Applied Radiation and Isotopes 137 (2018) 18–22

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Tritium internal dose estimation from measurements with liquid scintillators

A. Pántya[a,*], Á. Dálnoki[a], A.R. Imre[a,b], P. Zagyvai[a], T. Pázmándi[a]

[a] MTA Centre for Energy Research, P.O. Box 49, Budapest 1525, Hungary
[b] Budapest University of Technology and Economics, Department of Energy Engineering, Muegyetem rkp. 3, H-1111 Budapest, Hungary

HIGHLIGHTS

- Tritium activity concentration in PROCORAD urine samples have been determined.
- Sources of uncertainties of dose assessment have been analyzed.
- For final accuracy the assumptions used in the dose assessment can be more influential than the errors of the measured data.

ARTICLE INFO

Keywords:
Tritiated water
Activity concentration
MONDAL-3
PROCORAD
Urine

ABSTRACT

Tritium may exist in several chemical and physical forms in workplaces, common occurrences are in vapor or liquid form (as tritiated water) and in organic form (e.g. thymidine) which can get into the body by inhalation or by ingestion. For internal dose assessment it is usually assumed that urine samples for tritium analysis are obtained after the tritium concentration inside the body has reached equilibrium following intake. Comparison was carried out for two types of vials, two efficiency calculation methods and two available liquid scintillation devices to highlight the errors of the measurements. The results were used for dose estimation with MONDAL-3 software. It has been shown that concerning the accuracy of the final internal dose assessment, the uncertainties of the assumptions used in the dose assessment (for example the date and route of intake, the physical and chemical form) can be more influential than the errors of the measured data. Therefore, the improvement of the experimental accuracy alone is not the proper way to improve the accuracy of the internal dose estimation.

1. Introduction

Ionizing radiations may cause external or internal exposure to the human body. Radioactive material can get into the body by ingestion, inhalation and absorption through the skin or a wound. One of the important nuclides for internal contamination is tritium. Tritium is a pure β-emitter with a maximum energy of 18.6 keV and a mean energy of 5.7 keV; its physical half-life is 12.3 y (Chu et al., 1999).

For the great majority of radionuclides, the internal dose is estimated in two steps. First, the activity in the body is determined with direct or indirect monitoring methods. By direct measurements the radioactivity in the whole or part of the human body can be determined (in-vivo), while by the indirect method the radioactivity of physical and biological (in-vitro) samples representing the intake is assessed (for example urine, see Phukainen and Heikkinen, 2008; Kim et al., 2011; Kim and Kong, 2012, Dizman et al., 2015; Shen et al., 2015). In the second step, from measured data, considering some further assumptions on exposure conditions (time and route of intake, chemical form etc.), the intake value and from that the radiation dose can be estimated. In case of internal dose assessment of tritium, it is usually assumed that urine samples are obtained after the tritium concentration inside the body has reached equilibrium (see for example Kim et al., 2011; Kim and Kong, 2012). Incorporated tritiated water becomes uniformly distributed rapidly in body fluid, thus the tritium content of water present in different organs will be practically the same. A fraction of the tritium in tritiated water then becomes organically bound and its retention depends on the metabolic activity of various tissues in 2–4 h following the intake. After this transition period a urine sample is considered representative if it is collected for 24 h after the initial intake. For tritiated water (HTO) in the recommendations of the International Commission on Radiological Protection (ICRP, 1997) it is stated that 97% of the tritiated water activity is in equilibrium with body fluids and is retained with a biological half-life of 10 days, and the remaining 3% is assumed to be incorporated into organic molecules and retained with a half-life of 40 days. For organically bound tritium (OBT) e.g. tritiated thymidine, (Taylor, 2008) 50% of the total tritium was assumed to enter the total body water pool and the remainder was assumed to be incorporated into tissues by hydrogen–carbon bonding and be

* Corresponding author.
E-mail addresses: anna.pantya@energia.mta.hu, pantya.anna@energia.mta.hu (A. Pántya).

https://doi.org/10.1016/j.apradiso.2018.02.031
Received 7 September 2017; Received in revised form 18 February 2018; Accepted 26 February 2018
Available online 06 March 2018
0969-8043/

eliminated with a biological half-life of 40 d. The activity concentration of tritiated water in urine is the equilibrium value reached by the long-term (more than 4 h) equilibration of body fluids and soft tissue. Thus, analysis of tritiated water in a single sample of urine represents the activity concentration in body water at the time the sample was collected. Tritium measurement is carried out using liquid scintillation in most cases (ICRP, 1997; Phukainen and Heikkinen, 2008; Kim et al., 2011; Caridi et al., 2016).

It is commonly known that in internal dosimetry the uncertainty of the measurement itself is usually significantly lower that of the uncertainty of the model used for the dose estimation. Therefore, in the assessment of internal dose, the influence of the uncertainties due to these assumptions is predominant in comparison with the uncertainty of measured data. In this short paper we are not only going to demonstrate that improving the accuracy of the measurements is ineffective without improving the reliability of the assumptions used for internal dose calculations but also quantify and compare the effects of these two types of uncertainties.

2. Measurement methods and instruments

In various nuclear installations (including the MTA Centre for Energy Research) the routine monitoring of internal exposure of workers includes monitoring of tritium nuclide in urine. In our routine method for analyzing HTO in the urine, 10 ml Ultima Gold LLT scintillation cocktail and 1 ml volume of daily urine samples are used (Verrezen et al., 2008). The measuring time is 60 min, while the minimum detectable activity (MDA) for routine measurement is about 100 BqL^{-1}. For the OBT determination the samples are distilled before the measurement and original samples are also measured. Distillation process removes the color of the sample but not all the chemical impurities. Since distillation has been done under atmospheric pressure because of technical difficulties (instead of applying reduced pressure) one has to consider the high probability of the decomposition of some OBT material; this can lead to the overestimation of aqueous tritium and the underestimation of OBT, and thus a certain underestimation of the dose consequence as well. In case of high impurity concentrations, further cleaning is necessary because chemical impurities might have significant effect on the counting efficiency (Bergeron et al., 2012).

During our experiment, two types of vials were used: borosilicate standard 20 ml glass vials with low potassium content and standard 20 ml polyethylene vials. The samples were stored in scintillation vials for 2 h before the measurement. Background samples were made from distilled groundwater (taken from a deep driven well in Boconád, Hungary) with particularly low tritium concentration; these samples were used to check the experimental setup by determining its limit of detection for the actual circumstances. Background and quality controlled samples with known activity were also analyzed along with the urine samples. In order to prevent contamination we cleaned the measuring vials externally with acetone before placing them into the counter. Two types of liquid scintillation counter (Tri-Carb 2200 CA by Packard and 4810TR by Perkin-Elmer) were used for measurement of the samples with different energy window settings. The sensitivity of the two devices is different, thus the optimal measurement settings vary. The counting efficiency for the measurement was determined with two methods; standard addition (M1) and self-made color quench curve (M2). Color quench is an attenuation of the photons (light) emitted by the primary or secondary scintillator of the LSC cocktail. The counting efficiency can be determined using the relation between the quench indicating parameter tSIE and the counting efficiency, based on the quench correction (or quench calibration) curve. In the tSIE range relevant for us (250 − 500), quadratic fit proved to be sufficient enough ($R^2 = 0.9655$), although for the extension to higher values, higher order polynomials (at least 4th order ones) should be used. Both calculation methods require samples with known activity for comparison. The used parameters are summarized in Table 1.

Table 1
Parameters of the measurements.

Measurement geometry
10 ml Ultima Gold LLT scintillation cocktail and 1 ml urine sample
Types of vials
standard 20 ml polyethylene OR standard 20 ml low K content borosilicate glass
Types of equipment
TRICARB 2200 CA by Packard OR TRICARB 4810 TR by Perkin-Elmer
Window settings
I. cpmA: 1.5–5.5 keV OR II. cpmA: 0–18.6 keV
cpmB: 5.5–18.6 keV cpmB: 2–18.6 keV
Types of efficiency calculations
standard addition OR color quench curve fitting (separate ones for each window setting)
Measurement time
60 min

3. Results and discussion

As an example, the samples of PROCORAD (www.procorad.org) intercomparison were analyzed and shown here. PROCORAD organizes radioanalytical intercomparisons every year in order to evaluate the quality of medical analysis results and to promote a good laboratory practice (PROCORAD, 2017). These intercomparisons use urine samples containing radionuclides most often encountered in the course of controlling occupational exposure. Six samples were prepared for analyzing tritium in urine; as described in Table 2. Two of them (D and F) are selected for demonstrating our initial statement.

The reason behind these choices was that Sample D was a urine sample originating from a tritium contaminated man and Sample F contained tritium and tritiated thymidine (to represent organically bound tritium, see Taylor, 2008). Thus, separation of the two components in case of Sample F was necessary, and this sample had to be distilled as well.

Fig. 1 shows that the choice of equipment, type of the vial and the counting efficiency-determination method has different effect on the finally obtained efficiency values. Some of these effects are trivial, for example the effect of the type of vials (plastic or glass) or the type of the instrument (2200 or 4810), although the extent of these window effects are interesting (10% relative difference using different vials, 25% relative difference using different equipment). It is also interesting and less trivial that the effect of the counting efficiency determination method reached approx. 25% relative difference for sample E, while this difference was below 10% for the other samples. Since sample E is a water sample (with relatively low quenching), one might conclude that the anomalous (25%) difference is caused by the fact that this sample is outside of our quench correction curve or by the high uncertainty due to the fitting. But the quench curve is still valid in this range and the uncertainties are not higher than those for other samples; therefore this difference is rather a physical effect, than a numerical artifact. A common curve was fitted for the whole tSIE range (250 − 500) including low values for urine and higher values for pure water. This method gave low uncertainties for most of the samples, so this cannot explain the significant difference between results of (M1) and (M2) methods for sample E.

The activity concentration of sample D (contaminated urine) can be

Table 2
Sample for ^3H measurements used for the PROCORAD intercomparison in 2017.

Sample A	blank urine
Sample B	urine spiked with tritiated water, activity concentration < 10 $kBq\,L^{-1}$
Sample C	urine spiked with tritiated water, activity concentration < 10 $kBq\,L^{-1}$
Sample D	contaminated urine
Sample E	solution of tritiated water
Sample F	urine spiked with tritiated water and thymidine