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РАДІОБІОЛОГІЯ ТА РАДІОЕКОЛОГІЯ
RADIOBIOLOGY AND RADIOECOLOGY

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MODELING OF LONG-TERM RADIOLOGICAL AND TOXICOLOGICAL IMPACTS OF THE URANIUM MILL TAILINGS ON GROUNDWATER AND SURFACE WATER

Modeling predictions are presented of radionuclide transport processes in the zone of influence of the Zahidne uranium mill tailings situated at the Prydniprovsky Chemical Plant (PChP), Kamianske. The groundwater transport model was developed using the NORMALYSA software. Refined estimates of parameters of water exchange in the zone of uranium mill tailing (obtained from field studies and modeling of groundwater flow processes) were used to parameterize the model for radionuclide transport in groundwater. Calibration of the radionuclide transport model using monitoring data on radioactive contamination of groundwater in 2005 - 2021 allowed to estimate the sorption distribution coefficient (Kd) for the most hazardous contaminants ^{238,234}U isotopes (Kd = 8 ± 2 l/kg) and estimate the rate of uranium migration in groundwater. According to modeling, during the next 800 to 1100 years, uranium concentration in wells in the zone of influence of uranium mill tailing (at 500 - 800 m distance) will be determined mainly by the contamination of the alluvial aquifer, which was formed during the operation period of the uranium mill tailing. According to modeling predictions, usage of groundwater (partial drinking water consumption, irrigation) outside the PChP site downstream of the uranium mill tailing will result in doses exceeding the relevant reference level (annual effective dose > 1 mSv/year) in 380 - 440 years, while the toxicological impact will result in the exceeding of the acceptable hazard index for uranium (HI > 1) in 200 - 260 years. Modeling results indicate the importance of restricting the use of groundwater downstream of the uranium mill tailing within the PChP industrial site and, in the longer term, beyond its boundary. At the same time, contamination of the Konoplyanka River due to the migration of radionuclides from the uranium mill tailing does not pose unacceptable radiological and toxicological risks for the considered scenario (irrigation, fish consumption) due to the dilution of contaminants in surface waters.

Keywords: groundwater, groundwater modeling, Prydniprovsky Chemical Plant, risk assessment, uranium.

1. Introduction

Uranium mill tailings created at the beginning of the “nuclear era” in the 1950 and 1960s, when there was no relevant experience of environmental protection and established regulatory framework, in many cases have become over time sources of hazardous radiological and toxicological impacts on the population and environment. Justification of remedial measures for such uranium production legacy sites requires an assessment of dispersion of contaminants in groundwater and surface water [1, 2]. This article presents the results of predictive modeling of radionuclide transport in groundwater and radiological risk assessment for the Zahidne uranium mill tailing, which belongs to the infrastructure of the former industrial enterprise Prydniprovsky Chemical Plant (PChP), Kamianske. The PChP was one of the main enterprises where uranium raw materials for the nuclear program of the USSR were produced, and where operated a number of related chemical enterprises [3, 4]. Zahidne uranium mill tailing represents one of the most serious sources of radioactive and chemical contamination of groundwater within the PChP industrial site [5 - 8]. To substantiate the strategy for bringing Zahidne uranium mill tailing to an environmentally safe condition, it is necessary to understand the long-term risks to the environment and humans caused by releases of radioactive contaminants from the uranium mill tailing to groundwater and surface water and to evaluate the impact of possible remedial measures on radionuclide transport processes.

Screening radionuclide transport modeling analyses for the Zahidne uranium mill tailing were carried out by Skalskji et al. [9]. In the above case, the regional model of the PChP site was used for groundwater flow calculations. A simplified set of input data for the Zahidne uranium mill tailing was used in the IAEA MODARIA-II project as a test data set for comparing models for calculating dose impacts from uranium legacy sites on the environment [10]. Due to the lack of data, these modeling studies used expert estimates and literature data for main groundwater flow and radionuclide transport parameters.

In recent years, new data on the geological structure of the study area, as well as on groundwater level regime and contamination have been obtained [5 - 8]. As a result of calibration of the detailed cross-sectional groundwater flow model of the study site, the infiltration recharge rates and groundwater seepage rates in the zone of influence of the Zahidne uranium mill tailing were precised [11]. Also, potential scenarios for the usage of contaminated groundwater and surface water by the population in the zone of influence of the PChP industrial site were identified, and corresponding dose models were developed [12].

The above studies allowed us to perform a more detailed and accurate parameterization of the model of radionuclide transport from the Zahidne uranium mill tailing to groundwater and to the Konoplyanka River. To calibrate the transport model, we first used in this study a time series of observations of radioactive contaminants in groundwater for 2005 - 2021. The modeling analyses using the new data described above resulted in refined predictions of the impact of radionuclide transport processes from the Zahidne uranium mill tailing on the hydrogeological environment and on the Konoplyanka River, as well as allowed estimating the long-term radiological and toxicological impacts on the population, including scenarios of the implementation of remediation measures, which are presented below.

2. Materials and methods

2.1. Description of technogenic and hydrogeological conditions of the Zahidne uranium mill tailing

Technogenic conditions of the Zahidne uranium mill tailing are characterized based on the data of [5, 6, 12 - 14]. Zahidne uranium mill tailing was commissioned in 1949 at an early stage of establishing the PChP, and operated until 1954. Uranium mill tailing was constructed in a former clay pit at the edge of the 2nd terrace of the Dnipro River at the PChP Southern Site (Fig. 1).

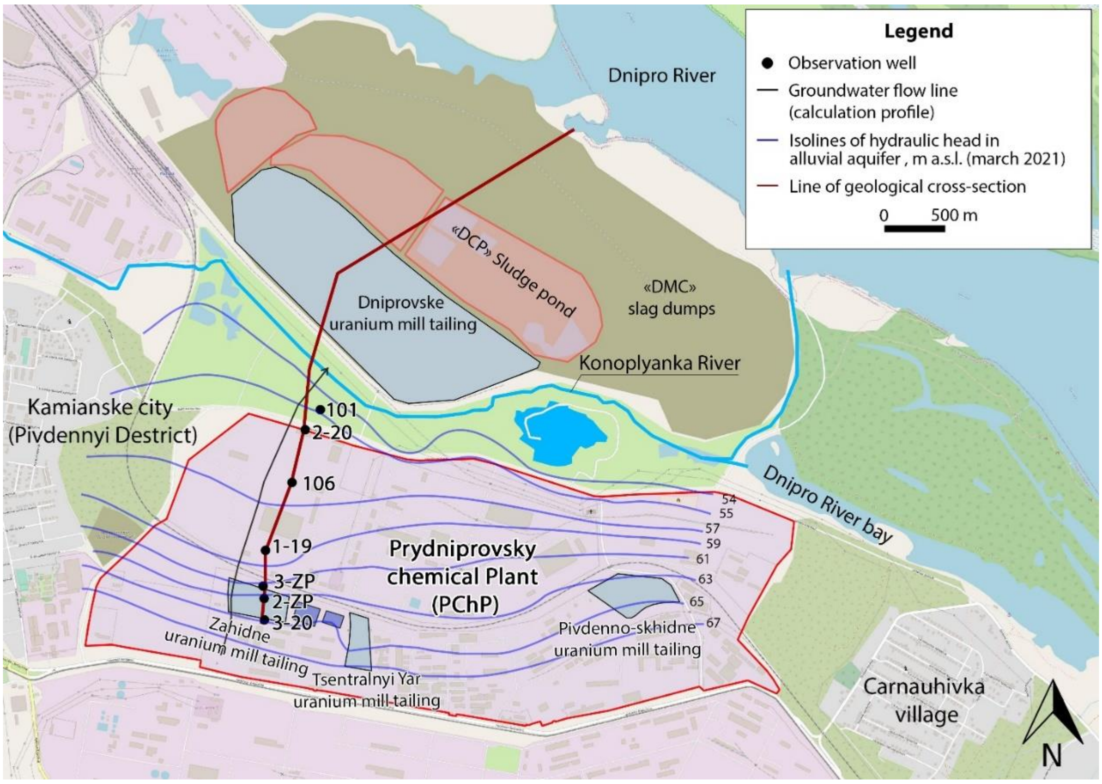


Fig. 1. Schematic map of the territory of the PChP showing the location of the Zahidne uranium mill tailings. (See color Figure on the journal website.)

From the side of the 1st terrace of the Dnipro River, the uranium mill tailing was confined by an earth-filled dam. Wastes from uranium ore leaching were transported to the uranium mill tailing both in a dry condition by conveyor (in the first years of operation) and in the pulp form by pipelines (in the subsequent period). The thickness of the tailings material layer within the facility varies from 1 to 12.5 m. The total volume of waste is estimated as 250000 m^3. In 2000, a protective soil cover composed of loam material (1.0 - 2.8 m thick) was installed in the northern part of the uranium mill tailing, and a system of drainage ditches was put in place to collect rainwater runoff. Before storage, the tailings material after the acid leaching (using nitric or sulfuric acid) was neutralized by adding lime or ammonia. As a result of the above technology, alkaline geochemical conditions (pH 8.5-9.5) were formed in the tailing material. The average content of ^{238}U in the tailings material is 1.7 kBq/kg, ^{226}Ra – 5.9 kBq/kg [13, 14].

The geological cross-section of the study site is composed of geological formations of Precambrian and Quaternary age. Hydrogeological conditions are characterized by the presence of aquifers in technogenic deposits (water-saturated wastes in the uranium mill tailing body – the perched groundwater aquifer) and in alluvial sediments and in the upper part of the underlying weathered crystalline rocks. The geological structure and hydrogeological conditions of the study area are described in more detail in [5, 9].

The alluvial aquifer in the zone of influence of Zahidne uranium mill tailing has been significantly impacted by infiltration of contaminated pore solutions from the uranium mill tailing body [5, 6, 8]. The uranium mill tailing represents a source of groundwater contamination by uranium isotopes (^{238,234}U), macroions (sulfate ion, magnesium, nitrates, ammonium), and toxic metals (manganese, lead) with significant exceedance of the maximum permissible concentrations for drinking water. Among the radionuclides, uranium isotopes show the highest mobility in groundwater in the area of influence of the Zahidne uranium mill tailing and other PChP facilities [5, 6, 15].

According to the monitoring data of 2005 - 2021, the maximum concentration of the sum of uranium isotopes (238, 234) in the aquifer in technogenic deposits in the Zahidne uranium mill tailing (source of migration) reached up to 1290 Bq/l in 2007, which is thousands of times higher than the drinking water safety standard for a mixture of uranium isotopes of 1 Bq/l. The plume of uranium migration in groundwater with a concentration of up to 100 Bq/l by the mixture of isotopes can be traced at a distance of about 170 m downstream from the uranium mill tailing (to well 1-2019). For other radionuclides of the ^{238}U decay series (^{226}Ra, ^{210}Po, ^{210}Pb), no clearly defined plumes of contamination are observed in groundwater in the alluvial aquifer [5 - 8].

Groundwater flowing from Zahidne uranium mill tailing discharges into the Konoplyanka River, which flows 1.2 km to the North of the site and subsequently joins the Dnipro River (see Fig. 1). The Konoplyanka River is characterized by an average yearly water flow rate of about 1.0 m^3/s [5].

2.2 Modeling of radionuclide transport processes

To parameterize the radionuclide transport model, we used data on water exchange parameters in the zone of influence of Zahidne uranium mill tailing obtained in recent years based on field studies and groundwater flow modeling (Table 1).

Table 1. **Hydrodynamic parameters used to parameterize the radionuclide transport model**

Parameter	Value	Evaluation method	Reference
Infiltration leakage rate from the tailing body to the aquifer, mm/year	50	Chlorine ion balance method	[7]
Infiltration recharge rate within the 1st terrace, mm/year	200	Water table fluctuation (WTF) method, calibration of groundwater flow model	[11]
Infiltration recharge rate within the 2nd terrace, mm/year	150	WTF method, calibration of groundwater flow model	[11]
Darcy flow velocity in the aquifer, m/year	19	Tracer test, calibration of groundwater flow model	[11]

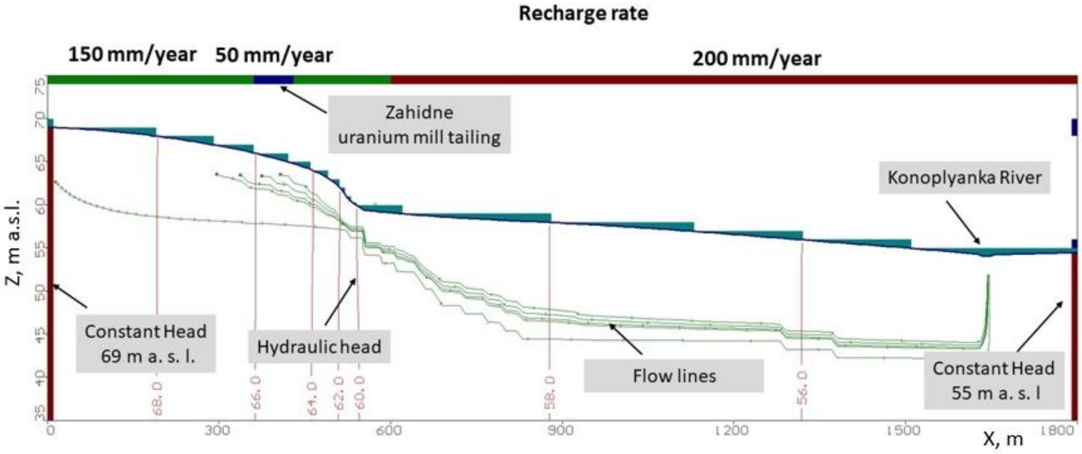


Fig. 2. Hydrodynamic flow net based on the calibrated groundwater flow model [11].
(See color Figure on the journal website.)

These estimates of the hydrodynamic parameters were precised by calibrating the cross-sectional groundwater flow model for Zahidne uranium mill tailings developed using Visual Modflow 3.0 software (https://www.waterloohydrogeologic.com/products/visual-modflow-flex/) [11]. The hydrodynamic flow net of the calibrated groundwater flow model, which served as the basis for the radionuclide transport modeling, is shown in Fig. 2.

Migration of ^{238}U decay series radionuclides along the groundwater flow path from the Zahidne uranium mill tailing towards the Konoplyanka River was modeled by the Ecolego 8.0 software using the NORMALYSA library (http://project.facilia.se/normalysa/software.html) [16]. Earlier a similar approach for modeling radionuclide transport in groundwater was used in [9, 17].

The model simulates the migration of radionuclides for the following decay chain: $^{238}U \rightarrow ^{234}U \rightarrow ^{226}Ra \rightarrow ^{210}Pb \rightarrow ^{210}Po$. The process of radionuclide migration in the soils of the vadose zone and in the aquifer (along the flow line) is described by the advection-dispersion equation taking into account radionuclide sorption described by the Kd model (linear equilibrium exchangeable sorption). The Ecolego software automatically takes into account radioactive decay and ingrowth of daughter radionuclides. For advection-dispersion modeling, the Ecolego "Transport Block" is used, which provides an accurate numerical solution of the advection-dispersion equation.

The radionuclide transport modeling assumed steady-state hydrodynamic conditions during the whole simulated time period (2000 years), which correspond to present-day conditions (see Table 1). We believe that this is a conservative assumption. At present time hydrodynamic conditions at the PChP industrial site are influenced by "technogenic recharge" of the aquifer caused by leakages from degraded water pipelines and storm runoff collectors. Such additional "technogenic" stress on the groundwater system causes higher groundwater flow velocity in the aquifer compared to natural undisturbed conditions [11], which would likely prevail in the long term after the PChP site is fully decommissioned.

2.3. Assessment of radiological and toxicological impacts

To predict radiological and toxicological impacts from the use of contaminated groundwater (from wells) and surface water (from the Konoplyanka River), the approach and mathematical models described in Zanoz et al. [12] were used. Modelled scenarios of the possible use of contaminated water resources in the PChP area by hypothetical "representative persons" from the local population are described in Table 2. By definition, a representative person is an exposed individual who is representative of the most highly exposed individuals in the relevant population group. As a radiation safety criterion, it was assumed that the annual effective dose for representative persons should not exceed 1 mSv/year. This criterion corresponds to the lower bound of reference levels for the "existing exposure situations" [18, 19], and it was used recently in a number of Ukrainian and international projects on safety assessment of nuclear legacy sites [7, 10, 20].

Table 2. **Description of scenarios of use (consumption) of contaminated groundwater and surface water in the zone of influence of the Zahidne uranium mill tailing [12]**

Scenario (SC) (source of water)	Exposure pathway (% of annual water or food consumption)		
	Drinking water	Irrigated vegetables	Fish
SC-1 (groundwater)			
Realistic	10 %	100 % (potato)	–
Conservative	25 %	100 % (potato), 50 % (vegetables)	–
SC-2 (Konoplyanka River)			
Realistic	–	100 % (potato)	100 %
Conservative	–	100 % (potato), 50 % (vegetables)	100 %

The first scenario (SC-1) assumes that contaminated groundwater from the unconfined aquifer collected from a well located in the zone of influence of the uranium mill tailing is used as irrigation water for growing potatoes and vegetables (e.g., tomatoes, cucumbers, etc.) in the garden. It is also assumed that groundwater is sometimes used for drinking (e.g., during gardening throughout the year). The second scenario (SC-2) considers the use of water from the Konoplyanka River, which is contaminated due to the discharge of contaminated groundwater into the river, for irrigation of a vegetable garden (similar to the first scenario), as well as consumption of fish from the river. Calculations were performed for "realistic" and "conservative" (or "worst case") sets of input parameters (see Table 2). The respective sets of parameters also differ in radioecological parameters (radionuclide accumulation coefficients in vegetables and fish, sorption coefficients of Kd distribution for soil; see Table 3 in [12]). In this study, the predictive modeling was limited to the calculation of radiation doses from contaminated groundwater and surface water for an adult.

Uranium in groundwater is a potential source of not only radiological but also hazardous toxicological impacts [21]. Toxicological risks to human health from uranium were assessed using the methodology developed by the U.S. Environmental Protection

Agency for assessing exposure to non-carcinogenic substances [22]. During the toxicological assessments, the calculated accumulation of toxic substances in the body of representative persons by oral ingestion was compared with the corresponding criterion of the reference dose (RfD) of the toxic element. Based on the Integrated Risk Information System database (https://www.epa.gov/iris) for uranium the RfD is 0.003 mg/(kg per day). The final calculation parameter was the daily intake of the toxic element per kilogram of body weight of a representative person, which is used to calculate the "hazard index" (HI):

$$HI = Dose / RfD.$$

If the calculated intake of a chemical exceeds the RfD value (HI > 1), it means that exposure to this chemical poses a risk to human health.

3. Results

3.1. Calibration of contaminant transport model and long-term predictions of radionuclide migration

The block diagram of the transport model developed using the NORMALYSA describing the migration of radionuclides from the Zahidne uranium mill tailing is shown in Fig. 3.

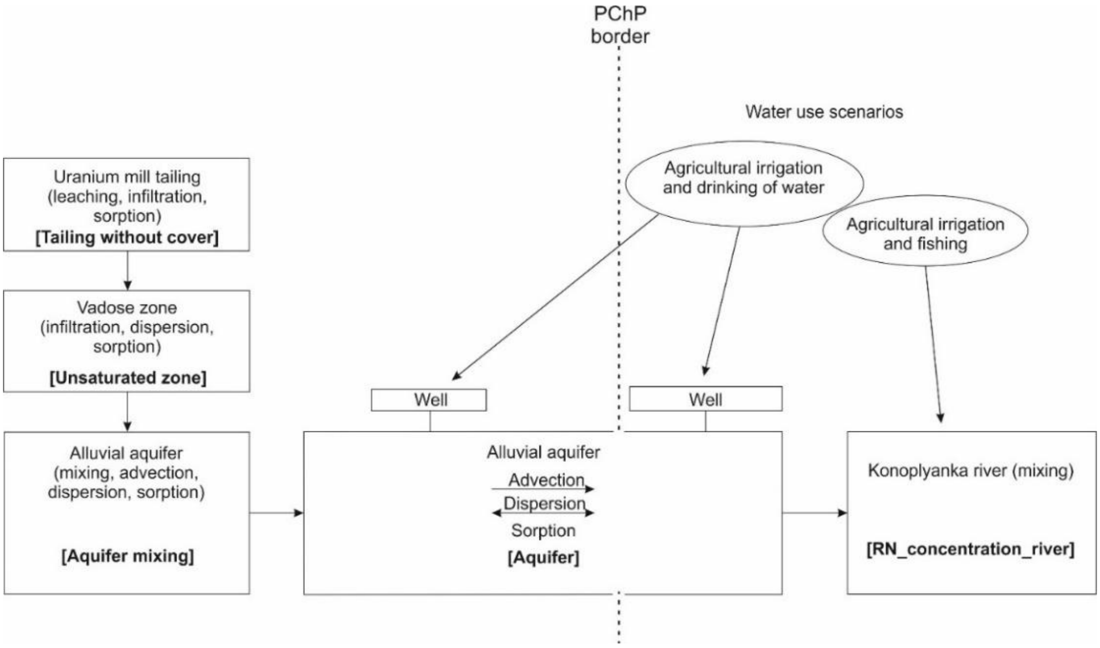


Fig. 3. Block diagram of the groundwater transport model for the Zahidne uranium mill tailings with the main components (compartments), description of the modeled radionuclide migration processes, and names of the modules from the NORMALYSA library used to model the corresponding compartment (in square brackets).

The arrows show transfers of parameters (concentrations, radionuclides fluxes in groundwater) between the main model compartments represented by the corresponding NORMALYSA modules. The NORMALYSA module "Tailings without cover" was used to model the leaching of radionuclides from the uranium mill tailing, which implements the mathematical leaching model described in [23]. The "Unsaturated zone" module models the vertical migration of radionuclides in the soils of the vadose zone. The aquifer area under the uranium mill tailing, where the vertical infiltration flux from the uranium mill tailing mixes with the horizontal groundwater flow, is modeled by the "Aquifer Mixing" module. The "Aquifer" module simulates the horizontal radionuclide transport with groundwater flow along the flow line in the direction of the Konoplyanka River. The concentration of radionuclides in the Konoplyanka River was determined based on the mixing model (mixing of groundwater discharge with the upstream river flow). The geometric, physical, and radiation characteristics of the tailing body and the parameters of the vadose zone are based on data of [13, 14].

Groundwater fluxes between the model compartments were determined according to the results of the Visual Modflow calculation [11].

The key parameters of the radionuclide transport model are sorption distribution coefficients (Kd-s) of radionuclides. The Kd values for the main model components (tailing body, vadose zone, aquifer) were assigned values based on previous studies and literature data, as well as based on model calibration (Table 3). The model calibration using the regime observations of radionuclide concentrations in groundwater was applied to estimate the Kd of uranium isotopes for the alluvial aquifer deposits, where a gradual propagation and dispersion of the uranium plume along the groundwater flow path was observed in 2005 - 2021. The model calibration results are described below.

Table 3. **Kd values for the main compartments of the radionuclide transport model, m^3/kg**

Radionuclide	Kd value, m^3/kg	Reference
Tailing body		
$^{238/234}U$	0.0052	[7]
^{226}Ra	0.8	[7]
^{210}Pb	1.8	[7]
^{210}Po	0.46	In-situ estimate
Vadose zone and aquifer		
$^{238/234}U$	0.008	Calibration of model
^{226}Ra	2.5	[24]
^{210}Pb	2	[24]
^{210}Po	0.21	[24]

The starting time of the modeling forecast was the year 2005, when regular monitoring groundwater observations were started at the Zahidne uranium mill tailing. The radionuclide concentrations in groundwater below the tailings were assumed to be equal to the maximum concentrations observed in the pore solutions in the perched groundwater within the tailings body (which is believed to be a conservative assumption).

For soils of the vadose zone, the initial contamination with radionuclide "i" ($C_{soil,i}$) was calculated according to the formula $C_{soil,i} = C_{pore,i} \cdot Kd_{soil,i}$, where $C_{pore,i}$ is the radionuclide concentration in tailing pore solutions (for 2005), $Kd_{soil,i}$ is the sorption Kd of the vadose zone soils. When setting the initial conditions in the radionuclide transport model, it was assumed that ^{210}Po activity is in equilibrium with ^{210}Pb.

The model was calibrated by selecting the Kd of uranium (in the range of values from 1 to 30 l/kg) and the horizontal hydro-dispersion parameter (α_L) (in the range from 10 to 100 m) minimizing the mean squared error of the calculated concentration of uranium isotopes in groundwater compared to the data of observations for 2005 - 2021 for monitoring wells located in the zone of influence of the uranium mill tailing. The smallest errors for tested values of the hydro-dispersion parameters were obtained in the range of uranium Kd values from 6 to 10 l/kg (Fig. 4). The results of the transport model calibration are presented in Fig. 5.

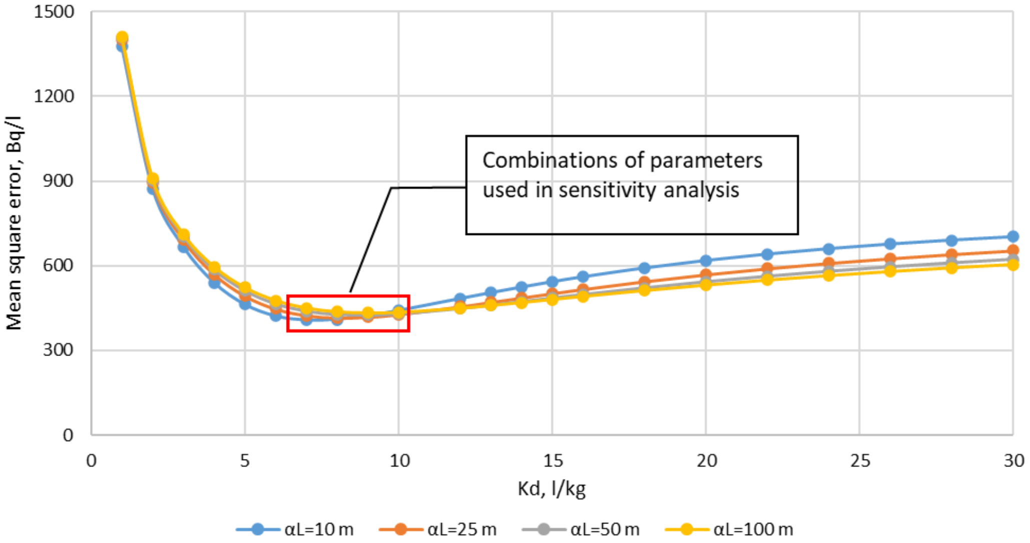
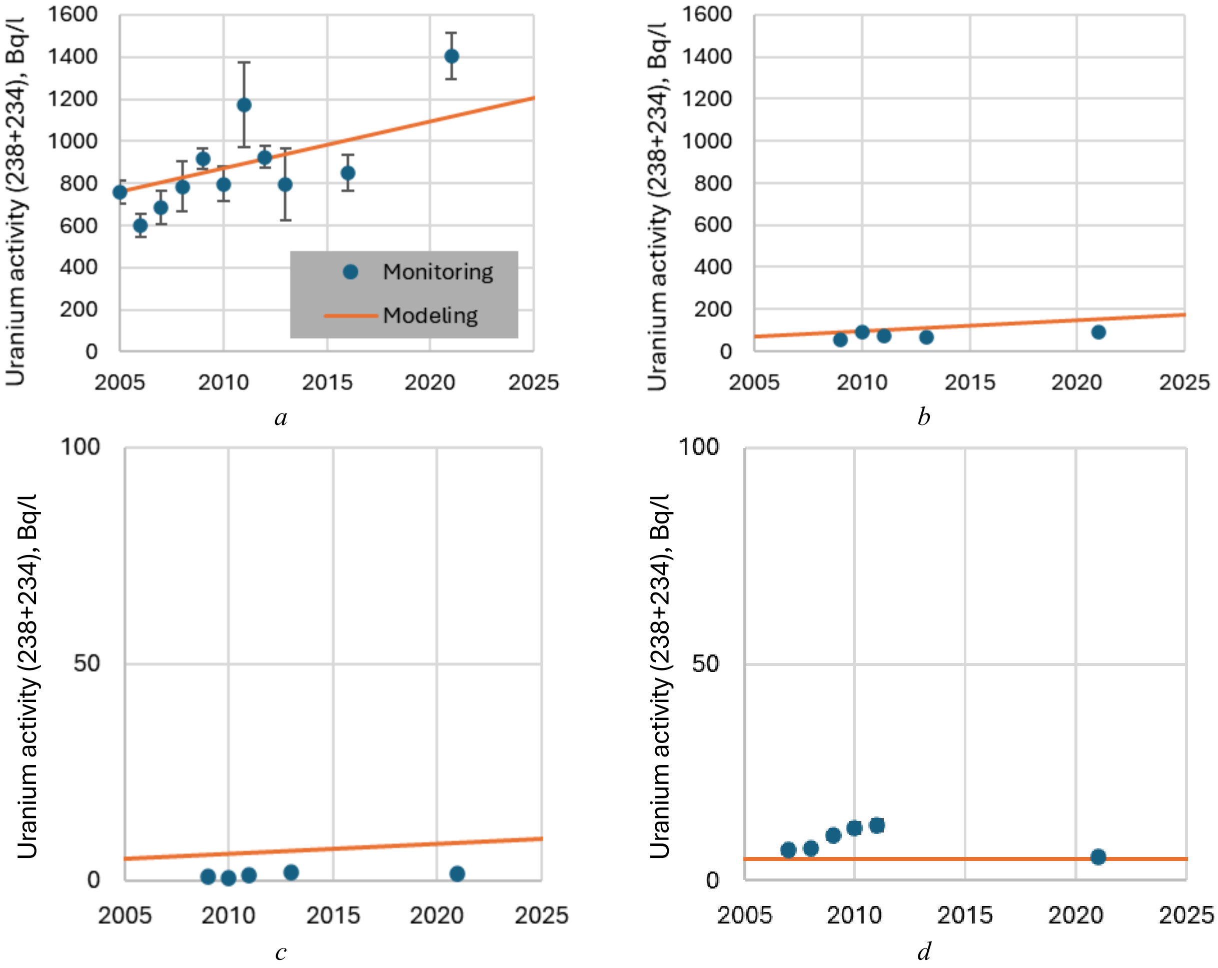


Fig. 4. Graph of the mean squared error (for well 3-ZP and well 1-19) of the modeling results compared to the monitoring data depending on the values of Kd and the dispersion parameter obtained in the course of groundwater transport model calibration. (See color Figure on the journal website.)

As a result of model calibration, the following "optimal" values of uranium transport parameters were selected for the alluvial aquifer: Kd = 8 l/kg, α_L = 25 m. In the absence of other data, the same values of Kd for uranium were used for the soils of the vadose zone. Fig. 5 shows graphs of $^{238,234}U$ concentrations in groundwater based on monitoring data and modeling results using the radionuclide transport model for selected "optimal" parameter values.

The Kd values of uranium obtained during the calibration correspond to the lower range of Kd values of this element known from the literature [21, 23, 24] and are close to the experimentally determined uranium Kd values for Zahidne uranium mill tailing [7, 13]. The high mobility of uranium in groundwater may be due to the fact that, according to geochemical modeling, this element migrates in the oxidation state +6 in the form of uranyl carbonate complexes (UO_2^{2+}), including negatively charged complexes [7].



a *b* *c* *d*

Fig. 5. Calibration graphs of the groundwater transport model for uranium migration in groundwater: *a* - well 3-ZP, *b* - well 1-19, *c* - well 106, *d* - well 2-20. Vertical bars show the analytical error of monitoring data. (See color Figure on the journal website.)

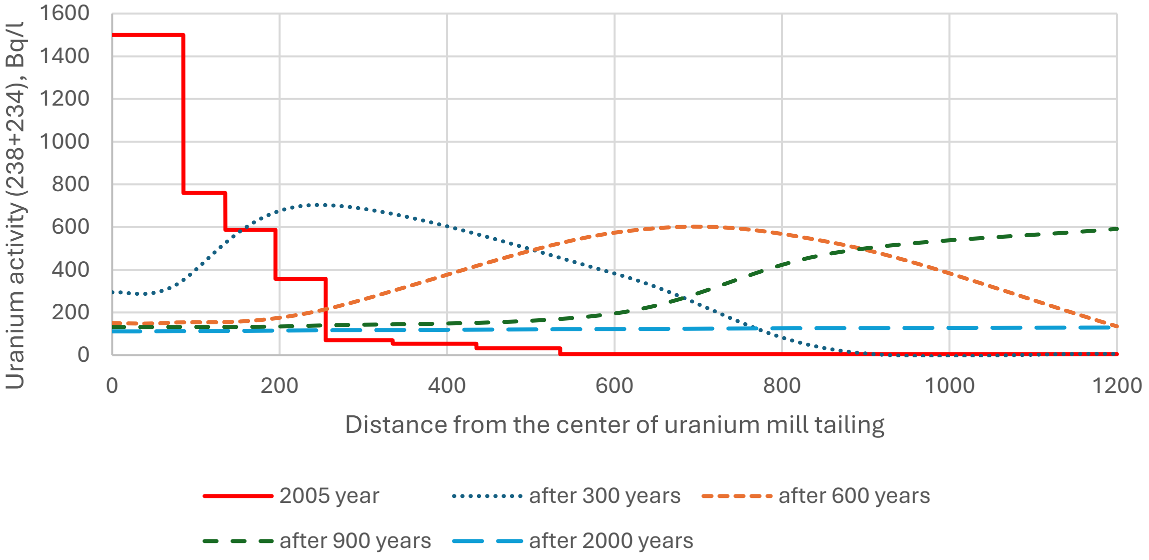
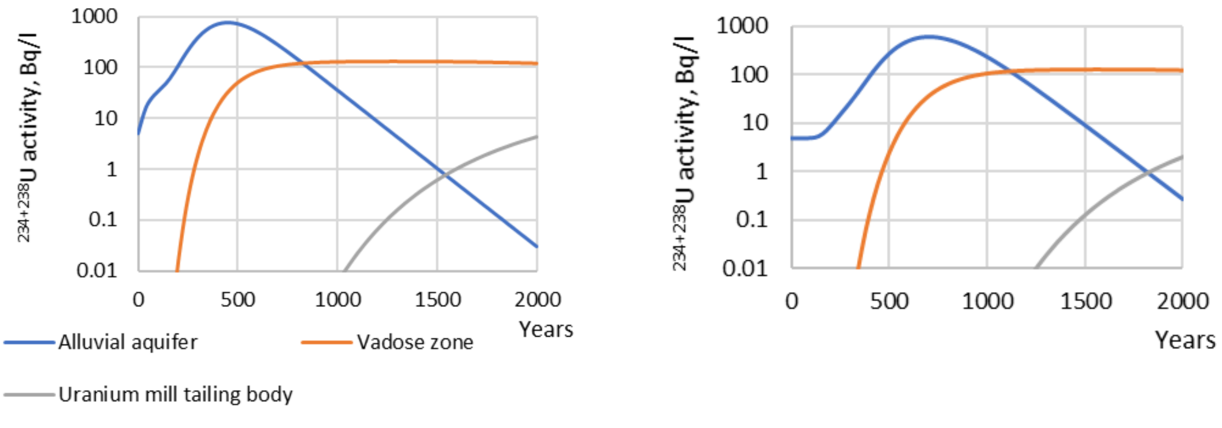


Fig. 6. Initial condition (2005) and long-term predictions of uranium plume migration in groundwater from the uranium mill tailings along the groundwater flow line towards the Konoplyanka River. (See color Figure on the journal website.)

The calibrated radionuclide transport model was used for long-term predictions of the migration of radioactive contaminants in groundwater from the Zahidne uranium mill tailing towards the Konoplyanka River. The simulated dynamics of movement of the plume of groundwater contaminated with uranium isotopes along the flow path is shown in Fig. 6. The predicted rate of uranium migration in groundwater is estimated at about 1 m/year, which is almost 100 times slower than the actual rate of groundwater flow (due to sorption).

According to modeling estimates, it will take thousands of years for the aquifer to gradually self-cleanse from uranium contamination (see Fig. 6).

Modeling results show that in the next 800 to 1100 years, uranium concentration in wells in the zone of influence of uranium mill tailing (wells 106 and 2-20) will be determined mainly by the source of contamination in the alluvial aquifer (as of 2005), which was formed during operation of the uranium mill tailing (Fig. 7). Contaminated soils of the vadose zone under uranium mill tailing and the tailing body have a relatively small impact on the predicted uranium concentration in downstream wells, which is due to the installation of an engineered soil cover tailing that minimizes infiltration of meteoric water through the tailing body and leaching of radioactive contaminants from the relevant compartments. This means that the retrieval of wastes from the Zahidne uranium mill tailing and their re-disposal at another storage site (e.g., Sukhachivske-2 or Dniprovske uranium mill tailing) will not be an efficient measure to minimize radioactive contamination of groundwater downstream of the uranium mill tailing.



a *b*

Fig. 7. Contribution of different contamination sources (as of 2005) to long-term groundwater contamination by uranium for wells at different distances from the uranium mill tailing: *a* - well 106 (500 m), *b* - well 2-20 (800 m). (See color Figure on the journal website.)

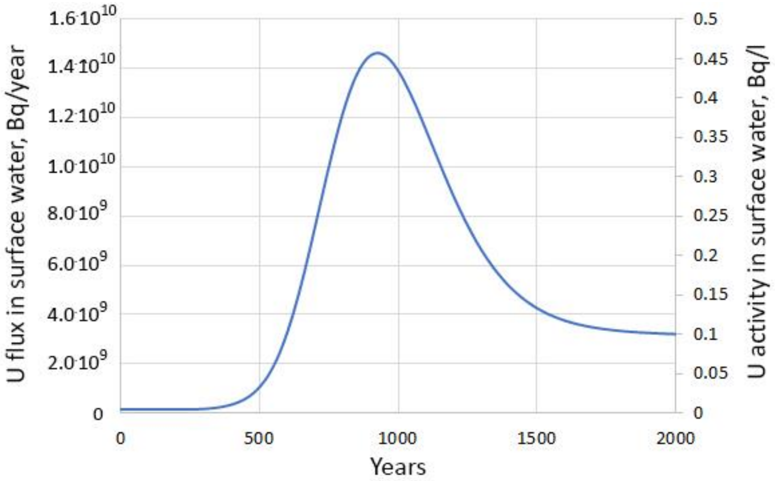


Fig. 8. Predicted uranium release from the Zahidne uranium mill tailings to Konoplyanka River and resulting concentration in river water.

The graph of the long-term transport of uranium by groundwater from the Zahidne uranium mill tailing to the Konoplyanka River is shown in Fig. 8. The yearly averaged water flow rate in the Konoplyanka River was assumed to be 1 m^3/s [5]. The graph in Fig. 8 accounts only for radionuclide transport in groundwater to the river from the Zahidne uranium mill tailing, and does not take into account other potential sources of contamination.

3.2. Sensitivity analysis of modeling results to Kd and α_L parameters

According to the results of radionuclide transport model calibration (see Fig. 4), there is a set of combinations of values of the Kd and the hydro-dispersion parameter (α_L) that provides close values of the mean squared error “model - observation” within 5 % (i.e., nearly equally good matching of modeling results and monitoring data). Therefore, calculations were performed to assess the sensitivity of the results of radionuclide transport predictions to the variation of the input values of Kd and α_L (see Fig. 5). The maximum calculated values of uranium concentration in the well 2-20 (outside the PChP territory) and in the Konoplyanka River and times when the corresponding values were reached were selected as “target parameters” of the sensitivity analysis (Fig. 9).

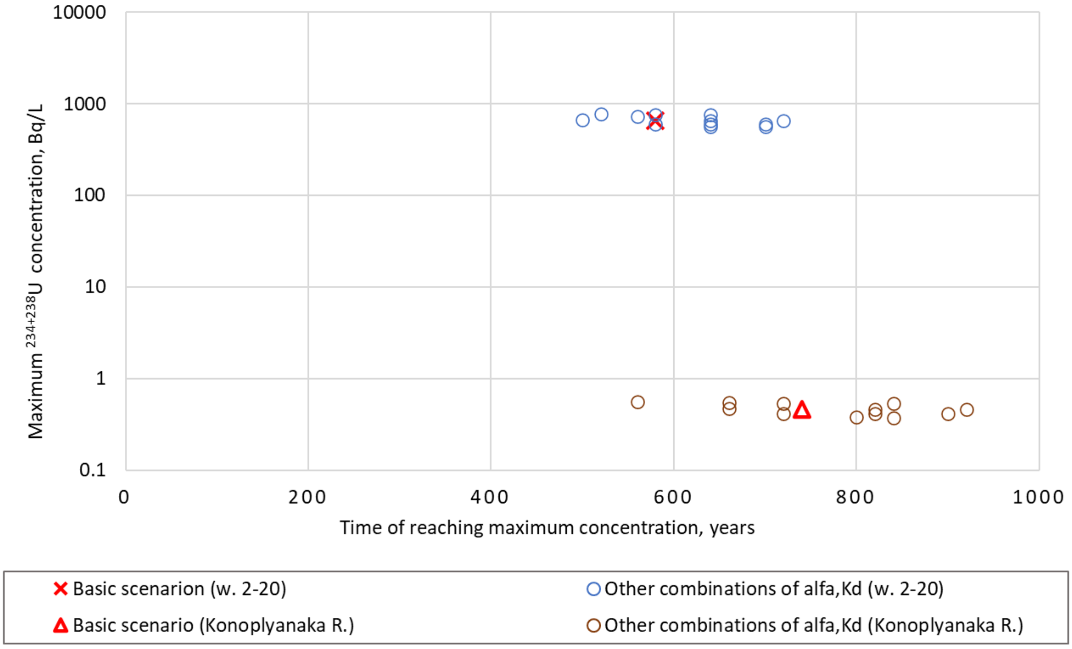


Fig. 9. The results of the model sensitivity analysis: maximum values of uranium concentration in water and time to reach the maximum for the well 2-20 and Konoplyanka River depending on the values of the parameters α_L (m) and Kd (l/kg). (See color Figure on the journal website.)

The time for reaching the maximum uranium concentration in the well 2-20 and Konoplyanka River for different combinations of transport parameters used in the sensitivity analysis varies by 20 - 25 %, and the values of maximum uranium activity values vary within 15 - 20 % compared to the “baseline forecast” (i.e., Kd = 8 l/kg, α_L = 25 m). For all combinations of parameters, the maximum predicted uranium concentrations in the Konoplyanka River are less than 1 Bq/l.

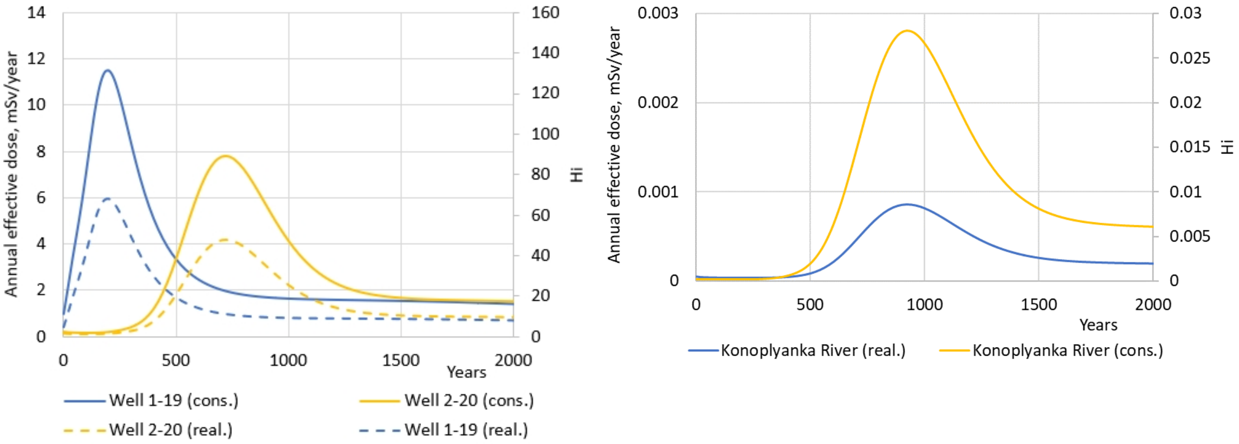
3.3. Assessments of long-term radiological and toxicological impacts

The predicted radionuclide concentrations in groundwater and in the Konoplyanka River obtained as a result of modeling (for the “baseline” set of transport parameters) were used to predict the dose impacts from using contaminated water according to the scenarios described in Table 2. The results of the dose calculations for the “realistic” and “conservative” input data sets are presented in Fig. 10 (doses are plotted in the left Y axes of the Figures). When interpreting the results, it should be taken into account that well 1-2019 is located within the 1st terrace at the PChP North site, and well 2-20 is located outside the PChP site downstream of the Zahidne uranium mill tailing.

Within the PChP industrial site, the predicted doses from groundwater use for 800 - 2000 years (depending on the selected input data set) exceed the reference level of 1 mSv/year (see Fig. 10, *a*, well 1-19). Outside the industrial site, downstream of the Zahidne uranium mill tailing, doses from water use do not exceed the criterion of 1 mSv/year for the next 380 to 440 years, but then doses from water usage exceed 1 mSv/y reference level due to the fact that the main front of uranium-contaminated groundwater extends beyond the PChP industrial site (see Fig. 10, *a*, well 2-20). The dose impacts (>1 mSv/year) from consumption of contaminated groundwater are mainly (>90 %) formed by ^{238}U and ^{234}U.

The dose calculations for the Konoplyanka River are shown in Fig. 10, *b*. These dose calculations take into account only the river water contamination by uranium formed by migration from Zahidne uranium mill tailing. Fig. 10, *b* shows that even for a conservative set of input data the predicted doses are less than 0.003 mSv/y, which is significantly less than 1 mSv/year.

As discussed, migration of uranium in groundwater poses not only radiological but also toxicological risks. Predicted values of the HI from uranium for the water use scenarios described in Table 1 are shown in Fig. 10 (HI values are plotted in the right Y axes of the Figures).



a *b*

Fig. 10. Predicted effective doses and HI from uranium for the scenarios of water usage from wells and for the Konoplyanka River: *a* - water usage from wells, *b* - water usage from the Konoplyanka River. (See color Figure on the journal website.)

Within the PChP industrial site, groundwater use is hazardous in terms of uranium concentration (HI > 1 for uranium) throughout the entire modeled time period. Outside the PChP industrial site, groundwater use becomes hazardous (HI > 1) due to uranium dispersion outside the industrial site according to the predictions in 200 - 260 years (depending on the selected scenario) (see Fig. 10, *a*).

At the same time, according to the modeling, uranium transport by groundwater to the Konoplyanka River does not lead to contamination of surface water at hazardous levels (HI values are less than 0.03, Fig. 10, *b*).

In fact, uranium concentrations in Konoplyanka River are influenced by a number of sources including groundwater transport from the adjacent Dniprovske tailings, which causes in current conditions ^{234,238}U activity concentration in river water of an order of 0.2 - 0.3 Bq/l [5, 12, 15]. Radiological impacts caused by the usage of water from the river considering scenarios listed in Table 2 and the present-day water contamination levels by uranium and other radionuclides were estimated in [12] as $3\ 10^{-3}$ – $3\ 10^{-2}$ mSv/y, while HI values for uranium were estimated as $7\ 10^{-3}$ – $2\ 10^{-2}$. Thus, the reference dose criteria (1 mSv/year) and toxicological impact criteria (HI < 1) will not be exceeded for the considered water usage scenarios for Konoplyanka River even when considering cumulative impacts due to the existing background contamination levels of river water and additional future impacts caused by radionuclide transport in groundwater from the Zahidne uranium mill tailings.

4. Conclusions

Calibration of the radionuclide transport model developed using the NORMALYSA software using data of observations of radioactive contamination of groundwater in 2005 - 2021 allowed us to estimate the sorption Kd for the most dangerous contaminants – ^{238,234}U isotopes (Kd = 8 ± 2·l/kg) and predict the rate of radioactive contaminant transport in groundwater, as well as to assess the long-term radiological and toxicological risks from groundwater contamination. Calculations show that groundwater contamination in the area between the Zahidne uranium mill tailing and the Konoplyanka River is a long-term problem. According to the predictions, the main front (>100 Bq/l) of uranium-contaminated groundwater reaches the Konoplyanka River in 580 (±120) years, and the contaminant’s flux into the river reaches its maximum values in 920 (±220) years. At the same time, in the next 800 to 1100 years, the uranium concentration in wells in the uranium mill tailing zone of influence (500 m to 800 m distances) will be determined mainly by the contamination source in the alluvial aquifer formed during the operation period of uranium mill tailing. Therefore, the retrieval of wastes from the uranium mill tailing and their re-disposal to another facility will not be an efficient measure in terms of minimizing radioactive contamination of groundwater.

Dose calculations show that contamination of groundwater within the PChP industrial site with ^{238}U decay series radionuclides is a source of long-term radiological and toxicological risks to humans, which exceed the relevant reference levels and/or hazard indices. Thus, the modeling results indicate the need for long-term restrictions on the use of groundwater downstream of the uranium mill tailings within the PChP industrial site, and in the longer term (>200 years) – beyond the PChP site. At the same time, contamination of the Konoplyanka River due to migration of radionuclides from the uranium mill tailing does not pose radiological and toxicological impacts in excess of considered reference levels due to dilution of contaminants in surface waters.

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МОДЕЛЮВАННЯ ДОВГОСТРОКОВИХ РАДІОЛОГІЧНИХ І ТОКСИКОЛОГІЧНИХ ВПЛИВІВ ВІД УРАНОВОГО ХВОСТОСХОВИЩА НА ПІДЗЕМНІ І ПОВЕРХНЕВІ ВОДИ

Представлено результати прогнозного моделювання геоміграційних процесів у зоні впливу уранового хвостосховища «Західне», що належить до Придніпровського хімічного заводу (ПХЗ), м. Кам’янське. При параметризації геоміграційної моделі використано уточнені параметри водообміну в зоні хвостосховища, отримані на основі польових досліджень і моделювання геофільтраційних процесів. Калібрування геоміграційної моделі на основі програми NORMALYSA з використанням даних спостережень за радіоактивним забрудненням підземних вод у 2005 - 2021 р. дало змогу оцінити сорбційний коефіцієнт (Kd) для найбільш небезпечних забруднювачів – ізотопів ^{238,234}U (Kd = 8 ± 2 л/кг) і спрогнозувати швидкість переносу урану в підземних водах. Згідно з моделюванням у найближчі 800 - 1100 років концентрація урану у свердловинах у зоні впливу хвостосховища (на відстані 500 - 800 м) буде визначатися головним чином осередком забруднення в алювіальному водоносному горизонті, що сформувався в період експлуатації хвостосховища. Відповідно, вилучення хвостів із хвостосховища, перезахоронення їх на інший пункт зберігання не буде ефективним заходом з точки зору мінімізації радіоактивного забруднення підземних вод. Згідно з прогнозами споживання підземних вод (часткове питне споживання, зрошення) за межами проммайданчику ПХЗ нижче за потоком від хвостосховища призведе до перевищення відповідного референтного рівня (доза опромінення >1 мЗв/рік) через 380 - 440 років, а токсикологічний вплив від урану призведе до перевищення припустимого індексу небезпеки для урану (HI > 1) – через 200 - 260 років. Результати моделювання свідчать про необхідність обмежень на використання підземних вод нижче за потоком від хвостосховища в межах промислового майданчику ПХЗ, а в довгостроковій перспективі – і за його межами. При цьому забруднення р. Коноплянка за рахунок міграції радіонуклідів із хвостосховища не являє неприйнятних радіологічних і токсикологічних ризиків для розглянутого сценарію (зрошення, споживання риби) внаслідок розбавлення забруднень у поверхневих водах.

Ключові слова: підземні води, гідрогеологічне моделювання, Придніпровський хімічний завод, оцінки ризику, уран.

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