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Gross alpha and beta activities in drinking water from Goiás state, Brazil

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ABSTRACT

Detection of gross alpha and beta radioactivity is important for a quick surveying of both natural and anthropogenic radioactivity in water. Furthermore, gross alpha and gross beta parameters are included in Brazilian legislation on quality of drinking water. In this work, a low background liquid scintillation spectrometer was used to simultaneously determine gross alpha and gross beta in samples of the public water supplies in the state of Goiás, Brazil, during 2010-2015. Sample preparation involved evaporation to concentrate the sample ten-fold. The results indicate that the water meets the radioactivity standards required by the regulations MS 2914/2011 of the Brazilian Department of Health. Concerning the high level of censored observations, a statistical treatment of data was conducted by using analysis methods of censored data to provide a reference value of the gross alpha and beta radioactivity in drinking water from the state of Goiás. The estimated typical activities are very low, 0.030 $Bq.L^{-1}$ and 0.058 $Bq.L^{-1}$, for gross alpha and gross beta, respectively.

Keywords: Water quality, radioactivity, liquid scintillation, censored data

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1. INTRODUCTION

Goiás is located in the Center-West of Brazil and is the most populous state of the region with a population of the about 6.7 million inhabitants (2016) [1]. The state occupies an area of 340,106,492 km^2 and has 246 municipalities and its capital is Goiânia. In 1987, one of most serious radiological accidents in Brazil occurred in Goiânia due to the rupture of the Cs-137 source capsule from the dismantle of a teletherapy unit that was abandoned in a radiotherapy institute partially demolished. It resulted in the death of four people and many others were injured by radiation; it also led to the radioactive contamination of parts of the city [2]. Since the radiological accident, the attention of the population in relation to the sources of radiation and its effects in the health is prominent.

The Brazilian legislation on quality of drinking water MS 2.914/2011 [3] provides the potability of the water considering the physical, chemical and radiological features. In case of radioactivity, the drinking water should be analyzed first for gross alpha and beta activities and the concentration values not exceeding 0.5 $Bq.L^{-1}$ for gross alpha and 1 $Bq.L^{-1}$ for gross beta, according to national and international recommendations [3,4]. If this values are overcome, analysis of present radionuclides should be made and the result will be compared with the reference levels from 1 $Bq.L^{-1}$ to Ra-226 and 0.1 $Bq.L^{-1}$ for Ra-228. Ra-226 and Ra-228 are products of the natural decay of U-238 and Th-232 series, respectively. Despite this regulation, this survey is not carried out routinely throughout Brazil due to the lack of availability of the radioanalysis service by the sanitation companies.

In this study, gross alpha and beta activities in natural waters of Goiás state, which are intended for human consumption, were determined by using liquid scintillation counting (LSC) method [5]. Low-level contaminants data set usually has censored data and multireporting limits, and this is the case of gross alpha and beta values in drinking water. Therefore, the statistical treatment of the data was conducted by using analysis methods of censored environmental data. These combine the values above the detection limit(s) with the information contained in the proportion of data below the detection limit(s), that are the censored data, in order to computing summary statistics [6]. The statistic methods for censored data assume that the data have a particular distribution and the environmental data are commonly modeled using a lognormal distribution.

2. MATERIALS AND METHODS

2.1. Sampling waters

Freshwater and groundwater (323 samples) obtained from 97 different municipalities of Goiás state were analyzed. This represents 39.4% of the cities of the state. As shown in Fig. 1, a wide range of cities distributed into all regions had water samples analyzed for gross alpha and beta activities. The water samples were collected in drinking water abstraction areas by Goiás Sanitation Company (SANEAGO) and also by Health Surveillance and Environmental Agency (SVISA) during the years 2010-2011 and 2013-2015.

Figure 1: *Locations of municipalities from Goiás as for natural water samples for gross alpha and beta activities survey*

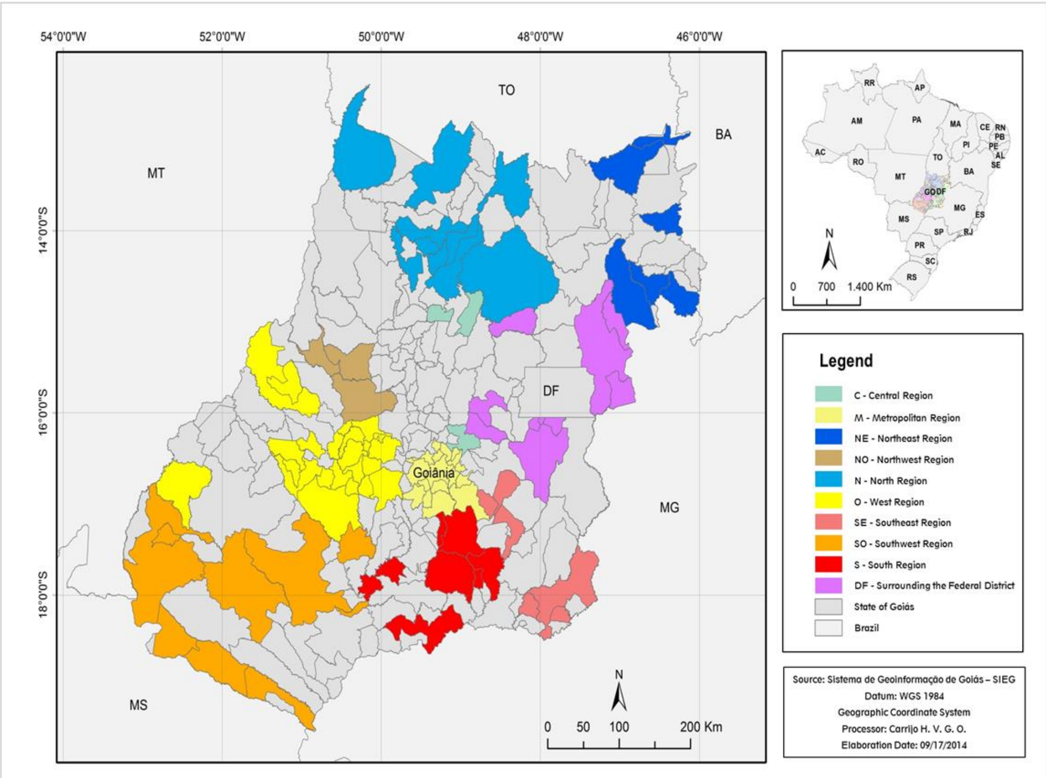
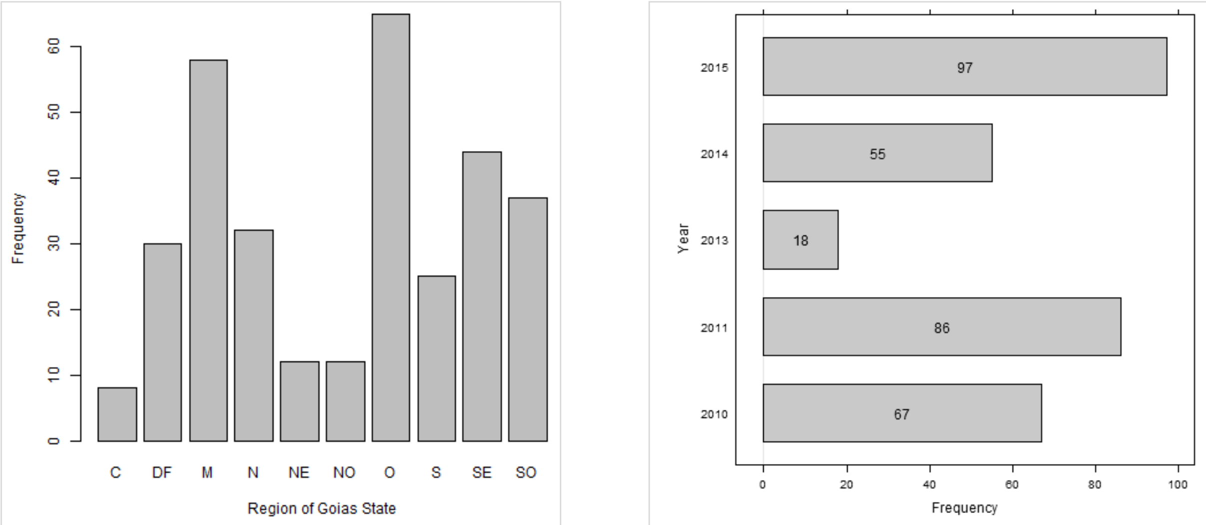


Figure 2: *Frequency of sampling by region and year*

(a) Sampling by region **(b)** Sampling by year



The sampling frequencies by region and year are shown in Fig. 2. To preserve the samples, HNO_3 was added to obtain pH ≤ 2, and the water samples were analyzed until one year from the collecting date [7].

2.2. Analytical Method and Equipments

The gross alpha and beta activities in water were determined by LSC method according to Mingote and Costa (2016) [5] in the Environmental Radioprotection Laboratory of the CRCN-CO/CNEN (Regional Center of Nuclear Sciences of Center-West / Brazilian Nuclear Energy Commission, Abadia de Goiás, Brazil). In summary, an aliquot (100 mL) of water sample was taken and evaporated to dryness on a hot plate in a glass beaker. The residue was dissolved in 8.0 mL of 0.1 mol. L^{-1} HCl and transferred to polyethylene scintillation vial (Perkin Elmer, USA). After then, 12 mL of scintillation cocktail (Optiphase HiSafe 3, Perkin Elmer, USA) were added to the vial, mixed thoroughly and the solutions were stored in the dark, under ambient temperature, for at least 24 hours to allow full decay of luminescence. The samples were counted in a low background liquid scintillation spectrometer (1220 Quantulus, Perkin-Elmer/Wallac, Turku, Finland) for 270 min to measure their gross alpha and gross beta activities. Am-241 and Sr-90/Y-90 were used as calibration standards, which were provided by IRD/CNEN (Radioprotection and Dosimetry Institute/Brazilian Nuclear Energy Commission, Rio de Janeiro, Brazil). The detection limits were calculated according to Currie (1968) [8]. The method is under the quality control through the participation of the laboratory in the National Intercomparison Program (PNI) coordinated by IRD/CNEN [9].

2.3. Statistical analysis

Statistical analyses were carried out using the R language of statistical computing [10]. The Nondetects And Data Analysis (NADA) for R package [11] was used for statistical analyses of the censored data. NADA estimates the mean, median and standard deviation using three methods: Kaplan-Meier, Regression on order statistics (ROS) and Maximum likelihood estimation (MLE) as described by Helsel (2012) [6].

3. RESULTS AND DISCUSSION

Methods specifically designed for handling censored environmental data were applied to data analyses. A summary of the censored data set is shown in Tab. 1. There are 3 different reporting limits (MDA) out of a total of 323 observations, corresponding to high censored datasets, 144 (44.6%) and 166 (51.4%) for gross alpha and gross beta, respectively (Tab. 2). The multiple reporting limits (MDA) raised due to changes over time in the methodology to gross alpha and beta activities determination [5].

Figure 3 shows the censored boxplots of the gross alpha and beta activities concentration in water. ROS method is used to estimate the percentiles in boxplots [6]. The horizontal line is drawn at the highest reporting limit in the data set. Percentiles above this line are unaffected by censoring.

Fig. 4 shows the censored boxplots grouped by region, the sampling frequency is shown in Fig. 2. In mostly of the regions, the median for gross alpha and beta values are lower than the respective maximum detectable activities. A summary of the censored data set grouped by region is shown in Tab. 2. Most of the water samples showed concentration activities below than the directive MS

2914/2011 [3], i.e. below 0.5 Bq.L^{-1} and 1.0 Bq.L^{-1} for gross alpha and beta, respectively. No sample presented higher value than directive for gross beta activity.

Table 1: Summary of censored data of gross alpha and beta activities (Bq) in drinking water from Goiás

Activity	MDA	n.cen	uncen	pexceed
Gross Alpha	0.01	55	1	0.560
	0.02	82	7	0.552
	0.03	7	171	0.529
Gross Beta	0.04	41	2	0.534
	0.06	82	38	0.511
	0.10	43	117	0.362

MDA = Minimum detectable activity, n.cen= quantity of censored data, uncen= quantity of uncensored data, pexceed= percent chance of exceedance the MDA.

Estimate for the median, mean and standard deviation of gross alpha and beta activities data using (lognormal) maximum likelihood estimation, Kaplan-Meier, and the robust ROS methods are shown in the Tab. 3. The recommended methods for estimation of summary statistics consider the amount of available data and the percent censored. Based on the percent of data censored, the sample size and the t to the distribution, Kaplan-Meier and MLE methods were chosen to gross alpha and beta activities, respectively. These estimated the typical values of 0.030 Bq.L^{-1} for gross alpha radioactivity and 0.058 Bq.L^{-1} for gross beta radioactivity in drinking water from Goiás. The values obtained are relatively low and comparable with some literature data from Iran [12], Bulgaria [13], Bosnia and Herzegovina [14], and Turkey [15, 16].

Only 1% (3 samples) of investigated waters had values that overcame the limit of 0.5 Bq.L^{-1} for gross alpha activity, from two sampling points of groundwater. A sample of 2011 presented gross alpha value (0.7 ± 0.1 Bq.L^{-1}), but for a new sampling campaign in 2014, this sampling point had alpha radioactivity of 0.20 ± 0.06 Bq.L^{-1}[5]. Another groundwater sampling point had gross alpha values of 1.9 ± 0.4 Bq.L^{-1}, 1.9 ± 0.4 Bq.L^{-1} and 2.4 ± 0.5 Bq.L^{-1} in collections conducted in July, September and October 2015, respectively. New analysis results for this point were not available until the close of this report, however, it is important to emphasize that this is not the only point of abstraction of water for the local population, and that the other points of capture of the city showed values near or below the limit of detection. The characteristic of the alpha spectrum of LSC of these samples suggests the presence of Ra-226 or natural uranium.

Table 2: Summary of censored data of gross alpha and beta activities (Bq.L^{-1}) in drinking water from Goiás as grouped by region

Activity	Region	n	ncen	pct.cen	min	max
	All	**323**	**144**	**44.6**	**0.01**	**2.40**
	C	8	1	42.5	0.02	0.70
	DF	30	7	23.3	0.01	0.32
	M	58	38	65.5	0.01	0.24
	N	32	13	40.6	0.01	0.45
Gross Alpha	NE	12	5	41.7	0.02	0.08
	NO	12	5	41.7	0.01	2.40
	O	65	30	46.2	0.01	0.19
	S	25	16	64.0	0.01	0.17
	SE	44	12	27.3	0.01	0.31
	SO	37	17	45.9	0.01	0.16
	All	**323**	**166**	**51.4**	**0.04**	**0.77**
	C	8	1	12.5	0.07	0.60
	DF	30	12	40.0	0.06	0.70
	M	58	42	72.4	0.04	0.20
	N	32	7	21.8	0.06	0.54
Gross Beta	NE	12	8	66.7	0.04	0.21
	NO	12	4	33.3	0.06	0.75
	O	65	40	61.5	0.04	0.51
	S	25	14	556.0	0.04	0.29
	SE	44	15	34.1	0.04	0.77
	SO	37	23	62.2	0.04	0.31

n= quantity of data, n.cen= quantity of censored data, pct.cen= percentage of censored data, min= minimum value of detectable activity, max= maximum value of detectable activity

Figure 3: *Censored plots for gross alpha and beta activities in drinking water from Goiás*

(a) Gross alpha activity (Bq.L^{-1}) **(b)** Gross beta activity (Bq.L^{-1})

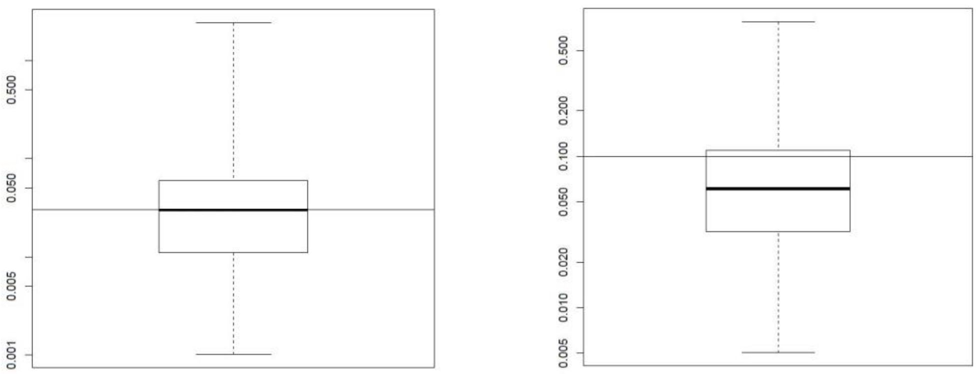


Figure 4: *Censored plots for gross alpha and beta activities in drinking water from Goiás grouped by region*

(a) Gross alpha activity (Bq.L^{-1}) **(b)** Gross beta activity (Bq.L^{-1})

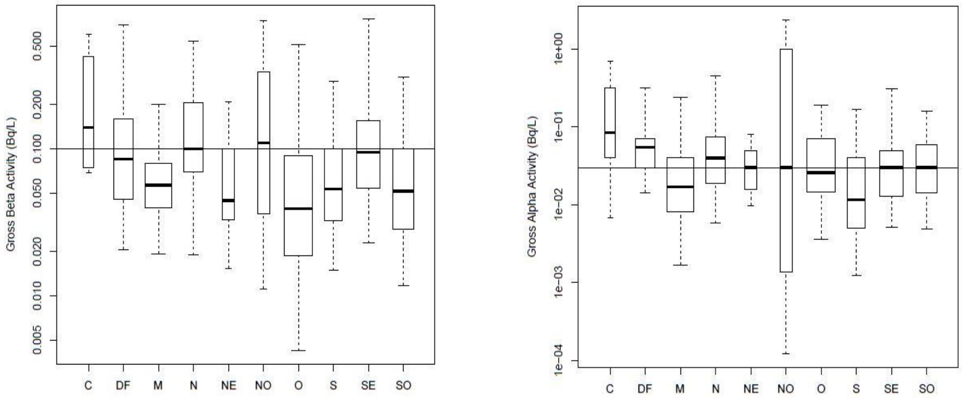


Table 3: Estimates for gross alpha and beta activities (Bq.L^{-1}) in drinking water from Goiás

Activity	Censored method	Median	Mean	sd
Gross Alpha	**Kaplan-Meier**	**0.030**	**0.068**	**0.21**
	ROS	0.030	0.068	0.21
	MLE	0.021	0.068	0.21
Gross Beta	Kaplan-Meier	0.060	0.105	0.12
	ROS	0.061	0.100	0.11
	MLE	**0.058**	**0.103**	**0.15**

sd= standard deviation. ROS= Regression on order statistics. MLE= Maximum likelihood estimation.

4. CONCLUSION

Considering the preoccupation of the local population about the sources of radiation since the radiological accident with Cs-137, and the importance of water for exposure to radiation, the presented study has social importance so as technical relevance. Natural waters coming from different regions of Goiás intended for human consumption were analyzed for gross alpha and beta radioactivities showing that the drinking water distributed by the water treatment of the public system are according the potability standards for radioactivity. It is recommended that the higher gross alpha values samples be analyzed for uranium and Ra-226.

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