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Some aspects of environmental hazard due to uranium mining in Ukraine

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Abstract. Some aspects of environmental hazard within uranium mining areas are considered. The uranium content in the environment components (rocks, soils, underground and surface waters) of the central part of the Ukrainian Shield within and beyond the uranium mining area is analyzed on the example of the Michurinske ore field. It is emphasized that man-made sources of natural origin should be considered more broadly than just waste dumps from uranium mining and processing enterprises. These are sources of ionizing radiation of natural origin, which have been subjected to concentration or their accessibility has been increased because of anthropogenic activity. Additional irradiation to the natural radiation background is formed. Waste dumps of uranium mining are considered as sources of potential dust pollution in the surface layers of atmosphere with fine dust containing uranium, its decay products and associated elements. The area of waste dumps is calculated using space images. Uranium accumulates in the dusty fraction, where its content is 0.01-0.06%. Taking into account the geological and geochemical characteristics of uranium deposits, radioactive elements, heavy metals and other associated elements of uranium mineralization are carried out of the dumps by winds and atmospheric waters with their subsequent migration into environment components.

A mathematical model of potential dust air pollution in the area of long-term operation of the oldest uranium mine is presented for the summer 2019. In total, 15 factors influencing the potential threat of air dust pollution are considered and analyzed. The mathematical model is developed on the basis of the method of discriminant functions. To assess the degree of the model parameters informativeness, one-factor covariance analysis is used. It allows assessing the degree of a single sign influence on the prediction result. The developed model takes into account the area of waste dumps, uranium content in the dust fraction and wind direction southeast and/or east as the most hazardous for the study area. The model allows determining correctly the level of potential threat of air dust pollution in $96.3\% \pm 3.6\%$ of all cases.

Key words: environmental hazard, uranium mining waste dumps, dust pollution, method of discriminant functions

Деякі аспекти екобезпеки внаслідок видобування урану

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Анотація. Розглянуто деякі аспекти екологічної небезпеки в районі видобутку урану та за його межами. Проаналізовано вміст урану в компонентах довкілля (породах, ґрунтах, підземних та поверхневих водах) центральної частини Українського щита на прикладі Мічуринського рудного поля. Наголошено, що техногенно-підсилені джерела природного походження слід розглядати ширше, ніж просто відходи урановидобувних і переробних підприємств. Це джерела іонізуючого випромінювання природного походження, які в результаті антропогенної діяльності були піддані концентруванню або збільшилася їхня доступність, внаслідок чого утворилося додаткове до природного радіаційного фону опромінювання. Породині відвали урановидобування розглядаються як джерела потенційної запиленості приземних шарів атмосфери дрібнодисперсним пилом, який містить уран,

продукти його розпаду та супутні елементи. Площа породних відвалів розраховується з використанням космічних знімків. Уран накопичується у пиловатій фракції, де його вміст складає 0.01-0.06%. З урахуванням геолого-геохімічних характеристик уранових родовищ радіоактивні елементи, важкі метали та інші елементи-супутники уранового зрудення виносяться з відвалів вітрами та атмосферними водами з подальшою їх міграцією у компоненти довкілля. Розроблено математичну модель потенційного пилового забруднення повітря в районі довготривалого функціонування найстарішої урановидобувної шахти, за літній період 2019 року. Загалом розглянуто та проаналізовано 15 факторів, що впливають на потенційну загрозу запиленості повітря. Математичну модель розроблено на базі методу дискримінантних функцій. Для оцінки ступеня інформативності параметрів моделі був використаний однофакторний дисперсійний аналіз, що дозволяє оцінити ступінь впливу окремо взятої ознаки на результат прогнозування. Розроблена модель, що враховує площу відвалів, вміст урану в пиловатій фракції та напрям вітру південно-східний та/або східний як найбільш небезпечний для досліджуваної території, коректно дозволяє визначити рівень потенційної загрози запиленості повітря у $96,3\% \pm 3,6\%$ усіх випадків.

Ключові слова: екобезпека, відвали урановидобування, пилове забруднення, метод дискримінантних функцій

Introduction.

The development of mineral deposits that contain radioactive elements can lead to radioactive contamination of the territory and the formation of man-made sources of natural origin, generating alpha, beta and gamma radiation. Issues related to low-level radioactive wastes management derived from exploration, mining and processing complexes of uranium deposits and deposits enriched in radioactive elements are widely discussed in the world, as evidenced, in particular, by materials of international symposia (since 1997) on Naturally Occurring Radioactive Materials (NORM), the last of which was held in 2019 in Denver, USA.

The primary hazard to humans and the environment from natural radionuclides is associated with Technologically Enhanced NORM (TENORM), as specified in Norms of Radiation Safety of Ukraine (NRBU, 1997; TENORM, 2007). Because of man-caused activities, radioactive substances are concentrated in TENORM or their availability is increased, due to which additional to the natural background radiation occurred.

If to think of radiation hazard within the areas where the sources of TENORM are developed, then uranium mining and processing waste dumps and dust pollution of the surface atmosphere have to be considered as an urgent regional problem. Dust pollution within mining sites and beyond is mainly due to ventilation, which creates a scattering halo up to 200 m, atmospheric dust caused because of the host rocks grinding, transportation, as well as due to the long-term storage of low-level waste dumps, pollution from which also requires in-depth study.

The largest number of known uranium deposits, numerous anomalies of uranium and thorium in crystalline rocks, anomalous concentrations of uranium (up to $9 \cdot 10^{-2}$ g/l) in underground waters are met in the central part of the Ukrainian Crystalline Shield (Belebtsev et al., 1995; Verkhovtsev et al., 2014, 2018; Bakarzhiev and Lysenko, 2018) and presented in the National Atlas of Ukraine, 2007 (Rudenko *Ed*). In terms of uranium resources and proven reserves,

Ukraine is among the top ten countries in the world and is a leader in Europe. A major share of deposits has insignificant uranium reserves, ranging from 1 to 5 thousand tons. The deposits are characterized by the complexity of tectonic structure and ore bodies' morphology. There are a lot of «windows» of ore-free rocks. Uranium concentrations are usually associated with hydrothermal metasomatic processes, discontinuous tectonics, and various exogenous (syngenetic), diagenetic, and epigenetic veins. Many deposits are genetically unique, so the search for their analogues in many cases did not lead to expected results. However, recent studies have significantly expanded the database of the Ukrainian uranium (thorium) deposits and manifestations (Mikhailichenko, 2018).

Sources of radiation exposure to the environment due to uranium mining are diverse and covered in many publications (Shumlyanskyi et al., 2003; Lyashenko et al., 2011, 2018; Dudar et al., 2015, 2018; Stankevich et al., 2016, 2018) and generalized in the IAEA publication on uranium exploration worldwide (IAEA, 2018). They are aerosol, dust, liquid, solid low-level wastes of mining and ore-processing complexes of uranium deposits and deposits, which are enriched in associated radioactive elements. Environmental components within and beyond areas, where the processes of extraction and processing of radioactive raw materials take place, are subjected to radioactive contamination of various level. Radioactivity caused by uranium ore mining requires special measures to protect the population and the environment in addition to generally accepted control that accompanies the extraction of other metals.

Research methods.

The study uses a comprehensive approach that includes data analysis and generalization on long-term environmental impact due to uranium mining; mineralogical and petrographic analysis of uranium ores and host rocks; method of discriminant functions; mathematical statistics for processing the results of measurements and modeling.

The purpose of the work is to overview and analyze the environmental impact of natural radiation sources within and beyond the uranium mining areas in Ukraine and develop a mathematical model of potential dust air pollution on the example of the oldest uranium mine in highly populated region.

posits (Michurinske and Tsentralne) connected under residential neighborhood which is perceived as a source of permanent potential environmental hazard. For half a century since the mine started prospecting, an underground labyrinth was created at depths of 160 to 650 m (Podulyakh, 2017). There are five vertical



Fig.1 Uranium mining site

Study area. In Ukraine uranium is mined in the vicinity of residential areas in the Kropyvnytskyi district, central Ukrainian uranium province. The territory of the Kropyvnytskyi city and its environs is located in the area within the tectonic node of deep faults that controls uranium mineralization. Natural and man-made factors of radiation hazards have become widespread here (Kalashnik, 2017; Dudar et al., 2019, 2020). The oldest uranium mine Ingulska site located in the south-east part of the city of the city of Kropyvnytskyi is especially worth attention as a unique existing mining area (Fig. 1).

The mine has been developing two remote de-

posits here and underground tunnel almost 6 km long, passing also under the river Ingul, dug for the connection of both uranium deposits.

After extraction, grinding and radiometric sorting the ore is loaded into the wagons and then transported for further processing to the town of Zhovti Vody, where the hydrometallurgical plant is located. A railway track and automobile roads are connected to the territory of the mine which is also proved to serve as a continuous source of the environmental pollution. Poor off-balance ore and barren rock are stored near the mine in waste rock dumps.

Analysis of uranium content in the environment components. Uranium content in the environment components is a very specific feature of the study area which stipulates its enhanced level of background radiation and development of technogenically enhanced sources of natural origin. Uranium in *rocks* of the earth's crust: 1) is available in mineral form (uranium minerals: uranium black, uraninite, nasturan, coffinite); 2) isomorphically included in the crystal lattices of highly radioactive non-uranium minerals (zircon, monazite, apatite, sphene); 3) is scattered in rocks or dissolved in water. The average uranium content in acid rocks $(1-6) \cdot 10^{-4}\%$, in alkaline – up to $30 \cdot 10^{-4}\%$ (Shumlyanskyi et al., 2003; Fomin et al., 2019). Compared with uranium clark in acidic igneous rocks, the average uranium content in the host rocks of the Michurinske ore field is 1.5-2 times higher (table 1).

The uranium content in *soils* varies between $(0.5-2.1) \cdot 10^{-4}\%$. The background is $1-1.5 \cdot 10^{-4}\%$. Uranium scattering halo (uranium content is more than $1.5 \cdot 10^{-4}\%$) is observed at the Michurinske deposit site. Capillary-diffusion rise of uranium-bearing waters, activated in fault zones, is the cause of the formation of uranium salt halos in the soils, as well as anomalies of radioactivity (at the level of 1 m from the earth's surface).

Table 1. Average content of uranium in crystalline rocks of the Michurinske ore field

№№	Roks	Samples-number	Uranium content, %
1	Gneiss fine-grained	46	0.0005
2	Gneiss coarse-grained	33	0.0007
3	Gneiss even-grained	15	0.0008
4	Trachytoid granite	42	0.0009
5	Pegmatite	23	0.0008

Availability of radioactive hydrochemical halos in *ground and surface waters* around uranium deposits is another characteristic feature of the study area. All aquifers of the area are fed by precipitation. The main aquifer that feeds the rivers is the one in fractured zone of the Precambrian crystalline rocks. Previous research proved the stability of radioactive hydrochemical halos: the content of radionuclides of uranium, radium and radon is stable over time (Sushchuk and Verkhovtsev, 2019; Fomin, 2019). So, there is a natural contamination of groundwater with radioactive elements. Beyond the ore deposit sites, contamination of uranium surface water is predominant.

The migration of natural radionuclides within the study area and beyond depends on general geomorphological and geochemical conditions. The mining site along with the wastes dumps is located on the left bank of the river Ingul at a distance of 150-200

m from the riverbed. The relief is lowered down from the absolute mark of +140 m on the eastern border of the mining site to +100 m (water inlet of the river Ingul). It means that the uranium salt halo is directed to the riverbed and covers a small area within the mining zone. This fact makes it possible to form surface runoff enriched in natural radionuclides of precipitation and meltwater infiltrated through dumps, as well as from highways and land terrain (Koshik et al., 2013; Dudar et al., 2011, 2015; Sushchuk and Verkhovtsev, 2019).

Availability of fracture zones and their density in host crystalline rocks (granites and gneisses), the presence or absence of uranium minerals, sulfides, iron minerals as well as penetration of gaseous fluids from the earth crust interiors influence on the conditions for radionuclides migration (shumlyanskyi et al., 2007; Fomin et al., 2019; Dudar et al., 2019, 2020). Vertical hydrogeochemical zoning has also been proved due to natural groundwater contamination with uranium, radium and radon. In the upper parts of geological cross-sections, where oxidative conditions prevail, groundwater is maximally saturated with uranium and minimally with radium.

Radon halos tend to fractured zones – faults, the so-called «emanating collectors». The amount of ura-

anium increases in fractured waters. Within zones of weathering crust development, especially in rocks with high uranium content, its concentration in fractured waters increases on average to $70 \cdot 10^{-6}$, reaching $(150-300) \cdot 10^{-6}$ g/l. That means, it increases 6-25 times compared to the content in waters of Quaternary sediments (Shumlyanskyi et al., 2003, 2007).

The concentration of ^{222}Rn in the indoor air is a very specific characteristic feature of the study region residential areas. In particular, it depends on the content of ^{238}U , ^{226}Ra and other radioactive components in the natural environment components - rocks, weathering crust of parent material, soils and groundwater, on ^{222}Rn emanation coefficient from the soil, on the soil properties and condition, on concentration of uranium anomalous in the earth crust. Geospatial analysis of radon-prone areas identification taking into account uranium content in the environment as well as spa-

tial density of faults and lineaments can be of great help for potential radon hazard sites study (Dudar et al., 2019, 2020). Radon-prone areas are considered in the EU Basic Safety Standards (EC Council Directive 2013/59/Euratom, 2014) and of great attention in the European Atlas of Natural Radiation (EU Joint Research Center, 2019).

Research material and discussions.

The uranium ores within the Michurinske ore field are mainly one-component and monomineral in composition. The presence of thorium and other radioactive elements is negligible (table 2.)

All the waste dumps of the Ingulska mine con-

position of rocks that after a series of underground explosions were crushed and removed to the surface), dust fraction (< 0.25 mm) and the content of uranium in it, which is carried by the wind according to the wind direction. It is also important to take into account the availability of residential areas and the distance to them from the SPZ of the mining site, and to analyze the wind rose and wind speed of the study area in general and on the example of a particular season (and / or year).

The presented study analyzed the potential threat of air dust pollution within and beyond the area of long-term operation of the Ingulska mine on the example of the wind rose of summer 2019. A total of

Table 2. Geochemical characterization of the Michrinske uranium deposit [Fomin, 2020]

Chemical elements	U	Th	V	Ni	Pb	Sr	Be	Zr
Content, g/t	5-1670	0.5-4.3	12-54	4.0-12.0	5-810	36-161	1-46	22-291

tain a low level of uranium (more than 0.01%). The content of uranium in the dust fraction exceeds the content of uranium in the total samples. The most radioactive are the samples from the foot of almost all waste dumps. The usual uranium content in the dust fraction (<0.25 mm) is 0.01-0.06%. (Shumlyanskyi et al., 2003; Lyashenko et al., 2011; Kovalenko, 2013).

Having analyzed the factors influencing the potential threat of air dust pollution within and beyond the sanitary protection zone (SPZ) of any mining site, it is possible to identify potentially hazardous conditions for any study area and predict measures to eliminate it, especially for residential areas. The authors have analyzed a number of man-made and climatic factors that should be first considered to tackle the task (tables 3, 4).

Of course, in any case, the characteristics of the waste dumps themselves are important - their area, mineral and chemical composition of the crushed rock substrate (which in principle corresponds to the com-

16 factors for 27 predicted situations were considered and analyzed. All situations were divided into 2 groups: group 1 - with a low level of potential dustiness threat; group 2 - with a high level of potential dustiness threat. The waste dumps area was calculated using the Sentunel-2 images data (as of 07.02.2019) and accurate terrain SRTM (2000) data, presented on the figure 2. It makes up 0.2650 km².

To reduce the potential air dustiness threat, the variants of reducing the waste dumps area by at least 2 times are being considered. The uranium content in the dumps dusty fraction is given based on published analyses results (Shumlyanskyi et al., 2003). Climatic data were taken from the World Weather site data (World Weather, 2019). The south-eastern and eastern wind directions (SE+E) are considered to be the most potentially threatening, as in this case the dustiness threatens the south-eastern and eastern outskirts of the city of Kropyvnytskyi located at a distance of 1.5 km to 5 km from the mine waste dumps.

Table 3. Technogenic and man-made factors influencing the potential dustiness threat

Threat level	Technogenic and man-made factors									
	within SPZ (mining site)						residential sites			
	Waste rock dump area, m ²		U in dusty fraction of waste rock, %		Fraction ≥ 0.25 in samples from dumps, %		Distance from SPZ, km		dose rate at height of 1 m, mkSv/g	
	A	A ¹	B	B ¹	C	C ¹	D	D ¹	E	E ¹
1	130 000- 160 000	1	< 0.005	1	< 5	1	>5	1	0.08-0.09	1
1	161 000-190 000	2	0.005-0.01	2	5-9	2	3,1-5	2	0.10-0.11	2
2	191 000-265 000	3	0.02-0.03	3	10-14	3	2,1-3	3	0.12-0.13	3
2	≥265 000	4	0.04-0.06	4	> 15	4	1-2	4	>0.13	4

Table 4. Climatic factors influencing the potential dustiness threat

Climatic factors																			
NW		N		NE		NE+E		S		SW		W		Wind speed, m/s				Precipitation	
F	F ¹	G	G ¹	H	H ¹	I	I ¹	J	J ¹	L	L ¹	M	M ¹	N	N ¹	O ¹	O ¹	P	P ¹
11-9	1	11-9	1	11-9	1	<2	1	<2	1	11-9	1	11-9	1	≤2	1	30-22	1	14-9	1
8-5	2	8-5	2	8-5	2	4-2	2	4-2	2	8-5	2	8-5	2	4-3	2	21-14	2	8-5	2
4-2	3	4-2	3	4-2	3	8-5	3	8-5	3	4-2	3	4-2	3	6-5	3	13-6	3	5-2	3
<2	4	<2	4	<2	4	11-9	4	11-9	4	<2	0	<2	0	≥6	4	<5	4	<2	4

The method of discriminant functions was used to objectify the determination of potential dustiness threat level. This method has a number of advantages: takes into account the variability of the model parameters; considers the signs as a whole and identifies the most significant of them; demonstrates the share of each sign in the final conclusion. Before the discriminant analysis, all signs were coded (tables 3 and 4) and put in accordance with the 16-dimensional vector, which takes into account the absence, presence, focus and magnitude of each sign.

One-factor analysis of variance was used to assess the informativeness level of the model parameters, for which the null hypothesis of no influence was put forward and evaluated. At the same time, an alternative hypothesis was put forward that the influence of the studied sign exists. In order to test the hypothesis, Fisher’s criterion (F) was calculated. Observed value of Fisher-test (F_o) was compared with Critical F-value (F_c), which depended on the level of significance p (<0.05). If F_o > F_c, then an alternative hypothesis was accepted, which allowed to assert with a probability of at least 0.95 about the influence of the studied sign on the prediction outputs. As a result, three statistically significant and non-correlated signs were identified, which allowed determining the level of potential dustiness threat (table 5).

These signs were further used as the main ones in the construction of the «decision rule» of the mathematical model for predicting the level of potential dustiness threat in the form of equations:

$$F_1(X) = 13.989 \cdot X_1 - 10.018 \cdot X_2 + 10.514 \cdot X_3 - 8.640,$$

$$F_2(X) = 43.502 \cdot X_1 - 35.830 \cdot X_2 + 33.664 \cdot X_3 - 64.748,$$

where X₁ - waste rock dumps area

X₂ - U in dusty fraction of waste rock

X₃ – wind direction – south-eastern and/or eastern

The obtained values of the variables F1 (X) and F2 (X) are compared with each other and under the

condition of F₁ (X) > F₂ (X) we can talk about a low level of potential dustiness threat.

The obtained coefficients and constants of discriminant equations reflect a linear regression set of relevant indicators that have the greatest impact on predicting the potential dustiness threat.

Based on the obtained value of discriminant functions, graphs of distribution of values F₁(X) and F₂ (X) were made up (Fig.3), where a) graph for group 1, b) graph for group 2.

The assessment of optimal assignment likelihood into groups, the usefulness of discriminant functions and the number of functions that have real meaning in determining the differences between groups were evaluated using canonical correlation coefficients. Having analyzed the obtained value of the canonical correlation coefficient (0.965), one can draw the following conclusion. There is a high positive relationship between the real process and the predicted values obtained through mathematical model, which is also confirmed by the high percentage of absorbing dispersion of this function (99.0%).

The assessment of the discriminant functions significance was verified by Wilkes λ-statistics (table 6), according to the formula:

$$\lambda_i^* = \prod_{i=k+1}^g \frac{1}{1 + \lambda_i} \tag{1}$$

where k is the number of calculated functions; λ_i is the eigenvalue.

Table 6. Significance of discriminant functions

Test of Function (s)	Wilks’ Lambda	df	Sign.
1 to 2	0.069	3	0.001

The λ-Wilkes test showed that the level of differences is quite significant (p = 0.001).

The accuracy of classification, according to the obtained model, is estimated on the basis of comparison of coincidences of the predicted and actual groups and presented in table 7.

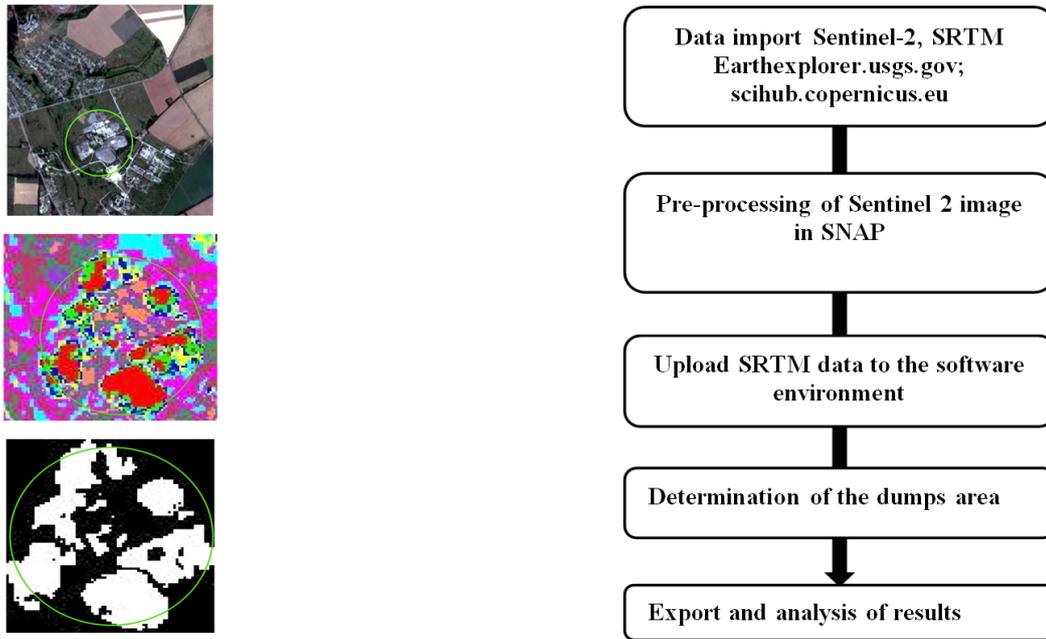


Fig. 2 Chain for waste dumps area determination

Table 5. Results from the estimation of the model parameters informativeness

Predictors included in the model	Observed F-value	Significance
Wind direction SE+E	141.667	0.001
Waste rock dumps area, m ²	95.368	0.009
U in dusty fraction of waste rock, %	104.212	0.001

Table 7. Classification accuracy

Pollution hazard	Predicted Group Membership		Total
	Group 1	Group 2	
Group 1	17	0	17
Group 2	1	9	10

Thus, the developed mathematical model allows correctly determining the level of potential dustiness threat in $96.3\% \pm 3.6\%$ of all cases.

Conclusions.

Studies of unique waste rock dumps of uranium mining in the Kirovohrad uranium ore subprovince allowed identifying them as sources of potential dustiness threat in the surface layers of atmospheric air with fine dust (less than 0.25 mm) containing uranium, its decay products and associated elements. Uranium mining dumps serve as a source of radioactive pollution of the environment, including residential areas. Uranium accumulates in the dusty fraction, where its content is 0.01-0.06%. Taking into account

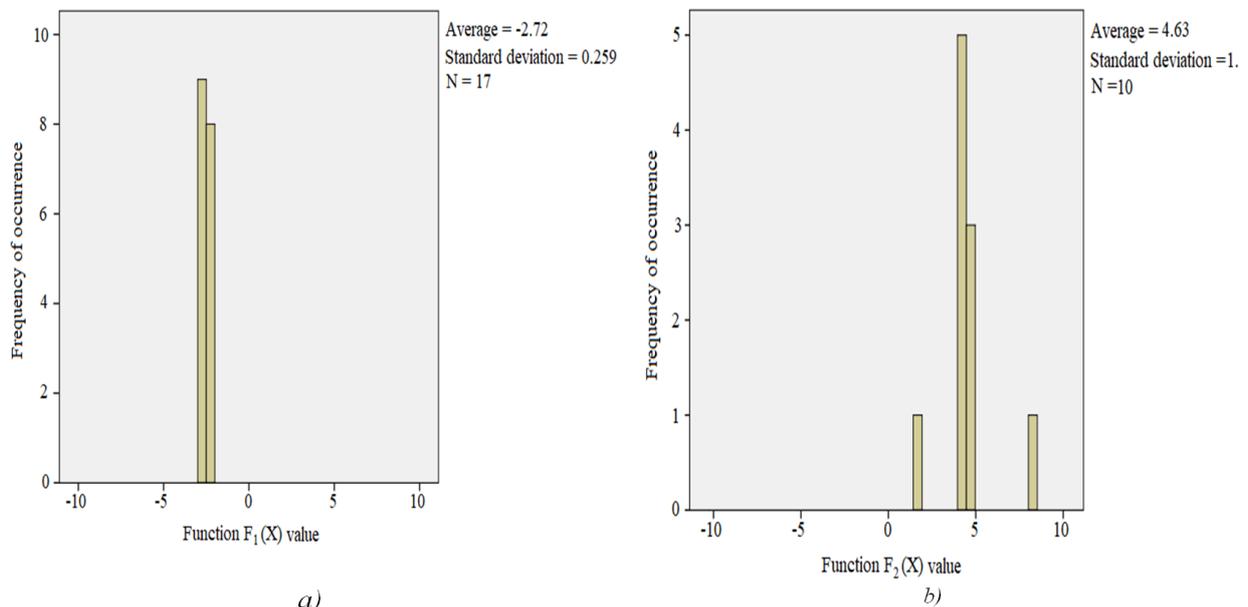


Fig. 3. Distribution of the discriminant function values: a) group 1; b) group 2

the geological and geochemical characteristics of uranium deposits, radioactive elements, heavy metals and other associated elements of uranium mineralization are carried out of dumps by winds and atmospheric waters with their subsequent migration into the environmental components.

The results of comprehensive research have identified the main ways for prediction the potential dustiness threat in the vicinity and beyond mining site of the Ingulska mine. The developed mathematical model based on the method of discriminant functions, taking into account the area of waste rock dumps, uranium content in the dust fraction and wind direction southeast and /or east, allows correctly determining the level of potential dustiness threat in $96.3\% \pm 3.6\%$ of all cases for the south-eastern and eastern outskirts of the city of Kropyvnytskyi.

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