University of West Hungary

Theses of doctoral (PhD) dissertation

Spreading of Radioactive Isotopes in the Environment (Cs-137-based Method for Surveying Soil Erosion)

Ervin Kiss

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University of West Hungary
Faculty of Forestry

Pál Kitaibel Doctoral School of Environmental Sciences
Geoenvironmental Sciences

Supervisor: Dr. Ferenc Divós

Choice of Topic, Objectives Set

The nuclear events of the past more than half century have started a process, a demand, to make the developments of nuclear events get publicity -- not only afterwards. As an effect of this, both the nuclear measurement methods and the detectors, measurement systems, used for measuring kept developing. Legitimate demand emerged that it should not be possible to keep possibly occurring nuclear events -- the pollution of which usually would not remain within the territory of the given country, but would along different transport routes reach further countries, territories -- quiet. As the measurement methods have been developing, as more and more sensitive detectors have been developed, and as, from these, national or international measurement networks have been formed, it is possible to trace the propagation and extent of any radioactive pollution, and by taking the meteorological conditions into calculation even the source of emission can be determined fairly well. However, systems developed can be resultfully employed not merely by the nuclear accidents and their aftermath, but also by surveying, detecting natural radioactive isotopes present. Knowledge gained about radioisotopes, both natural and artificial ones, in the natural and quasi-natural ecosystems, helps to better understand the ecological role of the given isotope, furthermore helps in tracing, modelling environmental processes, and in dealing with risks of the agent of a possible contamination. Among the artificial isotopes which got out in the environment, from the viewpoint of the biological effect of radiation, Cs-137 is one of the most important isotopes. It was produced in relatively large quantities during the detonations of atomic bombs, furthermore its physical half-life is ~30.2 years, so it survives in the environment for a long time.

On the European continent it was the deposition of radionuclides after the nuclear accident which occurred in the Chernobyl nuclear power plant in April 1986 that caused the higher radioactive pollution in the soil, plants, and other ecological systems. Cs-137 played a great role also in the remote effects of this pollution.

Global Cs-137 fallout by atmospheric nuclear weapon tests and the fallout originating from the Chernobyl accident also did reach Hungary's territory, which made the use of models based on measuring Cs-137 in soil erosion investigations possible also by us. Within the framework of the dissertation, one of the investigative models (the profile distribution based soil relocation rate estimation model) also used under international and homeland relations came, aligned with local conditions, to be applied in the Sopron Mountains, in an experimental area (Farkas-árok: Farkas Trench). Coupled to the application of the model, the objective of the dissertation is to survey and present the depth accorded distribution of Cs-137 in the soil of one of the water catchments (of Farkas Trench) in the Sopron Mountains, furthermore to present the spatial and temporal alteration of this depth distribution. A further objective by using these results is to determine, present the accumulation reference contents characteristic to the water catchment, the depth distribution function which could serve data for future investigations. Building upon these data, by means of the model based on the depth distribution of Cs-137, one is able to estimate the erosion conditions of the area, one can follow up the alteration of these erosion conditions, and one can give an evaluation relating to the adaptability of the model to the area.

Material and Method

Investigated Area, Sampling Plots

The Farkas Trench (0.63 sq. km), as investigated area, is located in the western part of the Sopron Mountains, is a southwest-northeast oriented trench along a branch of the Rák Brook. The elevation of the trench above sea level spreads from 401 m to 545 m, average slope inclination in the valley exceeds 21 %.

The sampling happened at the sampling plots along the slopes, defined in EOV coordinate system, at an adequate distance (min. 1.5 to 2 m) from the trees forming the forest stands, by means of a sampler of steel plate, down to a depth of 10 to 12 cm, at depth intervals of 2 cm. Any sampling to be performed along a grid of plots would have run into difficulties because of the terrain conditions, multi-plot sampling assigned along the slope was performed, instead. Preliminary sampling showed that a significant portion of the Cs-137 in the area is found in the topmost 10-cm layer of the soil, therefore sampling depth was adjusted accordingly. In 2001, at the reference plots, at a depth of 10 cm, there were one order of magnitude lower Cs-137 activity concentrations to be measured, than in the surface layer (0 to 2 cm). From the investigation area, in 2001, a total of 151 samples at 30 plots, then 9 years later, in 2010, 27 samples at 5 plots were collected.

Activity Measurement Method

Samples collected from the area were measured after braying, drying, pulverisation, sieving (<2mm). The samples were put into ½- or 1-liter Marinelli sample beaker dishes, and after this came determining the activity of the samples. The samples' Cs-137 activity was determined by gamma spectrometry at the University of West Hungary, in the Sopron laboratory of Hungary's Radiation Monitoring Detection and Verification System OSJER ("Országos Sugárzásfigyelő Jelző és Ellenőrző Rendszer"). Cs-137 activity was measured by a measurement device system comprising of a high-resolution ORTEC HPGe detector (Gem-10185), a high-voltage power supply unit, an amplifier, a low background lead chamber, and of a PC-attached multi-channel (8K) analyser card.

The Model Used for the Investigation

In the investigation area, the measurement of the spatial distribution of Cs-137 accumulation provides the basis for estimating erosion and sedimentation rates. Should the accumulated contents appear relatively drained, there it is to be presumed that some erosion have occurred, as well. The measure of erosion rate can be estimated from the measure of decrement rate of accumulated Cs-137 contents, by relating to the reference value. This measure represents an average value referring to the period of time between the fallout and the time of measurement. Where there has been deposition, an increase in accumulated Cs-137 contents will indicate it, in relation to the reference values. Average rate of deposition for the time period since the fallout can be estimated from the amount of excess Cs-137 accumulation contents.

As to estimate the erosion conditions of the investigation area, to convert the soil's Cs-137 accumulation contents into soil erosion rate, the profile distribution model was used. The model -- which in this environment contains several initial simplifications, e.g. that the fallout happened in a certain year -- was originally invented for measuring the Cs-137 originating from fallouts by nuclear weapon tests. The Cs-137 fallout duration following the Chernobyl accident and the Cs-137 profile generated after this fallout do eliminate, however, the reference time point, a mere hypothetical provision in the original model, and any uncertainty bound to it. In this case, in fact, temporally the fallout had lasted only for a short time, whereas in the original model the year 1963 stands for one certain year of a fallout time interval of several years. Thus, despite its simplicity, the model has its legitimacy for existence in this investigation area by the relatively simple, yet efficient estimation of soil erosion rates.

According to data in the literature, in the investigation area, the extent of deposition originating from Chernobyl was one order of magnitude greater than the activity concentration of nuclear weapon tests origin present in the soil at the time of the accident. Since it is all about the same isotope, the separation of the Cs-137 isotope deposited before, and, respectively, after 1986, and of their activity concentrations is not possible in retrospection, thus, the soil relocation conditions found during the investigation give information primarily about the period of time since 1986.

Results and their Evaluation

Within the framework of the dissertation, Cs-137 activity was investigated in the soil of a small water catchment (Farkas Trench) of the Sopron Mountains. Through the measurements, the distribution of Cs-137 by depth as well as the spatial and temporal alteration of this depth distribution were presented. The measurements verified that Cs-137 activity in the soil of the investigation area decreases by soil depth. On creating a graphic representation of this decrease in function of soil depth and by fitting a function to the data it became observable that the in-soil activity distribution alters accorded to a decreasing exponential function. From further analysis of the soil profile it was deducable that in any undisturbed soil profile in the area most (>90%) of the total Cs-137 activity is concentrated into the uppermost 10-12-cm layer of the soil.

In addition to the soil's Cs-137 contents, it was also investigated whether the isotope appears in living organisms. Within the framework of this, mushroom samples collected in the area were examined, in which the presence of Cs-137 was indicatable. The mushroom did take up and store away the Cs-137 present in the soil, so the isotope might enter the food chain of the forest also this way.

From the soil profile activity concentration data, the accumulated Cs-137 contents referring to the full profile of the lesser-sloped (slope inclination angle < 4 degrees) loci were determined, these were then used as reference levels for determining the soil movement conditions of the area (in 2001: 10569.9±380 Bq/m2, 9137±237.6 Bq/m2, 8909.8±363.1 Bq/m2; in 2010: 3887.7±288.4 Bq/m2). In the area of the Trench, at the sampling plots with greater than 4 degrees of slope inclination, Cs-137 accumulation contents varied between 988.9±413.8 Bq/m2 and 9419±253.1 Bq/m2. The values derived and their alterations may provide a reference basis for future erosion follow-up in this investigation area and its wider surroundings. Based on the activity concentration values, it can be stated that even several years after the pollution, forest soil still remains to be the main caesium store. It appeared in the 2010 measurements that the highest Cs-137 activity concentration had been shifting downwards from the upper layers, and the depth distribution of Cs-137 was (in function of slope inclination) altering along an increasing or stagnating function down to a depth of 4 to 6 cm, then along a decreasing exponential function. Based on the measurement results, the speed of the profile alteration is to be approximated as 0.44 to 0.66 cm/year.

In the investigation area, radioactive fallout originating from the Chernobyl accident caused approximately one order of magnitude greater Cs-137 content accumulation in the soil than the Cs-137 accumulation content, of nuclear weapon tests origin, present in the soil preceding the accident. Using these data, the profile distribution based soil relocation rate estimation model was aligned to the Cs-137 fallout conditions of the investigation area In the know of this model and of the Cs-137 accumulation contents of the sampling plots, soil erosion / deposition rates were estimated for these plots, which gives an image of the soil relocation conditions of Farkas Trench, and of their spatial alteration. According to the measurements, among the sampling plots, with the exception of one sample, there were only erosion plots. Samples close to the brook had smaller Cs-137 contents, which indicated greater soil movement. Erosion rate values for sampling plots varied between 0.15 and 28.55 t/ha/year, the deposition rate by the plot where there was depositing being 0.69 t/ha/year. By relating the values derived to the values derived by USLE model it was to be concluded that the soil erosion rates estimated by the profile distribution model do provide correct estimates of the extent of real soil erosion in the area. As a future research, through repeated investigation of the upper part of the area, analysis of samples, further, more accurate information could be acquired about the temporal alteration of erosion rates in the area.

Based on the measurement results it appeared that there is an interrelation between the surface (0 to 2 cm) activity concentration and the slope inclination of the sampling plot. When this same investigation was carried out for soil loss rate, results showed that the interrelation between soil loss rate and slope inclination does not, anyhow, show that close a connection in the investigation area.

Theses of the Dissertation

1. Measurements performed proved that the Cs-137 isotope is there, present in well measurable quantities in the soil and in living organism (mushroom) in the examined area (Farkas Trench / Farkas-árok), moreover that within the soil of the area, a significant part (> 90 %) of the total Cs-137 activity has concentrated to the top 10-12-cm layer of the soil. Measurements indicated that at the plots selected to be reference loci (slope < 4 degrees), the depth distribution of Cs-137 was shaped in accordance with an exponential function, and the Cs-137 profile of the other sampling plots (slope > 4 degrees) also neared to an exponential distribution.

2. Within the framework of the dissertation, the Cs-137 activity concentration values characteristic to the top (0-12 cm) soil layer of Farkas Trench, moreover the Cs-137 accumulation referring to this layer and the soil redistribution rates, were determined. The derived values and their alteration provide a picture of the Cs-137 activity values of the soil in the examined area, moreover a basis to relate to in future examination of erosion in the examined area and its wider surroundings.

3. Based on the measurements performed 9 years apart it could be confirmed that the shape of Cs-137 profile in the soil of the examined area does alter, and that by the 2010 measurements the maximal activity concentration was not to be found in the surface soil layer (as by the 2001 measurements), rather in a zone at 4-6 cm depth. Beneath that layer at 4-6 cm depth, the profile was still characterised by a decreasing exponential function, whereas the surface layers (0-6 cm) still showed either nearly stagnating (slope < 4 degrees), or increasing (slope > 4 degrees), activity concentration values in function of slope inclination. Based on the measurement results, the speed of alteration is estimated to be 0.44 to 0.66 cm/year.

4. In the area investigated, the radioactive fallout originating from the Chernobyl reactor accident caused approximately one magnitude-level greater Cs-137 accumulation in the soil than the Cs-137 accumulation originating from nuclear weapon tests, present preceding the accident, hence the Cs-137 which settled during the Chernobyl accident covered up the activity concentration originating from earlier fallout. Based on these data, the soil relocation rate estimation model based on profile-distribution, which is used in estimating erosion, was aligned for the period which has passed since 1986. Based on a comparison of soil erosion rate values referring to the area investigated with the estimate values derived by other models (USLE) it is to be concluded that the model based on profile-distribution can only be used for magnitude-approximations in the investigated area due to significant and altering vegetation cover as well as to significant slope inclinations in the area. The extent of erosion was overestimated by the values derived by the model based on profile-distribution.

5. Evidence was gained based on the measurement data that there is a connection between the surface (0-2 cm) activity concentration of the soil of a sampling plot

and the slope inclination of a sampling plot. On examining the same for soil loss rate, the results showed that the connection between soil loss rate and slope inclination does not appear so evident in the area investigated.

Publications

Ervin Kiss – Péter Volford (2013): Depth and Areal Distribution of Cs-137 in the Soil of a Small Water Catchment in the Sopron Mountains (Acta Silvatica et Lignaria Hungarica, Vol. 9 (2013) 147–159)

Kiss Ervin (2008): Környezeti minták radioaktivitása. Természet Világa 139. (2): 91.

Kiss Ervin (2003): Környezetünk nukleáris veszélyeztetettsége (Kankalin Környezettudományi Felsőoktatási Folyóirat (www.kankalin.bme.hu) 8/2003, Online: http://www.kankalin.bme.hu/Dok/Dip.doc

Kiss Ervin (2003): Cézium-137 az erdőben. Élet és Tudomány 58. (48): 1524 - 1525.

András Varga, Ágnes Matusik, Ervin Kiss, Ferenc Divós (2000): Use of medical CT to qualify defects in wood (Proceedings of the 12th International Symposium on Nondestructive Testing of Wood, Sopron 13-15 September 2000, pp.466)