



Assessment of tritium exposure through urine samples analysis from selected Indonesian radiation workers

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Abstract

In this study tritium exposure was assessed through urine samples analysis by the determination of tritium concentration and effective dose of selected 25 laboratory radiation workers in non-nuclear power plant (non-NPP) facility and 15 people of general public in Jakarta. The samples were mixed with activated charcoal, filtered, and distilled before analysis using liquid scintillation counter. It is revealed that the concentration of tritium levels in urine varied substantially depending not only on the job category but also their water intake, metabolism, and environment. These findings are important as reference data for evaluating the radiological impact on the population from future tritium contamination in Indonesia.

Keywords Tritium in urine · Internal dosimetry · Liquid scintillation counter · Urine bioassay · Radiation workers

Introduction

Tritium, identified by the symbol ^3H , is categorized as a radionuclide that originates from both natural sources and human activities. Tritium occurs naturally in small amounts in the earth's atmosphere through interaction between cosmic rays and atmospheric gases, nitrogen (^{14}N) and oxygen (^{16}O) [1, 2]. However, the anthropogenic tritium is mainly

sourced from the nuclear industry [3–5]. Compared to other radionuclides, nuclear industries release enormous amounts of tritium into the environment [6]. Tritium is applied to various everyday life areas, including self-lighting exit signs, watches, and other products that emit phosphorus lights when darkness emerges. Tritium is a β emitter with a physical half-life of 12.33 years and a biological half-life around 10 days. Its maximum range in air is 5 mm, 0.6 μm in tissue, and has maximum energy of 18.6 keV [3].

Ingestion of contaminated food and water, inhalation of contaminated water vapor and skin absorption are the ways that tritium can enter the body. Tritium behaves like hydrogen once it is inside the body and distributes throughout the tissues and organs. The internal tritium exposure releases low-energy beta particles that can harm neighboring tissues. The main form of tritium is tritiated water (HTO), although it may also be found as organically bound tritium (OBT). When HTO enters the body, equilibrium is quickly attained throughout the entire water pool of the body, and excretion occurs as HTO in sweat and exhaled water vapor and urine and feces [7].

Based on selected references [4, 8–11], several countries monitor the activity concentration and the effective dose of tritium in urine. Some studies estimated the tritium concentration for the general public as the baseline level, while in other studies, the radiation workers were evaluated for radiation protection purposes. In Indonesia, the previous

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study [12] estimated the internal radiation dose by tritium concentration in the urine of Nuclear Technology Center for Materials and Radiometry (PTNBR-BATAN) workers near TRIGA 2000 research reactor, Bandung.

In this study, the activity concentration of tritium in urine samples from The National Research and Innovation Agency employees was determined to estimate the levels of tritium (HTO) effective dose. Since HTO possesses identical chemical characteristic to ordinary water, it is typically encountered in both natural and occupational settings. In the event of an unintentional release of tritium, this information will be used as the baseline data for the dose assessment of those exposed in Indonesia. Furthermore, based on these measurement results, the effective dose of tritium was calculated in accordance with the recommendations of the International Commission on Radiological Protection (ICRP).

Materials and methods

Ethical clearance

This study was carried out with the approval of The Ethics Committee of the Faculty of Medicine, Syarif Hidayatullah State Islamic University Jakarta, No: 3674022P123442022070100001 and The Ethics Committee of National Research and Innovation Agency of Indonesia, No: 001/KE.05/SK/1/2023. Before sample collection, the participants written informed consent was obtained in accordance with the research ethics standards. Participants were also asked to fill in a form that contains data on age, gender, job category, and address.

Urine samples collection

The spot urine samples were obtained from radiation workers in non-NPP facilities and from administrative staff representing the general public during the period from January to March 2023. Most of the participants lived in Jakarta area. The age of the 40 participants ranged from 27 to 66 years old, including 18 females and 22 males. The participants involved in administration, calibration, chemical analysis, and health physics work were randomly selected as the research target. In this study, the participants were asked to collect urine in a 200 mL polyethylene jar. The urine samples were refrigerated at 4 °C, until sample pretreatment and measurement.

Urine samples preparation

The urine samples were prepared according to the following procedures. 25 mL of urine sample was mixed with 1.25 mg of powdered activated charcoal to be centrifugated

(3000 rpm, 30 min) to remove impurities and reduce foaming during distillation [11, 13]. Thereafter, for removing the charcoal and absorbed organic matter, the mixture was filtered on filter paper (Whatman® 42, GE, USA).

The filtered urine was transferred to a distillation flask, where it was mixed with 1.5 g of potassium permanganate (UPT BPPTK LIPI, Jakarta, Indonesia), 0.05 g of sodium hydroxide (Loba Chemie PVT Ltd, Mumbai, India), and heated. The chemical substances were used to destroy the organic compound found in urine [12].

Tritium measurement

The distilled urine sample and scintillation cocktail, Ultima Gold™ LLT (Perkin Elmer, Massachusetts, USA) were mixed in 8:12 ratio in a 20 ml polyethylene vial (Ø27×60.8 mm). The polyethylene vial (Perkin Elmer, Massachusetts, USA) was used because it has lower background than the glass vial and does not contain ⁴⁰K, so the polyethylene vial was preferred for low-activity sample counting [8, 14]. The vials were stored for 2–5 days in a dark place before measurement. Thereafter, all samples were counted using liquid scintillation counter (Quantulus 1220, Perkin Elmer Inc, USA) for 200 min by 3 cycles to obtain the tritium content in urine. The low-level count mode was chosen for counting and SQP(E) (Spectral Quench Parameter of the External Standard) was used as the quench indicator.

The efficiency of the measurement equipment was determined by utilizing ³H standards from Perkin Elmer for aqueous solution (Massachusetts, USA). The counting rate of a standard tritium sample was evaluated under the same measurement conditions. The efficiency of the method was estimated by using the following equation:

$$\varepsilon = \frac{R_s - R_b}{A_s} \times 100 \quad (1)$$

where ε is the counting efficiency (%), R_s is the counting rate of tritium standard samples (cpm), R_b is the background counting rate (cpm), A_s is the activity of tritium added to the standard sample (dpm). For this study, the method of external standardization was used.

The activity concentration of tritium in urine was determined by equation:

$$C = \frac{R_u - R_b}{\varepsilon \times V_d \times K} \quad (2)$$

where C is the tritium activity concentration in urine (Bq L⁻¹), R_u is the counting rate of the urine sample (cpm), V_d is the measuring volume of urine distillate (L), K is the coefficient of unit conversion between counts per minute and Becquerel (Bq) which is 60.

The minimum detectable activity (MDA) is an important element in the development of the LSC method. The MDA was calculated by the equation[15]:

$$MDA \sim \frac{2.71 + 4.65\sqrt{Rb \times T}}{T \times \epsilon \times Vd \times K} \quad (3)$$

where T is the counting time (minutes).

The parameter $R_b = 1.72$ cpm, $T = 600$ min, $V_d = 0.008$ L, $\epsilon = 51.7\%$. The MDA is 1.00 Bq L^{-1} .

The concentration of tritium inside the body was equivalent to the concentration in the urine. Based on ICRP, these data were used to calculate the annual effective dose of tritium received by the public by equation[16]:

$$H = 0.73 \times m \times \delta \times C \quad (4)$$

where H is the annual effective dose (Sv), 0.73 coefficient is the fraction of body water mass, m is body weight (kg), using a parameter for mean total body weight for man, 73 kg, and for woman, 60 kg, δ is the coefficient of the effective dose, which is $1.8 \times 10^{-11} \text{ Sv Bq}^{-1}$, C is the tritium activity concentration in urine (Bq L^{-1}), and the assumption of total body water volume per unit mass is 1 L kg^{-1} .

In this study, the background sample was obtained using twice distilled water with a low tritium content. The tritium standard solution was supplied from Perkin Elmer (Massachusetts, USA).

Results and discussion

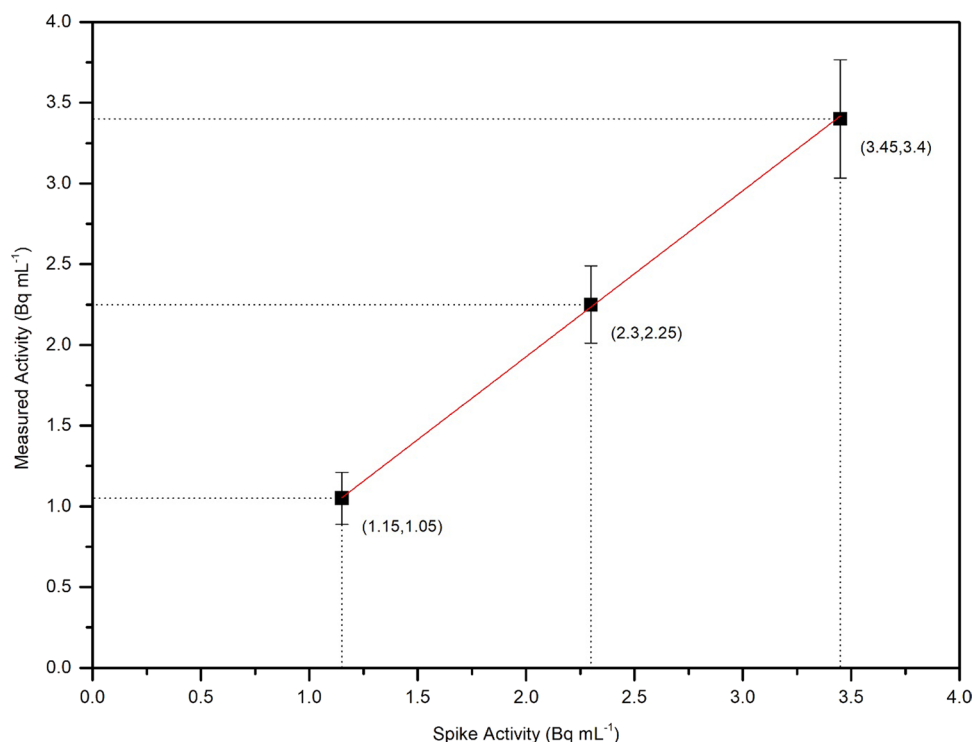
Accuracy of the method

The efficiency (5–350 channels) of the instrument was 51.7%, while the background value (5–350 channels) was 1.72 cpm. For ensuring the quality control of this technique, three urine samples from general public, not exposed to ^3H source, were intentionally contaminated with varying amounts of a standard tritium source. The spiked samples were treated as the samples (distillation and 8:12 mixing ratio) to verify the process. The results presented in Fig. 1 show there were no significant differences between the spiked values and the measured results for the samples.

Activity concentration of tritium in urine of selected radiation workers

The urine samples collected from 40 Indonesian random adults have been measured, and the results are presented in Fig. 2 for ^3H activity concentration in urine, Table 1 and Table 2 for ^3H concentration and effective dose by job category. From the Eq. (3), the MDA for this method was determined to be 1.00 Bq L^{-1} , as shown in Fig. 2. There were 20 (50%) of the results below the MDA. Therefore, the data results below MDA were excluded from the calculation. In this study, the range of the results of tritium concentration in urine sample was $1.04\text{--}3.49 \text{ Bq L}^{-1}$ and the average was

Fig. 1 Three urine samples with varying tritium amounts were spiked and analyzed for activity



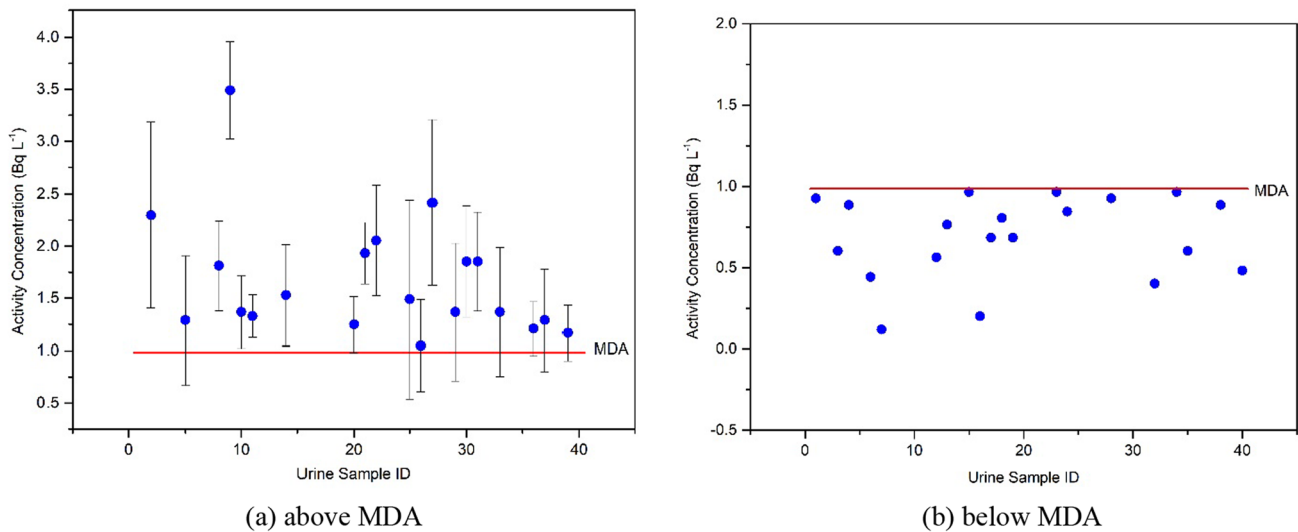


Fig. 2 Urinary activity concentration of tritium for each worker

Table 1 Activity concentration and effective dose of radiation workers based on job category

Job Category	Sample amount	Activity Concentration (Bq L ⁻¹)			Effective dose (nSv year ⁻¹)		
		Minimum	Maximum	Average	Minimum	Maximum	Average
Chemical Analysis	7	1.04	2.42	1.63 ± 0.48	0.83	1.97	1.41 ± 0.45
Health Physics	4	1.33	1.85	1.64 ± 0.26	1.28	1.47	1.42 ± 0.09
Calibration	3	1.29	3.49	2.36 ± 1.10	1.24	3.35	2.26 ± 1.06
General Public	6	1.17	1.93	1.39 ± 0.29	0.93	1.85	1.19 ± 0.35

Table 2 The activity concentration of HTO in urine samples collected from Indonesia and other countries

Year	Country	Sample	Sample amount	Activity (Bq L ⁻¹)	Effective Dose (nSv year ⁻¹)		References
					Male	Female	
2008	Finland	General Public	227	2.55	2.4		[11]
2010	Indonesia (PTNBR-Batan)	Radiation Workers	24	763	6.02		[12]
		Administration Staff	24	674	5.32		
2013	Korea	General public	50	2.8 ± 1.4	1.9 ± 0.7	2.4 ± 1.1	[9]
2015	China	General Public	27	2.09 ± 0.62	2.5 ± 1.7	2.9 ± 1.3	[10]
2015	Turkey	General Public	100	4.66 ± 1.94	4.56	3.54	[8]
2018	Sweden	General Public	37	< 2.1 (MDA)	22 (assumption)		[4]
2023	Indonesia	Radiation Workers in Non-NPP Facilities	14	1.79 ± 0.63	1.60 ± 0.63		This study
		General Public	6	1.39 ± 0.29	1.19 ± 0.35		

1.79 ± 0.63 Bq L⁻¹. These results also suggest that there are no significant differences related to the age and the gender for the tritium activity concentration.

In the results, there is one sample that is out of trend and has the highest ³H activity concentration (3.49 Bq L⁻¹). It was found that the participant was in fasting condition. The tritiated water (HTO) moves through the body fluids

circulation system before being eliminated. According to Butler and Leroy[17], the concentration of ³H in urine varies according to the amount of water consumed. If the individuals increase the amount of water consumed, the concentration of ³H in the urinary tract will diminish, while conversely, if the water intake level decrease, the concentration of ³H in the urine will increase.

The Table 1 summarizes the analytical results of the tritium level in urine and the effective dose of the workers. It revealed that the tritium activity concentration of radiation workers was slightly higher than the administration workers. However, the result show by the average value there are no significant differences in urine tritium values among workers in the same job categories. Based on the average value the job categories with higher urine tritium activity concentration were: calibration (2.36 Bq L^{-1}) > health physics (1.64 Bq L^{-1}) > chemical analysis (1.63 Bq L^{-1}) > administration (1.39 Bq L^{-1}).

Effective dose estimation

The internal dose from the participant was estimated using Eq. (4). The highest effective dose of tritium in urine was $3.35 \text{ nSv year}^{-1}$. Meanwhile, the average values of the effective dose received by the job category were: calibration ($2.26 \text{ nSv year}^{-1}$) > health physics ($1.42 \text{ nSv year}^{-1}$) > chemical analysis ($1.41 \text{ nSv year}^{-1}$) > administration ($1.19 \text{ nSv year}^{-1}$).

According to a Japanese study [18], 39% of the tritium ingested is derived from food, 52% from water that is consumed, and 9% from airborne particles. In Table 2, the tritium activity concentration and effective dose from several countries are listed. The results reveal that tritium level of Indonesians in this study, is the lowest among the other countries. Meanwhile, the previous study [12] shows that the Indonesian workers that work inside and near the research reactor have a significant tritium level compared to this study and the others, since tritium is the dominant radionuclide released from NPP. In such environment, tritium may have been released through both gaseous and liquid emissions originating from nuclear reactors before entering the body.

The differences observed of tritium activity concentration in the urine among the countries can be explained by the different environment and metabolism. It also has been widely studied that the biological half-time may vary depending on the ambient temperature range [17, 19]. As the ambient temperature rises, the concentration level of tritium in urine is decreased. In the comparison to other countries, it is reflected in Indonesia, which lies on the equator and where the climate is hot and humid.

The results of this study are also considered to be comparable to those detected in sea, rain, and tap water in Jakarta, Indonesia. Furthermore, the tritium level in Jakarta tap water was found to be low ($0.36 \pm 0.09 \text{ Bq L}^{-1}$) compared to the World Health Organization limit of $10,000 \text{ Bq L}^{-1}$ [20] (Table 3).

Based on ICRP [22] limit of $40,000 \text{ nSv}$, the results obtained were significantly below the dose limit for radiological exposure in the general population. However, in the event of tritium contamination in Indonesia, these findings

Table 3 The activity concentration of HTO in urine and environmental samples collected from Indonesia

Samples		Tritium concentration (Bq L^{-1})
Urine	Laboratory radiation workers	1.79 ± 0.63
	Administration staffs	1.39 ± 0.29
Tap water in sampling area		0.36 ± 0.09
Sea		$0.12\text{--}0.2$ [21]

can serve as reference data for evaluating the dose of radiation received by individuals, in the event of an unanticipated tritium leak.

Conclusion

The presented work found that the tritium concentration in urine of the Indonesian public differed from the results of other country results. According to this study results, the personal committed effective dose of the radiation workers in normal condition for tritium was lower than the annual effective dose limit recommended by ICRP. The results show that the concentration of tritium levels in urine varied substantially depending not only on their job category but also their metabolism, water intake and environment. However, further study is needed to check the tritium concentration and effective dose for Indonesian nuclear research reactor workers, periodically. As radiation workers in nuclear power plant are more likely to encounter tritium sources, it is important to implement appropriate protective measures and monitoring protocols to ensure their safety by regular monitoring.

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Declarations

Conflict of interest None of the authors of this article has any conflict of interest.

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