Simpósio Qualidade da Água e Radioatividade

A COMPARISON OF ANALYTICAL METHODS FOR SCREENING GROSS ALPHA AND BETA RADIOACTIVITY IN WATER BY LSC AND GFPC

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Introduction

Water is not only a resource; it is also a life source. Therefore, access to safe drinking water is essential to health, a basic human right and a component of effective policy for health protection.

The importance of water, sanitation and hygiene for health and development has been reflected in the outcomes of a series of international policy forums, most recently the UN General Assembly declared the period from 2005 to 2015 as the International Decade for Action, "Water for Life".

Access to safe drinking water is important as a health and development issue at a national, regional and local level. Therefore, the WHO "Guidelines for Drinking-water Quality" is addressed primarily to water and health regulators, policymakers and their advisors, to assist in the development of national standards.

Radiological aspects

The approach taken in the WHO Guidelines for controlling radiological hazards has two stages:

I. initial screening for gross alpha and/or beta activity to determine whether the activity concentrations (in Bq/litre) are below levels at which no further action is required; and

II. if these screening levels are exceeded, investigation of the concentrations of individual radionuclides and comparison with specific guidance levels.

Radiological aspects

The current Guidelines are based on:

- a recommended reference dose level (RDL) of the committed effective dose, equal to 0.1mSv from 1 year's consumption of drinking-water.

- This comprises 10% of the intervention exemption level recommended by the ICRP for dominant commodities (e.g., food and drinking-water) for prolonged exposure situations, which is most relevant to long-term consumption of drinking water by the public (ICRP, 2000).

Screening of drinking-water supplies

The process of identifying individual radioactive species and determining their concentration requires sophisticated and expensive analysis, which is normally not justified, because the concentrations of radionuclides in most circumstances are very low.

A more practical approach is to use a screening procedure, where the total radioactivity present in the form of alpha and beta radiation is first determined, without regard to the identity of specific radionuclides.

Screening of drinking-water supplies - National regulation

In Brazil, the Guideline for controlling radioactivity levels in drinking-water is established by Ministério da Saúde (MS Nº 2.914, 12 de Dezembro de 2011). According to this Guideline, the screening levels for drinking-water below which no further action is required are:

- 0.5 Bq/litre for gross alpha activity; and
- 1 Bq/litre for gross beta activity.

Objectives

The question that arises is "how to measure low alpha and beta activity concentration in water"?

- The classical approach consists of evaporating a known volume of water and measure the activity of the residue in a glass flow proportional counter;

- An attractive alternative to classical methods is the ultra-low level liquid scintillation counting (LSC) coupled to alpha–beta discrimination. This method allows rapid and simple determination of gross alpha and beta activities, which are simultaneously measured through alpha–beta discrimination technique.

Objectives

Objectives

The main aim of this presentation is to compare these two methodologies, considering an identical initial step of pre-concentration, in terms of:

1. Principles involved in the measurement (efficiency, measuring time, background radiation);

2. Minimal Detectable Activity (MDA);

3. Time to perform the analysis.

Gas Flow Proportional Counting (GFPC)

The GFPC presents the following characteristics for the determination of low gross alpha and beta activity in water: simultaneous alpha and beta counting using a discriminator; low background radiation; utilization of a gas, usually a P-10 mixture (10% methane and 90% argon). This gas is necessary to avoid the formation of secondary avalanches, which were not caused by the ionizing radiation.

The GFPC presents, however, the following limitations:

- Self-absorption (the radiation from alpha emitters having an energy of 8 MeV and from beta emitters having an energy of 60 keV will not escape from the sample if the emitters are covered by a sample thickness of 5.5 mg/cm2 (Standard Methods, 1985);

- Crosstalk correction method for differentiating beta from alpha radiation may not adequately protect from misidentification for samples containing very low-energy alpha or high-energy beta energies;

- As the gas flow proportional counting does not provide any spectroscopic information, it cannot be used to identify the radionuclides detected and the technique is not suitable for the detection of low energy beta emitters (< 200 keV).

Ultra low background Liquid Scintillation counting (LSC)

The wide popularity of liquid scintillation analysis is a consequence of numerous advantages, which are: high efficiencies of detection, improvements in sample preparation techniques, automation including computer data processing and the spectrometer capability of liquid scintillation analyzers permitting the simultaneous assay of different radionuclides.

The LSC presents, however, the following limitations:

- quenching,
- materials of vials that can interfere in the counting, and
- the alpha-peaks are rather broad due to a very poor energy resolution (compared to a spectrum measured with a semiconductor detector).

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Comparison between LSC and GFPC to measure gross alpha and gross beta in water

The question that arises is "what is the best choice for a large number of analysis"?

In order to answer this question, the following parameters will be discussed: analytical parameters, time and cost of analysis.

Experimental procedure

The equipment used for the measurement of gross alpha and beta activities were 1220 Quantulus™ Ultra Low Level Liquid Scintillation Spectrometer and low background gas flow proportional detector (10-channel Low-Level Planchet Counter LB 770 Berthold).

The initial step of pre-concentration of the water samples is the same for the two techniques and consists of concentrate 1 L of water on a hot plate, at a temperature of 80 °C, to a final volume of 50 mL.

Experimental procedure

- For the GFPC method, an aliquot of 4 mL of this final solution is heated (on infrared light) to dryness on a stainless steel planchet and is counted on a gas flow proportional counter.

- For the LSC method, an aliquot of 5 mL of the same final solution is mixed with 15 mL of the scintillation solution in the appropriate vial and is counted on a liquid scintillation counter.

Analytical Parameters

EFFICIENCY

Table 1. Efficiencies of GFPC and LSC

	Efficiency (cps.dps^{-1})	
Detector	alpha[1]	beta[1]
GFPC	0.21 ± 0.02	0.33 ± 0.03
LSC	0.77 ± 0.04	0.55 ± 0.05

[1] expanded uncertainty

COMPARISON OF EFFICIENCIES

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Detector	Efficiency ($cps.dps^{-1}$) alpha[1]	beta[1]
GFPC	0.21 ± 0.02	0.33 ± 0.03
LSC	0.77 ± 0.04	0.55 ± 0.05

[1] expanded uncertainty

It can be seen that LSC presents better efficiencies for both, alpha and beta counting.

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BACKGROUND

Table 2. Background for GFPC and LSC

Counting time (min)	GFPC[1]		LSC[2]	
	alpha (cpm)	beta (cpm)	alpha (cpm)	beta (cpm)
30	0.07 ± 0.05	0.61 ± 0.09	0.14 ± 0.03	1.7 ± 0.2
60	0.06 ± 0.04	0.6 ± 0.1	0.11 ± 0.06	1.7 ± 0.2
120	0.06 ± 0.03	0.6 ± 0.1	0.06 ± 0.03	1.7 ± 0.1
200	0.07 ± 0.03	0.6 ± 0.1	0.06 ± 0.02	1.7 ± 0.1
300	0.07 ± 0.03	0.6 ± 0.1	0.07 ± 0.01	1.77 ± 0.09
400	0.07 ± 0.03	0.6 ± 0.1	0.07 ± 0.01	1.76 ± 0.03

[1]average value and standard deviation of 10 detectors.
[2] average value and standard deviation of 7 background counting.

BACKGROUND

GFPC

counting time (min)	background (cpm)	
	alpha	beta
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LSC

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120	0.06 ± 0.03	1.7 ± 0.1
200	0.06 ± 0.02	1.7 ± 0.1
300	0.07 ± 0.01	1.77 ± 0.09
400	0.07 ± 0.01	1.76 ± 0.03

It is observed that the alpha background for counting times higher than 120 min are the same for the LSC and GFPC techniques, the corresponding uncertainties for the GFPC technique were similar , whereas the uncertainties and consequently precision for the LSC improve with the increase of the counting time.

BACKGROUND

GFPC

counting time (min)	background (cpm)	
	alpha	beta
30	0.07 ± 0.05	0.61 ± 0.09
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400	0.07 ± 0.01	1.76 ± 0.03

The beta background obtained for the LSC and GFPC techniques did not vary with the counting time, however the results obtained for the LSC are higher than the GFPC but with uncertainties decreasing with counting time.

Table 3. Figure of Merit obtained for LSC and GFPC

Detector	FOM[1,2]	
	alpha	beta
GFPC	7350	1815
LSC	98817	1779

[1] Figure of merit (counting efficiency[2]/background)
[2] Background values for a counting time of 120 min

An approach useful for comparison of sensibility for different methodologies is the Figure of Merit (FOM), which relates the square root of the efficiency with the background.

Table 3. Figure of Merit obtained for LSC and GFPC

Detector	FOM	
	alpha	beta
GFPC	7350	1815
LSC	98817	1779

These results showed that the LSC technique presents higher sensibility for alpha measurement.

Table 3. Figure of Merit obtained for LSC and GFPC

Detector	FOM	
	alpha	beta
GFPC	7350	1815
LSC	98817	1779

For the beta measurement the two techniques presented similar sensibilities

It is important to emphasize that the beta window used in the LSC technique was defined to take into account beta energies higher than 200 keV, since energies lower than that cannot be detected by the GFPC technique and the objective of this study is to compare both techniques.

MINIMUM DETECTABLE ACTIVITY

Table 4. Results of MDA for LSC and GFPC using different counting times

Counting time (min)	MDA ($Bq.L^{-1}$)			
	GFPC		LSC	
	Alfa	Beta	Alfa	Beta
30	0.204	0.419	0.068	0.34
60	0.142	0.285	0.043	0.24
120	0.101	0.203	0.023	0.17
200	0.082	0.157	0.018	0.13
300	0.066	0.131	0.015	0.11
400	0.057	0.113	0.013	0.09

Maximum permissible values (VMP) for alpha = 0.5 $Bq.L^{-1}$, beta = 1.0 $Bq.L^{-1}$)

Another parameter important in the comparison of different methodologies is the minimum detectable activity (MDA).

GFPC

Counting time (min)	MDA (Bq.L^{-1})	
	alpha	beta
30	0.204	0.419
60	0.142	0.285
120	0.101	0.203
200	0.082	0.157
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400	0.057	0.113

LSC

Counting time (min)	MDA (Bq.L^{-1})	
	alpha	beta
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60	0.043	0.24
120	0.023	0.17
200	0.018	0.13
300	0.015	0.11
400	0.013	0.09

The LSC technique is clearly more advantageous in terms of sensibility, especially for the measurement of alpha particles.

GFPC

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	alpha	beta
30	0.204	0.419
60	0.142	0.285
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400	0.013	0.09

For the beta measurement the two techniques presented similar MDA.

Table 5. Parameters for the determination of total alpha and beta activity in water by LSC

Sample concentration factor	20
Scintillation solution	Ultima Gold AB
Volume (ml)	5
Standard solution for calibration	^{241}Am and $^{90}Sr/^{90}Y$
Counting window	(alpha= 500–1000) and (beta = 500–1000)
Efficiency ($cps.dps^{-1}$)	(εalpha = 0.77 ± 0.04)[1] and
	(εbeta = 0.55 ± 0.05)[1]
Discrimination of alpha/beta (PSA)	100
Background (cpm)	(alpha = 0.07 ± 0.01)[2] and
	(beta = 1.76 ± 0.03)[2]
Counting time (min)	120
MDA ($Bq.L^{-1}$)	(alpha < 0.02) e (beta < 0.17)

[1] expanded uncertainty.
[2] average value and standard deviation for 7 background counts (DOQ-CGCRE-008).

Table 6. Parameters for the determination of total alpha and beta activity in water by GFPC

Sample concentration factor	20
Volume (ml)	4
Standard solution for calibration	^{241}Am and $^{90}Sr/^{90}Y$
Operating voltage (V)	1650
Efficiency ($cps.dps^{-1}$)	(εalpha = 0.21 ± 0.02)[1] and (εbeta = 0.33 ± 0.03)[1]
Correction	Spillover Correction
Background (cpm)	(alpha = 0.07 ± 0.03)[2] and (beta = 0.6 ± 0.1)[2]
Counting time (min)	120
MDA ($Bq.L^{-1}$)	(alpha < 0.10) and (beta < 0.20)

[1]expanded uncertainty.
[2] average value and standard deviation for 10 background counts.

Performance of GFPC and LSC techniques

The performance of the GFPC and LSC methods for the measurement of gross alpha and beta activity in water was evaluated by participating in Proficiency Tests organized by Instituto de Radioproteção e Dosimetria IRD/CNEN, which is available on a routine basis three times per year.

Table 7. Performance of GFPC and LSC methods for the determination of gross alpha activity in water, by using the normalized standard deviation (D)

		GFPC		LSC	
PT (month/ year)	Reference value ($Bq.L^{-1}$)	Measured value[1] ($Bq.L^{-1}$)	D[2] $D = \frac{(X - U}{\frac{su}{\sqrt{n}}}$	Measured value[1] ($Bq.L^{-1}$)	D $D = \frac{(X - U)}{\frac{su}{\sqrt{n}}}$
dez/09	0.600 ± 0.120	0.673 ± 0.032	1.06	0.619 ± 0.122	0.22
abr/11	0.420 ± 0.084	0.521 ± 0.017	2.09	0.458 ± 0.010	0.63
ago/11	0.860 ± 0.170	0.803 ± 0.033	-0.58	0.917 ± 0.050	0.48
abr/12	0.520 ± 0.100	0.482 ± 0.036	-0.65	0.524 ± 0.048	0.06
ago/12	1.050 ± 0.210	1.078 ± 0.034	0.23	1.042 ± 0.022	-0.06
dez/12	0.620 ± 0.120	0.627 ± 0.026	0.10	0.655 ± 0.007	0.50
abr/13	0.530 ± 0.110	0.536 ± 0.044	0.09	0.524 ± 0.019	0.18
ago/13	0.730 ± 0.150	0.626 ± 0.051	-1.20	0.689 ± 0.020	-0.47

[1]average value and standard deviation of 3 measurements.

[2] D: normalized standard deviation, X: average value of three independent measurements, U: reference value, su: reference value standard deviation and n: square root of the number of independent measurements.

PT (month / Year)	ALPHA				
	Reference value (Bq.L^{-1})	GFPC		LSC	
		Measured value[1] (Bq.L^{-1})	D	Measured value[1] (Bq.L^{-1})	D
dez/09	0.600 ± 0.120	0.673 ± 0.032	1.06	0.619 ± 0.122	0.22
abr/11	0.420 ± 0.084	0.521 ± 0.017	2.09	0.458 ± 0.010	0.63
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When *D* results are within the interval $-2 \leq D \leq +2$, the performance of the laboratory is considered good. Results included in the interval $-3 < D < -2$ or $+2 < D < +3$ are considered acceptable, but are within warning limits; data with $D \leq -3$ or $D \geq +3$ indicate that the measurement system is out of control and the performance is not acceptable.

The best performance for the determination of alpha activity in water was achieved by the LSC technique

Table 8. Performance of GFPC and LSC methods for the determination of gross beta activity in water, by using the normalized standard deviation (D)

		GFPC		LSC	
PNI (month/ year)	Reference Value ($Bq.L^{-1}$)	Measured value [1] ($Bq.L^{-1}$)	D^2 $D = \frac{(X-U)}{\frac{su}{\sqrt{n}}}$	Measured value [1] ($Bq.L^{-1}$)	D $D = \frac{(X-U)}{\frac{su}{\sqrt{n}}}$
dez/09	2.228 ± 0.458	2.197 ± 0.035	-0.35	1.876 ± 0.080	-1.09
abr/11	0.910 ± 0.182	1.144 ± 0.047	2.23	0.868 ± 0.001	-0.33
ago/11	1.38 ± 0.280	1.513 ± 0.075	0.82	1.569 ± 0.051	0.95
abr/12	0.960 ± 0.190	1.032 ± 0.025	0.66	1.013 ± 0.062	0.48
ago/12	3.080 ± 0.620	3.354 ± 0.098	0.76	3.051 ± 0.044	-0.07
dez/12	2.060 ± 0.410	2.007 ± 0.031	-0.23	2.025 ± 0.043	-0.15
abr/13	0.830 ± 0.170	0.953 ± 0.087	1.25	0.885 ± 0.053	0.56
ago/13	2.370 ± 0.470	2.267 ± 0.035	-0.38	2.279 ± 0.132	-0.34

[1]average value and standard deviations of 3 measurements.

PT (month / Year)	BETA				
	Reference value (Bq.L⁻¹)	GFPC		LSC	
		Measured value[1] (Bq.L⁻¹)	D	Measured value[1] (Bq.L⁻¹)	D
dez/09	2.228 ± 0.458	2.197 ± 0.035	-0.35	1.876 ± 0.080	-1.09
abr/11	0.910 ± 0.182	1.144 ± 0.047	2.23	0.868 ± 0.001	-0.33
ago/11	1.38 ± 0.280	1.513 ± 0.075	0.82	1.569 ± 0.051	0.95
abr/12	0.960 ± 0.190	1.032 ± 0.025	0.66	1.013 ± 0.062	0.48
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[1]average value and standard deviation of 3 measurements.

The results obtained for the determination of gross beta activity in water also showed that the best performance was achieved by the LSC technique.

Time of analysis

For the GFPC method, an aliquot of 4 mL of this final solution is heated (on infrared light) to dryness on a stainless steel planchet (the time required in this procedure is approximately **3 hours** for the evaporation of the sample).

The sample is counted three times on a gas flow proportional counter for **120 minutes**.

The planchets used must be counted empty, three times, before the analysis for the evaluation of the background, which will be subtracted from the final measurement. That means that the time necessary to perform the whole analysis is increased in **6 hours**.

Time of analysis

In this section, the time necessary for the determination of the gross alpha and beta activity using the two methodologies, excluding the initial step of pre-concentration, which is the same for both, will be compared.

Since, the GFPC allows the simultaneous determination of 10 samples, the comparison with the LSC will be done, considering the time spent for the measurement of 10 samples.

The initial step of pre-concentration of the water samples is the same for the two techniques and consists of concentrate 1 L of water on a hot plate, at a temperature of 80 °C, to a final volume of 50 mL.

Time of analysis

For the LSC method, an aliquot of 5 mL of the same final solution is mixed with 15 mL of the scintillation solution in the appropriate vial.

The time required for the homogenization and stabilization of the temperature in the vials is around **30 minutes**. The sample is counted on a liquid scintillation counter for **120 minutes.**

The vials used must be counted empty, before the analysis for the evaluation of the background, which will be subtracted from the final measurement.

That implies in an increase of **two hours** in the total time necessary to perform the analysis.

Table 9. Time necessary for the analyses of 10 samples by LSC and GFPC

Technique	Pre-concentration time (h)	Sample preparation time (h)	BG counting time (h)	Sample counting time (h)	Total time for 10 measurement
GFPC	16	3	6[1]	6	31 h
LSC	16	0.5	2[1]	20	38.5 h

[1] Background counting time adopted considering a good reproducibility of the system.

Although the LSC requires more time (around 25%) to perform the 10 analysis, it should be emphasized that it can measure 60 samples automatically, without human intervention.

In conclusion, the total number of analysis that can be performed is equivalent for the two techniques, considering that the technician, who will execute the analysis, works 8 hours per day.

Conclusion

Comparing the two techniques for the screening of total alpha and beta activity in water, it can be concluded that:

1. The LSC technique presented results more precise and accurate, considering all the parameters studied background, efficiency, sensibility and MDA.

2. As for the time necessary to perform 10 analysis, the GFPC technique requires 31 hours and the LSC 38.5 hours. However, the LSC can measure 60 samples automatically, without human intervention.

3. Although the cost of the LSC equipment is three times higher than the GFPC equipment, it offers a wider range of applications, such as determination of ^{3}H and ^{14}C. Another advantage of the LSC is that it is capable of measuring radionuclides with lower energies what is not possible with the GFPC. Finally, the LSC allows the identification of radionuclides alpha and beta emitters in the obtained spectra.

THANK YOU FOR YOUR ATTENTION

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