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Improvement of the quality of effective dose estimation by interlaboratory comparisons

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Radiation Protection Measurements Laboratory (RPLM) of the Institute of Atomic Energy POLATOM determines radionuclides in human urine to estimate the effective dose. Being an accredited laboratory, RPLM participated in interlaboratory comparisons in order to assure the quality of services concerning monitoring of internal contamination. The purpose of the study was to examine the effect of interlaboratory comparisons on the accuracy of the provided measurements. The results regarding tritium (^{3}H) and strontium (^{90}Sr) determination, obtained within the radiotoxicological intercomparison exercises, organized by PROCORAD, in 2005-2010, were analyzed and the methods used by the laboratory were verified and improved.

Key words: tritium, strontium, urine, intercomparisons, effective dose.

Introduction

Radiation Protection Measurements Laboratory (RPLM) of the Institute of Atomic Energy POLATOM performs regular monitoring of internal exposure to radiation for employees of the Institute and other occupationally exposed workers. RPLM is the only laboratory in Poland that provides such services, accredited by Polish Centre for Accreditation. Detection and determination of the radionuclide activities makes it possible to detect the internal contamination and to assess the effective dose.

The procedures used in the laboratory include *in vivo* and *in vitro* measurements. *In vivo* methods of radionuclide detection are provided by the use of a whole body counter and thyroid counter. *In vitro* measurements are carried out by determination of beta emitters (^{3}H, ^{90}Sr, ^{35}S, ^{32}P) and gamma emitters. In this study the results obtained in interlaboratory comparisons regarding radiochemical determination of beta emitters, namely tritium (^{3}H) and strontium (^{90}Sr), in urine are discussed [1].

Although tritium emits only low energy beta radiation and it is not dangerous externally, it is important to monitor the level of tritium exposure, because it becomes hazardous when ingested via water, food, inhaled or absorbed through the skin. Tritium combines with oxygen to form tritiated water (HTO). With an assumption that there is an equilibrium between tritiated water in urine and in the body water, it is possible to determine the activity concentration in body water by measuring the amount of tritiated water in a sample of urine collected in specific time [2].

Strontium ^{90}Sr is considered as highly radiotoxic nuclide due to its long physical half-life (28.6 years), a long biological half-life (49.3 years) and its chemical similarity to calcium. It enters the organism by ingestion with contaminated food (milk is one of the main ^{90}Sr sources). Strontium remaining in the organism is incorporated into bone tissue and bone marrow. Maximum permissible ingestion levels for strontium are very low, because the radioisotope may negatively influence health. Exposure to ^{90}Sr can be examined by urine analysis with the use of sensitive analytical methods [3, 6].

In order to assure the quality of services concerning the monitoring of internal contamination, RPLM takes part in interlaboratory comparisons. Yearly the laboratory participates in the radiotoxicological intercomparison exercises organized by PROCORAD (Association for the Promotion of Quality Control in Radiotoxicological Analysis). The use of real biological samples, which contain the radionuclides often encountered in occupational exposure, is the feature that distinguishes these intercomparisons. Participation in the exercise gives the opportunity to evaluate the quality of provided medical analyses and to validate the used methods.

Materials and methods

RPLM takes part in interlaboratory comparisons Procorad since 2005. Tritium (^{3}H) and strontium (^{90}Sr) are determined in urine samples.

Urine samples are provided by the organizer of the exercise. Mixed urine collected from various persons constitutes samples, which are delivered in polyethylene bottles to each laboratory. Each of the participants of the tritium exercise obtains five samples of natural human urine. Sample A is a blank sample, sample B and C are spiked with tritiated water, sample D is tritiated urine resulting from natural metabolism and sample F is urine spiked with tritiated thymidine. For ^{90}Sr determination there are three samples provided of approximately 500 ml urine brought at pH < 2 by addition of concentrated nitric acid (A – blank; B, C – spiked with radioactive strontium). For the preparation of urine samples stable chemical compounds corresponding to each radionuclide are used [5].

In order to determine the tritium content, the sample must be discoloured by active carbon. Next, sodium carbonate and sodium thiosulfate are added and then the distillation is carried out. The distillate is mixed with liquid scintillation cocktail, the beta activity of sample is measured with liquid scintillation counter (LSC) Tri- Carb 2900-TR. The measurement is carried out in energy range 0-18.6 keV [4].

As a part of interlaboratory exercise also tritiated thymidine in urine is determined (sample F). The activity of thymidine ($F_{Thymidine}$) is calculated as a difference between total activity (F_{Total}) and activity of tritium ($F_{Tritium}$). Total activity is measured directly, urine sample is mixed with liquid scintillation cocktail. In order to calculate the total activity of the sample, the efficiency of the measurement must be known. The quench curve, which assign dependence between the efficiency of measurement and tSIE (Transformed Spectral Index of External Standard), must be performed. Spiked urine, with defined activity, is mixed with scintillation cocktail in various proportion. Beta activity of the sample is measured in LSC, tSIE is defined during LSC measurement and then the efficiency of measurement is calculated.

The method of strontium (^{90}Sr) determination in urine consists in the measurement of activity of ^{90}Y which is in equilibrium with ^{90}Sr. Radioactive strontium and yttrium are coprecipiated with oxalates. Acid environment, strontium and calcium carrier support this process. Formed deposit is submitted to mineralization to eliminate organic residues. Obtained ash is dissolved in hydrochloric acid. The solution is submitted to extraction with bis(2-ethylhexyl)-orthophosphoric acid (HDEHP) in toluene. The complete extraction procedure is shown in Figure 1. The extract, which is obtained in the process, is evaporated on the counting disc. The activity of sample is measured in gas proportional counter Berthold LB510.

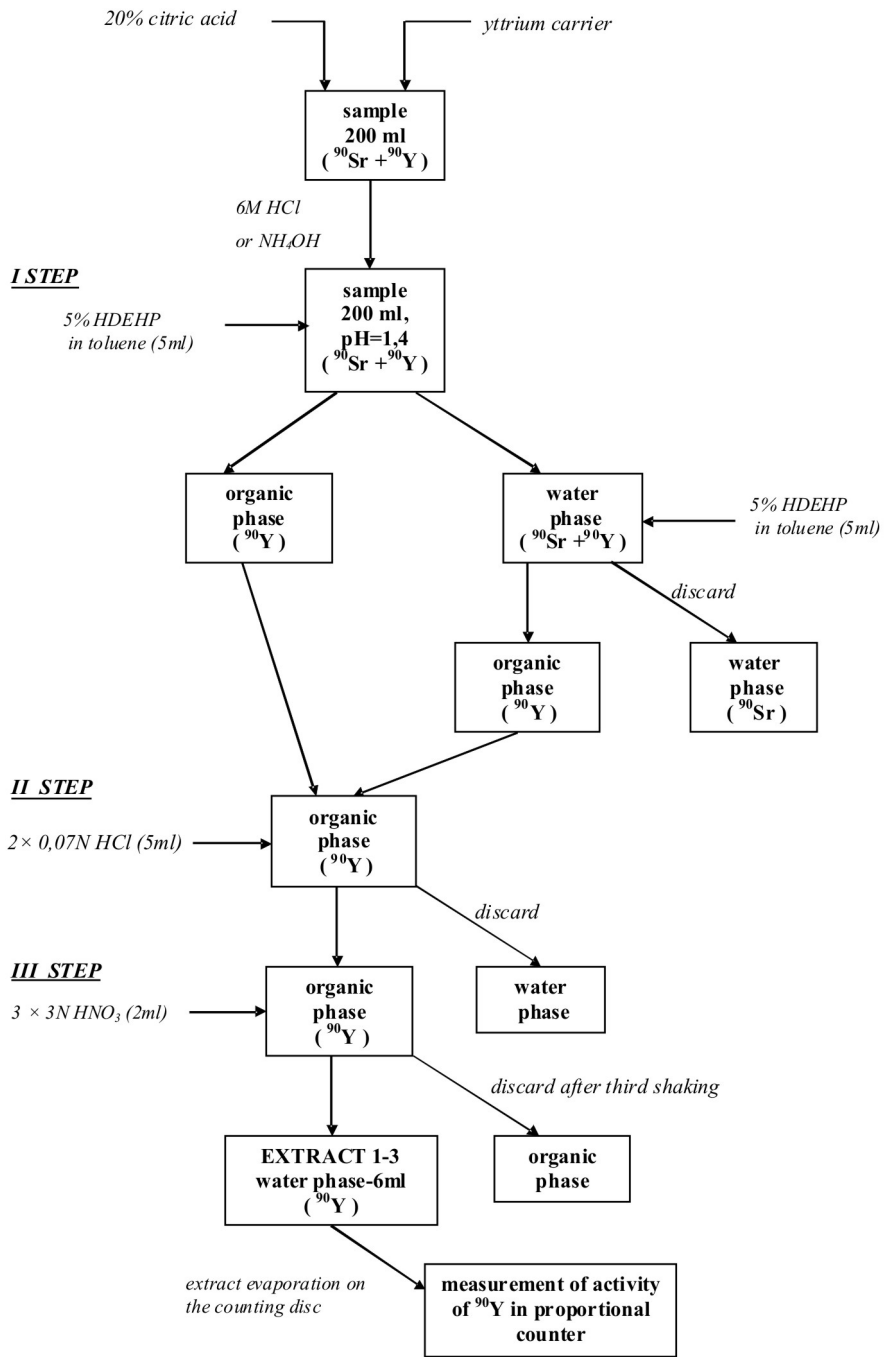

Figure 1. Extraction of ^{90}Y from water sample

Results and discussion

Results obtained during last six years of Procorad's intercomparisons regarding tritium determination are presented on Figure 2, with the relative bias on the axis of ordinates. The relative bias is calculated using the following equation [5]:

$$B = \frac{A_i - A_{ref}}{A_{ref}} \cdot 100\%$$

where:
A_i – measured activity of sample i [Bq · dm^{-3}],
A_{ref} – reference activity of sample i [Bq · dm^{-3}].

In the graph, the relative biases are presented for the results of the tritium exercises, achieved for each sample (A, B, C, D, F) in the years 2005-2010.

Conclusions drawn from the comparisons can have a significant effect on the maintenance of good laboratory practice. The results obtained in 2006 and 2007 showed

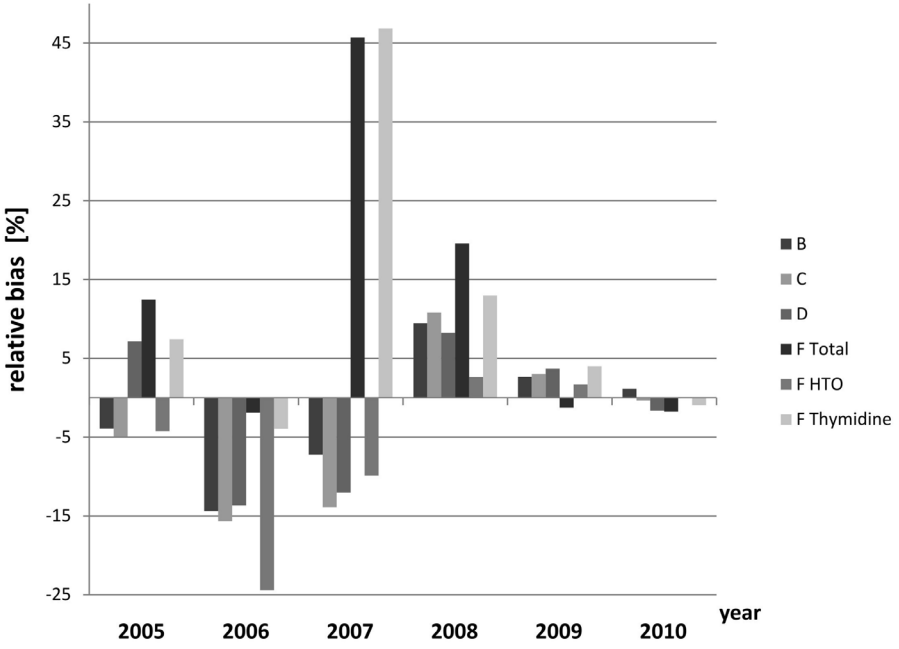

Figure 2. Relative biases for the results obtained by RPLM in interlaboratory comparison – tritium (HTO) in urine

that the efficiency of measurement applied to the calculations was too high (resulting in a negative bias). It was gathered that tritium standard solution became more concentrated as the result of long-lasting use. The value of efficiency influenced calculated results, in these case underrated the tritium activity in sample. Consequently, the effective doses, which could have been evaluated for contaminated persons during this period, were too low. Furthermore, the huge relative biases (~ 45%) for tritium determination in sample F, containing tritiated thymidine (F_{Total}, $F_{Thymidine}$), in 2007 caused that the sample preparation methods had to be reconsidered.

In 2008, a new standard solution has been introduced and also proportion, in which the distillate with liquid scintillation cocktail was mixed, was changed. The results weren't as good as it had been expected, therefore in the next years laboratory returned to the previous proportion.

Intercomparisons in 2009 and 2010 showed that laboratory performed determination of tritium activity in urine with better precision. The obtained results were very close to the reference activities.

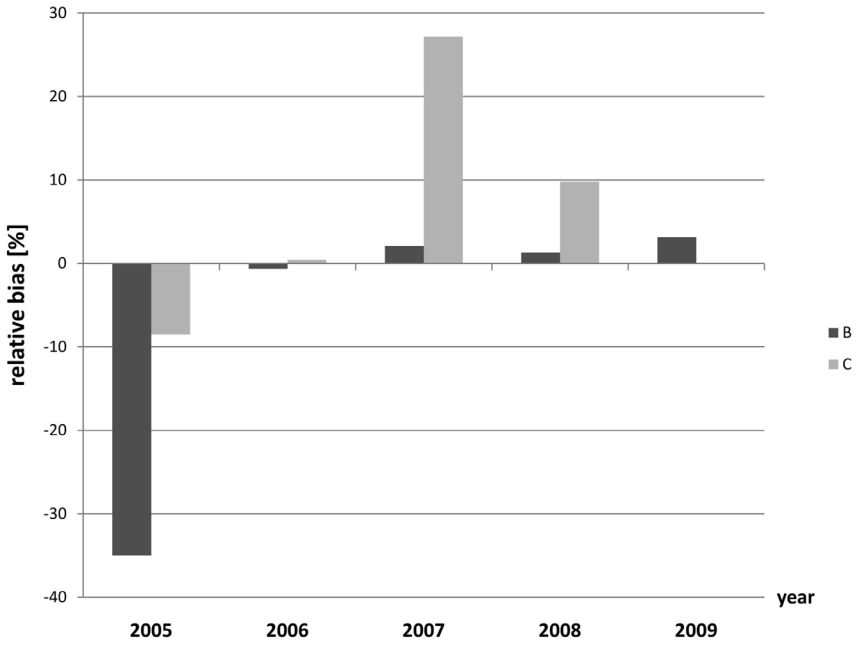

Figure 3. Relative biases for the results obtained by RPLM in interlaboratory comparison – strontium (^{90}Sr) in urine

Figure 3 shows the relative biases for results achieved in Procorad's intercomparisons regarding strontium (^{90}Sr) determination in urine samples.

Figure 3 shows that there are significant discrepancies between the relative biases for results gained in the years 2005-2009. In 2006, the activity of ^{90}Sr in urine was determined with high precision (B ~ 0.5%), while the value of relative bias characterizing some of the results obtained in 2005 and 2007 is 30%. The conclusion was drawn that the evaporation of the sample on the counting disc and the application of gas proportional counter to activity measurement could have been the source of uncertainty. For that reason, the decision regarding changes in the method of determination strontium activity in urine was made. A new method of strontium activity measurement in urine was introduced in 2010. The measurements are carried out in LSC, the extract obtained in process is mixed with scintillation cocktail. The next interlaboratory comparison will verify whether the new approach provides better accuracy.

Conclusions

The methods of tritium and strontium determination in urine were verified by participation in interlaboratory comparison. The exercises confirmed the reliability of the procedures used by RPLM laboratory. All of the results proved acceptable. Participation in the radiotoxicological intercomparison exercises organized by PROCORAD gave the possibility to improve the manner of determination of the amount of selected radionuclides. Analysis of tritium detection comparison lead to confirmation of laboratory precision. As a result of participation in strontium exercise the method of the measurement has been changed due to unsatisfactory repeatability of the results.

The exercises were valuable for the laboratory because of gaining experience and knowledge considering performed analyses. This improves the accuracy in determination of the real effective doses for employees and helps to protect their health. Furthermore, the described methods make it possible to estimate the effective doses for patients undergoing therapy using radioactive isotopes and for persons, who work with unsealed sources.

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