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Radiological Study of Water for Human Use and Consumption in Rural Areas of the Central Zone of the State of Veracruz, Mexico

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ABSTRACT

A study and spatiotemporal radiological characterization of water for human use and consumption in the main rural populations of the center of the State of Veracruz was carried out, covering 22 municipalities. The objective was to estimate the annual effective dose as a function of the concentration of gross alpha and beta activity. For this purpose, a low background proportional flux detection system calibrated with NIST-traceable radioactive standards was used. Sampling included only wells, springs, and lagoons in rural areas that supply water to these populations. The decision was based on the fact that these do not have a physicochemical treatment and was carried out during the dry and rainy seasons, which became factors of impact on the radiological material. The analysis included the results of 195 samples from 22 municipalities which showed ranges in the gross alpha of 0.052-0.95 BqL^{-1} with a mean of 0.376 ±0.101BqL^{-1} and a gross beta of 0.034-1.48 BqL^{-1} with a mean of 0.389 ±0.108 BqL^{-1}. The comparison of the values obtained with respect to those of other countries and their complement with analysis of variance showed that there was a significant difference, particularly, for the results of gross alpha in the municipality of Alto Lucero de Gutierrez Barrios and gross beta in Nautla and Tecolutla in dry and rainy seasons (at a probability of $p \leq 0.05$ with the Tukey-Kramer HSD statistical test). A correlation between gross alpha and gross beta was also performed with an r of -0.18 and -0.44 in dry and rainy seasons. This means that among the radionuclides, the major sources of beta radiation are uranium and thorium decay series radionuclides. For the determination of gross alpha, the municipalities in the mountainous zones showed lower values of this activity than the municipalities in the coastal zone. Gross alpha activity values of 0.95 ± 0.11 BqL^{-1} were detected in the municipality of Alto Lucero de Gutiérrez Barrios in the locality of Arroyo Agrio, which exceeded the limit of the Official Mexican Standard.

INTRODUCTION

While water is an essential element for the life of all living beings on the planet, it is also a fundamental human right (Rickert et al. 2016). It is estimated that the composition of water on planet Earth is 97.5% saline and only 2.5% corresponds to fresh water; of that 2.5%, almost 30% is groundwater, 70 % is in glaciers and other snow layers, and only less than 1 % is surface water found in rivers, lakes and other bodies of water (CONAGUA 2011). The above confirms that little water is applied for human consumption and use and that it meets the requirements and quality from the sanitary point of view. Including chemical, physical and biological factors; highlighting that radioactivity is a physical factor; taking into account that the monitoring of radioactive contamination has been established in several laws. All of this is a result of the citizenry's interest sparked by related human activity and accidents such as Chernobyl and Fukushima. This highlights the significance of population-wide water and food consumption being monitored for radioactivity.

According to data from the United Nations Scientific Committee (UNSCEAR 2000), the population of the planet receives a per capita radiation dose of 2.8072 mSv, of which 2.4 mSv are due to natural background, and 1.2 mSv are received through inhalation and 0.3 through ingestion. Despite this, studies of measurements made in Germany show that the average annual radiation exposure for children is 0.047

mSv, which is within the defined limits for alpha and beta activity concentration (Obrikat et al. 2004).

Italy presents studies of radioactivity in drinking water with values below the levels recommended by the WHO (Jia et al. 2009), as does Serbia (Jankovic et al. 2012).

In Pakistan, the values of mean concentrations of ^{226}Ra, ^{232}Th, and ^{40}K, were 11.3±2.3, 5.2±0.4, and 140.9±30.6 mBqL^{-1} (Fatima et al. 2007). In Kazakhstan, the calculated effective doses for adults resulting from consumption of the investigated waters are in the range from 1.0 to 18.7 μSv per year. These doses are lower than the WHO and IAEA reference value (100 μSv per year) for drinking water. (Yamamoto et al. 2010). In Tunisia, water analyses were found to be below the reference levels (Gharbi et al. 2010).

In South Africa, the minimum and maximum gross alpha activity obtained was 0.0041 BqL^{-1} and 0.0053 BqL^{-1} respectively, while the minimum and maximum gross beta activity obtained from the water samples was 0.0083 BqL^{-1} and 0.0105 BqL^{-1} respectively. The reference level used in this country of 1.38 BqL^{-1} for gross beta activity (Madzunya et al. 2020).

In Ghana, the radiation levels are within the natural background radiation levels found in the literature (Faanu et al. 2014).

MATERIALS AND METHODS

Study Area

The state of Veracruz measures approximately 71,826 km^2 and has a population of approximately 8,062,579 as of 2020 (INEGI. 2020). West Longitude (98°40'53.40" , 93°36'28.44") , North Latitude (17°08'13.20" , 22°28'18.48") 22 municipalities (Fig. 1).

Water Sampling

The samples were collected in rural areas of the central part of the state of Veracruz A total of 195 samples were collected in 22 municipalities, in dry and rainy seasons, where the different types of soil in the center of the state were considered. This was based on the fact that Veracruz is a state with a great diversity of types of geomorphology since it has been from coastal plains to significant mountains. To correctly locate the points, a Garmin GPS was used (GPSMAP64) and it was corroborated with UTM coordinates for geopositioning. The water was collected from supply sources such as aquifers, springs, and wells, using 3.7 L plastic containers with lids and covers, previously washed and rinsed with distilled water, then dried and marked with the sampling guide. The procedure for collecting the water was carried out according to the standard, fulfilling the following steps: the container was opened and submerged in water, which was allowed to run long enough so that the sample did not come from stagnant water. No reagents were added for the preservation or conditioning of the sample. Each sample was labeled appropriately and was not acidified since its radiological analysis did not exceed more than seven calendar days from the collection to receipt of the sample at the State Public Health Laboratory of Veracruz (Standard methods 2020).

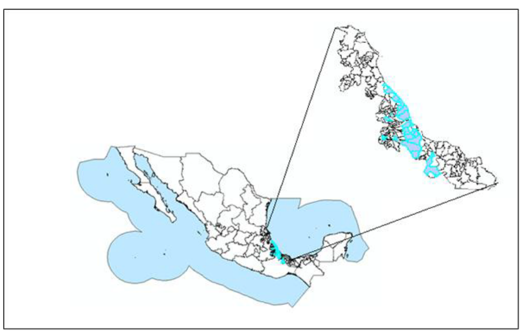

Fig. 1: Municipalities in the study area Veracruz, México.

Test Method

The methods were validated and authorized by the Federal Commission for the Protection of Sanitary Risks (COFEPRIS 2020). With respect to the measurement of conductivity, its value is determined with the volume to be used, its purpose is to control the mass thickness for every 20 cm^2 of counting area, methyl orange indicator solution is added at 0.05% and acidified to pH 4 with 1N nitric acid or at the change of color from orange to cinnamon yellow.

Determination of Gross Alpha and Beta Activities by a Gas Flow Proportional Counter (GFPC)

Gross alpha and beta activity measurements, called the thin source deposition method, were performed as a first step in the radiological characterization of water samples, and this method included the standards.

The procedure is accredited by Federal Commission for the Protection of Sanitary Risks (COFEPRIS. 2020). For gross alpha and beta analyses, 2000 mL of each water sample was evaporated at ≤90 °C (ensuring deposits with a mass thickness of less than 1,48E-2 g.cm^{-2}). The residue obtained was transferred to a stainless steel planchet (2 inches in diameter and 1/8 inch deep). Each planchet was measured for gross alpha and gross beta activity during an interval of 60 or 120 minutes per sample depending on the conductivity of the samples at the state public health laboratory of the State of Veracruz using a Canberra Tennelec low-background. The detector type was a gas flow proportional counter (GFPC) with a mixture of 90% argon and 10% methane (P-10) and an automatic transport of 50 samples. The high operating voltage of the detector was set at 1425 V. The background of each detector was determined by counting empty planchets for 60 min and 120 min.

The detectors were calibrated in 2020 for alpha and beta efficiency using ^{241}Am (11.0 Bq ± 1.44% at 1σ) and ^{90}Sr/^{90}Y (16.3 Bq ± 1.25% at 1σ) standard solution sources, which were supplied by the Eckert & Zeigler Company.

The gross alpha efficiency was 0,0405±2.96 % at 1σ to 0.1740 ± 1.90 % at 1σ, with a mass thickness (3,07 E^{-3}-1,48 E^{-2} g.cm^{-2}) and a mathematical model Ef=0,287 EXP(-139m) where *m* is the mass thickness of the deposit (g/cm^2) while the gross beta efficiency was 0,3322 ± 1,36% at 1σ to 0,3909 ± 1,34% at 1σ, with a mass thickness (3,03 E^{-3}-1,26 E^{-2} g/cm^2) and a mathematical model Ef=0,416 EXP (-17,4 m) where *m* is the mass thickness of the deposit (g/cm^2). The seven solutions were carried out with the same sample preparation procedure analyzed; evaporated on a planchet and counted by the Canberra Tennelec GFPC.

The gross alpha and beta activity concentrations can be obtained as given in Equation (1)

$$A = \frac{cpmt - cpmf}{Ef * V * 60} \quad \ldots(1)$$

Where *A* is the gross alpha activity concentration or the beta activity concentration of the sample (BqL^{-1}); *cpmt* is the gross sample count (alpha or beta); *cpmf* are background counts (alpha or beta); *Ef* is the counting efficiency (alpha or beta); *V* is the sample volume(L); 60 is the Becquerel conversion to disintegrations per minute.

The minimum detectable concentration (MDC) for the gross alpha or gross beta activity concentration is calculated as given in Equation (2) (Currie 1968):

$$\text{MDC} = \frac{4.66\ \sigma\ cpmf}{Ef * V * 60} \quad \ldots(2)$$

Where:

σ = *cpmf* background uncertainty (alpha or beta).

Estimation of the Annual Effective Dose

The annual effective dose (AED) associated with human use and consumption, specifically with the ingestion of water, was not estimated because we worked with a gross alpha and beta activity concentration.

The radiological criteria for water quality in Mexico were established by the Ministry of Health in the NOM-201-SSA1-2015 (2015) and NOM-127-SSA1-2021 (2021), which contemplates gross alpha radioactivity parameters with a limit of 0.5 BqL^{-1} and the gross beta radioactivity parameter with a limit of 1.0 BqL^{-1}. regulations published in the Official Journal of the Federation. These values are 0.5 BqL^{-1} for the gross alpha activity concentration and 1 BqL^{-1} for the gross beta activity concentration according to the IAEA (IAEA 2016) and World Health Organization (WHO 2017). However, the

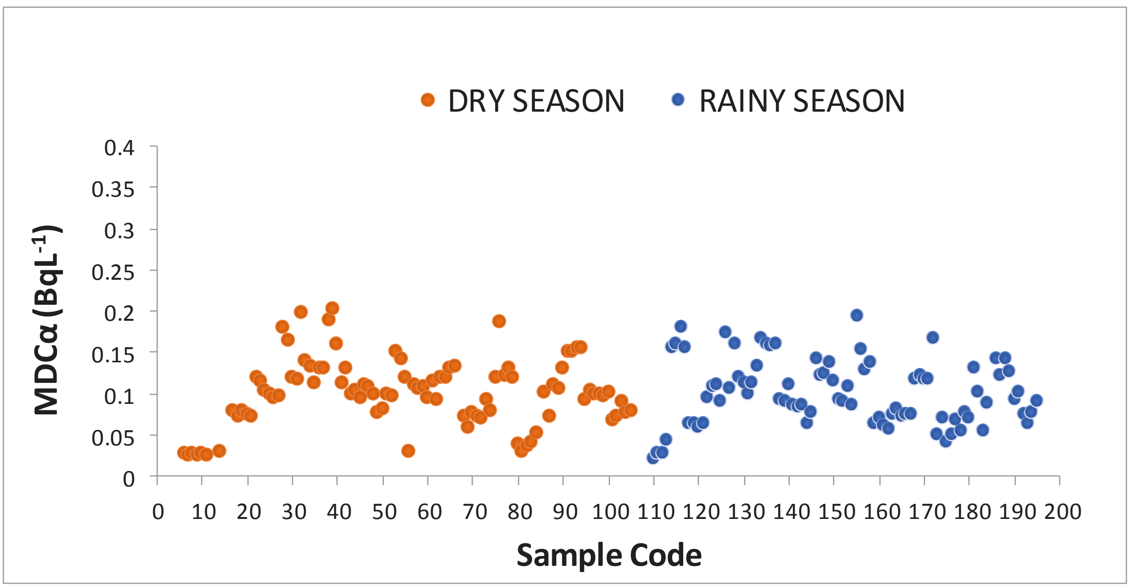

Fig. 2: The minimum detectable concentration of gross alpha in 2020.

annual effective dose value is as stated 0.1 $mSvy^{-1}$ and which is similar to the value recommended by the World Health Organization, WHO.

RESULTS AND DISCUSSION

The minimum detectable concentrations MDCs of gross alpha and beta were calculated according to equation 2, the minimum detectable concentrations for gross alpha were found in a range of 0.025-0.202 with an average value of 0.1009 BqL^{-1} for the dry season and the rainy season in a range of 0.022-0.193 with an average value of 0.1008 BqL^{-1} (Fig. 2 and Fig.3).

The statistical test for comparison of means (t-test) reports that there is no significant difference ($p> 0.05$) for the MDCs of alpha activity in the water samples in the dry and rainy seasons.

In Fig. 2, the values of the minimum detectable concentrations for the water sampled in the dry and rainy seasons for gross alpha and beta are of the order, with the MDC of the gross beta concentration for the dry season being 0.042-0.232 with an average value of 0.1226 BqL^{-1} and for the rainy season in a range of 0.049-0.232 with an average value of 0.1262 $BqL^{-1.}$ According to the statistical test of comparison of means (t-test), the MDCs of beta activity in the water samples during the dry and wet seasons do not differ significantly (p 0.05).

Determination of Gross Alpha and Beta Activity Concentrations

Of the 195 water samples analyzed in the dry and rainy seasons, only 16 samples exceeded the MDC_{α}. In the dry season, only 11 samples presented an alpha activity concentration higher than the MDC_{α} and 5 samples an alpha activity concentration higher than the MDC_{α} in the rainy season; 115 samples recorded a beta activity concentration higher than the MDC_{β}, of which 58 samples presented a beta activity concentration higher than the MDC_{β} in the dry season and 57 samples presented a beta activity concentration higher than MDC_{β} in the rainy season.

Table 1 presents the gross alpha and beta activity concentration values in the dry and rainy seasons, with mean values, range and minimum detectable concentrations, the mean value of alpha activity concentration in the dry season was 0.2409±0.0986 BqL^{-1}; with a range of 0.055-0.91 BqL^{-1} and for the gross beta activity concentration was 0.4208±0.1149 BqL^{-1} with a range of 0.034-1.41 BqL^{-1}, as for the rainy season the mean values for the gross alpha activity concentration were 0.5102 ± 0.1038 BqL^{-1} with a range of 0.052-0.95 BqL^{-1}, and for the mean gross beta activity concentration was 0.3563 ± 0.1008 BqL^{-1}, with a range of 0.025-0.25 BqL^{-1}. The performance of the statistical test (t-test) when performing the comparison of the gross alpha and beta activity concentrations in the dry and rainy seasons did not present a significant difference at a reliability value of ($p >0.05$).

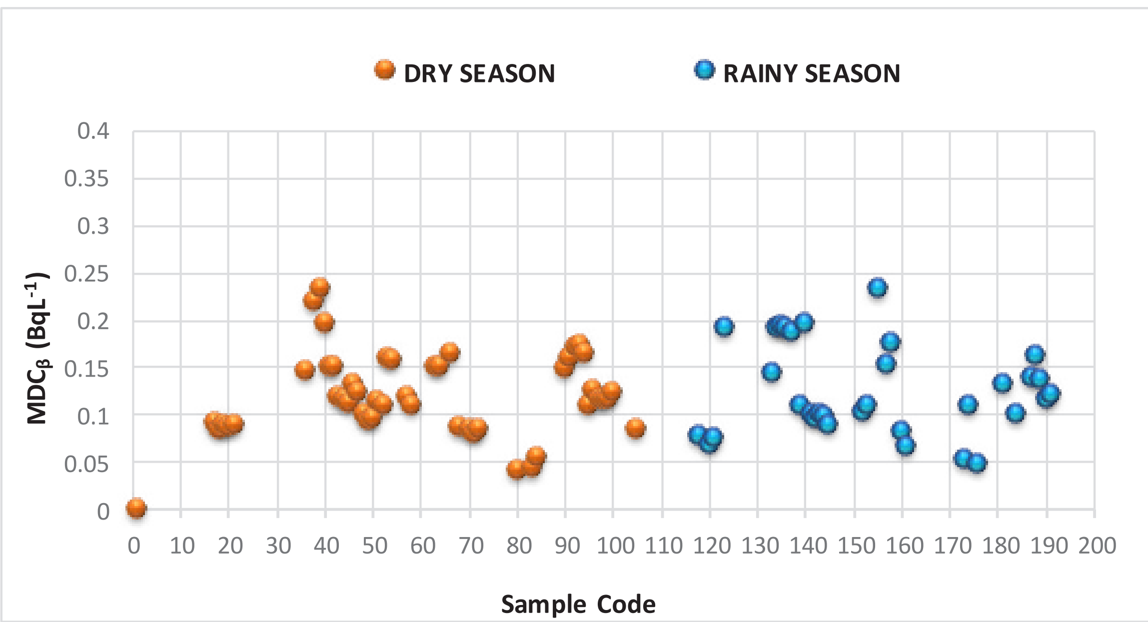

Fig. 3: The minimum detectable concentration of gross beta in 2020.

Although the results of the gross alpha activity concentration in the water samples taken in the municipality of Alto Lucero de Gutiérrez Barrios correspond to 56.25% of the total that was recorded, 45.45% of the samples in the dry season are from this locality, and the gross alpha activity concentration reached 80% with the samples taken in the rainy season. A gross alpha activity concentration of 0.95 BqL^{-1} in the rainy season is reported as a maximum reached, so follow-up measures should be considered at this location. Monitoring should also be carried out in the municipalities of Nautla and Tecolutla because they reached values of interest.

It was demonstrated that none of the samples collected in the dry and rainy seasons in this study reached the limit established by Mexican regulations of 1.0 BqL^{-1} for the gross beta.

Next, a comparative table was made of the mean values recorded from the different sampling sites in the 22 municipalities of the state of Veracruz, resulting in the construction of Table 2 for the determination of the concentration of gross alpha and beta activity. A comparative table of the concentration values of other studies in different countries was also elaborated (Table 3).

Table 1: Mean gross alpha and gross beta activity concentration of the sample with uncertainties.

Dry Season					Rainy Season				
Total Samples 105	Gross Alpha A_α	ΔA_α	Gross Beta A_β	ΔA_β	Total Samples 90	Gross Alpha A_α	ΔA_α	Gross Beta A_β	ΔA_β
Mean	0.2409	0.0986	0.4208	0.1149	Mean	0.5102	0.1038	0.3563	0.1008
Range	0.055-0.91	0.032-0.27	0.034-1.41	0.021-0.32	Range	0.052-0.95	0.03-0.14	0.057-1.48	0.025-0.25

Table 2: Mean values of the gross alpha and gross beta activity concentration in the dry and rainy stations.

Municipality	Latitude	Longitude	Mean Alpha in the dry station	Mean Beta in the dry station	Mean Alpha in the rainy station	Mean Beta in the rainy station
ALTO LUCERO DE GUTIERREZ BARRIOS	19.70641	-96.43159	0.418	0.596	0.625	0.755
BANDERILLA	19.58988	-96.94425	0.027	0.079	0.030	0.091
TECOLUTLA	20.25037	-96.94425	0.063	1.238	0.163	0.530
CAMERINO Z. MENDOZA	18.79346	-97.19874	0.076	0.088	0.063	0.081
COTAXTLA	18.82034	-96.34871	0.105	0.153	0.101	0.180
NAUTLA	20.15704	-96.71706	0.156	1.304	0.140	1.318
JAMAPA	19.03812	-96.22573	0.129	0.167	0.115	0.215
ACTOPAN	19.69853	-96.41605	0.160	0.190	0.161	0.191
VEGA DE ALATORRE	19.96973	-96.60164	0.104	0.121	0.095	0.146
JUCHIQUE DE FERRER	19.79197	-96.67081	0.091	0.103	0.078	0.097
LA ANTIGUA	19.36506	-96.36894	0.111	0.267	0.128	0.190
MEDELLIN	19.01771	-96.13743	0.105	0.127	0.094	0.115
NOGALES	18.82071	-97.16451	0.133	0.203	0.153	0.198
ORIZABA	18.86339	-97.07618	0.070	0.084	0.063	0.087
PASO DE OVEJAS	19.303	-96.439	0.086	0.142	0.075	0.150
TLALIXCOYAN	18.7439	-96.1375	0.136	0.354	0.128	0.398
PLAN DE LAS HAYAS	19.76176	-96.67541	0.040	0.044	0.052	0.068
PLAYA VICENTE	17.83254	-95.81519	0.098	0.482	0.068	0.378
SOLEDAD DE DOBLADO	19.04594	-96.42056	0.115	0.167	0.094	0.124
TIERRA BLANCA	18.63032	-96.15522	0.153	0.168	0.117	0.322
TRES VALLES	18.231	-96.132	0.099	0.118	0.097	0.119
YANGA	18.82543	-96.79071	0.077	0.175	0.077	0.164

Table 3: Gross alpha and gross beta activity concentrations in some countries.

Region/Country	A_α (BqL^{-1})		A_β (BqL^{-1})		References
	Mean	Range	Mean	Range	
Wisconsin/USA	-	0.037-5.320	-	0.0925-1.924	Arndt and West (2004)
Albania	-	0.01-0.126		0.029-0.884	Cfarku et al. (2014)
Saudi Arabia	3.15	-	5.39	-	Alkhomashi et al. (2016)
Balaton/Hungary	-	0.026-1.749	-	0.033-2.015	Jobbagy et al. (2011)
Galati/Romania	0.022	<0.006-0.0852	0.076	<0.025-0.435	Pintilie et al. (2016)
Jordania	-	0.26-3.58	-	0.51-3.43	Alomari et al. (2019)
Katsina/Nigeria	-	0.080-2.300	-	0.120-4.970	Muhammad et al. (2010)
Turkey	0.164	0.007-3.042	0.555	0.021-4.845	Taskin et al. (2013)
TurKey	0.0493	-	0.1284	-	Osmanlioglu et al. (2007)
Southern/Vietnam	0.183	0.024-0.748	0.152	0.027-0.632	Ho et al. (2020)
South Africa	-	0.0041-0.0053	-	0.0083-0.0105	Madzunya et al. (2020)
Veracruz /México	0.376	0.052-0.95	0.389	0.034-1.48	Present study

Annual Effective Dose Assessment for Ingestion of the Water Samples

The radionuclides causing the gross alpha and beta concentration results were not investigated in this work. The radionuclides that promote gross alpha activity in water are mainly due to uranium and its progeny, such as ^{226}Ra. The gross beta activity concentration is probably caused by the contribution of ^{40}K, ^{210}Pb, and ^{228}Ra. The sixteen samples with gross alpha activity concentration above the MDC and the ninety-five samples with gross beta activity concentration were contrasted to determine the strength of correlation between gross alpha and beta activity concentrations, presenting a negative Pearson correlation factor of -0.18 and -0.44 for the dry and rainy seasons respectively, which meant that there is a null correlation and that the radionuclides constituting this gross alpha and beta activity concentration are probably of natural origin such as mentioned by the Vietnamese author Ho et al. (2020) (Fig. 4 and Fig. 5).

During the research, the annual effective dose of the water samples was also evaluated with the respective compliance with national and international regulations. Therefore, the results presented are a reference that will allow an analysis to be made for decision-making, at some point there are significant changes in these values due to the contribution of radioactivity in the environment due to industrial and other anthropogenic activities in principle the great majority is below 0.1 mSv per year corresponding to not exceeding the guide value of the WHO.

CONCLUSIONS

The present work can be used as a reference in the concentration of gross alpha and beta activity today, since the only nuclear power plant in Mexico, Laguna Verde, is located in the state of Veracruz, as well as the different anthropogenic activities that involve the use of radioactive material and that can increase with their contribution the concentration of activity in the studied area.

Future estimates should be carried out in the study area where there were higher concentrations of alpha activity, as is the case of Alto Lucero de Gutiérrez Barrios, and determine the specific activity of its constituents. At this site, it was observed that during the rainy season most of the measurements exceeded the official Mexican standard and the WHO and IAEA levels, and their increases were concentrated forcefully due to runoff phenomena as manifested during the rainy season.

It was possible to achieve all the objectives that were established in this work, as well as to have made the comparison of the results in the dry season and rainy season, complying with the sampling design and statistical analysis that included the variables regarding the dilution factor and location of the samples. This allowed for the internal and external validity of the research.

The performance of the method used by the Radiological Control of the LESP with respect to the WHO recommendations, in this part, by using as indicators the values of 1 BqL^{-1} for gross beta activity and 0.5 BqL^{-1} for gross alpha activity was adequate, complying with the limit requested by the current standard.

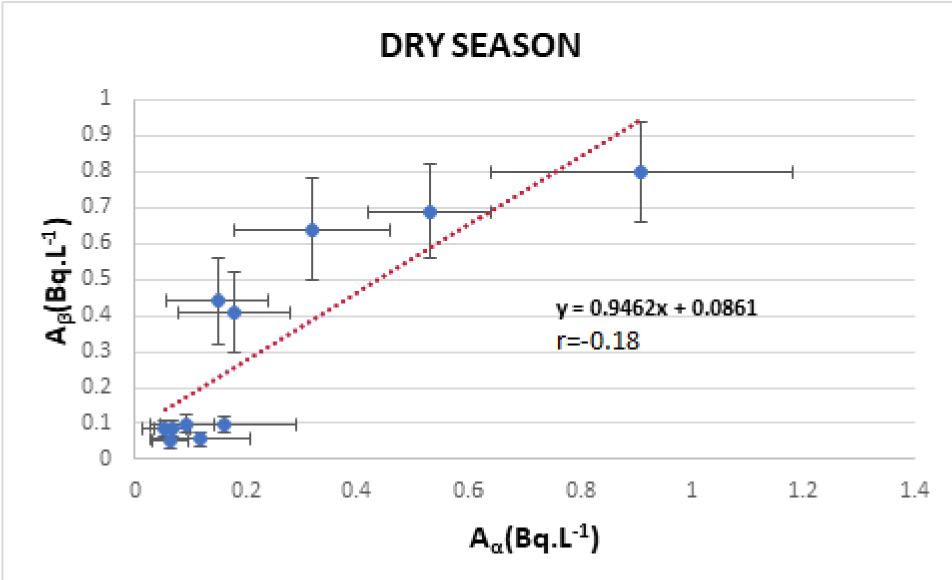

Fig. 4: The correlation function of the gross alpha and gross beta activity concentrations for the samples in the dry season of 2020.

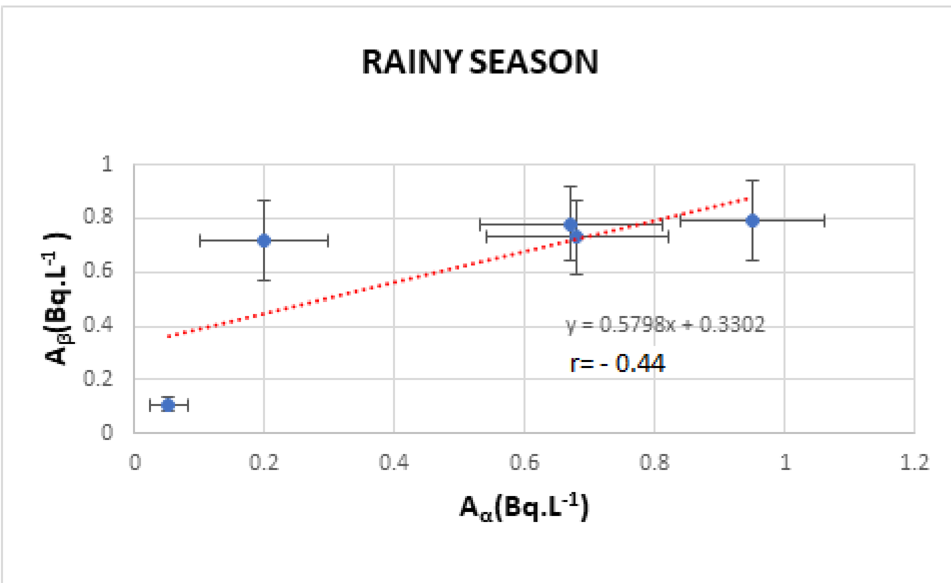

Fig. 5: The correlation function of the gross alpha and gross beta activity concentrations for the samples in rainy 2020.

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