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J Radioanal Nucl Chem. Author manuscript; available in PMC 2021 August 18.

Published in final edited form as:

Author manuscript

J Radioanal Nucl Chem. 2021 January 3; 327(1): 513-523. doi:10.1007/s10967-020-07493-y.

# Urine gross alpha/beta bioassay method development using liquid scintillation counting techniques

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# Abstract

In the case of a radiological or nuclear incident, valuable information could be obtained in a timely manner by using Liquid Scintillation Counting (LSC) technique through fast screening of urine samples from potentially contaminated persons. This work describes the optimization of LSC parameters on PerkinElmer (PE) Tri-Carb and Quantulus GCT series instruments to develop a rapid method for screening urine in an emergency response situation.

# Keywords

liquid scintillation counting; gross alpha/beta urine bioassay; Tri-Carb; Quantulus GCT; emergency response

# Introduction

Radionuclides are present in the environment from many sources. They originate from natural sources, from nuclear weapons testing fallout, and from nuclear and non-nuclear industry discharges. Because of their increased presence in the environment, ease of entry into the food chain, use in nuclear medicine applications, and potential release in radiological incidents, radionuclides pose a risk to humans [1]. The persistence of radionuclides varies. Their effects range from possible increased lifetime risk for various types of cancer to acute radiation syndrome and death [2]. Therefore, an important first step in radiobioassay is to determine the gross alpha and beta emission levels in urine of people contaminated with radionuclides. Liquid scintillation counting (LSC) can be a fast and effective technique for addressing these concerns, especially during an emergency response to a radiological incident when many people might be contaminated.

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Since the 1950s, after the discovery of scintillation in organic compounds under nuclear radiation, LSC has become a widely used technique for detection and quantitative measurement of radioactivity [3–5]. The scintillation process is different for alpha, beta, and gamma decay. Because of the unique pulse shape and slower pulse decay times of alpha detection in LSC, the technique can be used to distinguish alpha particles from most other nuclear decay radiations [6].

Radiochemists have used LSC for urine bioassay, primarily for tritium analysis [7–9]. Some groups of scientists applied LSC for urine gross alpha/beta bioassay as well [10,11]. We would like to continue these efforts.

Modern LSC instruments are equipped with a pulse decay discriminator (PDD) or pulse shape analysis (PSA), which allows simultaneous analysis of alpha and beta emissions and collection of the analytical results in different channels through multiple channel analyzer technology. These results are affected by sample matrix, PDD/PSA setting, type of cocktail, sample/cocktail volume, and count duration [6]. Our task was to optimize these parameters and validate the urine gross alpha/beta screening method on PE Tri-Carb and Quantulus GCT series instruments.

# Experimental

# **Reagents and materials**

We used Ultima Gold (UG), Ultima Gold XR (UGXR), Ultima Gold LLT (UGLLT), Ultima Gold AB (UGAB), and OptiSafe HiSafe-3 (OPHS-3) (PerkinElmer Company) for cocktail optimization, and 99% nitromethane (ACROS Organics) for quenching. Deionized (DI) water ( 18 MQ cm) from an Aqua Solutions Ultrapure Water System, Aqua Solutions, Inc., was used for all solutions. "Base urine" was collected according to Centers for Disease Control and Prevention Institutional Review Board (IRB) protocol 3994 through anonymous human donations and acidified to 1% HNO3. All radioactive source solutions were traceable to the National Institute for Standards and Technology (NIST) (Gaithersburg, MD, USA). Urine gross alpha/beta quality control materials (Low QC and High QC) were purchased from Eckert & Ziegler Analytics, Inc. Those QC materials were base urine (BU) spiked with Am-241 and Sr-90/Y-90 at low and high levels. Gross alpha/beta reference material (RM) and gross alpha/beta high calibration range material (HCR) were base urine spikes prepared in our laboratory with NIST-traceable reference solutions of Am-241 and Sr-90/ Y-90 at different levels. The reference solutions used for spiking were purchased from Eckert & Ziegler Analytics, Inc. Four gross alpha/beta standards for the limit of detection (LOD) determination were prepared in our laboratory by spiking diluted urine with Am-241 in the range of 0-25 Bq/L and Sr-90/Y-90 in the range of 12-65 Bq/L. Nine gross alpha/ beta proficiency testing (PT) materials (PT1-PT9), used for ongoing performance testing requirements, were prepared in our laboratory by spiking base urine at nine different levels with Th-230 (70-6300 Bq/L) and Sr-90/Y-90 (1680-108,000 Bq/L) NIST-certified standards provided by ERA, a Waters Company.

#### Instrumentation and labware

For LSC analysis, we used Tri-Carb 3110 #1 and #2, Tri-Carb 5110, and a Quantulus GCT6220 instrument, and 20-mL LSC plastic vials (all from PerkinElmer). The Quantulus GCT6220, a newer version of the Tri-Carb, has a guard compensation technology (GCT) option that can decrease background interference for alpha and beta nuclides. We used a high-precision analytical balance with an accuracy of 0.0001 gm (Mettler-Toledo, LLC). We also used 15 mL and 50 mL conical polypropylene tubes (Becton Dickinson) for different solution preparation, a Brinkman bottletop dispenser with capacity from 5 mL to 25 mL (Brinkman Instruments, Inc.) for cocktail dispensing, and four electronic pipettes with total volume range from 5  $\mu$ L to 5 mL (Eppendorf, Inc).

# **Results and discussion**

#### Method development

**Minimum detectable activity**—Minimum detectable activity (MDA) determination is a critical component of LSC method development. Currie L.A. proposed the following formula for radionuclide MDA determination [12], which was applied to LSC [13] as Eq. (1):

$$MDA \sim (2.71 + 4.56 \times B^{1/2}) / (T \times E \times V \times K)$$
<sup>(1)</sup>

where *B*-background (counts), *T*-count time in minutes, *E*-instrument efficiency, *V*-sample volume, K-coefficient for activity units connecting counts per minute (CPM) with Becquerel (Bq) or picocurie (pCi). LSC instruments provide MDA for each measurement, which makes it easier to optimize parameters such as sample volume (V) and count time (T).

**Sample volume**—For a 20 mL vial geometry we varied the amount of aqueous sample from 2 mL to 8 mL with the amount of cocktail correspondingly from 18 mL to 12 mL. The sample/cocktail volume depends on the type of cocktail, sample matrix, and amount of sample (some samples might be limited in available volume). Theoretically, the larger the sample, the lower the MDA. Therefore, preference should be given to higher sample volume. However, greater sample volume produces higher quench, which will decrease the efficiency and increase MDA. To evaluate this, we graphed MDA (Bq/L) versus urine sample volume (mL) (Fig.1). MDA decreased significantly as sample volume increased from 2 mL to 5 mL, after which minimal reduction in MDA occurred. Therefore, we chose a 5 mL urine sample size as an optimal volume with 15 mL of LSC cocktail, as seen with other work in this research area [14].

**PDD/PSA setting optimization**—We used standard procedures for PDD/PSA setting optimization [15]. Alpha and beta sources in appropriate matrices were analyzed at different PDD/PSA settings using "A/B Test" protocol provided by the instrument software. We used the instrument software to build a graph showing spillover of alpha in beta and beta in alpha versus PDD/PSA setting. Spillover curves can be seen by selecting "standard curve" option in software. The lowest spillover gave the optimal PDD/PSA setting. We used Sr-90/Y-90 as a beta source and Am-241 as an alpha source. We found the PDD/PSA setting for base urine

and deionized water matrices and chose the average setting as optimal for urine bioassay. Figure 2 shows typical alpha/beta spillover curves. For the new Quantulus GCT6220, we chose the double PSA setting. That setting allows the user to ignore alpha/beta events inside the chosen PSA range, where those events are mixed and small, and to count only the events outside this PSA range. All chosen PSA/PDD settings can be saved in the QuantaSmart software library. Table 1 shows the optimal PDD/PSA setting for each instrument.

**Quench (efficiency) curves**—We built quench curves according to the standard procedure [15]. We prepared 18 vials with 20 mL of cocktail each. Two of the 18 vials were left for background measurement. Eight vials were spiked with the same amount of Sr-90/Y-90 and eight other vials were spiked with the same amount of Am-241. Both spiked sets were quenched with different amounts of nitromethane, in the range from 0 to 0.3 mL [16], and analyzed at the optimal PDD/PSA setting on each instrument. The efficiency was calculated as found activity (counts per minute; CPM) divided by added activity (disintegrations per minute; DPM) for the given instrument and nuclide as shown in Eq. (2):

$$E = CPM/DPM$$
(2)

where E is instrument efficiency

The quench curve presents the relationship between efficiency and quenching factor tSIE (transformed spectral index of external standard), which is calculated to an external gamma source (Ba-133) and provided by the instrument software. The Sr-90 quench curves normally are fitted to third order polynomial equations and the Am-241 quench curves are fitted to exponential equations. Figures 3 and 4 show examples of Am-241 and Sr-90 quench curves for Tri-Carb 3110.

**Choice of cocktail**—We chose five commonly used cocktails for optimization: UG, UGLLT, UGXR, UGAB, and OPHS-3. PDD/PSA settings were optimized for each cocktail. The criteria for LSC cocktail choice were gross alpha/beta activities for known urine spikes (QC materials). UGAB and OPHS-3 cocktails gave better correlation between found and target activities (Fig. 5). We chose UGAB cocktail as optimal because it was created specifically for alpha/beta analysis [17].

**Sample analysis time**—Theoretically, the longer the counting time, the lower the MDA. However, time is critical in an emergency response situation. Data should be produced quickly, without compromising quality. Therefore, analysis time must be optimized for the expected application. To choose the optimal count time, we graphed MDA versus count time (Fig. 6). MDA decreases sharply as count time increases, from 1 minute (min) to 5 minutes (min), and then the MDA curve flattens. Based on those results, we set the count time for the urine bioassay method at 5 min.

**External standard analysis time**—To obtain a quench factor (tSIE), the sample is analyzed against a gamma external source (Ba-133) installed in these LSC instruments. The instrument software gives us the option to choose a count time in the range of 1 - 1

600 seconds (sec). To ensure adequate counting statistics, the external standard is typically counted to a 2 sigma counting error of 0.5%. This has worked well for Tri-Carb 3110 and 5110 (about 10–15 sec in most cases). Table 2 shows an example of the external standard count time effect on gross alpha/beta activity in QC samples on Tri-Carb 3110. However, for Quantulus GCT6220, which has the same external source with smaller activity than counters of Tri-Carb series, external standard count time, set at 2 sigma, takes about 5 min, which makes total analysis time 10 min longer. When time is critical, this is not optimal choice. Therefore, we evaluated different count time for external standard (5 sec, 10 sec, 60 sec, 240 sec, and 300 sec) on Quantulus GCT6220 and found that 60 sec can be optimal for this instrument.

**Turnaround analysis time**—Total analysis time consists of sample analysis time plus double external standard analysis time. The external standard analysis time is doubled because the sample will be counted for this time twice: with and without external standard to get the quenching factor (tSIE). For Tri-Carb 3110 and 5110, total analysis time will be about 6 min per sample. For Quantulus GCT6220, total analysis time will be about 7.5 min per sample. Therefore, during an 8-hour shift, 80 samples could be analyzed with each Tri-Carb counter and 64 samples could be analyzed with each Quantulus GCT6220.

**Region of interest**—To decrease the spillover between alpha and beta signals, we chose the region of interest (region in which the given nuclide will be counted) for each nuclide and each instrument based on alpha/beta spectra received at the optimal PSA/PDD settings. Table 1 shows the resultant parameters for all instruments.

The next step was to validate the method through accuracy, precision, range, linearity, nuclide stability, and LOD.

#### Method validation

**Accuracy and precision**—To demonstrate accuracy, we analyzed two different urine samples spiked with Am-241 or Th-230 (alpha source) and Sr-90/Y-90 (beta source) at three different levels on two days using two instruments with one replicate per instrument. The average between all measurements was the recovery for alpha and beta nuclides. Tables 3 and 4 present these results. Total recovery for alpha and beta nuclides was close to 100%.

To demonstrate precision, we characterized urine gross alpha/beta QC, reference material, and a high calibration range sample and compared the results with their target numbers. Table 5 shows the results found statistically (using SAS) during more than 50 runs on different days. The bias between found and target results is about  $\pm$  5% for gross alpha nuclides and  $\pm$  2% for gross beta nuclides, which confirms the reasonable calibration for all these LSC instruments (PSA/PDD optimization and quench curves).

**Range and linearity**—We showed the linearity for gross alpha in the range of LOD to 15 000 Bq/L using Am-241 and for gross beta of LOD to 1,000,000 Bq/L using Sr-90/Y-90 (Figs 7 and 8), with a regression coefficient  $R^2$  around 1. We also showed that 100-fold urine dilution will not affect the linearity and can be used if the sample with higher than this range activity is analyzed.

**Stability in urine matrix**—We proved the stability of Am-241 and Sr-90/Y-90 nuclides in urine matrix using urine gross alpha/beta QC samples during three freeze-thaw cycles, during at least 6 years at temperatures  $-70^{\circ}$ C, during 24 hours at room temperature, and during 24 hours in mixture with cocktail in the instrument at a temperature about 8°C. Tables 6 shows the results for alpha and beta nuclides using two instruments with one replicate per instrument, that confirm stability under chosen conditions.

We observed an increase in alpha signal with time (about 75% for 30 days) when the urine sample and LSC cocktail mixture was kept in plastic vials, whereas the beta signal decreased slightly (about 1–2% for 30 days). However, we did not observe such effect for the alpha signal in glass vials (Figs. 9 and 10). Therefore, glass vials would give better results for samples that need to be recounted in 24 hours or more.

**Limit of detection**—We determined method LODs for gross alpha and beta nuclides using Am-241 (alpha) and Sr-90/Y-90 (beta) emitters. Four LOD standards, close to the LOD, found as standard deviation multiplied three times (3SD) after 20 blank urine analyses, were produced. These LOD standards were analyzed at least 20 times on each instrument. LODs were found according to the Eq. (3) [18]:

Activity<sub>LOD</sub> = [Mean 
$$b$$
 + 1.645(SD  $b$  +  $I$ )]/[1 - 1.645S] (3)

where Mean *b*—blank average and SD *b*—standard deviation for blank, *I*—intercept and *S*— slope of the equation of standard deviation versus LOD standards activity (see Fig.11). Table 7 shows the LOD results. Quantulus GCT6220 gave slightly lower LOD for alpha and beta nuclides because it can reduce background interference. Tri-Carb 3110 and 5110 showed similar LOD results and were combined.

**Blank urine characterization**—Urine collected from 50 persons not exposed to radionuclides was screened for gross alpha/beta nuclides to find the average results on each instrument. Table 8 shows the results. Small levels of alpha (less than LOD) were observed in the blank urine, but beta nuclides were present in the range of 10–80 Bq/L, mostly from dietary potassium (K-40). This is in an agreement with other research data [11].

# Conclusion

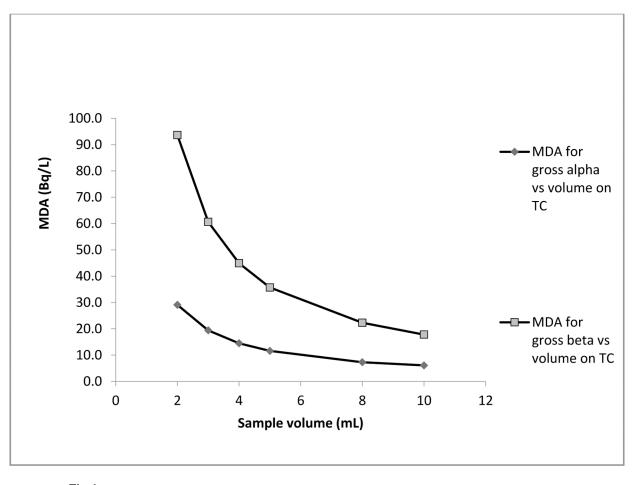
We developed and validated a urine gross alpha/beta screening method using Tri-Carb 3110, 5110, and Quantulus GCT6220. This method uses a 5 mL of urine sample with 15 mL of UGAB cocktail. With a sample analysis time of 5 min, we can determine gross alpha at LOD = 12.6 Bq/L and gross beta at LOD = 44.6 Bq/L. Having two Tri-Carbs 3110, one Tri-Carb 5110, and Quantulus GCT6220, we can analyze approximately 760 samples within 20 hours.

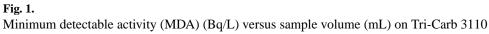
# References

 Centers for Disease Control and Prevention (2015) Radiation from the Earth (terrestrial radiation) Radiation and Your Health. https://www.cdc.gov/nceh/radiation/terrestrial.html. Accessed 28 May 2020.

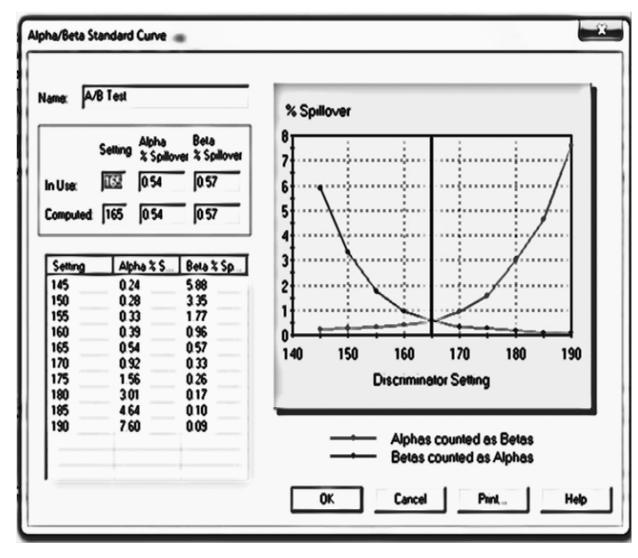
- 3. Schonhofer F (1998) The use of low-level liquid scintillation spectrometry for rapid measurement and decision making. Radioact Radiochem 9(3):18–24
- Kaihola L (2000) Radionuclides identification in liquid scintillation alpha-spectroscopy. J Radioanal Nucl Chem 243 (2):313–317
- Salonen L, Hukkanen H (1997) Advantages of low-background liquid scintillation alphaspectrometry and pulse shape analysis in measuring <sup>222</sup>Rn, uranium, and <sup>226</sup>Ra in groundwater samples. J Radioanal Nucl Chem 226:67–74
- 6. L'Annunziata M (ed) (2004) Handbook of Radioactivity Analysis, 2nd edn. Academic Press, New York
- 7. Shafer I, Herbert D, Zeiske U (2000) On low-level tritium measurements with LSC Quanulus. Appl Radiat Isot 53: 309–315 [PubMed: 10879878]
- Mingote RM, Barbeira PJS, Rocha Z (2006) Methodology for rapid tritium determination in urine. J Radioanal Nucl Chem 269(2):475–479
- 9. Puhakainen M, Heikkinen T (2008) Tritium in the urine in Finnish people. Radiat Prot Dosim 128(2):254–257
- Sas D, Janda J (2017) Rapid determination of gross alpha/beta activity in urine with LSC. J Radioanal Nucl Chem 311(1): 23–27
- Shoji M, Kondo T, Nakajima Tet al. (2007) Investigation of monitoring for internal exposure by urine bioassay in a biomedical research facility. Radiat Prot Dosim, pp 1–5, published by Oxford University Press
- 12. Currie LA (1968) Limits for quantitative detection and qualitative determination: application to radiochemistry, Anal Chem 40(3):586–593
- Erchinger JL, Aalseth CE, Bernacki BE, et al. (2015) Development of a low background liquid scintillation counter for a shallow underground laboratory. Appl Radiat Isot 105:209–218 [PubMed: 26334781]
- Preston R, Reese RP, Shanks ST (2007) Optimization of screening for radioactivity in urine by liquid scintillation. 53rd Radiobioassay and Radiochemical Measurements Conference, Jackson Hole, Wyoming, USA
- 15. Piraner O and Jones R (2009) Urine gross alpha/beta analysis by Liquid Scintillation Counting for emergency and terrorism preparedness and response in Eikenberg J, Jäggi M, Beer H, Baehrle H, eds, LSC 2008, Advances in Liquid Scintillation Spectrometry, University of Arizona
- Piraner O and Jones R (2020) The Effect of Quench Agent on Urine Bioassay for Various Radionuclides Using Quantulus 1220 and Tri-Carb3110. J Radioanal Nucl Chem 326(1): 657–663
- 17. [June 18, 2020] PerkinElmer Scintillation Cocktails and Consumables. www.perkinelmer.com/ content/relatedmaterials/CAT\_LSCCocktailsVials.pdf Accessed
- Clinical and Laboratory Standards Institute (2012) EP17-A2, Protocols for determination of limit of detection and limit of quantification, 2nd edn, V24, N34. https://community.clsi.org/media/ 1430/ep17a2\_sample.pdf. Accessed 26 May 2020.

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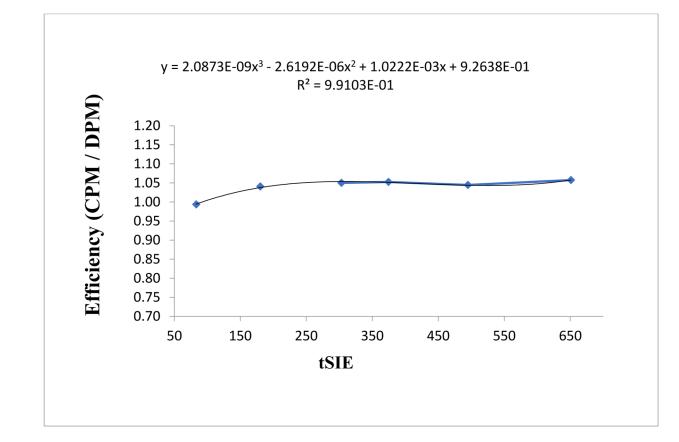


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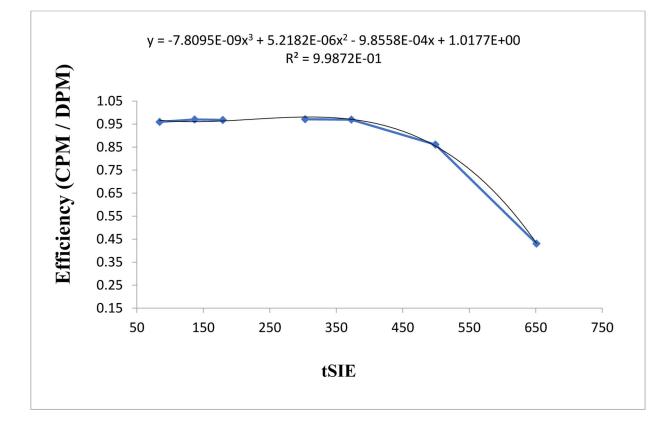
#### Fig. 2.

Example of alpha (Am-241) and beta (Sr-90/Y-90) discrimination in DI water on Tri-Carb 5110



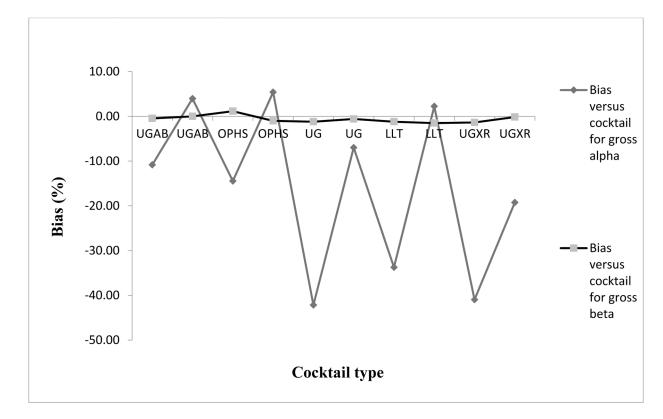
#### Fig. 3.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using nitromethane (0–0.3 mL) as a quench agent in the UGAB cocktail (20 mL) for Am-241 on Tri-Carb3110. Solid black line is the trendline



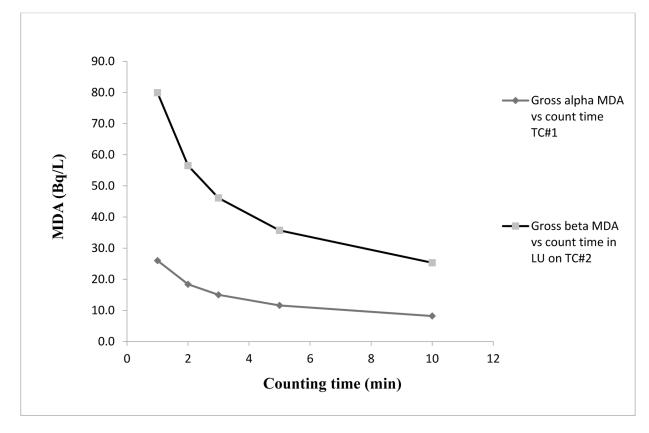
# Fig. 4.

Quench indicating parameter measurements in term of tSIE effect on Efficiency using nitromethane (0–0.3 mL) as a quench agent in the UGAB cocktail (20 mL) for Sr-90/Y-90 on Tri-Carb3110. Solid black line is the trendline



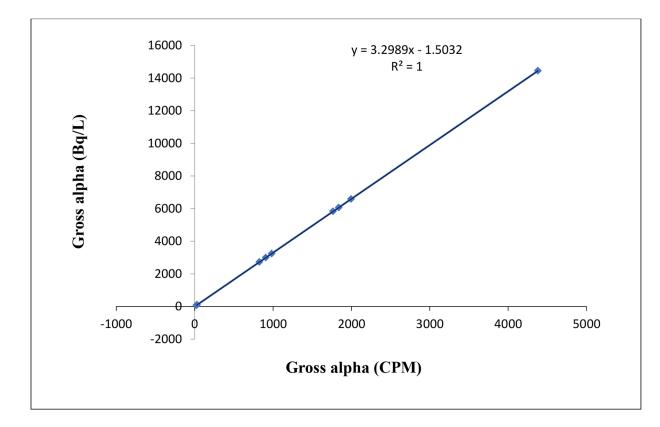
# Fig. 5.

The effect of liquid scintillation counting cocktail type on alpha/beta bias between found and target results using urine gross alpha/beta quality control (QC) materials



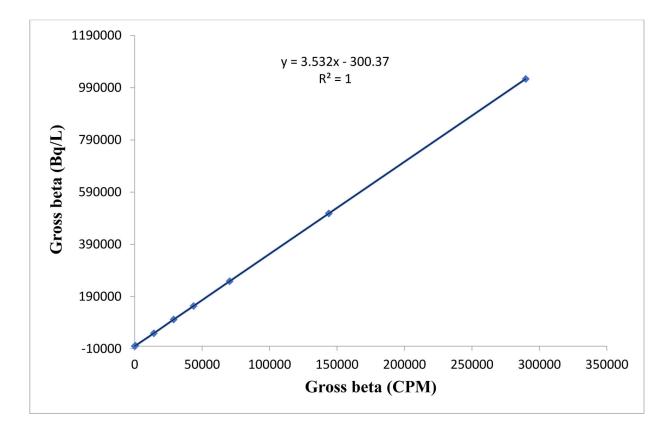
### Fig. 6.

Minimum detectable activity (MDA) (Bq/L) for gross alpha (Am-241) and beta (Sr-90/Y-90) versus counting time on Tri-Carb 3110



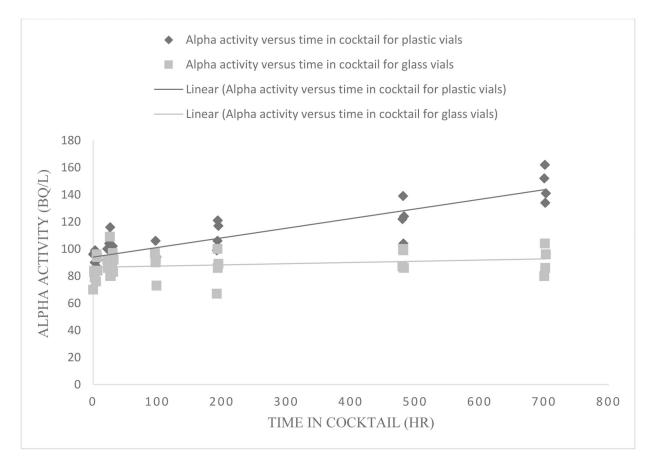
# Fig. 7.

Gross alpha activity in Bq/L versus counts per minute (CPM) signal on Tri-Carb 3110 for the range of LOD to 15 000 Bq/L



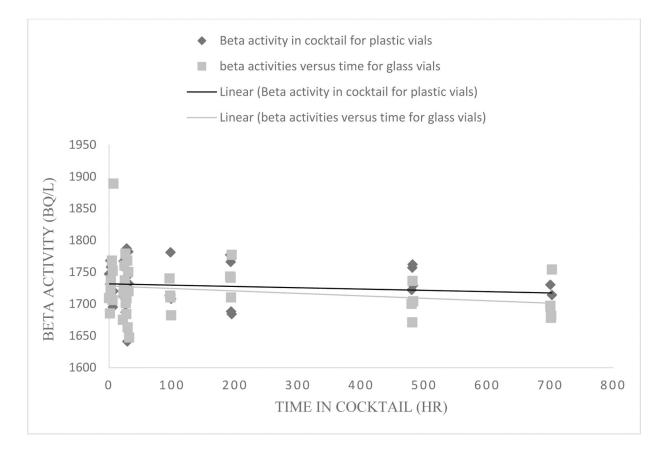
# Fig. 8.

Gross beta activity in Bq/L versus counts per minute (CPM) signal on Tri-Carb 3110 for the range of LOD to 1 000 000 Bq/L



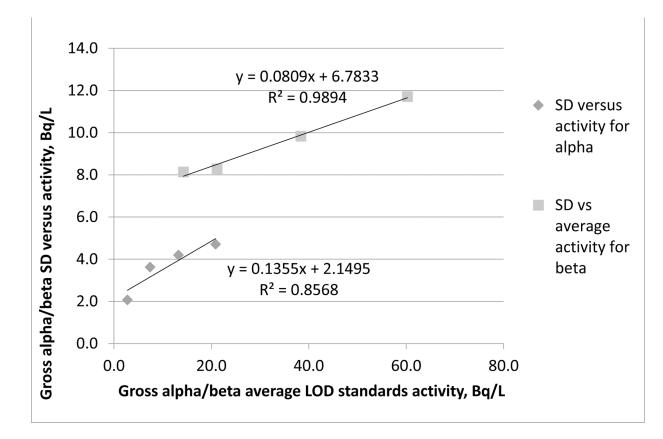
#### Fig. 9.

Stability of alpha nuclide in UGAB cocktail (Low QC) at 7–8°C in plastic and glass vials for 30 days



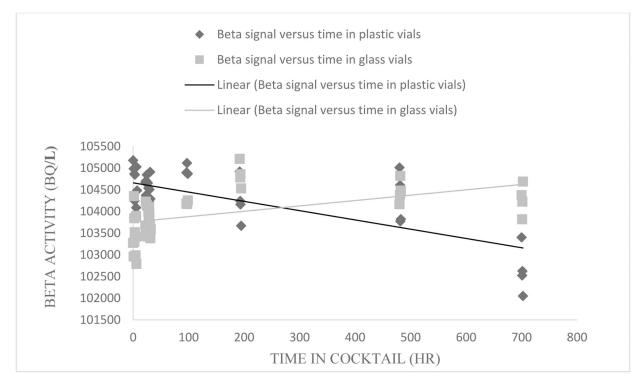
# Fig. 10.

Stability of beta nuclide in UGAB cocktail (Low QC) at 7–8  $^{\circ}\mathrm{C}$  in plastic and glass vials for 30 days



#### Fig.11.

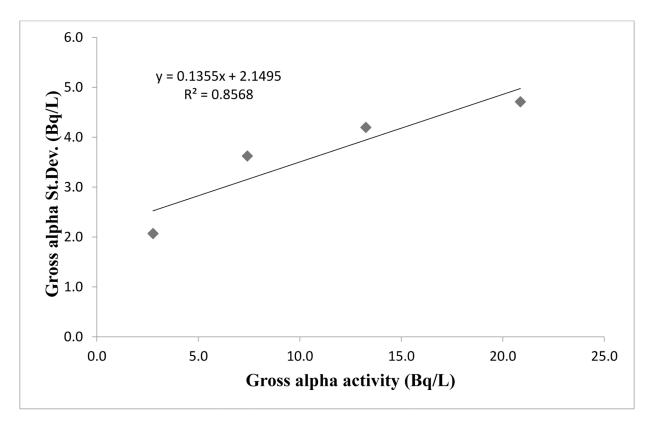
Standard deviation (SD) versus alpha/beta activity in LOD standards (60 analyses) for LSC instruments Tri-Carb series



# Fig. 12.

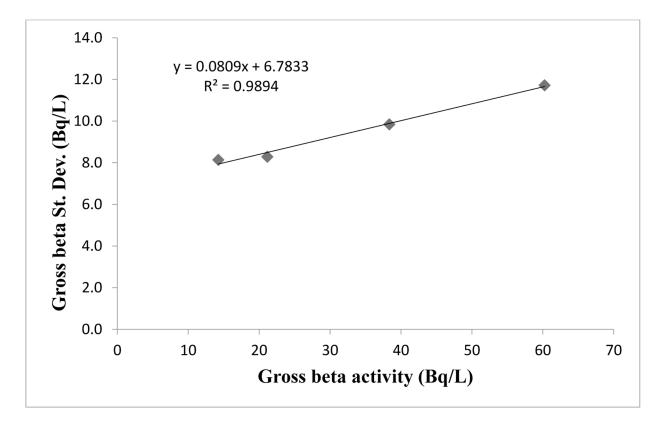
Stability of beta nuclide in UGAB cocktail (High QC) at 7–8°C in plastic and glass vials for 30 days

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# Fig. 13.

Standard deviation versus alpha activity in LOD standards (60 analyses) for LSC Instruments Tri-Carb series



# Fig. 14.

Standard deviation versus beta activity in LOD standards (60 analyses) for LSC instruments Tri-Carb series

# Liquid scintillation counting method parameters for different instruments

Parameter	Tri-Carb 3110 #1	Tri-Carb 3110 #2	Tri-Carb 5110	Quantulus GCT6220
PSA/PDD setting	125	165	135	120–170
Sample volume (mL)	5	5	5	5
Cocktail volume (mL)	15	15	15	15
Sample analysis time (min)	5	5	5	5
External std analysis time	2 Ω (15 sec)	2 Ω (15 sec)	2 Ω (10 sec)	60 sec
Alpha region of interest (keV)	0–300	0–200	0-1000	0–450
High energy beta region of interest (keV)	0–2000	0–2000	0-2000	0–2000
Low energy beta region of interest (keV)	0–18.6	0–18.6	0–18.6	0–18.6

Effect of external standard analysis time (Ext S T) on bias between found and target activity for urine gross alpha and beta in urine QC samples (example on Tri-Carb 3110 #1)

Quality control	Ext S T	Gross A	Bias	Gross B	Bias
			%		%
Low QC	2 Sigma	Am-241	-1.22	Sr-90/Y-90	0.06
High QC	2 Sigma	Am-241	-3.88	Sr-90/Y-90	-3.57
Low QC	5 sec	Am-241	14.63	Sr-90/Y-90	-4.57
High QC	5 sec	Am-241	-2.92	Sr-90/Y-90	-3.40
Low QC	10 sec	Am-241	21.95	Sr-90/Y-90	-1.56
High QC	10 sec	Am-241	-3.30	Sr-90/Y-90	-4.21
Low QC	60 sec	Am-241	9.76	Sr-90/Y-90	1.27
High QC	60 sec	Am-241	-2.87	Sr-90/Y-90	-3.58
Low QC	240 sec	Am-241	25.61	Sr-90/Y-90	-3.12
High QC	240 sec	Am-241	-5.67	Sr-90/Y-90	-3.67

Accuracy for gross alpha (Am-241) through spike recovery. All activities are in Bq/L

			Samp	le 1			
			Measured	activity			
	Instrument ID	Spike Target Activity (SD)	Day 1	Day 2	Mean	Recovery (%)	
Urine	TC3110 #1	0	2.6	-2	-0.2		
	TC3110 #2	0	0.7	-2	-0.2		
PT2	TC3110 #1	82 (0.0)	83.2	86.4	83.8	101	
	TC3110 #2	83 (9.0)	83	82.6	83.8	101	
PT6	TC3110 #1	2,000 (127)	3 100	2 870	2 990	99.7	
	TC3110 #2	3 000 (127)	2 970	3 010	2 990	99.7	
PT9	TC3110 #1	( 200 (220)	6 090	6 190	6.000	08.0	
	TC3110 #2	6 300 (230)	6 030	6 600	6 2 3 0	98.9	
			Measured	activity			
BU	TC3110 #1	0	3.3	0.7	0.5		
	TC3110 #2	0	-0.7	-1.4	0.5		
Low QC	TC3110 #1	917(97)	76.9	86	80.8		
	TC3110 #2	81.7 (8.7)	69.2	90.9	80.8	98.9	
High QC	TC3110 #1	5 200 (202)	5 140	5 120	5 220	08.0	
	TC3110 #2	5 290 (303)	5 230	5 410	5 230	98.9	
HCR	TC3110 #1	14,700 (240)	14 500	14 600	11000	101	
	TC3110 #2	14 700 (340)	14 800	15 300	14 800	101	
Overall rec	covery, %					99.7	
Standard d	eviation					1.05	

Accuracy for gross beta (Sr-90/Y-90) through spike recovery. All activities are in Bq/L

			Samj	ple 1					
		Measured activity							
Sample ID	Instrument ID	Spike Target Activity (SD)	Day 1 Day 2		Mean	Recovery (%)			
Urine	TC3110 #1	0	47	50	49.8				
	TC3110 #2	0	50	52	49.8				
PT2	TC3110 #1	1.740.(40)	1 750	1 790	1 780 102	102			
	TC3110 #2	1 740 (48)	1 780	1 790		102			
PT6	TC3110 #1	50,400 (450)	49 900	50 600	50.000	00.6			
	TC3110 #2	50 400 (452) 50 000 50 400		50 200	99.6				
PT9	TC3110 #1		104 000	106 000	104.000	00.0			
	TC3110 #2	105 000 (1 500)	103 000	105 000	104 000	99.0			
			Me						
BU	TC3110 #1	0	45	35	25.5				
	TC3110 #2	0	21	41	35.5				
Low QC	TC3110 #1	1 750 (42 2)	1 770	1 700		08.0			
	TC3110 #2	1 750 (43.3)	1 710	1 740	1 730	98.9			
High QC	TC3110 #1	105,000 (1,650)	105 000	104 000		00.0			
	TC3110 #2	105 000 (1 660)	105 000	104 000	104 000	99.0			
HCR	TC3110 #1		151 000	150 000		98.0			
	TC3110 #2	152 000 (2 200)	150 000	148 000	149 000				
Overall recov	/ery, %					99.5			
Standard dev	iation					1.47			

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#### Table 5

Urine gross alpha/beta low and high quality control (QC) materials, high calibration range (HCR), and reference material (RM) characterization during more than 50 runs on different days using all instruments

Sample ID	Nuclide	Mean, Bq/L	SD, Bq/L	Target, Bq/L	Bias, %
BU	Alpha	-1	3	0	
	Beta	47	12	50	
Low QC	Alpha	81.7	8.65	80	2.1
	Beta	1 750	43.3	1 740	0.6
High QC	Alpha	5 290	303	5 350	-1.1
	Beta	105 000	1 660	106 000	-0.9
HCR	Alpha	14 700	340	15 000	-2.0
	Beta	152 000	2 200	150 000	1.3
RM	Alpha	4 190	189	4 000	4.8
	Beta	49 500	1 150	50 000	-1.0

Alpha (Am-241)/Beta (Sr-90/Y-90) urine stability in QC samples. All activities are in Bq/L

Alpha stab	bility				
Quality material	1 (Low QC)				
Instrument ID	Initial measurement	Three freeze- thaw cycles	Bench-top stability	Processed sample stability	Long-term stability
TC3110 #1	82.7	80	75	86	76.6
TC3110 #2	77.2	83.4	80	80	80.3
Mean	80.0	81.7	77.5	83.0	78.5
difference from initial (%)	—	2.13	-3.13	3.75	-1.88
Quality material	2 (High QC)				
TC3110 #1	5 670	5 370	5 210	5 750	5 480
TC3110 #2	5 410	5 590	5 340	5 790	5 330
Mean	5 540	5 480	5 280	5 770	5 410
difference from initial (%)	—	-1.08	-4.69	4.15	-2.35
Beta stabi	ility				
Quality material	1 (Low QC)				
Instrument ID	Initial measurement	Three freeze- thaw cycles	Bench-top stability	Processed sample stability	Long-term stability
TC3110 #1	1 760	1 660	1 730	1 730	1 760
TC3110 #2	1 720	1 780	1 800	1 780	1 710
Mean	1 740	1 720	1 770	1 750	1 740
difference from initial (%)	_	-1.15	1.72	0.57	0.0
Quality material	2 (High QC)				
TC3110 #1	103 000	102 000	106 000	105 000	104 000
TC3110 #2	105 000	107 000	106 000	105 000	107 000
Mean	104 000	104 500	106 000	105 000	105 500
difference from initial (%)	—	0.48	1.92	0.96	1.44

Limits of detection (LOD) estimates for Tri-Carb instrument series and Quantulus GCT6220 in plastic vials with linear fit

Instrument/vial	Gross alpha LOD, Bq/L	p-value	Gross beta LOD, Bq/L	p-value
Tri-Carb series (60 runs)	12.6	0.025	44.6	0.275
Quantulus GCT6220 (20 runs)	7.58	0.002	40.3	0.021

Blank urine characterization for gross alpha/beta nuclides (average using 50 individual urine samples and four liquid scintillation counting instruments), by instrument and overall

	TC3110 #1		TC3110 #2 TC		TC5	TC5110		GCT6220		Overall	
	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta	Alpha	Beta	
Average (Bq/L)	2.49	38.5	1.56	33.5	1.39	41.2	0.71	30.4	1.54	35.9	
SD (Bq/L)	1.99	19.4	1.75	19.7	2.93	20.1	1.73	19.6	2.10	19.7	