

Content of ^{90}Sr and ^{137}Cs in Sediments and Soils from the Vicinity of the NPP Kozloduy during the Period 2008-2010

R. Kotova, R. Kamenova-Totzeva, V. Badulin

National Center of Radiobiology and Radiation Protection, 3 Sv. Georgi Sofijski Str., 1606 Sofia, Bulgaria

Abstract. The systemic radiation monitoring of the environment in the vicinity of the Kozloduy NPP is exceptionally significant part of the radiation protection. For monitoring purposes and in accordance with the control program annually samples are being analyzed to determine the content of anthropogenic long-lived radionuclides of a biological significance such as ^{90}Sr and ^{137}Cs . The goal of performing radio-ecological studies is to assess the impact of gaseous and liquid effluents during the normal operation of the nuclear power plant and to determine the potential radiological risk to the population in this region and to the population as a whole.

In this paper are presented the results from the analyses of the content of ^{90}Sr and ^{137}Cs in soils and bottom sediments at 17 sampling sites, located from 6 to 90 km around the Kozloduy NPP in four Bulgarian districts. The obtained results of the investigation and analyses for the period 2008-2010 are shown in tables and figures. Annual results are compared with those from previous years, as well as with data from the pre-operational period of the Kozloduy NPP.

Keywords: NPP Kozloduy, soils, bottom sediments, ^{90}Sr , ^{137}Cs .

1 Introduction

Since 1972 until now National Centre of Radiobiology and Radiation Protection (NCRPP) in Sofia carries out systemic radiation monitoring and control of environmental objects in the vicinity of the Kozloduy NPP [1-7].

The main goal of this work is to present the obtained results from the investigation of the content of anthropogenic long-lived radionuclides of a biological significance such as ^{90}Sr and ^{137}Cs in environmental objects. These radioisotopes should be most closely guarded against release into the environment. They both have half-lives of around 30 years, which is the worst range for half-lives of radioactive contaminants. In a study of bottom sediments important information is obtained about the accumulation of radionuclides in aquatic ecosystems and soils research provides information on the accumulation of radionuclides in the terrestrial ecosystem.

Bottom sediments and soils are being analyzed to determine the content of ^{90}Sr and ^{137}Cs for three years period 2008–2010. The samples were collected at 17 points in the surveillance zone – from 6 to 90 km in the region of the nuclear plant. Sampling sites are located in four Bulgarian districts – Vidin, Montana, Vratsa and Pleven. The annual results are compared with those from previous years, as well as with the data from the pre-operational period of the Kozloduy NPP.

2 Materials and Methods

Sites of sampling included in the monitoring program are selected on the basis of demographic and meteorological characteristics of the area and sampling points are located

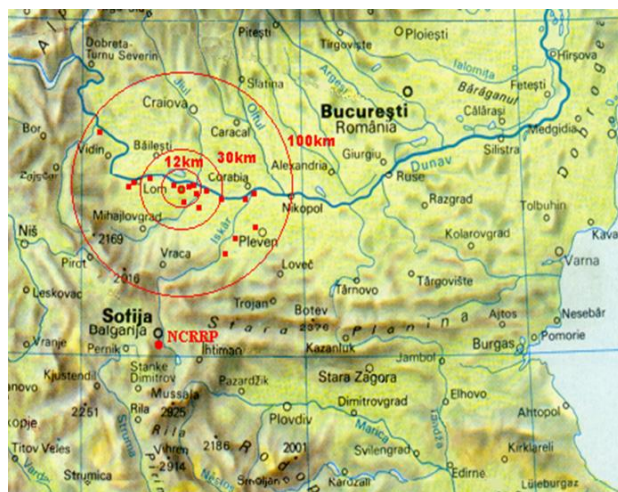


Figure 1. Sampling sites and points around the Kozloduy NPP.

at a distance of 6 km to 90 km from the NPP. They are shown in Figure 1.

Bottom sediments were collected from the internal rivers – Tsbritsa, Ogosta, Lom and Iskar and from the upper and lower stream of the Danube, to the location of the Kozloduy NPP. Uncultivated soils were examined as the content of ^{90}Sr and ^{137}Cs is defined in the surface layer of a thickness of 10 cm. The soils of the sampling points on the Danube river are soils of land flooded by the river. For the period 2008-2010 are analyzed as a total of 133 soils and bottom sediments (Table 1), sampling twice per year in spring and autumn. The total number of the analyses is 266.

After sampling the soils and bottom sediments are air dried, cleared out of stones, roots and other impurities.

Table 1. Analytical methods for analyses of environmental objects

Environmental objects	Number of samples	Gamma spectrometry of ^{137}Cs	Radiochemistry of ^{90}Sr	Total number of analyzes
Bottom sediments	55	IEC 61452 (1995-09)	ILVM ^a	110
Soils	78	IEC 61452 (1995-09)	ILVM ^a	156

Note: ^a ILVM – Internal laboratory validated method



Figure 2. Radiochemical prepared samples for ^{90}Sr measurement.

After that samples are grinded and sift through sieve with diameter of the holes 1 mm, homogenized and burned in a muffle furnace.

The radiation status of the studied environmental objects is determined by using methods of analysis presented in Table 1. For assessment of technogenic pollution of the soil and bottom sediment surface with ^{90}Sr is applied radiochemical separation according to the method of Bryant [8] with some modifications and radiometric measurement of ^{90}Sr by its daughter product ^{90}Y after equilibrium. 200 g soil sample is used for analysis. For sediment samples the quantity is the same. A known amount of stable Sr was added to each sample to determine the chemical yield. The ash was dissolved and alkaline earth metals separated by carbonate precipitation. Strontium was purified from other elements in the matrix using various precipitations such as fuming nitric acid, chromate, hydroxide and carbonate precipitation. Stable Y was added to the sample, which was allowed to stand for 18 days to reach equilibrium between ^{90}Sr and ^{90}Y . ^{90}Y was then precipitated as a hydroxide followed by oxalate precipitation. The measurement of prepared samples (Figure 2) is performed by low level background beta counting system with scintilla-



Figure 3. Gamma-spectrometric system.

tion detector. The instrumentation is calibrated with certified reference material and it has efficiency 48% for measurement of ^{90}Y . In standard conditions including 3000 s measuring time, the minimal detectable activity (MDA) is in ranges from 0.09 Bq/kg¹ to 0.15 Bq/kg¹. ^{137}Cs is determined by gamma-spectrometric analysis (Figure 3).

The accuracy of all applied analytical procedures is systematically checked by participation in national and international inter-laboratory comparisons organized by IAEA, IRRM, WHO and etc. [9,10].

3 Results and Discussion

The annual results from the analysis to determine of the activity concentration of ^{90}Sr and ^{137}Cs in bottom sediments are presented in Figures 4 and 5 and for soils samples – in Figures 6 and 7. The obtained results for the period 2008-2010 are shown and compared with the upper limit of natural background content of ^{90}Sr and ^{137}Cs in bottom sediments and soils with a bold red line on the same figures.

3.1 Bottom sediments

The annual content of ^{90}Sr (Figure 4) in bottom sediments from the Danube varies within the ranges from 0.21 ± 0.06 Bq/kg¹ (Ostrov, 2008) to 0.81 ± 0.09 Bq/kg¹ (Novo selo, 2009). The annual content of ^{137}Cs (Figure 5) is between 4.02 ± 3.87 Bq/kg¹ (Baykal, 2010) to 29.33 ± 4.70 Bq/kg¹ (NPP, 2008). In terms of the internal rivers the content of ^{90}Sr is in the range of 0.22 ± 0.08 Bq/kg¹ at the river Tsibritsa (2008) to 1.26 ± 0.14 Bq/kg¹ at the river Lom (2009). The annual content of ^{137}Cs is between 1.58 ± 0.71 Bq/kg¹ at the river Tsibritsa (2010) to 10.98 ± 0.81 Bq/kg¹ at the river Lom (2008).

In previous years [5-7] (2003-2007) the annual content of ^{90}Sr in bottom sediments from the Danube is in the ranges of 0.14 ± 0.05 Bq/kg¹ (Baykal, 2003) to 0.50 ± 0.08 Bq/kg¹ (NPP, 2006) and from 0.20 ± 0.05 Bq/kg¹ at the river Tsibritsa (2003) to 0.84 ± 0.08 Bq/kg¹ at the river Lom (Lom, 2007) for the internal rivers. The annual content of ^{137}Cs varies between 0.86 ± 0.25 Bq/kg¹ (Lom, 2007) to 40.97 ± 28.70 Bq/kg¹ (NPP, 2003) for the Danube and from 1.95 ± 0.91 Bq/kg¹ at the river Tsibritsa (2007) to 16.30 ± 15.41 Bq/kg¹ at the river Lom (2005) for the internal rivers [5,7].

The annual concentrations of ^{90}Sr in bottom sediments are in the range of values recorded in the previous periods [4-7] and are lower than the measured in pre-operational period of the plant – 5.4 ± 0.2 Bq/kg¹ for the internal rivers and 2.6 ± 0.6 Bq/kg¹ for the Danube [1]. In com-

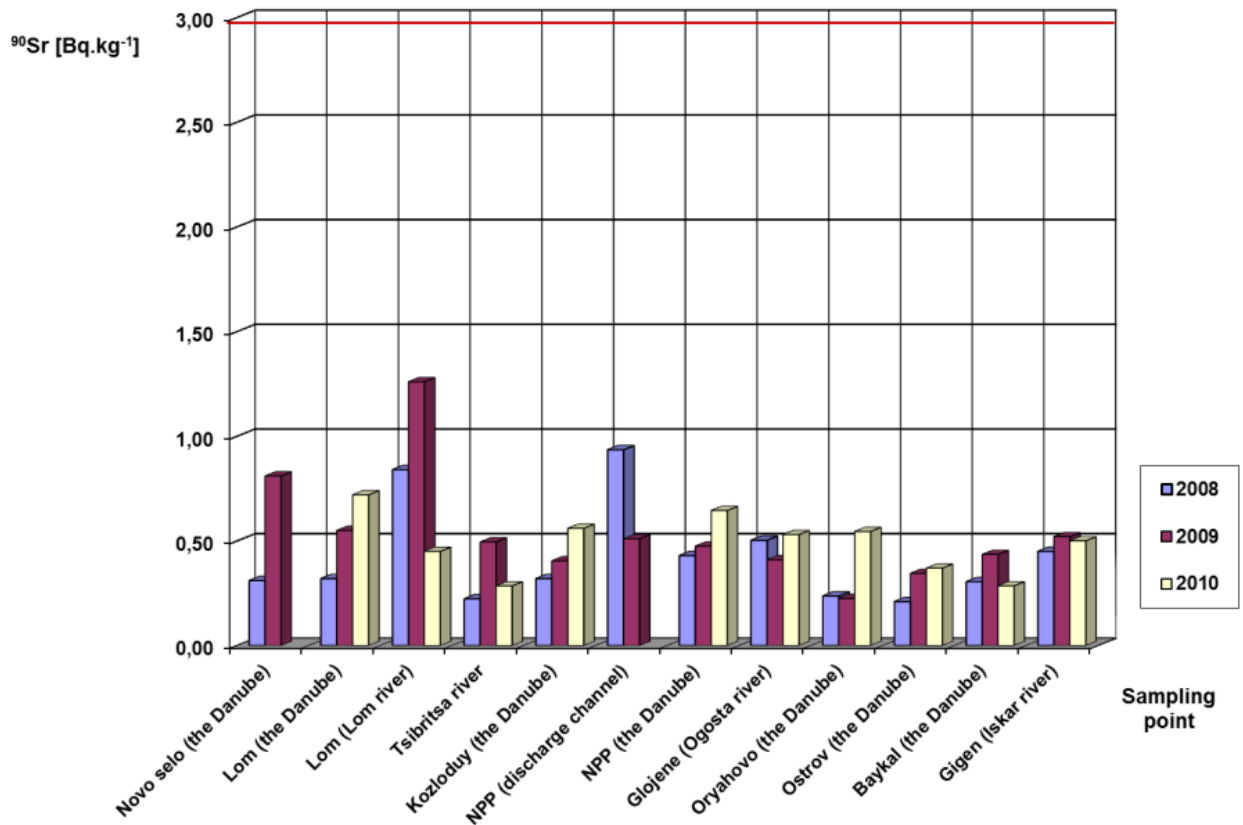


Figure 4. Annual content of ^{90}Sr in bottom sediments from the region of the Kozloduy NPP.

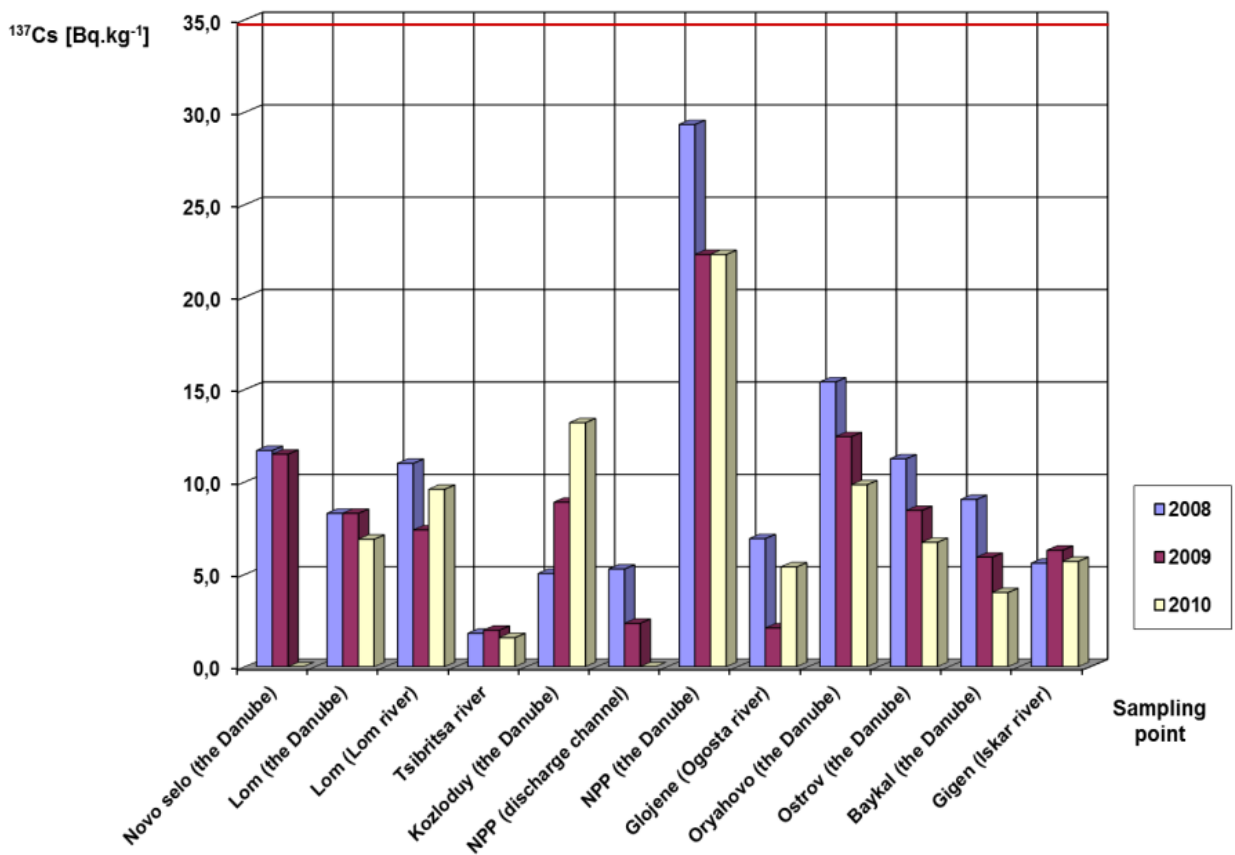


Figure 5. Annual content of ^{137}Cs in bottom sediments from the region of the Kozloduy NPP.

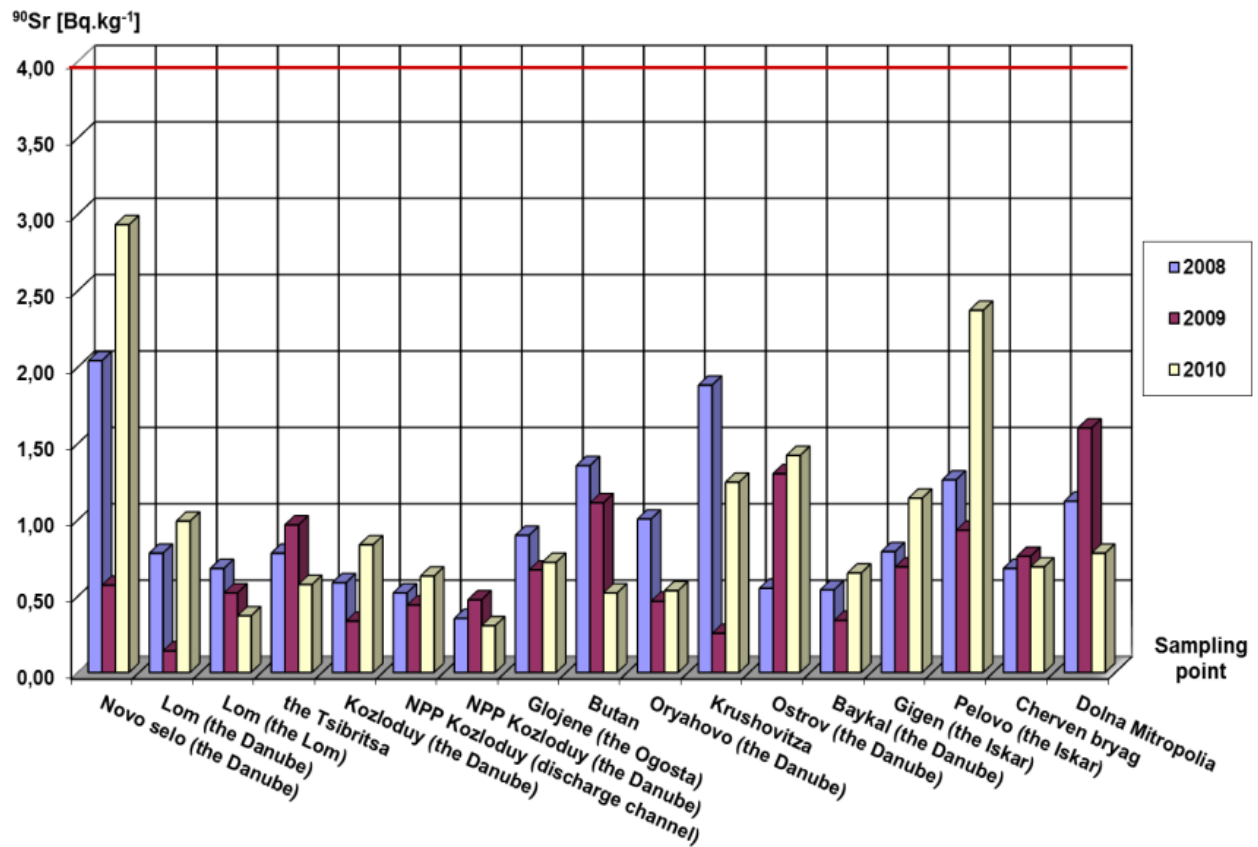


Figure 6. Annual content of ^{90}Sr in soils from the region of the Kozloduy NPP.

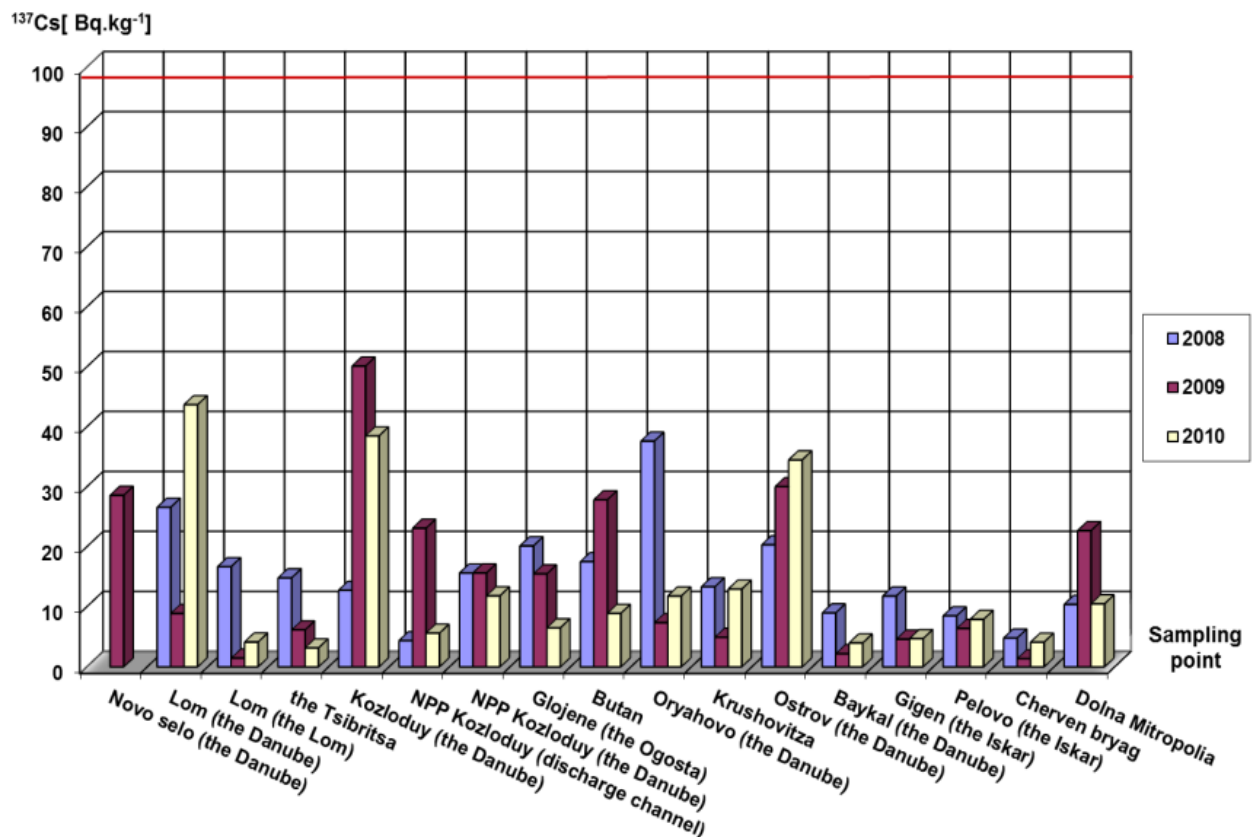


Figure 7. Annual content of ^{137}Cs in soils from the region of the Kozloduy NPP.

parison with ^{90}Sr the registered concentrations of ^{137}Cs in bottom sediments continue to remain higher as a result of the global fallout and the Chernobyl accident than the measured values in pre-operational period of the plant – $5.6 \pm 0.4 \text{ Bq/kg}^1$ for the internal rivers and $3.6 \pm 1.4 \text{ Bq/kg}^1$ for the Danube [1].

3.2 Soils

The annual content of ^{90}Sr in soils (Figure 6) from the region of NPP Kozloduy is between $0.14 \pm 0.06 \text{ Bq/kg}^1$ (Lom, the Danube, 2009) to $2.93 \pm 0.13 \text{ Bq/kg}^1$ (Novo selo, the Danube, 2010). The annual content of ^{137}Cs (Figure 7) varies in more widely ranges than ^{90}Sr – from $1.35 \pm 0.17 \text{ Bq/kg}^1$ (Cherven bryag, 2009) to $50.15 \pm 13.79 \text{ Bq/kg}^1$ (Kozloduy, the Danube, 2008).

In previous years [5-7](2003-2007) the annual content of ^{90}Sr in soils is in the ranges of $0.23 \pm 0.06 \text{ Bq/kg}^1$ (Baykal, the Danube, 2005) to $3.96 \pm 0.49 \text{ Bq/kg}^1$ (Novo selo, the Danube, 2004). The annual content of ^{137}Cs in soils also varies in more widely ranges from $2.09 \pm 0.27 \text{ Bq/kg}^1$ (Pelovo, the Iskar river, 2005) to $82.50 \pm 17.68 \text{ Bq/kg}^1$ (Novo selo, the Danube, 2004) [5,7].

The annual concentrations of ^{90}Sr in soils are in the range of values recorded in the previous periods [4-7] and are commensurable with the measured in pre-operational period of the plant – $5.0 \pm 0.4 \text{ Bq/kg}^1$ [1]. In comparison with ^{90}Sr the registered concentrations of ^{137}Cs in soils continue to remain higher as a result of the global fallout and the Chernobyl accident than the measured values in pre-operational period of the plant – $7.6 \pm 0.6 \text{ Bq/kg}^1$ [1].

4 Conclusions

- During the period 2008-2010 the obtained results for content of ^{90}Sr and ^{137}Cs in analyzed bottom sediments and soils are below the upper limit of the background content of these radionuclides in environmental objects.
- The results of the analysis of the content of ^{90}Sr and ^{137}Cs in soils in the area of the Kozloduy NPP show that there are no considerable quantitative changes registered, caused by radioactive gaseous discharges from the plant. They do not produce any noticeable changes of natural background [11], originating from atmospheric nuclear weapons tests and the Chernobyl accident. Therefore for the period 2008-2010 radioactive atmospheric gaseous discharges from the NPP did not influenced the concentration of radionuclides in objects from the terrestrial ecosystem.
- Do not register quantitative changes caused by liquid radioactive discharges from nuclear power plant over the content of ^{90}Sr and ^{137}Cs in the bottom sediments of the internal rivers and of the Danube in the period 2008–2010. The measured concentrations of ^{90}Sr and ^{137}Cs in bottom sediments attest to the lack of influence of liquid radioactive discharges

from the Kozloduy NPP on the content of ^{90}Sr and ^{137}Cs in objects from the aquatic ecosystem.

- Annual exposure of the critical group of members of the public resulting from liquid radioactive discharges, estimated as individual dose for screening purposes, does not exceed 10^{-6} Sv/a , therefore presenting little risk to the population even with conservative risk assessment estimates.

References

- [1] Petkov T., Zlatanava R., Kerteva A., Novakova E. (1976) Radioactivity of the environment in the region of the Kozloduy NPP in the pre-operational period. *Radiology and radiobiology*, **XV/1/** 51–54.
- [2] Vassilev G., Petkov T., Badulin V., Zlatanava R., Zotchev S. (1995) Main results of an environmental radioactivity monitoring in the region of the Kozloduy NPP throughout its operation (1974-1994): Report IAEA-SM-359/78P. International Symposium on Environmental Impact of Radioactive Releases, IAEA, Vienna, 1995.
- [3] Badulin V., Zlatanava R., Botzova L., et al. (1998) Radioactivity of environmental objects in the region of the Kozloduy NPP during 1997. Report IX Congress of the Bulgarian association of radiology, Sofia, 1998.
- [4] Totzeva R., Nyagolova N., Karaivanova R., Badulin V., et al. (2002) Content of strontium-90 in soils, grasses and sediments form 30 km area around the Nuclear Power Plant Kozloduy in 2001. International Symposium Ecology-2002, Bourgas, Electronic Journal, BG, 2002, pp. 71-75, ISBN 08619861.
- [5] Badulin V., Botzova L., Karaivanova R., Totzeva R., et al. (2003) Irradiation of the population in the area of the Kozloduy NPP during normal plant operation: 30 years monitoring, Man and Ionizing radiation. Jubilee Scientific Session on the Occasion of 40 years NCRRP 1963-2003, Sofia, November 2003.
- [6] Totzeva R., Kotova R. (2003) Content of ^{90}Sr in bottom sediments from the region of the Kozloduy NPP during the period 2002–2003, Man and ionizing radiation. Jubilee scientific Session on the occasion of 40 years NCRRP 1963-2003, Sofia, November 2003.
- [7] Kotova R., Badulin V., Karaivanova R., Totzeva R., et al. (2007) Monitoring of radioactivity levels in environmental objects from the region of Kozloduy NPP during 2001–2005, Ecology-2007, Bourgas, Electronic Journal, BG, 2007, pp. 190-205, Vol. 2, ISBN 978-954-9368-25-3.
- [8] Bryant F., Morgan A., Spicer G. (1959) The determination of radiostrontium in biological materials. Atomic Energy Research Establishment, AERE-R 3030, Harwell.
- [9] Angelova A., Totzeva R., Karaivanova R., et al. (1996) International intercalibration as a method of quality control of radiochemical analyzes. National Workshop with International Participation on Radioactivity in the Environment, Sofia, 1996.
- [10] Merešová J., Wätjen U., Altzitzoglou T. (2012) Evaluation of EC Interlaboratory Comparison on Radionuclides in Soil. JRC Scientific and Policy Reports, European Commission, 2012, ISBN 978-92-79-25111-5.
- [11] INTERNATIONAL ATOMIC ENERGY AGENCY (2000) Safety glossary, IAEA, Vienna, 2000.