Content and Behavior of Radiocesium in Undisturbed Bulgarian Soils

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Abstract

Results of the radiological monitoring of radiocesium content in soils from different regions of the country are presented and discussed in this work. Plain, hilly and mountain areas covering two thirds of the country are included in the study. The samples are analyzed by gamma-spectrometry. Some aspects of the fixation of Cesium in the soil are studied. Laboratory experiments were carried out trying to explain the quick distribution of Cs down the soil profile in 1986, immediately after the Chernobyl's contamination. It was established that the mobility of Cesium in soil is higher in the initial period after deposition and decreases with time.

Key words: Soil, Radioactivity, Gamma-Spectrometry

Introduction

The radioecological studies of earth's crust and especially soil are very important part of radioecology as they give basic data for estimation and prediction of radionuclide content in agricultural production and thus in food and man. Nuclear accidents, such as the Chernobyl accident, have released large amounts of radionuclides into the environment. The accident in Chernobyl nuclear power plant (ChNPP) caused the largest uncontrolled radioactive release into the environment dispersed on over the entire northern hemisphere It was estimated that about 85 PBq of ^{137}Cs were released in the accident (UNSCEAR, 2000). The radionuclides with long half-life like ^{137}Cs deposited after the accident remain in the environment, mainly in the soil, for decades and are relevant for the environmental monitoring. Analysis of radionuclide content of soil, plants, water and knowledge of the behavior of the radionuclides in soil–plant system provides an important part of a data basis for dose estimation (UNSCEAR, 1993 and UNSCEAR, 2000).

Bulgaria was among the European countries with relatively high contamination as a result of Chernobyl accident. The activity concentrations of man-made radionuclides in the Bulgarian soils increased considerably.(Tsvetkov, et al 2006.). Subject of the studies were undisturbed soils from high mountain areas, hills and plains (the region of Kozloduy NPP and the Danube river valley).(Staneva, et al., 2012; Yordanova, et al.2006; Staneva, et al., 2006). The areas around Kozloduy NPP are of special interest. Such data can be used as a baseline from which we can detect future release of radioactive nuclides.

The mobility of radiocesium in the soil is one of the most important factors controlling its long-term transfer to man via terrestrial pathways. That is why the knowledge about the behavior of this radionuclide in the soil is of significant importance, as the insufficient understanding of the fundamental processes taking place in the soil compartment causes large uncertainties on the environmental forecast.

Up till now, 30 years later, ^{137}Cs is still detected in all soil samples and represent a potential danger for the contamination of the plant production through the root feeding. Our work aims assessment of local background values for ^{137}Cs in soil on the base of regular radiological monitoring of virgin soils from high mountain areas, hills and plains covering most of the territory of Bulgaria.

Material and Methods

For the purposes of the radiation monitoring of the soils in our country sampling of undisturbed soils is done annually from one and the same sampling sites covering almost the whole area of the country. Sampling areas are specified considering the wind direction and difference in altitude so-that different soil types are included. According to the altitude of the investigated areas three groups have been defined: plains – mainly North Bulgaria, (around the NPP Kosloduy and along the river Danube – 40 sampling points *[sp]*); semi-mountainous (the Sofia plain – 10 sp, and the valleys of Struma and Mesta rivers (15 sp)) and mountains area - in South Bulgaria (the Rodopa mountain– 25 sp).(Yordanova I., et al.2014; Yordanova I., et al.2015)

The soil samples were taken according to the procedure defined in Bulgarian Governmental Standard BGS17.4.5.01-85 for soil sampling from permanent sites annually from one and the same sampling points from the soil layer 0-5 cm were collected. At some sites the samples are collected from up to 40 cm depth in layers of 5 cm.

The soil samples are homogenized, dried at 80ºC and sieved through a 2-mm mesh before measurement with a gamma-spectrometer. A Canberra high-purity germanium detector with 20% efficiency an energy resolution of 1.8 keV for ^{60}Co γ-ray energy line at 1332 keV was used. The measuring system included a multichannel analyzer DSA 1000(Canberra, USA). The spectrum was analyzed by GENIE-2000 software with measurement uncertainties less 10%. Typical counting times were 19–24 h. The ^{137}Cs concentrations in soil were obtained by measuring the activity at 661.62 keV.

For the laboratory experiments we have used – *Fluvisols* soil, widespread in our country. This soil was part of a long-term vegetation pot experiment for studying the uptake of cesium by plants from contaminated with radiocesium (^{134}Cs) soil. (Ts. Bineva et al. 2012; D. Staneva et al, 2009)

The soil was additionally contaminated with ^{137}Cs in chloride form. Desorption of Cs was done with a lactate-acetate buffer, used for determination of exchangeable forms of K^+ in the soil (Radov A. C., et al. 1985). Soil aliquots (2g) were suspended in desorption solutions and shaken. After centrifugation the supernatant was removed and analyzed for ^{137}Cs using gamma spectrometry.Different time periods and solid: liquid phase ratios were tested in order to find optimal conditions for desorption and determination of different Cs fixation-sites

Results and Discussion

Until 1986 the mean values for the specific activities of ^{137}Cs were as follows: Northern Bulgaria - 10 Bq.kg^{-1} and Sothern Bulgaria - 26 Bq.kg^{-1} (Najdenov M., 1986, Najdenov M., Staneva D., 1987). These values were received by calculating the arithmetic mean from the results for specific activities in surface soil samples for each area observed.. The mean square deviation at this averaging was up to 40%.

After the Chernobyl disaster in 1986 the radio-ecological status of the soils in Bulgaria concerning the men-made radioactivity has changed radically because of the massive deposition of radioactive elements

Results for Ceasium-137 activities in soils are presented on figures 1..

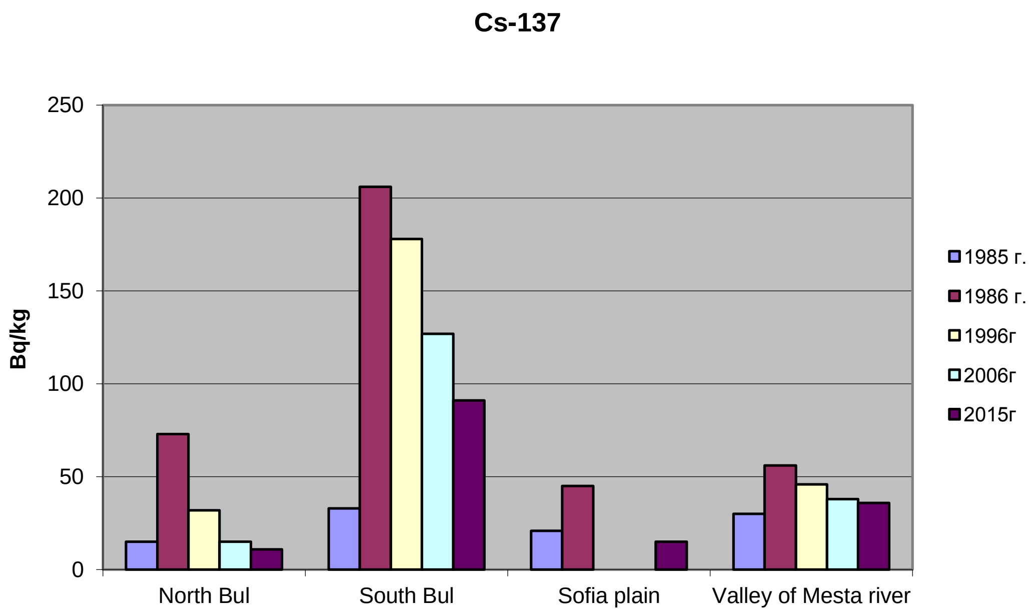

Fig. 1. *Average values for ^{137}Cs content in soils [Bq.kg^{-1} dry wt]*

It is to be noted that the surface pollution is very non- homogeneous and this brings about great variations in calculating the arithmetic mean values. For Ceasium-137 the variations are between 30% and 60% for the different areas. This is the reason why it is difficult to make categorical evaluations of an obvious increase or reduction of the contents of these radio-nuclides in the upper soil layers. The most probable reason for this non-homogeneity is the type of deposition in May 1986 after the Chernobyl's accident. It was wet deposition for a short time combined with heavy rainfall.

The specific activities of ^{137}Cs are four - five times higher in the soils from Southern Bulgaria than in those from Northern part. For SB the mean values vary in the years between 160 and 280 Bq.kg^{-1} , while for NB – between 40 and 60 Bq.kg^{-1}.This can be explained with the difference in altitude and what is more the radioactive cloud in 1986 passed twice the Southern part of the country and there the quantity of the rainfall was higher.(Yordanova I., et al. 2007)

We can compare these results with data from neighbouring country like Greece (Vosniakos F.K., et al. 2000) They publish results for mean concentration of ^{137}Cs in soils for the period 1986-1990 (44.3 ± 28.6 Bq.kg^{-1}). This difference in the level of contamination is caused by the difference in meteorological conditions, type of deposition and difference in altitude.

After 1996 the researches on status and dynamics of the radio-active pollutions of the soils in the country was directed mainly towards an expansion of the sites number for the purpose of encompassing a greater territory, whereby regions have been researched on, which have potentially been exposed to radio-active pollutions, as for instance the high mountainous soils of the Rila and Pirin mountains.Figure 2 shows the results obtained from the analyses of soils in Northern Bulgaria, the plain area (around the Nuclear Power Station Kosloduy and along the river Danube); areas along the rivers Struma and Mesta, and the region of the Rila and Pirin mountains.As becomes obvious, the contents of radioceasium in the soils of Northern Bulgaria and the valleys of the rivers Struma and Mesta are comparable while for the mountains soil (Rila and Pirin), relatively higher values for this radio-nuclide have been observed, which is natural due to the geographic situation.

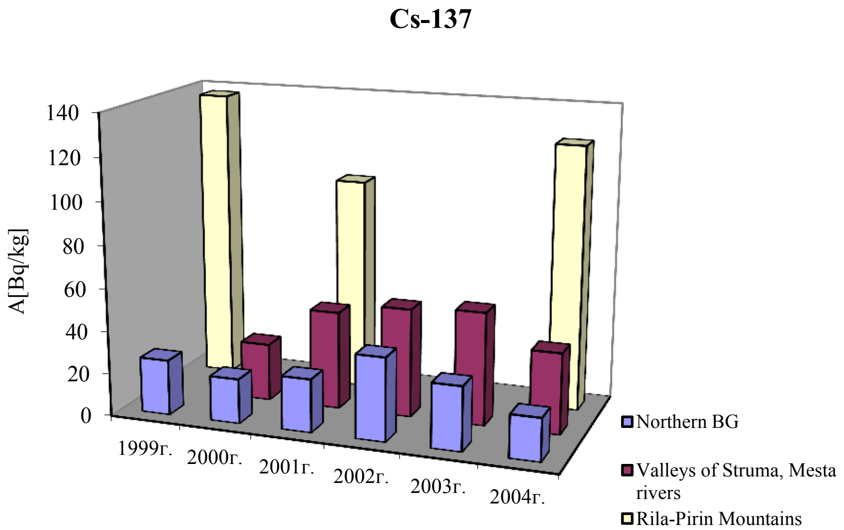

Fig.2. *Average values of Cs-137 content in soils [Bq.kg^{-1} dry wt] for the period of 1999–2004*

The results presented in fig.1 show that except for South Bulgaria (the Rhodope Mountains) the amount of ^{137}Cs in the surface soil layer of 0-5 cm for 2015 is comparable to that before the accident in Chernobyl's NPP,. This decrease is mainly due to the natural decay of cesium 137 and to a very small degree to migration processes in depth of the soil profile.

During the first months after the accident in 1986 we established that Chernobyl's Caesium indicated by ^{134}Cs had migrated down to a depth of 30 cm .(Tsvetkov Ts., et al 2006.), a fact reported by other authors too (Wolfgang S. and Kurt B., 1996). Fig. 3 shows the distribution in depth of Cs in three different soil types -*Luvic phaeozem, Phaeozem, Fluvisols (FAO classification*)

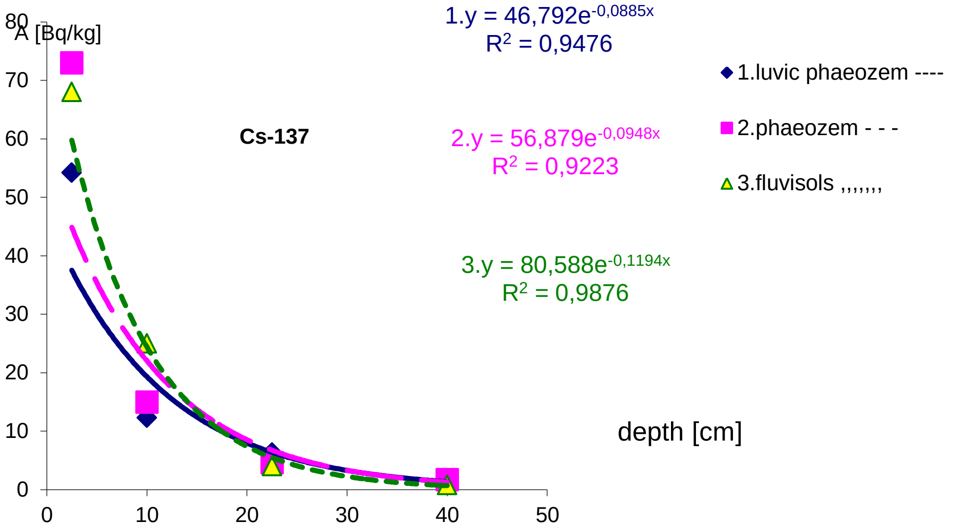

Fig. 3. *Distribution of ^{137}Cs in depth down the soil profile*

Our observation in the years following the Chernobyl accident did not show significant changes in this distribution of Cs down the soil profile and intensive migration (*Tsvetkov Ts., et al 2006*). For this reason we performed our laboratory experiment to study the behavior in soil of “old” and newly incorporated cesium. ^{134}Cs – as part of the long-term vegetation pot experiment was in the soil for more than ten years. ^{137}Cs was applied additionally and after twenty days desorption was carried out.

Based on the fact that Caesium and Potassium are analogues as chemical elements, we applied lactate-acetate buffer used to determine the exchange forms of Potassium. The ratio between solid and liquid phase was 1: 25, which is accepted in the methodology for defining the exchangable forms of Potassium (Radov A. C., et al. 1985). In this case our purpose was to study in more details the degree of fixation of a newly introduced Caesium or Caesium, which has stayed in the soil for years, for which we used *Fluvisols*. The results are presented in table 1.

Table 1. *Percentage of activity desorbed with lactate-acetate buffer*

.	1 hour shaking	2 hour shaking	3 hour shaking	4 hour shaking	5 hour shaking	6 hour shaking
^{137}Cs	40%	40%	50%	40%	50%	54%
^{134}Cs	4,6%	5,4%	4,5%	4,5%	3,1%	3,7%

With this experiment a considerable difference in behavior between a newly introduced (newcomer) and an “old” Caesium is displayed. In both cases at the end of the second hour a dynamic balance was established, where the degree of desorption did not change.Notwithstanding the established strong fixation of Caesium in the soil with time the plants grown on soil contaminated with ^{134}Cs absorb the element and with the years no considerable reduction of the transfer coefficients was to be observed (Bineva Tz., et al. 2005; Staneva D., et al. 2004).

Conclusion

As a conclusion we may say that:

The levels of ^{137}Cs established in the surface soil layer of 0-5 cm for 2015 are comparable to those detected before the accident in Chernobyl's NPP for North Bulgaria and the plain and hilly areas of South Bulgaria. This decrease is mainly due to the natural decay of ^{137}Cs;

In the region of Rodopa mountains in South Bulgaria the levels of ^{137}Cs are still relatively higher as the deposition of radionuclides in 1986 was more intensive on these areas.

The amount of radiocesium in the soil from the studied areas combined with the established immobilization of Cs with time are such that do not require additional initiatives for radiation protection.

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