HHS Public Access

Author manuscript

J Radioanal Nucl Chem. Author manuscript; available in PMC 2021 November 05.

Published in final edited form as:

J Radioanal Nucl Chem. 2021 August ; 330(1): 381–384. doi:10.1007/s10967-021-07950-2.

Limit of detection comparison on urine gross alpha/beta, H-3, and P-32 analysis between different liquid scintillation counters

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Abstract

As part of the Centers for Disease Control and Prevention's post-radiological/nuclear incident response mission, we developed rapid bioassay analytical methods to assess possible human exposure to radionuclides and internal contamination. Liquid scintillation counting (LSC) is a valuable analytical tool for the rapid detection and quantification of gross alpha/beta-emitting radionuclides in urine samples. A key characteristic of this type of bioassay method is its detection sensitivity for the priority threat radionuclides. We evaluated the limit of detection of selected LSC instruments to determine which instrument can be used when low-dose measurement is important.

Keywords

liquid scintillation counting; urine bioassay; Quantulus 1220; Tri-Carbs 3110 and 5110; Quantulus GCT6220; limit of detection

Introduction

Liquid scintillation counting (LSC) is a technique commonly used for rapid bioassays [1,2]. LSC efficiency for alpha and high-energy beta measurement is about 90 % to 100 %. The efficiency for low-energy beta radionuclides, such as tritium (H-3), is lower (5 % to 40 %), depending on quenching effects [3], and can be determined primarily by LSC.

Our laboratory uses Quantulus1220, Tri-Carb3110, Tri-Carb 5110, and Quantulus GCT6220 instruments from PerkinElmer. Each instrument type has advantages and disadvantages. The greater shielding of the Quantulus1220 reduces background interference, but Tri-Carb series instruments have higher capacity (420 samples versus 60 for Quantulus1220). Quantulus GCT6220, the newer version of Tri-Carb, provides high capacity and greater sensitivity by

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The authors declare that they have no competing financial interest.

reducing interferences from background radiation due to its guard compensation technology (GCT).

Limit of detection (LOD) is an important characteristic for each analytical method. It determines how well we can detect priority threat radionuclides using available urine samples collected days after a contamination incident and determine minimal count time to maximize throughput. This article presents details of LOD determinations for gross alpha/beta, H-3, and P-32 radionuclides for each listed instrument and compares the results using the simple “dilute and shoot” approach without additional urine purification or nuclide separation. Other bioassay laboratories can use this LOD information to decide which of these LSC instruments might best match their requirements for radiobioassy or population monitoring after a radiological or nuclear incident/accident.

Experimental

Reagents and materials

The Ultima Gold AB cocktail (UGAB) from PerkinElmer was used for gross alpha/beta analysis. For quench curves preparation, we used 99 % nitromethane and 99 % urea from ACROS Organics, and a black tea solution. For the black tea solution, we steeped 1 regular tea bag (Lipton black tea from any grocery store) in 200 mL of boiling water for 10 minutes then cooled the liquid and used it for quenching. Deionized water (≥ 18 MΩ·cm) from an Aqua Solutions Ultrapure Water System (Aqua Solutions) was used for all solutions.

Base urine was collected through anonymous human donations (in accordance with Centers for Disease Control and Prevention Institutional Review Board protocol 3994). The base urine was diluted approximately 4-fold to decrease gross beta activity normally present from potassium-40 (K-40) [4]. This is the common approach for LOD measurement when matrices contain the analyte of interest.

Four gross alpha/beta standards for LOD determination were prepared by spiking the diluted urine from 0 to 16 Bq/L americium-241 (Am-241) and from 0 to 90 Bq/L strontium-90/yttrium-90 (Sr-90/Y-90) at levels near the expected LOD. The expected LOD levels were estimated as standard deviation multiplied three times (3SD) after 20 blank urine analyses. For tritium LOD determination, four standards were prepared by spiking the diluted base urine with H-3 from 0 to 82 Bq/L. The phosphorus-32 (P-32) sample for LOD determination was prepared by spiking the diluted base urine with approximately 105 Bq/L P-32. Analyzing base urine many times by LSC for gross alpha/beta, we found that the range for LOD standards from 10 till 110 Bq/L looks reasonable for any beta nuclides. Table 1 shows the suggested LOD standards specific activities for each of the nuclides used for the LOD determination. All radioactive source solutions were traceable to the National Institute for Standards and Technology (Gaithersburg, MD, USA). All these solutions used for spiking were purchased from Eckert & Ziegler Analytics.

Instrumentation and labware—For this study, we used Quantulus1220, two Tri-Carbs 3110 (#1 and #2), Tri-Carb 5110, and Quantulus GCT6220 instruments (all from PerkinElmer). We also used 20-mL LSC plastic vials (PerkinElmer) for LSC analysis,

polyethylene bottles of different volume (Fisher Scientific), and a Brinkman bottletop dispenser with capacity from 5 mL to 25 mL (Brinkman Instruments) for cocktail dispensing. Additional supplies included four electronic pipettes with total volume range from 5 μL to 5 mL (Eppendorf) and a high-precision (0.0001 g accuracy) analytical balance (Mettler-Toledo).

LSC analysis and sample preparation for gross alpha/beta and H-3—For LSC analyses, we used all instruments in alpha/beta mode and our liquid scintillation counting approach [5, 6], which includes pulse shape analysis (PSA) or pulse decay discriminator (PDD) setting optimization and quench curves preparation. H-3 and gross alpha/beta analyses were combined in the same gross alpha/beta method which is recommended for urine screening. The quench curves for the listed radionuclides were prepared using the optimal quench agents. Those agents included nitromethane for Am-241 and Sr-90/Y-90 quench curves, black tea and 10% urea mixture for H-3 quench curves, and water and nitromethane or a black tea and nitromethane mixture (depending on the instrument) for P-32 quench curves [7]. The parameters such as sample analysis time, external standard analysis time, type of cocktail, and sample/cocktail volume were optimized for 20-mL vial geometry [5, 6]. In addition, a region of interest (region in which the given nuclide will be counted) was optimized based on spectra for each nuclide from each instrument. Table 2 shows the optimal parameters for each instrument.

Finally, for LOD determination, 5 mL of each urine LOD standard (blank and S1 through S3) was mixed with 15 mL of UGAB cocktail in 20 mL LSC plastic vials until the mixture was uniform. The LSC vials with solutions were placed on the LSC counter tray and LSC analysis was performed according to the chosen parameters.

Sample preparation for P-32—For short-lived radionuclides such as P-32 (half lifetime = 14.2 days), we used another approach for LOD standards preparation. First, we prepared the original diluted urine, spiked with P-32, (close to 550 mL) with approximately 105 Bq/L activity. We then used this urine, spiked with P-32, to prepare 20 samples by mixing 5 mL of sample with 15 mL of UGAB cocktail as LOD standard 4 (S4) and analyzed those on all instruments according to the chosen parameters in alpha/beta mode (see Table 2). After 10 days, when P-32 decayed to approximately 60 Bq/L, we prepared 20 other samples and analyzed those on each instrument as LOD standard 3 (S3). After an additional week, when the activity was around 45 Bq/L, we prepared and analyzed 20 samples as LOD standard 2 (S2). After another 2 weeks, we prepared 20 samples and analyzed those as LOD standard 1 (S1). After an additional month, when P-32 activity was close to 0 Bq/L, we prepared 20 samples and analyzed those as blanks.

Results and discussion

LOD calculation

To compare instruments, we calculated the LOD for gross alpha/beta, H-3, and P-32, using the suggested method as described in the Clinical and Laboratory Standards Institute method EP17-A2 [8]. Four LOD urine standards for alpha (Am-241), beta (Sr-90/Y-90), and H-3 were analyzed 20 times on each instrument on different days. For P-32, each standard was

analyzed 20 times on each instrument, each standard was taken from the same pool, but on different days and correspondingly with different activity level, as described in *Sample preparation for P-32* part. In total, we collected 20 data points on each standard for each nuclide and instrument. LODs were determined according to Eq. (1) [8]:

$$\text{Activity}_{\text{LOD}} = [\text{M}\,b + 1.645\,(\text{SD}\,b + I)] \;/\; [\,1 - 1.645\,S] \quad (1)$$

where M b – the blank average activity (Bq/L) and SD b – the standard deviation for blank average activity (Bq/L), I – the intercept and S – the slope of the curve of standard deviation (SD) versus LOD standards activity (Figs. 1 - 3 are shown as examples).

The calculated LOD results are summarized in Table 3. As indicated, the instrument with the most shielding (Quantulus1220) had a lower LOD for each nuclide due to the lower background. A Quantulus GCT6220 is comparable with Quantulus1220 but had a slightly higher LOD for the same nuclides. Tri-Carbs series instruments are comparable between each other; however, they have a higher LOD than the Quantulus1220 and Quantulus GCT6220. This means that GCT option added to Tri-Carb decreases the background providing a lower LOD.

Another important characteristic for the choice of the instrument is the analysis turnaround time (sample analysis time plus double external standard analysis time) [6]. The analysis time per sample is 5 minutes for all instruments. This analysis time provides reasonable minimum detectable activity and good counting statistics [5,6]. However, the external standard analysis time is different for Tri-Carb and Quantulus series instruments because the external standards in Tri-Carbs have higher activities than in the Quantulus instruments. Instruments Tri-Carbs 3110 and 5110 series have Ba-133 with activity about 20 μCi as external standard while Quantulus 1220 instrument has Eu-152 with 1 μCi activity as external standard and Quantulus GCT6220 instrument has Ba-133 with 1 μCi activity as external standard. As a result, for Tri-Carb 3110 and 5110, the external standard is typically counted to a 2 sigma error of 0.5% (normally shown as default option in the window for instrument parameters), which is in the range of 10 to15 seconds, compared with 5 minute for Quantulus 1220 and GCT6220 when the same 2 sigma error of 0.5% option is chosen. And this will increase the total analysis time till 15 minutes for these instruments. Therefore, the optimization of external standard analysis time was done for instruments Quantulus series and the optimal time was found as 1 minute [5,6]. Thus, the total analysis time for Tri-Carbs is approximately 6 minutes per sample and approximately 7.5 minutes per sample for the Quantulus instruments.

Conclusions

Quantulus1220 has the lowest LOD for all listed nuclides and it is the best option when a very low LOD is required. Quantulus GCT6220 gives slightly higher LOD than Quantulus 1220, but it has higher sample capacity (420 samples versus 60 for Quantulus 1220). The Quantulus GCT6220 will be the better option when numerous samples should be analyzed with the lower LOD. When a very low LOD is not an issue, Tri-Carb series instruments might be preferable because the sample analysis time is slightly less for these instruments.

Acknowledgements

The authors thank David Saunders for his advice during this work and Po-Yung Cheng for the LOD calculations.

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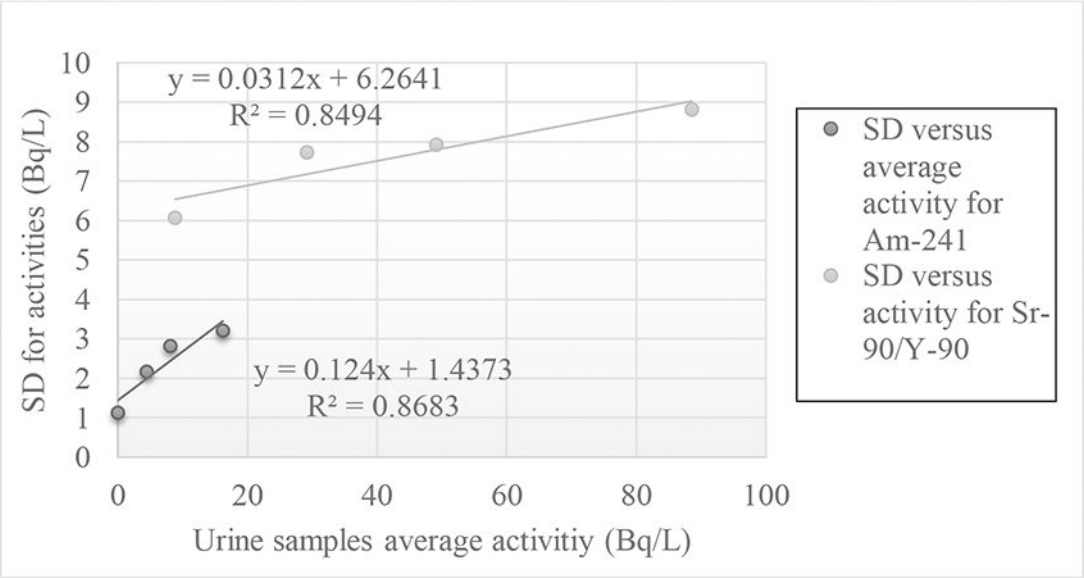

Fig.1.

Standard deviation (SD) versus alpha (Am-241)/beta (Sr-90/Y-90) activities in LOD samples (20 analyses) for Quantulus 1220

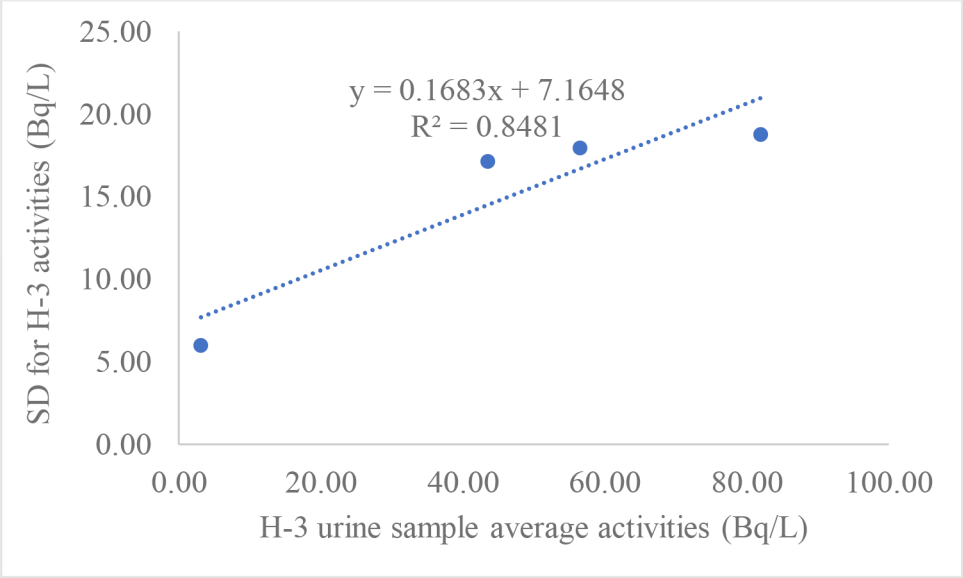

Fig.2.

Standard deviation (SD) versus H-3 average activities in LOD samples (20 analyses) for Quantulus GCT6220

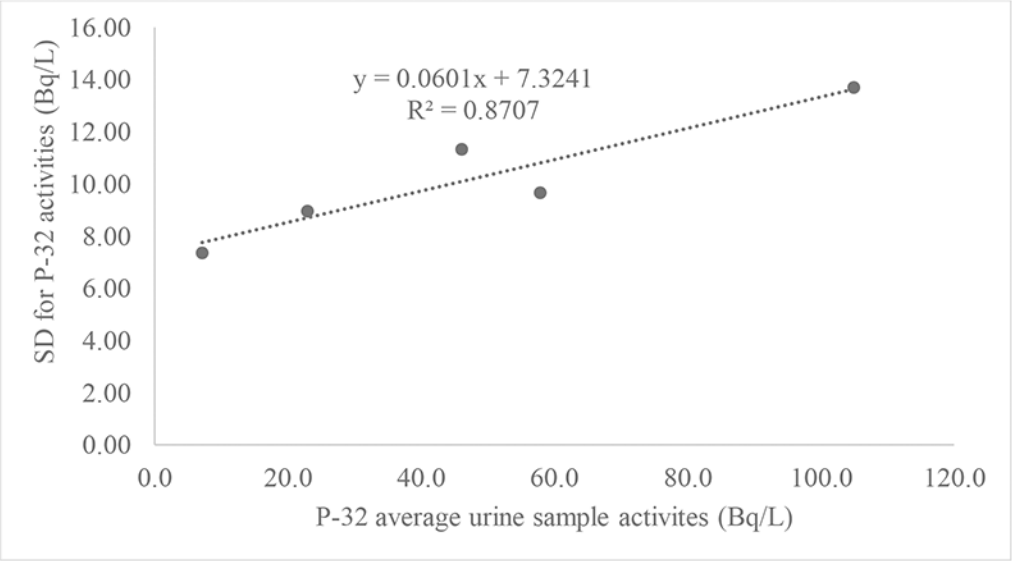

Fig.3.

Standard Deviation (SD) for P-32 activities versus P-32 average activities for LOD urine samples on Tri-Carb 5110

Table 1

Suggested gross alpha (Am-241)/beta (Sr-90/Y-90), H-3, and P-32 activities for LOD standards.

	Suggested activities for LOD standards (Bq/L)				
Nuclides	**Blank**	**S1**	**S2**	**S3**	**S4**
Am-241	0	4	8	16	
Sr-90/Y-90	10	30	50	90	
H-3	3	44	56	82	
P-32	10	23	46	58	105

Table 2

Liquid scintillation counting method parameters for different instruments

Parameter	Quantulus 1220	Tri-Carb 3110 #1	Tri-Carb 3110 #2	Tri-Carb 5110	Quantulus GCT6220
PSA/PDD setting	90	125	165	135	120–170
Sample volume (mL)	5	5	5	5	5
Cocktail volume (mL)	15	15	15	15	15
Sample analysis time (min)	5	5	5	5	5
External std analysis time	60 sec	2 Ω (15 sec)	2 Ω (15 sec)	2 Ω (10 sec)	60 sec
Alpha (Am-241) region of interest (channels or keV)	1-1024 channels	0–300 keV	0–200 keV	0–1000 keV	0–450 keV
High energy beta (Sr-90/Y-90 or P-32 region of interest (channels or keV)	1-1024 channels	0–2000 keV	0–2000 keV	0–2000 keV	0–2000 keV
Low energy beta (H-3) region of interest (channels or keV)	1-250 channels	0–18.6 keV	0–18.6 keV	0–18.6 keV	0–18.6 keV

Table 3

Gross alpha (Am-241)/gross beta (Sr-90/Y-90), H-3, and P-32 limits of detection (LOD) estimates for Tri-Carb instrument series, Quantulus 1220 and GCT6220 in plastic vials with linear fit

	LOD (Bq/L)			
Instrument	**Gross alpha**	**Gross beta**	**H-3**	**P-32**
Quantulus 1220	5.79	31.3	35.4	23.0
Tri-Carb 3110	12.6	44.6	48.2	37.8
Tri-Carb 5110*	-	-	57.6	34.1
Quantulus GCT6220	7.58	40.3	39.2	24.9

[1] Gross alpha/beta LOD for Tri-Carb5110 was close to Tri-Carb3110