DYNAMICS OF THE RADIOACTIVE POLLUTION IN THE SURFACE LAYER OF SOILS IN BULGARIA TWENTY YEARS AFTER THE CHERNOBYL NUCLEAR POWER PLANT ACCIDENT

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ABSTRACT

The twenty years period past after the contamination with radionuclides in 1986, as a result of the accident in the Chernobyl's NPP, allowed the accumulation of rich data base for the radiation status of the soils in Bulgaria.

Objective of many years studies were virgin soils from high mountain areas, hilly and flat (the region of Kozlodouy NPP and the Danube river valley). Ceasium-137 and strontium-90 were the main men-made radionuclides detected in the examined Bulgarian soils, few years after the accident. The content of ceasium-137 and strontium-90 in the soils from high mountain areas (Rodopa and Rila mountains) is several times higher then that in the soils from Northern Bulgaria and Sofia field. High non-homogeneity in the pollution within small areas (scores of square meters even) was determined. No significant horizontal redistribution was observed for the period after 1986. The tendency of changes in the radioactive status of the soils in Bulgaria after the accident at the Chernobyl Nuclear Power Plant is not due to trans-border transfer of radioactive materials or to any breakdown at the Kosloduy Nuclear Power Plant.

Key words: environmental radioactivity, soil, pollution, ceasium-137, strontium-90,

РЕЗЮМЕ

Изминалият двадесетгодишен период след масираното отлагане на радионуклиди през 1986 година, като резултат от аварията в Чернобилската АЕЦ, даде възможност да бъде натрупана богата база данни за радиационния статус на почвите в България.

Обект на дългогодишни изследвания бяха целинни почви от високопланински райони, полупланински и равнинни (района на АЕЦ "Козлодуй" и поречието на р. Дунав. Няколко години след инцидента цезий-137 и стронций-90 са основните техногенни радионуклиди се детектират в изследваните български почви. Съдържанието на цезий-137 и стронций-90 в почвите от високопланинските райони (Родопите и Рила планина) е неколкократно по-високо от това в Северна България и Софийското поле. Установенеа бе силна нехомогенност на замърсяването, дори в рамките на малки площи (десетки квадратни метра). За периода след 1986 година не се наблюдав значимо хоризонтално преразпределение. Тенденциите в изменението на радиационния статус на почвите в България след авариятав Чернобилската АЕЦ не се дължат на трансграничен пренос на радиоактивни материали или някакво нарушение в дейността на АЕЦ Козлодуй.

Ключови думи: радиоактивност, околна среда, почва, замърсяване, цезий-137, стронций-90.

Introduction

Continuous radiation control is performed in the country, since the end of the sixties because of the operation of the Bulgarian NPP "Kozlodouy",. Ceasium-137 and strontium-90 are the main men-made radionuclides detected in the soils. Until 1986 the mean values for the specific activities of the above mentioned radionuclides were as follows: Northern Bulgaria - ^{90}Sr - 4 Bq.kg^{-1} and ^{137}Cs - 10 Bq.kg^{-1}; Sothern Bulgaria - ^{90}Sr - 4 Bq.kg^{-1} and ^{137}Cs - 26 Bq.kg^{-1} [1, 2]. These values were received by calculating the arithmetic mean from the results for specific activities in soil samples from 25 different sampling points in Northern Bulgaria and 20 such in Sothern Bulgaria. The mean square deviation at this averaging was up to 40%.

The massive deposition of radioactive elements after the accident at the Chernobyl Nuclear Power Plant has changed radically the radio-ecological status of the soils in Bulgaria. After the first several months following the incident, essential for the radiation status of the soils were the contents of the long-lived isotopes of Ceasium-137 and Strontium-90. Until 1989 in the soils of some regions of the country, where the impact was greater one could observe rest quantities of up to 10-15 Bq.kg^{-1} Rutenium-106 and Antimonium-125, which are getting less in a natural way due to the shorter period of half-life time and which do not have an essential impact on the general pollution of the soils [3].

Material and methods

Object of our study are the two comparatively long-lived radioisotopes, namely Strontium-90 and Ceasium-137 not only because they represent a potential danger for the contamination of the plant production through the root feeding but also due to the fact that because of their long periods of half-live, it is possible to follow them for a longer period of time.

For the purpose of making an assessment of the upper strata pollution of the soil we have carried out analyses of soil samples from the soil layer 0-5 cm. Sampling points were specified considering the wind direction, and 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.

Sampling is done annually from one and the same sampling points. Depending on the place of taking the sample, three groups have been defined: Northern Bulgaria, which is a plain area (around the NPP Kosloduy and along the river Danube – 40 sampling points [sp]); semi-mountainous (the Sofia plain – 10 sp) and Southern Bulgaria (the Rodopa mountains area – 25 sp). The water catchment basin on the Beli Iskar river in the Rila mountains (5 sp) and the valleys of Struma and Mesta rivers (15 sp) were included to the investigated regions lately. In this way we have covered almost the whole area of the country which means that different soil types are included. In this paper we will not discuss in details the vertical distribution in soil profiles. That is why here we will not comment the effect of different soil types on migration processes. We have done investigation concerning these problems and the results are published [3, 4, 5].

The soil samples taken have been analyzed by the gamma-spectrometric method. A high-purity germanium detector with 20% efficiency was used. The measuring system included a multichannel analyzer DSA 1000. The experimental errors were less then 10%.

Radio-chemical determination of Strontium-90 has been carried out. [6]. ^{90}Sr was determined by its progeny ^{90}Y. The radiochemical procedure used is based on extraction of yttrium with TBP (tributylphosphate) and precipitation with oxalic acid. The measurement was done on low-background beta-measuring system, operating in an anti-coincident mode.

Results and discussion

To enable a more obvious depiction of the assessment of the changes in the radiation status of the soils we have presented averaged values for specific activities of the Ceasium-137 and Strontium-90 (figures 1 and 2 respectively) in soils from Northern Bulgaria and Southern Bulgaria for the first ten years following the accident (1986-1996). These values were received by calculating the arithmetic mean of the results from 40 soil samples from 40 different sampling points in Northern Bulgaria [NB}and 25 – from Southern Bulgaria [SB]. It is to be noted that the surface pollution is very non-homogeneous and this brings about great deviations in calculating the arithmetic mean values. For Strontium-90 the mean square deviations are between 20% and 80% for the different areas and for Ceasium-137 they are between 30% and 60%. 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 to 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

Table 1: Content of Cs-137 and Sr-90 in high mountainous soils from Rila mountains 1996 [in Bq.kg^{-1} layer 0–5 cm]

Site	Cs-137	Sr-90
1.Allinitsa	1208±1%	100
2. Allinitsa 2	298±1%	–
3. Near lake, at hut Moussala	1643±1%	–
4. Above the lake at hut Moussala	1123±1%	–
5. Borovets	1105±1%	49

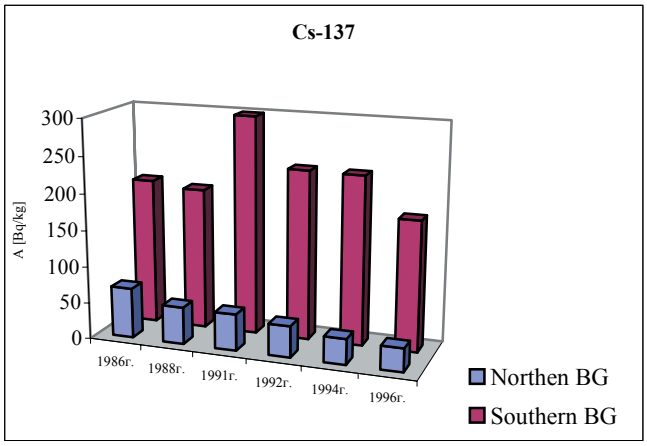

Fig.1: Average values of Cs-137 content in soils [Bq.kg^{-1} dry wt] for the period of 1986–1996

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.

We can compare these results with data from neighbouring country like Greece [7]. In the book cited, the authors report values of 44.3 ± 28.6 Bq.kg^{-1} for mean concentration of ^{137}Cs in Greek soils for the period 1986-1990. These concentrations are several times lower then those determined by us in the Southern part of Bulgaria. This difference in the level of contamination is caused by the difference in meteorological conditions, the type of deposition and difference in altitude.

In 1991 a tendency becomes evident of increase of the average values of the Ceasium-137 contents in the Rodopa mountains. This is most probably due to the fact that a more-active plant mass (pine needles) penetrates the soil, which is due to the specifics of the plant cover in this area. During the following years a small reduction of these values is observed, whereas the differences fall within the framework of the variations of unevenness of the pollution. During the last year the level of 1986 was achieved, whereas no impact of the plant mass, which has penetrated the soil afterwards, is to be noticed.

Several years later (1992-1994) a similar tendency is to be observed also with the Strontium-90 pollution in this area. For 1992 the values are as follows: 14,0 ± 4,9 Bq.kg^{-1} and for the year 1994 the values are 12,7 ± 6,7 Bq.kg^{-1}. The differences, when comparing with the preceding years, fall again within the framework of the non- homogeneity of the pollution. This displacement in the time regarding the two radio-nuclides is probably due to the difference in the forms of deposition and the methods of determination. The contents of Ceasium-137 have been determined in a non-destructive way, by gamma - spectrometric analysis, whereas for Strontium-90 a radio-chemical procedure has been applied, where the Strontium-90 is extracted from the soil by means of an acid. Probably with the time processes are taking place in the soils, which free the Strontium-90 from its acidic-non-soluble forms and this enables the detection of these forms by the method used.

The levels of Strontium-90 observed during the last

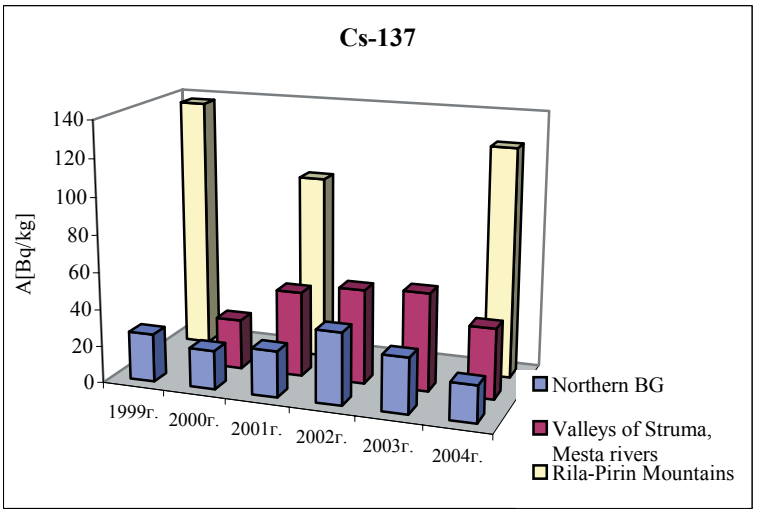

Fig.2: Average values of Cs-137 content in soils [Bq.kg^{-1} dry wt] for the period of 1999–2004

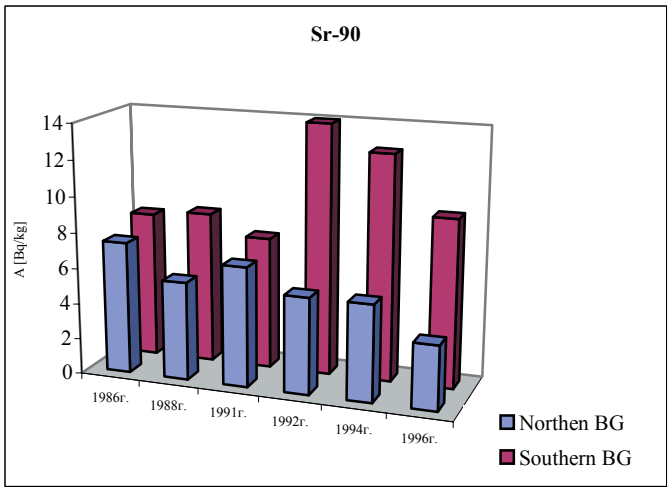

Fig.2: Average values of Sr-90 content in soils [Bq.kg^{-1} dry wt] for the period of 1986–1996

couple of years are slightly over those detected in the period 1986- 1991 (Figure 2)

Non- homogeneity of the Ceasium-137 and Strontium-90 isotopes distribution in the area observed becomes evident.

The comments on the mean values notwithstanding the great deviations when obtaining the latter, serves the purpose of information and is representative for the general status of the radio-active pollution of the areas under survey.

The results from the research on high mountainous soils (with an elevation of 1700-1900 meters altitude) have been presented in table 1. These samples have proved a strong non- homogeneity of the pollution, namely from 3 to 1700 Bq.kg^{-1} for Cs-137, whereas the highest values have been given in the table.

When studying the migration of the two radio-nuclides in depth of the soil profile during the first months after the accident in 1986 we established that radioceasium had penetrated down to a depth of 30 cm [3, 8], a fact reported by other authors too [9]. In the following years we have not observed significant change in this distribution; therefore the migration processes for ceasium have not

influenced its content in the surface soil layer.

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. Regions, which have potentially been exposed to radio-active pollution, have been researched on. An example for such regions are the high mountainous soils of the Rila and Pirin mountains, the areas along the rivers Struma and Mesta, etc.

Figure 3 shows the results obtained from the analyses of soils in Northern Bulgaria, the plain area (around the Kosloduy NPP 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.

The above mentioned expressed non- homogeneity of the pollution has been registered also during the last 10 years, that is 1997- 2006. The contents of Ceasium-137 vary within broad limits and no considerable change has been noted in the radiation status of the soils in the areas under observation.

Conclusions:

- In 1986 the radio-active status of the Bulgarian soils has strongly changed due to the accident at the Chernobyl Nuclear Power Plant.
- Ceasium-137 and Strontium-90 are the basic radio-nuclides to be detected in the soils until our last measurements.
- The contents of Ceasium- 137 and Strontium-90 in the soils of the Rila and Rodopa mountains are higher than these in Northern Bulgaria and the Sofia plain. The contents of Ceasium-137 is five- to six times higher at some places.
- In 1991 an expressed tendency of increase of the Ceasium-137 contents in the soils of the Rodopa mountain was observed, the same was observed in 1993 for Strontium-90 as well. It has to be noted that this change is not due to trans-border transfer of radio-active materials
- The tendency of changes in the radio-active status of the soils in Bulgaria after the accident at the Chernobyl Nuclear Power Plant is not due to trans-border transfer of radio-active materials or to any break-down at the Koslodouy Nuclear Power Plant.

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