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TOTAL ALPHA AND BETA DETERMINATION BY LIQUID SCINTILLATION COUNTING IN WATER SAMPLES FROM A BRAZILIAN INTERCOMPARISON EXERCISE

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ABSTRACT

This paper describes CNEN/LAPOC's participation in the Brazilian Intercomparison Exercise (PNI) for simultaneous determination of total radioactivity in water samples, which took place in August and December 2008. The Proficiency Test (PT) also involved a short description of the nuclear analytical technique employed, emphasizing sources of uncertainty. A Liquid Scintillation System (Packard TRICARB 2700) was used with appropriate corrections applied to final results, expressed as Bq L^{-1}. Participation and PT data provide independent information on performance of a Laboratory and have an important role in method validation; especially because it allows the assessment of method performance over an entire range of concentrations and matrices. PT is also an important tool to demonstrate equivalence of measurements, if not their metrological comparability, and to promote education and improvement of Lab practices.

1. INTRODUCTION

Water for human consumption is regulated under the Brazilian Health Ministry Norm 518 [1], which establishes procedures regarding water quality and drinking standards, such as radioactivity standards. As per the current Norm, maximum permitted value for alpha activity is set at 0.1 Bq L^{-1} and 1.0 Bq L^{-1} for beta activity. The guideline levels are calculated using a water consumption of 2 L d^{-1}, assuming that ^{226}Ra and ^{90}Sr are present as alpha and beta emitters, respectively. Further analyses are required only if guideline levels are exceeded [2].

Radiological examination of water requires a rapid screening technique that allows the determination of gross alpha and beta activities of each sample in order to decide if further radiological analyses are necessary. Water quality is an important concern in environmental studies because of its use for human consumption and ability to transport pollutants in the environment. Natural waters contain a number of both alpha (^{238}U, ^{226}Ra, and ^{210}Po) and beta emitters (^{40}K, ^{228}Ra, and ^{210}Pb) in broad concentration range, which may be responsible for a fraction, generally small, of the internal exposure by ingestion of natural radionuclides.

Many radionuclides are transferred to the water from the aquifer rock by erosion and dissolution mechanisms [3]. Radiological safeguards of drinking water are based on the control of natural and man-made radionuclide concentrations. Alpha emitters are the most dangerous radionuclides in case of ingestion. Two dangerous elements in this case are: radium, due to its similarity in behavior to calcium, an element commonly fixed in bones; and uranium, due to its commonly high solubility. In general, ^{214}Pb and ^{214}Bi are usually indicators of the presence of their progenitor, ^{226}Ra, which is originated by ^{238}U. Likewise, ^{228}Ac indicates the presence of its progenitor ^{228}Ra, originated by ^{232}Th [4].

Health effects from drinking water containing alpha and beta emitters in excess of the MCL (Maximum Contaminant Level) over many years may increase cancer risk. The “parent radionuclide” often behaves very differently from the “daughter” radionuclide in the environment. Because of this, parent and daughter radionuclides may have different drinking water occurrence patterns. For example, groundwater with high radium levels tends to have low uranium levels and vice-versa, even though ^{238}U is the parent of ^{226}Ra [5].

Protection of public health requires, among other things, the regulation of exposure to ionizing radiations through the consumption of drinking water. A full characterization of natural water samples in environmental levels is an expensive and complex operation. It is a well accepted cost-effective procedure to screen all samples for presence of gross activities, an estimation of the total activity of all alpha and high energy beta emitters.

In scientific terms, gross alpha activity is defined as the total activity of all alpha emitters once radon has been eliminated. Gross beta activity is defined as the total activity of all beta emitters excluding tritium, through ^{14}C and other soft-beta emitters are also excluded by most commonly used screening techniques [2].

In order to measure activity in water samples, fast and reliable nuclear techniques are necessary. Liquid Scintillation Counting is a rapid technique, gives spectral information, and is adequate to determine gross activities according to the World Health Organization guideline values [6]. According to a study performed by Forte et al (2007) [3], liquid scintillation counting proved to be very effective for both a quick screening of drinking waters and for single radionuclide determination, if suitable procedures are used.

2. MATERIALS AND METHODS

In general, for total alpha and beta determination employing a Liquid Scintillation System (LSC), an aliquot of 1 L of sample is used if the activity is known to be low. If a sample has high activity, an aliquot of 100 mL or less can be used.

For this study, 100 mL of samples were evaporated until dryness (avoiding boiling) and dissolved with 10 mL of 0.1M nitric acid. The solutions were then transferred to a liquid scintillation vial containing 10 mL of Packard AB Ultima Gold cocktail, and agitated during 3 minutes.

Some precautions were taken during sample preparation procedure. The evaporation was done in an exhaust hood to avoid incorporation of organic solvents. Automatic micro-pipettes were properly calibrated. Sample agitation was done by the same operator for the analyzed set.

Counting was performed under an adequate protocol adjusted to cover a broad range of channels (0 to 2 000). Water samples were generally counted for only 240 minutes. In order to determine the best counting configuration of the system, pulse shape discrimination was optimized as 128. If the sample was not counted immediately after preparation, it was stored under refrigeration.

A pure ^{237}Np standard was used to determine alpha efficiency and a standard of $^{90}Sr/^{90}Y$ was used to determine beta efficiency for the LSC analysis. In this paper, a Packard TRICARB 2700 LSC System [7] presented a background count of approximately 4 cpm in the alpha channel and 72 cpm in the beta channel (an average of past 10 background measurements). Efficiencies obtained were of approximately 1.0. The average detection limit was determined to be 0.8 Bq L^{-1} for this specific system.

The following equations, from 1 to 4, were used to calculate the alpha and beta efficiency and activity. It is important to note that the obtained beta efficiency was divided by 2, since counting rates referred to the sum of ^{90}Sr and ^{90}Y.

$$\text{Alpha Efficiency} = \frac{\text{Alpha Standard Counting on Alpha Channel} - \text{Alpha Channel Background}}{\text{Alpha Standard Activity x 60}} \quad (1)$$

$$\text{Alpha Activity} = \frac{\text{Sample Counting on Alpha Channel} - \text{Alpha Channel Background}}{\text{60 x Alpha Efficiency x Sample Volume}} \quad (2)$$

$$\text{Beta Efficiency} = \frac{\text{Beta Standard Counting on Beta Channel} - \text{Beta Channel Background} \div 2}{\text{Beta Standard Activity x 60}} \quad (3)$$

$$\text{Beta Activity} = \frac{\text{Sample Counting on Beta Channel} - \text{Beta Channel Background}}{\text{60 x Beta Efficiency x Sample Volume}} \quad (4)$$

Figure 1. Packard TRICARB 2700 Liquid Scintillation System

Figure 2. Addition of LSC cocktail to glass vial containing a spiked water sample.

3. RESULTS AND DISCUSSION

Table 1 summarizes the results from Liquid Scintillation Counting (LSC) obtained by the Poços de Caldas Laboratory (LAPOC) in spiked water samples from the Brazilian

Intercomparison Exercise (PNI), which took place in August and December 2008 and was organized by CNEN/IRD (Brazilian Nuclear Energy Commission/Dosimetry and Radioprotection Institute). Table 2 presents the parameters set up for the LSC Protocol employed.

According to the Official PNI report, which includes statistics prepared by CNEN/IRD, the results were within the interval of acceptable values, and also within the interval of good values between warning levels.

Results for total alpha and beta determination showed that LSC was an adequate radioanalytical technique for this type of measurement in water samples. It is routinely used at LAPOC to verify results obtained from other techniques, such as Low Background Gas Proportional Alpha and Beta Counting.

Table 1. Results for Total Alpha and Beta Radioactivity in Spiked Water Samples

	Alpha (BqL^{-1})	**Beta (BqL^{-1})**
Real Value (PNI August 2008)	1.290 ± 0.258	1.010 ± 0.202
LSC Value (Determined at LAPOC)	1.510 ± 0.076	1.016 ± 0.051
Real Value (PNI December 2008)	0.510 ± 0.102	1.319 ± 0.264
LSC Value (Determined at LAPOC)	0.530 ± 0.027	1.280 ± 0.064

Table 3 presents results from PNI Intercomparison Exercises carried out in August and December 2008. Total alpha and beta radioactivity results obtained by LSC are comparable to results obtained through employment of Gas Proportional Counting and Gamma Spectrometry.

Uncertainties were not reported, since they were not solicited at the time of reporting by the Organizing Agency (CNEN/IRD), which ultimately works with the standard deviation of all the measurements for their statistical official report.

Table 2. LSC Protocol Description for Total Alpha and Beta Determination

Parameter	Configuration
Count Time	240 min
Discriminator	128
Data Mode	CPM
Cycles	1
Background Subtract	No
All Region 2 Sigma?	No
Region A	0.0-2000.0 channel
Region B	0.0-0.0 channel
Alpha	0.0-2000.0 channel
QIP	SIS
ES Terminator	COUNT
% Reference?	No
# Counts/Vial	1
# Vials/Sample	1
Half Life Correction?	No
Region A	0.0
Region B	0.0
Ref Time	0:0:0
Luminescence Correction?	No
Heterogeneity Monitor?	No
Colored Samples?	No
Low Level/ HSCM – Count Mode?	No
Static Controller?	Yes
Coincidence Time (ns)?	18
Delay Before Burst?	Normal

Table 3. Results for Total Alpha and Beta Radioactivity using Different Nuclear Analytical Techniques

PNI August 2008	Alpha (BqL^{-1})	Beta (BqL^{-1})
Real Value	1.290 ± 0.258	1.010 ± 0.202
Liquid Scintillation Counting	1.510	1.016
Gas Proportional Counting	1.330	0.995
Gamma Spectrometry	1.300	1.042
PNI December 2008	**Alpha (BqL^{-1})**	**Beta (BqL^{-1})**
Real Value	0.510 ± 0.102	1.319 ± 0.264
Liquid Scintillation Counting	0.530	1.280
Gas Proportional Counting	0.450	1.070
Gamma Spectrometry	0.500	1.350

In order to promote Laboratory quality assurance, background, blank, and reference samples are routinely analyzed. In addition, there is continuous participation in Intercomparison Exercises organized by several Agencies.

Participation in PTs has been used as an effective tool for monitoring radioanalytical measurement performance and also to identify presence of variances in measurement results that are related to instrument performance and technical procedures.

The uncertainty reported for LSC measurements was considered to be approximately 5%, based on previous Laboratory experience.

4. CONCLUSIONS

Liquid Scintillation Counting (LSC) was an adequate nuclear technique to determine total radioactivity levels, as verified by this study. It is an important analytical tool as a broad range of equipment is ever more needed to attend increasing water consumption demands. When LSC equipment is available to a Laboratory, it can properly substitute more expensive nuclear equipment.

Proficiency Exercises provide a continuous, independent, third-part monitoring of all aspects of an analytical program for participating laboratories. They can verify accuracy, precision, and variations within radioanalytical techniques. As this matures, with information exchange and technical discussions, the quality of determinations is decreased. The Brazilian Intercomparison Program (PNI) specifically emphasizes the importance of participating in PTs, as the need for measurements in environmental samples is increasing.

Several intercomparison activities are currently in place at LAPOC to further improve Laboratory quality, scientific background, and personnel skills to fulfil requirements for accreditation.

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