SCREENING THE ACTIVITY OF INCORPORATED RADIONUCLIDES IN THE RESEARCH ORGANIZATION EMPLOYEES

Turlakov YuS, Grabsky YuV, Arefeva DV™, Shayakhmetova AA, Firsanov VB, Petushok AV

Scientific Research Institute of Marine and Industrial Medicine of the Federal Medical and Biological Agency, St. Petersburg, Russia

Studying the features of radiological situation in the workplace and assessing the individual effective doses in employees of research organizations working with open radiation sources (RS) are an urgent scientific task due to additional risks resulting from variability in the conditions and regimes of the technological operations. The study was aimed to assess the working conditions and intake of radionuclides by the employees of the V. G. Khlopin Radium Institute working with open RS. The data on exposure to work-related radiation factors were obtained by dosimetry, radiometry, and spectrometry. It was found that radiological situation in the employees' workplaces was characterized by the broad range of the gamma ambient dose equivalent rate values (0.10–122 µSv/h), alpha and beta working surface contamination, radioactive pollution of air in the working areas. In some individuals, spectrometry revealed the following: ¹²⁵I in the thyroid gland (up to 9,850 Bq), ⁹⁰Sr in the skeleton (up to 16,500 Bq), ¹³⁷Cs in the whole body (up to 1,100 Bq), etc. The findings can provide the basis for developing the measures to improve the quality of individual internal dose control and the efficiency of medical care provision to the research organization employees dealing with open RS.

Keywords: internal exposure, personnel, radiation factor, working conditions

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Correspondence should be addressed: Daria V. Arefeva

Yuri Gagarin prospect, 65, liter A, St. Petersburg, 196128, Russia; niipmm.210@gmail.com

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СКРИНИНГ АКТИВНОСТИ ИНКОРПОРИРОВАННЫХ РАДИОНУКЛИДОВ У ПЕРСОНАЛА НАУЧНО-ИССЛЕДОВАТЕЛЬСКОЙ ОРГАНИЗАЦИИ

Ю. С. Турлаков , Ю. В. Грабский, Д. В. Арефьева , А. А. Шаяхметова, В. Б. Фирсанов, А. В. Петушок

Научно-исследовательский институт промышленной и морской медицины Федерального медико-биологического агентства, Санкт-Петербург, Россия

Исследование особенностей формирования радиационной обстановки на рабочих местах и оценка индивидуальных эффективных доз облучения персонала научно-исследовательских организаций при выполнении работ с открытыми источниками ионизирующих излучений (ИИИ) является актуальной научной задачей в связи с наличием дополнительных рисков из-за вариативности условий и режимов выполнения технологических операций. Целью работы было оценить условия труда и поступление радионуклидов в организм персонала АО «Радиевый институт им. В. Г. Хлопина», работающего с открытыми ИИИ. Данные об уровне воздействия производственных факторов радиационной природы на персонал получены в ходе дозиметрических, радиометрических и спектрометрических измерений. Установлено, что радиационная обстановка на рабочих местах персонала характеризуется широким диапазоном уровней мощности амбиентного эквивалента дозы гамма-излучения (0,10–122 мкЗв/ч), наличием поверхностного загрязнения рабочих поверхностей альфа- и бета-частицами, наличием загрязнения радиоактивными веществами воздуха рабочих помещений. В результате спектрометрических исследований обнаружено наличие у отдельных лиц из персонала ¹²⁵ в щитовидной железе (до 9850 Бк), ⁹⁰Sr — в скелете (до 16 500 Бк), ¹³⁷Cs — во всем теле (до 1100 Бк) и др. Результаты исследований могут быть основой для разработки мероприятий по повышению качества контроля индивидуальных доз внутреннего облучения и эффективности медицинского обеспечения персонала научно-исследовательских организаций, имеющего контакт с открытыми ИИИ.

Ключевые слова: внутреннее облучение, персонал, радиационный фактор, условия труда

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Для корреспонденции: Дарья Владимировна Арефьева

пр. Юрия Гагарина, д. 65, литер А, г. Санкт-Петербург, 196128, Россия; niipmm.210@gmail.com

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Active development of the nuclear industry and nuclear medicine results in the increase in the volume of work with radioactive substances, including open radioactive sources. When carrying out such work, employees are exposed to a combination of harmful and/or dangerous work-related radiation factors: gamma ambient dose equivalent rate, alpha and beta surface contamination, radioactive air pollution in the work areas,

which increase the risk of the radionuclide intake by workers. Objective assessment of internal exposure and forecasting its effects on the human health are an urgent scientific issue that remains poorly understood despite the progress made by experimental studies in this area and clinical trials [1, 2].

The use of open radioactive sources (RS) in scientific research poses additional risks due to variability in the

ОРИГИНАЛЬНОЕ ИССЛЕДОВАНИЕ І ЗДРАВООХРАНЕНИЕ

conditions and regimes of the technological operations depending on the tasks to be performed. According to the production control data, individual effective doses from internal exposure in employees of the companies and organizations, whose professional activities involve dealing with the sources of ionizing radiation, do not exceed maximum permissible levels. However, in certain cases, especially when perforing some non-standard technological operations, or in emergency situations, the risk of intake and accumulation of radionuclides present in the working environment by workers increases. Depending on the route of intake and the radionuclide physical and chemical properties, the methods for assessment of the expected internal effective dose are distinguished that are currently based on the workplace dosimetric monitoring data or individual dosimetric control data (by direct and indirect measuring methods) [3].

During their professional activities, the employees are exposed to the so-called low doses, characterized by the risk of the stochastic effect emergence [4, 5]. Therefore, it is also necessary to take into account the type of radiation (alpha, beta, gamma, etc.), incorporated radionuclide half-lives, duration of the radionuclide retention in the body, and radionuclide localization in organs and tissues to predict the internal exposure effects on the human body.

The study was aimed to perform the radiation hygiene assessment of working conditions and intake of radionuclides by the employees of the V.G. Khlopin Radium Institute working with open radioactive sources.

METHODS

The system of individual dosimetric monitoring for internal exposure of the research organization personnel working with open radioactive sources (RS) was the object of the study.

The study was carried out in 2019–2021 in the V.G. Khlopin Radium Institute.

The following parameters were measured in order to obtain the baseline data on the characteristics of harmful and/or dangerous work-related radiation factors the surveyed personnel was exposed to:

- gamma ambient dose equivalent rate (ADER);
- spectral composition of the gamma-emitting radionuclides at workplaces (spectral measurements);
 - removable radioactive working surface contamination;
 - alpha and beta particles flux density;
- volumetric activity of radionuclides in the air of working premises.

Gamma ADER was measured using the DKS-AT1123 dosimeter (Atomtex; Republic of Belarus) in accordance with the operating manual and Methodological Guidelines MU 2.6.5.008-2016 "Monitoring of Radiation Environment. General Requirements". To get a picture of the radiation field formation in the workplace, the measurements were performed in every point at the following height from the floor: 0.1 m, 1 m, 1.4 m and 1.7 m. The levels of alpha and beta working surface contamination were measured with the MKS-AT1117M dosimeter-radiometer (Atomtex; Republic of Belarus) in accordance with the operating manual. The expanded uncertainty (k = 2) of measurement was considered equal to the relative error of the measuring tool assuming a normal distribution of readings.

Removable radioactive surface contamination was measured by the indirect method involving taking smears with subsequent determination of their activity in accordance with the regulatory documents [6, 7], measurement uncertainty

was calculated in accordance with the measurement procedure [7].

To define volumetric activity of radionuclides, air samples were collected in the working premises using the PA-300M-1 aspirator (Ecotech-Ural; Russia) with the aerosol filters AFA RSP-20 (Electroforming Technologies; Russia) and iodine filter AFA SRF-20 (Electroforming Technologies; Russia) in accordance with the aspirator operating manual, with subsequent measurement using the MKGB-01 RADEK spectrometer-radiometer (STC RADEK; Russia). The expanded uncertainty of measurement was calculated taking into account the error of the spectrometer sensitivity coefficient (8%), error of the ambient air sample volume (10%), and procedural error resulting from incosistency of the load composition (5%) that accounted for 16%.

Spectrometric study of employees engaged in working with open RS (45 people, of them 34 were assessed in 2019 and 21 were assessed in 2020) and the control group employees (10 people) was performed by the direct measurement of radionuclides in the human body or organ with the SEG-10P-02 (STC RADEK; Russia) and SICH-100 (Scientific Research Institute of Marine and Industrial Medicine of FMBA; Russia) whole body spectrometers. Inclusion criteria: working with open RS during the last year, prior to assessment. Control group inclusion criteria: no exposure to open RS when working. Measurements were performed by registration of the photon radiation emitted by human body. Measurements with the SICH-100 spectrometer were performed in the "linear longitudinal scan" mesurement geometry, and SEG-10P-02 used the "lung" and "thyroid" measurement geometries. The content of the 90Sr radionuclide in the bone tissue was defined based on the bremsstrahlung radiation spectrum with the energy range of 50-150 keV registered by two scintillation detectors with the CsI(TI) single crystals sized $\emptyset 150 \times 3$ mm, installed in the SICH-100 unit. The measurement duration of 30 min was used in both spectrometers.

Calibration of the SICH-100 spectrometer was performed using the UP-02T unified solid phantom of the whole body and the ARDF-11-C anthropomorphic whole body phantom with ⁹⁰Sr radionuclide in the skeleton. The UP-02T phantom consists of a number of PET blocks with the set of rod sources assembled into models, the phantoms of human body with the specified radionuclide content, on site. The ARDF-11-C phantom is a prefabricated unit consisting of the anthropomorphic models of organs (thyroid gland, liver, lung, kidney, etc.), skeleton, and integument manufactured from materials that mimic biological tissues, equivalent to human tissues in terms of interaction with ionizing radiation.

Calibration of the SEG-10P-02 whole body spectrometer in the "lung" measurement geometry was performed by direct calibration with the use of the FLT-05 chest phantom in combination with the standard phantom of an adult man (body weight 70 kg, height 170 cm). There was a polyurethane lung simulator equipped with the large RSs containing ⁶⁰Co, ¹⁵²Eu, ²⁴¹Am radionuclides in the chest phantom. Calibration was performed serially for each of the listed above radionuclides.

To measure the activity of ¹²⁵I in the thyroid gland, thin NaI(TI) detection unit previously calibrated based on the ¹³³Ba characteristic radiation using the phantom of thyroid PT-04T was used. The phantom was a model of human neck with two ¹³³Ba radionuclide sources in its cavity positioned similar to the thyroid lobes. Since the detection area of the unit included not only the thyroid gland, but also blood vessels of the neck, the energy spectrum of background radiation was measured in the forearm in order to take into account the contribution of these blood vessels.

Table 1. Gamma ADER, alpha and beta particles flux density measured at the μ employees' workplaces

Tasks performed at the workplace	Range of measured ADER values, µSv/h	Maximim flux density, particles/(cm² × min)	
		alpha particles	beta particles
Analysis of highly radioactive solutions, X-ray absorptiometry, working with samples under the fume hood or in the box	4,0–53,0	30	1830
Technological operations with highly radioactive solutions, packing and distribution of the ⁹⁰ Sr preparations	5,0–122,0	No data available	1100
Opening ampoules, dissolution, calibration of solutions and producing the sources	3,4–56,0	4,2	4000
Preparation of the simulant solution containing ⁹⁰ Sr, ¹³⁷ Cs, ²⁴¹ Am, ¹⁵² Eu and other isotopes. Extraction of ⁹⁰ Sr, ¹³⁷ Cs from the simulant solution. Preparation of the ²³⁸ U and ²³⁹ Pu nitric acid solution. Preparation of the ²³⁸ U and ²³⁹ Pu oxide powders, sorption extraction of impurities, packing	0,5–4,3	0,8	15
Packing ¹²⁵ I	0,9–18,2	< 0,1	1680
Extraction and chromatographic separation of products from the spent nuclear fuel and irradiated ²²⁶ Ra processing	0,4–4,6	29	32
Acceptance, chopping up, storage and transfer of the spent nuclear fuel and highly radioactive materials, nuclear waste repacking and discharge	0,1–0,6	0,4	30
Dissolution, precipitation, evaporation, clarification of the spent nuclear fuel components	0,1-0,2	< 0,1	200
Extraction, evaporation, solidification of nuclear waste	0,1-0,3	< 0,1	20
Technological equipment decontamination and repair, nuclear waste sorting, packing and preparation for removal	11,0–54,5	78	1680

Note: the values were calculated taking into account the expanded uncertainty of measurement (k = 2).

The expanded uncertainty for the confidence level of P = 0.95 and coverage factor of k = 2 for the SEG-10P-02 and SICH-100 spectrometers was calculated taking into account the errors of the spectrometer sensitivity coefficients defined based on the phantoms (8%), procedural error resulting from uneven distribution of activity in the human body or organs (10%), and procedural error resulting from incomplete equivalence of the phantoms and human body (20%), which accounted for 24%.

Statistical data processing was performed by standard methods used for analysis of the radiation hygiene and biomedical data. The mean, median, standard error of the mean, and significance of the differences were calculated for all parameters with the significance level set at p=0.95. All the calculations were performed using the modern software products (Microsoft Excel, v. Professional Plus 2010; USA) designed for working with spreadsheets, as well as for data visualization and analysis.

The employees' working conditions were assessed in accordance with the regulations [8].

RESULTS

When performing the radiation hygiene assessment of the employees' workplaces in the research organization, a total of 17 production premises were assessed where the work with open RS was carried out. The results of the gamma ambient dose equivalent rate and the alpha and beta particles flux density measurement at the employees' workplaces are provided in Table 1.

The highest gamma ADER values were observed when working with highly radioactive solutions. The maximum values of the alpha and beta particles flux density were registered when opening ampoules, perfoming dissolution and calibration of solutions, producing the sources. Surface contamination by alpha particles in the amount not exceeding the reference

values in accordance with the requirements of RSS-99/2009 was also observed in the workplaces. In addition, removable surface activity and volumetric activity of radionuclides in the air of working areas were measured due to surface contamination of the employees' workplaces.

Based on the results of measuring the levels of the alpha and beta working surface contamination in the employees' workplaces, removable surface activity was detected not exceeding the permissible values defined by the RSS-99/2009 [9]. The highest levels of the beta removable surface contamination (90Sr) were registered when opening ampoules, perfoming dissolution and calibration of solutions, producing the sources .

Based on the results of measuring the volumetric activity of radionuclides in the air of working areas, the highest values were registered when working with the ¹²⁵I and ¹³⁷Cs radionuclides (volumetric activity of ¹²⁵I was 240 Bq/m³, and volumetric activity of ¹³⁷Cs was 315 Bq/m³).

Based on the radiation monitoring data, the potential upper effective dose limit for the pesonnel performing technological operations was calculated in accordance with legislative requirements [8]. It was found that the working conditions of personnel working with open RS were within the range from acceptable (class 2) to dangerous (class 4) based on the characteristics of exposure to harmful and/or dangerous work-related radiation factors.

According to the data obtained in 2016–2019, the average values of the employees' individual effective doses from external exposure were 3.4 mSv per year, which was well below the dose limit established for the group A personnel. The highest values were registered in the leading industial engineers, and the average values for the four-year period were 7.5 mSv.

When performing the spectrometric study in some employees working with open RS, incorporated radionuclides were found in their bodies (Table 2). The table also presents

ОРИГИНАЛЬНОЕ ИССЛЕДОВАНИЕ І ЗДРАВООХРАНЕНИЕ

Table 2. Characteristics of the surveyed employees' internal exposure

Radionuclide	Radionuclide activity, Bq	Expected equivalent dose from internal exposure, mSv per year
¹³⁷ Cs	120–1100	0.001–0.004 (whole body)
⁹⁰ Sr	5200–16500	0.3; 0.9 (bone tissue)
²²⁶ Ra	360–770	0.47–1.00 (bone tissue)
²²³ Ra	550–2800	0.9; 4.8 (bone tissue)
125	2930–9850	0.18–2.1 (thyroid gland)
⁶⁰ Co	840	0.03 (lungs)

the values of the expected equivalent dose from internal exposure of the employees resulting from the radionuclide intake. Calculations were performed taking into account the spectrometry data of reference dose coefficients [10, 11].

In addition to radionuclides presented in Table 2, incorporated ²⁴¹Am was found in three employees. Due to the lack of calibration of the used measuring equipment, it was impossible to accurately measure activity of this radionuclide in the skeleton.

DISCUSSION

According to the results of the radiometric survey, radiological situation in the workplaces of the research organization employees working with open RS could be characterized as follows:

- broad range of the gamma ADER levels (0.10-122 µSv/h);
- alpha and beta working surface contamination (the following radionuclides were detected in smears from working surfaces: ¹³⁷Cs, ¹²⁵I, ²⁴¹Am, ¹⁵⁴Eu, ²⁴³Am with the ²³⁹Np decay product, and ²²³Ra with the ²¹¹Bi decay product);
- radioactive pollution of air in the working areas (including sich radionuclides as 125 I and 137 Cs).

In some individuals the following were found: ¹²⁵I in the thyroid gland (up to 9,850 Bq), ⁹⁰Sr in the skeleton (up to 16,500 Bq), ¹³⁷Cs in the whole body (up to 1,100 Bq). The finding of the ²²³Ra intake in employees stands out, since the ²²³Ra half life period is 11 days, and the ²²³Ra contribution to the internal exposure dose is much higher compared to other radionuclides.

The fact of radionuclide incorporation was confirmed by periodic spectrometry-based assessment of the same employees performed in 2019 and 2020. These employees should be considered a group at high risk of occupational, work-related and chronic diseases.

Currently, in the research organization under consideration, the expected effective doses from internal exposure in employees are monitored by computation (based on the dosimetry in worklaces), which is not fully effective when working with the large number of radionuclides, including the short-lived ones. That is why developing the programme of screening with the use of portable spectrometers is one of the ways to improve the efficiency of the internal dosimetry monitoring system in employees working with open RS. The studies are aimed to confirm the fact of the radionuclide intake and determine the radionuclide composition. In case

of radionuclide intake detection in the employee, assessment should be performed in the specialized institution using the high precision whole body radiation spectrometer. Such approach would provide the radiation safety service employees with the main instruments for effective implementation of the internal exposure monitoring programme [12–14].

The database was developed and registered in order to perform accumulation and processing of data on the levels of the employees' external and internal exposure in the planned exposure situations together with the results of the periodic medical examinations [15]. Introduction of the database in practice would make it possible to assess the general trends in occupational exposure of the staff and analyze the research results in order to reveal the correlation between the incidence of occupational, work-related, and chronic somatic disorders, as well as the emergence of medical contraindications for work, and the levels and duration of exposure to the harmful and/or dangerous work-related radiation factors and other factors (length of employment, age, gender, etc.).

CONCLUSIONS

(1) Working conditions of the research organization personnel working with open RS are within the range from acceptable (class 2) to dangerous (class 4) based on the characteristics of exposure to harmful and/or dangerous work-related radiation factors. Compliance with the established norms and radiaion safety regulations does not exclude radionuclide intake and incorporation in employees. (2) Regular spectrometry-based assessment of the V. G. Khlopin Radium Institute employees and the development of screening programmes (with the use of portable spectrometers), aimed at assessing radinuclide intake and, in case of confirming incorporation, performing indepth spectrometry-based assessment with the high precision whole body radiation spectrometer, are required for analysis of the dynamic changes in the individual effective doses from internal exposure. This will make it possible to improve the quality of the individual internal dose control and the efficiency of the radiation protection of workers. (3) Further studies are necessary aimed at assessing the trends and defining the correlation between the incidence of occupational and chronic somatic disorders, as well as the emergence of medical contraindications for work in the research organization employees working with open RS, and the levels and duration of exposure to harmful and/or dangerous work-related radiation factors and other factors.

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ОРИГИНАЛЬНОЕ ИССЛЕДОВАНИЕ І ЗДРАВООХРАНЕНИЕ

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Федеральное государственное унитарное предприятие научно-исследовательский институт промышленной и морской медицины Федерального медико-биологического агентства, Медико-дозиметрический регистр персонала группы А Акционерного общества «Радиевый институт им. В. Г. Хлопина», работающего с открытыми источниками ионизирующих излучений: Свидетельство о государственной регистрации базы данных № 2021620928 Российская Федерация, 05.05.2021.