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Technogenic radionuclides in undisturbed Bulgarian soils

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ABSTRACT

This work presents data for the activity concentration of artificial radionuclides in soils from different regions in Bulgaria for the period 1985–2011. The subjects of the study were undisturbed soils from mountainous, hilly, and plain areas. Special attention has been paid to the contamination with the long-lived technogenic radionuclides cesium-137, strontium-90, plutonium-238, and plutonium-239 + 240. ^{137}Cs was measured by low level gamma spectrometry and ^{90}Sr by radiochemical separation and low level beta counting. The isotopes of plutonium were measured by alpha-spectrometry after a radiochemical procedure of purification and concentration.

Cesium-137 and strontium-90 were the main technogenic radionuclides detected in the examined Bulgarian soils few years after the Chernobyl NPP accident. Their content in the soils from high mountain areas is several times higher than that in the soils from the plane areas in the northern part of the country. High heterogeneity in the pollution has been observed. Mean soil activity concentration levels of up to 250 Bq kg^{-1} for ^{137}Cs and 14 Bq kg^{-1} for ^{90}Sr were found in different years. The maximal values registered in separate soil samples were about 1300 Bq kg^{-1} (^{137}Cs) and 24 Bq kg^{-1} (^{90}Sr). The activity concentration levels for $^{239+240}$Pu varied between 0.1 and 3.7 Bq kg^{-1}. The results indicate that the radioactive pollution of the Bulgarian soils with artificial radionuclides is a result of global fallout and Chernobyl nuclear power plant accident.

1. Introduction

Technogenic radioactivity is due to radioactive materials which have been produced and released into the environment by human nuclear activity, including nuclear weapon testing, the operation of nuclear power plants, research reactors, and nuclear fuel reprocessing. Nuclear test explosions were carried out at a number of sites, mostly located in the northern hemisphere, between 1945 and 1980. 502 atmospheric tests with a total fission and fusion yield of 440 Mt were conducted. The total explosive yield of underground tests is estimated to be 90 Mt (UNSCEAR, 2008).

Nuclear accidents, such as the Chernobyl accident, have also released large amounts of radionuclides into the environment. The accident in the 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 and about 8 PBq of ^{90}Sr were released from the accident (UNSCEAR, 2008). The radionuclides with long half-life like ^{137}Cs, ^{90}Sr, ^{238}Pu, and $^{239+240}$Pu deposited after the accident will remain in the environment, mainly in the soil, for decades and will be relevant for environmental monitoring. Analysis of radionuclide content in soil, plants, and 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, 2000).

Bulgaria, South Bulgaria in particular, was among the European countries with relatively high contamination as a result of the Chernobyl accident. According to UNSCEAR (1988, Annex D, Table 11), the deposition of ^{137}Cs on the southern part of Bulgaria is 12 kBq m^{-2}, which is significantly higher than the deposition for countries like France (max 3.2 kBq m^{-2}), Belgium (max 0.84 kBq m^{-2}), Poland (max 5.2 kBq m^{-2}), Hungary (4.8 Bq m^{-2}) and Greece (8 kBq m^{-2}). In the same report (Annex I, Fig. VI) Southern Bulgaria is indicated as a territory with ^{137}Cs deposition density higher than 5 kBq m^{-2}, while for a large part of the European territory it is between 1 and 5 kBq m^{-2}. The activity concentration of man-made radionuclides in the Bulgarian soils increased considerably (Kinova, 1997; Pourchet et al., 1997; Tsvetkov et al., 2006; Zhiyanski et al., 2008). Up until now, 25 years later, ^{137}Cs and ^{90}Sr are still detected in all soil samples and represent a potential danger for the contamination of the plant production through root feeding. Our work aims the assessment of mean local background values for ^{137}Cs and ^{90}Sr in soil on the base of regular radiological monitoring of undisturbed soils from high mountainous, hilly, and plain areas in Bulgaria. The areas around Kozloduy NPP are of special interest. Such data can be used as a reference level from which we can detect eventual future release of radioactive nuclides.

2. Materials and methods

The sampling strategy chosen for the purposes of the radiological characterization of the soils in Bulgaria surveying the impact of NPP Kozloduy and the evolution of the territory in general corresponds to the orientated systematic strategy described in ISO, 18589-2 (2007).

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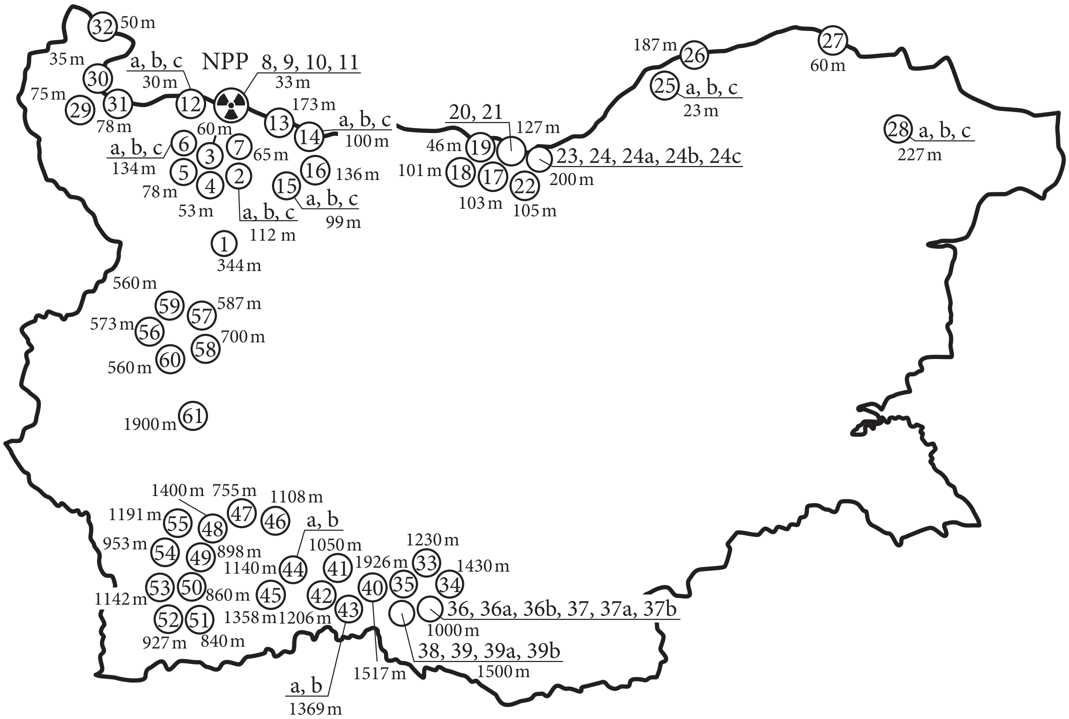

Fig. 1. Map of sampling site locations with site altitude above sea level.

Samples of undisturbed soils are collected annually from one and the same sampling site. Sampling areas were specified considering wind strength and direction, topography and difference in altitude. In this way regions with higher probability for contamination in case of accident in NPP Kozloduy or cross-border transfer of radionuclides were included. The location map of the area under study and sampling site

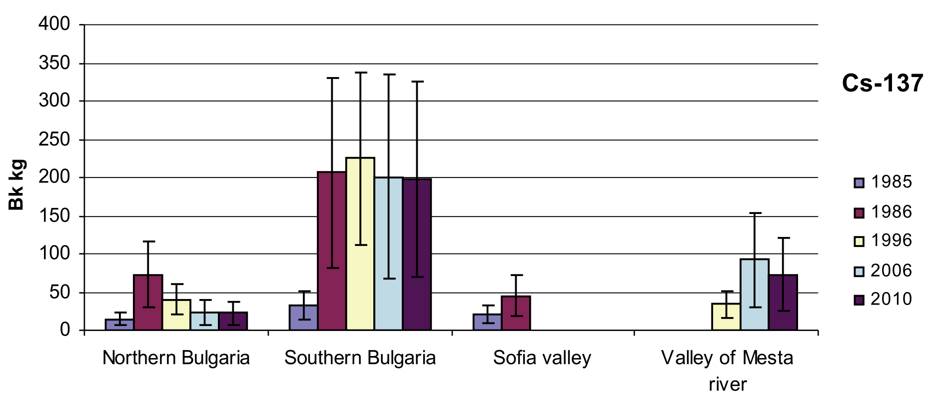

Fig. 2. Averaged values for ^{137}Cs activity in soil samples from different regions [Bq kg^{-1}] (1985–2010 calculated to reference date 1986).

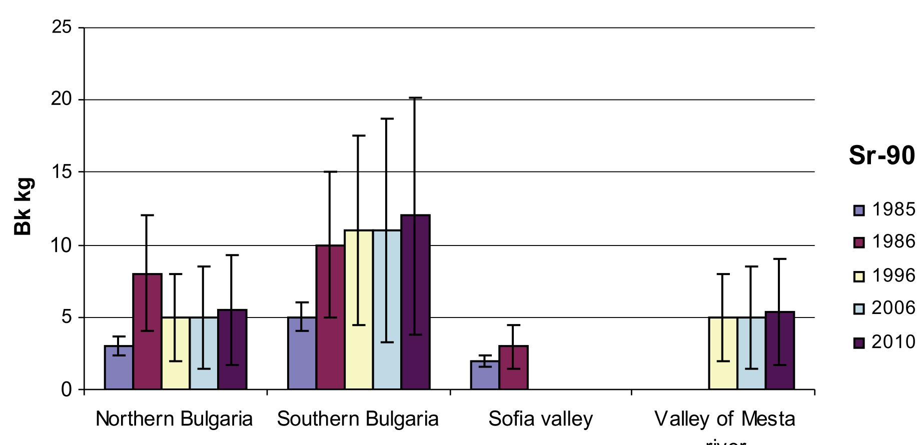

Fig. 3. Averaged values for ^{90}Sr activity in soil samples from different regions [Bq kg^{-1}] (1985–2010 calculated to reference date 1986).