MEASUREMENTS OF URANIUM RADIONUCLIDES IN URINE SAMPLES AND ASSESSMENT OF INTERNAL DOSES IN URANIUM-MINING INDUSTRY IN UKRAINE

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Abstract

The State Owned "Eastern Mining And Processing Complex" (SkhidGZK) was established in 1951. The SkhidGZK is the only enterprise in Ukraine engaged in the mining of uranium ore and in the production of a uranium oxide concentrate. Several thousand workers of the SkhidGZK have a risk of incorporation of uranium radionuclides.

The first attempts to determine content of uranium radionuclides in urine of the SkhidGZK personnel were started in 2009. 108 daily urine samples from 7 workers were collected. Measurements of ^{234}U and ^{238}U contents in samples were performed.

All results were in range from 3.6 to 766 mBq per sample. As expected, measurements demonstrate a steady correlation between contents of uranium radionuclides in urine. Average ratio ^{234}U:^{238}U is 1.004. Geometric means are 50.5 and 50.3 mBq per sample for ^{234}U and ^{238}U respectively.

Internal doses were calculated on the base of measurement results. Assessments of committed annual doses from inhalation intakes of uranium and thorium radionuclides (as well as 'accompanied' progenies in air) are very depend on assumption about Type of materials.

Keywords: uranium, individual monitoring, internal exposure.

1. INTRODUCTION

The State Owned "Eastern Mining and Processing Complex" (SkhidGZK) was established in 1951. Now it is the largest uranium mining enterprise in Europe. The SkhidGZK is the only enterprise in Ukraine engaged in the mining of uranium ore and in the production of a uranium oxide concentrate. Several thousand workers of the SkhidGZK have a risk of incorporation of uranium radionuclides.

The spectrum of ionizing radiation of uranium radionuclides significantly complicates the direct determination (by whole body counters) of these radionuclides in the human body. This leads to the use of measurements of uranium radionuclides in bioassay samples to identify their intakes and to assess the doses formed due to their incorporation.

Pilot project for measurements of uranium radionuclides in urine samples of the SkhidGZK personnel was started in 2009. It was a first attempt at the SkhidGZK to determine content of uranium radionuclides in urine of personnel and to perform internal dose assessments on the base of bioassay samples. A pilot measurement of uranium radionuclides in urine samples is the first step for the

organization of individual monitoring of internal exposure at the SkhidGZK.

2.   MEASUREMENTS OF URANIUM RADIONUCLIDES IN URINE SAMPLES

Urine samples were collected by seven workers from the calcination shop of the SkhidGZK. Urine was collected in hermetically sealed glass containers with a capacity of 2.0 L with the addition of 50 mL of nitric acid as a preservative. Sampling was carried at home during two days of rest after two or three working shifts. Average daily volume of bioassay sample was 1.5 L.

108 daily urine samples were collected during 11 months. Measurements of ^{234}U and ^{238}U contents in samples were performed. Summary of measurement results are represented in Figure 1.
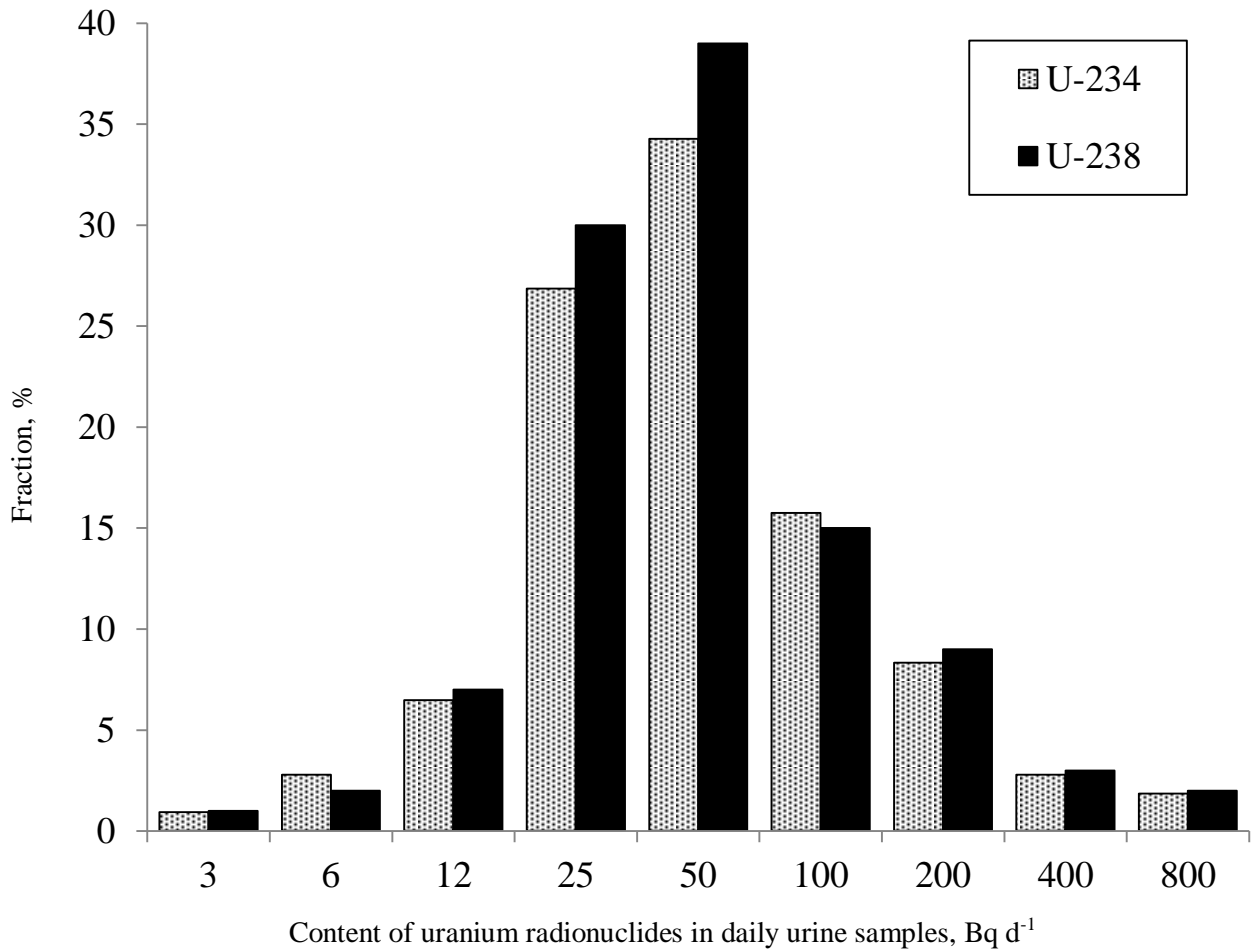

FIG.1. Distribution of measurements of ^{234}U and ^{238}U contents in daily urine samples, Bq d^{-1}

All results are in range from 3.6 to 766 mBq per sample. As expected, measurements demonstrate a steady correlation between contents of uranium radionuclides in urine. Average ratio ^{234}U:^{238}U is 1.004. Geometric means are 50.5 and 50.3 mBq per sample for ^{234}U and ^{238}U respectively. σ_g is 2.54 for both radionuclides.

3.   BASIC INITIAL DATA FOR ESTIMATION OF INTERNAL DOSES

The composition of natural uranium which is given in Table 1 was used to perform the dose calculations.

TABLE 1. COMPOSITION OF NATURAL URANIUM

Radionuclide	Mass fraction, %	Specific activity, kBq per g of U
^{234}U	0.0054	12.5
^{235}U	0.71	0.57
^{238}U	99.28	12.3

Intake of the above mentioned radionuclides is accompanied by progenies from radioactive decay chains of ^{235}U and ^{238}U (^{234}U is a progeny of ^{238}U). In addition, ^{232}Th occurs in uranium ores. For example, the ratio of the mass fraction of ^{232}Th to the natural uranium varies from 1.4 to 1.8 in the ores that are mined at the SkhidGZK. So, ^{232}Th and its progenies should be taken into account too.

Radon radionuclides and their progenies were excluded from the consideration. Therefore only the following radionuclides were considered: ^{238}U, ^{235}U, ^{234}U, ^{234m}Pa, ^{231}Pa, ^{234}Th, ^{232}Th, ^{231}Th, ^{230}Th, ^{228}Th, ^{227}Th, ^{228}Ac, ^{227}Ac, ^{228}Ra, ^{226}Ra, ^{224}Ra, ^{223}Ra.

For the calculations it was assumed that the equilibriums in all considered radioactive decay chains (^{235}U, ^{238}U, ^{232}Th) are reached. On this assumption relative activities of radionuclides (comparatively to ^{238}U) will be the values presented in Table 2 (all values are rounded to 2 significant digits).

TABLE 2. RATIO OF RADIONUCLIDE ACTIVITY TO ACTIVITY OF ^{238}U

Radionuclide	Ratio to ^{238}U
^{234}U, ^{234m}Pa, ^{234}Th, ^{230}Th, ^{226}Ra	1.0
^{232}Th, ^{228}Th, ^{228}Ac, ^{228}Ra, ^{224}Ra	0.22
^{235}U, ^{231}Pa, ^{231}Th, ^{227}Ac, ^{223}Ra	0.046
^{227}Th	0.045

Inhalation intakes of different type of chemical compounds were examined. Correspondence between the uranium compounds and their 'solubility' is the following [1, 2]:

- Type F materials: Deposited materials that are readily absorbed into blood from the respiratory tract deposited materials that are readily absorbed into blood from the respiratory tract (most hexavalent compounds, e.g. UF_6, UO_2F_2 and $UO_2(NO_3)_2$);

- Type M materials: Deposited materials that have intermediate rates of absorption into blood from the respiratory tract (less soluble compounds, e.g. UO_3, UF_4, UCl_4, and most other hexavalent compounds);

- Type S materials: Deposited materials that are relatively insoluble in the respiratory tract (highly insoluble compounds, e.g. UO_2 and U_3O_8).

Type M is the default type for uranium radionuclides. However, for specific works the appropriate

material type should be chosen. There are two recommended Activity Median Aerodynamic Diameters (AMADs) for workers: 1 μm and 5 μm. However, due to only mechanical influence to the rock AMAD can be larger, e.g. 10 μm. On the contrary, during works with thermal or chemical effects on materials AMAD can be much smaller. Therefore, the calculations were performed for a wide range of aerosol particle sizes (for AMADs from 0.01 to 10 μm).

4. MODELS AND APPROACHES FOR DOSE ESTIMATION

General models which were used for the modelling of biokinetic processes and for the dose assessment are described in [1, 2]. Using the data about the type of inhaled material (TM) and the size of aerosol particles (AMAD), the modelling of biokinetic processes and dynamics of daily urine excretion of ^{238}U were performed. Calculated excretion functions $u^{AMAD,TM}(t)$ are ratios of radionuclide activities that excreted with urine after time t after intake to the intake values. The excretion functions $u^{AMAD,TM}(t)$ for materials of Type F and Type M are shown in Figure 2.
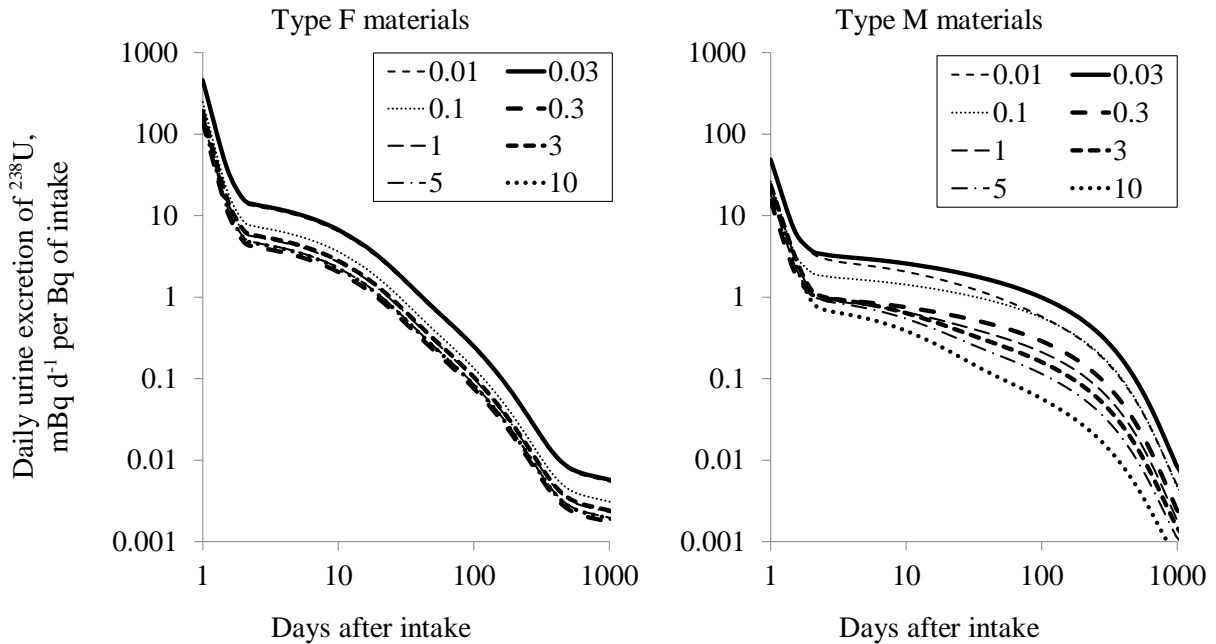

FIG.2. Daily urine excretion of ^{238}U (mBq d^{-1}) after acute inhalation intake of 1 Bq of ^{238}U (AMADs from 0.01 to 10 μm)

According to the radionuclides ratios during intakes estimations of the relative contribution of radionuclides to the total internal dose were performed. It should be taken into account that the structure of such contributions substantially depends on 'solubility' of compounds. Estimation for contributions of radionuclides to the total internal doses is represented in Table 3.

TABLE 3. CONTRIBUTION OF RADIONUCLIDES TO THE TOTAL INTERNAL DOSE, %

Radionuclide	Type F materials	Type M materials	Type S materials
^{230}Th	43 [a]	48	25
^{227}Ac	27	12	5.8
^{232}Th	10 [a]	11	9.9
^{228}Th	7.2 [a]	8.1	16
^{231}Pa	6.4 [a]	7.2	2.8
^{226}Ra	3.4 [a]	3.8	6.1 [a]
^{234}U	< 1	3.8	16
^{238}U	< 1	3.1	14

[a] Type M materials were used for dose estimation

The above described variants are provided to imagine how components of the formation of internal exposure depend on the types of compounds at the workplace. It is possible that measurement results are formed by different kinds of intake (chronic or acute) or/and by different types of compounds (slow or fast 'soluble'). For example, it is possible when there is a chronic intake of low values of Type M materials, against which there are substantially higher acute intakes of Type F materials.

As described above, daily urine excretion of ^{238}U were calculated according [1, 2]. For the purposes of retrospective dosimetry of internal exposure the 'effective dose per unit measurement' (DPUM) functions were used. The DPUM functions $z^{AMAD,TM}(t)$ obtained as follows [3]:

$$z^{AMAD,TM}(t) = e^{AMAD,TM} / u^{AMAD,TM}(t),$$

where $e^{AMAD,TM}$ is the effective dose per unit of inhalation of radionuclide calculated for the Type of material TM and AMAD, Sv·Bq^{-1};

$u^{AMAD,TM}(t)$ is the function of daily urine excretion of the radionuclide calculated for inhalation intake of 1 Bq of the radionuclide (for the same TM and AMAD), Bq·Bq^{-1}.

The functions $z^{AMAD,TM}(t)$ allows to convert the radionuclide content in the body (or in the sample) to the effective internal dose on the base of data about inhalation (TM and AMAD of inhaled aerosol, and time after intake). As shown in [3] and as seen in Figures 2 and 3, $z^{AMAD,TM}(t)$ functions have significantly less variability (in contrast to the $u^{AMAD,TM}(t)$ functions and to the $e^{AMAD,TM}$ values).

The functions $z^{AMAD,TM}(t)$ that are total effective internal doses per 1 mBq content of ^{238}U in daily urine samples, depending on time after acute intake, are shown in Figure 3 (for Types F and M). These functions take into account the contribution to the total effective internal doses of all radionuclides presented in Table 2.
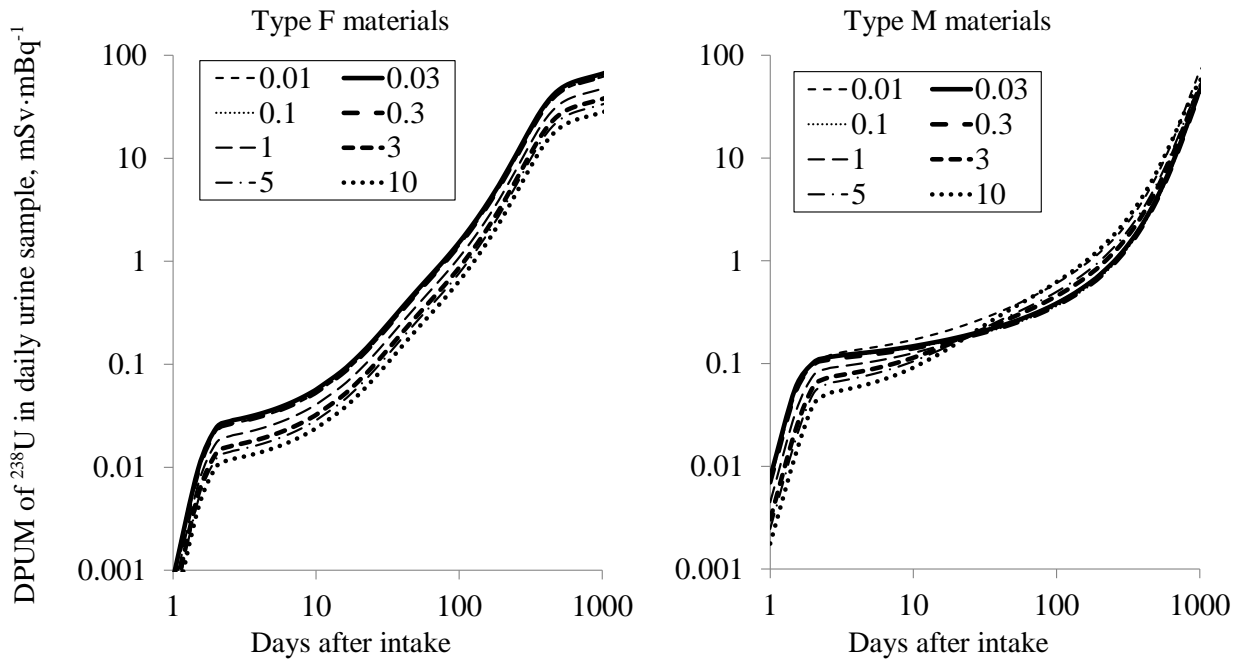
FIG.3. Total effective internal dose per unit content of ^{238}U in daily urine sample depending on time after acute intake (AMADs from 0.01 to 10 µm)

5. ASSESSMENT OF INTERNAL DOSES

Assessments of internal doses were performed using the IMIE software [4]. The IMIE software uses the DPUM functions for the retrospective approximation of dynamics of measured results and for the dose assessment. Example of such an approximation for Type F is represented in Figure 4.
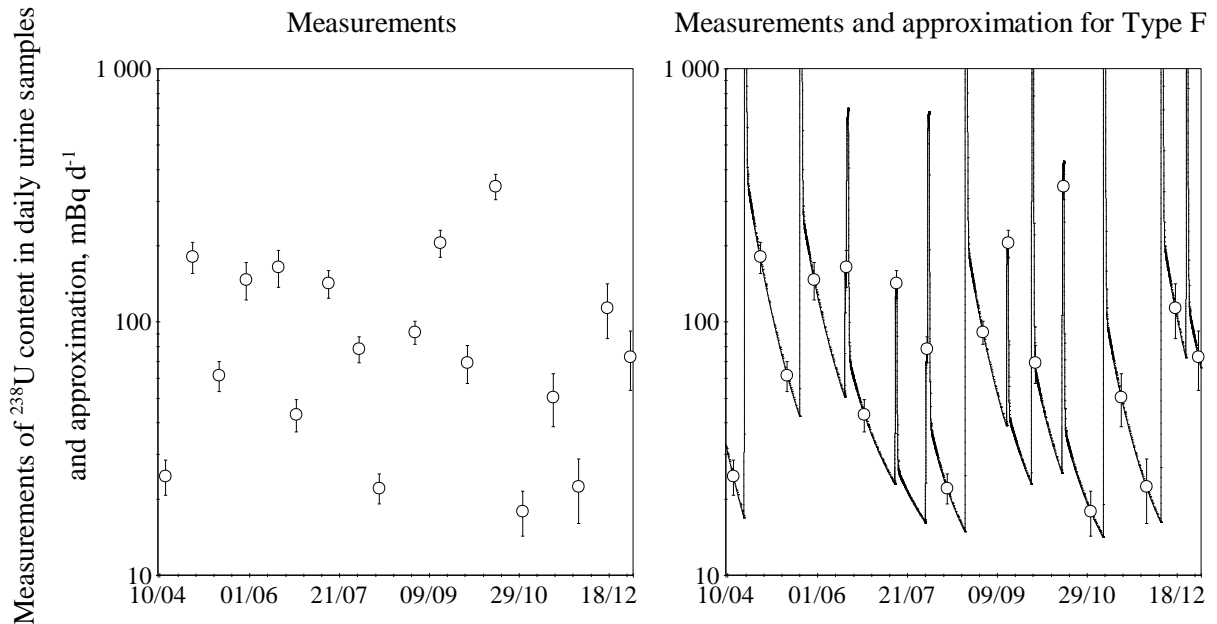

FIG.4. Measurements of ^{238}U content in daily urine samples and approximation, mBq d^{-1}

According to the measured dynamics of ^{234}U and ^{238}U in urine samples it seems unlikely that measured contents are the results of intakes of only slow soluble uranium compounds (type S). But such a

possibility should not be rejected, especially that such an assumption leads to internal doses of several tens of mSv.

The assumption about inhalation intakes of Type M materials is more likely for all workers. In such circumstances, most internal doses for examined workers are in the range from 10 mSv to 20 mSv (maximal dose is about 50 mSv).

The most likely assumption (for measured results and assumption about intakes of the same type compounds) is inhalation intakes of fast 'soluble' compounds (Type F). With this assumption the internal doses of 7 examined workers are in range from 8.3 mSv to 40 mSv. The average internal dose is about 20 mSv.

6.   CONCLUSION

1. Inhalation of Type F materials (fast 'soluble' compounds) is the most likely assumption about intake on the basis of 108 examined urine samples. With this assumption the internal doses of 7 workers from the SkhidGZK are in range from 8.3 mSv to 40 mSv. The average internal dose is 20 mSv.

2. Information about Type of inhaled material is the most important initial data that can significantly reduce the uncertainty of internal dose assessment for workers in the uranium mining and milling industry. Information about aerosol particle sizes (AMAD) at the workplaces has less significance.

3. The use of the 'effective dose per unit measurement' (DPUM) functions in a retrospective analysis helps to reduce uncertainty of internal dose assessments for workers in the uranium mining and milling industry. The uncertainty associated with lack of knowledge about AMAD almost completely eliminated by the use of the DPUM functions.

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