Screening procedure and criteria for tritium, gross alpha, and gross beta counting in urine samples for radiation emergency

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Abstract. Radiobioassay is generally performed for screening the internal contamination by alpha- and beta-emitting radionuclides to decide on medical treatment in a radiation emergency. In this study, the screening procedure and detection criteria for gross beta, tritium, and gross alpha are evaluated and verified using actinide radionuclide extraction techniques on urine samples. A portable liquid scintillation counter is introduced for on-site response in an emergency. The limit of detection is evaluated as a screening criterion for gross beta counting, with urine samples from three persons measured for validation. A working range considering the quenching parameter and criteria for tritium counting are also established. Moreover, a urine sample preparation procedure for gross alpha screening that eliminates interferences is proposed.

KEYWORDS: Gross alpha counting; gross beta counting; screening; radiation emergency; criteria

1 INTRODUCTION

In a radiation emergency, radiobioassay is conducted for screening the internal contamination and deciding on the medical intervention [1]. To rapidly assess contamination, screening techniques, such as gross alpha and beta were presented [2, 3], with the conventional method involving transferring collected samples to a laboratory for measurement. To expand the capacity and reduce the treatment time for gross beta screening, an on-site response system was recently proposed. Because the conventional screening techniques require modification to rapidly respond to an emergency, the criteria for gross beta screening are evaluated in this study. The utility of gross alpha screening techniques was demonstrated for highly contaminated cases [4]. However, if the internal contamination is in the mid-level radioactivity range, sample preparation is required to enhance the detection sensitivity and distinguish the contaminants from the background levels of naturally occurring radionuclides. Tritium in urine is characterized by relatively low emitting energy (E_{max} = 18.6 keV) in comparison with other beta emitting radionuclides. To improve the counting efficiency of tritium, the separation energy range for tritium must be considered in calibrating and counting samples. Additionally, screening criteria for tritium requires evaluation based on the committed effective dose.

In this study, a screening process involving extraction chromatography on urine samples for tritium, gross alpha, and gross beta during a radiation emergency (Fig. 1) is established. The screening results are validated using urine samples from volunteers and certified reference materials (CRMs). Additionally, the screening criteria for each method are discussed.

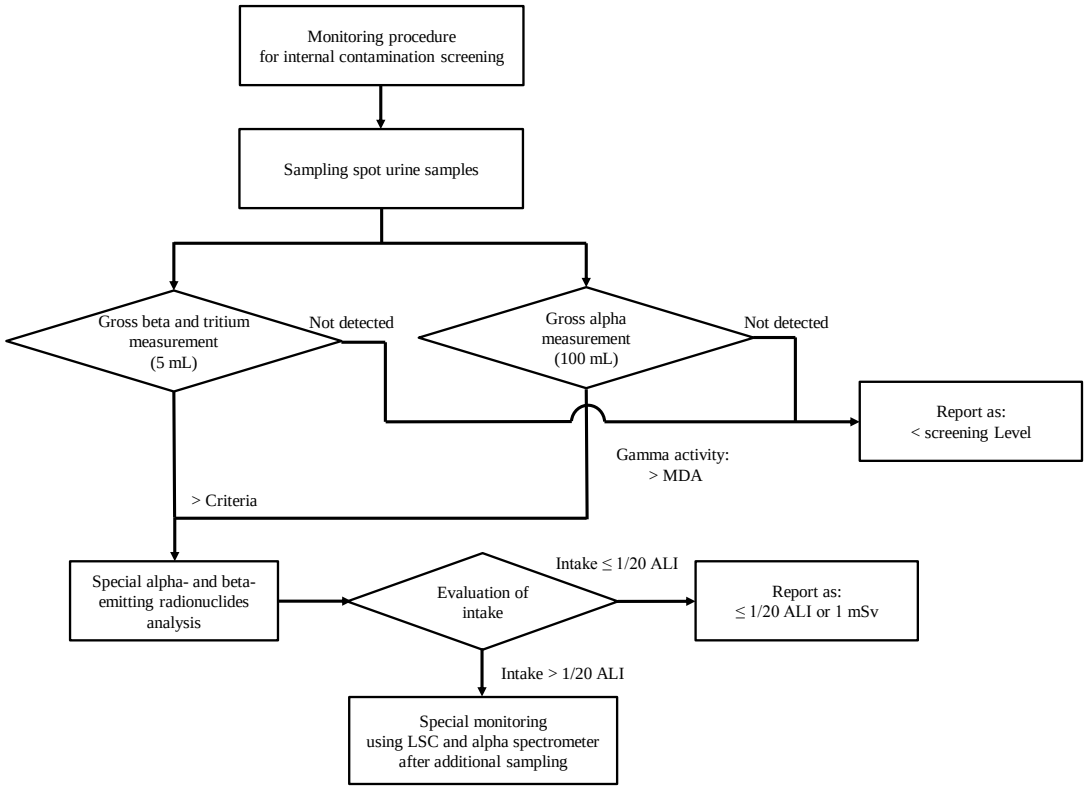

Figure 1: Internal contamination screening flow diagram for gross alpha- and beta-emitting and radionuclides including tritium.

2 MATERIAL AND METHOD

2.1 On-site screening criteria

2.1.1 Gross beta

The criteria for gross beta screening of urine samples were established. A portable liquid scintillation counter (Triathler LSC, HIDEX Co.) was used for on-site response in an emergency. The urine samples spiked with ^{90}Sr CRMs were mixed with a liquid scintillator (Ultima-gold, PerkinElmer Co.) at a 1:19 ratio, followed by evaluation of the total count rate and contamination level of beta energy bands. The internal dose by ^{90}Sr for humans was evaluated and discussed for reasonable criteria. Moreover, urine samples from three volunteers not associated with the radiation works were measured to verify the effectiveness of the screening method.

2.1.2 Tritium

The counting efficiency of the portable LSC and working range for the correction of the quenching parameter (QP) for tritium were estimated. The efficiency correction curve, involving the efficiency versus QP for a tritium quench standard set revealed a perfect correlation (R^2 = 0.99) (Fig. 2). The working range was also evaluated with other tritium certified reference quench standard sets. The counting results were compared with reference values to enable the selection of a working range with good accuracy. The urine samples spiked with CRMs were measured to verify the working range. Moreover, the screening criteria for tritium were evaluated based on the estimated internal dose.

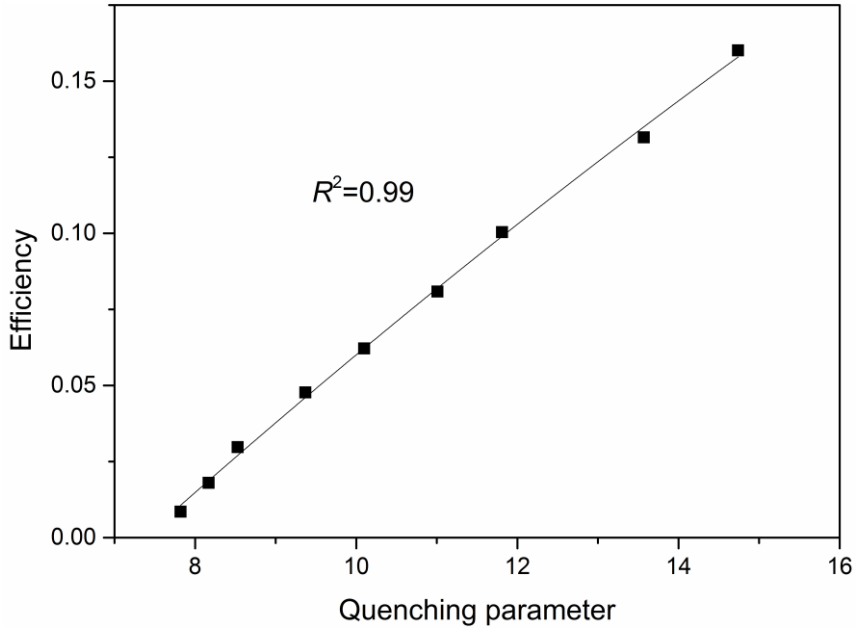

Figure 2: Quenching correction curve for tritium counting showing a perfect correlation between the efficiency and quenching parameter (QP).

2.2 2.2. Gross alpha screening using extraction chromatography

The urine sample preparation procedure for gross alpha screening is presented below. To eliminate interference, the raw urine samples were filtered using a 0.22 μm membrane filter. Concentrated nitric acid was added to the samples and then heated at 120 ℃ for 2 h for organic matter oxidation. Extraction chromatography was performed with an actinide resin (P,P'-di(2-ethylhexyl) methanediphosphonic acid; Eichrom Co.) involved. The urine samples were mixed with resin for a fixed duration and the resin was collected using a filtering set. Isopropanol was used to flush the alpha-emitting radionuclides from the resin. The solution from flushing was repeatedly dried after adding concentrated nitric acid and sulfuric acid for eliminating residual organic interferences. Approximately 2 mL of hydrochloric acid (0.5 M) was added to each dried residue and transferred into the counting vials. The liquid scintillator was mixed with samples in the counting vial to a final volume of about 20 mL [5]. A low background LSC (1220 Quantulus, PerkinElmer Co.) was used for analyzing the samples, with each sample counted 10 times considering statistical uncertainties. The screening criteria were then evaluated using counting results.

3 RESULTS AND DISCUSSION

3.1 Screening criteria

3.1.1 Gross beta

The counting efficiency of gross beta measurements exceeded 0.96 for ^{90}Sr reference samples, consistent with the criteria for the instrument [6]. Verification measurements using reference urine samples spiked with CRMs were conducted. The relative bias, precision, and root mean square errors (RMSE) for the measurements were 26%, 30%, and 40%, exceeding the criteria for radiobioassay performance testing [7]. The differences between our testing and the conventional approach are attributed to the quenching effect of the impurities and color of the urine samples. Generally, a quenching correction is applied for the quenching effect [8]. However, calibrating the portable LSC following each sample status for an on-site emergency response remains difficult. In addition, based on the internal dose from ^{90}Sr exposure, the 1 mSv for an adult in a day after intake is 96.9 dpm/mL. To satisfy the measuring condition for the MDA of the portable LSC, the counting time must exceed

60 min and sample volume must extend to 8 mL (Fig. 3) due to its relatively low shielding capacity. This renders its application for major screening during an emergency difficult.
In this study, the limit of detection (LOD) was used as a criterion for gross beta screening. The standard deviation for the sample counting results in comparison with the background counts was used to determine the presence of contaminants using the LOD equation [9]. Urine samples from three participants, who were unrelated to the radiation works, were analyzed to validate the criteria. Because the results were below the LOD level of 396 cpm (Fig. 4), this value is considered a reasonable screening criterion in an emergency.

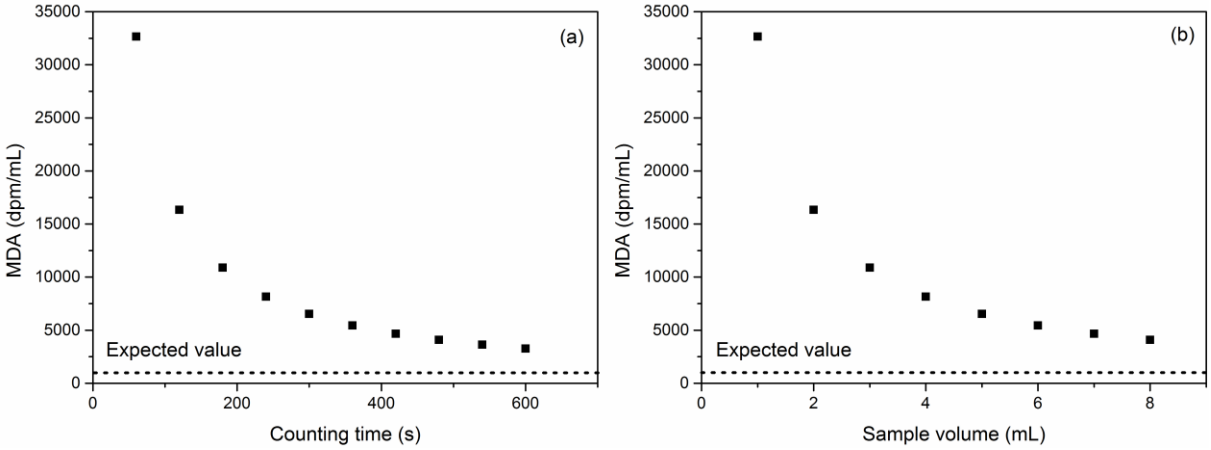

Figure 3: MDA variation against counting time (a) and sample volume (b) correction. The dotted lines represent the expected values for 1 mSv of ^{90}Sr.

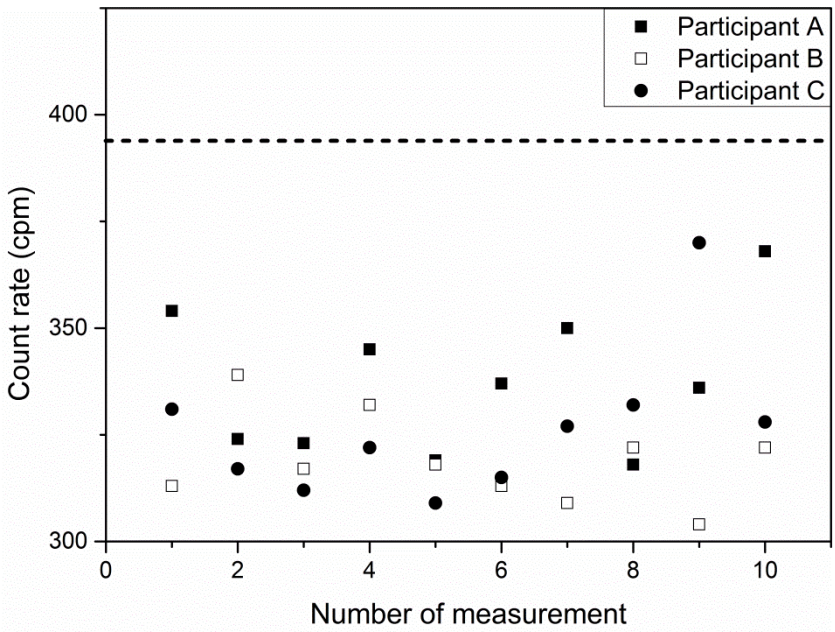

Figure 4: Comparison results of gross beta counting. The dotted line represents the LOD value (396 cpm).

3.1.2 Tritium

The working range including the detection efficiency calibration was estimated using a quenching standard set. The range was evaluated within 10% of the relative bias. The results are 10–15 for QP, with values below 10 showing poor accuracy due to impurities in the samples (Fig. 5). In counting samples below the working range, a sample preparation step is required to prevent the underestimated measurements due to interferences [8]. The quenching correction and working range were validated using urine samples of three participants spiked with CRMs. The counting results are consistent with reference values. (Table 1 and Fig. 5).

In addition, the counting condition was evaluated based on internal dose from tritium intake. Within four days of intake, the tritium activity corresponding to 1 mSv for an adult male was 60,000 dpm/mL. Therefore, the optimum counting time and sample volume were 120 s and 2 mL, respectively, according to the MDA of the counting condition (Fig. 6).

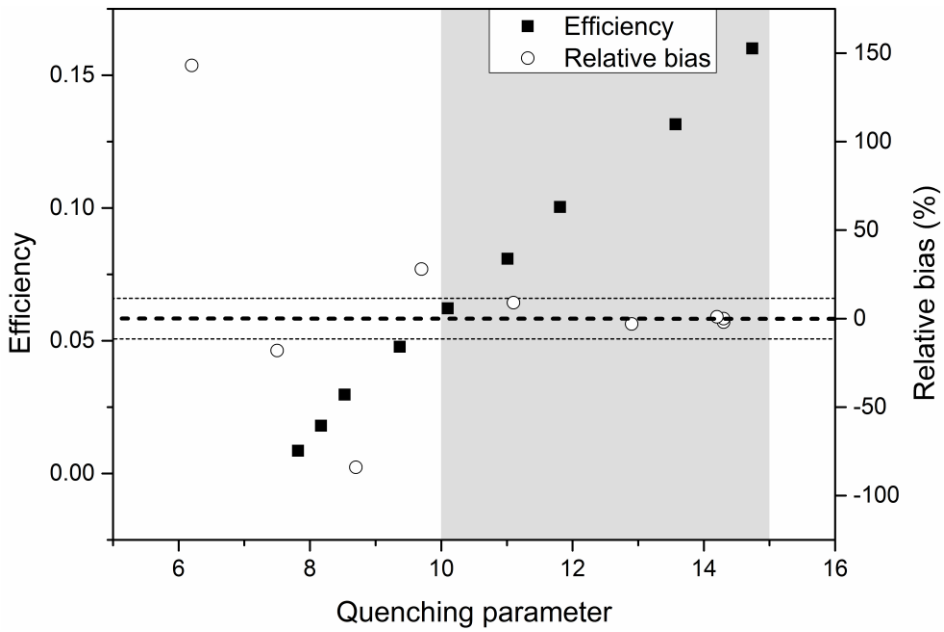

Figure 5: Comparison of the efficiency and relative bias against QP for reference samples. The dotted line represents the accuracy within 10%, while the shaded area covers the working range of 10–14.

Table 1: Validation results for tritium of reference urine samples.

Sample	QP (%)	Relative bias (%)	Relative precision (%)	RMSE (%)	Trueness
Participant A	12.3	4.4	2.6	5.1	Yes
Participant B	11.0	2.8	5.8	6.4	Yes
Participant C	11.4	3.5	6.6	7.5	Yes

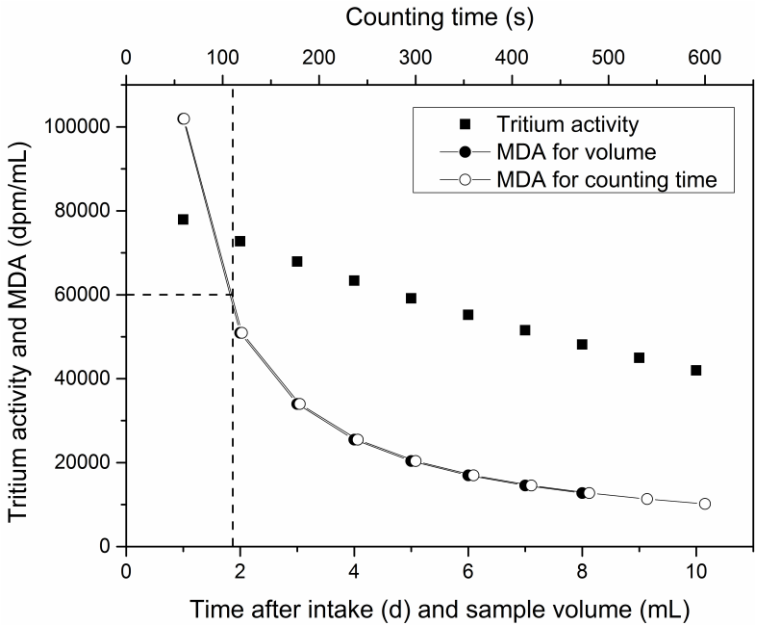

Figure 6: Comparison of tritium activity results following the time after intake and the MDA for the sample volume and counting time. The dashed lines represent the expected values according to tritium activity.

3.2 Gross alpha urinalysis

To evaluate the counting efficiency, reference samples were repeatedly measured. The urine samples collected from volunteers were spiked with ^{241}Am and ^{240}Pu CRMs. The counting results exhibited an efficiency of over 60%, and this approach may be used as an effective screening method during emergencies. Additionally, this procedure is advantageous for performing quantitative measurement through gross alpha counting. Generally, measurements involving extraction chromatography and alpha spectrometry are time-consuming, requiring over three days [10]. This procedure, including the LSC counting, was performed within 8 h. Moreover, the conventional gross alpha urinalysis exhibits a low counting efficiency, with difficulty to correct the radiation energy and quenching effect from raw urine sample interferences. This implies raw samples require purification to enhance the counting efficiency during an emergency.

The evaluated MDA shows values that were saturated to 100 Bq/L following the increase in the counted sample volume. A previous study indicates that the critical monitoring quantity for ^{241}Am, corresponding to 0.1 mSv is 0.0214 mBq/L [11]. Therefore, this is inappropriate for use as a screening reference value based on the internal dose. Additional preparation and alpha spectrometry are needed to enhance the MDA level for identification and quantitation.

The LOD was used to screen for positive and negative values of the gross alpha counting. Nine ultra-pure water samples and 11 urine samples collected from volunteers, who were unrelated with the radiation works, enabled the evaluation of the background level. All samples were prepared and counted using the same procedure. The average LODs of the ultra-pure water and urine samples were 0.65 cpm and 0.69 cpm, respectively. The values for the water and urine samples were rather similar. One hundred urine samples collected from 9 persons were measured for gross alpha counting (Fig. 7). Most of the results were below the gross alpha counting level within the LOD criteria. Some data (participants G–I) exceeded the LOD value, with these identified as samples from smokers based on interviews. Previous studies indicate that the background levels of naturally occurring radionuclides for smokers are relatively higher than those for others [12, 13].

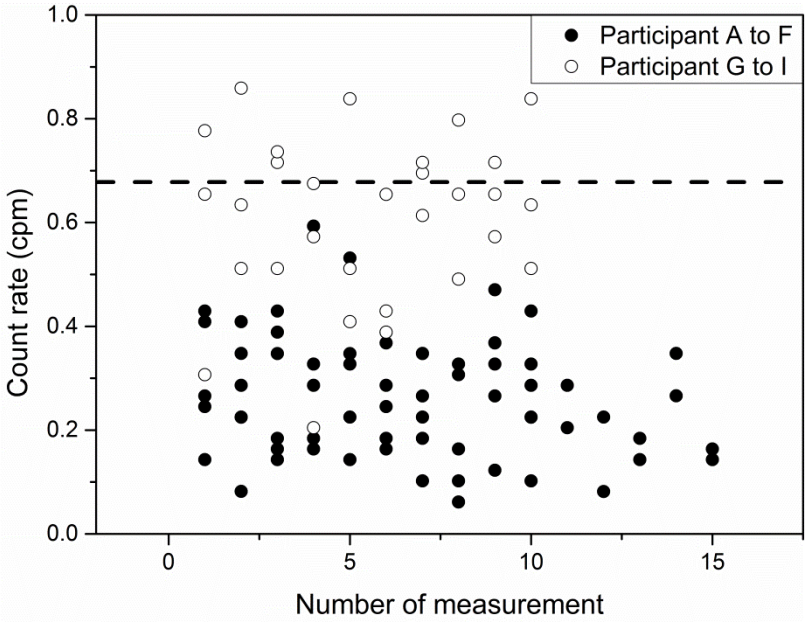

Figure 7: Counting results of urine sample from normal persons. The dotted line represents the LOD values of urine samples (0.69 cpm).

4 CONCLUSION

Radiation accidents require urgent attention to reduce the radiation risk. Generally, screening techniques are employed for classifying contaminants in an emergency situation. In this study, a screening procedure and criteria for identifying internal contamination by alpha- and beta-emitting radionuclides were proposed and verified. In particular, gross beta, tritium, and gross alpha counting after preparing urine samples were established classification criteria in comparison with background radiation. However, because a radiation accident generally increases the background level, it can trigger effects to determine the MDA and screening criteria according to the response sites. To enhance the counting efficiency and accuracy, the background shielding system needs to be studied using highly shielded counting instruments in the future. The background radiation levels for humans must be corrected to enhance the accuracy of gross alpha counting because naturally occurring radionuclides are the main source of background radiation in the body. Additionally, the quantities measured are limited for low level contamination. Correcting the effects based on lifestyle, such as smoking, is essential to reduce uncertainties in the results.

ACKNOWLEDGEMENTS

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