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SIMULATION OF SELF-PURIFICATION PROCESSES OF WATER MASSES FROM RADIOACTIVE SUBSTANCES

The rate of self-decontamination of water masses from radionuclides in the most contaminated water bodies in Ukraine is determined. It is shown that the content of ^{137}Cs in the water masses of the exclusion zone water bodies is reduced with halftime of decontamination of 9,6±1,4 years. The mechanism of slowing the rate of decontamination of water masses from radionuclides is considered. It is shown that two years after the radionuclide ingress in the water body there is a significant slowdown in the rate of purification of water masses.

Keywords: reservoirs, radionuclides, speed of purification, halftime of purification.

Prognostic assessment of water quality in the situation of anthropogenic impact, particularly when artificial radionuclides or radionuclides from natural but technologically amplified sources enter water bodies, is one of the most important tasks of protection of the environment. Contaminants can enter water bodies with aerosol deposition, come as a result of runoff from the area of water-collection or from waste waters of enterprises.

Currently, the most part of models describing the spread of contaminants in the water masses are based on the assumption that the change in the concentration of contaminants per time unit is proportional to their concentration and hydrodynamic flow characteristics [1-5]. In many cases it is impossible to consider all the factors influencing the concentration of contaminants in the water masses, so researchers use generalizations and simplifications to describe the processes when modeling.

One of the good examples of simplifying the description of hydrodynamic processes is the model of Y.A Egorov and S.V Kazakov [6-8], which predicts the dynamics of radioactive substances in water, sediment and biota. In this model, like in the most of others, the parameter describing the self-decontamination of water masses from radionuclides is constant. It is clear, that this value can be used for most freshwater ponds. However, the rate of decontamination from ^{137}Cs of the water masses of the Kiev reservoir in the period of 1987-1995, calculated for this model [9], was much smaller than the indicated by the authors of the model. Also, there is evidence that over time the slowing of self- decontamination of water masses from radionuclides was observed in the water bodies of the East Ural radioactive trace [10]. Therefore, the aim of our work was not only to determine the rate of self-decontamination of water masses from radionuclides but also to find the mechanisms of its deceleration.

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Methodological approaches to determination of the rate of self-decontamination of water masses from radionuclides. In the event when we can neglect the hydrological processes, including diluting of water masses in reservoirs with more clean waters of the tributaries, self- decontamination of water masses from the radio-nuclides is described by the following formula:

$$A(t) = A(0)\exp\left(\frac{-t \cdot \ln 2}{T_{1/2}}\right), \qquad (1)$$

where $A(t)$ — Activity of the radionuclide in the water masses at a moment of time t, Bq; $A(0)$ — the initial activity of the radionuclide in the water masses, Bq; t — Time of radionuclide presence in the water masses, days; $T_{1/2}$ — halftime of decontamination of the water masses from the radionuclide, days.

For watercourses and reservoirs formula (1) can be interpreted as follows: $A(0)$ and $A(t)$ - activity of radionuclides passing through the upper and lower section (on the inlet and outlet of a reservoir), t - time of the water masses from the top section to the bottom cross-section. Then the formula (1) takes the form

$$A(t) = A(0)\exp\left(\frac{-\ln 2}{K \cdot T_{1/2}}\right), \qquad (2)$$

where $A(0)$, $A(t)$ - Activity of radionuclides of the inflow and outflow; K=365,25/t - rate of water exchange, year^{-1}.

It is noted [7] that the calculation of the rate of self-decontamination from radionuclides of water masses in NPP cooling ponds, made by the time series of concentration of radionuclides in water and by water flow, is characterized by low accuracy due to significant variability in the magnitude of concentration. The calculation using the formula (2), which takes into account the total activity of the radionuclide averaged over a certain period of time will be more accurate.

Calculation of the parameters of self-decontamination of water masses in Dnipro reservoirs. Using data on the balance of ^{90}Sr and ^{137}Cs in the Kiev reservoir and in the whole of the Dnieper cascade, we determined the rate of self-decontamination of water masses from these radionuclides. Analysis of the data [11, 12] shows that for the period of 1987-1995 about 8-33% of ^{90}Sr that entered the Kiev reservoir were retained, which corresponds to the halftime of decontamination from 82 to 508, with an average value of 188±145 a day. In the downstream reservoirs (from Kanev to Kahovsk) about 21-67% of coming ^{90}Sr were retained, which corresponds to the halftime of decontamination 830±290 a day, i.e. self-decontamination of water masses of these reservoirs from ^{90}Sr is much slower than the Kiev one.

Thus, in the process of decontamination of water masses from ^{90}Sr two components were observed. Iterative methods found that the partial contribution of the fast component of purification of water masses from ^{90}Sr is 36%, partial halftime of decontamination - 50 days; for the second, slower components - 64% and 1200 days, respectively.

Water content of rivers is changed over the years, and accordingly the water exchange rate (i.e. the time of passage of water masses between the cross-sections) is changed. In the Kiev reservoir, depending on the dryness of the year, the water exchange rate is 6,7-15,6 year^{-1} [13]. Calculations show that with the decrease of water exchange in the reservoir the proportion of ^{90}Sr is increased, taken from the water masses with a “quick” component, and therefore, the overall share of this radionuclide removed from the water mass is going up, which leads to decrease in formal halftime of decontamination of water masses calculated in the one-component approach with formula (2).

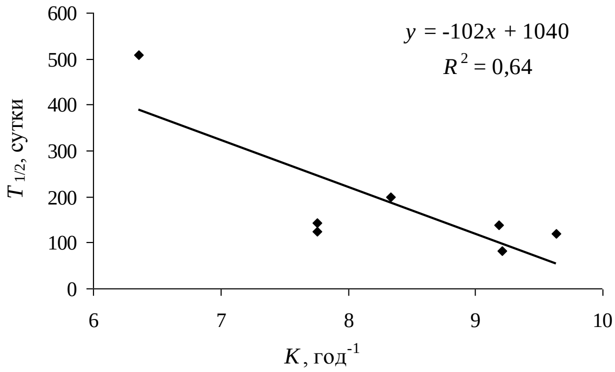

Fig. 1. Dependence of the rate of water exchange (K) halftime of decontamination ($T_{1/2}$) of water masses from ^{90}Sr (Kiev reservoir, 1987-1993.);
R - correlation coefficient

Dependence of halftime of decontamination of water masses from the water exchange rate is described best of all by a linear function (Fig. 1).

Earlier, on the basis of theoretical calculations, we showed [14] that the calculations in the one-component approach, carried out by the formulae (2) in the case of multi-component processes will give the values of half-reduction which are significantly different from the partial periods. Thus, the field data are in good agreement with theoretical calculations.

Note that ^{90}Sr migrates with the water masses mainly in the dissolved form, and these data refer to the dissolved form of the radionuclide.

In contrast with strontium, cesium isotopes migrate in the water masses in the dissolved and adsorbed (on suspended matter) forms in roughly equal proportions. In the Kiev reservoir halftime of decontamination of water masses from ^{137}Cs absorbed on suspensions is 27±5 days, and from the dissolved one - 111±39 days. During the passage of the water masses through the Kiev reservoir 53-85% is precipitated into the bottom sediments from hard flows ^{137}Cs and 12-33% of the dissolved form of this radio-nuclide. Average halftime of decontamination of water masses of the Kiev reservoir from ^{137}Cs is 60±14 days. In the range of water exchange characteristic to the Kiev reservoir it was not found any interconnections between the rate of water masses purification from ^{137}Cs and water exchange rate (Fig. 2 shows the linear regression formula).

Calculation of the rate of self-decontamination of contaminated water bodies of the exclusion zone. For water reservoirs, we calculated the rate of decontamination of water masses from radionuclides on the basis of the balance of the radionuclides in the ecosystem. For the closed-type bodies the rate of water self-decontamination of water masses can be estimated by the long-term trends of the average activity of radionuclides in water

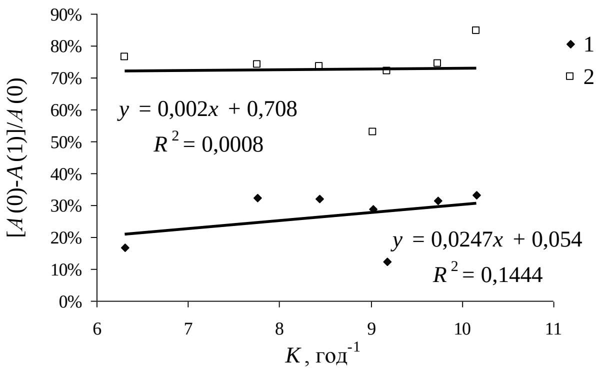

Fig. 2. Dependence of the fraction of ^{137}Cs, accumulated in the reservoir on the water exchange rate, ^{137}Cs the dissolved form (1) and ^{137}Cs in the form adsorbed on the suspensions (2) (Kiev reservoir, 1987-1993); A (0) - inflow of the radionuclide in the reservoir; A (1) – outflow of radionuclide from the reservoir; R - correlation coefficient

The halftime of decontamination of water masses from radionuclides for most polluted bodies of water exclusion zone were calculated using the data of GSNPP "Ecocenter" [15-17] for the period 1997-2012 years. The content of ^{137}Cs in the water mass was decreased with halftime of decontamination 9,6±1,4 year (weighted average 9,1±0,8 years) (Table. 1). About the same decrease rate was obtained for the content of ^{90}Sr in the water masses of lake Azbuchin and the Yanovsky backwater; in other reservoirs significant decrease in the concentration of ^{90}Sr in the water masses were not registered. Effective halftime of decontamination of water masses from radionuclides, taking into account the radioactive decay of isotopes, will be approximately 25% less

Simulation of self-decontamination of water masses in the event of single entry of radionuclides in the reservoir. The above estimate of the rate of self-decontamination of radionuclides from the Dnipro reservoirs and ponds of exclusion zone is based on the evidence about the long-term maintenance of ^{90}Sr and ^{137}Cs in the water masses. Now, based on the theoretical assumptions, we can define the rate of self-decontamination of water masses in the case of a single entry of radionuclides in the water body. In the calculations we made the following assumptions: 1) there are no chronic entry and discharge of radionuclides in the water body; 2) self-decontamination of water masses occurs only due to the sorption of radionuclides by the top "active" layer of silt and suspensions; 3) the rate of sedimentation and accumulation factors ($K_н$), i.e. the ratio of specific activity of radionuclides in sediments to its concentration in the water, is constant. The accepted assumptions allow us to make analytical calculations in the integral form. Radioactive decay and steady flow can be described by the following expression:

$$p_{эф} = p_0 + p_ф + p_с\,,$$

where $p_{эф}$ — effective speed of self-decontamination, s^{-1}; p_0 — the rate of self-decontamination of water masses from the radio-nuclide sorption due to suspensions and "active" layer of sediment, s^{-1}; $p_ф$ — speed of physical decay of the radionuclide, c^{-1}; $p_с$ — outflow rate of the radionuclide, s^{-1}.

The effective rate of self-decontamination is associated with the effective halftime of decontamination ($T_{эф}$) by the following expression:

$$T_{эф} = \ln 2 / p_{эф}\ .$$

Table 1. halftime of self-decontamination of water masses ($T_{1/2}$) of radionuclides of some reservoirs of the exclusion zone, years

Water reservoir	^{90}Sr	^{137}Cs
Lake Azbutchin	9,1±1,1	8,8±0,9
ChNPP cooling pond	—*	8,9±0,5
Yanivsky backwater	8,6±0,6	11,9±2,2
Lake Dalekoye	—	9,7±3,4
Lake Glubokoye	—	8,5±0,4

* «—» — value not defined

The model parameters were taken both from literature and the data obtained in the Kiev reservoir: 1) the area of silt accumulation is 0.5 of the water surface, average speed of silt accumulation is 0.8 cm / year, which is equivalent to 7 kg / m^2 with natural moisture or 1.4 kg / m^2 of dry weight per year; 2) for the sand the accumulation factor for ^{90}Sr ratio $K_н$=300 l/kg, ^{137}Cs - $K_н$ = 1000 l /kg; 3) for silt and sediment the accumulation factor ^{90}Sr $K_н$ = 2.000 l/kg, 137Cs - $K_н$ = 100000 l / kg; 4) The average depth of the water reservoir is 4 m. With these parameters, the accumulation of radionuclides in the sand can be neglected, while the error calculations for ^{137}Cs will not exceed 1% , and for ^{90}Sr - 15%.

For other freshwater reservoirs the above parameters can vary greatly. Thus, according to our estimates, in the cooling pond of Khmelnitsky NPP the average rate of silt accumulation (0.36 kg / m2 per year) is twice as low than in the Kiev reservoir, the coefficient of ^{137}Cs by suspensions $K_н$ = 270000 l /kg, and by silts - not more than 18,000 l / kg. Lake Beloye, located on the territory of Vladimiretskyi district of Rivne region, is characterized by rather small average speed of silt accumulation, and the integrated density of ^{137}Cs contamination of sands is higher than silts.

For analytical description of the model we use the following notation: A_0- a single entry in the water body of the activity of a radionuclide per unit of water area, Bq /m^2; V - volume of water per unit area, l / m^2; Δh - average speed of silt accumulation on the body of water, kg(m^2 year)$^{-1}$; z - thickness of "active" layer of sediment, kg/ m^2; $T_{1/2}$ - halftime of self-decontamination of water masses from radionuclides, years; $A_в(t)$, $A_д(t)$ - specific activity of water and sediments, respectively, at time t, Bq / liter (Bq / kg). Equal quantities $K_н$ for suspended matter and silt is identical to equality of specific activity of radionuclides in the suspended matter and the top "active" layer of silt.

A year after the entry the balance of radionuclides is described by the following equation :

$$A_0 = A_в(t)\cdot V + A_д(t)\cdot(\Delta h + z). \quad (3)$$

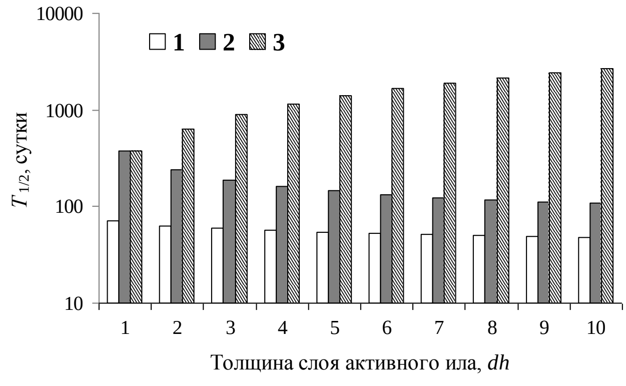

Fig. 3.
The dynamics of the speed of self-decontamination of water masses from ^{137}Cs (*dh* (≡Δh)) - the rate of silt accumulation):
1 - first year; 2 - second year; 3 - third year

We introduce the dimensionless quantity n, numerically equal to $z/\Delta h$; then equation (3) can be written as follows:

$$A_0 = A_в(t)\cdot V + A_д(t)\cdot\Delta h\cdot(1 + n). \quad (4)$$

By using the substitution $A_д = K_н \cdot A_в$, we exclude the specific activity of sediments from the equation (4) (substitution is valid for the upper layer involved in the exchange of silts at t >>1 day):

$$A_0 = A_в(t) \cdot V + A_в(t) \cdot K_н \cdot \Delta h \cdot (1+n), \qquad (5)$$

or

$$A_0/V = A_в(t) \cdot (1 + K_н \cdot \Delta h \cdot (1+n)/V). \qquad (6)$$

Using the equations (1) and (6), we get the value $T_{1/2}$ (years) for the first year:

$$T_{1/2} = \ln 2 \cdot [\ln(1 + K_н \cdot \Delta h \cdot (1+n)/V)]^{-1}. \qquad (7)$$

For subsequent years we consequently applied the above-described scheme, i.e. the equations (4)—(7) with taking into account burial (exclusion from further calculations) of the activity components of the bottom layer Δh.

At the selected options, depending on the power of the "active" layer of silt in the first year after entry of ^{137}Cs in the body of water its content in the water masses will be decreased by 35-195 times, corresponding to halftime of decontamination of water masses from the radionuclide 71-48 days (Fig. 3). For the second year the activity of water mass is reduced by 2-10 times with a halftime of decontamination of 380-108 days, and in the subsequent years halftime of decontamination will be 380-2670 days.

Fig 4. The dynamics of the speed of self-decontamination of water masses from ^{90}Sr (dh ($\equiv\Delta h$)) - the rate of silt accumulation)
1 - first year; 2 - second year; 3 - third year.

Calculations show that within two years after the entry of ^{137}Cs in the body of water, for any fixed layer the thickness of "active" silt, and halftime of decontamination of water masses do not change with time. Thus, at the thickness of the "active" layer $10\Delta h$, starting from the third year, the activity of ^{137}Cs in the water masses will be reduced twice in 7.3 years. Note, that while the first two years the rate of removal of radionuclides from the water mass is increased with the increasing of thickness of the "active" layer, the subsequent rate of purification of water masses is slowing down.

Purification of the water masses from ^{90}Sr occurs slower than from ^{137}Cs (Fig. 4). With thickness of the "active" layer of silt Δh the halftime of decontamination of water masses will be 1.5-3 years. With thickness of the "active" layer $10\Delta h$ the halftime of decontamination of water masses of ^{90}Sr in the first year is 160 days, for the second year - about 200 days, and for the third and subsequent years - about 9.2 years

Conclusions

The results of the simulation of speed of self-decontamination of water masses from radionuclides in the first year after inflow are in good agreement with those obtained taking the case of the cascade of the Dnieper reservoirs, and the results of subsequent years - with the rate of self-decontamination of water masses from radionuclides in the reservoirs of the exclusion zone. Accounting of disposal of radioactive masses in the bottom sediments can explain the slowdown of the process of self-decontamination of water masses from radionuclides.

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