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## Simulation of the shielding properties of environmental components against external radioactive radiation from the Ordovician *Dictyonema* shale

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This paper presents the results of simulating the conditions of absorption of natural radionuclides (uranium-238, thorium-232, potassium-40) in different environment media (water and quartz sand) to assess the effect of the shielding properties of environmental components on the intensity of external radiation exposure to the benthic biota. The source of radiation was the lower Tremadocian *Dictyonema* shale, which formed the bottom of the Baltic Paleobasin. Water and quartz sand, which was gradually deposited on the roof of the *Dictyonema* shale, were used in the simulation as media (materials) absorbing the primary radiation. The manuscript shows an experiment conducted in full-scale conditions considering a single exposure of a section of Ordovician rocks that includes highly radioactive *Dictyonema* shale. This is a small area, but the simulation results are also valid for larger areas as the Baltic Paleobasin. During the late Tremadocian — Floian, along with transgression, benthic biota began to emerge. The experiment shows quantitatively what dosage this biota could have experienced over tens and hundreds of thousands of years, until the *Dictyonema* shale was covered with a sufficiently thick layer of water-flooded sediments. Experimental data show that the presence of a 10 cm thick layer of water reaches absorption of 40 % of the integral flux of all gamma lines of natural radionuclides (NRN), and in a moist quartz sand layer of the same thickness the absorption value does not exceed 50 %. Thus, despite the screening effect of the environment, the benthic biota of Baltoscandia in the Early Ordovician could have been under significant radiation exposure for millennia. The main value of the experiment is that it was performed in situ, and exclusively natural media were used as modelling elements.

**Keywords:** *Dictyonema* shale, natural radionuclides, gamma radiation, absorption coefficient, radiation shielding properties, benthic biota.

## 1. Introduction

Geological formations with a high content of radioactive elements determine the ecological situation and potential radiological risks in particular zones. Radioactivity of rocks and soils is determined by the content of natural radionuclides (NRN). Among the main NRN common in the natural environment are  $^{40}\text{K}$ ,  $^{238}\text{U}$  and  $^{232}\text{Th}$ . The ways in which radioactive geological bodies have impacted the environment have been different over different periods of geological (historical) time. As a rule of thumb, during the modern geological section, highly radioactive bodies are covered with a multimeter layer of non-radioactive rocks (e. g., clays, sandstones, limestone, loose Quaternary sediments). The outputs of highly radioactive rocks on the surface are observed only in isolated outcrops and do not extend to any significant distances. An example is graptolite argillite (*Dictyonema* shale), well known in Estonia and Russia. It was formed during the Early Tremadocian on the territory of the Baltic Paleobasin (Männil, 1966; Soesoo and Hade, 2012). Nowadays, outcrops of *Dictyonema* shale can be found in the eastern part of the Baltic Klint (Ingrian Klint) (Lebedev et al., 2018).

The situation is different when considering the formation of radioecological locations at the geological time that sedimentation processes were taking place. Marine animals and plants could exist in the immediate vicinity of the roof of radioactive rocks, covered with an insignificant layer of seawater and unconsolidated sediments (sand, silt). In this situation, radioactive rocks at the bottom of the basin create a dose load on the organisms therein contained, but water and non-radioactive sediments gradually accumulated, reducing the received dosage until it was completely neutralized.

Our research describes an experiment conducted in full-scale conditions considering a single exposure of a section of Ordovician rocks that includes highly radioactive *Dictyonema* shale (Harrell et al., 1991; Saito and Jacob, 1995; Lee et al., 1998). This is a small area, but the simulation results are also valid for larger areas of land, which in the Ordovician Period represent a kind of “nuclear plate” similar in size to the Baltic Paleobasin. Along with the new ocean, benthic biota began to emerge. The experiment shows quantitatively what dosage this biota could have experienced over tens and hundreds of thousands of years, until the *Dictyonema* shale were covered with a sufficiently thick layer of water-flooded sediments (Ogendi et al., 2004; 2007).

This paper is devoted to modelling the absorption coefficient of radiation of natural radionuclides in different environments (water, quartz sand) in order to assess the effect of the shielding properties of environmental components on the intensity of external radioactive exposure for the benthic biota. The main contribution of the experiment is that it was performed in situ, and exclusively natural elements were used as components of the model.

## 2. Material and Methods

Calculation of the attenuation coefficient for a rather weak absorber — water — is not an easy task. But what about precipitation gradually accumulating at the bottom of the basin? It is rather problematic to calculate the coefficient of weakening of unconsolidated sediment, for example moist sand, because the input parameters are unknown. The density of quartz sand with water cannot be found in document directories because of the

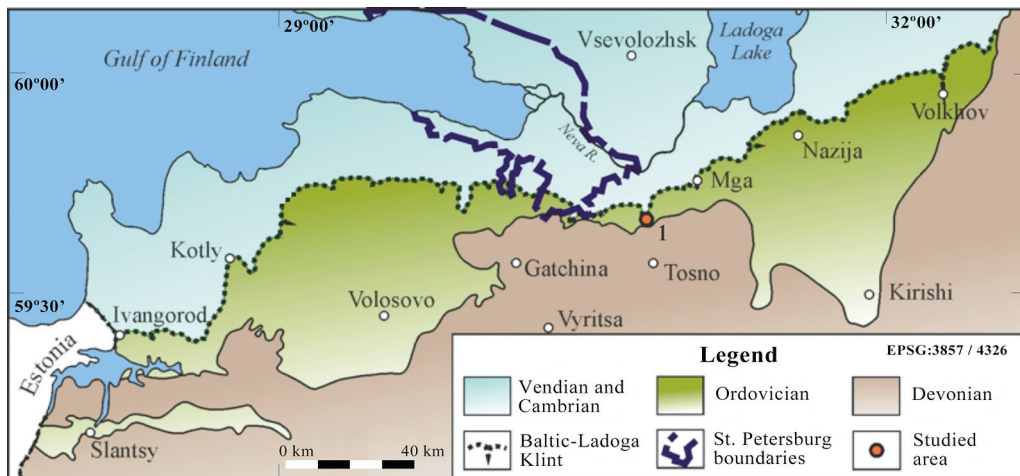


Fig. 1. Location of investigated outcrop of bituminous mudstones in the lower reaches of the Tosna River. Modified after Sokolova (1971)

high variability of such a value. For calculations, it would also be advisable to consider the presence of natural radioactivity in the overlapping sediments. As a practical example of such conditions, it is worth mentioning the presence of NRN in the glauconitic sands of Leetse formation covering argillites with a layer of up to 5 cm thick.

The tasks performed in this research included an experimental evaluation of the effect of shielding properties of environmental components. We determined the absorption coefficient of the primary radiation of natural radionuclides (uranium, thorium, potassium) contained in the *Dictyonema* shale. Water and sand were used as the environmental media absorbing the primary radiation, with which we covered the roof of the *Dictyonema* shale.

The outcrop of Ordovician deposit, in which measurements of the absorption coefficient of NRN radiation were made, is located about 40 km southeast of St. Petersburg near the settlement Ulyanovka in a small ravine on the left side of the Tosna River valley, 80 m north-northeast from the extreme northern point of the of St. Petersburg State University field camp (Fig. 1).

On the slope of the ravine the slope deposits were removed, as well as limestone and sandstone of the Leetse formation overlapping the shales. As a result, the shale roof was exposed in a section of about 90 × 95 cm (Fig. 2). Exposure dose measurements were performed with a SRP-68-01 radiometer (Electron, Ukraine), which fixed the integrated gamma radiation flux to within 1 μR/h.

Five measurements were made directly on the roof of the shale, one at the center of the cleared area and four measurements at a distance of 30 cm in different directions from the center. This was done in consideration of the potential anisotropy of the phenomenon. Orthogonal to the selected five points, new vertical measurements were made at a distance of 1, 3, 5, 7 and 10 cm from the surface of the roof. Later, a rectangular bath (small rectangular pool) with internal dimensions of 10 × 35 × 70 cm was constructed on the surface of the shale roof from limestone blocks connected by clay (Fig. 3). The self-activity of the bath's building material — limestone and clay — does not exceed 16 μR/h (Lebedev, 2012).

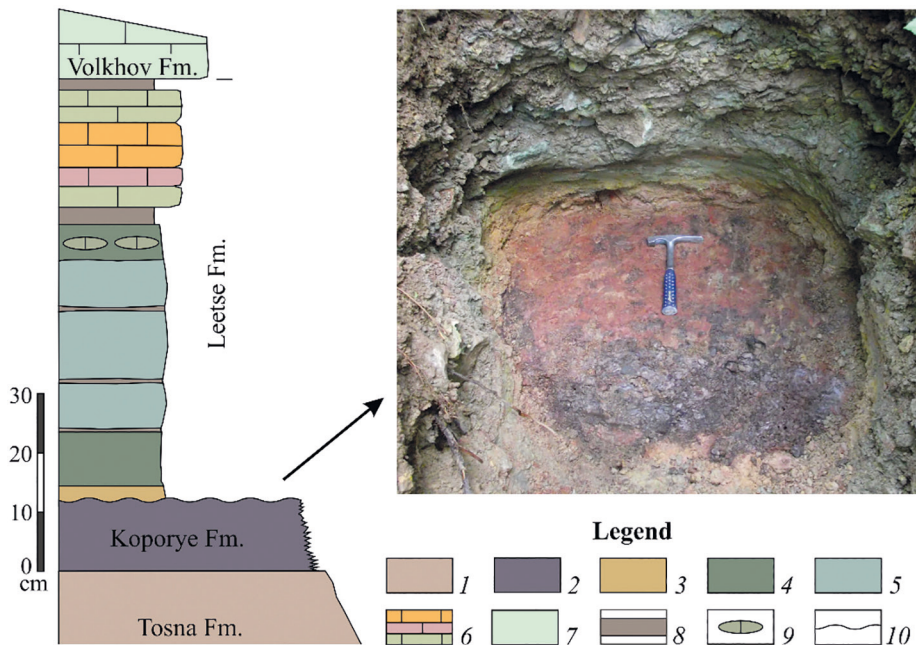


Fig. 2. Studied section on Tosna River (left) and the exposed upper boundary of the *Dictyonema* shale (right): 1 — *Obolus* sandstone; 2 — *Dictyonema* shale; 3 — basal quartz sandstone; 4 — weakly cemented glauconite sandstone; 5 — cemented glauconite sandstone; 6 — clayey glauconite limestone, variegated in colour; 7 — hard, bioclastic glauconite limestone; 8 — clay interbeds; 9 — limestone nodules; 10 — discontinuity surface



Fig. 3. Small pool on the roof surface of the *Dictyonema* shale filled with pink quartz sand

Then, we gradually filled the bath with fresh water at rising levels of 1, 3, 5, 7 and 10 cm in order to compare previous measures. Water surface activity was measured at three points for each level: at the center of the bath and 25 cm on each side from the center along the longitudinal axis of the bath. In total, 15 measurements were taken.

After the experimental measurements with water layers, the backfill of the experimental site was carried out using wet fine- and medium-grained quartz sand. To do so, we used the sand of the Sablinka and Tosna formations. The sand from the Sablinka formation, which is represented by almost pure quartz arenites with a small admixture of ferruginous minerals, has a radioactivity of 8–10  $\mu\text{R}/\text{h}$  (Lebedev, 2012). In contrast, the Tosna formation contains NRN, mainly uranium, which causes the level of radioactivity to reach up to 25  $\mu\text{R}/\text{h}$ . Thus, three different habitats of benthic biota were modelled: water; pure wet sand; and sand with a relatively small amount of NRN. With different layers of sand with thicknesses of 1, 3, 5, 7 and 10 cm, measurements of the surface activity of the sand were carried out according to the same scheme as for water.

After the measurements, the roof of the *Dictyonema* shale was re-cleared to sample for radionuclides. Five samples were taken with a weight of about 1 kg at the same points where surface activity on the shale roof in the air had previously been measured, and each sample was taken for the entire thickness of the shale layer. Samples were measured by a RADEK gamma spectrometer (RADEK, St. Petersburg) (see Section 3.3).

### 3. Results and discussion

To define the parameters of the model, we will assume that the bottom of the shallow basin, the surface source of radiation, is represented by the *Dictyonema* shale. Bituminous graptolitic argillite of the lower Tremadocian, better known as “*Dictyonema* shale”, is widely distributed in the northwest of the Russian Platform. These rocks were deposited in the epicontinental Baltic Paleobasin, which occupied vast continental territories of the Baltica in the Ordovician (Männil, 1966).

After the deposition of the *Dictyonema* shale, the basin experienced a regression, during which huge portions of the bottom turned into a flat land covered with a firm graptolite claystone containing in each ton more than a kilogram of toxic metals, not less than 80 g of uranium. In the late Tremadocian–early Floian, the roof of the graptolite claystone began to be covered again with water from a slowly transgressing basin in which a sort of condensed sedimentation was established. Marine biota began to develop in extensive and shallow waters, the bottom of which was a source of radioactive radiation. The dose loads on benthic biota created by the *Dictyonema* shale are described in detail in a previous study by the authors, see (Lebedev et al., 2018). Their studies take into account the real ratio of radioactive elements and their uneven distribution along the strike and thickness of the seams. In general, this dose in the air is about 600–1200 nGy/h.

#### 3.1. Calculation of the attenuation coefficient in water

Water, being the habitat of benthic biota, is the absorber of radiation, weakening its effect on living organisms and benthic plants. The question is to what extent the effect of weakening the radiation level is manifested while the water layer increases.

Gamma quanta possess the greatest penetrating ability. The gamma quantum interacts with the atoms of matter that gamma radiation passes through. These interactions are characterized by the fact that each gamma quantum is eliminated from the incident beam as a result of a single act. The law of attenuation of a monochromatic gamma-ray beam (gamma quanta with the same energy) has the following form (Galy and Magill, 2001):

$$I = I_0 e^{-\mu d}, \quad (1)$$

where  $I$  is the intensity of a beam of gamma rays transmitted through an absorber layer of thickness  $d$ ;  $I_0$  is the intensity of the incident beam of gamma rays;  $\mu$  is the linear attenuation coefficient, which is equal to the relative decrease in the intensity of the gamma-ray beam after passing through the absorber 1 cm thick. Thus, the beam intensity decreases exponentially with increasing thickness of the matter layer.

We emphasize that the above formula is valid only for a monochromatic beam of gamma radiation that normally penetrates on the surface of the absorber. The linear attenuation coefficient, according to its definition, has dimension  $\mu = S^{-1}$ . It depends on the energy of gamma radiation and on the material of the absorber (the density of matter).

The linear attenuation coefficient is the total coefficient that takes into account the attenuation of the gamma-ray beam due to three main processes: the photoelectric effect  $\tau_{pe}$ ; the Compton effect  $\tau_C$ ; and the pairing effect  $\tau_p$ . As follows:

$$\mu = \tau_{pe} + \tau_C + \tau_p. \quad (2)$$

Photoelectric interactions are dominant at low energy and pair production — at high energy, with Compton scattering being most important in the mid-energy range (Gilmore, 2008). The coefficient  $\tau_{pe}$  decreases sharply with increasing energy; the value of the coefficient  $\tau_C$  decreases slower than  $\tau_{pe}$ ; the coefficient of pair formation  $\tau_p$  increases with the energy increment starting from 1.02 MeV.

As the atomic number of the absorber material ( $Z$ ) increases, the photoelectric effect increases in proportion to  $Z^4$ , the Compton effect is proportional to  $Z$ , and the effect of pair formation is proportional to  $Z^2$ . In general, the penetrating power of gamma radiation increases with increasing energy of gamma quanta and decreases with increasing density of substance (absorber).

Figure 4 shows the results of our calculations to determine the absorption coefficient in water for the three most intense high-energy lines (the legend shows energy lines in kiloelectronVolts) of the daughter products of the decay of uranium-238 ( $^{238}\text{U}$ ).

As can be seen from the calculations, with a layer of water up to 10 cm thick, the radiation of the most intense and hard lines of the daughter decay products (DDP) of  $^{238}\text{U}$  will be attenuated by no more than about 40–60%. In this case, the gamma radiation of the rocks forming the bottom of the water basin will still have a significant effect on the benthic biota. In contrast, a half-meter layer of water will almost completely absorb the radiation from the bottom rock.

### ***3.2. Calculation of the absorption of gamma radiation from the Dictyonema shale***

To assess the absorbing properties of graptolite argillite, whose density is approximately  $2 \text{ g/cm}^3$ , we can compare them with the tabulated values of the linear absorption

coefficient of gamma radiation in concrete (Ragheb, 2006a; 2006b), which has an average density of  $2 \text{ g/cm}^3$  (the density of concrete is  $1.8\text{--}2.2 \text{ g/cm}^3$ ). Such estimations show that the half-attenuation layer in the studied rocks for the 609, 1120, and 1764 keV lines is approximately 3.6, 5.0 and 6.1 cm, respectively. Hence it is not difficult to calculate that 7/8 of the most intense high-energy line of gamma radiation of the DDP of a series of  $^{238}\text{U}$  will be absorbed in a layer of only 24.5 cm.

Therefore, it can be argued that the dosage amount created by the NRN of shale formation in the environment affects only its uppermost layer with a thickness of not more than 25 cm, which is also the absorber of the radiation of the underlying layers. We purposely did not consider the migration capabilities of radon, which can enter the surface through pores and cracks, dissolve in water, etc.; this is beyond the topic under discussion. Calculations in this case will be very approximate in view of the great variability of the influencing factors (e. g. pressure, temperature, fracturing of rocks). In addition, the direct effect of radon as an alpha-emitter is possible only if conditions are created for its accumulation in confined spaces.

### 3.3. Measurements of the specific activity of the bottom model

Specific sampling activity of the *Dictyonema* shale was determined using a RADEK gamma spectrometer (RADEK, St. Petersburg) at the Testing Laboratory of the Radiation Hygiene Department of the FGUZ Center for Hygiene and Epidemiology in the City of St. Petersburg. This gamma spectrometer is calibrated in units of radionuclide activity based on the standard measures of radionuclide activity  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$ .

Specific activity of the rock in Bq/kg was recalculated into the absorbed dose in the air as nanogray per hour (nGy/h) (Table 1) by considering the coefficients adopted in the reports of the United Nations Scientific Committee on the Effects of Atomic Radiation (United Nations..., 2000) and referring to the reports of the International Commission on Radiation Units and Measurements (International Commission..., 1994). The values of the conversion factors for  $^{238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  are 0.462, 0.604 and 0.0417, respectively.

Table 1. Specific activity and absorbed dose rate by sampling points

Sample	Activity, Bq/kg			Dose Rate, nGy/h			
	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$	$^{226}\text{Ra}$	$^{232}\text{Th}$	$^{40}\text{K}$	Sum
1	1240	100	2245	570	60	95	725
2	1575	25	1880	730	15	80	825
3	1280	50	1880	590	30	80	700
4	550	50	1985	255	30	85	365
5	1525	85	1950	705	50	80	840
<b>Average</b>	<b>1235</b>	<b>60</b>	<b>1990</b>	<b>570</b>	<b>35</b>	<b>85</b>	<b>690</b>
* $^{226}\text{Ra}$ line is measured for $^{238}\text{U}$							

### 3.4. Results of field measurements of the absorption coefficient

The data averaged over all the measurement points were used to determine the absorption coefficient of the primary radiation of natural radionuclides (uranium, thorium, potassium) in environmental media that absorb primary radiation (Fig. 4). For each testing case of the different absorbers, the measurement on the clean surface of the radioactive rock was equal to 1. The results of the measurements for different absorbing media (air; water; two types of wet sand: Sablinka formation — sand\_sb — and Tosna formation — sand\_ts —) at each layer are shown in Table 2.

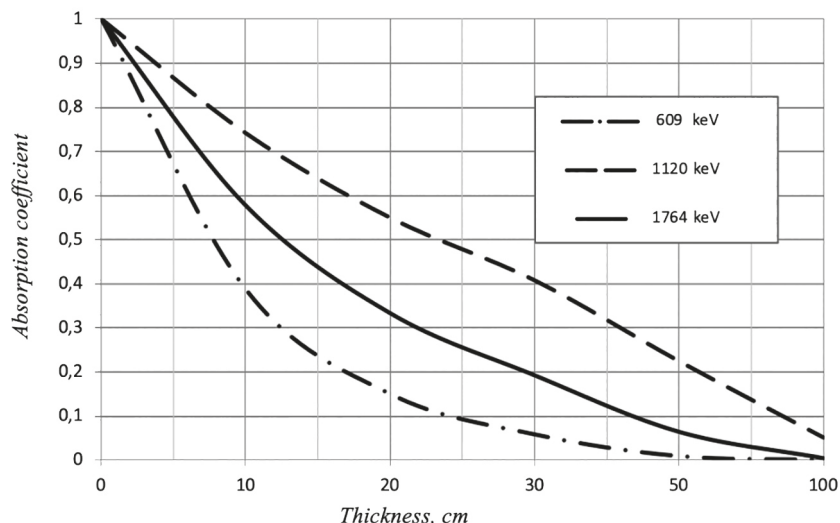


Fig. 4. Dependence of the absorption of gamma radiation in water

Table 2. Dependence of the absorption coefficient of the integral gamma-radiation flux of NRN of the *Dictyonema* shale on the layer thickness of different intermediate environmental media

<i>h</i> , cm	Absorption coefficient			
	<i>air</i>	<i>water</i>	<i>sand_sb</i>	<i>sand_ts</i>
0	1	1	1	1
1	0,99	0,95	0,89	0,94
3	0,98	0,83	0,81	0,88
5	0,96	0,78	0,78	0,80
7	0,94	0,75	0,66	0,68
10	0,92	0,6	0,56	0,58

The dependence of the absorption coefficient was approximated by a linear trend. As shown in Fig. 5, the presence of a 10 cm thick layer of water leads to the absorption



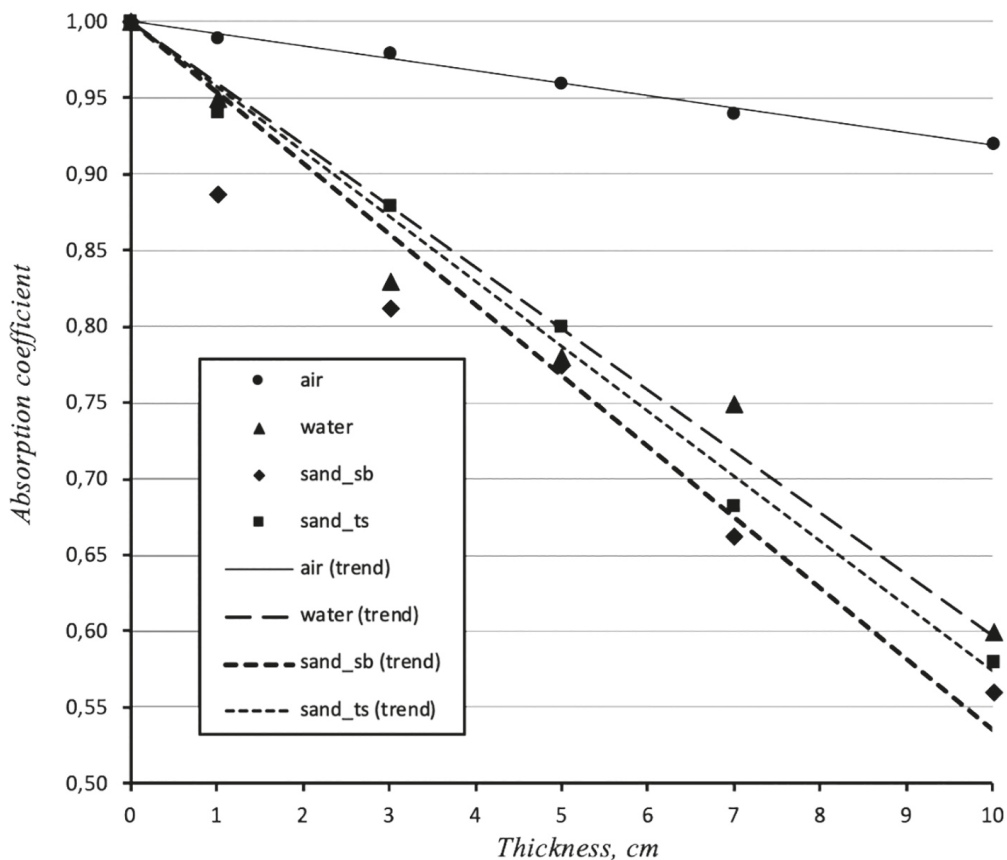


Fig. 5. Experimental dependence of the absorption coefficient on the thickness of the shielding layers of water and sand. The symbols “circle”, “triangle”, “square”, and “rhombus” show the results of independent measurements of the absorption coefficient of the integral gamma-radiation flux of NRN transmitted from an area source through layers of air, water or sand. The lines show the linear trends

of 40% of the integral flux of all gamma lines of NRN, which is in full agreement with the calculated data (Fig. 4). The coefficient of absorption of gamma radiation in moist quartz sand was surprisingly low: not more than 50% at a layer thickness of 10 cm. The results of measurements derived from the Tosna formation sand containing NRN were quite logical; its absorption coefficient occupies an intermediate value between water and pure quartz sand.

#### 4. Conclusions

Our aim was to present an experimental assessment of the effect of the shielding properties of environmental components that can substantially reduce the dose received by benthic biota from the radioactive bottom of the *Dictyonema* shale. We determined the absorption coefficient of the primary radiation of natural radionuclides (uranium, thorium, potassium) contained in the shale. Water and sand, which were gradually depos-

ited on the roof of the *Dictyonema* shale, represented environmental media absorbing the primary radiation.

The experimental data were obtained from absorbing layers of water and sand with thicknesses of 1, 3, 5, 7, and 10 cm. With a shielding layer thickness of 1 cm, the absorbed dose values are close in all environmental media, reaching an amount of about 5%. The presence of a 10 cm thick layer of water reaches absorption of 40% of the integral flux of all gamma lines of NRN. The absorption coefficient of gamma radiation in wet but not dense quartz sand at a layer thickness of 10 cm was surprisingly low, no more than 50%. The results of measurements with the Tosna formation sand containing NRN showed average values between water and the Sablinka formation sand: at a layer thickness of 10 cm, the absorption coefficient was 55%. Thus, despite the screening effect of the environmental media, benthic biota could have been exposed to radiation for thousands of years. The confirmation of this hypothesis indicates that most likely the radiation could have had a significant impact on the development of organisms and species in general. This fact should be considered in the paleoenvironmental reconstructions. Thanks to the results obtained in this research, a significant number of new hypotheses about the effect of radiation on organisms and their habitat can be formulated. For example, could the dramatic increase in radiation background have brought about the diversification of biota in the Ordovician? Is one of the phases of the dramatic increase in radiation background in the Ordovician associated with *Dictyonema* shale as rock? Or vice versa, did no adverse effect on biota occur? Currently, this topic has not been studied in detail and could be the key to understanding patterns of development of organisms in the Ordovician Period.

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### Моделирование экранирующих свойств компонентов природной среды как защиты от радиоактивного излучения ордовикских диктионемовых сланцев

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В данной статье представлены результаты моделирования условий поглощения природных радионуклидов (уран-238, торий-232, калий-40) в различных природных средах (вода и кварцевый песок) для оценки влияния защитных свойств компонентов окружающей среды на интенсивность внешнего облучения донной биоты. Источником излучения были нижнетремадокские диктионемовые сланцы, которые слагали дно Балтийского палеобассейна. Вода и базальный кварцевый песок, который постепенно накапливался на кровле диктионемовых сланцев, были использованы при моделировании в качестве среды (материалов), поглощающих первичное излучение. В статье описывается эксперимент, поставленный в натуральных условиях на одном обнажении с разрезом геологических пород ордовика, включающем высокорadioактивные диктионемовые сланцы. Это небольшой участок, однако результаты моделирования спра-

ведливы и для огромных пространств Балтийского палеобассейна. В позднем тремадоке — фло вместе с наступающим морем стала появляться бентосная биота. Эксперимент количественно иллюстрирует, какую дозовую нагрузку эта биота могла испытывать в течение десятков и сотен тысяч лет, пока диктионемовые сланцы не были перекрыты достаточно мощным слоем обводненных осадков. Экспериментальные данные показывают, что присутствие слоя воды толщиной 10 см приводит к поглощению 40 % интегрального потока всех гамма-линий природных радионуклидов, а во влажном, неплотном слое кварцевого песка такой же толщины величина поглощения не превышает 50 %. Таким образом, несмотря на экранирующее воздействие окружающей среды, бентосная биота Балтоскандии в ордовике могла подвергаться значительному радиационному воздействию в течение тысячелетий. Основная ценность эксперимента заключается в том, что он был выполнен *in situ*, а в качестве элементов модели использовались исключительно природные среды.

*Ключевые слова:* диктионемовые сланцы, естественные радионуклиды, гамма-излучение, коэффициент поглощения, радиационно-защитные свойства, бентосная биота.

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