HHS Public Access

Author manuscript

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

Published in final edited form as:

J Radioanal Nucl Chem. 2021 January 28; 327: 975–983. doi:10.1007/s10967-020-07557-z.

Universal use of alpha/beta mode in liquid scintillation counting analysis for both alpha/beta and single nuclide determination

Olga Piraner[*], **Robert L. Jones**

Centers for Disease Control and Prevention, National Center for Environmental Health, Division of Laboratory Sciences, Inorganic and Radiation Analytical Toxicology Branch, 4770 Buford Hwy, MS S110-5, Atlanta, GA 30341-3717

Abstract

Nuclear industry advancements and growing concerns about environmental contamination and terrorist activity have increased interest in quantifying radioisotopes in environmental and human samples. Increased presence in the environment, ease of entry into the food chain, nuclear medicine applications, and the possibility of radiological terrorism incidents can lead to human intake of these radionuclides [1,2]. A universal method to screen for and quantify individual radionuclides as well as both levels of alpha and beta emitters would address these concerns.

Keywords

Gross alpha/beta; Liquid scintillation counting; Quantulus1220

Introduction

Liquid Scintillation Counting (LSC) continues to be a very popular technique for detecting and quantifying radioactivity [3,4]. Modern liquid scintillation counters are equipped with pulse shape analysis (PSA) or pulse decay discriminator (PDD) and multichannel analyzers (MCAs). A PSA/PDD allows separation of alpha scintillation events from beta based on their peak shapes, and an MCA allows placement of radiation signals in different channels to create different spectra for alpha and beta emitters. Figure 1 provides examples of urine gross alpha (Am-241)/beta (Sr-90/Y-90) low and high QC materials spectra while Figures 2 and 3 provides examples of Cs-137, Co-60, and H-3 spectra in single urine spikes.

An appropriate PSA/PDD setting depends on sample quenching whereas sample quenching depends on sample matrix, LSC vials, and choice of cocktail [5]. Therefore, the optimal PSA/PDD setting should be determined for each matrix, vial type, and cocktail.

[*]Author to whom correspondence should be addressed: obp2@cdc.gov; Fax: +1 770 488 0509; Tel: +1 770 488 7301.

The authors declare that they have no competing financial interest.

Samples may contain a single radionuclide, therefore producing only beta or alpha emission. When using the highest PSA/PDD setting, all emission events are treated as beta while using the lowest PSA/PDD setting causes all emissions events to be treated as alpha. This approach leads to universal use of alpha/beta mode for alpha/beta and for single radionuclide analyses. With the correct PSA/PDD setting and quench curve, any individual radionuclide, such as ^{241}Am, ^{90}Sr, ^{60}Co, ^{137}Cs or ^{3}H, all of which have different LSC efficiencies, can be quantified using alpha/beta mode. In this work we studied the effect of PSA setting in alpha/beta mode on the analysis of double and single nuclide spikes.

Experimental

Reagents and materials

For gross alpha/beta analysis, we used Ultima Gold AB cocktail (UGAB) from PerkinElmer Company and 99% nitromethane from ACROS Organics as a quench agent. Deionized water (DI water) was used for all solutions (≥18 MΩ·cm, from an Aqua Solutions Ultrapure Water System, Aqua Solutions, Inc.). “Base urine” was collected through anonymous human donations according to the Centers for Disease Control and Prevention Institutional Review Board (IRB) protocol 3994 and acidified to 1% HNO_3. All radioactive source solutions were traceable to the National Institute for Standards and Technology (NIST) (Gaithersburg, MD, USA). Urine gross alpha/beta quality control (QC) LU-077203 and HU-077201 were purchased from Eckert & Ziegler Analytics, Inc. They are composed of base urine spiked with both Am-241 and Sr-90/Y-90 at low and high levels. BU-077401 is non-spiked base urine. Am-241, Sr-90/Y-90, Cs137, Co-60, and H-3 reference solutions were purchased from Eckert & Ziegler Analytics, Inc. We used them for PSA/PDD optimization and quench curves preparation. We spiked some individual urine samples with Am-241, Sr-90/Y-90, or mixtures of both these radionuclides at different activity levels. Fruit drink samples, spiked with Am-241, were received from the Department of Health, New York (DOH NY), Albany, NY as a part of Federal Drug Administration (FDA) “Menu 2010” exercise.

Instrumentation and labware

For this study, we used three ultralow level liquid scintillation spectrometers Quantulus1220 (Q#1,2,3) equipped with PSA and MCA (PerkinElmer); 20-mL LSC plastic vials (PerkinElmer) for LSC analysis; a high-precision analytical balance capable of accurately weighing 0.0001 gm (Mettler-Toledo, LLC) for quench curves and urine spikes preparation; 15-mL and 50-mL conical polypropylene tubes (Beckton Dickinson) for solutions preparation; Brinkman bottle top dispenser with capacity from 5 mL to 25 mL (Brinkman Instruments, Inc.) for cocktail dispensing; and four electronic pipettes with a total volume range from 5 μL to 5 mL (Eppendorf, Inc).

PSA optimization for gross alpha/beta mode

PSA optimization was performed using Am-241 as the alpha source and Sr-90/Y-90 as the beta source. Three urine samples (one spiked with the alpha source, one with the beta and one background) were analyzed at various PSA settings. We calculated spillover, or crosstalk, from alpha window to beta window and beta to alpha at each PSA setting and

plotted this data to yield a graph of spillover versus PSA setting (Fig. 4). The PSA at the lowest spillover was chosen as optimal.

The optimal PSA settings were established when Co-60, Cs-137 or H-3 was used as a beta source with Am-241 as an alpha source in base urine (Figs 5–7). We found the optimal PSA setting using the fruit drink as well (Fig. 8).

We used PSA setting 256 for pure beta emitting radionuclide sample analysis, and all events at this setting are counted as betas. We used PSA setting 1 for pure alpha emitting radionuclide sample analysis, and all events are counted as alphas.

Efficiency (quench) curves preparation

We built Am-241 and Sr-90/Y-90 quench curves at the optimal PSAs for urine and fruit drink as well as at PSA 1 for Am-241 and at PSA256 for Sr-90/Y90 as described in the 2009 publication by Piraner and Jones [6]. We used nitromethane, a commonly used quench agent [7,8], with Ultima Gold AB cocktail. Table 1 shows examples of Am-241 and Sr-90/Y-90 quench curves on Quantulus1220 #2 at different PSA settings. The quench curves on instruments Q#1 and Q#3 were similar to those on instrument Q#2.

Other LSC parameters and sample preparation

LSC parameters such as type of cocktail, sample/cocktail volume, sample analysis time, and external standard analysis time for each instrument were optimized according to the approach used previously for our gross alpha/beta urine analysis method [6]. We chose region of interest (ROI), region in which each nuclide will be counted, for alpha (Am-241) and beta (Sr-90/Y-90) sources on the base of their spectra to diminish the crosstalk between alpha and beta signals. Table 2 shows resultant parameters. Finally, we mixed 5 mL of urine samples with 15 mL of Ultima Gold AB cocktail until a uniform state was reached and placed prepared samples on counter tray for LSC analysis.

Results and discussion

The effect of a beta type nuclide on PSA setting

For all experiments, we used LSC instruments in alpha/beta mode. PSA optimization using different beta nuclides shows that the nature of the beta source does not affect the PSA setting. Similar results were obtained when Sr-90, Cs-137, Co-60, and H-3 were used as a beta source with Am-241 as an alpha source (Figs 4 –7). Sample matrix has the greatest effect on PSA setting. We found that the optimal PSA setting for one matrix will not be optimal for a different matrix, and alpha/beta crosstalk for more quenched samples is higher (Figs 4 and 8).

The effect of PSA settings on urine spikes analysis results

To show the effect of the PSA settings on LSC results, we used urine spiked either with Am-241 or Sr-90/Y-90 or with both nuclides. The activity correlation between found and target data for known urine spikes was evaluated. We characterized urine gross alpha/beta QC materials for gross alpha/beta activities at the optimal PSA setting for each instrument.

Table 3 shows the observed results. Bias between found and target activities is in the ± 2.5% range for both alpha and beta emitters, which validates the calibration for these instruments (PSA optimization and quench curves).

Using a PSA setting optimized for urine, we analyzed individual urine samples spiked with Am-241, Sr-90/Y-90, or mixtures of these nuclides at different levels. Table 4 shows the results from Quantulus #2. Other instruments (Q#1 and Q#3) produced very similar results. The data shows that the correlation between found and target results is in the range of ± 6% for double nuclide spikes and in the range of ± 5% for single nuclide spikes. The bias can be attributed to the effect of alpha/beta crosstalk. Urine, spiked with higher alpha and lower beta nuclides activities, gives positive bias for beta and negative bias for alpha activity and vice versa. Also, the individual urine samples are quenched differently and the optimal PSA setting chosen for base urine may not be optimal for the individual urine spikes. This indicates that the activity amount and different samples' quenching affects the crosstalk and causes biased results.

The next step was to analyze the same single nuclide urine spikes at PSA=256 (for gross beta only) and PSA=1 (for gross alpha only) and reprocess the results with either alpha or beta quench curves generated at the same PSA settings. Table 5 shows the results. The bias between found and target results for either alpha or beta activity in urine samples is in the range of ± 6%, which is comparable with the results for the same samples received at the optimal PSA setting for urine (Table 4).

As indicated, samples containing any individual alpha or beta emitting radionuclide do not require PSA optimization. A PSA of 256 should be used for a single beta radionuclide, a PSA of 1 should be used for a single alpha radionuclide, along with the radionuclide's corresponding quench curve.

Analysis of Am-241 fruit drink spikes (from FDA "Menu-2010" exercise) at different PSA setting

If the matrix is not urine, matrix-specific PSA optimization is necessary for a radionuclide mixture to decrease crosstalk between alpha and beta radionuclide emissions. Figure 8 shows that the optimal PSA setting for fruit drink is around 40. If the analyzed samples contain a single radionuclide, a PSA setting of either 1 or 256 can be used for analysis. Table 6 shows results for fruit drink spiked with Am-241, from the FDA "Menu 2010" exercise, analyzed at various PSA settings. As a result, the use of a non-optimal PSA setting for fruit drink spikes showed a bias of more than 50% because of alpha/beta crosstalk. When we get closer to optimal PSA for fruit drink, the bias for Am-241 decreases around PSA 40 and reaches a minimum, which is comparable with PSA 1. Therefore, for this analysis, a PSA setting of either 40 or 1 should be used for the best analytical results. The results presented are from Quantulus1220 #2.

Conclusion

An alpha/beta mode can be used for both alpha/beta and individual radionuclide analysis. PSA optimization is required for alpha/beta analysis to decrease the crosstalk between

alpha and beta radionuclide emission signals. The nature of beta nuclide does not affect PSA setting as much as sample matrix. The use of non-optimal PSA settings for LSC analysis results in higher alpha/beta crosstalk and, as a result, bias between found and target activities. The more quenched samples have a lower PSA setting and higher alpha/beta crosstalk.

Individual radionuclide analysis can be performed at the optimal PSA setting for a given matrix as well. However, PSA optimization is not necessary for such analyses because the activity of a single alpha emitting radionuclide can be determined at a pulse shape analysis value that classifies all events as alphas (for Quantulus1220, PSA=1), and that of a single beta emitting radionuclide can be determined at a pulse shape discrimination value that classifies all events as betas (for Quantulus1220, PSA=256). Alpha and beta quench curves (efficiency curves), used for activity calculations, should be generated at the PSA settings that will be used for analysis, and (if appropriate) with the radionuclide of interest.

Acknowledgements

The authors thank Dr. David Saunders for his past assistance with this manuscript preparation.

References

1. Radionuclides/Radiation Protection/US EPAhttps://www.epa.gov/radiation/radionuclides (accessed on 6/5/2020)
2. “Radiation from the Earth (Terrestrial Radiation)”, Radiation and Your Health, Centers for Disease Control and Prevention (2015) 12, 7 https://www.cdc.gov/nceh/radiation/terrestrial.html (accessed on 6/5/2020)
3. Shonhofer F (1998) The use of low-level liquid scintillation spectrometry for rapid measurement and decision making. Radioact Radiochem 9(3):18–24
4. Kaihola L (2000) Radionuclides identification in liquid scintillation alpha-spectroscopy. J Radioanal Nucl Chem 243 (2):313–317
5. Pujol L, Sanches-Cabeza JA (1997) Role of quenching on alpha/beta separation in liquid scintillation counting for several high capacity cocktails. Analyst 122:383–385
6. Piraner O, 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, the University of Arizona
7. Pates JM, Cook GT, MacKenzie AB, Passo CJ (1998) Implication of beta energy and quench level for alpha/beta liquid scintillation spectrometry calibration. Analyst 123: 2201–2207
8. L’Annunziata M (ed) (2004) Handbook of Radioactivity Analysis, 2nd edn, Academic Press, New York

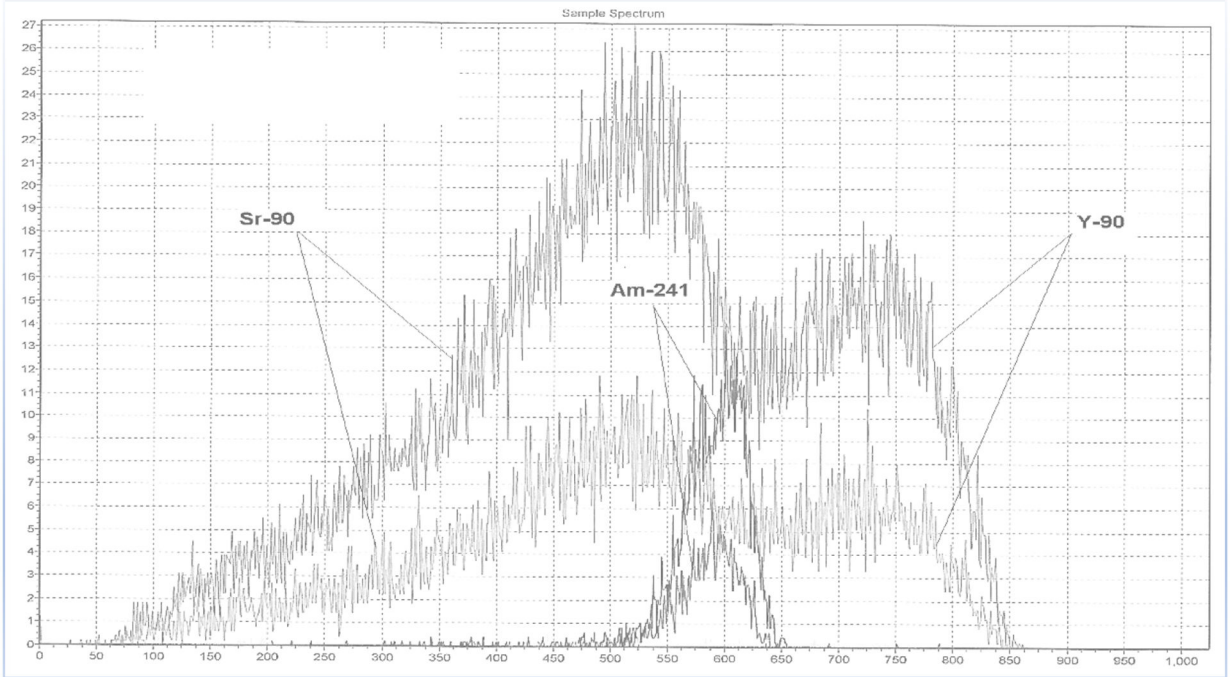

Fig. 1.

Urine gross alpha (Am-241)/beta (Sr-90/Y-90) low and high QC spectra received from Quantulus1220

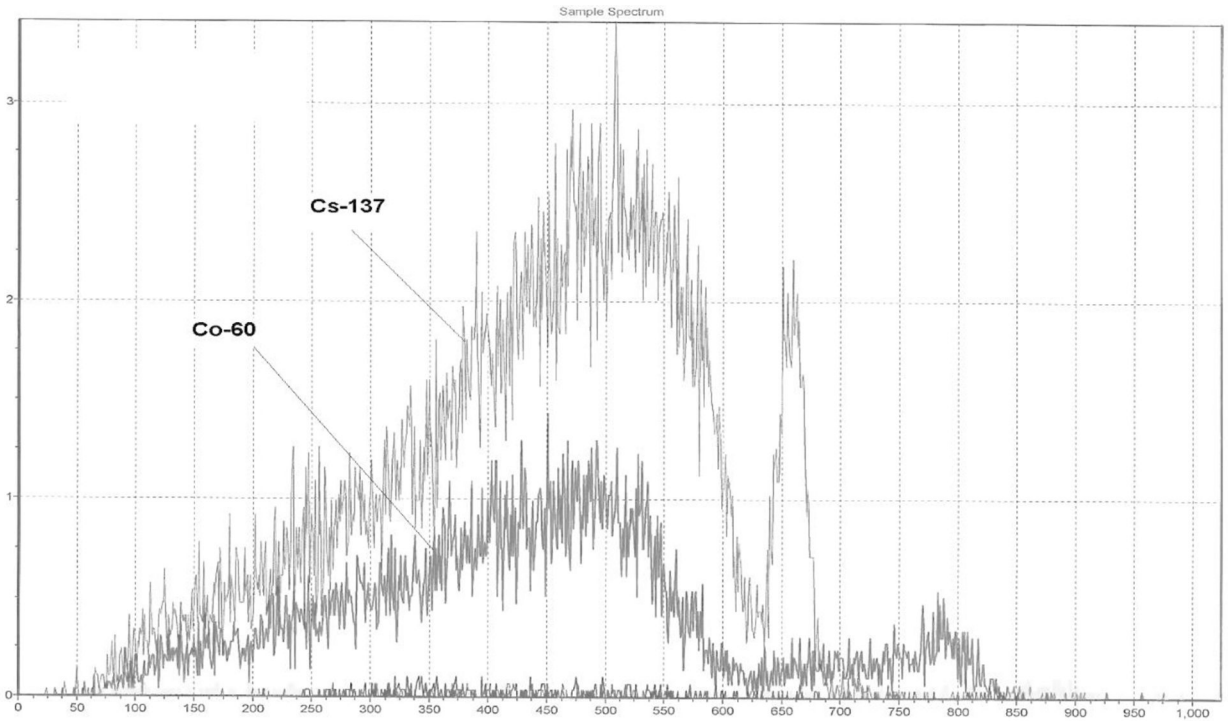

Fig. 2.
Urine Cs-137 and Co-60 single spike spectra from Quantulus1220

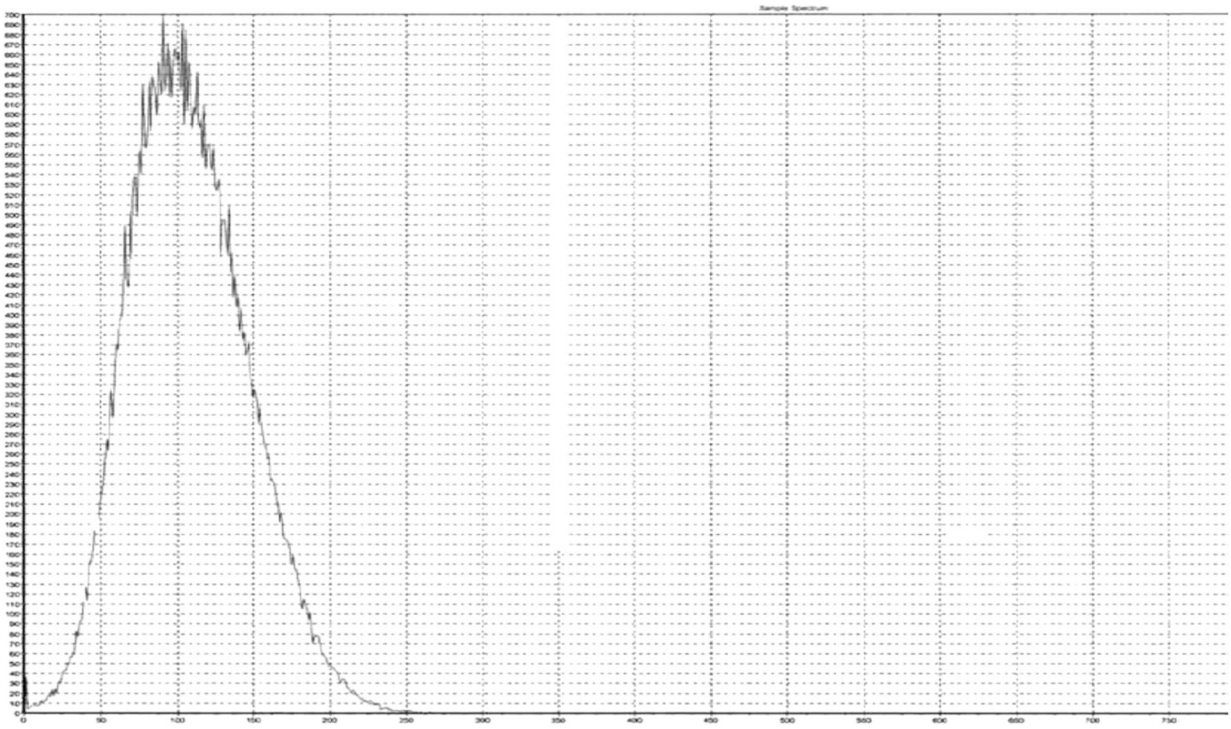

Fig. 3.
H-3 urine spike spectrum from Quantulus1220

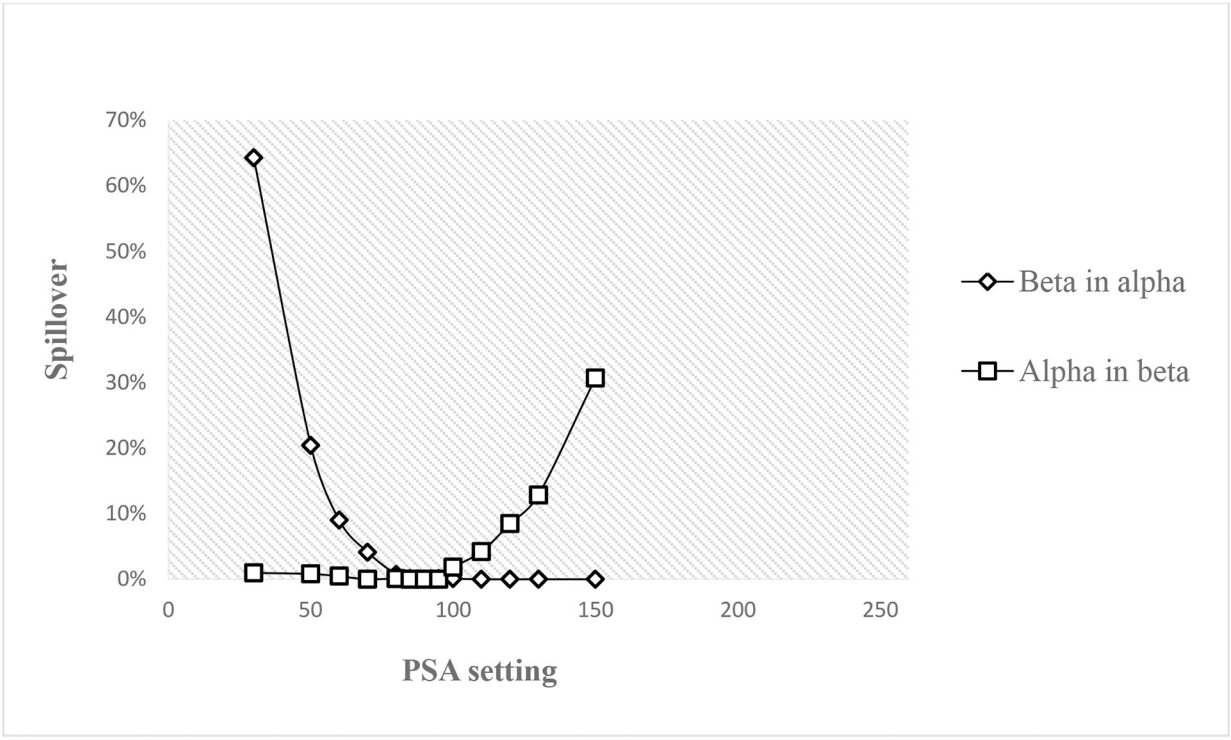

Fig. 4.
PSA optimazation on Quantulus1220 #2 using Am-241 and Sr-90/Y-90 as alpha and beta sources in urine matrix

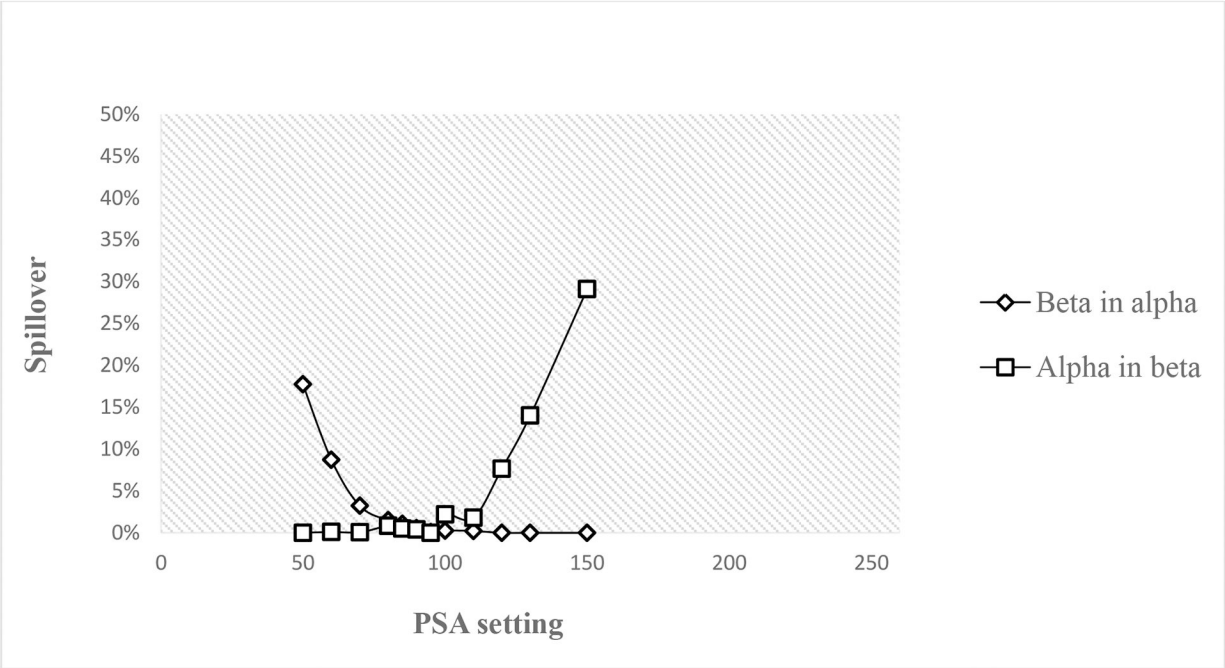

Fig. 5.
PSA optimization on Quantulus1220 #2 using Am-241 and Cs-137 as alpha and beta sources in urine matrix

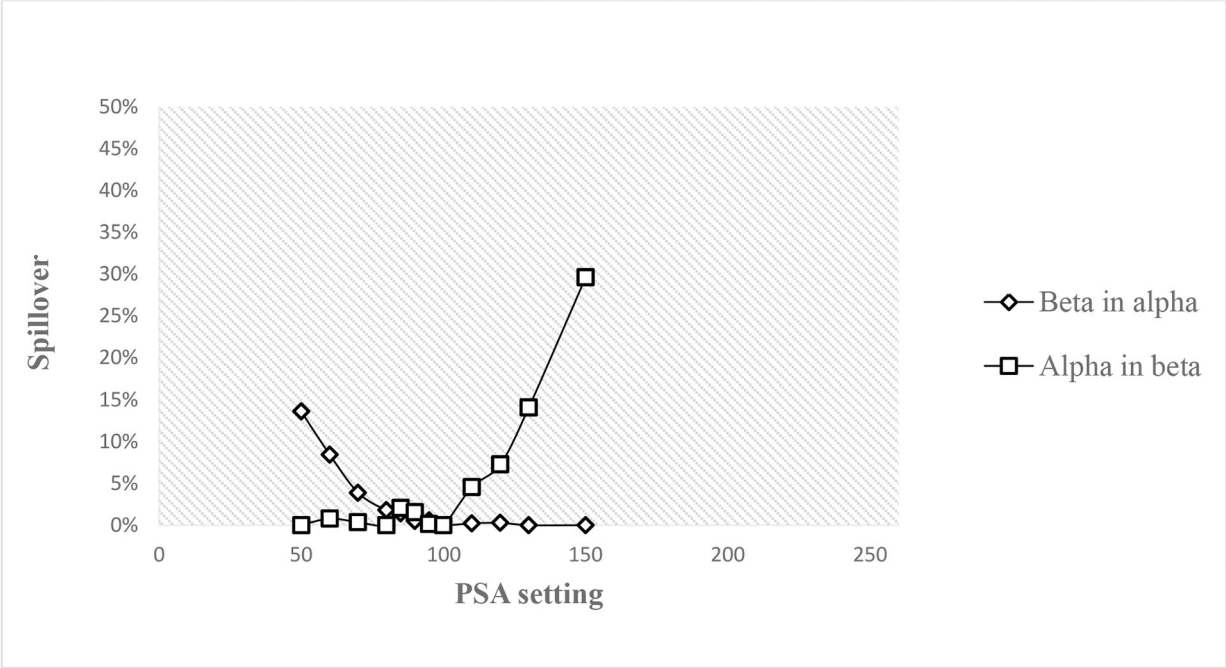

Fig. 6.

PSA optimization on Quantulus1220 #2 using Am-241 and Co-60 as alpha and beta sources in urine matrix

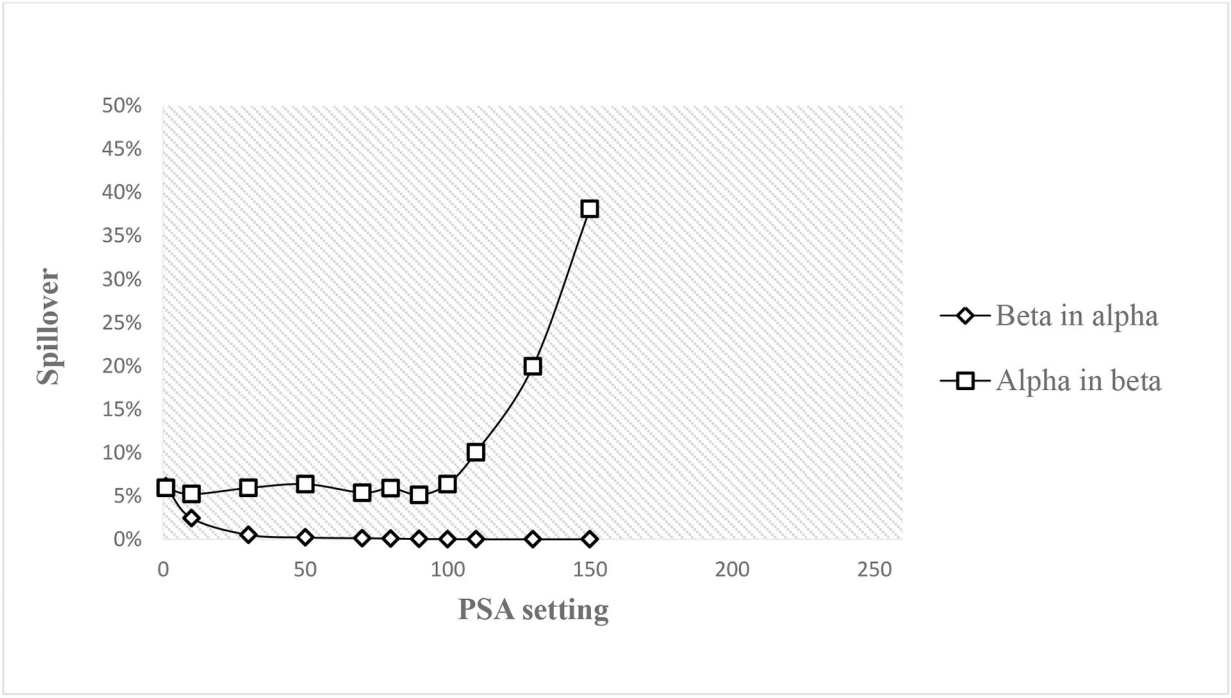

Fig. 7.
PSA optimization on Quantulus1220 #2 using Am-241 and H-3 as alpha and beta sources in urine matrix

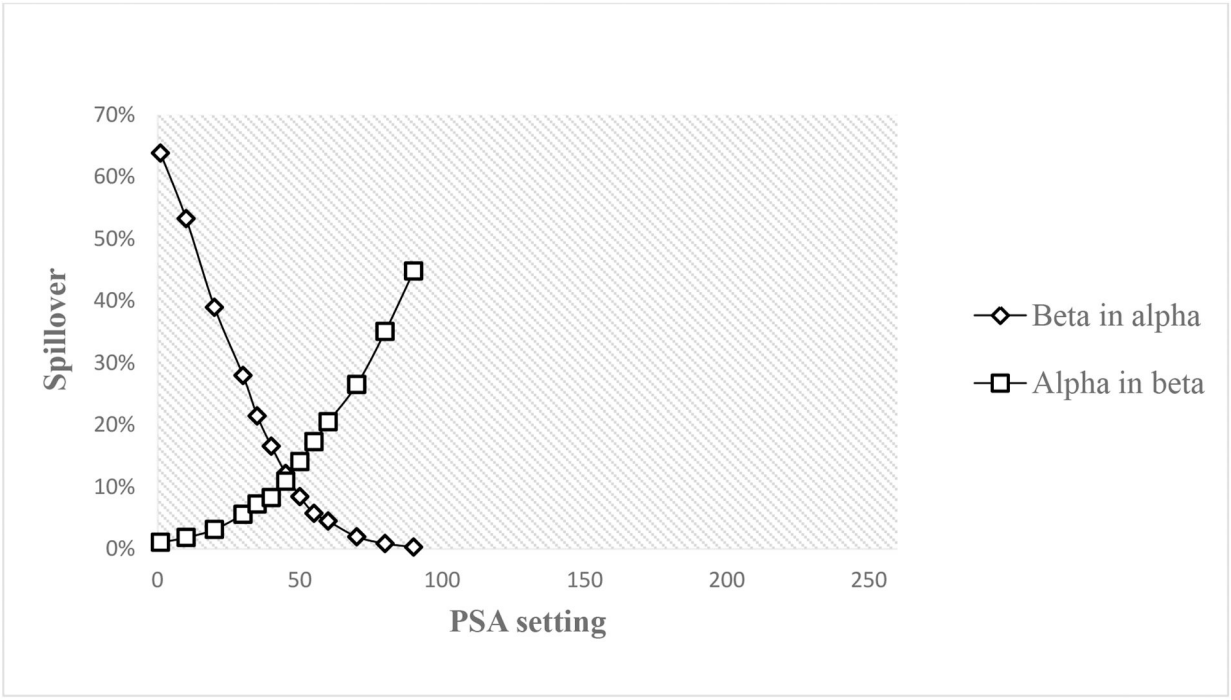

Fig. 8.

PSA optimization on Quantulus1220 #2 using Am-241 and Sr-90 as alpha and beta sources in fruit drink matrix

Table 1

Examples of Am-241 and Sr-90/Y-90 quench curves at different PSA settings on Quantulus1220 #2

Nuclide	PSA	Quench curves
Am-241	90	$y = -5.659E-10x^3 + 7.134E-07x^2 - 7.623E-05x + 9.581E-01$ $R^2 = 0.999$
Am-241	40	$y = -7.496E-10x^3 + 5.368E-07x^2 + 4.932E-04x + 6.266E-01$ $R^2 = 0.975$
Am-241	1	$y = -1.456E-09x^3 + 3.223E-06x^2 - 2.236E-03x + 1.547E+00$ $R^2 = 0.964$
Sr-90	90	$y = - 4.032E-08x^3 + 8.098E-05x^2 - 5.366E-02x + 1.267E+01$ $R^2 = 0.981$
Sr-90	40	$y = - 1.444E-08x^3 + 2.649E-05x^2 – 1.565E-02x + 3.911E+00$ $R^2 = 0.967$
Sr-90	256	$y = - 4.032E-09x^3 + 8.251E-06x^2 - 5.380E-03x + 2.070E+00$ $R^2 = 0.952$

Table 2

LSC method parameters for different Quantulus1220 instruments

Parameter	Quantulus1220 #1	Quantulus1220 #2	Quantulus1220 #3
PSA setting for urine gross alpha/beta bioassay	105	90	80
PSA setting for only beta emission events	256	256	256
PSA setting for only alpha emission events	1	1	1
Sample volume (mL)	5	5	5
Ultima Gold® AB volume (mL)	15	15	15
Sample analysis time (min)	5	5	5
External Std analysis time (min)	1	1	1
Alpha ROI (channels)	400–750	0–1024	400–750
High energy Beta ROI (channels)	1–1024	1–1024	1–1024
Low energy Beta ROI (channels)	1–250	1–250	1–250

Table 3

Mean and standard deviation (SD) data for base urine, low and high gross alpha/beta urine QC after 220 runs on different days using Quantulus1220 #1,2,3

QC sample	BU-077401		LU-077203		HU-077201	
	Alpha	Beta	Alpha	Beta	Alpha	Beta
Average (Bq/L)	0.7	60.8	1 010	12 000	2 560	30 400
SD (Bq/L)	1.09	9.77	43	289	79	647
Target Value (Bq/L)	N/A	N/A	1 020	12 300	2 560	30 300
Bias (%)			−0.98	−2.4	0.0	0.33

Table 4

Activity results with uncertainties for individual urine samples, spiked with Am-241 or Sr-90/Y-90 and with mixtures of these nuclides at various levels analyzed on Quantulus1220 #2 at optimal for urine PSA setting (PSA=90)

Sample ID	Found activities				Target activities		Bias	
	Gross Alpha (Bq/L)	Uncertainty (SD) (Bq/L)	Gross Beta (Bq/L)	Uncertainty (SD) (Bq/L)	Alpha (Bq/L)	Beta (Bq/L)	Alpha (%)	Beta (%)
Am50-U33	1 070	54.2	61	11.6	1 070	0	0.0	
Am50-U34	1 020	52.8	46	9.8	1 080	0	−5.6	
Am200-U35	4 120	106	35	8.4	4 300	0	−4.2	
Am200-U36	4 060	105	25	6.8	4 170	0	−2.6	
Sr50-U37	6	3.4	2 210	72.8	0	2 210		0.0
Sr50-U38	10	4.6	2 200	72.8	0	2 190		0.5
Sr200-U39	36	9.6	8 370	144	0	8 540		−2.0
Sr200-U40	26	8.0	8 150	140	0	8 460		−3.7
Am,Sr50-U41	1 070	54.4	2 140	71.8	1 080	2 210	−0.9	−3.2
Am,Sr50-U42	1 010	52.6	2 250	73.4	1 070	2 220	−5.6	1.4
Am,Sr200-U43	3 950	104	8 130	140	4 110	8 490	−3.9	−4.2
Am,Sr200-U44	3 930	103	8 130	140	4 190	8 430	−6.2	−3.6
Am50,Sr200-U45	1 140	56	8 190	142	1 120	8 410	1.8	−2.6
Am50,Sr200-U46	1 110	55.2	8 330	143	1 060	8 460	4.7	−1.5
Am200,Sr50-U47	3 900	104	2 300	74.4	4 160	2 170	−6.3	6.0
Am200,Sr50-U48	3 910	104	2 240	73.6	4 150	2 200	−5.8	1.8

Am50-U33, Am50-U34, Am200-U35, Am200-U36 – individual urine samples spiked with Am-241 at two levels.

Sr50-U37, Sr50-U38, Sr200-U39, Sr200-U40 – individual urine samples spiked with Sr-90/Y-90 at two levels.

AmSr50-U41, AmSr50-U42, AmSr200-U43, AmSr200-U44 – individual urine spiked with both Am-241 and Sr-90/Y-90 at the same two levels.

Am50Sr200-U45, Am50Sr200-U46 are individual urine spiked with Am-241 at low level and Sr-90/Y-90 at high level.

Am200Sr50-U47, Am200Sr50-U48 – are individual urine spiked with Am-241 at high level and with Sr-90/Y-90 at low level

Table 5

Activity results with uncertainties for individual urine samples, spiked with either Am-241 or Sr-90/Y-90 at various levels analyzed on Quantulus1220 #2 at PSA=1 or 256 setting

Sample ID	PSA	Found activities				Target activities		Bias	
		Gross Alpha (Bq/L)	Uncertainty (SD) (Bq/L)	Gross Beta (Bq/L)	Uncertainty (SD) (Bq/L)	Alpha (Bq/L)	Beta (Bq/L)	Alpha (%)	Beta (%)
Am50-U33	1	1 100	53.2			1 070	0	2.8	
Am50-U34	1	1 150	56.4			1 080	0	6.5	
Am200-U35	1	4 180	106			4 300	0	−2.8	
Am200-U36	1	4 130	105			4 170	0	−0.9	
Sr50-U37	256			2 300	76.6	0	2 210		4.1
Sr50-U38	256			2 280	76.0	0	2 190		4.1
Sr200-U39	256			8 870	151	0	8 540		3.9
Sr200-U40	256			8 470	144	0	8 460		0.1

Table 6

Gross alpha activity in FDA “Menu 2010” samples at different PSA setting

Sample ID	PSA setting	Measured			Target	
		Alpha (Bq/L)	Uncertainty (SD) (Bq/L)	Beta (crosstalk) (Bq/L)	Alpha (Bq/L)	Bias (%)
M3A002	90	272	13	145	389	−30.1
M3B001	90	200	12	192	433	−53.8
M3C001	90	147	11	139	299	−50.8
M3A002	80	348	15	67	389	−10.5
M3B001	80	323	15	107	433	−25.4
M3C001	80	195	12	99	299	−34.8
M3A002	70	384	16	41	389	−1.3
M3B001	70	344	15	95	433	−20.6
M3C001	70	222	13	74	299	−25.8
M3A002	60	345	15	41	389	−11.3
M3B001	60	398	17	46	433	−8.1
M3C001	60	237	12	59	299	−20.7
M3A002	50	386	16	14	389	−0.8
M3B001	50	422	18	49	433	−2.5
M3C001	50	267	14	52	299	−10.7
M3A002	40	395	16	7	389	1.5
M3B001	40	427	18	26	433	−1.4
M3C001	40	264	14	32	299	−10.5
M3A002	1	387	16		389	−0.5
M3B001	1	439	17		433	1.4
M3C001	1	268	13		299	−10.4

M3A002, M3B001, M3C001 are fruit drink samples spiked with Am-241 at different levels