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INVESTIGATION OF HEAVY METAL AND RADIONUCLIDE DISTRIBUTION IN SILT OF LAKE DIDŽIULIS

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Abstract. The article deals with research of the ecological situation in Lake Didžiulis, located nearby the highway Vilnius–Kaunas (Trakai District); the most polluted locations in the lake were identified. Research of heavy metals (HM) and radionuclides in the lake silt is described. To research HM concentrations and the specific activities of radionuclides, the samples were collected from 12 bores in the most typical locations of the lake. The estimated HM concentrations in the silt of Lake Didžiulis vary within a wide range: Cr from 6.9 to 45.2 mg/kg, Ni from 20.4 to 55.7 mg/kg, Mn from 236 to 1362 mg/kg, Cu from 7.1 to 80.9 mg/kg, Pb from 24.4 to 87.8 mg/kg, Zn from 13.0 to 75.8 mg/kg. The HM concentrations in the lake silt do not exceed the maximum allowed concentrations (MAC) in the soil, whereas the concentrations of manganese, copper and lead in some samples are close to MAC. The counted specific activities of ^{40}K fluctuate from 46 Bq/kg to 415 Bq/kg in the upper level silt and from 125 Bq/kg to 482 Bq/kg in the silt, which is found near the mineral soil. The specific activities of other radionuclides at the said depths of the silt range as follows: ^{226}Ra from 23 Bq/kg to 85 Bq/kg and from 28 Bq/kg to 93 Bq/kg, ^{227}Ac from 2.1 Bq/kg to17.0 Bq/kg and from 7.3 Bq/kg to 23.0 Bq/kg, whereas artificial ^{137}Cs from <0.6 Bq/kg to17.6 Bq/kg and from <0.5 Bq/kg to 12 Bq/kg.

Keywords: lake silt, Lake Didžiulis, heavy metals, radionuclides.

1. Introduction

Lakes are subject to decline due to the silt accumulating in them, the shores become overgrown with excessive vegetation, this causing a serious ecological problem – eutrophication takes place. Anthropogenic pollution contributes to rapid silting of lakes, even though this process also occurs naturally (Bakšienė, Ciūnys 2007; Ciūnys, Katkevičius 2008).

Poor ecological situation of lakes was predetermined by earlier easily entering untreated or insufficiently treated domestic wastewater, pollutants coming from livestock farms, arable fields and from atmosphere with precipitation (Ciūnys, Katkevičius 2004). Though at the present moment the anthropogenic and agrarian pollution of lakes has been decreased or suspended, its impact is being felt even now. Lentic ecosystems that have been polluted and which accumulated silt for many years are not able to self-purify from the accumulated contaminants. Therefore, one of the main measures for revival of silted lakes is partial or full removal accumulated silt. Findings of conducted studies make it possible to estimate the amount of the accumulated silt and its contamination as well as select technologies for its removal from the lake and opportunities of its further use or storage (Balevičius, Ciūnys 2006).

Elements referred to as heavy metals (HMs) are distinguished by their weight per 1 cm^3 exceeding 5 g. The list of HMs covers many elements, which are essential for living organisms, i.e. iron, manganese, copper, cobalt and etc. A part of them is called microelements. Their high concentrations, however, become hazardous to living organisms. Some 38 heavy metals occur in nature, but not all of them are regarded to be hazardous. Toxicity of an HM increases with the increment in its atomic weight (Mažvila 2001). HMs selected for studies are regarded to be hazardous to living organisms; therefore, it is important to determine their amount in the silt of the lake.

Both artificial and natural radionuclides occur on the Earth. Radioactive materials released to the atmosphere diffuse very quickly. The major part of radionuclides migrates in the land–water–flora–fauna system, thus entering lake sediments. It is very important that when using sapropel the amount of radionuclides in it would not exceed the background level of the environment (Butkus 2006).

According to the data identified by researchers, 0.15 to 4.4 cm of silt accumulates in the lake per year; thus, within 20 years it may constitute 3 to 88 cm (Ciūnys, Katkevičius 2008). According to this data, it is possible to estimate the environmental pollution caused in the corresponding year, since basing on the literature data, HM mobility is low deeper from the surface. In order to evaluate the pollution of the lake, it is important to select the sampling locations.

Research is aimed at determining concentrations of heavy metals (Cr, Cu, Mn, Ni, Zn, and Pb) and specific activities of natural (^{40}K, ^{226}Ra, ^{227}Ac) and artificial ^{137}Cs

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radionuclides in the silt of Lake Didžiulis and evaluating their distribution.

2. Methods for determination of heavy metals and radionuclides in the lake silt

Lake Didžiulis is located in south-eastern Lithuania, Trakai District, Lentvaris Eldership; its area amounts to 65.1 ha and the average depth is 6.5 m. The northern part of the lake is nearby the highway Vilnius–Kaunas–Klaipėda; and in the south-eastern part, Dėdeliškių village is located. In the southern part of the lake, Fabriko Stream flows in; and in the northern part, Malevankos Stream flows out (Fig. 1). In the past, the lake was polluted for several decades (until 2002) with Lentvaris township communal and industrial effluents from the Lentvaris Carpet Factory and other plants. At present, the pollution of Didžiulis Lake from Lentvaris has been terminated, but the effect of the past pollution is felt even now as regards the water quality and ecosystem of the lake. The diffuse pollution from the arable land located on the shores, farmsteads dispersed on the lake shore and highway has a certain effect on the ecological condition of the lake.

For identification of heavy metals and radionuclides, the silt samples were taken by a bog probe (a probe consists of a tip for silt-taking and bars of 0.8 m in length) from 12 bores made in different places (Fig. 1). Silt-taking sites were selected nearby Fabriko Stream – the lake inflow – and Melavankos Stream – the outflow; also at the western edge of the lake with a steep slope on its bank, and at the top of the slope the farms were formerly located and later – the car repairing workshops. Several bores were selected in the northern and north-eastern part of the lake, near the highway, and in the south-eastern part near the village of Dėdeliškių. Two samples were taken from one bore at a different depth (silt on the surface and near the mineral ground).

Concentrations of heavy metals in the sediments of Lake Didžiulis were determined by the method of absorption spectrometry. This method is based on element concentration measurements in a sample by using flame atomic absorption spectrometry.

Primarily, samples were dried at a temperature of 80–105 °C to eliminate humidity. The dried and granulated samples were burnt at a temperature of 450 °C for organic compounds to get burnt. Silt samples were burnt at a laboratory of the Vilnius Gediminas Technical University (VGTU). Then a sample of 0.5 g is weighed at an accuracy of 0.0001 g. The weighed sample is poured into a small plastic vessel; hydrogen peroxide H_2O_2 (2 ml) and concentrated 65% nitric acid HNO_3 (10 ml) is poured slowly. Mineralization is carried out in a microwave mineralizer Milestone Ethos. After mineralization, a vessel with a sample is cooled to 50–70 °C. Then the solution from the vessel is filtered through a glass filter into a 50 ml flask. After having filtered to the mark in the flask, it is dissolved with deionized water. Then it is measured with a metal concentration spectrometer Buck Scientific 210 VGP with the air–acetylene flame. All works for the preparation of a sample are carried out in the draught cabinet (Butkus *et al.* 2004).

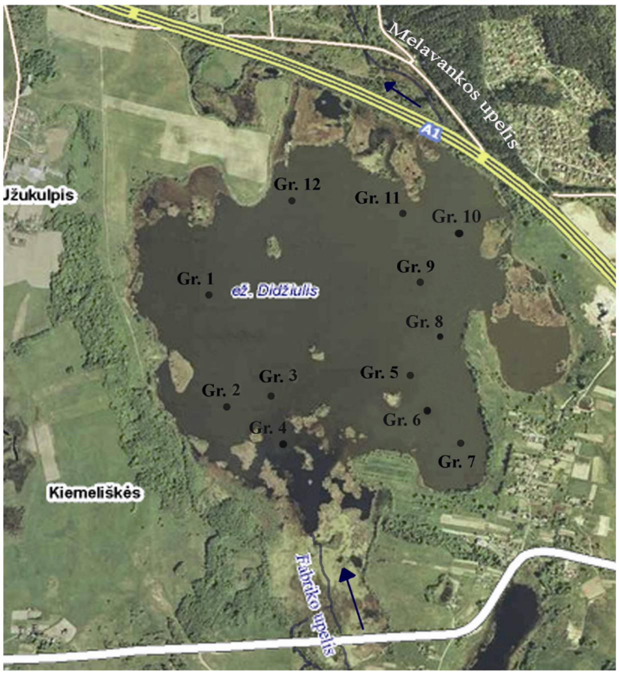

Fig. 1. Locality plan of Lake Didžiulis with bores

For producing a calibration curve, the comparative solutions without lanthanum and with lanthanum are used, as well as solutions of the used metals (Cr, Cu, Mn, Ni, Zn, Pb), prepared from standard metal solutions (approximately 1000 mg/*l* metal, 2% HNO_3).

HM concentration of the sample under study is calculated according to a formula:

$$W(M_e) = (C_{Me} \cdot f \cdot V)/W, \quad (1)$$

where $W(M_e)$ – metal concentration in a sample, mg/kg of dry weight; C_{Me} – metal concentration in a solution, mg/*l*; *f* – dilution factor; *V* – volume of solution, taken for analysis, *l*; W – sample weight, recalculated into dry sample weight, kg.

In the prepared lake silt sample HM concentrations were determined at the Laboratory of Environment and Workplaces Conditions of VGTU Institute of Environmental Protection according to the above presented methods.

Determination of radionuclides in sapropel samples is carried out by physical analysis methods, i.e. without the use of chemical reactions. The main methods are spectral and radiometric analyses. Spectral analysis is the determination of qualitative and quantitative composition of gaseous, liquid or solid substances when investigating the spectra of the object under analysis.

To determine the activity of radionuclides in samples, a semiconductor Ge (Li) spectrometer was used.

The efficiency of a semiconductor Ge (Li) gamma spectrometer used at the Department of Environmental Protection of Vilnius Gediminas Technical University for 662 keV energy is 0.26%, when a 52 ml cuvette is used (Butkus and Beinaravičius 2002; Butkus and Konstantinova 2005). The relation of background radiation peak area and measurement time for measured radionuclides is: ^{226}Ra – 0.0138; ^{214}Pb – 0.0605; ^{204}Pb – 0.0092; ^{208}Tl – 0.0053; ^{214}Bi – 0.0062; ^{137}Cs – 0.0029; ^{227}Ac (911 keV) – 0.0039; ^{227}Ac (969 keV) – 0.0018; ^{40}K – 0.0023.

The samples from the silt of Lake Didžiulis prepared for investigation (dried at a temperature of 80–105 °C) were measured in standard Denta cuvettes. Samples were measured for 24 hours. Weight of samples was approximately 35 g.

Radionuclides are identified according to the radiation energy, characteristic of each radionuclide: ^{214}Pb – 295 keV, 352 keV, ^{208}Tl – 583 keV, ^{214}Bi – 609 keV, ^{137}Cs – 662 keV, ^{227}Ac – 911 keV, 969 keV, ^{40}K – 1460 keV.

The specific activity of a radionuclide in a sample (Bq/kg) or volumetric activity in a sample (Bq/m^3) is calculated by identifying the radionuclide activity in a sample and sample weight or amount, according to a formula (Dimavičienė, Butkus 2007):

$$A = \frac{\frac{S}{t_1} - \frac{S_f}{t_f}}{\eta \cdot \varepsilon \cdot m}, \qquad (2)$$

where: S – radionuclide activity peak area in the measured substance, imp/s; S_f – background radiation peak area, imp/s; t – sample measurement time, s; ε – product of energy and geometric efficiencies of a semiconductor spectrometer; η – radionuclide decay quantum output; m – measured sample weight, kg.

The absolute error of specific activity shall be calculated according to a formula (Dimavičienė, Butkus 2007):

$$\Delta A = A\left(\frac{p}{100} + \frac{\Delta t}{t} + \frac{\Delta m}{m}\right), \qquad (3)$$

where: A – radionuclide specific activity in a sample, determined according to formula (2), Bq/kg; p – measurement relative error, determined by a spectrometer, %; Δt – measurement time error, s; Δm – sample weight determination error, kg.

3. Exploratory results of heavy metals in the lake silt and their analysis

Concentrations of heavy metals (Cr, Ni, Mn, Cu, Pb, Zn) were determined at different locations of the lake and silt depths.

HM concentrations determined in the surface layer of the lake silt (0.1–0.3 m from the silt top) and near the mineral ground (0.1–0.3 m from the mineral ground) are presented in Figs 2–3. A tendency is noticed that HM concentrations are higher in the deeper layer of the silt.

Figs 2–3 show that a rather significant difference between HM concentrations in the surface and deeper lake silt is in Bore 3, and this may occur due to the high thickness of the silt layer. Manganese, chromium and zinc concentrations are even twice as high in the near-bottom silt than in the surface silt.

While evaluating the surface silt research data, it is seen that the amount of manganese in different locations of the lake varies by up to 4 times, chromium by up to 6 times, nickel up to 2.5 times, copper even up to 9 times, lead up to 3 times, and zinc up to 5 times.

Exploratory results of heavy metals in the silt near the mineral ground also show that significant variation of concentrations exists in different locations of the lake.

Upon the evaluation of research data provided in Figs 2–3, it is possible to distinguish bores, which are studied in more detail (Bores 1, 4, 10 and 11). Those bores are made in the specific locations of the lake, i.e. where the highest and lowest afflux of pollution is probable. The distinguished Bore 1 is in the western part of the lake, where the former pollution with water run-off from the farms, later repairing workshops, located on the top of the slope, and arable lands has been possible. The second bore is not distinguished among the most specific bores,

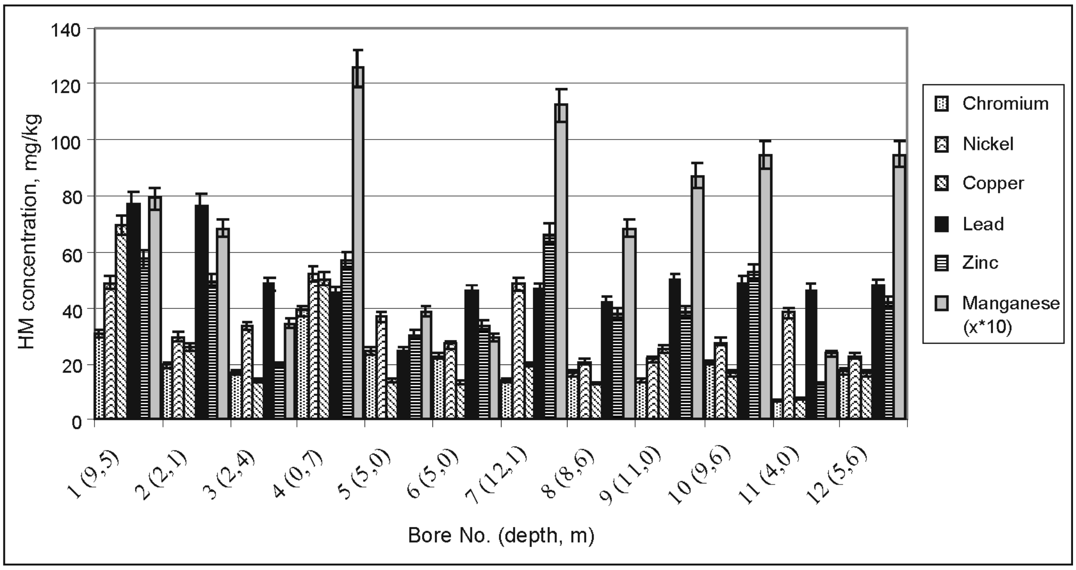

Fig. 2. Heavy metal concentrations in the surface silt of Lake Didžiulis

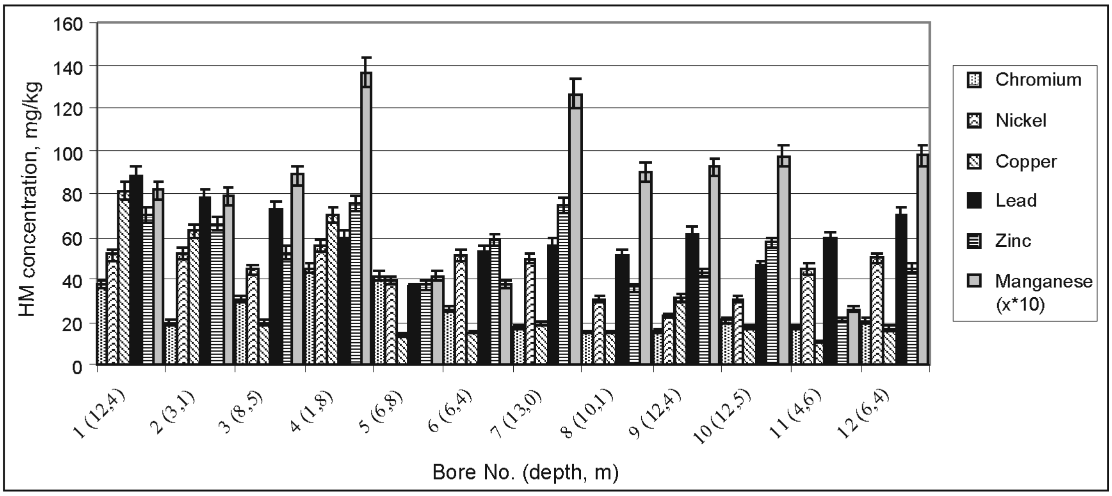

Fig. 3. Heavy metal concentrations in the silt of Lake Didžiulis near the mineral ground

since here concentrations of heavy metals in the silt at the surface and near the mineral ground are close, as well as Zn, Ni and Cu concentrations are close to the concentrations in the silt of the first bore, as they were influenced by the same sources as the first bore. Bore 4 was selected near the inflow (Fabriko Stream), which in the past brought many pollutants into the lake, and they entered with the effluents from the Lentvaris Carpet Factory. In the north-eastern part of the lake, Bores 10 and 11 are located nearby the highway Vilnius–Kaunas and the lake outflow (Melavankos Stream) (Fig. 1). According to the exploratory data of Bores 10 and 11, it is possible to estimate the pollution from the highway; however, the cleanest place of the lake is near the outflow, this being characteristic of lakes. The concentration values of heavy metals in these four bores are compared with the maximum allowable or background concentrations in the soil, as well as with the mean values identified for HM concentrations in the silt of Lake Didžiulis.

Fig. 4 presents the variation of chromium in the silt of the lake under study in the most specific locations of the lake.

The mean chromium concentration in the lake silt (22.9±4.3 mg/kg) is less than the background concentrations in soils (30.0 mg/kg); however, contamination with chromium is more significant in the western part of the lake (Bore 1) and near the inflow (Bore 4). Cr concentration in the deeper layers of those bores gets increased, and this could be affected by the former pollution entering with effluents the lake water and later also the silt. Cr concentrations, as provided in the foreign literature sources, fluctuate in the lake sediments from 14.3 to 110.0 mg/kg (Jha *et al.* 2002; Lee *et al.* 2007; Singh *et al.* 2007), i.e. up to 5 times higher than the established average concentration in the lake under study. Cr concentrations in the north-eastern part of the lake, at the lake outflow, are lower than the average chromium concentration in the explored lake and background concentration in the soil up to 4 times in the surface silt, and in the deeper existing silt up to 1.5 times. Thus Cr concentrations in the lake silt differ up to 6 times.

Fig. 5 presents the established nickel concentrations in the silt of the most typical locations of the lake.

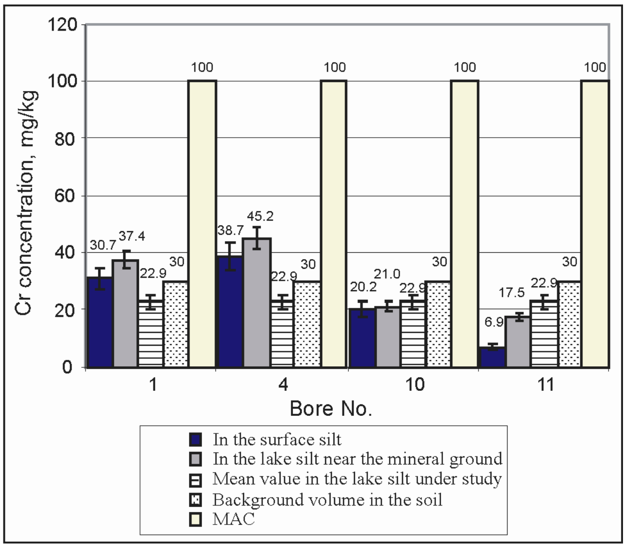

Fig. 4. Chromium concentration in the silt of the most specific locations of Lake Didžiulis

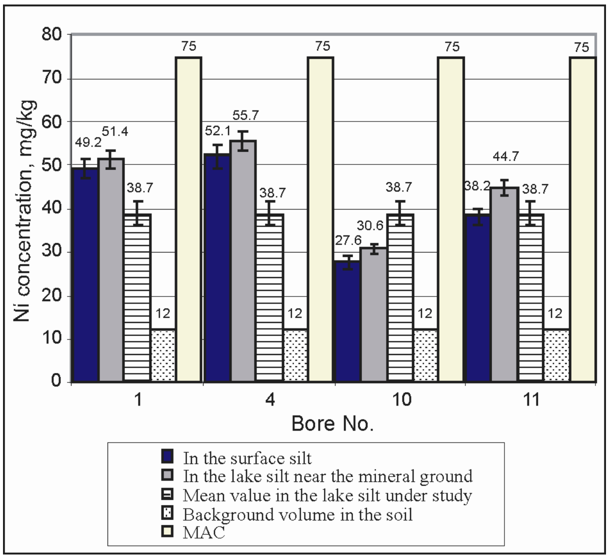

Fig. 5. Nickel concentration in the silt of the most specific locations of Lake Didžiulis

Nickel concentrations in the silt of the studied bores are just by up to 2 times lower than MAC and up to 5 times higher than the background concentration in soils. A tendency is observed that Ni concentrations in the deeper layers is higher, therefore, it is possible to consider that the former pollution is reflected in the lake silt. Taking into account that all studied lake silt samples contain considerably higher Ni amounts, as compared to the background volume in soils, this may show that soils accumulated nickel in lower amounts than the lake silt. Willing to use this silt for soil recultivation, it would be necessary to take into account the background volume and MAC of Ni in the soil, so that when mixing the fertilized soil with silt MAC would not be exceeded (75 mg/kg).

Ni concentrations in the lake silt in other countries, as provided in the literature, fluctuate from 17.7 to 49.0 mg/kg (Lee *et al.* 2007; Singh *et al.* 2007), they differ insignificantly from Ni concentrations established in the lake silt under study.

Fig. 6 shows the variation of manganese in the silt of the lake under study. Mn amounts, determined in the surface and near-bottom silt of the lake, are similar; Mn concentrations in the surface silt are up to 10% lower.

A significant increase in manganese concentration was observed near the inflow (Bore 4), which is very close to MAC (constituting about 90% of MAC). In Bores 1 and 10, manganese concentration is by 2 times higher than the background volume in the soil. The lowest manganese concentrations were determined in Bore 11; they by some 3 times are lower than the mean Mn concentration in the lake and even by some 6 times less than MAC. In the silt of the lake under study, the mean Mn concentration (771±52 mg/kg) is almost by two times higher than background concentration in soils (427 mg/kg); therefore before using the silt of this lake for soil improvement, it would be necessary to take into account that concentrations in the soil and lake silt would not exceed MAC. Mn concentrations in the lake silt of other countries, provided in the foreign literature, vary from 90 to 1370 mg/kg (Jha *et al.* 2002; Lee *et al.* 2007; Lepane *et al.* 2007; Singh *et al.* 2007).

The further considered copper variation in the lake silt under study is given in Fig. 7.

From the data provided in Fig. 7, it is seen that contamination with Cu is significant in the western part of the lake (Bore 1) and near the inflow (Bore 4). Copper concentration in those bores increases when going deeper and differs reciprocally by up to 30%. Cu concentrations, identified in the western part of the lake, are up to 4 times higher than the mean value in the silt of Lake Didžiulis and even up to 10 times higher than the background concentration in the soil. Such Cu concentrations could be predetermined by pollutants entering the lake with surface and underground wastewater. The untreated or not fully treated effluents discharged from the Carpet Factory into the Fabriko Stream could contain considerable Cu amounts, which formed during the carpet manufacturing process. Farms and repairing workshops that were located on the western slope in the past could also have had a negative effect on the contamination of the lake with HM. Cu concentrations in the north-eastern part of the lake are low, approximately by half less than the mean Cu concentration in the lake under study. Cu concentrations determined in the silt of the lakes outside Lithuania vary from 6.4 to 64.0 mg/kg (Lee *et al.* 2007; Lepane *et al.* 2007; Singh *et al.* 2007), i.e. up to 2 times higher than the established mean Cu concentration in Lake Didžiulis, but in Bore 1 the established Cu concentrations were higher than presented in the foreign literature.

Lead variation in the silt of the lake under study is given in Fig. 8.

The highest Pb concentrations were determined in Bore 1 both in the surface and deeper silt and they are close to MAC (making about 85% of MAC). Pb concentrations, found in Bore 1, are by 1.5 times higher than the mean concentration in the explored lake. In all samples studied, Pb concentrations are about 4 times higher than

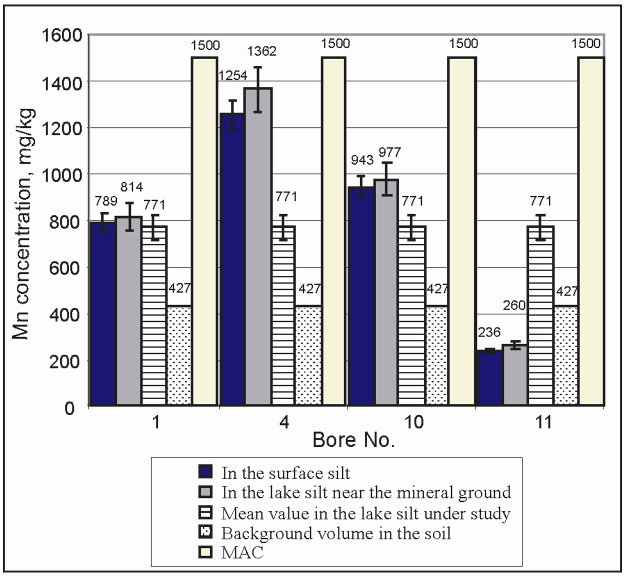

Fig. 6. Manganese concentration in the silt of the most specific locations of Lake Didžiulis

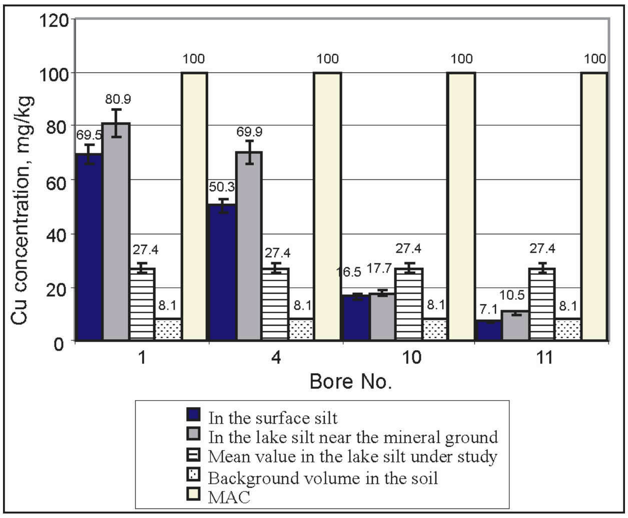

Fig. 7. Copper concentration in the silt of the most specific locations of Lake Didžiulis

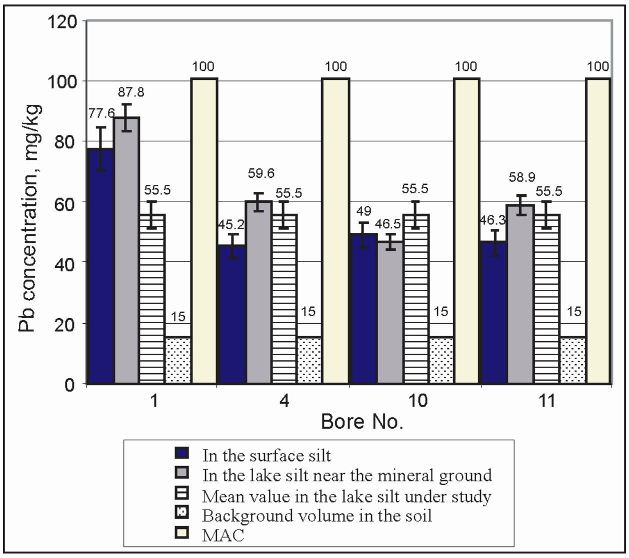

Fig. 8. Lead concentration in the silt of the most specific locations of Lake Didžiulis

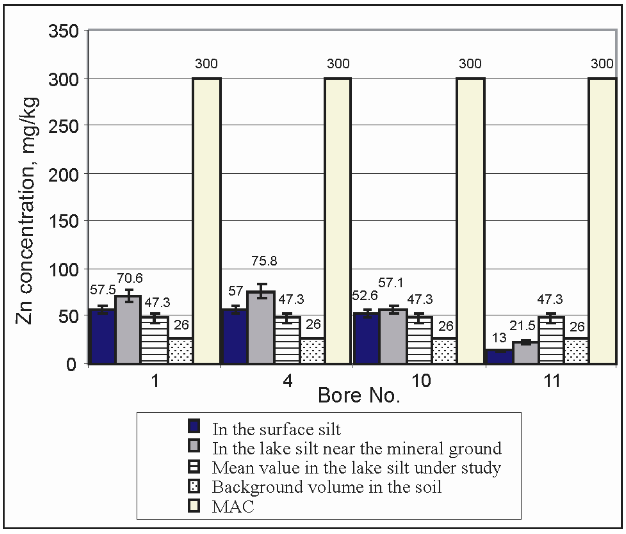

Fig. 9. Zinc concentration in the silt of the most specific locations of Lake Didžiulis

the background concentration in the soil. Pb amounts determined in the lake silt that were found in the foreign literature range from 18.0 to 167.4 mg/kg (Lee *et al.* 2007; Lepane *et al.* 2007; Singh *et al.* 2007), i.e. Pb concentrations up to 3 times lower and higher than the mean established in the silt of the lake under study as compared to the lowest and highest Pb amount, given in the literature. Pb could enter the lake with surface wastewater from the highway Vilnius–Kaunas; the same Pb concentrations could also be predetermined by Pb amounts entering with wastewater from motor repairing workshops, which were located on the lake slope, and arable lands. As known, considerable Pb amounts were previously released from transport, and this could have an effect on the higher Pb amounts prevailing in deeper layers. Also significant Pb amounts could enter with the water of the stream, into which untreated, later insufficiently treated domestic wastewater of the township of Lentvaris and industrial effluents from the carpet factory were discharged. However, evaluating the fact that lead amounts both in the surface and near-bottom silt are much higher than the background amount in the soil, it may be considered that lead is prone to accumulate in the lake silt.

Fig. 9 presents variation in zinc in the silt of the lake under study.

Zinc concentrations as compared to MAC are low (in some places lower up to 20 times); however, in the western part, near the lake inflow and in one bore in the eastern part, Zn concentrations are higher than the mean concentration in the lake and established in Bore 11. In the silt of Bore 10, located close to the outflow, established higher Zn amount might have been predetermined by pollution released from the highway Vilnius–Kaunas. The higher Zn concentrations in the deeper layers in the first bore and near the inflow could have been affected by the pollution in the past. Since Zn, like other explored HMs, established concentrations are higher than the background concentration in the soil (in this case around 3 times), it is possible to assume that HMs accumulate in the lake silt more considerably than in the soil. Zn concentrations in the silt of the lakes of other countries as presented in the literature, range from 23 to 190 mg/kg (Jha *et al.* 2002; Lee *et al.* 2007; Lepane *et al.* 2007; Singh *et al.* 2007), i.e. up to 4 times higher than the established mean Zn concentration in the silt of Lake Didžiulis.

Upon generalization of obtained results we see that concentrations of heavy metals do not exceed MAC in the soil. However, in certain silt samples from Lake Didžiulis, Mn, Cu and Pb concentrations are close to MAC, therefore prior to using the silt of that lake, the background HM concentrations should be estimated in the soil, onto which the silt woul be spread. HM concentrations in the silt of the lake under study are lower than the highest HM concentrations presented in the analyzed foreign literature. A tendency is also noticed that HM concentrations in the deeper layers are higher. It is possible to assume that this was owing to the pollution in the past, but knowing that the silt near the mineral ground has accumulated long ago when industrial and bigger pollution sources were not yet in existence, therefore, a presumption cannot be denied that silt mixing and possible migration deeper from the silt surface occurs due to water turbulence. From the results presented it is also seen that concentrations of heavy metals identified in the majority of samples are higher than the background concentration in the soil.

4. Radionuclide research results and their analysis

The specific activities of natural (^{40}K, ^{226}Ra, ^{227}Ac) and artificial ^{137}Cs radionuclides are identified in the surface silt and the silt near the mineral ground in the different locations of the lake (Figs 10–13).

Fig. 10 presents a variation in the specific activity of ^{40}K in the silt of Lake Didžiulis. ^{40}K specific activity is within the range of 46 Bq/kg to 415 Bq/kg in the surface silt and from 125 Bq/kg to 482 Bq/kg in the silt near the mineral ground.

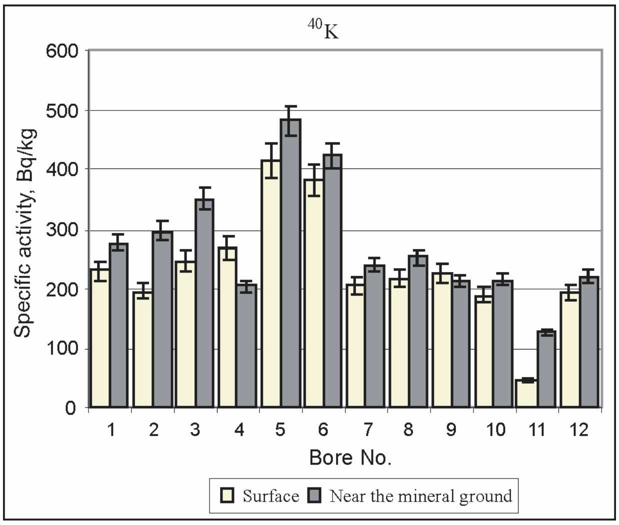

Fig. 10. ^{40}K specific activity in the silt of Lake Didžiulis

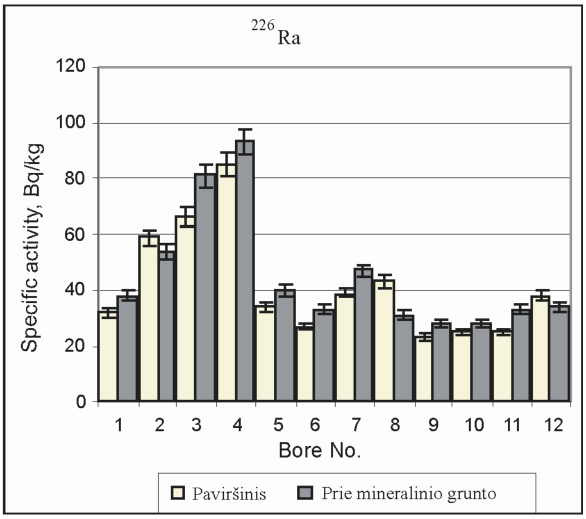

Fig. 11. ^{226}Ra specific activity in the silt of Lake Didžiulis

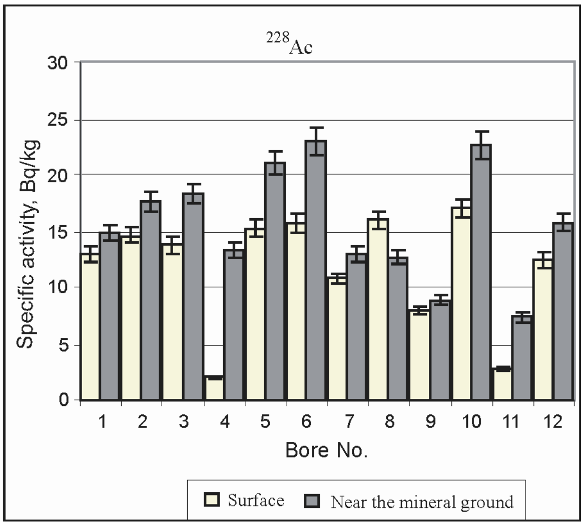

Fig. 12. ^{227}Ac specific activity in the silt of Lake Didžiulis

It is seen from the Figure that the highest specific activity was established in Bores 5 and 6. Such accumulation of potassium in those bores could be affected by the lake bottom relief and water turbulence. The mean ^{40}K specific activity in the lake silt equals 255±31 Bq/kg. In Bores 1, 2 and 3, a bigger difference is observed between the ^{40}K specific activity measured in the surface and deeper existing silt. This radionuclide could enter the lake due to the fertilization of arable lands with potassium fertilizers and farms around the lake, especially in the former years. ^{40}K specific activities provided in the analyzed foreign literature vary in the range from 221 to 326 Bq/kg (Khaten *et al.* 2005) and are close to the determined ^{40}K specific activities in the silt of Lake Didžiulis.

Fig. 11 presents a variation in ^{226}Ra specific activities in the silt of Lake Didžiulis. The specific activity of ^{226}Ra is within the range of 23 Bq/kg to 85 Bq/kg in the surface silt and from 28 Bq/kg to 93 Bq/kg in the silt near the mineral ground. ^{226}Ra specific activity found in the lakes of Lithuania fluctuates from 14 to 60 Bq/kg (Begy *et al.* 2009; Khaten *et al.* 2005).

When analyzing data provided in Fig. 11, it becomes clear that the highest ^{226}Ra specific activity was determined in Bores 3 and 4. Such increment in ^{226}Ra specific activity in the above bores may show that in the past this radionuclide would have been entering with untreated or insufficiently treated effluents from the industrial sector through the factory stream. The mean ^{226}Ra specific activity in the silt of the lake under study equals 43±16 Bq/kg. It is noted that ^{226}Ra specific activity in the silt existing deeper in Bores 2, 8 and 12 is lower, which is different from other bores. Such unevenness could have resulted from water mixing, which occurs in the lake and may also affect the silt.

Variation in ^{227}Ac specific activity in the silt of Lake Didžiulis is provided in Fig. 12. ^{227}Ac specific activity ranges from 2.1 Bq/kg to 17.0 Bq/kg in the surface silt and from 7.3 Bq/kg to 23.0 Bq/kg near the mineral ground existing in the silt.

It is seen from Fig. 12 that considerable differences exist between ^{227}Ac specific activities measured in the surface silt and the deeper existing silt. In the deeper existing silt, especially near the inflow (Bore 4), ^{227}Ac specific activity increases. The highest ^{227}Ac specific activity is measured in Bores 6 and 10, and this could be affected by the lake bottom relief and water turbulence. The mean ^{227}Ac specific activity in the silt of the lake under study equals 14±3 Bq/kg. Even though ^{227}Ac specific activity in the silt near the mineral ground (13,3 Bq/kg) is less than the mean, however, due to such difference with the activity determined in the surface silt (2.1 Bq/kg), it is possible to consider that in the past, the considerable amounts of pollutants were brought to the lake with the inflow.

A variation in the artificial radionuclide ^{137}Cs specific activity in the silt of Lake Didžiulis is presented in Fig. 13. The values of ^{137}Cs specific activity fluctuated from <0.6 Bq/kg to 17.6 Bq/kg in the surface silt and from <0.5 Bq/kg to 12 Bq/kg in the silt near the mineral ground. The ^{137}Cs specific activities in the silt of the lakes,

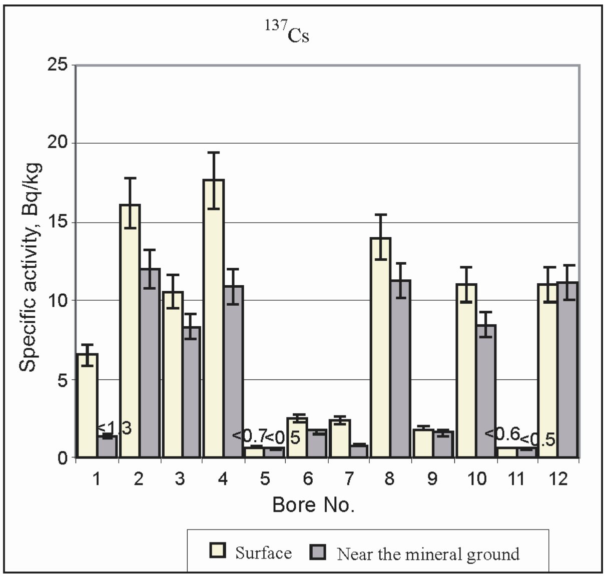

Fig. 13. ^{137}Cs specific activity in the silt of Lake Didžiulis

presented in the foreign literature, range from 2.3 to 12.0 Bq/kg (Arnaud *et al.* 2006; Khaten *et al.* 2005; Nomade *et al*. 2005), i.e. close to the determined ^{137}Cs specific activities in the silt of the lake under study.

When studying the distribution of the specific activity of artificial ^{137}Cs radionuclide in the lake silt, it is possible to notice that differently than the measured natural radionuclides, the specific activity of ^{137}Cs is higher in the surface silt, and deeper it reduces. The highest ^{137}Cs specific activity was measured in Bores 2 and 4, the mean ^{227}Ac specific activity in the silt of the lake under study equals 7±2 Bq/kg. Evaluating the fact that the speed of silt formation in the lakes of different sizes and in different locations of the lake is very uneaven, it is possible to assume that the silt, taken from the surface (0.1–0.3 m), could have been contaminated with ^{137}Cs due to the Chernobyl NPP accident. However, not possessing the silt accumulation data of this lake, it is difficult to estimate it precisely.

Upon generalization of the obtained results relating to the research of radionuclide specific activities, we see that natural radionuclides in the lake are distributed in the lake unevenly, and the highest activities are not determined in the locations of the lake with the maximum possible contamination. Comparing the specific activities of ^{40}K, ^{226}Ra and ^{137}Cs radionuclides, given in the foreign literature under study, we see that their values are close to those established in the silt of Lake Didžiulis. However, the highest specific activities of artificial ^{137}Cs radionuclides were established in the southern part of the lake, and in all places of the surface silt of the lake under study ^{137}Cs specific activity identified is higher. According to the data, provided in the literature, before the Chernobyl NPP accident (1981–1983) the ^{137}Cs contamination in the locality of the lake under study did not exceed 600 Bq/m^2, and after the accident in question (1988) 2000–4000 Bq/m^2 (Data… 1998; Izrael *et al.* 1997). This shows that in this locality, the predominant contamination with ^{137}Cs emerged after the Chernobyl NPP accident.

5. Conclusions

1. The established HM concentrations in the silt of Lake Didžiulis varied within a rather wide range: Cr – from 6.9 to 45.2 mg/kg, Ni – from 20.4 to 55.7 mg/kg, Mn – from 236 to 1.362 mg/kg, Cu – from 7.1 to 80.9 mg/kg, Pb – from 24.4 to 87.8 mg/kg, and Zn – from 13.0 to 75.8 mg/kg.
2. In certain cases, HM concentrations determined in the lake silt even make about 90% of MAC, however, do not exceed them, and in some locations – up to 10 times higher than the background amount in the soil; therefore, before using the silt of this lake it would be necessary to evaluate the background HM concentrations in the soil, onto which the silt would be spread.
3. Upon estimation of HM concentrations, established in the surface and deeper layers, it is seen that in deeper layers they are higher by up to 2 times.
4. The determined ^{40}K specific activities fluctuate from 46 Bq/kg to 415 Bq/kg in the surface silt and from 125 Bq/kg to 482 Bq/kg in the silt near the mineral ground. Radionuclide specific activities at the said silt depths vary as follows: ^{226}Ra – from 23 Bq/kg to 85 Bq/kg and from 28 Bq/kg to 93 Bq/kg; ^{227}Ac – from 2.1 Bq/kg to 17.0 Bq/kg and from 7.3 Bq/kg to 23.0 Bq/kg; and of artificial radionuclides ^{137}Cs – from <0.6 Bq/kg to17.6 Bq/kg and from <0.5 Bq/kg to 12 Bq/kg.
5. The mean specific activity of artificial ^{137}Cs radionuclide is 7±2 Bq/kg, but the specific activity is up to 5 times higher in the surface silt than in the silt near the mineral ground, this showing that the prevailing ^{137}Cs contamination in this locality occurred after the Chernobyl NPP accident.

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SUNKIŲJŲ METALŲ IR RADIONUKLIDŲ PASISKIRSTYMO DIDŽIULIO EŽERO DUMBLE TYRIMAS

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Santrauka

Straipsnyje nagrinėjama Didžiulio ežero šalia greitkelio Vilnius – Kaunas (Trakų raj.) ekologinė būklė, nustatytos ežero vietos, kur buvusi didžiausia tarša. Aptariami atlikti sunkiųjų metalų (SM) bei radionuklidų koncentracijų ežero dumble tyrimai. SM koncentracijoms ir radionuklidų savitiesiems aktyvumams tirti ėminiai buvo imti iš 12 gręžinių, parinktų būdingiausiose ežero vietose. Didžiulio ežero dumble nustatytas gana platus SM koncentracijų intervalas: Cr – 6,9–45,2 mg/kg, Ni – 20,4–55,7 mg/kg, Mn – 236–1362 mg/kg, Cu – 7,1–80,9 mg/kg, Pb – 24,4–87,8 mg/kg, Zn – 13,0–75,8 mg/kg. SM koncentracijos ežero dumble neviršija didžiausių leidžiamųjų koncentracijų (DLK) dirvožemyje, tačiau kai kuriuose bandiniuose mangano, vario ir švino koncentracijos yra artimos DLK. Nustatytieji ^{40}K savitieji aktyvumai svyruoja nuo 46 Bq/kg iki 415 Bq/kg paviršiniame dumble ir nuo 125 Bq/kg iki 482 Bq/kg esančiame arti mineralinio grunto. Kitų radionuklidų savitieji aktyvumai tuose dumblo gyliuose svyruoja taip: ^{226}Ra – nuo 23 Bq/kg iki 85 Bq/kg ir nuo 28 Bq/kg iki 93 Bq/kg, ^{227}Ac – nuo 2,1 Bq/kg iki 17,0 Bq/kg ir nuo 7,3 Bq/kg iki 23,0 Bq/kg, o dirbtinės kilmės ^{137}Cs – nuo <0,6 Bq/kg iki 17,6 Bq/kg ir nuo <0,5 Bq/kg iki 12 Bq/kg.

Reikšminiai žodžiai: ežero dumblas, Didžiulio ežeras, sunkieji metalai, radionuklidai.

ИССЛЕДОВАНИЕ РАСПРЕДЕЛЕНИЯ ТЯЖЕЛЫХ МЕТАЛЛОВ И РАДИОНУКЛИДОВ В ИЛЕ ОЗЕРА «ДИДЖУЛИС»

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Резюме

Анализируется экологическое состояние озера «Диджулис», которое находится возле магистральной дороги Вильнюс–Каунас (Тракайский район), идентифицируются места с наибольшим загрязнением, анализируются результаты исследований тяжелых металлов (ТМ) и радионуклидов в иле. Для исследования концентраций ТМ и специфических активностей радионуклидов пробы ила были взяты из 12 скважин, подобранных в характерных местах озера. Концентрации ТМ в иле озера «Диджулис» менялись в довольно широких интервалах: Cr – от 6,9 мг/кг до 45,2 мг/кг, Ni – от 20,4 мг/кг до 55,7 мг/кг, Mn – от 236 мг/кг до 1362 мг/кг, Cu –от 7,1 мг/кг до 80,9 мг/кг, Pb – от 24,4 мг/кг до 87,8 мг/кг, Zn – от 13,0 мг/кг до 75,8 мг/кг. Концентрации ТМ, находящихся в иле озера, не превышали предельно допустимых концентраций (ПДК) в почве, однако в некоторых пробах концентрации марганца, меди и свинца были близки к ПДК. Специфическая активность ^{40}K меняется от 46 Бк/кг до 415 Бк/кг на поверхности слоя ила и от 125 Бк/кг до 482 Бк/кг вблизи минерального грунта. Специфические активности других радионуклидов на определенных глубинах ила меняются таким образом: ^{226}Ra – от 23 Бк/кг до 85 Бк/кг и от 28 Бк/кг до 93 Бк/кг, ^{227}Ac– от 2,1 Бк/кг до 17,0 Бк/кг и от 7,3 Бк/кг до 23,0 Бк/кг, а радионуклидов искусственного происхождения: ^{137}Cs – от <0,6 Бк/кг до 17,6 Бк/кг и от < 0,5 Бк/кг до 12 Бк/кг.

Ключевые слова: ил озера, озеро «Диджулис», тяжелые металлы, радионуклиды.

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