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Evaluation of in vivo and in vitro dose detection limits for different radionuclides and measurement techniques

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HIGHLIGHTS

- Personal monitoring results depend on the accuracy of measurement instruments.
- Twelve radioisotopes were studied using gamma and alpha spectrometry.
- Detection limits were presented for direct and indirect monitoring methods.
- The calculated committed effective dose proved the adequacy of used methods.

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ABSTRACT

Personal monitoring programs for workers handling radioactive materials are influenced by numerous factors as the measurements of radioactivity in tissues or/and in excreta can be carried out using different techniques. This paper summaries the basic procedures needed for accurate and fast measurement of different radionuclides like ^{235}U, ^{234}U, ^{238}U, ^{226}Ra, ^{210}Po, ^{131}I, ^{99m}Tc, ^{134}Cs, ^{137}Cs, ^{57}Co, ^{58}Co, and ^{60}Co. Overviews of in vitro and in vivo monitoring methods are provided as well as methods used to calculate detection limits and internal radiation dose. For the radionuclides of interest, in vivo and in vitro detection limits were converted into committed effective doses to evaluate the applicability and limitations of the systems used at the laboratory. The results proved that the systems' sensitivity is suitable for use in routine monitoring of workers subject to risk of internal exposure from such radionuclides. Consequently, monitoring programs suggested by the Syrian internal dosimetry laboratory are suitable to detect committed effective doses even below 1 mSv in most cases

1. Introduction

Monitoring of workers occupationally exposed to the risk of internal contamination is mandatory to protect them against risks of incorporated radionuclides. There are two methods of quantifying how much radioactive material is in the human body. The first method is indirect measurement which involves the determination of radionuclide concentrations in materials separated from the body, like urine, for the purpose of assessing intakes and internal doses. This is referred to as in-vitro bioassay (IAEA, 2000). The second method is direct measurement of gamma or X-ray photons (including bremsstrahlung) emitted from internally deposited radionuclides. These methods are frequently named as organ activity measurements or whole body counting. This is referred to as in-vivo bioassay (IAEA, 1996). Actually, the selection of measures and programs requires decisions concerning methods, techniques, frequencies etc. and the performance criteria required for such measurements usually depend upon the purpose for the radiobioassay measurement (ISO, 2006, 2010b). However, in order to achieve consistency and reliability in the assessment of doses, the International Standard Organisation presented procedures and assumptions for the standardized interpretation of monitoring data (ISO, 2011).

Currently available internal dosimetry service in Syria is located at the Syrian Atomic Energy Commission (SAEC) and it consists, at present, of two laboratories: internal dosimetry laboratory (IDL) for bioassay analysis, and thyroid counter laboratory (TCL).

The IDL is monitoring radiation workers for possible internal deposits of different radionuclides from various activities. This paper investigates the detection limits of routine monitoring programs for the following radionuclides: ^{235}U ^{234}U, ^{238}U, ^{226}Ra, ^{210}Po, ^{131}I, ^{99m}Tc, ^{134}Cs, ^{137}Cs, ^{57}Co, ^{58}Co, and ^{60}Co.

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2. Materials and methods

2.1. In vitro monitoring

The main task of IDL is carrying out radiochemistry analysis of urine samples for the indirect monitoring of, in most cases, alpha emitter radionuclides. However, the analysis of alpha emitter radionuclides, such as ^{235}U ^{234}U, ^{238}U, ^{226}Ra, and ^{210}Po, requires chemical separation of radionuclide, sample-specific tracers to determine the chemical recovery, and long counting time on low-background alpha spectrometry system. Practically, alpha counting of ^{210}Po [5304.4 keV (100%)], ^{226}Ra [4784.4 keV (94.45%)], ^{234}U [4774.9 keV (72.5%), 4722.6 keV (27.5%)], ^{235}U [4395.2 keV (55%), 4364.1 keV (11%)], and ^{238}U [4197 keV (79%), 4147 keV (21%)] (OASIS, 1995) was carried out using an alpha spectrometer (Oasis, Oxford) with a passive ion-implanted silicon detector (active area 450 mm^2, detector resolution 17 keV at 5486 keV, 3.6 background counts per day and the minimum depletion thickness of 10^{-4} m).

On the other side, radionuclides that emit gamma ray, such as ^{131}I, ^{123}I, ^{99m}Tc, ^{137}Cs, ^{134}Cs, ^{57}Co, ^{58}Co, and ^{60}Co, were determined in urine samples by direct measurement using HPGe gamma spectrometry. Concentration of these radionuclides in urine samples were measured with a stationary unit (Fig. 1(a)) which consists of HPGe detector (EURISYS MESURES, France) 80% relative efficiency with high resolution (1.85 keV at 1.33 MeV) and low background lead shield, portable multichannel analyzer (DigiSpectrum, BRUKER Co.), and InterWinner-07 PC spectrometric software (EURISYS MESURES).

Efficiency (ε) calibration curve was established for the same geometry used for samples (Marinelli-type container of 1000 cm^3 capacity) using spiked water sample with standard mixed gamma Certified Reference Material QCY-48 (Amersham, UK) which is traceable to NIST, conforming with ANSI N42.22-1995. The prepared standard contains gamma emitters with energies covering the desired energy range, namely, ^{57}Co (122.1 keV, 136.47 keV), ^{60}Co (1173.24 keV, 1332.5 keV), ^{85}Sr (514.0 keV), ^{88}Y (898.05 keV, 1836.1 keV), ^{109}Cd (88.03 keV), ^{113}Sn (391.7 keV), ^{137}Cs (661.66 keV), ^{139}Ce (165.9 keV), ^{203}Hg (279.2 keV), and ^{241}Am (59.54 keV) (LNHB, 2015).

2.2. In vivo monitoring

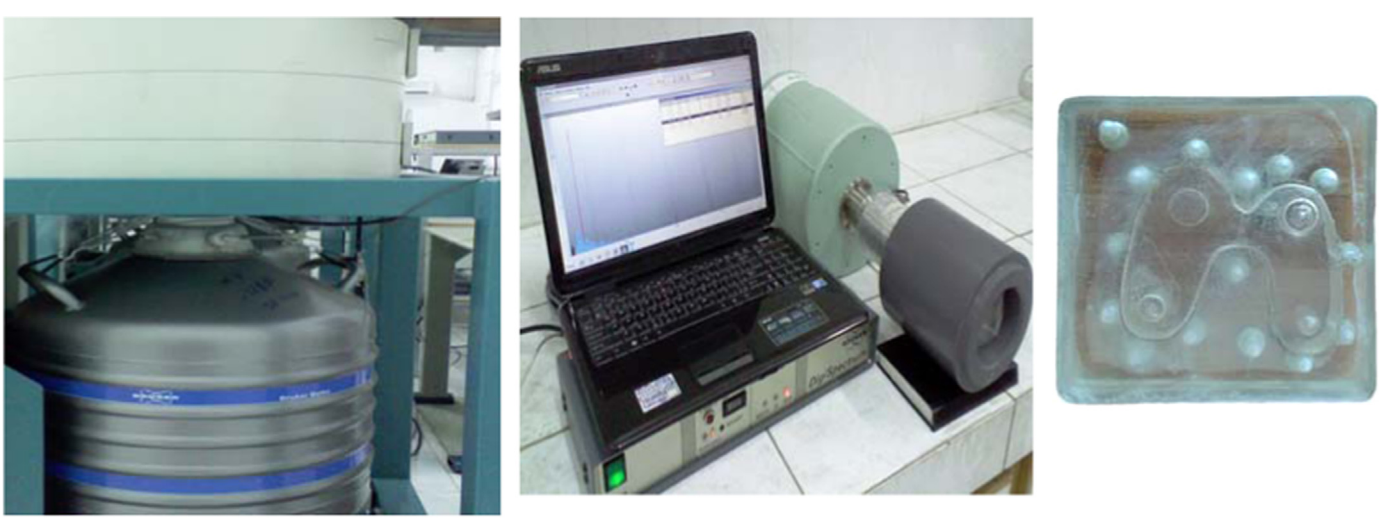
In vivo monitoring is feasible only for those radionuclides emitting penetrating radiation, usually gamma, that can escape from the body. The type of monitoring technique available at our laboratory is "thyroid counting". The measurements of iodine in thyroid were performed with a portable unit (Fig. 1(b)) which consists of HPGe detector (Eurysis Systems, France) with high resolution (1.85 keV at 1.33 MeV) and relative efficiency of 35%, portable multichannel analyzer (DigiSpectrum, BRUKER Co.) paired with a notebook computer, basic spectroscopy software (SpectraLineGP, LSRM Ltd.), and home-made thyroid phantom (Fig. 1(c)) for calibration. The phantom was made of Plexiglas with the dimensions of 10 cm × 10 cm × 3 cm and containing a thyroid-like shape whose the volume is 46.5 mL.

Table 1
Methods and intervals adopted for routine monitoring according to ICRP 78 and ISO 20,553.

Radionuclide	Absorption type	Monitoring interval (days)	Monitoring method
Uranium hexafluoride	F	90	Indirect (urine)
Uranium peroxide, uranium nitrate, Ammonium diuranate	F	30	
Uranium tetrafluoride, uranium trioxide	M	90	
Uranium octoxide, uranium dioxide	S	90	
^{226}Ra	M	180	
^{210}Po	F	90	
^{210}Po	M	90	
^{131}I	F	15	Indirect (urine) and direct (thyroid)
^{134}Cs	F	180	Indirect (urine)
^{137}Cs	F	180	
^{57}Co	M	90	
^{57}Co	S	30	
^{58}Co	M	90	
^{58}Co	S	90	
^{60}Co	M	90	
^{60}Co	S	90	

2.3. Individual monitoring program

The frequency of measurements, for the monitored workers, differs according to the task performed and to the operation place. Table 1 summarizes the detailed information for each concerned



(a) (b) (c)

Fig. 1. a) Stationary unit gamma spectrometry; b) Mobile detection unit for I-131 measurements; c) Thyroid phantom.