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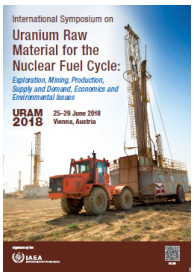

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ENVIRONMENT ASPECTS OF Th-230 ACCUMULATED IN RESIDUES COMPONENTS AT THE URANIUM PRODUCTION LEGACY SITE PRIDNEPROVSKY CHEMICAL PLANT

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INTRODUCTION

This paper presents the results of recent studies carried out at the former Uranium production facility Pridneprovsky Chemical Plant (PChP) in Ukraine and describes specific activity concentrations of radionuclides of U-Th decay series with specific focus on identified high activity concentrations of 230Th associated with different environment compartments, such as soils, aerosols, bottom sediments and U-production residues accumulated in the tailings and uranium extraction facilities remained at the PChP site.

The Production Association "Pridneprovskiy Chemical Plant"(PChP) was one of the largest facilities of the military complex of the former Soviet Union, where production of uranium for the Soviet atomic programme was carried out starting in 1949 till 1992. The plant is situated in a densely populated area in the industrial zone of Kamyanske town (formerly Dniprodzerzhynsk) and close located to the Dnieper River.

The processing procedures for uranium ores at the "PChP"were typical for technologies used in the former USSR, including grinding, hydrometallurgical extraction, sorption, radiochemical separation and purification of U-concentrate from radium and thorium impurities. Sulphuric and nitric acid were used for leaching of the uranium containing ores of different origin from deposits located in Ukraine and also Germany and Check republic. Since 1984 the phosphorus ore from Kazakhstan (Melovoe deposit), containing uranium were processed as well. The phosphorus ore processed at the site are considered as a main source of thorium-230, which currently dispersed at the PChP site.

Since 1992, when PChP uranium industrial production complex was ceased, no measures have been taken to decommission and remediate uranium production facilities. The ambitions remediation planning activities were initiated during recent years with financial assistance of EC and funded by also several national remediation projects. In order to provide safety assessment and collect data for remediation planning the extended site characterization and monitoring programs were carried out at the PChP legacy site during recent decade, including detailed spatial gamma-dose survey and assessment of contamination status of the environment by radionuclides of U-Th decay series and safety conditions at the uranium tailings facilities [1].
Specific interest to the specific activities and chemical speciation of 230Th at this site was determined by presents of huge amount of thorium containing materials at the site (up to 6 ton of thorium concentrate produced annually due to uranium ore purifying process) and its relatively high activity concentrations observed at the many locations of PChP site, which is exceeding level of exclusion from regulatory control such as 1Bq per gram, as well as its high radio-toxicities in the environment.

METHODS AND RESULTS

The methods used in this study consist of several components such as: historical analyses of the technologies, which determined existing state of thorium-230 in a complex of other radionuclides of U-Th series and its physical and chemical speciation in the affected environment; application of analytical methods used for determination of thorium isotopes and other radionuclides of U-Th decay series in soil, aerosols and tailings material samples using modern gamma and alpha-spectrometry methods with radiochemical supporting procedures [2] and also dose assessment methodology, providing site specific safety assessment and estimating

contribution of thorium presence in the present safety conditions forming at the legacy site and remediation strategy planning. Methods for remediation of the high contaminated environment as well as strategy for management of remediation wastes containing thorium and other radionuclides of 238U decay series are discussed as well.

Determination of the radioactivity of thorium isotopes in different environmental entities such as soil spill and sludge materials, dust, bottom sediment and water samples, which were collected at the PChP legacy site, have been carried out in the frame of state remediation and site specific monitoring projects.

In general, activity concentrations of thorium isotopes (230Th, 234Th, 232Th, 228Th) were quantified by gamma spectrometry analysis, using a high purity Germanium detector coaxial n-type HP Ge detector GMX (Ortex) with 40% relative efficiency and resolution of 1.8 keV for 1,332.5 keV 60Co energy. The detector efficiency was determined using a mixed standard solution of known activity containing 152Eu, 241Am, 226Ra in fixed sample geometry. The emission gamma spectrum was analysed using GammaVision32 application software in according to UNI 10797:1999 with sealing the beaker.

The concentrations of 230Th were measured directly from their gamma peak at 67.67 keV with emission probabilities 0.38%. Activity 234Th daughter radionuclide of the 238U was determined by its gamma peaks at 63.29 keV with emission probabilities of 3.7 %. For the determination of 232Th, assuming secular equilibrium in the sample, the gamma lines of 911.2 keV, 338.4 keV with emission probabilities of 26 %, 11 %, respectively.

The calibration quality control was carried out by means of a soil standard sample SRM (IAEA-434), IAEARGU-1 and IAEA-RGTh-1 whose concentrations of the main natural radionuclides have been certified by the IAEA in the same geometry the measurement. Quality checking of determination of Th isotopes in the UHMI laboratory has been regularly tested by IAEA Proficiency Tests (for instance as it was published in IAEA-CU-2008-03: "Determination of naturally occurring radionuclides in phosphogypsum and water").

To verify the accuracy of the determination of 230Th and other thorium isotopes by gamma spectrometry the alpha-spectrometry method with radiochemical pretreatment was used. Radio-analytical procedures consists of chemical dissolution of samples, chemical separation of thorium by ion-exchange method, preparation of counting source by electro-deposition method and measurement of thorium by alpha spectrometry method. Thorium in water sample was separated by co-precipitation method. For determining the chemical yield the 229Th tracers were used. The relative errors were lower than 20 % between both analytical techniques used. The basic set of monitoring data collection and site characterization analyses carried out in UHMI during recent decade, contains long-term time series specific activity concentrations of radionuclides of U-Th decay series (such as: 238U, 234U, 230Th, 226Ra, 210Pb, 210Po), containing in the U-ore raw materials, production residues and contaminated environment at the site, which may vary in wide range between less of 1.0 Bq/g to 3-4 thousands Bq/g.

Special attention in this study is given to the analyses of spill and sludge materials, containing high activity concentration of thorium, which are in large amount still accumulated in some buildings used for thorium removal and at the sludge pond accumulating complex compound of the thorium fractions before its transportation to the storage facility. Since different technologies used for purifying uranium concentrates from thorium impurities have been applied, different concentrations of thorium are contained in various objects of the uranium production legacy site, which vary from several Bq/g to hundreds of Bq/g in the environment.

The background activity concentrations of 230Th outside of PChP site vary in range 0.04-0.07 Bq/g. High activity concentrations of 230Th were found in many locations of the PChP legacy site and in particular in soils and also spill materials accumulated in the former U-extraction facilities and buildings used for removal and purifying of uranium concentrates from thorium contained impurities. In many locations high 230Th activities were observed in the residue production in range between several tens to several hundred Bq per gram. In some locations the horizons of soils with highest 230Th activities concentrations are located at the soils top surface, while at other places the layers with maximal activities are covered with clean soils with relatively low contamination and radionuclides of U-Th series identified on a depth up to 1.5 m. Results of vertical profile studies of radionuclide U-Th series of different origin are discussed in this report.

The ratios between 238U, 230Th and 226Ra in soils at the different locations, which may characterized the impacts of the uranium production residue by different origin and technologies applied for purifying uranium containing ores are presented in paper as well. Sufficient amount of residue materials from U-production, containing high activity concentrations of 230Th, were identified at several cells of sedimentation sludge ponds. In the lower layers of sludge materials accumulated in sedimentation pond, specific activities of thorium-230 were found in range from 10 to 170 Bq/g. Other naturally occurring radionuclides in the most of samples taken from the sludge columns were characterized by specific activity in range value 1Bq/g and less. Some by-products of uranium productions, containing thorium need for defining the regime of regulatory control, because thorium in sufficiently high activities identified in some NORM product, produced at the site is a significant limiting factor in further use of these materials. The surface layer of sludge in sedimentation pond also has high levels of radium-226 and other radionuclide and chemical pollutants.

Specific activity concentration in the spill materials remained on the floor and in some filled tanks with dry

residue of the complex radiochemical solutions 226Ra activity concentration may reach several hundred Bq per gram of dry samples. In the spill materials of "yellow cake"production found in several locations of the Building 103 high concentrations of Uranium were dominated reaching several thousand Bq per gram of dry samples and relatively low 230Th activity concentrations.

Specifically high activity concentrations of 230Th in spill materials were found in several locations of Building 104m which was used for thorium removal from phosphorus ores. Specific activities of 226Ra and 210Pb in such spill materials varied in relatively low value range from 2-3 Bq/g up to 20-30 Bq/g. The same samples were characterized Th-232 in range of activity concentrations between 2,3-4.4 Bq/g. In spite of relatively low activity concentration of uranium and radium isotopes were found, 230Th activity concentrations in such spill materials are reached in some locations 200-600 Bq/g. Such dispersed materials presented in the dust fractions and having very high thorium activity concentrations comparing with exemption level such as 1Bq/g, may pose a significant risk of inhalation exposure for remediation workers. Therefore detailed radiation protection plan and possible decontamination of these buildings before dismantling of most contaminated equipment and demolishing are strictly required.

The levels of Th-230 activity concentration in aerosols were measured just in limited number of samples collected in the contaminated buildings and at the surrounding areas. The results of the measurements showed that in the buildings where the uranium concentrate from phosphorus ore was purified, the content of thorium in dust and aerosols exceeded the activity of uranium and radium by 10-100 times. In other buildings of uranium extractions after leaching and purifying, the thorium-230 in the production residues and tailings materials were found in much more lower activity concentration with domination of radium-226.

The variety of conditions and forms of radionuclides of uranium-thorium series at various sites of uranium production heritage determines also the variety of radiological risks of irradiation for personnel in the territory of the former uranium production. At most sites and sites of the uranium object, the main contribution to the radiation dose is determined by direct gamma irradiation, which is mainly formed by radium 226.

The averaged doses for 1 hour presence in the most contaminated facilities were estimated in wide range between 3-10 μSv/h (at the locations with low concentration of the radioactive residues) and up to 1 mSv/h in some buildings near tanks where high activity concentrates of 226Ra in the residues is still stored. The main dose factor in such buildings is high gamma dose rate. In the facilities characterized by high thorium-230 contamination, the highest doses were created by gamma dose exposure as well and also relatively high Rn-222 ambient activity air concentration. In general contribution of inhalation exposure due to aerosol particles containing radionuclides of U-Th series is estimated as no more than 10% of the total exposure dose. However in some specific cases maintenance with spill materials contained thorium with high activity concentrations in spill and dust materials may created sufficient inhalation dose exposure for remediation workers.

In previous assessments at the stage of the safety assessment, it was assumed that the uranium, radium and thorium contents in dust and aerosols are approximately equilibrium. In fact, as has been shown by our investigations, the content of thorium-230 in dust and materials that can form aerosol contamination in some buildings and in storage facilities for the residues of thorium concentrates can be tens and hundreds of times higher than for equilibrium conditions. Therefore, the dose estimates of inhalation exposure, which were obtained at the stage of rehabilitation measures planning, taking into account the high radio-toxicity and dominance of thorium in some former uranium production facilities, can be significantly underestimated. The results of assessment and actions proposed to reduce risks are discussed as well.

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