A COMPARATIVE STUDY OF SEDIMENT TRACE METAL LEVELS IN UPLAND LAKES IN THE SOUTHERN AND NORTHERN CARPATHIANS OF ROMANIA

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"*Delight thyself also in the LORD: and he shall give thee the desires of thine heart. Commit thy way unto the LORD; trust also in him; and he shall bring it to pass"* (Psalm 37:4-5). I give all the glory and honour to the most high God for supernatural provision and for the successful completion of my Ph.D. Lord Jesus, I am very grateful indeed.

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Dedication

This research is dedicated to God; the power behind my success.

Declaration

I declare that the work presented in this thesis has not previously been submitted for a degree or similar award at Salford University or any other institution. To the best of my knowledge and belief, no material in this thesis has been previously published or written by another person, except where due reference is made.

Signed

Date

List of Abbreviations

ARM: Anhysteretic Remanent Magnetisation

BD: Bulk density

CRM: Certified Reference Materials

GMA: Geochemical analysis

ICP-OES: Inductively Coupled Plasma Optical Emission Spectrometer

LOI: Loss-on-ignition

MM: Mineral magnetic measurements

PS: Particle size

RD: Radiometric dating

SIRM: Saturation Isothermal Remanent Magnetisation

χ: Mass specific magnetic susceptibility

Abstract

The Carpathian Mountains in Romania hold around 150-200 glacial lakes and traverse a region where there are considerable environmental concerns. Despite a long tradition of palaeoecological study in the region, to date relatively little has been published on the alteration of their sediment characteristics due to recent human-induced environmental impacts. This research project has investigated the physical characteristics, the mineral magnetic properties and the trace metal levels of sediment cores from ten selected lakes in the southern and northern Carpathians of Romania in order to evaluate the possibility of using these lakes' sediment as records of recent human impacts and, in particular, trace metal deposition. Laboratory analysis has included sediment bulk density measurements, loss-on-ignition and laser diffraction based particle size determination, environmental magnetism and geochemical (ICP-OES) analysis. A single core from one of the south lakes (Lacul Capra) was radiometrically dated. There were distinct variations in catchment and lake area, the ratio of catchment area to lake size and in lake depth between both regions. The physical characteristics of the lake sediments demonstrated similar trends in their down core profiles in both regions, although the lakes from the south demonstrated a larger particle size range than those in the north. The environmental magnetism of the sediment cores demonstrated common characteristics in surface or near surface peaks magnetic concentration, but there were variations in the magnitude of the concentrations between both regions. The surface increase in concentrations indicated the influence of the atmospheric deposition of particulate deposition associated with fossil fuel combustion and vehicle emissions, but it may also be influenced by microbiological activities within the lakes' sediment. The geochemical analysis (EFs and down-core profiles) showed that the same trends in metal concentration were repeated across the lakes in both regions, but the south lakes displayed higher peak in Pb and Zn concentrations, than were found in the north lakes. The research project has demonstrated the likely influence of atmospheric particulate deposition on the sediments of the lakes from both regions and it has demonstrated spatial and temporal variations in trace metal levels in the lake sediments. It has thereby provided a preliminary database and an overview of palaeolimnological information in two regions of the Romanian Carpathians. Thus, it provides an addition to the records of recent pollution in Romania and a gateway to further investigations in the area of recent palaeoenvironmental change in this region.

CHAPTER 1: Introduction

1.1 General Introduction

This research project investigates the recent sedimentary records of some Romanian mountain lakes as a retrospective monitor of environmental change and human impacts and in particular for the possibility of the presence of trace metal deposition through atmospheric pollution. Past studies on mountain lakes show that they can act as sensitive monitors of environmental change and human impacts such as atmospheric pollution (e.g. Skjelkvale and Wright, 1998; Rose *et al.,* 2009). Mountain lakes can respond quickly to any direct anthropogenic influence in the catchment (Choudhary *et al*., 2009). Trace metals may be deposited on the surface of lake via aerial transport or may be supplied from the catchments via the weathering of rocks and other processes as well as from industrial sources (Smol, 2002; 2008). Regardless of how the metals have been transported to the body of the lake, a certain proportion of them ultimately become incorporated within the sediment column. The association of such metals with the lake sediments makes them useful materials for palaeolimnology. Palaeolimnology is the study of a lake's past environment through which the sequence of past environmental changes can be understood (Birks and Birks, 1980).

Researchers such as Bell and Walker (2005), Smol (2008), and Rose *et al.* (2009) have shown that palaeoenvironmental research can provide an insight into the extent, timing and causes of environmental changes, which have occurred in the past. Apparently, palaeo- studies have in the recent years gained increasing attention, because they allow the placing of contemporary environmental and climatic changes in a longer time perspective (Cohen, 2003). The work of Wohlfarth *et al.* (2001) and Feurdean (2004) show that a great deal of palaeoclimatic and palaeoenvironmental research has been carried out in areas around the North Atlantic region investigating Lateglacial and early Holocene climatic fluctuations (for example, Bond *et al*., 1997; Bjorck *et al*., 1998; von Grafenstein *et al*., 1999).Whereas, there are fewer records of such climatic and environmental change in eastern and south-eastern Europe particularly for Romania (Wohlfarth *et al.,* 2001; Feurdean, 2004). Buczko *et al*. (2009) clarified that despite the abundance of glacial lakes in the South Carpathians of Romania and the usefulness of palaeoecological studies few lake sediment analyses have been carried out in the region. The non-availability of such records makes it difficult to compare palaeoenvironmental development of this region with other regions such as the North Atlantic region (Feurdean, 2004). Although they would be ideal objects of palaeolimnological works with the many proxies applicable to them, glacial lakes in Slovakia and Romania as well as in Ukraine are seriously under-investigated (Buczko *et al.,* 2009).

Romania is known to have suffered elevated levels of atmospheric pollution resulting from past industrial emissions and mining operations (Oszlanyia *et al.*, 2004; Roberts, 2005). It has been observed that like other countries in Eastern Europe and the Soviet Union during the Communist era, Romania's public enterprises tended to pollute the environment without restrictions (Zinnes, 2004). Research has shown that since the late seventeenth century mining and industrial development in the area have created adverse environmental problems such as water and air pollution (Gregus, 1994; Wiersum, 1995; Oszlanyia *et al.*, 2004). Recent lake sediment research by Rose *et al.* (2009) on Lacul Negru located in the Carpathian Mountains of Romania (within the Retezat National Park) indicates that it may have been impacted by trace metal deposition since 16th century. The relative inaccessibility and distance from human activities of remote mountain lakes tend to make them relatively unpolluted systems compared with lowland lakes. In most cases, they are affected only by long-range transported atmospheric pollutants. However, even the low pollutant levels experienced by the most sensitive mountain lakes can cause measurable change in lake water chemistry and biology, to the extent that in some mountain regions these lakes are far from purity (Skjelkvale and Wright, 1998; Grimalt *et al*., 2001).

1.2 Introduction to the study area

Romania is located in the northeastern portion of the Balkan Peninsula. It is situated in the south-eastern part of Central Europe (Romania National Trust Office 1998) and it is the second largest country in the area, after Poland (Hitchins 1994; Roper 2000) (Figure 1.1).

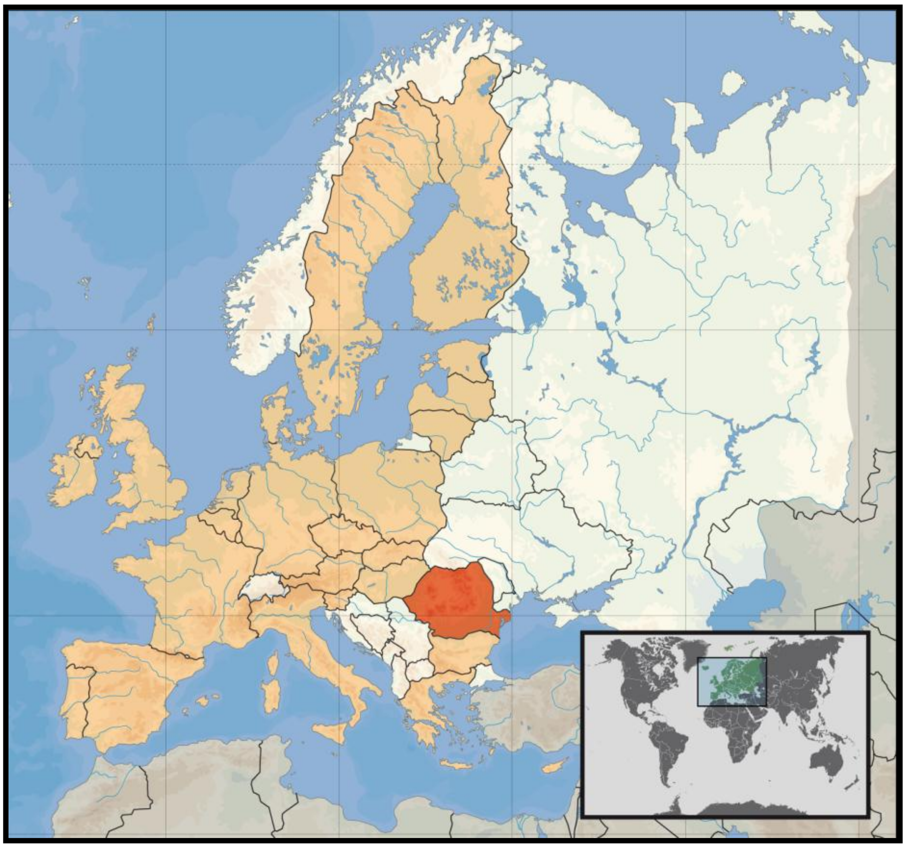


Figure 1.1: General location of Romania in Europe (Romania is highlighted and the other member states of the European Union are also indicated).
(Source: The European Environment Agency (EEA): www.eea.europa.eu)

Romania is transversed by a series of undulating mountains called the Carpathians. The Carpathians are dotted by around 150-200 glacial lakes (see Pisota, 1967; 1968; 1971). As Europe's largest mountain range the Carpathian Mountains have been described as a natural treasure of global significance (Bytnerowicz *et al.*, 2003). They support a wealth of natural diversity which is unparalleled in Europe; and a rich cultural heritage (Oszlanyia *et al.*, 2004) reflecting centuries of human settlement and history. The Carpathians cover an area of about 209 000 square kilometres (Bytnerowicz *et al.*, 2003) that extend over seven European countries; from Romania in the south, through Ukraine, Poland, Slovakia and Hungary to the Czech Republic and Austria in the north (Bodnariuc *et al.*, 2002; see Table 2.1).

According to the Carpathian Ecoregion Initiative (CEI, 2001) archaeological records show that man was harvesting wood from some parts of the Carpathians region from the Mesolithic age. Many migrating and colonising peoples, for example the Romans, Goths, Avars, Slavs and Magyars moved through the Carpathian region between 2nd and 16th

Century (Turnock, 2001). By the 19^{th} Century, the vast majority of the Carpathians belonged to the Austro-Hungarian Empire which collapsed at the end of World War I. After the Second World War, and with the support of the USSR, Communist governments gained power in all the Carpathian countries except Austria (CEI, 2001; Turnock, 2001). Romania became an independent state in 1990 follow the collapse of its Communist government. It is one of the newest member states of the European Union having joined in 2007.

History shows that industrialisation first came to the Carpathian Mountains with the expansion of the Prussian and Habsburg empires in the 18^{th} Century and expanded dramatically in the 19^{th} Century. During this time the Carpathians were exploited for coal and metal mining (for instance in regions of the Banat Carpathians in Romania) and for minerals (e.g. Dashava in Ukraine and Ploiesti in Romania) (Turnock, 2001). In the latter half of the 20^{th} Century, the effects of industry on the natural environment of the Carpathians became very obvious as a result of the discharge of industrial wastes into the environment (Zinnes, 2004). The Communist system's policy of central production was inimical to the natural environment with the attendant air and water pollution becoming significant in parts of the region (Zinnes, 2004). Sulphur emissions from factories, and acid rain originating from factory pollution (both from within and outside the Carpathian region) became a problem (Turnock, 2001). The end of the Communist systems led to a crash in industrial production, resulting in the reduction in emissions of pollutants (CEI, 2001).

Agricultural practices remain a vital part of life in the Carpathians (Roper, 2000). For instance, in some regions of the Romanian highlands farmers have increased the numbers of sheep they graze, as alternative forms of income in their villages have faded away. This has lead to over-grazing of the mountain pastures, reducing the unique biodiversity of these areas, and increased conflict with the carnivores of the forest (Roper, 2000). The consequence of the use of land for such agricultural practices has often led to deforestation. An attendant effect of deforestation can be accelerated erosion which makes flooding more likely after heavy rainfall (Turnock, 2002).

1.3 Rationale for the research

Researchers undertake comparative palaeolimnological study of lakes to cross reference the effects of local to regional scale issues and differentiate them from global issues (e.g. Reavie *et al.*, 1998; Rose *et al.*, 2009). The past 200 years has been described by Cohen (2003) as the period of greatest interest in understanding major human alterrations to the environment. Cohen (2003) also determined that until the 1960s, the number of lakes with regular monitoring programs for basic limnological parameters was very small. Examining the relevant literature suggest that to date very little research has been published about trace metals in Romanian Carpathian lake sediments. However, there is a long tradition in the region of research centred on climate and vegetation reconstruction; these studies are usually pollen based palaeoecological investigations. Nevertheless, Romania has been described as nearly a “white spot” on the palaeoecological map of Europe (Willis, 1994). The research by Bjorkman *et al*., (2002) and Buczko *et al.*, (2009) therefore suggest that additional palaeoenvironmental investigations are urgently needed in Romania. The rationale for this study is therefore that although there are many lakes in the Carpathians, relatively little recent palaeolimnological research has been undertaken. This research will therefore add to the records of recent environmental changes and it will open up Romania to further investigations in the area of palaeoenvironmental change.

A number of studies have focused on the sediments of high mountain lakes (e.g. AL: PE; AL: PE2; MOLAR; EMERGE) (Rose *et al.*, 2009). The Mountain Lake Research (MOLAR, 1999) project considered the most remote and least disturbed freshwater ecosystems in Europe, mainly located in the Alpine and Arctic regions. The project made it clear that although far from local sources of pollution, these lakes are threatened by the atmospheric deposition of pollutants (acidity and toxic air pollutants). MOLAR (1999) attributed the lakes’ sensitivity to these threats to the fact that many mountain lakes are susceptible to acidification because of the poor buffering capacities of the soil and rocks in the watersheds. Subsequently toxic trace metals and trace organics accumulate in the food chain more easily. Furthermore, some pollutants (e.g. mercury, volatile organics) accumulate preferentially in cold regions. The EU funded EMERGE programme included a pan-European assessments of the extent of acidification in mountain lakes located above the tree-line (Curtis *et al.*, 2005). Within the last two decades, this series of EU funded research programs has put forward the measurement of a range of contaminants and the preserved remains of key biological groups in dated sediment cores as their palaeolimnological standard (Rose *et al.,* 2009). High mountain lakes have been identified as sensitive to environmental change with the effects of air pollution and lake acidification having been recorded in many countries (Curtis *et al.,* 2005). EMERGE also shows that the Retezat Mountains of Romania (which is part of the Carpathian Mountain chain) was one of the four lake districts in Europe that exceed the critical loads of acidification (Curtis *et al*., 2005).

A further rationale for this study is therefore to extend the spatial range of the study of mountain lake sediments. Whereas, the Carpathian Mountains have been identified as susceptible to atmospheric pollution, to date there have been very few publications from this region with the only exception being Rose *et al.* (2009) at a site in the Retezat Mountains at the western extremity of the Carpathian Range in Romania. As the present study focuses on the southern and northern portions of the Romanian Carpathian Mountains, it explores the regional differences in the sedimentary properties of lakes in the regions. On the one hand historical mapping shows that a range of industry and extractive operations (e.g. ferrous and nonferrous mining, metal working, chemical processing, petroleum products) tend to be concentrated in the south of the country (e.g. Bucharest, Ploiesti). This differentiation may be reflected in the pattern of the atmospheric deposition of pollutants recorded in each region. This would support the Curtis *et al*., (2005) survey of the Retezat Mountains. On the one hand a wider regional flow and transboundary atmospheric deposition may obscure such inter mountain differences.

Oszlanyi *et al*. (2004) demonstrated that the social and economic transformations that started in early 1990s constitute threats to both the nature and the cultural heritage of the Carpathians. The Carpathians have been described as the back-bone of biodiversity richness and landscape variability in many European countries and that Romania consists of 52% of the Range (Oszlanyi *et* al., 2004). Such an invaluable area could therefore significantly benefit from palaeolimnological studies which could assist policy makers in assessing pollution levels and impacts. In Central Eastern Europe air and water pollution are considered the most serious environmental problems due to their impact on human health as well as the physical environment (Turnock, 2001). Thus, an additional rationale for this study is that an enhanced palaeolimnological insight into the recent lake sediments of the region will potentially inform environmental monitoring and decision making in one of the European Union's newest member states. Consequently, the findings of this research project will have an applied dimension in terms of providing a means of adding to recent records of environmental pollution affecting current (relatively sparse) proxy records both temporally and spatially.

1.4 Aims and Research Objectives

Review of the relevant literature suggests that very little research has been published concerning trace metals in recent lake sediments in the Romanian Carpathians. Indeed most of the available publications are palaeoecological investigations centred on longer term climate change and vegetation reconstruction based on the analysis of pollen and other biological remains. On the one hand part of the Romania Carpathian Mountain chain has been determined as one of the four lake districts in Europe that exceed the critical loads of acidification (Curtis *et al.*, 2005). While on the other hand the Romania Carpathian Mountains have great aesthetic value and are important in terms of biodiversity.

The aims of this research project are therefore: to investigate the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores from selected lakes in the northern and southern Carpathians of Romania in order to evaluate the possibility of using these lakes' sediments as records of recent human impacts (atmospheric delivery) and in particular trace metal deposition; and to determine whether there are differences between the sites in the south and in the north of the range based on these investigations.

In order to achieve the above aims; the research has the following objectives:

1. To determine the physical characteristics of the lake sediments.
2. To assess a mineral magnetic measurement approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants.

3. To determine the geochemistry of the sediments in order to assess the spatial and temporal variations in trace metal deposition.

The following hypotheses will therefore be tested:

Lake sediment records in the study regions can be used as a means of environmental reconstruction and readily provide an assessment of temporal and spatial variation in trace metal pollution.

There will be variations in the lake sediment record of atmospheric inputs between the south and the north of Romania Carpathians reflecting the regional distribution of industry.

1.5 Thesis structure

Chapter one introduces the subject of palaeolimnology. It also gives a brief introduction to the study area. This chapter illustrates the rationale for the study and sets out its aims and objectives. Chapter two reviews the relevant literature in terms of various aspects of the development of palaeolimnology. It examines the history of human influence on lakes and their surroundings, pollution history and implications and the specific roles of mountain lakes in all of the above. Inclusive in this chapter is also a review of literature on sampling techniques and laboratory procedures. Chapter three gives a brief description of the topography, geology, climate, vegetation and ecological importance of the Carpathian Mountains. The latter section relates to catchment and lake basin characteristics.

Chapter four (methodology) describes the field (sampling) and laboratory (analysis) procedures of this research. The field sampling aspect consists of site selection, coring and sample transportation to the laboratory. The laboratory section describes sequentially, the various analytical procedures employed in generating the data for this research. This aspect includes: loss-on-ignition, particle size analysis, environmental magnetism, geochemical analysis and radiometric dating. Particle size analysis, mineral magnetism and geochemical analysis constitute the principal part of the laboratory analysis hence;

more in-depth details are given about these in the review of literature. The statistical approach used to help the interpretation of the data is also included.

Chapter five consists of the main findings and the various data analyses. It includes the descriptions of the physical, mineral magnetic and geochemical characteristics of the lake sediments. Chapter six presents a discussion of the various investigations in this research and addresses the findings related to the specific research objectives. The different sections of the discussion chapter consider the physical characteristics of the lake sediments; the potential of using a mineral magnetic approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants. The subsequent sections look at the geochemistry of the sediments in order to examine the spatial and temporal variations in trace metal deposition. Finally, all data will be integrated to provide an overview. Chapter seven consists of key research findings, the conclusions to this research and further research requirements.

1.6 Summary

The introductory chapter has set out the theoretical framework for this study. It provided a background to the study area. Inclusive in the chapter is the rationale for the study plus the aims and objectives of the study. Romania has been described as nearly a “white spot” on the palaeoecological map of Europe (Willis, 1994); therefore the research hopes to add to the records of recent pollution and to open up Romania to further investigations in the area of recent palaeoenvironmental research. Bearing in mind the above rationale, the aims of this research are therefore: To investigate the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores from selected lakes in the northern and southern Carpathians of Romania in order to evaluate the possibility of using these lakes’ sediment as records of recent human impacts and in particular trace metal deposition. The latter part of this chapter illustrated the structure of the chapters in this thesis.

CHAPTER 2: Literature review

2.1 Introduction

This chapter reviews the relevant literature in terms of various aspects of the principles and development of palaeolimnology. It examines human influences on lakes and their surroundings, the use of lake sediments in the reconstruction of a pollution history, implications and the specific role of mountain lakes in all of the above. Inclusive in this chapter is also a review of literature on sampling techniques and laboratory procedures. The laboratory aspect includes: particle size analysis, loss on ignition, mineral magnetic measurements, geochemical analysis and radiometric dating. Particle size analysis, mineral magnetic measurements and geochemical analysis constitute the principal part of the laboratory analysis hence; more in-depth details are given about these in the review of literature.

2.2 Aspects of palaeolimnology

This section consists of an illustrative review of literature on the principles, development and applications of palaeolimnology. It is not exhaustive, but it contains a good number of references that explain the subject.

2.2.1 Principles of palaeolimnology

Lake sediments are formed from materials that originate from both within the lake system (autochthonous material) and from the in-wash of material from the lake catchments (allochthonous material) (e.g. Simpson, 2001; Smol, 2002; Smol, 2008; Cohen, 2003). Smol (2002) explains the attributes of accumulating sediments that make them useful archives in term of Law of Superposition. That is the progressive overlain of younger lake sediment material (in the upper strata) upon the older (deeper strata). Such lake sediment accumulation results in a depth-time profile which the palaeolimnologist can interpret through various analytical procedures such as density, loss-on-ignition, and particle size analysis, mineral magnetic analysis, geochemical analysis and radiometric dating.

Furthermore, research shows that some lake sediments contain alternate layers of dark and light sediment, which reflect winter and spring sediment inflows as well as the organic matter produced by algae blooms (Alexander and Charles, 1994).

As with peat deposits, lake sediments are ideal media for preserving a range of macroscopic and microscopic fossils, but a considerable amount of palaeoenvironmental information can be derived from the nature of the lake sediments (Smol, 2002). For example, in mid-latitude lake sequences, the climatic improvement at the end of the last cold stage is represented by the transition from minerogenic (inorganic) to organic deposits (Bell and Walker, 2005). This change reflects increased organic productivity within the lake ecosystem and also a reduction in the mineral in-wash as the catchment's slopes became stabilised by vegetation (Lowe and Walker, 1997; Bell and Walker, 2005). Simpson (2001) wrote further that the composition of the sediments is largely influenced by the geomorphology of the lake basins and the drainage basin and lake sediments may also be formed from wind blown atmospheric inputs from within the catchments or from beyond. The atmospheric inputs from beyond the catchment can consist of particulate material that contains some trace metals. The behaviour of such metals within the lake is considered in a later part of this chapter.

Pienitz and Lotter (2009) and Williamson *et al*. (2009) describe lakes as excellent indicators of environmental change because they can provide an insight into the effects and mechanisms of climate change and vegetation disturbance. By exploring the historical record offered in their sediments, lakes can provide natural archives for past environmental change. The study of lake sediments, (biotic and abiotic components) can help assess the general conditions of different physical, chemical, and biological systems (e.g. climate, nutrients, ecosystem functioning, contamination levels) (Pienitz and Lotter, 2009). Palaeolimnological approaches can provide a good reconstruction of climate variability and of the sensitivity of lacustrine ecosystems to such changes (Battarbee, 2005; Smol, 2008; Pienitz and Lotter, 2009). As a result of the capability of the lake sediment to store materials in long sequential order, lake sediment can also yield insights on the hydrological cycle and provide long records where other archives might not be adequate (e.g., trees rings and ice cores) (Pienitz and Lotter, 2009).

Lake basins entrap sediment and can contain a history of deposition spreading over thousands of years and in some lake sequences, such as those that accumulated in deep tectonic basins or volcanic crates, lake sediment records may extend over several glacial-interglacial cycles (Tzedakis *et al*., 1997; Reille *et al*., 2000). Many deep lake basins have been established as natural traps for sediment because the energy level within them are rarely high enough to transport material out once it has settled to the lake bottom (Smol, 2002; Bell and Walker, 2005). Lake margins are also important in the preservation of waterlogged archaeological sequences (Bell and Walker, 2005). Lakes provide opportunities for comparative environmental investigations using pollen, charcoal, chemistry, diatoms and magnetic properties (Bell and Walker, 2005). The increasing identification of sections of annually laminated sediments is also making an important contribution to the development of more precise land use and erosion chronologies (Hicks *et al.,* 1994).

Alexander and Charles (1994) described lake sediments as reservoirs of history because they can record what has occurred in their watershed and atmospheric deposition. Consequently, palaeolimnologist can reconstruct a sediment history by analysing the plant and animal remains preserved in the sediments. Shotbolt *et al.* (2005) state that lakes and reservoirs act as sinks for both catchments and atmospherically derived particulates and so their sediments can provide valuable information on temporal changes in these inputs and that the use of lake sediments as environmental archives is well established. The American Geophysical Union (1995) expressed the fact that important progress has been made toward recovering palaeoclimate information from lake sediments. It has been discovered that fine grained sediment (silt and clay) entrapped in the bottom of lakes and reservoirs preserve the historical nature and levels of many types of pollutants and that sediment cores can also be used to reconstruct lake pH conditions from planktonic material deposited with the sediment (Gem/Water, 2002).

2.2.2 Development and applications of palaeolimnology

Palaeolimnology is the study of a lake's past environment through which the sequence of past environmental changes can be understood (Birks and Birks, 1980). This approach attempts to identify the source of the changes and hence in some cases, open up the previously polluted environment to restoration measures. Environmental restoration of aquatic ecosystems is a global issue that is being addressed by regional and international legislation (Gell *et al.*, 2009). An example of the above is the European Water Framework Directive (WFD), that intends to return all water bodies in the European Union to good ecological status by 2015 (EU, 2000).

Effective management of lake resources requires long-term environmental data. The palaeolimnologist analyses and interprets the diverse information contained in the sedimentary records of lakes (Smol, 2002; 2008). Embedded in these sediments is a record of the organisms that lived in and around the lake, as well as representative data related to processes occurring in the lake, the composition of the lake's water, the conditions in its watershed, and the air above it (Hall and Smol 1996). Gell *et al.* (2009) reiterate that palaeolimnology attempts to identify long-term changes in the condition of lakes and wetlands, and can correlate changing conditions to environmental and climate factors at a range of related levels. Therefore, palaeolimnology can enable the determination of variation in the lake conditions in an historical manner (Haberle *et al.,* 2006; Gell *et al.,* 2009).

Hall and Smol (1996) observed that most environmental studies are undertaken after a problem has already been identified (such as the acidification of a lake or the development of algal blooms or fish kills). Environmental problems are best treated based on the historical knowledge of how and when the problems originally developed. Palaeolimnological studies have made a major contribution to the implementation of legislation, such as the Water Framework Directive (WFD) (Bennion and Battarbee, 2007), especially with respect to the major problems of acidification and eutorphication (Gell *et al*., 2009). In many European lakes (Gell *et al*., 2009) biological remains in sediment records combined with transfer functions have been employed to define ecological and chemical reference conditions, and assess deviation from the reference state (Bennion and Battarbee, 2007). The research of Gell *et al.* (2009) shows that eutrophication in some European lakes with long histories of agriculture in their catchments can date back to the Bronze Age (Bradshaw *et al*., 2005).

Within the field of palaeolimnology research shows that there has been a change of focus from the initial phase of predominantly descriptive studies to developing and improving the numerical foundations for quantitative palaeolimnology (e.g. Birks, 1998; Lotter and Birks, 2003; Bradshaw *et al.,* 2005; Pienitz and Lotter, 2009). Palaeolimnological approaches are now being used to tackle a large suite of environmental and management questions (Smol, 2002; 2008). Although much of the research thus far has been related to mainly lake eutrophication and acidification, palaeolimnological approaches can be used to study environmental issues such as erosion problems, the consequences of water level changes and impoundments, climatic change and pollution (Hall and Smol, 1996). An assessment of anthropogenic impacts on lake water quality involves analysis of recent and preindustrial sediment core samples. The differences between the recent and preindustrial inferences provide information on background or reference conditions (Hall and Smol, 1996; Smol, 2008). Environmental magnetism, discussed in another subsection of this chapter, is an important technique used in obtaining lake sediment palaeo- information. The level of trace metals (such as Lead: Pb) present in a lake can be employed as a measure of the level of anthropogenic activities going on around the lake (Miralles et al., 2006). Actually, Lead (Pb) is a common tracer of anthropogenic contamination widely investigated in numerous studies dealing with environmental quality and health care in Western Europe (Mielke *et al.,* 2005; Miralles et al., 2006). Anthropogenic lead emissions originate from power generation, ore smelting, automotive emissions, and natural sources such as volcanoes, hydrothermal vents has been investigated in various lake sediments (e.g. Renberg *et al.*, 2002; Winderlund *et al.,* 2002).

2.2.3 Palaeolimnology studies of human impact

Palaeolimnological research has assisted in documenting the history of human influence on lakes and their surroundings. Human land usage can result in catchment instability which can be reflected in changing sediment yields, often as a result of changes to the sediment delivery process (Foster *et al.,* 1988). High sediment yields may arise from a variety of human induced factors especially from deforestation practices and from a variety of processes relating to the planting and harvesting strategy adopted in agricultural land. High sediment yields can also emanate from some natural causes such as gullying and shallow land sliding (Foster *et al.*, 1988).

These historical deductions can help policy makers establish lake and ecosystem management objectives (Cohen, 2003). Human impacts on land/watersheds can include deforestation, urbanisation and intense grazing (for example). These activities can produce a clear signal of human disturbance in the environment which can be inferred from palaeolimnological archives (Cohen, 2003). Various studies have supported the above assertion. The research of Sandman *et al.* (1990) in Finland showed that ditching of peat lands meant to drain large areas for forestry and agriculture strongly accelerated the soil erosion recorded in the sedimentation rates. The Van der Post *et al.* (1997) study in the English Lake District showed that intense grazing greatly increased the rate of sedimentation in lake. Similar observations were made in semi-arid tropics by El-Daoushy and Eriksson (1998) who demonstrated that intense grazing greatly influenced accumulation rates in lakes. Intensive agriculture has led to increased accumulation rates in the northern part of Lake Tangayika (Cohen, 2003).

The growth in human population density, mechanised farming involving the use of fertilisers and the discharge of urban sewage into the lakes have resulted in increased eutrophication across the globe (Cohen, 2003). Palaeolimnology can help in defining the timing and size of human induced changes in lake trophic conditions (Wolfe *et al.*, 2001) and in determining realistic objectives for lake reconstruction (Bennion *et al.*, 1996).

2.2.4 Pollution: sediment based reconstruction and implications

Cohen (2003) described lakes as excellent sites for obtaining historical records of the atmospheric and watershed inputs of metal or organic pollutants. Lake sediments have accumulated a pattern of increasing metal and organic combustion by-product deposition in numerous lakes throughout the world as a result of the rapid expansion of heavy industry, internal combustion engines and power plants since the late nineteenth century (Cohen, 2003). Increases in the concentrations of trace metals, magnetic minerals and combustion derived particulates have been discovered in lake sediments throughout the world (e.g. Oldfield and Richardson, 1990; Wik and Renberg, 1996; Sandgren and Snowball, 2001; Hutchinson, 2005; Yang and Rose, 2005; Jumbe and Nandini, 2009a; Rose *et al*., 2009).

Trace metal-polluted sediments have become a global concern due to their increasing prevalence in ecosystems (Grandlic *et al*., 2006). Researchers (e.g. Audry *et al*., 2004) have shown that anthropogenic activities have caused important transformations in aquatic environments during the last 150 years and that trace metals are among the most widespread of the various pollutants originating from anthropogenic activities, particularly from mining and smelting waste sites (e.g. Salomons, 1995; Dar, 1997; Hochella *et al.,* 1999; von Braun *et al.,* 2002). There are coordinated efforts across the globe to keep the environment safe and fit (Gell *et al.*, 2009). For example, the 2000/60/EC Directive (WFD) in the field of the water policy focuses on maintaining and the improving of aquatic environment in the European Community by establishing a framework for the protection of surface water (Pertsemli and Voutsa, 2007). Trace metals have been identified as a group of pollutants that should be monitored in order to obtain a logical and up to date overview of quality status for surface water (Pertsemli and Voutsa, 2007). Sediments have been discovered to play an active role as a sink and possible source of trace metals (Salomons, 1995; Hochella *et al.,* 1999; Audry *et al.,* 2004).

Blake *et al.* (2007) shows that trace metals released from waste material have a tendency to associate with particulate matter (van den Berg *et al.* 2001; Audry *et al.* 2004). Therefore, this makes it a common occurrence to observe elevated concentrations of trace metals in accumulating lake sediments affected by urbanisation and industry (Foster and Charlesworth, 1996). Research shows that an undisturbed accumulated lake sediment can become an environmental concern (e.g. Becker *et al.,* 2001; Martin and Pedersen, 2002; Blake *et al.,* 2007), in the extreme, this can turn to a pollution store (Farmer, 1991).

The concentrations of trace metals in different ecological environments, particularly in water and sediments of lakes, are considered to be the major factor in environmental pollution (Vital and Statteger, 2000). Variable amounts of different pollutants are constantly being discharged into the environment, but heavy metals are regarded as serious pollutants of the aquatic environment (e.g Altindag and Yigit, 2005). This is attributable to their environmental persistence and their tendency to accumulate in aquatic organisms (Harte *et al*., 1991; Schurmann and Markert, 1998). Trace metals released into the environment can find their way into the aquatic phase as a result of direct input, atmospheric deposition, precipitation and erosion (Veena *et al.*, 1997). This might expose aquatic biota to excessive levels of such metals (Kalay and Canli, 2000). The high accumulation of trace metals in the aquatic organisms can result in serious ecological changes when they become biologically magnified through the food chain (Unlu and Gumgum, 1993; Altindag and Yigit, 2005). Human beings might consume metals through food webs. This might result in serious human health concern (Chen *et al.*, 2000). This implies that the high concentrations of trace metals for example Pb in a lake may affect or alter the ecosystem structure and food webs of the lake and may affect population density by inhibiting reproduction.

Reproduction is an important process for maintaining life continuity. Metals, as well as other pollutants, have been reported by Alquezar *et al.* (2006) to have the ability to reduce fish oocyte quantity and quality and may result in malformations and impaired development (Brooks *et al.*, 1997). Consumption of contaminated fish flesh has been reported to have effects on human health (Rowat, 1999; Suner *et al.*, 1999; Chan *et al.*, 2003). Pollutants can be transferred from parents to eggs, leading to reduced embryonic development (Miller, 1993). Eggs can get contaminated by direct uptake from the environment, and all these can have physiological effects on newly born hatchlings (Von Westernhagen, 1988; Alquezar *et al.*, 2006). Pollution can be counterproductive leading to ecological problems as well as wasting of resources (Turnock, 2001).

2.2.5 The behaviour of metals within lake sediment

Smol (2002) shows that once the trace metals get into a lake, they tend to be taken up by particles and deposited in sedimentary profiles. Consequentially, sediments often have trace metal concentrations several times higher than those in the water body above the sediments (Rognerud and Fjeld, 2001) provided the lake is not disturbed. Metals are regarded as a group of pollutants of high ecological relevance that are not removed from water by a natural process of purification. It has been discovered that within the lake system they are capable of redistribution among different components (Anu and Swarna 2009). Various processes occurring within the sediment are believed to influence the distribution or movement of the metals within sediment (e.g. Lockhart *et al*. 2000, Shotbolt *et al.* 2001). Such processes are considered as physical, chemical and biological activities, which include: pH, oxidative–reductive potential, dissolved oxygen, organic and inorganic carbon content, and the presence in water phase of some anions and cations (e.g. Boyle, 2002; Nguyena *et al*., 2005; Cevik *et al*., 2009; Anu and Swarna 2009). Generally, these diagenetic processes include desorption of metals (the accumulation of metals on the surface of the sediment particles or the lake water) and release to the porewater, chemical mobilisation of metals (changing from an adsorbed state on a surface to a gaseous or liquid state) and bioturbation (e.g. Oldfield and Wu, 2000; Boyle, 2001). These factors have been assessed as capable of affecting the credibility of sedimentary records as archive of trace metal deposits (e.g. Shotbolt *et al*. 2001; Boyle, 2002; Cohen, 2003).

Bioturbation may be defined as the stirring or mixing of sediment or soil by organisms, especially by burrowing, boring, or ingestion (Mifflin, 2010). This may include eating and excreting sediment (e.g. worms). Bioturbation aids the penetration of air and water and loosens sediment to promote movement of material across the water and sediment interface. It includes the physical processes such as traction, suspension and the chemical process of solution. The physical movement of particulate-associated metals may alter the lake sedimentary record (Bourdreau, 1999).

2.2.6 The mountain lake sediments as archive of atmospherically transported pollutants

Recent research funded by the European Union indicates that even the most remote lakes in Europe contain atmospherically transported pollutants and evidence is growing that climate change is beginning to have a significant impact (Battarbee, 2005). The combustion of coal and oil gives rise to the emission into the atmosphere of sulphur, nitrogen gases and fly ash particles (Battarbee, 2005). Evidence for the contamination of mountain lakes by these products can be demonstrated from the high concentration of non-marine sulphate and nitrate in the water column (Battarbee *et al.*, 2001) and the presence of fly ash particles, especially spheroidal carbonaceous particles (SCP) in the sediment (Rose *et al.*, 2002; Battarbee, 2005). The spatial variation in concentration of these substances is in good agreement with the distribution of industrialised regions within Europe and the temporal variation in the concentration of SCPs measured in sediment cores reflects the progressive industrialisation of Europe from the nineteenth century to the present day (Battarbee, 2005). It has been determined (Battarbee, 2005) that fish from the most remote sites in Europe, especially the high alpine zone carry substantial burdens of toxic metals in their tissues (Rognerud *et al.*, 2002).

Mountain lakes have been reported (e.g. Choudhary *et al.*, 2009) as responding quickly to any direct anthropogenic influence, or human-induced changes in the catchment. Graney and Eriksen (2004) applied a palaeolimnological approach in response to health concerns about current and past exposure to pollutants within Broome County, New York by determining historical records of anthropogenic activities as preserved in sediment cores. Choudhary *et al*. (2009) reported that topography influenced atmospheric transport and deposition of pollutants. Researchers (e.g. Battarbee *et al.,* 2002; Vreca and Muri, 2006; Choudhary *et al.,* 2009) show that these effects and changes are potentially recorded in the sediments, which can be used as means of reconstructing past conditions over different timescales.

It has been determined that remote mountain lakes can be indicators of atmospheric pollutants and its effects (Skjelkvale and Wright, 1998). MOLAR (1999) attributed the lakes' sensitivity to these threats to the fact that many mountain lakes are susceptible to acidification because of the poor buffering capacities of the soil and rocks in the watersheds. In addition to these, some pollutants (e.g. mercury, volatile organics) have been identified to accumulate preferentially in cold regions (MOLAR, 1999). The lakes' sensitive geologies, sparse soils and harsh climates can combine to produce sensitive ecosystems which can be stressed by atmospheric pollutants inputs resulting in detectable environmental change (Rose *et al.*, 2009). The progressive accumulation of the lake sediments can store a record of conditions within the lake systems and of contaminants deposited from the atmosphere. Such accumulation of materials can make them useful archive of atmospherically deposited pollutants and hence they can provide one of the few ways by which temporal trends in deposition can be identified in an historical context for contemporary measurements (Rose *et al*., 2009).

2.3 Review of sampling and analytical techniques

This section clarifies the sampling technique adopted for this research; it discusses the relevant sampling equipment and the underlining principles used in the research methodology for this study. The most common method of obtaining palaeolimnological data is by sediment coring. Cores are unit-dimensional columns of sediment of which the collection of several cores allow the study of spatial variability of sedimentary archives in a lake (Cohen, 2003). Core samplers can penetrate the sediment very deeply and consequently, they provide a cross-sectional slice of sediment layers and thus, information about the sediment deposition (Aaby and Digerfeldt, 1986; British Columbia, 1998). Palaeolimnological investigation in lake systems involves taking a sediment core sample from an appropriate area of a lake basin and slicing the sample into intervals, usually less than 1cm and often of 0.5cm or less so that higher temporal resolution in the analysis can be achieved (e.g. Simpson, 2001; Cohen, 2003; Hutchinson, 2005).

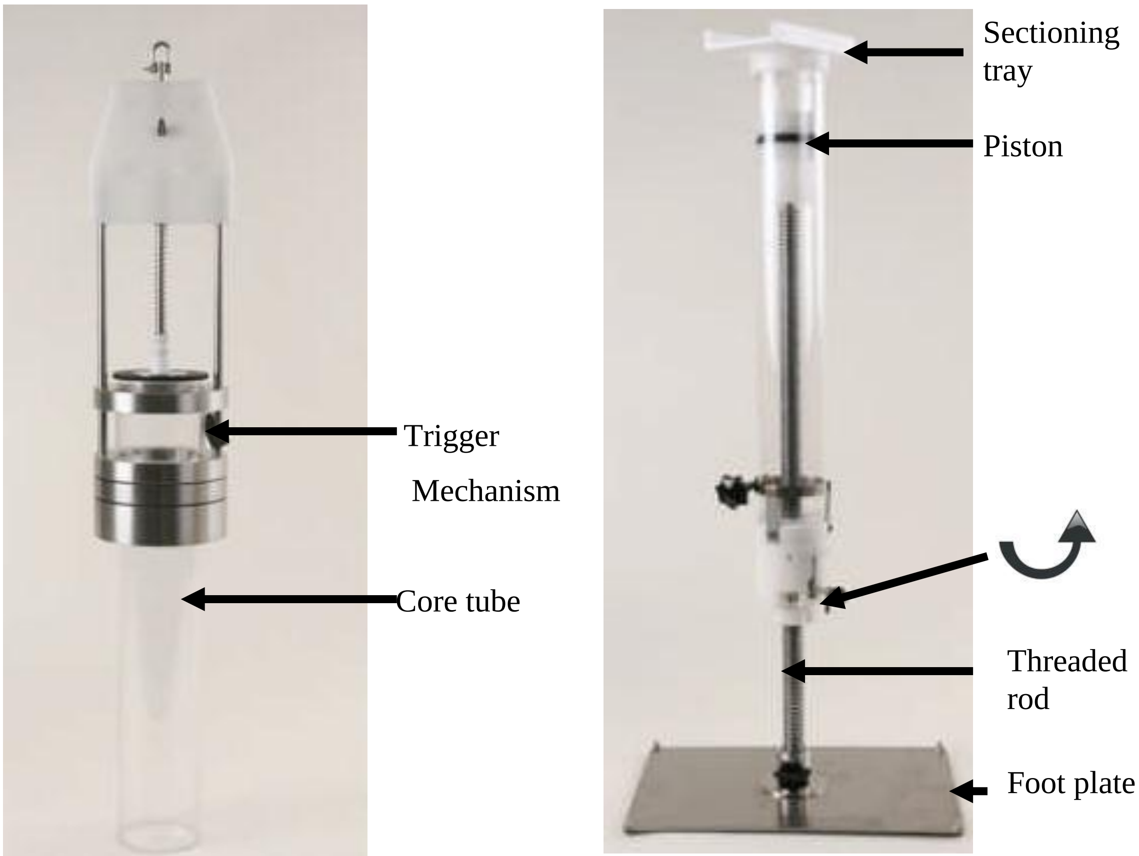
2.3.1 Common types of sampling equipment

A wide great variety of sampling strategies and techniques are used in palaeolimnology (Birks and Birks, 1980). A great deal of work has been undertaken on lake sediments sampling equipment. This section describes some commonly used samplers and the suitability of each for sampling in different conditions. British Columbia (2003) states that grab samplers can be used for collecting surface sediments, in order to determine the horizontal distribution of variables (across the bed of a lake). Core samplers can be used for collecting a depth profile of sediments, thereby providing material for determination of vertical distribution of variables. Hence, grab samplers are useful for assessing recent inputs of pollutants while core samplers (e.g. gravity corers) are better suited for assessing long-term (historical) inputs (British Columbia, 2003). Such sampling equipment is relatively easy to use and can collect a large quantity of sample. The type of sampler used at particular sites will vary depending on the purpose of the study and will be dictated by the project design (British Columbia, 2003; Birks and Birks, 1980). Gravity corers usually take 30–40 cm long cores of unconsolidated, surface sediment. Generally a valve at the top of the sampler closes by messenger, creating a vacuum seal that prevents the sediments from washing out (Aaby and Digerfeldt, 1986; British Columbia, 2003). The core tube is removed from corer for extrusion of sediment.

The research of Verschuren (1993) shows that piston corer features both the gentle, slow sediment penetration that is essential to prevent loss of surface sediments (Blomqvist,

1991) and minimum risk of incomplete recovery. Piston corers can be operated with push rods from lake-ice or a stable raft on shallow lakes (Verschuren, 1993). Mackereth corers can obtain long (6 m) continuous cores of soft sediment in water depths up to 100 m using lightweight equipment (Mackereth, 1958; Barton and Burden, 1979). An attractive feature of Mackereth corers is that the core tubes (and barrel) can be made of PVC water pipes, which are cheap, can, serve as permanent core retainers, and are easily sliced open. This makes the cores particularly suitable for palaeomagnetic work on lake sediments (Barton and Burden, 1979). The full description and functioning of the corer can be found in Mackereth (1958).

Gravity corers have been in use for a number of years (Renberg and Hansson, 2008). Coring is an important step in any palaeolimnological investigation because any failure during coring or sub-sampling cannot be amended by any subsequent analyses (Renberg and Hansson, 2008). In order to avoid contamination in pollution studies, the equipment is made of stainless steel and plastic (polyoxymethylene and polyethylene) which makes the equipment more durable (Renberg and Hansson, 2008). The HTH gravity corer consists of two main portions; the corer head (Figure 2.1a) and extrusion device (Figure 2.1b). The corer head consists of an open tube that is lowered into the sediment with a wire or rope, after which the tube is sealed by a closing mechanism before the corer is pulled up. The corer takes 30-40 cm long cores of unconsolidated, recent sediment. The core tube is removed from corer for extrusion of sediment. The tube is kept vertical in order not to disturb the sediment water interface. Once the core was disengaged from the corer, an extrusion was carried out to dislodge the water layer on top of the sediment. The extruding device (Figure 2.1b) consists of a piston that seals perfectly to the core tube wall, a threaded rod that can be mounted on a foot plate, and an extruder head with a stationary upper-half and a rotatable lower-half that is screwed along the rod.



(a) Corer head (b) Extrusion device

Direction of knob to raise sediment through core tube

Scale: The tube is about 50 cm long and 5 cm in diameter

Figure 2.1: HTH Gravity corer (Source: www.pylonex.com)

A gravity corer has been used for the sampling for this research. Collection of samples in the field follows standard sedimentological procedures while taking samples in the form of lake sediment cores. Standard air sealable plastic sample bags are suitable for collection and short term field storage of material intended for palaeolimnological analysis (Walden *et al*., 1999). During collection of samples, care must be taken to prevent contamination of the sample. Plastic sampling equipment must be used and the sampler must be conscious of contamination during sampling process (Dearing, 1999).

2.4 Laboratory analysis

Sediment samples from the studied lakes have been investigated using a wide range of analytical techniques; the results of which can be used to make inferences or deductions about the environmental conditions in the lake such as atmospheric inputs and sediment chronology (Simpson, 2001; Smol, 2002; Cohen, 2003; Bell and Walker, 2005; Hutchinson, 2005; Rose, *et al*., 2009). In palaeolimnology researchers carry out both physical and chemical analysis of lake sediments to exploit the wide ranges of environmental information potentially held within the sediments (Bengtsson and Enell, 1986). Sediment analysis is important in environmental control and can be more informative than other methods in reaching a comparative conclusion about environmental pollution and how eutrophication and pollution situation has developed in the ecosystem (Bengtsson and Enell, 1986).

2.4.1 Sediment organic content: Loss-on-ignition

The determination of loss-on-ignition is fundamental measurement that is highly appropriate to the characteristic of lake sediments (Matthiessen *et al*., 2005). It facilitates the measurement of organic and carbonates content. Different methods have been developed for the determination of organic matter content of sediments. Santisteban *et al*. (2004; Shotbolt *et al*. 2001) shows absolute carbon content can be determined either by dry combustion or wet oxidation followed by estimation of the evolved carbon dioxide or by the instrumental analysis of total organic carbon (Allen, 1989). These methods are time consuming and seem impracticable for a large number of samples (Shotbolt *et al*. 2001). Consequently, an easier and more rapid technique for determination of organic matter content is by measurement of mass loss on ignition. Loss-on-ignition can provide the most accurate estimate of organic matter content of sediments (Allen, 1989; Santisteban *et al*. (2004)).

Loss-on-ignition can be expressed per unit volume of fresh sediment and per dry weight of sediment or as sedimentation per m^2 and time (Bengtsson and Enell (1986). Most sediment is composed of a mixture of clastic silicates and oxides (sand, silt and clay fractions), organic material, carbonates and water. Quantitative determinations of such sediment parameters by means of loss-on-ignition are based on the sequential heating of the samples (Veres, 2002). Further details are given in chapter 3 (Methodology).

2.4.2 Particle size analysis of lake sediments

Sediment particle size analysis gives very diverse information about the sedimentation environment (Vaasma, 2008). Depending on the aims of research, different methods are used to describe particle size. For example, electron microscopic techniques enable the three-dimensional structure of particles or aggregates to be fixed (Konert and Vandenberghe, 1997; Kim *et al*., 2005; Vaasma, 2008). Light microscopic techniques allow the estimation of sediment composition and the selection of appropriate pre-treatment methods. Other research (Mikkelsen and Pejrup, 2001; Thonon *et al*., 2005) has shown that due to the fragile nature of aggregates, sometimes their properties have to be determined in situ.

Recent research (Vaasma, 2008) describe how despite being time consuming, the particle-size distribution of soil sediment record is often measured by a sedimentation method based on the Stokes' Law (Hiroaki *et al*., 1999). Recently, the application of a laser scattering method based on Mie scattering theory to determine the particle-size distribution has been developed (Horiba, 2009). The laser scattering method is more time and sample efficient than the sedimentation method, sieving, and manual microscopy processes as it uses shorter time and smaller amount of sample material for measurement (Hiroaki *et al*., 1999). The following subsection describes briefly the basic principles of a laser scattering method.

2.4.2.1 Basic principles of particle size analysis system

The particle size analyzer consists of at least one source of high intensity, monochromatic light, a sample handling system to control the interaction of particles and incident light, and an array of high quality photodiodes (Figure 2.3) to detect the scattered light over a wide range of angles (Hiroaki *et al*., 1999; Horiba, 2009). The instrument records angle and intensity of scattered light (Figure 2.2). The Partica LA-950V2 laser diffraction (Figure 2.2) is able to measure particle size from 10 nm to 3,000 µm.

Figure 2.2: The Partica LA-950V2 laser diffraction instrumentation (Source: Horiba, 2009)

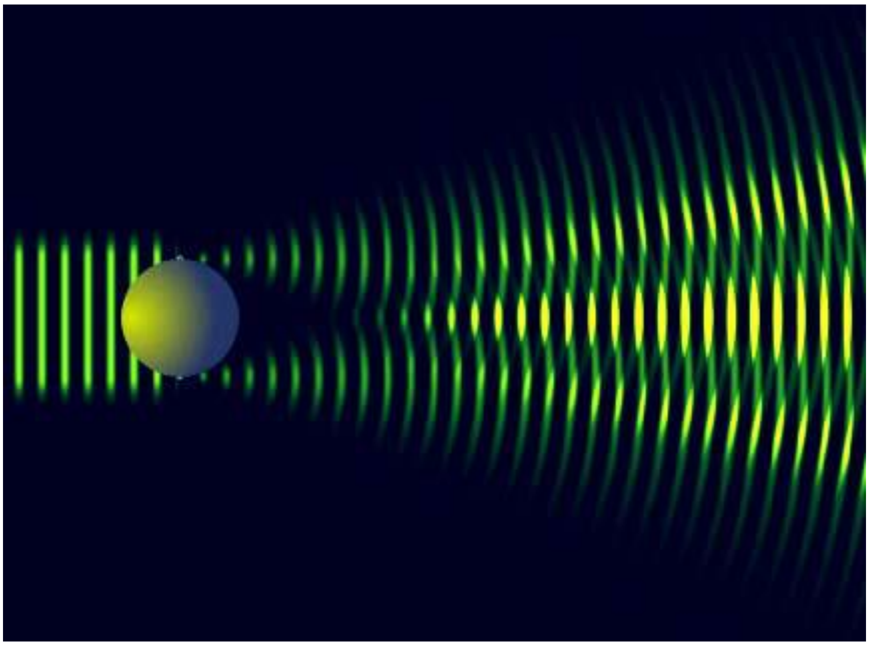


Figure 2.3: The diffraction pattern (right side) of a plane wave scattering off a spheroid (left side) (Source: Horiba, 2009).

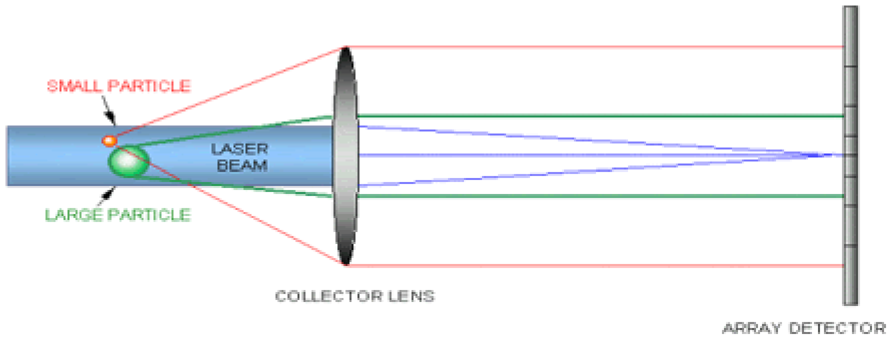


Figure 2.4: The diffraction pattern line spectra (Source: Horiba, 2009).

For particles larger than the wavelength of light, the light scatters from the edge of the particle at an angle which is dependent on the size of the particle (Figure 2.4). Larger particles scatter light at relatively smaller angles than light scattered from smaller particles (Figure 2.4). By observing the intensity of light scattered at different angles, the relative amounts of different size particles can be determined (Horiba, 2009). As the particles get close to or smaller than the wavelength of light, more of the light intensity is scattered to higher angles and back-scattered. The Mie Scattering Theory accounts for this different behaviour.

Horiba (2009) incorporates the full Mie Scattering Theory over the entire size range of interest. An array of detectors including high-angle and back-scatter detectors, and multiple light sources of different wavelengths are employed to provide an instrument that allows measurement of the full size range in one analysis. There is no need to combine results from two optical systems or analysis techniques. A detailed description of the analysis system can be found in Horiba (2009).

2.4.3 Environmental magnetism: principles and applications

The pioneering research of a Gustav Ising in the 1920s showed that clays deposited in ancient ice-lakes carried a stable natural remanent magnetisation (NRM), and hence a potential record of temporal changes in the direction and intensity of Earth's geomagnetic field (Sandgren and Snowball, 2001). The current applications of non-directional mineral magnetic techniques to lake-sediments originated through the work of John Mackereth who discovered that unconsolidated organic-rich sediments deposited in Lake Windermere also carried a stable NRM, and that it should be possible to establish regional palaeomagnetic secular variation (PSV) master curves, which could be used for relative dating and correlation (Mackereth, 1971; Sandgren and Snowball, 2001). Roy Thompson successfully applied the palaeomagnetic technique to several localities in northern Europe (Thompson, 1973; Sandgren and Snowball, 2001). Frank Oldfield (Oldfield, 1977) explored the use of lakes and their drainage basins as units of ecological study.

Thompson *et al.* (1975), while working on sediments deposited in Lough Neagh, Northern Ireland discovered that down-core magnetic susceptibility peaks were linked to periods of deforestation and the subsequent erosion of mineral soils. These magnetic susceptibility peaks could be traced from core to core, enabling correlation (Hay *et al*., 1997). The variations in the amplitude of the magnetic susceptibility record could be used as a proxy-indicator of the intensity of erosion. This observation (Sandgren and Snowball, 2001) opened the “flood-gates” for a variety of projects designed to qualify and quantify the source of the magnetic signal identified in lake-sediments and peat bogs (Hay *et al*., 1997).

Hu *et al*. (2003) working on “Magnetic responses to acidification in Lake Yangzonghai, SW China” concluded that in recent decades, magnetism is increasingly used for pollution studies (Petrovsk *et al*., 2000), because its measurements are fast, cost-effective, non-destructive (Dearing, 1999; Oldfield *et al*., 2003; Hutchinson, 2005), sensitive and informative (Dekkers, 1997). These attributes have been demonstrated by many researchers (Magiera and Strzyszcz, 2000; Shu *et al.*, 2001; Zhang *et al*., 2001; Knab *et al*., 2001; Schibler *et al*., 2002; Hu, *et al*., 2003; Hutchinson, 2005). Mineral magnetic measurements can be used to locate the pollution sources, to trace the transportation and distribution of pollutants, and to reconstruct the pollution history (Caitcheon, 1998).

Research by Hu *et al.* (2003) shows that magnetic approaches have been applied to many different targets for pollution studies; examples include soils (Jordanova and Jordanova, 1999), road dust (Hoffmann *et al*., 1999), airborne particles primarily from vehicles (Rose *et al*., 1999), and lakes (Zhang *et al*., 2001). Mineral magnetic techniques have been used to detect anthropogenic pollution caused by power plants (Heller *et al*., 1998; Kapicka *et* *al*., 2000; Hu *et al*., 2003). Fly ash and burned coal ash discharged by power plants are known to be always strongly magnetic, and can be easily detected by magnetic means (Hu *et al*., 2003). Detecting the amount of the airborne contaminants deposited in the sampled lakes has been one of the main focuses of this research. The research of Curtis *et al*. (2005) shows that high mountain lakes are sensitive to environmental change and also pointed out that the effects of air pollution and lake acidification have been recorded in many countries.

Various researchers have demonstrated that the field of environmental magnetism has been developed as a means of palaeoenvironmental or palaeoclimatic investigation which involve the magnetic analysis of sediments which characterises the sediments based on mineralogy, concentration, and grain-size variation of magnetic minerals which occur in them (Inouea *et al*., 2004; Hutchinson, 2005). Mineral magnetic properties have become an important phenomenon in sediment studies. Mineral magnetic properties of sediment have been used to determine the sources of sediment, and associated nutrients and contaminants in drainage basins (Caitcheon 1998; Hu *et al*., 2003). Caitcheon (1998) stated that determining the sources of sediment, and associated nutrients and contaminants, is an important issue for the management of water quality in river systems. It has been pointed out that dated sediment cores from channel and flood-plain deposits sampled at stream junctions can provide valuable information about longer term trends in source contributions (Caitcheon, 1998).

The research of Inouea *et al*., (2004) on environmental magnetism of brackish-water sediments from Lake Tougou-ike shows that magnetic properties of sediments enable environmental studies related to their origin, transportation, deposition and post-depositional process of the sediments. Investigations (Roberts *et al*., 1995; Inouea *et al*., 2004) showed that concentration of magnetic minerals often shows clear correlation with climate changes characterized in various timescales. For instance, variation of low-field magnetic susceptibility in deep-sea sediments or loess and palaeosol sediments can be linked with global glacial–interglacial cycles of 10,000–100,000 years (Inouea *et al*., 2004).

Booth *et al*. (2005) observed that mineral magnetic measurements become an important analytical tool when investigating the compositional properties of rocks, sediments and soils (Thompson and Oldfield, 1986; Walden *et al*., 1999; Maher and Thompson, 1999). Many of the studies have explored the relationship between mineral magnetic measurements and chemical/physical properties of sediments (e.g. Oldfield and Yu, 1994; Clifton *et al*., 1999; Chan *et al*., 1998; Booth, 2002). Based on these investigations, mineral magnetic measurements have been identified as a suitable tool for determining sediment origin (Oldfield and Yu, 1994; Booth, 2002), sediment transport pathways (Lepland and Stevens, 1996; Booth, 2002) and atmospherically deposited pollutants (Hu *et al*., 2003 and Rose *et al*., 2009). Therefore, mineral magnetic investigations can establish the presence of trace metals in lake sediment which then require further analytical technique to identify.

2.4.3.1 Magnetotactic bacteria

The interpretation of magnetic profiles as a record of erosion and deposition is said to be complicated (Cohen, 2003) due to the fact that certain redox sensitive bacteria precipitate magnetic minerals, especially magnetite (Oldfield and Wu, 2000). The amount of iron minerals and concentrations of minerals with different magnetic susceptibility properties is a function of variations in depth and intensity of erosion (Lott *et al*., 1994; Cohen, 2003). Research by several authors: (Blakemore, 1975; Heywood *et al.,* 1990; Mann *et al.*, 1990; Bazylinski *et al*., 1993; 1995; Oldfield and Wu, 2000; Cohen, 2003) reveal the capability of certain group of bacteria to be magnetically aligned (magnetotactic bacteria). Magnetotactic bacteria produce two general types of minerals as the mineral phases of their magnetosomes (Balkwill *et al*, 1980): iron oxides and iron sulphides. The iron oxides include only ferrimagnetic magnetite (Fe_3O_4) and the iron sulphides, ferrimagnetic greigite (Fe_3O_4) and non-magnetic pyrite (FeS_2) (Bazylinski *et al*., 1994). These intracellular particles confer a permanent magnetic dipole moment to the cell resulting in the cell's magnetotactic response, i.e. a motile, bio-magnetic compass (Bazylinski, 1996).

The magnetotactic bacteria are aquatic, able to align and navigate along the Earth's geomagnetic field lines. This behaviour is known as magnetotaxis (Bazylinski 1996; Blakemore 1975; 1982). Each magnetotactic cell represents a swimming permanent magnetic dipole that is, in effect, a motile bio magnetic compass (Frankel and Blakemore, 1980). Changes in environmental factors that control the productivity of magnetic bacteria in the lake can contribute to the variability of magnetic mineral concentrations that can be observed in the lake sediments (Kim *et al*., 2005). Magnetotactic bacteria could possibly account for the high concentration of magnetic grains in the dark, organic rich layers of lake sediment (e.g. Kodama *et al.*, 1998 in Kim *et al*., 2005). In the environmental magnetic studies of annually laminated sediments from Lake Ely, northeastern Pennsylvania, USA (Kim *et al*., 2005), it is indicated that bacterial magnetite is the dominant magnetic mineral in the lake sediment.

2.4.4 Trace metals analysis

Metals are widespread pollutants of great environmental concern (Jumbe and Nandini 2009a) as they are non-degradable, toxic and persistent with serious ecological ramifications on aquatic ecology (Chopra, 2009; Jumbe and Nandini, 2009b; Jumbe and Nandini, 2009c).

In order to undertake laboratory elemental analysis, the following spectrometric techniques are available: flame and furnace AAS (Atomic Absorption), ICP-MS (Inductively Coupled Plasma-Mass Spectrometry), and ICP-OES or AES (Optical Emission Spectrometer or Atomic Emission Spectroscopy). Each technique can be used to undertake metal analysis but their functionality varies in terms of easiness of operation, detection limit, cost effective, speed of analysis and number of samples (Thermo Elemental, 2001). In recent research, metal analysis has employed ICP-OES preceded by microwave-assisted digestion.

The use of closed vessel microwave-assisted digestion and the trace metal analytical methods chosen for this research can be demonstrated by literature. Evenset *et al.* (2007) quantified metals with Plasma emission spectrometry (ICP-AES) and plasma mass spectrometry (ICP-QMS) and confirmed analytical quality of the result through analyses of certified reference sediment material. The current study has employed a Varian 700-ES Series Simultaneous ICP-OES Spectrometer (ICP – Expert II Software) to quantify metals and did the quality control of the result through analyses of certified reference lake sediment material.

2.4.4.1 Microwave digestion of sediments for trace metals analysis

The accurate measurement of trace metal concentrations is an important goal in environmental monitoring and research, as many of these elements have been identified as potentially hazardous pollutants (Jumbe and Nandini, 2009c). The use of closed vessel microwave-assisted digestion (Das *et al*., 2001; Melaku *et al*., 2005) systems under high temperature and pressure for acid digestion has now become routine (Frank and Arsenault, 1996; Hassan, 2007), as it allows shorter digestion times and good recoveries, even for volatile elements. Microwave assisted acid digestion (Kingston and Haswell, 1997) has proved to be the most suitable method for the digestion of complex materials such as soils and sediments containing oxides, clay, silicates and organic substances. In addition, it reduces the risk of external contamination and requires smaller quantities of acids, thus improving detection limits and the overall accuracy of the analytical method (Bettinelli *et al*., 2000; Valeria *et al*., 2003; Jumbe and Nandini, 2009c). Moreover, the technique is safer, simpler and provides more controlled and reproducible conditions than hot plate or block digesters (Valeria *et al*., 2003).

2.4.4.2 Principles of ICP

Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES) is a fast multi-element technique with a dynamic linear range and moderate to low detection limits (~0.2-100 ppb) (Geibert and Eades, 2009). ICP gets elements to emit characteristic wavelength of specific light which can then be measured (Bradford and Cook, 1997). Up to 60 elements can be screened per single sample run of less than one minute and the samples can be analysed in a variety of aqueous or organic matrices (Geibert and Eades, 2009).

Standards and samples are acquired with the aid of an autosampler and are fed to argon plasma via a nebuliser and spray chamber using the integral peristaltic pump (Gaines, 2010). Several different nebulisers and spray chambers may be used and the choice depends largely on the nature and quantity of the samples to be analysed. Typically, this might be a concentric glass nebuliser and a cyclonic spray chamber which humidifies the carrier (nebuliser) gas before it is supplied to the nebuliser. This humidification prevents salts building up on the tips of the nebuliser and the injector component of the plasma torch and is particularly beneficial for samples with high concentrations of total dissolved solids (Thomas, 2001). A solid-state radio frequency (RF) generator supplies RF energy at 40MHz to a coil around the horizontally mounted quartz torch to maintain the argon plasma, the (relatively cool) end of which is removed by a shear gas - a thin 'wall' of high-velocity air. Light from the plasma and the various atomic emissions therein passes through windows viewing the plasma radially (side-on) into the thermally stabilised, argon-purged optical compartment housing an Echelle polychromator to separate the light into its component wavelengths. This polychromator comprises diffraction gratings, mirrors, lenses and visible and ultra-violet detector sets each comprising arrays of charge-coupled devices and allows simultaneous quantification of atomic emissions in the plasma (Thomas, 2001; Geibert and Eades, 2009; Gaines, 2010)

2.4.5 Radiometric dating

Lead-210 (half-life is 22.3/year) is a naturally produced radionuclide, derived from atmospheric fallout (termed unsupported ^{210}Pb). The naturally occurring radioisotope of lead (^{210}Pb) is mostly employed for establishing the depth-time scales of recent sediments, that is dating of sediments of up to about 150 years (Smol, 2008). Radiometric dating depends on fluctuations of ^{210}Pb and ^{137}Cs radioactivity in the sediment. Atmospheric ^{210}Pb is thought to be deposited onto surfaces (e.g. land and water) at a relatively constant rate (Appleby 1998).

The different techniques for assessing ^{210}Pb data and calculating a best chronology can be accessed in the literature (e.g. Appleby and Oldfield 1983; Oldfield and Appleby 1984; 1998; 2001). By employing ^{210}Pb dating methodology the dominant processes by which fallout is delivered to the lake sediment can be assessed; the seasonal effects on the uniformity of supply rates can be problem in dating mountain lakes. For example, winter separates the water column from the natural atmospheric ^{210}Pb flux; therefore, ^{210}Pb deposit onto the lake and its catchment during winter is bound up in snow and ice and released only at the time of the spring thaw (Appleby 2000).

2.5 Summary

This review of the literature has considered the principles, development and applications of palaeolimnology (specifically with reference to lake sediments records in upland areas of human impacts on the environment). From the literature review, it is evident that mountain lake sediments can accumulate atmospheric pollutants (that is they can act as its archive of past conditions in the environment). It has been determined that pollution of lake systems has been a focus of the environment and has involved local, regional and global efforts in an attempt to moderate level of contamination. A careful review of lake sediments sampling and analytical techniques has demonstrated the usefulness of the lake sediments sampling and analytical procedures for this research. It can be concluded that the research can therefore make a contribution to environmental information on impacts of people on their environment in this region.

CHAPTER 3: The Study Area

3.1 Introduction

A brief description of the topography, geology, climate, vegetation and ecological importance of the Carpathian Mountains is included in this chapter. Also included is an highlight of industrial centres in Romania. The latter section relates to catchment and lake basin characteristics. It employs in most cases literature based information about the sampled lakes and their watersheds. Most of the details were obtained through the surveys of Pisota (1967, 1968 and 1971); Mindrescu (2006) and Vespremeanu-Stroe *et al*. (2008).

3.2 The Carpathian Mountains

Romania is traverse by a series of undulating mountains called the Carpathians. As Europe's largest mountain range the Carpathian Mountains have been described as a natural treasure of global significance (Bytnerowicz *et al.*, 2003). They support a wealth of natural diversity which is unparalleled in Europe; and a rich cultural heritage (Oszlanyia *et al*., 2004) reflecting centuries of human settlement and history. The Carpathians cover an area of about 209, 000 km^2 (Bytnerowicz *et al.*, 2003) that extend over seven European countries; from Romania in the south, through Ukraine, Poland, Slovakia and Hungary to the Czech Republic and Austria in the north (Bodnariuc *et al.*, 2002; see Table 3.1). This portion of the thesis discusses the Carpathian Mountains under the following sub-headings: Topography, Geology, Climate, and Vegetation and Ecological importance.

3.2.1 Topography of the Carpathian Mountains

Bodnariuc *et al.* (2002) described the Carpathian Mountains as an almost semicircular shaped mountain belt that is 1500 km long and 50-150 km wide (Table 2.1), that curves through north-eastern Romania, western Ukraine, southern Poland, and Slovakia. Bytnerowicz *et al.* (2003) estimated the area covered by the Carpathian Mountains to be about 209,000 km^2 with Romania having 55.5% of the total area of the range; followed by

Poland with 19.6%; Slovakia 17.6%; Ukraine 10.3%; Hungary 4.3%; and the Czech Republic 1.3% (Table 3.1). The work of Oszlanyia *et al.* (2004) describe the Carpathians as the largest mountain range of Central Europe in terms of maximum elevation and the total span of the mountain chain, measuring approximately 1400 km in length. Geographically, the Carpathians can be divided into three parts (Figure 3.1): Western, Eastern and Southern Carpathians (Bodnariuc, *et al*., 2002; Oszlanyia, *et al*., 2004). The Carpathians can also be divided along their length into a discontinuous inner belt and a continuous outer belt that is present only in the West and East Carpathians. The East Carpathians include mountain ranges with elevations of up to around 1800 m while the Southern Carpathians of up to 2500 m (Fielitz and Seghedi, 2005). While the region's valleys owe their creation to rivers, former glaciers have carved out lakes at the highest points – there are 110 such lakes in the High Tatras alone (CEI, 2001).

Table 3.1: Carpathian facts and figures (Source: Bodnariuc *et al.*, 2002; CEI, 2001)

Total area	209 000 km^2
Dimensions	1500 km long, up to 350 km wide
Highest peak	2665 m, Gerlach in High Tatras, Slovakia
Carpathian countries (% of Carpathians)	Romania (55%), Slovakia (17%), Ukraine (11%), Poland (10%), Hungary (4%), Czech Republic (3%), Austria (<1%)
Source of major rivers	Vistula, Dnister, Prut, Aluta and tributaries (e.g. Tisza, Vag)
Key wildlife	8000 brown bears, 4000 wolves, 3000 lynx
Geology	Carpathian Flysh, with small areas of limestone and granite
Population	ca. 16-18 million
Main economic sectors	Agriculture, forestry, tourism, local industry, mineral exploitation
Protected area	16% is under some form of protection

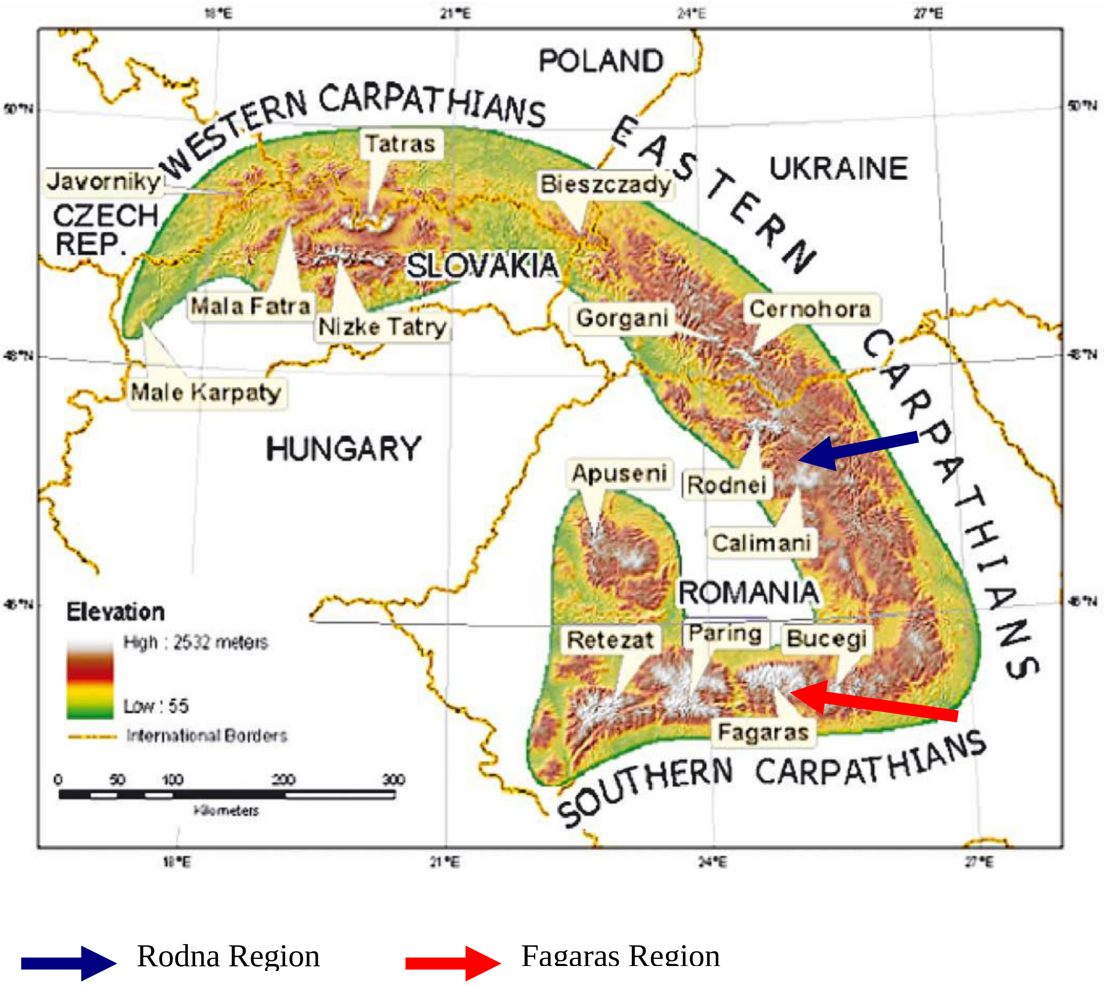


Figure 3.1: Geographical subdivisions of the Carpathian Mountains
(Source: Oszlanyia *et al.*, 2004)

3.2.2 Geology

Mindrescu *et al*. (2010a) describes the geology of the mountains of Romania as relatively young. They were folded and exhumed in the Cretaceous and the Cenozoic Alpine Orogeny. The structures of the 'inner' ranges are essentially Palaeogene, while the outer fold thrust belt (mainly the East Carpathians) is Miocene, with collision tectonics culminating 13–11Ma ago (Mindrescu *et al*., 2010b). Sandulescu (1994) described the geology of Romania as a predominantly alpine Carpathian Folded Belt (Orogen) (Figure

3.2) with the foreland consisting of several platforms, as well as the North Dobrogea Orogen (Figure 3.3). CEI (2001) described the geological formation of the Carpathian Mountains as composed mainly of sequences of sandy rocks, known as flysch formations, forming layers of alternating sandstone and shale. Other parts of the Carpathians are formed of limestone, or, as in the case of the Tatras, magmatic rock such as granite.

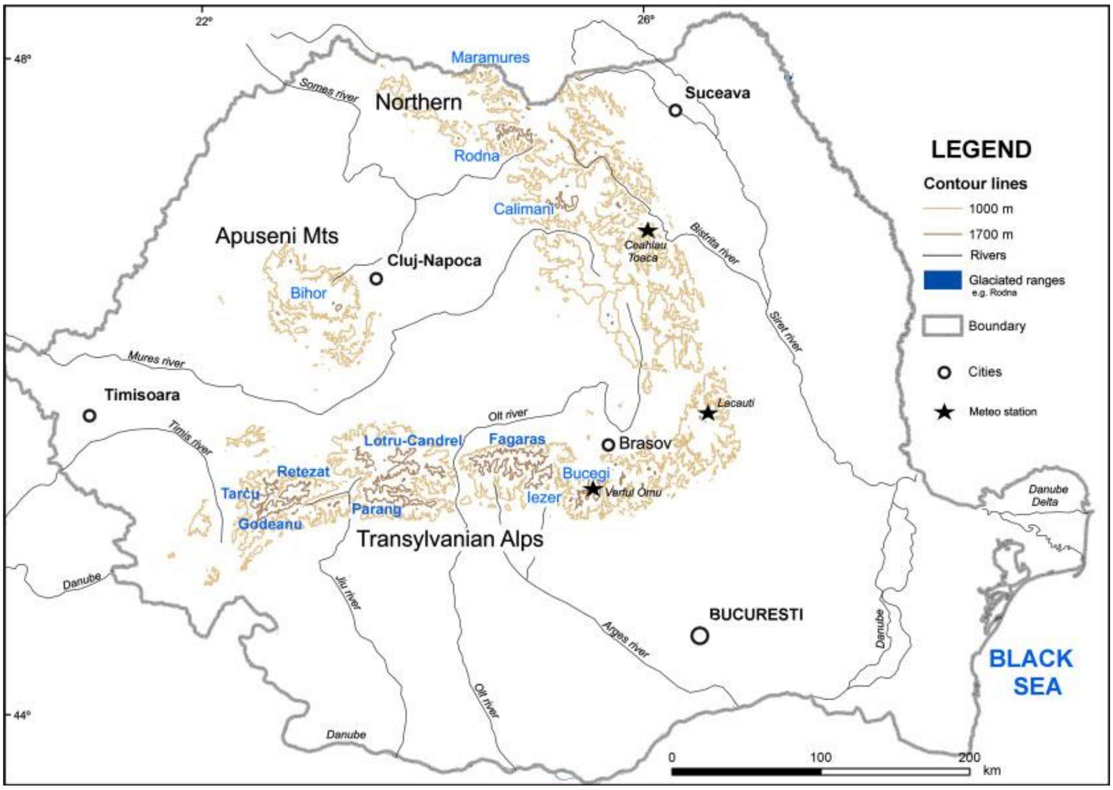
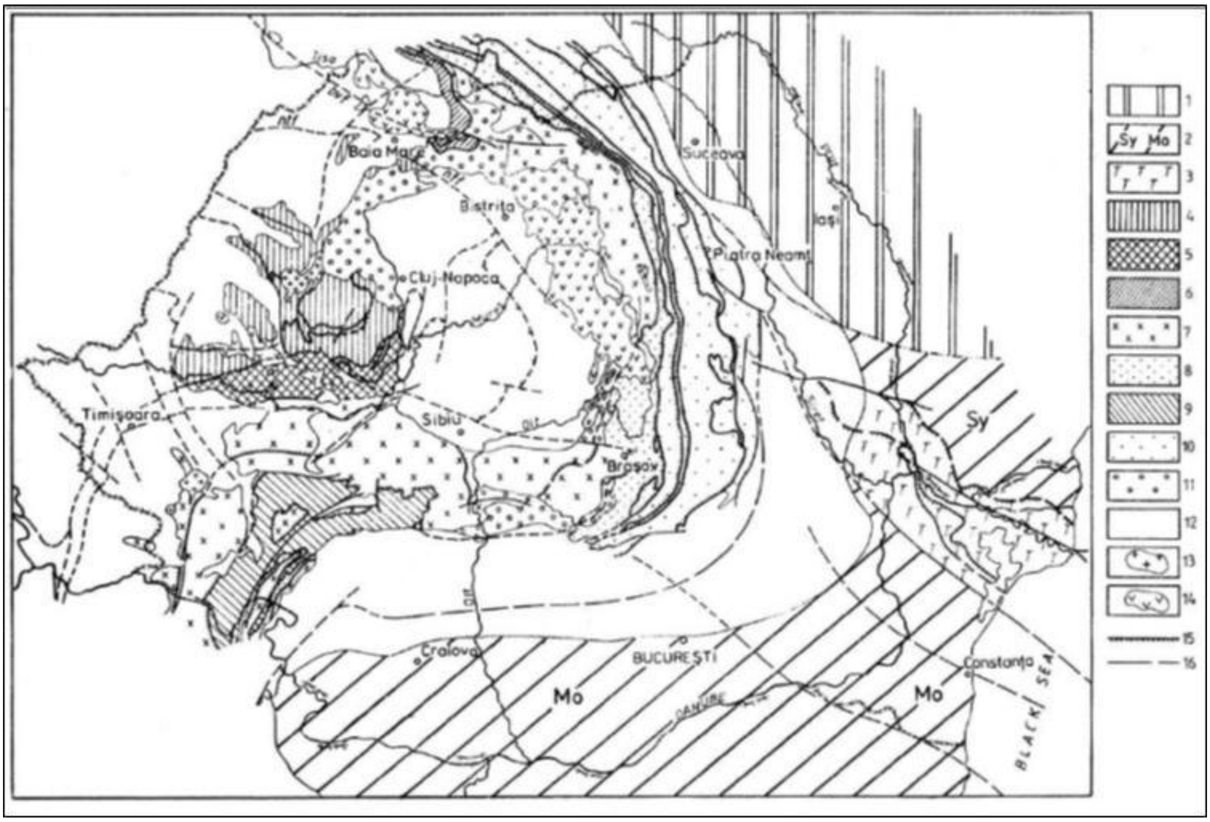


Figure 3.2: Romania, showing high ground, with names (in blue) of the main mountain ranges with cirques (Source: Mindrescu *et al.*, 2010b)



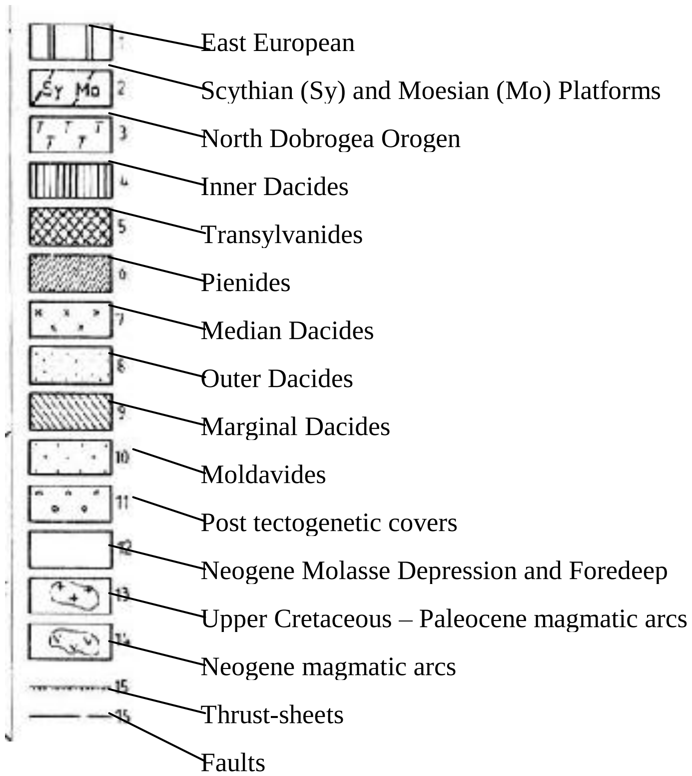


Figure 3.3: Tectonic sketch of Romania, Carpathian Foreland (Source: Sandulescu, 1994)

3.2.3 Climate

Feurdean (2004) and Bodnariuc *et al*. (2002) describe the Romanian climate as continental temperate varing across the country. Vespremeanu-Stroe *et al*. (2012) shows that the North Atlantic Oscillation (NAO), which occurs in particular during boreal winter and has almost a null signal in summer, is one of the most important modes of large scale climate variability in the Northern Hemisphere. Studies shows that changes in the NAO affects the temperatures, precipitation and wind speed in the Atlantic-European region (Kushnir, 1994; Hurell, 1995; Thompson and Wallace, 2000). A positive NAO index, aids westerly flow across the North Atlantic during winter; it moves relatively warm (and moist) maritime air over much of Europe, enhances precipitation over northern Europe while less precipitation over central and southern Europe (Hurrell, 1995; Rambu *et al*. 2002; Bojariu and Gimeno, 2003). The situation is reversed during the negative phase of the NAO (Vespremeanu-Stroe *et al*. 2012).

Figure 3.4 shows that in Romania, the strongest wind flows are recorded more often in an eastward and southward direction (Vespremeanu-Stroe et al. 2012). This study of the wind regime of Romania employed data from a 50 years period, collected from 167 meteorological stations and shows stronger winds in the area of the Fagaras Mountains, than the Rodna Mountains area (Figure 3.4). This tendency may enhance regional variations in atmospheric deposition and pollution levels between the south and north Romanian Carpathians.

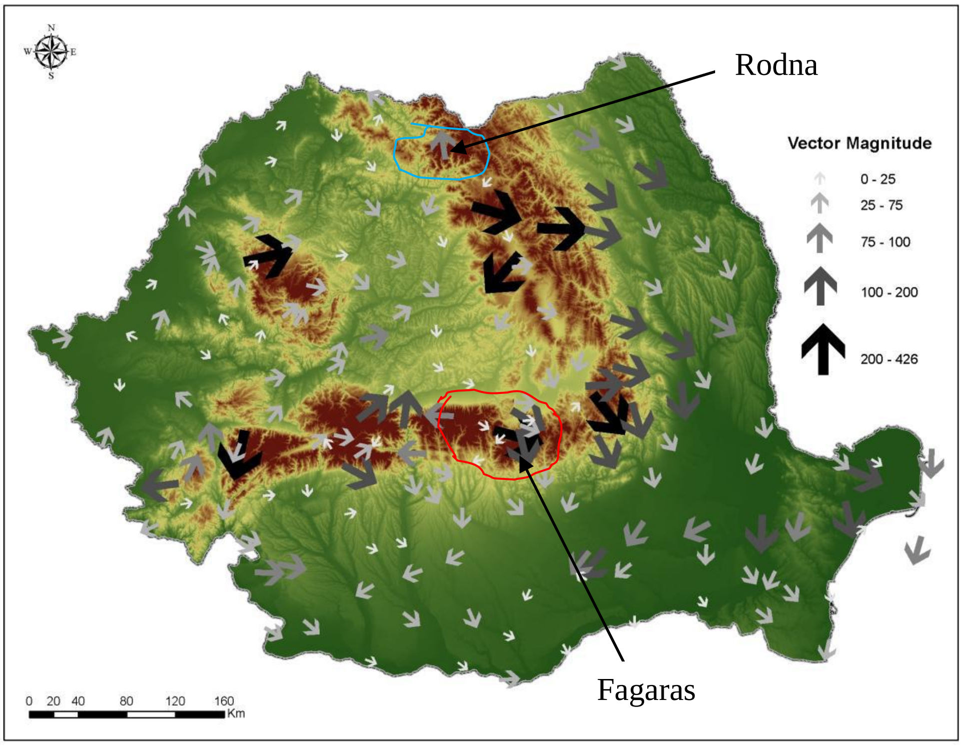


Figure 3.4: The wind vector magnitudes over the Romanian territory (Vespremeanu-Stroe *et al*. 2012).

The annual temperature range (Eastern Carpathian) is less than 19°C, from below -8°C in January to below 10°C in July. Mean annual temperature (1979–1999) was +0.1°C at Balea Lake (2038m altitude) in the Fagaras Mountains (Mindrescu *et al.*, 2010b). Mean annual temperature is around 8ºC and mean winter and summer temperatures are -3ºC and 12-13ºC, respectively (Feurdean, 2004). The Eastern Carpathian Mountains are relatively dry in the cold season, whereas in the warm season precipitation is more evenly spread across Romania’s mountains. In glaciated areas, precipitation occurs on more than 170 days a year, and snow lies on the ground for more than 150 days (Mindrescu *et al.*, 2010b).

Mean annual precipitation of Romania is about 1400mm (Bodnariuc *et al.*, 2002). The winter precipitation pattern over Romania is noted to be indirectly related to the NAO (Zaharia *et al*. 2002). In Romania, the winter NAO related signal is stronger in the extra-Carpathian regions, due to the orographic effects imposed by the Carpathian Mountains on atmospheric flow (Vespremeanu-Stroe *et al*. 2007; Vespremeanu-Stroe and Tatui, 2011). Yearly precipitation decreases in intensity from west to east, from over 600 mm to less 500 mm in the East Romanian Plain, under 450 mm in Dobrogea and about 350 mm by the Black Sea side, in the mountainous areas they reach 1000-1500 mm (Romanian Statistical Yearbook, 2013).

The north-western part of Romania experiences mild and moist climate, influenced by western oceanic air masses; the eastern part of Romania is under the influence of cold and dry air masses from the Russian Plain; warm air masses from sub-Mediterranean areas blow across the southwest region and the southeast is influenced by dry air masses from south-western Asia (Feurdean, 2004; Mindrescu *et al*., 2010a) (see Figure 3.4).

3.2.4 Vegetation

The vegetation in Romania is said to be characterised by the presence and the dominance of deciduous and thermophilous trees (Bodnariuc *et al.*, 2002; Feurdean, 2004). To the east and north-east are steppes, mixed coniferous /deciduous forests, deciduous forest and forest steppe which characterise the Eurasiatic Steppic Region (Bodnariuc *et al.*, 2002). To the west is the Atlantic Domain dominated by deciduous broad leaved trees. To the south of Romania, the Mediterranean Domain is mainly characterised by sclerophylous evergreen trees and shrubs (Bodnariuc *et al.*, 2002; Oszlanyia *et al*., 2004).

The forest vegetation cover is distributed unequally between the Carpathian countries from 29.5% cover in Hungary to almost 60% in Romania (Bodnariuc *et al.*, 2002; Mindrescu *et al.*, 2010b). The vegetation of the Carpathian Mountains ranges from alpine, to vast tracts of natural forest and rolling meadows grazed by cattle and sheep (Table 3.2). All sites in this study are above the tree line.

3.2.5 Ecological Importance of the Carpathian Mountains

According to Bytnerowicz *et al.* (2003) some of the important attributes of the Carpathian Mountains includes having some of the most beautiful areas in Europe; being of great potential for tourism and the economic utilization of forest resources. The Carpathian forests represent unique reservoirs of many endemic, rare and unusual plant and animal species which has led to the establishment of many national parks in Poland, Slovakia, Ukraine, and Romania (Bytnerowicz, *et al.*, 2002). The Carpathians are characterised by an exceptional richness of plant and animal species. More than a third of European vascular plant species grow in this area. Species endemic to the Carpathians, which form about 12% of the total flora, are of special importance (Bytnerowicz *et al.,* 2003).

The animals inhabiting the Carpathian Mountains include large predators such as brown bear (*Ursus arctos* L.), wolf (*Canis lupus* L.), and lynx (*Lynx lynx* L.) (Michalik, 1996). These mountains are also a site for endangered species such as the chamoix (*Rupicapra rupicapra* L.), the marmot (*Marmota marmota* L.) and the golden eagle (*Aquila chrysaetos* L.) (e.g. Michalik, 1996; Oszlanyia *et al*., 2004). The work of Oszlanyia *et al*. (2004) shows that the natural environment of the Carpathian Mountains are the richest in Europe in terms of species and ecological value and they constitute important parts of Europe's nature resources. Although the Carpathians are divided by political and ethnic frontiers (Oszlanyia *et al*., 2004; Feurdean, 2004) they still provide an excellent example of the possibility of protection and conservation of natural and cultural heritage (Oszlanyia *et al.*, 2004).

3.2.6 Romanian industrial centres

Figure 3.5 shows that generally, there are more industrial concentrations in the south than in the north of Romania; hence there is higher likelyhood of more pollution in the south than in the north. Nevertheless, the industries in neighbouring Hungary may serve as a pollution source for the north of Romania. This industrial mapping was done in the 1970s during the peak in industrial development of Romania.

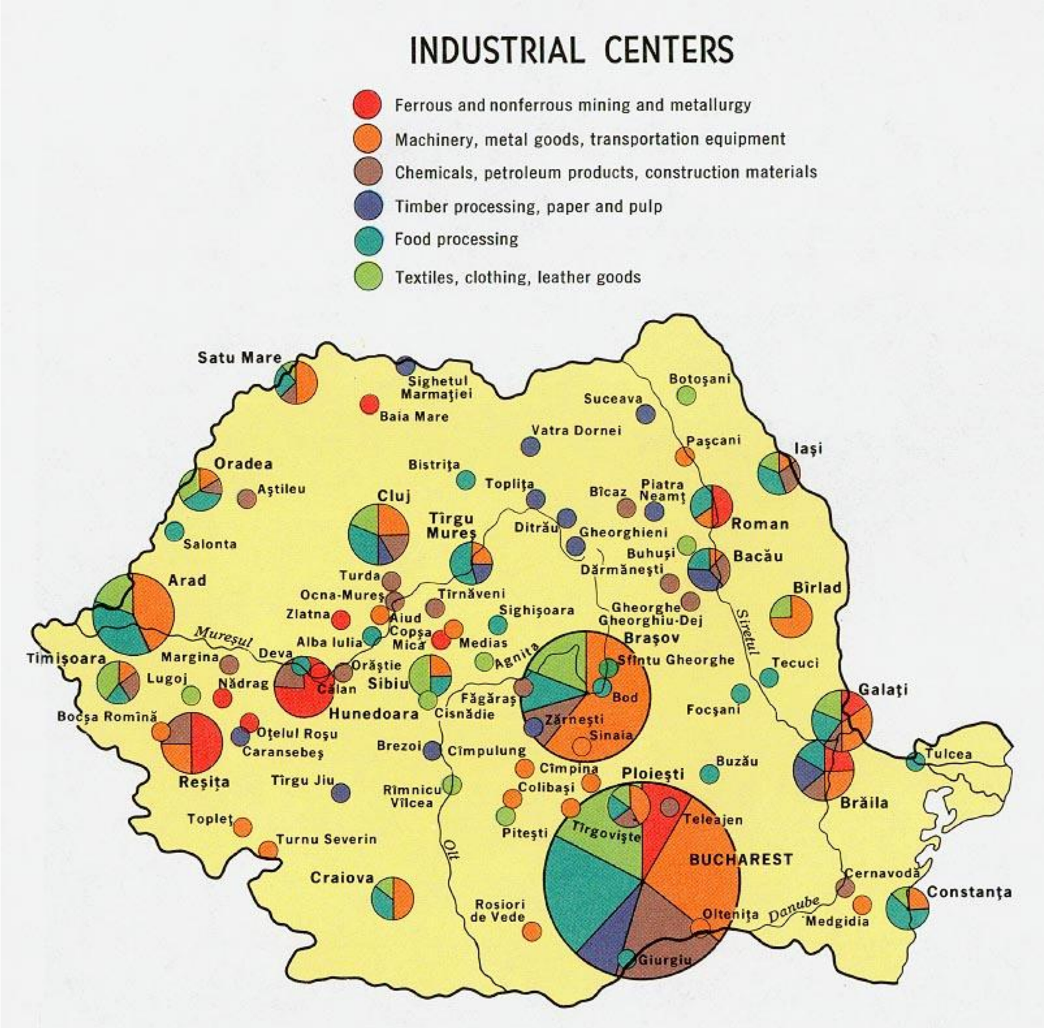


Figure 3.5: Romanian industrial centres (Perry-Castaneda Library Map Collection (2013) The University of Texas, Austin. http://www.lib.utexas.edu/maps/thematic.html. Accessed: 05-11-2013)

3.3 Catchment and lake basin characteristics

This section considers the catchment and lake characteristics in the two regions where sampling was carried out (the Fagaras and Rodna/Maramures regions) (Figure 3.6). The Fagaras region is located in the southern part of the Romanian Carpathians (the Transylvanian Alps). Four lakes were sampled in this region. These include: Balea, Caltun, Capra and Podragu Mare lakes (Figure 3.7). A catchment and lake characteristics summary is presented in Table 3.2. In this region the sampled lakes' maximum depths ranged from 8.6 - 16.5 m. Recovered sediment core lengths ranged from 0.20 - 0.32 m (Table 3.2). A more detailed description of the studied lakes is given in the latter part of this chapter.

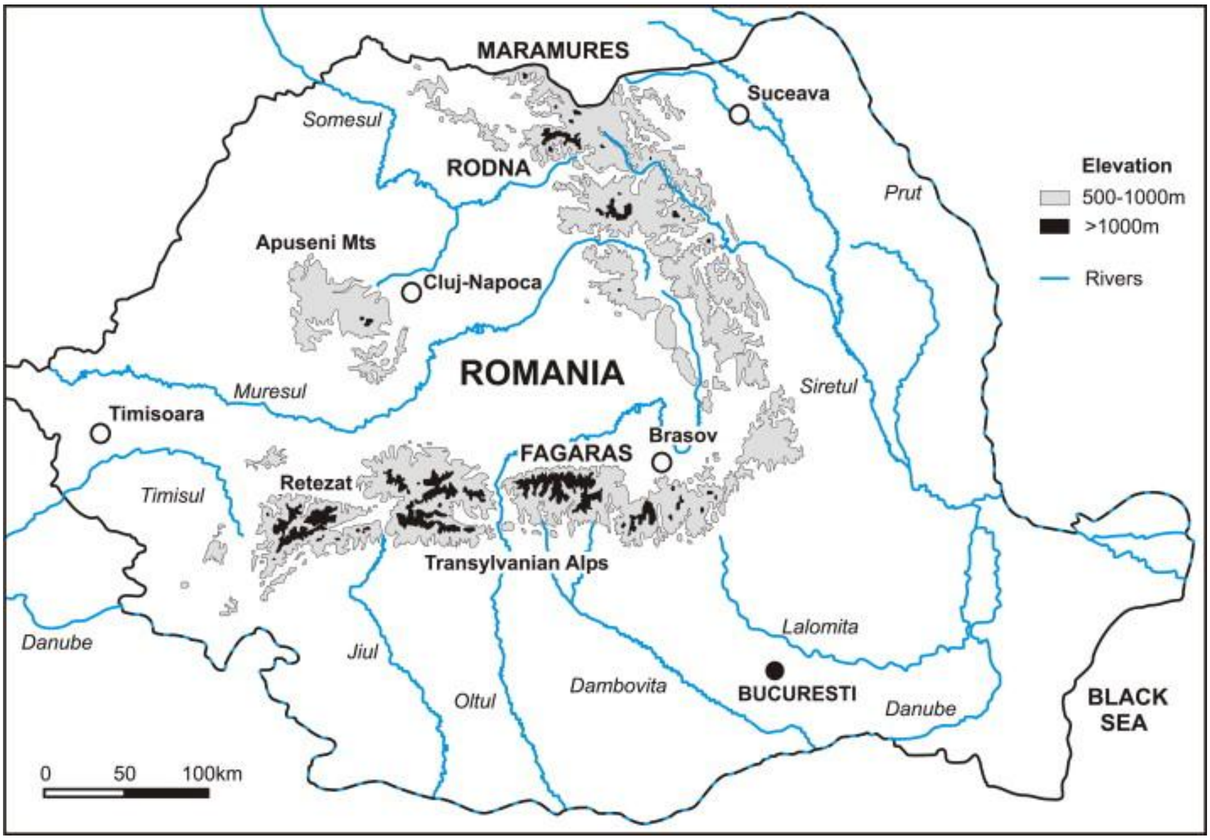


Figure 3.6: The position of the Fagaras, Rodna and Maramures Mountains in Romania (More detailed maps showing the relative positions of the sampled lakes in each of the regions are shown in Figures 3.7 and 3.8).

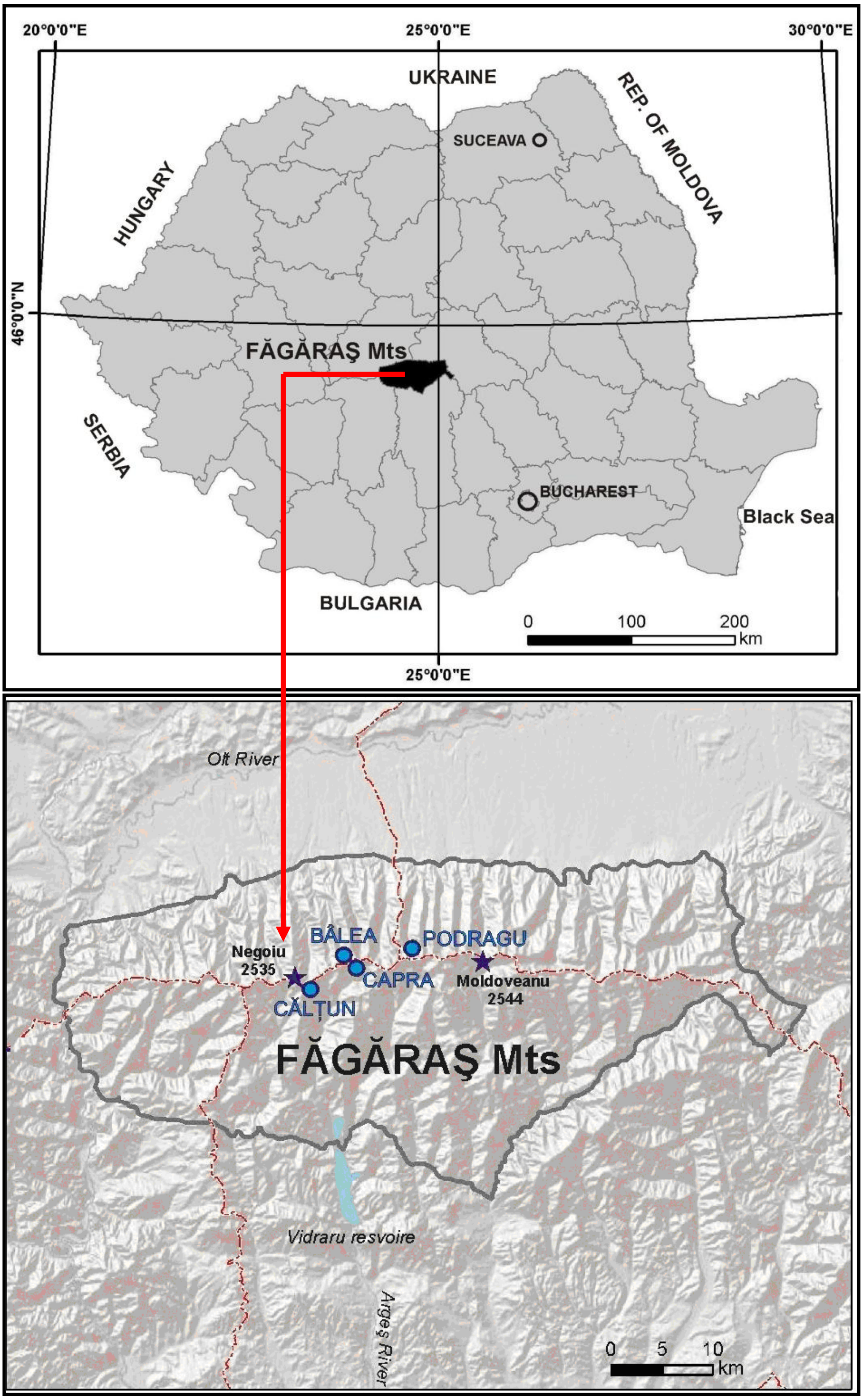


Figure 3.7: The position of the Fagaras Mountains and the relative positions of the sampled lakes.

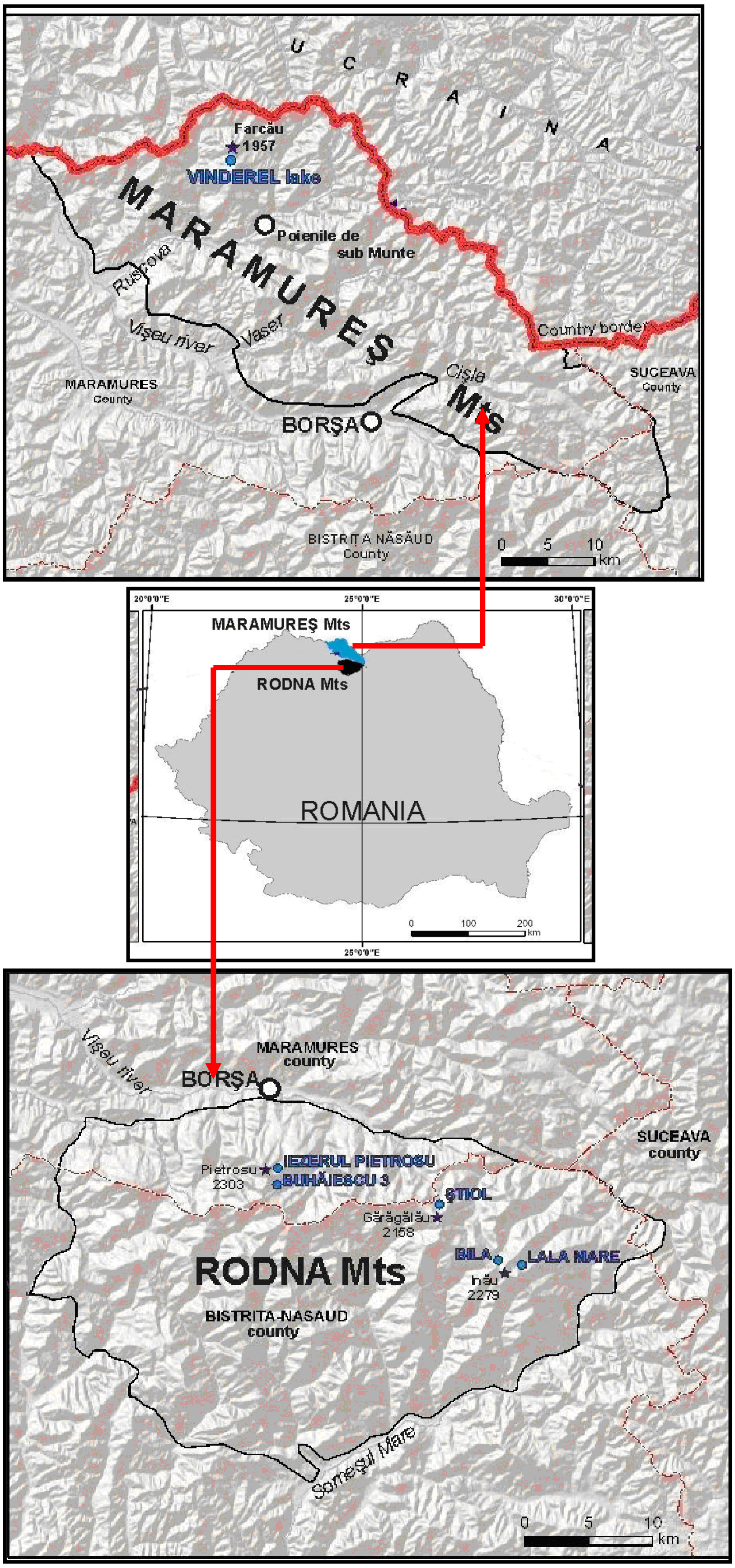


Figure 3.8: The positions of the Rodna and Maramures Mountains and the relative positions of the sampled lakes.

For the purposes of this study the Rodna region consists of the Rodna and Maramures Mountains located in the northern Carpathians. Six lakes were sampled from this region namely: Bila, Buhaiescu-3, Lala Mare, Pietrosul and Stiol (Rodna Mountains); Vinderel (Maramures Mountains) (Figure 3.8). A catchment and lake characteristics summary is presented in Tables 3.3. Although Stiol Lake had recently been artificially raised by illegal damming, all the lakes from this region were relatively shallow, ranging from 0.5 - 5.5 m in depth. Recovered sediment core lengths were also shallow (0.12 - 0.32 m) (Table 3.4).

3.3.1 Catchment and lake basin characteristics in the Fagaras region

These lakes were all located on similar geographical latitude and all the lakes were situated in classic glacial cirques. There is no marked difference in the altitudinal positions of the lakes as all were located above 2000 m. However, the lowest lake elevation is at 2035 m (Balea Lake) while the highest lake elevation is 2249 m (Capra Lake). There are distinct size variations in the catchment areas of the lakes; Podragu Mare lake has the largest catchment area of 55.2 ha followed by Balea lake (45.5 ha). The catchment areas of Capra and Caltun lakes are 29.7 ha and 18.6 ha respectively. Lake depths vary between 8.6 m and 16.5 m. The deepest lake is Podragu Mare (16.5 m) while the shallowest is Capra (8.6 m). The depth of Balea and Caltun lakes are 10.5 m and 13.0 m respectively. There are three estimates of lake area and lake maximum depth as shown in Table 3.2. In most cases estimates 1 and 2 are similar within each lake but estimate varies obviously (e.g depth in Balea Lake). Catchment and lake area[1] were determined from 2005 aerial photography. Lake depth (max)[1] and sediment core length were determined in the field. Lake area[2] and lake depth (max)[2] were products of Pişota (1971) survey while lake area[3] and lake depth (max)[3] were measured by Vespremeanu-Stroe *et al.* (2009). Lake area[1] was used to calculate catchment: lake ratio. This ratio ranged from 9.5-17.6. The maximum recovered sediment lengths are 0.32 m, 0.32 m, 0.30 m and 0.20 m from Balea, Capra, Caltun and Podragu respectively (Table 3.2).

Table 3.2: Summary of catchment, lake and sediment core data for the southern study area

Southern area	**Balea**	**Calţun**	**Capra**	**Podragu Mare**
Position	45° 36’ 13’’ 24° 37’ 07’’	45° 34’ 55’’ 24° 34’ 26’’	45° 36’ 03’’ 24° 37’ 46’’	45° 36’ 34’’ 24° 41’ 32’’
Catchment				
Geomorphology (position of lake)	within cirque, central	within cirque, central	within cirque, central	within cirque, central
Aspect	North	South	South	North
Area (ha)	45.5	18.6	29.7	55.2
Altitude range (m)	2035-2507	2139-2517	2249-2507	2066-2462
Land cover (% surface area)	grass (53) scree (26) rock (10) water (11) scrub (0)	scree (43) grass (28) rock (25) water (4) scrub (0)	grass (65) scree (20) rock (9) water (6) scrub (0)	grass (48) scree (25) rock (20) water (6) scrub (0)
Geology type[a] (% surface area)	moraine (53), paragneiss, mica schist, quartzite (41), amphibolites (6)	quartzite, paragneiss, mica schist (72), moraine (28)	moraine (60), amphibolites (21), paragneiss, mica schist, quartzite (19)	paragneiss, mica schist, quartzite (74), moraine (26)
Lake				
Altitude (m)	2035	2139	2249	2066
Area[1] (ha)	4.78	0.80	1.87	3.13
Area[2] (ha)	4.65	0.77	1.83	2.85
Area[3] (ha)	5.04	-	1.87	3.49
Depth (max)[1] (m)	10.5	13.0	8.6	16.5
Depth (max)[2] (m)	11.3	11.8	8.0	15.5
Depth (max)[3] (m)	16.9	-	13.1	18.7
Catchment:lake ratio	9.5	23.3	15.9	17.6
Sediment				
Core length (max) (m)	0.30	0.33	0.35	0.20

Note: Catchment and lake area[1] determined from 2005 aerial photography. Lake depth (max)[1] and sediment core length determined in the field. (Lake area[1] used to calculate catchment:lake ratio.) Lake area[2] and lake depth (max)[2]; Pişota (1971). Lake area[3] and lake depth (max)[3]; Vespremeanu-Stroe *et al.* (2008). Geological units determined from Sandulescu (1984).

The catchment geology of both Balea and Capra are dominated by moraine deposits. Capra’s geology is 60% moraine deposits while Balea geology is 53% moraine deposits. Caltun lake catchment is 72% quartzite, paragneiss, mica schist while Podragu Mare Lake has 74% paragneiss, mica schist plus quartzite. The land cover of all four catchments is predominantly grass except for Caltun where scree dominates (Table 3.2). A catchment map of each lake illustrating the topography of the area, a photo taken during the field trip illustrating catchment feature and a bathymetric map (where available) are presented below (Figures 3.8-3.16).

Balea Lake

Balea Lake is situated at an altitude of 2035 m. It is the largest lake and has an area of 4.78 ha and it has a maximum depth of 10.5 m (Table 3.2). The lake has a catchment: lake ratio of 9.5 which is the smallest in the Fagaras region. It is located in a cirque (Figure 3.9). It is the only lake where the catchment includes a road network. There is also a road tunnel and a hotel beside the lake. The lake experiences significant tourist number annually (Robert 2005) (Figure 3.10). The geology of Balea lake catchment consists of moraine (53%) paragneiss, micashists, micaceous quartzites (41%) and amphibolites (6 %).

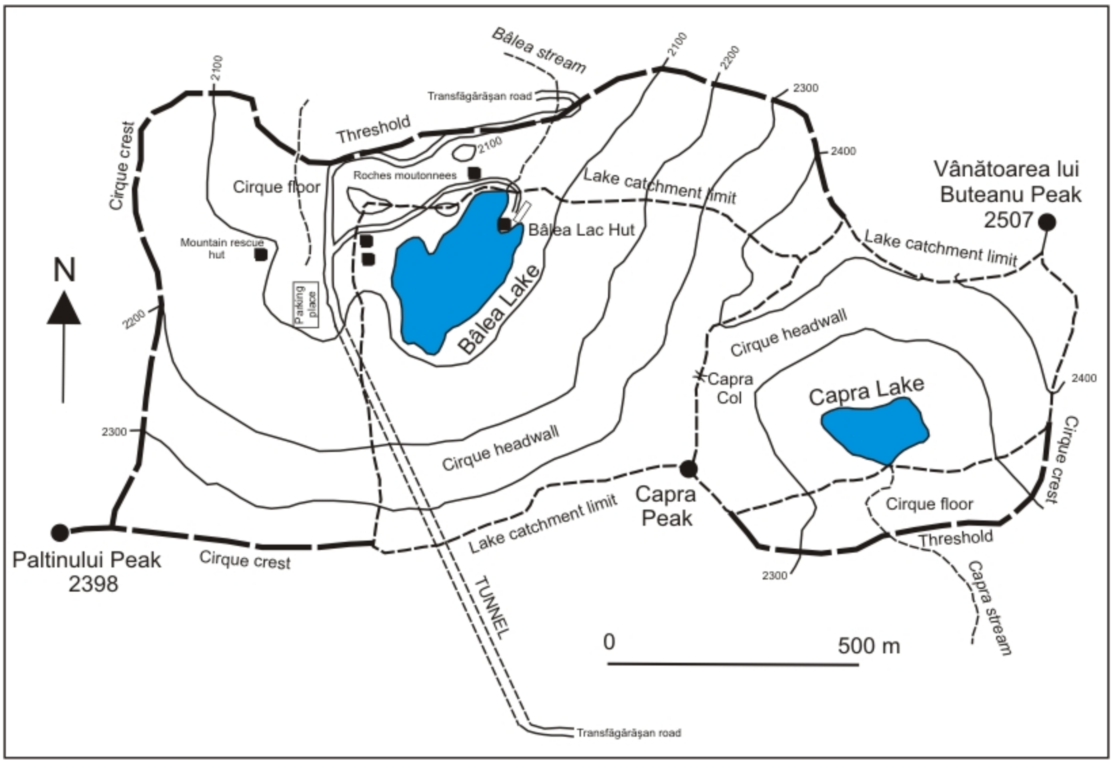


Figure 3.9: Balea and Capra lakes catchments maps (Mindrescu, 2001)

Figure 3.10: Balea Lake and catchment looking northeast (2007 field trip)

Note: The Transfagarasan road is located in the vicinity of Balea Lake. Balea Lake Chalet is situated on the lakeside. The catchment area has some year round snow patches.

Capra Lake

Capra Lake is situated in a cirque (Figures 3.8, 3.11 and 3.12) at an altitude of 2249 meters. It is the highest lake in the Fagaras region. The lake has an area of 1.87 ha and it has a maximum depth of 8.6 m (see Table 3.2). The lake has a catchment: lake ratio of 23.3 which is the largest in the Fagaras region. The geology of the Capra Lake catchment includes: 60% moraine deposits, 21% amphibolites and 19% paragneiss, mica schist plus quartzite. The land cover is predominantly grass used for grazing. There is no road in the catchment, but it is relatively close to the Fagaras Highway and is accessible by path (see the upper part of Figure 3.11). In Figure 3.12 an arrow indicates the deepest part of the lake from which the sediment cores were taken.

Figure 3.11: Capra Lake and catchment looking south (Source: http://www.testq.com)

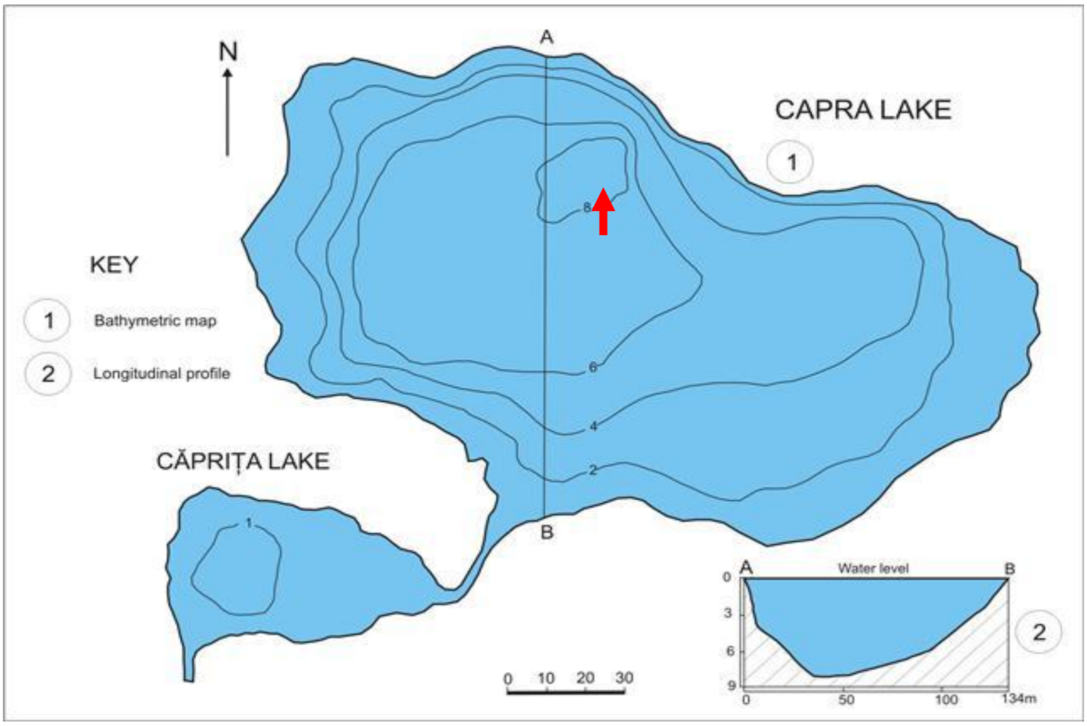


Figure 3.12: Capra Lake bathymetric map (Mindrescu, 2001). Arrow shows deepest part of lake.

Caltun Lake

The geology of Caltun Lake includes: quartzite, paragneiss, micashists (72%). There is 28% coverage of deposits such as moraine. The land cover comprises of dominant scree with a proportion of 43%, 28% grass, 25% rock and 4% water. Caltun Lake is situated in a cirque (Figures 3.13 and 3.14) at an altitude of 2139 m. It has an area of 0.8 ha and it has a maximum depth of 13.0 m (Table 3.2). The lake has a catchment: lake ratio of 15.9. The lake is relatively remote, but small mountain hut is close to the lake.

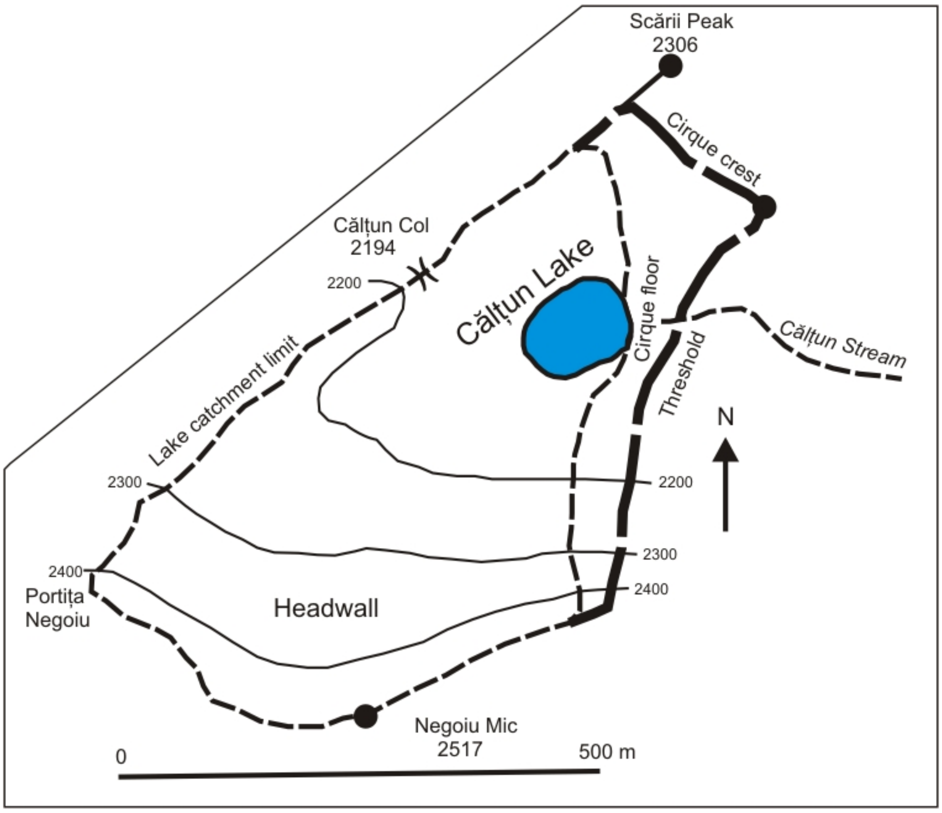


Figure 3.13: Caltun Lake catchment map (Mindrescu, 2001)

Figure 3.14: Caltun Lake and catchment (2007 field trip) looking north with scree dominating the land surface.

Podragu Mare Lake

Podragu Mare Lake is situated at an altitude of 2066 m. This lake has a surface area of 3.13 ha and a maximum depth of 16.5 m. The lake has a catchment: lake ratio of 17.6 which is the second largest in the Fagaras region.The geology of the catchment area of Podragu Mare Lake is predominantly metamorphic with 74% (paragneiss, mica schist, quartzite) and 26% moraine deposits. The land cover is 48% grass, 25% scree, 20% rock and 6% water (see Table 3.2). The lake is located within classic cirque (Figures 3.15, 3.16 and 3.17). A mountain hut is located near this lake, but there is no road access to the catchment (see Figure 3.16).

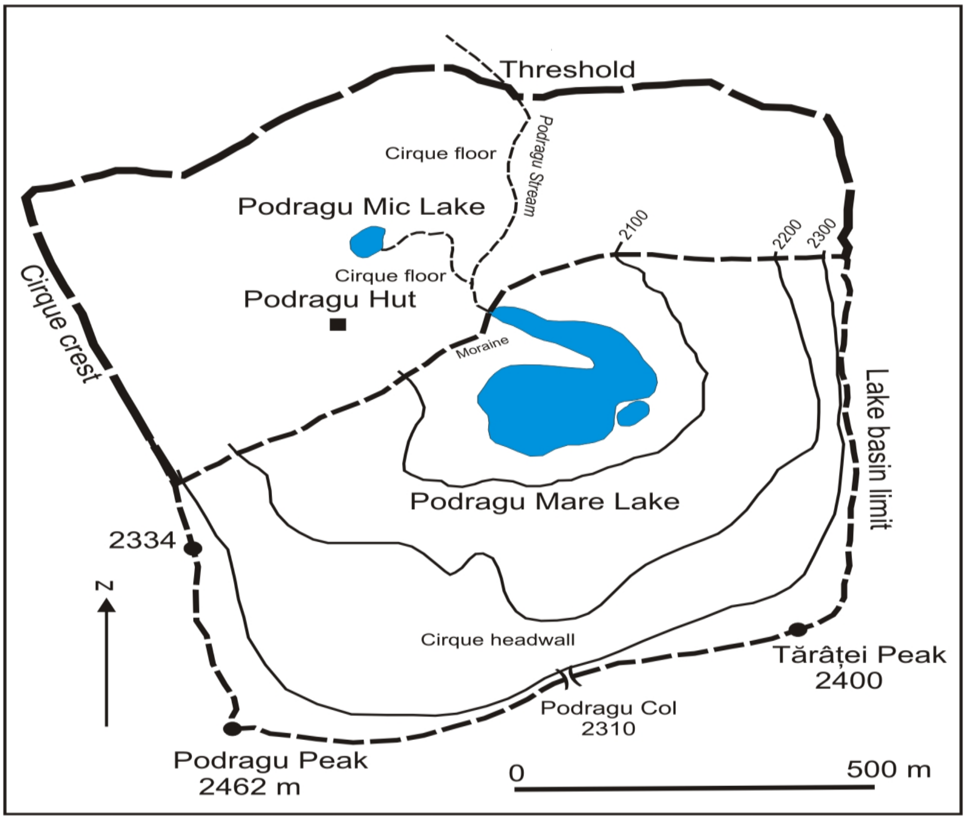


Figure 3.15: Podragu Mare Lake catchment map (Mindrescu, 2001)

Figure 3.16: Podragu Mare Lake and catchment (2007 field trip) looking east

➡ Hut

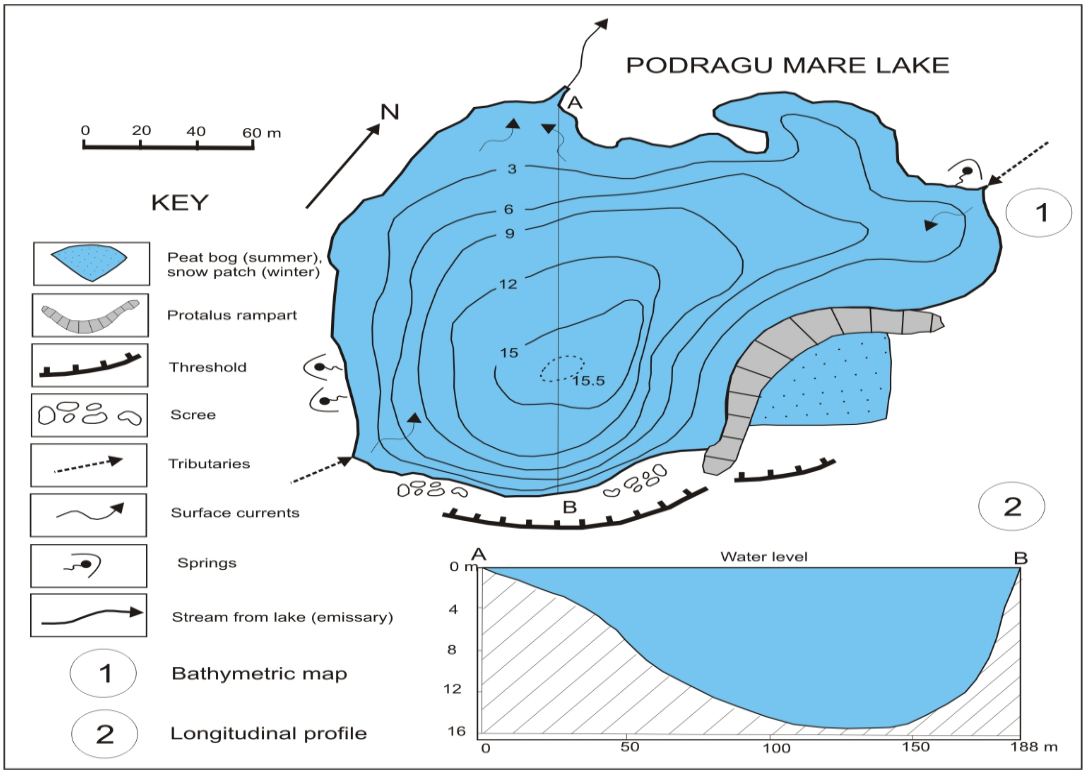


Figure 3.17: Podragu Mare Lake bathymetric map (Mindrescu, 2001)

3.3.2 Catchment and lake basin characteristics in the Rodna/Maramures region

The Rodna lakes are all located on similar geographical latitudes and each one of the lakes is located within a cirque except Vinderel which is located within a glacial col (see Tables 3.3a and 3.3b). The altitudinal positions of the lakes ranges from 1667 - 1840 m. The lowest is Stiol while Bila is at the highest altitude (Table 3.3a and 3.3b).

Table 3.3a: Summary of catchment, lake and sediment core data for the northern study area

Northern area	**Bila**	**Buhaiescu-3**	**Lala Mare**
Position	47° 31’ 58’’ N 24° 52’ 38’’ E	47° 35’ 14’’ N 24° 38’ 48’’ E	47° 31’ 41’’ N 24° 54’ 04’’ E
Catchment			
Geomorphology (position of lake)	within cirque, near headwall	within cirque, lowest glacial step	within cirque, lower, larger glacial step
Aspect	North	south east	north
Area (ha)	43.8	62.9	16.1
Altitude range (m)	1840-2279	1825-2250	1810-2160
Land cover (% surface area)	grass (70) scree (20) rock (9) water (1) scrub (0)	grass (40) scree (35) scrub (13) rock (10) water (2)	grass (64) scree (20) scrub (9) rock (3) water (4)
Geological units[a & b] (% surface area)	mica schist (88) moraine (12)	mica schist (92) moraine (8)	mica schist (80) moraine (20)
Lake			
Altitude (m)	1840	1825	1810
Area[1] (ha)	0.14	0.09	0.70
Area[2] (ha)	-	0.07	0.56
Depth (max)[1] (m)	0.5	0.5	1.6
Depth (max)[2] (m)	-	0.35	1.6
Catchment:lake ratio	312.9	698.9	23.0
Sediment			
Core length (max) (m)	0.23	0.12	0.18

Note: Catchment and lake area[1] determined from 2005 aerial photography. (Lake area[1] used to calculate catchment:lake ratio.) Lake depth (max)[1] and sediment core length determine in the field (** additional core). Lake area[2] and lake depth (max)[2]; Pişota (1968). Geological units determined from Sandulescu (1984).

Table 3.3b: Summary of catchment, lake and sediment core data for the northern study area

Northern area	**Pietrosul**	**Ştiol***	**Vinderel**
Position	47° 35’ 54’’ N 24° 38’ 52’’ E	47° 34’ 30’’ N 24° 48’ 55’’ E	47° 54’ 36’’ N 24° 27’ 26’’ E
Catchment			
Geomorphology (position of lake)	within cirque, central	within cirque, central	on the col, above the cirque
Aspect	North	north east	NA
Area (ha)	54.4	156*	5.1
Altitude range (m)	1835-2300	1671-2158	1684-1746
Land cover (% surface area)	grass (58) scree (21) rock (14) scrub (5) water (2)	grass (60) scrub (20) scree (14) rock (5) water (1)	grass (99) water (1) scrub (0) scree (0) rock (0)
Geological units[a & b] (% surface area)	mica schist (92) moraine (8)	gneiss with amphibolites and paragneisses (56) schist with limestone, green schist and dolomites (25) moraine (19)	sandstone and siltstone, diabase intrusions
Lake			
Altitude (m)	1835	1671	1684
Area[1] (ha)	0.41	1.06*	0.06
Area[2] (ha)	0.34	-	-
Depth (max)[1] (m)	2.3	5.0	5.5
Depth (max)[2] (m)	2.1	-	-
Catchment:lake ratio	132.7	147.2	85.0
Sediment			
Core length (max) (m)	0.12	0.18	0.16 (0.34)**

Note: Catchment and lake area[1] determined from 2005 aerial photography (except Ştiol* where this was calculated after lake enlargement by damming (see Mîndrescu *et al.* 2010)). (Lake area[1] used to calculate catchment:lake ratio.) Lake depth (max)[1] and sediment core length determine in the field (** additional core). Lake area[2] and lake depth (max)[2]; Pisota (1968). Geological units determined from Sandulescu (1984).

There are distinct size variations in the catchment areas of the lakes; Stiol lake has the largest catchment area of 156 ha followed by Buhaiescu-3 lake (62.9 ha). The catchment areas of Pietrosul and Bila lakes are 54.4 ha and 43.8 ha respectively. Lala Mare lake has a catchment area of 16.1 ha while Vinderel Lake has the smallest catchment area of 5.1 ha (Tables 3.3a and 3.3b). There are two estimates of lake area and lake maximum depth as shown in Tables 3.3a and 3.3b. Catchment and lake area[1] was determined from 2005 aerial photography (except Ştiol where this was calculated after lake enlargement by damming (see Mîndrescu *et al.* 2010a). Lake depth (max)[1] and sediment core length were determined in the field. Lake area[2] and lake depth (max)[2] were products of Pisota (1968) survey. Lake area[1] was used to calculate catchment: lake ratio. This ratio ranged from 23.0 - 698.9. Lake depth varies between 0.5 m and 5.5 m. The deepest lake is Stiol while the shallowest are Buhaiescu-3 and Bila. The recovered sediment core lengths were between 0.12m and 0.23 m.

The geological formations of the catchment areas are predominantly micachist and moraine deposits except at Stiol Lake where gneiss dominate. Another exception to the geological formations above is Vinderel Lake catchment which is underlain by sandstone and siltstone with diabase intrusions. The land cover of the catchments is predominantly grass and scree around the lakes (Tables 3.3a and 3.3b). A catchment map of each lake illustrating the topography of the area, a photo taken during the field trip illustrating catchment feature and a bathymetric map (where available) are presented below (Figures 3.18-3.28).

Bila Lake

This lake is situated on a glacial step within the Bila armchair cirque (Figure 3.18). The catchment area of Bila lake is 43.8 ha while the lake area is 0.14 ha (Table 3.3a). The geology of the lake catchment consists of 88% mica schist and 12% moraine deposit. The lake is not deeper than 0.50m. Most parts of the lake had a depth of less than 0.30m. The lake has a catchment: lake ratio of 312.9 which is the second largest in the Rodna/Maramures region. Its catchment is represented mostly by a cirque in cirque known as Fundu Bila. The land cover is 70% grass, 20% scree, 9% rock and 1% water.

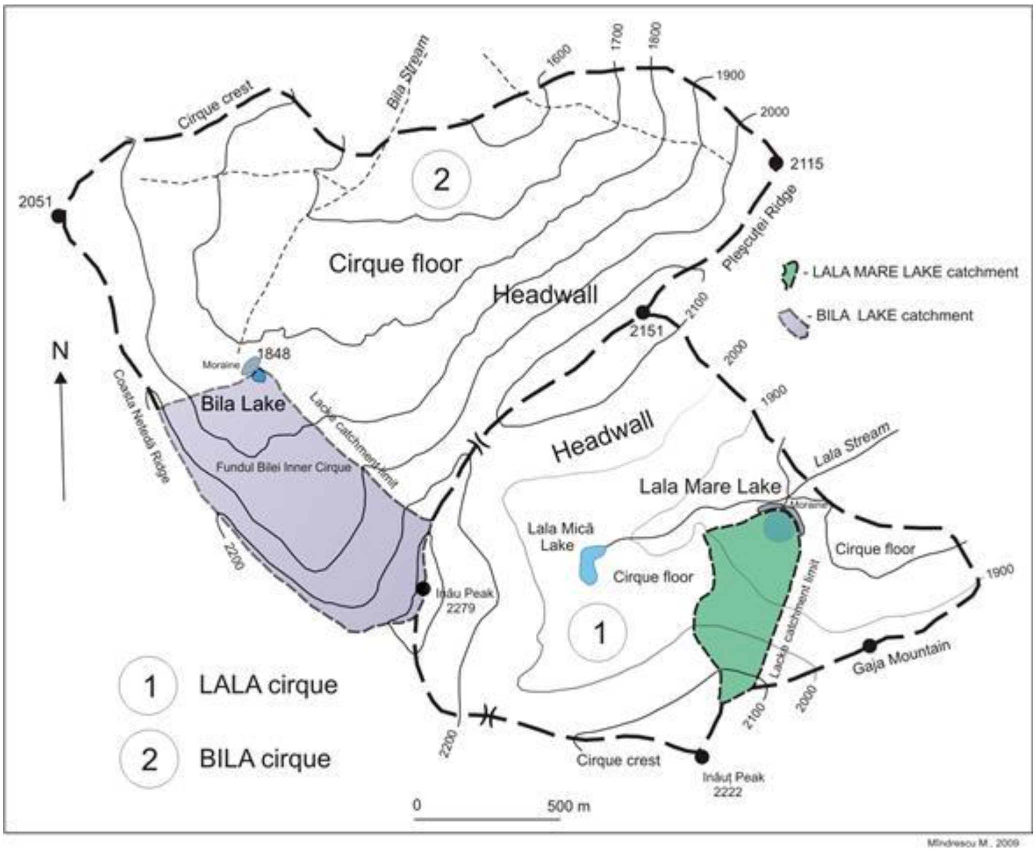


Figure 3.18: Lala Mare and Bila Lakes Catchment Map (Mindrescu, 2006)

Buhaiescu-3 Lake

Buhaiescu-3 lake is situated at an altitude of 1825 m. This lake has a catchment area of 62.9 ha but has a lake surface area of 0.09 ha and a maximum depth of 0.5 m (Table 3.3a). The lake has a catchment: lake ratio of 698.9 which is the largest in the Rodna/Maramures region. Buhaiescu-3 is the smallest sampled lake in the Rodna region. The geology of the catchment area of Buhaiescu-3 is 92% mica chist and 8% moraine deposits (see Table 3.3a, Figure 3.19). The lake is located within cirque. At the time of sampling Buhaiescu-1 and Buhaiescu-2 were ice bound (Figure 3.19). The land cover is 40% grass, 35% scree, 13% scrub, 10% rock and 2% water (Figure 3.20).

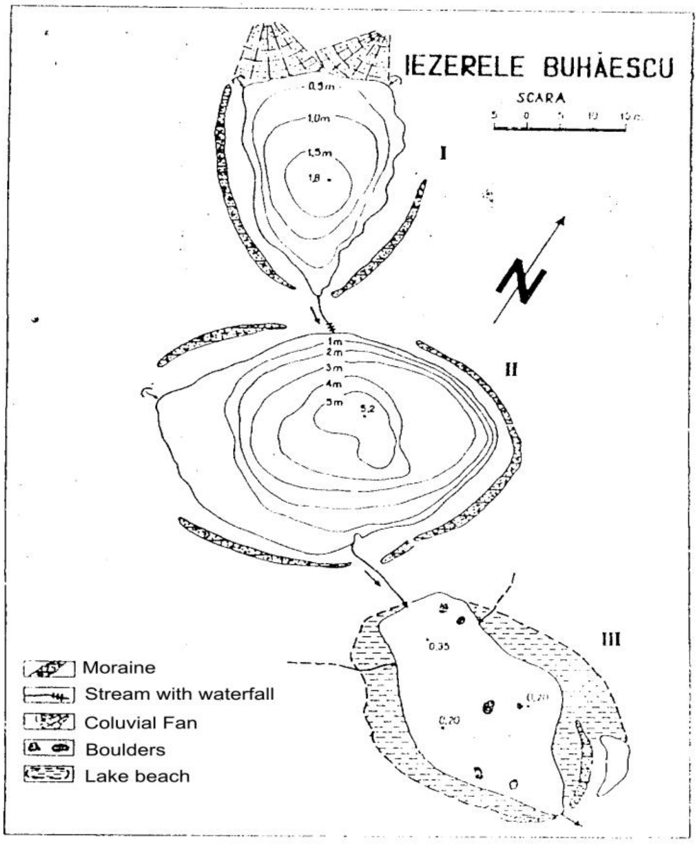


Figure 3.19: Buhaiescu: 1, 2 and 3. Bathymetric Map (Pisota, 1968)

Figure 3.20: Buhaiescu: 3 Lake and catchment looking north

Lala Mare Lake

The Lala armchair cirque is situated in the eastern section of the Rodna Mountains, between the peaks of Inau and Inauţ. This large cirque is located at the head of Lala valley at 1810 m altitude. The lake has a catchment: lake ratio of 23.0 which is the smallest in the Rodna/Maramures region It was covered by one of the biggest glaciers of the Rodna Mountains and Eastern Carpathians (Urdea, 2004). Its aspect, extent and altitude differ from alpine cirques in this area and consequently it has been classified as an armchair cirque (Pisota, 1968; Urdea, 2004). The geology of the catchment area of is 80% micashist and 20% moraine deposits (see Table 3.3a). The land cover is 64% grass, 20% scree, 9% scrub, 3% rock and 4% water (Figures 3.18, 3.21 and 3.22).

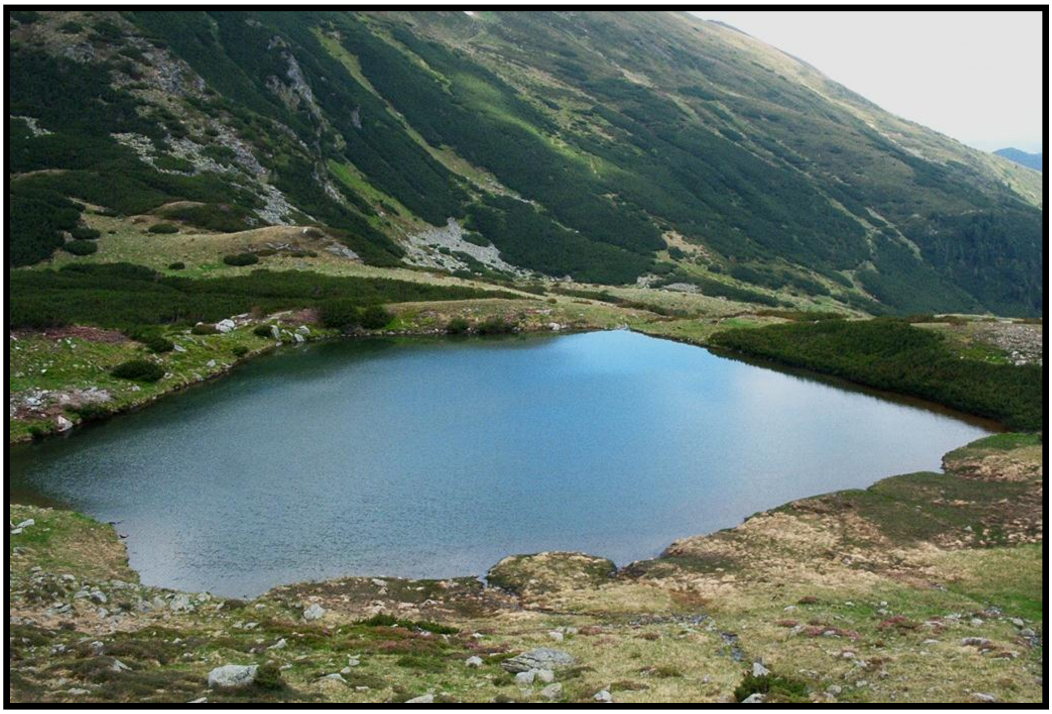


Figure 3.21: Lala Mare Lake and catchment (2006 field trip), looking north

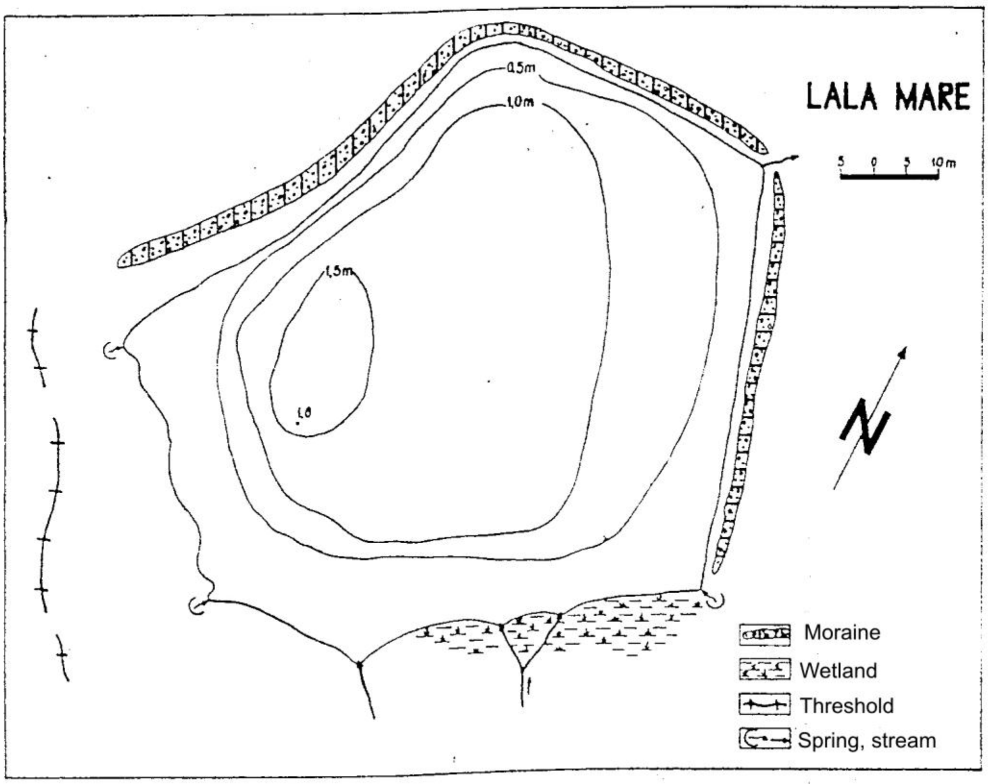


Figure 3.22: Lala Mare Lake bathymetric Map. (Pisota, 1968)

Pietrosul Lake

The lake is situated on a glacial step within the cirque at an altitude of 1835 m (Table 3.3b). The catchment area of Pietrosul lake is 54.4 ha while the lake area is 0.41 ha. It has a catchment: lake ratio of 132.7 (Table 3.3b). The geology is 92 % micashist and 8% moraine while the land cover of the catchment is predominantly grass (58%) with 21% scree, 14% rock, 5% scrub and 2% water (Figures 3.23 and 3.24). The lake maximum depth is 2.3 m.

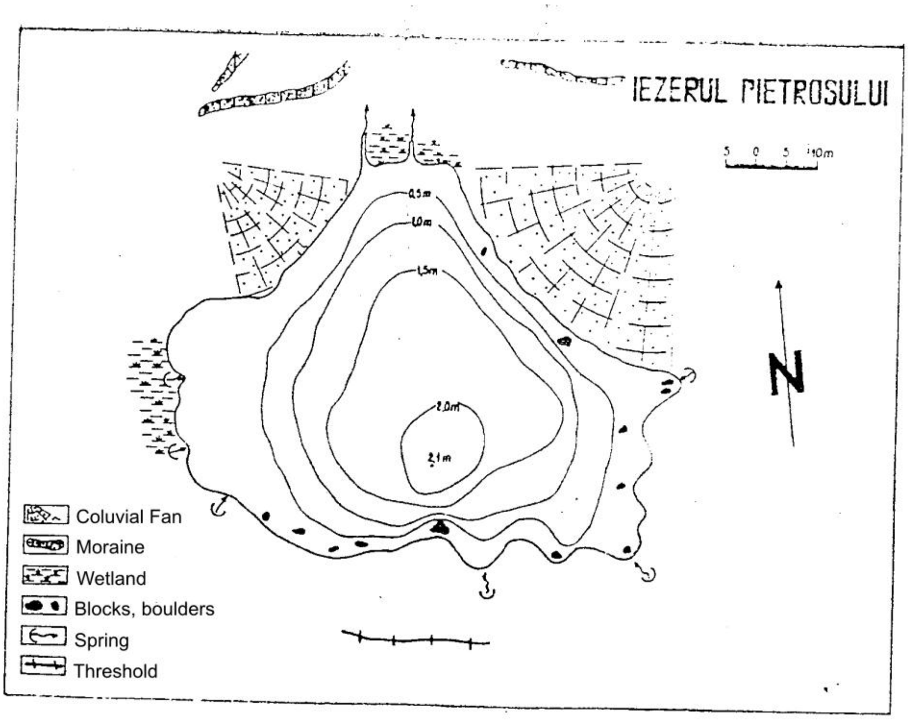


Figure 3.23: Pietrosul Lake bathymetric map (Pisota, 1968)

Figure 3.24: Pietrosul Lake and catchment looking northwards

Stiol Lake

Stiol Lake has a catchment area of 156 ha. While the original lake area was 0.06 ha (Pisota, 1967), as a result of artificial expansion the new lake area is 1.06 ha (Mindrescu, 2006) (Table 3.3b). The lake maximum depth is 5.0m. It has a catchment: lake ratio of 147.2. Although still relatively small, it is situated in one of the largest glacial cirques in the Romanian Carpathians, the Bistricioara Mare Cirque (Mîndrescu, 2006) (Figure 3.26). Being located at a relatively low altitude for a glacial lake, it lies en route a number of tourist paths. In 2002, the lake was illegally dammed. Consequently, the original glacial lake, of relatively small dimensions, has effectively been turned into a high altitude pond (Mindrescu *et al.,* 2010a). However, the original lake area was not disturbed by its enlargement. The geology is 56% paragneisses, 25% dolomites and 19% moraine while the land cover of the catchment is predominantly grass (60%) with 20% scrub, 14% scrub, 5% rock and 1% water (Figure 3.25).

Figure 3.25: Stiol Lake and catchment (2006 field trip) looking north towards the dam wall.

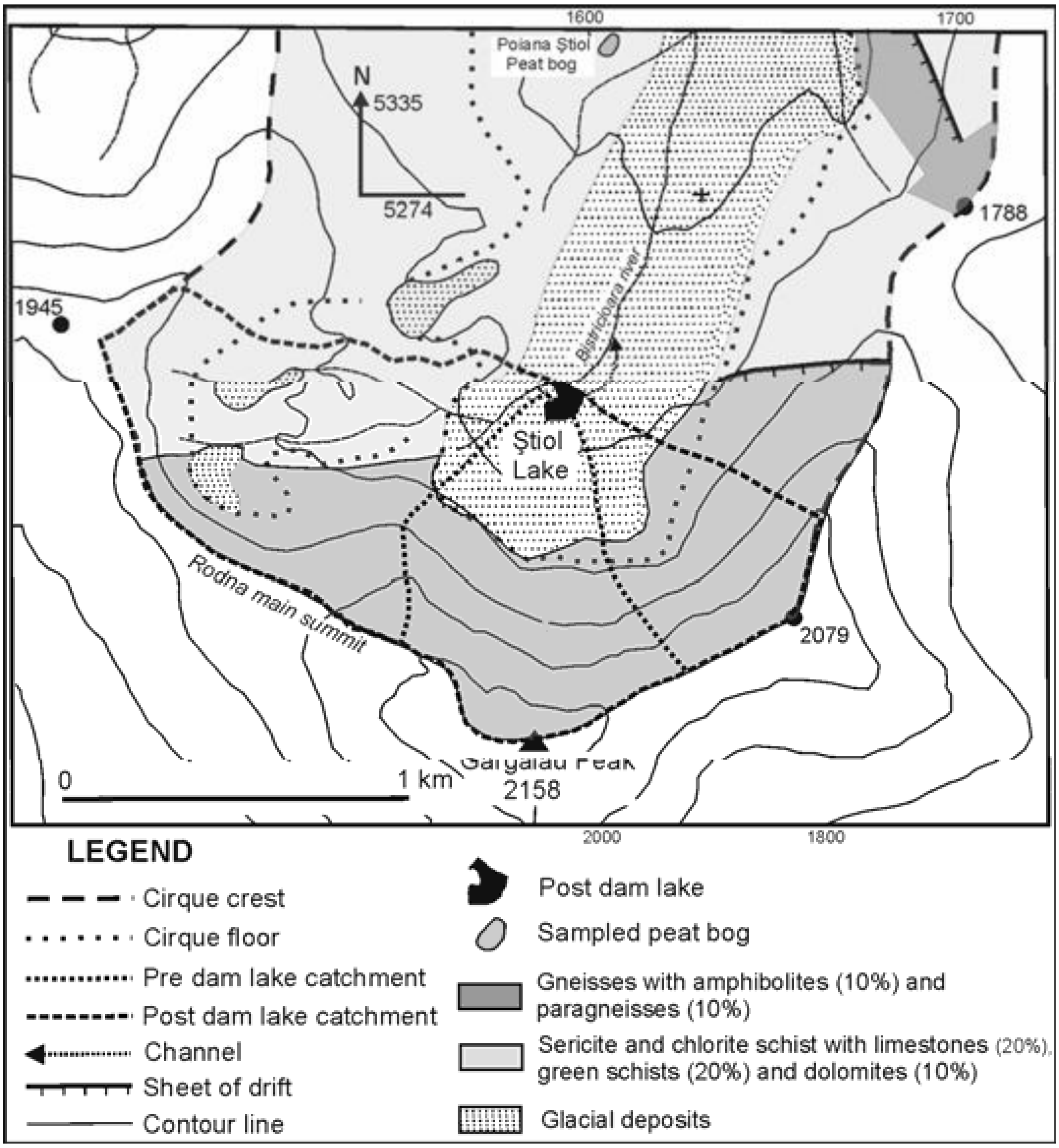


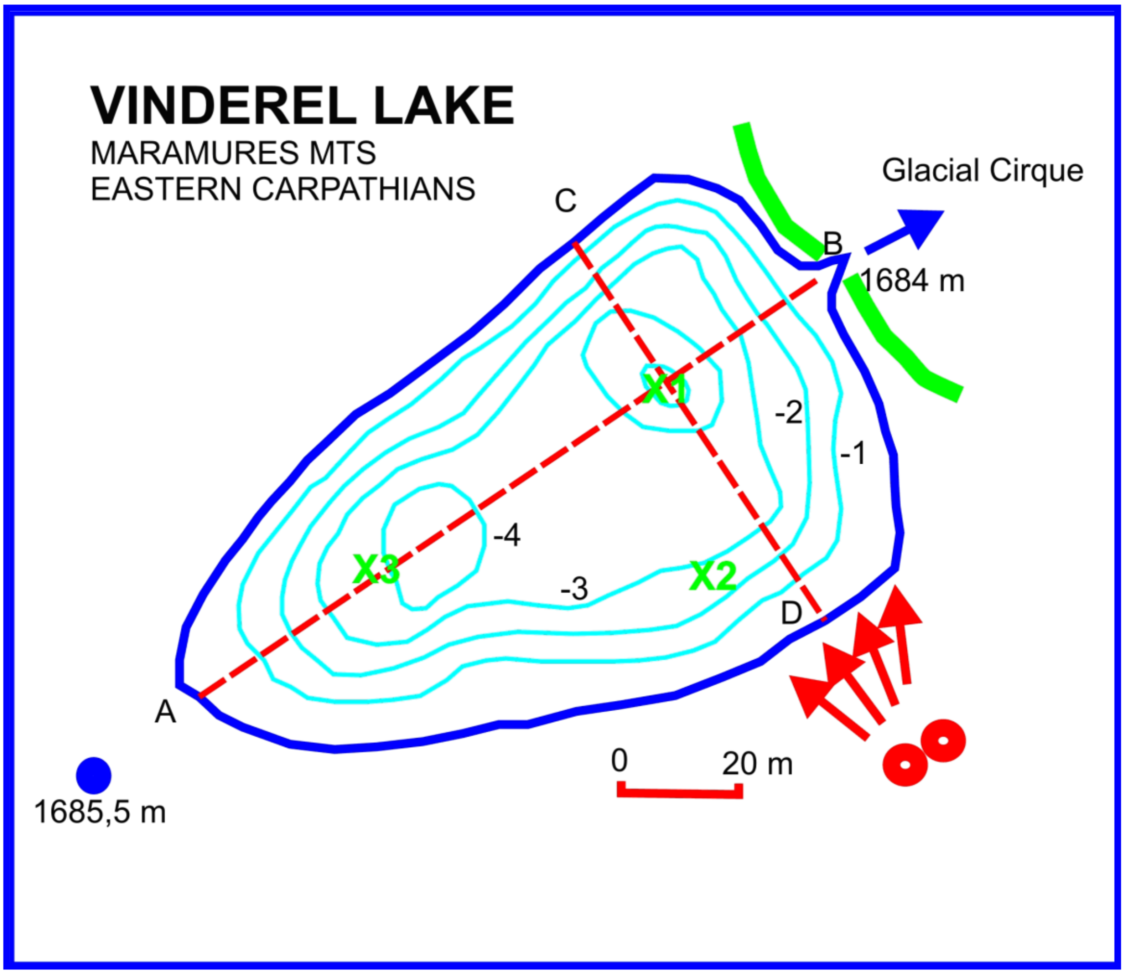
Figure 3.26 Stiol Lake Catchment: geomorphology and geology of the area (Source: Mîndrescu *et al.,* 2010b).

Vinderel Lake

Lake Vinderel is situated in the Maramures Mountains region between the two peaks of Farcau and Mihailecu in the Farcau Massif. This lake is the largest natural lake in the Eastern Carpathians (Mindrescu, 2001). It is located in a glaciated col (1684 m) (Figure 3.27). Vinderel Lake has a maximum depth of 5.5 m, a length of 140 m and a width of 80 m with an area 0.06 ha. It has a catchment: lake ratio of 85.0 (Table 3.3b). Only 18 glacial lakes from Southern Carpathians exceed Vinderel's surface area and only 23 the maximum depth (Mindrescu, 2001). Its geological formations consist of sandstone and siltstone, diabase intrusions. The land cover is 99% grass and 1% water (Figure 3.27).

Figure 3.27: Vinderel Lake and catchment (2008 field trip)

The vegetation cover of Vinderel lake catchment area is mainly grass. There is evidence of grazing pressure around the lake.



Key: Crest of the cirque's headwall

Banks of the lake

Profiles

Fan lake

Stream

Figure 3.28: Vinderel Lake bathymetric map (Mindrescu, personal communication)

3.4 Summary of the catchment and lake basin characteristics

All four lakes from Fagaras region were located in classic glacial cirques. There is no marked difference in the altitudinal positions of the lakes as all were located above 2000 m. However, the lowest lake elevation is at 2035 m (Balea) while the highest lake elevation is 2249 m (Capra). The six lakes from Rodna region are also all located within a cirque except Vinderel, which is located in a glacial col. The altitudinal positions of these lakes ranged from 1667 - 1840 m. The lowest is Stiol Lake (1671 m) while Bila Lake is the highest (1840 m). Thus it can be seen that even the lowest lake elevation in Fagaras region (2035 m) is higher than the highest lake elevation in Rodna region (1840 m).

There are distinct size variations in the catchment areas of the lakes in both the Fagaras and the Rodna areas. Podragu Mare lake (in the Fagaras region) has the largest catchment area of 55.2 ha followed by Balea lake (45.5 ha). Stiol lake (from Rodna) has the largest catchment area of 156 ha followed by Buhaiescu-3 lake (62.9 ha). Most of the lakes sampled in Rodna Mountains have larger catchment areas than their counterparts from the Fagaras Mountains (Tables 3.2 and 3.3) (Stiol Lake has the highest catchment area of 156 ha). The lakes in the Rodna Mountains have higher catchment: lake ratios than lakes in the Fagaras Mountains. The smallest of such ratios in Rodna Mountains is 23.0 (Lala Mare Lake) which is almost equal to the largest ratio 23.3 (Caltun Lake) in Fagaras Mountains. Buhaiescu-3 lake (Rodna region) has the largest catchment: lake ratio of 698.9.

The land cover of all four catchments from Fagaras region is predominantly grass except for Caltun where scree dominates (Table 3.2); while the catchment surface geology is predominantly moraine. In comparison with Fagaras region, the Rodna region’s land cover is predominantly grass and scree around the lakes (Tables 3.3a and 3.3b) while the geology formation of the catchment areas is predominantly mica chist and moraine deposits, except at Stiol Lake where gneiss dominates. Another exception to the geological formations above is Vinderel Lake catchment which consists of sandstone and siltstone with diabase intrusions.

The lakes from Fagaras region were generally deeper than the Rodna lakes with depth ranges from 8.6 - 16.5 m. Recovered sediment lengths were also greater with length ranges from 0.20 - 0.32 m (see Table 3.4). All the lakes from Rodna region were relatively shallow, ranging from 0.5 - 5.5 m. Recovered sediment lengths were also shallow (0.12 - 0.32 m). The variation in length of sediment cores might be partly due to the amount of material accumulated at the bottom of the lakes or due to the nature of the lake bottom substratum.

Table 3.4: Lake depths and sediment length summary for all sampled lakes.

Region	**Name of lake**	**Lake depth (max.) (m)**	**Regional mean (std. dev.)**	**Max sediment core length (m)**	**Regional mean (std. dev.)**
Fagaras	**Balea**	10.5	12.15 (3.4)	0.30	0.28 (0.06)
	Caltun	13.0		0.32	
	Capra	8.6		0.32	
	Podragu Mare	16.5		0.20	
Rodna	**Bila**	0.5	2.6 (2.2)	0.23	0.19 (0.08)
	Buhaiescu-3	0.5		0.12	
	Lala Mare	1.6		0.18	
	Pietrosul	2.3		0.12	
	Stiol	5.0		0.18	
	Vinderel	5.5		0.32	

CHAPTER 4: Methodology

4.1 Introduction

This chapter describes the field (sampling) and laboratory (analysis) approaches of this research. The field sampling aspect consists of site selection, coring and sample transportation to the laboratory. The laboratory section describes sequentially, the various analytical procedures employed in generating the data for this research. This includes: sediment bulk density measurements, loss-on-ignition, particle size analysis, mineral magnetic measurements, and geochemical analysis and in the case of one core radiometric dating. The physical characteristics of the lake sediments were determined with a view to determine if further analyses were necessary. Further analysis was made to assess a mineral magnetic measurement approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants. Another analysis was to determine the geochemistry of the sediments in order to assess the spatial and temporal variations in trace metal deposition.

The laboratory aspect of this research commenced working on the sediment samples already collected by Dr. S.M. Hutchinson from his field trips to Romania in 2006 and 2007. In summer 2008, we both made a further field trip to Romania and selected lakes were re-sampled to compare the results with the previous data. This also provided me with an opportunity to see at least a number of the field catchments (first hand) and to take sediment samples.

4.2 Field sampling

4.2.1 Site selection, coring and sample transport

Site selection was based on Pisota’s (1967, 1968 and 1971) surveys of the glacial lakes of various mountain areas in the Carpathians. They provide surveys of the characteristics of the studied lakes including a detailed bathymetry. Dr. Marcel Mindrescu of University of Suceava (Romania) acted as a guide to the lakes and provided additional local knowledge and logistical support. Certain criteria took pre-eminence while selecting the lakes for sampling. These included: size, depth and access. Care was taken to select lakes that were neither too small in size nor too shallow. In relatively small and shallow lakes there is an increased risk of sediment disturbance due to wind induced wave action and consequent turbulence. Accessibility of the lakes was a major factor in site selection due to the fact that the sampling gear and other required equipment had to be carried. Except for Lacul Balea the lakes were all located away from any road access.

Two regions in the Romania Carpathians were selected for a lake sediment-based study of key lakes. The North and South Carpathians (Rodna/Maramures and Fagaras regions respectively) were chosen for this study. The reason for studying two different regions was to be able to compare and contrast the situation in the two areas and to assess the extent of any regional variations; in terms of metals deposit into the lakes. The lakes in both areas are glacial lakes and all lie above the tree-line.

At all the sites lake sediment samples were taken using a gravity corer. The 37 sediment cores available for this thesis were collected using an HTH gravity corer (Renberg and Hansson 2008) (Figure 2.4) which was operated from an inflatable boat. With the aid of a depth sensor, most of the coring sites were located in the deepest part of each basin, in order to avoid steep parts of the lake bed and the possibility of sediment slumping. Multiple cores were taken from each lake. Details of the lengths of the cores are shown under Study area (Table 3.4). The replicability of the sampling technique can be demonstrated by the resampling of some of the previously sampled lakes (will be discussed under section 4.3.1).

After taken the sediment samples with the gravity corer, the core tube is removed from corer for extrusion of sediment. The tube is kept vertical in order not to disturb the sediment water interface. Once the core was disengaged from the corer, an extrusion was carried out to dislodge the water layer on top of the sediment. When fitted into the HTH extrusion system one 360^{o} turn gives a 5-mm thick sediment increment that is scraped off using a sectioning tray. In the field sectioning a 30 cm long core at 5mm interval took the team about 1 hour. Generally three or four persons are the optimum number for efficient sample extrusion and sectioning. Each core was sectioned into slices ranging from 0.25

cm to 1.0 cm and transferred into a pre-labelled PVC bags and transported to the Salford University Sediment Laboratory for analysis. All sediment samples were stored at 4^{o} C.

4.3 Laboratory analysis

4.3.1 Core/sample selection and analytical sequence

Three cores were taken from the deepest part of each lake at each year of sampling. Table 4.2 (Results) summarises core information for each lake. On the first field trip (summer 2006) all sediment samples were sliced into 1 cm sections. In 2007 and 2008, the resolution at which the sediment cores were sectioned was increased. An increased slicing resolution was made possible on these lakes during the last field trips with the availability of the HTH extrusion system. In each lake at least one key core from the summer 2007 trip was sectioned at an interval of 0.25 cm. The other cores from the lake were sectioned at 0.5cm intervals. All the lake sediment samples from the summer 2008 trip were sectioned at 0.25 cm. Based on field notes describing the position within the lake, the core length and any slicing deficiencies a key core from each lake was selected for laboratory analysis. Sediment sample slices were subdivided into subsamples (if enough) for subsequent laboratory analyses. The laboratory analyses consist of sediment density (wet and dry) measurements, loss-on-ignition, particle size analysis, mineral magnetic measurements and geochemical analysis. One core was submitted for radiometric dating. A full description of each of the above techniques can be found in subsequent subsections. The specific analyses undertaken on each key core is summarised in Table 4.1.

Table 4.1: Checklist of analysis undertaken on key cores

Region	Lake Name	Key core	Sample intervals(cm)	Analysis
North (Rodna/ Maramures Mountains)	Bila	LB 2	1	BD, LOI, PS, MM, GMA.
	Buhaiescu	LB-3 2	1	BD, LOI, PS, MM, GMA.
	Lala Mare	LLM 2	1	BD, LOI, PS, MM, GMA.
	Pietrosul	LP 1 (2006)	1	BD,LOI, MM, GMA.
		LP 1 (2008)	0.25	BD, LOI, PS, MM, GMA.
	Stiol	LS 2	1	BD, LOI, PS, MM, GMA.
	Vinderel	LV 3 (2006)	1	BD,LOI, MM, GMA.
		LV 1 (2008)	0.25	BD, LOI, PS, MM, GMA.
South (Fagaras Mountains)	Balea	LBa 1	0.5	BD,LOI, MM, GMA.
		LBa 4	0.25	BD, LOI, PS, MM, GMA.
	Caltun	LCt 2	0.5	BD, LOI, PS, MM, GMA.
	Capra	LCp 2	0.5	BD., LOI, MM, GMA, PS.
		LCp 3	0.25	BD, LOI, MM, GMA, RD.
	Podragu Mare	LPm 2	0.5	BD, LOI, PS, MM, GMA.

Key: BD= Bulk density, LOI: Loss-on-ignition, MM: Mineral magnetic measurements, PS: Particle size, GMA: Geochemical analysis, RD: Radiometric dating.

4.3.2 Bulk density measurements

For all lakes the sediment samples were stored in clearly labelled polyvinylchloride (PVC) bags in a refrigerator at a temperature of 4^{o}C prior to the bulk density analysis. From the key cores, using a pre-weighed 2 ml syringe, 1 ml of each slice was taken. The syringe and content (sediment sample) was then weighed on an electronic balance to allow the calculation of wet bulk density (gcm$^{-3)}$. Samples were weighed to 3 decimal places. The weighed sediment was transferred into a PVC bag. Both the weighed sediments and the residues were oven-dried for about 24 hours (in labelled PVC bags) at 40 degree Celsius (40 ^{o}C) to drive off as much as possible water, while avoiding thermally induced alteration of the magnetic mineral assemblage. At the completion of the drying stage, the sediment sample in each of the PVC bags was emptied into a pre-weighed disposable petri dish. The sediments were then weighed to permit the subsequent calculation of dry bulk density (dry mass per unit volume; gcm^{-3}).

4.3.3 Loss-on-ignition

Loss-on-ignition (LOI) was determined for one core per lake (key core) i.e sub- sampling of the same sediment core samples on which mineral magnetic and other analyses were performed (see Table 4.1), following the methods described by Bengtsson and Enell (1986). Loss-on-ignition (LOI) was determined at 1cm intervals. LOI is expressed as percentage of the weight of the dried sample and is used for estimating the organic matter content and help to characterize the sediment composition. The subsamples for LOI were taken from the previously oven-dried residues from the bulk density determination.

A batch of porcelain crucible (about 30 ml) was heated for 1 hour at 550^{o}C in a muffle furnace. The crucibles were allowed to cool down to room temperature in a desiccator and the weight of the crucible (A) was measured accurately on an electronic balance. About 0.2 g of sample was transferred to the crucible and immediately the weight of sample plus crucible (B) was measured. The crucible and content was placed in an air circulation oven at 105^{o}C and dry to constant weight overnight (for about 12 hours). The crucible and sample was allowed to cool to room temperature in a desiccator and the weight of the dry sample plus crucible (C) was measured. The crucible and the dry sample were placed in furnace for 2 hours at 550^{o}C. (In case the sample had a high organic content, a porcelain lid was placed on the crucible to prevent ash losses by violent burning of the sample). The crucible and the content were allowed to cool to room temperature in a desiccator. The weight of ash plus crucible (D) was measured (Bengtsson and Enell, 1986).

$$\text{Loss-on-ignition IG} = \frac{\text{C-D}}{\text{B-A}} \ \% \text{ DW}$$

4.3.4 Particle Size Analysis

The particle size analyser used was first of its kind at the University of Salford. Therefore, there is need to write step by step guide to sample preparation and analysis to make the laboratory procedures clear for incoming researchers. Particle size analysis was carried out on sub-samples from the key core from each lake at a 1 cm interval (see Table 4.1). The only exception was core Capra 3 (LCp 3). This core was set aside for radiometric dating. Particle size analysis (PSA) was not carried out on this core because there was insufficient sediment to do so. Consequently PSA was investigated using the second Capra core (LCp 2). The Partica LA-950V2 laser diffraction (chapter 2: Figure 2.2) was used to measure the particle size. Further information regarding sample pre-treatment for PSA can be found in Vaasma, (2008) and Horiba, (2009).

In order to determine the most appropriate sediment preparation and analytical technique to ensure repeatable results, five different sample pre treatments were assessed. The result of this trial analysis is set out below. The same weight of samples was taken from a common core section and analysed as follows:
(1) A dry sample (dry), (2) sample treatment with deionised water (wet), (3) by the addition of calgon only (C), (4) treated with hydrogen peroxide only (H) and (5) treated with hydrogen peroxide followed by few drops of calgon (H+C). All samples were subjected to the same analytical process. Each sample was allowed to circulate for 3 minutes in the instrument before taking any measurement. The importance of the 3 minutes circulation period was to ensure the equipment mixed and dispersed the sample

thoroughly. Three measurements were taken of each sample at 1 minute interval. Table 4.2 shows the D10 and D90 values for the various treatments.

Table 4.2: Trial particle size analysis (µm)

	1. Dry	2. Wet	3. C only	4. H only	5. H+C
D10	9.0848	10.9967	10.5159	9.6644	9.3306
D90	112.0445	65.9911	99.7903	65.4108	50.2382

In the first three treatments (dry, wet and calgon only), there was no removal of organic matter from the samples (i.e. no digestion). For the dry treatment, a sample of it was introduced into the analyser directly. The fine particles were partially suspended or clung to the upper part of the reservoir during analysis. The result shows a size range of 9.0848 to 112.0445 µm at D10 and D90 respectively. The upper limit was relatively coarse as the organic materials binding the particles together were not removed. For the wet treatment, the sediment was moistened in a clean beaker by the addition of deionised water before introducing it to the particle size analyser. The wet treatment resulted in a size range of 10.9967 to 65.9911 µm at D10 and D90, respectively. The size range was probably finer than the dry treatment because soaking the sediment in water has caused the particles to be more disaggregated. For the calgon only treatment, the sediment sample was moistened with calgon solution before introducing it to the analyser. The size ranged between 10.5159 to 99.7903 µm at D10 and D90, respectively, and similar to dry treatment. For hydrogen peroxide only and hydrogen peroxide plus calgon treatments, the samples were both digested in hydrogen peroxide before calgon solution was added to the last treatment in order to chemically disperse the sediments. The hydrogen peroxide oxidises the organic remains in the sediments. Treatment with hydrogen peroxide only showed a result that was similar to addition of water only. This suggested that the core sample selected was not highly organic. The treatment with H+C showed the size range from 9.3306 to 50.2382 µm at D10 and D90 respectively. It was therefore apparent that some pre-treatment of samples is required. H+C process was chosen.

The Horiba LA-950V2 (Figure 2.2) was linked to a PC. Before measurement commenced, the system, measurement, sample conditions and calculation functions were checked. A blank measurement was made for every sample to measure background conditions in order to confirm the degree of purity of the water used (from a stored source). Pure water (Option-Q labelled water) was re-circulated to rinse the equipment between samples. The blank measurement was subtracted from sample measurement to produce a scattering value excluding any background data. Sediment samples were pre-treated with 30% hydrogen peroxide (partial digestion) for the removal of organic matter or carbonates before carrying out the particle size distribution (PSD) analyses. 0.2 g of sample was weighed (to 3 decimal places) into a beaker. 30% hydrogen peroxide was added and left for 18 hours. The purpose of weighing the samples was to ensure that same amount of sediment material was treated for analysis. A few drops of calgon was added to the content of the beaker and stirred before introducing it to the Horiba LA-950V2 for analysis.

Each sample was allowed to circulate for 3 minutes before taking any measurement. Three measurements were taking for every sample at 1 minute interval. When the measurement is completed, the dedicated desktop displays the distribution graph, data table, measurement data and classification result data. The classification result data consists of surface area, mean size, variance, median size, mode size, standard deviation, geometric mean size.

4.3.5 Mineral magnetic measurements

After oven drying at 40°C, the sediment samples from each key core were carefully disaggregated and tightly packed into clean pre-weighed, polystyrene air tight (10 ml) container with cling film at 1 cm interval (when a core was sliced at 0.25 cm, all the four slices were separately packed before being loaded into the same pot). This was reweighed to permit the calculation of mass. The purpose of the cling film is to ensure the complete immobilisation of the samples which is essential for its accurate characterisations when passing through the analytical stages.

A review of the standard magnetic parameters used in this study can be found in Thompson and Oldfield (1986), Dearing (1999), Walden *et al.* (1999), Maher and Thompson (1999) and Evans and Heller (2003). Mass-specific magnetic susceptibilities

(X) were measured using Bartington MS2 System in the University of Salford's Sediment Laboratory. The laboratory-induced remanences investigated were anhysteretic remanent magnetisation (ARM) and saturation isothermal remanent magnetisation (SIRM). All remanences were measured using a minispin magnetometer at the University of Manchester's Geography Laboratory.

4.3.5.1 Mass-specific magnetic susceptibility (χ)

Mass specific magnetic susceptibility (χ) measurements were made by inserting a packed sample into a dual frequency susceptibility sensor (air-cored bridge system) connected to a digital meter (Bartington MS2 System). Calibration of the sensor was achieved using a calibration pot supplied with the Bartington Instrument. The oscillator circuit within the sensor generates a low intensity alternating magnetic field (0.1 mT). The introduction of the sample within the influence of this field causes a change in oscillator frequency, which is transferred to the meter, converted to a magnetic susceptibility and displayed in digital form. This procedure was followed for all samples at both low (χLF) and high frequency (χHF). This allows the calculation of frequency-dependent susceptibility (χfd). Frequency-dependent susceptibility was calculated from the difference in susceptibilities at low and high frequency measurements (see Dearing, 1999 for more details).

For both low and high magnetic susceptibility, measurements were repeated three times and an average value for each parameter recorded. Magnetic susceptibility measurements are simple to perform but the equipment is sensitive to background noise (Dearing, 1999).

4.3.5.2 Anhysteretic Remanent Magnetisation (ARM)

ARM was generated in the sediment samples by placing them in alternating field of 0-100 mT, which was steadily increased and then decreased and upon which a direct current field of 0.04 mT was superimposed. The 0.04 mT field is approximately equal to the Earth's magnetic field. The two magnetic fields were generated using a specially adapted Molspin AF demagnetiser. The sample was then transferred to a Minispin fluxgate magnetometer where its magnetic remanence was measured.

4.3.5.3 Saturation Isothermal Remanent Magnetisation (SIRM)

After the ARM measurement, the samples were subjected to a strong forward magnetic field 1000 mT (magnetically saturated), followed by reverse magnetic field of -20 mT, -40 mT, -100 mT and -300 mT. The required magnetic fields were generated using a Molspin pulse discharge magnetiser. After each step, the sample was quickly transferred from the pulse magnetiser, having received a pre-set pulse magnetic field, to a Minispin fluxgate magnetometer where its magnetic remanence at that stage was measured and recorded. Each magnetic field was generated by current-carrying coils using a pulse discharge system, which produces a uniform, repeatable short-duration field. This procedure was used to derive the SIRM of the samples and other parameters and ratio from the stepwise demagnetisation of their SIRM.

A minispin fluxgate magnetometer was used to measure the magnetic remanence of samples at each stage through the measurement sequence. The instrument was regularly and accurately calibrated against a known and stable standard. In simple terms, the magnetometer measures the magnetic field surrounding a sample using fluxgate detector and employs the principle that the strength of this field is proportional to the remanent magnetisation of the sample itself (Hutchinson, 1990; Dearing, 1999).

4.3.6 Geochemical analysis

Both the microwave system that was used for the digestion of the sediments and the ICP equipment used for the analysis were first of their kind at the University of Salford. Therefore, this section of the thesis attempts to give a summarised step by step guide to the laboratory procedures for carrying out sediment chemistry and hence constitute a quick guide for prospective researchers/students.

4.3.6.1 Sample digestion procedure

The analytical vessels were cleaned using a standard scientific dish washer (Miele Professional). The digestion vessel consists of a tube, a bung and a cap. The vessels allow for expansion of gases during digestion. Samples were oven dried overnight at 105 ^{o}C to remove the moisture. 0.2 g of the oven dried samples was weighed (to 4 decimal places) directly into each pre-cleaned vessel. 10 ml of concentrated HNO_3 (analytical grade) acid was added. In each batch of samples, a 'blank' and a 'reference' were included. They were processed in the same manner as the lake sediment samples but included no sample. The reference is certified reference materials (CRM).

A CEM MarsXpress microwave digestion system equipped with temperature and pressure regulation (through a sensor vessel) was used. When the instrument is switched on, a series of screens appear in succession and end in the Method main menu as shown below (Figure 4.1). From the main menu, the set-up key was pressed. A MarsXpress method was selected on the main screen of the instrument. The screen shows the following display (Figure 4.2).

CEM Method Menu

Edit/Create Method   Load Method

'Start' Current Method:

XXXXXXXX

Figure 4.1: Main menu of CEM MarsXpress microwave

Mars Setup Menu

Delete Method	Comm. Port Setup
System Variables	Information
Select Sensor	System History
Printer Setup	Select Language
Duotemp Setup	

Figure 4.2: Selection of digestion method from the main menu

With the aid of the operation manual and by following the instructions displayed on the screen appropriate temperature, pressure and timings can be selected and saved on the system with a method name. This allows the same set of parameters to be used for the whole series of the sample batches. The microwave parameters used for the digestion of the sediment samples in this study are as follows (Table 4.3) and were named Sediment General Method.

Table 4.3: Microwave oven digestion parameters

Parameter	**Quantity**
Power	1600 watts
Temperature	160 °C
Hold time	15 minutes
Cooling time	45 minutes

The next step was to ensure the microwave vessels were tightly sealed. The vessels were stacked in the microwave carousel in an even pattern, starting with the inner locations first. Then, the carousel was placed inside the microwave digestion system. Care was taken to make sure each vessel was placed inside a sleeve and that the composite sleeves were fully inserted into the receptacles of the turntable. The turntable key was pressed on the instrument to rotate the turntable drive lug so that the flat on the drive lug is parallel with the front of the cavity. Finally, the door of the instrument was closed and the method commenced. The microwave was also programmed to monitor the cooling down sequence for safety reasons.

After cooling, the digested material was filtered through ashless 2-3 µm slow speed filter cups to remove the silica content. The filtration was undertaken using a LabXpress Assembly consisting of turntable assembly; funnel and diaphragm pump assembly (60 Hz) into pre-washed 25 ml volumetric flasks. The filtrate in the 25 ml volumetric flasks was then diluted to mark with Option-Q labelled water. The Option-Q labelled water has the following qualities: in organics at 25 °C up to 18.2 MΩ-cm and Total Organic Carbon of 1–3 ppb. The filtrate obtained from each of the sediment samples was transferred into a 30 ml polyethylene bottle. The digests were stored at 4 °C until analysed.

4.3.6.2 Elemental Analysis using Varian 700-ICP-OES (Inductively Coupled Plasma Optical Emission Spectrometer)

5 ml of the digests / filtrates which were stored at 4 °C were pipetted into prewashed 25 ml volumetric flask and made up to the mark with Option -Q water. A subsample was transferred to a pre-labelled Falcon centrifuge tube. The tubes were capped and stored in a rack fitting the autosampler at 4 ^{o}C, ready for ICP-OES analysis.

In the analytical procedure the auto sampler takes up the sample and introduces it to the nebulizer through a peristaltic pump. The peristaltic pump ensures a constant flow of liquid, regardless of differences in viscosity between samples, standards, and blanks (Thomas, 2001; Gaines, 2010). The ICP-OES uses a nebulizer for converting a liquid into a fine spray that uses a gas as the driving force. It consists of a spray chamber that can allow only small droplets produced by the nebulizer to enter into the plasma. The chamber's material of construction (glass or polymers) as well as the sample matrix and the chemistry of the element will influence the drainage characteristics/washout time. Nevertheless, it is recommended that the drainage process is smooth and continuous.

More details on the basic principles of ICP can be found in Thomas (2001) and Gaines (2010).

The ICP-OES is switched on permanently. From the desk top pc dedicated to the equipment, clicking on the ICP-Expert II Software icon displays the overview of all instrument parameters (Figure 4.3). This allows the functionality of each of the components of the instrument to be checked before proceeding to the next stage of the analysis (creating a worksheet or a file).

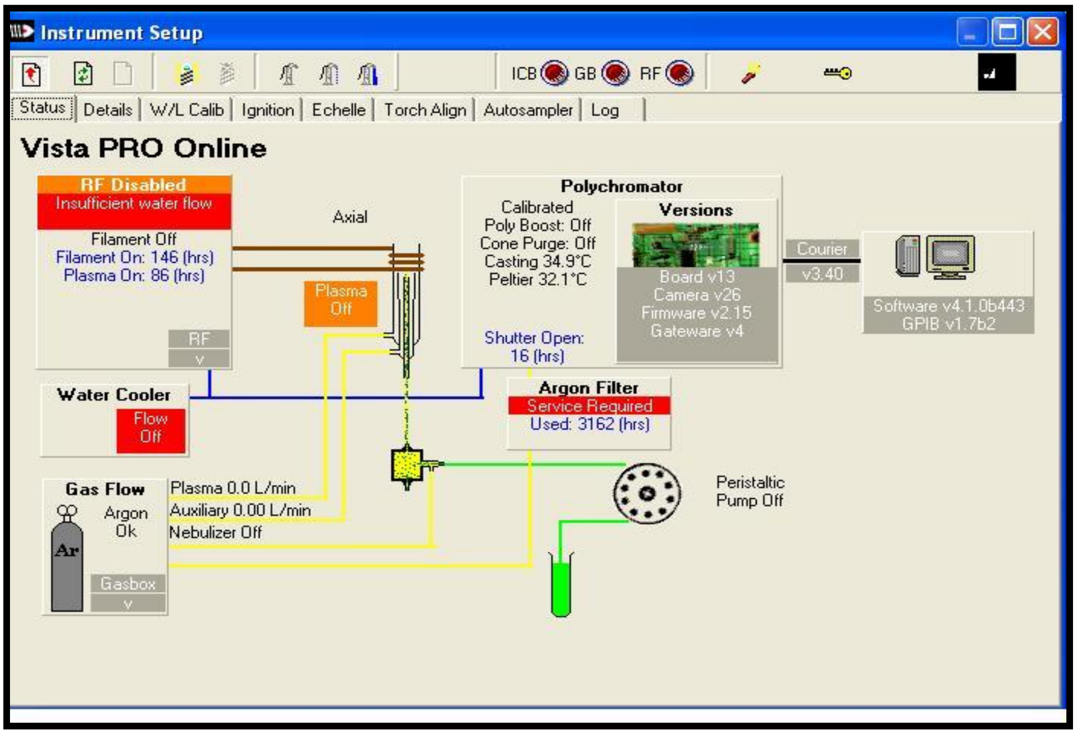


Figure 4.3: Status page of Varian 700-ICP-OES

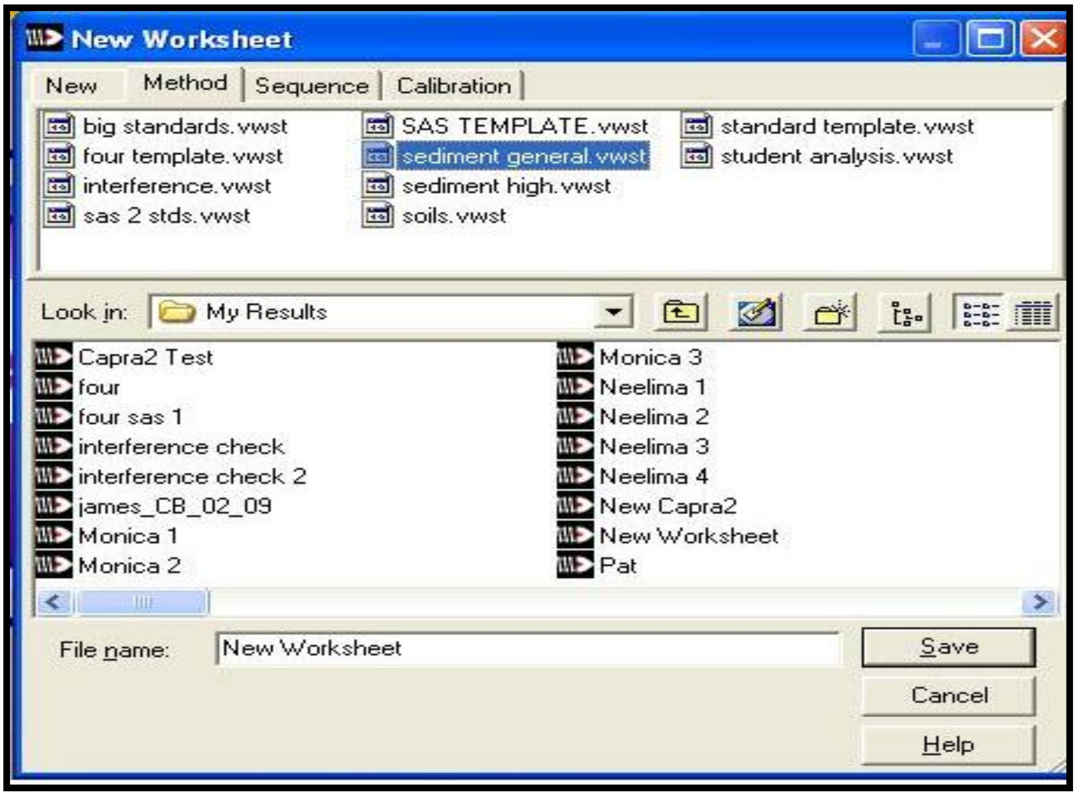


Figure 4.4: Creating a new worksheet from existing template on the Varian 700-ICP-OES

After checking the working condition of the components, such as the pump and the auto-sampler, a new worksheet was created by clicking on ‘new’. A new worksheet can equally be created from an existing template (Figure 4.4). The worksheet was given a new data filename which was confirmed by clicking ‘okay’. This was then saved.

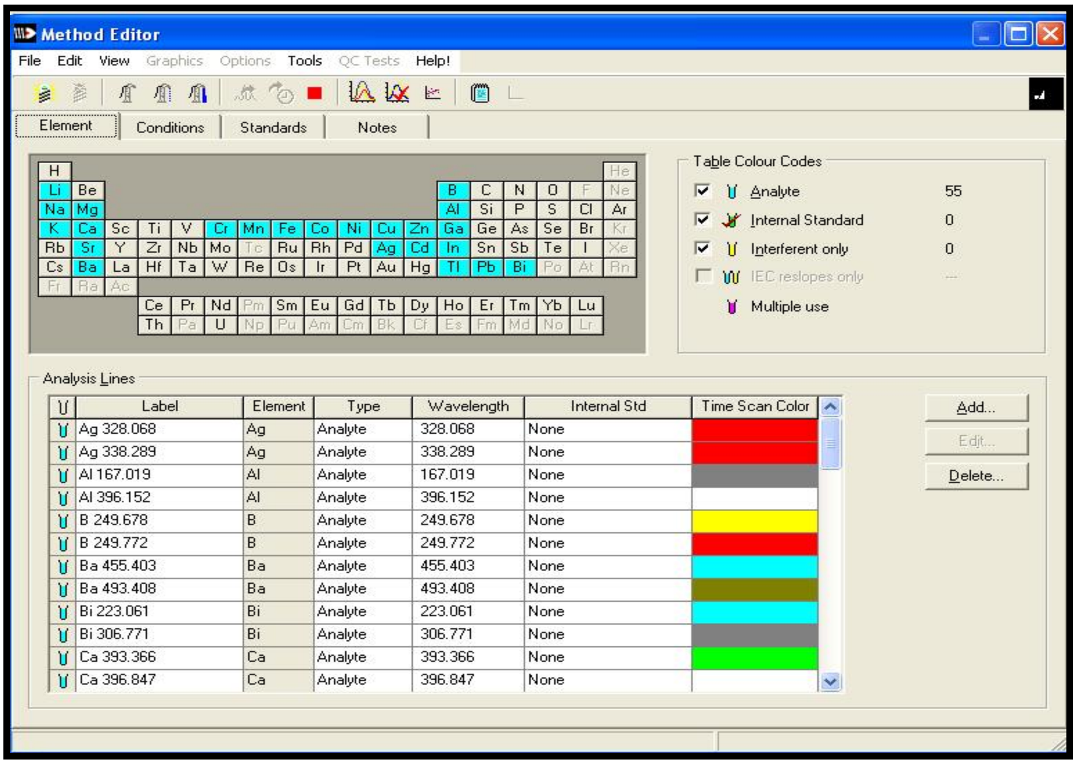


Figure 4.5: Selection of elements to be measured from the periodic table (Varian 700-ICP-OES)

The next task was to choose the elements to be measured from the periodic table. This was done via the Method Editor by clicking on the selected elements from the library (Figure 4.5). The library contains all the basic information about every element. Both atom and ion lines were used. Emission lines lying close to each other were avoided as this could give rise to spectral interferences. Choosing the required elements led to a table showing all the chosen analytical lines as shown above. Then ‘conditions’ was selected from the menu bar.

This allows the entering of the required parameters for the analysis like the pump flow, nebulizer flow fast pump, rinse time, etc. This was followed by the selection of element concentrations. Before the analysis of the sediment digests, a multi-element standard solution was used to calibrate the instrument. The standards range from 0.5ppm to 100ppm. A blank solution tube consisting of Option-Q water was included in the standards. The analytical standard used to calibrate the instrument includes: Ag, Al, B, Ba, Bi, Ca, Cd, Co, Cr, Cu, Fe, Ga, ln, K, Li, Mg, Mn, Na, Ni, Pb, Sr, Ti and Zn. The standard was supplied by Alfa Chemicals UK.

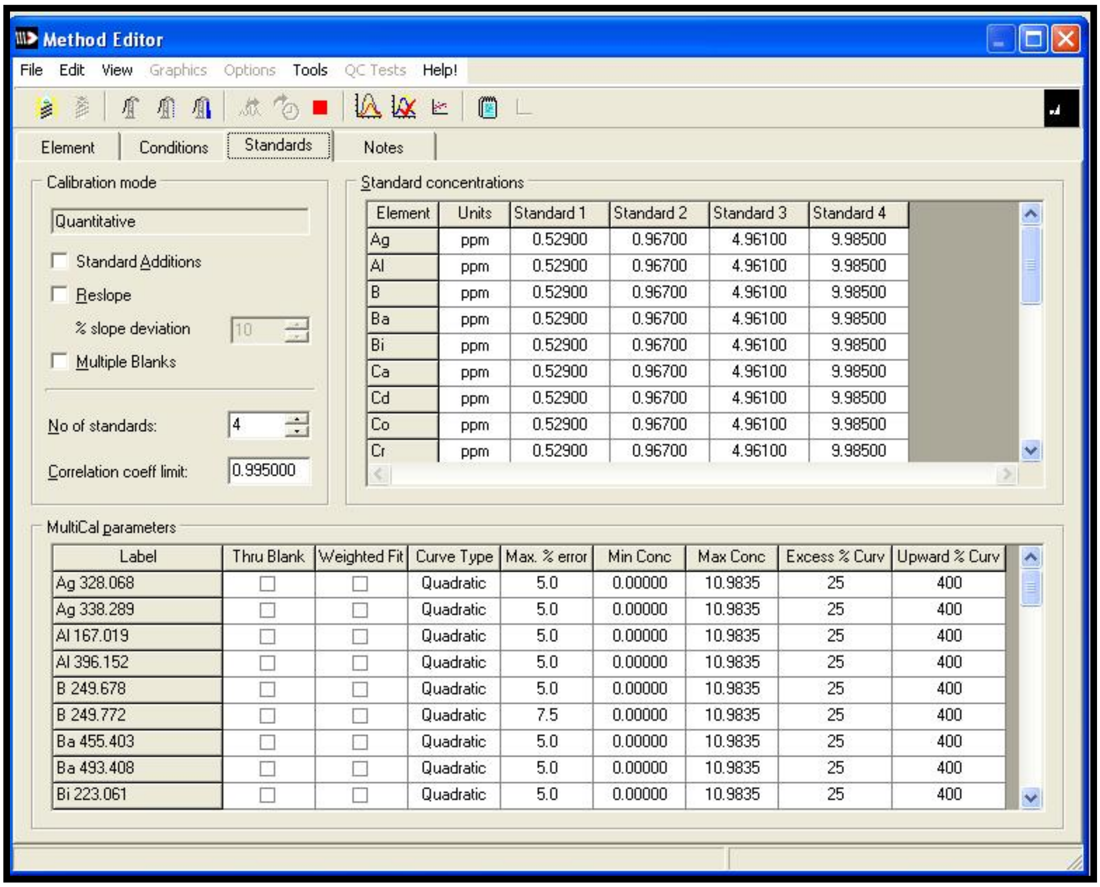


Figure 4.6: Selection of analytical parameters I (Varian 700-ICP-OES)

By clicking on the ‘standards’ from the menu bar, element concentrations were chosen in ppm. Using the ‘counter’ button the number of standards was chosen (Figure 4.6). A file was saved and the worksheet was later updated with changes.

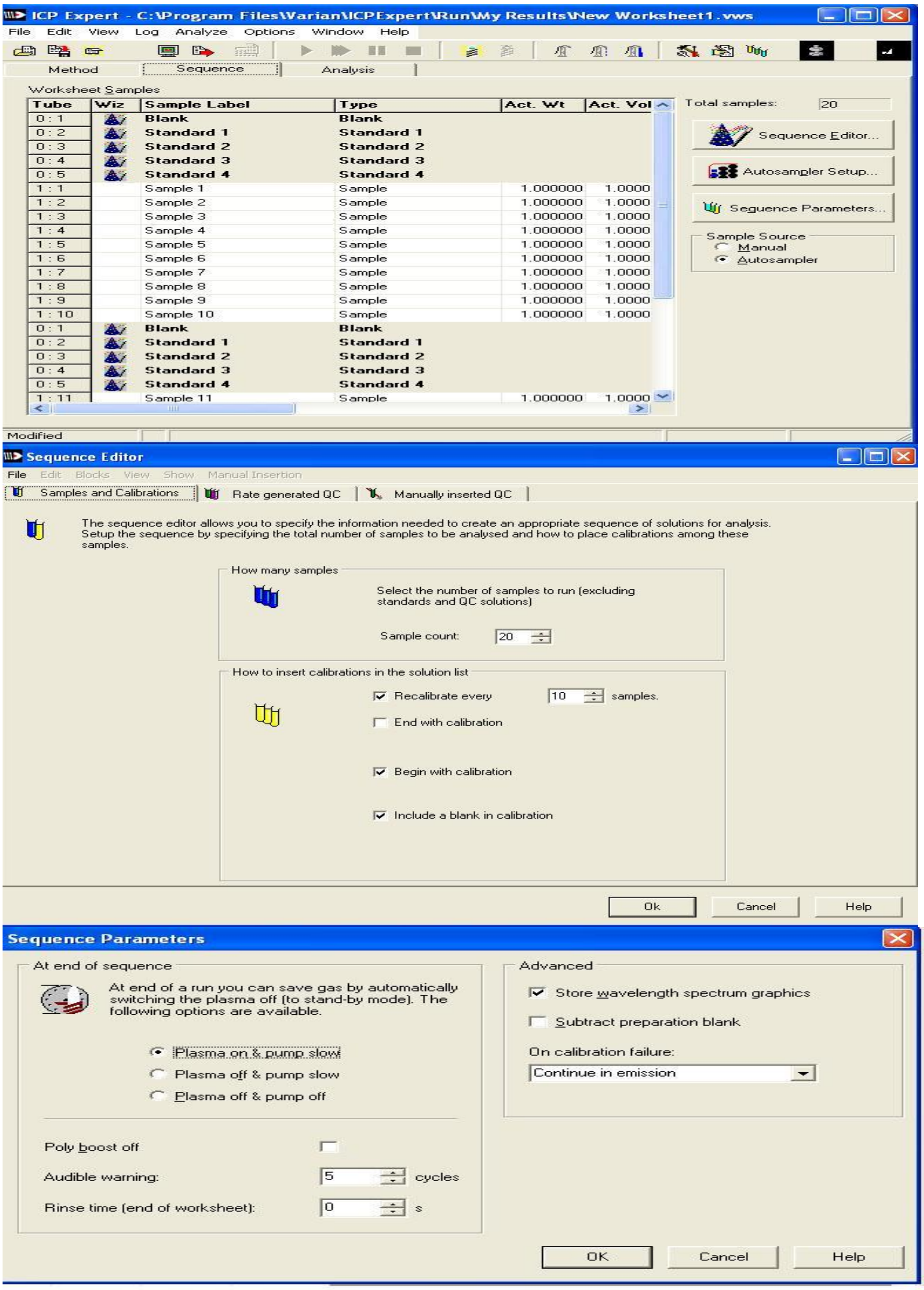


Figure 4.7: Selection of analytical parameters II (Varian 700-ICP-OES)

As shown above, by clicking on ‘sequence’, the details of the instrument, auto-sampler and samples were inputted (Figure 4.7). The argon gas pressure was switched on and the argon purge delay was completed within few minutes. The peristaltic pump tubing was clamped in place with open end inserted into deionised water. All solutions for analysis were highlighted in yellow (Figure 4.8). The extraction was checked for adequacy and then the plasma was ignited. The system was allowed to stabilise before proceeding on analysis by clicking on the green arrow.

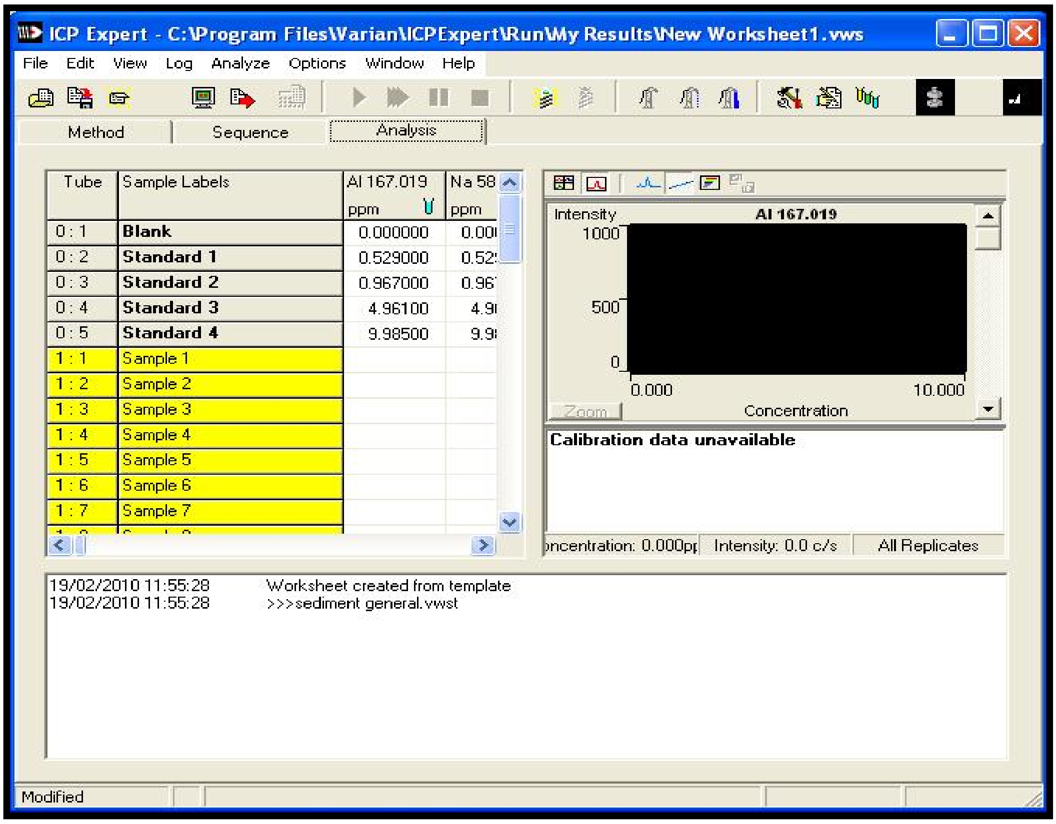


Figure 4.8: Generating results with Varian 700-ICP-OES

4.3.6.3 Validation of Elemental Analysis using Varian 700-ICP-OES method

This subsection discusses the steps taken to ensure that the data generated by ICP-OES analysis was both precise and accurate. This requires consideration of both the procedures used to prepare the sample for analysis and the setting up of the instrument. The functioning of an ICP-OES instrument will depend on the operating conditions and analytical parameters. The instrument's operating conditions such as the pump flow rate, nebulizer flow rate, pump, rinse time and sample uptake time were optimized (Table 4.4). For example, the rinse time of 10 s allowed the instrument to be thoroughly cleaned between sample uptake and 3 s replicate read time allowed each sample to be precisely measured. In the earlier part of this chapter the selection of analytical parameters (Varian 700-ICP-OES) has been described (Figures 4.7 and 4.8).

Table 4.4: ICP-OES instrumental parameters

Parameter	**Setting**
Power	1.25 kW
Plasma gas flow rate	15.0 Lmn^{-1}
Auxiliary gas low rate	1.5 Lmn^{-1}
Replicate read time	3 s
Sample uptake delay	35 s
Pump rate	12 s
Rinse time	10 s
Replicate	3

Research shows that measurement accuracy is best established through the analysis of a certified reference material (CRM) (Gaines, 2010). The reliability (accuracy) of the analytical method employed was evaluated by means of certified reference material LKSD-4 which is lake sediment. The CRM was subjected to the same digestion and analytical procedures as all the lake sediment samples. Table 4.5 summarises the certified and the measured values of a range of elements. The average recovery of each element (as a percentage) has also been calculated. Accuracy of measurement defines the closeness of the result of a measured value and a true value.

Although the recovery of Fe is relatively low, most elements the percentage recovery is within the range commonly reported in the literature (e.g. Melaku *et al.*, 2005). However it should also be noted that the certified values quoted will be total concentrations derived via a range of techniques (as specified in the documentations provided the CRM). In this study the digestion technique employed is effectively a partial digestion and so discrepancies between certified and measured values were anticipated.

Table 4.5: Percentage Recoveries of CRM (%)

Metal	wavelength	Certified Value	Measured Value	Average (%)
Co	228	11	10.5	96.8
Cr	267	21	17.3	82.4
Cu	324	30	29.6	98.7
Fe	234	2.7	2.2	80.4
Mn	257	430	374.4	87.1
Ni	231	32	30.8	96.3
Pb	283	93	94.4	101.5
Zn	213	189	171.2	90.6

Table 4.6 also demonstrates that appropriate steps were taken to ensure the quality of the data derived by ICP analysis. This table reports the mean concentrations (and the percentage of values under range) for 9 elements in the blank samples that were measured with each batch of sediment samples. It can clearly be seen that the pre-cleaning of all containers, the use of analytical grade acid and Option Q quality water has ensured that background levels of all elements have remained very low and that there is therefore little chance that sediment samples have experienced any contamination during their preparation for analysis.

Table 4.6: Blank HNO_3 measurement (% under range): values are in μg

Al	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn
0.046	-0.028	-0.005	-0.011	0.026	-0.004	-0.016	0.004	0.007
0.005	0.011	0.000	-0.002	0.018	0.018	0.008	0.006	-0.005
0.025	-0.011	-0.004	-0.015	0.018	-0.005	-0.010	-0.001	-0.002
0.019	-0.010	-0.007	-0.004	0.021	0.002	-0.005	0.001	0.004
-0.016	0.001	-0.013	-0.006	0.026	-0.017	0.003	-0.002	0.000
20%	60%	100%	100%	0%	60%	60%	60%	60%

4.3.7 Dating

The equipment for lake sediment dating was not available at the University of Salford sediment laboratory. Due to the high cost of lake sediment dating a single core LCp 3 from Capra Lake was sent out to be dated at the Environmental Change Research Centre, University College London. Core LCp 3 from Capra Lake was dated by gamma assay at the Environmental Change Research Centre, University College London. Sediment samples were analysed for ^{210}Pb, ^{226}Ra, ^{137}Cs and ^{241}Am by direct gamma assay in the Bloomsbury Environmental Isotope Facility (BEIF) using an ORTEC HPGe GWL series well-type coaxial low background intrinsic germanium detector (Appleby *et al*., 1986). Lead-210 was determined via its gamma emissions at 46.5 keV, and ^{226}Ra by the 295 keV and 352 keV gamma rays emitted by its daughter isotope ^{214}Pb following 3 weeks storage in sealed containers to allow radioactive equilibration. Lead-210 (half-life 22.3 years) is a naturally produced radionuclide, derived from atmospheric fallout (termed unsupported ^{210}Pb). Cesium-137 and ^{241}Am, were measured by their emissions at 662 keV and 59.5 keV. Cesium-137 (half-life 30 years) and ^{241}Am are artificially produced radionuclides, introduced to the study area by atmospheric fallout from nuclear weapons testing and nuclear reactor accidents around 1963. The absolute efficiencies of the detector were determined using calibrated sources and sediment samples of known activity. Corrections were made for the effect of self absorption of low energy gamma rays within the sample.

4.4 Summary

The methodology chapter described the sampling and analysis procedures of this research. The field sampling aspect consisted of site selection, coring and sample transportation to the laboratory. For the purposes of comparison research lakes were selected from two areas of the Romanian Carparthians. These are the Fagaras region (south) and the Rodna/Maramures regions (north). The site selection of the two areas was guided by previous work and local knowledge. The laboratory section described sequentially, the various analytical procedures employed in generating the data. This included: sediment bulk density measurements, loss-on-ignition, particle size analysis, mineral magnetic measurements and geochemical analysis (ICP-OES). A single core from one of the south lakes (Lacul Capra core LCp 3) was radiometrically dated (^{210}Pb). The laboratory techniques and procedures were validated to ensure accuracy and precision of data generated.

CHAPTER 5: Results

5.1 Introduction

This results chapter presents data generated in this study. The data generated include sediment density, loss-on-ignition, particle size determination (i.e. physical characteristics), mineral magnetic measurements, geochemical analysis and radiometric dating. The data on physical characteristics, mineral magnetic measurements and geochemical analysis were all generated in the University of Salford Sediment Laboratory. The instrumentation for lake sediment dating was not available at the University of Salford Sediment Laboratory. Due to the high cost of lake sediment dating a single core (LCp 3) from Capra Lake was sent out to be analysed at the Environmental Change Research Centre, University College London.

5.2 Physical characteristics of the lake sediments

This section describes the physical characteristics of the lakes sediments; first, from the Fagaras region followed by sites in the Rodna region. The physical characteristics of the sediments described include: wet and dry density, loss on ignition and particle size. It has been mentioned in an earlier chapter of this thesis that the reason for studying two different regions was to be able to compare and contrast the situation in the two areas and to assess the extent of any regional variations.

5.2.1 Density characteristics of lake sediments in the Fagaras region

Three core samples were taken from each lake sampled in the Fagaras region in the 2007 sampling trip. Density measurements were carried out on one core each for Caltun and Podragu Mare lakes while density measurements were carried out on two cores from Balea and Capra lakes. Within each core, the wet density profiles were clearly similar to and mirrored sediment dry bulk density. There are two versions of the profiles (Figures 5.1a and 5.1b), in Figure 5.1a the cores were all plotted to the same scale on all the axes while in Figure 5.1b the density axes were plotted to the maximum for each core in order to show any density fluctuations more clearly. All lakes demonstrated a decrease in density towards the surface (see Figure 5.1). The mean sediment dry density values of cores from the lakes in Fagaras region varied from 0.27 g cm^{-3} (Caltun Lake) to 0.83 g cm^{-3} (Balea Lake, LBa 4). The minimum dry density value was observed in Capra Lake (0.06 g cm^{-3}) while the maximum was observed in Balea Lake, LBa 4 (1.62 g cm^{-3}) (Table 5.1).

Table: 5.1 Mean and standard deviations (Stdev) of Fagaras lakes' sediment dry density (g cm^{-3})

Name	**Balea Lake**		**Caltun Lake**	**Capra Lake**		**Podragu Mare Lake**
	LBa 1	**LBa 4**	**LCt 2**	**LCp 2**	**LCp 3**	**LPm 2**
Mean	0.70	0.83	0.27	0.42	0.51	0.83
Stdev	0.20	0.32	0.08	0.12	0.16	0.24
Range	0.37-1.25	0.34-1.62	0.15-0.53	0.06-0.58	0.11-0.75	0.29-1.26

Balea Lake

The profiles of both cores LBa 1 and LBa 4 analysed for this lake are to some extent similar. A number of peaks and troughs were observed down the profile of Balea lake core 1 (LBa 1). In LBa 1 troughs were observed between the depths of 19 - 15 cm and 10 - 6 cm. The core peaked at 15 cm, 10 cm and 6 cm. LBa 1 demonstrated a surface decrease from the depth of 2 cm. The core (LBa 1) has a mean dry density value of 0.70 g cm^{-3} and a dry density value range of 0.37 - 1.25 g cm^{-3} (Table 5.1). From the bottom of Balea Lake core 4 (LBa 4-key core) all through to the surface shows series of fluctuations in sediment density. An increase in density was observed at the depths of 29 - 26 cm. There were also increases in density at the depths of 23 - 18 cm. These peaks were also alternated with troughs. Such peak and trough was also observed between the depths of 10 - 6 cm. The alternating peaks and troughs observed in LBa 4 might demonstrate an in wash of minerogenic materials. The core demonstrated a surface decrease from the depth of 2 cm (Figures 5.1a and 5.1b). The core (LBa 4) has a mean dry density value of 0.83 g cm^{-3} and a dry density value range of 0.34 - 1.62 g cm^{-3} (Table 5.1). The features between the depths of 19 - 15 cm and 10 - 6 cm were similar in both cores. Statistically, there was no significant correlation in the density of both cores; at 0.01 levels the correlation was 0.292.

Caltun Lake

A close examination of the profile (Caltun lake core 2, LCt 2) reveals some level of fluctuations down the core but, this is not as conspicuous as were observed in Balea and Podragu Mare lakes. There were series of inconspicuous increase and decrease in density down the core profile and a marginal surface decrease from the depth of 1 cm. The peaks and troughs are similar to Capra Lake being not very conspicuous (Figures 5.1a and 5.1b). The core (LCt 2) has a mean dry density value of 0.27 g cm^{-3} and a dry density value range of 0.15 - 0.53 g cm^{-3} (Table 5.1).

Capra Lake

Two cores of Capra Lake were analysed for sediment density. Although there was a series of inconspicuous peaks and troughs down the entire length of the core, however Capra lake core 2 (LCp 2) demonstrated a general surface decrease from the depth of 18 - 0 cm. The core (LCp 2) has a mean dry density value of 0.42 g cm^{-3} and a dry density value range of 0.06 - 0.58 g cm^{-3} (Table 4.4). Lacul Capra core 3 (LCp 3) also demonstrated a surface decrease from the depth of 18 - 0 cm. LCp 3 has a mean dry density value of 0.51 g cm^{-3} and a dry density value range of 0.11 - 0.75 g cm^{-3} (Table 5.1). Some level of fluctuations in density can be noticed on the profile but, this was not as conspicuous as were observed in Balea and Podragu Mare lakes (Figures 5.1a and 5.1b). The two Capra cores have a weak correlation in density (0.471 at 0.05 level).

Podragu Mare Lake

There was a series of peaks and troughs in the core profile of Podragu Mare Lake core 2 (LPm 2), for example, between the depths of 19 - 17 cm, 15 - 12 cm, 10 - 6 cm and 5 - 3 cm. Despite the numbers of the peaks and troughs the profile still demonstrated an obvious surface decrease in density from the depth of about 2.5 cm (Figures 5.1a and 5.1b). The core (LPm 2) has a mean dry density value of 0.83 g cm^{-3} and a dry density value range of 0.29 - 1.26 g cm^{-3} (Table 5.1).

The density profiles of Balea Lake (LBa 1, LBa 4) and Podragu Mare Lake (LPm 2) all demonstrated a number of peaks and troughs down the profiles. Examples can be found between the depths of 19 - 15 cm and 10 - 6 cm in LBa 1 and LBa 4 and between the depths of 19 - 17 cm, 15 - 12 cm, 10 - 6 cm and 5 - 3 cm in LPm 2. There was no clear demonstration of such peaks and troughs down the profiles of LCt 2, LCp 2 and LCp 3. The minimum dry density value was observed in Capra Lake (0.06 g cm-3) while the maximum was observed in Balea Lake, LBa 4 (1.62 g cm-3) (Table 5.1).

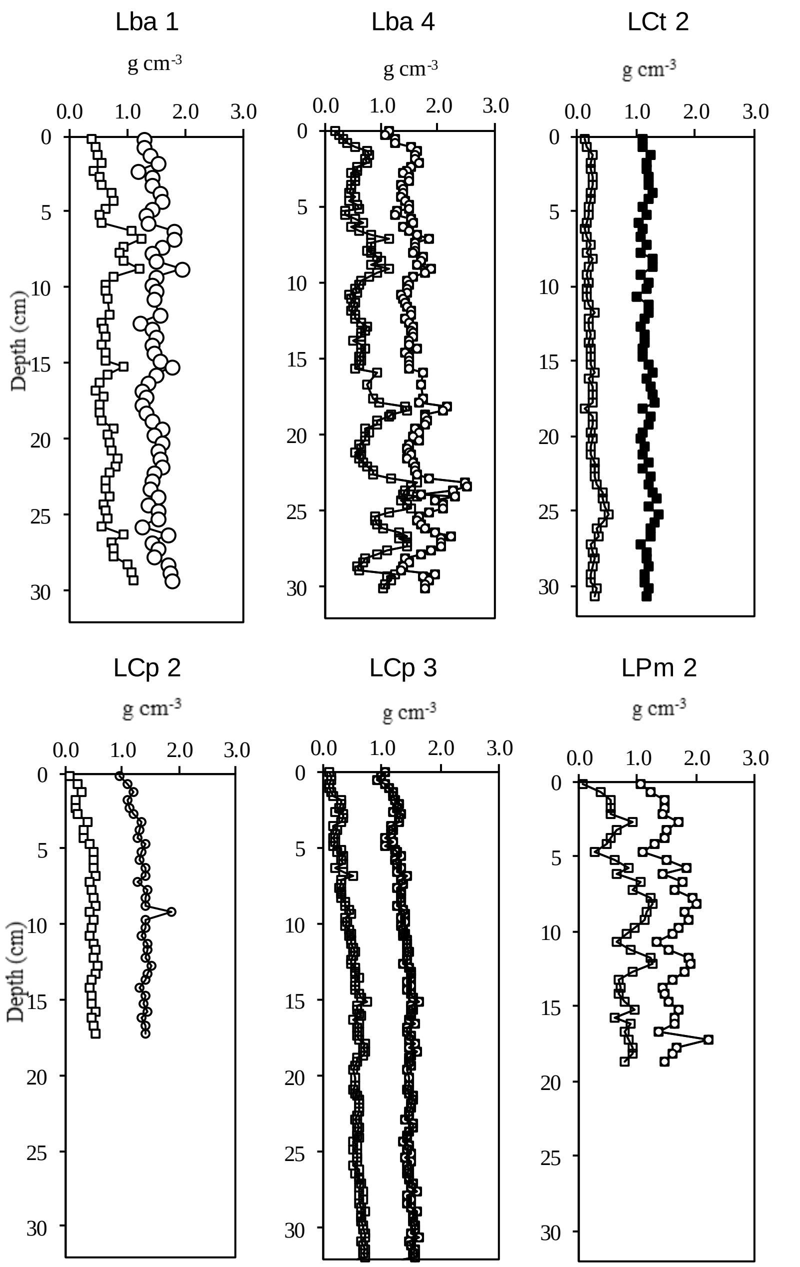


Figure 5.1a: Down core density profiles of Fagaras Lakes - Balea Lake core 4: LBa 4; Balea Lake core 1: LBa 1; Capra Lake core 3: LCp 3; Capra Lake core 2: LCp 2; Caltun Lake core 2: LCt 2 and Podragu Mare Lake core 2: LPm 2. (Note: The unit of density is in g cm^{-3}, the open square symbol profile represent the dry density while the open circle symbol represent the wet density).

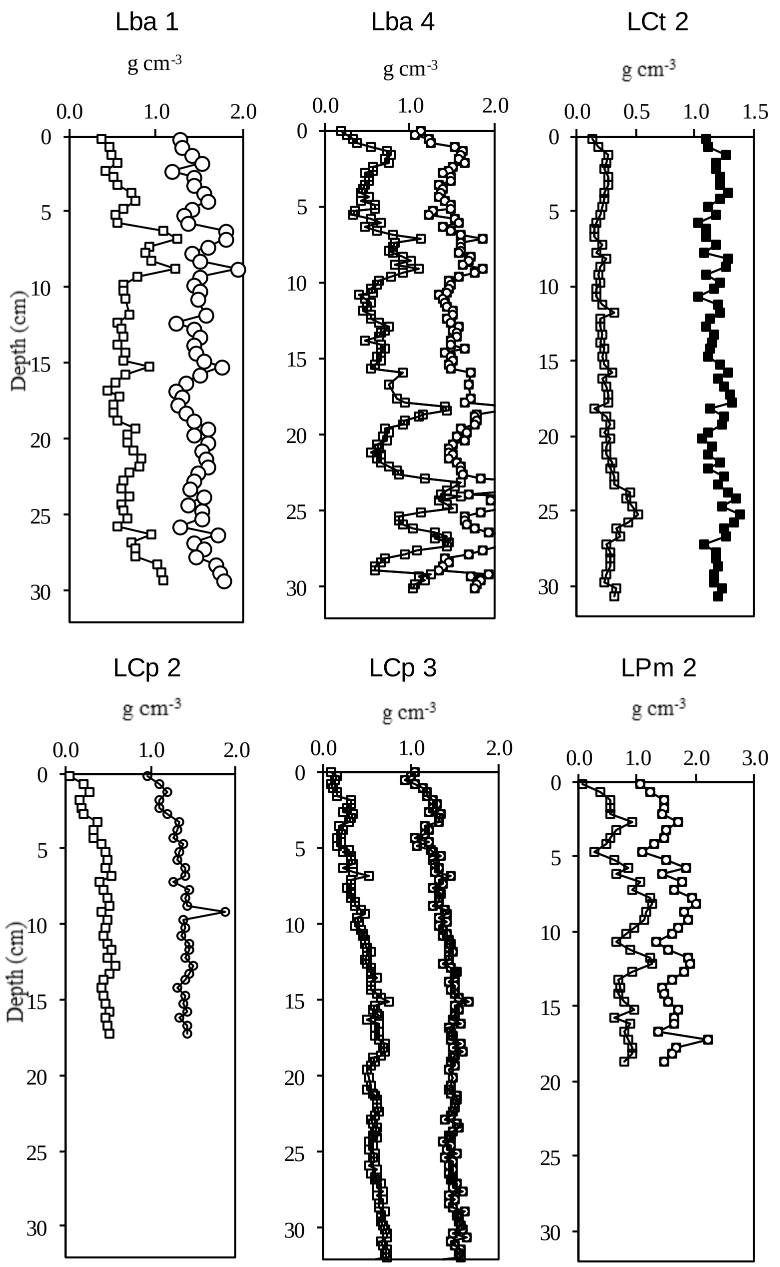


Figure 5.1b: Down core density profiles of Fagaras Lakes - Balea Lake core 4: LBa 4; Balea Lake core 1: LBa 1; Capra Lake core 3: LCp 3; Capra Lake core 2: LCp 2; Caltun Lake core 2: LCt 2 and Podragu Mare Lake core 2: LPm 2. (Note: 1. The unit of density is in g cm^{-3}, the open square symbol profile represent the dry density while the open circle symbol represent the wet density. 2. The density axes were plotted to maximum).

5.2.2 Density characteristics of lake sediments in the Rodna/Maramures region

Three core samples were taken from each lake sampled in the Rodna region in the 2006 sampling trip. Density measurements were carried out on all the three cores. A further three cores were also taken in both Pietrosul and Vinderel lakes in the 2008 sampling trip. Density measurements were carried out on one core for each of the two lakes sampled in 2008. There are two versions of the profiles (Figures 5.2a - 5.4a and Figures 5.2b - 5.4b), in Figures 5.2a - 5.4b the cores were all plotted to the same scale on all the axes while in Figures 5.2b - 5.4b the density axes were plotted to the maximum for each core in order to show fluctuations more clearly. In the Rodna Lake cores there were no clear demonstrations of peaks and troughs down their profiles. The mean dry density values of the lakes in the Rodna region varied from 0.44 g cm^{-3} (Lala Mare Lake, LLM 2) to 0.82 g cm^{-3} (Bila Lake, LB 2). The minimum dry density value was observed in Vinderel Lake (0.11 g cm^{-3}) while the maximum was observed in Bila Lake, LB 2 (1.21 g cm^{-3}) (Table 5.2).

Table: 5.2 Mean and standard deviations (Stdev) of Rodna lakes' sediment dry density (g cm^{-3})

Name	**Bila Lake**	**Buhaiescu-3 Lake**	**Lala Mare Lake**	**Pietrosul Lake**		**Stiol Lake**	**Vinderel Lake**	
	LB 2	**LB:3-2**	**LLM2**	**LP1 2006**	**LP1 2008**	**LS 2**	**LV3 2006**	**LV1 2008**
Mean	0.82	0.58	0.44	0.57	0.58	0.65	0.52	0.53
Stdev	0.27	0.24	0.07	0.27	0.18	0.25	0.12	0.13
Range	0.30-1.21	0.36-1.20	0.32-0.57	0.25-0.68	0.27-0.97	0.27-0.94	0.36-0.74	0.11-0.74

Bila Lake

Three cores from Bila Lake were subjected to density analysis (Bila lake cores LB1, LB 2 and LB 3). They all demonstrated a general decrease in sediment dry density towards the surface of the cores. Bila lake core LB1 demonstrated a general decrease in sediment dry density towards the surface of the core from the depth of 9.5 cm. Bila lake core LB 2 demonstrated a general decrease in sediment dry density towards the surface of the core from the depth of about 6.5 cm while Bila lake core LB 3 demonstrated a general decrease in sediment dry density towards the surface of the core from the depth of about 5 cm. A close examination of the profile shows some level of fluctuation density down each of the cores. The three cores of Bila Lake showed strong correlations in density at 0.01 level of significance (0.702: LB1/LB2; 0.575: LB1/LB3 and 0.712: LB2/LB3 respectively) There was a basal spike in density in LB 3 which was also observed in Buhaiescu-3 lake cores LB-3.2 and LB-3.3 (Figures 5.2a and 5.2b). The key core (LB 2) has a mean dry density value of 0.82 g cm^{-3} and a dry density value range of 0.30- 1.21 g cm^{-3} (Table 5.2).

Buhaiescu-3 Lake

All the three cores of Buhaiescu-3 Lake (LB-3.1, LB-3.2 and LB-3.3) subjected to density analysis demonstrated sediment density surface decrease from the depth of around 7.5cm. There was a strong correlation between cores 2 and 3 on a stastistical scale (0.809 at 0.01 level) of significance. Unlike Bila Lake, no obvious fluctuation was observed down the profiles. There was a basal spike in Buhaiescu-3 lake cores LB-3.2 and LB-3.3 (but penetrated below too) just like the basal spike in Bila lake LB 3 (Figures 5.2a and 5.2b). The core (LB-3.3) has a mean dry density value of 0.58 g cm^{-3} and a dry density value range of 0.36 - 1.20 g cm^{-3} (Table 5.2).

Lala Mare Lake

Three cores from Lala Mare Lake were subjected to density analysis (LLM 1, LLM 2 and LLM 3). There was no apparent change in down core density profiles of Lala Mare Lake (Figures 5.2a and 5.2b). Nevertheless, the three cores of Lala Mare Lake showed strong correlations in density at 0.01 level of significance (0.830: LLM1/LLM2; 0.882: LLM1/LLM3 and 0.884: LLM2/LLM3 respectively). The core (LLM 2) has a mean dry density value of 0.44 g cm^{-3} and a dry density value range of 0.32 – 0.57 g cm^{-3} (Table 5.2).

Pietrosul Lake

Five cores from Pietrosul Lake were subjected to density analysis (LP 1, LP 2 and LP 3: 2006; LP 1: 2008 and Russian corer 2008). There was a clear surface decrease in density from 3 cm for LP 2 and LP 3 both taken in 2006 whereas there was a clear surface decrease in density from 5 cm for Pietrosul lake sediments samples LP 1: 2006 and LP 1: 2008. At the depths of 5 - 7 cm and 7 - 9 cm troughs were observed in the profile of LP 1 taken in 2008. A peak was also observed at the depth of about 8 cm in the same core (Figures 5.3a and 5.3b). For Pietrosul lake sediments sample LP 1 taken with Russian corer in 2008 the surface decrease was observed from the depth of about 1.5 cm. The Russian corer sample was taken close to the lake margins. The wet and dry density (where measured) were similar. The cores (LP 1 : 2006 and LP 1 : 2008) have mean dry density values of 0.0.57 g cm^{-3} and 0.58 g cm^{-3}; dry density values range of 0.25 - 0.68 g cm^{-3} and 0.27 - 0.97 g cm^{-3} respectively (Table 5.2). There was no correlation in density in the cores of Pietrosul Lake except a weak correlation of 0.856 shown at 0.05 significant level between cores 1 and 3 taken in 2006.

Stiol Lake

All the three cores of Stiol Lake (LS 1, LS 2 and LS 3) subjected to density analysis demonstrated sediment density surface decrease from the depth of 4.5 cm. There was a basal increase in density in Stiol Lake core 3 (LS 3) between the depths of 8.5 - 6.5 cm (Figures 5.4a and 5.4b). The core (LS 2) has a mean dry density value of 0.65 g cm^{-3} and a dry density value range of 0.27 – 0.94 g cm^{-3} (Table 5.2). There were strong correlations in density at 0.01 significant level between LS1 and LS2 (0.940); also LS1 and LS3 (0.858).

Vinderel Lake

All the three cores from Vinderel Lake (LV 1, LV 2 and LV 3) sampled in 2006, as well as LV 1 sampled in 2008, demonstrated sediment density surface decrease from the depth of approximately 10 cm (Figures 5.4a and 5.4b). In both LV 3 sampled in 2006 and LV 1 sampled in 2008 there was a slight spike in density at the depth of 10 cm. The wet and dry density (where measured) were similar. There was a strong correlation in density at 0.01 level between LV1 and LV3 (0.878).

In this lake, the summer 2008 sampling (taken by British group) produced a core twice as long as the core from summer 2006 sampling (taken by Romanian group) and with the same gravity corer. This might be attributable to better skills on the part of the samplers. The cores (LV 3: 2006 and LV 1: 2008) have mean dry density values of 0.0.52 g cm^{-3} and 0.53 g cm^{-3}; dry density values range of 0.36 - 0.74 g cm^{-3} and 0.11 - 0.74 g cm^{-3} respectively (Table 5.2).

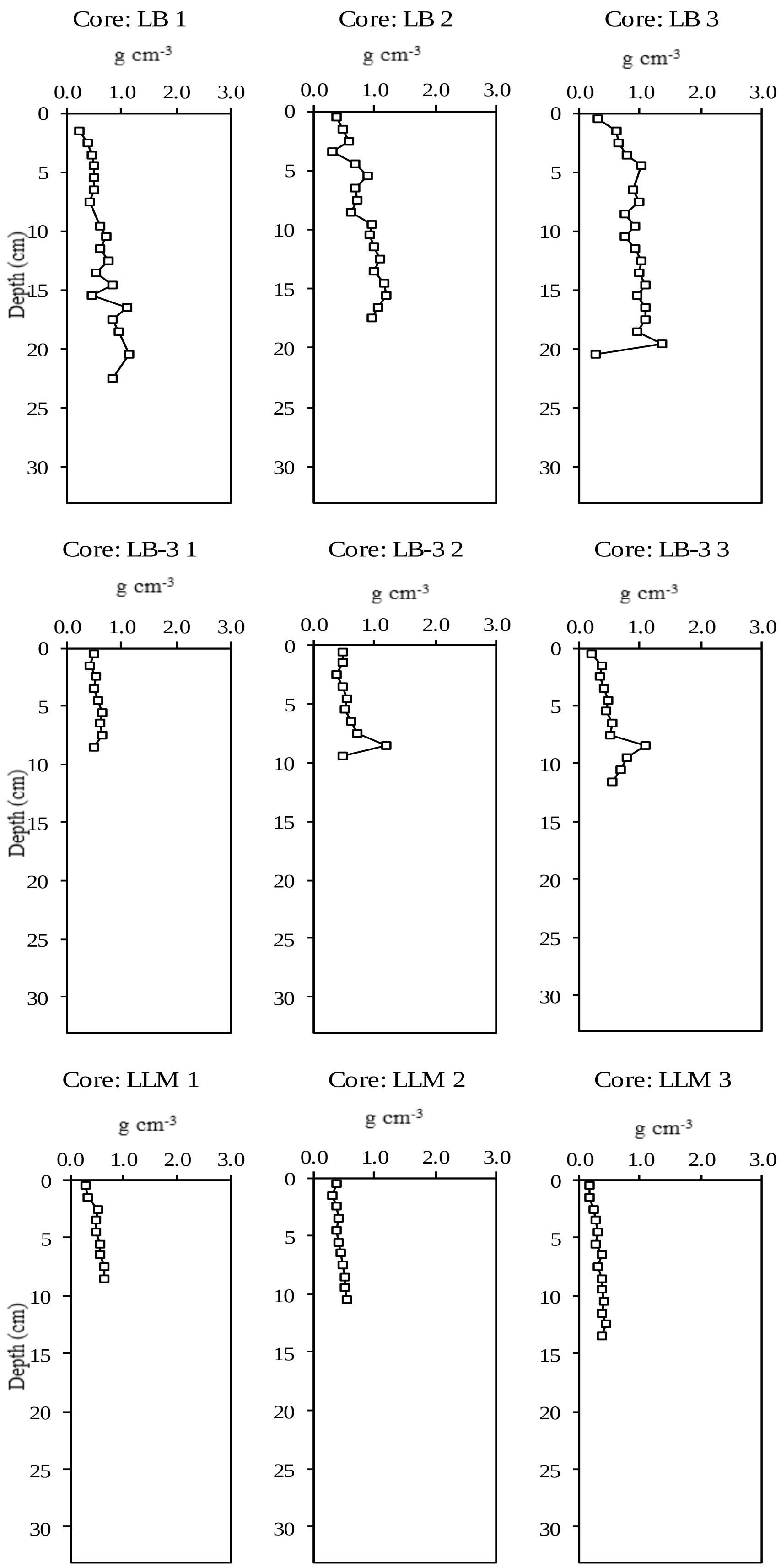


Figure 5.2a: Down core density profiles of Rodna Lakes (Lacul Bila cores - LB1, LB 2 & LB 3; Lacul Buhaiescu 3 cores - LB 3-1; LB 3- 2; LB 3-3 and Lacul Lala Mare cores - LLM 1, LLM 2 and LLM 3.

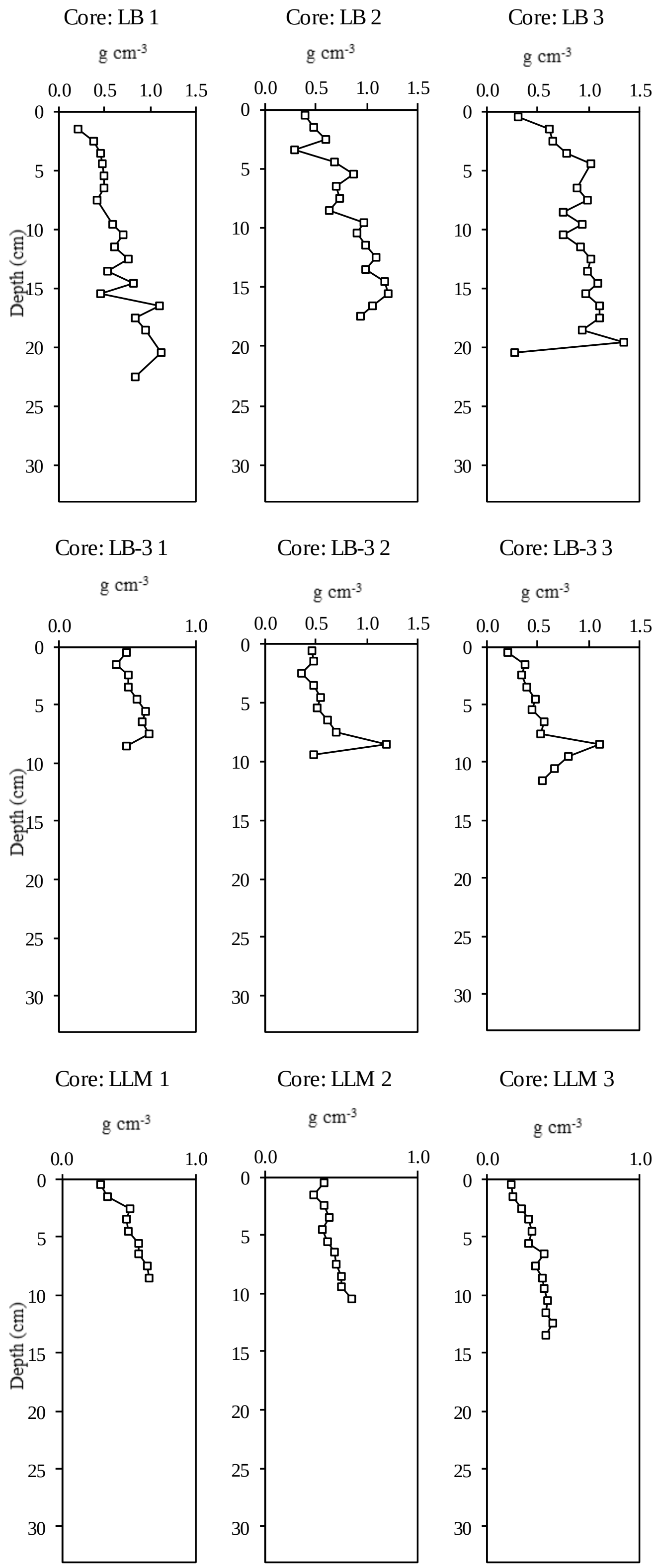


Figure 5.2b: Down core density profiles of Rodna Lakes (Lacul Bila cores - LB1, LB 2 & LB 3; Lacul Buhaiescu 3 cores - LB 3-1; LB 3- 2; LB 3-3 and Lacul Lala Mare cores - LLM 1, LLM 2 and LLM 3.

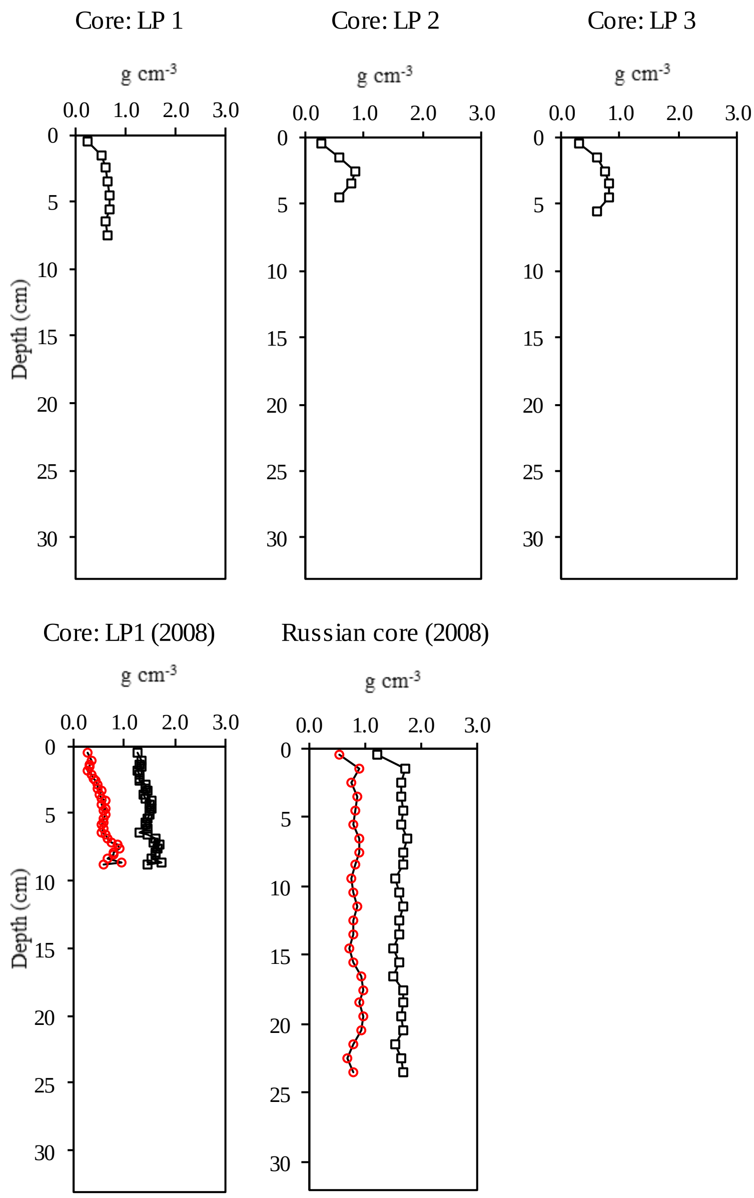


Figure 5.3a: Down core density profiles of Rodna Lakes (Lacul Pietrosul cores- LP 1, LP 2 & LP 3; LP 1 – 2008 and Russian corer 2008 All measurements are dry density except LP 1 sampled in 2008 in which case the wet density and dry density were made for comparison. All samples were taken in summer 2006 except LP1 (2008) and Russian corer (2008). (Note: The red symbol profile represents the dry density while the black symbol represents the wet density).

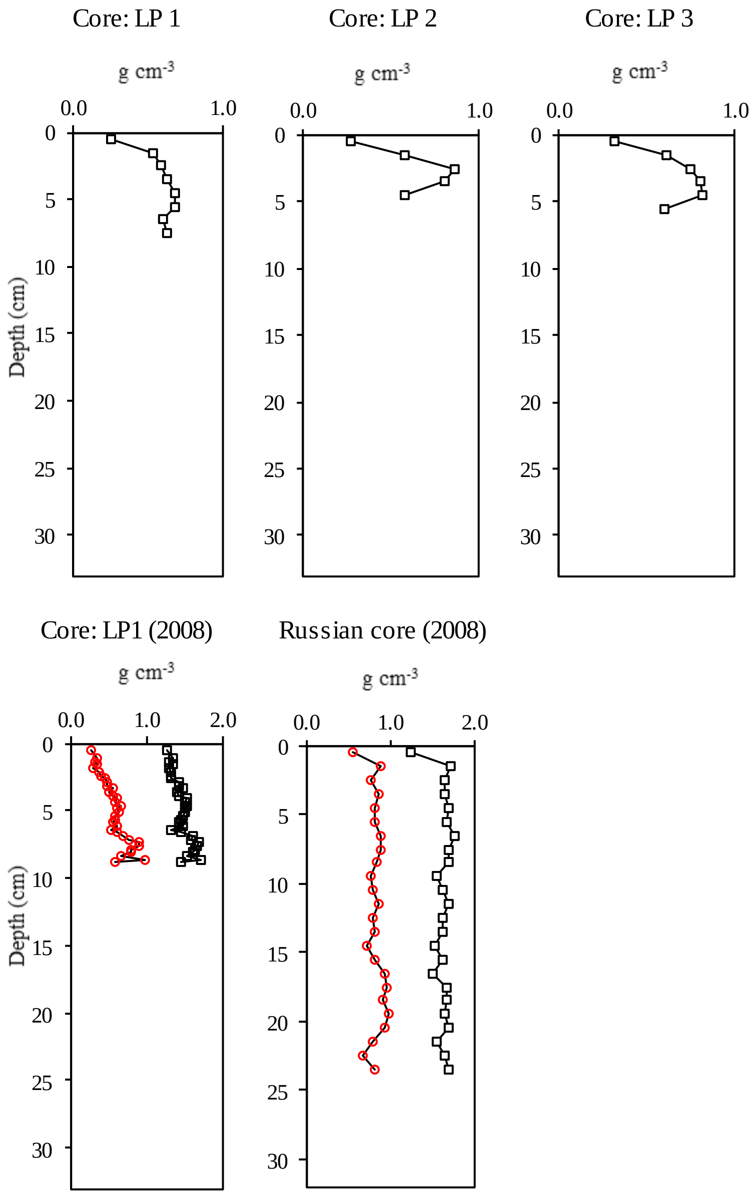


Figure 5.3b: Down core density profiles of Rodna Lakes (Lacul Pietrosul cores- LP 1, LP 2 & LP 3; LP 1 – 2008 and Russian corer 2008. All measurements are dry density except LP 1 sampled in 2008 in which case the wet density and dry density were made for comparison. All samples were made in summer 2006 except LP1 (2008) and Russian corer (2008). (Note: The red symbol profile represent the dry density while the black symbol represent the wet density).

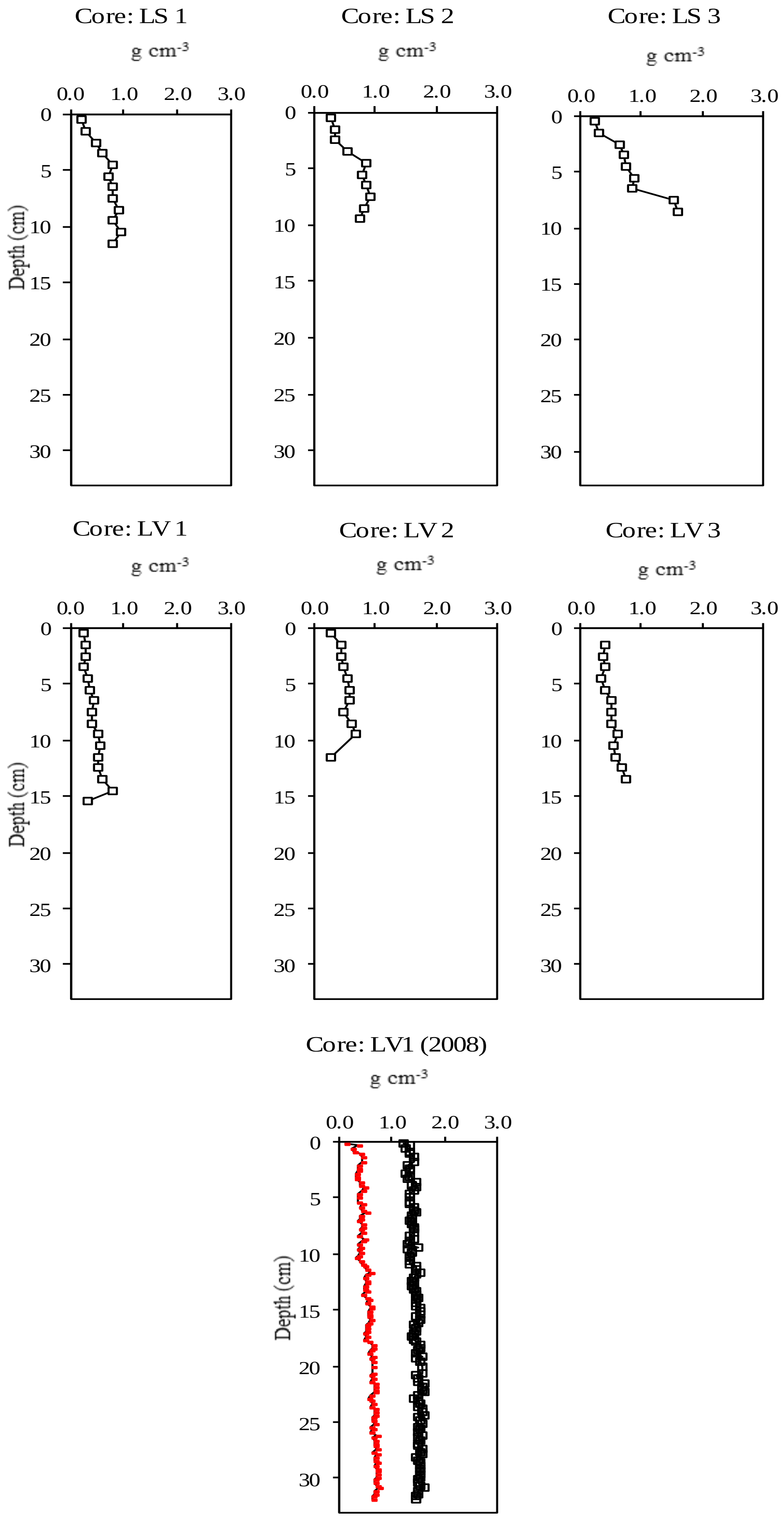


Figure 5.4a: Down core density profiles of Rodna Lakes (Lacul Stiol cores – LS 1, LS 2 & LS 3; Lacul Vinderel; cores- LV1, LV2 & LV3- 2006; LV 1- 2008; All measurements are dry density except LV 1 sampled in 2008 in which case the wet density and dry density were made for comparison. All samples were made in summer 2006 except LV1 (2008). (Note: The unit of density is in g cm^{-3}, the red symbol profile represent the dry density while the black symbol represent the wet density).

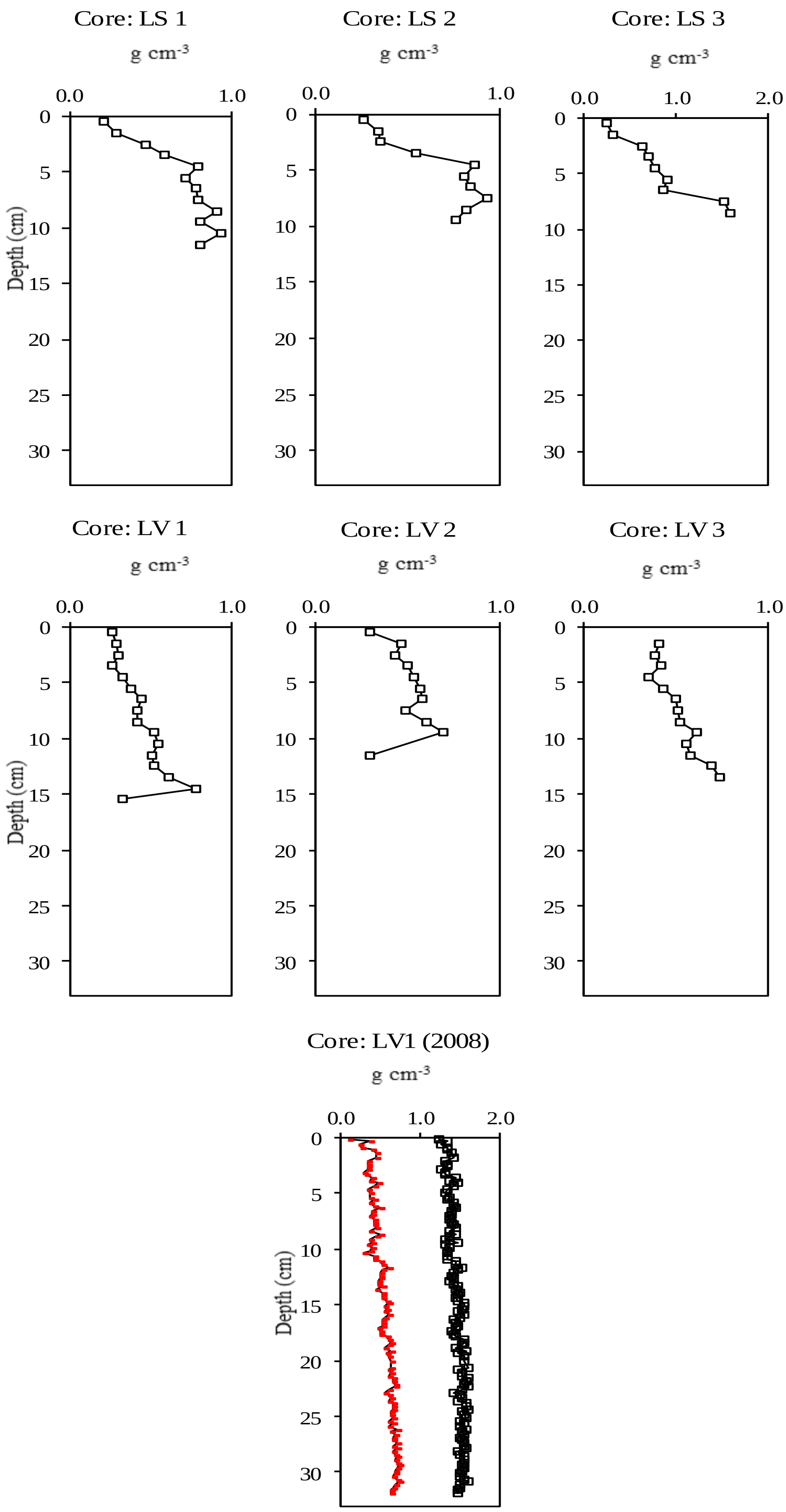


Figure 5.4b: Down core density profiles of Rodna Lakes (Lacul Stiol cores – LS 1, LS 2 & LS 3; Lacul Vinderel; cores- LV1, LV2 & LV3- 2006; LV 1- 2008; All measurements are dry density except LV 1 sampled in 2008 in which case the wet density and dry density were made for comparison. All samples were made in summer 2006 except LV1 (2008). (Note: The unit of density is in g cm^{-3}, the red symbol profile represent the dry density while the black symbol represent the wet density).

5.2.3 Loss-on-Ignition characteristics of lake sediments of the Fagaras region

Figure 5.5 shows the loss-on-ignition profiles for the main cores from the Fagaras region except Lacul Capra (core LCp 3) as this core was set aside for dating. However Capra 2 loss-on-ignition profile has been included in the analysis. The main findings are that loss-on-ignition profiles peak values vary from 15 - 22 %. The mean values of LOI for the Fagaras region lakes range from 10 - 15 % (Table 5.3). All lakes show an increase in loss-on-ignition towards the surface except Podragu Mare Lake (Figure 5.5).

Balea Lake key core (core LBa 4) demonstrated series of low LOI and presumably higher content of minerogenic material (e.g. between the depths of 25 - 20 cm, 19 - 12 cm and 12 - 5 cm). Within a small range a basal spike was observed Caltun Lake (core LCt 2) at the depth of about 27 cm. The down core profile was quite variable at almost every other centimetre interval but Caltun Lake (core LCt 2) showed a slight surface peak from the depth of 2.5 cm. Capra Lake (core LCp 2) demonstrated peaks between the depths of 17 - 13 cm and 11 - 9 cm. It demonstrated low LOI zones between the depths of 13 - 11 cm and 3 - 9 cm. Podragu Mare Lake (core LPm 2) demonstrated weak troughs between the depths of 17 - 14 cm and 9 - 5 cm. It showed a subsurface increase in LOI at the depths of about 8 - 4 cm.

Table 5.3: Mean and standard deviations (Stdev) of Fagaras lakes LOI (%)

Name	**Balea Lake (LBa 4)**	**Caltun Lake (LCt 2)**	**Capra Lake (LCp 2)**	**Podragu Mare Lake (LPm 2)**
Mean	9.58	14.55	13.81	12.29
Stdev	2.94	3.03	1.99	0.96
Range	3.79-14.95	7.65-21.59	10.46-17.21	10.98-14.68

On a statiscal scale, Balea Lake core 1 showed a weak positive correlation with Balea Lake core 4.

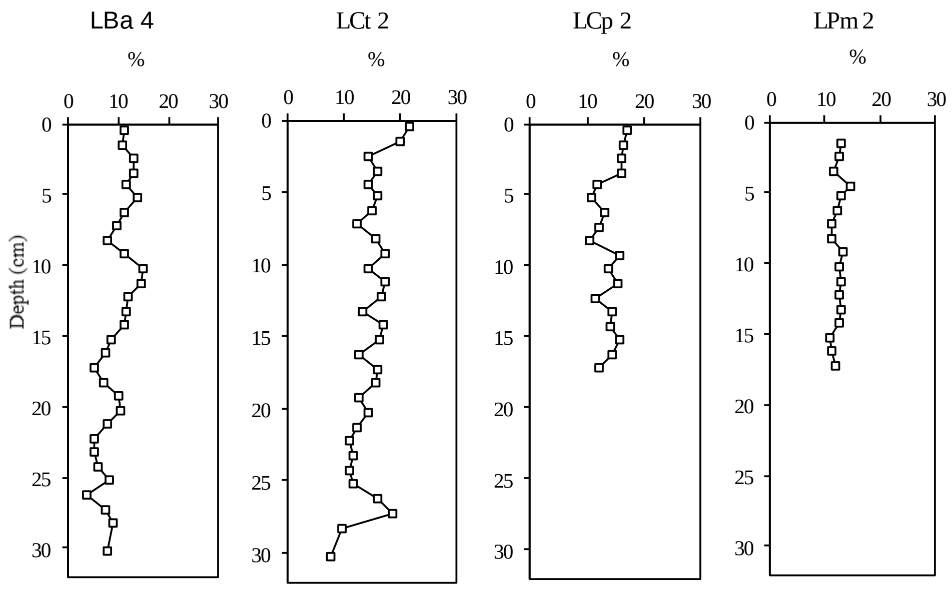


Figure 5.5: Down core variations of Fagaras lakes LOI profiles (Cores have been defined under density figures)

5.2.4 Loss-on-Ignition characteristics of lake sediments of the Rodna/Maramures region

Figure 5.6 shows the loss-on-ignition profiles for the six lakes from the Rodna region. The main findings are that the percentage of organic matter in the lake sediments is fairly high; the peak values varied from 14 - 25 % (Lala Mare Lake and Pietrosul Lake respectively). The core mean value of LOI for the Rodna region lakes ranges from 11 - 14 % (Table 5.4). All lakes showed a clear increase in loss-on-ignition towards the surface except Lala Mare Lake and Vinderel Lake.

In Bila Lake the steady rise for LOI starts from the depth of 9.5 cm and ranging from 14 % rises to 17 % at the surface depth of 0.5 cm. LOI for Buhaiescu-3 peaks from 8.5 cm at 6 % and rises to 21 % at the depth of 1.5cm and reaches 19 % at the surface depth of 0.5 cm. The fluctuations in the LOI profile of Buhaiescu-3 are quite obvious. There was no surface peak in LOI in Lala Mare Lake and there was no obvious fluctuation observed. Its LOI value ranges from 12 - 13 %. Both the 2006 and 2008 samples of Pietrosul Lake show obvious and very similar surface peak. They have the surface peak values of 21 % and 26 % respectively. Stiol Lake demonstrated a surface rise in LOI from a depth of 4.5 cm. There was no apparent change in any other portion of the profile. Both the 2006 and 2008 samples of Vinderel Lake demonstrated no very obvious surface increase. The 2008 core showed a small decrease in LOI at about a depth of 20 cm.

Table 5.4: Mean and standard deviations (Stdev) of Rodna lakes LOI (%)

Name	**Bila Lake**	**Buhaiescu-3 Lake**	**Lala Mare Lake**	**Pietrosul Lake**		**Stiol Lake**	**Vinderel Lake**	
	LB 2	**LB:3-2**	**LLM2**	**LP 1 (06)**	**LP1 (08)**	**LS 2**	**LV3 (06)**	**LV3 (08)**
Mean	14.21	14.94	13.70	10.94	13.43	10.80	13.97	13.01
Stdev	1.79	5.24	0.70	4.28	5.29	2.76	1.19	1.33
Range	11.00-16.69	6.39-21.53	12.70-14.38	7.65-21.06	9.20-25.62	9.03-16.52	11.97-15.83	9.24-15.99

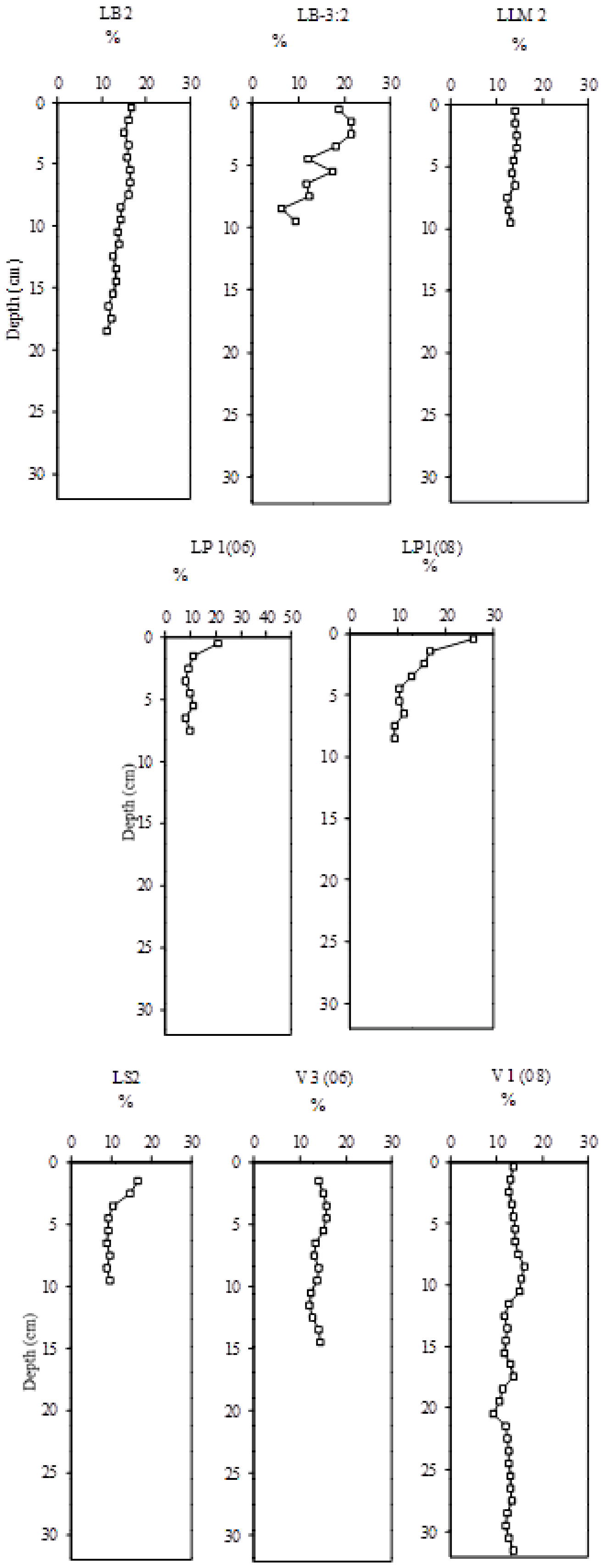


Figure 5.6: Down core variations of Rodna lakes LOI profiles (key cores) (Cores have been defined under density figures)

5.2.5 Particle size characteristics of lake sediments in the Fagaras region

Figures 5.7 - 5.9 show the particle size profiles for the main cores from the Fagaras region except Capra Lake (core LCp 3 which has been replaced by core LCp 2) as this core was set aside for dating. All cores were plotted to the same scale on both x-axis (concentration) and on the y-axis (depth) for the purpose of comparison (Figures 5.7 and 5.9.). In figure 4.31 each core was scaled to its maximum on the x-axis to show the intensity of the fluctuations of the particle size with depth. The peaks and troughs in the particle size profiles in all the lakes tallied with the peaks and troughs in each lake's respective sediment density.

Balea Lake

Balea Lake (core LBa 4) displayed alternating coarse and fine particle size layers with peaks in particle size at 26 cm, 23 cm and 18 cm and troughs at 25 cm and 21 cm respectively. This was followed by a zone of low particle size values between the depths of 14 - 3 cm. In LBa 4 there was a subsurface peak in particle size at the depth of 2.5 cm. LBa 4 has fluctuations in particle size distribution (profiles feature) greater than the other cores (Figures 5.7, 5.8 and 5.9). The core has a mean particle size value of 43.02 μm and a particle size value range of 20 - 104 μm (Table 5.5).

Caltun Lake

The whole core length of Caltun Lake (LCt 2) demonstrated troughs in particle size between 30 - 27 cm and from 27 - 22 cm. It also showed a surface decrease at around 6 - 0 cm. LCt2 demonstrated relatively fine particles compared to LBa4 and LPm2 (Figures 5.7, 5.8 and 5.9). The core has a mean particle size value of 32 μm and a particle size value range of 23 - 40 μm (Table 5.5).

Capra Lake

Capra Lake (core LCp 2) displayed a basal increase in particle size at 16 cm. It demonstrated a peak at 14 cm and a trough between the depths of 15 - 12 cm. There were no clear fluctuations between 12 - 6 cm which was followed by a spike at about 5 cm. It demonstrated a surface trough between the depths of 4 - 0.5 cm (Figures 5.7 and 5.9). The core has a mean particle size value of 24 µm and a particle size value range of 17 - 33 µm (Table 5.5). LCp 2 demonstrated relatively fine particles compared to LBa 4 and LPm 2.

Podragu Mare Lake

Podragu Mare Lake (core LPm 2) demonstrated clear troughs in particle size at the depths of 18 - 13 cm and 10.5 - 4.5 cm. It showed peaks in particle size at 16 cm, 13 cm, 10 cm and at about 3 cm. The core showed a surface decrease in particle size from the depth of about 3cm. The core has a mean particle size value of 50 µm and a particle size value range of 38 - 62 µm (Table 5.5).

All the four lakes from Fagaras demonstrated larger particle size than Rodna lakes. The mean value for the region varied from 24 - 50 µm and the actual particle size values ranged from 17 - 104 µm (see Table 5.5).

Table 5.5: Mean and standard deviations (Stdev) of Fagaras lakes particle size (µm)

Name	**Balea Lake (LBa 4)**	**Caltun Lake (LCt 2)**	**Capra Lake (LCp 2)**	**Podragu Mare Lake (LPm 2)**
Mean	43.02	31.89	24.15	49.93
STD Dev	23.72	3.83	4.33	6.79
Range	19.86-103.72	22.66-39.74	16.63-33.43	37.60-61.56

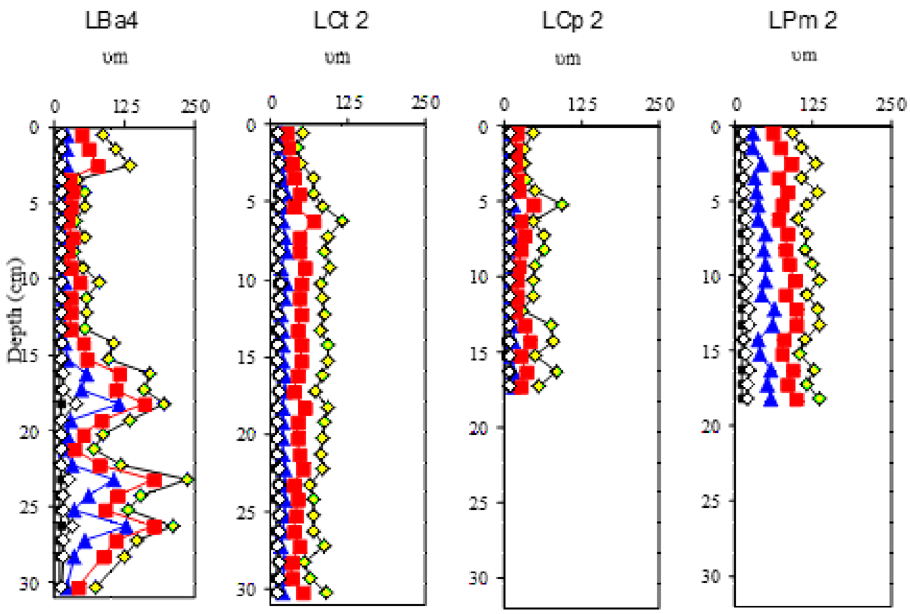


Figure 5.7 Particle size characteristics of Fagaras lakes in unified scale (black series represents D10, the white series represents D30, blue series represents D60, red series represents D80 and gold series represents D90). D represents diameter. (Cores have been defined under density figures)

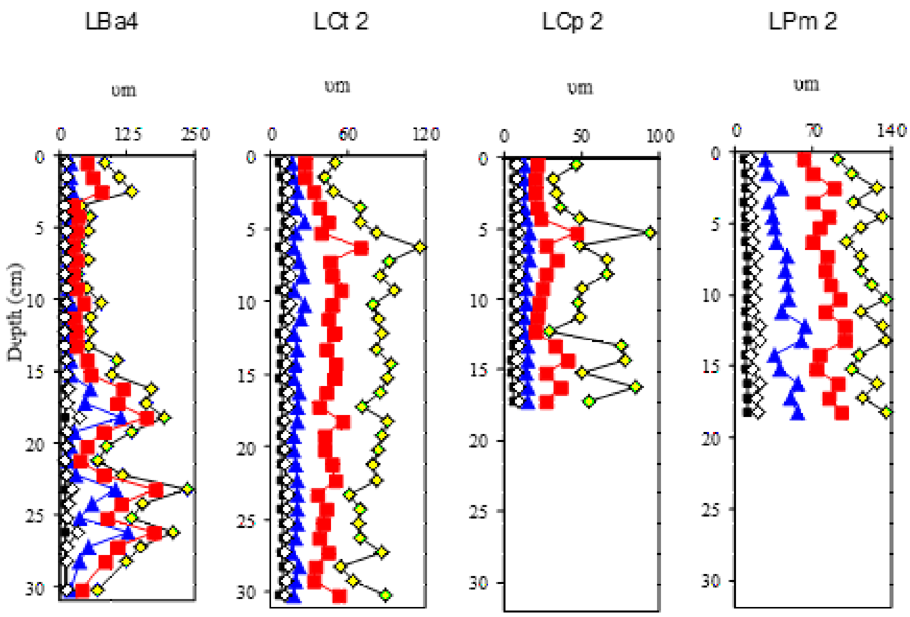


Figure 5.8: Particle size characteristics of Fagaras lakes individually scaled (black series represents D10, the white series represents D30, blue series represents D60, red series represents D80 and gold series represents D90). D represents diameter. (Cores have been defined under density figures)

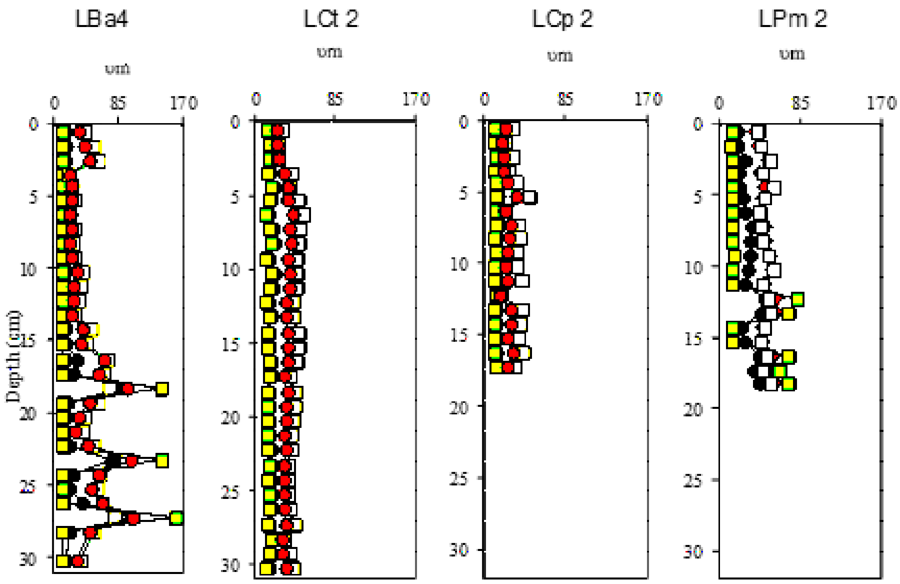


Figure 5.9: Particle size characteristics of Fagaras lakes (gold represents mode, black represents median, red represents mean, and white represents std. dev. particle size)

5.2.6 Particle size characteristics of lake sediments in the Rodna/Maramures region

Figures 5.10-5.12 show the particle size profiles for the main cores from the Rodna region. All cores were plotted to the same scale on both x-axis (concentration) and on the y-axis (depth) (Figures 5.10 and 5.12). In figure 5.11 each core was scaled to its maximum on the x-axis to show the intensity of the fluctuations of the particle size with depth. The peaks and troughs in the particle size profiles in all the lake tallied with the peaks and troughs in each lake respective density (Figure 5.2-5.4).

Bila Lake

Bila Lake (core LB 2) showed series of fluctuations in particle size distribution down core, but it exhibited a clear trough in particle size between the depths of 11 - 4 cm and peaks at 11 cm and 4 cm (Figures 5.10-5.12). It has a mean value of 25 µm in particle size and a particle size range of 9 -32 µm (Table 5.6).

Buhaiescu 3 Lake

Buhaiescu-3 Lake (core LB 3-2) showed a trough at the depth of 6.5 - 3.5 cm. No other clear feature was observed down the profile (Figures 5.10-5.12). It has a mean value of 37μm in particle size and a particle size range of 27 - 44 μm (Table 5.6). It has the next highest mean value of particle size after Pietrosul Lake (LP 1:2006).

Lala Mare Lake

Lala Mare Lake (core LLM 2) exhibited a surface increase in particle size from the depth of 2.5 cm but the remaining length of the core was characterised by fluctuations in particle size distribution with no clearly defined feature (Figures 5.10-5.12). It has a mean value of 27 μm in particle size and a particle size range of 24 - 30 μm (Table 5.6).

Pietrosul Lake

Pietrosul Lake (core LP 1) showed a subsurface peak at 3.5 cm but no extraordinary feature was shown more than that (Figures 5.10-5.12). It has a mean value of 39 μm in particle size and a particle size range of 31 - 52 μm (Table 5.6). It has the highest mean and the highest particle size range in the region.

Stiol Lake

Stiol Lake (core LS 2) exhibited a surface increase in particle size from the depth of 2.5 cm but the remaining length of the core was characterised by fluctuations in particle size distribution with no clearly defined feature (Figures 5.10-5.12). The lake has a mean value of 26 μm in particle size and a particle size range of 21 - 33 μm (Table 5.6).

Vinderel Lake

Vinderel Lake (core LV 3) showed a virtually flat down core particle size distribution (Figures 5.10-5.12). It has a particle size distribution with a range of 9.10 - 15 μm and 13 μm mean value of particle size (Table 5.6).

Each of LB 2, LB 3-2, LLM 2 and LS 2 demonstrated some level of relatively coarse particle size. The mean value for the region varied from 13 - 39 µm and the actual particle size values ranged from 9 - 52 µm (see Table 5.6). All the six lakes from Rodna demonstrated finer particle size than Fagaras lakes. Vinderel Lake stands out demonstrating relatively fine particle size (see Tables 5.5 and 5.6).

Table 5.6: Mean and standard deviations (Stdev) of Rodna lakes particle size (µm)

Name	**Bila Lake (LB 2)**	**Buhaiescu-3 Lake (LB:3-2)**	**Lala Mare Lake (LLM)**	**Pietrosul Lake (LP 1) (2006)**	**Stiol Lake (LS 2)**	**Vinderel Lake (LV 3) (2006)**
Mean	25.33	36.69	26.89	38.50	25.81	12.91
STD Dev	4.19	6.27	2.18	6.34	3.66	1.58
Range	8.66-32.26	27.26-44.26	23.91-30.09	31.27-51.88	21.34-32.63	9.86-15.33

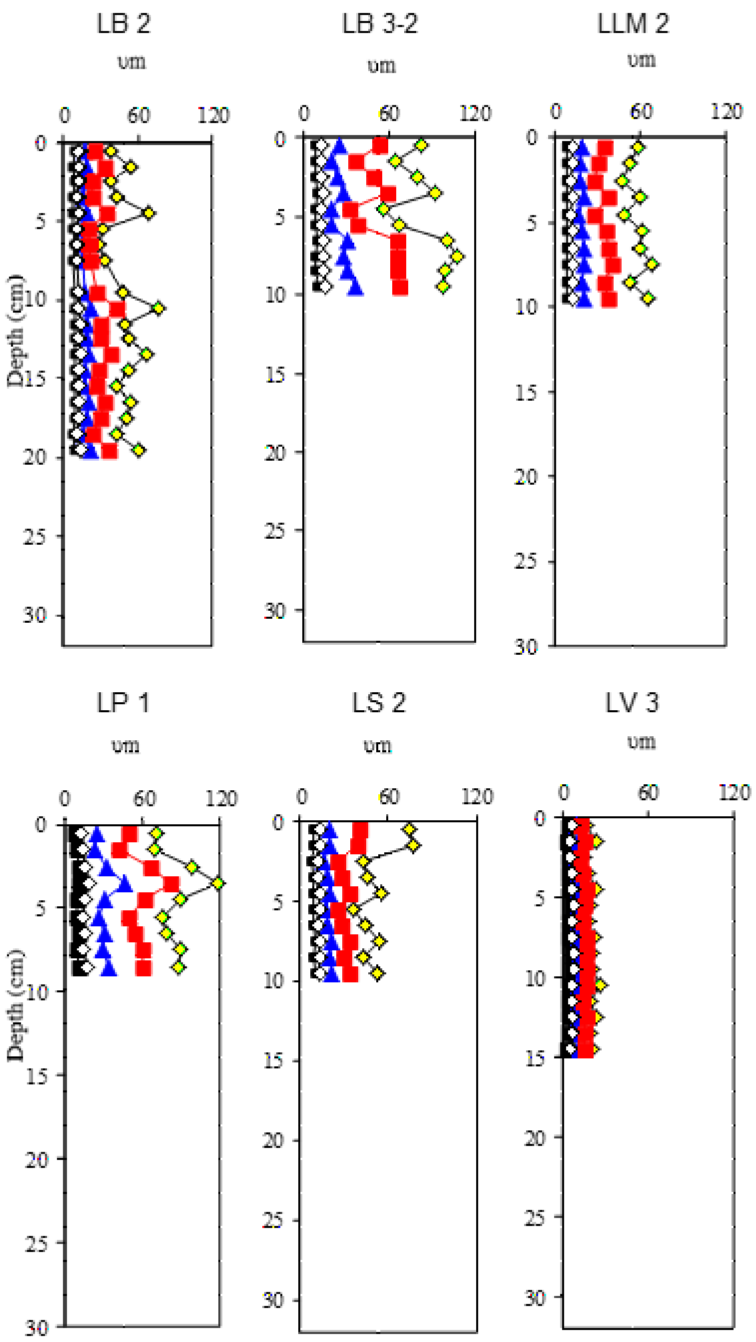


Figure 5.10: Particle size characteristics of Rodna lakes in unified scale (black series represents D10, the white series represents D30, blue series represents D60, red series represents D80 and gold series represents D90). D represents diameter. (Cores have been defined under density figures)

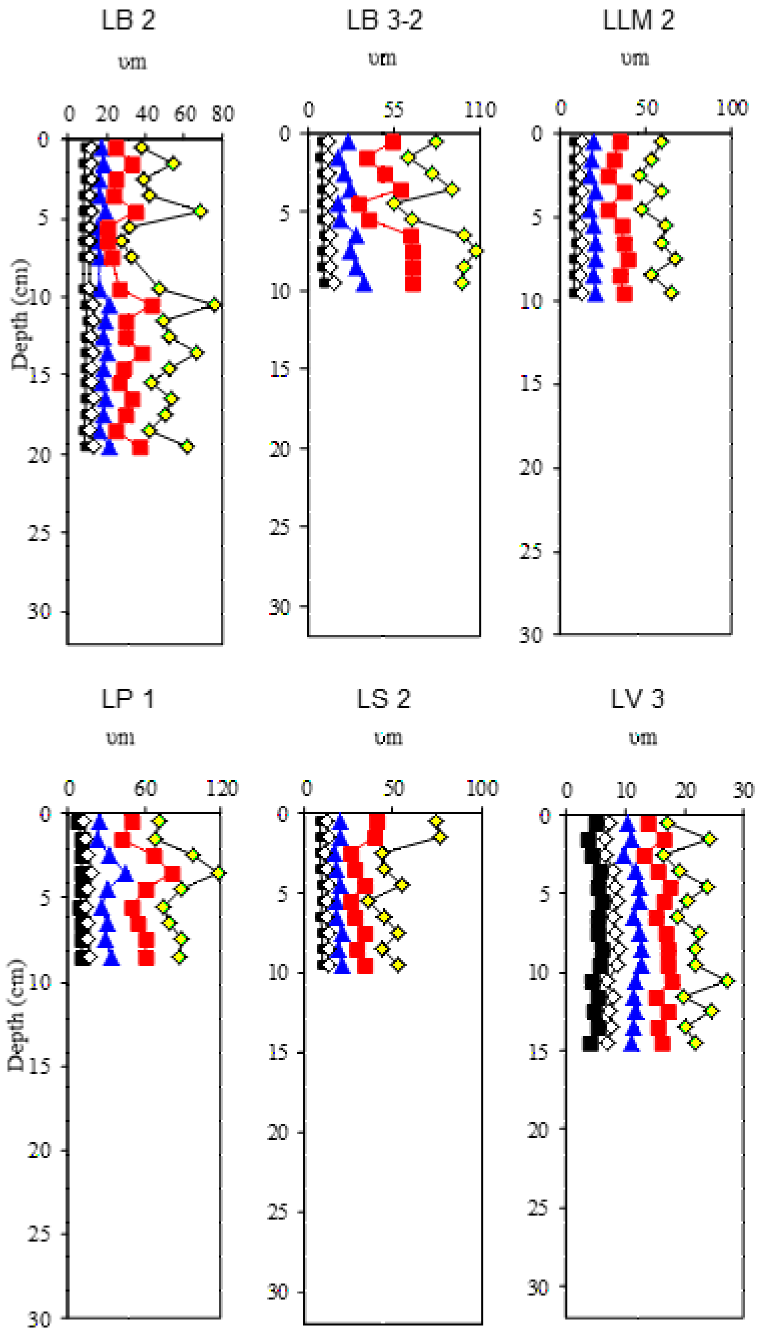


Figure 5.11: Particle size characteristics of Rodna lakes individually scaled (black series represents D10, the white series represents D30, blue series represents D60, red series represents D80 and gold series represents D90). D represents diameter. (Cores have been defined under density figures)

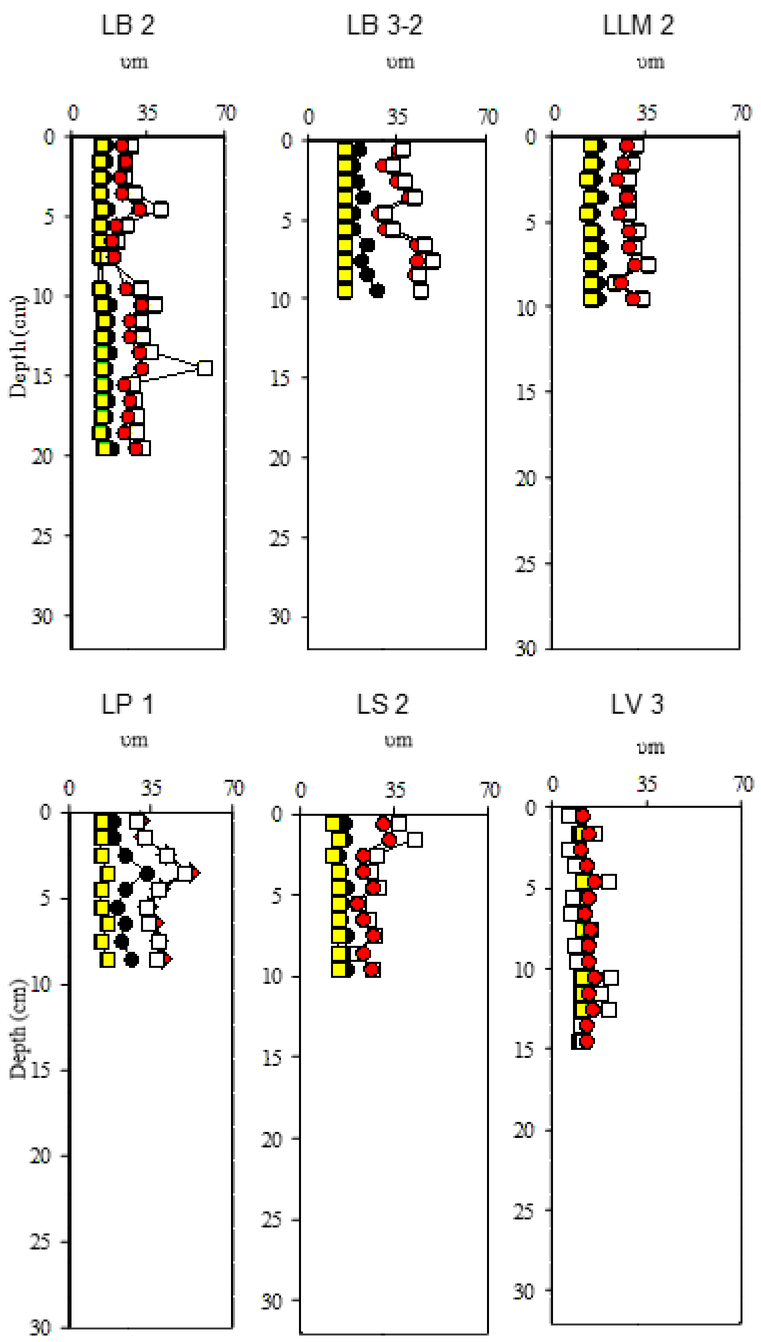


Figure 5.12: Particle size characteristics of Rodna lakes (gold represents mode, black represents median, red represents mean, and white represents std. dev. particle size)

5.2.7 Summary of lake sediments physical characteristics

All the lake sediment cores sampled demonstrated a decrease in sediment density towards the core surface. In the Fagaras region the minimum dry density value was observed in Capra Lake (0.06 g cm^{-3}) while the maximum was observed in Balea Lake, LBa 4 (1.62 g cm^{-3}) (Table 5.1). The mean sediment dry density values of cores from the lakes in Fagaras region varied from 0.27 g cm^{-3} (Caltun Lake) to 0.83 g cm^{-3} (Balea Lake, LBa 4). Statistically, there was significant correlation in the physical characteristics of Balea Lake core 4; at 0.01 significant level (the red figures in Table 5.7) but there was a weak correlation in Capra Lake (Table 5.7).

Table 5.7 Correlation of the physical characteristics of lake sediments in the Fagaras region

Balea Lake Core 4		Lba4 LOI	Lba4 Density	Lba4 PS Median	Lba4 PS Mean	Lba4 PS Mode	Lba4 PS Stddev	Lba4 D10	Lba4 D90
Lba4_LOI	Pearson	1	-.707**	-.485**	-.645**	-.347	-.638**	-.724**	-.707**
	Sig. (2-		.000	.008	.000	.065	.000	.000	.000
	N		29	29	29	29	29	29	29
Lba4_Density	Pearson		1	.614**	.681**	.552**	.608**	.766**	.639**
	Sig. (2-			.000	.000	.002	.000	.000	.000
	N			29	29	29	29	29	29
Lba4_PS_Median	Pearson			1	.914**	.966**	.745**	.792**	.674**
	Sig. (2-				.000	.000	.000	.000	.000
	N				29	29	29	29	29
Lba4_PS_Mean	Pearson				1	.825**	.945**	.897**	.901**
	Sig. (2-					.000	.000	.000	.000
	N					29	29	29	29
Lba4_PS_Mode	Pearson					1	.643**	.683**	.540**
	Sig. (2-						.000	.000	.003
	N						29	29	29
Lba4_PS_Stddev	Pearson						1	.845**	.955**
	Sig. (2-							.000	.000
	N							29	29
Lba4_D10	Pearson							1	.890**
	Sig. (2-								.000
	N								29
Lba4_D90	Pearson								1
	Sig. (2-								
	N								

Caltun Lake Core 2		LCt2 LOI	LCt2 Density	LCt2 PS Median	LCt2 PS Mean	LCt2 PS Mode	LCt2 PS Stdev	LCt2 PS D10	LCt2 PS D90
LCt2_LOI	Pearson	1	-.457*	-.410*	-.193	-.272	.016	-.401*	-.030
	Sig. (2-		.013	.027	.316	.154	.935	.031	.878
	N		29	29	29	29	29	29	29
lct2_Density	Pearson		1	.124	-.322	.115	-.420*	.201	-.362
	Sig. (2-			.520	.088	.553	.023	.297	.053
	N			29	29	29	29	29	29
LCt2_PS_Median	Pearson			1	.264	.895**	-.047	.951**	-.205
	Sig. (2-				.167	.000	.810	.000	.285
	N				29	29	29	29	29
LCt2_PS_Mean	Pearson				1	-.015	.923**	.094	.886**
	Sig. (2-					.938	.000	.629	.000
	N					29	29	29	29
LCt2_PS_Mode	Pearson					1	-.246	.941**	-.449*
	Sig. (2-						.198	.000	.015
	N						29	29	29
LCt2_PS_Stdev	Pearson						1	-.170	.946**
	Sig. (2-							.378	.000
	N							29	29
LCt2_PS_D10	Pearson							1	-.356
	Sig. (2-								.058
	N								29
LCt2_PS_D90	Pearson								1
	Sig. (2-								
	N								

Capra Lake Core 2		LCp2 LOI	LCp2 Density	LCp2 PS Median	LCp2 PS Mean	LCp2 PS Mode	LCp2 PS Stdev	LCp2 PS D10	LCp2 PS D90
LCp2_LOI	Pearson	1	-.418	-.453	-.228	-.371	-.186	-.385	-.183
	Sig. (2-		.095	.068	.378	.143	.476	.127	.483
	N		17	17	17	17	17	17	17
lcP2_Density	Pearson		1	.401	.405	.084	.302	-.167	.423
	Sig. (2-			.110	.107	.749	.239	.521	.090
	N			17	17	17	17	17	17
LCp2_PS_Median	Pearson			1	.775**	.449	.552*	.380	.784**
	Sig. (2-				.000	.071	.021	.133	.000
	N				17	17	17	17	17
LCp2_PS_Mean	Pearson				1	-.062	.929**	.056	.973**
	Sig. (2-					.813	.000	.831	.000
	N					17	17	17	17
LCp2_PS_Mode	Pearson					1	-.188	.637**	-.098
	Sig. (2-						.470	.006	.707
	N						17	17	17
LCp2_PS_Stdev	Pearson						1	.066	.843**
	Sig. (2-							.802	.000
	N							17	17
LCp2_PS_D10	Pearson							1	.010
	Sig. (2-								.969
	N								17
LCp2_PS_D90	Pearson								1
	Sig. (2-								
	N								

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

Podragu Mare Lake Core 2		LPM2 LOI	LPM2 Density	LPM2 PS Median	LPM2 PS Mean	LPM2 PS Mode	LPM2 PS Stdev	LPM2 PS D10	LPM2 PS D90
LPM2_LOI	Pearson	1	-.676**	-.375	-.326	-.150	-.003	-.488*	-.150
	Sig. (2-		.003	.138	.202	.565	.990	.047	.564
	N		17	17	17	17	17	17	17
LPM2_Density	Pearson		1	.582*	.485*	.195	.005	.694**	.216
	Sig. (2-			.014	.048	.453	.983	.002	.406
	N			17	17	17	17	17	17
LPM2_PS_Median	Pearson			1	.908**	.860**	.364	.950**	.614**
	Sig. (2-				.000	.000	.152	.000	.009
	N				17	17	17	17	17
LPM2_PS_Mean	Pearson				1	.762**	.711**	.857**	.883**
	Sig. (2-					.000	.001	.000	.000
	N					17	17	17	17
LPM2_PS_Mode	Pearson					1	.331	.729**	.529*
	Sig. (2-						.195	.001	.029
	N						17	17	17
LPM2_PS_Stdev	Pearson						1	.306	.951**
	Sig. (2-							.233	.000
	N							17	17
LPM2_PS_D10	Pearson							1	.556*
	Sig. (2-								.020
	N								17
LPM2_PS_D90	Pearson								1
	Sig. (2-								
	N								

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

In the Rodna region the minimum density value was observed in Vinderel Lake (0.11 g cm^{-3}) while the maximum was observed in Buhaiescu-3 Lake, LB: 3-2 (1.20 g cm^{-3}) (Table 5.2). The mean dry density values of the lakes in the Rodna region varied from 0.44 g cm^{-3} (Lala Mare Lake, LLM 2) to 0.65 g cm^{-3} (Stiol Lake, LS 2). Statiscal analysis of lakes from the Rodna region showed strong correlations between some of the physical characteristics of some of the lakes (red figures in Table 5.8).

Table 5.8: Correlation of the physical characteristics of lake sediments in the Rodna/Maramures region

Bila Lake Core 2		LB2 LOI	LB2 Density	LB2 PS Median	LB2 PS Mean	LB2 PS Mode	LB2 PS Stdev	LB2 PS D10	LB2 PS D90
LB2_LOI	Pearson	1	-.824**	-.526*	-.483*	-.495*	-.380	-.562*	-.392
	Sig. (2-		.000	.025	.042	.037	.119	.015	.107
	N		18	18	18	18	18	18	18
LB2_Density	Pearson		1	.247	.128	.201	-.001	.289	.143
	Sig. (2-			.322	.613	.424	.997	.244	.572
	N			18	18	18	18	18	18
LB2_PS_Median	Pearson			1	.824**	.828**	.442	.737**	.852**
	Sig. (2-				.000	.000	.066	.000	.000
	N				18	18	18	18	18
LB2_PS_Mean	Pearson				1	.717**	.837**	.482*	.919**
	Sig. (2-					.001	.000	.043	.000
	N					18	18	18	18
LB2_PS_Mode	Pearson					1	.529*	.812**	.605**
	Sig. (2-						.024	.000	.008
	N						18	18	18
LB2_PS_Stdev	Pearson						1	.319	.580*
	Sig. (2-							.196	.012
	N							18	18
LB2_PS_D10	Pearson							1	.339
	Sig. (2-								.168
	N								18
LB2_PS_D90	Pearson								1
	Sig. (2-								
	N								

Buhiescu 3 Lake Core 2		LB32 LOI	LB32 Density	LB32 PS Median	LB32 PS Mean	LB32 PS Mode	LB32 PS Stdev	LB32 PS D10	LB32 PS D90
LB32_LOI	Pearson	1	-.726*	-.606	-.565	-.258	-.495	-.607	-.497
	Sig. (2-		.018	.063	.089	.471	.145	.063	.144
	N		10	10	10	10	10	10	10
LB32_Density	Pearson		1	.260	.393	-.346	.381	.072	.427
	Sig. (2-			.468	.261	.328	.277	.844	.218
	N			10	10	10	10	10	10
LB32_PS_Median	Pearson			1	.900**	.530	.773**	.752*	.800**
	Sig. (2-				.000	.115	.009	.012	.005
	N				10	10	10	10	10
LB32_PS_Mean	Pearson				1	.157	.967**	.641*	.981**
	Sig. (2-					.666	.000	.046	.000
	N					10	10	10	10
LB32_PS_Mode	Pearson					1	-.023	.667*	-.019
	Sig. (2-						.950	.035	.959
	N						10	10	10
LB32_PS_Stdev	Pearson						1	.539	.987**
	Sig. (2-							.108	.000
	N							10	10
LB32_PS_D10	Pearson							1	.547
	Sig. (2-								.101
	N								10
LB32_PS_D90	Pearson								1
	Sig. (2-								
	N								

Lala Mare Lake Core 2		LLM2 LOI	LLM2 Density	LLM2 PS Median	LLM2 PS Mean	LLM2 PS Mode	LLM2 PS Stdev	LLM2 PS D10	LLM2 PS D90
LLM2_LOI	Pearson	1	-.737*	-.238	-.357	-.311	-.163	.553	-.506
	Sig. (2-		.015	.509	.311	.381	.652	.097	.135
	N		10	10	10	10	10	10	10
LLM2_Density	Pearson		1	.607	.470	.409	.050	-.047	.560
	Sig. (2-			.063	.170	.241	.891	.898	.092
	N			10	10	10	10	10	10
LLM2_PS_Median	Pearson			1	.826**	.842**	.342	.548	.810**
	Sig. (2-				.003	.002	.333	.101	.005
	N				10	10	10	10	10
LLM2_PS_Mean	Pearson				1	.721*	.794**	.330	.977**
	Sig. (2-					.019	.006	.352	.000
	N					10	10	10	10
LLM2_PS_Mode	Pearson					1	.269	.293	.768**
	Sig. (2-						.452	.411	.010
	N						10	10	10
LLM2_PS_Stdev	Pearson						1	.179	.723*
	Sig. (2-							.621	.018
	N							10	10
LLM2_PS_D10	Pearson							1	.161
	Sig. (2-								.657
	N								10
LLM2_PS_D90	Pearson								1
	Sig. (2-								
	N								

Pietrosul Lake Core 1 (2006)		LP1 LOI	LP1 Density	LP1 PS Median	LP1 PS Mean	LP1 PS Mode	LP1 PS Stdev	LP1 PS D10	LP1 PS D90
LP1_LOI	Pearson	1	-.912**	-.583	-.615	-.478	-.662	-.955**	-.553
	Sig. (2-		.002	.129	.105	.231	.074	.000	.155
	N		8	8	8	8	8	8	8
LP1_Density	Pearson		1	.413	.464	.233	.520	.794*	.415
	Sig. (2-			.309	.247	.579	.186	.019	.307
	N			8	8	8	8	8	8
LP1_PS_Median	Pearson			1	.965**	.740*	.920**	.700	.928**
	Sig. (2-				.000	.036	.001	.053	.001
	N				8	8	8	8	8
LP1_PS_Mean	Pearson				1	.570	.985**	.700	.991**
	Sig. (2-					.140	.000	.053	.000
	N					8	8	8	8
LP1_PS_Mode	Pearson					1	.474	.596	.466
	Sig. (2-						.235	.119	.244
	N						8	8	8
LP1_PS_Stdev	Pearson						1	.743*	.985**
	Sig. (2-							.035	.000
	N							8	8
LP1_PS_D10	Pearson							1	.634
	Sig. (2-								.091
	N								8
LP1_PS_D90	Pearson								1
	Sig. (2-								
	N								

Stiol Lake Core 2		LS2 LOI	LS2 Density	LS2 PS Median	LS2 PS Mean	LS2 PS Mode	LS2 PS Stdev	LS2 PS D10	LS2 D90
LS2_LOI	Pearson	1	-.812**	.098	.806**	-.575	.847**	-.229	.866**
	Sig. (2-		.008	.803	.009	.106	.004	.553	.003
	N		9	9	9	9	9	9	9
LS2_Density	Pearson		1	.401	-.483	.747*	-.711*	.578	-.568
	Sig. (2-			.250	.157	.013	.021	.080	.087
	N			10	10	10	10	10	10
LS2_PS_Median	Pearson			1	.515	.496	.154	.901**	.420
	Sig. (2-				.127	.145	.671	.000	.227
	N				10	10	10	10	10
LS2_PS_Mean	Pearson				1	-.157	.919**	.201	.990**
	Sig. (2-					.664	.000	.578	.000
	N					10	10	10	10
LS2_PS_Mode	Pearson					1	-.360	.657*	-.250
	Sig. (2-						.307	.039	.486
	N						10	10	10
LS2_PS_Stdev	Pearson						1	-.144	.937**
	Sig. (2-							.692	.000
	N							10	10
LS2_PS_D10	Pearson							1	.087
	Sig. (2-								.811
	N								10
LS2_D90	Pearson								1
	Sig. (2-								
	N								

Vinderel Lake Core 3 (2006)		LV3 LOI	LV3 Density	LV3 PS Median	LV3 PS Mean	LV3 PS Mode	LV3 PS Stdev	LV3 PS D10	LV3 PS D90
LV3_LOI	Pearson	1	-.631*	-.398	-.372	-.317	-.195	-.257	-.340
	Sig. (2-		.021	.178	.211	.291	.523	.396	.256
	N		13	13	13	13	13	13	13
LV3_Density	Pearson		1	.323	.495	.418	.439	.188	.347
	Sig. (2-			.282	.086	.155	.133	.538	.245
	N			13	13	13	13	13	13
LV3_PS_Median	Pearson			1	.497	.807**	.045	.789**	.341
	Sig. (2-				.084	.001	.884	.001	.254
	N				13	13	13	13	13
LV3_PS_Mean	Pearson				1	.571*	.872**	-.036	.933**
	Sig. (2-					.042	.000	.908	.000
	N					13	13	13	13
LV3_PS_Mode	Pearson					1	.205	.461	.502
	Sig. (2-						.503	.113	.081
	N						13	13	13
LV3_PS_Stdev	Pearson						1	-.396	.813**
	Sig. (2-							.180	.001
	N							13	13
LV3_PS_D10	Pearson							1	-.266
	Sig. (2-								.379
	N								13
LV3_PS_D90	Pearson								1
	Sig. (2-								
	N								

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

There are more fluctuations in the density down core profiles of most lakes in the Fagaras region than the Rodna region; for example Balea Lake in the Fagaras region. As shown above the highest dry density value recorded in the Rodna region was lower than the highest value recorded in the Fagaras region. The low density zones of each lake are also characterised by a relatively high organic content.

The main findings for loss-on-ignition are that the percentage loss-on-ignition in the lake sediments in Fagaras ranged from 4 to 22 % (Balea Lake and Caltun Lake respectively). The mean values of LOI for the Fagaras region lakes ranged from 10 to 15 % (Table 5.3). The percentage loss-on-ignition in the Rodna region lake sediments is higher than the Fagaras region; it ranged from 6 to 26 % (Buhaiescu-3 Lake and Pietrosul Lake respectively). The mean value of LOI for the Rodna lakes ranged from 11 to 14 % (Table 5.4). All lakes in both regions tend to show an increase in loss-on-ignition towards the surface except Lala Mare and Vinderel lakes.

All the four lakes from the Fagaras region demonstrated a larger particle size range than the sample in the Rodna region. The mean value for the region varied from 24 - 50 μm and the particle size values ranged from 17 - 104 μm (see Table 5.5). The mean value for the Rodna region varied from 13 - 39 μm and the particle size values ranged from 9 - 52 μm (see Table 5.6).

5.3 Mineral magnetic measurements

The purpose of mineral magnetic measurements is to characterise the magnetic mineral properties of the sediments and to determine their concentration and grain size variations in remanence carrying minerals. It has also been mentioned in chapter two that sediment magnetic parameters are almost never sensitive to one of the above-mentioned properties alone, but respond to several physical properties of a mineral grain.

5.3.1 Mineral magnetic measurements of lake sediments in the Fagaras region

Down core profiles of key magnetic parameters and selected ratios between them are shown for the Fagaras lakes in Figures 5.13-5.18. Susceptibility (χ), anhysteretic remanent magnetisation (ARM) and saturation isothermal remanent magnetisation (SIRM) are all measures of magnetic mineral concentration. The magnetic parameters χ, ARM and SIRM were shown for all lakes. The different ratios such as SIRM/ARM, SIRM/χ and ARM/χ; magnetic softness, magnetic hardness and backfields were also shown for all lakes. The key findings are that the profiles of: low frequency magnetic susceptibility (X), anhysteretic remanent magnetisation (ARM) and saturation isothermal remanent magnetisation (SIRM) are slightly comparable between cores from the same lake and are slightly comparable among the lakes in the southern region; all cores show a clear surface peak. At this peak there is also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters 'soft' and 'hard' (Figures 5.13-5.18). The χ values are relatively high except for Caltun Lake which has a relatively low value. The ARM and SIRM peak values for each core are both respectively comparable.

Balea Lake

Two cores LBa 1 and LBa 4 were characterised magnetically (Figures 5.13 and 5.14). Peaks and troughs in concentrations were observed down the profile of Balea lake core 1 (LBa 1). Between the depths of 9 - 5 cm, there was a trough in ARM, SIRM, SIRM/χ, and ARM/χ. There was a peak in SIRM/ARM and the parameter 'hard' at the same depths. The profiles of both cores LBa 1 and LBa 4 analysed for this lake are to some extent similar except that LBa 1 did not demonstrate a surface increase in the magnetic parameters χ, ARM and SIRM. From the bottom of Balea Lake core 4 (LBa 4) all through to the surface showed series of fluctuations in the sediment magnetic parameters χ, ARM, SIRM and the other ratios (for example between the depths of 23-18cm). These troughs were also alternated with peaks.

LBa 4 demonstrated a clear surface peak in χ, ARM and SIRM from 8 cm. The magnitudes of all the parameters differ in between LBa 1 and LBa 4. In all the parameters LBa 4 has higher values, for example LBa 1 has a SIRM surface value of 432.5 x 10^{-5} Am^2kg^{-1} while LBa 4 has a SIRM surface value of 1285.1 x $10^{-5}Am^2kg^{-1}$. In both cores the peaks and troughs in the ARM and SIRM were similar (occurred at corresponding depth) but such features were more distinct in LBa 4 than in LBa 1.

Caltun Lake

Magnetic susceptibility exhibited fluctuations in profile down core. Starting from the bottom to a depth 18.25 cm were series of fluctuations in magnetic susceptibility but a change was observed between the depths of 18.25 - 15.25 cm where there was a sort of steady flat state of magnetic susceptibility. The fluctuations resurged from 15.25-8.25cm while the surface peak emerged from 8.25cm upward. In terms of the magnetic parameters ARM, SIRM and Soft there were no visible changes from the bottom of the core to the depth of about 5cm. At 5 cm a clear surface peak was observed in these parameters. At this depth there was a change in magnetic hardness and a shift in the backfields. Caltun Lake displayed only weak surface peak in concentration and the sediments appeared relatively coarse grained (low SIRM/ARM) and magnetically 'soft' at the surface and magnetically relatively 'hard' down core (Figure 5.15). Although the surface values of χ, ARM and SIRM were relatively high when compared to the basal values, the surface peaks in the magnetic concentration parameters χ, ARM and SIRM were relatively small compared with the other lakes from the region.

Capra Lake

Capra lake core LCp 2 (Figure 5.16 demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of the core; for example from a depth of 3.5 cm. At this peak there was also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters 'soft' and 'hard'. Between the depths of 18 - 21 cm, there was a trough in χ, ARM, SIRM, SIRM/χ, ARM/χ and the soft parameter in Capra lake core LCp 2. There was a peak in SIRM/ARM and the hard parameter at the same depths. Core LCp 3 (from Capra lake) also demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of the core profiles; for example from the depth of 10.5 cm (Figure 5.17). At this peak there is also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters 'soft' and 'hard'. In core LCp 3, the highest value of ARM is 61.84 x $10^{-5}Am^2kg^{-1}$ while that of SIRM is 920.20 x $10^{-5}Am^2kg^{-1}$.

The surface peak in magnetic concentration parameters in core LCp 3 becomes obvious from the depth of 10.5 cm (Figure 5.17) while in the Capra lake core 2 (Figure 5.16), the surface peak becomes apparent from the depth of 3.5 cm. The difference in the levels of the surface peak might be due to the distribution of the magnetic materials within the lake. If such is due to differences in distribution of trace metals then, the geochemistry of the sediments will prove it. The ARM/χ and SIRM/χ values are low in contrast to relatively high values of SIRM/ARM (Figure 5.17). The reverse field ratios also show very little variation. The combination of these parameters indicates the predominance of relatively coarse grain ferromagnetic material at the upper surface of the lake.

Podragu Mare Lake

Core LPm 2 (from Podragu Mare Lake) showed a small peak at the base in the main magnetic parameters X, ARM and SIRM (Figure 5.18). The down core profile demonstrated a steady increase in the magnetic concentration parameters X, ARM and SIRM from the depth of 15 cm to the top of the core. There was a sharp surface spike from the depth of 1.5 cm in the magnetic concentration parameters χ, ARM and SIRM. There was a surface spike and also a basal spike at such depths mentioned above in the magnetic soft profile but a sharp correspondence decrease in magnetic hardness was not obvious. Down the profiles of the magnetic soft and hard were some corresponding decrease in magnetic softness and increase in magnetic hardness (for example at the depths of 2.25 cm, 9.25 cm and 15.25 cm).

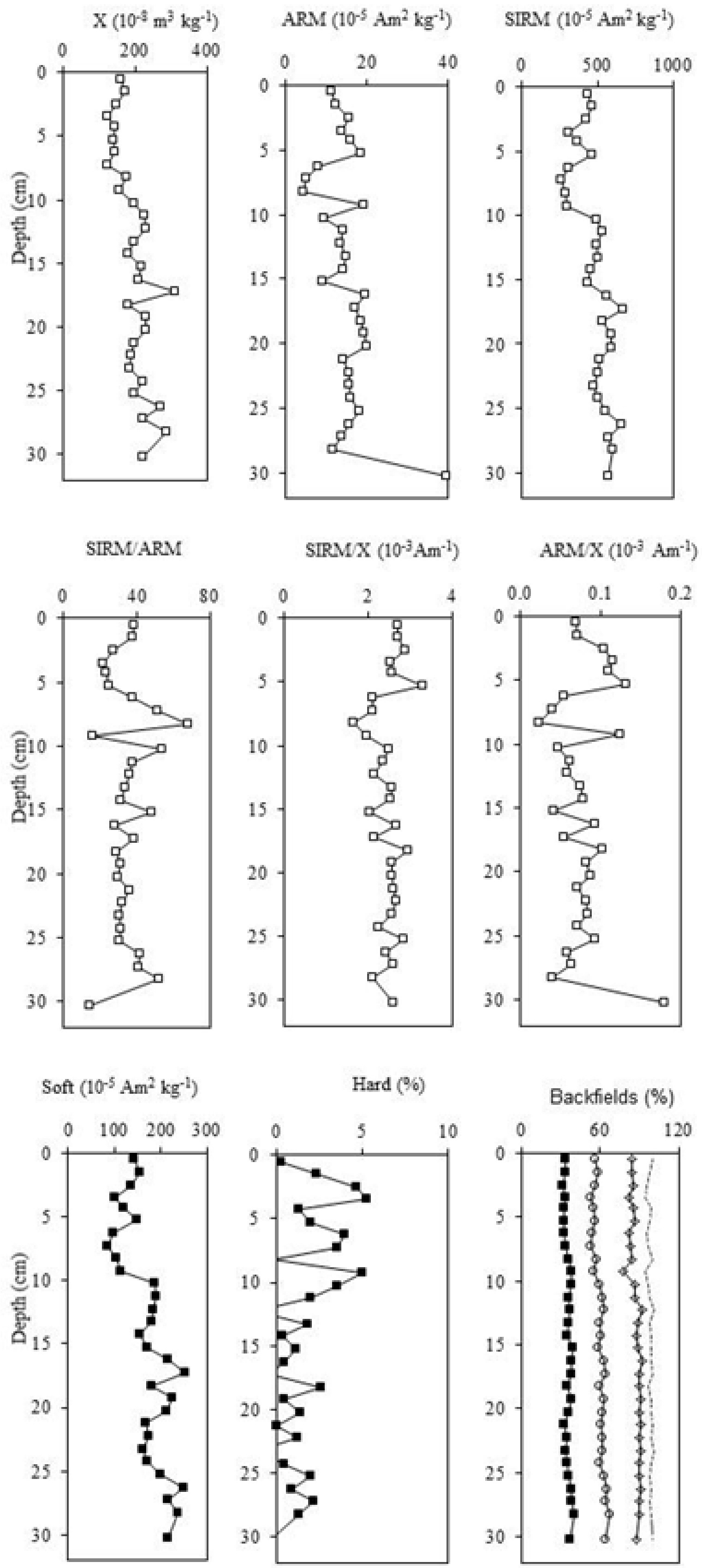


Figure 5.13: Down core variations of sediment magnetic parameters of Balea Lake (Core 1: LBa 1)

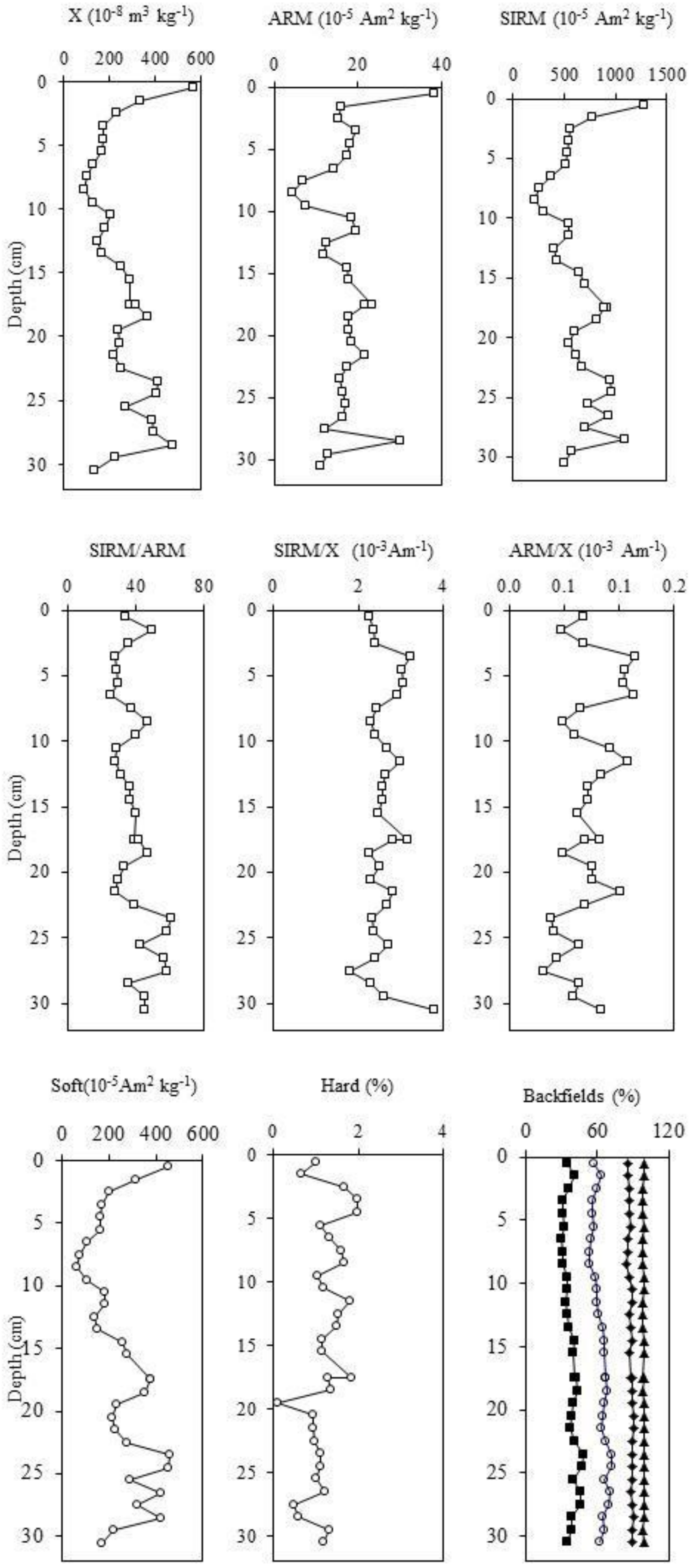


Figure 5.14: Down core variations of sediment magnetic parameters of Balea Lake (Core 4: LBa 4)

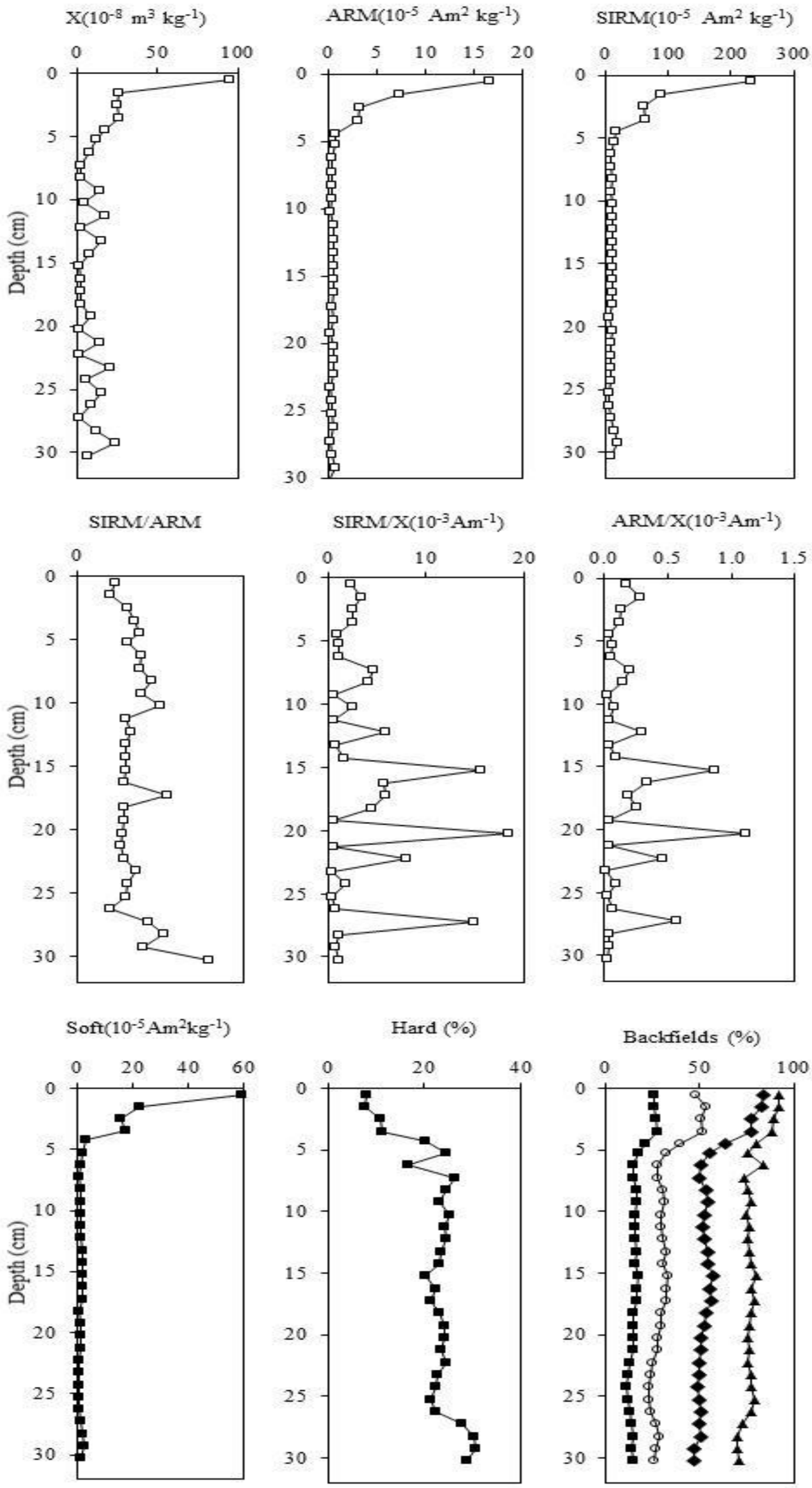


Figure 5.15: Down core variations of sediment magnetic parameters of Caltun Lake (Core 2: LCt 2)

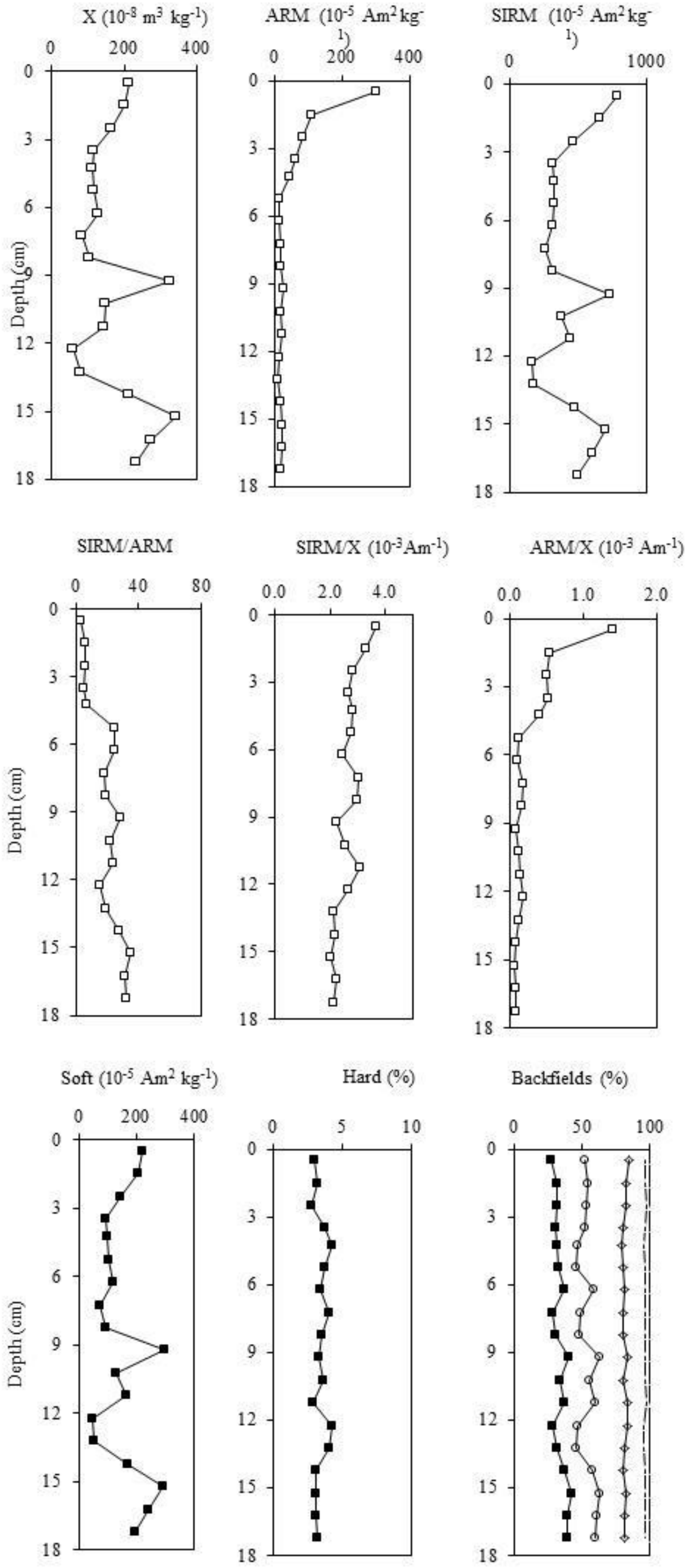


Figure 5.16: Down core variations of sediment magnetic parameters of Capra Lake (Core 2: LCp 2)

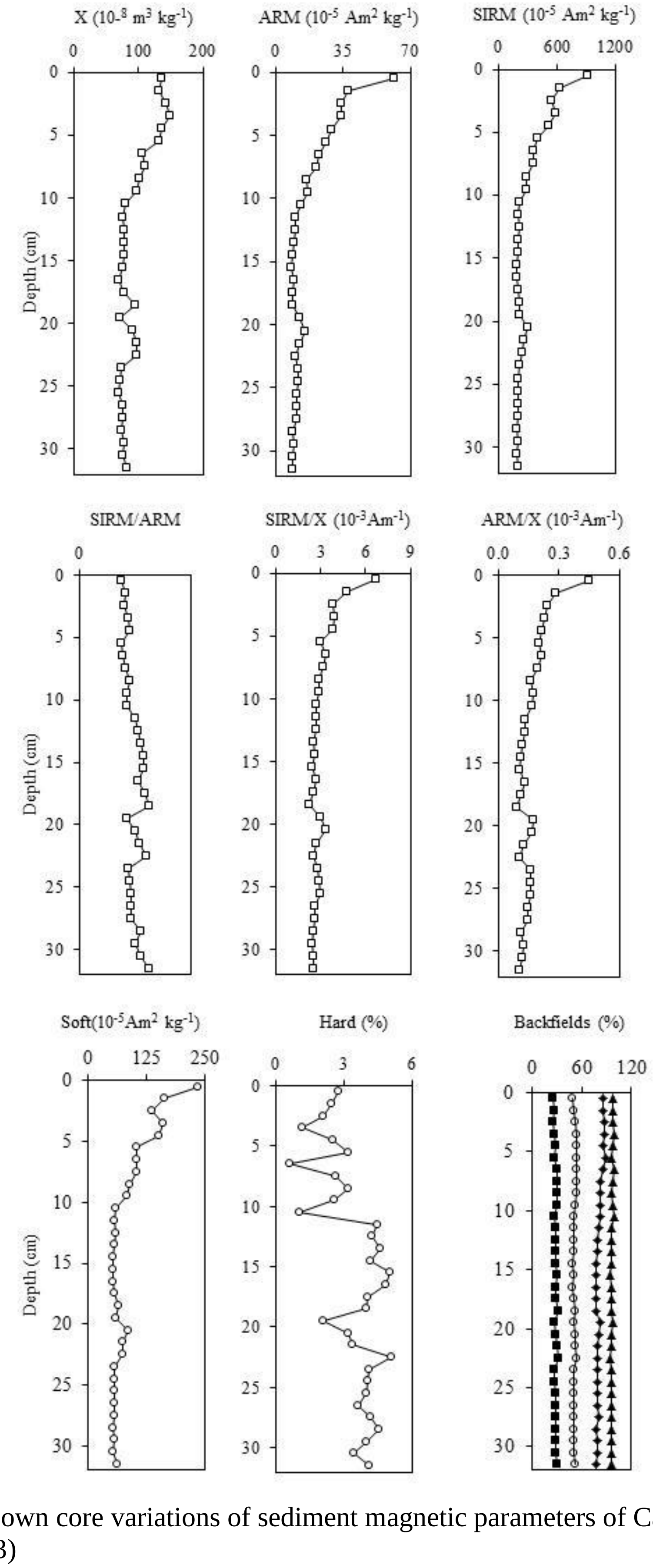


Figure 5.17: Down core variations of sediment magnetic parameters of Capra Lake (Core 3: LCp 3)

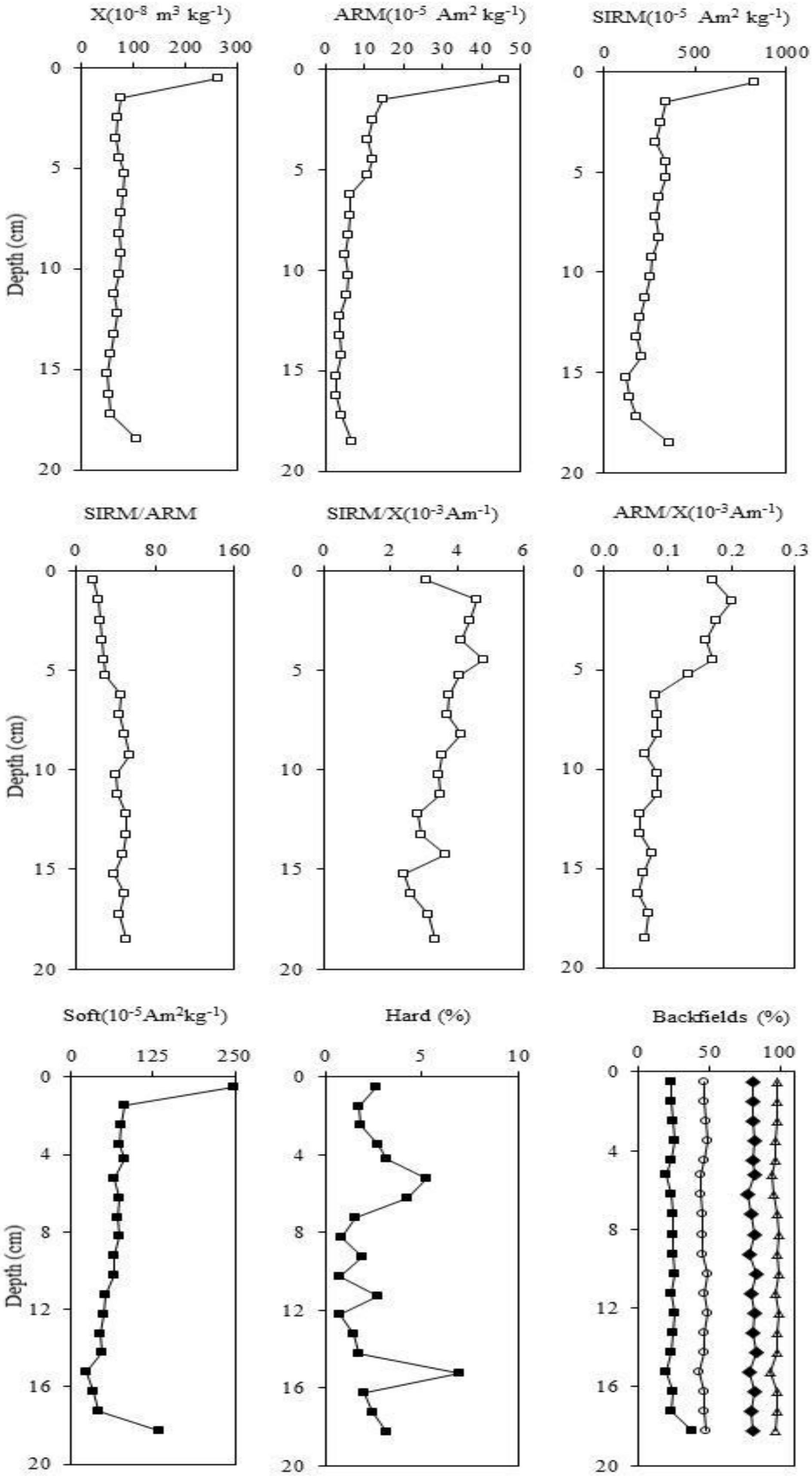


Figure 5.18: Down core variations of sediment magnetic parameters of Podragu Mare Lake (Core 2: LPm 2)

5.3.2 Mineral magnetic measurements of lake sediments in the Rodna/Maramures region

Down core profiles of key magnetic parameters and selected ratios between them are shown for the Rodna lakes in Figures 5.19-5.26.

Bila Lake

Bila Lake (core LB 2) demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of the core profiles; for example from the depth of 4.5 cm (Figure 5.19). At this peak there was also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’. These sediments appeared relatively coarse grained (low SIRM/ARM) and magnetically ‘soft’ at the surface and magnetically ‘hard’ down core. The surface value of SIRM/ARM ratio is 13.87 and the bottom value is 30.92.

Buhaiescu-3 Lake

In this short core (Buhaiescu-3 Lake core LB 3-2) magnetic concentration parameters χ, ARM and SIRM displayed only a small surface increase. Little fluctuation was observed at lower depths. The sediments are relatively coarse grained (low SIRM/ARM) and appeared magnetically ‘soft’ (Figure 5.20). The SIRM profile and the soft profile both have an “S” shape down their profiles.

Lala Mare Lake

Lala Mare Lake (core LLM 2) displayed no surface peak in concentration of the magnetic parameters χ, ARM and SIRM and the sediments appeared relatively coarse grained (low SIRM/ARM) and magnetically ‘soft’ throughout (Figure 5.21). Lala Mare Lake was located at an exposed site. It is relatively shallow therefore; surface sediments appear to have been affected by wind induced disturbance. Lala Mare Lake exhibits magnetic concentration values that are relatively low (e.g. the highest values of χ, ARM and SIRM are 17.0×10^{-8} m^3 kg^{-1}, 25.9×10^{-5} Am^2 kg^{-1} and 163×10^{-5} Am^2 kg^{-1} respectively.

Pietrosul Lake

LP 1 (sampled in 2006) demonstrated a surface peak of magnetic concentration parameters χ, ARM and SIRM from the depth of 1.5cm and a decrease in magnetic concentration down the profile (Figure 5.22). At this peak there was also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’. In Pietrosul lake core LP 1 sampled in 2008 (Figure 5.23), the surface increase commenced from the depth of 2.5 cm. Also at this peak there was a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’. The difference in the levels of the surface peak might be due to the distribution of the magnetic materials within the lake. There are apparent variations in the magnitude of the magnetic parameters X, ARM and SIRM in the Pietrosul lake cores sampled in 2006 and the one sampled in 2008. For example, the highest value of χ is 25.27 x 10^{-8} m^3 kg^{-1}, ARM is 21.17 x 10^{-5} Am^2 kg^{-1} while that of SIRM is 229.56 x 10^{-5} Am^2 kg^{-1} for Pietrosul lake core 1 sampled in 2006 whereas, the highest values of χ, ARM and SIRM for Pietrosul lake core 1 sampled in 2008 are 23.5 x 10^{-8} m^3 kg^{-1}, 11.3 x 10^{-5} Am^2 kg^{-1} and 134 x 10^{-5} Am^2 kg^{-1} respectively.

Stiol Lake

Stiol Lake (LS 2) demonstrated the input of magnetically clean, bank material which is effectively ‘burying’ the magnetically coarse grained peak (Figure 5.24). An observation of the Stiol Lake mineral magnetic profiles indicates that the subsurface peak may reflect the impact of the artificial rise in the lake’s level. The concentration values of the magnetic parameters χ, ARM and SIRM are relatively low compared to Pietrosul. Stiol is a coarse grain lake with a surface SIRM/ARM ratio of 6.5

Vinderel Lake

Two cores were magnetically characterised in Vinderel Lake. LV 3 (sampled in 2006) demonstrated a surface peak from the depth of 6.5 cm. At this peak there was also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’ except that the shift in the parameter hard was only clear towards the bottom. LV1 (sampled in 2008) displayed clear peaks in the concentration parameters χ, ARM and

SIRM from the depth of 6.5 cm. Both cores LV 3 (sampled in 2006) and LV1 (sampled in 2008) appeared relatively coarse grained (low SIRM/ARM) and magnetically ‘soft’ at the surface and magnetically ‘hard’ down core (Figures 5.25 and 5.26). Although, the sediment core sampled in summer 2008 was longer than the sample taken in 2006 there was a clear correlation between the two cores as shown by the profiles.

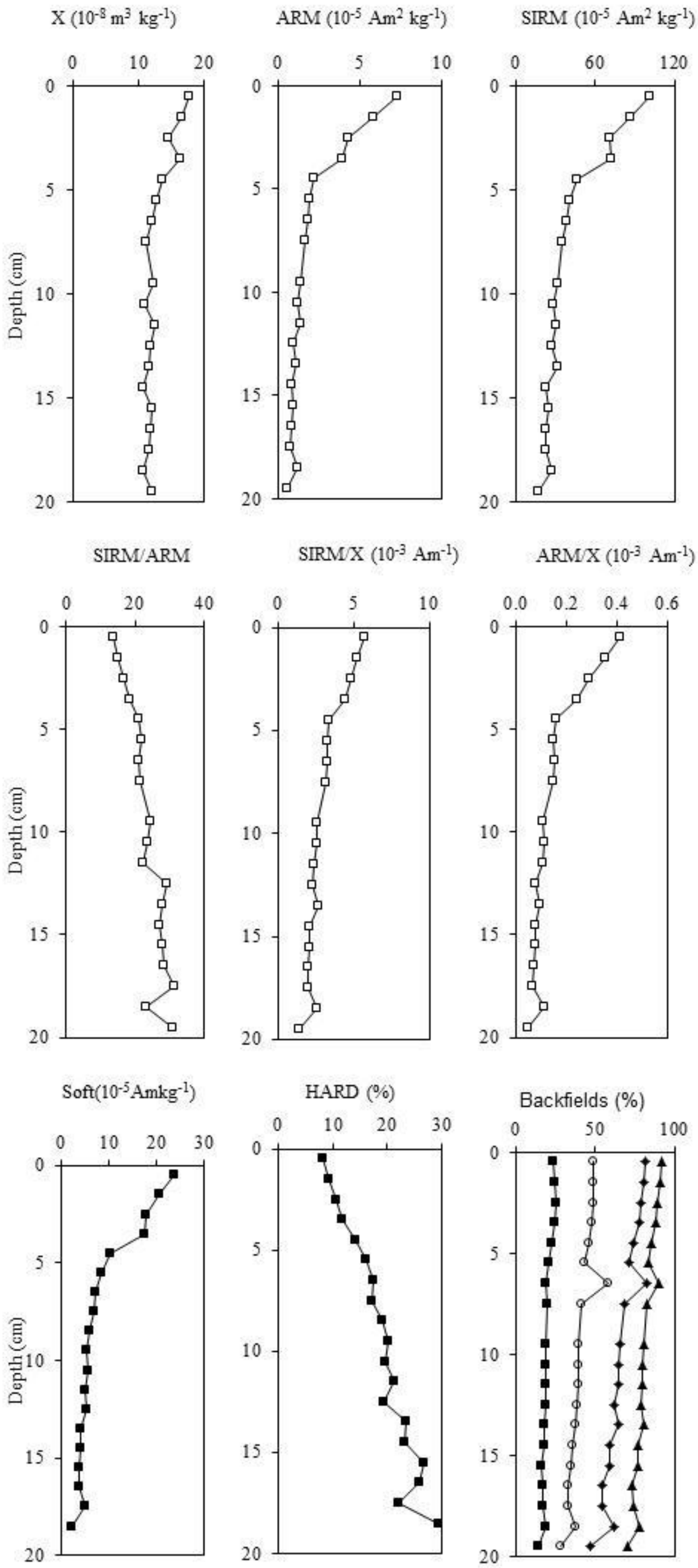


Figure 5.19: Down core variations of sediment magnetic parameters of Bila Lake (Core 2: LB 2) – sampled in 2006

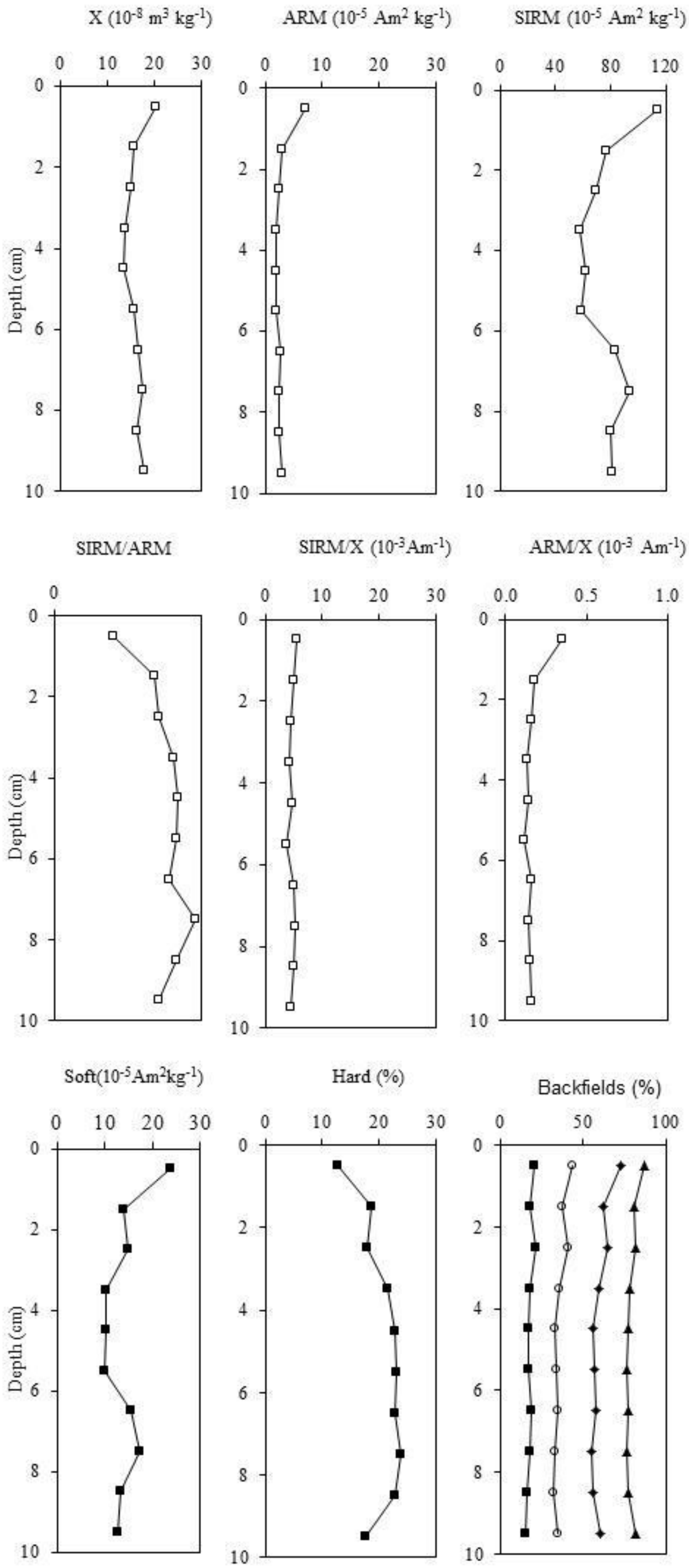


Figure 5.20: Down core variations of sediment magnetic parameters of Buhaiescu-3 Lake (Core 2: LB-3:2) – sampled in 2006

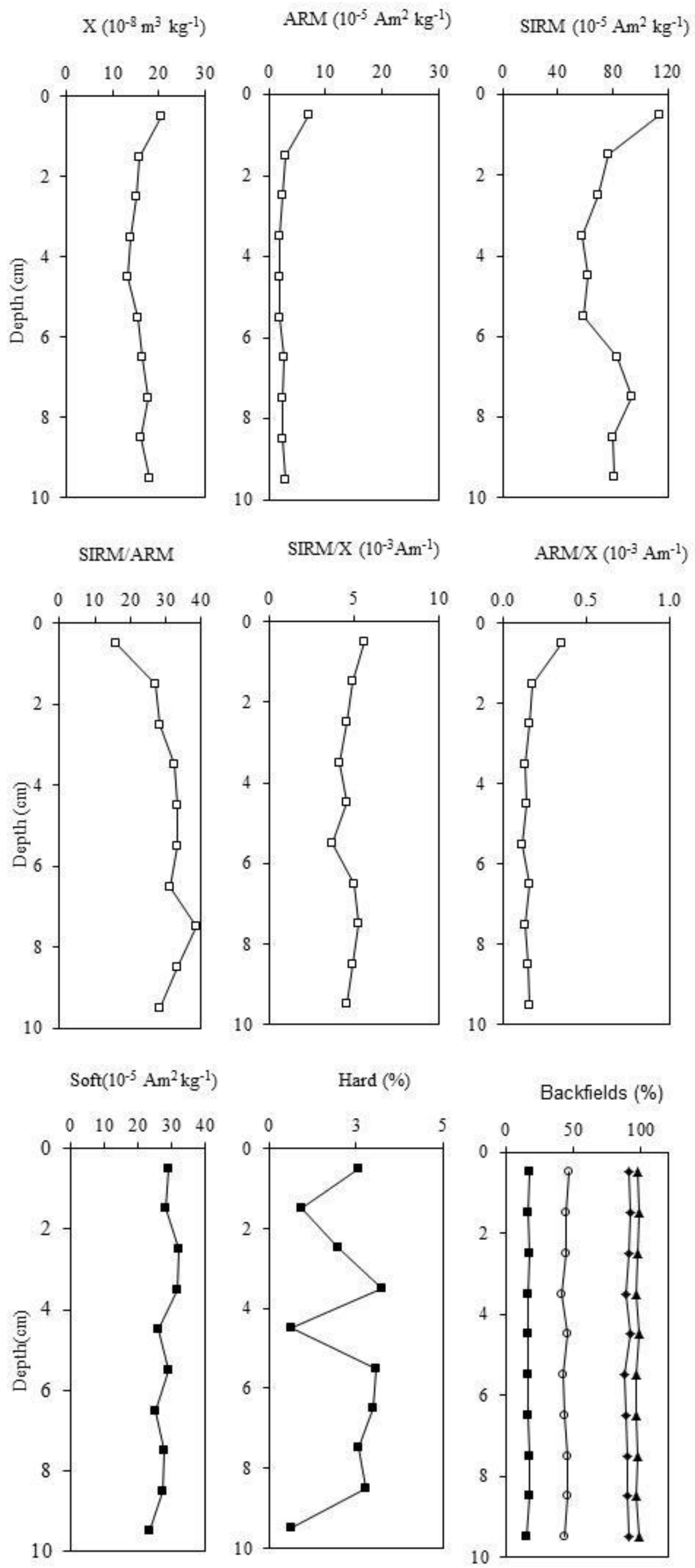


Figure 5.21: Down core variations of sediment magnetic parameters of Lala Mare Lake (Core 1: LLM 2) – sampled in 2006

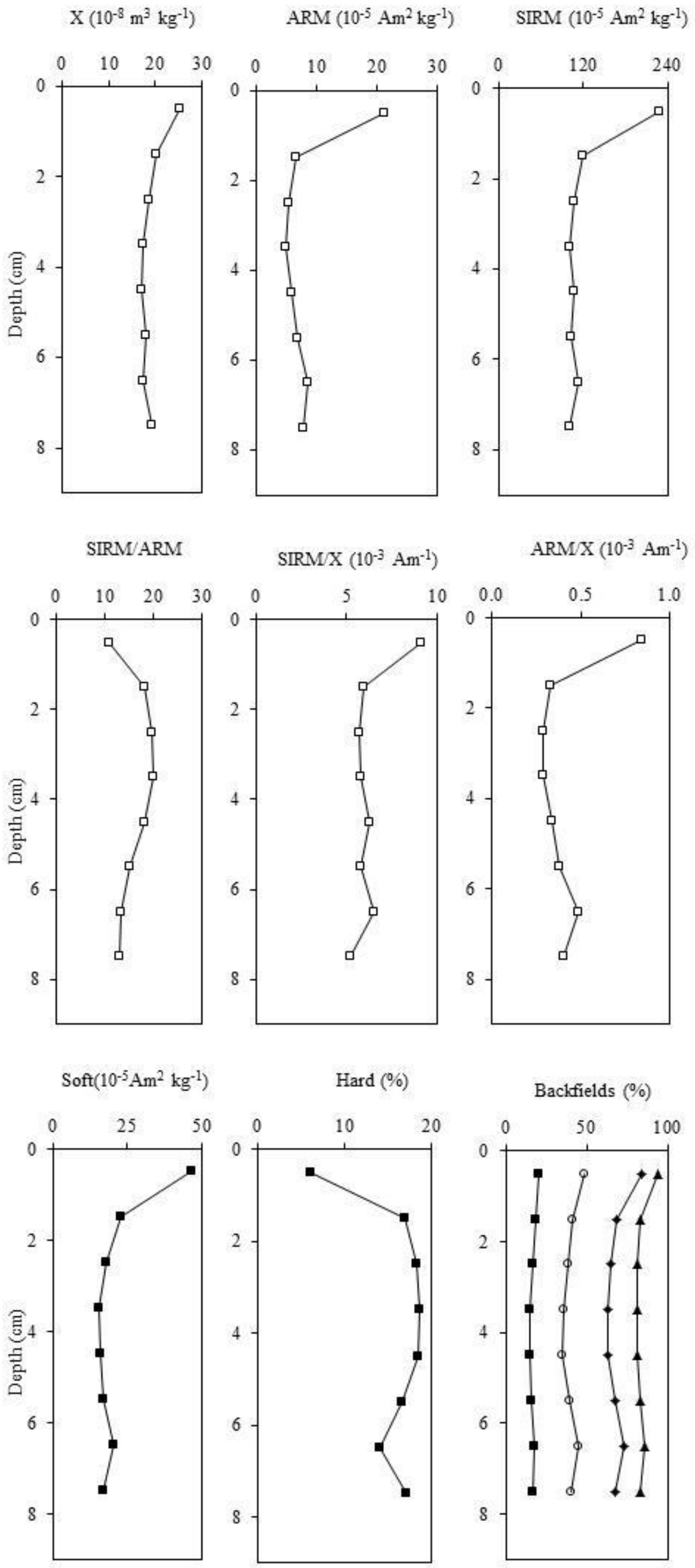


Figure 5.22: Down core variations of sediment magnetic parameters of Pietrosul Lake (Core 1: LP1) – sampled in 2006

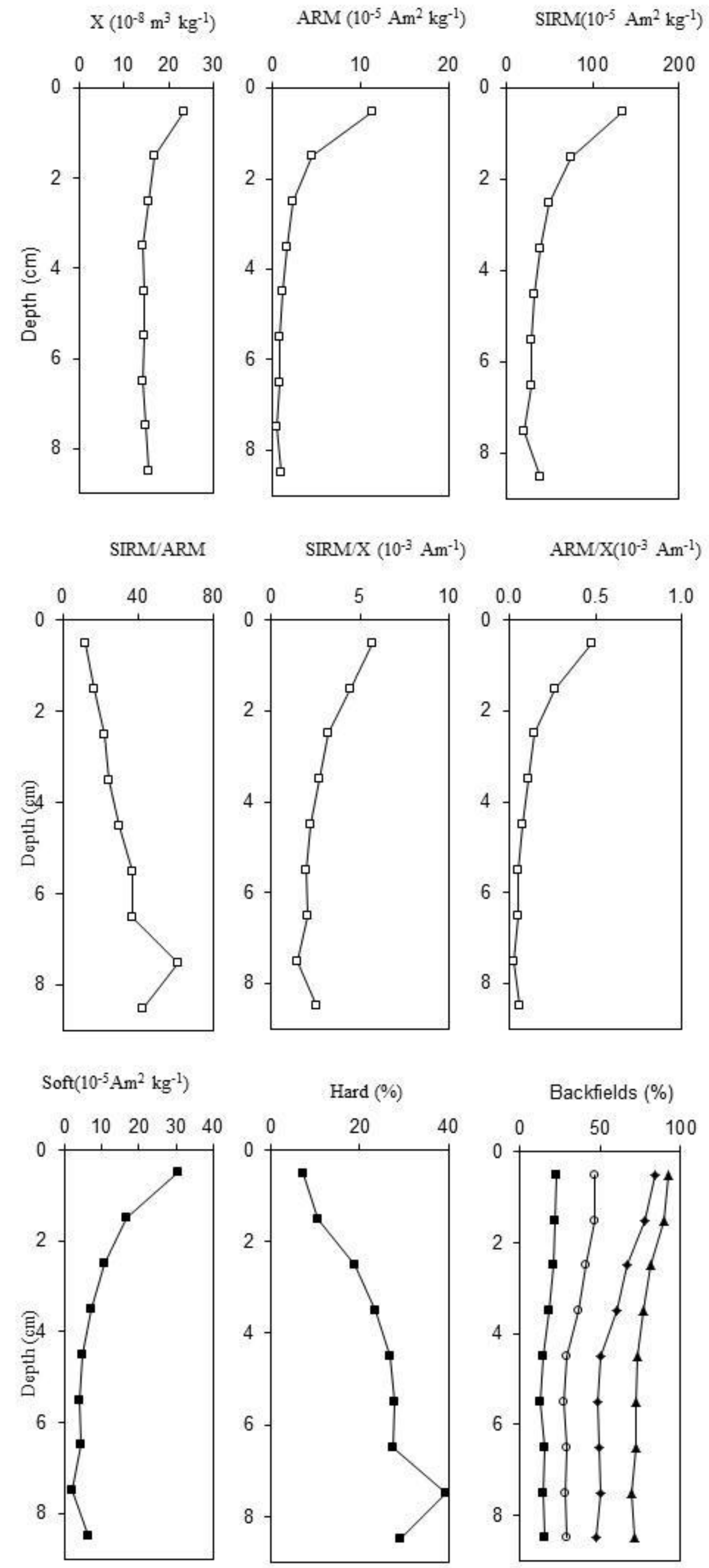


Figure 5.23: Down core variations of sediment magnetic parameters of Pietrosul Lake (Core 1: LP1) – sampled in 2008

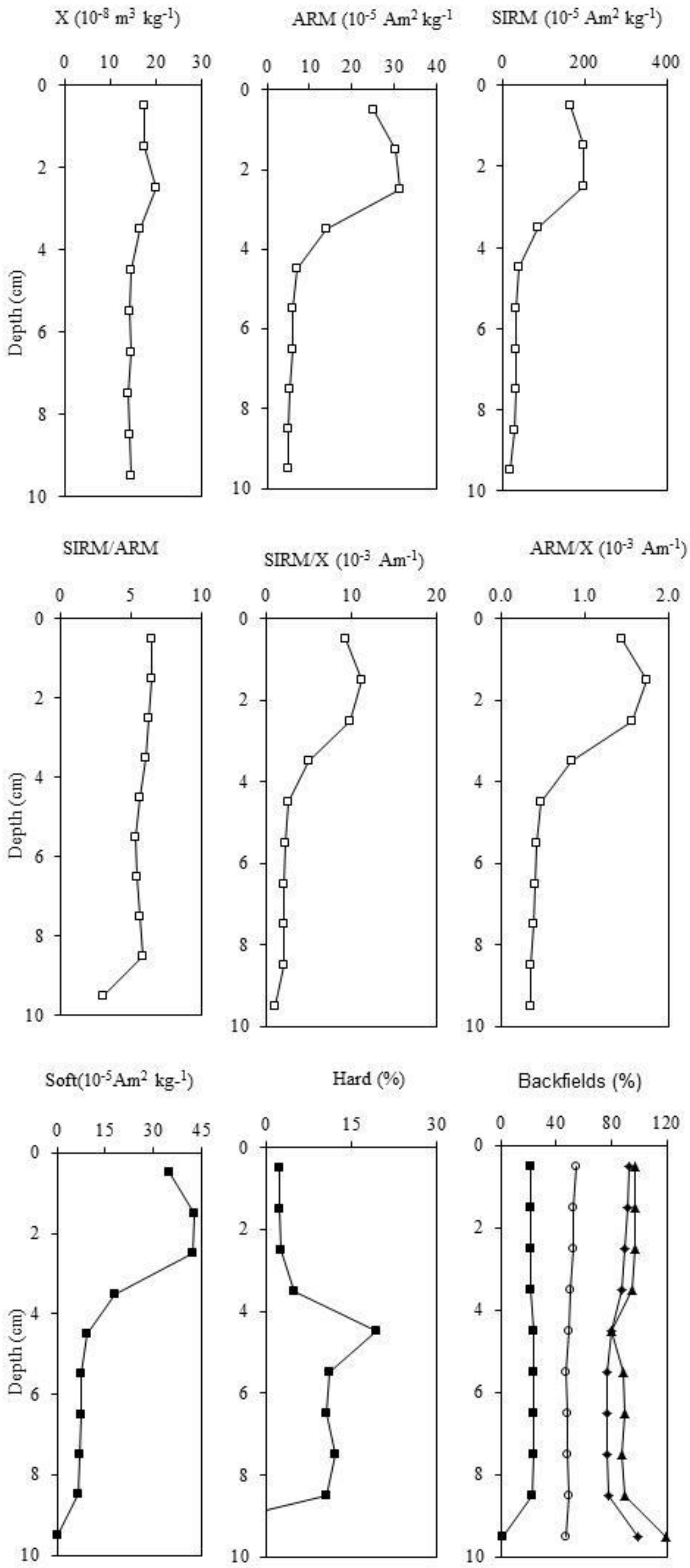


Figure 5.24: Down core variations of sediment magnetic parameters of Stiol Lake (Core 1: LS 2) – sampled in 2006

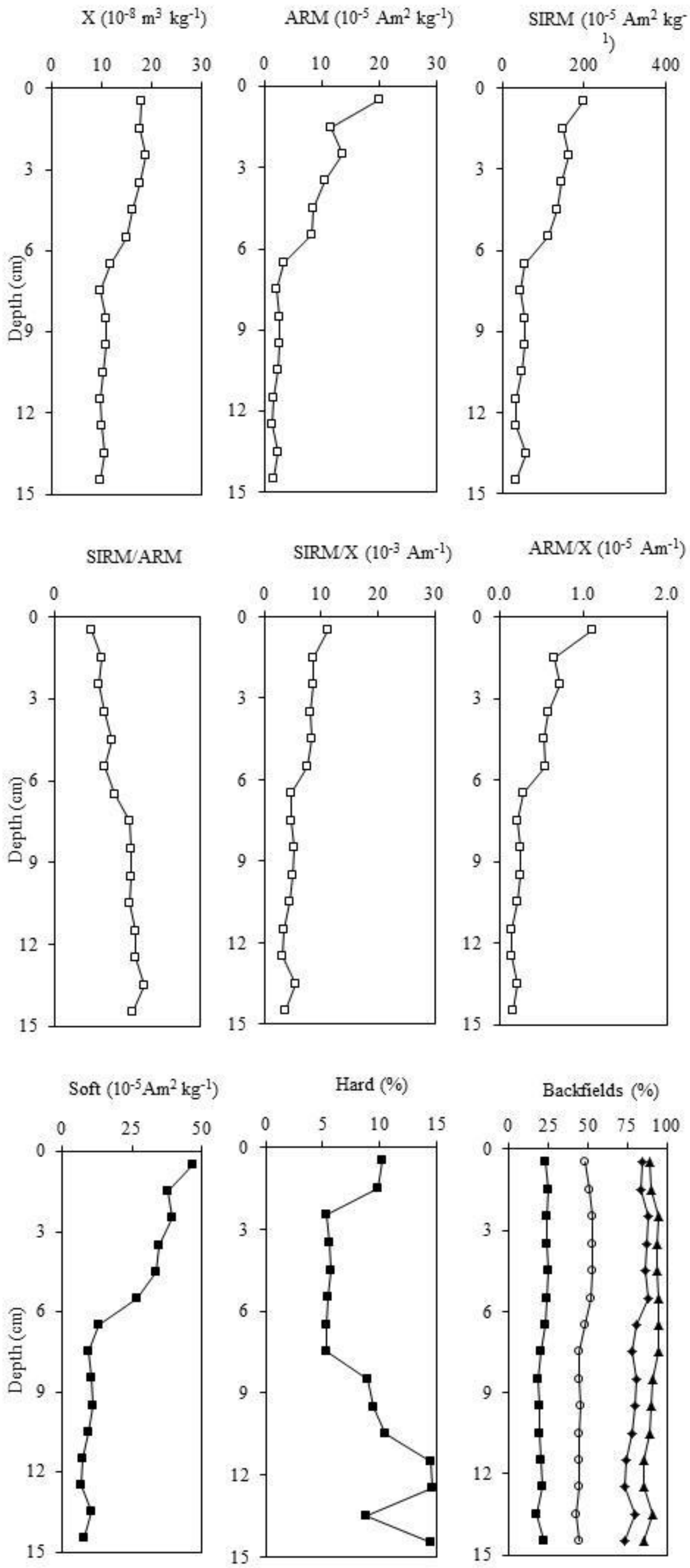


Figure 5.25: Down core variations of sediment magnetic parameters of Vinderel Lake (Core 3: LV 3) – sampled in 2006

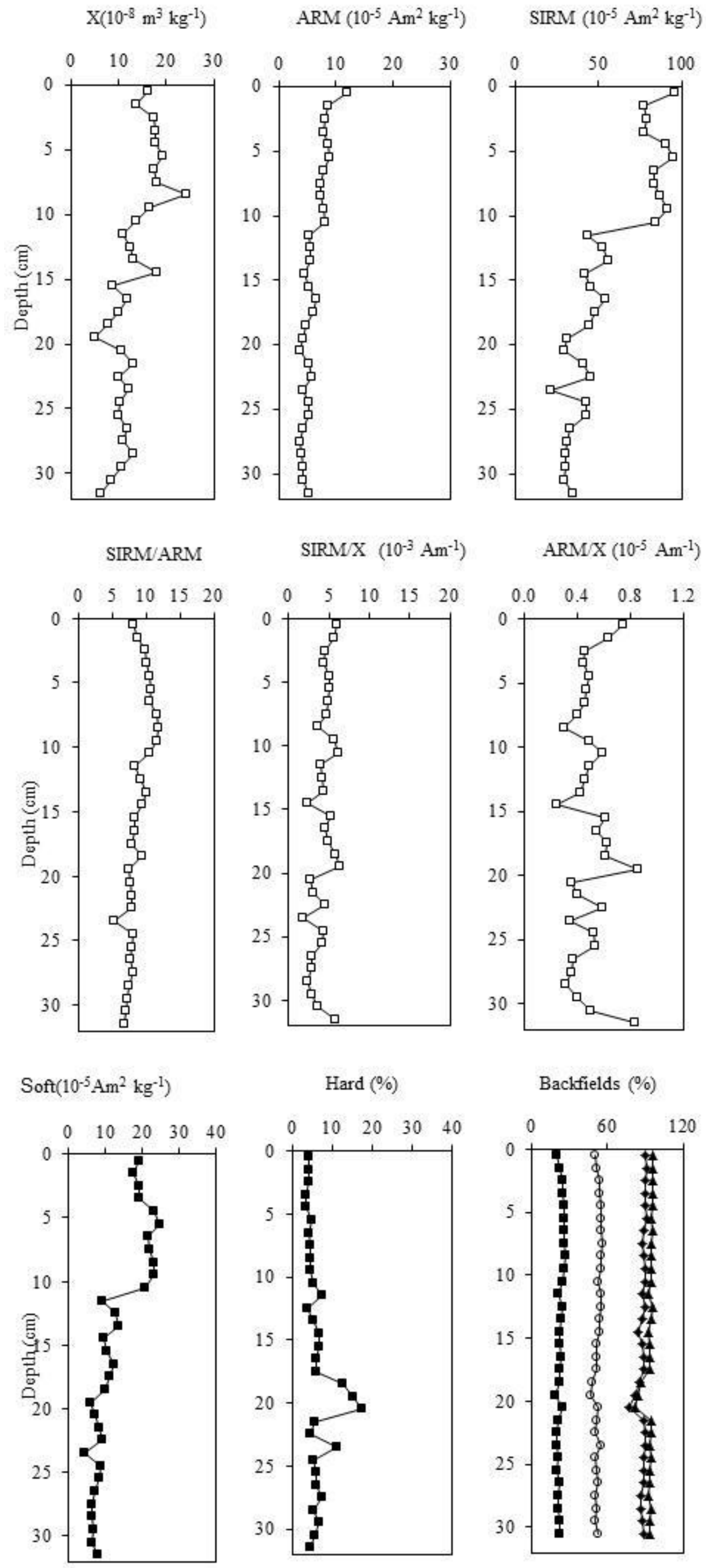


Figure 5.26: Down core variations of sediment magnetic parameters of Vinderel Lake (Core 1: LV 1) – sampled in 2008

5.3.3 Summary of the mineral magnetic measurements

The key findings of the magnetic measurements are that the magnetic concentration parameters χ, ARM and SIRM are similar between cores from the same lake (when the profiles were compared visually) and are comparable among the lakes in the Fagaras region except for Caltun Lake which had relatively low values for all magnetic concentrations. Although the core LBa 1 (from Balea Lake) did not demonstrate surface increase in the magnetic concentration parameters χ, ARM and SIRM, its peaks and troughs are slightly comparable to LBa 4 (from Balea Lake). When the data were subjected to correlation analysis, χ showed signicant correlation with ARM, SIRM, SIRM/ARM, ARM/X, SOFT, 20mT and 40mT in LBa 4 (0.626, 0.950, 0.557, -0.517, 0.941, 0.742 and 0.635 respectively). In LCp 3 (Capra Lake core 3) χ showed strong correlation with SIRM, SOFT, HARD, 20mT, 40mT and 300mT (0.896, 0.979, -0.632, 0.742, 0.822 and 0.632 respectively). While in LCt 2, (Caltun Lake core 2) χ correlated significantly with ARM, SIRM, SOFT, HARD, 20mT, 40mT, 100mT and 300mT (0.923, 0.934, 0.934, -0.681, 0.606, 0.587, 0.714 and 0.681 in order). χ showed strong correlation with ARM, SIRM, SOFT and 20mT LPm 2 (0.951, 0.953, 0.978 and 0.627 rerspectively). All correlations are at 0.01 significant levels (Tables 5.9a and 5.9b).

Table 5.9a: Correlation of magnetic parameters of lake sediments of the lakes in the Fagaras region

Balea Lake Core 4		Lba4_ X	Lba4_ ARM	Lba4_ SIRM	Lba4_ SIRM_ ARM	Lba4_ SIRM_ X	Lba4_ ARM_ X	Lba4_ SOFT	Lba4_ HARD	Lba4_ 20mT	Lba4_ 40mT	Lba4_ 100mT	Lba4_ 300mT
Lba4_ X	Pears	1	.626**	.950**	.557**	-.442*	-.517**	.941**	-.397*	.742**	.635**	.302	.397*
	Sig. (2-		.000	.000	.002	.016	.004	.000	.033	.000	.000	.112	.033
	N		29	29	29	29	29	29	29	29	29	29	29
Lba4_ ARM	Pears		1	.752**	-.256	.237	.293	.576**	-.083	.161	.154	.239	.083
	Sig. (2-			.000	.180	.216	.123	.001	.669	.405	.426	.213	.669
	N			29	29	29	29	29	29	29	29	29	29
Lba4_ SIRM	Pears			1	.431*	-.160	-.336	.965**	-.261	.701**	.626**	.345	.261
	Sig. (2-				.020	.406	.074	.000	.172	.000	.000	.067	.172
	N				29	29	29	29	29	29	29	29	29
Lba4_ SIRM_ ARM	Pears				1	-.625**	-.916**	.611**	-.287	.776**	.655**	.146	.287
	Sig. (2-					.000	.000	.000	.132	.000	.000	.449	.132
	N					29	29	29	29	29	29	29	29
Lba4_ SIRM_ X	Pears					1	.816**	-.263	.529**	-.440*	-.320	-.026	-.529**
	Sig. (2-						.000	.168	.003	.017	.091	.894	.003
	N						29	29	29	29	29	29	29
Lba4_ ARM_ X	Pears						1	-.496**	.408*	-.689**	-.573**	-.083	-.408*
	Sig. (2-							.006	.028	.000	.001	.670	.028
	N							29	29	29	29	29	29
Lba4_ SOFT	Pears							1	-.313	.851**	.773**	.429*	.313
	Sig. (2-								.098	.000	.000	.020	.098
	N								29	29	29	29	29
Lba4_ HARD	Pears								1	-.457*	-.439*	-.387*	-1.000**
	Sig. (2-									.013	.017	.038	0.000
	N									29	29	29	29
Lba4_ 20mT	Pears									1	.964**	.625**	.457*
	Sig. (2-										.000	.000	.013
	N										29	29	29
Lba4_ 40mT	Pears										1	.755**	.439*
	Sig. (2-											.000	.017
	N											29	29
Lba4_ 100mT	Pears											1	.387*
	Sig. (2-												.038
	N												29
Lba4_ 300m	Pears												1
	Sig. (2-												
	N												

Capra Lake Core 2		LCp2_ X	LCp2_ ARM	LCp2_ SIRM	LCp2_ SIRM_ ARM	LCp2_ SIRM_ X	LCp2_ ARM_ X	LCp2_ SOFT	LCp2_ HARD	LCp2_ 20mT	LCp2_ 40mT	LCp2_ 100mT	LCp2_ 300mT
LCp2_ X	Pearso	1	.177	.896**	.444	-.336	.014	.979**	-.632**	.742**	.822**	.403	.632**
	Sig. (2-		.496	.000	.074	.188	.957	.000	.007	.001	.000	.108	.007
	N		17	17	17	17	17	17	17	17	17	17	17
LCp2_ ARM	Pearso		1	.547*	-.630**	.675**	.975**	.277	-.340	-.398	-.022	.448	.340
	Sig. (2-			.023	.007	.003	.000	.281	.181	.114	.933	.072	.181
	N			17	17	17	17	17	17	17	17	17	17
LCp2_ SIRM	Pearso			1	.105	.098	.393	.951	-.719	.462	.698	.523	.719
	Sig. (2-				.689	.709	.119	.000	.001	.062	.002	.031	.001
	N				17	17	17	17	17	17	17	17	17
LCp2_ SIRM_ ARM	Pearso				1	-.719**	-.772**	.362	-.155	.799**	.516*	-.010	.155
	Sig. (2-					.001	.000	.154	.554	.000	.034	.970	.554
	N					17	17	17	17	17	17	17	17
LCp2_ SIRM_ X	Pearso					1	.725**	-.170	-.087	-.667**	-.358	.161	.087
	Sig. (2-						.001	.513	.740	.003	.158	.537	.740
	N						17	17	17	17	17	17	17
LCp2_ ARM_ X	Pearso						1	.111	-.214	-.537*	-.164	.343	.214
	Sig. (2-							.672	.409	.026	.529	.178	.409
	N							17	17	17	17	17	17
LCp2_ SOFT	Pearso							1	-.694**	.699**	.829**	.483*	.694**
	Sig. (2-								.002	.002	.000	.050	.002
	N								17	17	17	17	17
LCp2_ HARD	Pearso								1	-.469	-.714**	-.443	-1.000**
	Sig. (2-									.058	.001	.075	0.000
	N									17	17	17	17
LCp2_ 20mT	Pearso									1	.854**	.167	.469
	Sig. (2-										.000	.521	.058
	N										17	17	17
LCp2_ 40mT	Pearso										1	.423	.714**
	Sig. (2-											.091	.001
	N											17	17
LCp2_ 100mT	Pearso											1	.443
	Sig. (2-												.075
	N												17

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

Table 5.9b: Correlation of magnetic parametetrs of lake sediments of the lakes in the Fagaras region

Caltun Lake Core 2		LCt2_X	LCt2_ARM	LCt2_SIRM	LCt2_SIRM_ARM	LCt2_SIRM_X	LCt2_ARM_X	LCt2_SOFT	LCt2_HARD	LCt2_20mT	LCt2_40mT	LCt2_100mT	LCt2_300mT
LCt2_X	Pears	1	.923**	.934**	-.306	-.310	-.247	.934**	-.681**	.606**	.587**	.714**	.681**
	Sig. (2-		.000	.000	.107	.101	.197	.000	.000	.000	.001	.000	.000
	N		29	29	29	29	29	29	29	29	29	29	29
LCt2_ARM	Pears		1	.992**	-.357	-.077	-.008	.989**	-.746**	.658**	.666**	.796**	.746**
	Sig. (2-			.000	.057	.690	.966	.000	.000	.000	.000	.000	.000
	N			29	29	29	29	29	29	29	29	29	29
LCt2_SIRM	Pears			1	-.305	-.084	-.025	.999**	-.756**	.699**	.697**	.817**	.756**
	Sig. (2-				.108	.665	.897	.000	.000	.000	.000	.000	.000
	N				29	29	29	29	29	29	29	29	29
LCt2_SIRM_ARM	Pears				1	.027	-.150	-.309	.417*	-.167	-.203	-.283	-.417*
	Sig. (2-					.889	.438	.103	.024	.387	.290	.137	.024
	N					29	29	29	29	29	29	29	29
LCt2_SIRM_X	Pears					1	.969**	-.093	.152	-.093	-.079	-.106	-.152
	Sig. (2-						.000	.633	.431	.630	.685	.584	.431
	N						29	29	29	29	29	29	29
LCt2_ARM_X	Pears						1	-.035	.062	-.049	-.026	-.041	-.062
	Sig. (2-							.857	.751	.802	.895	.831	.751
	N							29	29	29	29	29	29
LCt2_SOFT	Pears							1	-.774**	.722**	.720**	.836**	.774**
	Sig. (2-								.000	.000	.000	.000	.000
	N								29	29	29	29	29
LCt2_HARD	Pears								1	-.816**	-.836**	-.903**	-1.000**
	Sig. (2-									.000	.000	.000	0.000
	N									29	29	29	29
LCt2_20mT	Pears									1	.992**	.960**	.816**
	Sig. (2-										.000	.000	.000
	N										29	29	29
LCt2_40mT	Pears										1	.973**	.836**
	Sig. (2-											.000	.000
	N											29	29
LCt2_100mT	Pears											1	.903**
	Sig. (2-												.000
	N												29

Podragu Mare Lake Core 2		LPM2_X	LPM2_ARM	LPM2_SIRM	LPM2_SIRM_ARM	LPM2_SIRM_X	LPM2_ARM_X	LPM2_SOFT	LPM2_HARD	LPM2_20mT	LPM2_40mT	LPM2_100mT	LPM2_300mT
LPM2_X	Pearso	1	.951**	.953**	-.499*	-.079	.394	.978**	-.011	.627**	.106	.282	.022
	Sig. (2-		.000	.000	.041	.764	.117	.000	.966	.007	.687	.272	.934
	N		17	17	17	17	17	17	17	17	17	17	17
LPM2_ARM	Pearso		1	.969**	-.729**	.129	.648**	.972**	.033	.549*	.174	.327	-.101
	Sig. (2-			.000	.001	.621	.005	.000	.899	.022	.505	.200	.701
	N			17	17	17	17	17	17	17	17	17	17
LPM2_SIRM	Pearso			1	-.639**	.226	.604*	.987**	-.030	.554*	.101	.271	-.068
	Sig. (2-				.006	.383	.010	.000	.909	.021	.699	.293	.796
	N				17	17	17	17	17	17	17	17	17
LPM2_SIRM_ARM	Pearso				1	-.477	-.927**	-.588*	-.259	-.123	-.233	-.262	.272
	Sig. (2-					.053	.000	.013	.316	.639	.369	.310	.291
	N					17	17	17	17	17	17	17	17
LPM2_SIRM_X	Pearso					1	.712**	.109	-.134	-.131	.071	.028	-.322
	Sig. (2-						.001	.677	.608	.615	.786	.914	.207
	N						17	17	17	17	17	17	17
LPM2_ARM_X	Pearso						1	.534*	.042	.147	.279	.228	-.293
	Sig. (2-							.027	.872	.575	.279	.380	.254
	N							17	17	17	17	17	17
LPM2_SOFT	Pearso							1	-.086	.656**	.175	.288	-.023
	Sig. (2-								.743	.004	.502	.263	.930
	N								17	17	17	17	17
LPM2_HARD	Pearso								1	-.634**	-.631**	-.395	-.122
	Sig. (2-									.006	.007	.116	.640
	N									17	17	17	17
LPM2_20mT	Pearso									1	.652**	.311	.199
	Sig. (2-										.005	.224	.444
	N										17	17	17
LPM2_40mT	Pearso										1	.543*	-.070
	Sig. (2-											.024	.789
	N											17	17
LPM2_100mT	Pearso											1	-.564*
	Sig. (2-												.018
	N												17

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

The magnetic concentration parameters χ, ARM and SIRM are similar between cores of the same lake and are slightly similar among most of the lakes in the Rodna region as well (compared based on the visual outlook of the profiles). All cores from Rodna region show clear surface peak in the magnetic concentration parameters χ, ARM and SIRM, except Stiol Lake which demonstrated subsurface peak and Lala Mare Lake which did not show any clear fluctuations in the magnetic concentration parameters χ, ARM and SIRM. All the lakes in Rodna region demonstrated significant correlations in all the magnetic parameters except LB-3 (Buhaiescu Lake 3) that showed correlations only in χ, ARM, SIRM, SOFT (0.815, 0.924 and 0.846) and LLM 2 (Lala Mare Lake core 2) that showed no signicant correlation in any of the parameters (Tables 5.10a-c). In all the sites that demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of their core profiles; at this peak there is also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’ (Figures 5.19-5.26). These features indicated both an increase in magnetic concentration, and a change in grain size and mineralogy; they demonstrated the possible influence of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions.

Table 5.10a: Correlation of magnetic parameters of lake sediments of the lakes in the Rodna/Maramures region

Bila Lake Core 1		LB1_X	LB1_ARM	LB1_SIRM	LB1_SIRM_ARM	LB1_SIRM_X	LB1_ARM_X	LB1_SOFT	LB1_HARD	LB1_20mT	LB1_40mT	LB1_100mT	LB1_300mT
LB1_X	Pearso	1	.937**	.955**	-.793**	.901**	.909**	.954**	-.769**	.875**	-.465	-.888**	-.897**
	Sig. (2-		.000	.000	.000	.000	.000	.000	.000	.000	.052	.000	.000
	N		18	18	18	18	18	18	18	18	18	18	18
LB1_A	Pearso		1	.992**	-.881**	.969**	.992**	.984**	-.823**	.910**	-.520*	-.931**	-.935**
RM	Sig. (2-			.000	.000	.000	.000	.000	.000	.000	.027	.000	.000
	N			18	18	18	18	18	18	18	18	18	18
LB1_SI	Pearso			1	-.891**	.985**	.989**	.997**	-.858**	.931**	-.551*	-.955**	-.958**
RM	Sig. (2-				.000	.000	.000	.000	.000	.000	.018	.000	.000
	N				18	18	18	18	18	18	18	18	18
LB1_SI	Pearso				1	-.927**	-.921**	-.892**	.905**	-.867**	.638**	.913**	.900**
RM_A	Sig. (2-					.000	.000	.000	.000	.000	.004	.000	.000
RM	N					18	18	18	18	18	18	18	18
LB1_SI	Pearso					1	.988**	.984**	-.908**	.940**	-.611**	-.974**	-.971**
RM_X	Sig. (2-						.000	.000	.000	.000	.007	.000	.000
	N						18	18	18	18	18	18	18
LB1_A	Pearso						1	.984**	-.866**	.927**	-.566*	-.955**	-.955**
RM_X	Sig. (2-							.000	.000	.000	.014	.000	.000
	N							18	18	18	18	18	18
LB1_S	Pearso							1	-.849**	.937**	-.539*	-.960**	-.965**
OFT	Sig. (2-								.000	.000	.021	.000	.000
	N								18	18	18	18	18
LB1_H	Pearso								1	-.834**	.833**	.919**	.885**
ARD	Sig. (2-									.000	.000	.000	.000
	N									18	18	18	18
LB1_2	Pearso									1	-.515*	-.973**	-.988**
0mT	Sig. (2-										.029	.000	.000
	N										18	18	18
LB1_4	Pearso										1	.669**	.599**
0mT	Sig. (2-											.002	.009
	N											18	18
LB1_1	Pearso											1	.996**
00mT	Sig. (2-												.000
	N												18
LB1_3	Pearso												1
00mT	Sig. (2-												
	N												

Buhiescu 3 Lake Core 2		LB32_X	LB32ARM	LB32_SIRM	LB32_SIRM_ARM	LB32_SIRM_X	LB32_ARM_X	LB32_SOFT	LB32_HARD	LB32_20mT	LB32_40mT	LB32_100mT	LB32_300mT
LB32_	Pearso	1	.815**	.924**	-.564	.659*	.736*	.846**	-.584	.278	.482	.533	.584
X	Sig. (2-		.004	.000	.090	.038	.015	.002	.077	.437	.158	.112	.077
	N		10	10	10	10	10	10	10	10	10	10	10
LB32_	Pearso		1	.850**	-.874**	.682*	.990**	.898**	-.826**	.535	.791**	.850**	.826**
ARM	Sig. (2-			.002	.001	.030	.000	.000	.003	.111	.006	.002	.003
	N			10	10	10	10	10	10	10	10	10	10
LB32_	Pearso			1	-.548	.894**	.810**	.946**	-.553	.397	.515	.544	.553
SIRM	Sig. (2-				.101	.000	.004	.000	.097	.256	.127	.104	.097
	N				10	10	10	10	10	10	10	10	10
LB32_	Pearso				1	-.380	-.899**	-.649*	.948**	-.523	-.881**	-.952**	-.948**
SIRM_	Sig. (2-					.278	.000	.042	.000	.121	.001	.000	.000
ARM	N					10	10	10	10	10	10	10	10
LB32_	Pearso					1	.692*	.856**	-.376	.410	.403	.398	.376
SIRM_	Sig. (2-						.027	.002	.284	.240	.248	.254	.284
X	N						10	10	10	10	10	10	10
LB32_	Pearso						1	.879**	-.847**	.572	.825**	.880**	.847**
ARM_	Sig. (2-							.001	.002	.084	.003	.001	.002
X	N							10	10	10	10	10	10
LB32_	Pearso							1	-.644*	.666*	.718*	.707*	.644*
SOFT	Sig. (2-								.045	.036	.019	.022	.045
	N								10	10	10	10	10
LB32_	Pearso								1	-.529	-.904**	-.956**	-1.000**
HARD	Sig. (2-									.116	.000	.000	0.000
	N									10	10	10	10
LB32_	Pearso									1	.836**	.721*	.529
20mT	Sig. (2-										.003	.019	.116
	N										10	10	10
LB32_	Pearso										1	.979**	.904**
40mT	Sig. (2-											.000	.000
	N											10	10
LB32_	Pearso											1	.956**
100mT	Sig. (2-												.000
	N												

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

Table 5.10b: Correlation of magnetic parameters of lake sediments of the lakes in the Rodna/Maramures region

Lala Mare Lake Core 2		LLM2_ X	LLM2_ ARM	LLM2_ SIRM	LLM2_ SIRM_ ARM	LLM2_ SIRM_ X	LLM2_ ARM_ X	LLM2_ SOFT	LLM2_ HARD	LLM2_ 20mT	LLM2_ 40mT	LLM2_ 100mT	LLM2_ 300mT
LLM2_ X	Pearso	1	.587	.657*	.391	-.093	-.296	.568	-.041	-.137	-.301	.007	.041
	Sig. (2-		.075	.039	.265	.799	.407	.087	.911	.707	.398	.984	.911
	N		10	10	10	10	10	10	10	10	10	10	10
LLM2_ ARM	Pearso		1	.927**	.165	.669*	.599	.701*	-.045	-.375	-.511	-.049	.045
	Sig. (2-			.000	.649	.034	.067	.024	.902	.286	.131	.893	.902
	N			10	10	10	10	10	10	10	10	10	10
LLM2_ SIRM	Pearso			1	.522	.688*	.435	.859**	.218	-.195	-.580	-.270	-.218
	Sig. (2-				.122	.028	.209	.001	.546	.590	.079	.451	.546
	N				10	10	10	10	10	10	10	10	10
LLM2_ SIRM_ ARM	Pearso				1	.293	-.212	.675*	.661*	.360	-.339	-.576	-.661*
	Sig. (2-					.412	.557	.032	.037	.307	.338	.082	.037
	N					10	10	10	10	10	10	10	10
LLM2	Pearso					1	.872**	.585	.291	-.131	-.448	-.322	-.291
	Sig. (2-						.001	.075	.414	.718	.194	.365	.414
	N						10	10	10	10	10	10	10
LLM2_ ARM_ X	Pearso						1	.251	-.051	-.320	-.282	-.027	.051
	Sig. (2-							.484	.888	.367	.429	.941	.888
	N							10	10	10	10	10	10
LLM2_ SOFT	Pearso							1	.458	.334	-.233	-.245	-.458
	Sig. (2-								.183	.346	.518	.495	.183
	N								10	10	10	10	10
LLM2_ HARD	Pearso								1	.502	-.317	-.844**	-1.000**
	Sig. (2-									.139	.372	.002	0.000
	N									10	10	10	10
LLM2_ 20mT	Pearso									1	.602	-.004	-.502
	Sig. (2-										.066	.992	.139
	N										10	10	10
LLM2_ 40mT	Pearso										1	.703*	.317
	Sig. (2-											.023	.372
	N											10	10
LLM2_ 100mT	Pearso											1	.844**
	Sig. (2-												.002
	N												

Pietrosul Lake Core 1 (2006)		LP1_X	LP1_ ARM	LP1_ SIRM	LP1_ SIRM_ ARM	LP1_ SIRM_ X	LP1_ ARM_ X	LP1_ SOFT	LP1_ HARD	LP1_ 20mT	LP1_ 40mT	LP1_ 100mT	LP1_ 300mT
LP1_X	Pearso	1	.911**	.933**	-.585	.805*	.835**	.949**	-.863**	.833*	.752*	.864**	.863**
	Sig. (2-		.002	.001	.128	.016	.010	.000	.006	.010	.031	.006	.006
	N		8	8	8	8	8	8	8	8	8	8	8
LP1_A RM	Pearso		1	.974**	-.764*	.935**	.985**	.973**	-.983**	.776*	.818*	.958**	.983**
	Sig. (2-			.000	.027	.001	.000	.000	.000	.024	.013	.000	.000
	N			8	8	8	8	8	8	8	8	8	8
LP1_SI RM	Pearso			1	-.608	.963**	.933**	.994**	-.949**	.777*	.752*	.916**	.949**
	Sig. (2-				.110	.000	.001	.000	.000	.023	.031	.001	.000
	N				8	8	8	8	8	8	8	8	8
LP1_SI RM_A RM	Pearso				1	-.575	-.833*	-.635	.791*	-.633	-.828*	-.820*	-.791*
	Sig. (2-					.136	.010	.091	.019	.092	.011	.013	.019
	N					8	8	8	8	8	8	8	8
LP1_SI RM_X	Pearso					1	.925**	.946**	-.934**	.695	.706	.884**	.934**
	Sig. (2-						.001	.000	.001	.055	.050	.004	.001
	N						8	8	8	8	8	8	8
LP1_A RM_X	Pearso						1	.934**	-.989**	.758*	.848**	.968**	.989**
	Sig. (2-							.001	.000	.029	.008	.000	.000
	N							8	8	8	8	8	8
LP1_S OFT	Pearso							1	-.959**	.841**	.810*	.942**	.959**
	Sig. (2-								.000	.009	.015	.000	.000
	N								8	8	8	8	8
LP1_H ARD	Pearso								1	-.823*	-.886**	-.988**	-1.000**
	Sig. (2-									.012	.003	.000	.000
	N									8	8	8	8
LP1_2 0mT	Pearso									1	.939**	.890**	.823*
	Sig. (2-										.001	.003	.012
	N										8	8	8
LP1_4 0mT	Pearso										1	.945**	.886**
	Sig. (2-											.000	.003
	N											8	8
LP1_1 00mT	Pearso											1	.988**
	Sig. (2-												.000
	N												8
LP1_3 00mT	Pearso												1
	Sig. (2-												
	N												

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

Table 5.10c: Correlation of magnetic parameters of lake sediments of the lakes in the Rodna/Maramures region

Stiol Lake Core 2		LS2_X	LS2_ARM	LS2_SIRM	LS2_SIRM_ARM	LS2_SIRM_X	LS2_ARM_X	LS2_SOFT	LS2_HARD	LS2_20mT	LS2_40mT	LS2_100mT	LS2_300mT
LS2_X	Pearso	1	.951**	.939**	.529	.905**	.920**	.929**	-.232	.093	.840**	.553	.232
	Sig. (2-		.000	.000	.116	.000	.000	.000	.518	.798	.002	.097	.518
	N		10	10	10	10	10	10	10	10	10	10	10
LS2_ARM	Pearso		1	.998**	.609	.989**	.995**	.992**	-.198	.147	.913**	.550	.198
	Sig. (2-			.000	.062	.000	.000	.000	.583	.684	.000	.100	.583
	N			10	10	10	10	10	10	10	10	10	10
LS2_SIRM	Pearso			1	.650*	.995**	.996**	.997**	-.154	.195	.925**	.514	.154
	Sig. (2-				.042	.000	.000	.000	.672	.589	.000	.129	.672
	N				10	10	10	10	10	10	10	10	10
LS2_SIRM_ARM	Pearso				1	.676*	.627	.699*	.589	.843**	.711*	-.233	-.589
	Sig. (2-					.032	.052	.025	.073	.002	.021	.518	.073
	N					10	10	10	10	10	10	10	10
LS2_SIRM_X	Pearso					1	.997**	.994**	-.129	.221	.940**	.501	.129
	Sig. (2-						.000	.000	.723	.540	.000	.140	.723
	N						10	10	10	10	10	10	10
LS2_ARM_X	Pearso						1	.991**	-.186	.160	.930**	.551	.186
	Sig. (2-							.000	.606	.658	.000	.099	.606
	N							10	10	10	10	10	10
LS2_SOFT	Pearso							1	-.085	.264	.926**	.453	.085
	Sig. (2-								.816	.461	.000	.188	.816
	N								10	10	10	10	10
LS2_HARD	Pearso								1	.903**	-.076	-.859**	-1.000**
	Sig. (2-									.000	.834	.001	0.000
	N									10	10	10	10
LS2_20mT	Pearso									1	.240	-.712*	-.903**
	Sig. (2-										.503	.021	.000
	N										10	10	10
LS2_40mT	Pearso										1	.500	.076
	Sig. (2-											.141	.834
	N											10	10
LS2_100mT	Pearso											1	.859**
	Sig. (2-												.001
	N												10
LS2_300mT	Pearso												1
	Sig. (2-												
	N												

Vinderel Lake Core 3 (2006)		LV3_X	LV3_ARM	LV3_SIRM	LV3_SIRM_ARM	LV3_SIRM_X	LV3_ARM_X	LV3_SOFT	LV3_HARD	LV3_20mT	LV3_40mT	LV3_100mT	LV3_300mT
LV3_X	Pearso	1	.920**	.970**	-.945**	.933**	.902**	.981**	-.432	.856**	.895**	.875**	.432
	Sig. (2-		.000	.000	.000	.000	.000	.000	.140	.000	.000	.000	.140
	N		13	13	13	13	13	13	13	13	13	13	13
LV3_ARM	Pearso		1	.980**	-.936**	.967**	.996**	.969**	-.277	.713**	.688**	.746**	.277
	Sig. (2-			.000	.000	.000	.000	.000	.360	.006	.009	.003	.360
	N			13	13	13	13	13	13	13	13	13	13
LV3_SIRM	Pearso			1	-.947**	.988**	.974**	.997**	-.374	.776**	.793**	.834**	.374
	Sig. (2-				.000	.000	.000	.000	.208	.002	.001	.000	.208
	N				13	13	13	13	13	13	13	13	13
LV3_SIRM_ARM	Pearso				1	-.931**	-.938**	-.952**	.491	-.845**	-.843**	-.856**	-.491
	Sig. (2-					.000	.000	.000	.088	.000	.000	.000	.088
	N					13	13	13	13	13	13	13	13
LV3_SIRM_X	Pearso					1	.973**	.977**	-.413	.717**	.755**	.846**	.413
	Sig. (2-						.000	.000	.160	.006	.003	.000	.160
	N						13	13	13	13	13	13	13
LV3_ARM_X	Pearso						1	.960**	-.296	.695**	.674*	.751**	.296
	Sig. (2-							.000	.326	.008	.012	.003	.326
	N							13	13	13	13	13	13
LV3_SOFT	Pearso							1	-.382	.820**	.825**	.835**	.382
	Sig. (2-								.198	.001	.001	.000	.198
	N								13	13	13	13	13
LV3_HARD	Pearso								1	-.446	-.631*	-.739**	-1.000**
	Sig. (2-									.127	.021	.004	0.000
	N									13	13	13	13
LV3_20mT	Pearso									1	.911**	.708**	.446
	Sig. (2-										.000	.007	.127
	N										13	13	13
LV3_40mT	Pearso										1	.902**	.631*
	Sig. (2-											.000	.021
	N											13	13
LV3_100mT	Pearso											1	.739**
	Sig. (2-												.004
	N												13
LV3_300mT	Pearso												1
	Sig. (2-												
	N												

**. Correlation is significant at the 0.01 level (2-tailed).

*. Correlation is significant at the 0.05 level (2-tailed).

There are obvious variations in the magnitude of the magnetic parameters in both regions. For example, the highest value of ARM is 61.84 x $10^{-5}Am^2kg^{-1}$ while that of SIRM is 1285.11 x $10^{-5}Am^2kg^{-1}$ (Fagaras Capra Lake Figure 5.17 and Fagaras Balea Lake Figure 5.14). From Rodna Pietrosul Lake (Figure 5.22), the highest value of ARM is 21.17 x 10^{-5} Am^2 kg^{-1} while that of SIRM is 229.56 x 10^{-5} Am^2 kg^{-1} but the fundamental principle is same.

5.4 Geochemical analysis of the lake sediments

The purpose of the geochemical analysis of the lake sediments is to determine the levels of trace metals stored into the lake sediments and hence assess down core variations in their levels. This analysis shows that across the lakes in the Fagaras region, Pb and Zn consistently demonstrated clear surface increase in concentration. In the Fagaras region the patterns of Pb and Zn concentrations in each lake are quite similar with corresponding peaks and troughs. Similar trends in metal concentration are repeated across most of the lakes in the Rodna region. Pb and Zn consistently show noticeable surface increase in most of the lakes. Higher trace metal concentrations were recorded in the Fagaras mountains than in the Rodna region for example, Balea Lake (from Fagaras region; Table 5.11) has a Pb and Zn magnitudes of 280 mg kg^{-1} and 320 mg kg^{-1} respectively while the highest level of Pb was recorded in Bila Lake (from Rodna; Table 5.12) with a value of 175 mg kg^{-1}. The highest level of Zn in Rodna region was recorded in Buhaiescu Lake with the value of 149 mg kg^{-1}. Therefore, the concentration of metal accumulation in Fagaras region was higher than in the Rodna lakes (see Figures 5.27-5.41).

5.4.1 Geochemical analysis of the lake sediments in the Fagaras region

The results of trace metal analysis of sediment profiles are presented below. In Lacul Podragu Mare however, at the depth of 12 cm to 11 cm a spike was observed on Zn profile without a corresponding spike on Pb profile. Other metals (e.g. Al, Co, Cr, Cu, Fe, Mn and Ni) either demonstrated an increase down the profiles (e.g. Al, Co and Cr in LBa 1) or marginal surface increases. For example, in Podragu Mare Lake Ni has a bottom concentration of 46 mg kg^{-1} at the sediment depth of 19 cm and at the surface depth of 1 cm Ni has a concentration of 51 mg kg^{-1}. Figures 5.27-5.32 show down core profiles of

Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations in each core. On the x-axis (concentration) each core was plotted to its maximum in order to make clear any fluctuations in concentrations of the metals but all cores were plotted to the same scale on the y-axis (depth). The variability in core depth shows the degree of core recovery.

Table 5.11: Mean and standard deviations (Stdev) of Fagaras lakes Pb and Zn concentrations (mg kg^{-1})

Name		**Balea Lake**		**Caltun Lake**	**Capra Lake**		**Podragu Mare Lake**
		LBa 1	**LBa 4**	**LCt 2**	**LCp 2**	**LCp 3**	**LPm 2**
Pb	Mean	204.47	176.65	96.88	113.36	119.48	91.01
	Stdev	40.69	51.08	28.80	43.89	60.12	29.36
	Min	108.94	82.14	47.74	81.96	30.83	91.01
	Max	277.85	278.79	163.28	251.98	262.10	170.21
Zn	Mean	277.06	226.70	128.90	127.35	115.48	124.45
	Stdev	65.19	69.45	17.83	38.72	45.82	35.33
	Min	135.33	103.78	89.04	98.95	26.18	124.45
	Max	401.00	375.46	179.34	240.55	229.09	232.39

Balea Lake

A series of peaks and troughs were observed down the Pb and Zn profiles of Balea lake core 1 (LBa 1). In LBa 1, there were peaks between the depths of 30 - 27 cm and the depths of 14 - 9 cm. There were increases towards the surface in Pb and Zn profiles from the depth of 8 cm. In this core, Al, Co, Cr, Cu, Fe, Mn, and Ni showed a down core increase from the depth of 31 - 16 cm. In Balea Lake core 4 (LBa 4) a series of peaks and troughs were observed down the Pb and Zn profiles. In this lake, there were peaks between the depths of 30 - 27 cm and the depths of 14 - 9 cm. There were clear surface peaks in Pb and Zn profiles from the depth of 8 cm. In this core also, Al, Co, Cr, Cu, Fe, Mn, and Ni showed a sort of down core increase from the depth of 31 - 16 cm. A sharp surface increase was observed from the depths of 2.5 cm upwards in the profiles of both Fe and Mn in LBa 4.

The down core profiles of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations for Balea Lake (both cores LBa 1 and LBa 4) are similar (Figures 5.27 and 5.28) except for the surface increase from the depths of 2.5 cm upwards in the profiles of both Fe and Mn in LBa 4. The magnitudes of all the parameters were only slightly different between LBa 1 and LBa 4. For example the maximum concentrations of Pb and Zn in LBa 1 were 278 and 401 mg kg^{-1} while those of LBa 4 were 279 and 375 mg kg^{-1} respectively (Table 5.11).

The Pb and Zn profiles for both cores were similar with corresponding peaks and troughs. The peaks and troughs in the Pb and the Zn profiles also correspond with the peaks and troughs in the sediment density, LOI and particle size. Despite the similarities in peaks and troughs of metal concentrations down the profiles there were noticeable variations in the quantity of the metal concentrations down the profiles of the two cores. For example, at the depth of 1 cm the LBa 1 has a Pb concentration of 261 mg kg^{-1} and Zn concentration of 375 mg kg^{-1} but at the same surface depth LBa 4 has concentrations of 279 mg kg^{-1} and 309 mg kg^{-1} of Pb and Zn respectively. In addition to the above, there seems to be clearer fluctuations in the concentrations of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn in LBa 4 between the depth of 31 cm and 24 cm.

Caltun Lake

Between the depths of 27-22 cm a sort of hollow shape was common to all the trace metal profiles in this lake (Figure 5.29). There was such a hollow shape in density profile at the corresponding depth and a reduction in the percentage of organic material (Figures 4.24 and 4.28). There was also a notch at the depth of 18 cm. A clear surface peak was not recorded in Caltun Lake for the down core concentrations of Al, Co, Cr, Fe, Mn and Ni. However for Pb and Zn and to some extent Cu, an obvious surface peak was observed from the depths of about 9 cm, 5 cm and 4 cm respectively. In Caltun Lake the surface concentrations of Pb and Zn were 163 mg kg^{-1} and 164 mg kg^{-1} respectively while in

Balea Lake (LBa 1) the surface concentrations of Pb and Zn were 261 mg kg^{-1} and 375 mg kg^{-1} respectively.

Capra Lake

The down core trace metal profiles of Capra Lake (core LCp 2) showed no apparent surface increase for Al, Co, Cr, Cu, Fe, Mn, and Ni. Rather, a surface decline in concentrations of Al, Co, Cu, Cr, Mn and Ni has been observed from about the depth of 5 cm (Figure 5.30). No striking features were observed in the Pb and Zn profiles of LCp 2 from the bottom to a depth of about 5 cm. However there has been a surface increase in the concentrations of Pb and Zn from the depth of 5 cm (Figure 5.30). There were no striking features observed in the metal profiles of LCp 3 from the bottom to the depth of about 16 cm. At the depth of 16.5 cm a sharp notch was observed in all the metal profiles in LCp 3.

The down-core trace metal profiles of Capra Lake (core LCp 3) also showed no apparent surface increase for Al, Co, Cr, Cu, Fe, Mn, and Ni. A surface decline in concentrations of Al, Co, Cu, Cr, Mn and Ni was observed from about the depth of 5 cm (Figure 5.31). The surface increase in the concentrations of Pb and Zn was observed from the depth of 12 cm. There were variations in the magnitudes of the metal concentrations down the profiles of the two cores. For example, at the depth of 1 cm the LCp 2 has a Pb concentration of 252 mg kg^{-1} and Zn concentration of 241 mg kg^{-1} but at the same surface depth LCp 3 has concentrations of 244 mg kg^{-1} and 183 mg $kg^{-1}$of Pb and Zn respectively.

Podragu Mare Lake

Podragu Mare Lake showed a surface increase from the depth of 10 cm which was more noticeable in the Pb and less in the Zn profiles, but not in Al, Co, Cr, Cu, Fe, Mn, Ni. Pb demonstrated an obvious decline in concentrations towards the bottom of the core (Figure 5.32). A slightly hollow shape was observed on the profiles of Cr, Fe, Ni, Pb and Zn between the depths of 10 - 4 cm. There was a spike in the profile of Zn at the depth of 11 cm. Podragu Mare lake had a maximum Pb concentration value of 170 mg kg^{-1} and a maximum Zn concentration value of 232 mg kg^{-1} (see Table 5.11).

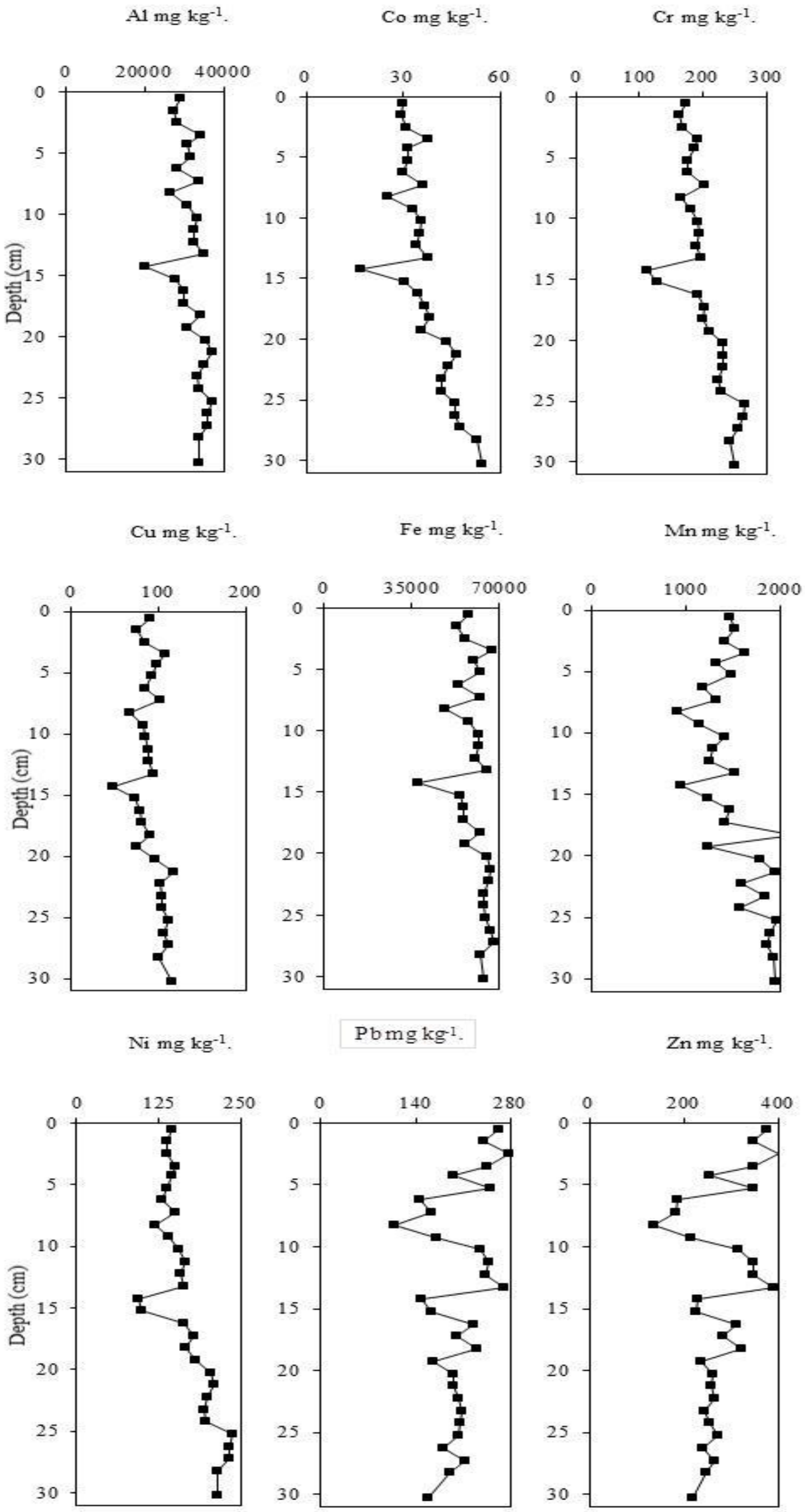


Figure 5.27: Down core variations of sediment metal profiles of Balea Lake (LBa 1)
Note: Metal concentration measurements are all in mg kg^{-1}.

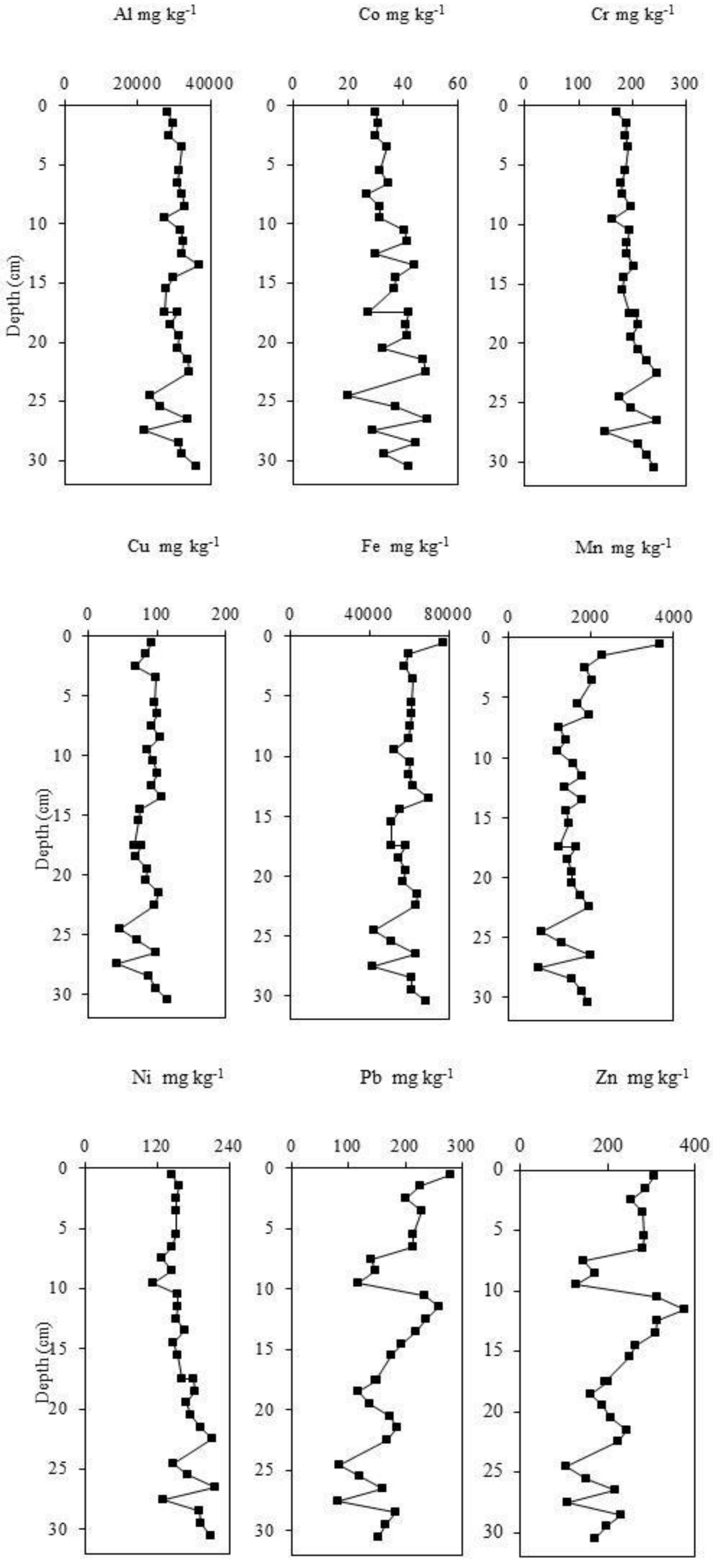


Figure 5.28: Down core variations of sediment metal profiles of Balea Lake (LBa 4) Note: Metal concentration measurements are all in mg kg^{-1}.

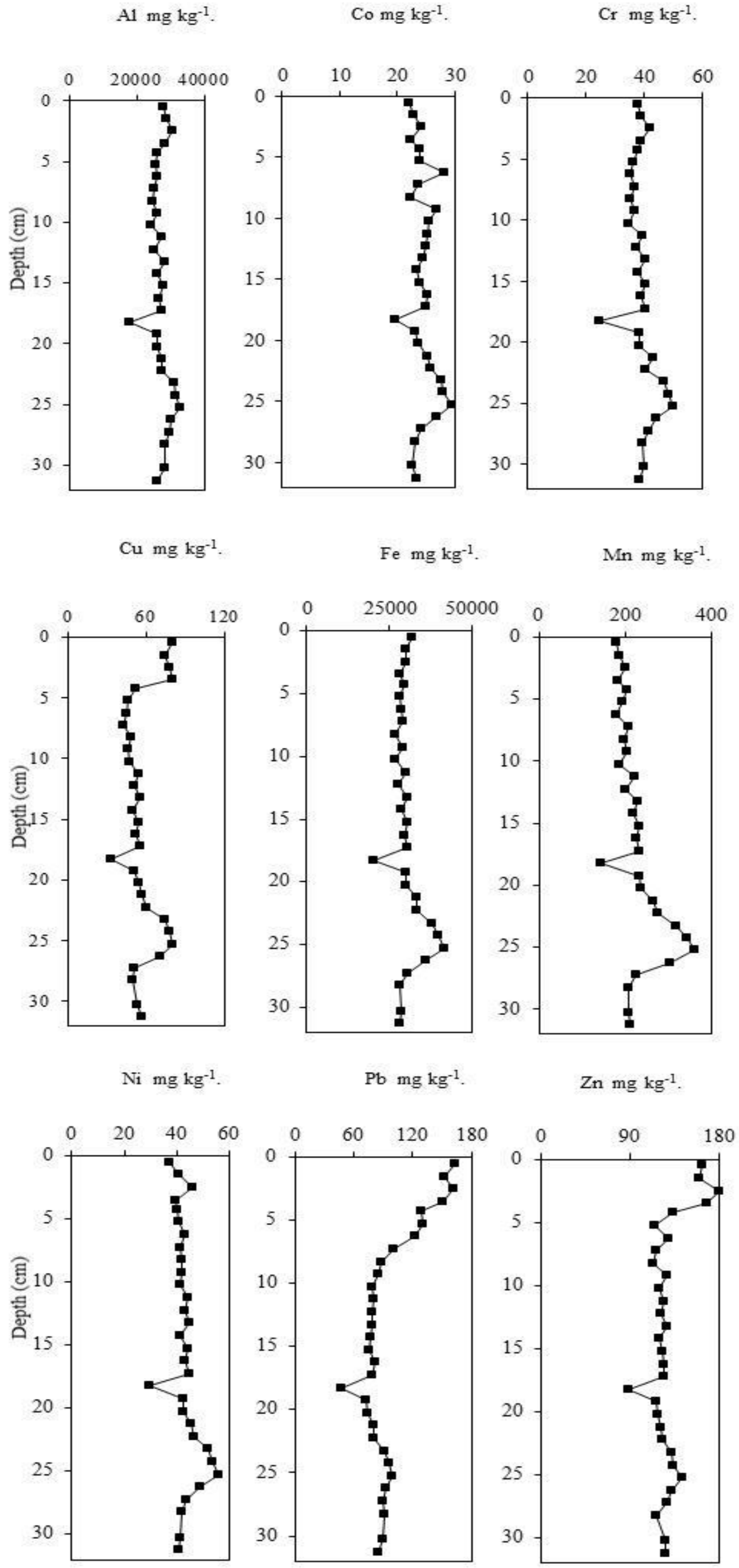


Figure 5.29: Down core variations of sediment metal profiles of Caltun Lake (LCt 2)

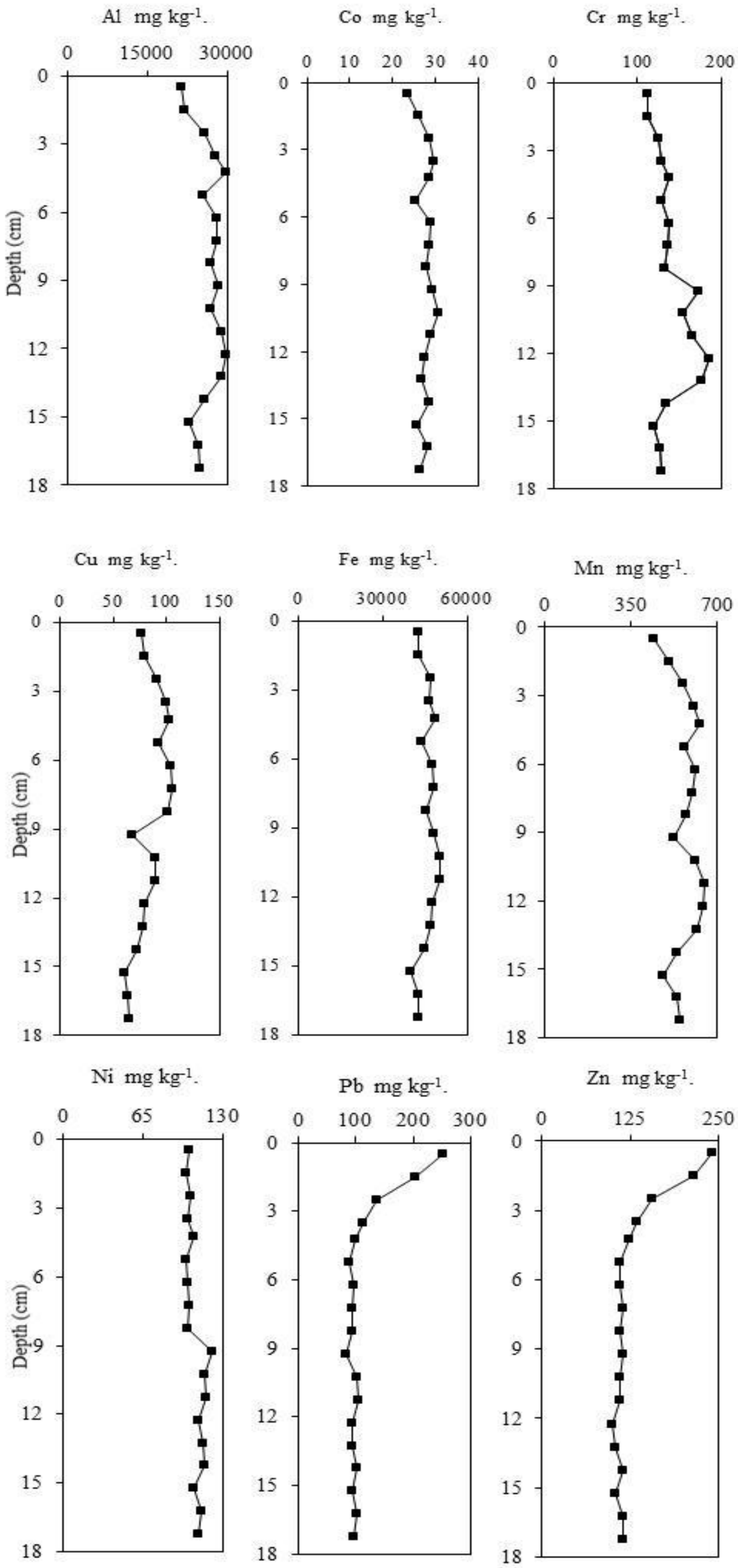


Figure 5.30: Down core variations of sediment metal profiles of Capra Lake (LCp 2)

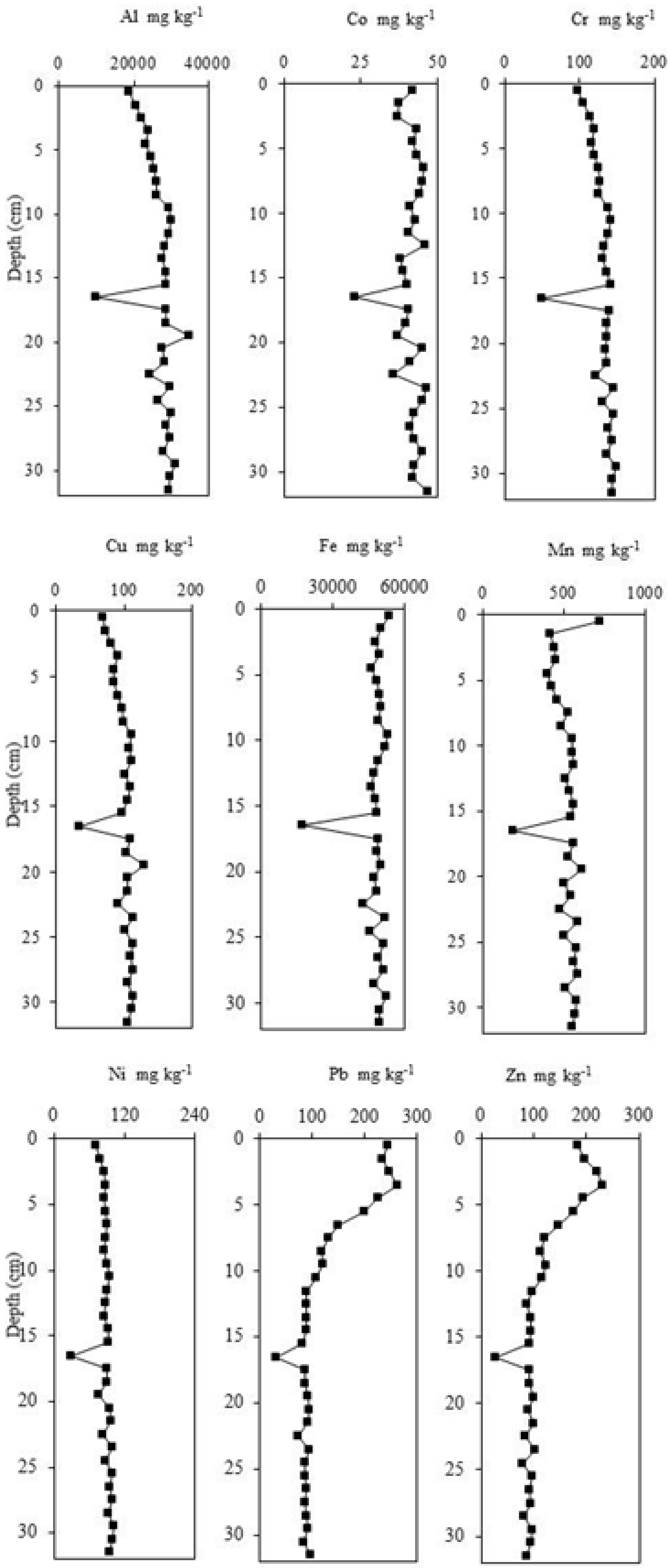


Figure 5.31: Down core variations of sediment metal profiles of Capra Lake (LCp 3)

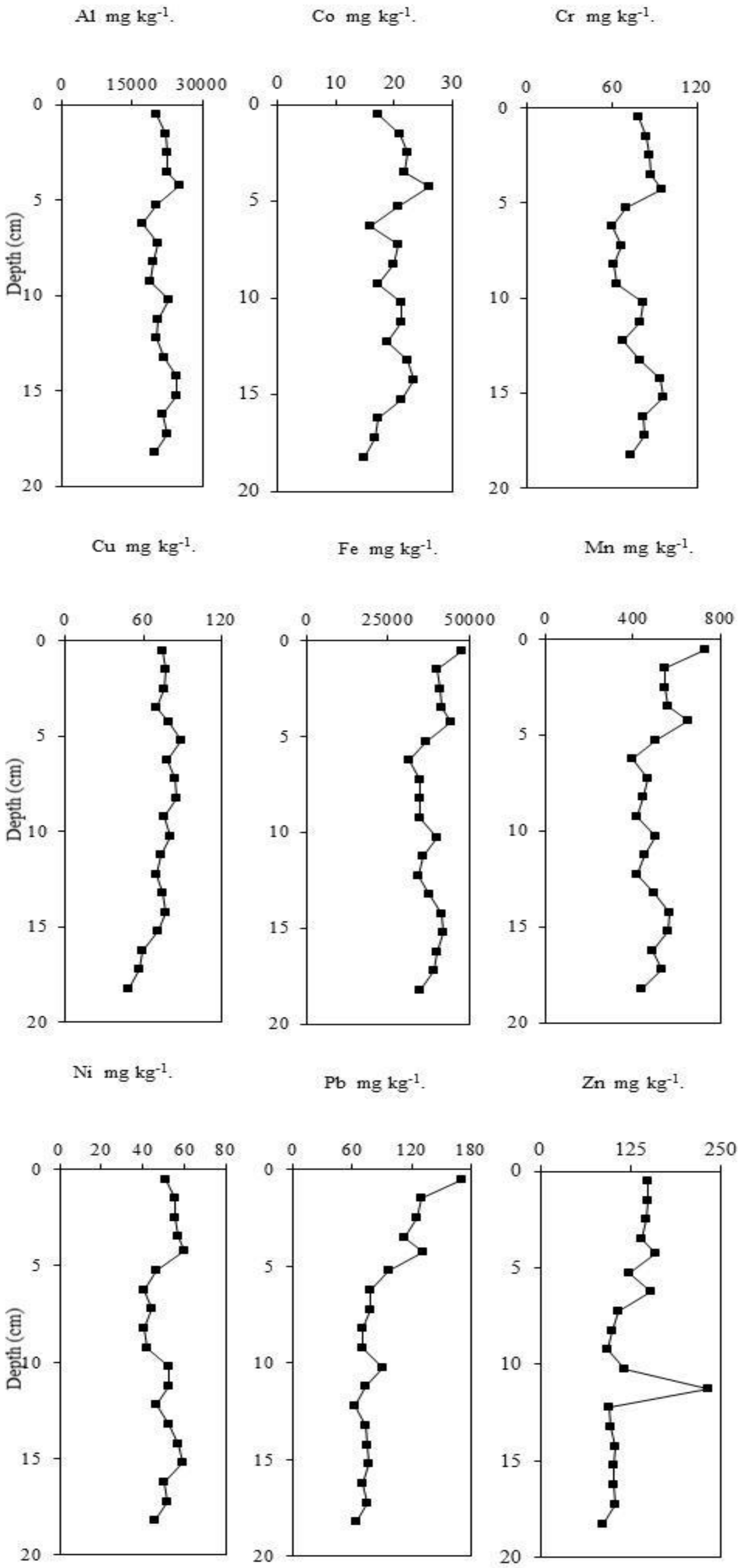


Figure 5.32: Down core variations of sediment metal profiles of Podragu Mare Lake (LPm 2).

5.4.2 Geochemical analysis of the lake sediments in the Rodna/Maramures region

The results of trace metal analysis of Rodna lake sediment profiles are presented below. The patterns of Pb and Zn concentrations in each lake where multiple cores were taken were quite similar with corresponding peaks and troughs. Figures 5.33- 5.41 show down core profiles of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations in each core. On the x-axis (concentration) each core was plotted to its maximum in order to make clear any fluctuations in concentrations but all cores were plotted to the same scale on the y-axis (depth). The variability in core depth shows the degree of core recovery. LB 2, LB-3:2, LP 1 (2006), LP 1 (2008), Russian corer, LV 3 and LV 1 cores show a general increase in metal concentration (especially Pb and Zn) towards the surface (Figures 5.33, 5.34, 536, 5.37, 5.38, 5.40 and 5.41). LLM 2 and LS 2 show no apparent general increase in metal concentration towards the surface (Figures 5.35 and 5.39). All the lakes sampled in Fagaras region exhibited more conspicuous surface peaks in Pb and Zn than their counterparts from the Rodna region. LB 2, LB-3:2, LP 1 and LV 1 cores show a clear surface peak for Pb and Zn from the depth of about 8 cm and a general decline towards the bottom. The highest value of Pb concentration (203 mg kg^{-1}) was recorded in Bila Lake (LB 2) while the highest Zn concentration (149 mg kg^{-1}) was recorded in Pietrosul Lake (LP 1, 2008). The mean value of Pb across the lakes ranged from 60 - 172 mg kg^{-1} while that of Zn was 86 - 123 mg kg^{-1}.

Table 5.12: Mean and standard deviations (Stdev) of Rodna lakes Pb and Zn concentrations (mg kg^{-1})

Name		**Bila Lake**	**Buhaiescu-3 Lake**	**Lala Mare Lake**	**Pietrosul Lake**		**Stiol Lake**	**Vinderel Lake**	
		LB 2	**LB-3:2**	**LLM 2**	**LP1 (2006)**	**LP1 (2008)**	**LS 2**	**LV3 (2006)**	**LV1 (2008)**
Pb	Mean	172.31	83.69	95.32	60.04	86.72	82.66	92.91	112.52
	Stdev	20.46	19.23	4.52	15.15	33.70	4.95	19.82	20.06
	Min	137.99	51.26	88.44	44.66	49.61	77.31	70.41	89.75
	Max	203.03	116.23	100.47	86.71	140.11	91.29	126.03	150.12
Zn	Mean	122.35	122.59	108.95	85.82	95.73	94.75	113.29	122.09
	Stdev	13.19	23.67	5.96	16.74	34.44	4.79	14.68	12.00
	Min	97.40	85.48	99.96	69.46	54.32	89.67	98.66	106.27
	Max	141.94	148.37	119.18	113.64	149.36	105.59	139.66	143.48

Bila Lake

The down core profiles of Cu, Fe, Mn, Ni, Pb and Zn concentrations for Bila Lake (LB 2) demonstrated slight near surface increase from about the depth of 15 cm (Figure 5.33). The down core profiles of all the metals (Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn) exhibited a sort of hollow shape between the depths of 12.5 - 5.5 cm. Between the depths of 16 - 13 cm a shallow cut was observed in all the metal profiles (Al, Co, Cr, Fe, Mn, Ni, Pb and Zn) except Cu. Bila Lake Pb concentration ranged from 138 - 203 mg kg^{-1} while the Zn concentration ranged from 97 - 142 mg kg^{-1}. It had a Pb mean value of 172 mg kg^{-1} which was the highest in Rodna region and a Zn mean value of 122 mg kg^{-1} (see Table 5.12).

Buhaiescu -3 Lake

From a depth of 8.5cm LB-3:2 showed surface increase in concentrations of Pb and Zn. At the depth of 1.5 cm there was a notch which was observed in all the metal profiles except Zn. LB-3:2 demonstrated a down core increase to Co, Cr, Fe and Mn. It also showed little basal spike in Co and Fe profiles (Figure 5.34). Its Pb concentration ranged from 51 - 116 mg kg^{-1} while the Zn concentration ranged from 85 - 148 mg kg^{-1}. It had a Pb mean value of 84 mg kg^{-1} and a Zn mean value of 123 mg kg^{-1} (see Table 5.12).

Lala Mare Lake

There was no clearly observable feature in the profiles (Figure 5.35). An examination of the trace metal profiles showed a slight hollow between the depths of 8.5 - 3.5 cm which was observed in all the metals (Al, Co, Cr, Fe, Mn and Ni except in Cu, Pb and Zn. Its Pb concentration ranged from 88 - 100 mg kg^{-1} while the Zn concentration ranged from 100 - 119 mg kg^{-1}. It had a Pb mean value of 95 mg kg^{-1} and a Zn mean value of 109 mg kg^{-1} (see Table 5.12).

Pietrosul Lake

Two cores (LP 1; 2006 and LP1; 2008) were geochemically analysed in Pietrosul Lake (Figures 5.36 and 5.37). LP 1 (2006) showed a clear surface increase to Pb and Zn from the depth of 3.5 cm. LP 1 (2008) showed a clear surface increase to Fe, Pb and Zn from the depth of 4.5 cm. Fe surface increase started from the depth of 3.5 cm while Pb and Zn peak started from 4.5 cm depth. At the depth of 7.5 cm, the core (LP 1 sampled in 2008) shows a basal spike for Al, Cr, Fe, Mn, Ni, Pb and Zn. From the depth of 2.5 cm, a surface decrease was observed in the profiles of Al, Cr and Ni. Its Pb concentration ranged from 45 - 87 mg kg^{-1} (LP1; 2006) and 50 - 140 mg kg^{-1} (LP 1; 2008) while the Zn concentration ranged from 69 - 114 mg kg^{-1} (LP1; 2006) and 54 - 149 mg kg^{-1} (LP 1; 2008). It had a Pb mean value of 60 mg kg^{-1} (LP1; 2006) and 87 mg kg^{-1} (LP 1; 2008) and a Zn mean value of 86 mg kg^{-1} (LP1; 2006) and 96 mg kg^{-1} (LP1; 2008). LP1 (2006) had the minimum mean value in Rodna region (see Table 5.12). The core taken with Russian corer in 2008 demonstrated no increase in density but it exhibited peaks and troughs between the depths of 23.5 - 13.5 cm.

Stiol Lake

Stiol Lake (Figure 5.39) did not demonstrate any obvious surface peak in any of the metals but did show a down core increase in Co, Cr, Cu, Mn and Ni. The concentration of Fe is virtually a straight line down the core length. Stiol Lake Pb concentration ranged from 77 - 91 mg kg^{-1} while the Zn concentration ranged from 90 - 106 mg kg^{-1}. It had a Pb mean value of 83 mg kg^{-1} and a Zn mean value of 95 mg kg^{-1} (see Table 5.12).

Vinderel Lake

Two cores LV 3 and LV 1 were geochemically analysed (Figures 5.40 and 5.41). LV 3 (sampled in 2006) demonstrated a slight increase in concentrations of Pb and Zn towards the surface from the depth of 7 cm. Vinderel Lake core LV 1 (sampled in 2008) showed a slight increase towards the surface for Pb and Zn from the depth of 14 cm. The down core profiles of Al, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn concentrations for LV 1 (sampled in 2008) demonstrated a common notch in all the metals at the depth of 4.5 cm (Figure 4.64). Also from the depth of 4.5 cm towards the surface the core showed a slight increase in the concentrations of Fe and Mn. Vinderel Lake Pb concentration ranged from 70 - 126 mg kg^{-1} (LV 3; 2006) and 90 - 150 mg kg^{-1} (LV 1; 2008) while the Zn concentration ranged from 99 - 140 mg kg^{-1} (LV 3; 2006) and 106 - 143 mg kg^{-1} (LP 1; 2008). It had a Pb mean value of 93 mg kg^{-1} (LV 3; 2006) and 113 mg kg^{-1} (LV 1; 2008) and a Zn mean value of 113 mg kg^{-1} (LV 3; 2006) and 122 mg kg^{-1} (LV 1; 2008) (see Table 5.12).

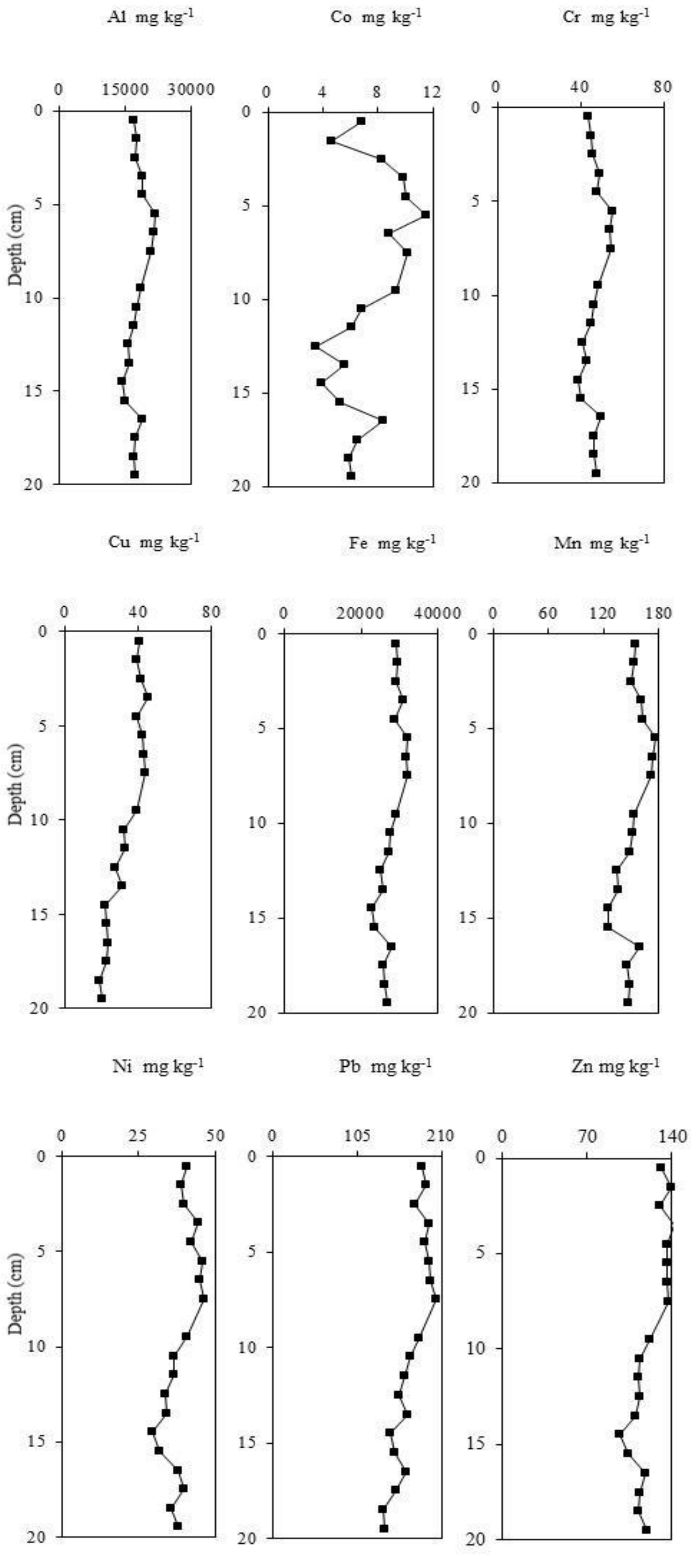


Figure 5.33: Down core variations of sediment metal profiles of Bila Lake- LB 2
Note: Metal concentration measurements are all in mg kg^{-1}.

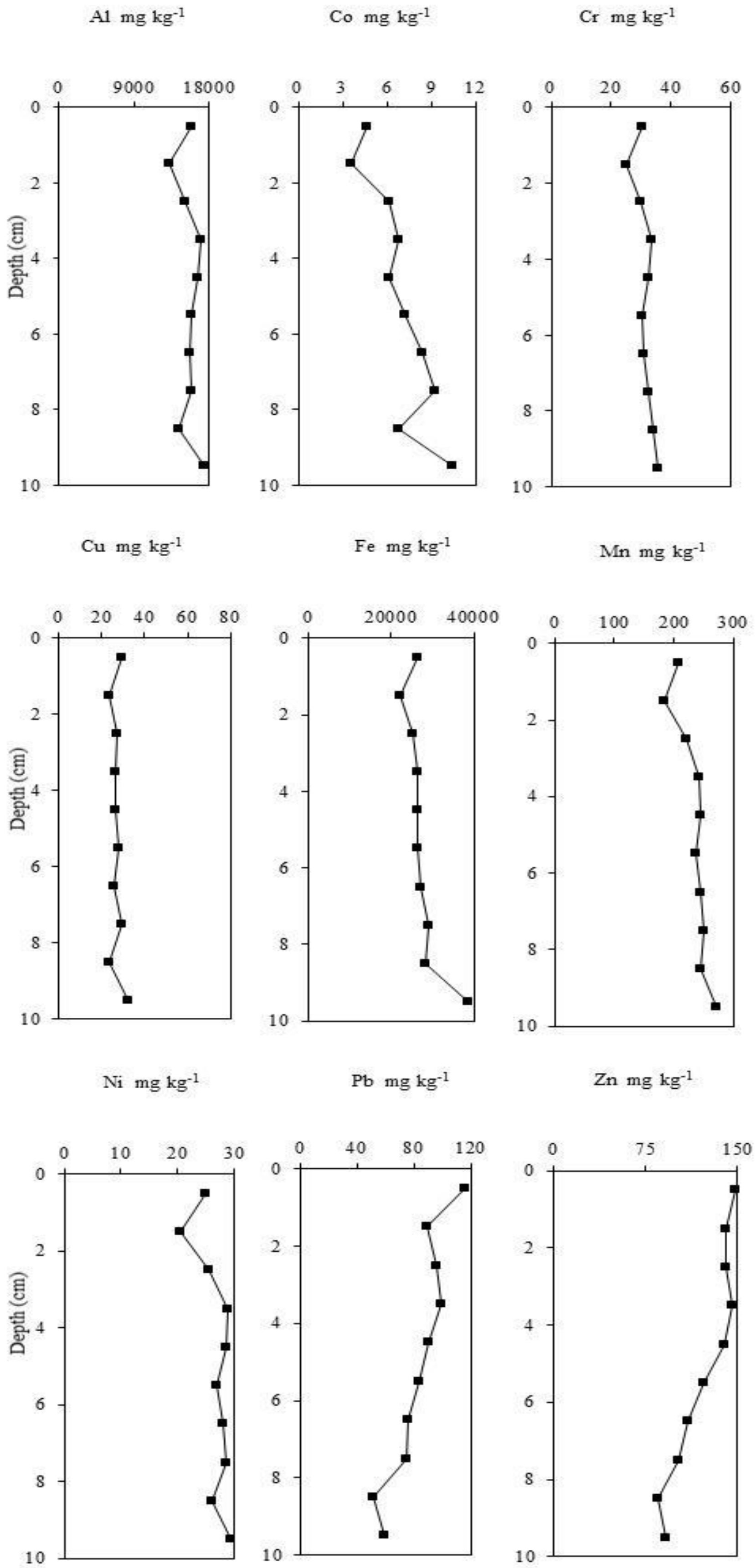


Figure 5.34: Down core variations of sediment metal profiles of Buhaiescu 3 Lake LB-3 2. Note: Metal concentration measurements are all in mg kg^{-1}.

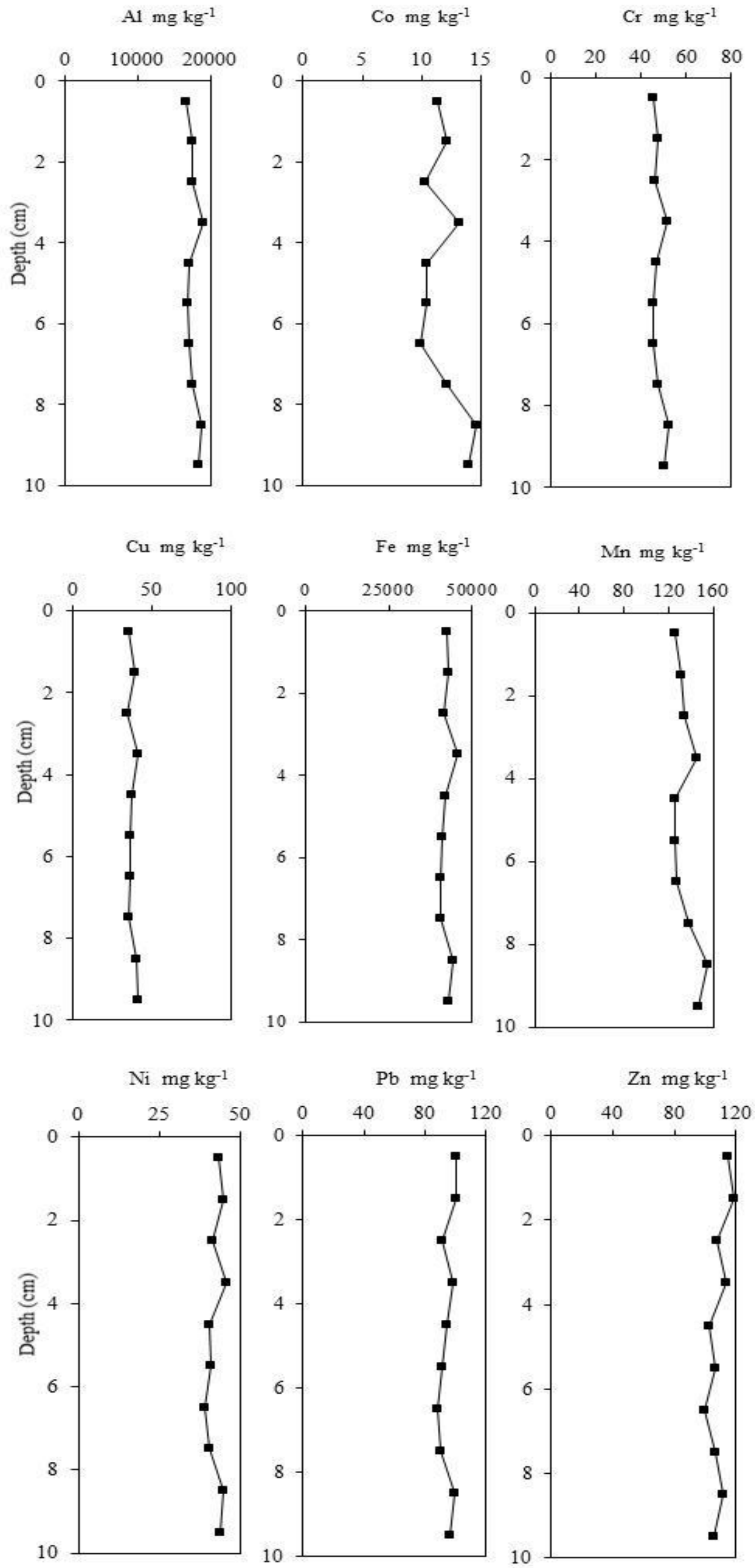


Figure 5.35: Down core variations of sediment metal profiles of Lala Mare Lake; LLM 2.

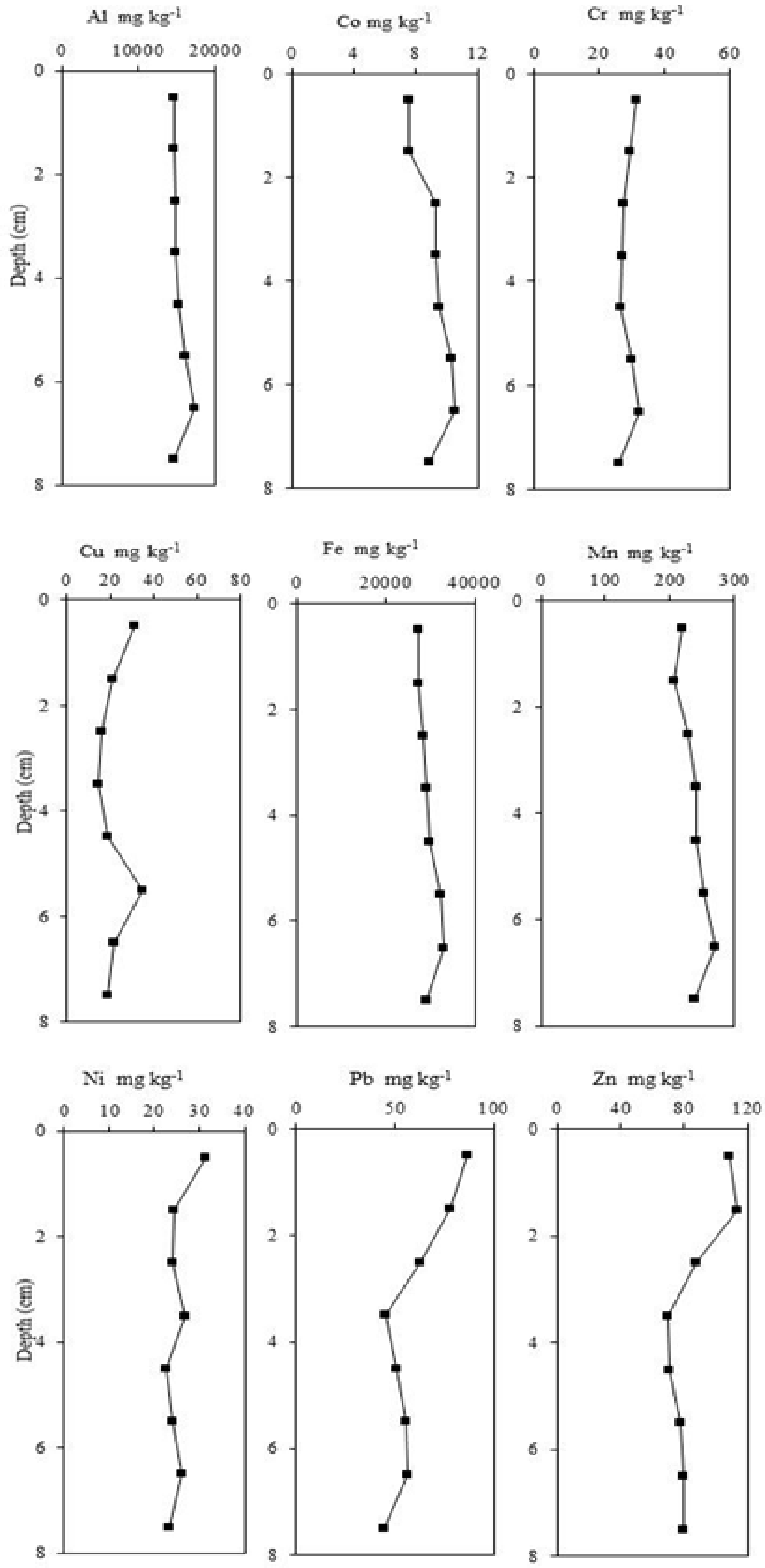


Figure 5.36: Down core variations of sediment metal profiles of Pietrosul; LP 1 (2006).

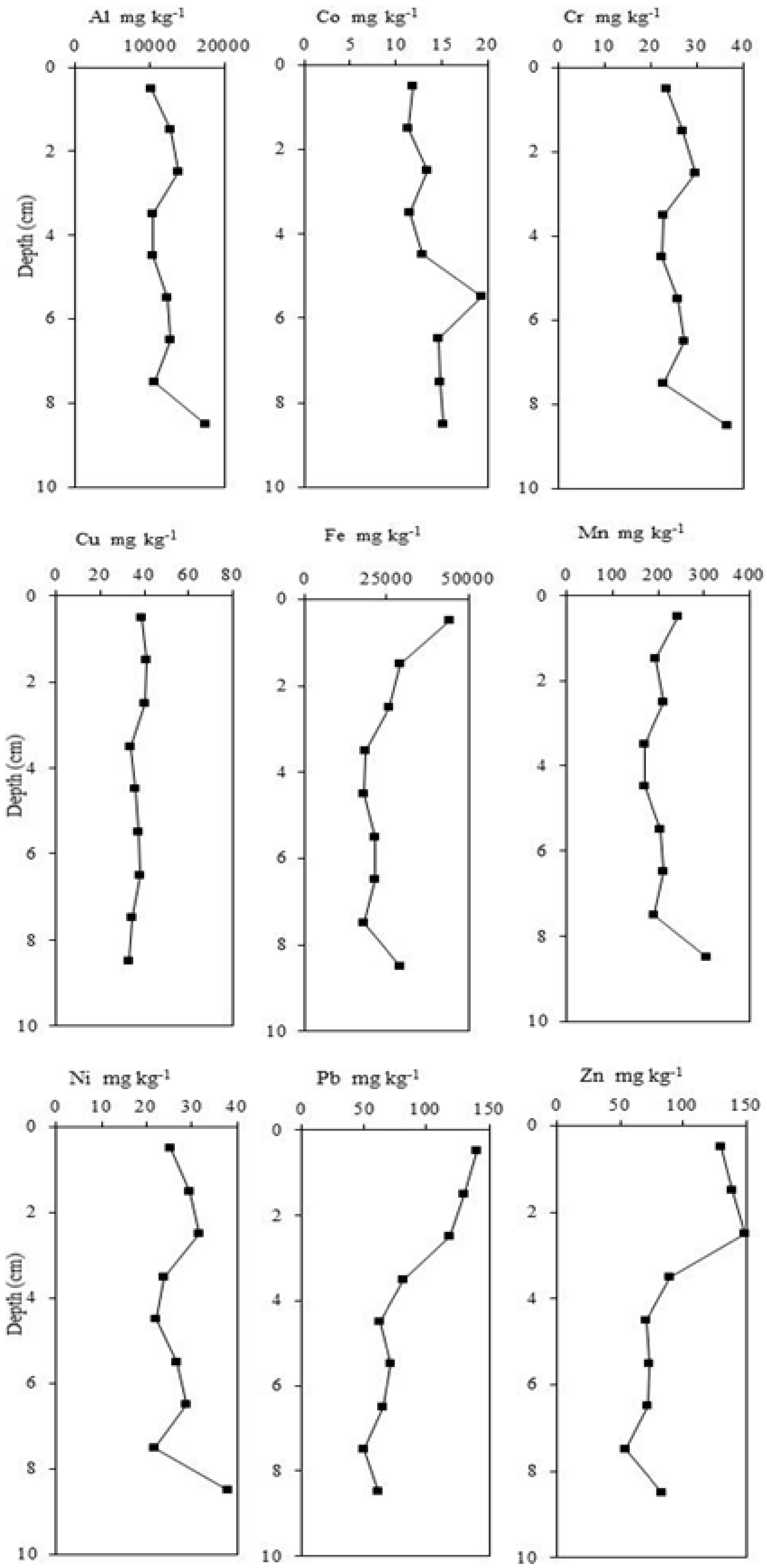


Figure 5.37: Down core variations of sediment metal profiles of Pietrosul; LP 1 (2008).

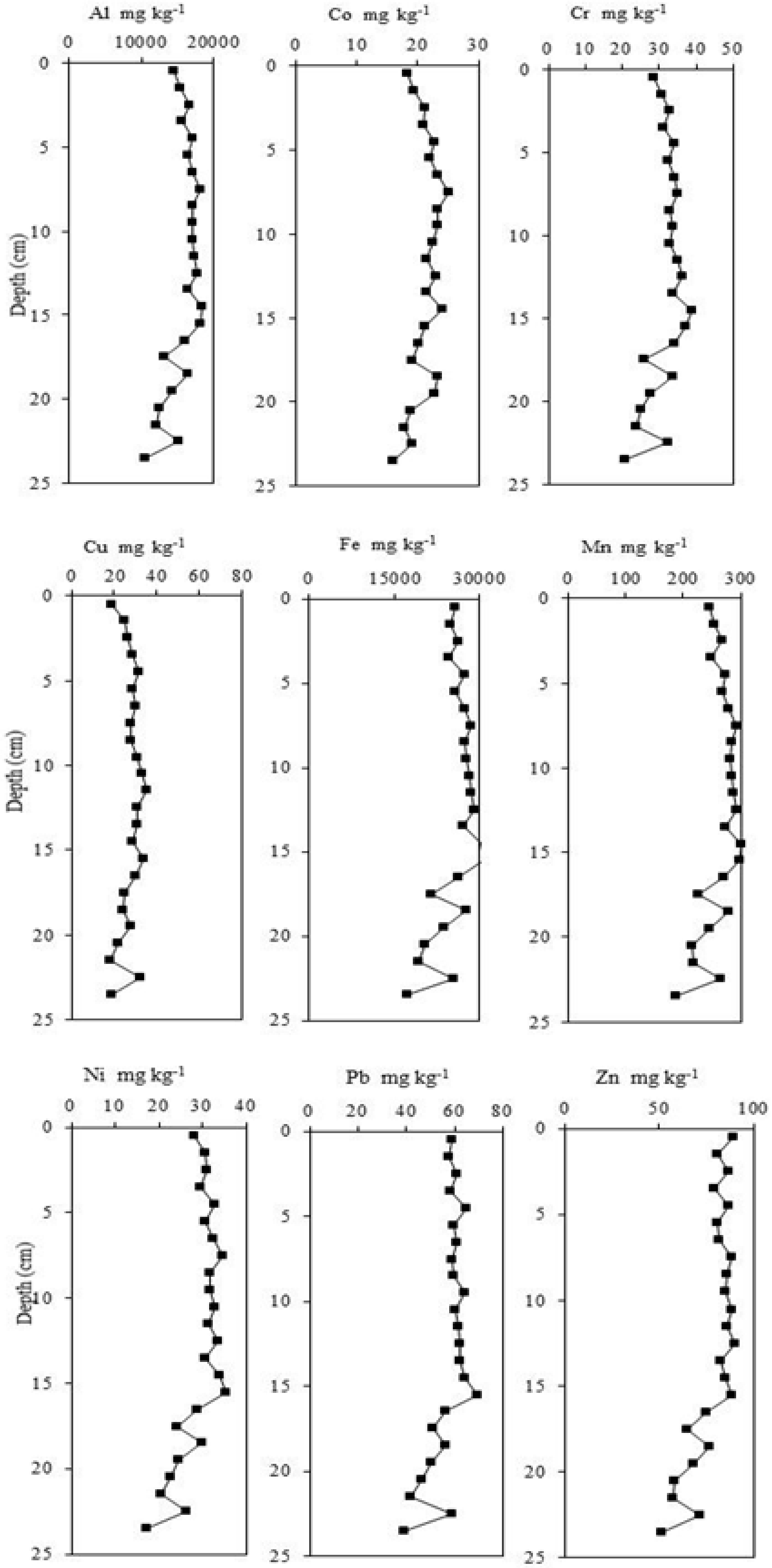


Figure 5.38: Down core variations of sediment metal profiles Pietrosul Russian core (2008).

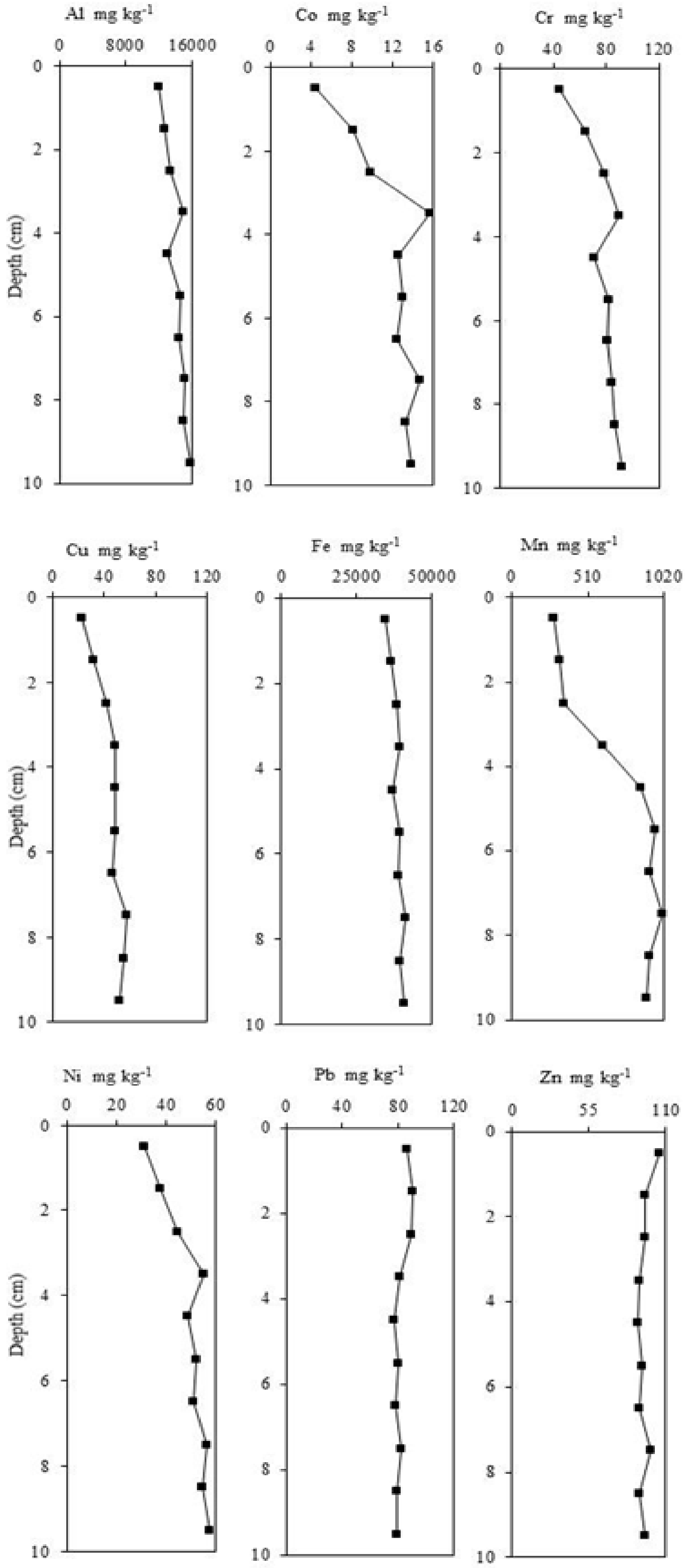


Figure 5.39: Down core variations of sediment metal profiles of Stiol Lake; LS 2.

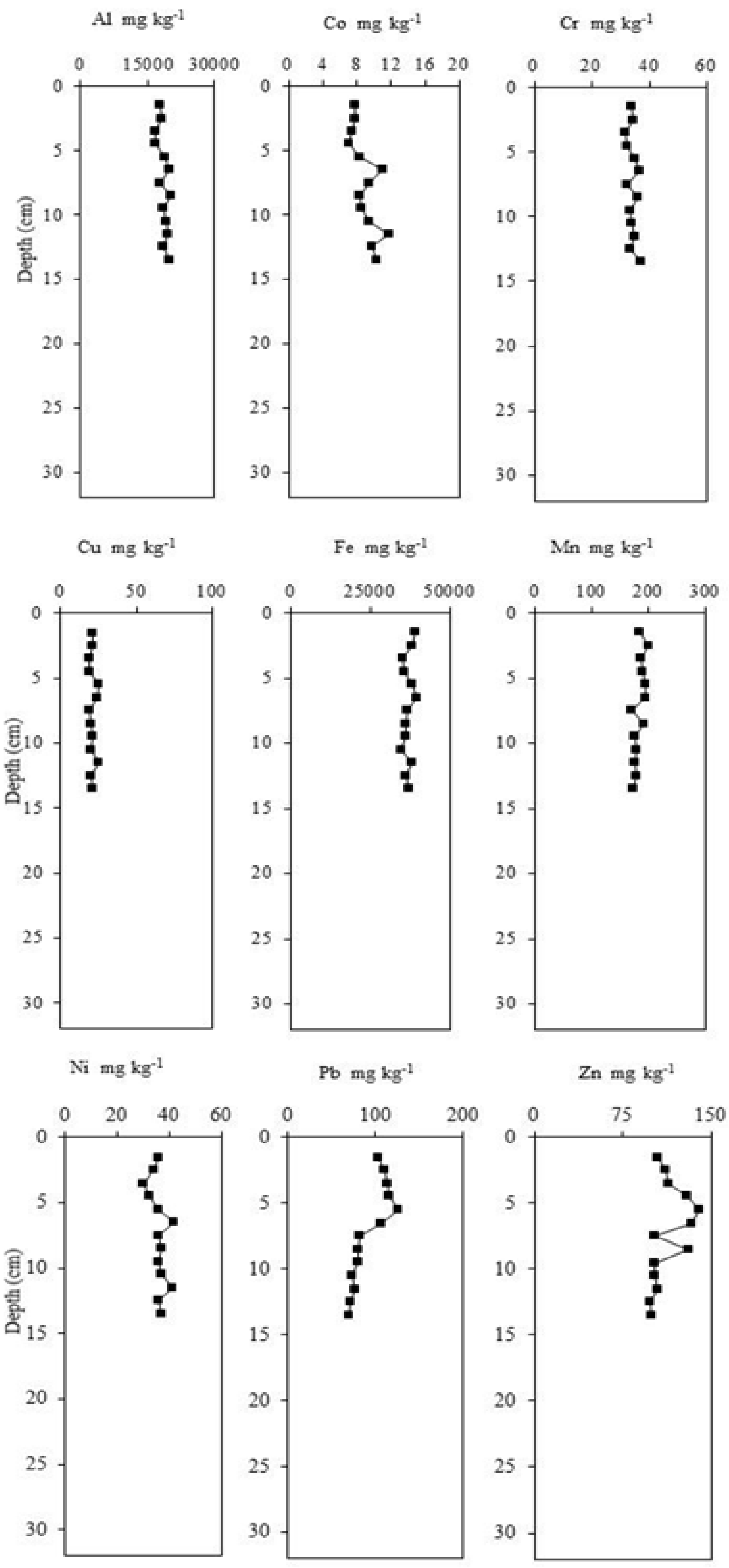


Figure 5.40: Down core variations of sediment metal profiles of Vinderel LV 3 (2006).

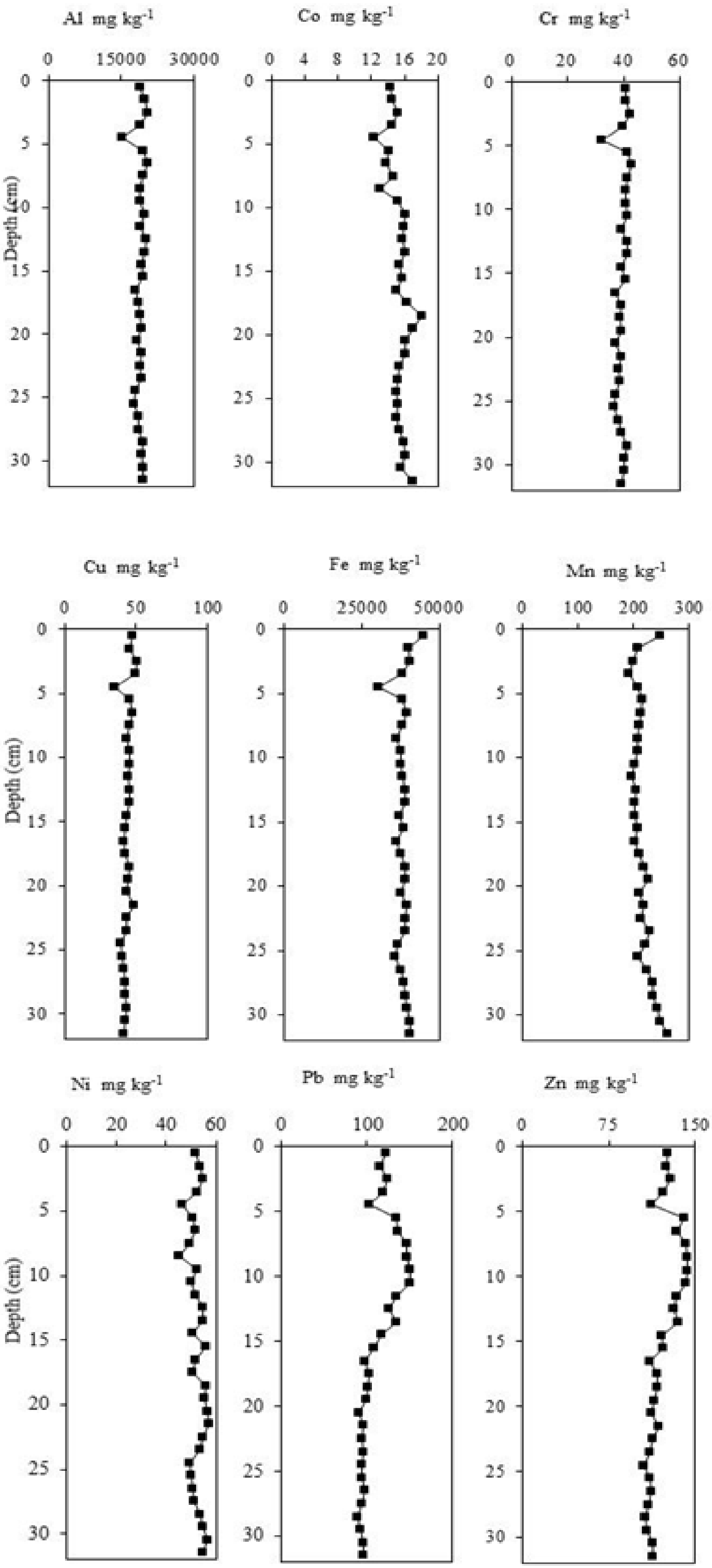


Figure 5.41: Down core variations of sediment metal profiles of Vinderel LV 1 (2008).

5.4.3 Summary of the geochemical analysis

The results of trace metal analysis of sediment profiles showed that the same trends in metal concentration were repeated across the lakes in the Fagaras region. Pb and Zn showed noticeable surface increases in all the lakes (profiles compared visually). A statistical approach showed that Al level in LBa 4 correlated significantly with every other metal analysed except Mn and Ni. Likewise Al level in LCp 3 correlated significantly with every other metal analysed except Cu and Ni. Also, Al level in LCt 2 correlated significantly with every other metal analysed except Pb. In LPm 2 Al level correlated significantly with every other metal analysed except Cu, Mn, Pb and Zn. In Fagaras region the patterns of Pb and Zn concentrations in each lake were quite similar with corresponding peaks and troughs (in all lakes with multiple cores). Pb and Zn demonstrated significant correlations in all lakes from Fagaras region except LPm 2 where there was no correlation between the two metals (Table 5.13).

Table 5.13: Correlation of metals of lake sediments of the lakes in the Fagaras region

Balea Lake Core 4		Lba4_ Co	Lba4_ Cr	Lba4_ Cu	Lba4_ Fe	Lba4_ Mn	Lba4_ Ni	Lba4_ Pb	Lba4_ Zn
Lba4_ Al	Pearso	.593**	.630**	.872**	.756**	.338	.434*	.489**	.527**
	Sig. (2-	.001	.000	.000	.000	.073	.019	.007	.003
	N	29	29	29	29	29	29	29	29
Lba4_ Co	Pearso	1	.765**	.448*	.353	.174	.779**	.070	.200
	Sig. (2-		.000	.015	.060	.365	.000	.717	.298
	N		29	29	29	29	29	29	29
Lba4_ Cr	Pearso		1	.396*	.327	.138	.942**	-.021	.052
	Sig. (2-			.034	.083	.475	.000	.914	.787
	N			29	29	29	29	29	29
Lba4_ Cu	Pearso			1	.817**	.508**	.196	.622**	.596**
	Sig. (2-				.000	.005	.309	.000	.001
	N				29	29	29	29	29
Lba4_ Fe	Pearso				1	.815**	.218	.761**	.682**
	Sig. (2-					.000	.257	.000	.000
	N					29	29	29	29
Lba4_ Mn	Pearso					1	.147	.730**	.618**
	Sig. (2-						.448	.000	.000
	N						29	29	29
Lba4_ Ni	Pearso						1	-.071	.024
	Sig. (2-							.715	.902
	N							29	29
Lba4_ Pb	Pearso							1	.965**
	Sig. (2-								.000
	N								29
Lba4_ Zn	Pearso								1
	Sig. (2-								
	N								

Capra Lake Core 2		LCp2_ Co	LCp2_ Cr	LCp2_ Cu	LCp2_ Fe	LCp2_ Mn	LCp2_ Ni	LCp2_ Pb	LCp2_ Zn
LCp2_ Al	Pearso	.656**	.778**	.466	.828**	.889**	.357	-.699**	-.689**
	Sig. (2-	.004	.000	.059	.000	.000	.160	.002	.002
	N	17	17	17	17	17	17	17	17
LCp2_ Co	Pearso	1	.415	.349	.758**	.628**	.424	-.581*	-.539*
	Sig. (2-		.098	.170	.000	.007	.090	.014	.026
	N		17	17	17	17	17	17	17
LCp2_ Cr	Pearso		1	-.041	.676**	.617**	.684**	-.520*	-.565*
	Sig. (2-			.875	.003	.008	.002	.032	.018
	N			17	17	17	17	17	17
LCp2_ Cu	Pearso			1	.543*	.613**	-.504*	-.130	-.092
	Sig. (2-				.024	.009	.039	.618	.725
	N				17	17	17	17	17
LCp2_ Fe	Pearso				1	.796**	.411	-.385	-.381
	Sig. (2-					.000	.102	.127	.132
	N					17	17	17	17
LCp2_ Mn	Pearso					1	.152	-.590*	-.614**
	Sig. (2-						.560	.013	.009
	N						17	17	17
LCp2_ Ni	Pearso						1	-.361	-.399
	Sig. (2-							.154	.113
	N							17	17
LCp2_ Pb	Pearso							1	.983**
	Sig. (2-								.000
	N								17
LCp2_ Zn	Pearso								1
	Sig. (2-								
	N								

Caltun Lake Core 2		LCt2_ Co	LCt2_ Cr	LCt2_ Cu	LCt2- Fe	LCt2_ Mn	LCt2_ Ni	LCt2_ Pb	LCt2_ Zn
LCt2_ Al	Pearso	.607**	.950**	.791**	.858**	.711**	.828**	.367	.640**
	Sig. (2-	.000	.000	.000	.000	.000	.000	.050	.000
	N	29	29	29	29	29	29	29	29
LCt2_Co	Pearso	1	.674**	.288	.735**	.714**	.842**	-.075	.121
	Sig. (2-		.000	.130	.000	.000	.000	.701	.530
	N		29	29	29	29	29	29	29
LCt2_Cr	Pearso		1	.717**	.941**	.876**	.920**	.142	.446*
	Sig. (2-			.000	.000	.000	.000	.462	.015
	N			29	29	29	29	29	29
LCt2_Cu	Pearso			1	.706**	.506**	.534**	.580**	.834**
	Sig. (2-				.000	.005	.003	.001	.000
	N				29	29	29	29	29
LCt2_Fe	Pearso				1	.931**	.902**	.109	.361
	Sig. (2-					.000	.000	.575	.054
	N					29	29	29	29
LCt2_ Mn	Pearso					1	.911**	-.219	.067
	Sig. (2-						.000	.255	.728
	N						29	29	29
LCt2_Ni	Pearso						1	-.058	.237
	Sig. (2-							.764	.216
	N							29	29
LCt2_Pb	Pearso							1	.841**
	Sig. (2-								.000
	N								29
LCt2_Zn	Pearso								1
	Sig. (2-								
	N								

Podragu Mare Lake Core 2		LPM2_ Co	LPM2_ Cr	LPM2_ Cu	LPM2_ Fe	LPM2_ Mn	LPM2_ Ni	LPM2_ Pb	LPM2_ Zn
LPM2_ Al	Pearso	.798**	.935**	-.161	.700**	.596*	.913**	.237	-.037
	Sig. (2-	.000	.000	.537	.002	.011	.000	.359	.889
	N	17	17	17	17	17	17	17	17
LPM2_ Co	Pearso	1	.672**	.257	.382	.401	.710**	.174	.166
	Sig. (2-		.003	.318	.130	.110	.001	.505	.524
	N		17	17	17	17	17	17	17
LPM2_ Cr	Pearso		1	-.344	.785**	.680**	.985**	.370	.178
	Sig. (2-			.176	.000	.003	.000	.144	.494
	N			17	17	17	17	17	17
LPM2_ Cu	Pearso			1	-.240	-.049	-.320	.098	.015
	Sig. (2-				.353	.850	.211	.709	.954
	N				17	17	17	17	17
LPM2_ Fe	Pearso				1	.957**	.761**	.737**	.113
	Sig. (2-					.000	.000	.001	.665
	N					17	17	17	17
LPM2_ Mn	Pearso					1	.673**	.833**	.200
	Sig. (2-						.003	.000	.442
	N						17	17	17
LPM2_ Ni	Pearso						1	.412	.246
	Sig. (2-							.100	.340
	N							17	17
LPM2_ Pb	Pearso							1	.399
	Sig. (2-								.113
	N								17
LPM2_ Zn	Pearso								1
	Sig. (2-								
	N								

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

Similar trends in trace metal concentration were repeated across most of the lakes in the Rodna region (compared based on the visual appearance of the profiles). Al level in LB 2 correlated significantly with every other metal analysed and a weak correlation with Pb. Al level in LB3-2 correlated significantly only with Ni; it showed weak correlation with Co, Cr and Mn but no correlation with Pb and Zn. Al level in LLM 2 correlated significantly with Cr, Cu, Mn and Ni, it showed weak correlation with Co and Fe but no correlation with Pb and Zn. In LP 1 Al level correlated significantly with Fe and Mn, it showed weak correlation with Co, Cr and Ni but no correlation with Cu, Pb and Zn. Al level in LS 2 correlated significantly with Co, Cr, Cu, Fe and Ni, it showed weak correlation with Mn and Pb but no correlation with Zn. Al level in LV 1 only demonstrated significant correlation with Co, Cr and Ni but no correlation with any other metal in the lake (Table 5.14). Pb and Zn consistently showed surface increase in most of the lakes. The patterns of Pb and Zn concentrations in each lake were quite similar with corresponding peaks and troughs. Higher metal concentrations were recorded in Fagaras than Rodna for example, Balea Lake had Pb and Zn peak concentrations of 279 mg kg^{-1} and 401 mg kg^{-1} respectively while the highest level of Pb was recorded in Bila Lake with the value of 203 mg kg^{-1} (Tables 5.11 and 5.12). The highest level of Zn in Rodna was recorded in Buhaiescu Lake with the value of 149 mg kg^{-1}. The Pb concentrations in Fagaras region range from 31 - 279 mg kg^{-1} while the Zn concentration range from 26 - 401 mg kg^{-1}. In Rodna region the Pb concentrations range from 45 - 203 mg kg^{-1} while the Zn concentrations range from 26 - 149 mg kg^{-1}. It can therefore be seen that the Fagaras lakes displayed higher peak in Pb and Zn concentrations than Rodna lakes which therefore have relatively low concentration of these elements

Table 5.14 Correlation of metals of lake sediments of the lakes in the Rodna/Maramures region

Bila Lake Core 1		LB1_Co	LB1_Cr	LB1_Cu	LB1_Fe	LB1_Mn	LB1_Ni	LB1_Pb	LB1_Zn
LB1_Al	Pearso	.798**	.963**	.750**	.912**	.970**	.864**	.582*	.741**
	Sig. (2-	.000	.000	.000	.000	.000	.000	.011	.000
	N	18	18	18	18	18	18	18	18
LB1_Co	Pearso	1	.725**	.722**	.767**	.823**	.928**	.633**	.704**
	Sig. (2-		.001	.001	.000	.000	.000	.005	.001
	N		18	18	18	18	18	18	18
LB1_Cr	Pearso		1	.655**	.828**	.921**	.773**	.500*	.617**
	Sig. (2-			.003	.000	.000	.000	.035	.006
	N			18	18	18	18	18	18
LB1_Cu	Pearso			1	.932**	.817**	.907**	.902**	.927**
	Sig. (2-				.000	.000	.000	.000	.000
	N				18	18	18	18	18
LB1_Fe	Pearso				1	.943**	.928**	.751**	.904**
	Sig. (2-					.000	.000	.000	.000
	N					18	18	18	18
LB1_Mn	Pearso					1	.912**	.627**	.818**
	Sig. (2-						.000	.005	.000
	N						18	18	18
LB1_Ni	Pearso						1	.775**	.881**
	Sig. (2-							.000	.000
	N							18	18
LB1_Pb	Pearso							1	.813**
	Sig. (2-								.000
	N								18
LB1_Zn	Pearso								1
	Sig. (2-								
	N								

Buhiescu 3 Lake Core 2		LB32_Co	LB32_Cr	LB32_Cu	LB32_Fe	LB32_Mn	LB32_Ni	LB32_Pb	LB32_Zn
LB32_Al	Pearso	.544	.648*	.683*	.625	.737*	.793**	.062	-.047
	Sig. (2-	.104	.043	.029	.053	.015	.006	.866	.898
	N	10	10	10	10	10	10	10	10
LB32_Co	Pearso	1	.673*	.795**	.835**	.902**	.758*	-.721*	-.825**
	Sig. (2-		.033	.006	.003	.000	.011	.019	.003
	N		10	10	10	10	10	10	10
LB32_Cr	Pearso		1	.638*	.691*	.872**	.835**	-.524	-.490
	Sig. (2-			.047	.027	.001	.003	.120	.150
	N			10	10	10	10	10	10
LB32_Cu	Pearso			1	.975**	.739*	.619	-.398	-.549
	Sig. (2-				.000	.015	.056	.255	.100
	N				10	10	10	10	10
LB32_Fe	Pearso				1	.791**	.620	-.541	-.659*
	Sig. (2-					.006	.056	.106	.038
	N					10	10	10	10
LB32_Mn	Pearso					1	.887**	-.616	-.642*
	Sig. (2-						.001	.058	.045
	N						10	10	10
LB32_Ni	Pearso						1	-.351	-.393
	Sig. (2-							.321	.261
	N							10	10
LB32_Pb	Pearso							1	.932**
	Sig. (2-								.000
	N								10
LB32_Zn	Pearso								1
	Sig. (2-								
	N								

Lala Mare Lake Core 2		LLM2_Co	LLM2_Cr	LLM2_Cu	LLM2_Fe	LLM2_Mn	LLM2_Ni	LLM2_Pb	LLM2_Zn
LLM2_Al	Pearso	.706*	.907**	.814**	.752*	.945**	.805**	.601	.308
	Sig. (2-	.023	.000	.004	.012	.000	.005	.066	.386
	N	10	10	10	10	10	10	10	10
LLM2_Co	Pearso	1	.825**	.794**	.626	.761*	.829**	.737*	.391
	Sig. (2-		.003	.006	.053	.011	.003	.015	.264
	N		10	10	10	10	10	10	10
LLM2_Cr	Pearso		1	.932**	.779**	.843**	.835**	.739*	.303
	Sig. (2-			.000	.008	.002	.003	.015	.395
	N			10	10	10	10	10	10
LLM2_Cu	Pearso			1	.928**	.731*	.896**	.892**	.541
	Sig. (2-				.000	.016	.000	.001	.106
	N				10	10	10	10	10
LLM2_Fe	Pearso				1	.637*	.905**	.888**	.671*
	Sig. (2-					.048	.000	.001	.034
	N					10	10	10	10
LLM2_Mn	Pearso					1	.778**	.587	.287
	Sig. (2-						.008	.074	.422
	N						10	10	10
LLM2_Ni	Pearso						1	.890**	.576
	Sig. (2-							.001	.081
	N							10	10
LLM2_Pb	Pearso							1	.656*
	Sig. (2-								.039
	N								10
LLM2_Zn	Pearso								1
	Sig. (2-								
	N								

Pietrosul Lake Core 1 (2006)		LP1_Co	LP1_Cr	LP1_Cu	LP1_Fe	LP1_Mn	LP1_Ni	LP1_Pb	LP1_Zn
LP1_Al	Pearso	.796*	.803*	.595	.943**	.883**	.757*	-.332	-.457
	Sig. (2-	.018	.016	.120	.000	.004	.030	.421	.255
	N	8	8	8	8	8	8	8	8
LP1_Co	Pearso	1	.847**	.827*	.921**	.974**	.675	-.768*	-.859**
	Sig. (2-		.008	.011	.001	.000	.066	.026	.006
	N		8	8	8	8	8	8	8
LP1_Cr	Pearso		1	.918**	.834*	.824*	.700	-.409	-.506
	Sig. (2-			.001	.010	.012	.053	.314	.201
	N			8	8	8	8	8	8
LP1_Cu	Pearso			1	.739*	.744*	.572	-.510	-.574
	Sig. (2-				.036	.034	.139	.197	.137
	N				8	8	8	8	8
LP1_Fe	Pearso				1	.967**	.753*	-.533	-.641
	Sig. (2-					.000	.031	.174	.087
	N					8	8	8	8
LP1_Mn	Pearso					1	.663	-.705	-.787*
	Sig. (2-						.073	.051	.021
	N						8	8	8
LP1_Ni	Pearso						1	-.192	-.398
	Sig. (2-							.648	.328
	N							8	8
LP1_Pb	Pearso							1	.960**
	Sig. (2-								.000
	N								8
LP1_Zn	Pearso								1
	Sig. (2-								
	N								

Stiol Lake Core 2		LS2_Co	LS2_Cr	LS2_Cu	LS2_Fe	LS2_Mn	LS2_Ni	LS2_Pb	LS2_Zn
LS2_Al	Pearso	.895**	.935**	.897**	.944**	.739*	.948**	-.658*	-.133
	Sig. (2-	.000	.000	.000	.000	.015	.000	.039	.715
	N	10	10	10	10	10	10	10	10
LS2_Co	Pearso	1	.936**	.941**	.888**	.762*	.980**	-.637*	-.441
	Sig. (2-		.000	.000	.001	.010	.000	.048	.202
	N		10	10	10	10	10	10	10
LS2_Cr	Pearso		1	.924**	.916**	.677*	.956**	-.588	-.283
	Sig. (2-			.000	.000	.031	.000	.074	.429
	N			10	10	10	10	10	10
LS2_Cu	Pearso			1	.942**	.823**	.974**	-.653*	-.324
	Sig. (2-				.000	.003	.000	.041	.361
	N				10	10	10	10	10
LS2_Fe	Pearso				1	.727*	.938**	-.509	-.071
	Sig. (2-					.017	.000	.133	.846
	N					10	10	10	10
LS2_Mn	Pearso					1	.807**	-.863**	-.311
	Sig. (2-						.005	.001	.382
	N						10	10	10
LS2_Ni	Pearso						1	-.691*	-.360
	Sig. (2-							.027	.307
	N							10	10
LS2_Pb	Pearso							1	.465
	Sig. (2-								.176
	N								10
LS2_Zn	Pearso								1
	Sig. (2-								
	N								

Vinderel Lake Core 3 (2006)		LV3_Co	LV3_Cr	LV3_Cu	LV3_Fe	LV3_Mn	LV3_Ni	LV3_Pb	LV3_Zn
LV3_Al	Pearso	.687**	.804**	.455	.351	-.098	.829**	-.480	.064
	Sig. (2-	.009	.001	.118	.240	.751	.000	.097	.835
	N	13	13	13	13	13	13	13	13
LV3_Co	Pearso	1	.488	.473	.333	-.510	.748**	-.581*	-.250
	Sig. (2-		.091	.102	.266	.075	.003	.037	.411
	N		13	13	13	13	13	13	13
LV3_Cr	Pearso		1	.556*	.502	.195	.723**	-.194	.076
	Sig. (2-			.049	.081	.524	.005	.525	.805
	N			13	13	13	13	13	13
LV3_Cu	Pearso			1	.899**	.380	.646*	.288	.343
	Sig. (2-				.000	.200	.017	.341	.251
	N				13	13	13	13	13
LV3_Fe	Pearso				1	.359	.542	.241	.195
	Sig. (2-					.228	.056	.428	.523
	N					13	13	13	13
LV3_Mn	Pearso					1	.045	.774**	.686**
	Sig. (2-						.884	.002	.010
	N						13	13	13
LV3_Ni	Pearso						1	-.266	.137
	Sig. (2-							.379	.655
	N							13	13
LV3_Pb	Pearso							1	.739**
	Sig. (2-								.004
	N								13
LV3_Zn	Pearso								1
	Sig. (2-								
	N								

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).
*. Correlation is significant at the 0.05 level (2-tailed).

5.5 Other characteristics: Dating (Lake Capra LCP 3)

5.5.1 Lead-210 Activity

Equilibrium depth of total ^{210}Pb activity with the supporting ^{226}Ra appears to occur at c 20 cm of the core (Figure 5.42a). Decline of unsupported ^{210}Pb activities, calculated by subtracting ^{226}Ra activity from total ^{210}Pb activity, shows different features in the core: unsupported ^{210}Pb activities decline more or less exponentially with depth in the top 10 cm of the core, suggesting sediment accumulation rates were relatively uniform in this section (Figure 5.42b); deeper than 10 cm, there are non-monotonic variations in unsupported ^{210}Pb activities with depth, suggesting variations in sediment accumulations.
Inventory of ^{210}Pb activity in the core worked out mean unsupported ^{210}Pb flux at 549 Bq m^{-2} yr^{-1}, implying that there was relatively high level of sediment focussing in the coring location.

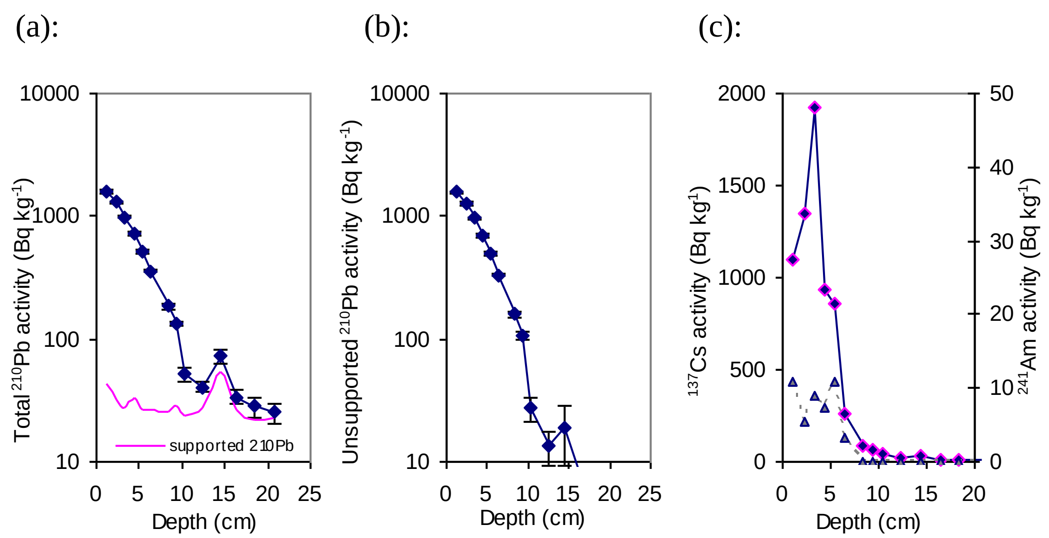


Figure 5.42: Fallout radionuclide concentrations in core LCP3 taken from Lake Capra, Romania, showing (a) total ^{210}Pb, (b) unsupported ^{210}Pb, and (c) ^{137}Cs (red symbols) and ^{241}Am (blue symbols) concentrations versus concentrations versus depth.

5.5.2 Artificial Fallout Radionuclides

The ^{137}Cs activity versus depth shows a well-resolved peak at 3.38 cm (Figure 5.42c) with extraordinary high level of ^{137}Cs activity. With the high level of ^{137}Cs activity, this peak can be confirmed to be derived from the 1986 Chernobyl accident. In the ^{241}Am record, there is a sharp increase in the ^{241}Am activities from 6.38 to 5.38 cm and reach the peak level at 5.38 cm, this peak should be derived from fallout maximum of the atmospheric testing of nuclear weapons around 1963. Comparing with the ^{241}Am record, the ^{137}Cs record shows that the ^{137}Cs activity derived from the 1986 Chernobyl fallout was so high that the peak of ^{137}Cs activity derived from the fallout maximum of the atmospheric testing of nuclear weapons around 1963 in the core was obscured.

5.5.3 Core Chronology

Core chronologies were calculated using the CRS dating model, as the non-monotonic variation in unsupported ^{210}Pb activity has precluded the use of the CIC model (Appleby 2001). The raw CRS model places the 1963 and 1986 layers at 5.3 and 3 cm, respectively, which are in good agreement with the ^{137}Cs and the ^{241}Am records of the core, implying the use of the CRS dating model for the core is reasonable. Core chronologies and sediment accumulation rates calculated from ^{210}Pb activities are shown in Appendix 15. It shows that sediment accumulation rates were relatively uniform in the last about hundred years or so with an average at 0.029 g cm^{-2} yr^{-1}, earlier than that, sediment accumulations varied in a significant high level (Figure 5.43), which may suggest sediment slumping during the time.

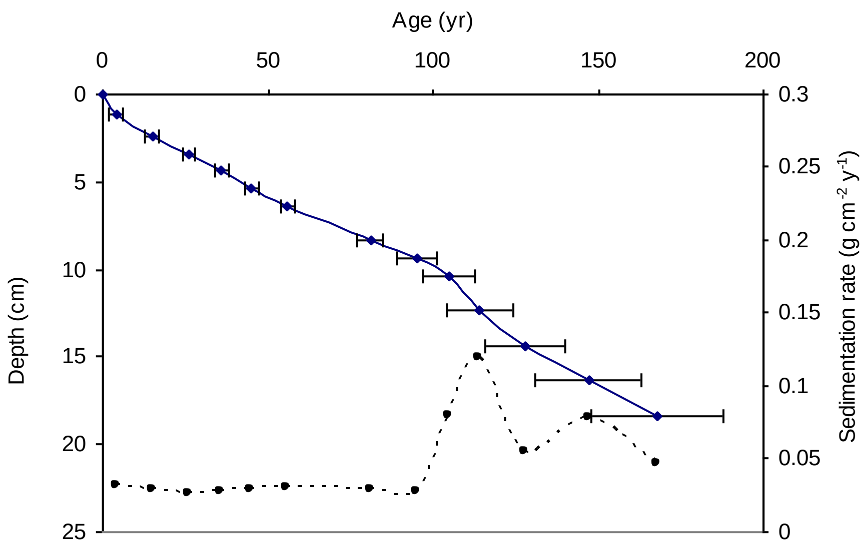


Figure 5.43: Radiometric chronology of core LCP3 taken from Lake Capra, Romania (the solid line is for age, the dashed line is for sedimentation rate).

5.6 Summary of results

Specific and detailed information about the study areas (Fagaras Mountains: south and Rodna/Maramures Mountains: north) were given in this chapter. It also described the data generated on dry bulk density, loss-on-ignition (LOI), particle size, mineral magnetic, geochemical analysis, and radiometric dating of LCp 3. The main findings were that the physical characteristics demonstrated similar trends in down core profiles both in the south and in the north. All the four lakes from the south demonstrated a larger particle size range than the samples in the north. The mineral magnetic measurements demonstrated common characteristics in magnetic concentration parameters X, ARM and SIRM (such as surface peak) but there are obvious variations in the magnitude of the magnetic parameters in both regions. The results of geochemical analysis (both the core profiles and the enrichment factors) showed that the same trends in metal concentration were repeated across the lakes in the South and in the North but the Fagaras lakes displayed higher peak in Pb and Zn concentrations than Rodna lakes. While core chronologies and sediment accumulation rates calculated from ^{210}Pb activities showed that sediment accumulation rates were relatively uniform in the last about hundred years or so with an average at 0.029 g cm^{-2} yr^{-1}, earlier than that, sediment accumulations varied in a significant high level, which may suggest sediment slumping during the time.

CHAPTER 6: Discussion

6.1 Introduction

This chapter presents a discussion of the various investigations in this research and addresses the findings related to the specific research objectives. This research project aims at investigating the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores from selected lakes in the northern and southern Carpathians of Romania in order to evaluate the possibility of using these lakes' sediments as records of recent human impacts and in particular trace metal deposition. Also to assess variations in the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores. The research objectives consist of the determination of the physical characteristics of the lake sediments with a view to determine if further analyses are necessary, the assessment of the potential of using a mineral magnetic approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants and the determination of the geochemistry of the sediments in order to assess the spatial and temporal variations in trace metal deposition. The following hypotheses were therefore tested: The Romanian Carpathians provide suitable sites for palaeolimnological investigation. Such sites record an atmospheric signal providing a retrospective monitor of recent human impacts and any other atmospheric signal identifiable through mineral magnetic and geochemical analyses. Regional variations can be observed (north versus south) in the mineral magnetic and geochemical materials deposits in the sediments hence the spatial and temporal variations in trace metal deposition could be assessed. The hypotheses were tested by subjecting the data to Pearson Correlation using SPSS. The study has therefore, considered the catchment characteristics and investigated the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores from selected lakes in the northern and southern Carpathians of Romania.

The different sections of the discussion chapter consider the physical characteristics of the lake sediments; the potential of using a mineral magnetic approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants. The subsequent sections look at the geochemistry of the sediments in order to examine the spatial and temporal variations in trace metal deposition. The later section integrates the above parameters in order to demonstrate the suitability of the study sites for sediment based study and environmental reconstruction with an assessment of the potential of using these lake sediments as records of temporal and spatial variation in trace metal pollution.

6.2 The physical characteristics of lake sediments and the catchment characteristics of lakes in the studied areas

Usually lake systems are linked to their catchments or drainage systems and to the atmospheric inputs above them as well (Smol, 2008). The materials present in the lake catchments can therefore determine the magnitude of the palaeolimnological changes that may occur within the lake. The physical and chemical conditions existing within a lake per time can be influenced by the ratio of catchment area to lake size (Enache and Prairie, 2000; Smol, 2008). In a lake with a very large catchment area, there may be tendency for such a lake to experience more in-wash of detrital materials from the catchment, than a lake with small catchment area. There might be more in-wash of soil from such catchment resulting in a rapid filling of the lake bed with soil particles. A lake in which the catchment land cover is predominantly grass might have more in-wash of organic materials than its counterpart in which the catchment is predominantly scree. In addition to these characteristics, a lake with a relatively shallow depth might experience more disturbances to the materials at the water sediment interface due to wave action.

There are distinct size variations in the catchment areas of the lakes from both the Fagaras region and the Rodna/Maramures region. There are also disparities in lake areas as there are differences in the lake depths. The effect of the variations in the catchment areas and the lake surface areas gives rise to differences in catchment: lake ratio (see Tables 3.2 and 3.3); for example, all the lakes from the North have larger catchment: lake ratio than the lakes from the South except Caltun Lake (from South) and Lala Mare Lake (from the North) has catchment: lake ratio of 23.3 and 23.0 respectively. This section considers, amongst other things, how the lake catchment characteristics might have impacted on the lake sediment physical characteristics (that is density, LOI and particle size). It has been mentioned in chapter 5 of this thesis that the reason for studying two different regions was to be able to compare and contrast the situation in the two areas and to assess the extent of any regional variations. Therefore, this section also looks into how these physical characteristics relate in the cores within a lake, how they relate in the lakes within a region as it also considers their regional differences.

6.2.1 The physical characteristics of lake sediments and the catchment characteristics of the lakes in the Fagaras region

Among the four lakes investigated in terms of their physical characteristics from the Fagaras region, there are two groups of lakes with contrasting characters. Balea Lake and Podragu Mare Lake demonstrated some conspicuous fluctuations down the cores in their physical characteristics that are similar. However, both Caltun Lake and Capra Lake demonstrated no conspicuous features down the cores in their physical characteristics. In the lakes where multiple cores were analysed in terms of their physical characteristics from this region, the cores showed similarities in their physical characteristics in terms of either surface increase or decrease (e.g. Balea Lake cores 1 and 4). Statistically, there was no significant correlation in the density of Balea Lake cores 1 and 4; at 0.01 significant level the correlation was 0.292 but there was a weak correlation between Capra Lake cores 2 and 3 at 0.05 significant level (0.471). The surface increase or decrease in the characteristics demonstrated the replicability of the measurements between the cores from same lake. All lakes demonstrated a decrease in density towards the surface (see Figure 5.1). The low density zones of each lake are also characterised by a higher organic content. This was statistically justified by strong negative correlations between the LOI and density of both Balea and Podragu Mare lakes respectively. The peaks and troughs in the particle size profiles in all the lakes tallied with the peaks and troughs in each lake's respective sediment density. These demonstrated the comparability of the measurements between the lakes from the same region.

The kind of fluctuations in sediment density demonstrated in Balea Lake core 4 (LBa 4) may signify changes in sediment composition such as particle size and organic matter content. There are corresponding fluctuations in LOI and particle size profiles especially at the middle portion of the length of the core (Figure 6.1a). The alternating peaks and troughs characteristic of LBa 4 physical characteristics (density, LOI and particle size) at points A, B, C, D, E, F and G appear to demonstrate an in-wash of minerogenic materials. Although, the differences in the density and LOI of Balea lake cores 1 and 4 have been well illustrated under section 5.2 in chapter 5 but it must be pointed out that at points A and B there are similarities in the features demonstrated by the two parameters in the two cores (top part of Figure 6.1a). It is most likely therefore, that the conspicuous fluctuations in physical characteristics recorded in core 4 might be the influence of organic material. It might also be due to the non uniform distribution of the in-wash of such material within the lake sediments – this can be a pointer to some spatial differences in the physical characteristics of the lake. In addition to the visual correlation made for the physical characteristics of the lakes, a statistical correlation was also calculated. There was a correlation between the density and LOI in Balea Lake. This further demonstrated the replicability of the cores within same lake.

Within the Fagaras region, Podragu Mare Lake demonstrated some features in its physical characteristics that are similar to Balea Lake. There were series of peaks and troughs in the density and particle size profiles of Podragu Mare Lake core 2 (LPm 2); both physical parameters demonstrated related peaks and troughs in the entire core depth except between 5 - 10 cm where the changes did not obviously correspond but at this depth, there were corresponding changes in density and LOI (Figure 6.1a). Statistical measurement did show a correlation in their physical characteristics (i.e. the relationship in the LOI, density and particle size of the lakes respectively). There was no material left to undertake the LOI measurements for the top slice of Podragu Mare Lake. Podragu Mare lake demonstrated a strong correlation between density and LOI (-0.676).

A close examination of the density, LOI and particle size profiles of Caltun Lake core 2 (LCt 2) reveals some level of fluctuations down the core but this is not as conspicuous as was the case in Balea and Podragu Mare lakes. There was a series of inconspicuous increase and decrease in the physical parameters down the core profile. There was a marginal surface decrease in density from the depth of 1 cm but the surface increase in LOI was about the depth of 3 cm whereas the surface decrease in particle size was from about 5 cm (Figure 6.1b). The peaks and troughs are similar to Capra Lake being not very conspicuous (Figure 6.1b). There was a spike in LOI at about the depth of 27 cm but there were no corresponding spikes in density and particle size - this might be due to in-wash of organic materials from the catchment rather than minerogenic inwash.

Although in Capra Lake core 2 (LCp 2) there were series of inconspicuous peaks and troughs in the density, LOI and particle size profiles down the entire length of the core, however this was not as conspicuous as were observed in Balea and Podragu Mare lakes (Figures 6.1a and 6.1b). There was a surface increase in LOI from the depth of about 5 cm and there were corresponding decrease in density and particle size from this depth as well. This demonstrates an in-wash of minerogenic materials. The features in Caltun and Capra lakes might signify a relatively rapid sedimentation rate. However, both Caltun lake and Capra lake did not demonstrate strong correlations in their physical characteristics as seen in Balea and Podragu lakes. Both the LOI and density of Caltun Lake exhibited weak correlation but there was no correlation in Capra lake LOI and density.

Balea Lake has a catchment: lake ratio of 9.5 which is the smallest in the Fagaras region. It is the only lake where the catchment includes a road network, a road tunnel and a hotel beside the lake. The lake experiences significant tourist number annually (Robert 2005). These factors might have contributed to the in-wash of materials from the catchment to the lake which might have resulted to the fluctuations experience in the physical characteristics. Balea Lake can be said to be consistent with Smol (2008) which states that lake systems are linked to their catchments or drainage systems and to the atmosphere above them as well. The other lakes from the Fagaras region that have larger catchment: lake ratio (e.g. Caltun Lake: 23.3, Podragu Mare Lake: 17.6 and Capra Lake: 15.9) did not demonstrate such fluctuations in physical characteristics that are attributable to catchment in-wash. That indicates probably the physical and chemical conditions existing within these lakes might not be so influenced by the ratio of catchment area to lake size contrary to Enache and Prairie (2000) and Smol (2008).

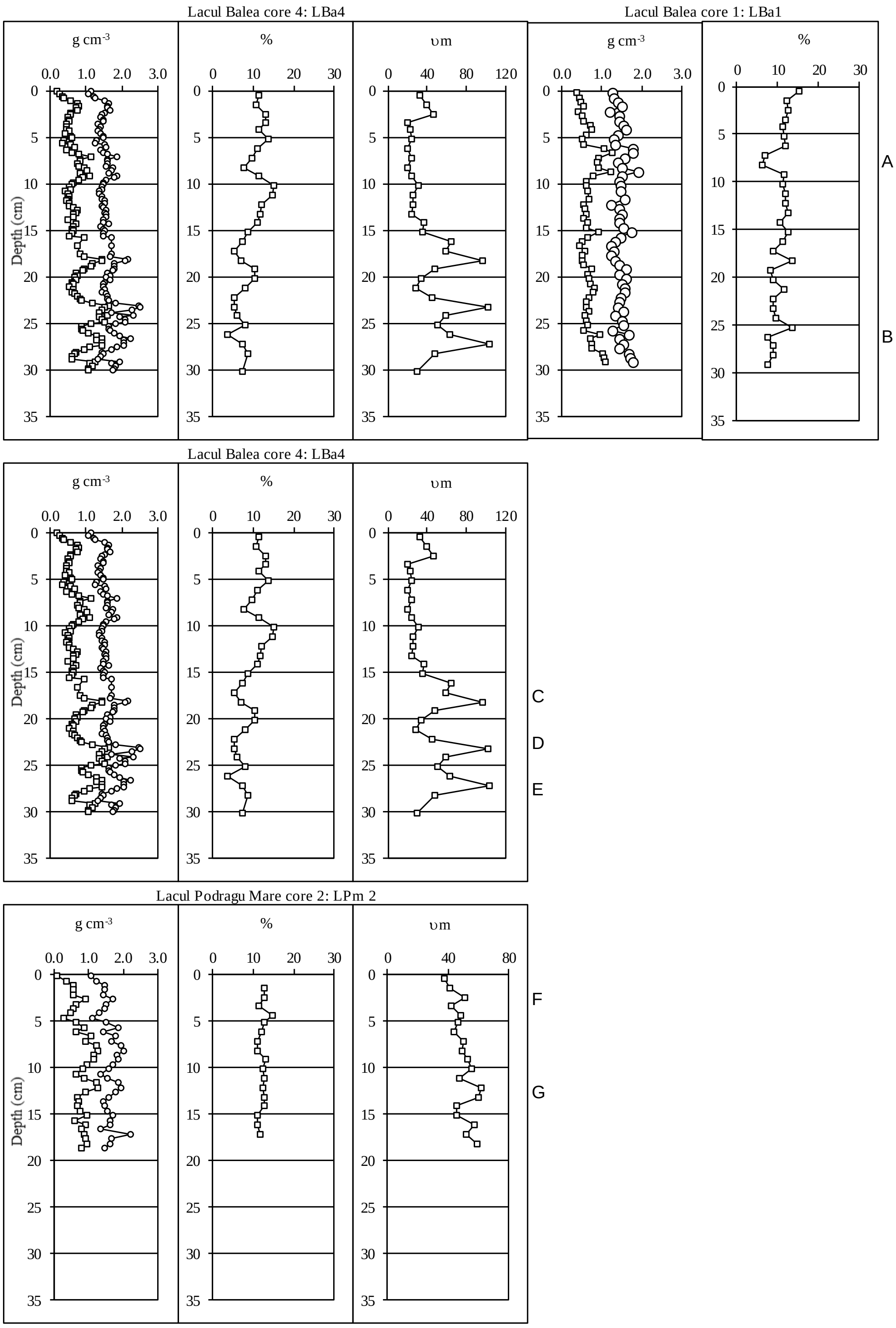


Figure 6.1a: Physical characteristics of lake sediments in the Fagaras region (dry sediment density, g cm^3; LOI, % and mean particle size, μm)

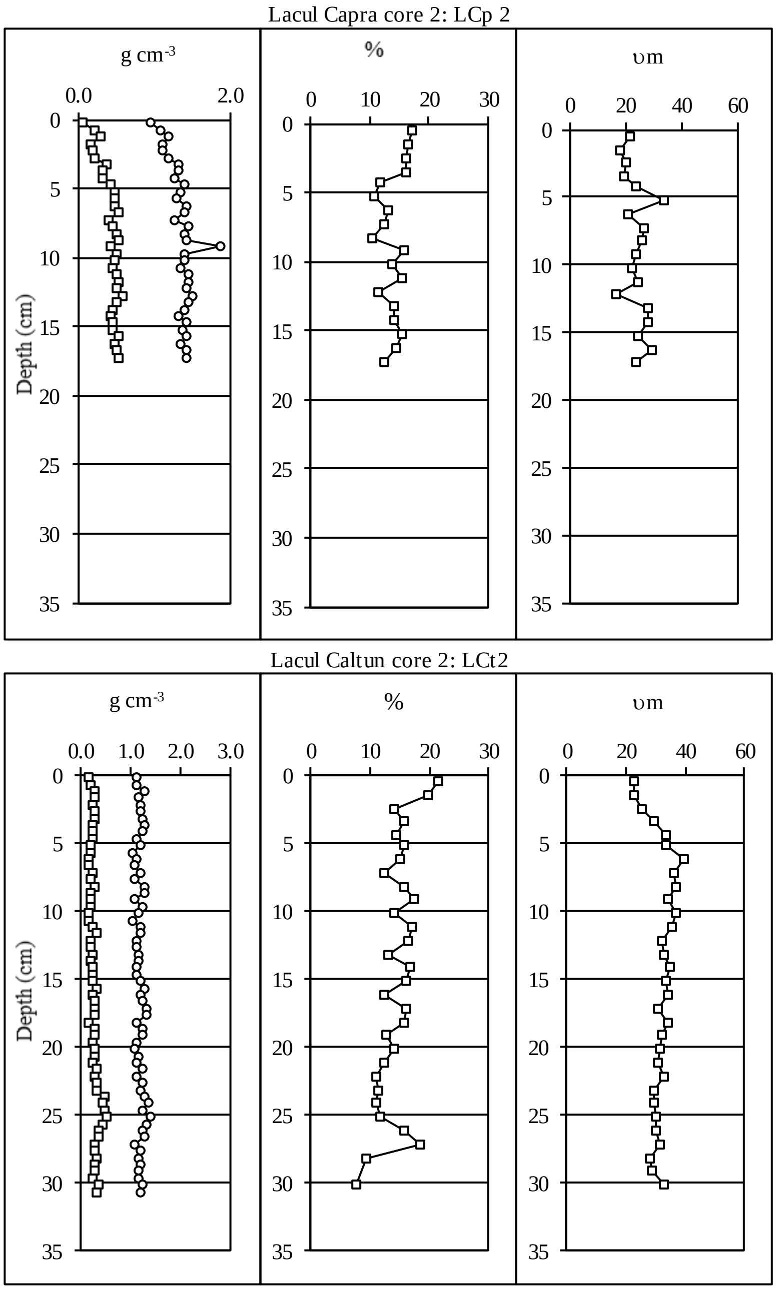


Figure 6.1b: Physical characteristics of lake sediments in the Fagaras region (dry sediment density, g cm^3; LOI, % and mean particle size, μm)

6.2.2 The physical characteristics of lake sediments and catchment characteristics of lakes in the Rodna/Maramures region

Apparently, there are clearer demonstrations of influx of materials in the Fagaras region than in the Rodna/Maramures region. Despite the less conspicuous fluctuations shown in the physical characteristics, the density, LOI and particle size measurements are replicable in the lake sediments in the Rodna/Maramures region as in Fagaras region. In the lakes where multiple cores were analysed in terms of their physical characteristics from this region, the cores showed similarities in their physical characteristics (e.g. Pietrosul Lake). This demonstrated the replicability of the measurements between the cores from same lake. All lakes demonstrated a decrease in density towards the surface. The low density zones of each lake are also characterised by a higher organic content. The peaks and troughs in the particle size profiles in all the lakes tallied with the peaks and troughs in each lake's respective sediment density (see chapter 5: Figures 5.2-5.4). These demonstrated the replicability of the measurements between the lakes from the same region.

Although it might not be very conspicuous, a close examination of the density, LOI and particle size profiles of Bila Lake shows some level of corresponding fluctuations down core (Figure 6.2a). There was a strong correlation between the density and LOI in Bila lake (the calculated 0.824) (Table 5.8). There was no distinguishable fluctuation in density profile of Buhaiescu-3 Lake. The fluctuations in the LOI profile of Buhaiescu-3 are quite obvious. The lake demonstrated corresponding change in the density, LOI and particle size between the depth of 5 - 10 cm (Figure 6.2a). However, there was only a weak statistical correlation between the LOI and density of Buhaiescu-3 whereas, there was no correlation between the LOI and particle size of Buhaiescu-3.

There was no obvious fluctuation in the physical parameters in Lala Mare lake (Figure 6.2a). Vinderel is similar to Lala Mare lake visually as there were no apparent corresponding changes in the physical parameters of Vinderel lake (Figure 6.2b). The above feature might imply rapid sedimentation with less organic in wash to the lakes.

Both Buhaiescu 3 Lake and Lala Mare Lake demonstrated a weak correlation between the density and LOI. However, both cores of Vinderel Lake demonstrated no correlation between density and LOI. Pietrosul Lake shows a surface decrease in density that corresponds with a surface increase in LOI and there was a change in particle size at the corresponding depth (Figure 6.2b). The correlation between the density and LOI in the Pietrosul Lake was strong. Stiol Lake demonstrated a surface decrease in density that corresponds with a surface increase in LOI but there was no corresponding change in particle size at the corresponding depth. Stiol Lake demonstrated a strong correlation between density and LOI; it also showed a strong correlation between LOI and particle size and a weak correlation in density and particle size. Stiol Lake is a special case as this lake has been artificially altered through damming.

The lakes from Rodna/Maramures region have larger catchment: lake ratios than most lakes from the Fagaras region. For example: Buhaiescu-3 Lake has a catchment: lake ratio of 698.9 which is the largest in the region; Bila Lake has a catchment: lake ratio of 312.9 which is the second largest in the region; Stiol Lake has a catchment: lake ratio of 147.2; Pietrosul Lake has a catchment: lake ratio of 132.7; Vinderel Lake has a catchment: lake ratio of 85.0. Lala Mare Lake has the smallest catchment: lake ratio in the region (23.0) whereas, from the Fagaras region, Caltun Lake has a catchment: lake ratio of 23.3 which is the largest in the region. Despite the enormous catchment: lake ratios the lakes did not demonstrate such fluctuations in physical characteristics that are attributable to catchment in wash that is consistent with their catchments. That means probably the physical and chemical conditions existing within these lakes might not be so influenced by the ratio of catchment area to lake size contrary to Enache and Prairie (2000) and Smol (2008). Climate condition might be parts of the influencing factors. Feurdean (2004) and Bodnariuc et al. (2002) described the Romanian climate as continental temperate that varies across the country. The north-western part experiences mild and moist climate, influenced by western oceanic air masses; the eastern part is under the influence of cold and dry air masses from the Russian Plain; warm air masses from sub-Mediterranean areas blow across the southwest region and the southeast is influenced by dry air masses from south-western Asia (Feurdean, 2004; Mindrescu et al., 2010a). Mean annual precipitation is about 1400mm (Bodnariuc et al., 2002). Mean annual temperature is around 8ºC and mean winter and summer temperatures are -3ºC and 12-13ºC, respectively (Feurdean, 2004).

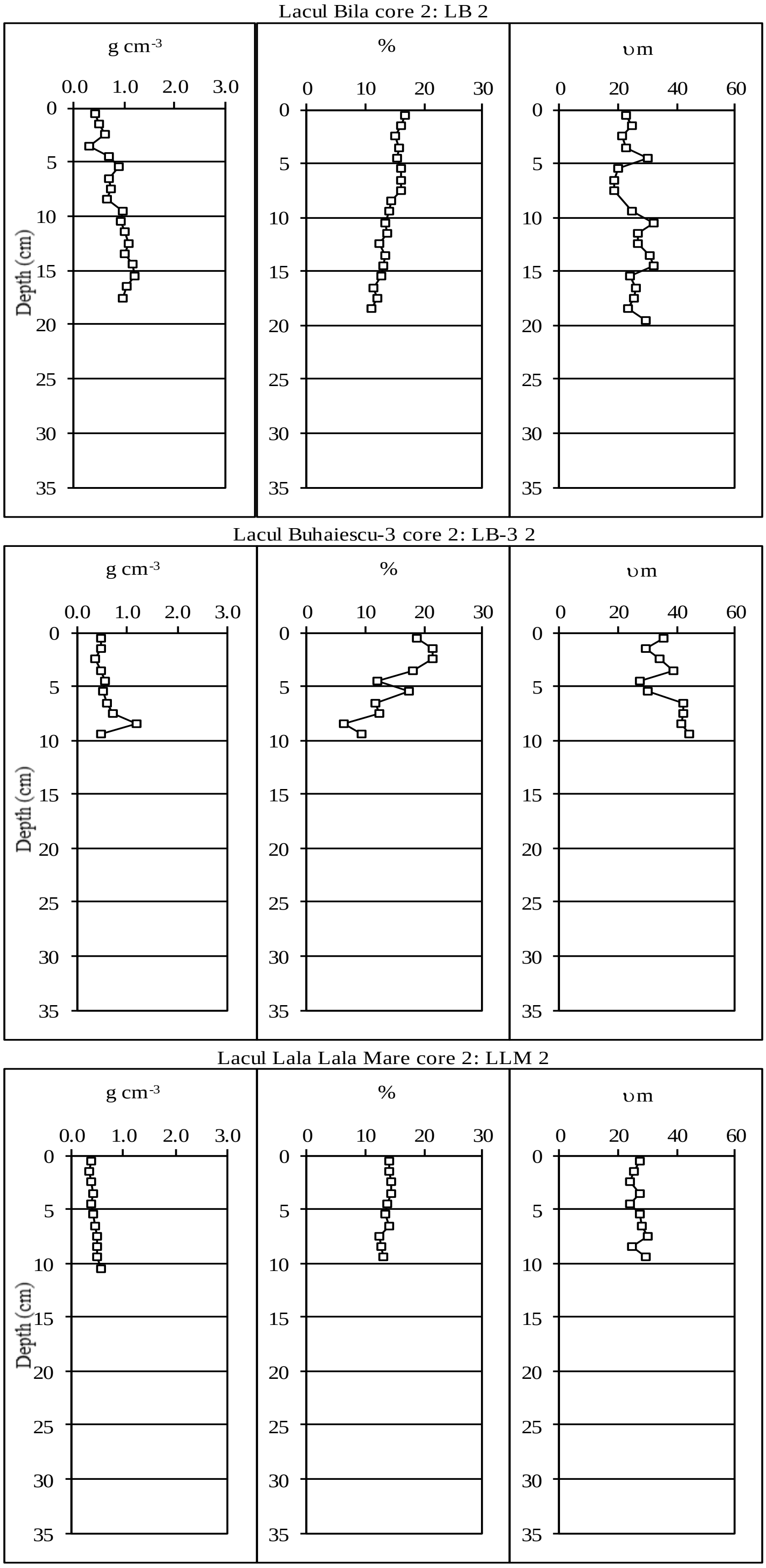


Figure 6.2a: Physical characteristics of lake sediments in the Rodna region (dry sediment density, g cm^3; LOI, % and mean particle size, μm)

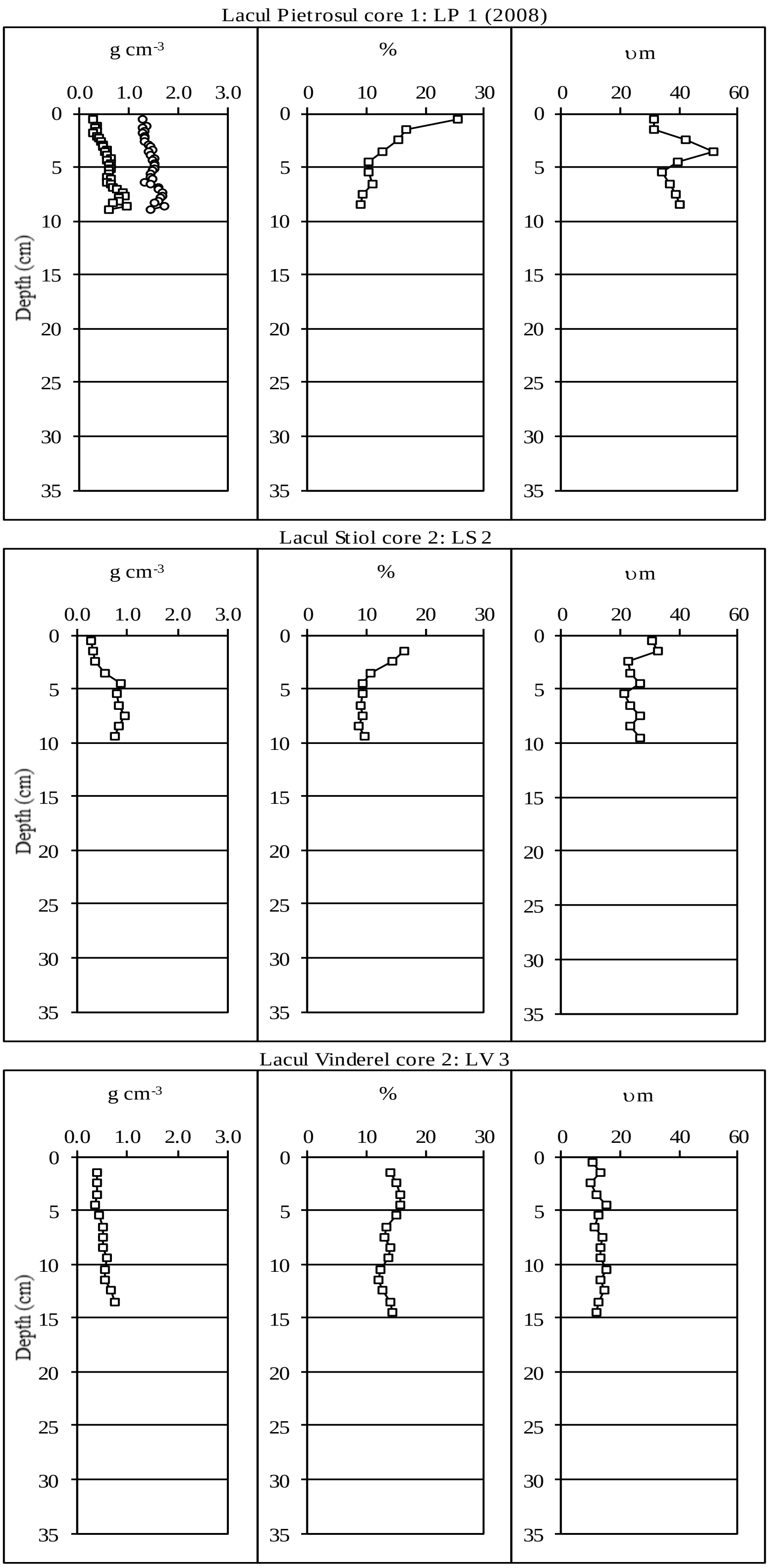


Figure 6.2b: Physical characteristics of lake sediments in the Rodna region (dry sediment density, g cm^3; LOI, % and mean particle size, μm)

6.2.3 Summary of the physical characteristics of lake sediments and the catchment characteristics of lakes in the studied areas

The physical characteristics of Balea lake core 4 from the Fagaras region showed an inwash of coarser material at the bottom of the core. There might be a deferentia deposition of materials within the lake sediments because Balea lake core 1 did not demonstrate same conspicuous features throughout the core length as in core 4. The differences in core features may also reflect variations in coring skills as the cores were taken by different persons. In addition, Balea lake core 1 was the first sample taken in that trip. Two groups of lakes with contrasting physical characteristics are obvious from the four lakes investigated from the Fagaras region. Balea Lake and Podragu Mare Lake demonstrated some conspicuous fluctuations down the cores in their physical characteristics that are similar. However, both Caltun Lake and Capra Lake demonstrated no conspicuous features down the cores in their physical characteristics. Statistical comparison supported the visual observation subgroupings of the lakes from Fagaras region. Both Caltun Lake and Capra Lake did not demonstrate strong correlations in their physical characteristics as seen in Balea and Podragu lakes. Both the LOI and density of Caltun Lake exhibited weak correlation but there was no correlation in Capra lake LOI and density.

In the lakes where multiple cores were analysed in terms of their physical characteristics from Fagaras region, the cores showed similarities in their physical characteristics (e.g. Balea Lake cores 1 and 4) in terms of either surface increase or decrease. The percentage of LOI at 550°C profiles for the various sediment cores studied on Svalbard show few consistent temporal patterns when plotted (Jones and Birks 2004; Brooks and Birks 2004; Boyle et al. 2004). This demonstrated the replicability of the measurements between the cores from same lake. All lakes demonstrated a decrease in density towards the surface. The low density zones of each lake were also characterised by a higher organic content. The peaks and troughs in the particle size profiles in all the lakes tallied with the peaks and troughs in each lake’s respective sediment density. These demonstrated the replicability of the measurements between the lakes from the Fagaras region.

The inconspicuous features demonstrated by Caltun Lake, Capra Lake and all the lakes from Rodna/Maramures region might be due to rapid sedimentation rate. These physical characteristics did show that the cores are replicable. In the lakes where multiple cores were analysed in terms of their physical characteristics from Rodna/Maramures region, the cores showed similarities in their physical characteristics (e.g. Pietrosul Lake). This demonstrated the replicability of the measurements between the cores from same lake. All lakes demonstrated a decrease in density towards the surface except Lala Mare lake. The low density zones of each lake are also characterised by a higher organic content. The peaks and troughs in the particle size profiles in all the lakes tallied with the peaks and troughs in each lake's respective sediment density. These demonstrated the replicability of the measurements between the lakes from the Rodna/Maramures region.

Although, the lakes from Rodna/Maramures region all have larger catchment areas (which mean that they have larger accumulation areas) than the lakes from Fagaras region, there was no conspicuous demonstration of in wash of materials into the lakes from this region. The physical and chemical conditions existing within these lakes have not been so influenced by the ratio of catchment area to lake size contrary to Enache and Prairie (2000) and Smol (2008). The differences in the LOI between the lakes from the two regions are not so enhanced (see chapter 5: Tables 5.3 and 5.4).

Although, some of the lakes from Rodna/Maramures region have stronger correlations between density and LOI than the lakes from the Fagaras region but they are not statistically strong because they have fewer data points per core. Density may flag up changes in the types of sediment composition such as particle size and organic matter content. Loss-on-ignition can be an accurate measurement of primary organic content of sediments, if the fine fraction is present in low percentages (Veres, 2002). Although loss-on-ignition can generally be regarded as an accurate measure of the organic mater content of sediment, the amount of fine fraction in the sediments is thought to be a limiting factor for an absolute organic determination (Veres, 2002).

The use of mean, median and mode in particle size distribution descriptions provide a generalized indication of down-core changes in depositional conditions and processes, but have been found not sensitive to non-normal or polymodal distributions (Beierle, *et al*., 2002). On the other hand, the surface plots of grain size data have been found to be useful in qualitative interpretation of the characteristics of the entire PSD and thus can provide important insights into depositional processes and changing environmental conditions; for example, when used in combination with conventional summary statistics (mean, median and mode), it increases the potential of using grain size as a palaeoenvironmental proxy for identifying changes in clastic and organic depositional processes in lake sequences (Beierle, et al., 2002). Particle size distribution is a fundamental property of sedimentary material and can be used to infer the provenance and history of the sediment transport (Buckley and Cranston, 1991). It has been determined that particle size can affect sediment-related analytical data because finer grained sediments possess larger specific surface areas, surface charges and cation exchange capacities, which enhance the extent of their preferential chemical adsorption (Booth *et al*., 2005). Therefore, the finer the sediment the greater is the concentration of both natural and anthropogenic pollutants such as trace metal concentration (Booth *et al*., 2005). This may result in non-uniform distribution of pollutants over the range of particle size classes causing variations in the chemical composition of sediment samples. A close look at the physicochemical profiles (Figures 6.1 and 6.2) of the lake sediments in this research show that in most cases, the zones of greater particle size correlate with the zones of low Pb and Zn accumulations while the zones of fine particle size correspond to the zones of high Pb and Zn accumulations (e.g. Balea lake from Fagaras region and Pietrosul lake from Rodna/Maramures region).

It was found out that mineral magnetic analysis and geochemical analysis of the lakes sediment show surface increase in concentrations of the metals. Previous research has discovered that heavy metals tend to accumulate in the surface soil horizons in soils with the tendency for concentration of these elements in fine soil particles (such as clay fraction particles less than 0.002 mm) (Shegunova and Atanassov, 2000). Rae (1997) has determined the preferred association of heavy metals with fine-grained sediments is actually a function of sediment mineralogy. This is due to the fact that the important carrier phases of heavy metals which are basically clay minerals and iron oxides are dominantly associated with finer fraction of sediments. This relationship may therefore reflect the important role of iron oxides in controlling heavy metal concentrations in addition to aluminium silicate minerals (Walden *et al*., 1999).

6.3 The potential of using a mineral magnetic approach as a retrospective tool for the assessment of human impacts on the studied lakes including atmospheric deposition of pollutants.

Environmental magnetism has become an important method for studying past global environmental changes in order to reconstruct the processes contributing to palaeoenvironmental change (Hu *et al*., 2002). It has been mentioned earlier in this thesis that the purpose of the mineral magnetic measurements can be to characterise the magnetic mineral content of the sediments and to determine their concentration and grain size variations in the remanence carrying minerals. Anhysteretic remanent magnetization (ARM) is sensitive to the fine magnetic fraction in lake sediment. It is common practice to use the ratio of ARM to isothermal remanent magnetization (IRM) or magnetic susceptibility as a means of inferring relative changes in magnetic grain-size (Hilton, 1986; Hu *et al*., 2002).

The technique of mineral magnetic measurement has been used for detecting anthropogenic pollution caused by power plants (Heller *et al.*, 1998; Kapicka *et al.*, 2000; Hu *et al.*, 2003). Fly ash and burned coal ash discharged by power plants are known to be always strongly magnetic, and can be easily detected by magnetic means (Hu *et al*, 2003). Stockhausen and Zolitschka, (1999) show that magnetic measurements of lake sediments reflect the type and the amount of magnetic grains transported to the lake from its catchment and show that: χ, ARM and SIRM are primarily sensitive to concentration variations of ferromagnetic minerals.

Most lakes investigated in this research demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of their core profiles with a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’ (Figures 5.13-5.26). The combination of these parameters appears to indicate the predominance of relatively coarse grain ferromagnetic material at the upper surface of the lakes. These features appear to indicate both an increase in magnetic concentration, and a change in grain size and mineralogy; they might demonstrate the influence of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions but, the surface increase in the magnetic parameters mentioned above might be also due to some micro-biological activities within the lake's sediment. Furthermore, a detrital input from the lakes catchment might be the source of magnetic minerals in the lakes sediments (e.g. Oldfield, 1991).

6.3.1 Integration of the mineral magnetic characteristics of lake sediments in the Fagaras and Rodna/Maramures regions

Variation in magnetic susceptibility (χ) can indicate changes in the concentration of magnetic minerals (Juyal *et al*., 2009). Most cores in this research show clear surface peak. The magnetic susceptibility (χ) values are relatively high (see chapter 5: Figures 5.13-5.26). Magnetic susceptibility is a measure of the ease with which a substance can be magnetised (Dearing, 1999). Susceptibility depends principally on the nature and concentration of magnetic minerals in a sample. Ferrimagnetic minerals generally dominate over paramagnetic minerals. ARM is said to be particularly sensitive to fine-grained ferromagnetic minerals such as single-domain magnetite; while all three parameters additionally respond to a minor extent to grain size variations and to changes in the magnetic mineralogy (Hutchinson, 1990; Hutchinson 2005). Zhang *et al.* (2001) classified the ratio parameters ARM/X and ARM/SIRM as grain size indicators that the higher the values, the finer the grain size of magnetite (Oldfield, 1994).

Down core profiles of χ, ARM, SIRM and IRM of Fagaras Lake (e.g. Balea lake core 4) show that the intervals of about 8.0 - 0 cm are characterized by significantly high concentration of magnetic minerals. The variations of the three concentration parameters are generally consistent, but they are not exactly proportional (see chapter 5: Figures 4.13-5.18). For example, the increase of ARM is not so enhanced compared to χ and SIRM at 8.0 - 0 cm. In the Rodna region (e.g. Vinderel Lake), down core profiles of χ, ARM, SIRM and IRM show that the intervals of about 6.0 - 0 cm are characterized by significantly high concentration of magnetic minerals (see chapter 5: Figures 5.19-5.26). As in the Fagaras lakes, the variations of the three concentration parameters are generally consistent, but they are not exactly proportional. These signify the grain-size dependency of the magnetic concentration parameters. Relatively coarse magnetic grains may be dominant at the surface compared to the other sandy layers (Inouea *et al., 2004)*.

The detrital input from the lake catchment can also be a source of magnetic minerals in the lake sediments; this has been researched by Oldfield (1991). It might be that the magnetic minerals within the lake sediments could originate from other sources such as diagenesis. The authigenic/diagenetic processes in organic-rich sediments could produce magnetic minerals such as greigite (Zhu *et al.*, 2001). This has been considered in 2.2.5. Research shows that the responses of greigite and magnetite to varying external magnetic fields are similar (e.g. Hu et al., 2002). Another probable source of magnetic minerals in the lake sediments can be due to the action of certain redox sensitive bacteria (magnetotactic bacteria) that are capable of precipitating magnetic minerals, especially magnetite (Oldfield and Wu, 2000; Cohen, 2003). Research by several authors, (Blakemore, 1975; Heywood *et al.,* 1990; Mann *et al.,* 1990; Bazylinski *et al*., 1993; Bazylinski *et al*., 1995; Oldfield and Wu, 2000; Cohen, 2003) reveal the capability of certain group of bacteria to be magnetically aligned (magnetotactic bacteria). Magnetotactic bacteria produce two general types of minerals as the mineral phases of their magnetosomes (Balkwill *et al*; 1980): iron oxides and iron sulphides. The iron oxides include only ferrimagnetic magnetite (Fe_30_4) and the iron sulphides, ferrimagnetic greigite (Fe_30_4) and non-magnetic pyrite (FeS_2) (Bazylinski *et al.,* 1994). These intracellular particles confer a permanent magnetic dipole moment to the cell resulting in the cell's magnetotactic response, i.e. a motile, bio magnetic compass (Bazylinski, 1996). The above extra likely sources of magnetic materials were not exploited in the laboratory in this research. It has been mentioned that environmental magnetism is an important method for studying past global environmental changes in order to reconstruct the processes contributing to palaeoenvironmental change. Therefore, it is recommended that the palaeolimnologist derive a complete understanding of the potential origin of the magnetic signal observed (Hu et al., 2002).

It has been determined that most lakes investigated in this research demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper part of their core profiles with a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’ (see chapter 5: Figures 5.13-5.26). Statistically there was a trend in the surface concentrations of the magnetic parameters. When the data were subjected to correlation analysis, χ, showed significant correlation with ARM, SIRM, SIRM/ARM, ARM/ χ, SOFT, 20mT and 40mT in LBa 4; in LCp 3 χ, showed strong correlation with SIRM, SOFT, HARD, 20mT, 40mT and 300mT. While in LCt 2, χ, correlated significantly with ARM, SIRM, SOFT, HARD, 20mT, 40mT, 100mT and 300mT. χ, showed strong correlation with ARM, SIRM, SOFT and 20mT LPm 2 (Fagaras region). All the lakes in Rodna region demonstrated significant correlations in all the magnetic parameters except LB-3 that showed correlations only in χ, ARM, SIRM, SOFT and LLM 2 that showed no signicant correlation in any of the parameters. The combination of these parameters indicates the predominance of relatively coarse grain ferromagnetic material at the upper surface of the lake. These features indicate both an increase in magnetic concentration, and a change in grain size and mineralogy; they demonstrate the influence (probably) of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions. It is therefore evident from the above that there was apparent demonstration of influence of atmospheric particulate pollution on the sediments of the lakes both from the Southern region and the Northern region of the Romanian Carpathian Mountains.

Such features that indicate both an increase in magnetic concentration, a change in grain size and mineralogy and the demonstration of the likely influence of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions, have been identified across Europe (*e.g.*, Oldfield and Richardson, 1990; Hutchinson, 1995; Rose *et al.*, 2009) and in other part of the world such as Himalayas in India (e.g. Alagarsamy, 2009). The low susceptibility zone in all the lakes (in both regions) has a coarser size distribution than elsewhere in the cores. The obvious peaks and troughs shown in the particle size correspond to the troughs and peaks in the magnetic susceptibility (Figures 5.7-5.26). All three parameters are said to decrease with increasing grain size if grain sizes are larger than grains at the superparamagnetic/single-domain boundary (Oldfield, 1994; Stockhausen and Zolitschka, 1999). ARM/SIRM has been reported to be a non-linear function of concentration with low concentrations acquiring a relatively higher ARM/SIRM than high concentrations (Stockhausen and Zolitschka, 1999). Dia- or paramagnetic minerals may also influence ARM /χ and SIRM/χ. In addition, the latter is particularly sensitive to the presence of the iron sulphide greigite (e.g. Snowball, 1991).

It has been stated earlier in this chapter that anhysteretic remanent magnetization (ARM) is sensitive to the fine magnetic fraction in lake sediment. It has been stated as well that palaeolimnologists often use the ratio of ARM to isothermal remanent magetization (IRM) as a means of inferring relative changes in magnetic grain-size. The ratio of ARM to magnetic susceptibility can be used as well (e.g.Hu *et al*., 2002). Researchers flagged it up that this method gives reliable results only when one magnetic mineral is present within the sediment, or when one magnetic mineral predominates. Such ratios might not correctly demonstrate changes in relative magnetic grain-size when two or more magnetic minerals make substantial magnetic contributions (Hilton, 1986).

There are obvious demonstrations of temporal and spatial variations in the mineral magnetic characteristics of the studied sites. The magnetic characteristics are similar between cores from the same lake and are comparable among the lakes in the Fagaras region except for Caltun Lake which had relatively low values for all magnetic concentrations. In addition to the small SIRM value, there are no apparent changes noticeable in magnetic fluctuations down the depth of the lake. Although the core LBa 1 (from Balea Lake) did not demonstrate surface increase in the magnetic concentration parameters χ, ARM and SIRM, its peaks and troughs are slightly comparable to LBa 4 (from Balea Lake). The changes in magnetic fluctuations noticeable in the two cores are more distinct in LBa 4 than LBa 1 (Figure 6.3). The SIRM profiles of Caltun, Capra and Podragu Mare lakes demonstrated rapid and smooth sedimentation. The magnetic concentration parameters χ, ARM and SIRM are similar between cores of the same lake and are similar among most of the lakes in the Rodna/Maramures region as well. All cores show clear surface peak in the magnetic concentration parameters χ, ARM and SIRM, except Stiol Lake which demonstrated subsurface peak and Lala Mare Lake which did not show any clear fluctuations in the magnetic concentration parameters χ, ARM and SIRM (Figure 6.4). The surface values of SIRM are relatively low and the fluctuations down the depth of the lakes are not absolutely conspicuous. In all the sites that demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of their core profiles; at this peak there is also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters ‘soft’ and ‘hard’ (see chapter 5: Figures 5.13-5.26). These features indicated both an increase in magnetic concentration, and a change in grain size and mineralogy; they demonstrated the possible influence of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions.

Although there are variations in the magnitudes of the magnetic parameters in both regions (e.g. SIRM); for example, the highest value of SIRM in the Fagaras region being 1285.11 x $10^{-5}Am^2kg^{-1}$ (Balea Lake) while the highest value of SIRM in the Rodna region is 229.56 x 10^{-5} Am^2 kg^{-1} (Pietrosul lake). If SIRM is taken as an indicator of level of pollution of the lakes in the two regions; then it is apparent that the lakes in the Fagaras region are more polluted than the lakes in the Rodna region. It is therefore evident from the above that there was obvious demonstration of influence of atmospheric particulate pollution on the sediments of the lakes both from the Southern region and the Northern region of the Romanian Carpathian Mountains. Therefore, the use of mineral magnetic approach for the study of Romanian lake sediments was appropriate.

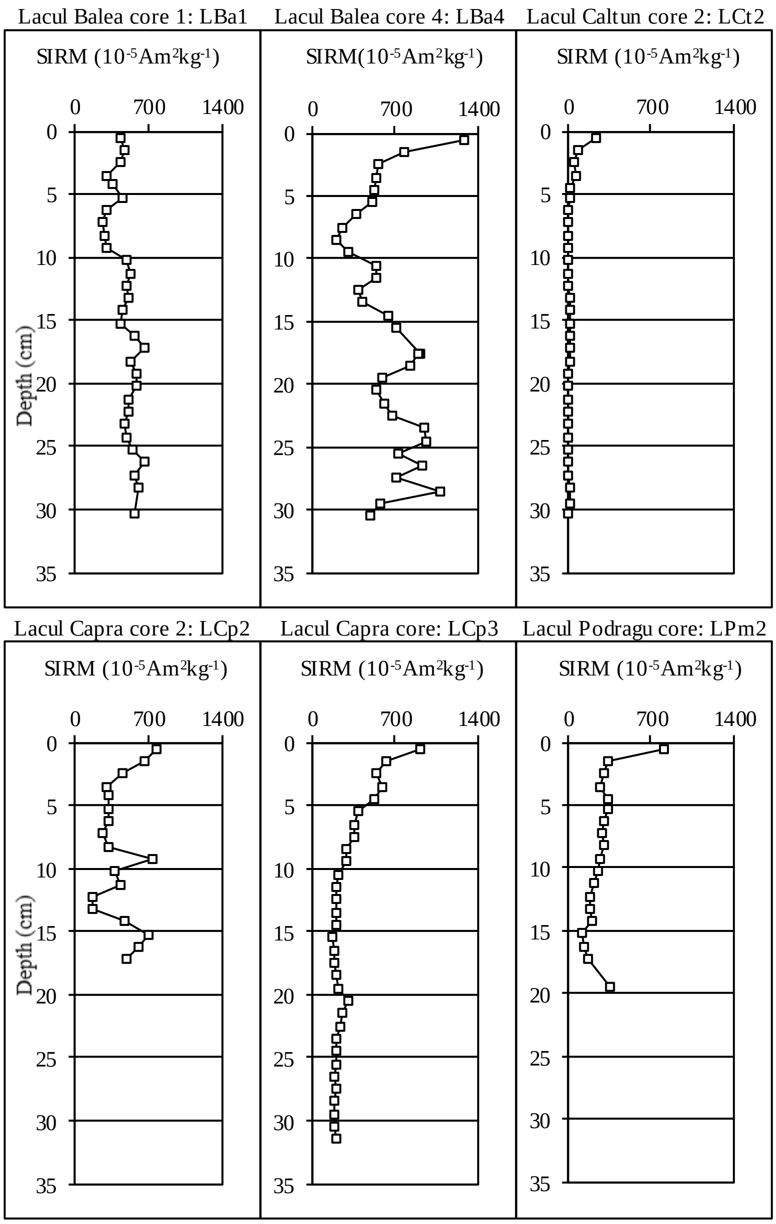


Figure 6.3: SIRM profiles of lake sediment cores in the Fagaras region

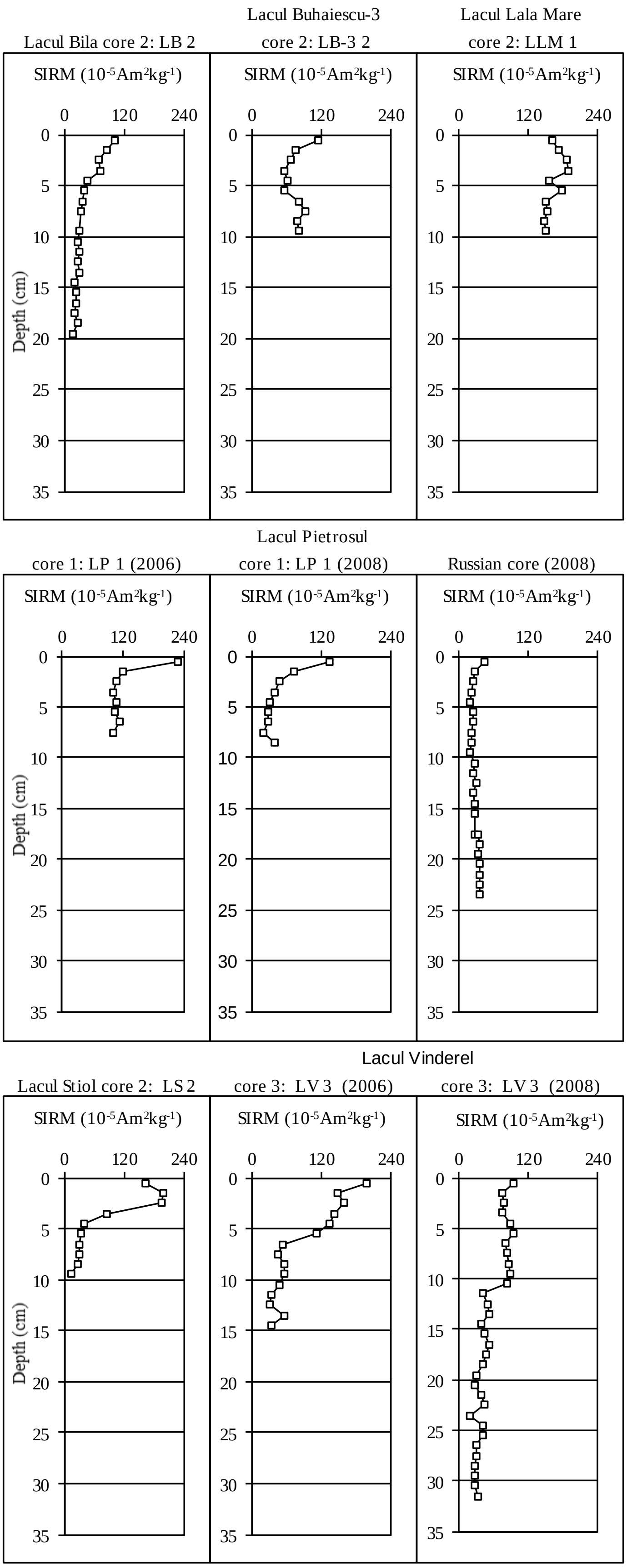


Figure 6.4: SIRM profiles of lake sediment cores in the Rodna region

6.4 Geochemistry: Assessment of contamination levels (the levels and the variations of the trace metals)

It has been mentioned earlier in this thesis that trace metal-polluted sediments have become a global concern due to their increasing prevalence in ecosystems (Grandlic *et al*., 2006). Researchers (e.g. Audry *et al*., 2004) have demonstrated that anthropogenic activities have caused important transformations in aquatic environments within the recent years and that trace metals are among the most widespread of the various pollutants originating from anthropogenic activities (e.g. Salomons, 1995; Dar, 1997; Hochella *et al.,* 1999; von Braun *et al.,* 2002). The concentrations of trace metals in different ecological environments, particularly in water and sediments of lakes, are considered to be the major factor in environmental pollution (Vital and Statteger, 2000).
This section examines the concentrations of trace metals in the lakes sediment in the Fagaras and Rodna/Maramures regions, in a comparative manner in order to assess their regional levels. It considers first the enrichment factors then, the down core variations of sediment lead (Pb) and zinc (Zn) profiles in the studied areas.

6.4.1 Enrichment factors

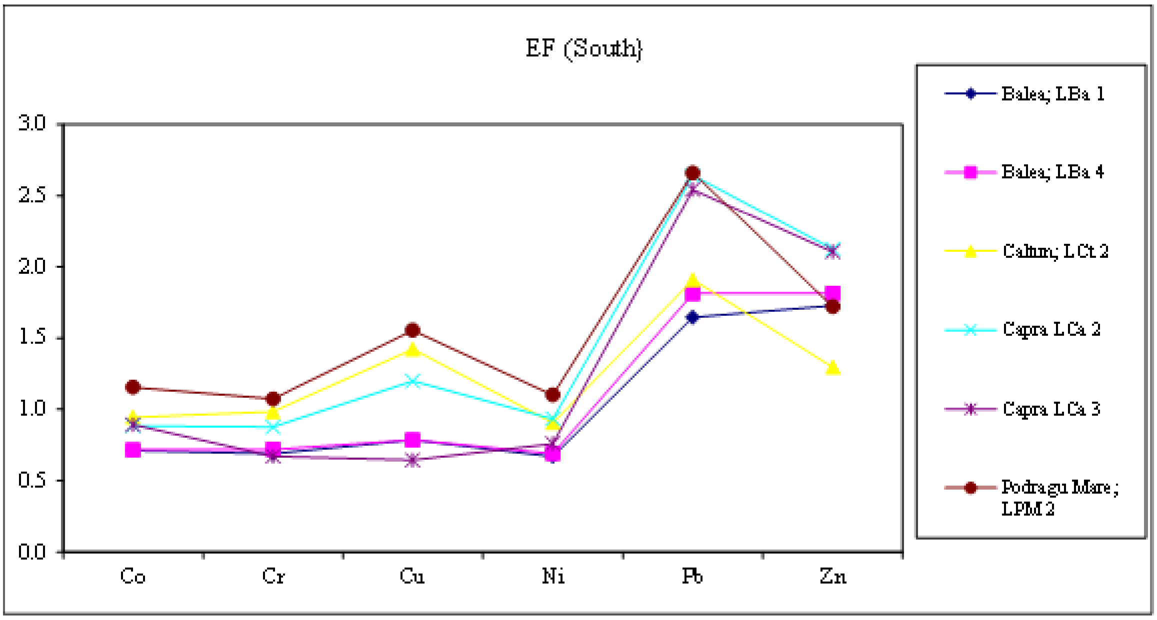
Enrichment Factors (EFs) are used as a means of identifying and quantifying human interference with global element cycles (Karageorgis *et al*., 2009). The details of the various approaches employed to quantify the EF can be found in Reimann and de Caritat (2000, 2005). The EFs of metals from each lake was estimated by dividing the surface (top) value for each metal by the bottom (background) value. When the value derived from such calculation is 1 or less than 1 then, at such point the lake sediment was not enriched. Therefore, the higher the calculated value in contrast to 1 then, the greater the enrichment.

Table 6.1: Fagaras Region (South) lakes Enrichment Factor (EF)

Name of lake	Depth	Metals					
		Co ($mgkg^{-1}$)	**Cr** ($mgkg^{-1}$)	**Cu** ($mgkg^{-1}$)	**Ni** ($mgkg^{-1}$)	**Pb** ($mgkg^{-1}$)	**Zn** ($mgkg^{-1}$)
Surface							
Balea; LBa 1	0.50	29.9	172.2	91.1	144.3	261.3	375.2
Balea; LBa 4	0.50	30.2	172.3	91.1	143.8	278.8	308.5
Caltun; LCt 2	0.50	22.0	37.8	80.0	37.0	163.3	163.7
Capra LCa 2	0.50	23.4	112.5	76.9	103.1	252.0	240.6
Capra LCa 3	0.50	41.9	96.6	67.4	71.8	243.9	183.4
Podragu Mare; LPM 2	0.50	17.3	78.5	74.8	50.6	170.2	148.3
Background							
Balea; LBa 1	30.25	42.0	249.5	116.0	215.7	158.9	217.1
Balea; LBa 4	30.50	42.1	239.6	116.1	208.3	153.9	170.3
Caltun; LCt 2	31.25	23.3	38.4	56.3	40.8	85.6	126.4
Capra LCa 2	17.25	26.4	128.4	64.2	110.5	95.4	113.4
Capra LCa 3	30.50	47.0	144.0	104.7	94.7	95.9	87.2
Podragu Mare; LPM 2	18.25	15.0	73.1	48.1	46.0	64.0	86.2
EF							
Balea; LBa 1		0.7	0.7	0.8	0.7	1.6	1.7
Balea; LBa 4		0.7	0.7	0.8	0.7	1.8	1.8
Caltun; LCt 2		0.9	1.0	1.4	0.9	1.9	1.3
Capra LCa 2		0.9	0.9	1.2	0.9	2.6	2.1
Capra LCa 3		0.9	0.7	0.6	0.8	2.5	2.1
Podragu Mare; LPM 2		1.2	1.1	1.6	1.1	2.7	1.7

Table 6.2: Rodna Region (North) lakes Enrichment Factor (EF)

Name of lake	Depth	Metals					
		Co ($mgkg^{-1}$)	**Cr** ($mgkg^{-1}$)	**Cu** ($mgkg^{-1}$)	**Ni** ($mgkg^{-1}$)	**Pb** ($mgkg^{-1}$)	**Zn** ($mgkg^{-1}$)
Surface							
Bila; LB 2	0.5	6.8	43.9	41.0	41.0	186.4	131.7
Buhaiescu-3; LB-3 2	0.5	4.7	30.4	29.1	24.9	116.2	148.4
Lala Mare; LLM 2	0.5	11.3	45.4	35.3	43.4	100.3	115.2
Pietrosul; LP 1(06)	0.5	7.6	31.6	31.4	31.3	86.7	108.7
Pietrosul; LP 1(08)	0.5	12.0	23.4	38.6	25.3	140.1	129.5
Stiol; LS 2	0.5	4.5	45.1	22.7	31.5	87.0	105.6
Vinderel LV 3(06)	1.5	7.8	33.8	21.4	36.0	103.6	105.1
Vinderel LV 1(08)	0.5	14.3	40.7	48.1	51.7	122.7	126.6
Background							
Bila; LB 2	19.5	6.1	47.4	20.4	38.2	140.2	119.1
Buhaiescu-3; LB-3 2	9.5	10.4	35.4	32.1	29.4	59.5	92.1
Lala Mare; LLM 2	9.5	13.9	50.4	40.8	43.6	97.1	105.6
Pietrosul; LP 1(06)	7.5	8.8	26.3	18.8	23.3	44.7	79.3
Pietrosul; LP 1(08)	8.5	15.2	36.5	33.2	37.9	61.1	83.6
Stiol; LS 2	9.5	13.9	92.2	52.3	57.4	79.2	95.6
Vinderel LV3(06)	13.5	10.4	37.3	20.5	37.1	70.4	99.4
Vinderel LV1(08)	30.5	16.9	39.2	41.2	55.0	95.8	112.9
EF							
Bila; LB 2		1.1	0.9	2.0	1.1	1.3	1.1
Buhaiescu-3; LB-3 2		0.4	0.9	0.9	0.8	2.0	1.6
Lala Mare; LLM 2		0.8	0.9	0.9	1.0	1.0	1.1
Pietrosul; LP 1(06)		0.9	1.2	1.7	1.3	1.9	1.4
Pietrosul; LP 1(08)		0.8	0.6	1.2	0.7	2.3	1.6
Stiol; LS 2		0.3	0.5	0.4	0.5	1.1	1.1
Vinderel LV 3(06)		0.8	0.9	1.0	1.0	1.5	1.1
Vinderel LV 1(08)		0.8	1.0	1.2	0.9	1.3	1.1



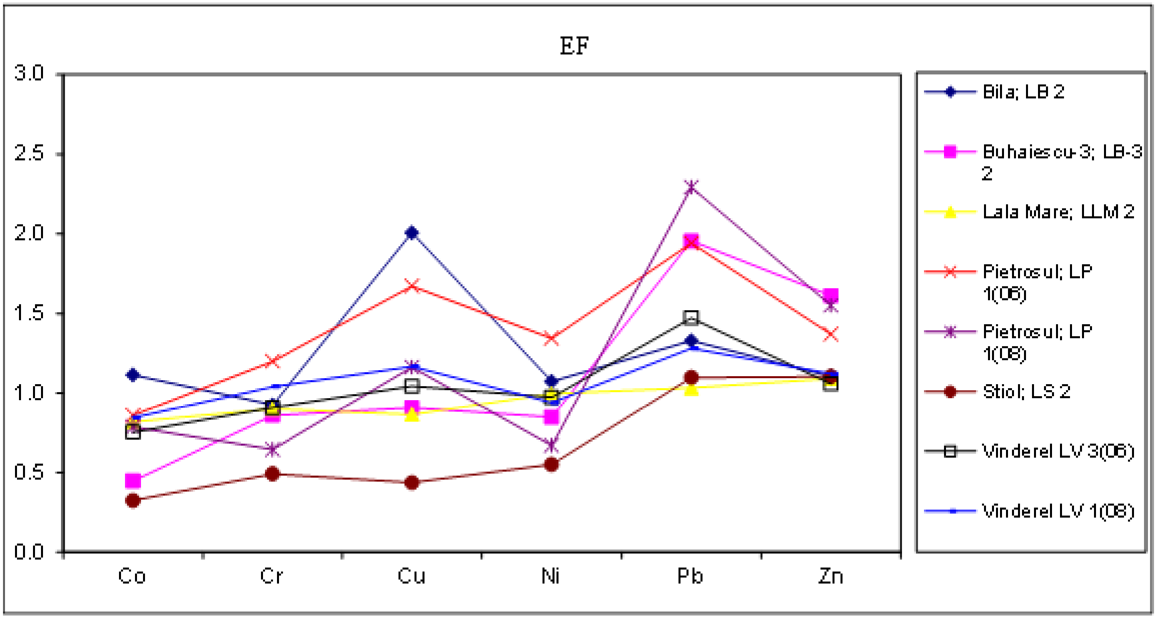


Figure 6.5: South and North lakes Enrichment Factor (EF) compared

Although there are differences in the magnitudes, all the lake sediments in the South are enriched with Pb and Zn. Podragu Mare Lake has the highest level of enrichment in Pb with an EF of 2.7. Capra Lake follows with an EF of 2.6 (LCp 2) and 2.5 (LCp 3); followed by Caltun Lake which has an EF of 1.9. Balea Lake was the least enriched lake in the South; LBa 4 has an EF of 1.8 and LBa 1 with an EF of 1.6. In terms of Zn accumulation in the South lakes LCt 2 was the least enriched with an EF of 1.3 while the highest enrichment was recorded in LCp 3 with an EF of 2.1. Only Podragu Mare Lake and Caltun Lake were sparingly polluted with Cu with EF of 1.6 and 1.4 respectively. As per the other metals present in the lakes they were represented in low magnitudes (Figure 6.5 and Table 6.3). Only Podragu Mare Lake was enriched in Co (1.2), Cr (1.1) and Ni (1.1). The EF for Pb in the sediments of Koumoundourou Lake in Greece was 10.2 (Karageorgis *et al*., 2009). This implies that the pollution level of Koumoundourou Lake in Greece is about three times Podragu Mare Lake's in Romania (Podragu Mare Lake has the highest level of enrichment in Pb with an EF of 2.7). This implies that the lakes in this region are relatively clean. A statistical approach showed that Al inflow into LBa 4 correlated significantly with every other metal analysed except Mn and Ni. Likewise Al inflow into LCp 3 correlated significantly with every other metal analysed except Cu and Ni. Also, Al inflow into LCt 2 correlated significantly with every other metal analysed except Pb. In LPm 2 Al inflow correlated significantly with every other metal analysed except Cu, Mn, Pb and Zn. Al inflow into these lakes might be through atmospheric or catchment supply (see Table 5.13).

Unlike in the South where all lakes have possibly been enriched above the background level by Pb and Zn only three lakes in the North (Buhaiescu Mare, Pietrosul and Vinderel) show enrichment in Pb (Figure 6.5 and Table 6.4). Only two lakes (Buhaiescu Mare and Pietrosul) show enrichment in Zn in the North. Bila and Pietrosul demonstrate enrichment in Cu. The EF in every other lakes fall below 1.5. Pietrosul Lake has the highest level of enrichment in Pb with an EF of 2.3. Buhaiescu Mare Lake follows with an EF of 2.0 while Vinderel Lake has an EF of 1.5. Pietrosul Lake recorded the highest enrichment in Zn with the value of 1.6 and for Cupper enrichment; Bila and Pietrosul have the value of 2.0 and 1.7 respectively. Pietrosul Lake seems the most polluted of all the lakes from the North. Statistically, Al inflow into LB 2 correlated significantly with every other metal analysed and a weak correlation with Pb. Al inflow into LB3-2 correlated significantly only with Ni; it showed weak correlation with Co, Cr and Mn but no correlation with Pb and Zn. Al inflow into LLM 2 correlated significantly with Cr, Cu, Mn and Ni, it showed weak correlation with Co and Fe but no correlation with Pb and Zn. In LP 1 Al inflow correlated significantly with Fe and Mn, it showed weak correlation with Co, Cr and Ni but no correlation with Cu, Pb and Zn. Al inflow into LS 2 correlated significantly Co, Cr, Cu, Fe and Ni, it showed weak correlation with Mn and Pb but no correlation with Zn. Al inflow into LV 1 only demonstrated significant correlation with Co, Cr and Ni but no correlation with any other metal in the lake. There was significant correlation between Pb and Zn in LB 2, LB3-2, LP 1 and LV 1 and a weak correlation in LLM 2 but no correlation in LS 2 (see Table 5.14).

The surface enrichment (by Pb and Zn) in all the four lakes from the south and three lakes from the north of Romania are consistent with the observation of Rose, *et al.* (2009) on Lacul Negru in the Retezat National Park of Romania where all trace metals demonstrated surface enrichment. It has been observed across much of Europe that trace metals have demonstrated consistent surface enrichment; the feature that is attributable to an increase in atmospherically deposited contamination resulting from increasing industrial emissions (Fernandez *et al.,* 2000; Rose *et al.,* 2009). Podragu Mare Lake and Caltun Lake (from the South); Bila Lake and Pietrosul Lake (from the North) were surface enriched by Cu. The surface enrichment by Cu demonstrated by Podragu Mare Lake, Caltun Lake and Bila Lake were not as conspicuous as the surface enrichment by Pb and Zn; such differences in the proportion of surface peaks have been documented in Lacul Negru in the Retezat National Park of Romania (Rose *et al.*, 2009). It is not unlikely that some common sources (presumably far away) are responsible for the deposition of trace elements at the South and North and probably with the Retezat National Park of Romania as well (Rose *et al.,* 2009). In Europe some studies have focused on the sediments of mountain lakes (e.g. AL: PE; AL: PE2; MOLAR; EMERGE). The Mountain Lake Research (MOLAR, 1999) project considered the most remote and least disturbed freshwater ecosystems in Europe, mainly located in the Alpine and Arctic regions. The project established that the lakes from these regions are contaminated by long distant travelled atmospheric pollutants (MOLAR, 1999).

6.5 Down-core variations of sediment lead (Pb) and zinc (Zn) profiles in the studied areas

The other metals (except Pb and Zn) represented in the lake sediments in this research are present only in minute concentrations. Lead is a stable metal in lake sediment (Renberg *et al*., 2002). Researchers such as Veron *et al.,* (1999); Zietz *et al.,* (2003); Henry *et al.,* (2004); Lakind *et al.,* (2004); Pereira *et al.,* (2004); Mielke *et al.,* (2005) and Miralles *et al*., (2006) have identified lead (Pb) as a common tracer of anthropogenic contamination widely investigated in numerous studies dealing with environmental quality and health care in Western Europe. This part of the thesis considers the Pb and Zn variations in the lake sediment.

Similar trends in metal concentration were repeated across the lakes in the Fagaras region. Pb and Zn profiles showed noticeable surface increases in all the lakes. In Fagaras region the patterns of Pb and Zn concentrations in each lake were quite similar with corresponding peaks and troughs. The trends in metal concentrations (surface enrichment) demonstrated by the lakes from the south and north Carpathians of Romania are consistent with other parts of Europe. For example, a sediment core from Lake Trenntsee in northeastern Germany was dated radiometrically to gain a chronology of the observed depth profiles of heavy metal concentration. The upper part of the sediment core demonstrated elevated Pb and Zn concentrations. These were attributed to elevated anthropogenic emissions reaching the environment after World War II (Suckow *et al.,* 2001). Similar trends in metal concentration were repeated across most of the lakes in the Rodna region. Pb and Zn consistently showed surface increase in most of the lakes. The patterns of Pb and Zn concentrations in each lake were quite similar with corresponding peaks and troughs.

Higher metal concentration levels were recorded in Fagaras than Rodna for example, Balea Lake had Pb and Zn surface concentrations of 279 mg kg^{-1} and 401 mg kg^{-1} respectively while the highest level of Pb was recorded in Bila Lake with the value of 203 mg kg^{-1} (Figures 6.6 and 6.7). The highest level of Zn in Rodna was recorded in Buhaiescu Lake with the value of 149 mg kg^{-1}. Fagaras lakes obviously have higher values in Pb and Zn concentrations than Rodna lakes which therefore have relatively low concentration of these elements. Metals are naturally occurring in the environment, and therefore the local geology can affect metal concentrations in soil and sediments. Research shows that differences in geology complicate comparisons of metal levels between different areas. A common method to interpret results from metal analyses is therefore to compare levels in pre-industrial sediment (WHO, 2007).

A range of researchers have studied the pollution history of aquatic ecosystem by core sediments (e.g. Karbassi *et al.,* 2005; Mohamed, 2005; Rose *et al.,* 2009). Many researchers have used sediment cores to study the behaviour of metals (e.g. Bellucci, *et al.,* 2003). The sediment history broadly reflects the contamination history of an area. As a result of urbanization and industrial development environmental pollution has become a major concern (Alemdaroglu *et al*., 2003). Trace metals have been identified as a group of pollutants that should be monitored in order to obtain a logical and up to date overview of quality status for surface water (Pertsemli and Voutsa, 2007). Sediments have been discovered to play an active role as a sink and possible source of trace metals (Salomons, 1995; Hochella *et al.,* 1999; Audry *et al.,* 2004). Metals are regarded as a group of pollutants of high ecological relevance that are not removed from water by a natural process of purification e.g. Hu *et al*., (2002) and Smol, (2008). Studies of the sediments of Lake Ontario have shown that heavy metals such as copper (Cu) and lead (Pb) are associated with fine grain particulate matter and these particles are transported by currents and settle to the bottom sediments of the lake; sediment cores taken from these areas show enrichment of heavy metals in the top 10 cm (Gem/Water, 2002).

The radiometric dating of a single core of Capra lake core 3 (LCp 3) suggested that sediment accumulation rates were relatively uniform up to the depth of about 10 cm but deeper than 10 cm there are variations in sediment accumulations (Figure 5.42b). Such observations have been consistent with the situation in the Lacul Negru in the Retezat National Park of Romania (Rose *et al.,* 2009). Core chronologies and sediment accumulation rates calculated from ^{210}Pb activities suggested that sediment accumulation rates were relatively uniform in the last about hundred years with an average at 0.029 g cm^{-2} yr^{-1}, earlier than that, sediment accumulations varied in a significant high level (Figure 5.43), which may suggest sediment slumping during the time. The raw CRS model places fallout maximum of the atmospheric testing of nuclear weapons around 1963 at 5.3 cm and the 1986 Chernobyl accident at 3 cm.

Sediment cores from seven European mountain lakes collected as part of a study of palaeolimnogical records of climate change (the MOLAR project) were dated radiometrically by ^{210}Pb. Although the lakes were remotely located yet, the ^{210}Pb record of three of the lakes demonstrated significant increase in sedimentation since ca 1950 (Appleby, 2000). Dating of sediment cores using ^{210}Pb and ^{137}Cs can reveal a large range of sediment accumulation rates (Curtis *et al*., 2010).

The nonferrous metal industry is one of the largest anthropogenic sources for atmospheric Zn. Generally, the European emissions of this metal have decreased during recent years

(Dietz *et al*., 1998). Thus the fluxes of Zn in Romanian lakes reflect the discharge pattern for these metals quite well. Atmospheric lead has the potential for long-range transportation resulting in transport at a regional level (e.g. WHO, 2007). Lead (Pb) is being widely investigated in studies connected with environmental quality and health care because it has been identified as a common tracer of anthropogenic contamination (Miralles *et al*., 2006 and WHO, 2007). The source of anthropogenic lead emissions is traceable to power generation, ore smelting, automotive emissions, and natural sources such as volcanoes, hydrothermal vents (Miralles *et al*., 2006). Renberg *et al*. (2000, 2002) and Winderlund *et al*. (2002) investigated Pb deposition in various lake sediments in Sweden and hence concluded that lake sediments are custodial of anthropogenic lead.

In some studies from Arctic areas an increasing Pb concentrations was measured after the introduction of leaded gasoline on the northern hemisphere (WHO, 2007), with a concurrent reduction after the phasing out of leaded gasoline (AMAP, 2004). In other studies no clear temporal trends for Pb have been identified (Outridge *et al*., 2002; Rognerud *et al*., 1993). The natural background levels of Pb can be high in some lake sediments (Skotvold and Savinov 2003), and the natural levels may be masking any changes superimposed by anthropogenic inputs. Another explanation for the low Pb input may be that such lake might have mainly received atmospheric inputs from areas with low Pb emissions. It has been documented that high emissions of Pb have occurred in Western-Europe, (Dietz *et al*., 2004) but the results for the other metals (Zn, Cu, Co, Ni and Cr) suggested that Romanian upland lakes have not been largely polluted. However, no clear increases in concentrations from pre-industrial times have been identified for these metals in other studies (Rognerud and Fjeld, 2001). It is therefore likely that the anthropogenic impact is so small that it is overwhelmed by the natural background levels. The results suggest a continuing input of Pb to Fagaras lake sediments such as Balea Lake and Podragu Mare Lake.

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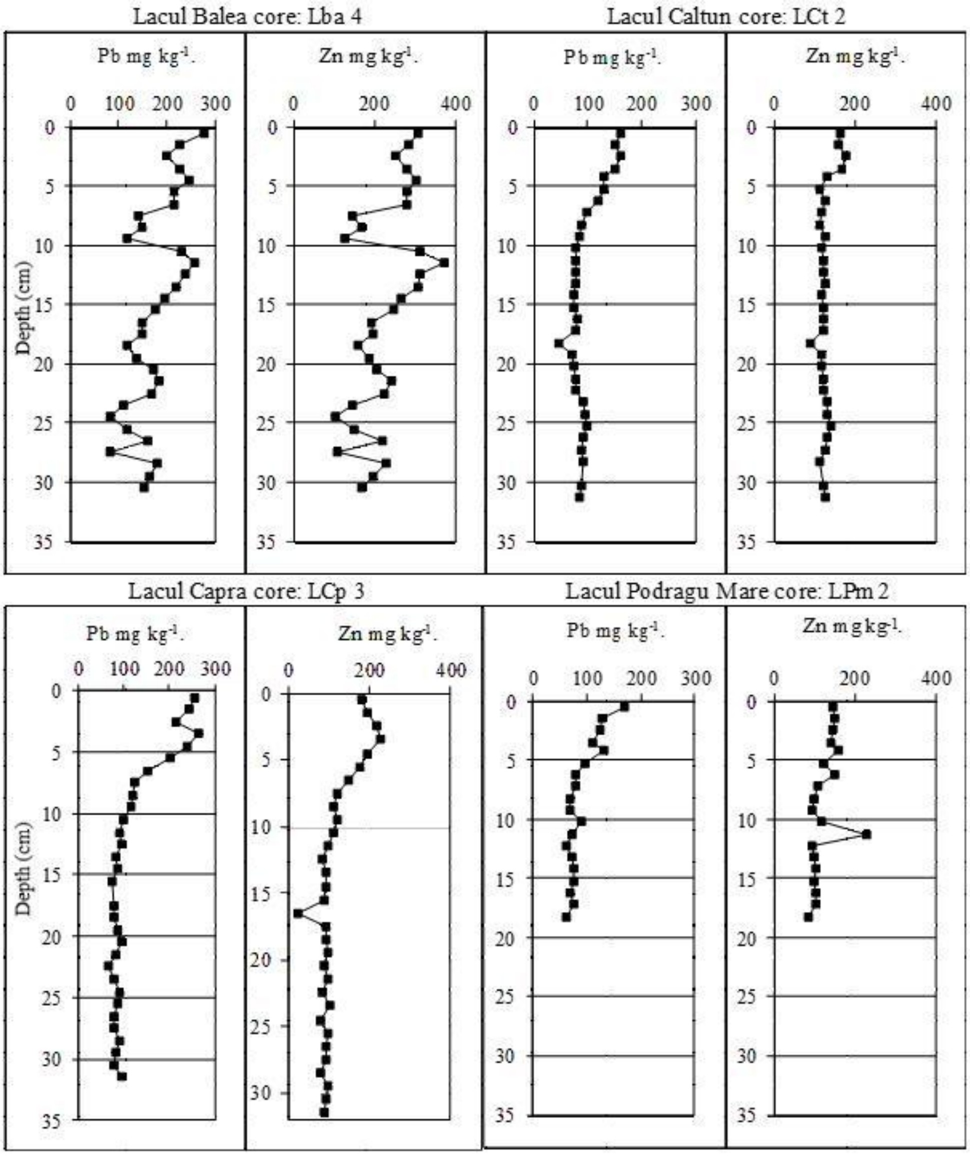

Figure 6.6: Down core variations of sediment lead (Pb) and zinc (Zn) profiles of Fagaras main cores

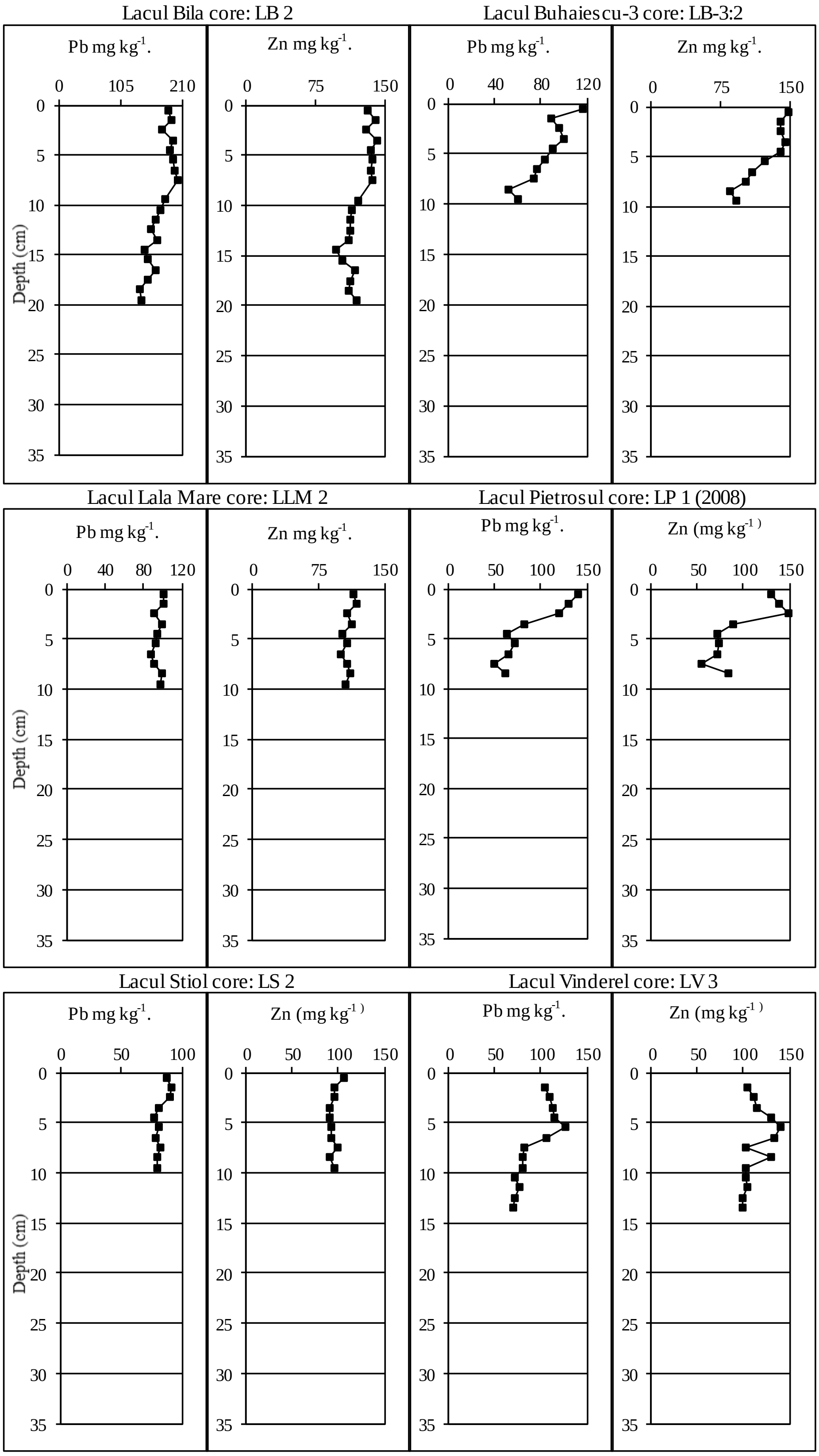

Figure 6.7: Down core variations of sediment lead (Pb) and zinc (Zn) profiles of Rodna main cores

6.6 Integration of lakes sediment characteristics to provide an overview

In this section, the physical characteristics, the mineral magnetic characteristics and geochemical characteristics were brought together for all sites in both regions and analysed using PAST Principal Componet Analysis recommended for such analysis by Hammer (2001). As a statistical technique principal components analysis (PCA) finds hypothetical (supposed) variables or components which form the basis for as much of the variance in multidimensional data as possible (e.g. Davis 1986, Harper 1999). The PAST principal components analysis (PCA) of the south and north of Romania sampled lakes showed that most of the variance is accounted for by the first two components. Component 1 accounted for about 47% while component 2 accounted for about 17% (Table 6.3). Pooling together components 1 and 2 gives 64% which is about two third of the variance.

Table 6.3: Eigenvalue and percentage variance follows 21 variables for all sites

PC	Eigenvalue	% variance
1	9.82	46.78
2	3.58	17.04
3	**2.17**	**10.33**
4	1.29	6.16
5	0.91	4.35
6	0.74	3.54
7	0.61	2.90
8	0.54	2.57
9	0.30	1.44
10	0.25	1.21
11	0.20	0.93
12	0.16	0.78
13	0.14	0.68
14	0.10	0.46
15	0.06	0.29
16	0.05	0.22
17	0.02	0.12
18	0.02	0.08
19	0.01	0.05
20	0.01	0.03
21	0.00	0.02

Plotting the two key components (Figure 6.8) illustrates the potential of the study's control dataset. It distinguishes Balea Lake, Capra Lake, and Podragu Mare Lake from Caltun Lake and every other lake from the north. It does not clearly distinguish the south from the north.

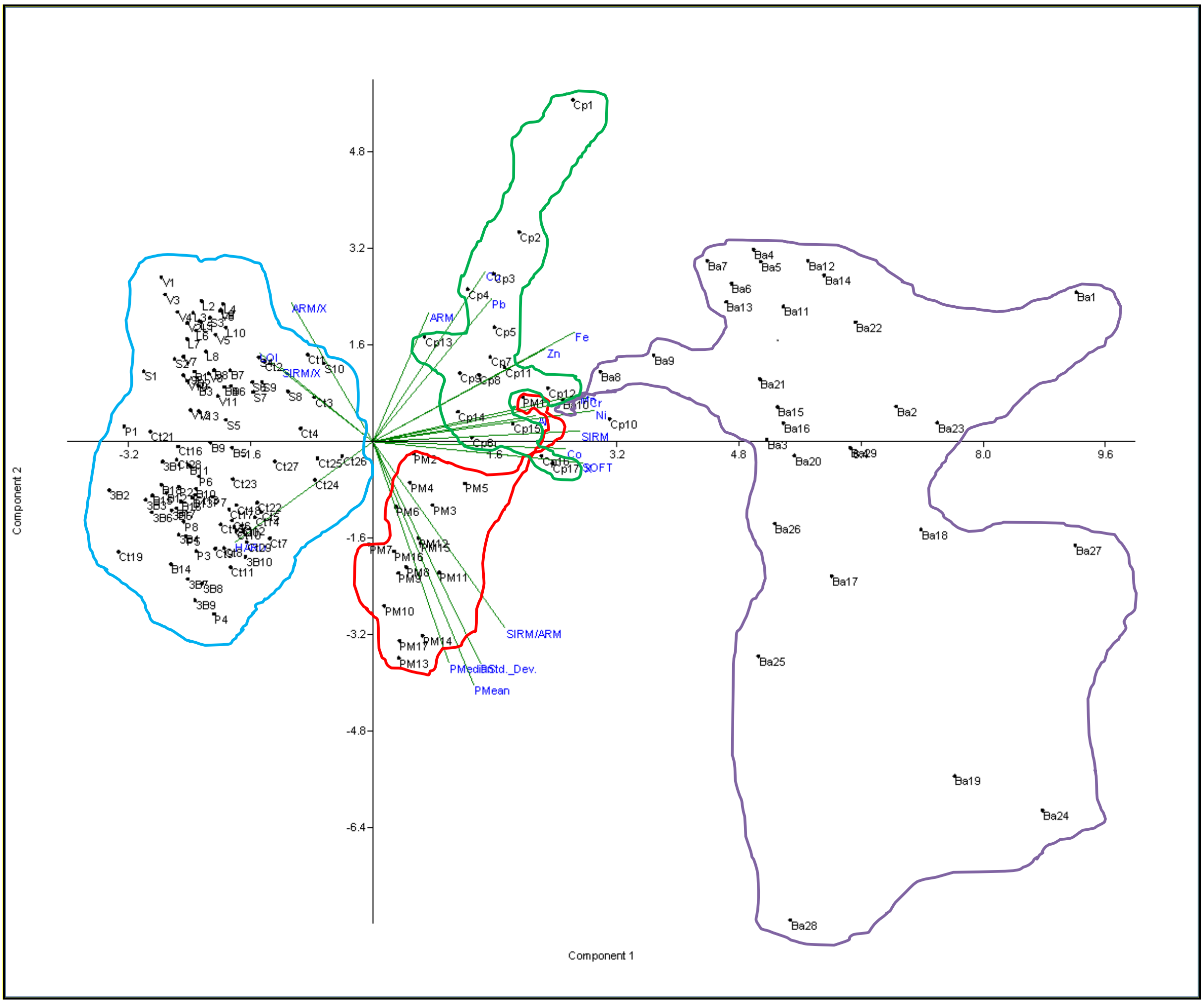

Figure 6.8: South and North principal component analysis biplot for components 1 and 2. Key: Purple (Balea Lake), Green (Capra Lake), Red (Podragu Mare Lake) – all from the south and Blue: Caltun Lake from the south; Bila, Buhaiescu 3, Lala Mare, Pietrosul, Stiol and Vinderel Lakes from the north.

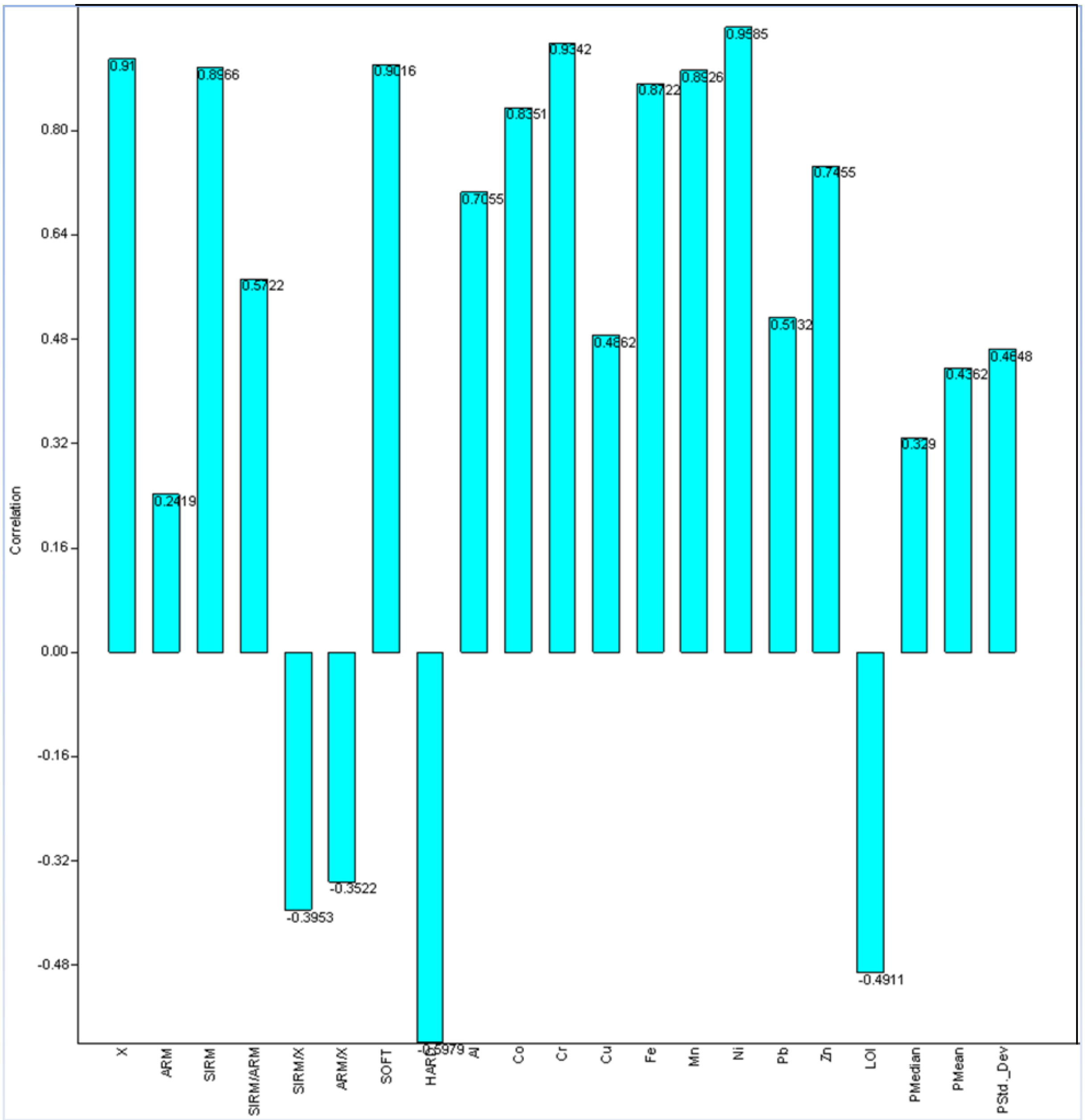

Figure 6.9: Illustration of the most important variables along component 1

Figure 6.9 illustrates the most important variables of the first component in the PCA. While the mineral magnetic parameters ARM/X, RISM/X and Hard, and sediment LOI are useful discriminators, it is apparent that lead levels are not. This implies that atmospheric input of contaminants does not adequately differentiate between the study sites in the south and those in the north of the Romanian Carpathians.

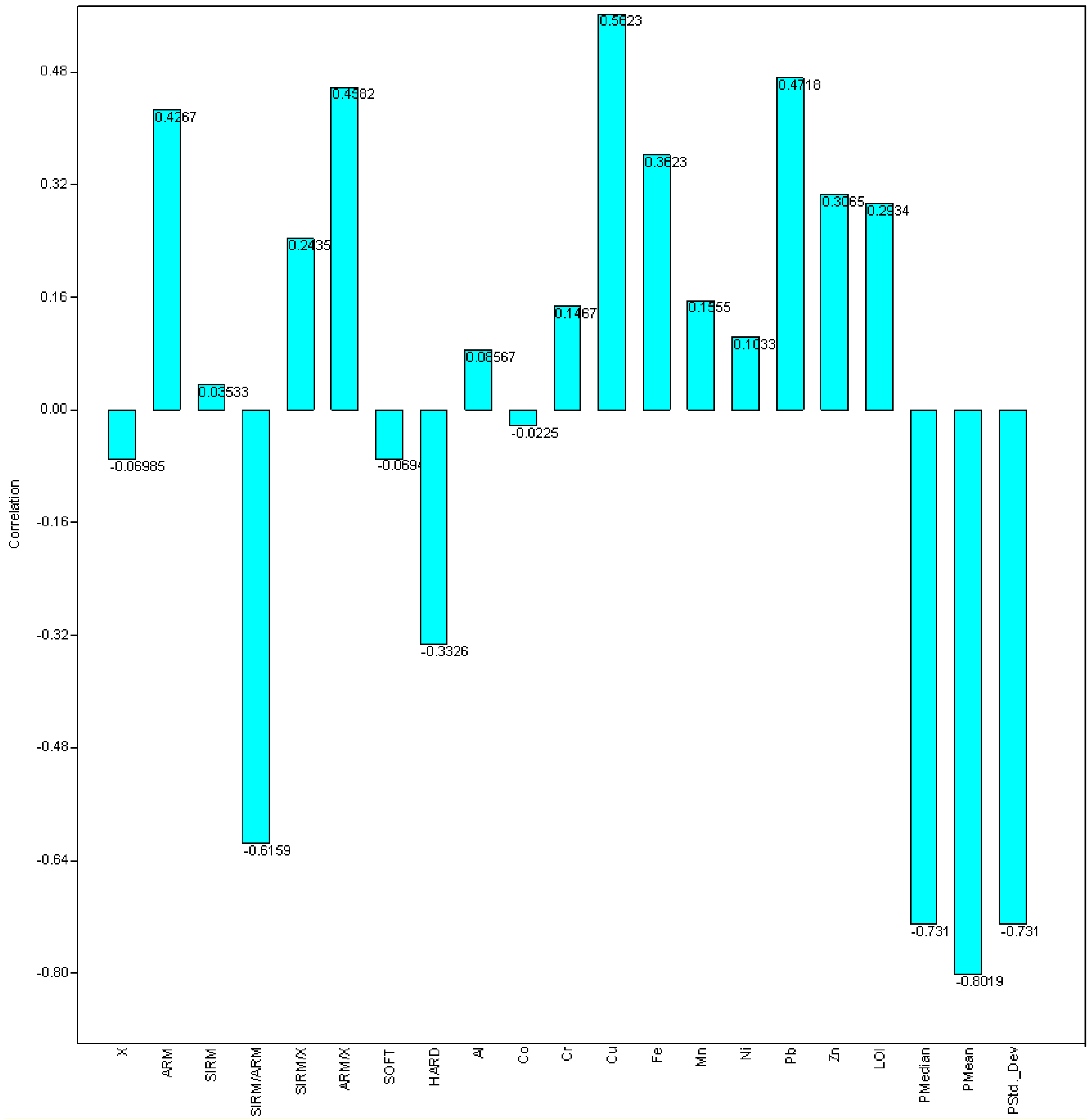

Figure 6.10: Illustration of the most important variables along component 2

6.7 Summary

The discussion chapter has considered the various investigations in this research and has addressed the findings related to the specific objectives of the study. The discussion focused on the different research objectives which consisted of the determination of the physical characteristics of the lake sediments, the assessment of the potential of using a mineral magnetic approach as a retrospective tool for the assessment of human impacts on these lakes including atmospheric deposition of pollutants and the determination of the geochemistry of the sediments in order to assess the spatial and temporal variations in trace metal deposition. The later part of this chapter integrated the above parameters and demonstrated the suitability of the study sites for sediment based study and environmental reconstruction. The sites do not allow a site by site statistical comparison as they have different characteristics. The depths are not the same. Nevertheless, it is possible to look at some lakes across the regions that show correlations in their physical, magnetic and chemical characteristics individually. It is clear that all the north sites cannot be directly compared with all the south sites. Some can be compared visually at least in terms of surface increase or decrease in the profiles. Therefore this research has been an addition to the records of recent pollution in Romania and a gateway to further investigations in the area of palaeoenvironmental change.

CHAPTER 7: Key research findings, conclusions and future work

7.1 Key research findings

The key research findings present the summarised version of the outcome of this research. It considers the lake catchment and sediment physical characteristics, the mineral magnetic characteristics and the geochemical characteristics of the lake sediments. Two regions in the Romania Carpathians were selected for a lake sediment based study of key lakes. All four lakes from Fagaras region were located in classic glacial cirques. There is no marked difference in the altitudinal positions of the lakes as all were located above 2000 m. However, the lowest lake elevation is at 2035 m (Balea) while the highest lake elevation is 2249 m (Capra). The six lakes from Rodna region are also all located within a cirque except Vinderel, which is located in a glacial col. The altitudinal positions of these lakes ranged from 1667 - 1840 m. The lowest is Stiol Lake (1671 m) while Bila Lake is the highest (1840 m). Thus, it can be seen that even the lowest lake elevation in Fagaras region (2035 m) is higher than the highest lake elevation in Rodna region (1840 m).

There are distinct size variations in the catchment areas of the lakes in both the Fagaras and the Rodna areas. Podragu Mare lake (in the Fagaras region) has the largest catchment area of 55.2 ha followed by Balea lake (45.5 ha). Stiol lake (from Rodna) has the largest catchment area of 156 ha followed by Buhaiescu-3 lake (62.9 ha). Most of the lakes sampled in Rodna Mountains have larger catchment areas than their counterparts from the Fagaras Mountains (Tables 3.2 and 3.3) (Stiol Lake has the highest catchment area of 156 ha). The lakes in the Rodna Mountains have higher catchment: lake ratios than lakes in the Fagaras Mountains. The smallest of such ratios in the Rodna Mountains is 23.0 (Lala Mare Lake) which is almost equal to the largest ratio 23.3 (Caltun Lake) in the Fagaras Mountains. Buhaiescu-3 lake (Rodna region) has the largest catchment: lake ratio of 698.9.

The lakes from Fagaras region were generally deeper than the Rodna lakes with depth ranges from 8.6 - 16.5 m. Recovered sediment lengths were also greater with length ranges from 0.20 - 0.32 m (see Table 3.4). All the lakes from Rodna region were relatively shallow, ranging from 0.5 - 5.5 m. Recovered sediment lengths were also shallow (0.12 - 0.32 m). The variation in length of sediment cores might be partly due to the amount of material accumulated at the bottom of the lakes or due to the nature of the lake bottom substratum.

The land cover of all four catchments from Fagaras region is predominantly grass except for Caltun where scree dominates (Table 3.2); while the catchment surface geology is predominantly moraine. In comparison to the Fagaras region, the Rodna region's land cover is predominantly grass and scree around the lakes (Tables 3.3a and 3.3b) while the geology formation of the catchment areas is predominantly mica chist and moraine deposits, except at Stiol Lake where gneiss dominates. Another exception to the geological formations above is Vinderel Lake catchment which includes of sandstone and siltstone with diabase intrusions.

All the lake sediment cores sampled demonstrated a decrease in sediment density towards the core surface. In the Fagaras region the minimum dry density value was observed in Capra Lake while the maximum was observed in Balea Lake, LBa 4 (Table 5.1). In the Rodna region the minimum density value was observed in Vinderel Lake while the maximum was observed in Buhaiescu-3 Lake, LB: 3-2 (Table 5.2). There are more fluctuations in the density down core profiles of most lakes in the Fagaras region than the Rodna region; for example Balea Lake in the Fagaras region. The highest dry density value recorded in the Rodna region was lower than the highest value recorded in the Fagaras region. The low density zones of each lake are also characterised by a relatively high organic content. All the lakes in both regions tend to show an increase in loss-on-ignition towards the surface. The percentage loss-on-ignition in the Rodna region lake sediments is higher than the Fagaras region (Tables 5.3 and 5.4). All the four lakes from the Fagaras region demonstrated a larger particle size range than the sample in the Rodna region (see Tables 5.5 and 5.6).

Although the core LBa 1 (from Balea Lake) did not demonstrate surface increase in the magnetic concentration parameters χ, ARM and SIRM, its peaks and troughs are comparable to LBa 4 (from Balea Lake). Therefore, the key findings of the magnetic measurements are that the magnetic concentration parameters χ, ARM and SIRM are similar between cores from the same lake and are comparable among the lakes in the Fagaras region except for Caltun Lake which had relatively low values for all magnetic concentrations. The magnetic concentration parameters χ, ARM and SIRM are similar between cores of the same lake and are similar among most of the lakes in the Rodna region as well. All cores show clear surface peak in the magnetic concentration parameters χ, ARM and SIRM, except Stiol Lake which demonstrated subsurface peak and Lala Mare Lake which did not show any clear fluctuations in the magnetic concentration parameters χ, ARM and SIRM. In all the sites that demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of their core profiles; at this peak there is also a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters 'soft' and 'hard' (Figures 5.13-5.26). These features indicated both an increase in magnetic concentration, and a change in grain size and mineralogy; they demonstrated the possible influence of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions. There are obvious variations in the magnitude of the magnetic parameters in both regions.

Many studies have explored the relationship between mineral magnetic measurements and physicochemical properties of sediments (e.g. Clifton *et al*., 1999; Chan *et al*., 1998). Based on the previous research, mineral magnetic measurements have been identified as a suitable tool for determining sediment provenance (Oldfield and Yu, 1994; Booth *et al*., 2005). Mineral magnetic characteristics serve to determine the sediment transport pathways (Lepland and Stevens, 1996). Mineral magnetic measurements serve as a proxy for geochemical, radioactivity, organic matter content and particle size data (Hutchinson and Prandle, 1994; Clifton *et al*., 1999; Zhang *et al*., 2001). Research has identified that anhysteretic remanent magnetisation (ARM) measurements can be used to reflect the concentration of fine-grained magnetite in the clay fraction and susceptibility of ARM (χARM) was strongly associated with clay and fine silts, and saturated isothermal remanent magnetisation (SIRM) was strongly associated with very fine to medium silts (e.g. Oldfield and Yu, 1994; Clifton *et al*., 1999). It has been established that high magnetic concentration measurements can be associated with large amounts of fine-grained sediments and an inverse relationship with coarse grained sediments (Booth *et al*., 2005). Research shows that changes in environmental factors that control the productivity of magnetic bacteria in the lake can contribute to the variability of magnetic mineral concentrations that can be observed in the lake sediments (e.g. Kim *et al*., 2005). Magnetotactic bacteria could possibly account for the high concentration of magnetic grains in the dark, organic rich layers of lake sediment (e.g. Kodama *et al.*, 1998 in Kim *et al*., 2005). In most of the lakes studied for this research (Figures 7.1 and 7.2) the zones of large particle size are met with low magnetic signals and vice versa (e.g. Caltun Lake from Fagaras region and Pietrosul Lake from Rodna/Maramures region).

The enrichment factors (EFs) of metals from each lake was estimated by dividing the surface (top) value for each metal by the bottom (background) value. Although there are differences in the magnitudes, all the lake sediments in the South are enriched with Pb and Zn. Only Podragu Mare Lake and Caltun Lake were sparingly enriched with Cu. The other metals present in the lakes were represented in low magnitudes (Figure 5.7 and Table 5.1). Only Podragu Mare Lake was enriched in Co, Cr and Ni. Unlike in the South where all lakes have been enriched by Pb and Zn only three lakes in the North (Buhaiescu Mare, Pietrosul and Vinderel) show enrichment in Pb (Figure 6.5 and Table 6.4). Only two lakes (Buhaiescu Mare and Pietrosul) show enrichment in Zn in the North. Bila and Pietrosul demonstrate enrichment in Cu. Pietrosul Lake seems the most contaminated of all the lakes from the North. The patterns of Pb and Zn down core concentrations in each lake were quite similar with corresponding peaks and troughs. Higher metal concentrations were recorded in Fagaras than Rodna.

There are no signs of direct catchment supply of the trace metals into the lakes both in the South and the North. Therefore, the trace metals are most likely to be industrially derived particulate deposition onto the Romania Carpathian lakes. Such deposit can only be made possible through the atmosphere before subsequent incorporation into the lake sediments. The radiometric dating of Capra Lake core 3 (LCp3) showed evidence of atmospheric inputs resulting from Pb and Zn inputs from 1960 upwards. Record showed the input was as a result of industrial emissions (Turnock, 2001). The industrial activities in Romania started rising from about 1960s and reached to a peak in about 1990s and then declined. Therefore, between 1960s and 1990s there was increase in iron production, energy production, coal mining, petrochemical industry, oil production and gas production with attendant atmospheric pollution. The increase in the Pb levels in all the lakes in the South show that the lake sediments can be interpreted as records of atmospheric deposition. Whereas in the north, the Pb levels of the lakes were not as high as in the south.

Trace metals vary significantly (e.g. in terms of magnitude) in the lake sediment of south and north of Romania; especially in the case of Pb and Zn (see Figures 7.1 and 7.2). This variation could be due to particle size effects or anthropogenic influences. It has been discovered that fine sediments with abundant clay minerals such as iron oxides or manganese oxides as well as organic matter often show higher metal concentrations than the coarse sediments (Rae, 1997). It has been determined also that higher concentrations of ferrimagnetic minerals are found in the south lake sediments and lower concentration in the north lake sediments of Romania. The higher χ values in the southern Carpathians than in the northern region demonstrated the variations in the strength of super paramagnetic grains in the samples from either region.

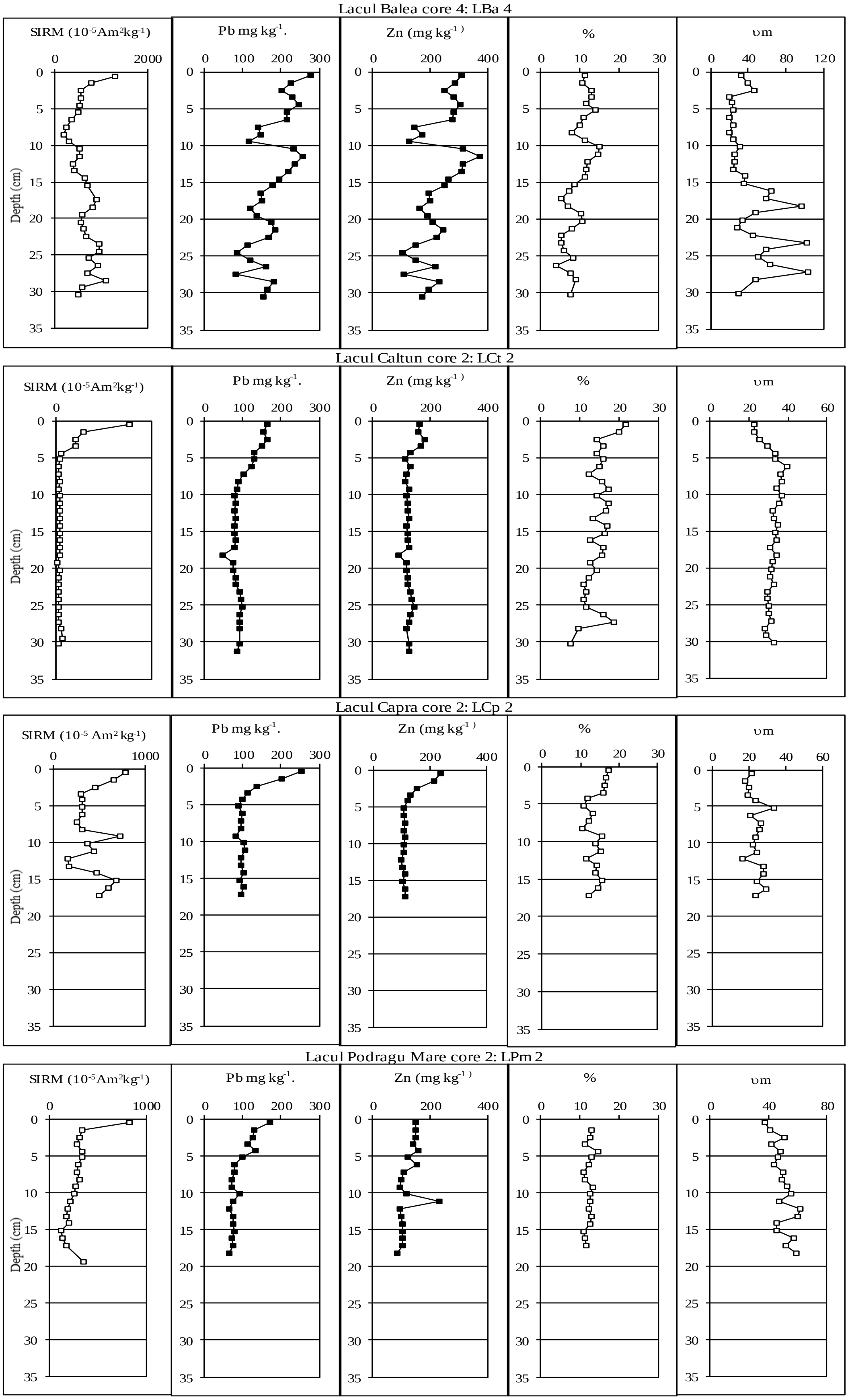

Figure 7.1: Down core variations of sediment SIRM, lead (Pb), zinc (Zn), LOI and mean particle size profiles of Fagaras cores

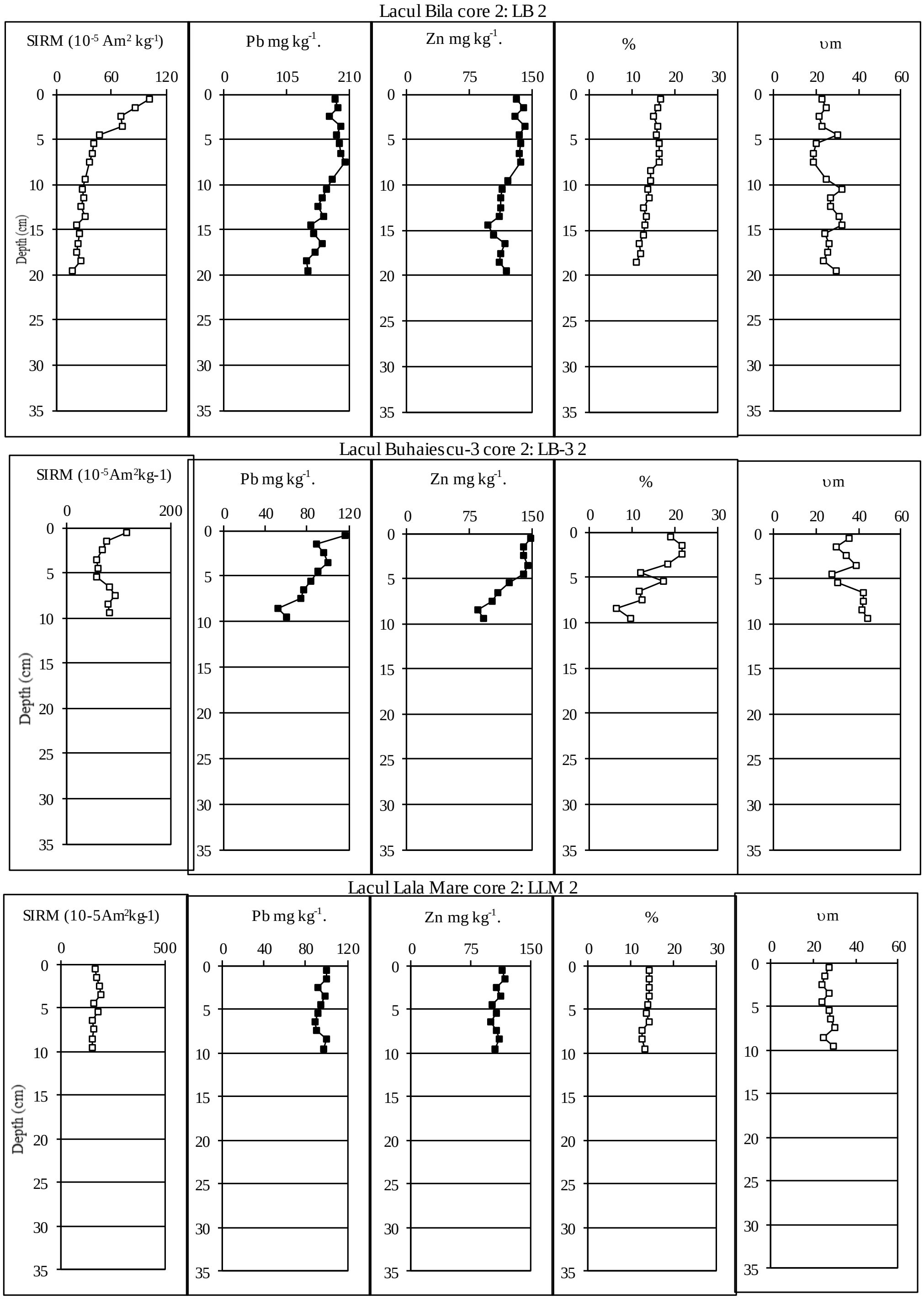

Figure 7.2a: Down core variations of sediment SIRM, lead (Pb), zinc (Zn), LOI and mean particle size profiles of Rodna cores

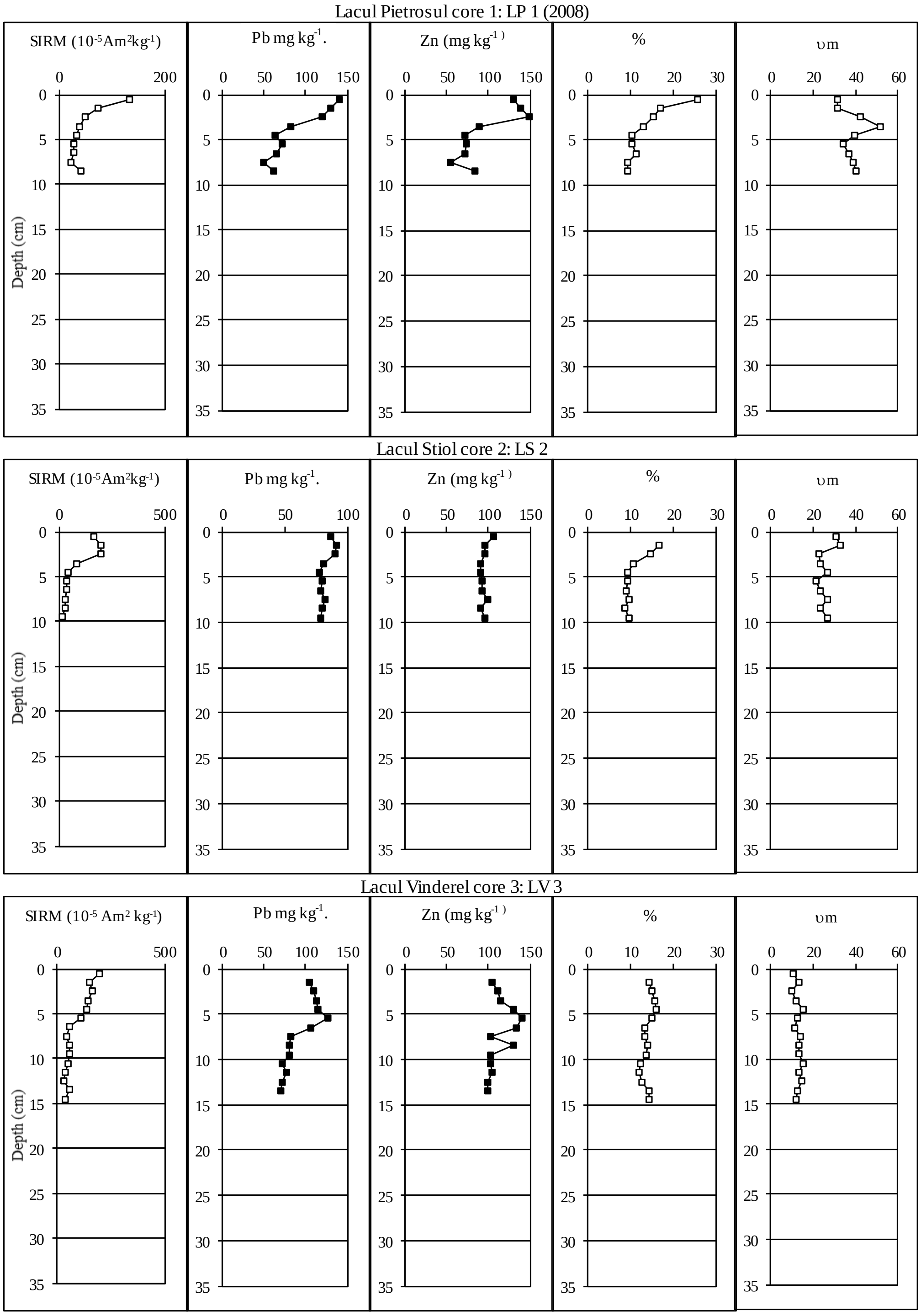

Figure 7.2b: Down core variations of sediment SIRM, lead (Pb), zinc (Zn), LOI and mean particle size profiles of Rodna cores

7.2 Conclusions

From the literature review, it is evident that mountain lake sediments can accumulate atmospheric pollutants (that is they can act as its archive of past conditions in the environment). It has been determined that pollution of lake systems has been a focus of attention to the environment and it has involved the local, regional and global efforts in an attempt to moderate it below the hazard level. A careful review of lake sediments sampling and analytical techniques has shown that the lake sediments sampling and analytical procedures for this research can be concluded to be appropriate. A brief discussion of the study areas (the Romanian Carpathian Mountains) showed that the area is environmentally significant because of its aesthetic value and tourism potential. It can be concluded that the research can contribute to environmental information on impacts of people on their environment.

The aims of this research project are: To investigate the physical characteristics, the mineral magnetic properties and trace metal levels of sediment cores from selected lakes in the southern and northern Carpathians of Romania in order to thereby evaluate the possibility of using these lakes' sediment as records of recent human impacts and in particular trace metal deposition. There are distinct size variations in the catchment areas of the lakes from both the Fagaras region and the Rodna/Maramures region. There are also disparities in lake areas as there are differences in the lake depths and catchment: lake ratio. The highest dry density value recorded in the Rodna region was lower than the highest value recorded in the Fagaras region. The low density zones of each lake are also characterised by a relatively high organic content. The percentage loss-on-ignition in the Rodna region lake sediments is higher than the Fagaras region; all lakes in both regions tend to show an increase in loss-on-ignition towards the surface. All the four lakes from the Fagaras region demonstrated a larger particle size range than the sample in the Rodna region. The mean value for the region varied from 24 - 50 μm and the particle size values ranged from 17 - 104 μm (see Table 5.5). The mean value for the Rodna region varied from 13 - 39 μm and the particle size values ranged from 9 - 52 μm (see Table 5.6).

Most lakes investigated in this research demonstrated an increase in the magnetic concentration parameters χ, ARM and SIRM in the upper most part of their core profiles with a corresponding decrease in the SIRM/ARM ratio and a shift in the parameters 'soft' and 'hard' (Figures 5.13-5.26). The combination of these parameters indicates the predominance of relatively coarse grain ferromagnetic material at the upper surface of the lake. These features indicate both an increase in magnetic concentration, and a change in grain size and mineralogy; they demonstrate the influence (probably) of the atmospheric deposition of particulate pollution associated with fossil fuel combustion and vehicle emissions. It is therefore evident from the above that there was apparent demonstration of influence of atmospheric particulate pollution on the sediments of the lakes both from the Southern region and the Northern region of the Romanian Carpathian Mountains. There are obvious variations in the magnitude of the magnetic parameters in both regions. For example, the highest value of ARM is 301.01 x $10^{-5}Am^2kg^{-1}$ while that of SIRM is 1285.11 x $10^{-5}Am^2kg^{-1}$ (Fagaras Capra Lake Figure 5.16 and Fagaras Balea Lake Figure 5.14). From Rodna Pietrosul Lake (Figure 5.22), the highest value of ARM is 21.17 x 10^{-5} Am^2 kg^{-1} while that of SIRM is 229.56 x 10^{-5} Am^2 kg^{-1} but the fundamental principle is same. Therefore this research justified the mineral magnetic approach as a potential tool for a retrospective assessment of human impacts on the studied lakes including atmospheric deposition of pollutants. However, the research by Kim *et al*., (2005) shows that the changes in environmental factors that control the productivity of magnetic bacteria in the lake can contribute to the variability of magnetic mineral concentrations that can be observed in the lake sediments.

This research project has shown that the same trends in trace metal concentration are repeated across the lakes in the South and in the North; that is relatively clean sediment towards the bottom of the cores and metal enrichment towards the surface of the cores. Pb and Zn consistently show obvious surface increase in all the lakes. The patterns of Pb and Zn concentrations in each lake are quite comparable. Core chronologies and sediment accumulation rates calculated from ^{210}Pb activities (Capra Lake, core LCp 3) shows that sediment accumulation rates were relatively uniform in the last about hundred years or so with an average at 0.029 g cm^{-2} yr^{-1}, earlier than that, sediment accumulations varied in a significant high level, which may suggest sediment slumping during the time.

Despite the comparability mentioned above, diffrences were noticed in the concentrations of the trace metals stored in the lake sediments. Higher trace metal concentrations were recorded in Fagaras than Rodna for example, Balea Lake had Pb and Zn peak concentrations of 279 mg kg^{-1} and 401 mg kg^{-1} respectively while the highest level of Pb was recorded in Bila Lake with the value of 203 mg kg^{-1} (Tables 5.7 and 5.8). The highest level of Zn in Rodna was recorded in Buhaiescu Lake with the value of 149 mg kg^{-1}. The Pb concentrations in Fagaras region range from 31 - 279 mg kg^{-1} while the Zn concentration range from 26 - 401 mg kg^{-1}. In Rodna region the Pb concentrations range from 45 - 203 mg kg^{-1} while the Zn concentrations range from 26 - 149 mg kg^{-1}. It can therefore be seen that the Fagaras lakes displayed higher peak in Pb and Zn concentrations than Rodna lakes which therefore have relatively low concentration of these trace metals. However, in consideration of the above, whereas it can be concluded that there are spatial and temporal variations in trace metal deposition between the lake sediments in the regions, statistical PCA does not wholely distinquish sites in the south from those in the north (Figure 6.8).

One of the rationales for this study is that although there are many lakes in the Carpathians, relatively little recent palaeolimnological research has been undertaken. Therefore this research is an addition to the records of recent pollution in Romania and a gateway to further investigations in the area of palaeoenvironmental change. The mountain lakes of the Romanian Carpathians have the potential to provide a sedimentary record of recent environmental change. Therefore, the research has demonstrated the influence of atmospheric particulate pollution on the sediments of the lakes both from the southern region and the northern region of the Romanian Carpathian Mountains. The study sites are suitable for sediment based study and environmental reconstruction. The lake sediments from the sites can be used as records of temporal and spatial variation in trace metal pollution.

Another rationale for this study is to extend the spatial range of the study of mountain lake sediments. Whereas, the Carpathian Mountains have been identified as susceptible to atmospheric pollution, to date there have been very few publications from this region with the only exception being Rose *et al.* (2009) at a site in the Retezat Mountains at the western extremity of the Carpathian Range in Romania. In Central Eastern Europe air and water pollution are considered the most serious environmental problems due to their impact on human health as well as the physical environment (Turnock, 2001). Thus, a further rationale for this study is that an enhanced palaeolimnological insight into the recent lake sediments of the region will potentially inform environmental monitoring and decision making in one of the European Union's newest member states. Consequently, the findings of this research project have an applied dimension in adding to recent records of environmental pollution affecting current (relatively sparse) proxy records both temporally and spatially.

7.3 Future work

The present study took sediment samples from ten lakes. Accessibility of the lakes was a major factor in site selection due to the fact that the sampling gear and other required equipment had to be carried. Except for Balea Lake the lakes were all located away from any road access. In future more sites (more lakes) from the two regions could be sampled and other regions might also be investigated. However, within the two areas optimal lakes were selected within a spatial range that was accessible in the time period available for the field work at each site. Furthermore, the two regions were carefully selected to provide geographically contrasting sites where little or no previous palaeolimnolgical work had been undertaken. Access to these mountain regions in the Carpathians can also be restricted e.g. Retezat Mountains. This research considered the recent changes that occur in the lake sediments and hence the relatively short cores taken were sufficient for that purpose. Recent lake sediment research by Rose *et al.* (2009) on Lacul Negru located in the Carpathian Mountains of Romania (within the Retezat National Park) indicates that it may have been impacted by heavy metal deposition since 16^{th} century. Therefore, longer sediment cores can be taken from the lakes in order to extend the study of the palaeolimnological changes in the sediments to longer time perspective. However, such sampling would involve more personnel to carry the sampling equipment required to do such sampling.

It can be deduced from the earlier discussion in this research work that the interpretation of sediment trace metal levels might be improved on through more detailed assessment of trace metal inputs pathways onto the lake sediments. The catchment trace metal input and the atmospheric deposition of metals onto the lake sediments could be more intensively investigated by fuller examination of the catchment and eventually fuller determination of the catchment metal inputs onto the lake sediments. It is recommended that the soil in the lakes catchment be sampled and subject to the same laboratory analysis as the lake sediment for fuller interpretations of the data.

More analysis could be undertaken on the organic contaminants of the sediments. The profiles from Spheroidal Carbonaceous Particles (SCPs) analysis of lake sediments can help further understanding of the magnetic concentrations of the lake sediments (Kodama *et al*., 1997). SCP records can show strong correlations with fossil fuel consumption even in remote lakes (Doubleday *et al*., 1995; Cohen, 2003). Also, the records of Polycyclic Aromatic Hydrocarbon (PAH) can be compared between lakes to understand the timing and the level of increases in fossil fuel emissions (Cohen, 2003).

Hu *et al*., (2002) states that it is necessary to derive a complete understanding of the potential origins of the magnetic signal observed in lake sediment. Therefore, laboratory analysis might be considered on the microbiological activities in the lake and the lake sediments to understand the contribution of the microbes to the magnetic sensitivity of the lake sediments. The dating in this research was limited to one core because of the cost of dating. Additional dating of cores from additional site would have provided a useful added insight to this study.

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Appendices

Appendix 1: Publications/ Presentations

Journal paper

Akinyemi, F.O., Hutchinson, S.M., Mindrescu, M. & Rothwell, J.J. (2013) Lake sediment records of atmospheric pollution in the Romanian Carpathians. *Quaternary International* 293:105-113

Conference papers

Simon M. Hutchinson, Olusola Akinyemi, Marcel Mindrescu & James J. Rothwell (2011) An overview of the recent palaeolimnology of selected lakes in the Romanian Carpathians. *PAGES 1st Carpathian Balkan Workshop* Suceava, Romania.

Simon M. Hutchinson, Olusola Akinyemi, Marcel Mindrescu & James J. Rothwell (2010) The atmospheric particulate pollution record of mountain lakes in the Romanian Carpathians. 1st Forum Carpaticum on the Integration of nature and society towards sustainability. Krakow, Poland.

S.M. Hutchinson, O. Akinyemi, M. Mindrescu, J.J. Rothwell (2009) Assessing the impacts of atmospheric particulate pollution on mountain lakes in the Romania Carpathians. *11th International Symposium Paleolimnology*, Guadalajara, Mexico.

Simon M. Hutchinson, Olusola Akinyemi, Marcel Mindrescu & James J. Rothwell (2008) Assessing the potential of lake sediment records of environmental change in the Romanian Carpathians. Preliminary data from the Rodna and Maramureş Mountains. *IAG Regional Conference on Geomorphology Landslides, Floods and Global Environmental Change in Mountain Regions*. Brasov, Romania.

Appendix 2: Density tables of lake sediments in the Fagaras region

Fagaras Mountains (2006)					
LAKE NAME: CALTUN LAKE; LcT2					
Depth			**BD**	**BD**	
Top	bot	mid	Wet	Dry	
1	1.5	1.25	1.260	0.278	
1.5	2	1.75	1.175	0.256	
2	2.5	2.25	1.190	0.245	
2.5	3	2.75	1.211	0.277	
3	3.5	3.25	1.221	0.268	
3.5	4	3.75	1.280	0.235	
4	4.5	4.25	1.219	0.238	
4.5	5	4.75	1.120	0.215	
5	5.5	5.25	1.187	0.210	
5.5	6	5.75	1.039	0.177	
6	6.5	6.25	1.105	0.148	
6.5	7	6.75	1.094	0.159	
7	7.5	7.25	1.189	0.225	
7.5	8	7.75	1.081	0.177	
8	8.5	8.25	1.281	0.263	
8.5	9	8.75	1.272	0.205	
9	9.5	9.25	1.098	0.181	
9.5	10	9.75	1.224	0.211	
10	10.5	10.25	1.171	0.171	
10.5	11	10.75	1.025	0.174	
11	11.5	11.25	1.207	0.219	
11.5	12	11.75	1.210	0.319	
12	12.5	12.25	1.135	0.199	
12.5	13	12.75	1.099	0.197	
13	13.5	13.25	1.168	0.229	
13.5	14	13.75	1.141	0.204	
14	14.5	14.25	1.126	0.241	
14.5	15	14.75	1.123	0.226	
15	15.5	15.25	1.209	0.246	
15.5	16	15.75	1.276	0.311	
16	16.5	16.25	1.201	0.216	
16.5	17	16.75	1.257	0.262	
17	17.5	17.25	1.301	0.268	
17.5	18	17.75	1.314	0.269	
18	18.5	18.25	1.125	0.152	
18.5	19	18.75	1.244	0.259	
19	19.5	19.25	1.225	0.281	
19.5	20	19.75	1.121	0.234	
20	20.5	20.25	1.072	0.289	
20.5	21	20.75	1.141	0.255	
21	21.5	21.25	1.113	0.251	

21.5	22	21.75	1.222	0.310	
22	22.5	22.25	1.123	0.293	
22.5	23	22.75	1.248	0.316	
23	23.5	23.25	1.206	0.326	
23.5	24	23.75	1.291	0.457	
24	24.5	24.25	1.351	0.428	
24.5	25	24.75	1.235	0.469	
25	25.5	25.25	1.389	0.530	
25.5	26	25.75	1.326	0.439	
26	26.5	26.25	1.256	0.346	
26.5	27	26.75	1.264	0.368	
27	27.5	27.25	1.075	0.256	
27.5	28	27.75	1.190	0.291	
28	28.5	28.25	1.178	0.297	
28.5	29	28.75	1.203	0.283	
30	30.5	30.25	1.234	0.342	
30.5	31	30.75	1.199	0.318	
				0.27	Mean
				0.08	STD Dev
				0.15	Min
LAKE NAME: PODRAGU; LPm2					
Depth			**BD**	**BD**	
Top	bot	mid	Wet	Dry	
1	1.5	1.25	1.486	0.56	
1.5	2	1.75	1.485	0.56	
2	2.5	2.25	1.441	0.56	
2.5	3	2.75	1.710	0.92	
3	3.5	3.25	1.522	0.65	
3.5	4	3.75	1.470	0.55	
4	4.5	4.25	1.301	0.48	
4.5	5	4.75	1.105	0.29	
5	5.5	5.25	1.509	0.63	
5.5	6	5.75	1.860	0.87	
6	6.5	6.25	1.424	0.66	
6.5	7	6.75	1.786	1.08	
7	7.5	7.25	1.659	0.91	
7.5	8	7.75	1.941	1.22	
8	8.5	8.25	2.034	1.25	
8.5	9	8.75	1.826	1.16	
9	9.5	9.25	1.878	1.15	
9.5	10	9.75	1.706	0.96	
10	10.5	10.25	1.601	0.83	
10.5	11	10.75	1.349	0.642	
11	11.5	11.25	1.554	0.894	
11.5	12	11.75	1.875	1.224	
12	12.5	12.25	1.928	1.256	
12.5	13	12.75	1.796	0.934	
13	13.5	13.25	1.602	0.695	
13.5	14	13.75	1.434	0.716	

14	14.5	14.25	1.483	0.697	
14.5	15	14.75	1.535	0.775	
15	15.5	15.25	1.711	0.962	
15.5	16	15.75	1.641	0.61	
16	16.5	16.25	1.636	0.896	
16.5	17	16.75	1.355	0.777	
17	17.5	17.25	2.223	0.874	
17.5	18	17.75	1.664	0.924	
			Mean	0.83	
			STD Dev	0.24	
			Min	0.29	
			Max	1.26	
Lacul Belea: Core LBa1					
Depth			**BD**	**BD**	
Top	bot	mid	Wet	Dry	
0	0.5	0.25	1.275	0.369	
0.5	1	0.75	1.289	0.451	
1	1.5	1.25	1.400	0.483	
1.5	2	1.75	1.516	0.565	
2	2.5	2.25	1.183	0.419	
2.5	3	2.75	1.425	0.505	
3	3.5	3.25	1.422	0.553	
3.5	4	3.75	1.546	0.725	
4	4.5	4.25	1.599	0.762	
4.5	5	4.75	1.404	0.623	
5	5.5	5.25	1.318	0.529	
5.5	6	5.75	1.350	0.552	
6	6.5	6.25	1.792	1.078	
6.5	7	6.75	1.786	1.252	
7	7.5	7.25	1.581	0.921	
7.5	8	7.75	1.415	0.882	
8	8.5	8.25	1.499	0.94	
8.5	9	8.75	1.927	1.218	
9	9.5	9.25	1.495	0.777	
9.5	10	9.75	1.432	0.633	
10	10.5	10.25	1.486	0.633	
10.5	11	10.75	1.465	0.653	
11.5	12	11.75	1.568	0.697	
12	12.5	12.25	1.220	0.548	
12.5	13	12.75	1.430	0.601	
13	13.5	13.25	1.494	0.627	
13.5	14	13.75	1.421	0.552	
14	14.5	14.25	1.446	0.637	
14.5	15	14.75	1.551	0.623	
15	15.5	15.25	1.749	0.926	
15.5	16	15.75	1.486	0.644	
16	16.5	16.25	1.335	0.524	
16.5	17	16.75	1.232	0.446	
17	17.5	17.25	1.300	0.583	
17.5	18	17.75	1.238	0.504	

18	18.5	18.25	1.329	0.513	
18.5	19	18.75	1.423	0.559	
19	19.5	19.25	1.593	0.763	
19.5	20	19.75	1.437	0.668	
20	20.5	20.25	1.591	0.68	
20.5	21	20.75	1.508	0.734	
21	21.5	21.25	1.568	0.828	
21.5	22	21.75	1.577	0.797	
22	22.5	22.25	1.464	0.693	
22.5	23	22.75	1.427	0.621	
23	23.5	23.25	1.389	0.61	
23.5	24	23.75	1.535	0.685	
24	24.5	24.25	1.349	0.591	
24.5	25	24.75	1.514	0.621	
25	25.5	25.25	1.529	0.666	
25.5	26	25.75	1.262	0.563	
26	26.5	26.25	1.692	0.946	
26.5	27	26.75	1.428	0.712	
27	27.5	27.25	1.533	0.762	
27.5	28	27.75	1.443	0.768	
28	28.5	28.25	1.677	1.016	
28.5	29	28.75	1.720	1.068	
29	29.5	29.25	1.779	1.094	
			Mean	0.70	
			STD Dev	0.20	
			Min	0.37	
Lacul Capra; Core LCp2			Max	**1.25**	
Depth			**BD**	**BD**	
Top	bot	mid	Wet	Dry	
0	0.5	0.25	0.964	0.06	
0.5	1	0.75	1.094	0.21	
1	1.5	1.25	1.200	0.29	
1.5	2	1.75	1.097	0.17	
2	2.5	2.25	1.112	0.18	
2.5	3	2.75	1.192	0.22	
3	3.5	3.25	1.332	0.37	
3.5	4	3.75	1.316	0.33	
4	4.5	4.25	1.267	0.33	
4.5	5	4.75	1.393	0.42	
5	5.5	5.25	1.340	0.48	
5.5	6	5.75	1.300	0.49	
6	6.5	6.25	1.415	0.481	
6.5	7	6.75	1.409	0.535	
7	7.5	7.25	1.269	0.407	
7.5	8	7.75	1.458	0.455	
8	8.5	8.25	1.408	0.492	
8.5	9	8.75	1.417	0.521	
9	9.5	9.25	1.877	0.425	
9.5	10	9.75	1.389	0.50	
10	10.5	10.25	1.410	0.47	
10.5	11	10.75	1.350	0.44	

11	11.5	11.25	1.442	0.50	
11.5	12	11.75	1.455	0.53	
12	12.5	12.25	1.416	0.50	
12.5	13	12.75	1.495	0.58	
13	13.5	13.25	1.441	0.52	
13.5	14	13.75	1.401	0.45	
14	14.5	14.25	1.315	0.43	
14.5	15	14.75	1.416	0.45	
15	15.5	15.25	1.380	0.46	
15.5	16	15.75	1.437	0.521	
16	16.5	16.25	1.344	0.468	
16.5	17	16.75	1.420	0.5	
17	17.5	17.25	1.417	0.524	
			Mean	**0.42**	
			STD Dev	0.12	
			Min	0.06	
			Max	0.58	

Appendix 3: Density tables of lake sediments in the Rodna/Maramures region

Lacul Stiol; Core LS 1				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	7.098	5.51
1	2	1.5	9.445	7.86
2	3	2.5	15.494	13.90
3	4	3.5	19.465	17.88
4	5	4.5	26.564	24.97
5	6	5.5	23.686	22.10
6	7	6.5	25.874	24.28
7	8	7.5	26.414	24.82
8	9	8.5	30.344	28.75
9	10	9.5	27.040	25.45
10	11	10.5	31.003	29.41
11	12	11.5	26.714	25.12
			Mean	20.84
			STD Dev	7.88
			Min	5.51
Lacul Stiol; Core LS 2			**Max**	**29.41**
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	8.890	7.30
1	2	1.5	11.237	9.65
2	3	2.5	11.690	10.10
3	4	3.5	18.024	16.43
4	5	4.5	28.653	27.06
5	6	5.5	26.678	25.09
6	7	6.5	28.136	26.55
7	8	7.5	31.083	29.49
8	9	8.5	27.146	25.56
9	10	9.5	25.454	23.86
10	11		Mean	20.11
11	12		STD Dev	8.39
			Min	7.30
			Max	29.49
Lacul Stiol; Core LS 3				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	8.475	6.89
1	2	1.5	10.831	9.24
2	3	2.5	21.398	19.81
3	4	3.5	23.724	22.13
4	5	4.5	25.623	24.03
5	6	5.5	30.143	28.55
6	7	6.5	28.586	27.00

7	8	7.5	50.860	49.27
8	9	8.5	53.120	51.53
9	10		Mean	26.49
10	11		STD Dev	15.42
11	12		Min	6.89
			Max	51.53
Lacul Pietrosul; Core LP 1				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	8.287	6.70
1	2	1.5	17.510	15.92
2	3	2.5	19.380	17.79
3	4	3.5	20.823	19.23
4	5	4.5	22.381	20.79
5	6	5.5	22.421	20.83
6	7	6.5	20.088	18.50
7	8	7.5	20.759	19.17
8	9		Mean	17.37
9	10		STD Dev	4.60
10	11		Min	6.70
11	12		**Max**	20.83
Lacul Pietrosul; Core LP 2				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	9.177	7.59
1	2	1.5	19.291	17.70
2	3	2.5	28.712	27.12
3	4	3.5	26.731	25.14
4	5	4.5	19.456	17.87
5	6			
6	7		Mean	19.08
7	8		STD Dev	7.70
8	9		Min	7.59
9	10		**Max**	27.12
10	11			
11	12			
Lacul Pietrosul; Core LP 3				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	10.761	9.17
1	2	1.5	20.586	19.00
2	3	2.5	24.787	23.20
3	4	3.5	26.953	25.36

4	5	4.5	27.341	25.75
5	6	5.5	20.045	18.46
6	7		Mean	20.16
7	8		STD Dev	6.21
8	9		Min	9.17
9	10		**Max**	25.75
10	11			
11	12			
Lacul Buhaiescu-3; Core LB-3 1				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	16.334	14.74
1	2	1.5	13.986	12.40
2	3	2.5	17.004	15.41
3	4	3.5	16.830	15.24
4	5	4.5	18.854	17.26
5	6	5.5	21.205	19.62
6	7	6.5	20.066	18.48
7	8	7.5	21.781	20.19
8	9	8.5	16.334	14.74
9	10		Mean	16.45
10	11		STD Dev	2.59
11	12		Min	12.40
			Max	20.19
Lacul Buhaiescu-3; Core LB-3 2				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	15.654	14.06
1	2	1.5	15.982	14.39
2	3	2.5	12.088	10.50
3	4	3.5	15.711	14.12
4	5	4.5	17.956	16.37
5	6	5.5	17.109	15.52
6	7	6.5	20.417	18.83
7	8	7.5	23.531	21.94
8	9	8.5	39.805	38.22
9	10	9.5	15.727	14.14
10	11		Mean	17.81
11	12		STD Dev	7.80
			Min	10.50
			Max	38.22
Lacul Buhaiescu-3; Core LB-3 3				

Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	6.928	5.34
1	2	1.5	12.559	10.97
2	3	2.5	11.200	9.61
3	4	3.5	13.224	11.63
4	5	4.5	15.791	14.20
5	6	5.5	14.805	13.22
6	7	6.5	18.487	16.90
7	8	7.5	17.409	15.82
8	9	8.5	36.855	35.27
9	10	9.5	26.641	25.05
10	11	10.5	22.288	20.70
11	12	11.5	18.278	16.69
12	13	12.5	22.172	20.58
			Mean	16.61
			STD Dev	7.65
			Min	5.34
Lacul Lala Mare; Core LLM 1			**Max**	**35.27**
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	9.663	8.07
1	2	1.5	11.366	9.78
2	3	2.5	17.054	15.46
3	4	3.5	15.900	14.31
4	5	4.5	16.463	14.87
5	6	5.5	18.972	17.38
6	7	6.5	18.916	17.33
7	8	7.5	21.153	19.56
8	9	8.5	21.605	20.02
9	10		Mean	15.20
10	11		STD Dev	4.07
11	12		Min	8.07
			Max	20.02
Lacul Lala Mare; Core LLM 2				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	12.774	11.18
1	2	1.5	10.673	9.08
2	3	2.5	12.824	11.23
3	4	3.5	13.934	12.34
4	5	4.5	12.324	10.73
5	6	5.5	13.801	12.21
6	7	6.5	15.217	13.63
7	8	7.5	15.665	14.08
8	9	8.5	16.790	15.20
9	10	9.5	16.767	15.18
10	11	10.5	18.819	17.23

11	12		Mean	12.92
			STD Dev	2.39
			Min	9.08
			Max	17.23
Lacul Lala Mare; Core LLM 3				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	5.527	3.94
1	2	1.5	5.858	4.27
2	3	2.5	7.686	6.10
3	4	3.5	8.936	7.35
4	5	4.5	9.920	8.33
5	6	5.5	9.003	7.41
6	7	6.5	12.326	10.74
7	8	7.5	10.782	9.19
8	9	8.5	12.236	10.65
9	10	9.5	12.335	10.75
10	11	10.5	13.246	11.66
11	12	11.5	12.801	11.21
12	13	12.5	14.498	12.91
13	14	13.5	12.855	11.27
			Mean	8.98
			STD Dev	2.82
			Min	3.94
			Max	12.91

Lacul Vinderel 1				
Depth				**BD**
Top	bot	mid	mass	Dry
0	1	0.5	8.733	0.26
1	2	1.5	9.756	0.29
2	3	2.5	10.105	0.30
3	4	3.5	8.985	0.27
4	5	4.5	10.747	0.32
5	6	5.5	12.785	0.39
6	7	6.5	14.770	0.44
7	8	7.5	13.741	0.41
8	9	8.5	13.876	0.42
9	10	9.5	17.237	0.52
10	11	10.5	18.245	0.55
11	12	11.5	16.833	0.51
12	13	12.5	17.540	0.53
13	14	13.5	20.438	0.62
14	15	14.5	25.825	0.78
15	16	15.5	10.913	0.33
Lacul Vinderel 2				
Depth				**BD**
Top	bot	mid	mass	Dry

0	1	0.5	9.730	0.29		
1	2	1.5	15.392	0.46		
2	3	2.5	14.411	0.43		
3	4	3.5	16.596	0.50		
4	5	4.5	17.945	0.54		
5	6	5.5	18.987	0.57		
6	7	6.5	19.385	0.58		
7	8	7.5	16.431	0.49		
8	9	8.5	20.008	0.60		
9	10	9.5	23.174	0.70		
11	12	11.5	9.762	0.29		
Lacul Vinderel 3						
Depth						**BD**
Top	bot	mid	mass 1	mass 2	mass	dry
1	2	1.5	8.125	5.627	13.752	0.41
2	3	2.5	6.843	6.180	13.023	0.39
3	4	3.5	7.455	6.449	13.904	0.42
4	5	4.5	6.111	5.694	11.805	0.36
5	6	5.5	7.520	6.708	14.228	0.43
6	7	6.5	8.346	8.265	16.611	0.50
7	8	7.5	8.630	8.464	17.094	0.51
8	9	8.5	8.489	8.832	17.321	0.52
9	10	9.5	7.946	12.488	20.434	0.62
10	11	10.5	8.366	10.061	18.427	0.56
11	12	11.5	7.408	11.741	19.149	0.58
12	13	12.5	8.448	14.652	23.100	0.70
13	14	13.5	7.596	17.016	24.612	0.74
					Mean	0.52
					Stdev	0.12
					Min	0.36
					Max	0.74
Lacul BILA LB1						
Depth						**BD**
Top	bot	mid	mass 1	mass 2	mass	Dry
1	2	1.5	3.974	2.989	6.963	0.21
2	3	2.5	5.770	6.949	12.719	0.38
3	4	3.5	6.155	8.966	15.121	0.46
4	5	4.5	5.136	10.606	15.742	0.47
5	6	5.5	6.089	10.068	16.157	0.49
6	7	6.5	6.753	9.867	16.620	0.50
7	8	7.5	5.699	7.888	13.587	0.41
9	10	9.5	5.775	13.706	19.481	0.59
10	11	10.5	6.234	17.030	23.264	0.70
11	12	11.5	7.157	13.205	20.362	0.61
12	13	12.5	5.411	19.870	25.281	0.76

13	14	13.5	4.954	12.411	17.365	0.52
14	15	14.5	5.769	21.255	27.024	0.81
15	16	15.5	7.019	8.355	15.374	0.46
16	17	16.5	7.436	29.261	36.697	1.11
17	18	17.5	6.971	20.672	27.643	0.83
18	19	18.5	6.921	24.447	31.368	0.94
20	21	20.5	6.433	30.977	37.410	1.13
22	23	22.5	7.838	19.953	27.791	0.84
					Mean	0.64
					Stdev	0.25
					Min	0.21
Lacul BILA LB 2					Max	1.13
Depth				**BD**		
Top	bot	mid	mass	Dry		
0	1	0.5	13.078	0.39		
1	2	1.5	16.088	0.48		
2	3	2.5	19.841	0.60		
3	4	3.5	9.888	0.30		
4	5	4.5	22.859	0.69		
5	6	5.5	29.065	0.88		
6	7	6.5	23.349	0.70		
7	8	7.5	24.327	0.73		
8	9	8.5	20.905	0.63		
9	10	9.5	32.144	0.97		
10	11	10.5	30.363	0.91		
11	12	11.5	33.184	1.00		
12	13	12.5	36.242	1.09		
13	14	13.5	32.829	0.99		
14	15	14.5	38.871	1.17		
15	16	15.5	40.330	1.21		
16	17	16.5	35.104	1.06		
17	18	17.5	31.415	0.95		
			Mean	0.82		
			Stdev	0.27		
			Min	0.30		
Lacul Bila LB 3			Max	1.21		
Depth				**BD**		
Top	bot	mid	mass	Dry		
0	1	0.5	10.136	0.31		
1	2	1.5	20.382	0.61		
2	3	2.5	21.770	0.66		
3	4	3.5	25.902	0.78		
4	5	4.5	34.031	1.03		
6	7	6.5	29.408	0.89		
7	8	7.5	32.671	0.98		
8	9	8.5	25.112	0.76		
9	10	9.5	31.063	0.94		
10	11	10.5	24.983	0.75		
11	12	11.5	30.836	0.93		
12	13	12.5	33.841	1.02		

13	14	13.5	32.875	0.99
14	15	14.5	36.121	1.09
15	16	15.5	32.216	0.97
16	17	16.5	36.654	1.10
17	18	17.5	36.823	1.11
18	19	18.5	31.413	0.95
19	20	19.5	44.960	1.35
20	21	20.5	9.383	0.28

Appendix 4: Loss-on-ignition tables of lake sediments in the Fagaras region

LOCATION: 114 Cockcroft Building							
DATE:19-10-2009							
CORE NAME: CAPRA							
No	**Description**	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI (g100g^{-1})
1	LcP2:0-1						
2	LcP2:1-2	6.52	6.73	0.21	6.70	0.17	16.52
3	LcP2:2-3	6.98	7.18	0.20	6.81	0.17	16.26
4	LcP2:3-4	6.51	6.71	0.21	6.68	0.17	16.06
5	LcP2:4-4.5	6.43	6.63	0.20	6.61	0.18	11.89
6	LcP2:5-5.5	6.32	6.52	0.21	6.50	0.19	10.69
7	LcP2:6-6.5	6.58	6.78	0.20	6.75	0.18	13.28
8	LcP2:7-7.5	6.95	7.15	0.20	7.13	0.18	12.31
9	LcP2:8-8.5	6.57	6.77	0.20	6.75	0.18	10.46
10	LcP2:9-9.5	6.40	6.60	0.21	6.57	0.17	15.71
11	LcP2:10-10.5	6.24	6.44	0.20	6.41	0.17	13.77
12	LcP2:11-11.5	6.21	6.42	0.21	6.39	0.18	15.42
13	LcP2:12-12.5	6.88	7.08	0.20	7.06	0.18	11.56
14	LcP2:13-13.5	6.48	6.69	0.20	6.66	0.17	14.29
15	LcP2:14-14.5	6.27	6.48	0.20	6.45	0.17	14.08
16	LcP2:15-15.5	6.40	6.60	0.21	6.57	0.17	15.63
17	LcP2:16-16.5	6.73	6.93	0.20	6.90	0.17	14.54
18	LcP2:17-17.5	6.95	7.15	0.20	7.13	0.18	12.31
						Mean	13.81
DATE:19-10-2009							
CORE NAME: BALEA LAKE							

No	Description	Crucible (g)	Crucible+Oven dried Sample (g)	Oven dried Sample (g)	Crucible+Ignited Sample (g)	Sample after Ignition (g)	LOI (g100g^{-1})
65	LBa1:0-1	6.58	6.78	0.21	6.75	0.17	15.29
66	LBa1:1-2	6.72	6.93	0.20	6.90	0.18	12.39
67	LBa1:2-3	6.59	6.80	0.21	6.77	0.18	12.68
68	LBa1:3-4	6.39	6.59	0.20	6.57	0.17	11.81
69	LBa1:4-4.5	6.59	6.80	0.21	6.77	0.19	11.17
70	LBa1:5-5.5	6.48	6.69	0.21	6.67	0.18	11.64
71	LBa1:6-6.5	6.56	6.76	0.20	6.73	0.18	11.87
72	LBa1:7-7.5	6.77	6.97	0.20	6.96	0.19	6.84
73	LBa1:8-8.5	6.38	6.59	0.20	6.57	0.19	6.40
74	LBa1:9-9.50	6.87	7.08	0.21	7.06	0.18	11.45
75	LBa1:10-10.5	6.39	6.59	0.20	6.57	0.18	11.34
76	LBa1:11.5-12	6.39	6.59	0.20	6.57	0.17	11.81
77	LBa1:12-12.5	6.38	6.58	0.20	6.55	0.18	12.05
78	LBa1:13-13.5	6.45	6.65	0.20	6.63	0.18	12.52
79	LBa1:14-14.5	6.82	7.02	0.21	7.00	0.18	10.67
80	LBa1:15-15.5	6.72	6.92	0.20	6.90	0.17	12.73
81	LBa1:160-16.5	6.59	6.80	0.21	6.77	0.19	11.17
82	LBa1:17-17.5	6.45	6.65	0.21	6.64	0.19	8.83
83	LBa1:18-18.5	6.33	6.54	0.21	6.51	0.18	13.56
84	LBa1:19-19.5	6.79	6.99	0.20	6.97	0.18	8.30
85	LBa1:20-20.5	6.30	6.51	0.21	6.49	0.19	8.91
86	LBa1:21-21.5	6.81	7.01	0.20	6.99	0.18	11.75
87	LBa1:22-22.5	6.70	6.91	0.20	6.89	0.19	8.94
88	LBa1:23-23.5	6.41	6.61	0.20	6.60	0.18	9.08
89	LBa1:24-24.5	6.53	6.73	0.21	6.71	0.19	9.45
90	LBa1:25-25.5	6.44	6.64	0.20	6.61	0.17	13.70
91	LBa1:26-26.5	6.39	6.60	0.21	6.58	0.19	7.64
92	LBa1:27-27.5	6.15	6.36	0.21	6.34	0.19	8.86
93	LBa1:28-28.5	6.25	6.46	0.21	6.44	0.19	8.84
94	LBa1:30-30.5	6.30	6.51	0.21	6.49	0.19	7.49
						Mean	10.64

DATE:19-10-2009

CORE NAME:CALTUN LAKE

No	Description	Crucible (g)	Crucible+Oven dried Sample (g)	Oven dried Sample (g)	Crucible+Ignited Sample (g)	Sample after Ignition (g)	LOI (g100g^{-1})
	LcT2:0-1	6.43	6.63	0.20	6.59	0.16	21.59
19	LcT2:1-2	6.45	6.65	0.20	6.61	0.16	19.90
20	LcT2:2-3	6.95	7.16	0.21	7.13	0.18	14.11
21	LcT2:3-4	7.07	7.27	0.20	7.23	0.17	15.83
22	LcT2:4-5	6.88	7.08	0.20	7.05	0.17	14.40
23	LcT2:5-5.5	6.50	6.67	0.17	6.64	0.14	15.93
24	LcT2:6-6.5	6.40	6.54	0.14	6.52	0.12	15.01
25	LcT2:7-7.5	6.44	6.64	0.21	6.62	0.18	12.34

26	LcT2:8-8.5	6.58	6.76	0.18	6.73	0.15	15.70
27	LcT2:9-9.5	6.50	6.69	0.19	6.66	0.16	17.32
28	LcT2:10-10.5	6.57	6.77	0.20	6.74	0.17	14.19
29	LcT2:11-11.5	6.47	6.67	0.20	6.64	0.17	17.13
30	LcT2:12-12.5	5.65	5.82	0.17	5.79	0.15	16.60
31	LcT2:13-13.5	6.38	6.58	0.20	6.55	0.17	13.23
32	LcT2:14-14.5	6.80	7.00	0.20	6.96	0.17	16.80
33	LcT2:15-15.5	6.86	7.07	0.21	7.03	0.17	16.19
34	LcT2:16-16.5	6.62	6.83	0.21	6.80	0.18	12.45
35	LcT2:17-17.5	6.98	7.19	0.21	7.16	0.17	15.98
36	LcT2:18-18.5	6.38	6.58	0.20	6.55	0.17	15.68
37	LcT2:19-19.5	6.51	6.71	0.20	6.69	0.17	12.65
38	LcT2:20-20.5	6.39	6.60	0.20	6.57	0.17	14.16
39	LcT2:21-21.5	6.62	6.82	0.20	6.80	0.18	12.43
40	LcT2:22-22.5	6.57	6.77	0.20	6.75	0.18	11.00
41	LcT2:23-23.5	6.61	6.82	0.21	6.80	0.18	11.52
42	LcT2:24-24.5	6.46	6.67	0.20	6.64	0.18	11.08
43	LcT2:25-25.5	6.81	7.01	0.20	6.99	0.18	11.75
44	LcT2:26-26.5	6.84	7.04	0.21	7.01	0.17	15.88
45	LcT2:27-27.5	6.72	6.93	0.21	6.89	0.17	18.63
46	LcT2:28-28.5	6.53	6.73	0.21	6.71	0.19	9.45
47	LcT2:30-30.5	6.50	6.71	0.21	6.70	0.19	7.65
						Mean	14.55
DATE:19-10-2009							
CORE NAME:PODRAGU MARE							
No	**Description**	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
48	LPm2:1-2	6.65	6.86	0.21	6.83	0.18	12.79
49	LPm2:2-3	6.47	6.66	0.18	6.63	0.16	12.68
50	LPm2:3-4	6.37	6.58	0.21	6.55	0.19	11.39
51	LPm2:4-5	6.78	6.98	0.21	6.95	0.17	14.68
52	LPm2:5-5.5	6.45	6.66	0.21	6.64	0.18	12.91
53	LPm2:6-6.5	6.75	6.96	0.21	6.93	0.18	12.16
54	LPm2:7-7.5	6.78	6.98	0.21	6.96	0.18	11.07
55	LPm2:8-8.5	6.42	6.62	0.21	6.60	0.18	11.16
56	LPm2:9-9.5	6.54	6.74	0.20	6.72	0.17	13.13
57	LPm2:10-10.5	6.62	6.83	0.21	6.80	0.18	12.45
58	LPm2:11-11.5	6.36	6.56	0.20	6.53	0.17	12.74
59	LPm2:12-12.5	6.65	6.85	0.20	6.83	0.18	12.40
60	LPm2:13-13.5	6.65	6.86	0.21	6.83	0.18	12.79
61	LPm2:14-14.5	6.47	6.66	0.18	6.63	0.16	12.68
62	LPm2:15-15.5	6.63	6.84	0.21	6.82	0.18	10.98
63	LPm2:16-16.5	6.80	6.99	0.20	6.97	0.17	11.15
64	LPm2:17-17.5	6.81	7.01	0.20	6.99	0.18	11.75
						Mean	12.29

DATE27-10-2009							
CORE NAME: BALEA LAKE 4							
No	**Description**	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
1	LBa4:0-1	6.7	6.9	0.21	6.87	0.18	11.26
2	LBa4:1-2	6.1	6.3	0.20	6.29	0.18	10.68
3	LBa4:2-3	6.1	6.3	0.20	6.31	0.18	12.97
4	LBa4:3-4	6.1	6.3	0.20	6.23	0.17	13.02
5	LBa4:4-5	6.1	6.3	0.20	6.24	0.18	11.53
6	LBa4:5-6	6.4	6.6	0.20	6.57	0.18	13.83
7	LBa4:6-7	6.4	6.6	0.20	6.58	0.18	11.02
8	LBa4:7-8	6.5	6.7	0.20	6.72	0.18	9.79
9	LBa4:8-9	6.5	6.7	0.20	6.69	0.19	7.86
10	LBa4:9-10	6.3	6.5	0.20	6.46	0.18	11.26
11	LBa4:10-11	6.5	6.7	0.20	6.68	0.17	14.95
12	LBa4:11-12	6.6	6.8	0.21	6.79	0.18	14.64
13	LBa4:12-13	6.3	6.5	0.20	6.44	0.17	11.99
14	LBa4:13-14	6.7	6.9	0.21	6.87	0.18	11.75
15	LBa4:14-15	6.3	6.5	0.20	6.45	0.18	11.13
16	LBa4:15-16	6.1	6.3	0.21	6.30	0.19	8.71
17	LBa4:16-17	6.6	6.8	0.21	6.74	0.19	7.26
18	LBa4:17-18	6.4	6.6	0.21	6.58	0.20	5.32
19	LBa4:18-19	6.4	6.6	0.21	6.56	0.19	7.05
20	LBa4:19-20	6.4	6.6	0.20	6.55	0.18	10.23
21	LBa4:20-21	6.7	6.9	0.20	6.84	0.18	10.51
22	LBa4:21-22	6.0	6.2	0.21	6.19	0.20	7.88
23	LBa4:22-23	5.9	6.1	0.21	6.13	0.19	5.31
24	LBa4:23-24	6.7	6.9	0.20	6.87	0.19	5.26
25	LBa424-25	6.6	6.8	0.20	6.83	0.19	6.08
26	LBa4:25-26	6.7	7.0	0.21	6.94	0.19	8.19
27	LBa4:26-27	6.7	6.9	0.20	6.93	0.19	3.79
28	LBa4:27-28	6.3	6.5	0.21	6.49	0.19	7.49
29	LBa4:29-30	6.1	6.4	0.21	6.34	0.19	8.86
30	LBa4:30-31	6.3	6.5	0.20	6.49	0.18	7.88
						Mean	9.58

Appendix 5: Loss-on-ignition tables of lake sediments in the Rodna/Maramures region

Lacul Pietrosul; Core LP 1								
Depth								
top	Bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI (g100g-1)
0	1	0.5	6.47	6.66	0.19	6.62	0.15	21.06
1	2	1.5	6.57	6.77	0.20	6.75	0.18	11.31
2	3	2.5	6.33	6.54	0.21	6.52	0.19	9.34
3	4	3.5	6.50	6.71	0.21	6.70	0.19	7.65
4	5	4.5	6.50	6.70	0.20	6.69	0.18	9.53
5	6	5.5	6.44	6.63	0.20	6.61	0.18	11.08
6	7	6.5	6.67	6.87	0.20	6.85	0.18	8.09
7	8	7.5	6.52	6.72	0.20	6.70	0.18	9.49
							MEAN	10.94
Rodna Mountains (2006)								
Lacul Stiol; Core LS 2								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI (g100g-1)
0	1	0.5						
1	2	1.5	6.5242	6.73	0.21	6.70	0.17	16.52
2	3	2.5	6.3788	6.58	0.20	6.55	0.17	14.47
3	4	3.5	6.5777	6.79	0.21	6.76	0.19	10.59
4	5	4.5	6.5628	6.77	0.20	6.75	0.19	9.38
5	6	5.5	6.3266	6.54	0.21	6.52	0.19	9.44
6	7	6.5	6.5584	6.76	0.20	6.74	0.18	9.03
7	8	7.5	5.7421	5.95	0.21	5.93	0.19	9.45
8	9	8.5	6.9311	7.13	0.20	7.11	0.18	8.73
9	10	9.5	6.5092	6.71	0.20	6.69	0.18	9.56
Rodna Mountains (2006)								
Lacul Buhaiescu-3; Core LB-3 2								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI (g100g-1)
0	1	0.5	6.51	6.71	0.21	6.67	0.17	18.97
1	2	1.5	6.5062	6.71	0.21	6.67	0.16	21.45

2	3	2.5	5.7421	5.95	0.20	5.90	0.16	21.53
3	4	3.5	6.5777	6.78	0.20	6.74	0.17	18.22
4	5	4.5	6.5087	6.72	0.21	6.69	0.18	11.99
5	6	5.5	6.473	6.68	0.21	6.65	0.17	17.31
6	7	6.5	6.8103	7.01	0.20	6.99	0.18	11.75
7	8	7.5	6.5767	6.78	0.20	6.75	0.18	12.35
8	9	8.5	6.8105	7.01	0.20	7.00	0.19	6.39
9	10	9.5	6.5252	6.73	0.21	6.71	0.19	9.45
Rodna Mountains (2006)								
Lacul Lala Mare; Core LLM 2								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
0	1	0.5	6.54	6.74	0.20	6.71	0.17	14.19
1	2	1.5	6.3935	6.5975	0.20	6.57	0.18	14.22
2	3	2.5	6.5242	6.72	0.20	6.70	0.17	14.38
3	4	3.5	6.646	6.85	0.21	6.82	0.18	14.35
4	5	4.5	6.8734	7.07	0.20	7.05	0.17	13.87
5	6	5.5	6.5907	6.79	0.20	6.76	0.17	13.56
6	7	6.5	6.9523	7.16	0.21	7.13	0.18	14.11
7	8	7.5	6.4774	6.68	0.20	6.65	0.18	12.49
8	9	8.5	6.6096	6.81	0.20	6.79	0.18	12.70
9	10	9.5	6.4027	6.61	0.20	6.58	0.18	13.12
Rodna Mountains		**2006**						
CORE NAME: Lacul Vinderel 3								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
0	1	0.5						
1	2	1.5	6.609	6.81	0.20	6.78	0.17	14.21
2	3	2.5	6.786	6.99	0.20	6.96	0.17	15.04
3	4	3.5	6.525	6.73	0.20	6.70	0.17	15.74
4	5	4.5	7.065	7.27	0.20	7.23	0.17	15.83
5	6	5.5	6.777	6.98	0.20	6.95	0.17	14.96
6	7	6.5	6.392	6.60	0.21	6.57	0.18	13.29
7	8	7.5	6.376	6.58	0.20	6.55	0.17	13.23
8	9	8.5	6.653	6.85	0.20	6.82	0.17	14.00
9	10	9.5	6.365	6.57	0.21	6.54	0.18	13.70
10	11	10.5	6.435	6.64	0.21	6.62	0.18	12.34

11	12	11.5	6.245	6.45	0.20	6.42	0.18	11.97
12	13	12.5	6.874	7.08	0.20	7.05	0.18	12.64
13	14	13.5	6.570	6.77	0.20	6.74	0.17	14.19
14	15	14.5	6.880	7.08	0.20	7.05	0.17	14.40
Rodna Mountains		**2006**						
CORE NAME:LACUL BILA LB1								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
0	1	0.5	6.39	6.60	0.20	6.56	0.17	16.69
1	2	1.5	6.594	6.80	0.20	6.77	0.17	16.09
2	3	2.5	6.695	6.90	0.20	6.87	0.17	14.96
3	4	3.5	6.594	6.80	0.20	6.77	0.17	15.94
4	5	4.5	6.671	6.88	0.21	6.84	0.17	15.57
5	6	5.5	6.512	6.71	0.20	6.68	0.17	16.29
6	7	6.5	6.719	6.93	0.21	6.89	0.17	16.25
7	8	7.5	6.854	7.06	0.20	7.02	0.17	16.11
9	10	9.5	6.563	6.77	0.20	6.74	0.18	14.33
10	11	10.5	6.778	6.98	0.21	6.95	0.18	14.20
11	12	11.5	6.567	6.77	0.20	6.74	0.17	13.53
12	13	12.5	6.538	6.74	0.20	6.71	0.18	13.86
13	14	13.5	6.623	6.83	0.21	6.80	0.18	12.45
14	15	14.5	6.878	7.09	0.21	7.06	0.18	13.28
15	16	15.5	7.065	7.27	0.20	7.24	0.18	13.08
16	17	16.5	6.512	6.71	0.20	6.69	0.17	12.65
17	18	17.5	6.621	6.82	0.20	6.80	0.18	11.53
18	19	18.5	6.706	6.92	0.21	6.89	0.18	12.11
19	20	19.5	6.568	6.77	0.20	6.75	0.18	11.00
Rodna Mountains		**2008**						
CORE NAME:LACUL PETROSUL LP1								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
0	1	0.5	6.33	6.53	0.20	6.48	0.15	25.62
1	2	1.5	6.6463	6.85	0.21	6.82	0.17	16.80
2	3	2.5	6.5207	6.73	0.20	6.69	0.17	15.40
3	4	3.5	6.6534	6.86	0.21	6.83	0.18	12.79
4	5	4.5	6.5941	6.80	0.21	6.78	0.18	10.32
5	6	5.5	6.5536	6.75	0.20	6.73	0.18	10.33
6	7	6.5	6.7954	6.99	0.20	6.97	0.17	11.15

7	8	7.5	6.6573	6.86	0.20	6.84	0.18	9.26
8	9	8.5	6.4331	6.64	0.20	6.62	0.18	9.20
Maramuris Mountains		**2008**						
CORE NAME: Lacul Vinderel 1								
Depth								
top	bot	mid	**Crucible (g)**	**Crucible+Oven dried Sample (g)**	**Oven dried Sample (g)**	**Crucible+Ignited Sample (g)**	**Sample after Ignition (g)**	LOI ($g100g^{-1}$)
0	1	0.5	6.79	6.99	0.20	6.96	0.18	13.85
1	2	1.5	6.72	6.92	0.21	6.90	0.18	13.23
2	3	2.5	6.34	6.54	0.21	6.52	0.18	12.72
3	4	3.5	6.66	6.87	0.21	6.84	0.18	13.32
4	5	4.5	6.50	6.71	0.21	6.68	0.18	13.69
5	6	5.5	6.36	6.56	0.21	6.53	0.18	14.22
6	7	6.5	6.37	6.57	0.21	6.54	0.18	14.25
7	8	7.5	5.65	5.85	0.21	5.82	0.18	14.69
8	9	8.5	6.52	6.73	0.21	6.70	0.17	15.99
9	10	9.5	6.58	6.78	0.21	6.75	0.17	15.29
10	11	10.5	6.00	6.20	0.20	6.17	0.17	14.95
11	12	11.5	6.44	6.64	0.21	6.62	0.18	12.66
12	13	12.5	6.39	6.59	0.20	6.57	0.17	11.81
13	14	13.5	6.63	6.84	0.21	6.81	0.18	12.41
14	15	14.5	6.07	6.27	0.21	6.25	0.18	12.00
15	16	15.5	6.56	6.76	0.20	6.73	0.18	11.87
16	17	16.5	6.18	6.38	0.20	6.35	0.17	13.21
17	18	17.5	6.20	6.41	0.21	6.39	0.18	13.64
18	19	18.5	6.87	7.08	0.21	7.06	0.18	11.45
19	20	19.5	6.50	6.70	0.20	6.68	0.18	10.84
20	21	20.5	6.60	6.80	0.21	6.79	0.19	9.24
21	22	21.5	6.38	6.58	0.20	6.55	0.18	12.05
22	23	22.5	6.42	6.62	0.20	6.60	0.17	12.52
23	24	23.5	6.15	6.35	0.20	6.33	0.18	12.80
24	25	24.5	6.72	6.92	0.20	6.90	0.17	12.73
25	26	25.5	6.05	6.26	0.20	6.23	0.18	13.07
26	27	26.5	6.43	6.64	0.20	6.61	0.18	13.08
27	28	27.5	6.33	6.54	0.21	6.51	0.18	13.56
28	29	28.5	6.33	6.54	0.20	6.51	0.18	12.47
29	30	29.5	6.10	6.30	0.20	6.27	0.18	12.05
30	31	30.5	6.85	7.05	0.20	7.03	0.17	12.85
31	32	31.5	6.75	6.95	0.20	6.92	0.17	13.91

Appendix 6: Particle size characteristics tables of lake sediments in the Fagaras region

Romanina Lakes Project					
Fagaras Mountains			2007		
CORE NAME:LACUL BALEA LBa4					
Depth	Lba 4	Lba 4	Lba 4	Lba 4	Lba 4
	D10	D30	D60	D80	D90
0.5	8.0758	12.0128	20.70	48.28	84.9207
1.5	8.1999	11.9738	20.73	61.03	109.652
2.5	8.0925	12.0353	22.43	77.16	134.4177
3.5	7.6943	10.8214	15.86	24.15	39.4661
4.25	7.4533	10.9832	17.30	31.64	54.9578
5.25	8.2138	11.5358	17.19	29.14	52.7279
6.25	7.5322	10.588	15.30	22.61	36.5369
7.25	7.9945	11.4173	17.43	30.56	52.7811
8.25	8.046	11.0413	15.61	22.40	34.5886
9.25	8.4285	11.7221	17.30	28.63	50.1581
10.25	7.6179	11.4756	19.46	43.40	78.2498
11.25	7.4508	10.6702	16.05	27.03	55.4593
12.25	7.4818	10.7016	16.20	28.10	56.9031
13.25	7.3224	10.7188	16.60	28.77	52.1286
14.25	7.955	11.4725	18.71	51.09	105.4949
15.25	8.198	11.9472	20.71	56.31	95.7831
16.25	8.9449	14.6863	55.33	116.45	171.5781
17.25	8.6638	13.5837	47.01	108.26	159.9787
18.25	10.3037	36.6871	113.87	160.12	195.7571
19.25	8.7178	12.8355	26.46	81.27	133.5213
20.25	8.3858	12.174	20.45	49.01	87.1575
21.25	7.7652	11.1573	17.43	35.07	71.3807
22.25	8.3435	12.8625	31.05	79.69	118.2911
23.25	10.2885	23.8167	104.49	176.70	239.3585
24.25	9.1382	15.0092	61.49	112.86	155.1866
25.25	8.6311	13.2451	34.82	87.98	132.5925
26.25	9.7727	32.085	126.63	175.63	211.4665
27.25	9.3443	14.3237	52.42	107.94	149.1486
28.25	8.4792	13.0651	34.92	84.94	123.4687
30.25	8.4319	12.2635	20.19	41.28	71.4293
Mean	8.37	13.96	34.47	67.58	103.82
STD Dev.1	0.79	6.10	30.25	45.80	55.32
STD Dev.2	1.57	12.19	60.50	22.40	34.59
Min	7.32	10.59	15.30	176.70	239.36

Romanina Lakes Project					
Fagaras Mountains		**2007**			

CORE NAME: Lacul Capra 2					
Depth	LCp 2	LCp 2	LCp 2	LCp 2	LCp 2
	D10	D30	D60	D80	D90
0.5	6.0398	8.9241	13.5365	21.6914	47.0116
1.5	6.293	9.2036	13.6558	19.8862	31.9881
2.5	6.7837	9.5996	13.8593	19.8046	33.7352
3.5	5.9503	9.0559	13.9200	21.4981	37.1379
4.25	6.9058	10.0195	14.9150	23.6227	49.4656
5.25	6.3545	10.0198	17.3737	46.6449	94.5051
6.25	6.3342	9.6940	15.4176	27.0053	49.0957
7.25	6.697	10.1577	16.6302	34.5115	66.3918
8.25	6.3043	9.4370	14.7683	27.3497	66.3653
9.25	6.3455	9.5216	14.7453	24.5299	50.4647
10.25	6.0575	9.1161	14.1182	23.0903	48.8348
11.25	6.2293	9.1534	13.8018	21.8333	49.3687
12.25	6.1622	9.1778	13.7514	19.8220	29.6751
13.25	6.3389	9.6645	15.6154	32.6876	76.2831
14.25	5.7851	9.2428	16.2512	40.8407	78.5642
15.25	5.5494	8.8064	14.5285	26.8857	50.6385
16.25	6.3674	9.6068	15.5166	36.3806	84.9252
17.25	6.1869	9.5731	15.3766	27.4521	55.2402
Mean	6.26	9.44	14.88	27.53	55.54
STD Dev.1	0.33	0.39	1.11	7.75	18.63
STD Dev.2	0.66	0.77	2.23	15.50	37.26
Min	5.55	8.81	13.54	19.80	29.68
Max	6.91	10.16	17.37	46.64	94.51

		Lacul Caltun; Core LcT 2			
	LCt 2	LCt 2	LCt 2	LCt 2	LCt 2
Depth	D10	D30	D60	D80	D90
0.5	6.7934	10.1135	15.6229	26.3885	50.189
1.5	8.2602	11.7352	17.3303	26.4755	42.2779
2.5	9.2764	13.4189	20.9183	32.6608	48.0436
3.5	7.7869	11.4373	18.5212	36.8655	70.3327
4.5	10.1941	14.9686	25.8032	44.4465	69.6397
5.25	8.4824	12.2791	19.5402	38.1325	82.5108
6.25	6.7745	10.1459	18.2307	69.9288	116.3769
7.25	9.2564	13.2614	21.5861	45.1842	91.7658
8.25	10.0566	14.5889	24.7049	47.0580	85.5323
9.25	6.6701	10.0985	17.5787	54.0300	96.9649
10.25	10.4445	15.2268	26.0260	47.3059	80.3268
11.25	9.4565	13.7405	22.7278	45.1315	84.508
12.25	6.8132	10.2349	17.4384	49.0843	87.1471
13.25	8.4305	12.3343	20.3708	43.5579	81.9106
14.25	7.9113	11.5509	19.2445	49.9950	93.313
15.25	6.9988	10.5310	18.0375	49.3242	91.5581
16.25	9.3055	13.2862	21.3174	43.1612	84.9021
17.25	9.3235	13.3017	20.9503	38.0438	70.991
18.25	7.1325	10.8815	19.8890	55.5850	91.6852
19.25	7.8392	11.2860	18.0063	42.2386	86.7377

20.25	7.8491	11.2882	18.0455	42.2999	83.8888
21.25	7.1625	10.7044	18.3990	46.7929	80.095
22.25	8.0251	11.8442	20.7298	50.3476	82.4453
23.25	9.5444	13.4727	21.0352	36.3353	61.8115
24.25	7.8787	11.9481	20.6597	42.4413	69.274
25.25	9.2218	13.1531	21.1654	40.0701	68.4004
26.25	8.7423	12.4374	19.6126	37.3656	69.8811
27.25	7.1145	10.4415	16.9180	44.8185	87.1928
28.25	9.8314	13.9226	21.5805	34.3208	54.5192
29.25	8.6722	12.2562	18.7677	32.7592	64.3997
30.25	6.8314	10.3941	18.2313	52.7217	90.2015
Mean	8.33	12.14	19.97	43.38	78.03
STD Dev.1	1.16	1.51	2.48	8.85	15.94
STD Dev.2	2.31	3.02	4.97	17.69	31.88
Min	6.67	10.10	15.62	26.39	42.28
Max	10.44	15.23	26.03	69.93	116.38

	Lacul Podragu; Core LPm2				
Depth	LPm 2	LPm 2	LPm 2	LPm 2	LPm 2
0.5	D10	D30	D60	D80	D90
1.5	8.043	13.1492	27.6708	60.3043	91.2833
2.5	8.3162	13.1793	28.9967	69.785	104.6995
3.5	8.9371	14.6901	41.8762	88.3596	128.5086
4.5	8.096	13.1905	29.6911	68.8488	105.9866
5.25	8.7701	13.684	32.5938	84.1638	133.2723
6.25	9.3719	15.0766	35.6531	75.2635	113.4404
7.25	9.6448	15.9514	36.9281	69.7358	99.6653
8.25	10.5823	18.1625	47.0094	82.0538	113.4806
9.25	10.3504	17.4501	45.3797	80.9521	113.1803
10.25	10.0731	17.7787	47.1784	85.7846	123.4701
11.25	9.8335	16.2957	48.2647	93.348	135.9865
12.25	9.738	15.9065	41.2665	80.0249	113.7852
13.25	11.2561	22.7906	62.9475	98.6012	132.1838
14.25	11.217	21.2446	58.8697	98.0123	136.0507
15.25	9.0636	14.3276	35.5551	76.1799	111.6504
16.25	9.6809	16.1873	39.4264	72.9369	104.2347
17.25	11.3088	21.2537	56.2403	92.7617	127.6916
18.25	10.44	19.2041	49.987	83.7627	115.0445
19.25	10.1355	19.9065	56.5208	96.0175	135.4837
Mean	9.73	16.81	43.27	81.94	117.85
STD Dev.1	1.02	2.97	10.55	10.92	13.57
STD Dev.2	2.03	5.94	21.09	21.83	27.14
Min	8.04	13.15	27.67	60.30	91.28
Max	11.31	22.79	62.95	98.60	136.05

Fagaras Mountains		**2007**		
CORE NAME:LACUL BALEA LBa4				

Depth	Lba 4	Lba 4	Lba 4	Lba 4
	Median	Mean	Mode	Std. Dev.
0.5	16.6368	33.369	12.43	39.89
1.5	16.4741	39.6124	12.23	51.23
2.5	17.0237	46.1657	12.39	60.32
3.5	13.8693	20.0887	12.40	19.32
4.25	14.659	23.6942	12.39	23.84
5.25	14.8434	24.4108	12.44	27.27
6.25	13.478	19.8615	12.38	21.60
7.25	14.9109	24.5873	12.42	27.91
8.25	13.8454	20.7304	12.42	25.57
9.25	14.9919	24.3485	12.46	28.09
10.25	15.8544	31.2816	12.41	38.40
11.25	13.8631	25.8408	12.36	35.98
12.25	13.9412	25.5003	12.36	33.80
13.25	14.1738	24.0588	12.37	29.01
14.25	15.3564	36.6475	12.38	49.10
15.25	16.4386	35.6319	12.40	42.18
16.25	31.2731	65.5329	12.42	71.79
17.25	24.8059	59.6638	12.39	66.23
18.25	91.4039	96.6245	141.91	74.24
19.25	18.7909	47.9098	12.44	58.79
20.25	16.5943	33.8183	12.45	40.58
21.25	14.7501	28.8717	12.38	37.09
22.25	20.0712	45.5943	12.41	50.86
23.25	76.7903	101.9032	142.48	95.87
24.25	26.9038	59.0539	12.44	61.41
25.25	21.374	50.4408	12.42	57.49
26.25	37.0032	63.3117	12.40	63.25
27.25	102.6235	103.7164	162.72	80.46
28.25	21.1636	47.9703	12.40	52.52
30.25	16.6299	30.2157	12.47	34.73

CORE NAME: Lacul Capra 2				
Depth	LCp 2	LCp 2	LCp 2	LCp 2
	Median	Mean	Mode	Std. Dev.
0.5	11.7422	21.4848	10.8231	30.0134
1.5	11.9685	18.0498	10.8695	21.8686
2.5	12.2473	20.4969	12.2785	29.7682
3.5	12.0284	19.6154	10.8588	26.1206
4.25	12.9861	23.9038	12.3201	34.0930
5.25	14.1049	33.7361	12.2984	47.0069
6.25	13.0943	21.3380	12.3110	22.9002
7.25	13.8489	26.9512	12.3123	34.6089
8.25	12.6149	26.2946	10.8586	38.0540
9.25	12.6767	23.7089	12.2909	34.0049
10.25	12.1507	22.7850	10.8494	32.9742
11.25	11.9962	24.3380	10.8472	39.4565
12.25	12.027	16.6251	12.2916	16.5418
13.25	13.1161	28.3495	10.8705	39.6523

14.25	13.1541	28.5393	10.8203	36.9950
15.25	12.2022	24.4952	10.8191	34.8485
16.25	13.0123	29.7951	10.8539	41.6441
17.25	13.0251	24.1396	12.3088	32.3630

		Lacul Caltun; Core LcT 2		
	LCt 2	LCt 2	LCt 2	LCt 2
Depth	Median	Mean	Mode	Std. Dev.
0.5	13.3897	22.9737	12.3368	28.6943
1.5	15.089	22.6619	14.0990	25.2704
2.5	17.8189	25.2213	14.2554	23.9963
3.5	15.3929	29.6736	12.4109	38.0736
4.5	21.0764	33.7664	16.1908	36.3158
5.25	16.4154	33.4354	14.1011	45.4341
6.25	14.1881	39.4315	10.8359	51.1371
7.25	17.8382	36.4276	14.1700	47.2146
8.25	20.1942	37.1438	16.1650	45.0461
9.25	14.0703	34.1507	10.8523	44.2533
10.25	21.2787	36.9518	16.2057	43.0755
11.25	18.767	35.3909	14.2244	43.3499
12.25	14.1269	31.9903	10.8710	40.6626
13.25	16.7645	32.6149	14.0842	39.5544
14.25	15.6643	34.9739	12.3876	45.4759
15.25	14.553	33.7126	12.2948	44.7463
16.25	17.7468	34.5692	14.1836	43.4724
17.25	17.6382	31.1942	14.2057	37.0229
18.25	15.4367	34.3225	12.3077	41.3275
19.25	15.0005	32.1332	12.3780	41.5604
20.25	15.0066	31.7627	12.3742	40.8207
21.25	14.7671	30.9045	12.3005	37.5291
22.25	16.4354	32.8731	12.3969	38.0754
23.25	17.7852	29.3411	14.2198	33.1878
24.25	16.6789	29.6496	12.4430	33.1938
25.25	17.5888	30.2817	14.1621	33.6683
26.25	16.5163	30.2174	14.1022	36.9246
27.25	14.0401	31.5580	12.2834	41.2047
28.25	18.4071	28.1177	16.1580	30.4069
29.25	16.0324	28.5595	14.1058	35.3521
30.25	14.5206	32.6838	10.8832	40.0366

	Lacul Podragu; Core LPm2			
	LPm 2	LPm 2	LPm 2	LPm 2
	Median	Mean	Mode	Std. Dev.
0.5	20.3204	37.5996	14.1292	40.0865
1.5	20.3948	41.3454	12.4614	44.6809
2.5	26.3636	51.3015	14.1146	53.6013
3.5	20.8701	41.7086	14.0999	45.5504
4.5	21.7418	48.6562	14.0885	57.5644
5.25	24.8761	46.3096	14.1862	48.96
6.25	26.6531	43.5905	14.2379	42.3261
7.25	34.0205	50.2512	14.247	45.1184
8.25	32.3486	49.5126	14.2282	45.619
9.25	33.4912	52.8471	16.1843	51.0917

10.25	31.7602	55.7464	14.1512	56.9602
11.25	28.1174	47.9065	14.1873	46.5311
12.25	48.4466	61.5624	82.5453	51.8904
13.25	43.9363	60.4139	72.4426	53.5272
14.25	23.7304	45.6716	14.1121	48.1755
15.25	28.1434	45.4507	14.2202	43.9738
16.25	42.3468	57.8209	72.3058	50.7895
17.25	37.1817	51.9693	63.2119	45.8424
18.25	41.7814	59.0967	72.3164	54.327

Appendix 7: Particle size characteristics tables of lake sediments in the Rodna/Maramures region

Romanina Lakes Project				
Rodna Mountains		**2006**		
CORE NAME:LACUL BILA LB1				
Depth	Median	Mean	Mode	Std. Dev.
0.5	15.1695	22.6724	14.1138	27.08
1.5	15.6718	24.7705	12.4653	24.7852
2.5	14.6081	21.7	12.4921	24.3804
3.5	14.0378	22.8999	12.4175	29.4303
4.5	16.0564	30.5609	14.0905	41.1764
5.5	13.3039	19.967	12.3838	25.3779
6.5	13.7175	18.6598	12.4599	21.3986
7.5	14.1221	18.8648	12.4607	16.2995
9.5	14.6454	24.5695	12.4578	31.5107
10.5	17.4492	32.2583	14.1369	38.1816
11.5	16.8143	26.5416	14.2347	31.643
12.5	16.097	26.6354	14.136	32.407
13.5	17.346	30.5771	14.1625	36.3157
14.5	15.5663	32.0592	14.0854	61.1233
15.5	15.6993	24.1019	14.1332	28.3451
16.5	16.7575	26.5132	14.1664	29.318
17.5	15.7952	25.4247	14.0968	29.7172
18.5	14.2774	23.2448	12.4436	30.1268
19.5	18.0011	29.2834	14.2134	32.4548

	Lacul Buhaiescu-3; Core LB-3 2			
Depth	Median	Mean	Mode	Std. Dev.
0.5	20.0876	35.4931	14.1704	37.0304
1.5	17.2044	29.3942	14.1574	33.2329
2.5	19.1627	34.5359	14.1736	37.7475
3.5	21.4293	38.9489	14.2046	41.2758
4.5	17.0198	27.2602	14.1965	30.0649

5.5	17.6064	29.8912	14.1869	32.6699
6.5	22.9834	42.6784	14.1881	45.7419
7.5	20.6819	42.6573	14.1412	48.4854
8.5	22.8122	41.7983	14.1584	43.04
9.5	27.0376	44.2566	14.2597	44.1664

		Lacul Stiol; Core LS 2		
Depth	Median	Mean	Mode	Std. Dev.
0.5	16.4637	30.8783	12.4862	36.8392
1.5	16.6579	32.6315	14.0942	42.3344
2.5	14.294	23.0293	12.427	28.7554
3.5	15.4245	23.5394	14.0844	25.9408
4.5	17.1096	26.9277	14.1777	28.8895
5.5	15.5618	21.3374	14.1529	21.7176
6.5	15.4008	23.2452	14.0988	25.4618
7.5	17.5375	26.7344	14.2226	27.6142
8.5	16.7381	23.264	14.2034	20.9738
9.5	17.6399	26.5277	14.234	27.0431

	Lacul Lala Mare; Core LLM 2			
Depth	LLM 2			
	Median	Mean	Mode	Std. Dev.
0.5	16.8472	27.6051	14.1506	30.9125
1.5	15.7951	25.7557	14.0873	29.665
2.5	15.1134	23.9104	12.4779	27.7898
3.5	17.2934	27.5621	14.1399	28.3771
4.5	14.8626	23.988	12.4457	28.3025
5.5	16.331	27.7018	14.0933	31.6107
6.5	17.7448	28.3187	14.1816	30.2029
7.5	17.0764	30.0857	14.1232	35.2111
8.5	16.5006	24.7229	14.1222	22.532
9.5	17.0965	29.2218	14.1519	33.4918

	Lacul Pietrosul; Core LP 1			
Depth	LP 1			
	Median	Mean	Mode	Std. Dev.
0.5	19.3123	31.2683	14.1252	29.352
1.5	19.2306	31.4171	14.2983	32.4399
2.5	23.9406	42.0354	14.2376	41.7641
3.5	33.4708	51.8758	16.1786	49.7354
4.5	23.9099	39.4768	14.2483	38.2867
5.5	20.6866	33.9578	14.2213	33.5896
6.5	24.3665	37.1079	16.2271	34.6332
7.5	22.5058	38.7686	14.2087	38.6288
8.5	26.5993	40.5721	16.2217	37.695

CORE NAME: Lacul Vinderel 3				

Depth	Median	Mean	Mode	Std. Dev.
0.5	9.3623	10.5215	9.4661	5.8228
1.5	9.4723	13.3314	10.7621	15.6657
2.5	8.6639	9.864	9.4004	5.7175
3.5	10.3959	12.007	10.7957	7.6589
4.5	10.8998	15.1513	10.8253	20.2934
5.5	11.1616	12.7285	10.8703	7.3659
6.5	10.2221	11.6035	10.7883	6.6571
7.5	10.7763	13.8685	10.8181	13.5055
8.5	11.4764	13.2933	12.2959	8.2101
9.5	11.2314	13.1137	10.872	8.4065
10.5	9.9781	15.3282	10.7613	21.075
11.5	10.1887	13.3751	10.7673	17.1971
12.5	10.2088	14.9276	10.7812	20.7266
13.5	10.1959	12.3958	10.7833	10.0911
14.5	9.5917	12.2078	10.767	10.9897

Romanina Lakes Project					
Rodna Mountains		**2006**			
CORE NAME:LACUL BILA LB1					
Depth	D10	D30	D60	D80	D90
0.5	9.0764	12.2026	17.0965	24.5341	37.9976
1.5	8.1185	11.8067	18.5426	32.5777	54.8736
2.5	8.5973	11.6932	16.5308	24.194	38.7021
3.5	8.0895	11.1375	15.9348	24.008	42.3934
4.5	8.208	12.0056	19.0749	35.0161	68.3475
5.5	7.8812	10.7172	14.8985	20.8166	31.767
6.5	8.3909	11.2091	15.1911	20.3237	28.0092
7.5	8.0271	11.1867	15.9684	22.5963	33.1461
9.5	8.3282	11.5517	16.7561	26.105	47.674
10.5	8.8656	12.8544	21.2586	42.9599	75.8236
11.5	9.8766	13.315	19.2188	29.2042	48.8036
12.5	9.3257	12.672	18.4892	29.4273	52.7705
13.5	9.4475	13.2013	20.6328	37.7209	66.9221
14.5	8.995	12.2572	17.8644	28.521	52.1782
15.5	9.2733	12.505	17.8215	26.5654	43.2705
16.5	9.1424	12.8372	19.5687	32.412	53.8069
17.5	8.8418	12.2495	18.3081	29.6786	50.4851
18.5	8.2888	11.3665	16.2012	24.1644	42.1644
19.5	9.4175	13.4683	21.4538	37.1894	61.1246
Mean	8.75	12.12	17.94	28.84	48.96
STD Dev.1	0.58	0.81	1.96	6.18	12.88
STD Dev.2	1.16	1.62	3.91	12.36	25.76
Min	7.88	10.72	14.90	20.32	28.01
Max	9.88	13.47	21.45	42.96	75.82

	Lacul Buhaiescu-3; Core LB-3 2				
Depth	D10	D30	D60	D80	D90
0.5	9.3745	13.9443	25.9558	54.0803	82.5066
1.5	8.4039	13.1217	20.3869	37.2788	64.9265
2.5	9.6629	13.838	23.9641	49.811	80.5601

3.5	9.936	14.6264	28.0799	59.4439	92.628
4.5	9.7083	13.2676	19.7308	32.7297	56.1011
5.5	9.7511	13.4596	20.8952	38.6816	67.0936
6.5	10.2336	15.069	31.6402	66.1494	100.5271
7.5	9.7547	14.1709	27.9303	66.96	107.903
8.5	9.7532	14.7172	31.8155	66.9568	100.0383
9.5	10.7217	16.6324	36.7249	67.7833	98.746
Mean	9.73	14.28	26.71	53.99	85.10
STD Dev.1	0.59	1.04	5.62	13.69	17.71
STD Dev.2	1.19	2.09	11.25	27.38	35.42
Min	8.40	13.12	19.73	32.73	56.10
Max	10.72	16.63	36.72	67.78	107.90

	Lacul Lala Mare; Core LLM 2				
	LLM 2				LLM 2
Depth	D10	D30	D60	D80	D90
0.5	8.4761	12.486	20.0566	35.0519	58.9213
1.5	8.4071	12.0295	18.5243	31.2903	53.4381
2.5	8.2683	11.7203	17.4616	27.9496	46.8826
3.5	8.5399	12.6438	20.9839	37.7279	59.3934
4.5	8.0564	11.4728	17.2193	28.1509	48.148
5.5	8.045	11.9987	19.6209	36.2103	61.2732
6.5	9.0568	13.1361	21.3152	37.4157	59.4828
7.5	8.0759	12.2942	20.9026	39.9456	67.8184
8.5	7.9457	12.0331	19.8077	34.6077	53.6529
9.5	8.3996	12.5306	20.5921	37.6859	65.2291
Mean	8.33	12.23	19.65	34.60	57.42
STD Dev.1	0.33	0.49	1.46	4.16	6.84
STD Dev.2	0.66	0.97	2.92	8.31	13.69
Min	7.95	11.47	17.22	27.95	46.88
Max	9.06	13.14	21.32	39.95	67.82

	Lacul Pietrosul; Core LP 1				
	LP 1				LP 1
Depth	D10	D30	**D60**	D80	D90
0.5	8.4147	13.1	25.0667	49.0971	71.3026
1.5	10.5251	14.5158	22.9096	42.081	68.8146
2.5	10.6373	15.6651	32.8049	66.7009	98.0247
3.5	11.434	18.6749	45.6714	81.1687	117.7139
4.5	10.4376	15.6647	31.8118	61.0398	89.1055
5.5	10.0728	14.5465	26.1448	50.2013	75.3433
6.5	10.8274	16.3678	31.0048	54.7402	79.058
7.5	10.3034	15.078	30.0879	60.2241	88.4251
8.5	11.2503	17.1612	34.5626	60.8014	87.154
Mean	10.43	15.64	31.12	58.45	86.10
STD Dev.1	0.87	1.63	6.68	11.38	15.17

STD Dev.2	1.75	3.26	13.36	22.77	30.33
Min	8.41	13.10	22.91	42.08	68.81
Max	11.43	18.67	45.67	81.17	117.71

		Lacul Stiol; Core LS 2			
Depth	D10	D30	**D60**	**D80**	D90
0.5	8.6669	12.3338	19.7309	40.4451	74.5617
1.5	9.0271	12.6362	19.7792	39.5102	77.1696
2.5	8.0103	11.2015	16.3614	25.246	43.4448
3.5	8.6485	12.0262	17.7742	27.9496	45.2429
4.5	9.298	13.0602	20.0912	33.7981	55.1402
5.5	9.1535	12.4075	17.5782	25.0877	36.0204
6.5	8.3336	11.8784	17.8195	27.9278	44.459
7.5	9.7005	13.4704	20.5366	33.5409	53.2633
8.5	9.3438	12.9886	19.3169	29.3962	43.6126
9.5	9.7233	13.5292	20.6449	33.4236	52.2893
Mean	8.99	12.55	18.96	31.63	52.52
STD Dev.1	0.57	0.74	1.47	5.45	13.55
STD Dev.2	1.14	1.47	2.93	10.90	27.10
Min	8.01	11.20	16.36	25.09	36.02
Max	9.72	13.53	20.64	40.45	77.17

CORE NAME: Lacul Vinderel 3					
Depth	D10	D30	D60	D80	D90
0.5	4.8979	7.2343	10.558	13.8314	17.0235
1.5	3.498	6.5935	11.1819	16.6097	24.0876
2.5	4.4415	6.6443	9.8259	13.0552	16.3181
3.5	5.5749	8.108	11.6613	15.2812	19.2607
4.5	5.392	8.2303	12.454	17.3099	23.968
5.5	6.0349	8.7571	12.5185	16.4494	20.6137
6.5	5.4335	7.9476	11.4745	15.0205	18.8248
7.5	5.4623	8.2122	12.2552	16.8244	22.646
8.5	6.0843	8.9385	12.9067	17.128	21.8626
9.5	5.828	8.6613	12.6886	16.976	21.7842
10.5	4.2465	7.0822	11.7457	17.7887	27.3671
11.5	5.4288	7.9121	11.4637	15.2192	19.7392
12.5	4.661	7.4594	11.837	17.0324	24.4201
13.5	5.1811	7.7849	11.5545	15.595	20.2099
14.5	3.9547	6.8688	11.1843	16.0167	21.8222
Mean	5.07	7.76	11.69	16.01	21.33
STD Dev.1	0.77	0.76	0.82	1.34	2.95
STD Dev.2	1.54	1.51	1.65	2.68	5.89
Min	3.50	6.59	9.83	13.06	16.32
Max	6.08	8.94	12.91	17.79	27.37

Appendix 8: Magnetic measurements

X

X = (raw Xlf / mass) x 10
Units = 10^{-8} m^3 kg^{-1}

Xfd

Xfd = ((raw Xlf – raw Xhf) / raw Xlf) x 100
Units = %

ARM

ARM = raw ARM / mass
Units = 10^{-5} Am^2 kg^{-1}

SIRM

SIRM = raw SIRM / mass
Units = 10^{-5} Am^2 kg^{-1}

SIRM / X

SIRM / X = mass SIRM / mass Xlf
Unit = 10^{-3} A m^{-1}

ARM/ X

ARM / X = mass ARM / mass Xlf
Unit = 10^{-3} A m^{-1}

SIRM / ARM

SIRM / ARM = mass SIRM / mass ARM
Unit = none

Soft

Soft = (((raw SIRM - raw_20)/2) / mass)
Units = 10^{-5} Am^2 kg^{-1}

HIRM

HIRM = (((raw SIRM + raw_300)/2) / mass)
Units = 10^{-5} Am^2 kg^{-1}

(Hard (%)

Hard = 100 – (((((raw_300 x -1)/ raw SIRM) + 1) / 2) x 100)

Units = %)

Plot-able backfields (all as %)

-20 mT

((((raw_20 x -1) / raw SIRM) +1) / 2) x 100

-40 mT

((((raw_40 x -1) / raw SIRM) +1) / 2) x 100

-100 mT

((((raw_100 x -1) / raw SIRM) +1) / 2) x 100

-300 mT

((((raw_300 x -1)/ raw SIRM) + 1) / 2) x 100

Appendix 9: Mineral magnetic characteristics tables of lake sediments in the Fagaras region

CAPRA					DATE:21-05-2008			
CALCULATIONS								
			FREQUENCY DEPENDENT SUSCEPTIBILITY					
No	Description	Weight of Sample (g)	Raw Low Frequency K	Raw High Frequency K	Mass Spec Low Freq X=(rawXlf/mass)x10	Mass Spec High Freq X=(rawXlf/mass)x10	Xfd ={(K_{lf}-K_{hf})/K_{lf}} x100	Depth
1	LcP2:0-1	1.667	35.75	36.06	214.457	216.317	-0.867	0.5
2	LcP2:1-2	3.808	76.32	77.03	200.420	202.285	-0.930	1.5
3	LcP2:2-3	5.445	89.82	89.05	164.959	163.545	0.857	2.5
4	LcP2:3-4	7.386	84.6	83.25	114.541	112.713	1.596	3.5
5	LcP2:4-4.5	4.946	56.25	56.98	113.728	115.204	-1.298	4.25
6	LcP2:5-5.5	5.31	61.78	62.45	116.347	117.608	-1.084	5.25
7	LcP2:6-6.5	5.587	71.33	72.26	127.671	129.336	-1.304	6.25
8	LcP2:7-7.5	5.012	42.32	42.58	84.437	84.956	-0.614	7.25
9	LcP2:8-8.5	5.126	53.97	55.05	105.287	107.394	-2.001	8.25
10	LcP2:9-9.5	5.14	68.7	169.58	133.658	329.922	-146.841	9.25
11	LcP2:10-10.5	5.657	83.32	83.7	147.287	147.958	-0.456	10.25
12	LcP2:11-11.5	5.585	90.05	99.65	161.235	178.424	-10.661	11.25
13	LcP2:12-12.5	5.532	33.28	32.89	60.159	59.454	1.172	12.25
14	LcP2:13-13.5	6.938	54.3	52.81	78.265	76.117	2.744	13.25
15	LcP2:14-14.5	5.017	107.1	109.98	213.474	219.215	-2.689	14.25
16	LcP2:15-15.5	4.728	163.17	164.54	345.114	348.012	-0.840	15.25
17	LcP2:16-16.5	4.621	127.1	128.65	275.049	278.403	-1.220	16.25
18	LcP2:17-17.5	5.85	136.2	136.83	232.821	233.897	-0.463	17.25
BALEA					DATE: 22-05-2008			
CALCULATIONS								
			FREQUENCY DEPENDENT SUSCEPTIBILITY					
No	Description	Weight of Sample (g)	Raw Low Frequency K	Raw High Frequency K	Mass Spec Low Freq X=(rawXlf/mass)x10	Mass Spec High Freq X=(rawXlf/mass)x10	Xfd ={(K_{lf}-K_{hf})/K_{lf}} x100	Depth
65	LBa1:0-1	4.906	79.38	77.51	161.802	157.990	2.356	0.5
66	LBa1:1-2	6.204	107.41	104.24	173.130	168.021	2.951	1.5
67	LBa1:2-3	5.878	87.35	85.72	148.605	145.832	1.866	2.5
68	LBa1:3-4	6.812	82.52	80.89	121.139	118.746	1.975	3.5
69	LBa1:4-4.5	5.208	75.25	75.85	144.489	145.641	-0.797	4.25
70	LBa1:5-5.5	4.349	61.29	61.29	140.929	140.929	0.000	5.25
71	LBa1:6-6.5	7.992	116.12	114.6	145.295	143.393	1.309	6.25
72	LBa1:7-7.5	8.517	106.47	106.64	125.009	125.208	-0.160	7.25
73	LBa1:8-8.5	8.317	147.35	143.09	177.167	172.045	2.891	8.25
74	LBa1:9-9.50	9.053	140.96	138.8	155.705	153.319	1.532	9.25
75	LBa1:10-10.5	5.106	101.38	100.92	198.551	197.650	0.454	10.25
76	LBa1:11.5-12	5.77	130.65	128.2	226.430	222.184	1.875	11.25
77	LBa1:12-12.5	6.492	149.55	148.74	230.360	229.113	0.542	12.25
78	LBa1:13-	5.433	108.08	106.22	198.932	195.509	1.721	13.25

	13.5							
79	LBa1:14-14.5	6.173	111.06	109.98	179.913	178.163	0.972	14.25
80	LBa1:15-15.5	7.352	159.18	157.81	216.513	214.649	0.861	15.25
81	LBa1:160-16.5	4.337	91.42	93.21	210.791	214.918	-1.958	16.25
82	LBa1:17-17.5	5.896	184.32	185.3	312.619	314.281	-0.532	17.25
83	LBa1:18-18.5	5.204	94.15	97.44	180.919	187.241	-3.494	18.25
84	LBa1:19-19.5	6.709	155.26	155.26	231.420	231.420	0.000	19.25
85	LBa1:20-20.5	6.482	149.35	148.15	230.407	228.556	0.803	20.25
86	LBa1:21-21.5	7.798	154.2	151.75	197.743	194.601	1.589	21.25
87	LBa1:22-22.5	7.531	142.45	141.06	189.152	187.306	0.976	22.25
88	LBa1:23-23.5	6.7	124.56	121.38	185.910	181.164	2.553	23.25
89	LBa1:24-24.5	6.546	145.56	139.42	222.365	212.985	4.218	24.25
90	LBa1:25-25.5	6.958	135.99	136.47	195.444	196.134	-0.353	25.25
91	LBa1:26-26.5	9.072	246.14	245.84	271.318	270.988	0.122	26.25
92	LBa1:27-27.5	8.602	189.63	189.72	220.449	220.553	-0.047	27.25
93	LBa1:28-28.5	9.74	278.33	269.96	285.760	277.166	3.007	28.25
94	LBa1:30-30.5	5.629	124.43	121.61	221.052	216.042	2.266	30.25
CALTUN					DATE: 23-05-2008			
CALCULATIONS								
			FREQUENCY DEPENDENT SUSCEPTIBILITY					
No	**Description**	**Weight of Sample (g)**	**Raw Low Frequency K**	**Raw High Frequency K**	**Mass Spec Low Freq X=(rawXlf/mass)x10**	**Mass Spec High Freq X=(rawXlf/mass)x10**	**Xfd ={(K_{lf}-K_{hf})/K_{lf}} x100**	**Depth**
19	LcT2:1-2	5.286	13.92	10.63	26.334	20.110	23.635	1.5
20	LcT2:2-3	5.476	13.58	10.06	24.799	18.371	25.920	2.5
21	LcT2:3-4	5.02	12.92	9.15	25.727	18.227	29.152	3.5
22	LcT2:4-5	5.207	8.94	5.6	17.169	10.755	37.360	4.5
23	LcT2:5-5.5	2.54	3.00	2.23	11.811	8.780	25.667	5.25
24	LcT2:6-6.5	2.758	2.23	2.48	8.086	8.992	-11.211	6.25
25	LcT2:7-7.5	3.76	0.66	3.65	1.755	9.707	-453.030	7.25
26	LcT2:8-8.5	2.534	0.64	2.4	2.526	9.747	-285.938	8.25
27	LcT2:9-9.5	2.812	4.10	2.47	14.580	12.198	16.341	9.25
28	LcT2:10-10.5	3.474	1.41	3.43	4.059	9.557	-135.461	10.25
29	LcT2:11-11.5	3.38	5.94	3.32	17.574	9.142	47.980	11.25
30	LcT2:12-12.5	3.266	0.60	3.09	1.837	9.737	-430.000	12.25
31	LcT2:13-13.5	3.167	4.77	3.18	15.062	10.452	30.608	13.25
32	LcT2:14-14.5	3.243	2.30	3.31	7.092	10.176	-43.478	14.25
33	LcT2:15-15.5	3.336	0.25	3.3	0.749	9.562	-1176.000	15.25
34	LcT2:16-16.5	3.226	0.65	3.19	2.015	11.221	-456.923	16.25
35	LcT2:17-17.5	3.442	0.9	3.62	2.615	10.343	-295.556	17.25

36	LcT2:18-18.5	5.996	4.92	3.56	8.205	6.171	24.797	18.25
37	LcT2:19-19.5	3.352	0.2	3.7	0.597	13.992	-2245.000	19.25
38	LcT2:20-20.5	4.137	5.69	4.69	13.754	11.627	15.466	20.25
39	LcT2:21-21.5	3.951	0.46	4.81	1.164	15.819	-1258.696	21.25
40	LcT2:22-22.5	4.923	10.04	6.25	20.394	11.883	41.733	22.25
41	LcT2:23-23.5	4.521	2.31	5.85	5.109	20.239	-296.104	23.25
42	LcT2:24-24.5	6.205	9.31	9.15	15.004	14.279	4.834	24.25
43	LcT2:25-25.5	6.234	5.06	8.86	8.117	10.218	-25.889	25.25
44	LcT2:26-26.5	4.755	0.27	6.37	0.568	6.814	-1100.000	26.25
45	LcT2:27-27.5	3.586	4.12	3.24	11.489	9.760	15.049	27.25
46	LcT2:28-28.5	3.976	2.66	3.5	6.690	7.646	-14.286	28.25
47	LcT2:30-30.5	3.381	1.21	3.04	3.579	0.000	100.000	30.25
PODRAGU					DATE: 23-05-2008			
CALCULATIONS								
			FREQUENCY DEPENDENT SUSCEPTIBILITY					
No	Description	Weight of Sample (g)	Raw Low Frequency K	Raw High Frequency K	Mass Spec Low Freq X=(rawXlf/mass)x10	Mass Spec High Freq X=(rawXlf/mass)x10	Xfd $=\{(K_{lf}-K_{hf})/K_{lf}\}$ x100	Depth
48	LPm2:1-2	7.198	53.96	52.97	74.965	73.590	1.835	1.5
49	LPm2:2-3	7.198	50.47	50.33	70.117	69.922	0.277	2.5
50	LPm2:3-4	6.7	45.44	44.84	67.821	66.925	1.320	3.5
51	LPm2:4-5	6.758	48.22	47.7	71.352	70.583	1.078	4.5
52	LPm2:5-5.5	5.761	47.43	47	82.329	81.583	0.907	5.25
53	LPm2:6-6.5	8.301	67.30	65.46	81.075	78.858	2.734	6.25
54	LPm2:7-7.5	8.431	64.47	63.51	76.468	75.329	1.489	7.25
55	LPm2:8-8.5	7.533	55.40	55.47	73.543	73.636	-0.126	8.25
56	LPm2:9-9.5	8.615	64.58	63.34	74.962	73.523	1.920	9.25
57	LPm2:10-10.5	6.547	48.27	48.1	73.728	73.469	0.352	10.25
58	LPm2:11-11.5	7.572	48.11	48.53	63.537	64.091	-0.873	11.25
59	LPm2:12-12.5	7.689	52.43	51.5	68.188	66.979	1.774	12.25
60	LPm2:13-13.5	8.559	52.42	52.36	61.245	61.175	0.114	13.25
61	LPm2:14-14.5	5.266	29.28	29.38	55.602	55.792	-0.342	14.25
62	LPm2:15-15.5	9.098	44.17	43.72	48.549	48.055	1.019	15.25
63	LPm2:16-16.5	7.013	37.13	37.05	52.945	52.830	0.215	16.25
64	LPm2:17-17.5	6.183	34.35	33.56	55.556	54.278	2.300	17.25

Appendix 10: Geochemical analysis of the lake sediments in the Fagaras region

Romanina Lakes Project									
Fagaras Mountains		2006							
CORE NAME:LACUL BALEA LBa1									
Depth	**Co**	**Cr**	**Cu**	**Mn**	**Ni**	**Pb**	**Zn**	**Fe**	**Al**
0.5	29.9158	172.1984	91.0923	1465.4470	144.3348	261.3089	375.1508	58007.5781	28906.4605
1.5	29.4688	163.8859	74.7370	1527.0262	138.6234	240.8662	348.2855	52686.4259	27573.7343
2.5	31.0341	167.6501	84.2971	1418.2467	138.5476	277.8469	400.9987	56656.5452	28424.0891
3.5	37.6337	190.9496	107.5270	1621.5887	152.0306	244.8112	347.0779	66763.1894	34284.0528
4.25	31.3132	185.8197	98.7902	1329.9349	144.7031	196.3700	252.2625	59419.3693	30866.7007
5.25	31.7739	175.1697	92.0153	1477.6654	137.9810	250.9130	348.0037	62346.8137	31665.0123
6.25	29.9493	175.6569	84.0054	1184.1169	130.7202	145.3783	185.6331	53353.2220	28327.4463
7.25	36.1777	202.5607	102.7185	1331.4061	150.2162	162.8434	183.9015	62309.2044	33691.7784
8.25	25.1942	165.6129	67.7329	921.0612	119.8203	108.9418	135.3310	48337.7299	26415.1137
9.25	33.0475	182.1514	82.2711	1144.7007	140.8122	170.0563	214.2500	57472.7700	30964.6127
10.25	35.6651	191.8126	85.6428	1412.7472	156.8783	235.2092	315.6561	61897.0092	33458.0921
11.25	35.1865	195.4688	88.3806	1295.0724	165.8155	248.1063	345.8720	61845.8517	32642.3049
12.25	34.3840	190.1915	88.0212	1256.1950	159.1809	242.5850	344.9734	60653.6326	32526.3188
13.25	38.0080	197.4332	93.7813	1520.4963	163.7047	268.5643	388.7004	65379.9020	35072.9167
14.25	16.9839	111.1472	48.4498	951.8869	94.2870	149.3724	229.9692	37832.9656	19949.9536
15.25	30.2228	127.3565	73.5969	1229.4794	98.5109	163.8263	226.9661	54300.0000	27873.5472
16.25	34.4869	192.3988	78.6003	1460.6357	163.6162	224.0619	312.4361	55935.2509	30134.7656
17.25	36.8383	201.6137	81.0865	1408.2017	178.9991	200.2196	282.9776	55464.9133	29877.5578
18.25	38.4859	199.8171	90.0709	2152.5860	166.6128	228.8978	321.0265	62565.4070	34398.6192
19.25	35.8624	211.1910	74.5742	1230.9780	180.7484	166.7308	237.9507	56583.0544	30814.7675
20.25	43.5455	232.6877	96.3236	1779.7444	205.1110	196.0100	260.6752	65086.6584	35436.2219
21.25	46.5191	233.1028	117.1669	1950.5234	209.0209	194.5357	258.3510	66748.7685	37063.6084
22.25	43.8187	232.2894	102.4348	1596.8548	200.2949	203.3628	263.0107	65688.4811	34988.2849
23.25	42.0258	224.1205	103.2579	1842.9076	194.4378	207.0021	244.0267	63927.7443	33159.7383
24.25	41.7678	228.8390	105.0068	1576.0133	197.1354	204.4593	253.0227	63854.4241	33835.3930
25.25	46.3821	267.4645	111.3503	1965.0922	237.5523	201.5996	270.1464	64171.8594	37035.0199
26.25	45.9048	263.5637	106.9100	1898.1978	234.3631	179.2042	238.6696	66340.7209	35760.0463
27.25	47.7531	256.2257	110.8529	1854.5104	233.5627	212.1817	264.1726	68073.7316	35893.8478
28.25	52.9040	241.5275	101.1722	1936.0788	214.4869	189.8405	245.1818	62168.1961	33871.4749
30.25	54.7045	249.5404	116.0483	1949.9420	215.6519	158.8700	217.1193	63868.7993	33911.9147

Romanina Lakes Project									
Rodna Mountains		2006							
CORE NAME:LACUL BALEA LBa4									
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	30.1614	172.3259	91.0571	3686.3888	143.7716	278.7948	308.5405	76895.1791	28359.8518
1.5	31.2022	190.6161	83.0866	2278.2338	155.5871	227.3195	288.1505	59403.7387	29712.9888
2.5	29.9136	187.5254	69.6195	1848.3916	152.3192	200.5073	252.7850	57510.7945	28775.313
3.5	34.0502	192.5214	98.3492	2032.0045	152.9736	228.6906	280.5426	62260.8091	32089.2016
4.5	32.0704	182.8460	100.5278	2064.4856	145.8546	247.3038	305.3605	62210.2880	29353.8086
5.5	31.8184	188.4514	95.9728	1678.6699	152.1357	213.9738	282.9268	61521.9017	31495.0321
6.5	34.8973	180.0173	100.9236	1967.3368	145.6955	214.5023	279.9492	61376.9996	30906.2365
7.5	26.9478	182.4775	91.1751	1228.9730	127.1974	139.7718	145.1665	60368.5092	32211.8405
8.5	31.3225	197.9455	104.7642	1424.5205	144.8227	147.6054	171.8678	60022.3015	33068.8559
9.5	31.3602	162.5504	86.3503	1200.2261	112.2849	117.1063	128.3888	52292.8455	27423.0635
10.5	40.2852	195.6708	93.6479	1591.4847	154.6508	233.4601	314.1684	60636.7371	31833.0986
11.5	41.6229	189.3265	100.6141	1808.0588	153.5365	258.2635	375.4618	59903.5294	32641.0588
12.5	30.1149	188.8502	91.2571	1384.3410	151.9951	237.7854	313.4641	61591.5033	32036.9826
13.5	44.2204	204.0551	106.2600	1786.7859	166.1066	219.4204	311.2748	70231.3503	36723.7370
14.5	37.1101	185.5583	74.3029	1409.3312	146.8699	194.7181	265.4231	55545.0266	29766.4172
15.5	36.8297	182.8284	73.1703	1488.1816	153.0803	176.3324	249.8323	51385.3722	27641.8919
17.5	27.5338	193.9769	67.2654	1224.6957	161.7486	147.4546	195.3082	51315.0177	27411.1332
17.5	42.3293	206.6711	77.6994	1641.1296	180.0473	150.1318	199.9395	58323.5506	30986.6435
18.5	41.0703	212.5068	68.9568	1434.8412	182.8151	118.3173	162.1320	54824.7169	29071.3934
19.5	41.8038	197.7224	86.7198	1541.9659	167.5904	137.2990	189.3431	57974.3976	31201.1701
20.5	32.5637	211.5480	84.4535	1561.4903	175.3934	173.9772	208.8257	56651.6681	30935.7989
21.5	47.4220	228.2141	101.7049	1753.2705	192.1711	185.7177	243.7632	64091.4546	33747.1795
22.5	48.6563	244.9727	96.7471	1966.9004	212.5974	168.3826	224.9088	63186.4449	34147.7781
23.5	45.5764	212.9549	85.2030	1443.3748	185.1879	111.1953	148.6098	53192.1824	29298.8018
24.5	20.0583	177.3723	44.8906	827.6100	145.9812	84.1143	103.7805	42248.5562	23505.8307
25.5	37.0983	198.9291	71.2273	1319.9081	171.6780	119.5809	151.2852	51302.2326	26366.5831
26.5	49.1338	246.3311	98.5103	1991.8341	216.3002	161.2730	218.6447	63047.8208	33903.3898
27.5	28.8261	150.7735	40.9834	753.7243	131.0040	82.1384	109.5332	41296.4531	21891.9908
28.5	44.7064	212.4917	87.8948	1544.9202	189.5027	182.5989	231.0761	61179.7284	31451.5245
29.5	32.9985	227.0581	97.9902	1786.2813	193.7368	164.5268	196.8418	60955.7532	32268.5551
30.5	42.1361	239.6398	116.0657	1924.7729	208.2626	153.9174	170.3253	68733.1010	35930.2915

	Lacul Caltun; Core LCt 2								
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	21.9785	37.8122	79.9915	177.4072	36.9983	163.2766	163.7189	31854.6946	27707.6357
1.5	22.9072	38.9989	74.1003	187.4751	40.7741	152.7157	159.3836	30038.4068	28495.7918
2.5	24.1755	42.0950	77.7626	201.2240	45.8577	162.3479	179.3367	30202.2574	30420.2076
3.5	22.1705	38.8087	80.8756	181.0934	39.4009	151.2874	167.8994	28201.0110	27995.1947
4.25	23.9871	37.7352	51.7005	202.8727	40.0467	129.5458	132.8889	29685.2290	25910.9632
5.25	24.0017	36.3271	46.1070	193.7437	40.5111	130.5349	114.7623	28306.3657	25412.2266
6.25	28.1793	35.1419	45.1139	178.1632	43.3760	121.9518	129.7195	28514.6174	26001.5047
7.25	23.5699	36.8270	42.5754	208.1977	41.2573	100.0327	116.3714	29100.7871	24865.1281
8.25	22.2882	34.9782	48.8504	196.8599	42.1122	87.3627	113.9908	26935.6244	24277.4685
9.25	26.9159	36.9652	45.9553	202.1657	42.0347	84.9930	127.8387	29193.7896	26049.3490
10.25	25.4193	34.5081	47.1919	184.3085	41.6159	79.2525	119.0319	26627.6261	24165.5287
11.25	25.1938	39.3177	53.9417	220.4954	44.2447	80.0785	123.6122	29870.8379	27130.7314
12.25	25.0177	37.1403	50.3990	199.5239	43.3052	79.0784	121.7004	27650.6101	25125.0000
13.25	24.4558	40.6060	55.1385	228.3351	44.7998	79.3652	127.6103	30501.4443	28320.1812
14.25	23.3285	37.8378	50.2893	217.8251	41.3628	76.6388	118.9993	28820.9000	25783.4182
15.25	23.9638	40.4687	53.9626	232.3434	43.9983	76.1380	123.1723	30324.2020	27598.0824
16.25	25.3382	38.9209	52.4475	224.2378	43.3019	81.2571	123.7259	29712.7056	26373.2984
17.25	24.9221	40.2591	55.6588	233.0322	45.0839	79.2944	124.6600	30554.2579	27250.4866
18.25	19.7194	24.8306	32.8819	143.5899	29.3998	47.7438	89.0377	20342.9705	17451.9558
19.25	23.1471	38.3445	51.2307	232.6390	42.5284	73.0813	116.5404	29881.4088	25975.4991
20.25	23.5473	38.2995	54.8605	236.5051	42.8007	73.6031	118.7574	30237.3576	25650.8542
21.25	25.1033	42.8900	57.0456	264.2694	45.6209	79.8149	121.6388	33142.2934	27402.3909
22.25	25.6451	40.6475	60.4058	273.7231	46.5343	79.7881	122.7475	33320.5862	27132.0061
23.25	27.6444	46.5226	74.1270	315.8585	51.8677	90.7657	132.8213	37920.9397	30818.0394
24.25	27.8775	48.2216	78.1269	341.4532	53.7394	96.2929	134.2684	39999.4432	31532.6281
25.25	29.5443	49.8034	80.3956	361.1159	55.9199	98.9686	143.0383	41765.1886	32787.5429
26.25	26.8292	44.2535	70.4156	302.2272	48.8159	92.1521	132.7808	36288.0249	29748.3521
27.25	24.0803	41.5288	50.5655	223.8146	43.7865	89.7469	128.0134	30370.1532	29332.0369
28.25	23.1086	39.2097	50.2013	205.6324	41.8414	90.7545	115.9558	28165.6741	27979.3949
30.25	22.4926	39.8406	53.7334	208.1776	41.5808	89.9589	125.5216	28461.8373	28161.5059
31.25	23.2638	38.4283	56.2875	210.4439	40.8024	85.6011	126.3912	28299.4843	26051.6331

Romanina Lakes Project									
Fagaras Mountains		2007							
CORE NAME: Lacul Capra 2									
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	23.3622	112.4883	76.9070	445.9544	103.0556	251.9825	240.5543	42573.6234	21314.5280
1.5	26.1785	113.0384	78.6674	505.1791	100.5174	203.3163	214.2279	42777.4419	22157.3837
2.5	28.4330	125.3695	90.9948	566.9738	103.7978	135.8211	154.8316	46921.2704	25929.3124
3.5	29.6187	129.6102	99.3688	610.4129	102.1717	112.421	133.4104	46609.5442	27759.4765
4.25	28.4100	138.8007	102.6935	636.14	106.2179	99.86029	122.4963	48746.8472	29727.4357
5.25	25.3381	128.2003	91.5986	569.0215	100.5608	87.31799	109.8100	43487.0434	25650.4812
6.25	28.9611	138.6273	103.2440	617.3918	101.6374	96.6909	111.0662	47509.3648	28000.0000
7.25	28.6023	136.2871	105.7522	600.6729	103.0919	94.96635	113.7882	47880.0753	27998.0041
8.25	27.6621	131.5660	101.4193	576.9146	101.0729	93.28187	109.7466	45581.5073	26889.3316
9.25	29.2548	173.5024	67.5546	523.1169	121.5869	81.96283	114.3891	48199.3405	28408.0935
10.25	30.6942	155.3079	89.3694	613.0587	116.0289	100.9481	109.8437	50470.7561	26835.7109
11.25	28.9377	164.6240	89.8274	654.6606	117.0694	104.0458	110.3731	50313.1683	29001.1041
12.25	27.4524	186.5660	79.7973	644.5606	110.7683	93.29065	98.9515	47645.5592	29703.4187
13.25	26.8771	176.9825	77.6878	621.2965	113.8212	93.79298	102.8545	47006.5356	29100.0353
14.25	28.5539	134.6328	72.4712	540.5764	115.7124	101.7475	114.9117	45029.9499	25919.3609
15.25	25.6366	119.1212	59.8069	483.5442	106.4695	92.85415	104.3298	40004.0821	22775.1545
16.25	28.0330	126.7472	63.3882	540.3078	112.9122	100.7698	113.3967	42808.7664	24773.9194
17.25	26.4331	128.4414	64.2127	549.3194	110.4861	95.36798	113.3501	42781.2643	24792.2752

Romanina Lakes Project									
Fagaras Mountains		**2007**							
CORE NAME: Lacul Capral 3									
Depth	**Co**	**Cr**	**Cu**	**Mn**	**Ni**	**Pb**	**Zn**	**Fe**	**Al**
0.5	41.9638	96.6468	67.4382	719.5847	71.8496	243.8646	183.3897	53553.9941	18532.9142
1.5	37.3322	104.8424	71.4300	419.0541	79.0616	233.5290	196.5536	49992.8632	20458.0848
2.5	37.0486	113.5070	80.8544	439.3928	85.4163	246.0937	219.7613	47760.1021	21866.6525
3.5	43.2787	120.2968	91.0214	448.8875	86.8711	262.0997	229.0910	49571.7241	23666.8716
4.5	41.9665	116.3431	85.0086	401.0987	85.4043	224.7545	193.5815	46104.8127	23245.3165
5.5	43.5458	119.6021	85.4526	422.4263	88.0621	200.3184	175.9521	48231.3158	24718.6842
6.5	45.6142	125.3629	90.5977	458.4030	89.3818	149.9140	147.3900	49782.7598	25315.5196
7.5	45.3035	127.0469	96.7604	527.2671	87.0536	130.9387	121.6795	49976.2307	26090.0097
8.5	44.0431	125.2258	99.4768	489.4046	85.6166	117.5765	113.4716	49106.8172	25930.5797
9.5	41.1717	138.7712	110.3709	557.5862	90.6156	119.1003	122.7829	53096.4060	29193.7227
10.5	42.7621	140.8103	106.7890	552.0156	93.7874	107.2893	114.4170	51909.8481	30128.3072
11.5	40.7120	137.1521	109.9403	562.8612	90.5259	89.5327	98.5679	48905.9021	29200.6609
12.5	46.1410	132.9556	101.6550	513.0023	87.8126	88.0385	85.6842	47239.9310	28061.749
13.5	37.9089	130.8071	108.3954	535.1376	85.6032	89.0558	94.7321	45998.8106	27342.0933
14.5	38.9167	135.7833	104.9317	562.3833	91.8256	89.3050	95.6022	48102.0556	28786.5556
15.5	40.0614	142.5041	97.6878	543.7009	93.0025	80.9932	91.1956	48172.5550	28767.8989
16.5	23.2949	49.9052	34.1393	190.0579	28.5962	30.8296	26.1757	17127.6865	9792.35145
17.5	40.5929	140.4418	108.9718	562.7319	90.7071	85.5102	91.7888	48902.9496	28668.0542
18.5	39.6670	135.8953	103.5096	525.8748	89.6107	87.2971	92.4003	48479.7725	28794.2073
19.5	37.1702	135.9208	129.5861	615.5495	76.1849	91.2014	99.4363	50147.6196	34847.3472
20.5	45.2912	133.4057	104.1227	504.5874	93.4299	94.5253	89.5232	47329.6888	27646.762
21.5	40.8819	135.2737	105.3858	543.6544	96.3332	91.6146	99.1859	48563.4733	28175.2596
22.5	35.9005	121.9326	91.3475	474.7856	83.2108	72.7367	84.1218	42405.7801	24339.5794
23.5	46.5744	145.8653	112.3539	586.1481	100.1129	93.4749	101.8801	52155.4857	29826.4266
24.5	45.1886	129.8782	101.7579	499.9430	86.7334	85.4374	79.2903	45778.2713	26574.6237
25.5	42.4418	144.4053	112.4094	578.8688	98.6287	85.5763	96.3039	51406.6022	29937.5306
26.5	41.1258	137.8022	108.3096	558.9049	95.1531	87.5850	92.9779	49247.3417	28727.5196
27.5	42.6099	143.7300	113.1153	583.6782	98.5182	86.6148	95.8477	51473.9505	29751.6867
28.5	45.2357	136.7053	104.0287	515.2472	92.2533	88.1639	81.7660	47407.1744	28054.8565
29.5	42.2742	148.6226	112.7942	581.7636	101.3343	91.7164	97.9481	52502.3585	31206.8986
30.5	42.1255	142.7118	111.0316	573.2063	98.3635	82.7232	95.6139	49797.5276	29572.1315
31.5	47.01945	143.9852	104.6653	552.0907	94.71676	95.9187	87.18736	49271.7662	29476.3116

Lacul Podragu; Core LPm2									
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	17.2626	78.4612	74.8030	734.3040	50.5927	170.2102	148.2800	47924.9638	20176.95747
1.5	20.8893	84.5192	77.0204	544.7896	55.1119	129.9441	149.7063	39986.5385	22066.55012
2.5	22.1903	86.0984	75.8197	550.2187	55.3299	125.6794	147.1885	40982.5844	22273.41893
3.5	21.6482	86.9542	70.6634	559.2185	57.0629	112.5384	140.4171	41364.6030	22301.98488
4.25	25.9897	94.9557	79.5098	656.8908	60.0891	131.2839	160.5126	44242.5862	24940.97701
5.25	20.8041	70.3120	88.7656	505.2646	46.7055	97.5642	123.7632	36653.4446	20256.72108
6.25	15.9909	59.9568	78.4337	397.3098	40.4692	78.7366	152.6097	31351.1905	17218.19561
7.25	20.5712	66.0771	84.8185	467.3664	44.3016	78.9535	108.6818	34896.7391	20413.1013
8.25	19.9432	60.9570	85.0886	445.3225	40.2785	70.0006	100.1198	34864.9733	19489.69474
9.25	17.2423	63.2132	76.0194	418.4850	41.8982	71.0070	93.3205	34626.8648	18780.40235
10.25	21.1644	81.4031	81.4128	505.0838	52.2765	91.5057	116.9928	39989.7352	22763.05879
11.25	21.2004	79.1442	73.6303	455.2255	52.1370	74.0334	232.3859	35755.2339	20361.46993
12.25	18.8438	68.1040	70.1114	418.8896	46.6632	62.5838	95.9643	34448.3672	20004.3579
13.25	22.3108	79.1228	74.4103	499.6780	52.1379	72.8366	98.1767	37533.1563	21719.89381
14.25	23.5000	93.3521	77.2986	566.2718	57.0513	75.0890	102.8597	41583.1332	24248.30786
15.25	21.1372	95.8956	71.4001	559.2408	59.1723	76.9835	101.4916	42203.8234	24284.23532
16.25	17.1041	81.5367	59.6498	487.9956	50.2386	70.7615	102.5297	39970.5603	21577.51662
17.25	16.6469	83.2233	57.3771	530.4668	51.6731	75.4949	103.2620	39027.0220	22340.73889
18.25	14.9509	73.1108	48.1476	443.5927	45.9867	63.9943	86.2433	35003.6814	19772.98081

Appendix 11: Geochemical analysis of the lake sediments in the Rodna region

Romanina Lakes Project									
Rodna Mountains		2006							
CORE NAME:LACUL BILA LB1									
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	6.7861	43.9038	41.0039	154.3738	41.0015	186.3669	131.7335	29244.0949	17020.8723
1.5	4.6063	44.9185	39.1760	152.8004	39.1525	191.1296	139.7554	29603.8484	17642.4060
2.5	8.2429	45.5731	41.5629	149.7980	39.9839	176.7083	129.8014	29071.6822	17287.6720
3.5	9.8269	49.2918	45.4101	160.6796	44.7619	195.6847	141.9414	30782.9449	18830.7934
4.5	10.0291	47.7886	39.5350	162.4224	42.2684	188.9967	135.6589	28581.5349	18724.8424
5.5	11.5479	55.2304	42.1275	177.4128	46.0047	194.4263	136.4565	32136.8195	21772.3854
6.5	8.7752	54.2712	42.8757	173.7162	45.1701	197.0414	135.4163	31608.5462	21315.3856
7.5	10.1644	54.3196	43.9978	172.1111	46.5409	203.0271	137.0225	31942.8055	20747.4347
9.5	9.3657	48.8024	39.1812	153.4585	40.9651	182.2740	121.6168	29060.5349	18390.0655
10.5	6.8704	46.3334	32.4333	152.1583	36.8296	172.2689	113.8087	27434.3178	17423.5665
11.5	6.1188	44.9502	33.0843	147.9355	36.6703	165.0079	112.8681	27251.7614	17075.6195
12.5	3.5265	40.7180	27.4492	134.6233	33.7006	157.5007	113.5216	25088.3008	15518.3844
13.5	5.5456	42.8978	31.5499	136.0954	34.4649	168.3479	110.5671	25777.3399	15950.6158
14.5	3.8534	38.7041	22.4564	124.6060	29.5095	147.0618	97.3954	22632.2060	14379.8233
15.5	5.2844	40.2871	22.6702	125.8933	31.8236	151.4985	104.4878	23429.8608	14897.4010
16.5	8.3690	50.0830	23.8547	159.3553	37.9783	165.4645	118.0452	28101.6018	18780.0343
17.5	6.4801	46.0917	23.1868	145.1913	39.6931	152.8195	113.3907	25830.0683	17360.9339
18.5	5.8806	46.2326	18.9737	148.1817	35.4896	137.9859	112.0594	25962.3823	17074.4918
19.5	6.0915	47.4482	20.4350	146.5027	38.1965	140.2322	119.1201	26678.7943	17244.5706

Lacul Buhaiescu-3; Core LB-3 2									
Depth	**Co**	**Cr**	**Cu**	Mn	Ni	Pb	**Zn**	**Fe**	Al
0.5	4.6528	30.4061	29.1081	208.2706	24.8671	116.2252	148.3699	26222.2736	15972.0425
1.5	3.5260	25.2409	23.7861	184.9366	20.6203	88.9839	140.1757	22156.9483	13179.9930
2.5	6.1167	29.8490	26.9760	222.4295	25.4505	96.4209	140.7437	25071.1349	15127.6433
3.5	6.7680	33.2596	26.1526	243.0367	28.7602	99.7987	145.5108	26430.4087	16946.1538
4.5	6.1299	32.3151	26.3225	245.7653	28.6317	90.0666	139.8561	26268.6132	16650.6898
5.5	7.2433	30.1756	27.5867	236.3756	26.9372	83.9967	122.5606	26216.3889	15898.5000
6.5	8.3539	31.0205	25.7636	244.6866	27.8889	76.4284	109.1883	27169.8977	15622.2645
7.5	9.2219	32.2564	29.2207	251.3197	28.4473	74.2687	101.9204	28896.2529	15963.7002
8.5	6.7307	34.1761	23.1946	244.9301	26.0147	51.2619	85.4789	28282.8947	14337.2451
9.5	10.4218	35.4371	32.0788	272.7511	29.3594	59.4541	92.0986	38421.9388	17430.1587

Lacul Lala Mare; Core LLM 2									
Depth	Co	Cr	Cu	Mn	Ni	Pb	Zn	Fe	Al
0.5	11.3428	45.3772	35.2554	125.7243	43.4017	100.2590	115.2046	42628.8434	16647.5284
1.5	12.1405	47.7012	38.7587	131.5934	44.7858	100.4707	119.1822	43041.9771	17448.7705
2.5	10.2297	46.2124	33.8799	133.7272	41.1977	91.4538	108.1933	41694.8892	17455.5419
3.5	13.1641	51.1840	40.9708	145.7582	45.9398	99.0870	113.3715	45565.4206	18927.7453
4.5	10.4244	46.5868	37.3655	125.0333	40.5453	94.3151	102.2084	42080.3571	17068.4335
5.5	10.4381	45.4962	36.3875	126.2391	40.9495	91.9977	106.7941	40933.8948	16910.0218
6.5	9.8981	45.5974	36.2572	126.4159	38.9582	88.4388	99.9638	40326.1136	17042.1492
7.5	12.0780	47.4306	35.2701	138.6746	40.6499	90.5342	106.9605	40501.4245	17533.8319
8.5	14.5928	52.3151	39.8707	155.4924	44.7819	99.5892	112.0070	44211.6965	18747.1000
9.5	13.8723	50.3950	40.8191	146.3735	43.6265	97.0734	105.6226	42820.4114	18315.7254

Lacul Pietrosul LP 1 2006									
Depth	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Al
0.5	7.5901	31.5597	31.4051	27293.7633	218.3086	31.3465	86.7063	108.7148	14605.2239
1.5	7.5901	29.5410	21.2106	27293.7633	206.7809	24.4323	77.8470	113.6365	14605.2239
2.5	9.2806	27.7226	15.7453	28282.4427	228.1817	23.8963	62.9347	88.0566	14929.7604
3.5	9.2782	27.2071	14.3073	29052.1362	240.2369	26.6609	45.5039	69.4580	15014.5939
4.5	9.4906	26.7970	18.8814	29972.7048	240.5336	22.6019	50.5727	70.3529	15313.2239
5.5	10.2642	30.2207	34.7297	32423.5399	253.8018	24.1451	55.6899	77.4214	16161.8926
6.5	10.5557	32.4054	21.9376	33198.4869	271.0019	26.2656	56.4136	79.5895	17312.9729
7.5	8.8381	26.2965	18.7788	29223.7483	237.7647	23.3208	44.6622	79.3448	14760.7997

Lacul Pietrosul LP 1 2008									
Depth	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Al
0.5	11.9810	23.4395	38.6195	44493.1267	244.2533	25.2965	140.1101	129.5416	10247.4371
1.5	11.4538	27.1382	40.8228	29177.8755	194.5615	29.6738	129.7024	139.0958	12752.9705
2.5	13.4333	29.6702	40.0104	25833.1149	211.5356	31.7142	119.1355	149.3627	13859.6549
3.5	11.5935	23.0045	33.4004	18642.4475	168.9763	23.9995	81.7844	89.6065	10462.7257
4.5	12.9669	22.4669	35.7187	18252.4096	172.0892	22.1127	62.6958	70.7133	10566.8072
5.5	19.4432	25.8937	37.6503	21555.1381	206.9038	26.9104	70.9779	73.6524	12436.2579
6.5	14.6661	27.3461	37.9525	21685.3277	212.4907	28.8226	65.3736	71.7080	12811.9414
7.5	14.7738	22.8124	34.1983	17840.4829	192.1560	21.9245	49.6138	54.3214	10663.7951
8.5	15.2399	36.4717	33.2026	29134.9686	305.1879	37.8740	61.1195	83.5555	17524.4079

Lacul Pietrosul LP 1 2008 Russian									
Depth	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Al
0.5	18.2993	28.5503	19.1212	25614.6581	245.6480	28.1211	58.7751	88.8875	14366.0928
1.5	19.4460	30.8428	24.7363	24842.8121	252.6979	30.6677	57.5315	80.7313	15420.1283
2.5	21.1222	32.9565	26.6860	26167.9329	268.9170	31.2253	61.2126	87.0357	16602.7496
3.5	21.0834	31.1692	28.7304	24502.0197	247.7283	29.5646	58.0498	78.9441	15561.8829
4.5	22.6810	34.2413	31.6045	27342.2264	271.9397	32.7991	64.7301	86.9117	17157.9602
5.5	21.8979	32.6295	28.8996	25630.4958	268.1564	30.7419	59.5652	80.6497	16358.7567
6.5	23.2729	34.3381	30.0195	27349.8856	278.6785	32.5543	60.9949	82.0755	17160.4119
7.5	25.0482	35.1509	27.9018	28567.1465	292.2289	34.7953	58.9002	88.6878	18104.1084
8.5	23.2100	32.9393	27.9586	27519.4286	283.0607	31.9579	59.9457	85.6136	17021.4286
9.5	23.2450	33.6980	30.7554	27778.7181	281.9971	31.7293	63.9491	85.1186	17018.7292
10.5	22.4861	32.9222	33.2950	28176.7698	284.3917	32.7648	60.2885	88.7333	17100.6132
11.5	21.5693	34.8989	35.3369	28469.7857	286.3533	31.3566	61.3351	85.9539	17390.4662
12.5	23.1804	36.1557	30.9926	29003.4080	292.5590	33.7013	62.2535	89.9270	17688.9606
13.5	21.5007	33.5969	30.9375	27078.3235	273.2713	30.6874	62.6248	82.2510	16379.2536
14.5	24.1834	39.0272	28.8284	30171.6805	299.3741	33.8220	64.2473	84.8721	18434.5320
15.5	21.3610	37.0909	33.5088	30697.6987	296.6684	35.3510	69.8559	88.2520	18118.2008
16.5	20.2824	34.1129	30.2922	26301.6117	270.1852	28.7697	56.6006	74.7675	15933.9321
17.5	19.2291	26.1279	24.4965	21402.0386	225.1154	24.2749	50.8820	65.0293	13064.6362
18.5	23.4561	33.5272	24.4552	27760.2971	278.9672	30.0212	56.0547	76.7530	16342.7359
19.5	22.6947	27.8159	27.6652	23831.5417	244.7634	24.4402	50.3520	68.6176	14232.4987
20.5	18.8073	24.9994	21.7713	20480.8427	215.7899	22.7697	46.4096	58.3208	12479.2135
21.5	17.9711	23.7899	18.5116	19306.4815	219.1418	20.4387	41.9537	57.2865	11987.5579
22.5	19.1883	32.6221	32.3058	25521.5584	266.3032	26.3857	59.1558	71.3351	15070.0649
23.5	16.0575	20.7957	18.7448	17315.8799	187.2491	17.3207	39.0158	51.4075	10581.2901

Lacul Stiol; Core LS 2									
Depth	Co	Cr	Cu	Mn	Ni	Pb	Zn	Fe	Al
0.5	4.4766	45.1387	22.7420	282.6234	31.4989	87.0057	105.5865	34399.4038	11947.5180
1.5	8.2002	64.1138	31.1405	321.7979	37.9857	91.2923	95.5137	36545.7090	12638.8060
2.5	9.8338	78.4453	41.2958	343.8879	44.8904	89.8591	95.1915	38485.7298	13403.1792
3.5	15.7310	89.4019	48.7545	612.8647	55.0561	81.4127	91.2478	39425.4376	14894.4120
4.5	12.6141	70.7428	48.4487	867.8684	48.6842	77.3116	89.6691	37153.9855	13079.3478
5.5	13.0248	81.6955	48.2605	965.4350	52.2621	80.3585	92.6316	39241.4648	14559.9670
6.5	12.4806	80.7111	46.4045	920.8999	50.9301	78.2065	91.4177	39053.4900	14433.4512
7.5	14.6721	83.6026	57.0372	1007.1755	56.1614	82.0727	99.3402	41523.5375	15051.9881
8.5	13.2205	86.3926	54.9053	923.5506	54.6297	79.8071	91.2240	39537.6242	14886.7724
9.5	13.8678	92.1555	52.2795	907.7760	57.4237	79.2342	95.6480	40795.1638	15842.0669

Rodna Mountains		**2006**							
CORE NAME: Lacul Vinderel 3									
Depth	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Al
1.5	7.8021	33.8031	21.4054	39041.3748	182.7036	36.0453	103.6466	105.1204	17892.8415
2.5	7.8892	34.3421	20.7638	38362.6227	200.5269	34.3862	110.0552	111.3414	18354.3153
3.5	7.4503	31.6191	18.7130	35183.0791	186.7567	29.8491	113.2322	114.0832	16777.2436
4.5	7.0355	32.0783	19.2410	35561.4985	187.6121	32.4031	115.5067	129.3390	17010.6648
5.5	8.2886	34.8796	25.3059	38325.6349	194.1637	36.0097	126.0262	139.6596	18870.6217
6.5	10.9832	36.6493	23.8735	39583.2012	192.8846	42.0095	106.4397	133.7392	20106.9891
7.5	9.4815	32.4138	18.9920	36817.9873	169.4617	36.2149	81.1452	102.0451	17978.1058
8.5	8.3642	36.1723	19.9983	36301.2566	192.5998	37.3391	80.3246	130.3031	20149.4158
9.5	8.5165	33.5303	21.1476	36373.4031	176.6173	35.8954	80.4681	101.9323	18621.6960
10.5	9.4784	33.5688	19.8380	34748.2487	178.5103	36.8783	72.4124	102.3643	19110.5517
11.5	11.7636	34.7385	24.9019	38332.3360	175.9297	41.2494	76.6185	104.7723	19438.1649
12.5	9.7740	33.0893	19.8311	36460.0687	178.4360	35.8723	71.4798	98.6558	18630.7045
13.5	10.3689	37.2745	20.5233	37035.7852	173.1142	37.0506	70.4124	99.3521	19965.9241

Romanina Lakes Project									
Rodna Mountains		**2008**							
CORE NAME: Lacul Vinderel 1									
Depth	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	AL
0.5	14.3019	40.7331	48.0828	44724.9287	247.1227	51.6784	122.6711	126.5922	19071.7681
1.5	14.3687	40.8264	45.7247	40185.7783	207.8886	53.7675	116.0286	125.7029	19847.0704
2.5	15.2472	42.3409	50.8272	40341.3485	198.2489	54.7681	123.9807	128.5359	20351.3828
3.5	14.3860	39.8650	50.1762	38196.0105	190.6916	52.5770	119.1274	122.5776	19064.2021
4.5	12.3633	32.4624	35.4615	30598.3364	208.6616	46.4654	103.7056	112.7313	15328.7375
5.5	14.0649	41.3132	46.0279	38373.2278	217.0711	50.7644	134.9770	140.8713	19674.6219
6.5	13.6859	42.6928	47.7222	39789.0120	214.3808	51.9015	136.2950	134.2042	20474.6653
7.5	14.7008	41.3877	45.3500	38108.2354	209.0192	49.6500	147.1292	142.1304	19417.3397
8.5	13.0304	40.6835	43.4834	36027.0069	208.5780	45.3670	146.0550	143.2173	18964.9083
9.5	15.1585	40.7682	45.3046	37469.8357	207.7107	52.3991	150.1185	143.4795	19081.3967
10.5	16.0209	41.1872	45.4560	37874.0547	201.8917	50.0033	149.7570	141.8439	19928.8812
11.5	15.9802	39.3675	44.7406	37950.9011	196.5845	51.9299	135.2698	134.1954	19003.2348
12.5	15.7739	41.1626	45.0862	39312.6457	204.9009	54.7861	125.4913	132.2308	20129.5455
13.5	16.0892	41.0714	45.9849	39021.0481	202.8418	54.8908	133.9973	135.5486	19802.4055
14.5	15.4057	38.9054	42.9700	37203.42302	202.4170	50.3495	117.3604	121.6123	19255.3644
15.5	15.6534	40.6442	42.2667	38499.38935	207.1116	55.8738	107.4115	122.1611	19683.8056
16.5	15.0067	36.8293	41.0533	36272.41147	201.5610	51.6691	97.9281	111.3509	17889.6795
17.5	16.2483	39.0667	42.8356	37760.46305	210.0640	50.7903	102.8478	117.2022	18506.4559
18.5	18.1055	38.5962	45.2081	38897.27783	219.2151	55.9302	100.8478	117.2485	18873.2910
19.5	17.0617	39.2790	44.6577	39283.6355	226.5840	55.6661	99.1227	115.1759	19153.0296
20.5	16.1551	37.0593	43.1963	37844.43409	210.5985	56.8194	89.9918	112.7911	18216.8142
21.5	16.1235	39.0246	48.2866	39358.21343	218.4958	57.0486	95.7878	118.7494	19262.5899
22.5	15.4212	38.0557	43.1406	38987.84804	211.8576	54.6478	93.4680	114.0503	19050.7315
23.5	15.2172	38.5224	43.3259	39010.61629	228.6716	53.8099	96.7209	111.2838	19119.7735
24.5	14.9537	36.9859	39.6458	36690.45667	221.6575	49.3929	94.2155	106.2740	18006.5574
25.5	15.1459	36.7282	39.7447	35770.44476	206.4951	50.0311	94.0166	110.2822	17667.5036
26.5	15.0560	37.9852	41.4205	37436.33763	223.5491	50.7594	98.3220	112.5554	18742.7277
27.5	15.3553	39.1374	42.5148	38577.6693	235.2373	50.9831	94.2870	109.8251	18708.8482
28.5	15.8131	41.4236	41.9680	38989.78507	233.6343	53.6171	89.7550	107.3931	19563.0609
29.5	16.0382	40.0209	43.0499	39736.73184	242.4593	55.0314	92.4767	108.2041	19369.3552
30.5	15.5329	40.2334	42.6212	40556.70907	249.0086	56.4870	95.7165	114.0939	19433.8094
31.5	16.9452	39.1823	41.2026	40398.9547	261.0747	55.0414	95.7970	112.9159	19515.2439

Appendix 12: Fagaras Region (South) lakes Enrichment Factor (EF)

Name of lake	Depth	Metals					
		Co	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Surface							
Balea; LBa 1	0.50	29.9	172.2	91.1	144.3	261.3	375.2
Balea; LBa 4	0.50	30.2	172.3	91.1	143.8	278.8	308.5
Caltun; LCt 2	0.50	22.0	37.8	80.0	37.0	163.3	163.7
Capra LCa 2	0.50	23.4	112.5	76.9	103.1	252.0	240.6
Capra LCa 3	0.50	41.9	96.6	67.4	71.8	243.9	183.4
Podragu Mare; LPM 2	0.50	17.3	78.5	74.8	50.6	170.2	148.3
Background							
Balea; LBa 1	30.25	42.0	249.5	116.0	215.7	158.9	217.1
Balea; LBa 4	30.50	42.1	239.6	116.1	208.3	153.9	170.3
Caltun; LCt 2	31.25	23.3	38.4	56.3	40.8	85.6	126.4
Capra LCa 2	17.25	26.4	128.4	64.2	110.5	95.4	113.4
Capra LCa 3	30.50	47.0	144.0	104.7	94.7	95.9	87.2
Podragu Mare; LPM 2	18.25	15.0	73.1	48.1	46.0	64.0	86.2
EF							
Balea; LBa 1		0.7	0.7	0.8	0.7	1.6	1.7
Balea; LBa 4		0.7	0.7	0.8	0.7	1.8	1.8
Caltun; LCt 2		0.9	1.0	1.4	0.9	1.9	1.3
Capra LCa 2		0.9	0.9	1.2	0.9	2.6	2.1
Capra LCa 3		0.9	0.7	0.6	0.8	2.5	2.1
Podragu Mare; LPM 2		1.2	1.1	1.6	1.1	2.7	1.7

Appendix 13: Rodna Region (North) lakes Enrichment Factor (EF)

Name of lake	Depth	Metals					
		Co	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
Surface							
Bila; LB 1	0.5	6.8	43.9	41.0	41.0	186.4	131.7
Buhaiescu-3; LB-3 2	0.5	4.7	30.4	29.1	24.9	116.2	148.4
Lala Mare; LLM 2	0.5	11.3	45.4	35.3	43.4	100.3	115.2
Pietrosul; LP 1(06)	0.5	7.6	31.6	31.4	31.3	86.7	108.7
Pietrosul; LP 1(08)	0.5	12.0	23.4	38.6	25.3	140.1	129.5
Stiol; LS 2	0.5	4.5	45.1	22.7	31.5	87.0	105.6
Vinderel LV 3(06)	1.5	7.8	33.8	21.4	36.0	103.6	105.1
Vinderel LV 1(08)	0.5	14.3	40.7	48.1	51.7	122.7	126.6
Background							
Bila; LB 1	19.5	6.1	47.4	20.4	38.2	140.2	119.1
Buhaiescu-3; LB-3 2	9.5	10.4	35.4	32.1	29.4	59.5	92.1
Lala Mare; LLM 2	9.5	13.9	50.4	40.8	43.6	97.1	105.6
Pietrosul; LP 1(06)	7.5	8.8	26.3	18.8	23.3	44.7	79.3
Pietrosul; LP 1(08)	8.5	15.2	36.5	33.2	37.9	61.1	83.6
Stiol; LS 2	9.5	13.9	92.2	52.3	57.4	79.2	95.6
Vinderel LV3(06)	13.5	10.4	37.3	20.5	37.1	70.4	99.4
Vinderel LV1(08)	30.5	16.9	39.2	41.2	55.0	95.8	112.9
EF							
Bila; LB 1		1.1	0.9	2.0	1.1	1.3	1.1
Buhaiescu-3; LB-3 2		0.4	0.9	0.9	0.8	2.0	1.6
Lala Mare; LLM 2		0.8	0.9	0.9	1.0	1.0	1.1
Pietrosul; LP 1(06)		0.9	1.2	1.7	1.3	1.9	1.4
Pietrosul; LP 1(08)		0.8	0.6	1.2	0.7	2.3	1.6
Stiol; LS 2		0.3	0.5	0.4	0.5	1.1	1.1
Vinderel LV 3(06)		0.8	0.9	1.0	1.0	1.5	1.1
Vinderel LV 1(08)		0.8	1.0	1.2	0.9	1.3	1.1

Appendix 14: ^{210}Pb concentrations in core LCp3 taken from Capra Lake, Romania

Depth	Dry Mass	Pb-210						Cum Unsupported	
		Total		Supported		Unsupp		Pb-210	
cm	g cm^{-2}	Bq Kg^{-1}	±	Bq Kg^{-1}	±	Bq Kg^{-1}	±	Bq m^{-2}	±
1.13	0.1418	1605.05	48.35	44	10.12	1561.05	49.4	2365.7	128.4
2.38	0.4564	1299.85	32.19	32.1	4.48	1267.75	32.5	6799.1	283
3.38	0.7594	991.79	27.57	27.41	4.46	964.38	27.93	10159.8	343.4
4.38	1.0154	720.57	22.98	33.02	3.72	687.55	23.28	12254.3	366.4
5.38	1.2855	517.1	19.95	26.33	3.53	490.77	20.26	13830.7	379.8
6.38	1.599	356.57	7.7	26.23	1.36	330.34	7.82	15101.4	388.9
8.38	2.3377	185.14	8.68	25.67	1.83	159.47	8.87	16834.5	402.6
9.38	2.7322	134.34	6.38	28.09	1.49	106.25	6.55	17351.6	405.4
10.38	3.1267	51.59	6.17	24.13	1.63	27.46	6.38	17581.3	406.4
12.38	4.0705	40.71	4.06	27.11	1.11	13.6	4.21	17767.5	409.1
14.38	5.1678	73.14	9.57	53.99	2.66	19.15	9.93	17945.4	414.5
16.38	6.4036	33.86	4.62	26.41	1.29	7.45	4.8	18098.6	428.1
18.38	7.705	28.45	5.35	21.9	1.4	6.55	5.53	18189.5	432.9
20.88	9.2453	25.42	4.7	22.62	1.29	2.8	4.87	18257.5	440.1

Appendix 15: Artificial fallout radionuclide concentrations in sediment core LCP3

Depth	Cs-137		Am-241	
Cm	Bq Kg^{-1}	±	Bq Kg^{-1}	±
1.13	1099.9	18.02	10.8	3.46
2.38	1342.84	13.64	5.55	2.05
3.38	1926.19	15.39	9.02	2.05
4.38	931.46	10.12	7.42	1.66
5.38	856.47	9.71	10.85	1.74
6.38	262.81	2.42	3.29	0.58
8.38	82.17	1.98	0	0
9.38	62.54	1.41	0	0
10.38	41.85	1.44	0	0
12.38	21.31	0.78	0	0
14.38	27.19	1.72	0	0
16.38	15.39	0.82	0	0

18.38	6.28	0.79	0	0
20.88	5.55	0.72	0	0

Appendix 16: ^{210}Pb chronology of core LCP3 taken from Lake Capra, Romania

Depth	Drymass	Chronology			Sedimentation Rate		
		Date	Age				
Cm	g cm^{-2}	AD	yr	±	g cm^{-2} yr^{-1}	cm yr^{-1}	± %
0	0	2007	0				
1.13	0.1418	2003	4	2	0.0318	0.166	3.1
2.38	0.4564	1992	15	2	0.0282	0.103	3.8
3.38	0.7594	1981	26	2	0.0262	0.094	4.4
4.38	1.0154	1971	36	2	0.0273	0.104	5.2
5.38	1.2855	1962	45	2	0.0283	0.097	6.4
6.38	1.599	1951	56	2	0.03	0.086	6.8
8.38	2.3377	1926	81	4	0.0283	0.075	13.2
9.38	2.7322	1912	95	6	0.0274	0.069	19.2
10.38	3.1267	1902	105	8	0.0799	0.179	32.9
12.38	4.0705	1893	114	10	0.1188	0.233	43.4
14.38	5.1678	1879	128	12	0.0554	0.095	61.8
16.38	6.4036	1860	147	16	0.0784	0.124	80.2
18.38	7.705	1839	168	20	0.046	0.073	99.4