



# FALLOUT $^{137}\text{Cs}$ AND NATURAL $^{40}\text{K}$ AS TRACERS OF TOPSOIL DEVELOPMENT DURING SLOPE PROCESSES – A CASE STUDY FROM THE DAUGAI ENVIRONS, SOUTHERN LITHUANIA

IEVA BAUZIENE, JONAS MAZEIKA and ZANA SKURATOVIC  
*Nature Research Centre, T. Sevcenkos 13, LT-03223 Vilnius, Lithuania*

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**Abstract:** Investigations of soil erosion by  $^{137}\text{Cs}$  method brings uncertainties of different nature. The estimation of the  $^{137}\text{Cs}$  local inventory is associated with problems of data comparison and instrumental errors. In order to avoid systematic errors, the rate of soil erosion determined by the  $^{137}\text{Cs}$  method is compared with the values obtained for other radioactive elements. Soil parameters must be regarded too. The aim of the work was to test the character of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  distribution in the topsoil vertical section for determination erosion-accumulation type and ratio in different time spans. The topsoil thickened by different types of erosion-accumulation processes was sampled at a 2-3 cm interval. Soil samples were analysed by means of scintillation gamma spectrometry. The relationship between  $^{137}\text{Cs}$  and  $^{40}\text{K}$  inventories was weakest in the topsoil formed by mixing of soil material during the installation of artificial drains. Based on climatic characteristics, variations of theoretical soil accumulation rate in the last 50 years were calculated for the topsoil accumulated predominantly by water erosion.  $^{40}\text{K}$  and  $^{137}\text{Cs}$  correlation in the bottom of vertical section of topsoil or arable horizon are closest and this section may be used as a complementary parameter determining the local inventory value.

**Keywords:**  $^{137}\text{Cs}$  method, colluvial topsoil, erosion-accumulation rate, climatic indexes.

## 1. INTRODUCTION

The  $^{137}\text{Cs}$  method applied in many studies allowed determining the soil erosion rate (Bujan *et al.*, 2003; Ritchie and McCarty, 2003). The  $^{137}\text{Cs}$  method has some advantages over the direct measurement of erosion rates at the study plots. Erosion rates based on  $^{137}\text{Cs}$  method are often similar to soil losses directly measured in experimental sites (Golosov, 2003). Estimates based on  $^{137}\text{Cs}$  determination are retrospective and therefore avoid the need for establishment of long-term monitoring programs.  $^{137}\text{Cs}$  data can be obtained on the basis of a single site and the estimated rate of soil redistribution relates to

the past 50 years (short-term measurements may be unrepresentative) and reflects the integration of several landscape transformation processes, water and wind erosion, tillage effects etc. (Porto *et al.* 2001).

The erosion processes in hilly morainic agrolandscape of the East Lithuania region were carried out in the middle of 20 century and empiric equations for erosion processes were calculated (Racinskas, 1990). Since then there measurements of erosion have not been conducted. The  $^{137}\text{Cs}$  method was used for general description of geomorphologic processes in agro-landscape during establishing the Daugai Regional park.

The main sources of errors emerging in application of the  $^{137}\text{Cs}$  method for determination of the soil erosion rates are local reference values for an area (Porto *et al.*

Corresponding author: I. Bauziene  
e-mail: [ieva.bauziene@geo.lt](mailto:ieva.bauziene@geo.lt)

2001) and the assumption of an even  $^{137}\text{Cs}$  distribution in the arable horizon and even susceptibility to erosion of various soil fractions.

To avoid systematic errors, the soil erosion rate determined by  $^{137}\text{Cs}$  method is frequently compared with the values obtained by other radioactive isotopes (Zapata 2003, Poręba *et al.* 2006, Bluszcz *et al.* 2007, Zukova, 2009).

The processes of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  sorption in the soil and in other objects of nature are comparable (Davis, 1963) yet their sources are different.  $^{40}\text{K}$  is a natural radioactive isotope. Both  $^{137}\text{Cs}$  and  $^{40}\text{K}$  mainly are immobilized by certain clay minerals, such as vermiculite and illite, in the interlayers of crystalline lattice. Part of  $^{40}\text{K}$  is transferred to the soil with fertilizers. In all cases, the content of radioactive potassium is directly proportional to the total potassium. In Lithuania, similarly to Russia, the average content of  $^{40}\text{K}$  in *Albeluvisols* amounts to about 500 Bq/kg. It is exceeding the world's average by 1.7 times (Mazeika, 2002, Lunev and Orlov, 2009). The high dependence on soil fertilization causes high fluctuations of  $^{40}\text{K}$  inventories in time and space which are the main limitations of  $^{40}\text{K}$  use as tracers of geomorphologic processes.

The fallout of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in Lithuania in 1980 and 1990 was evaluated in two different ways: in 1980 for 0-20 cm layer of arable soil and in 1990 for 0-5 cm in natural landscape (Mažeika, 2002).

The initial assumption of the investigation was that the character of vertical distribution pattern of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in the profiles of accumulated topsoil depends on type of erosion and accumulation. The aim of the present work is to test the vertical  $^{137}\text{Cs}$  and  $^{40}\text{K}$  distribution in the profiles of accumulated topsoil for investigation of erosion-accumulation rates in different periods of past 50 years.

## 2. STUDY OBJECTS AND METHODS

The study site is situated in the southern part of Lithuania. The study objects are located at the gently sloped accumulation areas of eroded soil material from hills of different size north of Daugai settlement and motor road Varena-Alytus (Fig. 1).

The content of  $^{137}\text{Cs}$  in the topsoil is comparable with the average value for the territory of Lithuania,  $10.1 \pm 6.0$  Bq/kg (Mazeika, 2002) and in arable horizon of hilly moraine landscape 5.8-13.2 Bq/kg and the fallout of  $^{137}\text{Cs}$  after the Chernobyl incident has not changed of southern-eastern Lithuania (Lubytė and Antanaitis, 2004). According the data of three neighbouring meteorological stations at that station there was no rain in the period of April 26 1986 till May 12 1986.

The main part of farmlands at the study site is represented by grasslands and pastures. The altitude of hills is 15 and 10 m, slope angle 7-9 degrees and the expositions of the steepest slopes are eastern and southern. *Albeluvi-*



Fig. 1. Location of study site

*sols* are typical for the territory. The thickness of the arable horizon in the region is about 20 cm, but on steep slopes the surface Ap horizon it is not continuous. Soil erosion products accumulate in a, by comparison thick, topsoil (Table 1) generating topsoil of sandy loam texture, much thicker than on the slope. Sampling was performed at the thickest topsoil which was found by probe.

Land drainage in this territory was carried out in 1986-1987. The drainage channel is located about 20 m from the Daugai 1 soil study site. This area has no underground drainage system. On the other hand, underground drainage is installed in the sites of profiles 2 and 3. In order to avoid proximity to it, soil samples were taken not from the lowest points of the relief but at points located more than 20 m away from the drainage system. Profiles 2 and 3 are located in one trough. Profile Daugai 3 is in close proximity to an arable field (Fig. 2).

Three soils were investigated to the depth of the subsoil horizon. Excavation walls were sampled in 2-3 cm thick slices to a depth of 50-70 cm. The bulk density was measured using 40 mm diameter rings. The difference between the sampling interval and bulk density measuring interval was not expected to distort the calculations of the  $^{137}\text{Cs}$  inventory at a sampling point because no layers were observed in the field. In addition, laboratory inspection of dried samples proved the homogeneous character of the topsoil (Table 1). The 2 cm sampling interval was used and bulk density was estimated by using air-dry weight of the core samples (Du and Walling, 2011).

Homogenized samples were dried, put into 0.5 l Marinelli vials and analyzed with scintillation gamma spectrometry at the Radioisotope Research Laboratory of Nature Research Centre. Up to six samples from each profile for quality assurance were additionally examined by high resolution gamma spectrometry system equipped with an HPGe detector of type GWL-120-15-LB-AWR (manufacturer – ORTEC®). The minimal detectable specific activity of radionuclides concern in the soil for

**Table 1.** Description of investigated deluvial (colluvial) soil and features of the first buried horizon.

Number	Depth (cm)	Munsell colour code	Bulk density (g·cm <sup>-3</sup> )	Notes
Daugai1	0-8	4/2 2.5Y	1.32	Unicoloured. Boarder detected by reducing of roots density.
	8-14	4/2 2.5Y	1.33	Weaker structure (more sand), slightly cemented by ochre in lower part.
	14-30	4/2 2.5Y	1.33-1.35	1-5% ochre spots and coprolites. Few coal (brand) residues. Boarder detected by colour.
	30-36	4/3 2.5Y	1.31-1.34	0-3% ochre spots and coprolites. Boarder detected by colour.
	36-40	5/3 2.5Y	1.31-1.33	0-3% ochre spots and coprolites. Few coal (brand) residues. Boarder detected by colour.
	40-62	4/2 2.5Y	1.31-1.34	0-3% ochre spots and coprolites. Few coal (brand) residues. Boarder detected by colour.
	62-78	5/2 2.5Y	1.62	0-3% ochre spots and coprolites. Lamellae of coal (brand) residues. Buried horizon overlaying hard illuvial horizon.
Daugai2	0-8	3/2 10YR	1.28	Unicoloured. Coal (brand) residues mostly in 2-5 cm layer. Boarder detected by reduction of roots density.
	8-15	4/2 10YR	1.36	Hardness increasing, coprolites in pores and root canals (2%), pseudomorphs (5/6 7.5YR). Coal (brand) residues mostly in 10-12 cm layer, (0.06% by mass). Boarder detected by appearing of decayed stems and increased amount of pseudomorphs.
	15-25	5/3 10YR	1.36	5% coprolites in pores and pseudomorphs (5/6 7.5YR). Wavy boarder.
	25-35	3/1 ir 3/2 10YR	0.45	15% peat lenses with white mycelia. Coal (brand) residues, in 28...30 cm, 0.30%. Residues of mezzo-fauna. Mixed horizon of colluvial material and buried histosol.
	35-45	3/3 10YR	-	Increasing the part of peat lenses.
	>45	2.5/2 7.5Y	-	Buried <i>Histosol</i> . Peat
Daugai3	0-8	4/1 10YR	1.16	Unicoloured. Coal (brand) residues mostly in 6-9 cm layer, 0.05%. Fauna residues, liming traces. Boarder detected by reducing of roots density.
	8-17	5/2 10YR	1.41	Few coal (brand) residues. Boarder detected by hardness reducing.
	21-27	5/2 10YR	1.33	Pseudomorphs in pores and root cannels (3%), (6/6 7.5YR). Few coal (brand) residues. Boarder detected by colour.
	27-33	5/3 10YR	1.12	Soft. Pseudomorphs and coprolites in pores and root cannels (5-7%), (6/6 7.5YR). Boarder detected by colour and hardness.
	33-36	5/4 10YR	1.15	Soft, containing organic matter residues, overlaying hard illuvial horizon.

particular geometry and reasonable counting time (about 80 ks) with scintillation gamma spectrometry was 2 Bq/kg for <sup>137</sup>Cs and 30 Bq/kg for <sup>40</sup>K. Uncertainties of determinations were usually within an interval (with 95% of confidence) of 8% for <sup>137</sup>Cs and 4% for <sup>40</sup>K.

The <sup>137</sup>Cs inventory at a sampling point was calculated by simple equation (Sutherland, 1992):

$$C_{inv} = \sum_{i=1}^n C_i \times BD_i \times ID_i \quad (2.1)$$

where:  $C_{inv}$  is the <sup>137</sup>Cs inventory in soil profile (Bq/m<sup>2</sup>),  $C_i$  is <sup>137</sup>Cs specific activity (Bq/kg) in single (i) layer,  $BD_i$  – bulk density of single layer (kg/m<sup>3</sup>) and  $ID_i$  – thickness (m) of single (i) layer. The same value of  $BD_i$  was attributed to several layers of similar morphological features (Table 1).

The inventory for 1 cm sub-layer (kBq/m<sup>2</sup>), <sup>137</sup>Cs as well as of <sup>40</sup>K, was counted as:

$$C_{inv} = \sum_{i=1}^n C_i \div 1000 \times BD_i \div 100 \quad (2.2)$$

The elevated atmospheric fallouts of <sup>137</sup>Cs in Lithuania were observed in the periods 1956-1959, 1962-1963 and in 1986. Regarding low <sup>137</sup>Cs reference values, global fallouts prevail at the investigated site. Because of rain

deficit after the Chernobyl accident radioactive contamination in 1986 affected forested and laky areas (Mazeika, 2002; Lubyte and Antanaitis, 2004).

### 3. EVALUATION OF ERODIBILITY AND ERO-SIVITY FACTORS AT THE STUDY SITE

The evaluation of erosivity and erodibility factors was conducted for a general description of the Daugai study site. Standardised approach on soil erosion represents the Universal soil Loss Equation (USLE). It was designed to estimate annual erosion for long term periods. Annual soil loss depends on soil erodibility (texture and parent material), slope, slope length, cover management, and rainfall erosivity factors. This model is used for soil erosion risk assessment in Europe (Wischmeier and Smith, 1978; Van der Knijff *et al.* 2000).

The morphometric parameters of landforms and lithological composition predetermine susceptibility of a territory to erosion referred to as potential erodibility. Potential erodibility was defined using an empirical formula conformed to water erosion type in loamy *Albeluvisols* (Racinskas, 1990). Considering low average humus content in this region (<1%) it was simplified:

$$E = (0.09L + 1.62i - 8.7) \quad (3.1)$$



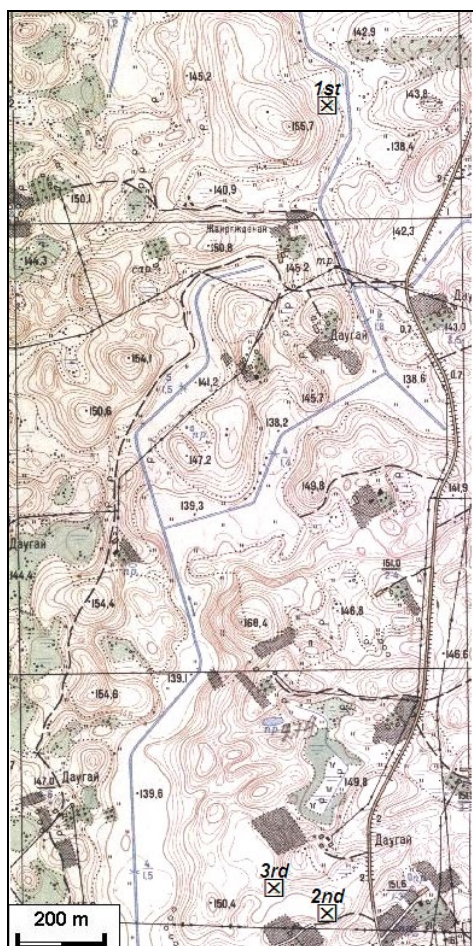


Fig. 2. A fragment of hypsometric map showing localization of deluvial-colluvial soil profiles

where  $E$  is the potential annual erodibility,  $L$  slope length (meters) and  $i$  is the slope expressed in degrees.

For Daugai 1<sup>st</sup>, 2<sup>nd</sup> and 3<sup>rd</sup> profiles potential erodibility was respectively 1.4, 0.7 and 1.1 mm per year. Within the European scale (Le Bissonnais *et al.*, 2001) the erosion risk in the studied territory can be classified as high and medium.

Temporal variation of erosion and accumulation intensity depends on climate and land use. Water erosion for the explored slope is initiated mainly by the amount and intensity of precipitation. Land use on the examined slopes in 40 year period was not intensive and grasslands and pastures prevail.

According to the long-term data from the nearest meteorological stations (Alytus and Meteliai), in 1951-1985, the average annual amount of precipitation was 628 mm; during the warm season 431 mm (68.6%); the maximum, of 80 mm on the average, was observed in July.

Climate conditions favourable for erosion in different time spans were determined in three different ways. The first, most readily available and widely used in long-term

investigations was rainfall erosivity or Modified Fournier Index, MFI<sub>j</sub> (mm/year) (Arnoldus, 1980).

$$MFI_j = \frac{\sum_{i=1}^{12} P_i^2}{P_j} \quad (3.2)$$

where  $j$  indicates years,  $i$  months and  $P_i$  monthly precipitation (mm/month). As MFI is a generalizing index, complementary indexes of precipitation intensity and snow cover were calculated based on empirical investigations of soil erosion in East Lithuanian Region.

The second and third way of erosivity evaluation was related to the intensity of erosion in different seasons. The most favourable climatic conditions for erosion occur in the warm season with frequent and intensive rainfalls and in spring when large water supplies are stored in the snow. In summer, soil erosion is induced by rainfalls with intensity >0.3 mm/min and lasting longer than 10 min. In these cases, the infiltration capacity of the soil becomes very low what leads to topsoil erosion (Racinkas, 1990). In the majority of meteorological stations, the intensity of precipitation is not measured. For this reason, the sum of >6 mm of precipitation in 12 hours was used as threshold for erosion initiation.

In the present study it is assumed that erosion is initiated by climatic conditions when precipitation value exceeds 3/2 of the median or 3rd quartile.

In the time frame 1955-2008, 3rd quartile of MFI values was exceeded in 1956, 1960, 1972, 1973, 1974, 1980, 1986, 1987, 1989, 1993, 1998, 2001, 2005 and 2007, i.e. 4 times in 1963-1985 and 8 times after 1986. Thus in 22 years after the 1986, the frequency of extreme values of MFI index doubled in comparison with the previous years (Fig. 3).

Total sum of >6 mm precipitation index temporal variation is different compared to MFI. In warm seasons of 1955-2008, the sums of intensive (>6 mm) precipitation exceeded the 3/2 of median in 1960, 1974, 1986, 1989 and 2005; five times in total; 3rd quartile was exceeded 13 times: in 1956, 1960, 1962, 1966, 1972, 1974, 1978, 1986, 1987, 1989, 1995, 2005 and 2007. The high total of precipitation in 1986, 1987 and 1989 is considered as a particular interval of <sup>137</sup>Cs depletion in the soil profile. In 1963-1985 the frequency of intensive (>6 mm) precipitation was 1.5 times as low as in 1986-2008. (Fig. 3).

The thickness of snow cover formed a week before the melting of the continuous snow cover, i.e. index of water storage in the snow, was a complementary criterion of erosion risk. In 1960, the values of all used climatic criteria were extreme yet in 1980 the MFI index was slightly lower than 3rd quartile though the snow cover was extremely thick (Fig. 3). For this reason, the year 1980 is classified as a year of high erosion risk.

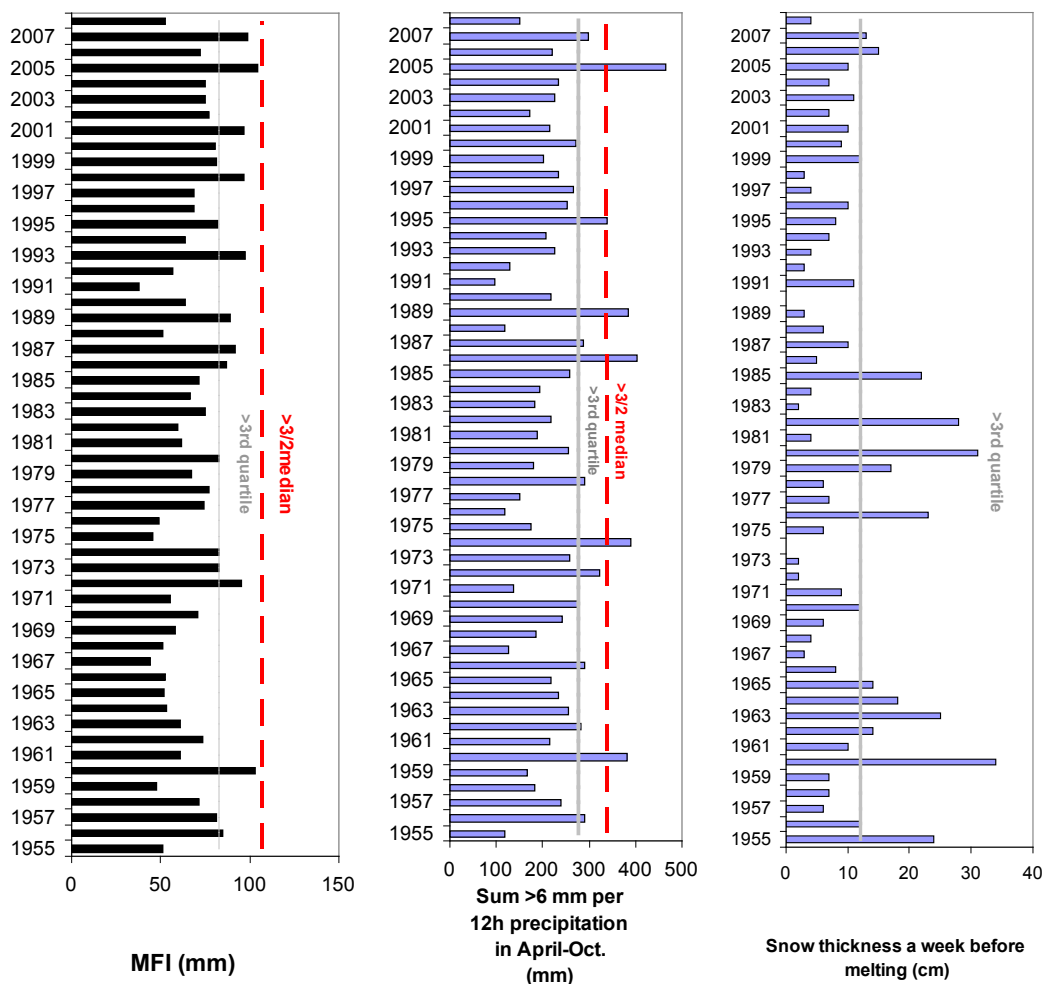


Fig. 3. Climatic indexes during 1955-2008 (data of Alytus and Meteliai meteorological stations).

Soil accumulation rate ( $H_j$ ) for particular year at a particular point (mm/year) was calculated by the following formula:

$$H_j = MFI_j \frac{H}{\sum_{i=j}^{j+t} MFI_i} \quad (3.3)$$

where  $MFI_j$  is the Modified Fournier Index for a particular year (mm/year),  $H$  the distance between two neighbouring  $^{137}\text{Cs}$  depletions in soil profile (corresponding to the time span between two high erosion years,  $t$ ),

$\sum_{i=j}^{j+t} MFI_i$  is the sum of yearly  $MFI$  for time span  $t$ .

#### 4. RESULTS AND DISCUSSION

The detectable activity of  $^{137}\text{Cs}$  in accumulated topsoil of Daugai area appears at a depth: in 1<sup>st</sup> profile – down to

27 cm, in 2<sup>nd</sup> – down to 36 cm, and in 3<sup>rd</sup> – down to 33 cm.

Local  $^{137}\text{Cs}$  inventory values for the 1<sup>st</sup> profile was 2374 and 1844 Bq/m<sup>2</sup> respectively. It is close to the reference inventory value (2508±475 Bq/m<sup>2</sup>) of south-eastern Lithuania. The inventory value of the 2<sup>nd</sup> profile was twofold and higher and equal to 4483 Bq/m<sup>2</sup>.

$^{137}\text{Cs}$  specific activity and inventory values of 1<sup>st</sup> and 2<sup>nd</sup> profiles are similar though erosion potential (Racinskas, 1990) differs by a factor of two and more. There are peat lenses in the 25-45 cm layer of the 2<sup>nd</sup> profile, so that a great part of the soil material is mixed with peat. This material can be derived from drainage systems construction works in 1987. That is why the 2<sup>nd</sup> deluvial-coluvial soil profile contains 100%  $^{137}\text{Cs}$ . Because of slope processes the material of 2<sup>nd</sup> topsoil is not supportive for erosion-accumulation rate study.

In the 3<sup>rd</sup> profile, the highest inventory value is twice as high as in the 1<sup>st</sup> and 2<sup>nd</sup> profiles. The higher initial  $^{137}\text{Cs}$  inventory value in the 3<sup>rd</sup> profile is predetermined

not by more intensive erosion but by a finer texture and (or) higher selective sorption capacity, reflected by a higher  $^{40}\text{K}$  activity. The fallout  $^{137}\text{Cs}$  in the 3<sup>rd</sup> profile is growing with depth, together with increasing  $^{40}\text{K}$  activity. Gradual accumulation of soil material in the 3<sup>rd</sup> profile takes place where hard plough pad had not formed. The absence of plough pad reflects the bulk density – in the lower part of the colluvial layer (27-33 cm) it is smaller and the soil is softer than in the upper part. Thus, there are no traces of clay illuviation (Table 1). The increased  $^{40}\text{K}$  activity in lower part (Fig. 4) probably reflects reduction of clay minerals surface because of tillage (Chizhikova, 1994) or mixture with underlying horizon rather than accumulation of fine material.

The  $^{137}\text{Cs}$  distribution in a 27 cm thick Daugai 1<sup>st</sup> topsoil horizon has four relative minimum values at the depths of 19, 17, 9-6 and 1 cm (Fig. 4). It is assumed that the material above the depth of approximately 19 cm might have accumulated after 1960. This depth corresponds to the first from the bottom of colluvial deposits depletion of  $^{137}\text{Cs}$  of an accumulated soil sub-horizon as a result of highest climatic indexes in 1960 compared to the whole period of 1955-2008 (Fig. 3).

The accumulation rate weighted using the MFI and  $>6$  mm precipitation sum climatic indexes ranges from 2 to 10 mm per year (five times).

The highest accumulation rates weighted according to MFI occurred in 1980, 1986, 1987 and 1993 (about 6 mm per year). About 5 mm per year were accumulated in

1960, 1972-1974, 1999 and 2002. The highest accumulation rates weighted according to  $>6$  mm index (about 9 mm per year) occurred in 1974, 1980 and 1985. The time span 1980-1986, when  $>6$  mm were accumulated every year forms an exception. The strongest erosion due to intensive cultivation may have taken place in the period 1980-1986, because this was a period of extensive agriculture in this territory and all over Lithuania that caused technogenic (tillage) erosion (Fig. 5).

$^{137}\text{Cs}$  and  $^{40}\text{K}$  inventories in the investigated topsoils are interrelated. It is commonly accepted that  $^{137}\text{Cs}$  activity is proportional to the  $^{40}\text{K}$  activity, because these elements compete for the same sites of selective sorption.

There is a positive correlation between  $^{137}\text{Cs}$  and  $^{40}\text{K}$  inventories in the 1<sup>st</sup> profile and it is particularly strong in the 3<sup>rd</sup> profile of Daugai accumulated topsoil ( $R^2=0.98$ ). Inventories of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in the 2<sup>nd</sup> topsoil are distributed in a chaotic way (Fig. 6). In Daugai topsoil the correlation between  $^{137}\text{Cs}$  and  $^{40}\text{K}$  inventories is strongest ( $R^2>0.98$ ) in the case of the soil material mixed by tillage (3<sup>rd</sup> profile) or sheet erosion, gradual translocation toward slope (1<sup>st</sup> profile) (Fig 6).

On the analogy of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  activities differ between species of fungi fruit body samples only in a limited range (Mietelski *et al.*, 2010), in case  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in accumulated topsoil can be presumed wider limits of  $^{40}\text{K}$  inventory in upper sub-horizons of topsoil and reverse dependence for  $^{137}\text{Cs}$  inventory (Fig. 6: hollow and full marks). It can be explained by mixture accumulated topsoil upper sub-horizons of different soil horizons.

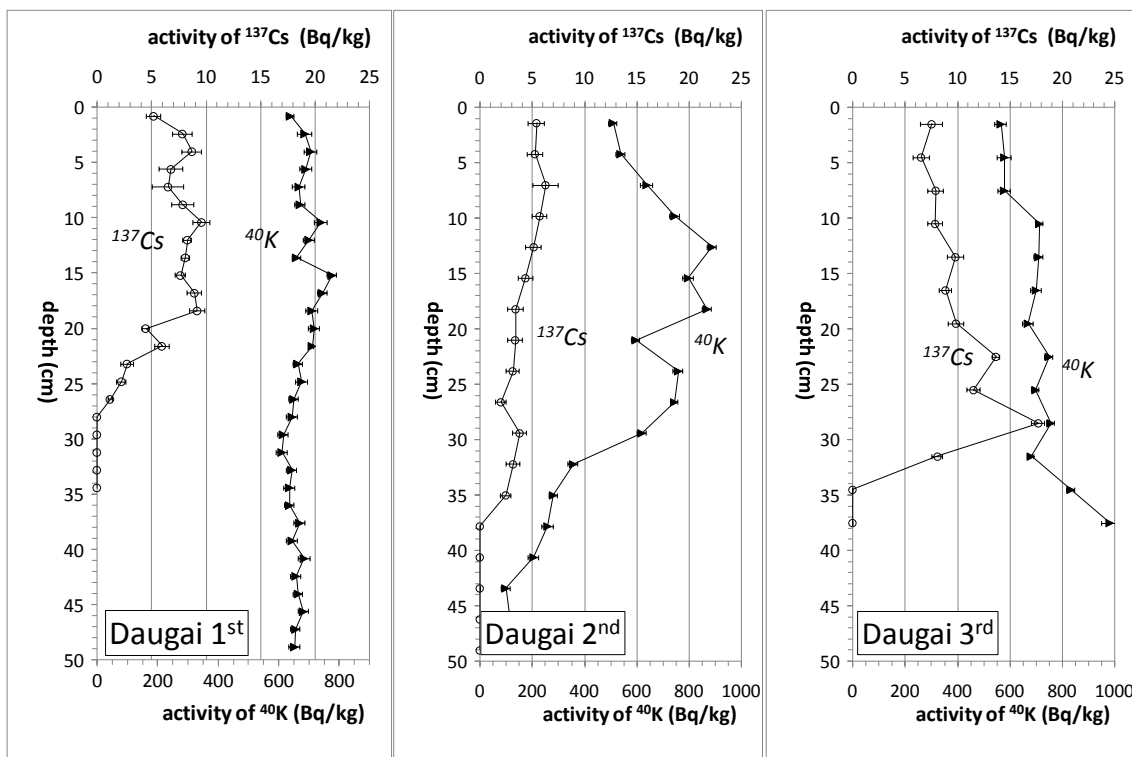


Fig. 4. Distribution of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in topsoil vertical sections and intervals of 95% confidence.

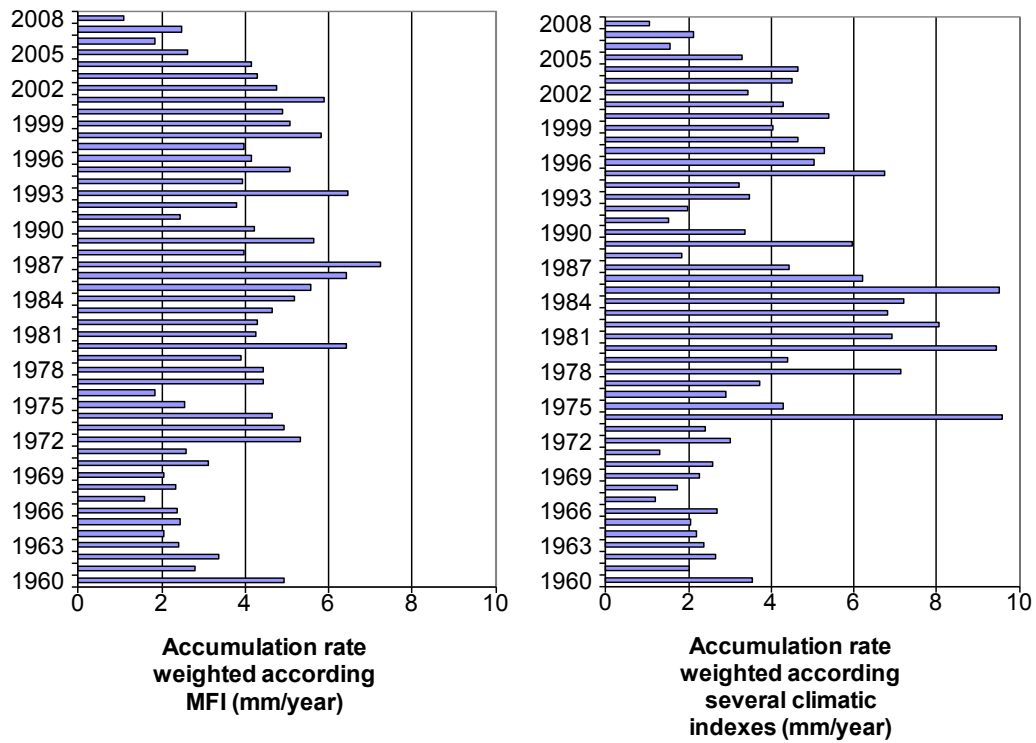


Fig. 5. Potential accumulation rate weighted according MFI and sum of >6 mm in warm period mm/year.

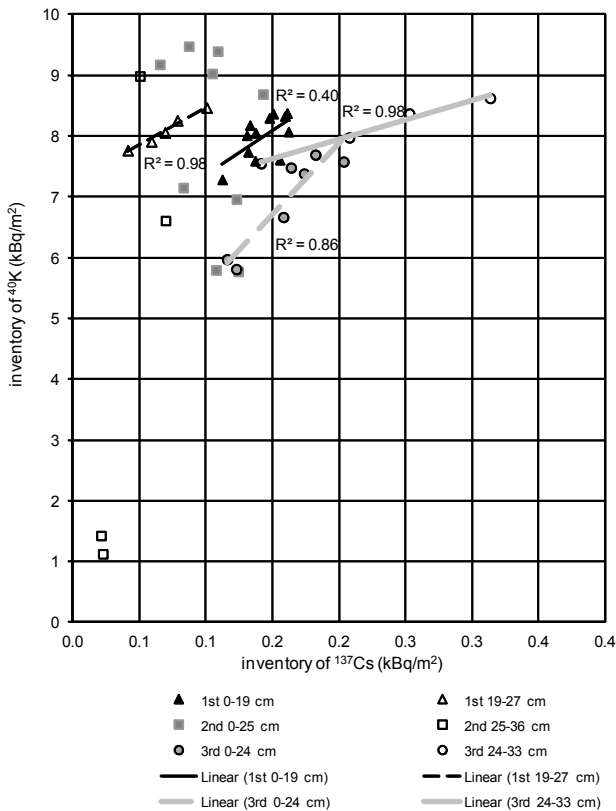


Fig. 6. Correlation between <sup>137</sup>Cs and <sup>40</sup>K inventories of topsoil for 1 cm sub-layers.

Erosion research methods can not be simply scaled up and down and with great range of possible conditions a single relationship between soil loss and slope length can not exist (Verheijen *et al.*, 2009). For this reason the scale of all the methods must be defined individually. In the case of applying universal equations (USLE, RUSLE), erosion rates are overestimated (Van Oost *et al.* 2000). Both universal equations and erosion potential (Racinskas, 1990) counts the possibility of erosion. The inventory of radionuclides in the accumulated soil material demonstrates actual erosion consequences. Measuring the erosion-accumulation by <sup>137</sup>Cs inventory distribution in the accumulated topsoil profile shows relative fluctuations of large scale soil and the actual erosion on the slope.

In our investigation the highest accumulation rate of eroded soil material in comparison with the average is 45 and 57% depending on climate indices (MFI alone or regional erosivity indexes). Though using radioactive elements for soil material accumulation studies is perspective for local soil and relief evolution and climate change studies.

## 6. CONCLUSIONS

Using the <sup>137</sup>Cs method alone reflects the integration of several landscape transformation processes and combination of <sup>137</sup>Cs and <sup>40</sup>K isotope in vertical section of accumulated topsoil carries out the information about



dominating origin of the accumulated soil material and in the case water erosion it is an opportunity to calculate changes of erosion-accumulation in different time spans.

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