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JOURNAL OF ENVIRONMENTAL ENGINEERING AND LANDSCAPE MANAGEMENT
ISSN 1648–6897 print/ISSN 1822-4199 online
2012 Volume 20(1): 42–48
doi:10.3846/16486897.2012.657775

EVALUATION OF EXPOSURE OF LAKE DRŪKŠIAI BIOTA REFERENCE ORGANISMS USING PROBABILISTIC METHODS

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Submitted 17 Aug. 2010; accepted 07 Apr. 2011

Abstract. External, internal and total exposure dose rates received by freshwater biota reference organisms of Ignalina NPP cooling basin Lake Drūkšiai were evaluated by applying ERICA computer code and probabilistic methods. These investigations were based on measurements of radionuclide activity in bottom sediments and water of Lake Drūkšiai (mean values, standard deviations, minimal and maximal values), and ecotoxicologic investigation data published during 1989–2003. The dose rate distributions for every investigated radionuclide and reference organism (average, minimum and maximum dose rate values, 5 and 95 order percentiles) were estimated. In the case of technogenic radionuclides originating from Ignalina NPP, the total dose rate values for Lake Drūkšiai reference organisms do not exceed the maximum permissible dose rate of 10 μGy h^{-1}, which is currently recommended for technogenic radionuclides in the European Union. Due to the ionising radiation of natural radionuclides, the exposure dose rate of reference organisms is much larger if compared with technogenic ones. The largest exposure dose rate caused by ^{238}U amounts to 7.2 μGy h^{-1} for insect larvae, and 41.8× μGy h^{-1} – for vascular plants.

Keywords: freshwater ecosystem, reference organisms, exposure dose rate, probabilistic methods.

1. Introduction

Assessment of environmental impact by Ignalina NPP on radioecological changes in Lake Drūkšiai – which is the Ignalina NPP cooling basin – was carried out (Šiluminė energetika 1989; Marčiulionienė *et al.* 1992; Mažeika 1998) taking into account the future development of the nuclear energy sector.

The environmental impact assessment report (Poveikio aplinkai... 2006) considering Lake Drūkšiai biota (fauna and flora) habitats indicates the frequency of cytogenetic damage (deviations in chromosome structure and number), which – due to specific impact caused by radionuclides in Lake Drūkšiai – is slightly greater than the environmental level. According to ecotoxicologic investigation of Lake Drūkšiai water and bottom sediments, this lake can be attributed to the category of low-toxic water basins. However, long-term (1989–1996) ecotoxicologic investigations of Ignalina NPP effluents demonstrate that these effluents are more or less dangerous to hydrobionts (Poveikio aplinkai... 2006).

An important problem was to determine the relationship between exposure dose rates of Lake Drūkšiai biota resulting from natural and technogenic radionuclides. One article (Nedveckaite *et al.* 2007) indicated that in Lake Drūkšiai, the dose rate of submerged hidrophytes resulting from technogenic discharged radionuclides is substantially lower as compared with the ionising radiation exposure of a natural background radionuclide (^{238}U, ^{226}Ra, ^{210}Po). It should be stressed that up to now, no such estimations were undertaken regarding other Lake Drūkšiai organisms.

As indicated in another article (Marčiulionienė 2007), during a short time period (2–4 days), radionuclides in freshwater ecosystems are equally distributed amongst water, bottom sediments and plants. This conditions a decrease of the radionuclide activity in water, which becomes an insufficiently informative component in the evaluation of the radioecological state. The long-term contamination of the freshwater ecosystem is the best reflected by bottom sediments, which become the radionuclide accumulation medium.

Mathematical models are developed for radionuclide distribution and evaluation of biota exposure regularities in freshwater ecosystems. They were especially widely applied for evaluation of radioecological consequences and in forecasting of biota contamination and radionuclide migration in ecosystems after the Chernobyl NPP accident (Kryshev 2008).

Although mathematical simulations of radionuclide transfer in different ecosystems were used for several decades, results of simulations have numerous uncertainties. For this reason, along with the deterministic exposure evaluation, close attention is paid to the exposure analysis using probabilistic methods. For the exposure evaluation, probabilistic methods were also recently applied in Lithuania (Nedveckaite *et al.* 2007, 2010; Nedveckaitė 2004).

Investigations based on radionuclide accumulation in hydrobionts and intended for the parameter evaluation of separate radionuclide transfer both in terrestrial (Butkus 2005, 2006, 2009; Butkus, Pliopaitė-Bataitienė 2006) and freshwater ecosystems should also be mentioned (Marčiulionienė *et al.* 1992; Čepanko *et al.* 2006). By combining probabilistic models and data obtained by radionuclide measurements, the most possible consequences of different decisions can be determined and various contamination scenarios can be verified. Probabilistic models enable forecasting the most probable future reality. The first step is to determine constraints of variable values (minimum and maximum). The second step is distribution of probabilistic leverages according to the maximum permissible dose rate value. In the European Union, the largest permissible exposure dose rate for biota is 10 μGy h^{-1} (ERICA Assessment 2007). This standard is currently recommended as the largest permissible radiation dose rate from exposure to technogenic radionuclides.

The aim of the work is to evaluate the exposure dose rate of reference biota of the Ignalina NPP cooling basis – Lake Drukšiai (within the range of ERICA computer code valid in the European Union) by applying probabilistic methods based on experimental data accumulated during 1989–2003 and taking into account ecotoxicologic investigation of Lake Drūkšiai water and bottom sediments.

2. Methods

Data on radionuclide activity concentrations in lake water and bottom sediments were chosen to evaluate the impact that radionuclides of natural and technogenic origin have on Lake Drūkšiai biota (Table 1).

ERICA computer code, which evaluates the most probable exposure dose rates of organisms by applying probabilistic methods, was used. This software that interacts with most of databases and modules referring to the experimental data of radionuclide activity concentration measurements in the environment, allows for evaluation of biota activity concentrations, and external, internal and total exposure dose rates.

The following dependences are most frequently used for estimation of internal and external absorbed dose rates:

$$D_{\text{int}}^{b} = \sum_{i} C_i^b * DCC_{\text{int},i}^{i}\,; \quad (1)$$

$$D_{ext}^{b} = \sum_{z} v_z \sum_{i} C_{zi}^{ref} * DCC_{ext,zi}^{b}\,; \quad (2)$$

$$D = \left[DCC_{\text{int}}^{FW} \cdot CR + 0.5 \cdot (1 + K_d^{DW} \cdot \rho_{sed}) \cdot DCC_{ext}^{DW} \right] \cdot C_{water}, \quad (\mu\text{Gy}\cdot\text{h}^{-1}), \quad (3)$$

where: D_{int}^{b} is the absorbed internal exposure dose rate of the *b*-th reference organism, μGy h^{-1};

C_i^b is the mean activity concentration of the *i-th* radionuclide in the *b*-th reference organism (Bq kg^{-1});

$DCC_{\text{int},i}^{b}$ is the dose conversion factor for the internal exposure defined as the ratio between the dose rate and the activity concentration of radionuclide *i* in organism *b* (μGy h^{-1})/(Bq kg^{-1});

D_{ext}^{b} is the external exposure dose rate in *b-th* reference organism, μGy h^{-1};

v_z is the time fraction when organism *b* is in habitats *z*;

C_{zi}^{ref} is the mean concentration of radionuclide *i* in the environment *z* (Bq kg^{-1} in case of bottom sediments or Bq l^{-1} in case of water);

Table 1. Radionuclide activity concentrations in Lake Drūkšiai bottom sediments (Bq/kg dry weight) and water (Bq/*l*)

Nuclide	Mean	S. D.	Apex	Distribution*	Range min	Range max	References
Natural radionuclides				**Sediments**			
^{238}U	30		30	TR	10	150	Šiluminė energetika 1989
^{226}Ra	48	38		LN	12	120	
^{210}Pb	48	38		LN	3	150	
^{210}Po	48	38		LN	12	120	
^{232}Th	50		50	TR	10	100	
^{40}K	504	206		LN	110	1860	
Technogenic radionuclides				**Sediments**			
^{54}Mn	14	28		LN	0.04	88	Paškauskas and Mažeika 1997; Marčiulionienė 2007
^{60}Co	46	45		LN	3.4	170	
^{90}Sr	28	13.6		LN	8.0	83	
^{134}Cs	0.7		0.7	TR	0.26	7.8	
^{137}Cs	150	110		LN	13	440	
^{239}Pu	0.9	1.2		LN	0.01	2.4	
				Water			
^{3}H	8	4.7		LN	0.05	20	Mažeika 2002
^{14}C	0.012	0.012		LN	0.008	0.014	

*LN – Lognormal

*TR – Triangular

$DCC^{j}_{ext,zi}$ is the dose conversion factor for the external exposure defined as the ratio between the dose rate and the activity concentration of radionuclide i in organism b.

ERICA software (ERICA 2007), in which the Monte Carlo probabilistic simulation uses available input data distributions, was chosen to evaluate the exposure of biota reference organisms. The result of such simulation is the probabilistic distribution of the dose rate that facilitates evaluation of the most and the least probable (but possible) distribution values. In this work, due to a certain radionuclide present in the environment of the freshwater system, the finite points of the probabilistic distribution were estimated using the following dose rate D (μGy h^{-1}) definition.

The value of the dose rate D not only depends on the reference organism species and the habitat of the freshwater ecosystem organism but also on the dose conversion factor (*DCC*), the ratio of concentrations *CR* (*equation 4*) and values of the distribution coefficient K_d (*equation 5*) (D-ERICA 2007). Values of these parameters were estimated as indicated in the FASSET (Fasset 2003) database (μGy h^{-1})/(Bq kg^{-1}) in case of bottom sediments or Bq l^{-1} in case of water.

$$CR\ (l\ kg^{-1}) = \frac{Activity\ concentration\ in\ biota\ whole\ body\ (Bq\ kg^{-1} fresh\ weight)}{Activity\ concentration\ of\ filtered\ water\ (Bq\ l^{-1})}; \tag{4}$$

$$K_d = \frac{Activity\ concentration\ in\ sediment\ (Bq\ kg^{-1} dry\ weight)}{Activity\ concentration\ in\ water\ (Bq\ l^{-1})}. \tag{5}$$

3. Results and discussion

Investigations performed in Lake Drūkšiai demonstrated that the largest total dose rate (internal and external) found in tested reference organisms results from natural radionuclide activity concentrations. The following hydrobionts undergo the largest exposure: crustacean, insect larvae, vascular plants, gastropod and bivalve molluscs (Fig. 1). Out of all natural radionuclides, the reference organisms mentioned above receive the largest total dose rate from the ionizing radiation impact of ^{238}U and its decay product ^{226}Ra (Fig. 2).

It can be noted that the total dose rate value of vascular plants compared with that of other reference organisms is the largest and reaches 4.18 μGy h^{-1} (Fig. 1); 98% of the dose rate is related to ionizing radiation of ^{238}U and its decay products (Fig. 2). Ordinarily, the exposure to natural radionuclides is not limited.

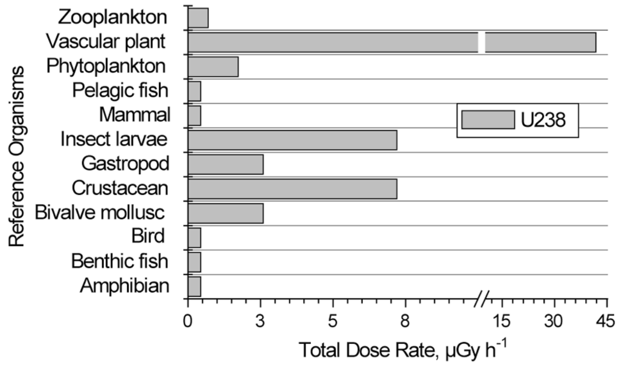

Fig. 1. The total exposure dose rate of reference organisms resulting from ^{238}U in Lake Drūkšiai

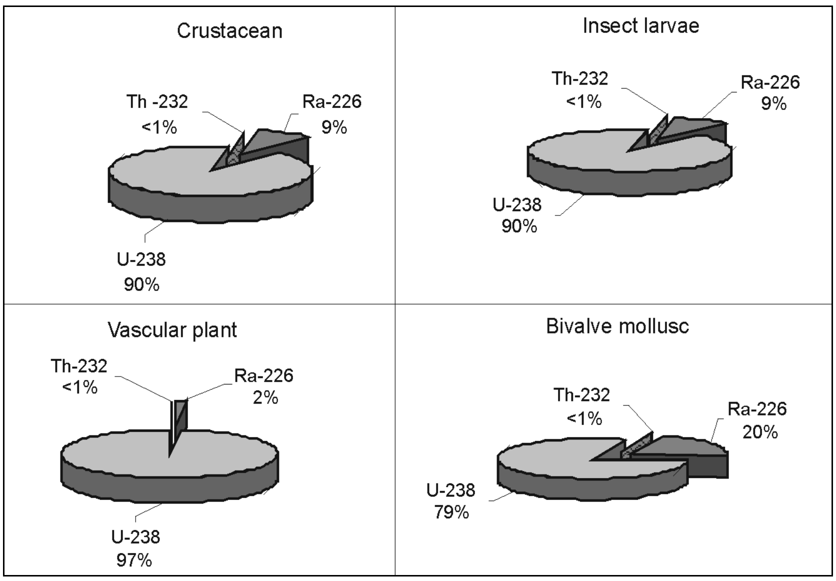

Fig. 2. The total exposure dose rate of reference organisms having received the largest exposure to natural radionuclides in Lake Drūkšiai

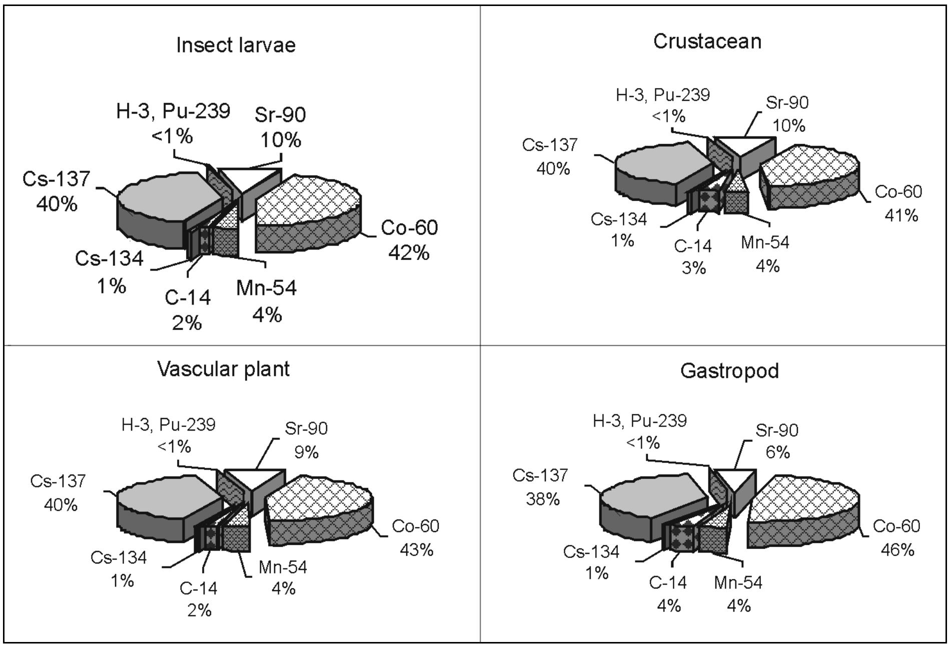

Fig. 3. The total exposure dose rate of reference organisms resulting from ionising radiation of technogenic radionuclides in Lake Drūkšiai

Out of all measured technogenic radionuclides (^{134,137}Cs, ^{90}Sr, ^{54}Mn, ^{60}Co, ^{239}Pu, ^{3}H, ^{14}C), the highest activity concentrations in Lake Drūkšiai were obtained in case of ^{137}Cs and ^{90}Sr. Insect larvae, crustacean, vascular plants, gastropod and bivalve molluscs undergo the largest exposure to technogenic radionuclides among all of the reference organisms (Fig. 3).

The obtained results – distributions and their statistical parameters – are presented in Figs 4 and 5. Based on the experimental data presented in Table 1, ERICA code, which applies probabilistic methods (Monte Carlo simulation), evaluates the distribution of total dose rate of reference organisms for each presented radionuclide; determines average, minimal and maximal values; and the 5th and the 95th order percentiles. The simulation results demonstrate that in Lake Drukšiai, the total dose rate value of reference organisms does not exceed the largest permissible value of 10 μGy h^{-1}, which is indicated in the EU recommendations for technogenic radionuclides. The distribution of the total dose rate (μGy h^{-1}) resulting from ionising radiation of ^{90}Sr, ^{137}Cs and ^{238}U estimated for vascular plants and insect larvae in Lake Drūkšiai is shown in Figs 4 and 5.

Various chemicals, acids and alkaline solutions, weak organic acids, heavy metals and dry materials that remain after artesian water vaporization at the Ignalina NPP, make their way to Lake Drūkšiai together with industrial rainwater sewerage (IRS) effluents and radionuclides. Although such wastes are attributed to comparatively low-toxicity effluents, their danger to living organisms was clearly determined. The laboratory investigation of the impact of IRS effluents on the radionuclide accumulation in water plants demonstrated that these effluents increase the ^{137}Cs accumulation in water plants (Marčiulionienė 2003). For evaluation of the radioecological state of Lake Drūkšiai, research data of 1988–2003

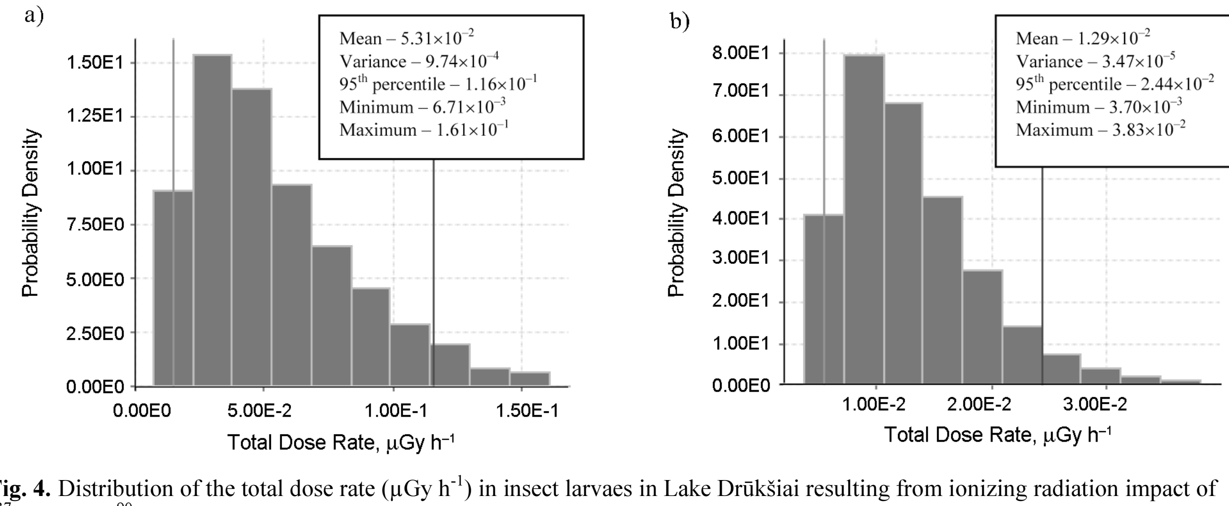

Fig. 4. Distribution of the total dose rate (μGy h^{-1}) in insect larvaes in Lake Drūkšiai resulting from ionizing radiation impact of ^{137}Cs (a) and ^{90}Sr (b)

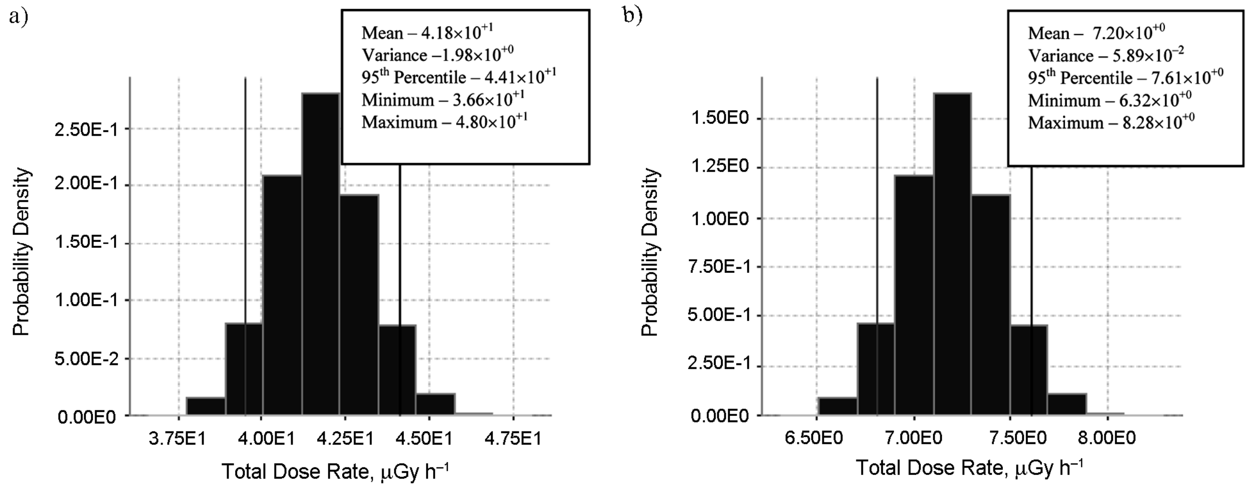

Fig. 5. Distribution of the total dose rate (μGy h^{-1})in vascular plants (a) and insect larvae (b) in Lake Drūkšiai resulting from ionizing radiation impact of ^{238}U and its decay products

were generalised: activity concentration values of ^{137}Cs and ^{90}Sr in plants and especially in bottom sediments of Lake Drūkšiai were higher than those in the Ignalina NPP effluent channels; whereas on the contrary, activity concentration values of ^{60}Co and ^{54}Mn were lower in Lake Drūkšiai than in the Ignalina NPP effluent channels (Marčiulionienė 2007).

In the distribution of ^{137}Cs in Lake Drūkšiai, where the activity concentration of this radionuclide is up to 56 times higher than in plants, bottom sediments play the main role. The largest accumulation of ^{90}Sr was determined in plants, where the activity concentration of this radionuclide was up to 3 times higher than in bottom sediments (Marčiulionienė 2007). Therefore, as compared with other technogenic radionuclides, the largest exposure dose in reference organisms was identified to result from ^{137}Cs and ^{90}Sr (Fig. 3). Out of the technogenic radionuclides, ^{90}Sr and ^{137}Cs belong among the most biologically toxic radionuclides as their relatively low concentrations can disorder the main functions of organisms (Čepanko *et al.* 2006).

Both the bottom sediments and plants, in which average values of activity concentrations of these radionuclides in most cases were similar, play an important role in the distribution of ^{60}Co and ^{54}Mn (Marčiulionienė 2007).

It should be noted that in the water ecosystem, uranium concentrates mostly in some plant species (Verkhovskaia *et al.* 1972). Accumulation of uranium by submerged plant species is 4–5 times larger as compared with air-water ecological plant species (Lavrova *et al.* 2003). In molluscs, uranium mostly concentrates in shells, while only small amounts are found in soft tissues. Since uranium is not a biogenic element, its accumulation in water plants under conditions of radioactive contamination can reach large values due to uranium adsorption by plant surface.

Water plants play an important role in regulation of water quality. In the case of water filtering by water plants, mechanical, biological and physical–chemical water cleaning takes place, i.e., organic, mineral and radioactive materials are absorbed onto stems and leaves of plants. These and radioactive elements, are accumulated in leaves and stems of water plants. Water plants, which have a large surface per weight unit, can absorb a large amount of radionuclides from the water environment (Marčiulionienė *et al.* 1992). Due to a larger water plant surface per weight unit, since vascular plants are also attributed to submerged plants, their exposure to ^{238}U and its decay products is the largest (Figs 1 and 2).

4. Conclusions

1. Referring to data of the radioecological research and ecotoxicologic investigations carried out in Lake Drūkšiai during 1989–2003, and with the help of probabilistic methods and ERICA computer code, it was determined that the dose rate values above the 95% confidence interval for reference organisms do not exceed the maximum permissible exposure dose rate level of 10 μGyh^{-1}, which is currently indicated in the EU recommendations for technogenic radionuclides.

2. In Lake Drūkšiai, the exposure dose rate received by reference organisms (vascular plants, insect larvae, crustacean, gastropod, bivalve molluscs and etc.) resulting from the impact of ionizing radiation of natural radionuclides is larger than that resulting from technogenic ones.

3. The largest exposure dose rates in reference organisms resulting from natural radionuclides were determined in the case of ^{238}U and its decay products (insect larvae – 7.2 μGy h^{-1}, vascular plants – 41.8 μGy h^{-1}). In the case of technogenic radionuclides, both ^{137}Cs (insect larvae – 5.31×10^{-2} μGy h^{-1}, vascular plants – 2.61×10^{-2} μGy h^{-1}) and ^{90}Sr (insect larvae – 1.29×10^{-2} μGy h^{-1}, vascular plants – 6.16×10^{-3} μGy h^{-1}) were the largest.

4. Out of all of the investigated reference organisms, the largest exposure dose rate was determined in benthic organisms (insect larvae, crustacean, gastropod and bivalve molluscs) and vascular plants. It should be noted that vascular plants undergo the largest exposure.

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DRŪKŠIŲ EŽERO BIOTOS TESTINIŲ ORGANIZMŲ APŠVITOS VERTINIMAS TAIKANT TIKIMYBINIUS METODUS

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Santrauka

Straipsnyje nagrinėjama Drūkšių ežero hidrobiontų (testinių organizmų) apšvitos dozės galia (μGy h^{-1}), kuriai vertinti taikant tikimybinius metodus, naudota įteisinta Europos Sąjungoje ERICA kompiuterinė programa. Skaičiavimams naudoti radionuklidų aktyvumo koncentracijų Drūkšių ežero dugno nuosėdose ir vandenyje 1989–2003 m. matavimų duomenys (vidutinės reikšmės, standartiniai nuokrypiai, mažiausios ir didžiausios reikšmės). Taikant tikimybinius metodus (Monte Karlo modeliavimą) kiekvienam testiniam organizmui ir radionuklidui įvertintas dozės galios pasiskirstymas, vidutinė, mažiausia ir didžiausia apšvitos vertės bei 5 ir 95 eilių procentilės. Nustatyta, kad technogeninių radionuklidų atveju Drūkšių ežero testinių organizmų apšvitos bendrosios dozės galios vertės (95 % pasikliautinumo dydis) yra mažesnės už šiuo metu technogeniniams radionuklidams Europos Sąjungoje rekomenduojamas didžiausias galimas 10 μGy h^{-1} dozės galios vertes. Testinių organizmų apšvitos dozės galia dėl gamtinių radionuklidų poveikio yra kur kas didesnė nei dėl technogeninių. Pažymėtina, kad gamtinių radionuklidų atveju didžiausia apšvitos dozės galia yra nulemta ^{238}U, pavyzdžiui, vabzdžių lervoms apšvita siekia 7,20×10^{0} μGy h^{-1}, induotiesiems vandens augalams – 4,18×10^{+1} μGy h^{-1}.

Reikšminiai žodžiai: gėlavandenė ekosistema, testiniai organizmai, apšvitos dozės galia, tikimybiniai metodai.

ОПРЕДЕЛЕНИЕ МОЩНОСТИ ДОЗЫ ОБЛУЧЕНИЯ РЕФЕРЕНТНЫХ ВИДОВ ОРГАНИЗМОВ ОЗЕРА ДРУКШЯЙ С ИСПОЛЬЗОВАНИЕМ ВЕРОЯТНОСТНЫХ МЕТОДОВ

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Резюме

С помощъю компьютерной программы ERICA и использованием вероятностных методов исследована мощность дозы облучения гидробионтов в озере Друкшяй. Для расчетов использованы измерения концентрации активности радионуклидов в воде и донных отложениях озера Друкшяй (среднее значение и стандартное отклонение; минимальные и максимальные значения) путём анализа литературных данных за 1989–2003 гг. С использованием вероятностных методов (метод Monte Carlo) подсчитаны распределения мощности дозы от каждого заданного радионуклида со средним, минимальным и максимальным значениями, рассчитаны 5-й и 95-й перцентили. Установлено, что в случае техногенных радионуклидов суммарная мощность дозы облучения гидробионтов (за пределами 95-го перцентиля) не превышает максимальной нормы облучения в 10 μGy h^{-1}, которая рекомендована Европейским Союзом как максимально возможная в случае техногенных радионуклидов. Мощность дозы облучения референтных видов организмов от природных радионуклидов значительно выше, чем от техногенных. Максимальная мощность дозы облучения в случае природных радионуклидов вызвана ^{238}U, например, 7,20×10 μGy h^{-1} для личинок насекомых, 4,18×10^{+1} μGy h^{-1} для водных сосудистых растений.

Ключевые слова: пресноводная экосистема, гидробионты, мощность дозы облучения, вероятностные методы.

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