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CLUJ-NAPOCA
Faculty of
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FALLOUT RADIONUCLIDES USED FOR THE QUANTITATIVE ASSESSMENT OF SOIL REDISTRIBUTION

- Summary -

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CLUJ-NAPOCA - 2013

This PhD thesis has been performed with the financial support of the project **"DOCTORAL STUDIES FOR EUROPEAN PERFORMANCES IN RESEARCH INOVATION (CUANTUMDOC)"** POSDRU/107/1.5/S/79407.

The **"DOCTORAL STUDIES FOR EUROPEAN PERFORMANCES IN RESEARCH INOVATION (CUANTUMDOC)"** project POSDRU/107/1.5/S/79407, is a strategic project which has as general objective „*The implementation of the managerial, research and educational strategies for the improvement of the initial training of the future researchers through doctoral studies program, according to the Bologna process, by the development of the research specific proficiency, but also of some other common outcomes: research management, linguistic and communication skills, scientific documentation, elaboration, publication and scientific communication, modern ITC equipment utilization, entrepreneurship transfer of the research results. The human capital development for research and innovation will contribute to long-term doctoral training at European Union level with interdisciplinary concern. The PhD students financial support will provide their participation to national doctoral programs, as well as the research internship in European Union universities and research institutes. The project main objective is the training of a young researcher adapted to the new economic and technological environment having theoretical, practical, economic and managerial knowledge, which will be able to promote the sustainable development and the environmental protection principles.* "

Strategic project funded for period 2010 - 2013

Project funding: **16.810.100,00** RON

Beneficiary: "Gheorghe Asachi" Technical University of Iaşi

Partner: University „Babeş Bolyai" of Cluj-Napoca

Project manager: Prof. univ. dr. ing. Mihai BUDESCU

Partner project manager: Prof. univ. dr. ing. Alexandru OZUNU

List of contents

Abstract 1

Chapter 1. Introduction 2

1.1. General background and research motivation 2

1.2. Aim of the work 5

1.3. Outline of the thesis 6

Chapter 2. Fallout radionuclides (FRNs) as tools in soil erosion research 8

2.1. The use of ^{137}Cs as radiotracer of soil movement 8

2.2. The use of $^{210}Pb_{ex}$ as a soil and sediment tracer 14

2.3. The use of ^{7}Be in soil erosion studies 18

2.4. The use of fallout radionuclides as erosion tracers in Romania 21

Chapter 3. Gamma-ray spectrometry – basic principles 23

3.1. Gamma-emission and radioactive decay 23

3.2. Gamma-ray interactions with matter 25

3.3. Detector response and the shape of the gamma-spectra 27

3.4. Gamma-spectrometric system 28

3.5. Spectrum analysis 30

Chapter 4. The effects of tillage practices on soil erosion and deposition rates using ^{137}Cs and $^{210}Pb_{ex}$ 33

4.1. Introduction 33

4.2. Materials and methods 35

4.2.1. Study site description 35

4.2.2. Soil sampling in the cultivated field 36

4.2.3. Sampling in the reference areas 37

4.2.4. Laboratory analysis 38

4.3. Results and discussions 39

4.3.1. Inventories of ^{137}Cs and $^{210}Pb_{ex}$ at the reference sites 39

4.3.2. Inventories of ^{137}Cs and $^{210}Pb_{ex}$ at the study site 42

4.3.3. Correlation between the radionuclide inventories and the soil texture parameters 44

4.3.4. Soil redistribution rates obtained by means of radiometric models 44

4.4. Conclusions 48

Chapter 5. Uncertainty related to input parameters in radiometric derived-soil redistribution rates model: The case of undisturbed soils 50
5.1. Introduction 50
5.2. Materials and methods 51
5.2.1. Study site and sampling design 51
5.2.2. Laboratory analysis 53
5.2.3. Diffusion and Migration Erosion Model for uncultivated fields 54
5.3. Results and discussions 55
5.3.1. Application of the convection-diffusion equation for the ^{137}Cs depth profile in the reference site 55
5.3.2. Assessment of erosion and deposition rates and uncertainties related to the input parameters of the radiometric model 58
5.3.2.1. The assessment of soil redistribution rates in Jucu area using the DMM 60
5.3.2.2. Reference inventory value 61
5.3.2.3. Chernobyl contribution 62
5.3.2.4. Diffusion coefficient and convection velocity 63
5.3.2.5. Particle size correction factors 64
5.3.3. The application of the ROMSEM model 65
5.3.4. Correlation between ^{137}Cs and the physicochemical soil properties 66
5.4. Conclusions 68

Chapter 6. Spatial distribution of ^{7}Be in soils after heavy rains 70
6.1. Introduction 70
6.2. Materials and methods 71
6.2.1. Study site and sampling 71
6.2.2. Gamma-spectrometric measurements 74
6.3. Results and discussion 75
6.3.1. ^{7}Be depth profile 75
6.3.2. ^{7}Be spatial variability 77
6.4. Conclusions 79

Chapter 7. Specific issues in gamma-ray spectrometry 80

7.1. Measurement validation through intercomparison 80
7.1.1. Introduction 80
7.1.2. Preparation of soil samples used for the intercomparison 82
7.1.3. Results and discussions 83

7.1.3.1. Gamma-spectrometric measurements of soil samples within the radiometric laboratory from Cluj-Napoca 83
7.1.3.1.1. Methodology and equipment 83
7.1.3.1.2. Mass activity determination in soil samples 84
7.1.3.1.3. Internal validation procedure 85
7.1.3.1.4. Experimental approach for self-absorption correction of low-gamma energy of ^{210}Pb 86
7.1.3.2. Gamma-spectrometric measurements within the IAEA Terrestrial Environment Laboratory from Seibersdorf (Austria) 88
7.1.3.2.1. Mass activity determination in soil samples 88
7.1.3.2.2. The determination of the soil elemental composition 91
7.1.3.2.3. Corrections performed in gamma spectrometric measurements 93
7.1.3.3. Determination of ^{210}Pb and ^{226}Ra in soil samples using alpha-ray spectrometry and liquid scintillation counting 99
7.1.3.3.1. Materials and methods 99
7.1.3.3.2. Analysis of ^{226}Ra by alpha-ray spectrometry 105
7.1.3.3.3. Analysis of ^{210}Pb by liquid-scintillation counting 107
7.1.3.4. The evaluation of the intercomparison results 110
7.1.4. Conclusions and recommendations 116

7.2. The calibration of the HPGe detectors from Cluj-Napoca radiometric laboratory 117
7.2.1. Introduction 117
7.2.2. Results and discussions 118
7.2.2.1. Energy and peak width calibration 118
7.2.2.2. Efficiency calibration 119
7.2.2.2.1. Efficiency calibration of the Ortec GEM detector 123
7.2.2.2.2. Efficiency calibration of the Ortec GMX detector 125
7.2.2.3. Natural background control 130
7.2.2.4. External validation through participation in the IAEA Worldwide Proficiency Test (PT) 133
7.2.3. Conclusions and recommendations 135

Chapter 8. General conclusions and future directions 137

Author's declaration 141

References 143

Keywords: *soil erosion, fallout radionuclides, cesium-137, lead-210, beryllium-7, net erosion rate, conversion models, reference inventory, high resolution gamma spectrometry, measurement validation*

Summary of the PhD thesis

Chapter 1. Introduction

The conservation of soil resources is critical to human well-being as consequences of increasing demands for food production in the context of a continuous increasing world population and climate change (*Nguyen et al., 2012*). The present interdisciplinary study comes into prominence with the aim of the application and development of nuclear-based techniques for the assessment of soil erosion and deposition processes on the degraded fields from Romania.

Aim of the work

The thesis concerns the applications of fallout radiotracers (^{137}Cs, ^{210}Pb$_{ex}$ and ^{7}Be) for the assessment of soil erosion and deposition and also specific problems related with the main spectrometric technique employed for the radionuclide determination in soil samples.

The **first major goal** was to test and improve fallout radionuclide (FRN) methodologies for the estimation of soil redistribution to cultivated and uncultivated fields from NW Romania. The specific objectives of the work performed on the application of FRNs to soil erosion study can be summarised as:

(i) to simultaneously use ^{137}Cs and ^{210}Pb$_{ex}$ inventories for the medium-term and long-term assessment of soil erosion and deposition rates on Romanian cultivated soils and also for determining the effects of changes in tillage practices on soil erosion magnitudes in our country;

(ii) to estimate the soil erosion and deposition rates affecting Romanian pastureland using the ^{137}Cs method, and to highlight the impact of the input parameters (e.g. the reference inventory, the Chernobyl contribution, diffusion coefficient and the convective velocity for ^{137}Cs, particle size correction factor) used in the Diffusion and Migration Model (DMM) for uncultivated fields on the derived estimates;

(iii) to verify one of the key assumptions underlying the application of ^{7}Be technique, i.e. the uniformity of radionuclide deposition associated with erosive events (heavy rainfalls).

The **second major goal** of the thesis deals with specific issues of the radiometric techniques used for the determination of FRNs in soil samples, namely gamma-ray spectrometry. Although many spectrometric techniques (gamma spectrometry, alpha spectrometry and liquid scintillation counting) are available for the determination of the total ^{210}Pb and ^{226}Ra (needed to determine the excess ^{210}Pb), ^{137}Cs and ^{7}Be are commonly measured in environmental samples only by means of gamma spectrometry. One of the greatest advantages of this technique is that all radionuclides of interest can be determined by only one spectra acquisition, making possible the quick determination of FRNs within a large number of samples that are needed in soil redistribution research. Yet, the reliability, accuracy and precision in measurement is of high importance within the scientific community, and erroneous results in FRN activities can lead to major discrepancies in assessed erosion rates. In this respect, the following specific objectives were highly important for this work:

(i) internal check and external validation of the gamma-measurements performed within the radiometric laboratory of the Faculty of Environmental Science and Engineering (FESE) through

intercomparison of the radionuclide activities determined by gamma spectrometry and/or alternative methods (alpha- and beta-spectrometry) in the same soil samples between 5 international laboratories;

(ii) the efficiency calibration of the high-purity germanium gamma-detectors (HPGe) from the faculty laboratory and further validation of the results through participation to the worldwide *Proficiency Test* organized by the International Atomic Energy Agency (IAEA).

The thesis is structured in 8 chapters: one introductory part, two chapters which present literature data regarding the application of fallout radioisotopes in soil erosion studies and the basic principles of gamma-spectrometry, four chapters which describe the original contributions of the thesis and one final chapter which summarises the major conclusions and presents aspects of future research in the field. Alpha- and beta-spectrometric analysis and parts of the gamma-measurements were carried out by the author during a research stage of 7 months at the *IAEA Seibersdorf Laboratories*, Austria.

Chapter 4. The effects of tillage practices on soil erosion and deposition rates using ^{137}Cs and ^{210}Pb$_{ex}$

As a result of change in the government policy at the beginning of 1990s, the Romanian land resource was reassigned to the original landowners resulting in a great number of small farms and the division of the agricultural land into narrow plots and fields, favouring the increase of soil erosion extent and magnitude in many Romanian territories. The objectives of this work are: (i) to assess the soil erosion and deposition rates on Romanian cultivated soils using both ^{137}Cs and ^{210}Pb$_{ex}$ (excess lead) inventories in bulk and incremental soil core samples, (ii) to study the effects of tillage practices on soil erosion pattern, and (iii) to determine the potential of ^{210}Pb$_{ex}$ as tracing technique (in complement to the ^{137}Cs method) to estimate soil redistribution magnitude.

The agricultural area under investigation is located in the central part of the Transylvanian Plain, in 'Mureş' watershed (N46°36', E24°06'). The soil type in this area is a Cambic Chernozem sandy textured clay, with a medium slope gradient of 10%.

According to information provided by local farmers, this field was cultivated across the slope by private owners from 1946 to 1959. From 1959 to 1991, after the institution of the local agricultural cooperative (CAP), all agricultural plots were ploughed together up and down slope increasing soil erosion processes.

The technique of the radiotracer is based on a comparison between the radionuclide inventories in the study area and the mean inventory in a flat 'reference area', not affected by soil redistribution processes. Collection of soil samples was undertaken at the beginning of October 2011 during two field campaigns, following the way of two transects in the study field. The first reference site selected was a courtyard of an abandoned house covered by generous vegetation and the second reference site was selected in a relatively flat area of a country path under consistent grass cover, on the back of the hill. The activities of ^{137}Cs and ^{210}Pb$_{ex}$ in soil samples were determined by non-destructive gamma spectrometry using an 'N' type high purity germanium detector (34% relative efficiency). The self-absorption correction factor was

experimentally determined and applied for the total ^{210}Pb, considering the lower gamma line at 46.5 keV. $^{210}Pb_{ex}$ was determined as difference between the activities of the total ^{210}Pb and ^{226}Ra from soil. For physicochemical characterization additional top soil samples were collected.

The ^{137}Cs depth profiles showed similar patterns for the two reference areas selected. The ^{137}Cs areal concentration for the incremental soil core of the first reference site totalled 6640 Bq m^{-2} (Figure 1a). Most of the ^{137}Cs content (92%) was found in the first 15 cm of the soil profile and its presence was not detected below 30 cm depth. For the second reference site, the ^{137}Cs inventory had a value of 4560 Bq m^{-2} (Figure 1b).

The mean reference inventory for the 10 soil profiles was determined at 5460 ± 880 Bq m^{-2}. The incremental and bulk reference inventories ranged from 4560 Bq m^{-2} to 6950 Bq m^{-2}, and indicated an admissible degree of spatial variability in ^{137}Cs areal activities (the coefficient of variation (CV): 16%). Analyses have also been made to determine the $^{210}Pb_{ex}$ activities in the incremental soil core from the first reference site, but the ^{226}Ra values (which gave the supported ^{210}Pb) were higher or equal compared with the total ^{210}Pb, resulting in no $^{210}Pb_{ex}$. The presence of abundant vegetation, the lack of significant fresh $^{210}Pb_{ex}$ inputs or increased radium activities in soil, and thereby increased concentrations of supported ^{210}Pb, can clearly limit the use of $^{210}Pb_{ex}$ in soil redistribution investigations, if the reference value cannot be determined. The measured $^{210}Pb_{ex}$ inventory at the second reference site was 9640 Bq m^{-2}, 95% confidence.

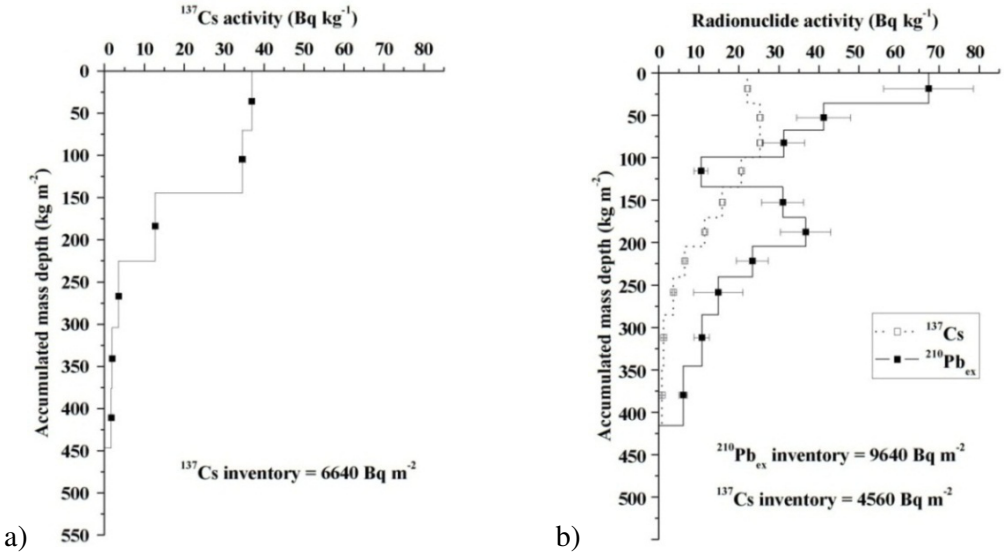


Figure 1. a) ^{137}Cs depth distribution at the first reference site; b) ^{137}Cs and $^{210}Pb_{ex}$ depth distributions at the second reference site.

Inventories of ^{137}Cs and $^{210}Pb_{ex}$ at the study site highlighted different values for the two transects (Figure 2), mainly caused by the field topography and different ploughing practices of the plots where the soil cores were collected; however they had similar trend along the slope. Most of the ^{137}Cs inventories (except for the two higher points of each transect) were above the

reference value, on both transects, reflecting potentially an accumulation of soil and associated ^{137}Cs. However, most of the $^{210}Pb_{ex}$ inventories were smaller than the reference value (except for sampling points located at the bottom of the field), showing different temporal characteristics of soil redistribution comparing with ^{137}Cs. The levels of $^{210}Pb_{ex}$ and ^{137}Cs inventories measured at the different sampling points within the field were influenced by the change in tillage practices associated with the liquidation of the local CAP in 1991. In the last 21 years, the field was cultivated in narrow plots across the field (except two wider plots located in the higher and the lowest part of Transect 1) and hence reducing the soil loss.

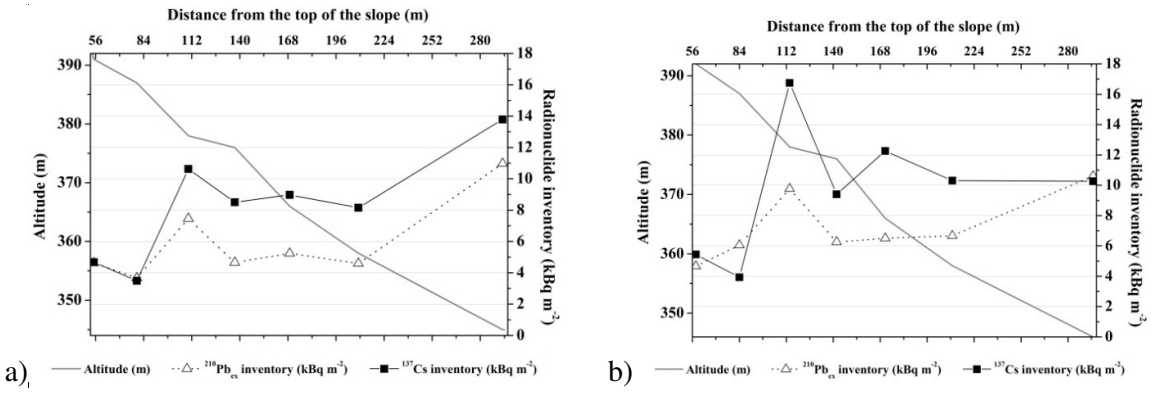


Figure 2. a) ^{137}Cs and $^{210}Pb_{ex}$ inventories (kBq m^{-2}) for Transect 1; b) ^{137}Cs and $^{210}Pb_{ex}$ inventories (kBq m^{-2}) for Transect 2;

Significant positive Pearson's correlations were found between the ^{137}Cs and $^{210}Pb_{ex}$ inventories ($r = 0.76$, $p = 0.002$), and between the medium sand content (with the diameter from 0.25 mm to 0.5 mm) and the ^{137}Cs ($r = 0.71$, $p = 0.004$) and $^{210}Pb_{ex}$ ($r = 0.62$, $p = 0.019$) inventories, reflecting the increased inventory at depositional sites, which are sand rich.

The conversion of $^{210}Pb_{ex}$ and ^{137}Cs inventories into soil redistribution rates (t ha^{-1} yr^{-1}) was done using two radiometric models employed in cultivated fields for ^{137}Cs: Mass Balance Model 2 (MBM2) and Mass Balance Model 3 (MBM3), and for $^{210}Pb_{ex}$: Mass Balance Model (MBM) and Mass Balance Model with Tillage (MBMT) *(Zapata, 2002)*.

Table 1. Parameters used in radiometric conversion models

Parameters	Transect 1	Transect 2
Bulk density (kg m^{-3})	1570	1623
Particle size factor P	1.258 - 2.108	0.737 - 2.391
Particle size factor P'	0.366 - 0.854	0.469 - 0.832
Reference inventory ^{137}Cs (Bq m^{-2})	5460	5460
Reference inventory $^{210}Pb_{ex}$ (Bq m^{-2})	9640	9640
Proportional factor	0.7	0.7
Relaxation depth (kg m^{-2})	4	4
Sampling year	2011	2011
Tillage depth (kg m^{-2})	314	324.6
Tillage constant (kg m^{-2} yr^{-1})	258.6	363.6

The ^{137}Cs method gives an estimation of the soil redistribution processes for the last 25 years, considering the high contribution of the Chernobyl Cs (~73%) in the area. The radical changes in the Romanian farming practices, starting at the beginning of the 1990s, led to most plots from the study site being ploughed across the field, resulting in erosion reduction. The results were in the same order of magnitude when comparing the values obtained by applying the two radiometric models for ^{137}Cs. The high erosion rate values determined by the $^{210}Pb_{ex}$ technique reflect particularly well the up and down ploughing practices from the time of the local agricultural cooperative (CAP), starting in the late 1950s. The resulting net erosion rates from $^{210}Pb_{ex}$ measurements, for both transects demonstrated moderate to high risk for surface erosion, averaged over the last ~ 60 years.

The combined application of ^{137}Cs and $^{210}Pb_{ex}$ measurements represents the first attempt to quantify changes in soil erosion magnitude in Romania. Long-term estimates provided by $^{210}Pb_{ex}$ revealed moderate to high risk of soil erosion, mostly due to the improper cultivation practices up and down slope during the local agricultural cooperative, integrating a period of 30 years (1959 - 1991). Along with the changes in the cultivation system (across the slope) at the beginning of 1990's, the erosion risk decreased and the middle-term redistribution rates provided by the ^{137}Cs approach showed a preponderant accretion process in the field. The values derived from the two models were comparable and also showed a good correlation with the measured ^{137}Cs inventories. The study highlighted very well the influence of the cultivation practices on soil erosion processes given by the combined use of the two radiotracers with different origins and half-lives. The results demonstrate a close relationship between the measured inventories of ^{137}Cs and $^{210}Pb_{ex}$ and the derived soil redistribution rates provided by both radiometric methods.

Chapter 5. Uncertainty related to input parameters in radiometric derived-soil redistribution rates model: The case of undisturbed soils

There is still a lack of information and guidance for an accurate determination of some of the parameters involved in the radiometric models of soil redistribution. This is especially the case when a nuclear accidental release has a major contribution to the overall ^{137}Cs inventory in soil, which could be a noticeable source of uncertainty in erosion and sedimentation radiometric-derived evaluation. The objectives of our study were:

(i) to apply the convection-diffusion equation for the ^{137}Cs soil depth profile in a Romanian reference site using the least square fitting procedure for the determination of the effective diffusion coefficient and the convective velocity of both Chernobyl and bomb-originated cesium;

(ii) to highlight the impact of the input parameters (the reference inventory, the Chernobyl contribution, the diffusion coefficient, D and the convective velocity, v for ^{137}Cs, the particle size correction factor) used in the Diffusion and Migration Model (DMM) for uncultivated fields;

(iii) to assess the soil erosion and deposition rates affecting Romanian pastureland using ^{137}Cs.

Soil samples were collected in a degraded pastureland from Cluj county, Somes watershed, located at the west extremity of the Transylvanian Plain (N46º52', E23º45'). This grassland has a complex topography with prominent acclivities and average slope of 10%. Two sampling

campaigns were conducted in November 2010 and July 2011. Twelve soil cores were taken to 40 cm along two parallel transects. To establish the mean reference value for the ^{137}Cs inventory, five sampling points (one incremental and 4 composite soil from 3 nearby sampling points) were randomly considered in a flat undisturbed grassed terrace situated about 500 m west from our study site. Two HPGe detectors, one 'N' type and one 'P' type with relative efficiencies of 34% and 30% respectively, were employed for the gamma analysis. Several soil physicochemical properties were determined by analysing additional soil samples corresponding to each soil core, such as texture, pH, oxidation-reduction potential (ORP), electrical conductivity (EC), total dissolved solids (TDS), water content (%), bulk density and humus content (%).

The convection-diffusion model was fitted to our available experimental data set to describe the ^{137}Cs behaviour and penetration in soil after both contaminations occurred (firstly in 1963, and subsequently in 1986). The ^{137}Cs inventory for the investigated area was approximated based on the model of *Sarmiento and Gwinn (1986)*. A ^{137}Cs areal activity of 766 Bq m^{-2} could be expected for the nuclear-bomb deposit at the time of sampling. The difference of the inventory between the total experimental content of ^{137}Cs within the incremental soil profile and the nuclear tests represents the expected Chernobyl derived ^{137}Cs, reaching 1823 Bq m^{-2}. The variation of these parameters was limited to a maximum of 5% for the fitting procedure.

Applying the least square fitting, the v_2 = 0.350 cm yr^{-1} and D_2 = 0.321 cm^2 yr^{-1} values were obtained for the depth migration of bomb-derived cesium, and v_1 = 0.097 cm yr^{-1} and D_1 = 0.15 cm^2 yr^{-1} for the Chernobyl cesium distribution in soil. The depth migration of ^{137}Cs with distinctive origin appeared to be quite different. As highlighted by Figure 3, the deeper ^{137}Cs peak profile which appeared around 15-20 cm indicates a broader longitudinal dispersion of nuclear tests-derived cesium, while the peak associated to the Chernobyl event was found close to the soil surface at 2.5-5.0 cm.

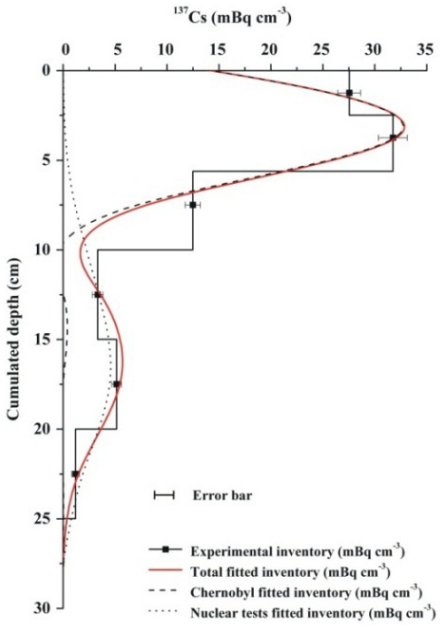


Figure 3. Experimental depth distribution of ^{137}Cs in the reference site, Chernobyl and the nuclear tests fitting curves

The two transects (T1 and T2) do not show similar patterns of downslope variation in cesium inventories (Figure 4). The distinction is mostly due to the field topography, with prominent acclivities and falls in the north-eastern part of the study field (first transect).

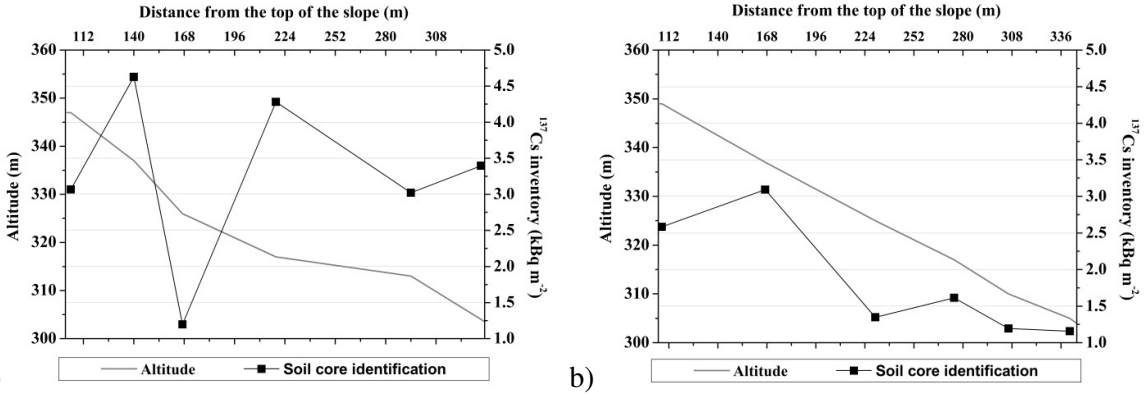


Figure 4. a) ^{137}Cs inventories (kBq m^{-2}) for T1; b) ^{137}Cs inventories (kBq m^{-2}) for T2.

The averaged ^{137}Cs reference site inventory of the five composite soil profiles was measured by *Iurian et al. (2012)* at 3160 ± 867 Bq m^{-2} (areal inventory ± standard deviation, 2σ confidence interval), with a coefficient of variation (CV) of 27%. The key parameters needed for running the DMM radiometric model for cesium are: (i) the reference inventory (Bq m^{-2}), (ii) the migration velocity, v (kg m^{-2} yr^{-1}) and the effective diffusion coefficient, D (kg^2 m^{-4} yr^{-1}), (iii) the Chernobyl contribution (%) to the total ^{137}Cs inventory in the study area, (iv) the particle size correction factors (P and P'), and (v) the relaxation depth (H), for which the input value of 5 kg m^{-2} is considered for undisturbed fields *(Walling et al., 2002)*. These parameters have been determined using our experimental data set, apart from the H value, which is difficult to determine empirically as the current ^{137}Cs fallout flux is negligible, being impossible to replicate the past situation during the main period of fallout.

Because of the preponderant Chernobyl ^{137}Cs in the investigated area relative to the nuclear tests-derived ^{137}Cs (ratio between Chernobyl ^{137}Cs and bomb-derived ^{137}Cs about 4:1), it can be considered that the results derived from the ^{137}Cs data modelling give estimates of the yearly average soil redistribution rates over the 1986-2010 period (2010 being the sampling year).

Table 2. List of parameters used in the radiometric conversion model (DMM)

Parameter	Value
^{137}Cs inventories (Bq m^{-2})	1157 - 4629
Particle size correction factor P	1.01 - 1.67
Particle size correction factor P’	0.33 - 0.76
Reference inventory ^{137}Cs (Bq m^{-2})	3160
Relaxation depth, H (kg m^{-2})	5
Sampling year (yr)	2010
Chernobyl contribution (%)	72%
Distribution coefficient D (kg^2 m^{-4} yr^{-1})	28.2
Convection velocity v (kg m^{-2} yr^{-1})	2.012

Table 3. Assessment of soil redistribution rates

Assessment of soil redistribution rates	Value
Gross erosion rate (t ha^{-1} yr^{-1})	-12.2
Mean erosion rate in the eroding areas (t ha^{-1} yr^{-1})	-16.2
Mean deposition rate for the depositional areas (t ha^{-1} yr^{-1})	4.2
Net erosion rate (t ha^{-1} yr^{-1})	-6.6
Sediment delivery ratio (%)	54%

The author demonstrated that neglecting the contribution of only one of these input parameters, the final results can be significantly over- or under- estimated (Figure 5).

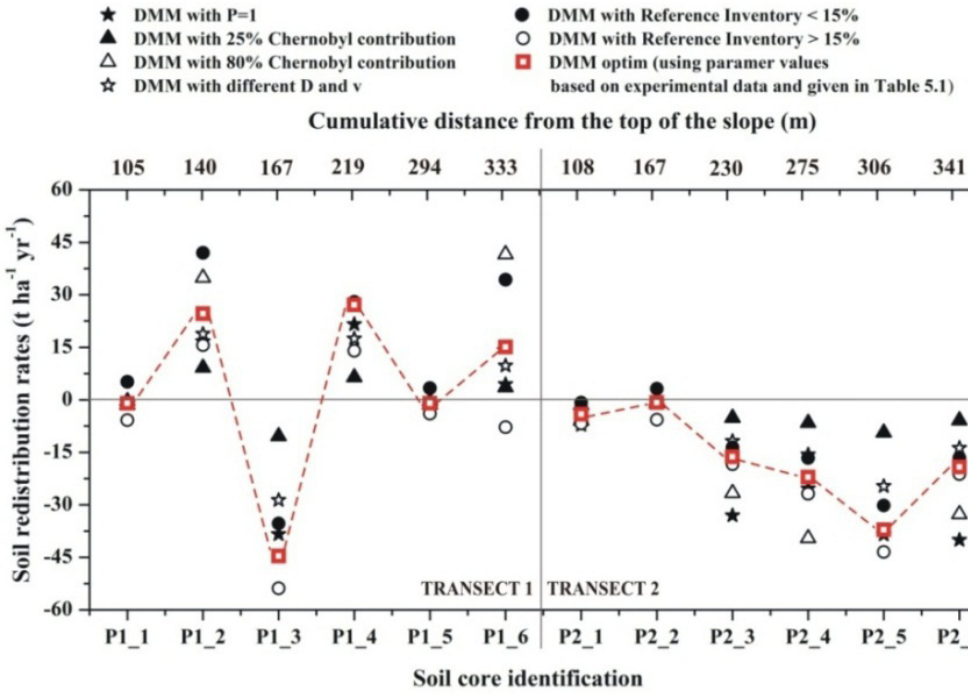


Figure 5. Sensitivity analysis of the input parameters in DMM radiometric derived-soil redistribution rates model (negative values of the soil redistribution rates represent net erosion and positive values net sedimentation)

The Romanian Soil Erosion Model (ROMSEM) (*Moţoc et al., 1979*) gives estimates of the water erosion rates (t ha^{-1} yr^{-1}) based on: rainfall record, land use information, field observations and topographic data. The ROMSEM calculated value of -4.17 t ha^{-1} yr^{-1} is comparable with the medium-term net erosion rate obtained with the DMM.

Soils of the investigated area are all fine textured, however a differentiation could be made between the two transects. The humus content of the collected soil samples from Jucu area varied from low (about 2%) to high (about 5%), with lower values recorded for the second transect, where also the erosion processes appeared to be marked. Using the statistical software GraphPad Prism 6, the Pearson's correlation coefficients between ^{137}Cs total inventories (0–40 cm) and the different phisicochemical parameters measured in the 12 soil samples, showed that

the ^{137}Cs inventory was significantly positive correlated (0.05 level) with the humus content (r = 0.63) and the ORP (r = 0.59) and negatively correlated (r = -0.60) with the pH.

By application of the diffusion-convection equation for the ^{137}Cs depth profile in our reference area the experimental results were well fitted. An estimation of the bomb-derived ^{137}Cs amount was determined according to the precipitation regime and the geographical location of our investigated area. Thus, the Chernobyl contribution in the selected reference site was determined at 72% of the total ^{137}Cs inventory. The effective diffusion coefficient and the convective velocity of ^{137}Cs for undisturbed soil were statistically derived for both Chernobyl and nuclear tests using the convection-diffusion equation. The assessment of net soil erosion rate for Romanian pastureland using the DMM for ^{137}Cs was -6.6 t ha^{-1} yr^{-1}. This result is comparable with the predicted medium-term soil erosion rate obtained by ROMSEM.

As highlighted in this contribution, the DMM is very sensitive to the different input parameters and particular care is required in their estimation. The predicted erosion rates are positively related to D, v and Chernobyl contribution, but inversely related to the particle size correction factors. As suggested by the findings of our study, integrating a way in the conversion models to assess the impact of the uncertainty around the model's parameters will be a big asset. Confidence intervals should be established for the reliable estimates of the derived redistribution rates and upper and lower bounds should be considered and integrated in the conversion models.

Chapter 6. Spatial distribution of ^{7}Be in soils after heavy rains

The experiment and gamma-measurements of soil samples were carried out by the author at the *IAEA Soil and Water Management & Crop Nutrition Laboratory* from Seibersdorf, Austria, over a period of 4 months research stage, during which Ms. Andra-Rada Iurian has worked under the supervision of Mr. Long Nguyen, Mr. Gerd Dercon and Mr. Joseph Adu-Gyamfi.

The spatial uniform distribution of the pre-existing ^{7}Be and the one associated with the rainfall event represent key assumptions in soil erosion studies. Due to the limited number of samples that must be measured in a relatively short time because of the rapid decay of beryllium-7, these assumptions are not verified for the reference site. The objective of the present research was to assess the spatial variability of ^{7}Be after two consecutive heavy rainfalls within a small plot in Seibersdorf experimental field.

The study field which meant to be validated as reference site was selected in a flat area, unvegetated and undisturbed since the last tillage in autumn 2011. The study site was situated 45 km south of Vienna (N47°58'; E16°30'), Lower Austria, on the experimental field of the IAEA Laboratory from Seibersdorf. After an extended period with rains of relative small intensities and with no evident erosive influence on soils, two consecutive storm events occurred in Seibersdorf area at the end of July 2012 producing a total rainfall of 81 mm in 6 days. Considering the regional and synoptic patterns characterised by *Seibert et al. (2007)*, which can cause the appearance of heavy precipitation events in Austria, and knowing that high rainfall amounts of 33.6 mm accumulated in 12 hours in 20-21 of July and 28.2 mm in 3 hours in 25 of July, respectively, these events could come under heavy rainfalls.

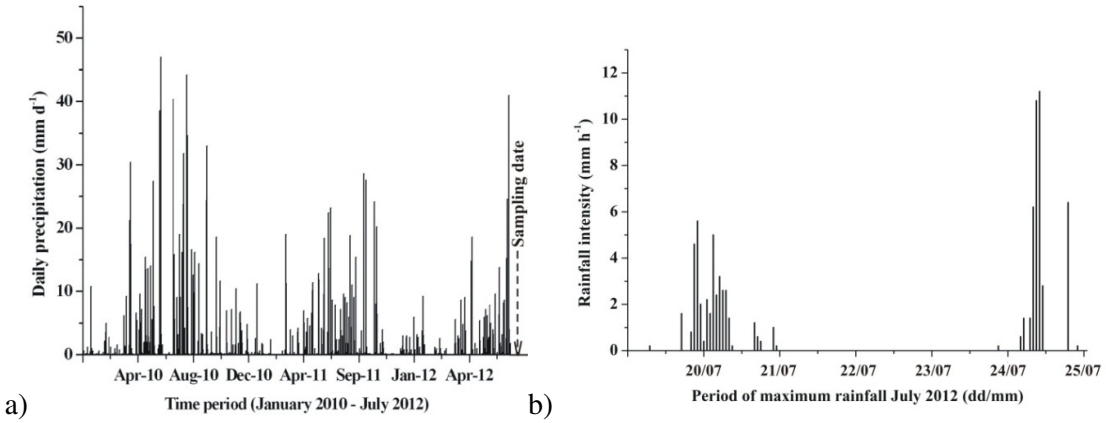


Figure 6. a) Daily precipitation record (mm d^{-1}) for the study site during the period January 2010 - July 2012; b) hourly precipitation data for the period with maximum rainfall intensities

In order to reach all detectable ^{7}Be, the soil profiles were collected to a depth of 40 mm. All soil cores were sampled using the Fine Soil Increment Collector (FSIC), developed and tested by the IAEA Laboratory from Seibersdorf *(Mabit et al., 2013)*.

Figure 7. Soil core inside the stainless steel tube of the FSIC: (a) during sampling, and (b) during the incremental scraping

Ten soil cores were collected from the investigated site. For one incremental soil core, 2.5 mm slices were considered, while the other cores were sectioned in 20 mm slices to avoid the ^{7}Be dilution in the whole soil mass. Gamma-spectrometric analyses were performed using a HPGe Ortec detector, with 110% relative efficiency.

The ^{7}Be depth profile in Seibersdorf soil showed an exponential decrease until 25 mm, as expected (Figure 8), with maximum activity (13.7 Bq kg^{-1}) recorded in the first 2.5 mm layer. The relaxation mass depth (h_0) for the investigated site was obtained from the exponential equation fitting (Figure 8) and had a value of 6.9 kg m^{-2}. An average inventory of 312 Bq m^{2} was calculated for the 10 points within the selected area. This mean value corresponded to a total rainfall of 295 mm in the last 5 months prior sampling, as this represents the timing in which the highest ^{7}Be activity at soil surface will decay until the minimum detectable activity (MDA).

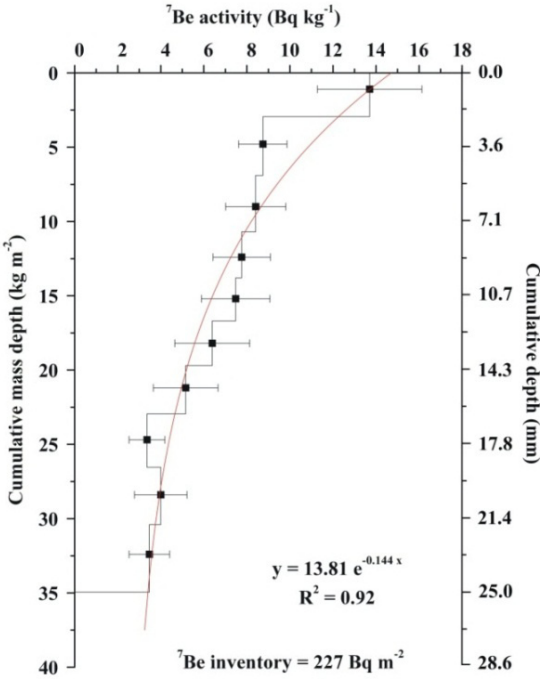


Figure 8. The vertical distribution of ^{7}Be in soil. Error bars represent relative combined uncertainties at 95% confidence level.

Table 4. ^{7}Be inventories (Bq m^{-2}) for the sampling points and the summary statistics

Soil profile identification	^{7}Be inventory (Bq m^{-2})	Combined standard uncertainty (Bq m^{-2})
P1	227	84
P2	302	45
P3	280	47
P4	329	51
P5	232	50
P6	332	70
P7	294	60
P8	392	81
P9	334	50
P10	401	73
Aritmetic mean (Bq m^{-2})	**312**	
Standard deviation (STD)	**58**	
Coefficient of variation (CV%)	**19%**	

The resulted coefficient of variation (CV) has an acceptable value of 19% for a flat undisturbed field with no evidence of soil movement after the occurrence of heavy rains. Thus, the experimental site from Seibersdorf can be considered as suitable reference site for further investigation on soil erosion using ^{7}Be.

The use of fallout ^{7}Be in soil and sediment dynamics is still in its incipient phase and further work is required to fully exploit its potential. This study presents preliminary results on spatial

variability of ^{7}Be in soil, in order to validate a key feature of the radiotracer behaviour, which has to be considered when using it as tracer of soil movement.

The coefficient of variation of 19%, obtained for a small plot showed an acceptable degree of variability. It also demonstrates the suitability of the experimental field as reference site within forthcoming soil redistribution researches. However, there is a need to better understand the ^{7}Be depositional flux in this area and to provide further validation of the assumptions related with the use of ^{7}Be in soil redistribution studies under wider fields.

Chapter 7. Specific issues in gamma-ray spectrometry

Measurement validation through intercomparison

The analytical performance of the present laboratory procedure employed for gamma measurements was not externally evaluated through participation in worldwide proficiency tests or intercomparison exercises. However, in 2007 efforts have been made to validate the incipient methodologies used for gamma-measurements *(Timar et al., 2007)*. Thus, an intercomparison exercise was conducted between international radiometric laboratories, with particular interest in a suite of natural radionuclides (including the anthropogenic ^{137}Cs) in soil samples in order to validate the accuracy of the gamma-measurements performed in the faculty laboratory, as external validation procedure. Two laboratories from Hungary *(Central Radiological Laboratory, Hungarian Agricultural Authority, Budapest and Institute of Radiochemistry and Radioecology, University of Pannonia, Veszprem)* and one radiometric laboratory from Austria *(Division of Physics and Biophysics, Department of Materials Research and Physics, University of Salzburg)* were involved in this intercomparison. Additionally, the samples were measured within the *IAEA Terrestrial Environment Laboratory from Seibersdorf* during the research stage performed by Ms. Andra-Rada Iurian. Three types of measurements were performed at the IAEA Laboratories for these intercomparison exercise: (i) two gamma-measurements in different sample containers (cylindrical, B87 and disk, B5), and (ii) alpha- and beta analysis as alternative method for ^{226}Ra and ^{210}Pb determinations.

This intercomparison exercise aimed to: (i) check the accuracy of the analytical results produced by the FESE Laboratory from Cluj-Napoca and used by the author to derive soil erosion estimates for the Transylvanian Tableland, (ii) find remedial actions for the gamma-procedure employed by the faculty laboratory, where failures in measurements performance is detected, (iii) check the comparability of the measurement results within participant laboratories, and (iv) check the most suitable analytical technique for the determination of the proposed radionuclides in soil samples.

The samples to be measured contained soil from an undisturbed court yard of an old house from Mureş area (Romania), and represented a whole soil core, until 40 cm, with 5 cm layers, used as reference soil profile within a study of soil erosion assessment (*Iurian et al., 2013)*. The soil was air-dried and then oven-dried 24 hours at 105^{0}C. After manually grinding and sieving to 1 mm, the material was enclosed in plastic containers (named B5 disk geometry) with 72.9 mm external diameter, 1.16 mm wall and bottom thickness, and 14 mm height, sealed with electrical

band and stored for minimum three weeks to reach equilibrium between ^{226}Ra, ^{222}Rn and their progenies. Eight soil samples circulated amongst the participant laboratories for the determination of ^{137}Cs, ^{226}Ra, ^{210}Pb, ^{40}K and ^{228}Ac mass activities (Bq kg^{-1}, dry weight) using the methods applied in each laboratory routine work, basically gamma-ray spectrometry.

Two Ortec HPGe detectors are currently in use within the Alpha and Gamma Spectrometric Laboratory from Cluj-Napoca: (i) one Ortec P-type coaxial detector (GEM30P4) with Al window, 30% relative efficiency and a resolution of 1.85 keV at 1.33 MeV line of ^{60}Co, and (ii) one Ortec N-type coaxial detector (GMX30P4-ST) with Be window, 34% relative efficiency and a resolution of 1.92 keV at 1.33 MeV line of ^{60}Co. The gamma-ray spectrometry facilities also incorporate passive background shielding (old lead tower of 8 cm thickness with an additional Cu shield inside the tower, of 3 mm).

The gamma-measurements performed within the faculty laboratory employed a direct calibration method and certified reference materials sources of known activity (IAEA-375, IAEA-312, IAEA-447). The disadvantage of the method is that only the radionuclides present in the reference material can be measured. Corrections were made for background and for decay with the sampling day (October 1st, 2011). An additional self-absorption correction was employed for ^{210}Pb using an experimental approach. The internal validation procedure employed two distinctive approaches: (1) replicate measurements of soil samples, using the same direct calibration method and the same reference source, and (2) measurements of secondary certified standard samples with similar composition and geometry.

The experimental approach employed for self-absorption correction of low-gamma energy of ^{210}Pb relies on the transmission procedure initially proposed by *Cutshall et al. (1983)* and modified by *Khater and Ebaid (2008)*. The self-absorption factor used in the ^{210}Pb activity determination, with respect to the IAEA-447 reference material sample was given as the ratio between the relative transmission factor for the IAEA-447 and the relative transmission factor for each soil sample. Neglecting the self-absorption effect for the low gamma-energy of ^{210}Pb, would result in the underestimation of its activity concentration in soil samples from Zau by about 16.5%.

The gamma-measurements of the eight soil samples used in the intercomparison exercise were carried out by the author at the *IAEA Terrestrial Environment Laboratory* from Seibersdorf, Austria, over a period of three months research stage, during which Ms. Andra-Rada Iurian worked under the supervision of Ms. Alessia Ceccatelli, PhD. The activity of natural-occurring radionuclides (^{210}Pb, ^{226}Ra, ^{228}Ac, ^{40}K, ^{235}U) and anthropogenic ^{137}Cs in soil samples was determined using two broad-energy Canberra detectors, BE5030P and BE2825, with carbon-epoxy window, shielded with 15 cm of lead. Both detectors were efficiency calibrated with a volumetric radioactive source of mixed radionuclides, in B87 cylindrical geometry (inside diameter 55.91 mm, wall thickness 1.34 mm, bottom wall 1.82 mm) with density 0.985 g cm^{-3}, certified by the Czech Metrology Institute. For the internal validation of the present measurements, the IAEA-447 reference material was used.

To take account of the effect of sample geometry when determining the sample activity for a specific radionuclide, the soil samples were measured both in the original B5 geometry and in B87 geometry. Comparing the two measurements, higher discrepancies were highlighted by ^{226}Ra due to the strong correction for the interfering peak of ^{235}U and the high statistic uncertainty which accompanied the ^{235}U activity determination by gamma spectrometry.

To account for differences between the automated and manually evaluation of the photopeak efficiency at different energies, activity results using the LabSOCS automated derived efficiencies were also reported. The relative bias (%) between the calculated mass activity of each radionuclide using the experimental true photopeak efficiency, and the activity obtained with the LabSOCS efficiency ranged between -14% (for ^{40}K measured with BE2825 detector) and 12% (for ^{210}Pb), for all samples and each geometry (B87 and B5).

Decay-corrections have been applied for the multigamma standard solution with the reference time given in the Certificate during the efficiency calibration procedure, and for the soil samples with the reference date of the field sampling (October 1st, 2011). Corrections for geometry and self-attenuation effect, true coincidence summing corrections for standardised solution and samples and corrections for background and interfering radionuclides were also considered.

The elemental composition and mass attenuation coefficients of the soil used in intercomparison are required to compute the correction factors for radionuclide activity determinations. The XCOM: Photon Cross Section Database *(Berger and Hubbell, 1987)* was further on used to calculate the total attenuation coefficients for the soil samples from Zau. The evaluation of the soil matrix composition was performed by X-ray fluorescence spectrometry using the X-Press 3635 and EDXRF spectrometer (Spectro XLab 2000) from the *IAEA Physics Section Laboratory, NA Division of Physical and Chemical Sciences* from Seibersdorf, during the research stage of Ms. Andra-Rada Iurian at the IAEA Laboratories.

The ^{210}Pb and ^{226}Ra content in soil samples was independently evaluated by liquid-scintillation counting (LSC) and alpha spectrometry, as alternative methods to gamma-ray spectrometry. These measurements were carried out by the author at the *IAEA Terrestrial Environment Laboratory* from Seibersdorf, over a period of three months research stage, during which Andra-Rada Iurian worked under the supervision of Mr. Kis-Benedek Gyula, PhD.

The method presented here is commonly applied in the practice of the IAEA Terrestrial Environmental Laboratory *(Shakhashiro et al., 2012; IAEA, 2010)* and it is based on a simple but efficient simultaneous separation of Pb and Ra from Ba *(Sill, 1987; Vajda et al. 1997).* After the radiochemical separation procedure, ^{226}Ra is determined by isotope dilution alpha spectrometry and ^{210}Pb by liquid scintillation counting. Sr Spec resin was chosen for the simultaneous separation of lead and radium. In the case of lead, the chemical recovery was checked gravimetrically, using Pb^{2+} inactive carrier in the form of $Pb(NO_3)_2$ solution, while for ^{226}Ra analysis, the ^{225}Ra (daughter of ^{229}Th) has been used. The determination of chemical recovery is possible via ^{217}At at 7.07 MeV using ingrowth decay equations, as ^{217}At shows no interferences with other alpha-peaks *(Jia et al., 2006, 2007).*

Two replicates were prepared for each of the four soil samples considered for radiochemical determinations, together with one IAEA soil reference material (IAEA447) for quality assurance and one blank sample for background check. Alpha-spectrometic system (Ortec OCTETE, with PIPS detectors) and Canberra Genie 2000 software were used for the determination of radium activity in soil samples. The ^{225}Ac activity reached a maximum after 17.4 days, growing into a maximum of 44% of the initial ^{225}Ra spike.

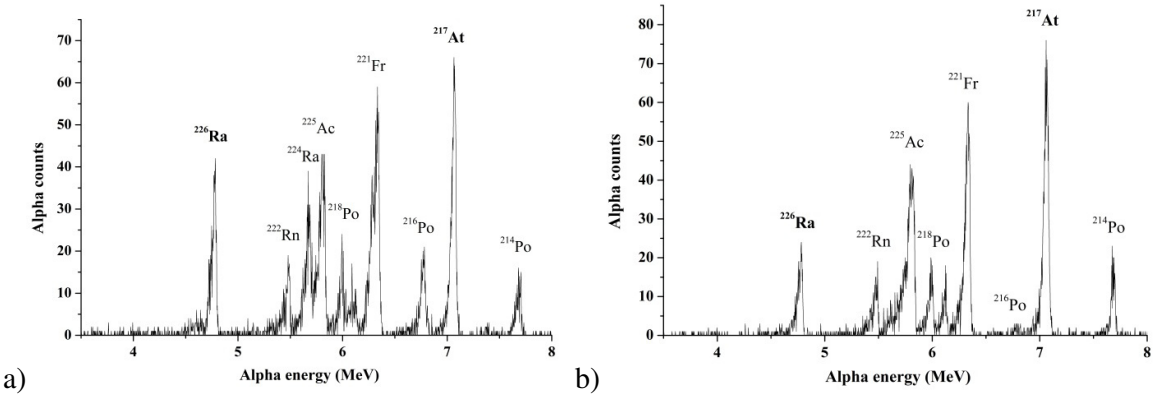


Figure 9. Alpha spectra of radium in a soil sample using ^{225}Ra tracer and registered: (a) 3.7-8 days after the Ra/Th separation; (b) recounted about 17 days after separation

Beta-spectrometric analyses of ^{210}Pb sources were performed with Wallac Quantulus 1220 Liquid Scintillation Spectrometer. Three running measurements were performed (200 minutes / measurement) for samples and background using the double window method. This method allowed us to correct the contribution of ingrowing ^{210}Bi to the total ^{210}Pb region (Figure 10).

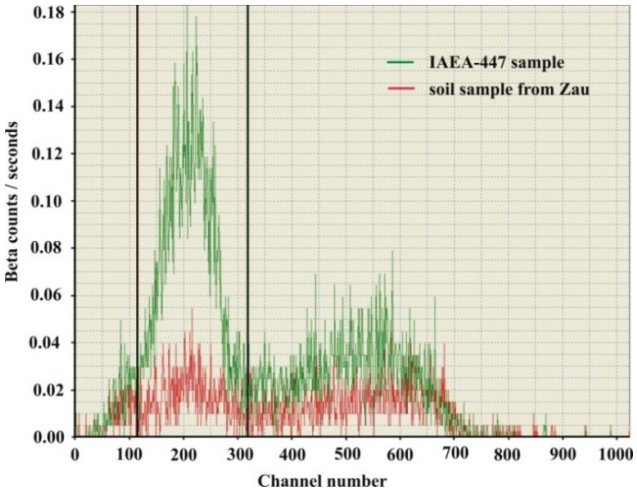
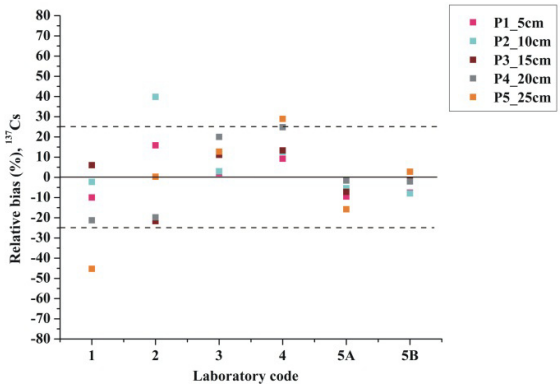


Figure 10. Beta spectrum of ^{210}Pb with double window selection

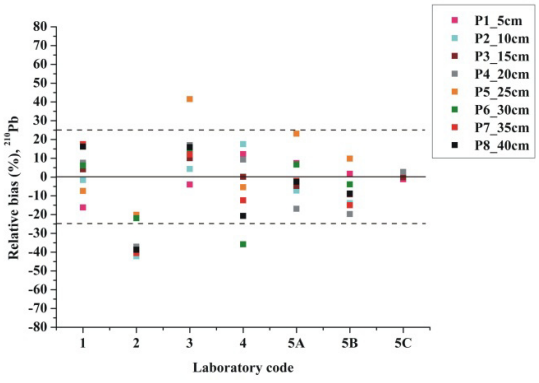
The results reported by five international radiometric laboratories for ^{137}Cs, ^{210}Pb, ^{226}Ra, ^{40}K and ^{228}Ac mass activities in eight soil samples were evaluated for intercomparison. Each

participant laboratory gave values of gamma-spectrometric measurements for the same soil samples in B5 geometry, but implying different methods for the efficiency calibration of their gamma detectors. The data of all participants were firstly evaluated for the relative bias between the analyst value and the reference value, expressed as percentage. As there were no assigned activity values for this intercomparison exercise, the reference values were calculated for each radionuclide in each soil sample as the arithmetic average of all reported activity values. The Maximum Acceptable Relative Bias (MARB) was set to 25% for all radionuclides in all samples.

Figures 11 show the final results of the laboratories as a relative bias (%) to the proposed mean reference values for the radionuclides of interest in all soil samples. The laboratory names were codified with Arabic numbers, and the IAEA different techniques were additionally codified with uppercase letters: 5A code for the gamma results in disk geometry (B5), 5B code for the gamma data in cylindrical geometry (B87) and 5C code for alpha- and beta-determinations. The laboratory from Cluj-Napoca received code 1. As it can be seen, the least problematic determinations were those of ^{228}Ac and ^{40}K. In the case of ^{137}Cs, the reported values showed higher relative bias for samples with lower activities. ^{210}Pb was not as problematic as expected due to the problems related with the correction of self-attenuation effect at low energies. Only one laboratory had a systematic discrepancy compared with the proposed mean values due to the omission of self-attenuation correction in activity determination.



a)



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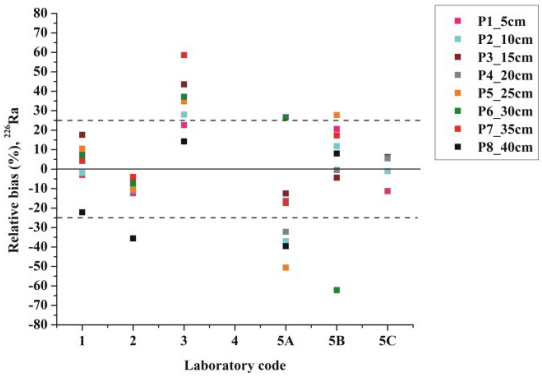

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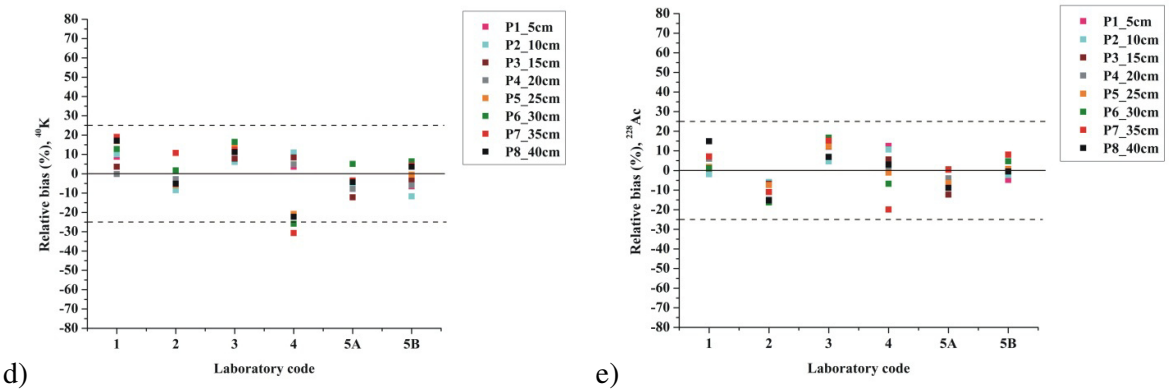

Figure 11. Individual results of each sample measured by each laboratory, given as bias relative to the averaged reference value for (a) ^{137}Cs, (b) ^{210}Pb, (c) ^{226}Ra, (d) ^{40}K, (e) ^{228}Ac. The established MARB is represented dotted line.

The most problematic results of the present intercomparison were found for ^{226}Ra. Only 68% of the reported values were situated within the MARB. Yet, successfully results were only obtained by the first two laboratories and also by the IAEA for B87 geometry. One laboratory did not report any values for radium. However, the most consistent results reported for ^{210}Pb and ^{226}Ra evaluated data were derived from alpha-ray spectrometry and liquid scintillation counting.

The individual Z-score was calculated for each radionuclide to assess the Cluj-Napoca laboratory performance in terms of accuracy. This procedure was accepted as a standard for ISO/IUPAC *(ISO, 1997)*. The target standard deviation was established to a fixed value of 10%. The Z-score performance of the laboratory is presented in Figure 12, for each considered radionuclide. The dotted lines represent the acceptable and questionable limits of performance.

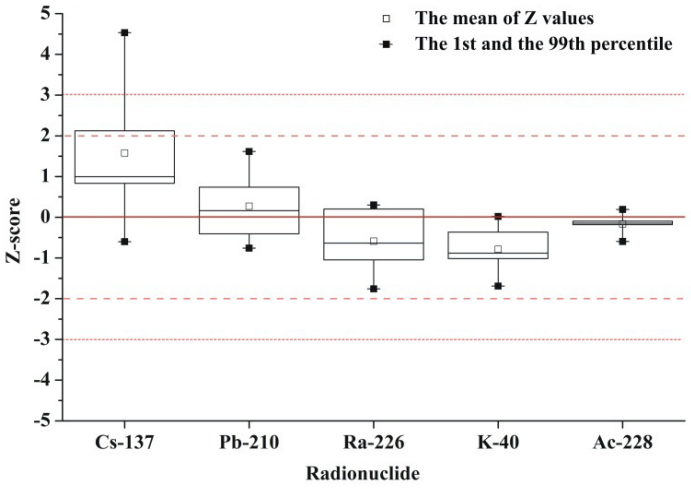

Figure 12. Z-score evaluation for the laboratory from Cluj-Napoca

The measurements performed by the faculty laboratory show overall acceptable results for accuracy. From all the radionuclide data reported by the Cluj-Napoca laboratory, 34 results (92%) were acceptable ($Z \leq 2$), 2 results were questionable and one not-acceptable. The results obtained for ^{137}Cs, ^{210}Pb and ^{226}Ra validate the gamma-spectrometric measurements of soil samples, performed for the radiometric modelling of soil erosion.

Overall, only 13% from a total of 206 evaluated results were out of the maximum acceptable relative bias of 25%. ^{40}K and ^{228}Ac showed very small differences relative to the mean reference value, while the higher discrepancies were found for ^{226}Ra, due to the many uncertainties related with its accurate determination through gamma-ray spectrometry. Still, satisfactory results can be obtained for all radionuclides, if all the corrective measures are being considered for the activity determinations. In the case of soil redistribution studies, dealing with hundreds of samples, the simultaneous determination of radiotracers and quicker procedures at low costs are optimum and can be assured only by means of gamma-ray spectrometry. In this consideration, the quality control procedures, participation in proficiency tests and intercomparation exercises are at maximum importance to assess the validity of the gamma-results. The Z-score analysis of the results reported by the laboratory from Cluj-Napoca within this intercomparison was used to evaluate the laboratory performance for accuracy and showed a normalized average analytical performance of 92%.

The involvement of the participant laboratories and the technical staff that took part in this inter-laboratory comparison and contributed with their time and facilities to the present work is highly acknowledged.

The calibration of the HPGe detectors from Cluj-Napoca radiometric laboratory

The calibration procedure is presented with the aim to be used for radionuclide analysis within the faculty laboratory for: (i) activity measurements of gamma emitting radionuclides in environmental samples with different matrix comparing with the reference material, or (ii) measurements of short-live radionuclides, which were not possible using the direct calibration method. This represents the first attempt to experimentally calibrate these detectors in efficiency. It has to be mentioned that previous work carried out by our research group used computed efficiencies for the energies of interest using a Monte Carlo Code, namely GES-Gamma Electron Efficiency Simulator, version 2.7 *(Fulea, 2009)*.

As both detectors have been previously calibrated in energy, our objective was to verify the stability of the system energy calibration and to correct if necessary. The energy calibration checking was performed using a certified water-equivalent ^{152}Eu source *(Sahagia and Grigorescu, 1992)* and one ^{241}Am point source. Perfect correlation between energy and channel number was found for both detectors at the time of measurement (April 2013) and there was no need for correction. The variation of the peak resolution has been also investigated through short time measurements of ^{152}Eu and ^{241}Am certified sources and a good dependence has been found between the resolution of both detectors and the gamma-energy. After several years of continuous running, the detectors resolution is still in the guaranteed values. The efficiency calibration of HPGe detectors has been performed with RGU-1 and RGTh-1 certified materials

provided by the IAEA, which have been prepared using natural U and Th ore, respectively, diluted with floated silica powder. Our calibration method also included pure crystals of KCl. The aluminium sample containers were very well sealed with glue and electrical band, to prevent radon and thoron losses. For background corrections, an empty geometry was measured in the same conditions for a longer period. The true photopeak efficiency was obtained after correcting for coincidence summing (where needed) using the available code EFFTRAN *(Vidmar, 2005, Vidmar et al., 2011)*. The efficiency fitting curve was described for the GEM detector by a 4-order polynomial function to fit the natural logarithm of efficiency to the logarithm of energy.

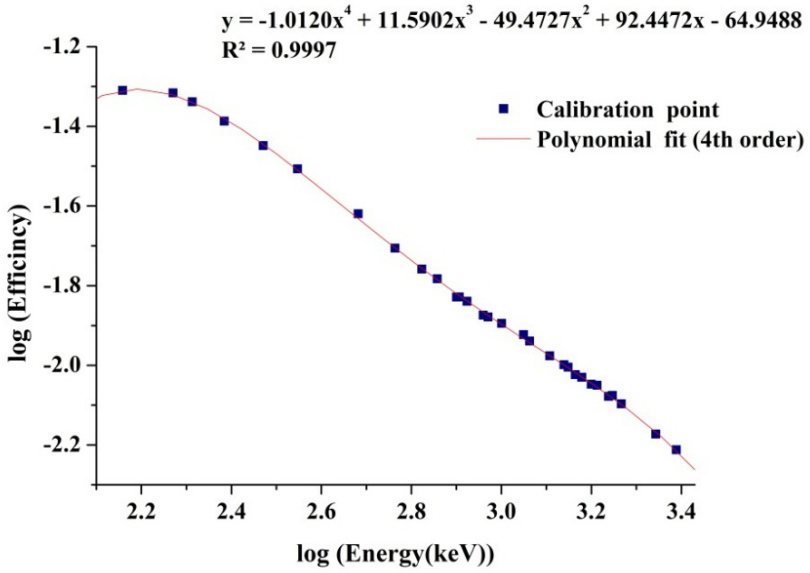

Figure 13. The efficiency curve of the Ortec GEM detector (144 – 2447 keV)

A very good fit (R^2=0.9997) could be obtained for the efficiency curve, using the three natural radioactive sources within an energy range from 144 keV to 2447 keV. One IAEA reference material, IAEA-375 soil with known activity for ^{137}Cs and ^{40}K has been used for the *internal validation* of the efficiency calibration curve. The results obtained for the two radionuclides contained by the IAEA-375 reference material, at different gamma energies, showed a good agreement in comparison with the certified values and therefore validate the experimental calibration curve for GEM detector.

The efficiency fitting curve for the GMX detector is given in Figure 14 as a 4-order polynomial function in natural logarithm scale using the Excel chart function Trendline. Two calibration curves were fitted: one in the low energy range (< 250 keV) and the second in the high energy scale (> 250 keV) of the spectrum, with a common point between. This method allowed us to obtain a better fitting for both efficiency curves. The *internal validation* of the efficiency calibration curve was performed with three IAEA reference materials: IAEA-447 soil-moss, IAEA-375 soil, IAEA-372 hay. The validation results obtained for reference materials are

satisfactory and therefore validate the experimental calibration curves for the entire spectra region and also for different matrix samples.

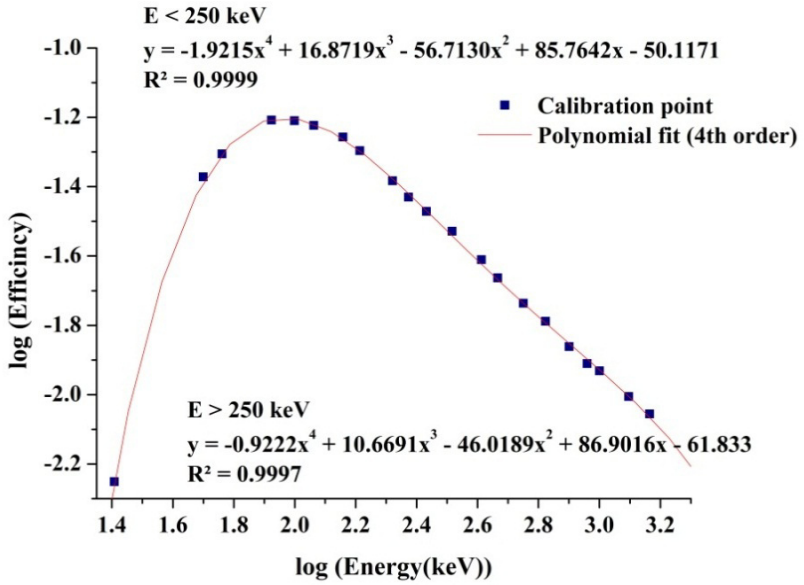

Figure 14. The efficiency curve of the Ortec GMX detector (25.5 – 1461 keV)

In addition to the internal laboratory validation, the proposed efficiency calibration procedure and the laboratory performance in radiometric measurements was verified through participation in Proficiency Test (*IAEA-TEL-2012-03 worldwide open proficiency test on the determination of radionuclides in water, hay and soil*) organized by the IAEA Laboratories from Seibersdorf. Ms. Andra-Rada Iurian was involved in all gamma-measurements, spectra and data analysis performed for this worldwide Proficiency Test. In this external performance evaluation, each participating laboratory received 5 standard samples (3 water bottles, one soil box and one hay box), accompanied by an information sheet and a report form. The performance of the participant laboratories was evaluated for accuracy and precision relative to the target values established by the IAEA. This represented the first participation of our laboratory in a worldwide proficiency test employing gamma measurements of environmental samples with different matrix (water, hay, soil). The reported results were in close agreement with the target activity values provided by the IAEA, having a normalized average analytical performance of 92.9% and also showing reasonably good precision evaluated as the relative standard uncertainty. This external evaluation emphasis the fact that the GMX N type spectrometric system used for the gamma measurements was correctly calibrated in energy and efficiency. Only the value reported for ^{210}Pb exceeded the MARB given by the IAEA, but it was still below the 25% relative difference. The influence of soil composition on the efficiency transfer factors at lower energies could be one of the reasons for discrepancy in ^{210}Pb result.

A method of efficiency calibration for the two HPGe gamma-ray detectors of the FESE laboratory was described, as significant part of environmental radioactivity measurements. For

practical and cost saving purposes, the calibration involved certified natural sources which contained U and Th radioactive series in equilibrium, namely the IAEA RGU-1, RGTh-1 and additionally KCl crystals. The present study represents the first attempt for an experimental efficiency calibration of the gamma detectors from the Cluj-Napoca FESE Laboratory.

The internal validation results obtained through gamma measurements of the reference materials (IAEA-447, IAEA-372 and IAEA-375) showed very good agreement with the reference values for different matrix materials. Moreover, the calibration was externally validated through participation in the *IAEA-TEL-2012-03 Proficiency Test*, organized by the IAEA Laboratories from Seibersdorf. The overall results of the Cluj-Napoca laboratory had a normalized average analytical performance of 92.9%.

Chapter 8. General conclusions and future directions

The potential of fallout radionuclides for soil erosion assessment has attracted the attention of the research community over the last decades. The key findings of the thesis are listed below.

(1) The present thesis aimed to further explore the ^{137}Cs applications in soil erosion for the Romanian landscape. Thus both ^{137}Cs and $^{210}Pb_{ex}$ have been used to assess the magnitude of soil redistribution for an intensive cultivated field, located on the Transylvanian Plain, Mureş county. Combining the two radiotracers, a long-time and medium-time retrospective assessment of the soil redistribution rates could be obtained and reflected well the changes occurred in Romanian agriculture system in 1991. A limitation of the lead approach in soil erosion studies could be identified within this application, as $^{210}Pb_{ex}$ was not present in one reference site, with abundant vegetation cover.

(2) Another application concerned the ^{137}Cs approach used to derive soil erosion estimates on a degraded pastureland from Jucu area, Cluj county. The diffusion-convection equation and the least square fitting were applied for the ^{137}Cs depth profile in our reference area. This alternative method has been used to statistically determine the diffusion and migration coefficients for both Chernobyl and bomb-derived cesium, parameters required for the running of Diffusion and Migration Radiometric Model. The net soil erosion rate derived from the model was comparable with the predicted medium-term soil erosion rate obtained by the conventional approach (ROMSEM equation). The sensitivity analysis of the parameters required by the radiometric model showed that this can be very sensitive to the different input parameters and particular care must be considered in their estimation, especially in areas affected by Chernobyl fallout.

(3) Preliminary results were also obtained regarding the application of ^{7}Be to assess short-time changes of soil redistribution. One of the key requirements of any radiometric modelling is the spatial uniform distribution of the radiotracer. In the case of beryllium, this assumption is commonly not verified due to the short half-live of ^{7}Be and the great number of samples that have to be measured in a short time. The coefficient of variation of 19%, obtained for an experimental plot after two consecutive storm events at the end of July 2012, showed an

acceptable degree of variability for ^{7}Be and therewith a way of quantifying the uncertainties of erosion estimates.

(4) Moreover, the thesis emphasises the importance of accurate and precise gamma-measurements involved in soil erosion radiometric modelling. Thus, an intercomparison exercise was organized between 5 international laboratories for radionuclide determination in soil samples from Zau reference area (Mureș county). Different procedures were employed for the gamma spectrometric evaluation of soil samples by the different laboratories, but also alternative methods (alpha-spectrometry and liquid scintillation counting) were applied. Integrating all data, ^{226}Ra showed to be the most problematic radionuclide when gamma-spectrometry is employed in its determination. Overall, a normalized average analytical performance of 92% was achieved by the Cluj-Napoca laboratory, confirming the reliability of the gamma-measurements performed in soil samples.

(5) Measurements of short-lived radionuclides in soil (e.g. ^{7}Be) were not possible in the faculty laboratory, based on the direct calibration method used for ^{137}Cs, ^{210}Pb and ^{226}Ra. Thus, a method of efficiency calibration for the two HPGe gamma-ray detectors of the FESE laboratory was proposed, as significant part of all environmental radioactivity measurements. This represents the first attempt to experimentally calibrate these detectors in efficiency. For practical purposes, the calibration involved certified natural sources which contained U and Th radioactive series in equilibrium, namely the IAEA RGU-1, RGTh-1 and additionally KCl crystals. The method has been internally validated using IAEA reference materials (IAEA-447, IAEA-372 and IAEA-375) and externally validated through participation in the *IAEA-TEL-2012-03 Proficiency Test*, with very good results. The success of this calibration represents a step forward for future applications of short-life radiotracers in soil redistribution studies in Romania.

Although the FRN technique for the assessment of soil redistribution magnitude has been widely applied in recent years, a future goal still remains to further validate the assumptions relying on the technique in different environments and landscapes, to refine the modelling and its input parameters and to further exploit its potential *(Guzmán et al., 2013).* Moreover, uncertainties should be accounted and integrated in the software tool modelling within a confidence interval and considering the variation of the input parameters.

List of relevant publications

Published books

Iurian A-R, Cosma C, Hofmann W, 2012. Chernobyl radionuclides in environmental samples from Romania&Austria, LAP LAMBERT Academic Publishing GmbH & Co., Germany, ISBN 978-3-8465-8293-0, Ed. Tatiana Melnic, 73 pp.

Published ISI articles

Mabit L, Meusburger K, **Iurian AR**, Owens PN, Toloza A, Alewell C, 2013. Sampling soil and sediment depth profiles at a fine-resolution with a new device for determining physical, chemical and biological properties: the Fine Increment Soil Collector (FISC), *J Soils Sedim* (accepted manuscript), **Impact Factor 2012: 1.965**

Iurian A-R, Toloza A, Adu-Gyamfi J, Cosma C, 2013. Spatial distribution of ^{7}Be in soils of Lower Austria after heavy rains, *J Radioan Nucl Chem*, DOI: 10.1007/s10967-013-2683-8, **Impact Factor 2012: 1.467**

Iurian AR, Mabit L, Begy R, Cosma C, 2013, Comparative assessment of erosion and deposition rates on cultivated land in the Transylvanian Plain of Romania using ^{137}Cs and $^{210}Pb_{ex}$, *J Environ Radioactiv*, 10.1016/j.jenvrad.2013.02.009, **Impact Factor 2012: 2.119**

Iurian AR, Begy R, Cătinaş I, Cosma C, 2012. Results of medium-term soil redistribution rates in Cluj county, Romania, using ^{137}Cs measurements, *Procedia Environ Sci* 14:22-31 **(ISI Proceedings)**

Cosma C, **Iurian AR**, Nita CD, Begy R, Cîndea C, 2012. Indicators of the Fukushima radioactive release in NW Romania, *J Environ Radioactiv* 114:1-6, **Impact Factor 2011: 1.339**

Cosma C, **Iurian AR**, Niţă DC, Begy R, Cîndea C, 2011. Considerations about the presence of Fukushima radionuclides in the NW part of Romania, *Rom J Phys* 56(9-10): 1999-2007, **Impact Factor 2010: 0.340**

Iurian AR, Hofmann W, Lettner H, Türk R, Cosma C, 2011. Long term study of Cs-137 concentrations in lichens and mosses, *Rom J Phys* 56(7-8): 983-992, **Impact Factor 2010: 0.340**

Articles under review in ISI journals

Iurian A-R, Mabit L, Cosma C, 2013. Uncertainty related to input parameters in radiometric derived-soil redistribution rates model: The case of undisturbed soils, *J Soils Sedim,* **Impact Factor 2012: 1.965** (submitted July 2013)

Mabit L, Benmansour M, Abril JM, Walling D, Meusburger K, **Iurian AR**, Bernard C, Tarján S, Owens PN, Blake WH, Alewell C, 2013, Fallout Pb-210 as a soil and sediment tracer in the terrestrial environment: a review, *Earth-Sci Rev,* **Impact Factor 2012: 7.339** (submitted March 2013)

References (selected)

Ayub JJ, Di Gregorio DE, Velasco H, Huck H, Rizzotto M, Lohaiza F, 2009. Short-term seasonal variability in ^{7}Be wet deposition in a semiarid ecosystem of central Argentina, *J Environ Radioactiv* 100(11): 977-981.

Begy R, Cosma C, Timar A, 2009a. Recent changes in Red Lake (Romania) sedimentation rate determined from depth profiles of ^{210}Pb and ^{137}Cs radioisotopes, *J Environ Radioactiv* 100(8): 644-648.

Berger MJ, Hubbell JH, 1987. XCOM: Photon cross sections on personal computers. NBSIR 87-3597, http://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html (last accessed June 2013).

Blake W, Walling DE, He Q, 1999. Fallout beryllium-7 as a tracer in soil erosion investigations, *Appl Radiat Isot* 51(5): 599-605.

Currie LA, 1968. Limits for qualitative detection and quantitative determination - application to radiochemistry, *Anal Chem* 40: 586-593.

Cutshall NH, Larsen IL, Olsen CR, 1983. Direct analysis of ^{210}Pb in sediment samples: Self-absorption corrections, *Nucl Instrum Methods* 206(1): 309-312.

Debertin K, Helmer R, 1988. Gamma- and X-ray Spectrometry with Semiconductor Detectors, North-Holland, Amsterdam, pp.367.

De Cort M, Dubois G, Fridman SD, Germenchuk MG, Izrael YA, Janssens A, et al. 1998, Atlas of Caesium Deposition on Europe after the Chernobyl Accident. EUR Report 16733. EC, Office for Official Publications of the European Commission Communities, Luxembourg.

Fulea D, Cosma C, 2009. Monte Carlo sampling for gamma and beta detectors using a general purpose PC program. *Radiat Meas* 44: 278–282.

He Q, Walling DE, 1996. Interpreting particle size effects in the adsorption of ^{137}Cs and unsupported ^{210}Pb by mineral soils and sediments, *J Environ Radioact* 30: 117-137.

IAEA, 2010. Analytical Methodology for the Determination of Radium Isotopes in Environmental Samples, Analytical Quality in Nuclear Applications Series No. 19, Printed by the IAEA in Austria

Ionita I, Margineanu RM, 2000a. Application of ^{137}Cs for measuring soil erosion/deposition rates in Romania, *Acta Geol Hisp* 35(3-4): 311-319.

Ionita I, Margineanu RM, Hurjui C, 2000b. Assessment of the reservoir sedimentation rates from ^{137}Cs measurements in the Moldavian Plateau, *Acta Geol Hisp* 35(3-4): 357-367.

ISO, 1997. Proficiency testing by interlaboratory comparisons - Part 1: Development and operation of proficiency testing schemes, Guide 43-1.

Iurian AR, Begy R, Cătinaş I, Cosma C, 2012. Results of medium-term soil redistribution rates in Cluj county, Romania, using ^{137}Cs measurements, *Procedia Environ Sci* 14:22-31.

Iurian AR, Mabit L, Begy R, Cosma C, 2013, Comparative assessment of erosion and deposition rates on cultivated land in the Transylvanian Plain of Romania using ^{137}Cs and $^{210}Pb_{ex}$, *J Environ Radioactiv*, 10.1016/j.jenvrad.2013.02.009.

JCGM, 2008. JCGM 100:2008. GUM 1995 with minor corrections. Evaluation of measurement data - Guide to the expression of uncertainty in measurement; http://www.bipm.org/en/publications/guides/

Jia G, Torri G, Innocenzi P, Ocone R, Di Lullo A, 2006. Determination of radium isotopes in mineral and environmental water samples by alpha-spectromtry, *J Radioanal Nucl Chem* 267(3): 505-514.

Jia G, Torri G, Ocone R, 2007. Determination of radium isotopes in soil samples by alpha-spectrometry, *J Radioanal Nucl Chem* 273(3): 779-783.

Khater AEM, Ebaid YY, 2008. A simplified gamma-ray self-attenuation correction in bulk samples, *Appl Radiat Isotopes* 66: 407–413.

Knoll GF, 2000. Radiation Detection and Measurement, 3rd Edition, Wiley, pp. 802.

LNHB (Laboratoire National Henri Becquerel), LARA Database, 2008 CEA-R-6201, ISSN 0429-3460, http://laraweb.free.fr/. (last accessed June 2013)

Mabit L, Benmansour M, Walling DE, 2008a. Comparative advantages and limitations of Fallout radionuclides (^{137}Cs, ^{210}Pb and ^{7}Be) to assess soil erosion and sedimentation, *J Environ Radioactiv* 99:1799-1807.

Mabit L, Bernard C, Makhlouf M, Laverdière MR, 2008b. Spatial variability of erosion and soil organic matter content estimated from ^{137}Cs measurements and geostatistics, *Geoderma* 145(3-4):245-251.

Mabit L, Chhem-Kieth S, Toloza A, Vanwalleghem T, Bernard C, Amate JI, González de Molina M, Gómez JA, 2012. Radioisotopics and physicochemical background indicators to assess soil degradation affecting olive orchards in southern Spain, *Agr Ecosyst Environ* 159:70-80.

Mabit L, Meusburger K, **Iurian AR**, Owens PN, Toloza A, Alewell C, 2013. Sampling soil and sediment depth profiles at a fine-resolution with a new device for determining physical, chemical and biological properties: the Fine Increment Soil Collector (FISC), *J Soils Sedim (accepted manuscript)*

Moţoc M, Stanescu P, Taloiescu I, 1979. Metode de estimare a eroziunii totale şi a eroziunii efluente pe bazine hidrografice mici, Buletin ICPA, Bucureşti (in Romanian).

Murray AS, Marten R, Johnston A, Martin P, 1987. Analysis for naturally occurring radionuclides at environmental concentrations by gamma spectrometry, *J Radioanal Nucl Chem* 115: 263–288.

Nguyen L, Zapata F, Lal R, Dercon G, 2012, Role of Nuclear and Isotopic Techniques in Sustainable Land Management: Achieving Food Security and Mitigating Impacts of Climate Change. In Lal R and Stewart BA (Eds.), Advances in Soil Science, World soil resources and food security, Chapter 8; 345-418.

Piton F, Lépy M-C, Bé M-M, Plagnard J, 2000. Efficiency transfer and coincidence summing corrections for γ-ray spectrometry, *Appl Radiat Isotopes* 52(3): 791–795.

Popa N, Filiche E, Petrovici G, Margineanu RM, 2011. Using caesium-137 techniques to estimate soil erosion and deposition rates on agricultural fields with specific conservation measures in the Tutova rolling hills, Romania. In Dercon G (Eds), IAEA-TECDOC-1665 Impact of Soil Conservation Measures on Erosion Control and Soil Quality:259-277.

Poręba GJ, Bluszcz A, 2007. Determination of the initial 137Cs fallout on the areas contaminated by Chernobyl fallout, *Geochronometria* 26:35-38.

Porto P, Walling DE, Callegari G, Catona F, 2006. Using fallout lead-210 measurements to estimate soil erosion in three small catchments in southern Italy, *Water, Air, and Soil Poll: Focus* 6: 657–667.

Ritchie JC, McHenry JR, 1990. Application of Radioactive Fallout Cesium-137 for Measuring Soil Erosion and Sediment Accumulation Rates and Patterns: A Review, *J Environ Qual* 19:215-233.

Sahagia M, Grigorescu EL, 1992. Water-equivalent solid standard sources*, Nucl Instrum Meth A* 312: 236-239.

Sarmiento J, Gwinn E, 1986. Strontium 90 fallout prediction, *J Geophys Res* 91(C6):7631-7646.

Schuller P, Iroumé A, Walling DE, Mancilla B, Castillo A, Trumper RE, 2006. Use of beryllium-7 to document soil redistribution following forest harvest operations, *J Environ Qual* 35:1756-1763.

Seibert P, Frank A, Formayer H, 2007. Synoptic and regional patterns of heavy precipitation in Austria, *Theor Appl Climatol* 87:139-153.

Shakhashiro A, Tarjan S, Ceccatelli A, Kis-Benedek G, Betti M, 2012. IAEA-447: A new certified reference material for environmental radioactivity measurements, *Appl Radiat Isotopes* 70: 1632–1643.

Sill C, 1987. Determination of radium-226 in ores, nuclear wastes and environmental samples by high resolution alpha-spectrometry, *Nucl Chem Waste Man* 7: 239-256.

STAS 7107/1-76, 1976. Institutul de studii şi proiectări pentru îmbunătăţiri funciare al României. Determinarea conţinutului de materie organică (in Romanian).

STAS 1913/5-85, 1985. Institutul de studii şi proiectări pentru îmbunătăţiri funciare al României. Determinarea granulozităţii (in Romanian).

Sutherland RA, 1991. Examination of caesium-137 areal activities in control (uneroded) locations, *Soil Technol* 4: 33-50.

Sutherland RA, 1996. Cesium-137 soil sampling and inventory variability in reference samples; literature survey. *Hydrol Process* 10:34-54.

Timar A, Cosma C, Benea V, Begy R, Jobagy V, Szeiler G, Barbos D, Fulea D, 2007. Estimation of environmental radionuclide concentration in soils, a comparison of methods for the annual radiation dose determination in luminescence dating, *Studia Universitatis, Babes-Bolyai, Geologia* 52(1):80-81.

Vajda N, LaRosa J, Zeisler R, Danesi P, Kis-Benedek G, 1997. A novel technique for the simultaneous determination of ^{210}Pb and ^{210}Po using a Crown Ether*, J Environ Radioactiv* 37(3): 355-372.

Vidmar T, 2005. EFFTRAN - A Monte Carlo efficiency transfer code for gamma-ray spectrometry, *Nucl Instrum Meth A* 550(3): 603-608.

Vidmar T, Kanisch G, Vidmar G, 2011. Calculation of true coincidence summing corrections for extended sources with EFFTRAN, *Appl Radiat Isotopes* 69: 908–911.

Wallbrink PJ, Murray AS, 1994. Fallout of ^{7}Be in south-eastern Australia, *J Environ Radioactiv* 25(3): 213-228.

Walling DE, He Q, 1999. Improved models for estimating soil erosion rates from cesium-137 measurements, *J Environ Quality* 28:611-622.

Walling DE, He Q, Appleby PG, 2002. Conversion models for use in soil-erosion, soil redistribution and sedimentation investigations. In: Zapata F. (Ed.), Handbook for the Assessment of Soil Erosion and Sedimentation using Environmental Radionuclides. Kluwer, Dordrecht, The Netherlands, pp. 111-164.

Walling DE, Collins AL, Sichingabula HM, 2003. Using unsupported lead-210 measurements to investigate soil erosion and sediment delivery in a small Zambian catchment, *Geomorphology* 52:193–213.

Walling DE, 2013. Beryllium-7: The Cinderella of fallout radionuclide sediment tracers?, Hydrol Process Special Issue: Introduction to the special issue 'Tracer Applications in Sediment Research' 27(6): 830-844.

Zapata F (Ed.), 2002. Handbook for the Assessment of Soil Erosion and Sedimentation Using Environmental Radionuclides, Kluwer, The Netherlands, pp. 215.