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URANIUM MINING AND MILLING SITES AS SOURCES OF TECHNOLOGICALLY-ENHANCED NATURALLY OCCURRING RADIOACTIVE MATERIALS

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Data on the uranium deposits database for the identification of radioactive material and sources of natural radiation within the Ukrainian Crystalline Shield are provided. The territories of uranium ore mining and processing are considered from the point of technologically enhanced naturally occurring radioactive materials view according to the proposed classification of uranium legacy sites. Measures to refer the uranium mining and processing sites to the "legacy" category of Ukraine are presented.

INTRODUCTION

In terms of Radioactive Source Security Assessment it is considered a relatively new approach that assesses the national policies, commitments, and actions to secure radioactive sources and prevent dirty bomb real possibilities [1]. This approach makes considering the concept of radiological terrorism as use or threat of use for terrorist purposes of radioactive substances and materials designed to be used as a damaging factor of ionizing radiation.

Substances containing natural radioactivity are known as naturally occurring radioactive material (NORM). Last year the 25th international symposia on NORM, their origin, distribution in the world, impact on the environment and humans was held (since 1997) [2]. These symposia bring together experts from around the world, who especially emphasize the aspects of human intervention (first of all, mining operations), which lead to the concentration of radioactive substances in technologically-enhanced naturally-occurring radioactive material (TENORM). Because of this, additional to the natural radiation background exposure occurs [3, 4].

Uranium mining and processing sites, where radioactive raw material have been extracted and processed by different ways without corresponding mandatory safety measures, are then transferred to the category of uranium legacy sites, which are left as a legacy to the next generations. Currently, such areas fall under the scope of European Safety Standards (BSS – EU Directive 59/2013) as radiation-hazardous sites that require certain measures regarding possible ways of their environmental remediation and are therefore controlled at the level of EU environmental legislation [5]. This applies to twelve European countries (not including the eastern countries of the former USSR), where such territories exist and are waiting for their time, or have been fully or partially reclaimed: Bulgaria, Estonia, Spain, Germany, Poland, Portugal, Romania, Slovenia, Hungary, France, of the Czech Republic and Sweden [6–8].

To date, uranium is almost not mined in Europe, except Ukraine. The legislation opportunities provide different responses to uranium legacy sites management in the listed above countries, depending, in particular, on the environmental standards adopted in each country [9–11]. However, public opposition made it possible to examine and restore each site from the point of both radiation and general environmental hazards to public health and ecosystem restoration.

1. DATABASE OF URANIUM DEPOSITS FOR IDENTIFICATION OF RADIOACTIVE MATERIAL

There are many places around the world where radioactive materials that have gone through the nuclear fuel cycle are produced, used and stored. Ukraine has the largest amount of uranium ore resources in Europe and has accumulated a lot of radioactive materials and wastes from the uranium ore production and processing (tails, technological structures, and radioactive landfills) associated with the former USSR [12]. This inventory is a real risk and there is a recognized need to be able to identify the "radioactive material story" for the purpose of nuclear forensics – its origin, composition, place of extraction or production.

Illegal incident and trafficking of radioactive materials around the world are recorded in a special database – Incident and Trafficking Database (ITDB), which was created by the IAEA after several serious cases of smuggling in 1995 [13, 14]. Information that is collected and analyzed is sent to IAEA member states and relevant international organizations. Participation in this program is voluntary. Currently, 134 states, including Ukraine, provide their data to this information base.

So, for a long time since 1995, more than 3,100 cases of unauthorized trafficking of nuclear and radiation materials were recorded in the ITDB database [14]. In addition, there are regular cases of theft of radioactive materials used in medicine and industry, which can be used to create a "dirty bomb".

As it is well known, "dirty bombs" are a potential weapon of terrorists. They are not used by regular armies in the world. "Dirty bombs" are conventional explosives combined with radioactive material that can be used by terrorists under certain conditions to cause panic among the population [1, 15]. The uranium mining and milling sites along with waste dumps and available radioactive equipment can also be considered as a source of potential radiation risk if to consider a “dirty bomb case” [12].

Mineral composition, localization and distribution of ore mineralization, chemical composition of ores can be used as identification signs for nuclear forensics tasks. Uranium ores from the three deposits – Central, Michurinske, and Vatutinske – were examined for their identification signs. Among these three deposits, the ores of the Vatutinske deposit are characterized by a sharply increased content of Ra, Th, Pb, Po, and U, while the Central deposit is turned out to have the lowest content of those elements. These significant differences in the ores’ chemical composition can serve as one of the important identification (characteristic) signs [16, 17].

According to the sampled data, an average content of uranium in the Vatutinsk deposit is 0.13%. The presence of neodymium in the ores can be considered as one of the signs of the Vatutinske deposit uranium ores. Especially neodymium (Nd) accumulates in nenadkevite (0.12…0.8%) and branerite (0.58%). The average content of uranium in the Michurinsky deposit ores is 0.08%, and the content of thorium there is 0.0056%. That is, uranium ores are almost thorium-free. Zirconium can be considered one of the main typomorphic elements of uranium ores. The highest content in ores is noted for cerium ($107.9 \cdot 10^{-4}$%) and neodymium ($59.1 \cdot 10^{-4}$%). However, due to the higher content in uranium minerals – branerite (0.69% cerium, 0.47% neodymium) and uranium ferropseudobrookite (0.41 and 0.21%, respectively), these elements can be identifying signs in uranium ores processing products. The average content of uranium in the Central deposit is 0.1%. Thorium content is negligible (0.002%). Uranium titanosilicate (8…15% SiO_2) was also found. The ores are characterized by titanium oxide (TiO_2) (twice as much as in the Michurinske deposit) and zirconium oxide ZrO_2 (almost twice as much). The content of neodymium in the ore exceeds its content in the ore of the Michurinske deposit [17, 18].

Such studies are very important from the point of environmental and radiation safety for uranium mining and milling sites as well as for uranium legacy sites. Identified impurities are expected in production waste, which are considered as man-made and technologically enhanced naturally occurring radioactive materials, and may affect the components of the environment.

2. URANIUM LEGACY SITES IN UKRAINE

Ukraine has accumulated a great deal of research and practical experience and a significant amount of archival material on geology of uranium and related elements in the deposits of the Ukrainian Crystalline Shield (UCS) and its slopes [19–21].

The Ukrainian Shield occupies the axial (northern, central, and southwestern) part of the territory of Ukraine, stretching from northwest to southeast for almost 1,000 km. According to the latest data, a total of 357 radioactive sites (uranium and thorium) have been counted within the territory of the UCS to date, including 39 ore deposits, 298 ore occurrences, 20 ore manifestations of mineralization [22]. Our research is concentrated in the central part of the UCS, where the largest number of natural uranium deposits among the listed ones was discovered and studied.

However, due attention was not and is not being paid to the issues of radiation and environmental contamination of territories where there are sources and potential risk from TENORM. Such territories often contain waste from mining and primary processing of ores, specific impurities of chemical, including radioactive elements are found. They should be considered as sources of TENORM, which can be potentially used as radioactive material for making a dirty bomb and have an impact not only on the environment components, but also serve as a terrorist threat.

Due to certain objective reasons (which are not considered here), in numerous European publications (IAEA, UMREG, etc.) regarding the territories affected as a result of mining and processing of radioactive raw materials, uranium legacy sites of Ukraine are met extremely rarely [23]. This study draws attention to the radiation and environmental safety for the potential uranium legacy sites of Ukraine, where, since the Second World War until now, uranium mining and milling is in progress.

All uranium facilities in the country were operated and still are under operation according to the standards that do not meet the level of protection required by the European standards. And these facilities also fall within the scope of international safety standards as sites of radioactive contamination. In the event that the requirements for bringing the site to a safe state (by an operator of active production) are not fulfilled, then all sites must be transferred to a category of legacy – “existing exposure”. Legacy sites require identification according to certain criteria established by the Regulatory Authority, which must determine compliance with requirements and criteria for security management at legacy sites and planning measures to bring them to a safe state.

In Ukrainian legislation, there is no clear definition of “legacy site”, as well as fixed procedures for bringing the site to a safe state. Therefore, this study emphasizes the necessity of science-based measures to determine the territory of uranium legacy in Ukraine based on the signs of potentially hazardous objects (post-uranium legacy sites).

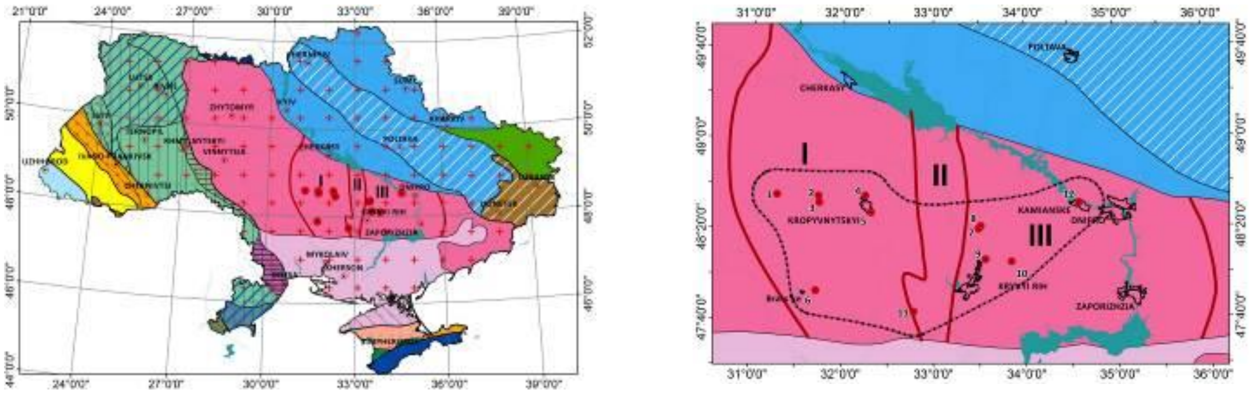
The classification of uranium legacy sites was proposed in [23] based on types of activities and signs of potentially hazardous objects having TENORM (Table 1).

Table 1

Uranium mining and milling facilities legacy sites of Ukraine

Legacy sites of depleted uranium deposits		
Name of legacy site	Location	Notes
Pervomaiske deposit	Central part of the UCS	Depleted through underground mining
Zhovtorichenske deposit	Central part of the UCS	Depleted through underground mining
Devladivske deposit	Southern Slope of the UCS	Depleted through underground leaching
Bratske deposit	Southern Slope of the UCS	Depleted through underground leaching
Legacy sites of operating uranium mines		
Ingulska mine	Central part of the UCS	Operates two deposits
Smolinska mine	Central part of the UCS	Operates Vatutinske deposit
Novokostyantynivska mine	Central part of the UCS	Operates Novokostyantynivske deposit
Legacy sites of milling facilities		
Prydniprovskyi Chemical Plant	Dnipropetrovska oblast	Closed in 1990, waiting for remediation
Zhovti Vody “Skhid GZK”	Dnipropetrovska oblast	Operating facilities

The research area of approximately 260 km length and of 125 km width is shown on Fig. 1,a,b. It includes the main uranium ore mining and milling facilities discovered and operated since 1945 when the Pervomaiske deposit was first discovered. All identified areas fall into the central part of the Ukrainian Shield.



a *b*

Fig. 1. Location of uranium legacy sites in the Central part of the Ukrainian Shield: I – Ingulskyi megablock; II – Kremenchuk-Inguletska interblock suture zone; III – Prydniprovskyi megablock

To consider the concept of a uranium legacy site (“radiation legacy sites” and/or “uranium legacy sites”) two aspects have to be taken into account: the first – anthropogenic activity either in the past or bearing traces of long-term affect – “affected by past practices”, where the level of radioactivity exceeds the background (equivalent dose rate reaches 350 μSv/h); the second – environmental components are characterized by a high content of uranium (rocks – up to 40…53 g/t; soils – up to $(0.5…1.9)\cdot10^{-4}$%; water – $5\cdot10^{-6}…9\cdot10^{-2}$ g/l), its decay products, and associated elements. However, each outlined location differs in terms of the potential risk rate to the environment, including sources of TENORM and approach to environmental monitoring and further remediation techniques.

The area under consideration is potentially radon-hazardous. For each territorial unit the question of its zoning according to the level of radon hazard is an issue of the near future. The priorities should be considered within the boundaries of municipalities or integrated territorial units. Determination of radon risk rate should become the basis for educational and awareness-raising work for the population about the radon potential of their residential territories.

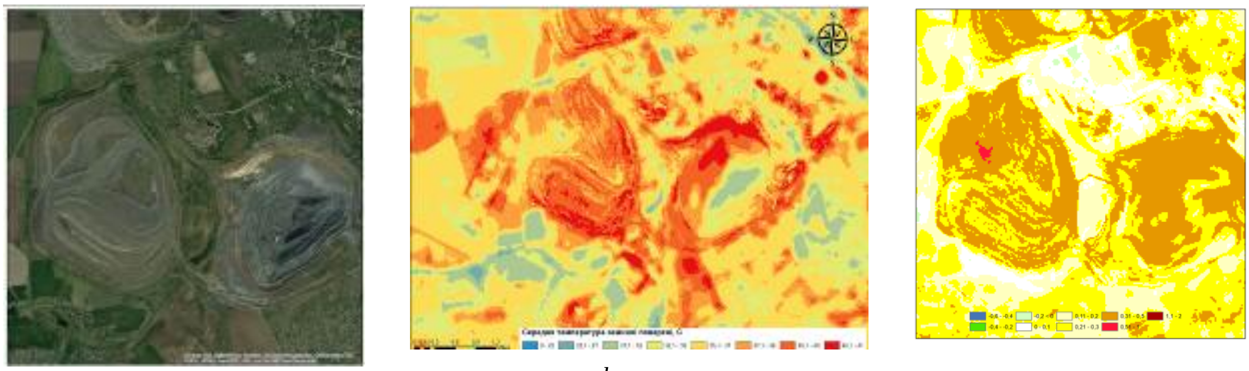
It should be noted that each location of potential uranium legacy site in Ukraine is a unique one if to consider its location in densely populated areas of predominantly fertile black soil, where agricultural activity is widely developed. Let's consider the territory of the unique Pervomaiske deposit site.

The Pervomaiske deposit site (*legacy site of worked-out uranium deposit of iron-carbonate-uranium ores*) is shown in Fig. 2. The deposit itself was opened right after the war in 1945 in the northern part of the city of Kryvyi Rih. Further study confirmed the discovery of the first large uranium deposit in the former USSR. Uranium ores were mined in 1968, but the mine continued to operate for rich iron ores.

Fig. 2. Location of the Pervomaiske deposit in the outskirts of the Kryvyj Rih city

At the Pervomaiske deposit, uranium mineralization was completely localized in commercial iron ores. It was found out that the ores had a carbonate-hematite-magnetite composition having interspersed uranium minerals. Therefore, these iron ores are simultaneously classified as uranium. Their composition is relatively simple: iron ore minerals (30…75%), carbonates and silicates (20…70%).

We conducted remote studies of the long-term dynamics of the earth's surface temperature within the area of research characterized by high radon rate (see Fig. 1). Data of the long-wave infrared range (8…14 μm) of the Landsat-8/OLI satellite, obtained for the period of 2013–2019, were used to obtain images of the land surface temperature (LST) distribution. The method of determining the temperature of the earth's surface is described in [24, 25]. In general, mining territories ate noted to have high average annual temperature and average annual growth of the LST, which is mentioned in [26]. A clear correlation of the depleted iron ore quarries and the uranium mining wastes with the zones of high average annual temperature and average annual growth of the LST at the Pervomaiske deposit is shown in Fig. 3.



a *b* *c*

Fig. 3. Location of the worked out Pervomajske deposit (a); average land surface temperature (b); annual average growth of the land surface temperature (c)

A detailed study of the mineralogy and petrography of ores and host rocks is of great importance for the development of technological schemes for the enrichment of uranium ores, their hydrometallurgical processing and subsequent impact on the environment components and methods of the territory reclamation. Unfortunately, geological information on uranium ores deposits was not sufficiently summarized and published due to the special regime of secrecy for radioactive raw material resources and remained scattered in various classified reports in the former USSR archives. So, the specified uranium deposits sites have to be additionally studied in terms of their radioecological situation in order to classify them as uranium legacy sites.

Uranium tailings available within uranium legacy sites in Ukraine are a waste byproduct (tailings) of uranium mining. In mining, raw uranium ore is brought to the surface and crushed into fine sand. The valuable uranium-bearing minerals are then removed via heap-leaching with the use of acids or bases, and the remaining radioactive sludge, called "uranium tailings", is stored in huge impoundments. Uranium tailings contain over a dozen radioactive nuclides, which are the primary hazard posed by the tailings. The most important of these are thorium-230, radium-226, radon-222 (radon gas) and the daughter isotopes of radon decay, including polonium-210 [27–29]. A lot of this waste is alpha particle-emitting matter from the decay chains of uranium and thorium.

Radioactivity is mainly associated with the uranium-238 family. The level of radioactivity of waste is different for different deposits and depends mainly on the initial content of uranium in primary ore, physical and chemical properties of the ores and host rocks, the meteorological conditions of the area, as well as the activity of geochemical processes occurring within the deposit site before its development [29, 30]. We are talking here about natural leaching, which causes the formation of different ratios of uranium to its decay products.

Based on the analysis of the situations at the proposed uranium legacy sites, a summary of the measures for classifying the sites in the "legacy" category was made, which are presented in Table 2. These measures are worth considering them to be used for future development of quantitative criteria for classifying the sites in the "legacy" category.

Table 2

Measures to refer the site to the "legacy" category of Ukraine

No.	Measures to refer the site to the "legacy" category
1	Determination of the equivalent dose rate of gamma radiation on the territory of a site – more than a background value (more than 0.15…0.20 μS/h)
2	Determination of radionuclides ^{238}U - ^{234}U - ^{230}Th - ^{226}Ra - ^{210}Po - ^{210}Pb, ^{230}Th in environmental components – soil, water, aerosols
3	Determination of the Rn-222 volume concentrations within the former industries sites, industrial and residential buildings, in residential areas
4	Determination of Rn-222 exhalation on the surface of tailings and wastes dumps, contaminated mining and other industries sites
5	Determination the accompanying elements content, the concentration of which exceeds the background for each site – for example, V, Sc, Cr, As, Ni for the territories of uranium mining sites for the deposits of albitite formation
6	Determination of radionuclides and toxic elements content in aerosols that are collected at industrial sites, personnel workplaces, residential areas and areas of earthworks and construction works

The programs of international technical cooperation aimed at providing assistance in implementation of rehabilitation projects are actively being developed in uranium related companies of the EU countries. The analysis suggests that effectiveness of such projects largely depends on the availability of appropriate national environmental safety strategies, regulatory requirements and regulatory mechanisms, as well as experience in managing similar projects in accordance with international standards. Such activity requires a lot of financial support and fruitful cooperation between governmental institutions (including the research ones), the EU, international organizations, SE "ShidGZK", local self-government bodies and non-governmental organizations.

CONCLUSIONS

Drawing on case studies, mining and milling facilities sites where TENORM sources are available, should be considered more broadly than just uranium waste. These are sources of ionizing radiation of natural origin that were subjected to concentration or their accessibility was increased as a result of human activities (mineral raw materials mining, milling, enrichment; water treatment and purification, etc.). As a result additional to natural radiation exposure is formed. In the future, due attention should be paid to the issues of environmental and radiation safety of territories where sources of naturally occurring radioactive materials are available.

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ТЕРИТОРІЇ ВИДОБУВАННЯ І ПЕРЕРОБЛЕННЯ УРАНОВОЇ РУДИ ЯК ТЕХНОГЕННО-ПІДСИЛЕНІ ДЖЕРЕЛА РАДІАЦІЇ ПРИРОДНОГО ПОХОДЖЕННЯ

Т.В. Дудар

Наведено дані щодо бази даних уранових родовищ для ідентифікації радіоактивного матеріалу та джерел природної радіації в межах Українського кристалічного щита. Території видобування і перероблення уранової сировини розглядаються, з точки зору техногенно-підсилених джерел радіоактивності природного походження, за запропонованою класифікацією потенційних об'єктів уранової спадщини. Представлено заходи щодо визначення об'єктів уранової спадщини України.