BABEȘ-BOLYAI UNIVERSITY

FACULTY OF ENVIRONMENTAL SCIENCE AND ENGINEERING

DOCTORAL THESIS

ABSTRACT

DOCTORAL SUPERVISORS

Conf. Habil. Dr. Alida-Iulia Gabor

Prof. Univ. Dr. Constantin Cosma

PH.D. STUDENT:

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CLUJ-NAPOCA

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Applications of ^{210}Pb and ^{137}Cs in environmental studies

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Keywords

^{210}Pb dating method, ^{137}Cs, alpha spectrometry, gamma spectrometry, ^{137}Cs database, ^{137}Cs and ^{210}Pb inventories and fluxes, dating studies, sedimentation rates, anthropic influences, Transylvania, Danube Delta.

Contents

I. INTRODUCTION

The ^{210}Pb dating method was first used in 1963 to date glacier ice by Goldberg (Goldberg, 1963). Lake sediments were first dated in 1971 (Krishnaswamy et al., 1971) using this method, followed by marine sediments in 1972 (Koide et al., 1972). Since then sediment deposits from a range of environments, including lakes, food plains, wetlands, reservoirs, estuaries, deltas and costal marine environments (Mabit et al., 2014) have been studied using the ^{210}Pb radioisotope. Understanding the processes undergoing on river floodplains prove to be of increasing interest because of the load of overbank sediment budget that can be dated by excess ^{210}Pb (Du and Walling, 2012). Additionally, sediments are excellent archives for the preservation of environmental changes over the past century. Besides determining sedimentation rates, ^{210}Pb geochronologies can be used to reconstruct heavy metal or organic compound contaminations (Hosono et al., 2016). In the last 25 years, records stored in lake sediments have become an important source of historical information on the impact of human activity on the environment during the past 100-200 years (Begy et al., 2016a).

The first ^{210}Pb dating studies bound to Romanian territories appeared in the 21st century when near-shore sections of the Black Sea coast (Aycik et al., 2004) as well as sediment cores from the anoxic zone of the Western Black Sea (Florea et al., 2011) were analyzed to determine chronologies and sedimentation rates. The construction of the Iron Gates on the Danube River leads to the retention of approx. 80% of solid material and ^{210}Pb geochronologies show that the Orşova Bay (Danube River) is the main sediment trap of the area (McGinnis et al., 2006). The Red Lake, being a natural barrage lake from the Oriental Carpathians, was prone to a series of studies involving the ^{210}Pb radionuclide. In 2008, its geochronology and sedimentation rates were established for the first time (Begy et al., 2009 a). It was estimated that the lake will disappear in 195 years (Begy et al., 2009 b), the dam-lake will fill up in 20 ± 8 years and in 81 ± 30 years only 20% of the Red Lake's surface will remain (Begy et al., 2015 a). The St. Anna Lake of volcanic origin (Harghita County, Romania) has been dated and sedimentation rates were calculated to assess the process of eutrophication (Begy et al., 2011). Fire events were related to magnetic and geochemical parameters using the ^{210}Pb, ^{137}Cs and ^{14}C dating methods in two lakes situated in the Rodna Mountains (Hailuc et al., 2016 a) and the controlling factors in the sedimentation dynamics were established in the Ighiel Lake, Apuseni Mountains (Haliuc et al., 2016 b).

I.1. Aim of the thesis and overview of the contents

The thesis is presenting the general aspects of the ^{210}Pb dating method as well as original research data published in five scientific articles I have authored and co-authored during my PhD studies.

The overall structure of the thesis contains an **Introduction**, followed by **Chapter II (Radioactive dating using ^{210}Pb and ^{137}Cs)** that focuses on lead, mainly the ^{210}Pb isotope: on its formation, occurrence and migration, possible measurement techniques and uses. The radiochronology by calculation of sedimentation rates and ages of the sediment layers is also discussed. Radiomarkers and the potential validation possibilities are described focusing on ^{137}Cs. A brief description of ^{137}Cs formation, occurrence and main sources is also included in this part.

Chapter III (Experimental Techniques) describes the gamma and alpha spectrometric measurements used in the present thesis and their application in the Constant Rate of Supply (CRS) model. The description of the equipment used and the measuring process is presented. Information on the gamma and alpha spectrometric systems on which the measurements of the radionuclides of interest have been carried out, along with the alpha source preparation using various acids are presented as well. Two of the main problems associated with the extraction of ^{210}Po are presented. Because the classical leaching procedure takes up to three weeks, an improved procedure for ^{210}Po extraction has been developed by changing the acids and their concentrations in the leaching procedure and decreasing thereby the time of the source preparation to a few days including the measuring of the samples. Repeatability tests for both classical and improved procedures were carried out on standard IAEA materials for validation and the improved method was successfully applied on deltaic sediments (Begy el al., 2015 b). Another goal of this work was to assess the systematic error derived from the ^{210}Po content in the residue silicates. Analyses were carried out using four leaching procedures on ten sediment sample of different geographic origin; one of the digestion methods is including HF, known to dissolve even the silicate content of the samples.

Chapter III (Dating studies) describes the studies which were undertaken to improve and asses the methodology of the dating studies, but also includes dating applications on different geographic regions. Because ^{137}Cs is an important radiomarker bound to the overall ^{210}Pb dating method, a study regarding this isotope was conducted over Transylvania Region, Romania (Begy et al., 2017). An introduction to the radionuclide inventory calculations needed for age-depth model applications is presented by showing ^{137}Cs and ^{210}Pb fluxes and inventories that were calculated from the activity concentrations of soil and peat cores from seven different locations of Romania (Begy et al., 2016 a). The ^{210}Pb dating method was applied on three lakes from the Danube Delta. Parameters such as water content, porosity, bulk density, loss on ignition (organic matter, carbonate and total carbon content) were calculated and radionuclide concentrations were established to determine the ages and sedimentation rates of sediment cores (Begy et al., 2015 c; Begy et al., 2016 b). Several anthropic and natural effects on the sedimentation rates are discussed. An important find was the observation of increased sedimentation rates in the past 30 years in the Danube Delta. Therefore the sedimentation rates for four lakes situated in different geographical locations have been further analyzed (Simon et al., 2017).

At the end of the thesis **Final conclusions** are drawn and further possible studies are discussed.

II. RADIOACTIVE DATING USING ^{210}PB AND ^{137}CS

Among its existing 35 isotopes, there are four main lead-isotopes present in the environment: ^{208}Pb (52%), ^{206}Pb (24%), ^{207}Pb (23%) and ^{204}Pb (1%), their concentration differing in different rock formations. The first three isotopes are the end-products of the ^{238}U, ^{235}U and, respectively, ^{232}Th decay chains, while ^{204}Pb is the only primordial isotope with natural abundance. Only ^{202}Pb ($T_{1/2}$ = 52.5 ka), ^{205}Pb ($T_{1/2}$ = 17.3 Ma) and ^{210}Pb ($T_{1/2}$ = 22.2 y) have longer half-lives, the other isotopes have half-lives in the range of a few hours to fractions of seconds.

^{210}Pb ($T_{1/2}$ = 22.23 years) is part of the ^{238}U decay chain and it decays through β-emissions (E_{max} = 17 keV [84%] and E_{max} = 61 keV [16%]) followed by γ-emission (46.53 keV [5%]). 19.8% of the decay occurs at the ground level of the ^{210}Bi isotope, and 80.2% have as a result an excited nucleus, which stabilizes by emitting the low energy gamma rays and an internal conversion of electrons leads to the ^{210}Bi isotope. ^{210}Bi ($T_{1/2}$ = 5.01 days) has a much higher β-radiation (E_{max} = 1161.5 keV [100%]) than its parent. As a result, the α-emitting (E_{α} = 5.503 MeV) ^{210}Po ($T_{1/2}$ = 138.37 days) is produced.

^{210}Pb, ^{210}Bi and ^{210}Po are omnipresent in the environment (rocks, soils, sediments, atmosphere and natural waters) as a result of the decay and deposition of ^{226}Ra (^{222}Rn); the latter found often in elevated concentrations. The daughter radionuclides can often be detected at some distance from the supposed sites of release, due to the extreme mobility of ^{222}Rn gas. ^{210}Pb and its decay products diffuse through many surfaces (fissures, aquifers etc.), where the long lived progenies of the ^{238}U decay chain can be produced. The disequilibrium between ^{210}Pb and its parent nuclide, ^{226}Ra ($T_{1/2}$ = 1600 years), also arises through the diffusion of ^{222}Rn ($T_{1/2}$ = 3.8 days). A fraction of the ^{222}Rn atoms diffuse into the atmosphere and its decay products (mainly ^{210}Pb and its granddaughter ^{210}Po, since the other intermediates are relatively short lived and show less importance in the radionuclide transport) are absorbed onto aerosols and dust particles and are then removed from the atmosphere by wet and dry deposition. The process is summarized in the *Fig. II.1*.

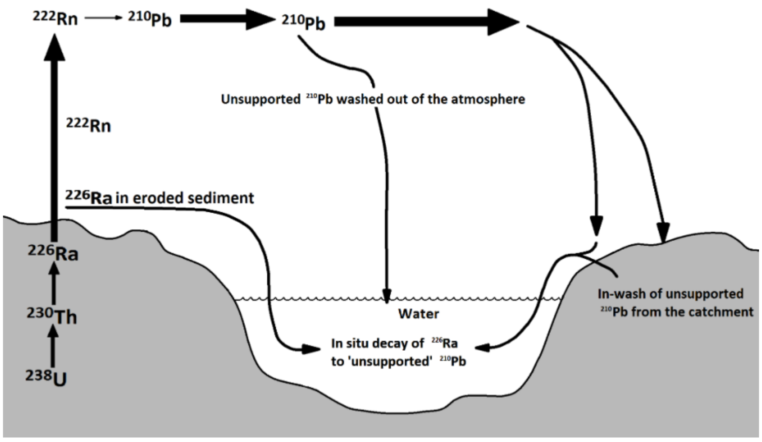

Fig. II.1 – The ^{210}Pb pathways (after Oldfield and Appleby, 1984)

Consequently, the surface soil layer contains a higher ^{210}Pb level than the one expected from the equilibrium with ^{226}Ra. The fraction of ^{210}Pb which is in equilibrium with ^{226}Ra is called supported ^{210}Pb (or ^{210}Pb in situ), while the excess to the equilibrium is called unsupported ^{210}Pb (or excess ^{210}Pb – $^{210}Pb_{ex}$). The latter decreases with the half-life of ^{210}Pb and is the basis for environmental applications.

For a reliable depth determination of soil and sediment matrices, it is fundamental to accurately determine both supported and unsupported components of ^{210}Pb. The measurement of the unsupported ^{210}Pb requires the determination of both total ^{210}Pb and ^{226}Ra. The latter being in secular equilibrium with the supported ^{210}Pb, the calculation being made as follows: $^{210}Pb_{ex}$ = $^{210}Pb_{total}$ – ^{226}Ra ($^{210}Pb_{sup}$).

Several analytical techniques are available for the measurement of the ^{210}Pb. However, in this study, gamma spectrometric and alpha spectrometric measurements were used for the supported and, respectively, total ^{210}Pb content determination.

^{226}Ra and ^{137}Cs radionuclide content measurements were carried out using an ORTEC Digiart gamma spectrometer with an N-type GMX HPGe semiconductor detector (resolution for ^{60}Co at 1.33MeV of 1.92KeV with 0.5 mm Be window for low energies, and the relative efficiency of 34.2%) having a passive shielding of 10 cm thick lead and 3 cm copper. “Sarpagan” type geometry was used with cylindrical boxes of 7 cm diameter and 1 cm height. Samples were put into sealed aluminum boxes and stored for at least 21 days to ensure the equilibrium between ^{222}Rn and ^{226}Ra. The Gamma Vision 32 was used to analyze the peaks of interest and each measurement had at least 80,000 s counting time; background measurements were carried out for at least 500,000 s. The activity concentrations for each sample were determined using the relative method, by comparing the sample spectra to reference materials and all measurements were background corrected.

For the total ^{210}Pb measurements, dried, homogenized samples were added ^{209}Po tracer (E_α = 4.9 MeV) and put to acidic digestion using HNO_3, HCl and H_2O_2. Ascorbic acid was added to eliminate interfering ions and the pH of the solution was adjusted to be in the 0.5-1 range using distilled water, NaOH or HCl are added. High nickel content stainless steel discs were cleaned using HCl and ethanol and spontaneous depositions were made from the samples in a heating oven (3 h, 82°C).The alpha spectrometric measurements were carried out using an Ortec Soloist PIPS detector, having a resolution of 19 keV. Data acquisition was made using an ASPEC-927 Dual Multichannel.

Various ^{210}Pb dating models based on several assumptions related to the initial activities of ^{210}Pb, ^{210}Pb flux or accumulation rate (Appleby & Oldfield, 1978) exist, which make it possible to reconstruct variations in the sedimentation rate throughout time. The model used in this study is the Constant Rate of Supply model (also known as Constant Flux model – CF). It has as fundamental hypotheses an efficient transfer from the water column to the sediments and a constant $^{210}Pb_{ex}$ flux of the sediment surface and no diffusion throughout time. The chronologies are generated by comparing the overall core ^{210}Pb inventory to partial inventories below depth *i*, by integrating the supported ^{210}Pb concentration, the bulk density and the thickness of each sediment layer, obtaining the mass sedimentation of each layer and the ages at the limit of the sediment layers (Mabit et al., 2014).

The mass accumulation rates and the initial $^{210}Pb_{ex}$ concentrations may change over time in the layers, but must be inversely proportional (Appleby, 2001). The increase of sediment supply does not always imply the higher $^{210}Pb_{ex}$ fluxes in the upper sediment layer. In other words, the sedimentation rate and the initial $^{210}Pb_{ex}$ concentration can be variable or irregular throughout time.

The accumulated deposit below the layer *i* can be written as:

$$A_i = A_0 e^{-\lambda t} \qquad \text{(II.1)}$$

The age of the layer *i* can be calculated as follows:

$$t_i = \frac{1}{\lambda} \ln \frac{A_0}{A_i} \quad \text{(II.2)}$$

The mass accumulation rate for layer *i* can be calculated using:

$$r_i = \frac{\lambda A_0 e^{-\lambda t}}{C_i} = \frac{\lambda A_i}{C_i} \quad \text{(II.3)}$$

The CRS model can be and has been validated by several independent methods and is therefore the most applied dating method. It can be used if sedimentation rates vary significantly and if the sediment was prone to mixing (Szarlowicz et al., 2013).

In this study, the artificial ^{137}Cs (E_γ = 661 keV) is used in validating the ^{210}Pb dating method, since precise dates are known when these radionuclides presented global and regional fallouts. Two peaks are monitored according to depth: the nuclear weapon tests from 1956 and the Chernobyl accident in 1986 and therefore two maximums should be visible in each core. It is to be noted, that 86% of the ^{137}Cs content deposited in 1956 has already decayed, making it challenging to check the correctitude of the dating method.

III. EXPERIMENTAL TECHNIQUES

In this chapter, two often encountered problems associated with the total ^{210}Pb measurements through ^{210}Po are discussed: the first one treats the reduction of the digestion time through the development of new leaching recipe and applying it on deltaic sediments (*Subchapter III.1*), while the second (*Subchapter III.2*) analysis the ^{210}Po trapped in the crystal structures of sediment and their influence on the ^{210}Pb dating procedure.

III.1. An improved method for ^{210}Po determination

Over time, a series of ^{210}Pb determination techniques have been developed by using its progeny, ^{210}Po. Measurements are carried out using alpha spectrometry, which has a series of advantages if samples with low-level activities are analyzed: high sensitivity, low intrinsic detector background and the elimination of the possible interferences by chemical separation. Also, the use of a tracer makes the method even more reliable. A proper polonium source for alpha spectrometry must be sufficiently active to achieve good statistical accuracy and the polonium layer must be thin and homogeneously distributed (Begy et al., 2015b).

Following the procedure developed by Edgington and Robbins (1975) for alpha source preparation using acidic leaching presented in the introduction, the entire process, from sample to source lasts 4–5 days. Therefore an alternative method was developed having a similar yield and producing good spectra in the shortest period of time possible. The differences between the original are the concentrations and quantities of the used acids: 10 ml 35% HCl, 30 ml 6N HCl and 1.5 ml 35% H_2O_2.

For the testing of the polonium extraction yield, three samples with high organic material content were subjected to both classical and improved procedures. After filtration, 30-40% of the initial mass was left as residue, which was then re-digested using the classical procedure. Although the residual mass was significant, the polonium yield was 92%. Applying this method, the digestion time can be reduced to 3-4 h.

A comparison for four standard IAEA reference materials was carried out (IAEA-447, IAEA-312, IAEA-385 and IAEA-327) for the two methods. Results show, that values obtained by both methods are in good agreement with the certified values of the reference materials. The repeatability test carried out on seven IAEA-385 samples shows similar values within the margin of errors.

Applying the improved method on high silicate content samples, the residue content is high, and only a fraction of polonium is digested. Neither the classical, nor the improved method digest the silicate content fully, therefore a third digestion method was introduced containing 5 ml 70% HF, 8 ml 65% HNO_3, 25 ml 35% HCl and 2 ml 35% H_2O_2, this method having a 95% yield for ^{210}Po.

III.2. ^{210}Po incorporated in residual crystals

Another frequently encountered issue is the ^{210}Po generated from ^{210}Pb absorbed on clays, oxides, hydroxides and organic matter in environmental samples (mostly soil and sediments). Under right circumstances, manganese and iron oxides are exchanged with lead (which then decays to polonium), as well as calcium and potassium containing clays (IAEA, 2006).

Oxides, hydroxides and organic matter are soluble in either HNO_3, HCl or H_2O_2, and ^{210}Po can easily be extracted from these minerals. A fraction of the silicate content (orthosilicates) can be decomposed by acids (as an example by HCl to form silicic acids and salts), while the other fraction can only be decomposed by the additional use of HF (Jeffery et al., 1989).

Organic material can be oxidized as CO_2. Acids with a high rate of oxidation are HNO_3, $HClO_4$ and H_2SO_4, while those non-oxidizing are HCl, HF, HBr and H_3PO_4. HNO_3 is the most commonly used compound for organic matter destruction, since it decomposes organics, sulfides, selenides, tellurides, arsenides, sulphoarsenides and phosphates. It also dissolves the majority of metals with the exception of Au and Pt. Because of its high oxidative properties, $HClO_4$ can be used to decompose organic matter and sulphides. Most formed perchlorates are soluble in water, exception being those of K, Rb and Cs. Having a higher boiling point than HF, HNO_3 and HCl, it can be also used to drive these acids off. H_2SO_4 is less used because of the interferents created by SO_4 in the analytical procedures. Aqua regia (3:1 HCl:HNO_3) is effective against organic material , sulfides of As, Se, Te, Bi, Fe and Mo, arsenides, selenides, tellurides, sulfosalts, native Au, Pt and Pb, natural U-oxides, Ca-phosphates, most sulfates (except barite) and some silicates (eg. zeolites). Being a strong acid, HCl dissolves carbonates, phosphates, borates and sulphates (except barite). HF is used to decompose organic matter, oxides and sulphides, as well as silicates. It is most effective in breaking Si-O bonds to SiF_4, which then volatilize upon heating. To prevent the precipitation of insoluble solids, evaporation by the $HClO_4$ can be applied (Mudroch et al., 1996).

The most commonly used method for ^{210}Po extraction implies the use of aqua regia or a mixture of HF, $HClO_4$, HNO_3 and HCl acids in different quantities and concentrations, depending on the objective of the analysis. Sample to reagent ratio, grain size distribution heating temperatures, pressure and digestion times may vary, but the goal is to provide the complete – or at least partial –leaching. Digestion using HNO_3 alone digests polonium incompletely (Card and Bell, 1985), while the use of HNO_3 and HCl is more common (Vaaramaa et al., 2010).

The effects of different leaching procedures on the ^{210}Pb dating method were determined using four different leaching procedures were tested. The obtained residue after the leaching procedures can contain a certain amount of ^{210}Pb (Edgington and Robbins, 1975; Macklin Rania et al., 2014), therefore it was predigested using the HF digestion method (Aalto and Nittrouer, 2012) known to provide a complete digestion.

Ten lacustrine sediment samples with various genesis types from different areas of Romania were chosen for this study (Table III.2.1)

Table III.2.1 – Location and characteristics of the analyzed lakes

Location	Lake	Genesis	Geology
Eastern Carpathians, Rodna Montain Narional Park	Buhăescu Lake	Glacial lakes	Metamorphic rocks: chlorite schist, sericite-chlorite schist, mica schist, various types of gneiss, amphibolite, carbonate rocks; Sedimentary deposits: carbonate clays, limestone, clay, conglomerate and sandstone.
	Știol Lake		
	Muced Lake		
Central Eastern Carpathians	Sfânta Anna Lake	Volcanic lake	Volcanic rocks: high-K dacite with amphibole and biotite.
	Red Lake	Barrier lake	Sedimentary rocks: crystalline and clayey schists, limestone, andesite, dolomite, microcrystalline rocks, scree, sandstone, conglomerates.
Eastern Romania, Romanian Plain	Sărat Lake	Oxbow lake	Sedimentary deposits: loess deposits, pale yellow and dust-like soils, sandstone.
Danube Delta	Merhei Lake	Deltaic lakes	Sedimentary deposits: sand, sludge other rocks transported by the Danube from its catchment area.
	Uzlina Lake		
	Iacob Lake		
	Cruhlig Lake		

The four leaching recipes were tested following Benedik and Vrecek, 2001 (No1), Edgington and Robbins, 1975 (No2), Macklin Rania et al., 2014 (No3) and an own recipe (No4). 0.3-1 g of sediment per sample in two ways: with an aliquot of 0.3 ml ^{209}Po tracer and without any tracer, in order to avoid the contamination of the residue content. A summary of the used acids and their quantities is visible in *Fig. III.2.1*.

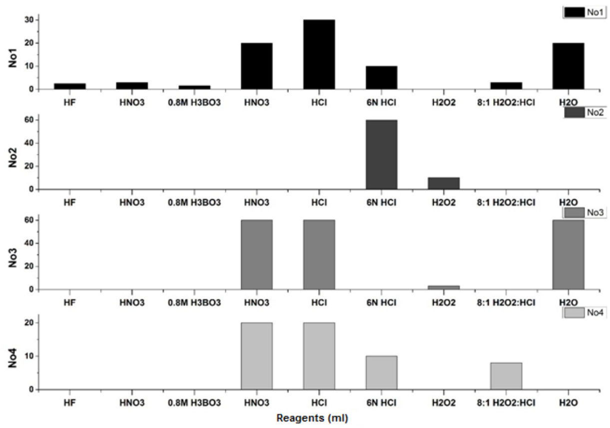

Fig. III.2.1 – Summary of the used acids in each leaching procedure

Results show, that only one of the lakes (Muced Lake) can be leached completely using each of the leaching procedures (activity concentrations being in the interval of confidence), since it is in an early peat-bog state.

The mass-percentage of the leached samples (*Fig. III.2.*2) shows that method No1 provides a complete digestion, not depending on the mineralogical composition of the sample. The second best method proved to be No3: the least amount of residue remains in case of a volcanic lake (13%), followed by the glacial lakes (32%), while the most unleached sediment remains in case of the deltaic lakes (57%). The most residue (61%) remained after applying method No2.

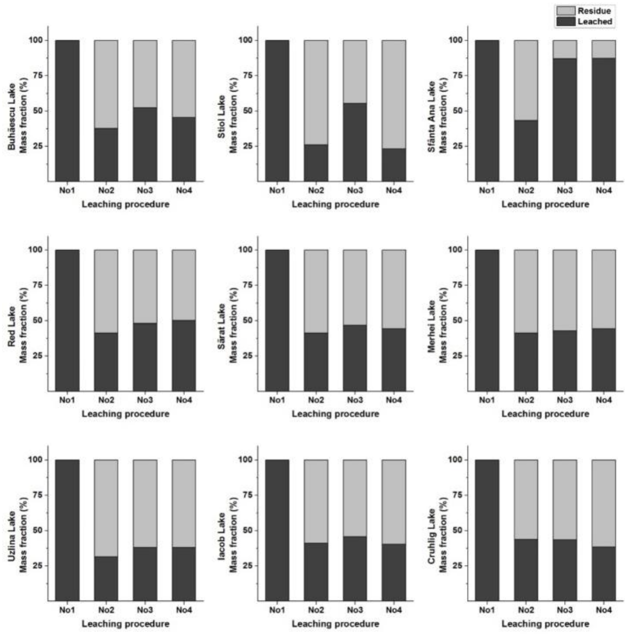

Fig III.2.2 – Mass percentage of the leached samples

Total activity concentrations were calculated from the activities measured after applying each digestion method and the HF method on the residue. Results obtained by method No1 were considered to be leached completely and the activity concentrations of the other methods were compared to these. Without leaching the ^{210}Po content of the residue, method No3 provides a 73% leaching, whereas methods No2 and No4 assure 56%

and, respectively 59% digestion. 28% of the ^{210}Po fraction restrained in the crystalline structure of the minerals remains after the No4 leaching procedure, while only 23% remain in case of No3 and 21% in case of No2.

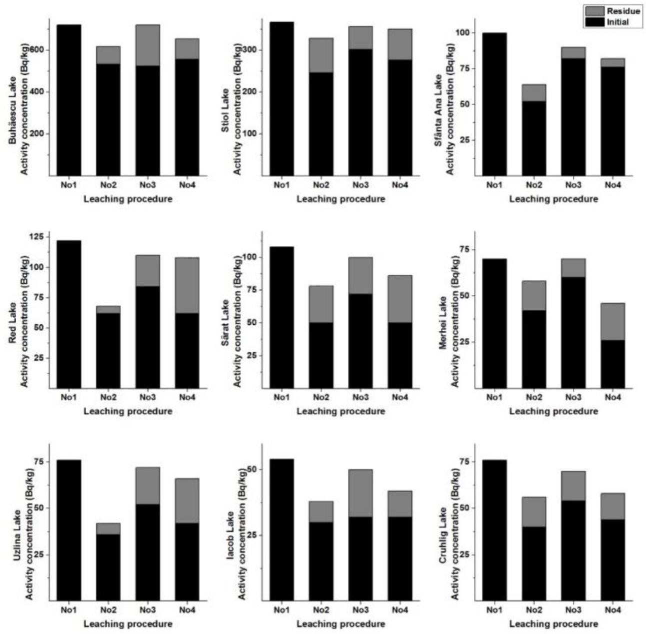

Fig III.2.3 – Activity concentrations after each leaching procedure

After applying the CRS model on the sediment samples an average of 9% relative uncertainty it is visible, being below the 2σ confidence level of the measured activities

IV. DATING STUDIES

Because of its pathways, ^{210}Pb is distributed through atmospheric circulation and is deposited then on Earth’s surface by wet and dry deposition. Soils, sediments, snow, ice and water reservoirs have proven to collect ^{210}Pb, which can provide information about the changes in these environments for a period of over 200 years (Walling and He, 1999a,b; Walling et al., 2003; Mabit et al., 2008).

One of the most important parameters of the ^{210}Pb cycle in the environment is its atmospheric flux, varying from 0.1 Bq m^{-2} yr^{-1} to 360 Bq m^{-2} yr^{-1} (Mabit et al., 2014). The ^{210}Pb deposition to the surface of the Earth is higher over continents than oceans and varies with season, longitude, local meteorological and geological conditions (Preiss et al., 1996; Winkler and Rosner, 2000; Caillet et al., 2001; Baskaran et al., 2011; Mabit et al., 2014). For determinations of ^{210}Pb flux, common procedures like the collection of wet and dry deposition during periods long enough to accommodate seasonal and episodic variations, are applied

IV.1. ^{137}Cs contamination over Transylvania region (Romania) after Chernobyl Nuclear Power Plant Accident

. ^{137}Cs is an artificial radionuclide and originates in the environment from nuclear weapon testings and nuclear accidents with known dates. Atmospheric inventory values at the time of the Chernobyl accident were measured, and values over 1480 kBq m^{-2} have also been reported in the proximity of the accident (De Cort et al., 1998). By the year 2017, these values have decayed to half the initial ones, since no sources have been reported for the past 30 years in the European region. An extensive mapping of ^{137}Cs activity concentration was carried out in the central, northern and western areas of Romania to establish the distribution of the radioisotope one half-life after the nuclear accident.

A total of 153 soil-samples were taken during 7 different campaigns over five years (2010-2015) from the Transylvania region: Eastern Carpathians (14-34), Transylvanian Plateau (1-13; 50-61; 105-124), Southern Carpathians (35-49) and two sampling campaigns which included sampling points throughout the entire Transylvania (62-104, 125-153) (*Fig. IV.1.1*). Additionally, 13 samples were taken from the Moldova-Ukraine border and the Danube Delta for data comparison.

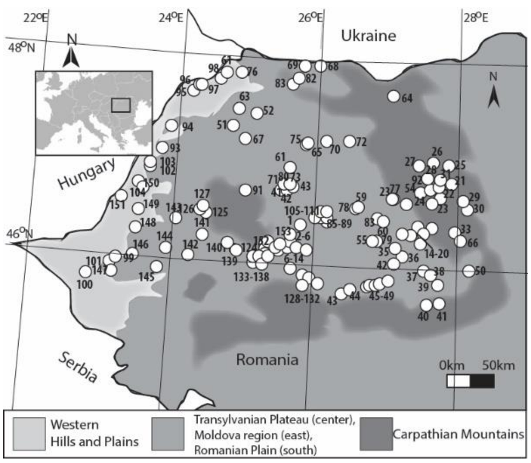

Fig. IV.1.1 – Location of sampling points

The radioactive deposition caused by the Chernobyl nuclear accident was mainly dry and uniformly distributed over the affected area, but due to the wet deposition isolated cases with high level concentrations occurred (De Cort et al., 1998).The obtained ^{137}Cs data showed surface activities in the range of 0.4 ± 0.1 to

301.1 ± 3.0 kBq·m^{-2}. The arithmetic mean of the values was 20.5 kBq·m^{-2}, while the geometric mean was 8.3 kBq·m^{-2}. Two hotspots could be observed, Iezeru-Ighiel (I) and Tulgheș (II) (*Fig. IV.1.2*). The location of these hotspots was in good concordance with the direction of the radioactive cloud movement simulations, which had a north-eastern to south-western direction from Chernobyl. These reached the areas from Romania in the zones with higher surface activities, producing probably wet precipitations as well.

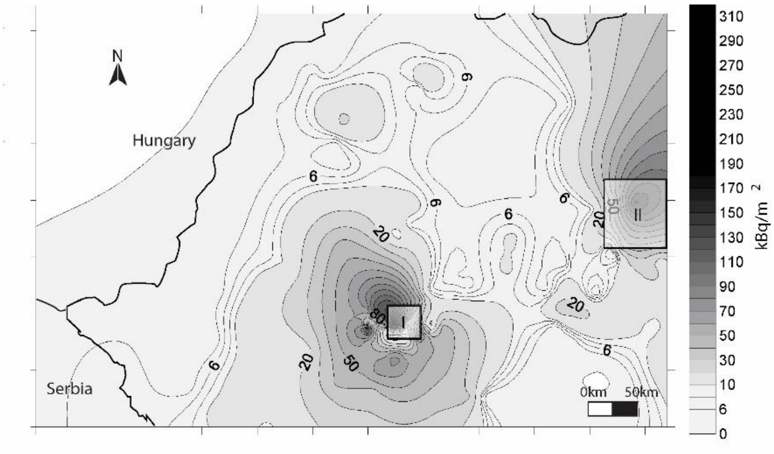

Fig. IV.1.2 – Distribution of surface activities

Considering Transylvania's geomorphology, the territory can be classified into three sub-regions: the Western Plain and Hill, the Transylvanian Plateau and the Carpathians. These areas can be well differentiated, taking into consideration their mean altitude and annual precipitation levels. The Western Hills and Plain had the lowest surface activity values for ^{137}Cs (2.6 ± 0.1 kBq m^{-2}, with values in the 0.4-6.3 kBq m^{-2} range). Characteristic values for the Transylvanian Plateau were 0.4-78.3 kBq m^{-2}, with a geometrical mean of 6.0 ± 0.1 kBq m^{-2}. Geometric mean value for the Carpathian Mountains was 18.3 ± 0.6 kBq m^{-2}, with minimum value of 0.7 kBq m^{-2} and a maximum of 301.1 kBq m^{-2}. Lowest values were observed in the Southern Chain (0.7-84.2 kBq m^{-2}). The Eastern and Western Carpathians displayed larger average values: 12.0 kBq m^{-2} (2.2-124.0 kBq m^{-2}) and, respectively, 28.9 kBq m^{-2} (5.8-301.1 kBq m^{-2}).

Taking the cardinal regions into consideration, it was noted that the southern region of Transylvania received the most amount of ^{137}Cs (on average 16.3 kBq m^{-2}), while the northern and western regions were the least exposed (2.9 kBq m^{-2}). Analyzing the samples according to their slope exposure, those with an eastern exposure received on average 27.8 kBq m^{-2}, the south-western, northern, western and south-eastern slope exposures received on average values in the 11.5-14.7 kBq m^{-2} range, the lowest values being registered on slopes with southern, north-western and north-eastern slope exposures (6.9-7.9 kBq m^{-2}).

VI.2. Atmospheric flux, transport and mass balance of ^{210}Pb and ^{137}Cs (Begy et al., 2017)

Because ^{210}Pb fluxes and inventories are two of the main measured parameters on which the ^{210}Pb method relies on, ^{210}Pb and ^{137}Cs fluxes and inventories were calculated. Radioisotope depositions of peat and soil cores from six different NW regions of Romania were analyzed in a total of 12 cores. Because SE Romanian territories were suspected to have received lower depositions of the nuclides of interest, a comparison between the two areas to quantify these differences (*Fig. IV.2.1*).The spatial distribution of the ^{210}Pb radionuclide is analyzed in the bed of the Red Lake, Romania.

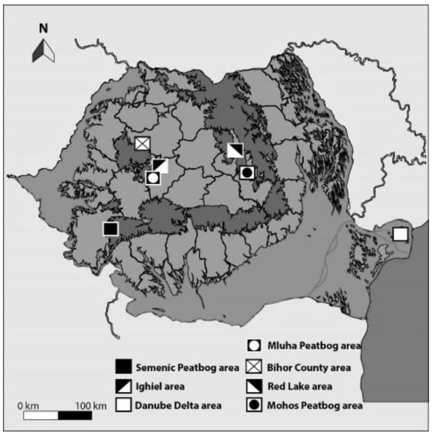

Fig. IV.2.1 – Sampling locations

Results show, that^{210}Pb flux distribution shows maximum values in the Semenic Peatbog area, since the presence of thermal waters and transversal faults is accentuated in that area; values gradually decrease to the eastern (Danube Delta area) and northern side of the country (*Fig. IV.2.2*).

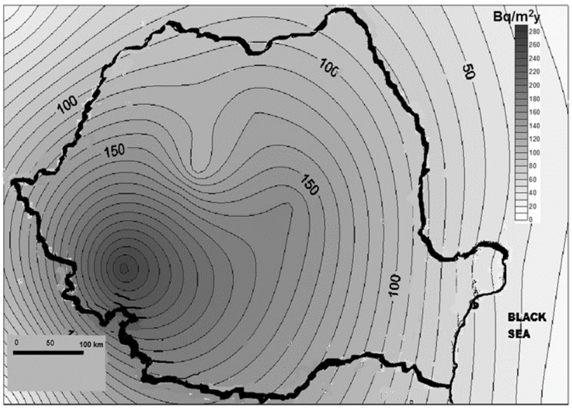

Fig. IV.2.2 – ^{210}Pb flux map of Romania

^{210}Pb transport and mass balance of the Red Lake sediments show that the southern part of the lake is exposed to considerably larger ^{210}Pb fluxes, the maximum value being 628 ± 94 Bq m^{-2} yr^{-1}. The minimum value (126 ± 29 Bq m^{-2} yr^{-1}) is registered on the eastern branch of the lake, where the water exchange is very fast, since the travelled distance of the water is only 440 m, therefore the deposition of ^{210}Pb is limited. The mean flux measured in the sediments (309 ± 48 Bq m^{-2} yr^{-1}) is 2.4 times larger than the atmospheric fallout of ^{210}Pb (129 ± 2 Bq m^{-2} yr^{-1}) and only 0.84% of the ^{210}Pb flux measured in the catchment area reaches the sediment.

IV.3. Dating studies applied to the Danube Delta

Being one of the largest floodplains of Europe, the Danube Delta encloses hundreds of shallow lakes. The characteristics of these are largely influenced by seasonal and interannual water fluxes caused by the transport of channels, flooding and the water flow under the reed beds. The variability of the conditions in the lakes leads to differences in plant, fish and plankton communities (Oosterberg et al., 2000; Buijse et al., 2002). Additionally, the natural habitats are exposed to man-made stress factors such as eutrophication (Coops et al., 1999) and geomorphological impacts, outstandingly since 1970, because of dike and dam construction, fishing, agricultural activities in the delta (Cremer et al., 2004) and contaminated water inflow. The water circulation of the delta is complex, since many major and minor river channels connect, having temporally and spatially changing water levels, caused by factors such as precipitation, evaporation, periodic floods and droughts. The dynamics is also influenced by erosion and sedimentation processes which are able to shape the landscape. Abiotic processes tend to trigger responses in plant growth by producing reed-beds and forests along the rivers.

Anthropogenic influences have always been present in the Danube Delta region. In the past, traditional activities have taken place such as hunting, fishing and trapping, but only for own consumption; reed harvesting; collecting of herbs; horticulture; beekeeping, all within the ecological capacity of the system. In the last century however, these activities have become more dominant and increased, some such as fishing, reed harvesting and agriculture being developed on an industrial scale (WWF, 2007).

The Danube River is regulated with hundreds of dams and reservoirs, over 150 being constructed on the Romanian portion of the river. These reservoirs are presently storing up to 22 10^9 m^3 water. Dams constructed on the tributaries of the Danube reducing the sediment quantity considerably by trapping sediments in the reservoirs (Constantinescu et al., 2015). Before the construction of the dams after 1950 on the Jiu, Olt, Argeș, Ialomița and Siret rivers, these provided one third of the sediment load; afterwards measurements show a decreasing of 69% for the Jiu River, 67% for the Argeș River and 48% for each Siret and Prut rivers (Rădoane, 2008). Additionally, more than 600 dams were built on the Bulgarian tributaries following World War II. The sediment discharge has decreased therefore to 9.1% of its original quantity (4.4 10^6 t yr^{-1}) (Levashova et al., 2004) and the suspended sediment is 60% less than in the pre-dam conditions (McCarney-Castle et al., 2012). However, the major impact on the reduction of the sediment load was the construction of the Iron Gates I and II Dams and Hydorenergetic Power Plants, which, combined with the other man-made water regulating structures, reduced the sediment load from 1846 kg s^{-1} (1840-1970) to 962 kg s^{-1}(1971-2000) (Bondar, 2008).

For this study, a total of 13 sediment samples were taken from three lakes: the Merhei Lake (ME15, ME16, MEII19, MEII20 and MEII21), the Matița Lake (MA18, MA20 and MAII17) and the Cruhlig Lake (CR1, CR2, CRII1, CRII2 and CRII3). Sampling locations and sampling points are shown in *Fig. IV.3.1* and *Fig. IV.3.2*.

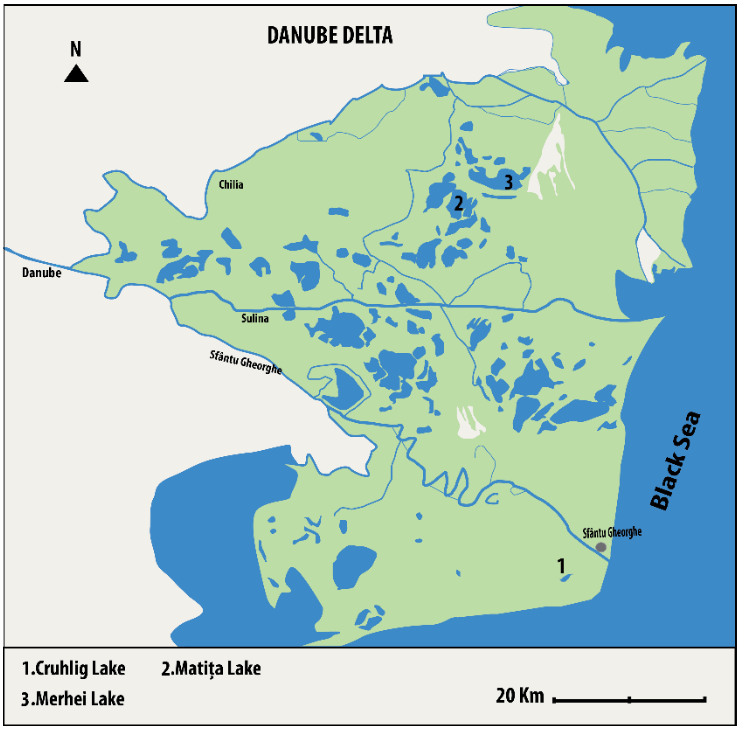

Fig. IV.3.2 – Sampling locations

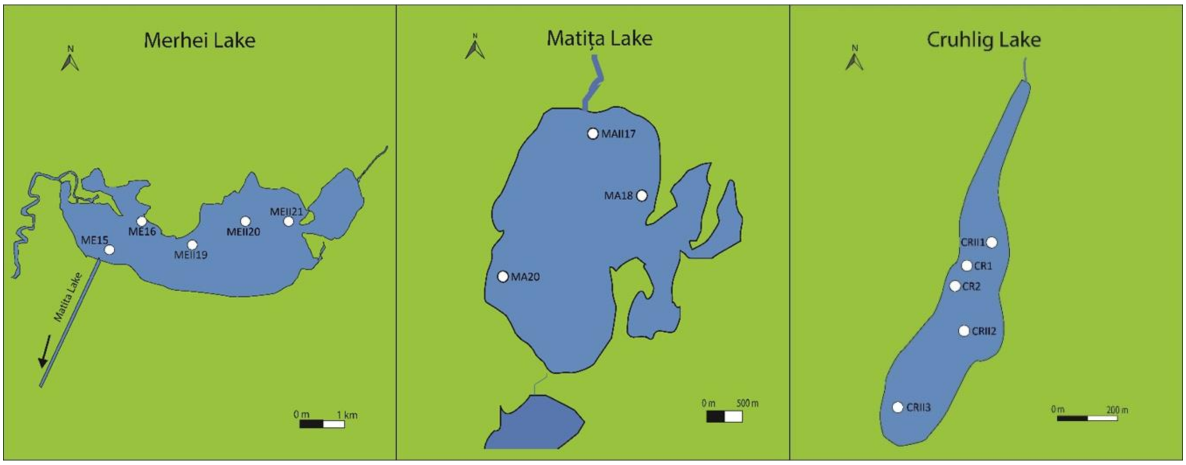

Fig. IV.3.2 – Sampling points

The next four subchapters aim to discuss the four of the main possible causes for changes in sedimentation rates. Several flooding events with extremely high debits have been recorded over the past few decades, phenomena which are able to shape the morphologies of the lakes by eroding and depositing significant amounts of sediment. Changes in the grain sizes and composition of the sediments can be due to the existence of strong aeolian activities, able to generate an intake in the lakebeds and eutrophication can generate an excess of deposed organic material. Anthropic activities, especially the construction of dams, can restrict the sediment income and can lead to erosion processes along the main paths of water flows.

Flooding events

The multiannual debit of the Danube River is constantly changing, depending on the period length taken into consideration. The multiannual mean for 122 years was 5420 m^3 s^{-1} at the Orșova hydrometric station. Generally, the minimal flow of the Danube is registered at the beginning of spring, in autumn or winter, and the lowest debits are measured in the winter periods with very low temperatures, when the formation of ice is high. In the 1965-2012 period 24 floods were registered with higher debits then 10,000 m^3 s^{-1}. These values were measured at Baziaș station and are summarized in *Table IV.3.1*.

The three analyzed lakes are not equally susceptible to the above mentioned flooding events. The Matița-Merhei lake system is susceptible to a series of the above mentioned floods. Both Matița and Merhei have six inflow channels, which account for a complex water circulation. The flooding events of the last 10-15 years are visible in the increased sedimentation rates of all cores, and, additionally, the eastern part of the lake-system received an increased sediment amount in the 1976-1988 period, where 8 major flooding events took place. According to the sedimentation rates of the Merhei Lake, the western part of the lake (ME15 and ME16) receives an order of magnitude lower sediment quantity and is not exposed to the major sediment income. On the other hand, the cores situated in the eastern part of the lake show much more variation and significantly increased sedimentation periods in the last 30 years.

The Cruhlig Lake, being the most secluded of the analyzed lakes, is situated in the south of the Danube Delta below the Sf. Gheorghe Branch. In the northern part of the lake, both eastern and western banks (CR1, CR2 and CRII1), most of the early flooding events before 1970 can be detected. These changes the layout of the lake in such manner that CRII1 and CRII3 receives twice as much sediment as before the construction of the Iron

Gates (Begy et al., 2016b), the southern part of the lake being more susceptible to the floodings from the late eighties and nineties. The spatial distribution of the sedimentation rates reflect the distribution of the sediment deposition processes toward the eastern bank of the lake where sediment deposition rates at CRII1and the CRII3 sampling points were found to be 7.25 times larger (Begy et al., 2016b).

Table IV.3.1 – Maximum debits (ANAR, 2012)

No.	Year	Max. debit (m^3 s^{-1})
1	1965	12,250
2	1966	10,810
3	1967	11,050
4	1968	10,500
5	1970	13,040
6	1974	12,100
7	1975	12,150
8	1976	11,400
9	1977	12,200
10	1979	10,900
11	1980	11,900
12	1981	14,800
13	1982	10,500
14	1987	11,610
15	1988	12,690
16	1998	10,280
17	1999	11,100
18	2000	12,000
19	2004	10,800
20	2005	12,900
21	2006	15,800
22	2009	10,700
23	2010	13,350
24	2011	10,200

Outer sediment source

The similarities in water content, porosity and dry bulk density as well as LOI values (*Fig. IV.3.3*), the lack of peaks in the total ^{210}Pb concentration and the slow decreasing of the ages in concordance with depth in case of the MEII19, MEII20 and MEII21 cores of the Merhei Lake lead to the conclusion of the existence of an outer source of sediment supply.

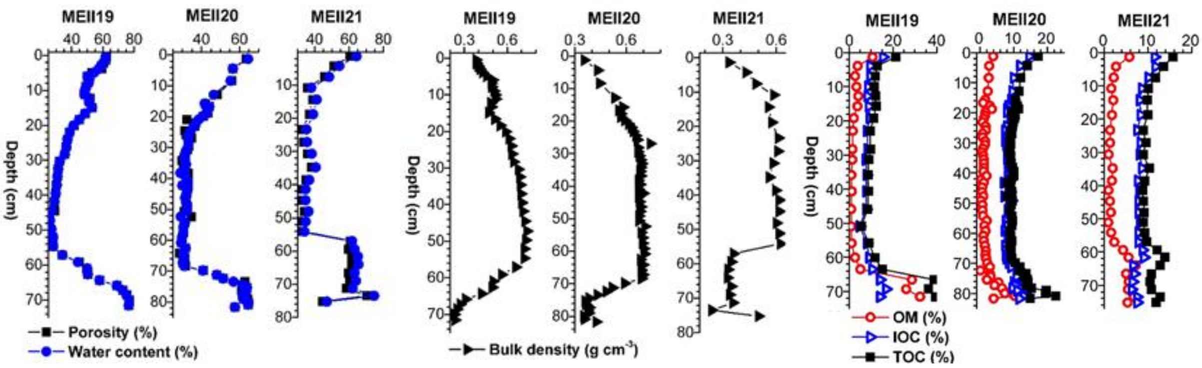

Fig. IV.3.3 – Water content, porosity, bulk density and LOI values for the analyzed cores

Grain size analysis was carried out on the cores, to determine if there is any similarity in the grain size distribution of the cores and to see whether there is a correlation between the three cores. Results are depicted in *Fig. IV.3.4.*

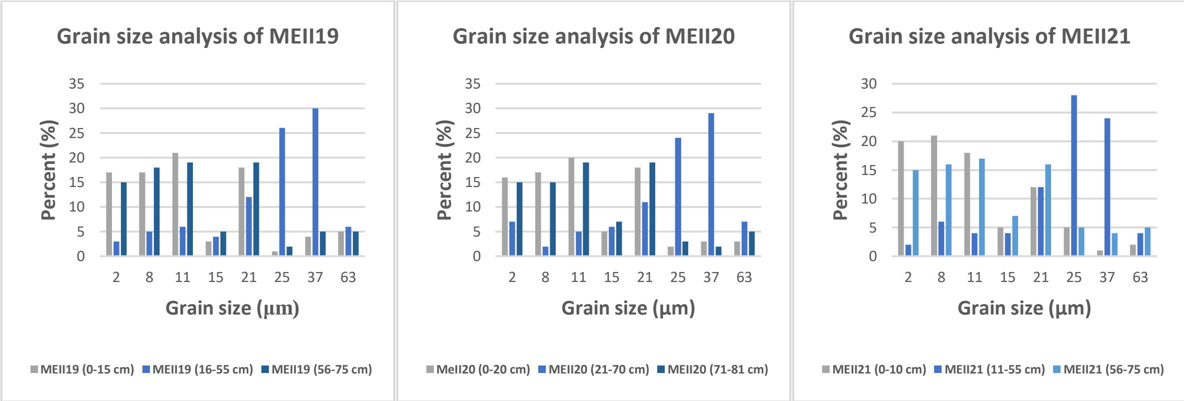

Fig. IV.3.4 – Grain size distribution

Data presented show good agreement in the grain size distribution of the first and last sections of the cores and distributions show good similarity in the three cores, proving that the sediment in the middle sections has the same source.

The Letea Dunefield was formed when the Sf. Gheorghe and Sulina branches of the Danube built wave-dominated lobes by longshore drift (Gioşan et al., 2005) and is one of the largest aeolian landforms from the Black Sea area (Preoteasa et al., 2009). It consists of three parabolic N-S oriented features. It is composed of allochtonous clastic sands and has a local contribution of the secondary distributaries of the Sulina Branch, its outer ridges are made out of Danube sands characterized by mica and quartz (Panin, 1989), with grain sizes between 0.22-0.26 mm for the marine sand and <0.20 mm for the Danubian sand (Preoteasa, 2008; Vespremeanu-Stroe et al., 2016). Wind conditions are predominant from the northern direction with a bimodal distribution of NW-NE (Preoteasa and Vespremeanu-Stroe, 2004) which changes in the summer months to S-SE (Giosan et al., 1999). The average annual wind speeds of 5-6.5 m s^{-1} (Giosan et al., 1999). The last aeolian activity, which shaped the land formation, occurred 240-70 yrs ago (Preoteasa et al., 2009).

Taking into consideration the last intense aeolian activity, the predominant wind directions and the reported grain sizes in the literature, the deposition of the middle section of each core could originate from the Letea Barrier. However, further investigations should be done.

Eutrophication

The eutrophication of the delta led to changes in flora and fauna (especially fish composition) (Buijse et al., 2002). As this is a problem that mostly occurs during the summer period when higher concentrations of N and P are available (a percentage of it entering the delta as pollutants), the algae reduce the penetration of light. However, the massive eutrophication starting in the 1960's is fully remediated to the present and eutrophication has no impact in the present on the analyzed lakes (Gastescu, 2009).

From the analyzed lakes, only one sediment core present higher organic material content: MA18, from the western near-shore part of the Matița lake presents an OM content with an average of 10% throughout the entire core. According to Oosterberg et al. 2000, the sampling area of the MA18 core is characterized by submerged aquatic vegetation and turbid water. Some cores of the Merhei Lake present increased sections of OM near the SW and NW inflow channels (ME15 and MEII19), where both floating and submerged aquatic vegetation can be temporarily observed (Oosterberg et al., 2000).

Anthropic influences – dam construction

Among the numerous anthropic constructions on the Danube Delta river basin, the most significant is the construction of the Iron Gates dam. The implementation of the Iron Gates Hydroelectric and Navigation System on the Danube River was based on an Agreement concluded in 1963 between Romania and Yugoslavia that led to the construction of the upstream dam Iron Gate 1 in 1972 (located at km 942 of the Danube) and the Iron Gate 2 in 1986 localized 68 km downstream. The lower system is composed of two dams: the lower one on the main branch of Danube at km 862 and the upper one on the Gogoșu secondary branch, km 875 and of a separate navigation lock between them through the Ostrovul Mare Island.

The total drainage area upstream of the Iron Gate I is 577,000 km^2, representing 250-300 km upstream the Danube River. The annual water flow of the Danube River is 110-220 10^9 m^3, while daily discharges range between 1500 and 15000 m^3 s^{-1}. The suspended sediment concentrations in the Danube River are in the 10^{-3} to 10^{-1} kg m^{-3} range, while the sediment volumes entering the reservoir are considerably larger: 7-30 million tons per year (Laszlo, 2007).

The dams interrupted the natural sediment transport in the Upper Danube. The Iron Gate dams retain approximately two-thirds of the suspended solids. Therefore, sediment delivery to the Delta decreased from 53 to 18 million tone yr^{-1} (Dutu et al., 2014), resulting in severe coastal erosion (Sommerwerk et al., 2009). A significant reduction of the Danube's sediment discharge is visible after the construction of the Iron Gate barrages (Opreanu, 2010), which is also proof of the increasing riverbed erosions in the last decades. Samples collected upstream of the Iron Gates 1 and downstream from the Iron Gates 2 predominantly indicate sediment erosion, while samples taken from the Iron Gates 1 and 2 reservoir lakes are attesting deposition. It is notable, that the Iron Gate 2 lake's intake of sediments is less, because most of it is already retained by the first barrage (Opreanu et al., 2007). In the delta, sedimentation being present at the sea mouths of each branch (Chilia, Sulina, Sf. Gheorghe). Some ambiguous results have been obtained using various sedimentation and erosion identifying methods, meaning that there is a possibility for the alternation of erosion and sedimentation patterns according to the fluctuations of the hydrologic parameters.

Assuming that there should be an observable decreasing in the sedimentation rates of the lakes after the implementation of the two dams, the sedimentation rates of each core were analyzed for the period of before and after 1972. However, the delta is a dynamic environment and the decreasing or increasing of the sedimentation rates cannot be attributed to a single event but rather to the sum of events that take place in the delta and in the catchment area of the Danube River. *Table IV.3.2* sums up the sedimentation rates before (1940-1972) and after (1972-1983) the construction of the Iron Gates. Increasing tendencies after 1972 are marked with blue, while decreasing ones with red.

From the analyzed lakes, the Matița Lake is the only one, with a visibly increasing sedimentation rate after 1972, values increasing 1.95 times for mass and 1.85 times for linear sedimentation, meaning that the lake has either not been affected by the sediment retention of the Iron Gates or that multiple sediment transporting and depositing phenomena have occurred in the area of the lake.

The sedimentation tendencies are similar for the Merhei Lake: sedimentation shows increasing for the MEII20 and MEII21 cores of 1.25 times for mass and 1.48 times for linear sedimentation, while MEII19 is the only core of the Merhei Lake to experience decreasing in both mass and linear sedimentation. The sampling points from the western part of the lake show relatively constant values for both periods of before and after 1972.

Table IV.3.2 – Average sedimentation rates before and after the construction of the Iron Gates

Lake	Core	Mass sedimentation ($g\ cm^{-2}\ yr^{-1}$)		Linear sedimentation ($cm\ yr^{-1}$)	
		Before 1972	**After 1972**	**Before 1972**	**After 1972**
Matița	MA18	0.21	**0.38**	0.22	**0.41**
Merhei	MA20	0.29	**0.58**	0.32	**0.69**
	MAII17	0.14	**0.27**	0.30	**0.45**
	ME15	0.10	0.11	0.32	0.32
	ME16	0.04	0.02	0.17	0.14
	MEII19	0.84	**0.53**	1.20	**0.67**
	MEII20	0.22	0.23	0.27	**0.44**
	MEII21	0.17	**0.26**	0.41	**0.55**
Cruhlig	CR1	0.10	**0.07**	0.63	**0.20**
	CR2	0.09	**0.05**	0.59	**0.22**
	CRII1	0.45	**0.11**	0.30	**0.21**
	CRII2	0.28	**0.14**	0.10	**0.08**
	CRII3	0.26	**0.12**	0.18	**0.14**

The Cruhlig Lake, due to its secluded position in the Danube Delta, was influenced by a decreasing in both mass and linear sedimentation rates with an average of 54% and, respectively, 56%.The most affected sampling point is CRII1 with a decreasing of 75.25%, being situated in the proximity of the channel connecting the lake to the Sf. Gheorghe branch. CR2, CRII2 and CR3 show similar decreasing patterns (51.48%, 51.25% and 54.92%), while CR1 shows the least changes (Begy et al., 2016b).

IV.4. Anthropic influences on the sedimentation rates of lakes situated in different geographic areas (Simon et al., 2017)

Four lakes from different geographic areas of Romania were chosen (*Fig. IV.4.1*). Two lakes were analyzed from the Carpathian region with different formation histories: the St. Anna Lake being formed in an inactive volcanic crater by surface run-offs (one core: SzA), while the other one – the Red Lake – was formed by the collapse of rocks during a landslide (six cores: three from the lake itself LR3, LR4 and LR5, and three from the dam-lake (G1.1, G1.2 and G1.3), one artificial retention lake from the Transylvanian Plateau: Vârșolț Lake (one core: V1C), and one dynamically changing lake from the Danube Delta region: Matița Lake (three cores: MA18, MA20. MAII17).

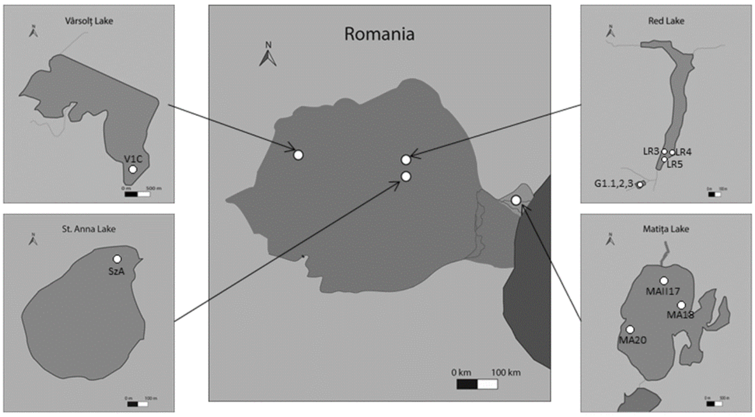

Fig. IV.4.1 – Sampling sites and locations of the sampled sediment cores

All these lakes are analyzed using the ^{210}Pb dating method, by emphasizing the period of the last 30 years, when Romania witnessed a regime change from communism to democracy. After the 1989 revolution citizens got their lands back, and, in order to get to a fast financial reward, people started to engage in unauthorized lumber processing all over the country, which then altered the entire hydrological cycle. Besides this, a series of anthropic events influence the changes in the sedimentation rates. Some of these are the agricultural practices, industrialization, the dramatic urbanization, the growth of deforestation in the last decade. These are all activities which contribute to the phenomenon of global warming. Lacustrine navigation can also be a potential source of physical and chemical pollution along with the uncontrolled tourism – this being mostly characteristic to the St. Anna Lake and to the Red Lake (Gomoiu, 1996). The Danube Delta has been greatly influenced by hydro technical works – especially the building of the Iron Gate Hydro Power and Navigation System in the 1972-1978 period – especially Iron Gate I, which has a backwater effect of 310 km upstream and a drainage area of 577,000 km^2. These diverted a great amount of sediment from the delta – and the channel, dams and polders built for the alteration of the natural water flow. The suspended sediment concentration carried by the Danube River is in the $10^{-3} – 10^{-1}$ kg m^{-3}, while the carried sediment is in the $7 – 30 \cdot 10^6$ tons yr^{-1} (BabicMladenovic et al., 2013). Over-fishing can also lead to the disruption of the natural equilibrium of the lakes (Sakan et al., 2011; Vukovic et al., 2014).

In case of the Red Lake, sedimentation rates started growing at the beginning of the nineties in such manner, that those of the dam lake increased on average 1.88 times, while those of the lake itself 2.81 times (*Fig. IV.4.2*). The eastern part of the lake shows an increasing tendency (from 0.38 ± 0.04 g cm^{-2}yr^{-1} to 1.76 ± 0.21 g cm^{-2} yr^{-1}) until the end of the nineties, followed by a series of low and high sedimentation periods. Outstanding values can be observed between 2000-2009, when values grow 4.05 times compared to the prior period. In case of the dam lake a relatively constant 0.33 ± 0.04 g cm^{-2} yr^{-1} sedimentation rate is visible until the nineties. Begy et al. (2009b) found an average of 1.32 ± 0.31 g cm^{-2} yr^{-1} and, respectively, 1.68 ± 0.43 for cm yr^{-1} for the 1991-2009 period. The latter period with increased sedimentation rates is attributed to the increased quantity of suspended sediment and increased woodwork in the drainage basin, which leads to an accentuated soil erosion.

Similar tendencies are observable in the sedimentation rates of the St. Anna Lake (*Fig. IV.4.3*): the incipient 0.06 ± 0.01 g cm^{-2} yr^{-1} in 1980 increases doubly (0.11 ± 0.01 g cm^{-2} yr^{-1}) to 2002, the maximal value of 0.16 ± 0.02 g cm^{-2} yr^{-1} is accounted to the 2002-2005 period.

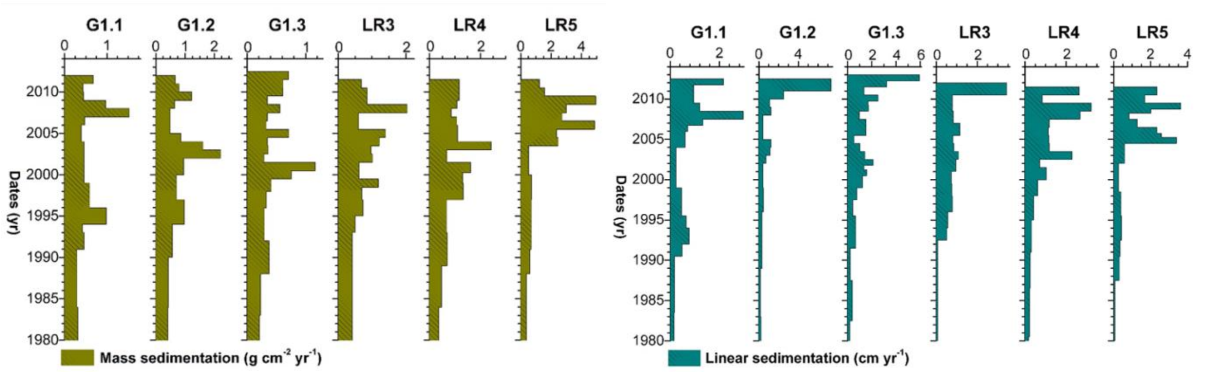

Fig. IV.4.2 – Mass and linear sedimentation rates for the Red Lake and its dam lake

Both Red Lake and St. Anna Lake are affected by the massive deforestation from the 2000-2006 period, when 4295 ha of woods were cut in Harghita County, from which approximately 33.34 ha can be found in the catchment area of the Red Lake. According to Enea et al., 2012 the soil erodibility in the catchment area of the Red Lake had values from 0.83 tons ha^{-1} yr^{-1}, up to 1.14 tons ha^{-1} yr^{-1}, with isolated, extreme values reaching 5.28 tons ha^{-1} yr^{-1}. The deforestation sources are close to the drainage basins (often upstream, in hidden sites) of the main rivers and the cut tree logs are transported illegally to the water courses. Local authorities noticed the silting probabilities of the lakes, and have therefore built three dams on the Oii, Rosu and Licas streams) in the nineteen sixties for retaining the increased alluvium in the Red Lake. These have a relatively small capacity in comparison to the Red Lake, two of them having almost reached their maximal retention capacity. A minor factor for the increased sedimentation rates is the lack of roots in the deforested areas, grains being able to enter the river basin by surface runoffs caused by precipitation, the melting of snow, wind etc. (Gancz et al., 2014).

The Matița Lake receives its sediment through more channels and is part of a system composed of several lakes (*Fig. IV.4.3*). The MAII17 is situated in the northern part of the lake at mouth of the main inflow channel, so the transport of the sediments is much more accentuated then the deposition. This is the explanation for the relatively constant 0.35 ± 0.03 g cm^{-2} yr^{-1} sedimentation rates until 2006, preceded by a growth of 6.61 times. The maximum value is visible in the 2009-2010 period. The western MA18 sampling point shows outstanding values (1.59 ± 0.21 g cm^{-2} yr^{-1}) for the 1980-1984 period, followed by a relatively constant sedimentation with an average of 0.59 ± 0.06 g cm^{-2} yr^{-1} until 2005. The maximum value (3.14 ± 0.42 g cm^{-2} yr^{-1}) is visible in 2012. The sedimentation periods of the MA20 sampling point (situated in the eastern part of the lake) can be separated in two: namely the 1980-1996 period, when the average sedimentation was 0.52 ± 0.06 g cm^{-2} yr^{-1}, and the 1998-2012 period, when the average was 1.16 ± 0.21 g cm^{-2} yr^{-1}.

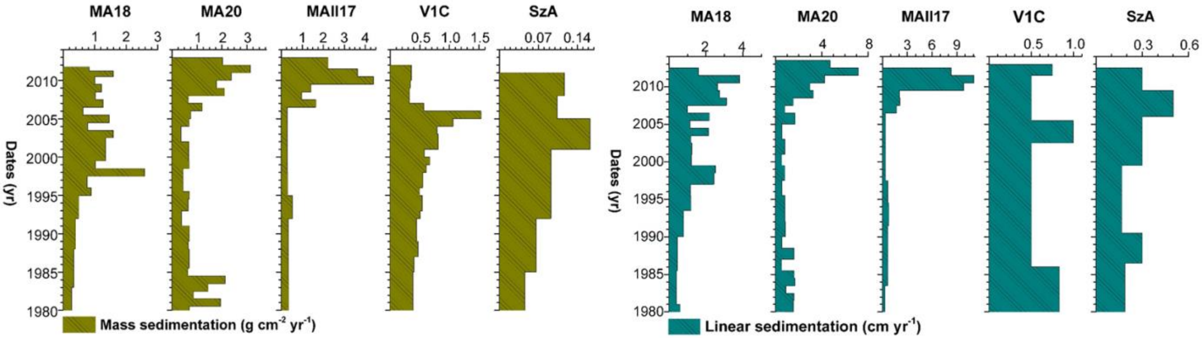

Fig. IV.4.3– Mass and linear sedimentation rates for the Matița Lake, St. Anna Lake and Vârșolț Lake

An increasing tendency of sedimentation rates is visible in case of the Vârșolț Lake until 2006, its average being 0.53 ± 0.05 g cm^{-2} yr^{-1}. In the period of 2006-2007 the sedimentations are maximal: 1.53 ± 0.18 g cm^{-2} yr^{-1}, when solid discharge reached the highest annual average (15.31 kg s^{-1}) (Moigrădean, 2013).This is followed by a constant period of 0.39 ± 0.04 g cm^{-2} yr^{-1}. Linear sedimentation rates show three outstanding periods: namely 2010- 2011 (0.75 ± 0.08 cm yr^{-1}), 2002-2004 (1.02 ± 0.12 cm yr^{-1}) and 1980-1984 (0.83 ± 0.09 cm yr^{-1}). The silting characteristics of the Vârșolț Lake measured in the 1979-2009 period (Moigrădean, 2013) show that the process was accelerated in the first 12 years (0.39 m^3 yr^{-1}), and that they considerably diminished in the next 18 years (0.19 m^{3}yr^{-1}) – also visible in both mass and linear sedimentation rates – as consequence of the reduction of the local erosion rate, changes appeared in the land-use, the reduction of land degradation by the land-washing phenomena. Also, significant work has been undertaken in the improvement of river-beds (Șerban et al., 2010).

CONCLUSIONS

^{210}Pb dating method accompanied by the ^{137}Cs radiomarker is a chronological technique of great importance in environmental studies regarding records stored in lake sediments and peat bog accumulations. This method has proven to be useful in the radiochronology of young sediments (up to 200 years old) and the determination of their accumulation rates. Measurement of alpha emitting radionuclides consists of a rather complex chemical preparation and separation process. ^{210}Pb can be quantified via its ^{210}Po daughter by alpha spectrometry. In this thesis a faster ^{210}Po extraction method was developed. It was shown that four of the most used leaching recipes yield results with uncertainties can be quoted on a 95% confidence level. This thesis has presented some of the most used applications of ^{137}Cs and ^{210}Pb radionuclides. Measurement of radionuclide concentrations and calculation of ^{137}Cs and ^{210}Pb inventories and fluxes on different regions of Transylvania, Romania, have been carried out. Data presented here demonstrates that the ^{210}Pb dating method, along with the application of the CRS model and the measurement of the ^{137}Cs radiomarker, can be successfully applied on lakes of various origin such as deltaic lakes, volcanic lakes, glacial lakes, metamorphic lakes and artificial lakes.

As far as methodological applications are concerned, the most commonly used digestion recipe for the preparation of ^{210}Po sources is time-consuming, therefore a more time-effective method was developed where the leaching time can be reduced from 2 weeks to 3-4 hours, while the chemical recovery remains high and good spectra resolution is obtained. When tested on IAEA reference materials, results were in good agreement in case of the two procedures. The best results were obtained in case of samples with high organic material content (peat samples). This extraction method can be used efficiently on medicinal plants, vegetables, tobacco, fertilizers, mushrooms, lichens or animals (bioaccumulation in fish and other marine organisms, herbivores, etc.), when samples need to be analyzed in a short period of time.

Due to experimental limitations and the extreme toxicity of HF, four alternative digestion methods have been tested to determine what acid combination is the most suitable to extract the ^{210}Po trapped inside the crystalline structure of the sediment grains. Sediment samples from ten lakes of five different genesis types (glacial, volcanic, barrier, oxbow and deltaic) with various volcanic, metamorphic and sedimentary geology were analyzed in this comparative study. All analyzed methods have yielded relative uncertainties of 8-9% in the CRS model and all provide a complete extraction of polonium in early peat-bog state lakes. It was concluded that sediments originating from volcanic lakes and lakes with sedimentary and metamorphic mineralogical composition can be digested thoroughly using less harmful acids.

Since ^{137}Cs nuclide is one of the most often used radiomarkers in recent chronological studies measurements that resulted in a ^{137}Cs database over the Transylvania Region of Romania have been carried out. The results of this study also serve as important input information regarding the pollution caused by the 1986 Chernobyl Nuclear Power Plant accident in the region.

Being essential parameters in the ^{210}Pb dating procedure, ^{137}Cs and ^{210}Pb fluxes and inventories were calculated for soil and peat cores from different locations, covering the whole area of Romania. Western Romania was represented by samples from the Occidental Carpathians (two soil samples Ighiel and Băița-Plai and two peat bog samples Mluha and Semenic), Central Romania was represented by Oriental Carpathian samples (soil samples from the Red Lake region and the Mohoș peat bog) and Eastern Romania was characterized by soil samples from the Danube Delta. From the analyzed areas, the Danube Delta shows the least flux and inventory, while peat cores in general have the highest activity concentrations.

In this thesis, lakes from both the fluvial (Matița and Merhei lakes) and marine (Cruhlig Lake) regions of the Danube Delta were investigated using the ^{210}Pb dating method, enabling therefore a representative study of the area. These lakes are susceptible to a series of natural and anthropogenic phenomena, which influence the sediment intake. The 24 major floodings in the past 50 years caused significant changes in the sediment deposition pattern and, as measured in case of the Merhei Lake, an additional intake can originate from aeolian activities. As a precursor of the eutrophication phenomena, higher organic material content characterized by submerged vegetation and turbid water can be observed seasonally. The construction of the Iron Gates Hydroenergetic Power Plant (which also serves as a dam) in 1972 lead to the trapping of an yearly amount of millions of tons of sediment, decreasing therefore the sediment income in the delta area. However, the lakes from the marine delta are prone to multiple sediment transporting and depositing phenomena and show therefore increasing tendencies, thus other natural (e.g. floods, storms, riverbank erosion) and anthropogenic (e.g. channel cutting, dam construction) factors may also influence the sediment income. This data serves as a starting point in understanding the diverse sedimentation patterns in deltaic environments.

Because of the increased sedimentation rates observed in the past decades in the Danube Delta area, a preliminary study aimed at analyzing the tendencies all over Romania was undertaken. The western area was represented by the Vârșolț Lake (Apuseni Mountains, Occidental Carpathians), while for the central region St. Anna Lake was chosen along with the Red Lake and its dam lake (Oriental Carpathians) while for the eastern part the Matița Lake (Danube Delta) was considered. Each of these lakes is influenced differently by natural and anthropic factors. The sedimentation patterns as well as their quantity and quality is influenced by the location of the downstream hydrographical basins characterized by residential, touristic or agricultural areas. All regions are characterized by a rather unstable climatic equilibrium as the physico-chemical and biological quality of the water and the quantity of the transported insoluble material are influenced by the hydrological conditions such as droughts, heavy rainfalls, floods, soil-washings. The sedimentation rates have increased multiply (up to eightfold) in the past decade compared to ones registered in the early eighties. Possible reasons vary from region to region, but include climatic changes, increase of the precipitation pattern and the higher presence of floodings in the catchment areas of the lakes. Another possible cause can be an increase in deforestation.

Results presented in this thesis focus on Transylvania (the central and north-western regions of Romania) and as a comparison, the Danube Delta (eastern area). However, for a complete assessment of Romania, the southern areas could be analyzed. Oltenia (south to the Western and Southern Carpathians) and Muntenia (south to the Eastern Carpathians) are areas characterized by farming regions in the floodplain of the Danube River, having decreasing altitudes from north to south. The lakes in these regions are of various origin and geological composition. The characteristic lakes are glacial and oxbow with predominantly sedimentary and metamorphic rocks, which – if analyzed – could give new insight on the ^{210}Pb and ^{137}Cs inventories and fluxes as well as on the chronology and sedimentation rates of southern Romania.

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