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RADIOISOTOPES OF CHERNOBYL FALLOUTS — RADIATION CONDITION OF SOILS AND POTENTIAL RISKS GROUNDWATER QUALITY ESTIMATION

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Abstract. The results of the behavior of ^{137}Cs and ^{90}Sr of Chernobyl fallouts in different types of soils distributed over a wide territory in the contaminated Belarusian areas including 30 km zone around the Chernobyl are presented in the paper. The advective-dispersion equation was applied to quantify rates of vertical migration of radioisotopes through the soil profile and to predict the time of maximum (peak) activity transport of the radioisotopes in groundwater. Monitoring data of ^{137}Cs and ^{90}Sr-activity in groundwater and deeper aquifer horizons (Quaternary, Paleogene and Upper Cretaceous) allowed to estimate their contemporary contamination in comparison with drinking water standard in Belarus and background activity before accident. It was established that ^{137}Cs and ^{90}Sr- activity in groundwater remains high, especially if the non-threshold hypothesis of dose-effect relationship in the context of impact of low levels radiation on the population is taken into account.

Key words: caesium-137, strontium-90, migration in soils, groundwater, aquifer horizons.

Abstract. W pracy przedstawiono wyniki zachowania się ^{137}Cs i ^{90}Sr z opadu Czarnobyla w glebach rozprzestrzenionych na szerokim obszarze skontaminowanej powierzchni Białorusi, wraz z 30-kilometrową strefą wokół Czarnobyla. Do oceny tempa pionowej migracji izotopów radioaktywnych w profilu glebowym i do przewidywania czasu maksimum aktywności transportu izotopów radioaktywnych w wodach podziemnych zastosowano równanie adwektywno-dyspersyjne. Dane z monitoringu aktywności ^{137}Cs i ^{90}Sr w wodach podziemnych i głębszych horyzontach wodnych (czwartorzęd, paleogen i górna kreda) pozwoliły na ocenę ich współczesnej kontaminacji w odniesieniu do standardu wód pitnych na Białorusi i aktywności tła przed katastrofą. Ustalono, że aktywność ^{137}Cs i ^{90}Sr w wodach podziemnych pozostaje wysoka, zwłaszcza gdy bierze się pod uwagę bezprogową hipotezę związku dawka – efekt w kontekście oddziaływania niskiego stopnia radiacji na społeczeństwo.

S³owa kluczowe: cez-137, stront-90, migracja w glebach, wody podziemne, poziomy wodne.

INTRODUCTION

Data of ^{137}Cs and ^{90}Sr-activity depth profiles distribution in mineral and organic soils of an aeration zone and the groundwater contamination have been obtained by the authors during the monitoring in the period from 1986 to 2002 in the eastern and the southeastern areas of Belarus (Gomel and Mogilev regions, Figs.1 and 2). Contamination levels in the investigated soils (types of these soils and their spread in the areas are shown in Figure 3) by ^{137}Cs had varied from 7 to 17560 kBq m^{-2} and ^{90}Sr from 22.2 to 1085 kBq m^{-2} in these areas.

SAMPLING

The soil samples were taken from the surface up to the groundwater level, typically within intervals of 0–1 cm before 10 cm depth and 0–5 cm further down to groundwater level. Contamination levels were determined by summarizing activ-

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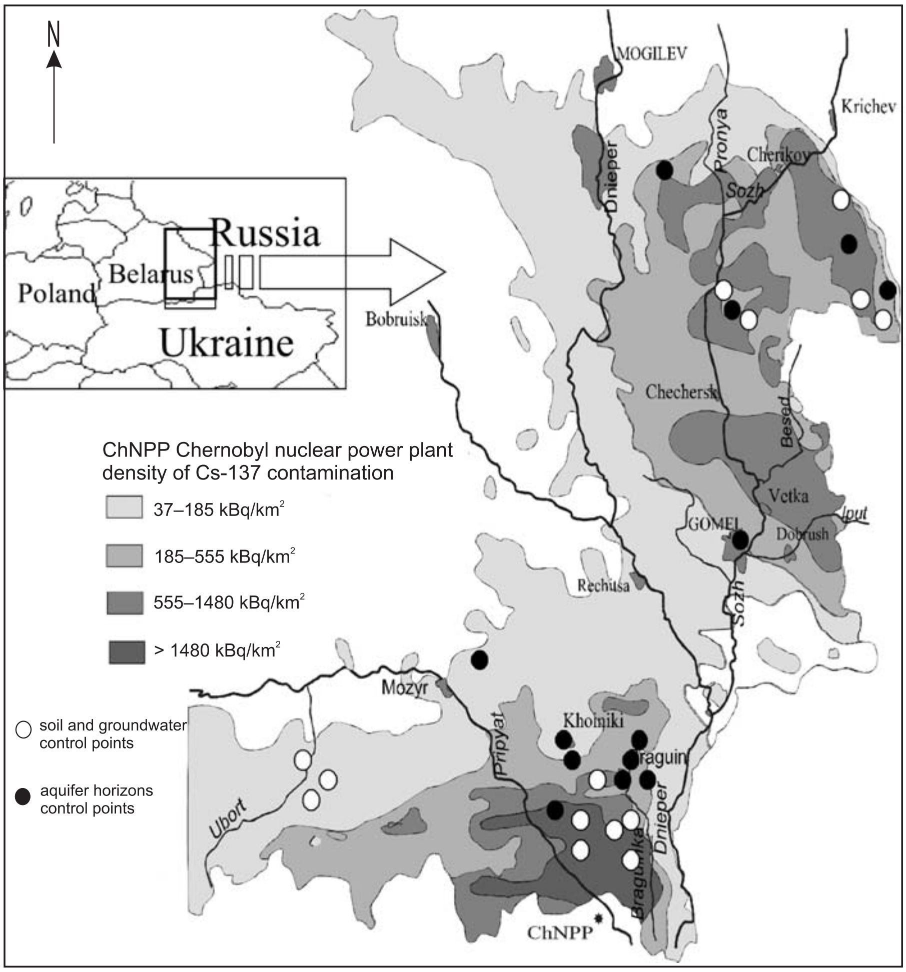

Fig. 1. The map-scheme of caesium-137 contamination of the southeastern part of Belarus and control points

ity of radioisotopes within the profile. Sampling of groundwater was carried out directly from the soil holes. As a rule, groundwater was found at the depth from 35 cm to 3 m. Aquifer horizons such as Quaternary, Paleogene and Upper Cretaceous were studied by sampling waters from hydrogeological boreholes, which are used for water supply systems as well.

METHODOLOGY

Generally in the mathematical modelling, the vertical migration of radioisotopes with concentration, C (Bq cm^{-3}), within a soil profile is characterized by the advection-dispersion equation:

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} - \lambda C \qquad [1]$$

where:

v (cm year^{-1}) – effective velocity,

D (cm^2 year^{-1}) – dispersion coefficient to describe these two processes — dispersion and diffusion of the profile,

λ – decay constant of the radioisotope,

t – time since deposition of the radioisotope to the soil,

x – depth of soil.

On the basis of our empirical data distribution of ^{137}Cs and ^{90}Sr-activity along soil profiles, derived due to the solution of equation [1] can be approximated by a Gaussian profile (Kudelsky *et al*., 1996) for an impermeable upper boundary (i.e. dispersion can take place only downwards from soil surface):

$$C = \frac{M}{2\sqrt{\pi D t}}\left\{\exp\left(-\frac{(x - vt)^2}{4Dt}\right) + \exp\left(-\frac{(x + vt)^2}{4Dt}\right)\right\} \qquad [2]$$

where:

t – time since fallout began,

M (Bq) – amount of ^{137}Cs or ^{90}Sr present,

x – depth.

It was assumed that the radioisotopes are deposited at time $t=0$ in the plane $x=0$ (an "instantaneous plane source"), and that the source is translated a distance vt from the surface after time t.

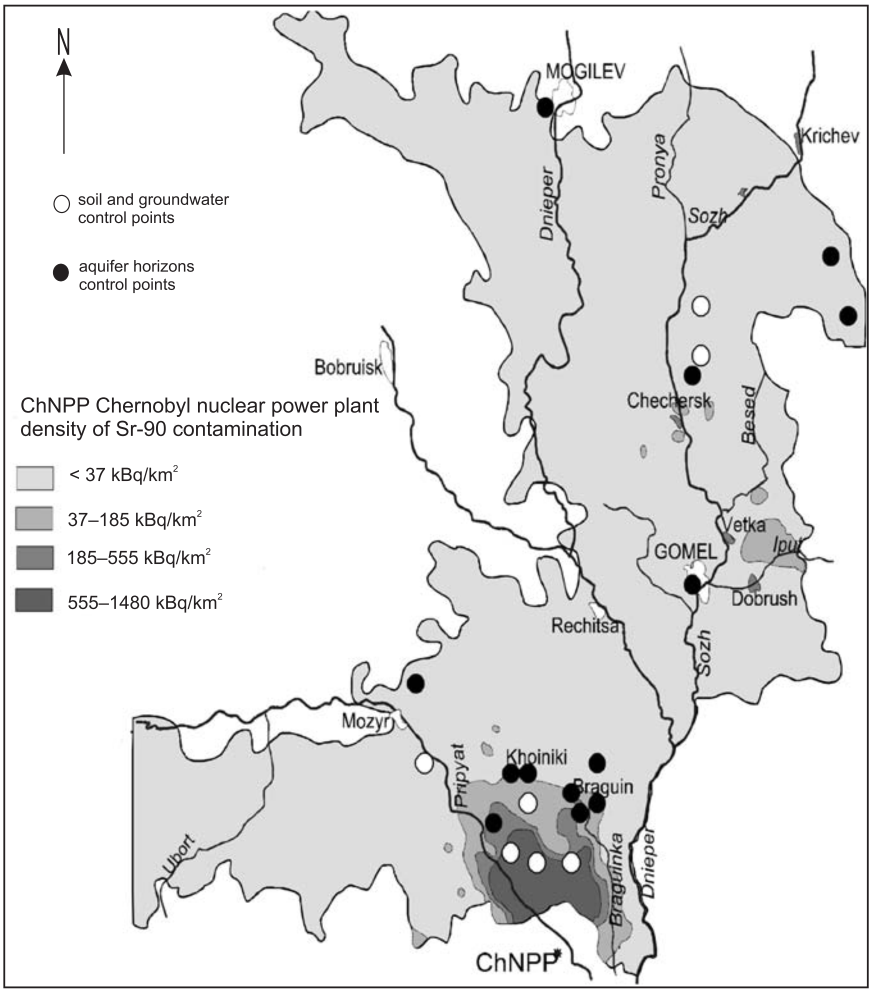

Fig. 2. The map-scheme of strontium-90 contamination of the southeastern part of Belarus and control points

Radioactive decay affects only the amount of activity present. It has no influence onto the transport (migration) parameters. Due to this reason, a decay were ignored in the solution of equation [1]. The decay of radioisotopes, however, should be taken into account for practical purposes.

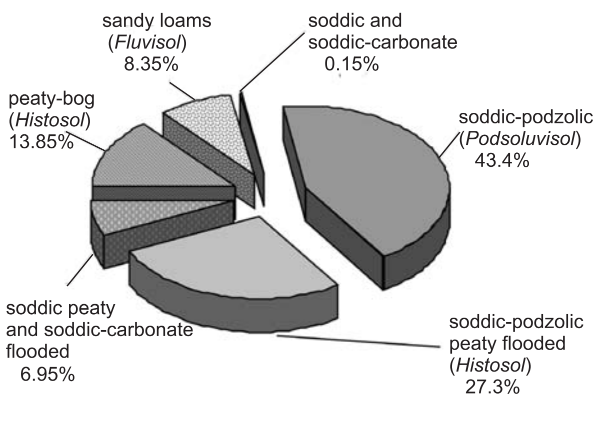

Fig. 3. The different sorts of mineral and organic soils widespread in the areas

The Half-Width at Half-Maximum (HWHM) simple method (Smith, 1999) was applied for the assessment of transport parameters (see: Figure 4).

The parameter values v and D were can be evaluated from following formulas:

[3]

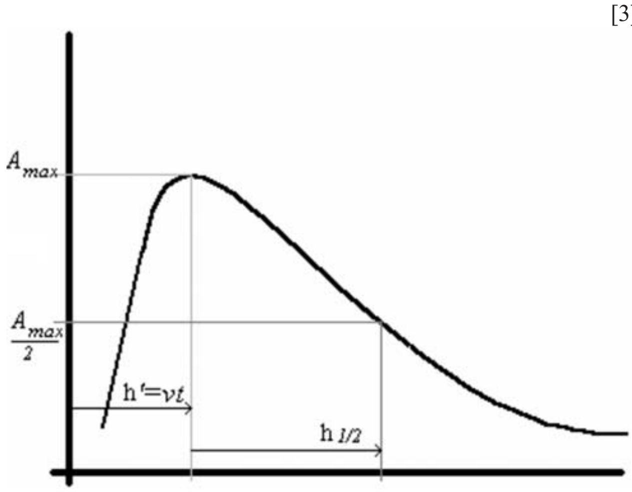

Fig. 4. Graphical illustration of (HWHM) method

$$D = \frac{h_{1/2}^2}{4t \ln 2}$$

$$v = \frac{h'}{t} \qquad [4]$$

where:

$h_{1/2}$ – distance between the peak in activity and the depth at which the activity concentration falls to half of its maximum value. The advection velocity is calculated from the depth, $h^{'}$, of the maximum of the activity profile, as shown in Figure 4.

The solution (2) can be used to predict the time of maximum (peak) activity transport of the radioisotopes into groundwater by putting in the depth of occurrence of the groundwater instead of x coordinate – the depth of soil interval. Thus, we also can easily predict the ^{137}Cs and ^{90}Sr- activity at the depth of occurrence of groundwater in any time after accident.

RESULTS

Using the obtained information, the following results may be shown:

1. During 16 years after the Chernobyl accident 95–98 % of ^{137}Cs inventory retains in the upper 0–5 cm layers or rarely within 0–20 cm independently of ^{137}Cs fallouts density. The vast majority of ^{90}Sr-activity (95–98%) is observed within the layer 0–15 cm or 0–25 cm (Krasnoselie, the sand dune).

2. The linear migration rate (v) and diffusion coefficient (D) of ^{137}Cs vary in wide limits. They are: 0.11–2.66 cm year^{-1} and 0.01–1.4 cm^2 year^{-1}, respectively. The migration parameters of ^{90}Sr-activity depth profiles are: v (0.14–7.14 cm year^{-1}) and D (0.01–19 cm^2 year^{-1}).

In the unsaturated mineral soils classified as soddic-podzolic (*Podzoluvisol*[3]) located on floodlands the significant statistical tendency of decreasing migration parameters of ^{137}Cs over time had been observed (R^2 = 0.58–0.77), that can be explained as an irreversible sorption (fixation) of ^{137}Cs by solid phase of soils and the radioisotope diffusion into interplanar spaces of clay minerals (Comans *et al*., 1990; Hilton, 1994). This tendency takes place in other types of soils such as peaty and sandy loams (*Histosol and Fluvisol*), see Figure 5.

Other tendency has been noticed for the soddic-podzolic sand of saturated soils of lake catchments. In this case, the increasing migration parameters of ^{137}Cs over time have been determined as well. The migration occurred possibly due to the intensive washing out of these soils by atmospheric precipitation and as a result of a combination of two different processes, as – convection and diffusion transfer.

When comparing ^{137}Cs behavior against that of ^{90}Sr in the unsaturated mineral soddic-podzolic (*Podzoluvisol*) soils, the increasing migration parameters of ^{90}Sr over time (R^2 = 0.7–0.9) can be observed. The migration parameters in soddic-alluvial sandy loam soils (*Fluvisol*) of flood-lands are already increasing over time. At the same time, migration parameters of ^{90}Sr in some saturated peaty-bog soils (*Histosol*) decrease or remain relatively permanent. On the contrary to ^{137}Cs, ^{90}Sr remains fixed in soils as a result of interaction with the organic matter, rather than with the mineral.

It should be noticed that the solution advective-dispersion equation was based on *a priori* assumption that convection and diffusion rates of migration of radioisotopes in soil profile are constant over time. According to the obtained results, however, this fact is not proved. The better possible decision is to use an average value of migration parameters for studied period *in situ* or an empirical functional dependence (for example as in the case illustrated in Fig. 5) to predict ^{137}Cs and ^{90}Sr distribution along soil profile at any time in the future.

The equations for calculation of average values of migration parameters are shown below:

$$D_{average} = \sum_{1}^{n} D_i t_i \Big/ \sum_{1}^{n} t_i \qquad [5]$$

$$D_{average} = \sum_{1}^{n} v_i t_i \Big/ \sum_{1}^{n} t_i \qquad [6]$$

where:

D_i , v_i – dispersion coefficient and advective rate during duration t_i;

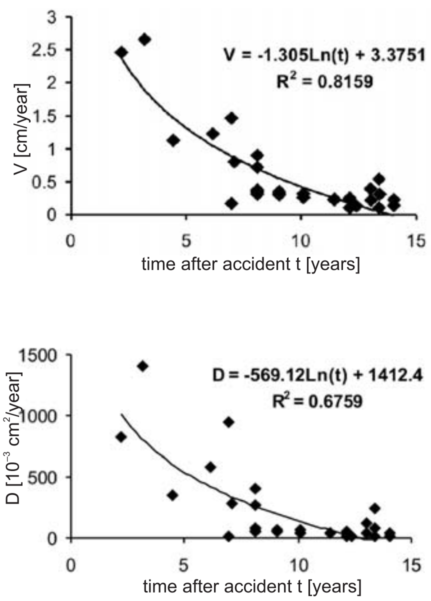

Fig. 5. Decreasing migration parameters of ^{137}Cs over time after Chernobyl accident

[3] *Podzoluvisol, Fluvisol, Histosol – FAO UNESCO classification of soils*

n – number of duration from which summarizing of the common migration time of radioisotope.

3. The obtained prediction of the time of the maximum (peak) activity transport of ^{137}Cs and ^{90}Sr into groundwater level are: this time varies from 30 to > 1,000 years for ^{90}Sr (more than period half-life = 28 year) and for ^{137}Cs — from 62 to > 1,000 years (more than period half-life = 32 year). The evaluation of the radioisotopes long-term transfer from soil to groundwater is demonstrated by the example of ^{90}Sr-activity depth profiles distribution located in 30 km zone on sand dune (see Fig. 6). Here the highest values of migration parameters ($v = 7.14$ cm year^{-1} and $D = 19$ cm^2 year^{-1}) were established and a groundwater was revealed on the depth of about 35 cm.

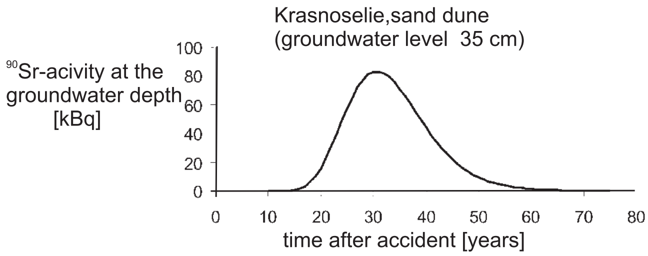

Fig. 6. The prediction the time of maximum (peak) activity transport in groundwater

4. According to the data from 2002 year, these values of contemporary contamination of groundwater by ^{137}Cs in the South-eastern areas of Belarus (0.02–0.58 Bq l^{-1}) are relatively low even in the areas with a high density of Chernobyl fallouts. The ^{137}Cs migration via soil depth profiles is generally controlled by the content of fraction of clay minerals (in particular, the illite clay particles). The clays can fix ^{137}Cs, that is the reason of significant reduction in mobility of this isotope in the soils. The ^{90}Sr activity in groundwater varies from 0.012 to 1.205 Bq l^{-1} (Radin) and 2.226 Bq l^{-1} (Kulagin). According to the data from the period 1991–2001, the strong correlation dependence ($R^2 = 0.78$) between ^{90}Sr-activity in the groundwater and an inventory of this radioisotope in the soil profile has been established.

The ^{137}Cs contamination of the aquifer horizons limits 0.095–0.98 Bq l^{-1} at the depth of 40–98 m (the Paleogene aquifer horizon) and from 0.035 to 0.445 Bq l^{-1} at the depth of 40–100 m (the Upper Cretaceous aquifer horizon). The ^{90}Sr contamination of aquifer horizons varies from 0.028 to 1.783 Bq l^{-1} at the depth of 4–6 m (Quaternary) to 0.095–0.482 Bq l^{-1} at the depth 40–98 m (the Paleogene aquifer horizon) and 0.002–0.048 Bq l^{-1} at the depth 40–100 m (the Upper Cretaceous aquifer horizon).

The Maximum Permissible Concentration (MPC) according to the drinking-water standard in Belarus is: for $^{137}Cs = 10$ Bq l^{-1} and for $^{90}Sr = 0.37$ Bq l^{-1} but the radioactive contamination in the waters of the river Pripyat before the Chernobyl accident oscillated from 0.003 to 0.018 Bq l^{-1} for ^{90}Sr and from 0.006 to 0.066 Bq l^{-1} for ^{137}Cs.

The latter values result from nuclear weapons testing in the atmosphere. The contemporary ^{137}Cs and ^{90}Sr contamination in the groundwater corresponds to the intervals of 0.02–0.58 and 0.012–2.206 Bq l^{-1} at the depth above 2 m and in the aquifer horizons (to 0.445–0.98 Bq l^{-1} and 0.482–1.783 Bq l^{-1} at the depth above 100 m). It can be noticed that contemporary levels are quite high, especially if the non-threshold hypothesis of dose-effect relationship in the context of impact of low levels radiation on the population is taken into consideration.

CONCLUSIONS

In accordance with the results of modelling of the ^{137}Cs and ^{90}Sr migration in depth of the soil profiles, a potential hazard of an appearance of the extreme activity in the groundwater is not observed in the most investigated cases.

In conclusion, it should be noticed that the changes in vertical migration parameters in time of ^{137}Cs directly depend on water-physical conditions of soils and the specific sorption process of the radioisotope. These facts should be taken into account in a long-term prediction of the radiation condition of the soils and the groundwater in the contaminated areas after the Chernobyl accident.

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