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Study of Gross alpha beta radioactivity content in commercial water brands by liquid scintillation

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This study presents the gross alpha beta radioactivity of bottled water samples taken from twenty different commercial brands to assess the safety of drinking-water with respect to its radionuclide content. In Mexico, a limit of 0.5 and 1.0 Bq/L is established for Gross alpha and beta radioactivity in drinking-water is established by the official Mexican standard NOM-127-SSA1-2021. The analysis was carried out using the liquid scintillation method, making a concentration of 10 to 1 to obtain a minimum detectable concentration of 0.25 and 0.5 Bq/L respectively. The obtained results indicate that in all cases the bottled waters comply with the applicable standard within the official NOM. These results indicate that the effective annual dose would be less than 0.1 mSv, providing evidence that the radiological risk for the consumer is negligible according to this norm.

Keywords: Radionuclides; Gross alpha beta radioactivity; liquid scintillation; radiological risk.

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1. Introduction

Radioactivity is a natural phenomenon commonly found in the environment stemming from various natural and human-induced sources. Groundwater, surface water, and seawater have been observed to serve as reservoirs for radionuclides, which may originate from these both natural processes and human activities. Causing, these water bodies, to potentially act as harboring vessels of a spectrum of radionuclides ranging from those naturally occurring to those synthesized by human endeavors.

Natural radionuclides such as ^{40}K, ^{3}H, and ^{14}C, as well as those originating from the decay series of ^{238}U, ^{232}Th, and ^{235}U, including ^{226}Ra, ^{228}Ra, ^{234}U, ^{238}U, ^{210}Po, and ^{210}Pb, are present in water due to desorption from the soil wash-off by rainwater, or release from technological processes involving Naturally Occurring Radioactive Materials (NORM) [1]. These processes may include the mining and processing of mineral sands or the production and use of phosphate fertilizers [2]. Radionuclides such as transuranium elements like Am, Pu, Np, Cm, and radioisotopes like ^{3}H, ^{14}C, and ^{90}Sr can also be present in natural waters due to human activities.

Some quantities of radionuclides are discharged into the environment as part of authorized routine releases from nuclear fuel cycle facilities -specific radionuclides employed in medical and industrial applications released into the environment after use. Adding to these, because historical fallout, contamination due to historical fallow resulting from atmospheric propagation due to nuclear device explosions and other accidents, such as those happening in events like Chernobyl and Fukushima.

Another cause of radionuclide activity concentration in water bodies has been explained based on local geological characteristics and different climatic conditions. Adding to this variability, the releases from nuclear installations during planned, existing, and emergency exposure situations lead to site specific changes in the increasing concentration of radionuclides, which can pose potential risks to human health and its surroundings.

One critical aspect for ensuring water quality for human consumption is the monitoring and analysis of its radioactivity content. International guidelines advocate for an annual equivalent dose not exceeding 0.1 mSv [3]. In alignment with this objective and considering average radionuclide concentrations in water, the World Health Organization (WHO) has recommended limits of 0.5 Bq/L for gross alpha radiation and 1.0 Bq/L for gross beta radiation recommended values that were adopted as limiting values within the Official Mexican Standard- NOM-127-SSA1-2021 [4].

In recent years, the consumption of bottled water has surged in Mexico and globally. Mexico, alongside Thailand, has become one of the largest consumers of bottled water in the world, with an average per capita consumption of 274 liters per year [5]. This accelerated demand has prompted various commercial firms to market bottled water extensively, resulting in a substantial supply in the market. To assess the safety of water brands in Mexico, a study of 20 of the most consumed bottled-water brands was carried out to characterize their radioactive content. For this, a liquid scintillation measurement method was performed to measure the levels of radioactivity within the samples. The findings revealed that the water from the selected brands adheres to the radioactivity limits established in the aforementioned Mexican standard.

FIGURE 1. Liquid scintillation system Hidex® 300 SL.

2. Methodology

2.1. Instrumentation

The measurements were conducted using the Hidex® model 300 SL low-background liquid scintillation system (LS) provided by the Environmental Radiological Surveillance Laboratory (LVRA) (Fig. 1). This system comprises a set of three photomultiplier tubes. The counting system is operated with its dedicated software, `MikroWin` [6].

The characteristics and performance of the system are as follows.

The system's features and performance are outlined below:

a) Sample capacity, 20mL/7mL: 40/96.

b) Counting efficiency ^{3}H/^{14}C:70/96 (per cent).

c) Background ^{3}H in water (CPM): 12.

d) Dimensions, W/H/D (cm): 52/68/63.

e) Weight (kg): 125.

Counting efficiency

a) For ^{3}H and typical > 96 (per cent) for ^{14}C with unquenched samples.

b) > 35 (per cent) for 3H quenched (8 mL water sample + 12 mL "AquaLight cocktail").

c) Alphas: (^{210}Po, ^{234}U/^{238}U, ^{241}Am, ^{222}Rn, ^{226}Ra) > 95 (per cent).

Typical background

$<$ 3 CPM with 8 mL water + 12 mL "AquaLight cocktail" Low Level cocktail. *Background value measured using window with 25%.

Counting efficiency

< 0.3 CPM for alphas (with a/b separation option).

2.2. Sampling

Twenty distinct brands available in the Mexican market, predominantly in the country's central region, were chosen. Three different one-liter bottles were tested for each selected brand.

From each of the one-liter bottles 200 mL of water were extracted for concentration, counting and analysis [7].

2.3. Optimization of LSD counting conditions

The operational parameters of the liquid scintillation (LS) system were obtained following the manufacturer's guidelines to ensure effective alpha-beta separation (Fig. 2). In these systems, the distinction between alpha and beta regions is achieved through pulse duration, in contrast to other systems where discrimination is based on pulse height.

Alpha-beta (AB) calibration was performed using an alpha-active standard source solution as a reference. The ini-

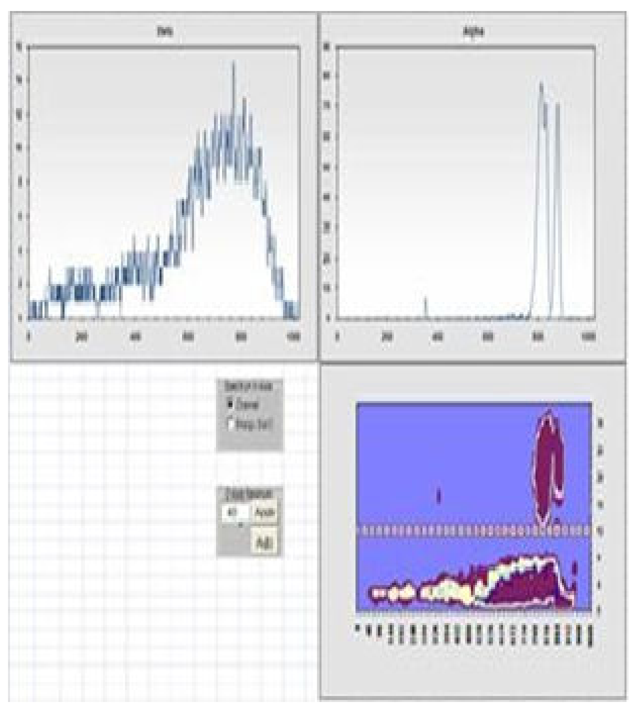

FIGURE 2. Alpha beta Discrimination in Hidex® System.

TABLE I. Results of gross alpha beta measurements in different commercial brands of bottled water.

Commercial Brand	Gross alpha	Error %	Gross beta	Error %
1 Fiji	4.0E-01	±6.0	< 4.2E-01	
2 Evian	2.3E-01	±7.0	3.6E-01	±9.0
3 Liverpool	3.7E-01	±6.0	4.3E-01	±9.0
4 Great Value	< 2.7E-01		< 2.9E-01	
5 Bonafont Sport	< 1.6E-01		3.1E-01	±8.0
6 Voss	< 1.9E-01		< 2.5E-01	
7 Wi on	< 1.8E-01		< 2.1E-01	
8 Bioleve	< 1.9E-01		< 2.1E-01	
9 Ciel	< 1.9E-01		< 2.3E-01	
10 Members Mark	< 1.9E-01		< 1.9E-01	
11 Santa María	< 1.6E-01		< 2.1E-01	
12 Gerber	< 1.5E-01		3.7E-01	±9.0
13 Skarch	< 1.5E-01		< 1.9E-01	
14 E-pura	< 2.4E-01		< 2.5E-01	
15 Bonafont	< 2.1E-01		< 2.5E-01	
16 We are w. people	2.8E-01	±6.0	4.0E-01	±9.0
17 Vitawa	< 1.5E-01		< 1.7E-01	
18 Zoe	< 2.0E-01		< 2.5E-01	
19 Happy baby	< 1.5E-01		4.5E-01	±8.0
20 Bluebay	< 1.5E-01		< 1.7E-01	

tial alpha/beta parameters set by the manufacturer are documented in the Quality Assurance (QA) report accompanying the instrument. They can serve as the baseline for the optimization of the calibration process.

The Pulse-Shape Discrimination (PLI) optimization was obtained by the 2D plot generated by MikroWin® acquisition software, as depicted at the bottom of Fig. 2. Calibration was shown to be successful as the PLI line was appropriately positioned between the beta belt and alpha islets.

2.4. Method of measurement

Measurements were made based on the ISO 11704 Standard titled “Water quality-Gross alpha and gross beta activity-Test method using liquid scintillation counting” (2018) [7]. The samples were taken by concentration through evaporation; reducing from 200 to 20 grams of water, further introduced into a 20 mL low-background vial. To this, 8 grams of the water sample and 12 mL of scintillator cocktail were added.

The utilized cocktail for this experiment was “Ultima Gold TM AB” from PerkinElmer. The efficiency applied in the measurement was obtained using the triple-to-double coincidence ratio (TDCR) method, based on a free-parameter model describing the process of light emission and detection in a scintillation counter. Implementing this method requires an LS counter equipped with three photomultiplier tubes (PMT) labeled A, B, and C and suitable coincidence electronics.

With a substantial number of detected events, the triple-to-double coincidence counting rates ratio (T/D) converges towards the ratio of calculated counting efficiencies (ϵ), denoted as $\epsilon T(\lambda)/\epsilon D(\lambda)$, where these efficiencies are dependent on the free parameter λ. Consequently, the experimental information allows for the derivation of the free parameter, enabling the calculation of the counting efficiencies [8].

3. Results

The samples underwent a counting duration of 3600 seconds to achieve a minimum detectable concentration (MDC) [9], equivalent to at least 50% of the limit values stipulated in the Mexican standard (0.5 Bq/L for Alpha and 1.0 Bq/L for Beta). The individual results for each sample of every brand were averaged when at least one of the three samples yielded a value above the minimum detectable concentration (MDC). The highest value among the samples was reported in cases where the reported result was below the MDC.

The results of the measurements of alpha and beta Gross are presented in Table I.

4. Discussion

In every instance, the average values and those falling below the minimum detectable concentration were consistently found to be below the minimum values prescribed by the relevant official Mexican standards. This observation implies that the purification procedures applied to bottled water exert a diminishing effect on the mineral content present in these products. These minerals likely contribute to natural radioactivity’s alpha and beta components in the examined waters.

Water bottle brands exhibiting comparable or lower quantification levels concerning gross alpha include “Bonafont Sport”, “Gerber”, “Skarch”, “Vitawa”, “Happy baby”, and “Bluebay”. Notably, including two baby water brands (“Happy Baby” and “Gerber”) underscores the implementation of highly effective purification processes.

Regarding Beta quantification, the brands with the lowest data were “Vitawa” and “Bluebay”.

It’s noteworthy that “Bonafont”, with its two lines “Bonafont” and “Bonafont Sport” shows minimal variations in alpha and beta quantification.

Interestingly, despite being among the most expensive in this analysis, the “Evian” brand does not demonstrate any additional benefits in terms of alpha or beta quantification when compared to the more affordable water brand, “Members Mark”.

The average values and those falling below the minimum detectable concentration in the analyzed bottled water samples consistently remain within the limits set by the corresponding Mexican official regulations. This observation implies that bottled water production’s purification process contributes to reducing minerals, likely contributing to the alpha and beta components of natural radioactivity in these waters.

It is crucial to emphasize that the radioactivity levels observed in the analyzed samples adhere to established safety standards, marking a positive outcome for the health and well-being of consumers. However, further research and in-depth analysis are essential to elucidate the specific purification processes responsible for mineral reduction and to comprehend how this might affect the long-term quality of bottled water.

A diverse range of results is evident in quantifying gross alpha and beta in coarse waters. Hence, correlating these results with physicochemical parameters is essential to attribute the variations to the presence of minerals.

Notably, the World Health Organization's (WHO) guidelines for gross radioactivity in water are not strict limits; instead, they serve as indicators. If these guideline values are surpassed, further investigation into the involved radionuclides is recommended, and additional studies should be conducted if remediation becomes necessary.

Specifically, if the gross beta value surpasses the recommended limit, the contribution from 40K should be discounted first, and if the recommended values are, still exceeded, additional scrutiny must be done.

In summary, the values defined as limits in Mexican legislation should not be considered absolute limits.

5. Conclusions

The quantification of gross Alpha Beta results consistently in 20 water bottle brands in Mexico was found to fall below the limit values established in the official Mexican standard, aligned with the guideline values recommended by the World Health Organization (WHO). The calculated annual dose to the population remains below 0.1 mSv, signifying a negligible radiological risk for the population consuming from these water brands. From a radiological standpoint, it can be asserted that these water brands are safe for consumption.

Although all commercial waters conform to official Mexican regulations, measuring their physicochemical parameters and considering their origin is imperative to enhance result correlation, especially given the circulation of mineral waters in the market.

Further work for future analysis involves periodic assessments throughout the year to determine if purification methods sustain their efficiency consistently over time and as the environmental conditions change.

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