought. It was in the 1970's that the problem of acid deposition in Scandinavian lakes was first recognised. The reports of acidification to Scandinavian and North American rivers led to claims that industrial countries were damaging rural ecosystems many hundreds of kilometres distant. In the United Kingdom these claims were contested for a time by the Central Electricity Generating Board, (Davidson *et al* 1992, 114). Scientific work on declining fish populations in Scotland stimulated this research further (cf. Harriman *et al* 1981; Harriman *et al* 1986 and Maitland *et al* 1987).

A comprehensive and detailed study on the acidification of lakes and reservoirs throughout the upland regions of the United Kingdom was compiled by Battarbee and co-workers (1988) to investigate and identify the scale and geographical extent of this problem. The timing and rate of acid deposition were addressed by four main hypotheses. These were: (i) Lakes were naturally acidic; (ii) acidification had occurred slowly over hundreds to thousands of years, (iii) acidification had occurred only recently (since 1800) but was the result of a combination of land-use or land management practices (e.g. burning, grazing, afforestation and decrease in agricultural liming); and (iv) acidification had occurred recently (since 1800) and was the result of an increase in acid deposition from the combustion of fossil fuels.

This research was undertaken at a time when there was increasing scepticism both in scientific and government circles as to the validity of the 'acid rain hypothesis'. This scepticism came about by the attraction of an alternative 'land-use hypothesis' which lay the blame at the door of past and present day land-use practices. Previous work by palynologists and palaeoecologists has shown that acidification was occurring over a period of millennia rather than over the last few decades which added more fuel to the debate, (cf. Barber 1981; Moore 1975,1984; Pennington 1981; 1984, Pakarinen 1983 and Norton and Khal 1991).

Over 40 lochs throughout Scotland have now been examined for evidence of recent lake acidification and consequently, the extent of the problem is now understood. The most strongly affected areas are Galloway, Arran (Battarbee *et al* 1988) the Trossachs (Kreiser *et al* 1990) and Rannoch Moor (Flower *et al* 1988). The onset of contamination is clearly registered in the lead profiles, which are often dated to the beginning of the 19th century reaching a maximum in the mid 20th century. Cu and Zn follow similar but not identical historical patterns (Battarbee and Elliot 1994). The increase in the Pb curve towards the surface represents the first clear impact of atmospheric contamination, and in most sites studied, this point is dated to about 1800 in both Scotland and Wales (Battarbee 1988, 57). This date was observed as pre-dating the beginning of acidification in some regions by about 50 years based on ^{210}Pb chronologies which were supported by palynological, diatom and historical land-use information. At most sites the general trend is towards a maximum Pb increase in the mid-twentieth century before falling somewhat in recent years. Alterations in pH based on fluctuations or decline in the diatom flora reflect post-1850 acidification at many Scottish sites. As a result of this work Battarbee *et al* (1988) are able to conclude that throughout Scotland near-surface concentrations of trace metals such as Pb, Zn and Cu are high in the south and central areas of Scotland, but decrease in concentration towards the north (Battarbee *et al* 1988; Rippey 1990).

Recent research by Williams (1991) at Loch Dee, in Galloway, SW Scotland, recorded fly-ash within the near surface of the core (pulverised high combustion power-station fuel) using scanning electron microscopy. This work also identified both pre- and post-industrial age increases in heavy metal influx and detectable Fe- oxide concentrations in the uppermost 8 cm of sediment. The lead and zinc inventories in this loch were seen to be closely related to early mineral mining and smelting dating to about 1500 AD. A contributing factor to the total lead inventory is the more recent (post-1920s) emissions of lead derived from petrol. Source apportionment study on sediment from Loch Lomond (Sugden *et al.* 1991) using the isotopic ratios ^{206}Pb/^{207}Pb established that lead contamination from heavy industry and coal combustion has declined in recent years, but lead derived from car exhausts has increased since the 1930s. In a comparative study using total Pb influx and ^{206}Pb/^{207}Pb ratios (Sugden et al 1991) at remote peat deposits in North Uist, Outer Hebrides showed that there has been a significant increase in atmospheric deposition of lead commencing around 1900 and going on to 1950 with 2.8 to 4.8 mg m^{-2} yr^{-1} respectively. From 1950-1988 there has been a decline in the deposition of lead from 4.8 to 3.4 mg^{-2} yr^{-1}.

Trends in lead deposition in ombrotrophic peat at Flanders Moss in Stirlingshire between the years 1900-1950 showed an increase from 10.2 to 20.8 mg m^{-2} yr^{-1} respectively. From 1950 to 1988 the lead influx increased to a maximum of 30.8 mg m^{-2} yr^{-1}. This increase was attributed to closer proximity to domestic, industrial and transport sources in the heavily populated Central Belt of Scotland.

The discussion above has reviewed the wider context of atmospheric deposition within Northern Europe in relation to Scotland. It is clear from what has been discussed that there are broad geographical trends in lead deposition dominated by the position of large areas of industry and urban conurbation. The discussion will now examine how the lochs in the study area relate to the wider problem of long distant transport of atmospheric lead deposition.

9.4.4 Atmospheric fall-out deposition on Islay (*c.*1800 AD-present)

The preceding discussion has shown that European industrialisation has caused a conspicuous enhancement of Pb, Zn and Cu within the uppermost regions of loch sediment in Scotland. As Battarbee and Elliot (1994, 127) have stated:

> "Although only a small fraction of potential sites have been studied (in Scotland) it is apparent that, despite their relative remoteness from major industrial and urban centres, few have been uninfluenced by human activity over the last century. Some lowland lochs have been severely enriched, many upland sites have been strongly acidified, and a few have suffered from massive soil inwash associated with poor forestry management. All have been contaminated by air pollutants of various kinds"

It can be clearly demonstrated that such a case exists for Islay which is situated well away from the more industrialised Central Belt of Scotland but which has most probably been affected by the industrial belt of Northern Ireland. Nevertheless the lochs examined have received an atmospheric derived Pb and Zn component. Our attention can now turn to the lochs and examine these heavy metal trends in closer detail.

9.4.5 Loch Leathann

Hypotheses relating sedimentary trace metal profiles to anthropogenic pollution can best be validated by studying the relationship between temporal enrichment and certain aspects of regional industrial development (Williams 1991). By examination of the geochemical profiles at Loch Lossit and Leathann (Figure 7.19 and 8.20) it is possible to resolve the elements into two distinct groups on the basis of their down-core enrichment patterns. The first includes Ca, Mn and Fe which are derived principally from the lithosphere and Pb, Zn and Cu which are derived predominantly from the atmosphere. The intention here is to examine the broader chemiostratigraphic trends of Pb, Zn and Cu to assess the overall implications of the results.

Table 9.1 shows the maximum soil concentration of Pb, Zn and Cu recorded in the soil adjacent to Loch Leathann which was determined as 22, 24 and 5 ppm respectively. These values are all below the mean concentration proposed in normal 'unpolluted' soils (see Table 9.2). The total Pb concentration within the loch sediment was calculated at 205 mg kg^{-1} which exceeds the maximum soil Pb concentration (55 ppm) by some 26.8%. In the light of these results we can reject a direct terrestrial source as being responsible for the near-surface increase in Pb within the Leathann profile.

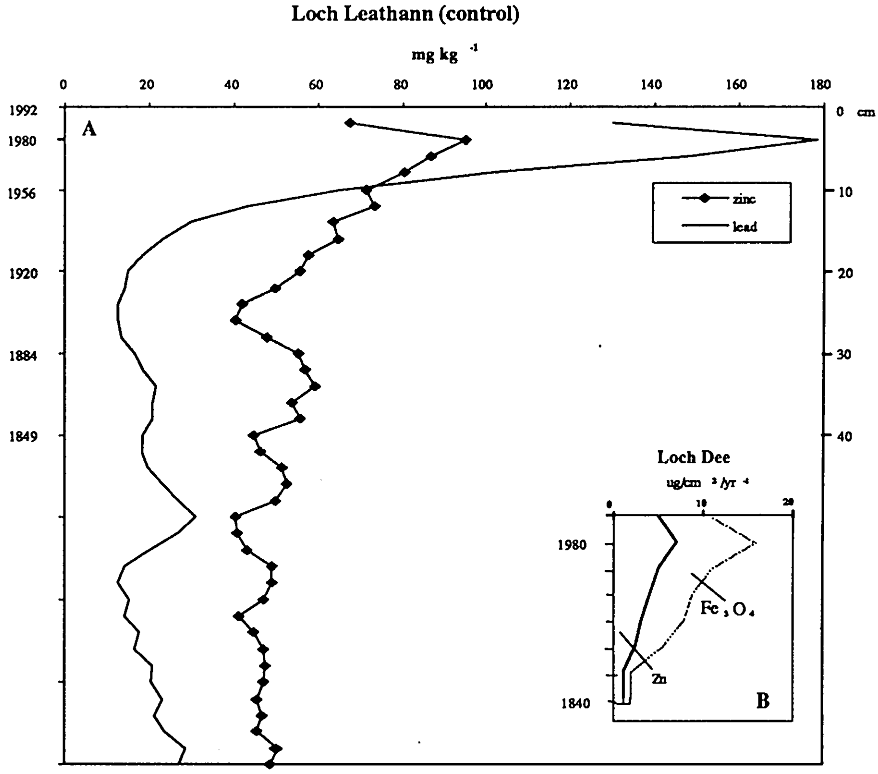


Figure 9.4 (A) Variation of Zn and Pb influx at Loch Leathann (smoothed). The inset (B) shows the variation of Zn and Fe_3O_4 influx to Loch Dee, Galloway over the same period (after Williams 1991).

However for the deeper sediment (below *c.* 20 cm) the baseline Pb concentration is established as 22 mg kg^{-1} and comprises an intrinsic mineral lead component that is derived principally from terrestrial sources. Based on the annual sedimentation rate of 2.8 mm yr^{-1}, the data in Figure 9.4 shows that up until the *c.*1920 (corresponding with 20 cm in depth) the Pb and Zn profile has steadily fluctuated until this depth. From this point there is a rapid rise in both these metals. There is a striking similarity between the take-off position of increased Pb influx (20 cm) and the Zn and Fe-oxide profile from Loch Dee, Galloway, SW Scotland.

Williams (1991) also recorded a maximum Zn and Pb concentration of 290 mg kg^{-1} and 287 mg kg^{-1} at around 1980 using a ^{210}Pb-derived chronology. Hematite (Fe_3O_4) was seen to peak at around 1980 and was attributed to fly ash from power stations within the industrial Central Belt of Scotland.

9.4.6 Loch Lossit

Moving now to Loch Lossit, the depth versus concentration profiles for Cu, Pb and Zn (Figure 9.2) share the same trend observed in the Loch Leathann profile, with an increase in Pb, Zn and Cu towards the uppermost sections of the profile. Based on the sedimentation rate of 1.6 mm yr^{1} a steady increase in all three metals occurs towards the upper-surface of the core starting at around 1867.

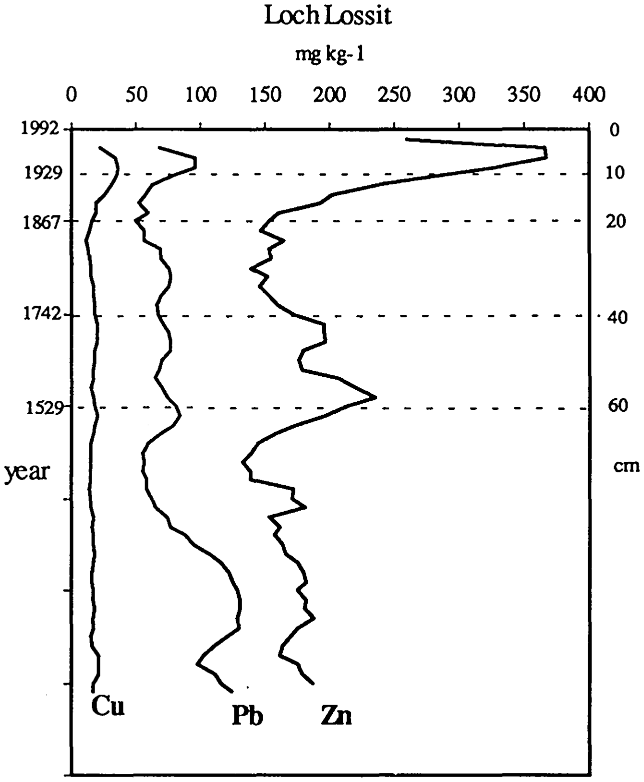


Figure 9.6 Loch Lossit ~ Influx concentration profiles showing the near-surface increases of Cu, Pb and Zn due to atmospheric deposition since the late 19th century. The Inset shows the Pb and Zn concentration (µg g^{-1}) from the undated Ness-1 core from Loch Stiapavat, Isle of Lewis, Outer Hebrides (after Entwhistle *et al* 1995).

The maximum concentration in all three metals corresponds to the 5-6 cm depth section which dates to around 1961. From this position there is a rapid decline in concentration in all three metals that is coincident with the 2-3 cm depth and dates to *c.*1980. Indeed, the decrease in

Pb concentrations at this depth of the core correlates well with improvements in atmospheric quality over recent years (compare with Rippey 1990 and Entwhistle *et al* 1995, and inset in Figure 9.6 below). What is interesting when comparing these curves with the Loch Leathann Pb and Zn curves (Figure 9.1) is that the 'take-off' point of Pb and Zn in the Loch Lossit core occurs some 53 years earlier. An atmospheric derived influx of Cu, Pb and Zn combined with an earlier intrinsic terrestrial component from mid-19th century land improvement within the catchment may possibly account for the trends within the concentration profiles.

Improvement by land drainage and tree planting commenced around *c.*1842 (*New Statistical Account* 1845). If ploughing and planting has occurred over soils that contain pockets of Pb mineralisation then we might expect to see a chemical ‘response’ particularly in the Fe and Mn profiles. Arguably, lead mining cannot be ruled out as a possible source. The Ordnance Survey map of Islay (Second Edition 1882) records the name of Glasgo Beag and the presence of a mine shaft. This alone cannot be used as evidence that the mine was in operation at the time the map was re-drawn. The mine is not recorded on the First Edition Ordnance Survey map of 1856. We can be more certain that the mine dates to the late 18th century from the reference in the collection of mining papers published in *The Book of Islay* (Smith 1896).

Deeper in the core a small subsidiary increase in Cu, Pb and Zn occurs at around 60 cm which dates by linear extrapolation to *c.*1529. This date is well within the 'historical sphere' of lead mining activity in north-east Islay. The rise in all three metals at this position is augmented by a slight rise in Fe and the Fe:Mn ratio to 1.3. This might be taken to suggest an increase of Fe-rich material from catchment soils. Paradoxically, there is a lack of other critical supporting data at this particular depth in the profile to be absolutely certain that the calculated increase is a direct result of local lead mining and processing. The much larger protracted increase in the Pb curve towards the base of the profile at 102 cm could be attributed to early mining activity within the catchment. Based on linear extrapolation of the annual rate of sedimentation of 1.6 mm yr^{-1}, this provides a date of approximately 1367. This would predate the earliest historical account (1511-12) of mining in Islay by 144 years. A possible alternative to this hypothesis is an erosion event within the catchment that has promoted the influx of Zn and Pb-rich material into the loch. However, at this position, only Fe supports the rise in the Pb and Zn curve. There is no concerted rise in magnetic susceptibility or organic carbon as defined by the loss on ignition curve as one might have expected.

9.4.7 Loch Bharradail

Moving now to Loch Bharradail, the core has a maximum Pb concentration of 925 mg kg^{-1} at 48 cm depth. This is 720 mg kg^{-1} higher than the surface Pb maximum of 205 mg kg^{-1} at 1-2 cm depth calculated for Loch Leathann. If we accept that the near-surface Pb maximum of 205 mg kg^{-1} in this loch is the result of atmospheric pollution, then this value can be used to establish an Atmospheric Enrichment Factor (AEF). This formula is a modification of a similar Sediment Enrichment Factor model proposed by Jennet *et al* (1980). Using the 1-2 cm surface Pb concentration of 640 mg kg^{-1} in the Bharradail core minus the 'atmospheric component' of 205 mg kg^{-1} from the Loch Leathann surface maximum, this provides a Pb concentration value of 435 mg kg^{-1}. An AEF for the near surface region of the Bharradail core can now be established from:

surface Pb concentration in control (Leathann) - surface Pb concentration (Bharradail)
background Pb concentration (Bharradail)
=205 mg/kg^{-1} Pb - 640 mg/kg^{-1} Pb
= 434 mg/kg^{-1} /346 mg/kg^{-1} Pb
= 1.26 (AEF)

The AEF provides a very crude index to the level of atmospheric Pb enrichment at the near surface of Loch Bharradail. For Loch Lossit the same equation provides an AEF of 2.53 which is a product of a lower baseline and subsurface maximum Pb concentrations. The estimated base-line concentration of 346 mg/kg (64 cm depth) for the Loch Bharradail core is high in comparison to other calculated background concentrations observed in Lochs Leathann, Finlaggan and Lossit and at other lochs elsewhere in Scotland.

Baseline and maximum Pb concentrations from five other locations in Scotland reflect the level of Pb enrichment that Loch Bharradail has received from lead mining. Among the lochs not in Islay, only Loch Tay in Perthshire has a record of extensive mining recorded within its catchment. Lead and copper were mined between the years 1861-1865 along its southern shore and further afield along its headwaters at Tyndrum, Perthshire (lead and silver) between the years 1850-1885 (Burt *et al.* 1981). This has resulted in a maximum of 500 mg kg^{-1} Pb at 13-14 cm depth.

Location	Baseline Pb concentration	Maximum Pb concentration
Loch Leathann*	**20** (22)	**205** (3-4)
Loch Lossit*	**43** (18)	**135** (102-104)
Loch Finlaggan*	**42.5** (12)	**236** (18-20)
Loch Bharradail*	**346** (68)	**925** (108-110)
Round Loch of Glenhead †	**34** (22)	**396** (6-7)
Loch Lomond†	**15** (28)	**152** (7.5-9)
Loch Leven†	**10** (14)	**48** (0-1)
Loch Coire nan Arr†	**22** (9)	**51** (3-4)
Loch Tay†	**16** (60)	**500** (13-14)

Table 9.3 Comparison between baseline and maximum Pb concentrations between the four lochs in this study and lochs elsewhere in other regions of Scotland (data on lochs denoted † are after Farmer, *et al* 1993, 211). Bold values are in mg kg^{-1} with depth (cm) in parentheses.

9.4.8 Implications of the environmental record at Loch Bharradail

At Loch Bharradail a hiatus occurs at 64 cm depth that is marked by a noticeable decrease in all metals at this position in the core. Leading up to this event there appears to be a steady decline in the metal influx. Two intermittent subsidiary peaks in Pb and Cu occur which might reflect a chemical response to episodes of short-term lead extraction within the catchment. To some extent the increases are supported by an increase in Fe and to a lesser extent Mn. A steady rise in magnetic susceptibility is also coincident with these events. This suggests an input from local mining but paradoxically, the LOI curve shows little variance in organic carbon which remains constant throughout the profile at around 40%. As a result of disturbances to the regolith within the catchment one might have expected a decrease in organic carbon as a direct response to a greater influx of mineral matter. Alternatively, what we might be seeing here is an artefact of diagenetic alteration where Mn and to some extent Fe has scavenged Pb that has resulted in the formation of the subsidiary peaks.

Following the hiatus at 64 cm depth, there is a rapid increase in Pb, Zn, Cu, Ca and Fe concentrations at around 40 cm depth. Alongside these increases is a noticeable rise in magnetic susceptibility which points to an increase in allochthonous material. The near-surface increases of Fe and Mn alongside magnetic susceptibility could be due either to disturbance to spoil heaps as part of reworking or removal for track repair or as a result of diagentic alterations within the sediment. Following the construction of the leat (Plate 12, Appendix 1) which is assumed to be related to the late 18th century period of 'shaft and adit' mining at South Ardachie, a substantial increase in Pb-rich mine drainage was discharged directly into the loch. Based on the ^{210}Pb-derived sedimentation rate of 2.8 mm yr^{-1}, the period commencing in *c.*1760 through to *c.*1850 sees a protracted rise throughout the chemical inventory. We know from the mining reports compiled in the *Book of Islay* (Smith 1896) that the mine was fully operational in 1770 and probably continued well into the 1800s if we accept the contemporary account by Alexander Shirrif the visiting engineer from Wanlockhead who commented on the extent of the ore deposits at the site. In the absence of a supporting radiocarbon chronology, the dates prior to 1850 should be viewed as tentative, dates after this period are deemed to be more reliable.

The cessation of mining sometime in the early to mid-19th century led to a reduction in the influx of Pb and to a lesser extent Cu. This may have resulted in changes in the pH history of the loch as pH is seen to decline dramatically and then recover to almost the level previously recorded in the profile. It is a matter of speculation whether mine drainage acted as a buffer against increased acidification that initiated a trend towards eutrophication. As high discharges from the mine ceased this may have promoted a general increase in the biomass resulting in the highly eutrophic state that we see today. However the pH values have to be taken as an estimate only owing to the lack of reliable diatom inventories.

9.4.9 Implications for the study area

The results from lochs Lossit and Leathann show reliable evidence for increased atmospheric contamination towards the upper regions of the cores, with a post-1980 decline in the influx of Pb, Zn and Cu. These trends are in keeping with heavy metal inventories seen elsewhere in lochs and in ombrotrophic peat deposits in the western regions of Scotland. The Loch Lossit sediment has received high lead-rich material as a result of mining at an earlier period and a tentative date has been proposed placing this event in the mid-14th century. As we have seen this predates the earliest historical accounts for lead mining on Islay. The geochemical record at Loch Bharradail shows good conformity between the historical and environmental record since the early 18th century at least. A longer history of mining is suggested but is difficult to interpret owing to the lack of supporting chronological data. However, we can be certain that reduction in the buffering effects caused by the input of metalliferous-rich mine drainage and possibly land use practices (ploughing and peat extraction) has led to an increase in the trophic status of the loch. This is marked by the high density of floating and littoral aquatic vegetation that we see at the present time.

9.5 THE MECHANISMS BY WHICH HEAVY METALS ARE RETAINED IN SEDIMENT AND THE DEGREE OF POST-DEPOSITION MOVEMENT

9.5.1 Introduction

In order to make an objective assessment between natural and anthropogenic influences recorded in the geochemical profiles, the mechanisms by which heavy metals are retained in sediment and the degree of post-deposition movement needs to be assessed. Moreover, the processes likely to control their vertical distribution have to be examined in the light of the geochemical evidence. To some extent this approach has been found to be problematic as Engstrom and Wright (1984, 11-12) have stated:

"In view of the potential chemical changes in lake water, and in particulate matter during initial deposition, chemical profiles of lake sediments are difficult to interpret. Yet there remains the challenge to develop further understanding of the stratigraphic variations in chemical components for environmental reconstruction" (Engstrom and Wright 1984, 11-12).

Since this statement was made a major advancement regarding the mechanisms of chemical deposition have been made based on tracer studies using $^{207/206}$Pb (cf. Petit *et al.* 1984; Sturges *et al.* 1987; Shirahata *et al* 1980; Flegal *et al* 1989 and Ritson *et al* 1990). It is now possible, using ^{210}Pb-dated cores, combined with Pb isotopic data, to isolate and distinguish anthropogenic lead sources and also to establish the source of particulate matter and track its transportation between various locations within a lake (Ritson *et al* 1994). More recently, the adoption of step-wise sequential extraction techniques in sample preparation, allows the partition of different chemical phases between trace metals and particulate matter (Tessier *et al* 1979, 1985, Vuorela *et al* 1980). In some cases sequential methodologies might be favoured over analyses based on 'total' metal concentrations as they offer the major advantage of simulating to a certain extent, various environmental conditions to which the sediment may be subjected.

A major constraint in using sequential extraction techniques is that the chemical sequences formulated are time consuming and complex in terms of preparation and final analyses. Owing to the time needed to examine the samples within the four cores in this study, a digestion method was employed based only on the 'total' chemical component within a given sediment sample.

9.5.2 The mechanics of sediment formation

The formation of lake sediment is dominated by physical processes, the relative importance of which is particularly influenced by catchment topography, local climatic conditions, shape of the lake basin, orientation and size (Sly 1978). Glacial history, drainage processes (e.g. inflow and outflow) and local biota are seen as additional factors in the development of a sediment profile (Forstner 1977).

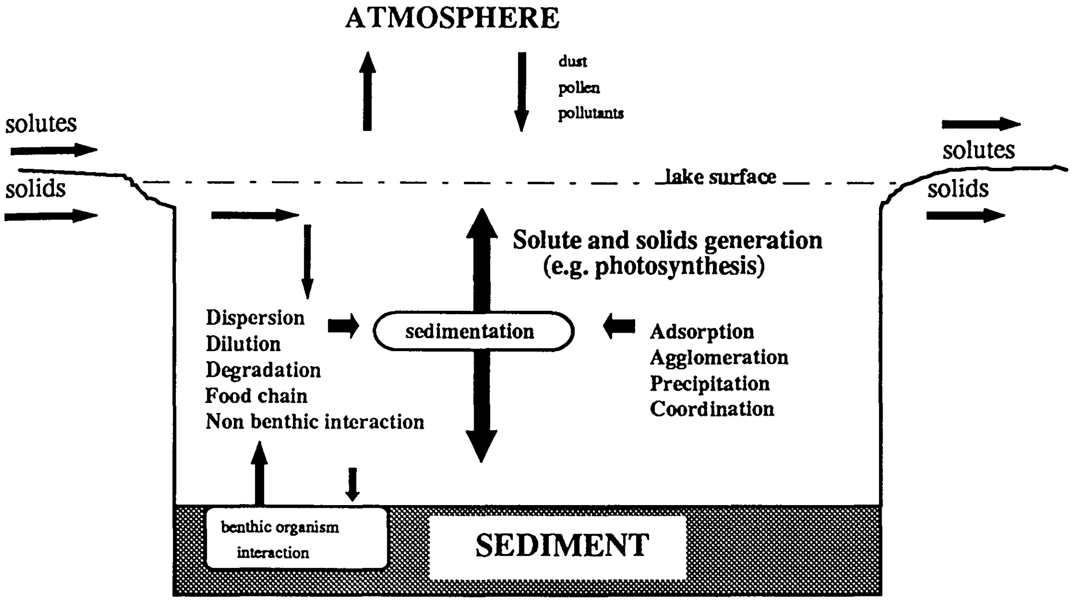


Figure 9.3 A simplified model of sediment-water interaction in a lake, after Sly (1978)

The composition of lake sediment is partly inorganic and partly organic and is either formed in the lake by living and physical-chemical processes (autochthonous), or introduced from outside the lake (allochthonous). The latter are derived from pathways outside the lake, such as catchment run-off, detrital vegetation matter and atmospheric input from precipitation such as pollen and spores, dust and man-made pollutants (Bennett *et al* 1992 and Coker *et al* 1978). A lake has been described as a 'mirror of its environment' acting as a 'sink' for incoming materials (Stumm and Baccini 1978). This description over-simplifies the mechanisms that govern how metals are incorporated into the sediment. In some cases the relative contributions of catchment and atmospheric sources are not fully understood and in others, post-deposition processes can modify the stratigraphic accuracy of the record.

9.5.3 Diagenetic (in situ) alterations in lake sediment

Sub-surface enrichment in fresh water sediment may be due to two mechanisms: (1) increased anthropogenic trace metal loading of sediments in recent years, (2) diagenetic and/or upward migration of chemical species in the pore waters (Digerfeldt, 1975; Levinson 1974; Kemp *et* *al* 1976a; Malcolm 1981 and Jennet *et al* 1987). Forstner (1977) has examined the mechanisms that can influence the distribution of major, minor and trace elements in 74 lakes from around the world. He concluded that lacustrine accumulation and dilution of metals is controlled chiefly by sorption processes between mineral and organic materials. Importantly these factors include precipitation of carbonate minerals, Fe/Mn-oxides and hydroxides. The sorption of metals by clay minerals is dependent on pH values, Fe-and Mn-hydroxides, carbonates and other organic substances. These factors proposed by Forstner, are well cited by others (cf. Stumm and Baccini (1978); Jackson *et al* (1978); Sly (1978) Engstrom and Wright (1984) and Young and Harvey (1992a 1992b). Elsewhere, researchers have reported a decrease in metal concentration in the sub-surface region of sediment cores and there have been suggestions put forward to account for these trends (cf. Galloway and Likens 1979; Norton *et al* 1981, Farmer *et al* 1980 and Brugmann and Hennings 1982). Krom (1976) and Davison *et al* (1985) have reported reducing conditions as the most likely in sediments from a lake rich in organic matter. Under such conditions and in the presence of sulphide ions, Fe, Mn, Cu, Zn and Pb will precipitate as sulphides.

In summary the most favoured mechanisms leading to the remobilization of metals towards the near-surface of freshwater sediment are:

- Increased eutrophication leading to a decrease in oxygen and consequent dissolution of Fe/Mn oxides
- Increase in the influx of acid waters lowering pH and the subsequent desorption reactions/dissolution of carbonates.
- Complexion with organic compounds
- Oxidation in the surface layers of the cores leading to the remobilisation of Mn towards this zone (i.e. the sediment water interface or redox boundary)

9.5.4 Implications for the lochs in the study area

With reference to the geochemical summary diagrams from Loch Lossit and Loch Finlaggan (Figures 7.20 and 5.42 respectively) these data suggests that in two cases we can possibly see the controlling influence (scavenging) of Mn and Fe. Firstly this phenomenon is apparent in the Loch Lossit core at 50 cm depth. At this position there is also a reduction in pH (units) which is coincident with the start of the Mn maximum (2700 mg kg^{-1}) and a similar increase in Zn, Cu and Ca at this depth.

Secondly a similar case is suggested in the Loch Finlaggan core where at 22 cm depth, the Mn maximum (1480 mg kg^{-1}) is coincident with concerted peaks in Pb, Zn and Fe. However, at this position there is no reduction in pH. Collectively these results suggest that Mn is a controlling factor at these depths. It is noteworthy that in all the chemical profiles constructed in this study, Mn does not form a peak in concentration at the surface of the cores. This is of importance as this shows that where Pb, Zn and Cu concentrate towards the near-surface (e.g. Loch Leathann and Lossit) they are doing so independently of the controlling agents mentioned above. From the above discussion, it is clear that there are several perameters affecting the chemical stratification in lake sediment. It must also be stressed at this juncture that no two lakes will be alike in terms of sediment formation and chemical record. The geomorphology, geology and hydrological input combined with interactions of land-use history, man induced and other natural agencies (catchment-erosion and atmospheric fall-out deposition) within the catchment will have a direct bearing on the geochemical composition of the lake sediment.

9.6 RADIOMETRIC DATING: AN ASSESSMENT OF ITS RELIABILITY

9.6.1 Introduction

It was stated in the original aims that the integrity of constructed time-frames was fundamental if we are to establish the link between the environmental, historical and archaeological record and to date periods of (unknown) mining activity. This section will now examine the implications of the ^{14}C ^{210}Pb and ^{137}Cs inventories from the Bharradail, Lossit and Leathann cores. Owing to the inverted nature of the ^{210}Pb curve in Loch Finlaggan, an annual rate of sedimentation could not be calculated.

9.6.2 The limitations of ^{14}C Dating at Loch Bharradail

It was concluded in Chapter 6 Section 6.6.4 that the likely reason for the apparent invariability in the age range within the ^{14}C age versus depth curve in the Loch Bharradail sediment (Figure 6.11 and Table 6.3) was the effect of fossil carbon (graphite) that has been reworked from deeper terrestrial deposits. A combination of lead mining, peat extraction and ploughing in the catchment could have caused these results. This has seriously restricted the use of ^{14}C as a means of additional support to the ^{210}Pb derived annual rate of sedimentation calculated for this core. ^{14}C of sediments cannot generally identify terrestrial inwash to a lake unless a sudden or major change in the amount of terrestrial input occurs, the most common example being the mobilisation of top soil during clearance, which manifests itself as an inversion in the dating of lake cores (Fowler *et al* 1986, 42).

In the light of these events, the scale of impact caused by mining is likely to be more acute than the clearance of woodland in the traditional sense (i.e. clear felling followed by grubbing out the roots). Mine spoil is continually exposed and often poorly vegetated, (due to its toxicity) is often reworked and prone to continued chemical weathering and erosion. At best the Loch Bharradail ^{14}C profile can only tell us that older carbon has been entering the loch on a large scale resulting in an overlap in the calculated ^{14}C ages. (see Chapter 6, Figure 6.11). These data also suggest that there has been a reduction in the amount of 'old' carbon entering the loch possibly as a result of the cessation of lead mining. This assumption is based on the depth/time curve above the ^{14}C inflection at 60 cm depth. From this position upwards there is a noticeable steepness in the curve back towards the near-surface of the core at the post-1950 'bomb' position at 1-5 cm. The geochemical record and the ^{14}C results supports the theory that there has been a protracted input of 'mine-derived' carbon into the loch basin throughout the historic period and this at least provides us with qualitative evidence to suppose that lead mining was occurring much earlier than the documented mining from the 18th century at South Ardachie.

In the light of Loch Bharadail's interesting geochemical profile it is unfortunate that the results from radiocarbon dating at Loch Bharradail are of limited use and can not be used to support the ^{210}Pb chronology. On the basis of the palynological record for the region, it is proposed that pollen analyses used as a supporting 'proxy' dating method would also have been of limited use. The palaeoecological record suggests (unsurprisingly) that the landscape was cleared of nearly all of its primary woodland much earlier than the period of interest in this study.

9.6.3 ^{210}Pb and ^{137}Cs Inventories

The ^{210}Pb dating method has been successfully used in numerous geochronological, palaeolimnological and ecological studies (cf. Chapter 3, Section 3.8.). For age calculations based on ^{210}Pb measurements, the deposits should contain perfectly preserved stratigraphic sequences unaffected by any external and internal processes. External processes might include physical alteration caused by perturbation of the lake bed such as lateral reworking or slumping, wave action and sediment focusing. Internal process might include biological alteration caused by benthic invertebrates or chemiostratigraphic alteration via diagenesis (Benoit and Hemond 1991 and 1993). Varvas *et al* (1993, 34) have argued ideal conditions do not usually exist in nature and therefore it is necessary to examine the results carefully in every case, and if possible compare them with results obtained by other independent dating methods.

Depending on the nature of the sediment, these might include varves (annual laminations), pollen analyses, historically documented erosion events, chemical influx from recorded events (point-source influx) or the distribution of artificial radio nuclides (^{137}C Cs, ^{90}Sr, $^{239/240}$Pu) produced from nuclear weapons testing or accidental release from nuclear power stations (Ritchie and McHenry 1990; Srdoc 1992; Kasuumagar 1992; Kada and Hiet 1992).

The main sources of radiocaesium in Scottish freshwater lochs are fallout from nuclear weapons testing and from the Chernobyl nuclear reactor accident (Bryant *et al.* 1993). Of significance is the maximum atmospheric deposition peak of radiocaesium in 1959-1963. The Chernobyl accident in May 1986 led to the deposition of radionuclides on the ground by wet and dry deposition processes. The particle reactive nature of many of the Chernobyl-derived fallout radionuclides such as ^{137}C and ^{134}Cs, resulted in their rapid fixation within the upper horizons of soil and lacustrine sediment (Irlweck 1991). Clarke *et al* (1988) using rainfall and air concentration data estimated total deposition of this nuclide over the United Kingdom. Along with high incidence of ^{137}C deposition fall out in SW Scotland, Islay was calculated to have received an estimated ^{137}C total deposition of 1-5 k Bq m^2.

Recent studies have shown (Bryant *et al* 1993) that the Chernobyl-derived ^{137}C has resulted in extremely high near-surface concentrations in Scottish lochs that are well above the values usually attributed to the weapons testing maximum. Bryant *et al* (1993,158 Figure 6) calculated the component sources for Loch Lomond and established a near-surface maximum of 1482 Bq kg^{-1}. (at 2-3 cm depth) compared to a lower weapons testing maximum of 931 Bq kg^{-1} at 5-6 cm depth. Such profiles are not always as well defined and a number of mechanisms can arise which may limit the use of ^{137}C for chronological support.

The use of ^{137}Cs as a dating tool is only valid if the radiocaesium has been deposited rapidly from the water column to the sediment and has not undergone any subsequent remobilisation or redistribution. Unlike ^{210}Pb, radiocaesium tends to be more mobile and susceptible to remobilisation in pore waters when certain conditions prevail. For example, Davis *et al* (1984) and Bryant *et al* (1993) have established the importance of sediment composition in determining the behaviour of radiocaesium in lacustrine sediments. In organic-rich lake sediment (>20%), ^{137}C was found to have a tendency to be mobile resulting from poor adsorption due to low clay mineral content in sediment.

9.6.4 Implications of radionuclide influx between the lochs in the study area

It has been demonstrated above that there is good conformity with the linearly-extrapolated ^{210}Pb dates and recent trends in post-1800 deposition of Pb and Zn in Lochs Leathann and

Lossit. An assessment can now be made regarding their respective ^{137}Cs inventories which have been used as additional support alongside the ^{210}Pb curves. The ^{210}Pb and ^{137}Cs inventories have been critical in establishing the degree of sediment disturbance that may have occurred within the uppermost region of the cores. Deviation from the expected trend in exponential fall-off can be taken to suggest an alteration or an increase in dry mass sedimentation rate when examined alongside the metal and physical property profiles. Appleby and Oldfield (1980) have recognised chemical disequilibrium at the sediment water interface as a possible mechanism for a departure from exponential decay. It has been mentioned above that the redox boundary can be an area within cores where radioisotopes and metals can be prone to remobilization or chemical precipitation.

Among the four lochs examined in this study, the core from Loch Finlaggan has displayed what can be termed an 'inverted' ^{210}Pb decay profile and as a result of this no chronology could be established. It will be recalled that assumptions regarding the recent and past depositional trends at Finlaggan have been severely hampered by sediment disturbance in the upper regions of the core. The Finlaggan ^{210}Pb curve (Figure 5.16) is a good example of sediment disturbance, where below 3 cm depth, the curve displays a rapid inflection away from an exponential fall-off pattern corresponding with the principle of increasing age with depth (Oldfield and Petit 1980). Furthermore the ^{210}Pb$_{excess}$ concentration in the 0-1 sediment section is extremely low (maximum concentration 61.1± 8.5 Bq kg^{-1}) in comparison to Loch Bharradail and Loch Lossit where 338.5 ±28.4 and 230 ± 10.6 Bq kg^{-1} have been attained respectively.

Examination of the relative rates of atmospheric influx of ^{210}Pb$_{excess}$ to the sediment can provide an estimate of annual flux of ^{210}Pb to the sediment surface, based on the premise of steady-state conditions. Working on ombrotrophic peat deposits, Sugden *et al* (1991) calculated the ^{210}Pb flux at Flanders Moss, Stirlingshire, at 108 Bq m^{2} yr^{-1} which is 28% lower than 150 Bq m^{2} yr^{-1} estimated for North Uist, in the Outer Hebrides. The disparity between the values was attributed to greater and more frequent rainfall at the latter site. Bryant *et al* (1993) calculated the ^{210}Pb flux at Loch Lomond as 105 Bq m^{2} yr^{-1} which is in the same order of magnitude as the figure obtained at only one of the sites in this study.

The calculated ^{210}Pb flux for the lochs investigated in this study (column 4 in Table 9.4) imply that the ^{210}Pb flux is highly variable and governed by annual precipitation, catchment size and water residence time. The method and formula used to establish the data in Table 9.4 and 9.5 are described in Chapter 3, Section 3.8.7. Calculated ^{210}Pb flux within the study area shows that Loch Finlaggan has received the lowest ^{210}Pb and ^{137}Cs flux in comparison to Lochs

Leathann, Bharradail and Lossit. The former also attains the lowest ^{210}Pb and ^{137}C surface maxima among the group of lochs investigated.

Location	**Surface max. ^{210}Pb excess (Bq kg^{-1})**	**^{210}Pb total inventory (Bq m^{-2})**	**^{210}Pb flux (Bq m^{-2} yr^{-1})**
Loch Leathann (2.8)	217.8 ±17.9	2069	64.2
Loch Lossit (1.6)	320 ± 10.6	2695	83.7
Loch Finlaggan (na)	61.1 ± 8.5	1920	59.5
Loch Bharradail (2.8)	338.5 ± 28.4	4507	140

Table 9.4 Surface maximum of ^{210}Pb excess, total ^{210}Pb inventory, ^{210}Pb flux corrected to 1:9:92. The ^{210}Pb-derived annual rate of sedimentation is shown in parentheses as mm yr^{-1}.

Location	**L/C ratio**	**Surface Max.^{137}C (Bq kg^{-1})**	**^{137}C total inventory (Bq m^{-2})**	**^{137}Cs (Bq m^{-2} yr^{-1})**
Loch Leathann	4.7	361.4	1956	60.8
Loch Lossit	7.1	290.5	2640	82.6
Loch Finlaggan	9.6	61.11	748	23.2
Loch Bharradail	149.2	338.5	3193	99.2

Table 9.5 Surface maximum of ^{137}C, total ^{137}C inventory decay corrected to 1:9:92. (note: L/C = Loch Catchment ratio).

The relationship between catchment size and radionuclide flux is evident from the results attained at Loch Leathann, Lossit and Bharradail. There is good conformity between the range of values attained from the calculated ^{210}Pb$_{total}$ inventory and ^{137}Cs flux at Lochs Lossit, Leathann and Bharradail. However, the calculated ^{137}Cs flux at Loch Finlaggan does not conform with these trends. This disparity might be explained as an effect of taphonomic processes at deposition as both ^{210}Pb and ^{137}Cs have failed to be adsorbed into sediment. Both the ^{210}Pb and the ^{137}Cs total inventory are extremely low and one can only speculate whether this is due to sediment formation processes controlled by the physical nature of the loch. Examination of multiple cores would be needed to resolve this issue and is outwith the boundaries of this research.

Lochs Bharradail and Leathann both have an annual rate of sedimentation of 2.8 mm yr^{-1} which enables both ^{137}Cs deposition curves to be plotted simultaneously against the ^{210}Pb-derived time-frame. Firstly dealing with Loch Bharradail, radiocaesium has been detectable to 45 cm depth which according to steady state conditions dates to 1832. This reflects the mobility of ^{137}Cs in pore waters. As already mentioned this radionuclide is a product of the 1950s and 1960s period weapons testing era. The Bharradail core has a surface maximum of 706 Bq kg^{-1} at 0-1 cm depth. A subsidiary peak from 5-7 cm depth occurs with 370 Bq kg^{-1} which spans the period 1974-1967. The atmospheric weapons testing maximum is usually taken as *c.*1963 which according to the calculated ^{210}Pb chronology places this year at 8 cm depth. In the light of these results it appears that there is some disparity between the ^{210}Pb-derived time-frame and the deposition of the subsidiary peak (which is taken as the 1963 weapons testing maximum). This can only be explained by a) the ^{210}Pb-derived time-frame is over-estimating the annual rate of sedimentation, which in this case would be lower (approximately 2.3 mm yr^{-1}) or b) ^{137}Cs has been relocated upwards towards the surface of the profile due to diagenetic processes occurring in the upper region of the profile.

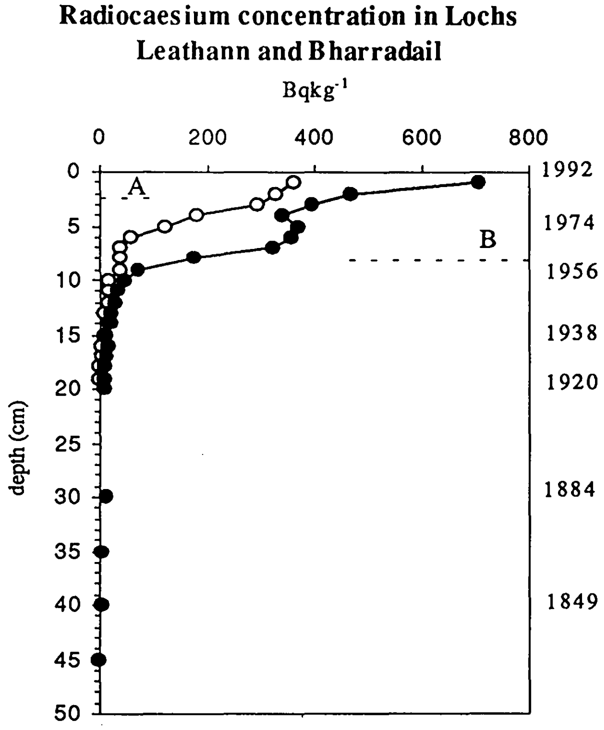


Figure 9.7 Radiocaesium concentration and ^{210}Pb-derived date in Lochs Leathann (o) and Bharradail (•) on 1.9.92. (A) marks the relative position of the Chernobyl event of 1986 and (B) marks the position of 1963 which forms the maximum period of ^{137}Cs weapons testing fall-out deposition.

Loch Leathann has a lower ^{137}C concentration and the decay profile is essentially more linear than the Bharradail ^{137}C profile. The reduction in ^{137}C at Loch Leathann may be attributed to a smaller catchment to loch ratio (4.7) which is 11.8 % smaller than the Loch Bharradail catchment ratio calculated as 39.8. The most probable explanation for the lack of correspondence between atmospheric deposition and sedimentary trends of ^{210}Pb and ^{137}Cs is that a substantial portion of these radionuclides will be derived from the watershed. Reservoir, water residence and the loch:catchment ratio contribute in a major way to the rate that these isotopes are incorporated into sediment (McCall *et al* 1984).

In summary the data has allowed the author to assess the overall integrity of the upper-surface of the cores. It is concluded that the quality of the ^{210}Pb and ^{137}Cs data is sound given the limitations of the dating model that has been applied.

9.7 Final Conclusions

It will be recalled in Chapter 1 that a conceptual 'geochemical yardstick' theory was envisaged to estimate the ‘environmental impact’ resulting from heavy metal contamination in the uppermost region of the loch sediment against the chemical record of a loch with no mineralisation within its catchment. Issues arising from this approach will now be summarised.

There are potentially three major routes that heavy metals may be derived and concentrated in freshwater sediment. These are terrestrial, atmospheric and *in-situ* diagenetic alteration. The scale of the first component is governed by the physical and chemical processes that have occurred and are ongoing in the lake’s watershed. Weathering of surrounding soils, mineral extraction, past and present farming practices, deforestation and afforestation are all important factors controlling the amount of heavy metals that have and continue to enter into sediment. The second component is a global phenomenon that has left no loch in Scotland unaffected (Battarbee and Elliot 1994). The third component is governed by internal forces such as redox boundaries, biological activity and eutrophic status.

Lake sediments are by their nature a dynamic medium, often very unstable and undergoing many alterations during the history of their formation. The relative stability of the uppermost sediment sections will to a large extent be governed by internal chemical and external forces. As we have seen the influx of radionuclides and their ability to remain unaffected by the physiochemical nature in the topmost sediment sections allows critical assessments to their use as a dating tool and indicator of the degree of sediment disturbance.

The integrity of a chosen ^{210}Pb-dating model can be seen to be reliable if a) there is additional chronological support and b) that sediment mixing is negligible. From the standpoint of soil contamination, the conceptual environmental pathway model (Figure 2.7) and results of geochemical survey have established that there is localised heavy metal contamination controlled by the distribution of sulphide-rich spoil heaps.

The major environmental problem of mine tailings is that they possess unfavourable porosity, aeration, water infiltration and percolation properties, along with a high bulk density and an absence of structural aggregates. The consequence is that waste materials representing past eras of mining contain high residual quantities of toxic metals often in excess of 1% weight of elements such as Pb, Zn and Cu (Johnson 1994). Heavy metals leached from the spoil heaps or from ore processing enter streams and adits along with those entering the loch via ground water or surface run-off. Ultimately, these can be incorporated into loch sediment under the mechanisms described in Section 9.5 above. As a general rule, if the sedimentation rate in the loch is fairly constant, the relative metal distribution will reflect mainly the natural metal distribution in the catchment area (Quarfort 1977, 66).

However the metal concentration profiles may become distorted by fluctuations in the sediment rate and the increase of the metal burden that is a direct result of mining activities in the catchment. Moreover, over the historic period we may expect to see intermittent increases in the Ca, Mn and Fe profiles caused by land use, such as ploughing, afforestation and natural erosion in the catchment soils. The latter may significantly increase magnetic susceptibility values. Similarly we may also expect to see variability in the LOI profile as a result of higher mineral influx (cf. Finlaggan Marsh at 150 cm and Loch Leathann at 23 cm depth). Fluctuations in the geochemical profiles may also occur as a result of intermittent heavy metal influx, and such events could be attributed to short phases of 'early' lead mining.

It is argued that post-1850's atmospheric pollution has affected the lochs in the study area resulting in significant near-surface increases in Pb, Zn and Cu. At Lochs Lossit and Leathann this has led to marked increases in Zn and Cu that are significantly higher than proposed baseline concentrations. In assessing the impact of mining from earlier periods (e.g. pre late 18th-early 19th century episodes) the evidence confirms that lead mining was occurring at the mines of Glasgo Beag (near Loch Lossit) and South Ardachie (near Loch Bharradail) in the 16th and 17th century. The environmental record from Loch Lossit suggests that mining was occurring as early as the 14th century (*c.*1367). The depth versus metal concentration profiles (Figures 5.42, 6.22, 7.19 and 8.20 respectively) show the general trends in concentration and these have been 'zoned' according to major increases across respective chemical inventories.

Bearing in mind the limitations of extrapolating ^{210}Pb-derived dates beyond *c.*1850, the dates should be viewed as 'suggested' rather than absolute. Table 9.6 below combines all the major Pb peaks from the four lochs with suggested minimum ages at a given depth. This data shows the maximum Pb values in relation to their respective corresponding baseline values and the calculated 'Above Baseline Enrichment' (ABE) ratio above these values. Within the limitations of the ^{210}Pb dating method it has to be concluded that the dates associated with these mining events are not in conflict with the historical record.

Location	Depth (cm)	Pb conc. mg kg^{-1}	Baseline mg kg^{-1}	Suggested date
Loch Lossit	62	85	43 (0.5)	*c.*1529
" "	106	130	43 (0.6)	*c.*1367
Loch Bharradail	38	800	68 (0.9)	*c.*1849
" "	90	695	68 (0.9)	*c.*1634
" "	108	757	68 (0.9)	*c.*1563
Loch Finlaggan	20	182	42.5 (0.7)	*c.*1830*
Finlaggan Marsh	125	48	n/a-	*c.*1586*

Table 9.6 Summary table of the major Pb influx events and associated 'suggested' minimum dates. (*) Note the Finlaggan dates are very tentative and were calculated using proxy data such as ^{137}Cs-derived sediment rate, recent afforestation event and 1850's onset in pollution. The values in parenthesis are calculated as a ratio of 'above baseline enrichment' (ABE) established from V-B/V where V is the observed Pb value and B is the baseline value respectively.

Finally, the results of geochemical analysis on sediment sections from Lochs Lossit, Bharradail and Finlaggan Marsh have established that earlier periods of lead exploitation can be identified within the environmental record. The evidence from Loch Lossit strongly suggests that lead was being mined on Islay as early as the mid-14th century. In the Loch Bharradail core, there is a significant correlation between the geochemical profiles and the ^{210}Pb-derived chronology. The calculated ^{210}Pb dates support an increase in lead exploitation during the late 16th to early 17th century. The environmental impact of late 18th century period of mining activity is also represented within the geochemical record of the upper regions of the core. There is also a very good relationship between this environmental record and the contemporary 18th century accounts that describe the presence of "1,000 early workings" within the study area.

CHAPTER 10: CONCLUSIONS

10.1. Introduction

In the absence of documentary evidence about lead mining in NE Islay before the 16th century, an analogue approach has been used to identify early lead mining in the region. The main objectives in this study have been:

- To record the archaeological evidence of 18th and 19th century lead mining
- To assess the principal sources of heavy metal contamination in the study region
- To determine the possible routes of heavy metals into loch sediment
- To identify the mechanisms by which heavy metals are retained in sediment and the degree of post-deposition movement within sediment cores
- To develop a chronology for mine-derived heavy metal contamination within loch sediment and assess its reliability.

These objectives have been essential to establish the link between the environmental, historical and archaeological record and to date the periods of earlier (unknown) mining activity. The results obtained from geochemical analyses of loch sediment confirm that the north-east region of Islay indeed contains a longer history of lead mining that pre-dates the earliest historical account of 1511-12. The implementation of a multi-disciplinary methodology has allowed a historical and environmental perspective of lead mining in the region to be better understood. The study has highlighted and tried to overcome some of the problems encountered in the recovery, analysis and dating of freshwater sediment. A broad-based integrated systems-approach has led to a greater understanding of a lesser-known region of lead mining in Scotland.

10.2 General review

Chapter 1 stressed the need to develop an approach that would allow the author to gain an insight into the importance of lead at a significant period in the history of the Lordship of the Isles (13-15th century). The historically important site of Finlaggan formed a backdrop to the study owing to its political importance during these centuries. Assuming the ease with which lead could be exploited, and given the importance of galena as a secondary source of silver, it was therefore considered that local ore deposits could not have gone unnoticed. This chapter further stressed the merits of examining freshwater sediment with the aim of monitoring the contaminatory effects from mining events within the catchment of three freshwater lochs. It was explained that freshwater sediment can often retain a reliable environmental record of events that have occurred within the catchment. However, a major constraint encountered at Loch Finlaggan has been sediment disturbance. This restricted the calculation of a time-frame based on ^{210}Pb with additional support from ^{137}Cs. Without reliable isotopic inventories, assumptions regarding specific events that has occurred within a given catchment can only be tentative. The geochemical 'yardstick' theory was explained; whereby the relative scale of mine-derived contamination from the last recorded period of lead mining would be used to assess the level of contamination from small-scale mining episodes deeper in the sedimentary profiles. The scale and duration of these earlier stratified events could then assessed in relation to Loch Leathann a control site where no lead mineralisation is recorded in its catchment.

Chapter 2 provided a brief account of the history of non-ferrous metal mining in Great Britain and focused on Scotland in particular, in order to underline the importance and technical developments of this industry. A review of the principal methods in mining, refining and smelting not only highlighted the technical aspects of the industry, but also assessed its contaminatory nature. This provided the backdrop to the problems of heavy metal contamination at and around abandoned mines. The contaminatory effects of the heavy metals Pb, Zn and Cu were examined in relation to the terrestrial environment, drawing on data from research undertaken at sites with a long history of metal extraction. This section concluded that in Scotland only limited work has been undertaken on the environmental impact of mining owing to the smaller regional scale of Scottish mining operations (this author predicts that in the future this situation will radically alter as the problem of coal mining-related pollution becomes more acute). Section 2.4 discussed the effects of non-ferrous metal mining on the fluvial environment and established that lacustrine sediment is a powerful medium with which to examine long and short-term environmental changes alongside the history of pollution under the general term of historical monitoring. A conceptual model was formulated to relate the three principal environmental gradients of heavy metals into loch sediment in the study area. The model predicted that the principal environmental pathways of heavy metals into loch sediment were primarily, fluvial (via streams and seepage-run-off) mining (direct input via leats and mine adits) and recent (post 1850's) atmospheric fall-out deposition.

At the outset of any experiment or study, a research design with a clear objective is an important criterion, without which the study may not be successful. Chapter 3 set out in clear terms the methods and objectives that were to be employed in this study. It was further demonstrated that the loch catchment unit as a basis of study formed an integral part of this research. Importantly, a systems approach was developed to gain an environmental and historical perspective of lead mining in the study area. Wet and dry methods of sediment handling maximised the use of different sediment sections. Experimental design and laboratory control to minimise contamination was fundamental for ensuring that the final result was to be meaningful and unbiased. Therefore, more time was spent on the decontamination of glassware and equipment than on direct analyses themselves to ensure confidence in the final results.

In relation to fieldwork, a geographical and archaeological approach ensured that the catchment of a given loch was understood, both in terms of its geomorphology and archaeology. The latter was seen to be more extensive than was originally thought and archaeological field survey has brought to light many new features previously unrecorded in the landscape. Conversely, the results of field survey have established that the industry was extensive in some parts and more intensive in others. As with any project that investigates the history of a tract of landscape, more questions are often raised than can ever be answered. This aspect of the study established that mines dated to the 18th century are dove-tailed within an existing framework of agricultural settlement and confirms the author's notion of 'farmer-miners' who were engaged in a dual economy. Mulreesh forms the only 19th century mining complex in the region. Here miners were recruited from the mainland and housed off-site in purpose built houses. It is at Mulreesh that we also see the introduction of steam power and a hydraulically operated ore washing plant which highlights the level of financial investment at this particular mine. It was argued that shipment costs of lead were cheaper (as ballast) than the overland routes to the Port of Leith which emerged as the main lead export route to the Low Countries in the 17-18th centuries.

Sections 5.5-5.6 in Chapter 5 examined sediment recovered from Loch Finlaggan and Finlaggan Marsh. Unfortunately, the cores recovered from the loch were degraded. The core selected for analyses was seen to be affected by mechanical or biological mixing that limited its use in the calculation of an annual rate of sedimentation using ^{210}Pb. ^{137}Cs has been less affected by mixing and some useful qualitative data was obtained from the Loch Finlaggan core. With respect to the site of Finlaggan Marsh, a 4m long sequence of sediment did provide some interesting data concerning the effects of local lead mining. Other important physical and chemical data were obtained to show that as with many stratified peat or lake deposits in Scotland, the upper sediment sections contain a significant inventory of post-1850 industrially-derived atmospheric pollution. Indirectly this was used as a proxy dating method alongside a recorded period of afforestation that occurred in 1982 in the Loch Finlaggan catchment.

The 18th century mine known as South Ardachie is linked directly to Loch Bharradail by a large drainage leat. This has led to a huge increase in the lochs heavy metal burden, owing to the influx of metalliferous-rich mine drainage. Chapter 6 discussed the results of geochemical and archaeological survey within the catchment of this loch. It is concluded that two periods of mining activity can be observed that pre-date the peak mining period of the late 18th century. The long history of mining and other land use activities in the catchment have seriously affected the potential for dating the sediment using ^{14}C owing to the influx of 'old' reworked carbon.

Loch Lossit formed the subject of Chapter 7. This loch is suggested to contain the earliest record of lead mining subject to the limitations of the ^{210}Pb dating method. The sediment contained a strong indication of atmospheric pollution which is also mirrored in Loch Leathann which was chosen as a control. It will be recalled that in order to assess the relative scale of soil and sediment contamination at a given site, a 'control' environment is needed that is geologically similar, with reduced or no known record of heavy metal contamination. Chapter 8 discussed the usefulness of Loch Leathann as a control site. Both sediment and catchment soils proved adequate for this purpose and both recorded 'background' Pb, Zn and Cu concentrations. These were seen to be well within the 'normal' range recorded elsewhere in the literature concerned with heavy metal contamination of soil and freshwater lake sediment. The historical record of atmospherically-derived pollution has been highlighted within this loch and the calculated concentration profile for Pb and Zn shows close conformity with the wider geographical trends of atmospheric pollution recorded in another remote loch in the on the mainland and in the Western Isles of Scotland. It was further concluded that at the present, no loch remains unaffected by the onset of post-*c.*1850s atmospherically-derived pollution within this region.

In Chapter 9, the evidence obtained from geochemical analyses of the four lochs was examined collectively alongside other wider environmental issues, such as contemporary soil contamination and recent atmospherically-derived Pb and Zn. Consideration of the limitations of the dating methods employed was also assessed accordingly. The overall success of the dating method employed governs the level of confidence in the final interpretation. It was commented earlier in this thesis that in order to obtain the widest regional perspective of small-scale mining environmental impact, only replicate cores were likely to be sufficient. The viability of multiple-core analyses can only be speculated on. Time and cost ultimately governs the limits of what can be achieved within the framework of study. Finally, the results of the timing of historic mining have been integrated into tabular form based on data calculated from the ^{210}Pb-dated depth versus concentration profiles. The overall success of this thesis is reflected in the quality of environmental data obtained from the study region confirms that lead mining occurred earlier than the historical documentary record would suggest.

10.3 Future and ongoing research

This research has brought to light the potential for future work within the study area. Among the most important will be the implementation of radiocarbon dates on specific sediment sequences at Finlaggan Marsh. This will provide the archaeologists working at Finlaggan with a clearer picture of the age when the marsh began to form and address important environmental questions such as the palaeoecological and palyhydrological changes that have been observed within the pollen and stratigraphic record. This thesis has also stimulated further archaeological interest in the industrial heritage in the study region which until now has been neglected. It is envisaged that the Washings complex at Mulreesh be given some form of protection through Historic Scotland to safeguard what is left of these unique remains.

A collaborative project involving the author and Dr Jon Sadler of the Department of Geography, University of Birmingham, is currently underway to identify deformities in *Chirinomidaea* larvae. Sediment sections from Loch Bharradail and Leathann are being examined to assess if *Chirinomidaea* can be used as an indicator of heavy metal contamination in lacustrine sediment.

Plate 1. A Tertiary basalt dyke seen in section at Ballygrant limestone quarry, NE.Islay. Note the sharp boundary between the blue Dalradian limestone and the olivine basalt.

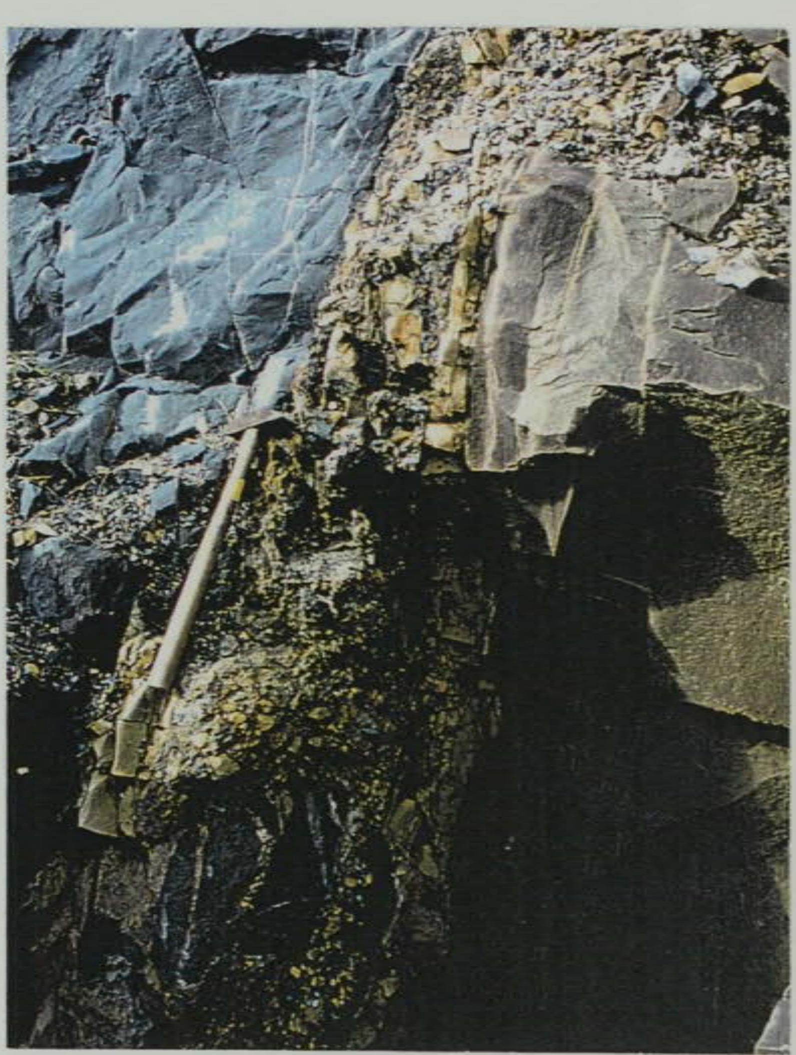


Plate 2. A close-up of a metamorphic contact zone between the Tertiary dyke and the Dalradian limestone. The zone contains a breccia deposit of galena, basalt and limestone.

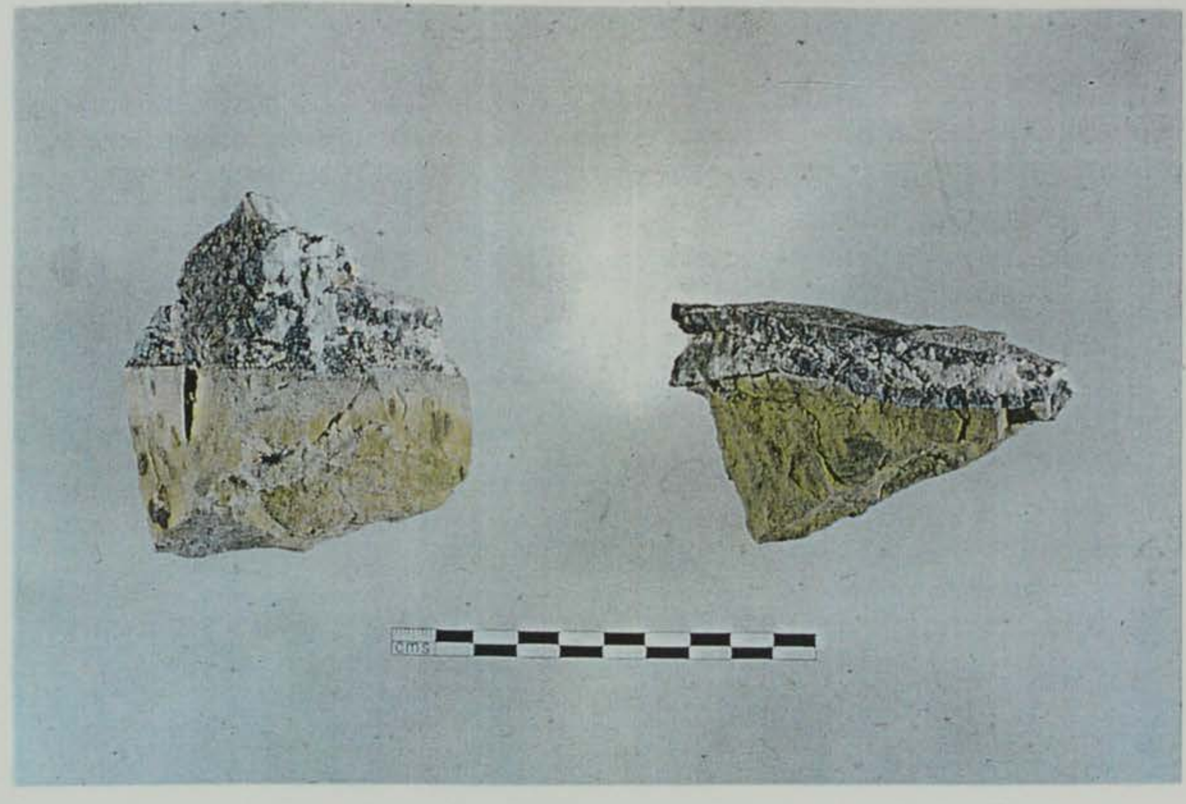


Plate 3. A close-up of lead ore recovered from the contact zone in Plate 2.

Plate 4. Close-up of a typical piece of 'gangue' or waste rock taken from a spoil heap from the Robolls mine close to Loch Finlaggan. Iron pyrites is a common guest constituent in spoil heaps and is easily weathered to form extremely acidic solutes.

Plate 5. The lead-smelting furnace at Freeport, 1772. This site is now probably the site of the Coal Isla whisky distillery situated on the Sound of Islay.

Plate 6. A Cornish beam engine-house and cottage at the Mulreesh mine taken from the south, (Feature 3 in Figure 5.4).

Plate 7. Spoil tips surrounding the entrance of the North Shaft at Mulreesh, (see Figure 5.4).

Plate 8. The remains of a rectangular turf building surrounded by vegetated spoil heaps at South Ardachie mine close to Loch Bharradail, (Feature 11 in Figure 6.2).

Plate 9. The gravity coring device used in this study. The core tube shown is 2m long with a diameter of 7.9 cm.

Plate 10. Finlaggan Marsh (taken from the south in the spring) with the archaeological site of *Eilean Mor* (Finlaggan) in the distance, (See Figure 5.29).

Plate 11. General view of Loch Finlaggan taken from the south, (see Figure 5.14).

Plate 12. The leat flowing from the South Ardachie mine into Loch Bharradail, (see Figure 6.2)

Plate 13. A general view of Loch Bharradail taken from the eastern shore, (see Figure 6.1 and 6.6).

Plate 14. A general view of Loch Lossit taken from the south-west, (see Figure 7.1 and 7.5).

Plate 15. A general view of Loch Leathann taken from the north-east. This loch has no mining in its catchment and was chosen as a control, (see Figure 8.1 and 8.5).

Appendix 2 Loch Finlaggan and Finlaggan Marsh

DRL Soil Geochemical data (Portnealon Mine)				
coordinate	coordinate	ppm	ppm	ppm
x	y	Pb	Zn	Cu
690	70	0	0	0
650	10	0	0	0
610	130	0	0	0
560	170	15	12	2
510	210	23	13	2
470	240	76	15	7
470	270	102	56	10
370	310	170	114	28
320	350	400	146	57
280	380	2280	295	142
230	420	470	118	63
180	450	253	117	39
130	490	87	161	18
770	160	49	98	5
720	200	41	51	5
670	240	39	47	4
620	270	29	41	3
570	310	22	20	3
520	340	86	41	6
470	380	174	108	14
430	420	460	152	54
370	460	270	144	31
330	490	374	85	47
270	520	180	78	37
230	470	0	0	0
200	600	0	0	0
840	250	69	39	6
800	300	44	64	6
760	330	87	37	13
730	350	163	79	27
670	380	25	28	5
650	410	84	298	20
600	460	137	100	23
850	490	5000	185	1150
490	530	250	129	49
440	570	233	197	44
380	600	181	118	41
330	640	154	115	25
270	680	126	90	13
910	360	47	39	5
870	390	0	0	0
810	420	72	21	6
750	450	24	12	0
690	500	0	0	0
630	520	145	53	12
560	570	331	174	45
490	620	243	170	34
430	650	163	109	33
380	680	176	115	32
320	720	171	108	15
930	520	48	68	3
950	580	147	25	5
900	590	54	19	5

Table 1. Soil geochemical data from the Portnealon mine in the catchment of Loch Finlagggan. (Figures 5.10, 5.11& 5.12)

DRL Soil Geochemical Data (Portnealon Mine)				
coordinate	coordinate	ppm	ppm	ppm
840	600	126	219	12
800	610	129	225	11
740	640	132	152	19
690	690	138	158	31
640	730	170	227	32
590	760	137	126	33
810	530	136	141	32
470	860	116	108	29
420	900	158	147	25
1090	578	67	66	11
1040	610	34	88	7
990	640	41	21	3
940	680	76	36	6
890	710	60	27	7
820	760	173	122	21
790	790	197	163	30
750	820	119	126	26
690	860	142	148	27
650	890	87	92	22
590	920	163	136	32
550	960	114	116	29
500	990	219	163	25
1240	790	39	12	3
1180	820	63	31	4
1120	850	51	25	3
1070	870	99	87	6
1010	890	126	99	8
950	920	216	153	20
900	940	220	270	42
840	970	228	173	31
780	1000	122	106	44
730	1020	108	123	25
670	1050	237	113	42
620	1070	133	127	27
580	1090	142	113	22

Table 1. Cont. Soil geochemical data from the Portnealon mine in the catchment of Loch Finlagggan. (Chapter 5 Figures 5.10, 5.11 & 5.12)

Appendix 2 Loch Finlaggan and Finlaggan Marsh

	Lead			Zinc			Copper	
cm	mg kg^{-1}	3 Period Ave.	cm	mg kg^{-1}	3 Period Ave.	cm	mg kg^{-1}	3 Period Ave.
82	106		82	96		82	26	
80	154	131.33	80	193	142.33	80	25	25.33
78	134	142.33	78	138	165.33	78	22	23.00
76	139	132.33	76	165	153.67	76	25	24.00
74	124	129.33	74	158	158.00	74	36	32.33
72	125	124.33	72	151	156.67	72	26	29.33
70	124	128.33	70	161	169.67	70	25	25.33
68	136	125.67	68	197	169.67	68	29	27.67
66	117	130.67	66	151	177.33	66	25	26.33
64	139	123.33	64	184	164.00	64	33	30.33
62	114	129.00	62	157	180.00	62	26	28.33
60	134	119.00	60	199	168.00	60	33	30.67
58	109	119.33	58	148	167.00	58	27	29.00
56	115	112.67	56	154	151.00	56	27	27.00
54	114	117.33	54	151	139.67	54	27	27.00
52	123	121.67	52	114	144.33	52	29	28.33
50	128	125.67	50	168	149.00	50	30	29.67
48	126	125.67	48	165	166.00	48	30	30.00
46	123	119.33	46	165	163.00	46	29	29.33
44	109	112.33	44	159	157.00	44	30	29.67
42	105	107.67	42	147	157.67	42	27	28.00
40	109	117.00	40	167	168.00	40	28	27.67
38	137	128.33	38	190	175.00	38	35	32.67
36	139	134.67	36	168	177.00	36	30	31.67
34	128	126.67	34	173	161.00	34	27	28.00
32	113	122.33	32	142	155.67	32	22	23.67
30	126	117.33	30	152	149.67	30	23	22.67
28	113	117.33	28	155	145.67	28	31	28.33
26	113	120.00	26	130	148.67	26	27	28.33
24	134	136.67	24	161	137.33	24	27	27.00
22	163	177.67	22	121	167.00	22	28	27.67
20	236	182.33	20	219	166.00	20	43	38.00
18	148	163.67	18	158	167.33	18	37	39.00
16	107	121.67	16	125	141.33	16	32	33.67
14	110	100.00	14	141	125.33	14	33	32.67
12	83	94.00	12	110	122.00	12	27	29.00
10	89	101.33	10	115	130.67	10	28	27.67
8	132	118.33	8	167	141.00	8	38	34.67
6	134	133.33	6	141	150.00	6	35	36.00
4	134	130.67	4	142	148.33	4	35	35.00
2	124	119.67	2	162	151.00	2	25	28.33
1	101		1	149		1	27	

Table 2. Loch Finlaggan Core 2 Geochemical data.
(Chapter 5 Figures 5.18, 5.19 & 5.20)

	Calcium			Manganese			Iron
cm	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}
82	5		82	763		82	3600
80	25	18.33	80	1138	996.33	80	2950
78	25	29.33	78	1088	1138.00	78	3650
76	38	32.67	76	1188	1146.33	76	4350
74	35	34.33	74	1163	1154.67	74	3050
72	30	33.33	72	1113	1142.00	72	2800
70	35	36.33	70	1150	1175.33	70	2900
68	44	42.33	68	1263	1183.67	68	4300
66	48	48.67	66	1138	1250.33	66	3500
64	54	45.67	64	1350	1217.00	64	3500
62	35	40.33	62	1163	1225.33	62	2950
60	32	28.67	60	1163	1154.67	60	3700
58	19	20.00	58	1138	1138.00	58	3500
56	9	15.33	56	1113	1121.33	56	3850
54	18	17.33	54	1113	1092.00	54	3000
52	25	21.00	52	1050	1112.67	52	4050
50	20	23.00	50	1175	1129.33	50	3850
48	24	21.00	48	1163	1162.67	48	3200
46	19	17.00	46	1150	1104.33	46	3500
44	8	11.67	44	1000	1058.33	44	3500
42	8	10.00	42	1025	1041.67	42	3550
40	14	13.33	40	1100	1066.67	40	3250
38	18	12.67	38	1075	1066.67	38	4700
36	6	15.33	36	1025	1079.33	36	3100
34	22	13.67	34	1138	1037.67	34	3350
32	13	14.33	32	950	1029.33	32	3150
30	8	11.33	30	1000	987.67	30	3150
28	13	10.00	28	1013	1008.67	28	3250
26	9	12.00	26	1013	1250.33	26	3350
24	14	17.67	24	1725	1354.33	24	3600
22	30	27.33	22	1325	1466.67	22	4600
20	38	34.33	20	1350	1329.33	20	4950
18	35	33.00	18	1313	1196.00	18	4100
16	26	33.00	16	925	1171.00	16	3250
14	38	32.33	14	1275	1179.33	14	4100
12	33	33.67	12	1338	1312.67	12	3300
10	30	36.00	10	1325	1337.67	10	3050
8	45	39.33	8	1350	1350.00	8	4750
6	43	44.33	6	1375	1371.00	6	3600
4	45	44.00	4	1388	1362.67	4	3450
2	44	38.00	2	1325	1271.00	2	3450
1	25		1	1100		1	3350

Table 2 .Cont. Loch Finlaggan Core 2 Geochemical data.
(Chapter 5 Figures 5.21,5.22 & 5.23)

	LOI	Mineral	Pore water
cm	%	%	%
82	18	82	70
80	21	79	70
78	19	81	72
76	20	80	71
74	19	81	71
72	19	81	71
70	19	81	70
68	18	82	70
66	19	81	69
64	19	81	69
62	19	81	69
60	18	82	68
58	19	81	66
56	18	82	67
54	18	82	66
52	18	82	66
50	18	82	66
48	18	82	66
46	14	86	66
44	17	83	64
42	18	82	65
40	16	84	62
38	17	83	63
36	21	79	64
34	16	84	64
32	16	84	64
30	15	85	65
28	21	79	70
26	21	79	69
24	24	76	66
22	19	81	67
20	20	80	67
18	17	83	64
16	18	82	66
14	25	75	72
12	27	73	72
10	23	77	68
8	24	76	69
6	28	72	72
4	24	76	69
2	24	76	69
1	23	77	69

Table 2. Cont. Loch Finlaggan Core 2 physical properties.
(Chapter 5 Figures 5.24,5.25 & 5.28)

Appendix 2 Loch Finlaggan and Finlaggan Marsh

Depth	Pellet Wgt.	$^{210}Pb_{excess}$			Pellet wgt.	^{137}Cs	
cm	k gms	Bq kg^{-1}	P. wgt x activity		k gms	Bq kg^{-1}	P. wgt.x activity
1	0.01419	61.1	0.867009		0.01419	78.2	1.109658
2	0.01545	40.5	0.625725		0.01545	55.9	0.863655
3	0.01448	27.7	0.401096		0.01448	43.3	0.626984
4	0.01433	46	0.65918		0.01433	47.6	0.682108
5	0.01524	28.7	0.437388		0.01524	28	0.42672
6	0.01528	31	0.47368		0.01528	23.3	0.356024
7	0.01472	8.9	0.131008		0.01472	16.4	0.241408
8	0.015	28.8	0.432		0.015	12.4	0.186
9	0.01511	11.4	0.172254		0.01511	8.5	0.128435
10	0.01588	28	0.44464		0.01588	10.3	0.163564
11	0.01536	31.8	0.488448		0.01536	9.3	0.142848
12	0.01513	18.2	0.275366		0.01513	8.3	0.125579
13	0.01516	14.9	0.225884		0.01516	3.9	0.059124
14	0.01514	27.8	0.420892		0.01514	5.9	0.089326
15	0.01509	13.8	0.208242		0.01509	6.9	0.104121
16	0.01509	13.8	0.208242		Sum	5.305554	
17	0.01509	13.8	0.208242		inventory	748	
18	0.01509	13.8	0.208242		flux	23.2	
19	0.01509	13.8	0.208242				
20	0.01509	13.8	0.208242				
21	0.01509	13.8	0.208242				
22	0.01509	13.8	0.208242				
23	0.01509	13.8	0.208242				
24	0.01509	13.8	0.208242				
25	0.01509	13.8	0.208242				
26	0.01509	13.8	0.208242				
27	0.01509	13.8	0.208242				
28	0.01509	13.8	0.208242				
29	0.01509	13.8	0.208242				
30	0.01509	13.8	0.208242				
31	0.01509	13.8	0.208242				
32	0.01509	13.8	0.208242				
33	0.01509	13.8	0.208242				
34	0.01509	13.8	0.208242				
35	0.01509	13.8	0.208242				
36	0.01509	13.8	0.208242				
37	0.01509	13.8	0.208242				
38	0.01509	13.8	0.208242				
39	0.01509	13.8	0.208242				
40	0.010987	15.9	0.1746933				
41	0.010987	15.9	0.1746933				
42	0.010987	15.9	0.1746933				
43	0.010987	15.9	0.1746933				
44	0.010987	15.9	0.1746933				
45	0.010987	15.9	0.1746933				
46	0.010987	15.9	0.1746933				
47	0.010987	15.9	0.1746933				
48	0.010987	15.9	0.1746933				
49	0.010987	15.9	0.1746933				
50	0.0995	6.1	0.60695				
		Sum	13.614503				
		inventory	1920				
		flux	59.7				

Table 3. ^{210}Pb and ^{137}Cs inventory and flux data. (Table 9.4 page 296)

	$^{210}Pb_{excess}$		1 sigma	^{137}Cs	1 sigma
cm	Bq kg^{-1}	LNAO	Bq kg^{-1}	Bq kg^{-1}	Bq kg^{-1}
1	61.1	4.112512	8.5	78.2	1.7
2	40.5	3.701301974	8.9	55.9	1.2
3	27.7	3.321432413	10.5	43.3	1.1
4	46	3.828641396	10.3	47.6	1.3
5	28.7	3.356897123	9.7	28	0.9
6	31	3.433987204	9.5	23.3	1.1
7	8.9	2.186051277	9.1	16.4	1.4
8	28.8	3.360375387	11	12.4	0.8
9	11.4	2.433613355	11.1	8.5	0.8
10	28	3.33220451	10.9	10.3	0.8
11	31.8	3.45946629	13.6	9.3	0.8
12	18.2	2.901421594	11.2	8.3	1.8
13	14.9	2.701361213	10.5	3.9	0.7
14	27.8	3.325036021	12.1	5.9	0.7
15	13.8	2.624668592	9.2	6.9	1.3
40	15.9	2.766319109	11.7	0	0
50	6.1	1.808288771	9.6	0	0

Table 4. $^{210}Pb_{excess}$ and ^{137}Cs activities with 1 sigma error. LNAO is the calculated log normal $^{210}Pb_{excess}$ activity. (Figures 5.16 & 5.17)

Appendix 3 Loch Bharradail

DRL Soil Geochemical data (South Ardachie Mine)				
coodinate	**coordinate**	**ppm**	**ppm**	**ppm**
x	y	Pb	Zn	Cu
120	20	25	30	2
120	90	55	45	4
120	140	105	290	12
120	200	40	90	12
120	250	320	245	22
120	310	45	30	4
120	360	50	30	2
120	430	125	130	12
120	480	65	75	2
120	540	45	45	8
120	590	40	130	6
120	640	45	60	6
120	700	25	20	2
400	10	60	35	8
400	80	35	85	12
400	120	650	265	56
400	180	60	98	6
400	240	15	5	2
400	300	250	120	6
400	340	95	70	8
400	410	200	106	8
400	460	60	70	12
400	530	50	130	16
400	580	20	30	2
400	640	30	15	2
400	700	10	40	4
700	10	30	55	16
700	60	80	65	14
700	130	150	120	58
700	180	65	210	10
700	250	70	100	10
700	290	225	150	14
700	350	180	85	44
700	410	115	20	2
700	470	100	80	6
700	520	80	55	6
700	580	30	25	6
700	640	35	55	6
999	10	40	160	14
999	60	20	60	6
999	120	30	15	4
999	170	160	100	8
999	230	170	230	12
999	290	160	260	10
999	340	145	120	16
999	400	65	55	8
999	460	30	55	6
999	520	35	40	2
999	570	135	380	10
999	640	15	35	2
999	750	15	25	2

Table 1.Loch Bharradail Catchment. DRL Soil Geochemical Survey Data (Chapter 6. Figures 6.3, 6.4 & 6.5)

Depth	lead		Depth	Zinc		Depth	Copper	
cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.
135	813		135	530		135	28	22.00
134	758	773.67	134	465	503.33	134	24	21.00
132	750	751.00	132	515	486.67	132	14	20.00
130	745	716.00	130	480	483.33	130	25	23.00
128	653	697.67	128	455	473.33	128	21	22.67
126	695	677.00	126	485	470.00	126	23	23.33
124	683	674.33	124	470	453.33	124	24	23.33
122	645	636.00	122	405	425.00	122	23	23.00
120	580	606.00	120	400	395.00	120	23	24.00
118	593	640.33	118	380	421.67	118	23	24.33
116	748	659.67	116	485	425.00	116	26	24.67
114	638	641.33	114	410	425.00	114	24	26.33
112	538	700.33	112	380	436.00	112	24	27.67
110	925	735.33	110	518	424.33	110	31	27.00
108	743	757.00	108	375	432.67	108	28	24.00
106	603	661.33	106	405	403.33	106	22	21.00
104	638	651.33	104	430	440.00	104	22	19.33
102	713	584.67	102	485	423.33	102	19	19.67
100	403	562.00	100	355	421.67	100	17	20.33
98	570	469.33	98	425	376.67	98	23	20.33
96	435	438.33	96	350	400.00	96	21	21.67
94	310	503.33	94	425	400.00	94	17	22.67
92	765	583.33	92	425	420.00	92	27	24.67
90	675	695.00	90	410	383.33	90	24	23.67
88	645	656.00	88	315	378.33	88	23	22.67
86	648	568.67	86	410	311.67	86	24	23.00
84	413	568.67	84	210	345.00	84	21	21.33
82	645	512.00	82	415	311.67	82	24	20.00
80	478	517.00	80	310	340.00	80	19	17.67
78	428	402.00	78	295	298.33	78	17	17.00
76	300	353.67	76	290	293.33	76	17	17.00
74	333	343.67	74	295	301.67	74	17	16.00
72	398	335.33	72	320	285.00	72	17	14.67
70	275	323.67	70	240	266.67	70	14	13.33
68	298	297.67	68	240	231.67	68	13	13.00
66	320	395.33	66	215	226.67	66	13	14.33
64	568	393.67	64	225	240.00	64	13	17.33
62	293	498.67	62	280	295.00	62	17	19.67
60	635	539.33	60	380	336.67	60	22	20.67
58	690	653.33	58	350	373.33	58	20	21.00
56	635	646.67	56	390	398.33	56	20	21.00
54	615	630.00	54	455	415.00	54	23	22.00
52	640	655.00	52	400	428.33	52	20	23.00
50	710	711.00	50	430	426.67	50	23	26.33
48	783	774.33	48	450	358.33	48	26	26.33
46	830	801.00	46	195	356.67	46	30	27.67
44	790	784.33	44	425	356.67	44	23	27.67

Table 2. Loch Bharadail Core 2 Geochemical data (Chapter 6 Figures 6.12, 6.13 & 6.14)

Depth	lead		Depth	Zinc		Depth	Copper	
cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.
42	733	748.67	42	450	441.67	42	30	30.00
40	723	761.33	40	450	458.33	40	30	29.67
38	828	800.33	38	475	468.33	38	30	24.67
36	850	742.67	36	480	455.00	36	29	22.33
34	550	693.33	34	410	458.33	34	15	20.33
32	680	663.33	32	485	450.00	32	23	22.67
30	760	701.67	30	455	456.67	30	23	24.00
28	665	660.00	28	430	456.67	28	22	26.00
26	555	636.67	26	485	466.67	26	27	25.00
24	690	627.67	24	485	473.33	24	29	22.67
22	638	651.00	22	450	435.00	22	19	19.00
20	625	636.00	20	370	416.67	20	20	18.67
18	645	663.33	18	430	401.67	18	18	18.67
16	720	675.00	16	405	421.67	16	18	20.33
14	660	699.67	14	430	436.67	14	20	22.67
12	719	672.00	12	475	463.33	12	23	24.33
10	637	686.00	10	485	490.00	10	25	23.33
8	702	649.67	8	510	501.67	8	25	22.67
6	610	645.67	6	510	506.67	6	20	22.33
4	625	612.67	4	500	498.33	4	23	23.33
2	603	632.67	2	485	410.00	2	24	
1	670		1	245		1	23	

Table 2. Cont. Loch Bharadail Core 2 Geochemical data
(Chapter 6. Figures 6.12. 6.13 & 6.14)

Depth	Calcium		Depth		Manganese	Iron		
cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	x 10	3 Period Ave.
135	175		135	1350		1825	18250	
134	153	163.67	134	1350	1337.67	2320	23200	21150.00
132	163	158.00	132	1313	1321.00	2200	22000	22516.67
130	158	151.33	130	1300	1271.00	2235	22350	21050.00
128	133	147.33	128	1200	1225.00	1880	18800	20116.67
126	151	139.00	126	1175	1146.00	1920	19200	19066.67
124	133	116.33	124	1063	1075.33	1920	19200	16633.33
122	65	91.00	122	988	1008.67	1150	11500	17066.67
120	75	83.33	120	975	962.67	2050	20500	15000.00
118	110	102.67	118	925	979.33	1300	13000	17166.67
116	123	114.33	116	1038	962.67	1800	18000	17400.00
114	110	114.00	114	925	967.00	2120	21200	18966.67
112	109	117.67	112	938	979.33	1770	17700	20033.33
110	134	122.00	110	1075	1129.33	2120	21200	20000.00
108	123	116.67	108	1375	1091.67	2110	21100	19266.67
106	93	97.00	106	825	1041.67	1550	15500	19733.33
104	75	71.00	104	925	908.33	2260	22600	17666.67
102	45	56.67	102	975	812.67	1490	14900	16233.33
100	50	60.00	100	538	762.67	1120	11200	14966.67
98	85	63.33	98	775	700.33	1880	18800	16533.33
96	55	60.00	96	788	729.33	1960	19600	16533.33
94	40	62.67	94	625	779.33	1120	11200	17700.00
92	93	66.00	92	925	808.33	2230	22300	17300.00
90	65	74.33	90	875	883.33	1840	18400	20100.00
88	65	68.00	88	850	862.67	1960	19600	18033.33
86	74	64.00	86	863	825.33	1610	16100	16900.00
84	53	65.33	84	763	788.00	1500	15000	14900.00
82	69	63.00	82	738	738.00	1360	13600	14266.67
80	67	68.33	80	713	692.00	1420	14200	14133.33
78	69	66.33	78	625	650.33	1460	14600	14766.67
76	63	64.00	76	613	608.67	1550	15500	13300.00
74	60	64.67	74	588	596.33	980	9800	12833.33
72	71	63.67	72	588	571.33	1320	13200	11983.33
70	60	63.00	70	538	592.00	1295	12950	13403.33
68	58	54.33	68	650	596.00	1406	14060	12303.33
66	45	51.00	66	600	612.67	990	9900	10320.00
64	50	52.67	64	588	575.33	700	7000	11733.33
62	63	68.67	62	538	667.00	1830	18300	14800.00
60	93	81.33	60	875	754.33	1910	19100	18566.67
58	88	89.00	58	850	841.67	1830	18300	17666.67
56	86	90.67	56	800	812.67	1560	15600	16893.33
54	98	98.00	54	788	779.33	1678	16780	16960.00
52	110	107.67	52	750	833.67	1850	18500	17760.00
50	115	110.00	50	963	837.67	1800	18000	18400.00
48	105	111.00	48	800	875.33	1870	18700	18466.67
46	113	110.33	46	863	842.00	1870	18700	19400.00
44	113	106.33	44	863	858.67	2080	20800	20200.00
42	93	95.00	42	850	896.00	2110	21100	20300.00
40	79	70.67	40	975	929.33	1900	19000	20533.33
38	40	63.67	38	963	942.00	2150	21500	21300.00
36	72	61.67	36	888	883.67	2340	23400	20866.67
34	73	83.33	34	800	837.67	1770	17700	19900.00
32	105	98.67	32	825	829.33	1860	18600	18933.33
30	118	111.00	30	863	804.33	2050	20500	19400.00
28	110	107.00	28	725	779.33	1910	19100	18566.67

Table 2 Cont. Loch Bharradail Core 2 Geochemical data, Chapter 6. Figures 6.15, 6.17 & 6.16)

Depth	Calcium		Depth		Manganese	Iron		
cms	mg kg^{-1}	3 Period Ave.	cms	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	x 10	3 Period Ave.
26	93	105.33	26	750	791.67	1610	16100	17933.33
24	113	106.33	24	900	991.67	1860	18600	19166.67
22	113	105.33	22	1325	1158.33	2280	22800	20116.67
20	90	105.33	20	1250	1308.33	1895	18950	19816.67
18	113	109.00	18	1350	1721.00	1770	17700	18416.67
16	124	107.33	16	2563	1721.00	1860	18600	17266.67
14	85	98.00	14	1250	1671.00	1550	15500	17133.33
12	85	81.67	12	1200	1325.00	1730	17300	19583.33
10	75	80.00	10	1525	1233.33	2595	25950	21483.33
8	80	78.33	8	975	1229.33	2120	21200	22983.33
6	80	80.00	6	1188	1133.67	2180	21800	22200.00
4	80	86.00	4	1238	1221.33	2360	23600	23233.33
2	98	100.33	2	1238	1229.67	2430	24300	21743.33
1	123		1	1213		1733	17330	

Table 2 Cont. Loch Bharradail Core 2 Geochemical data. Chapter 6. Figures 6.15. 6.17 & 6.16)

Appendix 3 Loch Bharradail

Depth	Pore water	depth	Magnetic Susceptibility			
cm	%	cm	mean	3Per.Ave/21.56	cm	pH
135	84	132	21325	1806	1	5.15
134	84	130	22689	1689	10	4.87
132	84	128	21220	1615	20	4.6
130	84	125	20284	1531	30	4.17
128	85	121	19231	1337	40	4.5
126	85	119	16795	1371	50	4.53
124	85	116	17226	1207	60	4.57
122	85	114	15157	1379	70	4.52
120	85	112	17321	1397	80	4.56
118	85	108	17551	1522	90	4.52
116	85	106	19115	1607	100	4.4
114	85	102	20179	1604	110	4.59
112	82	100	20143	1545	120	4.72
110	82	98	19407	1582	132	4.7
108	85	96	19871	1417		
106	85	94	17802	1303		
104	85	90	16366	1202		
102	84	88	15097	1326		
100	86	86	16661	1326		
98	86	84	16658	1419		
96	86	82	17822	1387		
94	85	74	17419	1610		
92	85	71	20217	1445		
90	85	68	18147	1354		
88	85	65	17011	1195		
86	85	56	15009	1144		
84	85	52	14373	1133		
82	85	50	14237	1184	**Depth**	**Pore water**
80	85	45	14867	1067	cm	%
78	85	40	13398	1029	38	85
76	85	37	12929	961	36	84
74	86	35	12076	1074	34	86
72	86	32	13493	987	32	86
70	85	30	12391	828	30	87
68	85	28	10405	941	28	87
66	85	26	11815	1185	26	87
64	85	24	14879	1484	24	86
62	85	22	18643	1412	22	85
60	86	20	17741	1351	20	85
58	85	18	16965	1356	18	85
56	85	16	17029	1419	16	86
54	86	14	17826	1470	14	84
52	86	12	18463	1475	12	84
50	86	11	18527	1549	10	85
48	87	8	19458	1613	8	85
46	86	6	20255	1620	6	85
44	85	5	20353	1639	4	84
42	85	4	20583	1700	2	84
40	85	2	21347	3	1	

Table 2 Cont. Loch Bharradail Core 2 Physical properties 6.20, 6.21 & 6.22)

Appendix 3 Loch Bharradail

	$^{210}Pb_{excess}$		1 sigma	^{137}Cs	1 sigma
cm	Bq kg^{-1}	LNAO	Bq kg^{-1}	Bq kg^{-1}	Bq kg^{-1}
1	338.5	5.824524092	28.4	706.5	9.2
2	289.2	5.66711849	27.8	466.5	6.3
3	338.7	5.82511476	31.9	396.9	5.9
4	293.7	5.682558838	24.5	341	5.1
5	278.4	5.629058928	26	370.2	5.8
6	261	5.564520407	26.9	357	5.3
7	215.6	5.373424839	19.8	324.1	4.4
8	144.4	4.972587226	18.9	177.3	2.9
9	138.8	4.933034048	18.8	69.5	2.2
10	121.9	4.803201036	17.8	47.4	2
11	100.9	4.614129927	13.2	35.5	1.6
12	97.8	4.582924577	16.6	28.5	1.5
13	94	4.543294782	17	22.8	1.3
14	70.4	4.254193263	16.9	18.4	1.5
15	90.7	4.507557357	16.5	9.8	1.1
16	81.1	4.395682961	14.8	14.8	1.5
17	91.3	4.514150788	15.9	11.8	1.2
18	100.6	4.611152258	16.4	9.2	1.3
19	78.4	4.361823927	15.6	7.2	1.3
20	62.2	4.130355	19.9	8.2	1
30	49	3.891820298	20.1	12.8	2.6
35	24.6	3.202746443	13.5	6.1	1.4
40	44.1	3.786459782	14.9	4.1	1.7
45	57.3	4.048300624	15.1	1.4	1.1
50	42.6	3.751854253	12.4	0	0

Table 3 $^{210}Pb_{excess}$ and ^{137}Cs activities with 1 sigma error
LNAO is the calculated log normal $^{210}Pb_{excess}$ activity
(Chapter 6. Figure 6.8, 6.9 & 6.10)

Appendix 3 Loch Bharradail

Depth	Pellet Wgt.	$^{210}Pb_{excess}$		Depth	Pellet wgt.	^{137}Cs		
cm	k gms	Bq kg^{-1}	P. wgt.x activit	cm	k gms	Bq kg^{-1}	P. wgt.x activity	
1	0.0052	338.5	1.7602	1	0.0052	706.5	3.6738	
2	0.00565	289.2	1.63398	2	0.00565	466.5	2.635725	
3	0.00559	338.7	1.893333	3	0.00559	396.9	2.218671	
4	0.00684	239.7	1.639548	4	0.00684	341	2.33244	
5	0.0058	278.4	1.61472	5	0.0058	370	2.146	
6	0.00496	261	1.29456	6	0.00496	357	1.77072	
7	0.00884	215.6	1.905904	7	0.00884	324.1	2.865044	
8	0.00773	144.4	1.116212	8	0.00773	177.3	1.370529	
9	0.00721	138.8	1.000748	9	0.00721	69.5	0.501095	
10	0.00738	121.9	0.899622	10	0.00738	47.4	0.349812	
11	0.00813	100.9	0.820317	11	0.00813	35.5	0.288615	
12	0.00809	97.8	0.791202	12	0.00809	28.5	0.230565	
13	0.00696	94	0.65424	13	0.00696	22.8	0.158688	
14	0.00818	70.4	0.575872	14	0.00818	18.4	0.150512	
15	0.00812	90.7	0.736484	15	0.00812	9.8	0.079576	
16	0.00812	81.1	0.658532	16	0.00812	14.8	0.120176	
17	0.00846	91.3	0.772398	17	0.00846	11.8	0.099828	
18	0.00815	100.6	0.81989	18	0.00815	9.2	0.07498	
19	0.00817	78.4	0.640528	19	0.00817	7.2	0.058824	
20	0.00813	62.2	0.505686	20	0.00813	8.2	0.066666	
21	0.00813	62.2	0.505686	21	0.00813	8.2	0.066666	
22	0.00813	62.2	0.505686	22	0.00813	8.2	0.066666	
23	0.00813	62.2	0.505686	23	0.00813	8.2	0.066666	
24	0.00813	62.2	0.505686	24	0.00813	8.2	0.066666	
25	0.00813	62.2	0.505686	25	0.00813	8.2	0.066666	
26	0.00813	62.2	0.505686	26	0.00813	8.2	0.066666	
27	0.00813	62.2	0.505686	27	0.00813	8.2	0.066666	
28	0.00813	62.2	0.505686	28	0.00813	8.2	0.066666	
29	0.00813	62.2	0.505686	29	0.00813	8.2	0.066666	
30	0.00419	49	0.20531	30	0.00419	20.1	0.084219	
31	0.00419	49	0.20531	31	0.00419	20.1	0.084219	
32	0.00419	49	0.20531	32	0.00419	20.1	0.084219	
33	0.00419	49	0.20531	33	0.00419	20.1	0.084219	
34	0.00419	49	0.20531	34	0.00419	20.1	0.084219	
35	0.00803	24.6	0.197538	35	0.00803	6.1	0.048983	
36	0.00803	24.6	0.197538	36	0.00803	6.1	0.048983	
37	0.00803	24.6	0.197538	37	0.00803	6.1	0.048983	
38	0.00803	24.6	0.197538	38	0.00803	6.1	0.048983	
39	0.00803	24.6	0.197538	39	0.00803	6.1	0.048983	
40	0.00649	44.1	0.286209	40	0.00649	4.1	0.026609	
41	0.00649	44.1	0.286209	41	0.00649	4.1	0.026609	
42	0.00649	44.1	0.286209	42	0.00649	4.1	0.026609	
43	0.00649	44.1	0.286209	43	0.00649	4.1	0.026609	
44	0.00649	44.1	0.286209	44	0.00649	4.1	0.026609	
45	0.00698	57.3	0.399954	45	0.00698	1.4	0.009772	
46	0.00698	57.3	0.399954	46	0.00698	1.4	0.009772	
47	0.00698	57.3	0.399954	47	0.00698	1.4	0.009772	
48	0.00698	57.3	0.399954	48	0.00698	1.4	0.009772	
49	0.00698	57.3	0.399954	49	0.00698	1.4	0.009772	
50	0.00532	42.6	0.226632	50	0.00532	0	0	
		Sum	**31.956837**			**Sum**	**22.640175**	
		total inventory	**4507**	Bq m^2 yr^{-1}		**total inventory**	**3193**	Bq m^2 yr^{-1}
		flux	**140**	Bq m^2 yr^{-1}		**flux**	**99.2**	Bq m^2 yr^{-1}

Table 4. Calculated inventories and flux for ^{210}Pb and ^{137}Cs (Table 9.4 and 9.5 pages 296 and 297).

Appendix 4 Loch Lossit

DRL Soil Geochemical data (Loch Lossit catchment)				
coordinate	coordinate	ppm	ppm	ppm
x	y	Pb	Zn	Cu
50	20	75	140	28
50	70	35	35	14
50	130	80	355	64
50	180	480	250	6
50	240	80	35	4
50	300	5	5	2
50	360	45	135	10
50	410	10	20	2
50	530	15	40	2
50	640	25	45	6
50	710	25	50	6
50	760	60	70	6
50	830	30	40	8
50	880	25	110	18
50	940	80	80	12
210	10	15	50	18
210	70	90	190	22
210	130	900	485	36
210	180	130	520	14
210	240	235	240	10
210	300	20	40	2
210	350	60	135	8
210	410	35	55	4
350	10	80	160	36
350	70	220	1900	16
350	120	60	125	6
350	190	95	1000	14
350	240	750	200	18
350	300	19	345	10
350	350	270	20	16
350	410	145	20	6
350	470	20	40	2
350	530	70	20	4
350	590	35	20	2
350	700	20	70	4
350	820	35	65	10
350	930	40	35	2
500	10	75	360	10
500	70	95	340	14
500	120	140	90	10
500	180	35	80	6
500	240	20	240	4
500	300	880	300	30
500	360	600	360	60
500	410	30	410	2
650	10	70	290	22
650	70	65	80	32
650	120	30	15	2
650	180	150	180	16
650	240	200	25	6
650	300	650	325	16
650	360	105	80	18
650	410	20	70	6

Table 1. DRL soil geochemical data surrounding Loch Lossit (Chapter 7. Figures 7.2, 7.3 & 7.4).

		Lead	Zinc	Copper
X coordinate	Y coordinate	ppm	ppm	ppm
650	470	5	15	2
650	530	5	5	2
650	640	25	50	8
650	760	5	5	2
650	870	5	5	2
800	10	40	85	52
800	70	30	65	6
800	120	15	10	4
800	180	35	15	10
800	240	5	30	6
800	300	45	25	26
800	360	1180	360	62
800	420	200	125	18
930	10	5	5	2
930	70	5	5	8
930	130	5	5	2
930	180	30	25	10
930	530	35	30	18
930	580	20	75	6
930	750	5	5	2
930	810	5	10	4
930	870	50	30	6
930	920	5	5	1
1220	10	60	120	16
1220	60	40	80	10
1220	580	5	5	2
1220	650	25	75	18
1220	700	15	5	2
1220	750	10	10	2
1220	820	25	65	6
1220	870	25	5	2
1220	920	15	5	2

Table 1. Cont. DRL soil geochemical data surrounding Loch Lossit (Chapter 7. Figures 7.2, 7.3 & 7.4).

Appendix 4 Loch Lossit

Depth	Pellet Wgt.	$^{210}Pb_{excess}$			Pellet wgt.	^{137}Cs		
cm	k gms	Bq kg^{-1}	P. wgt x activity		k gms	Bq kg^{-1}	P. wgt.x activity	
1	0.0092	230	2.116		0.0092	290.5	2.6726	
2	0.01186	168.8	2.001968		0.01186	192.3	2.280678	
3	0.00832	128.8	1.071616		0.00832	122.1	1.015872	
4	0.01005	133.3	1.339665		0.01005	113.9	1.144695	
5	0.00816	133.4	1.088544		0.00816	114.7	0.935952	
6	0.0115	89.8	1.0327		0.0115	74.9	0.86135	
7	0.00774	110.5	0.85527		0.00774	79.6	0.616104	
8	0.00762	85.4	0.650748		0.00762	61	0.46482	
9	0.00889	89	0.79121		0.00889	61.8	0.549402	
10	0.01109	76	0.84284		0.01109	51.2	0.567808	
11	0.01007	73	0.73511		0.01007	43.7	0.440059	
12	0.00968	53.3	0.515944		0.00968	35.3	0.341704	
13	0.01105	47.5	0.524875		0.01105	31.1	0.343655	
14	0.01088	25	0.272		0.01088	24.7	0.268736	
15	0.01074	15.6	0.167544		0.01074	25.5	0.27387	
16	0.01156	33.7	0.389572		0.01156	25.4	0.293624	
17	0.01106	23.3	0.257698		0.01106	24.3	0.268758	
18	0.01022	30.5	0.31171		0.01022	24	0.24528	
19	0.01159	10.8	0.125172		0.01159	23.4	0.271206	
20	0.01213	30.5	0.369965		0.01213	29.1	0.352983	
21	0.01215	20	0.243		0.01215	28.1	0.341415	
22	0.01196	14.8	0.177008		0.01196	22.8	0.272688	
23	0.01162	11.8	0.137116		0.01162	17.1	0.198702	
24	0.00928	17.4	0.161472		0.00928	19.6	0.181888	
25	0.00884	22.9	0.202436		0.00884	16.6	0.146744	
26	0.00896	1.6	0.014336		0.00896	17.2	0.154112	
27	0.00896	1.6	0.014336		0.00896	17.2	0.154112	
28	0.01141	11.4	0.130074		0.01141	22.9	0.261289	
30	0.00966	12.4	0.119784		0.00966	18.2	0.175812	
31	0.00966	12.4	0.119784		0.00966	18.2	0.175812	
32	0.00608	17.8	0.108224		0.00608	12.3	0.074784	
33	0.00681	17.8	0.121218		0.00681	12.3	0.083763	
34	0.00681	17.8	0.121218		0.00681	12.3	0.083763	
35	0.00681	17.8	0.121218		0.00681	12.3	0.083763	
36	0.00681	4.5	0.030645		0.00681	14.4	0.098064	
37	0.00681	4.5	0.030645		0.00681	14.4	0.098064	
38	0.00681	4.5	0.030645		0.00681	14.4	0.098064	
39	0.00681	4.5	0.030645		0.00681	14.4	0.098064	
40	0.00651	30.7	0.199857		0.00651	19.8	0.128898	
41	0.00651	30.7	0.199857		0.00651	19.8	0.128898	
42	0.00651	30.7	0.199857		0.00651	19.8	0.128898	
43	0.00651	30.7	0.199857		0.00651	19.8	0.128898	
44	0.00651	30.7	0.199857		0.00651	19.8	0.128898	
45	0.00831	18.5	0.153735		0.00831	22.2	0.184482	
46	0.00831	18.5	0.153735		0.00831	22.2	0.184482	
47	0.0067	13.4	0.08978		0.0067	24	0.1608	
48	0.0067	13.4	0.08978		0.0067	24	0.1608	
49	0.00805	19.1	0.153755		0.00805	27.3	0.219765	
50	0.00593	16	0.09488		0.00593	29.2	0.173156	
		Sum	**19.108905**			**Sum**	**18.718034**	
		inventory	**2695**	Bq m^{2} yr^{-1}		**inventory**	**2640**	Bq m^{2} yr^{-1}
		flux	**83.7**	Bq m^{2} yr^{-1}		**flux**	**82**	Bq m^{2} yr^{-1}

Table 2. ^{210}Pb and ^{137}Cs inventory abd flux data (Chapter 9. Tables 9.4 & 9.5, pages 296 & 297)

Appendix 4 Loch Lossit

	$^{210}Pb_{excess}$		1 sigma	^{137}Cs	1 sigma
cm	Bq kg^{-1}	LNAO	Bq kg^{-1}	Bq kg^{-1}	Bq kg^{-1}
1	61.1	4.112512	8.5	78.2	1.7
2	40.5	3.701301974	8.9	55.9	1.2
3	27.7	3.321432413	10.5	43.3	1.1
4	46	3.828641396	10.3	47.6	1.3
5	28.7	3.356897123	9.7	28	0.9
6	31	3.433987204	9.5	23.3	1.1
7	8.9	2.186051277	9.1	16.4	1.4
8	28.8	3.360375387	11	12.4	0.8
9	11.4	2.433613355	11.1	8.5	0.8
10	28	3.33220451	10.9	10.3	0.8
11	31.8	3.45946629	13.6	9.3	0.8
12	18.2	2.901421594	11.2	8.3	1.8
13	14.9	2.701361213	10.5	3.9	0.7
14	27.8	3.325036021	12.1	5.9	0.7
15	13.8	2.624668592	9.2	6.9	1.3
40	15.9	2.766319109	11.7	0	0
50	6.1	1.808288771	9.6	0	0

Table 3. $^{210}Pb_{excess}$ and ^{137}Cs activities with 1 sigma error
LNAO is the calculated log normal $^{210}Pb_{excess}$ activity
(Chapter 7. Figures 7.7, 7.8 & 7.9).

Appendix 4 Loch Lossit

Depth	lead		Zinc		copper		Calcium		Manganese	
cm	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.
122	123		180		23		20		1503	
120	120	124	185	187.00	15	16.67	75	70.00	2398	2059
118	118	116	178	178.67	15	16.67	65	61.00	2275	2306
116	135	112	198	176.00	20	21.67	70	58.33	2245	2103
114	95	98	160	161.67	15	21.00	48	46.67	1790	1993
112	105	102	170	163.33	30	21.00	57	47.00	1945	1787
110	93	110	155	170.00	18	16.00	35	44.33	1625	1838
108	108	121	165	176.00	15	15.00	49	47.33	1945	1844
106	130	129	190	188.67	15	16.67	49	44.33	1963	1909
104	125	129	173	181.33	15	16.67	44	47.33	1818	1990
102	133	131	203	182.00	20	17.67	40	51.00	2190	2109
100	128	130	168	176.00	15	17.00	58	58.67	2319	2256
98	133	129	175	181.67	18	17.00	55	59.33	2258	2277
96	130	126	185	180.00	18	16.00	63	58.33	2255	2216
94	123	123	185	176.00	15	16.00	60	55.67	2135	2123
92	125	116	170	166.00	15	17.00	52	49.00	1980	2088
90	120	108	173	163.67	18	18.00	55	48.33	2150	2388
88	103	94	155	157.00	18	17.00	40	46.67	3033	2454
86	100	88	163	162.00	18	17.00	50	51.00	2180	2425
84	80	77	153	153.67	15	16.00	50	50.67	2063	2178
82	85	74	170	181.00	18	17.00	53	51.67	2290	2174
80	65	65	138	171.00	15	15.33	49	50.00	2168	2304
78	73	63	235	172.67	18	15.33	53	49.67	2455	2295
76	58	58	140	139.33	13	14.33	48	48.67	2263	2339
74	58	59	143	139.33	15	15.00	48	45.33	2298	2300
72	58	55	135	133.33	15	15.00	50	44.00	2340	2276
70	60	56	140	139.33	15	15.00	38	39.00	2190	2186
68	48	55	125	144.33	15	15.00	44	40.00	2028	2092
66	60	59	153	158.67	15	15.00	35	41.33	2058	2045
64	58	69	155	173.67	15	16.67	41	46.67	2050	2070
62	60	78	168	197.00	15	18.33	48	51.33	2103	2130
60	90	84	198	213.67	20	20.00	51	58.00	2238	2180
58	85	81	225	235.33	20	18.33	55	58.67	2198	2196
56	78	74	218	220.33	20	16.67	68	61.33	2153	2286
54	80	70	263	206.00	15	15.00	53	52.67	2508	2168
52	65	65	180	178.33	15	16.67	63	52.33	1843	2244
50	65	68	175	176.67	15	16.67	42	51.33	2380	2261
48	65	70	180	180.00	20	18.33	52	57.33	2560	2503
46	73	76	175	196.67	15	18.33	60	64.00	2568	2591
44	73	77	185	196.00	20	20.00	60	64.00	2645	2691
42	83	76	230	196.00	20	20.00	72	63.33	2860	2716
40	75	72	173	172.67	20	20.00	60	56.67	2643	2687
38	70	67	185	159.33	20	18.33	58	50.00	2558	2550
36	70	66	160	153.67	20	18.33	52	47.33	2448	2443
34	60	69	133	145.33	15	16.67	40	46.67	2323	2320
32	68	75	168	152.00	20	16.67	50	50.00	2188	2129
30	78	77	135	139.33	15	15.00	50	49.33	1875	2086
28	80	75	153	154.33	15	15.00	50	53.67	2195	2078
26	73	69	130	152.67	15	14.33	48	49.67	2163	2249
24	73	69	180	165.33	15	12.67	63	46.33	2388	2127
22	60	57	148	146.33	13	12.00	38	35.33	1830	1994
20	75	57	168	153.00	10	13.67	38	29.00	1763	1655
18	35	50	123	160.33	13	16.33	30	25.67	1373	1532
16	60	60	168	192.67	18	18.67	19	23.33	1460	1448
14	55	52	190	201.67	18	19.33	28	19.67	1510	1521
12	65	58	220	236.67	20	25.00	23	20.00	1593	1484
10	35	63	195	285.00	20	31.00	8	23.33	1348	1594
8	73	79	295	326.67	35	35.33	29	33.67	1840	1645
6	80	96	365	366.67	38	36.33	33	40.00	1748	1827
4	83	96	320	366.00	33	33.67	39	36.67	1893	1943
2	125	96	415	366.00	38	33.60	48	36.00	2188	1884
1	80		363		30		23		1570	

Table 4. Geochemical data from Loch Lossit Core 2 (Chapter 7. Figures 7.10 &7.14)

Apendix 5: Loch Leathann

DRL Soil Geochemical Data Loch Leathann catchment				
coordinate	coordinate	pm	ppm	ppm
x	y	Pb	Zn	Cu
70	40	0	0	0
70	100	50	40	6
70	160	30	30	4
70	220	20	20	4
70	280	20	25	4
70	330	40	55	4
70	390	50	75	4
70	450	10	10	2
70	500	15	15	2
70	560	10	5	2
70	620	5	15	2
70	680	15	15	2
370	30	5	5	2
370	90	30	25	4
370	150	25	35	6
370	200	30	35	4
370	260	55	60	6
370	320	5	5	2
370	490	15	5	2
370	550	5	5	2
370	600	5	5	2
370	660	35	15	4
650	30	10	5	4
650	90	50	35	3
650	150	25	45	22
650	200	25	35	62
650	250	40	60	14
650	310	35	0	8
650	370	15	35	2
650	430	15	10	2
650	490	5	10	2
650	550	15	5	2
650	600	10	15	2
650	660	20	15	2

Table 1. DRL Soil Geochemical Survey data. Loch Leathann catchment (Figures 8.2, 8.3 & 8.4).

Apendix 5: Loch Leathann

Depth	lead		Zinc		Copper		Calcium	Depth	Manganese
cm	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	3 Period Ave.	mg kg^{-1}	cm	mg kg^{-1}
80	18	27.00	42	49.67	8	7.63	120	80	800
79	25	27.00	49	49.67	9	7.67		79	1050
78	38	28.67	58	49.67	6	7.33	145	78	1675
77	23	28.67	42	48.67	7	7.00		77	850
76	25	26.00	46	45.00	8	7.67	105	76	950
75	30	26.00	47	47.00	8	7.67		75	1025
74	23	25.33	48	47.67	7	7.33	140	74	1000
73	23	23.00	48	46.33	7	6.67		73	1050
72	23	25.33	43	46.67	6	7.33	130	72	925
71	30	23.67	49	47.00	9	8.00		71	1075
70	18	22.00	49	47.00	9	8.00	115	70	925
69	18	21.33	43	45.67	6	8.00		69	900
68	28	20.33	45	45.67	9	7.00	135	68	1175
67	15	19.33	49	47.33	6	7.67		67	985
66	15	15.00	48	47.00	8	7.00	130	66	1000
65	15	16.67	44	47.33	7	7.33		65	975
64	20	16.67	50	46.33	7	6.67	135	64	1025
63	15	16.67	45	46.00	6	6.33		63	950
62	15	16.00	43	48.33	6	7.00	85	62	900
61	18	17.00	57	47.00	9	7.00		61	1125
60	18	17.00	41	48.33	6	7.33	90	60	825
59	15	14.33	47	42.67	7	6.00		59	925
58	10	11.67	40	42.33	5	6.33	95	58	800
57	10	12.67	40	46.67	7	7.33		57	825
56	18	12.67	60	50.67	10	8.33	140	56	875
55	10	12.67	52	53.00	8	8.67		55	1000
54	10	10.00	47	53.33	8	8.67	100	54	925
53	10	11.67	61	49.33	10	8.00		53	1150
52	15	11.67	40	56.00	6	9.33	90	52	850
51	10	20.00	67	50.00	12	8.33		51	1250
50	35	25.00	43	50.00	7	10.67	95	50	900
48	30	31.00	40	40.67	13	8.33	90	48	950
46	28	27.00	39	50.00	5	8.33	110	46	725
44	23	23.00	71	52.67	7	6.00	105	44	800
42	18	19.67	48	51.33	6	6.00	105	42	875
40	18	18.67	35	46.33	5	4.67	85	40	700
38	20	18.67	56	44.67	3	4.67	110	38	900
36	18	21.00	43	56.00	6	4.33	80	36	675
34	25	21.00	69	53.67	4	4.33	120	34	925
32	20	21.67	49	59.33	3	3.67	98	32	825
30	20	18.33	60	57.00	4	3.67	120	30	1000
28	15	16.67	62	55.33	4	4.00	120	28	1050
26	15	13.33	44	48.00	4	4.00	65	26	800
24	10	12.67	38	40.67	4	4.00	25	24	625
22	13	12.67	40	42.00	4	4.33	65	22	775
20	15	14.33	48	50.00	5	5.00	80	20	900
18	15	15.00	62	55.67	6	5.33	110	18	1200
16	15	18.33	57	57.67	5	6.33	125	16	1150
14	25	23.33	54	64.67	8	7.00	140	14	1150
12	30	30.00	83	63.67	8	8.00	145	12	1150
10	35	43.33	54	73.67	8	9.00	150	10	1375
8	65	65.00	84	71.33	11	9.67	185	8	1850
6	95	101.67	76	80.67	10	10.67	155	6	1875
4	145	148.33	82	87.00	11	11.67	100	4	1825
2	205	178.33	103	95.00	14	13.33	113	2	2000
1	185		100		15		90	1	1850

Table 2. Loch Leathann Core 2 Sediment geochemical data. (Chapter 8. Figures 8.10-8.15)

Apendix 5: Loch Leathann

Depth	Iron		Depth	LOI	Mineral	Pore water	Mag/Sus		
cm	mg kg^{-1}	3 Period Ave.	cm	%	%	%	cgs	cm	pH
80	1100		80	23.6	76.4	79	10.4	1	5.16
78	1380	1296.67	78	23.1	76.9	79	3	10	5.4
76	1410	1410.00	76	23.9	76.1	79	9.9	20	5.48
74	1440	1390.00	74	24.9	75.1	79	5.5	30	5.31
72	1320	1326.67	72	25.9	74.1	81	3.8	40	5.57
70	1220	1383.33	70	26.3	73.7	81	2.2	50	5.57
68	1610	1426.67	68	25.5	74.5	80	7.2	60	5.6
66	1450	1446.67	66	25.4	74.6	80	7.1	70	5.39
64	1280	1386.67	64	24.7	75.3	80	9.5	80	5.42
62	1430	1303.33	62	24.3	75.7	80	4.1		
60	1200	1263.33	60	24.2	75.8	79	7.1		
58	1160	1410.00	58	23	77	79	10.7		
56	1870	1410.00	56	22.7	77.3	79	6.6		
54	1200	1460.00	54	22.3	77.7	79	12.1		
52	1310	1236.67	52	22.8	77.2	78	11		
50	1200	1336.67	50	21.9	78.1	78	6.7		
48	1500	1246.67	48	22.3	77.7	78	4.9		
46	1040	1243.33	46	23.1	76.9	80	6		
44	1190	1093.33	44	25.9	74.1	80	6.4		
42	1050	1106.67	42	24.2	75.8	80	6.7		
40	1080	1193.33	40	21	79	80	6.3		
38	1450	1236.67	38	23.9	76.1	80	4.4		
36	1180	1426.67	36	22.4	77.6	79	2.9		
34	1650	1323.33	34	24.5	75.5	81	5.5		
32	1140	1423.33	32	30.6	69.4	81	3.2		
30	1480	1410.00	30	25.7	74.3	82	9.5		
28	1610	1476.67	28	25.2	74.8	82	7.1		
26	1340	1306.67	26	20.3	79.7	79	14.7		
24	970	1123.33	24	14.6	85.4	73	12.4		
22	1060	1093.33	22	21.2	78.8	77	5.3		
20	1250	1186.67	20	21.6	78.4	80	15.1		
18	1250	1243.33	18	23.4	76.6	81	13.4		
16	1230	1233.33	16	28.2	71.8	82	10.2		
14	1220	1243.33	14	27.9	72.1	82	22.7		
12	1280	1260.00	12	27.1	72.9	81	15.9		
10	1280	1346.67	10	31	69	84	12.5		
8	1480	1400.00	8	32.2	67.8	85	22		
6	1440	1380.00	6	31.5	68.5	85	14.1		
4	1220	1313.33	4	28.6	71.4	83	10		
2	1280	1226.67	2	25	75	80	18.6		
1	1180		1	23.7	76.3	80	17.2		

Table 2. Cont. Core 2 Geochemical data, physical properties, magnetic susceptibility and pH values. (Chapter 8.Figures 8.14-8.20)

Apendix 5: Loch Leathann

	$^{210}Pb_{excess}$		1 sigma	^{137}Cs	1 sigma
cm	Bq kg^{-1}	LNAO	Bq kg^{-1}	Bq kg^{-1}	Bq kg^{-1}
1	217.8	5.383577	17.9	361.4	4.9
2	234.4	5.457029	24.3	328.1	4.5
3	190.1	5.24755	18.9	295.5	4.6
4	124.1	4.821088	23.5	180.4	3.5
5	89.7	4.496471	22	123.5	2.4
6	54.1	3.990834	19.7	60.8	2.2
7	44	3.78419	15.1	39.6	1.7
8	37.7	3.62966	13.4	37.9	1.5
9	40.3	3.696351	12.7	38.7	1.3
10	38.4	3.648057	15.1	18.6	1.4
11	34.4	3.538057	11.7	18.1	1.1
12	40.8	3.708682	17.8	15.5	1.3
13	20	2.995732	14.2	7.6	1.5
14	16.3	2.791165	16	22	1.4
15	20.9	3.039749	17.2	14.2	1.3
20	7.1	1.960095	10.1	5.9	1.1
25	25.2	3.226844	17.7	5.9	1.1
30	18.3	2.906901	10.6	0	0
40	16.4	2.797281	11.3	0	0
50	0.6	-0.510826	10.2	9	7

Table 3. $^{210}Pb_{excess}$ and ^{137}Cs activities with 1 sigma error. LNAO is the calculated log normal $^{210}Pb_{excess}$ activity. (Chapter 8 Figures 8.7 & 8.9).

Depth	Pellet Wgt.	$^{210}Pb_{excess}$			Pellet wgt.	^{137}Cs	
cm	k gms	Bq kg^{-1}	P. wgt.x activity		k gms	Bq kg^{-1}	P. wgt.x activity
1	0.00864	217.8	1.881792		0.00864	361.4	3.122496
2	0.00806	234.4	1.889264		0.00806	328.1	2.644486
3	0.00745	190.1	1.416245		0.00745	295.5	2.201475
4	0.0059	124.1	0.73219		0.0059	180.4	1.06436
5	0.00732	89.7	0.656604		0.00732	123.5	0.90402
6	0.00797	54.1	0.431177		0.00797	60.8	0.484576
7	0.00797	44	0.35068		0.00797	39.6	0.315612
8	0.00813	37.7	0.306501		0.00813	37.9	0.308127
9	0.00819	40.3	0.330057		0.00819	38.7	0.316953
10	0.00715	38.4	0.27456		0.00715	18.6	0.13299
11	0.00951	34.4	0.327144		0.00951	18.1	0.172131
12	0.00821	40.8	0.334968		0.00821	15.5	0.127255
13	0.00813	20	0.1626		0.00813	7.6	0.061788
14	0.00814	16.3	0.132682		0.00814	22	0.17908
15	0.00811	20.9	0.169499		0.00811	14.2	0.115162
16	0.00811	7.1	0.057581		0.00811	5.9	0.047849
17	0.00811	25.2	0.204372		0.00811	5.9	0.047849
18	0.00811	18.3	0.148413		0.00811	5.9	0.047849
19	0.00811	16.4	0.133004		0.00811	5.9	0.047849
20	0.00853	0.6	0.005118		0.00853	5.9	0.050327
21	0.00853	0.6	0.005118		0.00853	5.9	0.050327
22	0.00853	0.6	0.005118		0.00853	5.9	0.050327
23	0.00853	0.6	0.005118		0.00853	5.9	0.050327
24	0.00853	0.6	0.005118		0.00853	5.9	0.050327
25	0.00855	15.7	0.134235		0.00855	5.9	0.050445
26	0.00855	15.7	0.134235		0.00855	5.9	0.050445
27	0.00855	15.7	0.134235		0.00855	5.9	0.050445
28	0.00855	15.7	0.134235		0.00855	5.9	0.050445
29	0.00855	15.7	0.134235		0.00855	5.9	0.050445
30	0.00837	30.7	0.256959		0.00837	5.9	0.049383
31	0.00837	30.7	0.256959		0.00837	5.9	0.049383
32	0.00837	30.7	0.256959		0.00837	5.9	0.049383
33	0.00837	30.7	0.256959		0.00837	5.9	0.049383
34	0.00837	30.7	0.256959		0.00837	5.9	0.049383
35	0.00837	30.7	0.256959		0.00837	5.9	0.049383
36	0.00837	30.7	0.256959		0.00837	5.9	0.049383
37	0.00837	30.7	0.256959		0.00837	5.9	0.049383
38	0.00837	30.7	0.256959		0.00837	5.9	0.049383
39	0.00837	30.7	0.256959		0.00837	5.9	0.049383
40	0.00837	16.4	0.137268		0.00837	5.9	0.049383
41	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
42	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
43	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
44	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
45	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
46	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
47	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
48	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
49	0.008938	16.4	0.1465832		0.008938	5.9	0.0527342
50	0.00914	0.6	0.005484		0.00914	0.9	0.008226
			14.6676888			13.8718138	
		inventory	2069	Bq m^{2} yr^{-1}	inventory	1956	Bq m^{2} yr^{-1}
		flux	64.2	Bq m^{2} yr^{-1}	flux	60.8	Bq m^{2} yr^{-1}

Table 4. ^{210}Pb and ^{137}Cs inventory and flux data (Chapter 9. Table 9.4 & 9.5, pages 296 & 297).

Loch Bharradail Core 2 XRD data				
Sample (cm)	**No.**	**match**	**XRD Ref.**	**Mineral**
1-2	1	5.9	7-32	Muscovite
1-2	2	4.4	33-1161	Quartz
1-2	3	3.2	19-814	Muscovite
1-2	4	3.1	5-490	Quartz
10	5	6.0	24-594	Lepidolite
10	6	6.0	19-814	Muscovite
10	7	5.9	33-1161	Quartz
10	8	5.4	5-490	Quartz
10	9	4.2	6-263	Muscovite
36	10	7.8	19-814	Muscovite
36	11	6.1	6-263	Muscovite
36	12	5.1	33-1161	Quartz
36	13	3.8	5-490	Quartz
60	14	6.9	19-814	Muscovite
60	15	6.2	6-263	Muscovite
60	16	5.8	7-32	Muscovite
60	17	5.8	33-1161	Quartz
60	18	5.4	5-490	Quartz
64	19	8.8	7-32	Muscovite
64	20	6.4	19-814	Muscovite
64	21	4.7	33-1161	Quartz
64	22	4.2	5-490	Quartz
66	23	5.4	5-490	Quartz
66	24	5.2	33-1161	Quartz
66	25	4.1	19-814	Muscovite
66	26	3.4	7-32	Muscovite
80	27	6.0	6-263	Muscovite
80	28	4.8	5-490	Quartz
80	29	4.4	33-1161	Quartz
80	30	4.2	19-814	Muscovite
80	31	2.8	7-32	Muscovite
135	32	5.7	19-814	Muscovite
135	33	4.1	5-490	Quartz
135	34	4.0	6-263	Muscovite
135	35	3.7	33-1161	Quartz

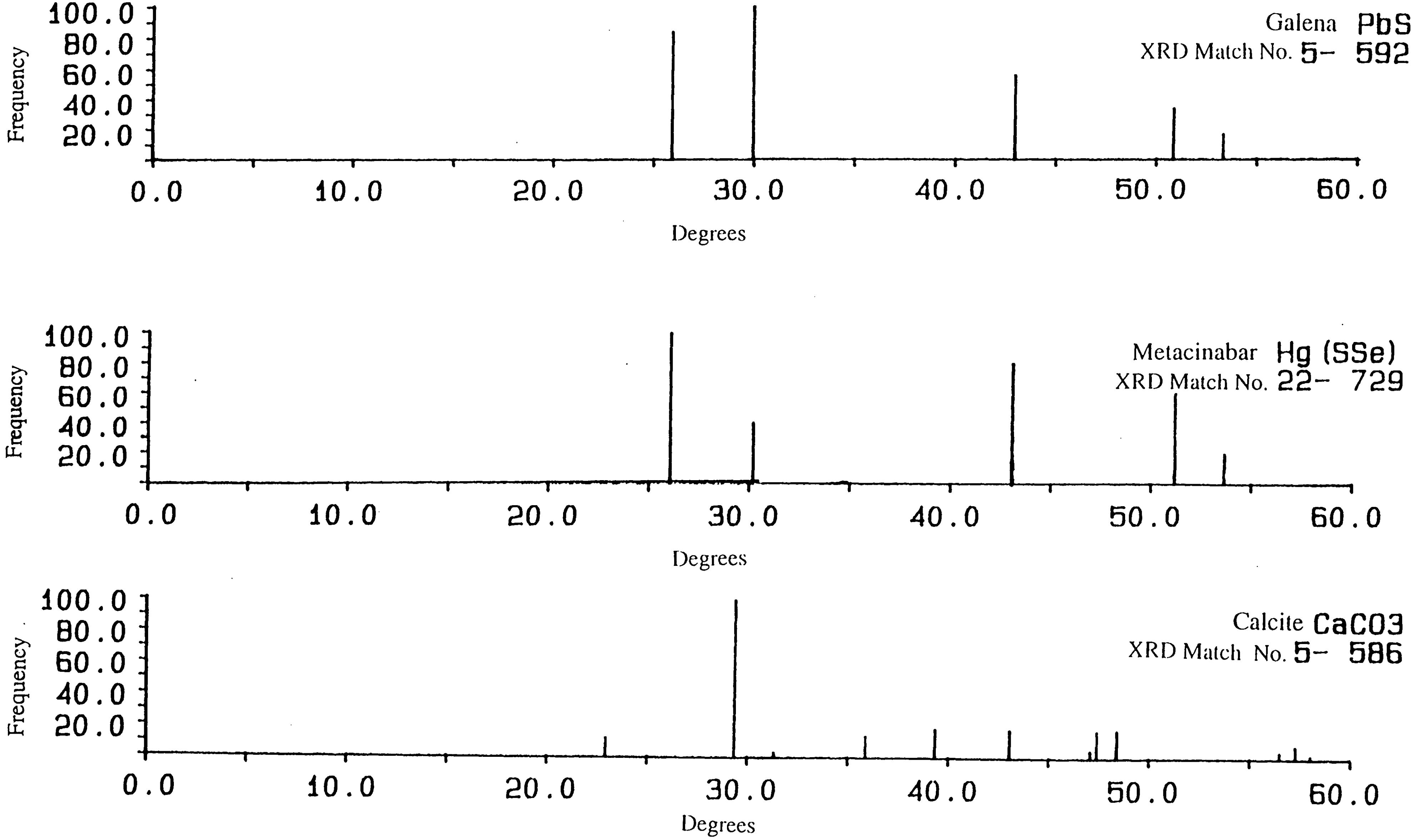
Table 1 X-Ray diffraction match-score data for specific sediment sections in the Loch Bharradail Core 2 (Chapter 6 Figure 6.7).

Loch Leathann XRD Data				
Sample (cm)	**No.**	**match**	**XRD Ref.**	**Mineral**
2	1	6.1	6-263	Muscovite
2	2	4.8	33-1161	Quartz
2	3	4.7	5-490	Quartz
20	4	4.5	5-490	Quartz
20	5	4.3	33-490	Quartz
60	6	7.3	6-263	Muscovite
60	7	4.7	19-814	Muscovite
60	8	4.4	5-490	Quartz
60	9	4.3	33-1161	Quartz
80	10	7.4	6-263	Muscovite
80	11	4.9	33-1161	Quartz
80	12	4.8	5-490	Quartz
80	13	4.2	19-814	Muscovite

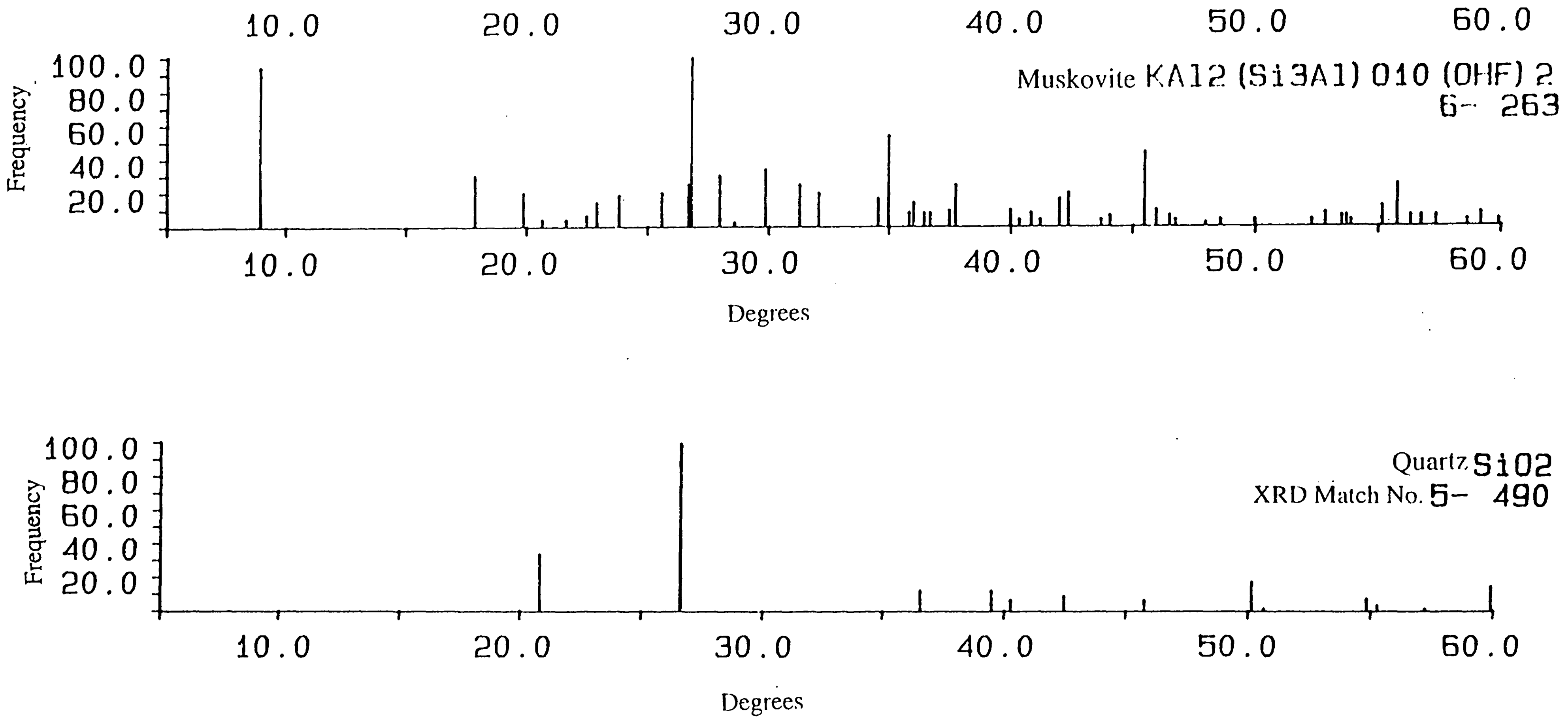
Table 2. X-Ray diffraction match-score data for specific sediment sections in Loch Leathann Core 2. (Chapter 8 Figure 8.6)

South Ardachie Leadmine Grab Sample XRD data				
Fraction	**Number**	**match**	**XRD Ref.**	**Mineral**
1	1	3.7	5-592	Galena
1	2	2.1	22-729	Metacinabar
2	3	3.4	5.586	Calcite
2	4	2.8	5-592	Galena
2	5	2.2	24-27	Calcite
2	6	2.2	4-593	Cerianite
2	7	2.1	22-729	Metacinabar
3	8	2.6	26-801	Pyrite
3	9	2.6	5-592	Galena
3	19	2.6	5-566	Sphalerite

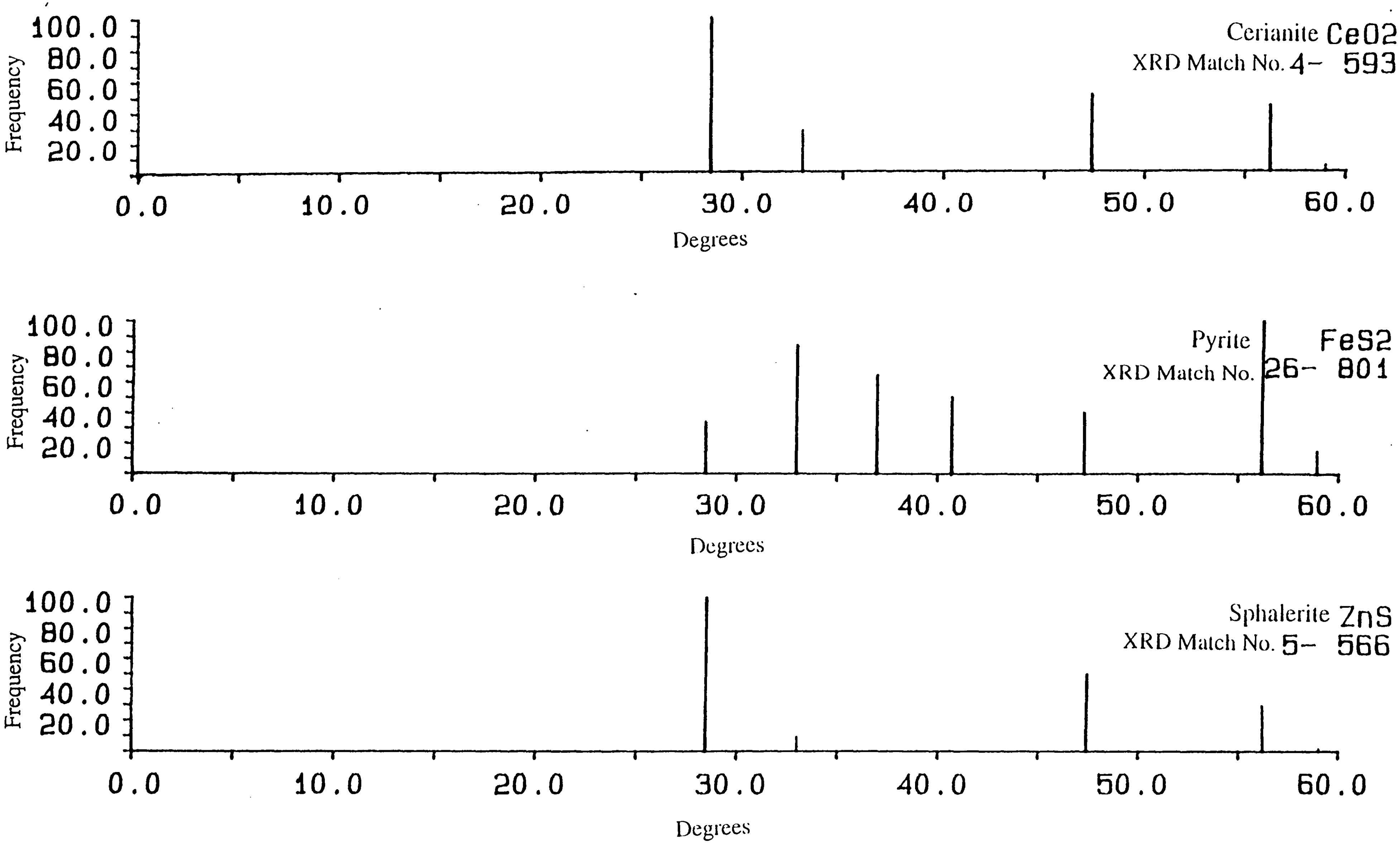
Table 3. X-Ray diffraction match-score data obtained from three mineral fractions obtained from a grab sample taken from a tailings heap at the South Ardachie leadmine in the catchment of Loch Bharradail. Only data with computor match scores above 2.0 are shown. These data represent the principle mineral constituants of the samples.



X-ray diffractogram of frequency versus degrees for Galena, Metacinabar and Calcite in Table 3, Appendix 6.



X-ray diffractogram of frequency versus degrees for Muskovite and Quartz in Table 1, Appendix 6.



X-ray diffractogram of frequency versus degrees for Cerianite, Pyrite and Sphalerite in Table 3 Appendix 6.

List of Abbreviations and glossary

General

BGS	British Geological Survey
CRPB	Clyde River Purification Board
DRL	Domego Resources Limited
MLURI	Macaulay Land Use Research Institute
NMS	National Museums of Scotland
RTZ	Rio Tinto Zinc Limited
SHMP	Southern Hebrides Mesolithic Project
SSGA	South of Scotland Geochemical Atlas
RCAHMS	The Royal Commission on The Ancient and Historical Monuments of Scotland

Scientific

AAS	Atomic Absorbance Spectrophotometry
AEF	Atmospheric Enrichment Factor
ABE	Above Baseline Enrichment
TLP	Total Land Pollen
LPAZ	Local Pollen Assemblage Zones
XRD	X-Ray Diffraction Analyses
LOI	Loss on ignition
Bq^{-1}	1 Bequerel = 1 disintegration per second
cgs	Counts per gram per second
δ	delta
σ	sigma
χ	Magnetic Susceptibility
λ	Lambda is the decay constant
VMS	Volume Magnetic Susceptibility

Botanical

Artemisia (Wormwoods/Mugwort)
Alnus (Alder)
Betula (Birch)
Casteneae sativa (Sweet chestnut)
Cyperaceae (Sedge family)
Caryophyllaceae (Pinks)
Corylus avellana (Hazel)
Taraxacum (Dandelion)
Equisetum (Horse-tail)
Eriocolon (Pipewort)
Ericacea (Heather)
Empetrum (Crowberry)
Filupendula (Meadow sweet)
Filicales (Fern type)
Fraxinus (Ash)
Gramineae (Grass family)
Isoetes lacustris (Quillwort)
Ranunculaceae (Crowfoots/Spearworts)
Sphagnum (Moss)
Selaginella (Fern)
Salix (Willow)
Typhacea (Reed mace or bull-rush)
Lactuceae (Wild lettuce)
Myriophyllum (Water Milfoil)
Pteridium (Bracken)
Picea (Spruce)
Pinus (Pine)
Potamogeton natans (Broad leaved pondweed)
Plantago maritima (*Sea Plantain*)
Phragmites australis (Common reed)
Quercus (Oak)
Juniperus (Juniper)
Rumex acetosa (Sorrel)

Glossary of mining terms

Adit	-a horizontal tunnel or sough driven into the hillside.
Barmaster	-the representative of the Crown, responsible for the administration of -mining -law, measure ore and checking lengths and weights.
Barmoot	-the lead miner's court and appointed jury-(regulatory body)
Bing	-large pieces of ore requiring only limited dressing.
Bole	-a primitive smelting hearth often on a hilltop
Bouse	-ore that has yet to be dressed
Dyke	-intrusive Tertiary rock formation often giving rise to 'contact' mineralisation.
Calcite	-calcium carbonate ($CaCO_3$).
Calcopyrite	-copper ore (CuS)
Contact Zone	-the region of mineralisation between two geological boundaries.
Cross-cut	-a passage cut through solid rock to link to mineral vein
Cupola	-a reverberatory furnace for smelting lead ore
Cupellation	-method of extracting silver from the litharge of molten lead.
Deads	-useless stone from a vein or working
Engine-shaft	-a larger shaft equipped with winding machinery rather than a windlass
Galena	-the mineral lead sulphide (PbS) primary lead ore.
Fire setting	-a mining technique using fires to heat the rock face followed by rapid -quenching and cooling with water to induce large stresses and thus cause -shattering of rock.
Gangue	-minerals which occur with the ore but which have no value in their own right.
Muscovite	-a white mica from the clay illite.
Jig	-a device for agitating a bed of crushed ore in water so that heavier -sulphides and gold sink to the bottom.
Kibble	-a large bucket used to lift ore up the shaft.
Leach	-dissolution of certain components of the ore or rock.
Load	-measure of lead ore, being 9 dishes *c*.3-4 loads to a ton.
Level	-horizontal tunnel, adit, sough or gate.
Leat or Lade	-dyke-like gully to carry water to or from the mine.
Metamorphism	-a change in texture or composition of a rock by heat, moisture and pressure.
Mineral	-a rock substance of regular and definite composition.
Ore	-mineralised rock which could be mined at profit.
Overshot-wheel	-gravity-fed water wheel for ancillary workings such as crushing and sorting.
Outcrop	-surface exposure of rock.
Pig	-block of cast lead with smelter's name or mining company.
Pyrite	-accessory mineral in sulphide rock (FeS_2). Common on spoil tips.
Rake	-a mineral vein, vertically sided and of various length and width.
Scots hearth	-Traditional air-blast ore hearths for primary and secondary smelting.
Stamp mill	-crushing device consisting of heavy vertical rods which mechanically lifted and dropped on the ore.
Sole	-the floor of the mine or lowest level worked.
Stope	-hole from which ore has or is in the process of being extracted.
Sumpter pot	-receptacle for containing molten lead at the base of the ore hearth.
Strike	-a horizontal line drawn on a plane at right angles to the direction of dip.
Syncline	-a trough-shaped fold.
Sphallerite	-zinc ore (ZnS) associated alongside Pb ore.
Tailings	-fine material from which the valuable constituents have been removed.
Washings	-Hydraulically driven stamp or mill for crushing lead ore.

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