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Gross alpha/beta radioactivity and radiation dose in thermal and non-thermal spas groundwaters

Daniel Marcos Bonotto *

Departamento de Petrologia e Metalogenia, Universidade Estadual Paulista (UNESP), Câmpus de Rio Claro, Av. 24-A No.1515, C.P. 178, CEP 13506-900, Rio Claro, São Paulo, Brazil

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A B S T R A C T

Gross alpha and gross beta activities have been determined in thermal and non-thermal spas groundwaters (75) occurring at São Paulo and Minas Gerais states in Brazil as they are ingested in public places, bottled and used for bathing purposes, among other. The samples provided from springs and pumped tubular wells drilled at different aquifer systems inserted in Paraná and Southeastern Shield hydrogeological provinces. The WHO guideline reference value proposed in 2011 for the drinking water quality was never reached for the gross alpha activity (0.5 Bq/L) but it was exceeded in 13 groundwater samples for the gross beta activity (1 Bq/L). Available activity concentration data of the natural radionuclides ^{40}K, ^{228}Ra (^{232}Th-daughter), ^{238}U and descendants (^{234}U, ^{226}Ra, ^{222}Rn, ^{210}Pb, ^{210}Po) allowed calculate the total Committed Effective Dose (CED) based on a drinking water ingestion rate of 2 L/day. The WHO reference level of 0.1 mSv per year for the CED was surpassed in a high number of water sources (62 (83%) or 41 (55%), disregarding radon), denoting the relevance of the radiological surveys detailing as much as possible the dissolved radionuclides present in potable waters, despite the analytical difficulties and costs involved.

1. Introduction

The groundwater quality is associated to the geochemical context of the related aquifers and because some rock-types contain relatively high concentrations of natural radioelements, then, hazard effects into human health may be caused by the ingestion of drinking water providing from wells utilized in water-supply systems (Cantor, 1997; Hopke et al., 2000). Therefore, the radiological characterization of drinking waters has been a topic of concern of several organizations worldwide. For instance, the US Environmental Protection Agency proposed Maximum Contaminant Level (MCL) of 148 Bq/L for ^{222}Rn, 30 μg/L for uranium and 185 mBq/L for combined Ra (^{226}Ra+^{228}Ra) (USEPA, 2000). Among other uses, such standards permitted the data evaluation of hydrogeochemical surveys of fresh groundwater from fractured crystalline rocks in North Carolina (Vinson et al., 2009) and of principal drinking-water aquifer systems of the United States (Szabo et al., 2012).

On the other hand, many countries have adopted the guidance levels proposed by WHO (2011) for ingestion of radionuclides in drinking water. WHO (2011) proposed an effective dose of 0.1 mSv as an annual limiting value based on the ingestion of 2 litres of water per day. WHO (2011) has also recommended that more sophisticated and time-consuming procedures for determining the dissolved radionuclides content only should be adopted when the results of a preliminary screening are positive. In general, for practical purposes, the guidance levels of 0.5 Bq/L for gross alpha and 1 Bq/L for gross beta have been used as maximum activity concentration values to routine operational limits for existing or new water supplies (WHO, 2011).

The radiological criteria for drinking water quality in Brazil are defined by Rule No. 2914 (12 December 2011) of the Health Ministry, which establishes that the identification of the dissolved radionuclides and the measurement of their activity concentration in waters should be only performed when the values found in them are greater than 0.5 Bq/L and 1 Bq/L, respectively, for the gross alpha and beta activity concentration. Thus, these guidance levels are the same of those recommended by WHO (2011).

^{40}K is the only radioactive isotope of potassium and is released to water bodies as a consequence of water/soil-rock interactions (Davis, 1963). Thorium has been considered a highly insoluble element in water due to its presence in minerals of difficult dissolution (Langmuir and Herman, 1980). Uranium tends to be mobile under conditions present at the earth's surface; its concentration generally ranges from 0.1 to 10 μg L^{-1} in rivers, lakes and groundwaters (Fritz and Fontes, 1980; Ivanovich

* Corresponding author.
E-mail address: danielmarcosbonotto@gmail.com.

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Table 1
Gross alpha/beta activities and activity concentration of ^{40}K, ^{222}Rn, ^{220}Rn, ^{226}Ra, ^{228}Ra, ^{210}Po, ^{210}Pb, ^{238}U and ^{234}U in spas groundwaters from southeastern Brazil.

Sample Code-No.	Gross alpha[1] (mBq/L)	Gross beta[1] (Bq/L)	^{40}K[2] (mBq/L)	^{222}Rn[3] (Bq/L)	^{220}Rn[3] (mBq/L)	^{226}Ra[4] (mBq/L)	^{228}Ra[4] (mBq/L)	^{210}Po[5] (mBq/L)	^{210}Pb[5] (mBq/L)	^{238}U[6] (mBq/L)	^{234}U[6] (mBq/L)
ALS-1	<1.0	0.24	150.1	1.4	52	156.6	<9.2	9.69	0.33	0.25	0.89
GIO-2	12.0	0.21	53.0	1.7	<4	323.6	42.5	46.32	0.25	4.22	20.24
JUV-3	62.0	0.24	32.4	0.02	<4	94.0	123.2	5.31	1.02	1.74	4.11
PLA-4	<1.0	0.21	111.3	24.4	270	146.2	<7.4	5.53	0.99	2.98	24.64
POL-5	<1.0	0.20	274.5	4.8	200	219.2	85.8	1.28	0.98	7.44	86.90
VIT-6	2.0	0.34	365.8	53.3	63	281.9	84.2	12.13	0.42	12.90	52.10
BOI-7	4.0	0.30	231.0	74.7	52	62.6	<7.3	0.09	30.38	0.99	4.01
PTA-8	18.0	0.18	127.8	23.2	70	83.5	<6.8	1.62	0.24	0.37	2.25
VIL-9	428.0	0.86	96.0	104.3	226	856.1	1942.5	16.67	4.93	6.57	24.05
PDE-10	2.1	0.72	149.3	5.6	255	208.8	<6.2	0.84	7.31	0.12	0.22
SIL-11	<1.0	0.14	71.4	22.1	15	125.3	10.8	0.28	6.54	0.87	1.84
FIL-12	1.0	0.27	89.3	7.8	200	114.8	<5.4	3.98	0.70	1.61	3.35
BEL-13	<1.0	0.29	62.5	8.3	67	104.4	<6.1	2.36	6.85	2.60	4.22
SER-14	2.0	0.12	121.6	5.0	<4	156.6	<6.8	0.32	5.57	1.61	4.90
COM-15	69.0	0.33	18.1	6.0	15	480.2	6.2	0.22	2.89	1.98	4.52
LIN-16	6.0	0.27	110.5	7.4	226	323.6	<7	43.86	3.49	0.74	2.80
CUR-17	<1.0	0.55	102.1	9.2	89	114.8	128.3	4.62	0.98	0.25	0.37
SJO-18	1.7	0.38	28.7	10.0	7	187.9	44.7	1.56	3.47	0.05	0.08
SCA-19	<1.0	0.52	82.6	10.8	15	177.5	181.1	3.92	5.56	0.12	0.24
ITA-20	19.5	0.40	77.6	5.8	26	156.6	43.9	0.91	34.45	0.25	0.55
SLU-21	<1.0	0.49	32.1	25.0	181	114.8	274.7	61.41	0.38	0.05	0.12
SAT-22	<1.0	0.53	37.1	31.4	240	104.4	<10	53.96	21.71	0.07	0.08
BRU-23	<1.0	0.51	51.3	30.2	418	114.8	<11.3	30.43	0.07	0.12	0.44
LAN-24	<1.0	0.43	26.2	6.2	15	198.4	<9.7	13.82	0.83	0.02	0.05
SAA-25	5.2	0.44	80.6	23.9	26	104.4	5.2	0.14	32.02	0.12	0.32
SBE-26	<1.0	0.47	103.8	10.3	56	187.9	<9.1	9.26	12.15	1.12	1.42
BIO-27	<1.0	0.47	122.8	2.4	<4	146.2	<6.9	0.89	2.94	0.07	0.14
JOR-28	<1.0	0.48	11.7	0.5	<4	104.4	185.0	24.58	1.03	0.25	0.39
ADB-29	<1.0	0.40	10.0	2.7	26	83.5	<6.7	28.18	0.18	4.22	5.99
CGO-30	1.5	0.44	10.9	3.4	26	208.4	<8.8	10.59	0.14	11.66	11.77
SRC-31	<1.0	0.50	9.2	3.6	26	125.0	113.5	4.77	3.50	1.49	1.56
SEI-32	<1.0	0.42	8.1	2.4	<4	167.0	<7.4	4.92	3.29	0.25	0.37
BMU-33	1.8	0.32	85.6	1.3	26	80.0	3.4	5.42	0.28	59.77	130.89
LA1-34	216.0	0.60	156.2	8.2	78	334.1	213.8	35.23	0.54	0.25	0.86
LA2-35	25.9	0.57	138.1	10.3	259	448.9	191	4.00	5.04	1.24	1.43
LA3-36	55.4	0.71	125.0	13.5	237	396.7	237.6	22.77	19.78	0.25	0.41
LA4-37	30.1	0.50	76.7	9.6	81	448.9	112.4	19.22	3.68	0.10	0.26
LA5-38	2.5	0.30	146.2	7.9	26	41.8	106.6	0.28	9.36	0.25	0.77
LA6-39	19.5	1.40	122.8	8.5	44	313.2	118.8	1.59	7.83	1.74	1.96
SL7-40	5.7	0.47	553.5	1.6	22	313.2	139.1	3.86	1.68	0.74	2.20
SL5-41	19.6	0.47	560.8	1.1	48	240.1	448.1	0.67	3.30	0.50	0.67
SL6-42	9.5	0.95	503.6	4.1	41	396.7	708.5	0.60	5.75	1.36	2.11
SL3-43	20.1	0.84	320.8	4.4	78	459.4	575.6	2.23	8.32	0.99	1.30
SL4-44	20.8	0.61	556.9	4.0	33	250.6	176	7.10	0.44	0.50	1.38
SL1-45	9.8	0.55	481.0	0.9	33	271.4	161.8	4.41	2.09	0.37	0.71
SL10-46	117.0	1.66	665.4	4.8	41	647.3	1915.9	1.36	8.74	0.25	0.51
SL9-47	8.4	0.80	488.8	3.6	52	229.7	379.2	6.73	0.60	1.86	1.82
ROR-48	47.1	0.56	53.6	1.5	92	323.6	634.4	26.96	0.63	0.25	0.37
REW-49	37.9	1.14	20.4	4.6	333	177.5	592.6	2.13	4.03	0.10	0.16
CAF-50	6.0	2.09	25.1	3.0	100	208.8	466.5	1.92	4.09	0.05	0.06
FEP-51	51.2	1.65	184.1	13.4	67	177.5	164.9	0.20	30.58	0.12	0.38
MAR-52	<1.0	1.42	364.1	1.6	218	187.9	121.6	0.26	19.83	0.10	0.17
SLI-53	82.5	1.64	53.6	0.8	<4	208.8	479	3.73	3.17	0.12	0.13
GFL-54	28.2	4.78	588.7	3.0	895	1284.1	3544.4	9.28	15.2	0.62	1.15
VEM-55	22.5	5.22	581.7	4.8	314	636.8	3899.1	31.64	0.30	0.05	0.31
MAY-56	12.1	0.62	336.5	35.3	444	407.2	<7.5	93.28	13.94	0.09	0.41
EGU-57	38.2	4.58	595.7	10.1	492	2672.6	46.9	3.40	54.83	0.74	1.43
VIO-58	<1.0	0.51	431.9	45.9	418	281.9	256	119.52	9.62	0.25	0.26
DPE-59	5.8	0.50	459.8	40.7	63	323.6	402.4	27.96	52.73	0.50	0.98
BZA-60	156.0	4.32	704.5	6.9	385	2912.8	1811.8	401.49	2.06	0.74	3.32
DXE-61	<1.0	0.78	599.8	8.0	192	563.8	796.5	4.60	20.13	0.62	0.84
LEO-62	<1.0	1.72	160.4	18.4	81	156.6	276.9	26.95	4.64	7.32	9.80
ISA-63	36.0	1.53	647.3	5.3	148	1106.6	753.8	21.63	16.61	0.25	1.52
QUI-64	<1.0	0.21	249.4	8.4	52	313.2	<6.4	6.82	3.34	2.23	2.81
NOV-65	<1.0	0.28	192.0	16.2	96	219.2	49.1	13.37	10.15	0.37	1.31
MAC-66	9.0	0.31	289.0	2.0	<4	104.2	40.3	19.77	0.56	0.25	0.70
SIN-67	<1.0	0.25	267.3	35.5	522	135.7	9.7	101.81	15.04	0.74	1.32
FRA-68	<1.0	0.17	20.6	23.2	866	240.1	<6.9	11.97	0.16	2.48	3.89
PEB-69	48.1	0.08	310.2	17.6	292	342.9	21.2	4.70	1.55	0.25	0.56
RIV-70	<1.0	0.34	260.3	36.3	81	156.6	<6.6	27.60	18.09	0.12	0.23
SMA-71	<1.0	0.42	292.1	10.4	178	135.7	<7.6	24.16	5.42	0.05	0.21
SJO-72	<1.0	0.39	296.3	41.1	89	156.6	<7.6	5.93	6.80	0.74	1.35

(*continued on next page*)

Table 1 (*continued*)

Sample Code-No.	Gross alpha[1] (mBq/L)	Gross beta[1] (Bq/L)	^{40}K[2] (mBq/L)	^{222}Rn[3] (Bq/L)	^{220}Rn[3] (mBq/L)	^{226}Ra [4] (mBq/L)	^{228}Ra [4] (mBq/L)	^{210}Po [5] (mBq/L)	^{210}Pb [5] (mBq/L)	^{238}U [6] (mBq/L)	^{234}U [6] (mBq/L)
AMO-73	<1.0	0.46	46.9	31.4	15	156.6	<7.8	175.13	7.04	0.01	0.02
DBJ-74	31.0	0.31	246.6	112.5	141	375.8	475.4	7.77	25.79	1.98	3.41
AJU-75	<1.0	0.64	416.0	7.0	26	291.8	588.3	7.79	12.14	0.74	2.22

[1] Analytical uncertainty ±10–15% corresponding to 1σ standard deviation.
[2] Calculated from total K content as reported by Bonotto (2016), using the factor of 27.9 Bq of beta activity per gram of total potassium (WHO , 2011).
[3] Data reported by Bonotto (2014).
[4] Data reported by Bonotto (2015).
[5] Data reported by Bonotto and Oliveira (2017).
[6] Data reported by Bonotto (2017).

and Harmon, 1992). WHO (2011) also defined guidance levels in drinking water for ^{228}Ra (^{232}Th-daughter), ^{238}U and daughters like ^{234}U, ^{226}Ra, ^{210}Pb, and ^{210}Po, whose activity concentration evaluation involves sophisticated and time-consuming procedures and are only performed if the gross alpha and beta measurements exceed the guidance levels of 0.5 and 1 Bq/L, respectively.

Under this perspective, it has been recognized the importance of the disintegration of ^{40}K and a large number of natural radionuclides belonging to the ^{238}U and ^{232}Th decay series. However, because of the analytical and economic constraints, only a few investigations have been conducted focusing the radiological quality of waters in view of an integrated approach. For such purpose, Bonotto and Bueno (2008) and Bonotto (2011) reported a large radionuclides database for major sandstone aquifer systems of the Paraná sedimentary basin, South America. Such types of aquifers have been recognized of importance on the generation of enhanced radioactivity levels worldwide mainly due to ^{222}Rn and radium isotopes ^{226}Ra and ^{228}Ra (Vengosh et al., 2009).

Despite the importance of the porous flow in sandstone aquifers, water-rock interactions processes taking place in fractured rock aquifers are also relevant for the transfer of natural radionuclides to the liquid phase. This paper describes a novel gross alpha/beta database for groundwaters exploited by thermal and non-thermal spas used for therapeutic and leisure purposes in the Brazilian states of São Paulo (SP) and Minas Gerais (MG). Radiation dose calculation has been also done taking into account the ^{40}K, ^{228}Ra, ^{238}U, ^{234}U, ^{226}Ra, ^{210}Pb, and ^{210}Po activity concentration data obtained from previous studies held by Bonotto (2014, 2015, 2016, 2017) and Bonotto and Oliveira (2017). The acquired gross alpha/beta and Committed Effective Dose (CED) databases in the sedimentary/fractured rock aquifers allowed perform a comparative evaluation of the performance of both indices for assuring the radiological quality of the waters.

2. Materials and methods

The groundwater samples (75) in this study were taken from the same springs and pumped tubular wells reported by Bonotto (2014, 2015, 2016, 2017) and Bonotto and Oliveira (2017). The water sources occur in different geological contexts associated to distinct aquifer systems in the Paraná and Southeastern Shield hydrogeological provinces (Mente, 2008) at the following spas in SP and MG states: Águas de São Pedro (3), Águas da Prata (7), Águas de Lindóia (7), Serra Negra (8), Lindóia (2), Termas de Ibirá (5), Águas de Santa Bárbara (1), Lambari (6), São Lourenço (8), Cambuquira (6), Caxambu (10), Poços de Caldas (6), Pocinhos do Rio Verde (4) and Araxá (2). The codes adopted by Bonotto (2014, 2015, 2016, 2017) and Bonotto and Oliveira (2017) for their identification will be also used in this paper. The hydrochemical database is very suitable for the purpose of this paper as there is a wide range of values for pH (4.2–9.6), redox potential Eh (−159 to +112 mV), electrical conductivity (20–6390 μS/cm), and Total Dissolved Solids (11–2898 mg/L) (Bonotto, 2016).

The analytical procedure reported by Bonotto et al. (2009) was used for providing the measurements of the gross alpha/beta activities in the

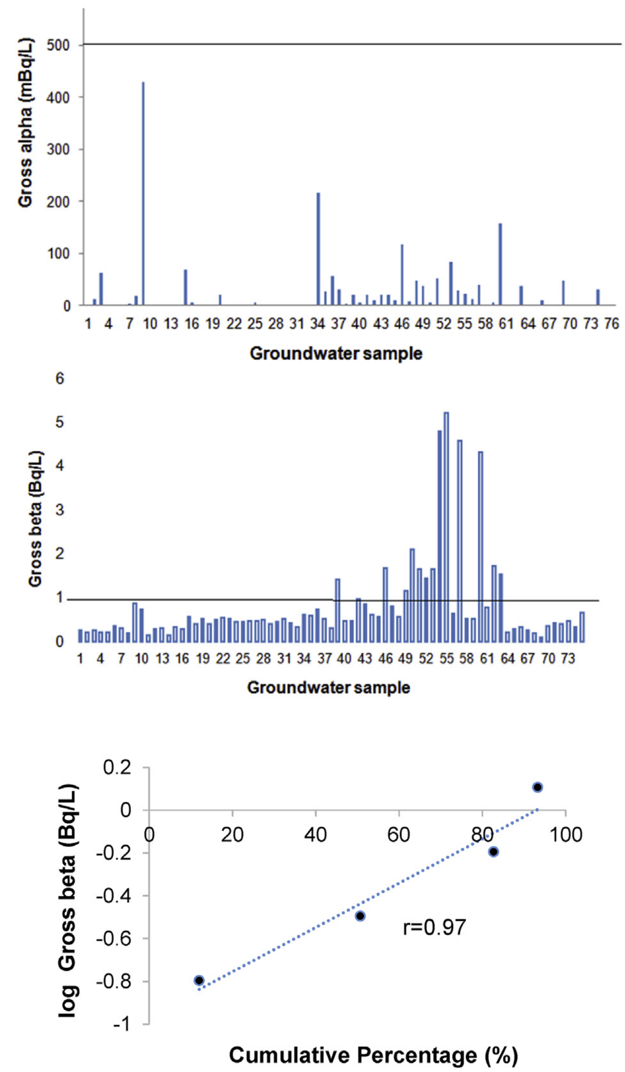

Fig. 1. (top) The gross alpha activity of each spa groundwater from southeastern Brazil, (middle) the respective gross beta activity, and (bottom) the logarithm of the gross beta activity plotted on a probability graph.

waters, which consisted on a combined gamma-alpha spectrometry technique. Each groundwater sample (1 litre) was stored in a polyethylene bottle, filtered through a 0.45 μm Millipore membrane, evaporated until a final volume of 12 mL, inserted in a cylindrical borosilicate glass vial, and submitted to the non-destructive gamma rays spectrometry technique through a 3"×3" well-type NaI(Tl) scintillation detector. The γ-spectrometry allowed the identification and quantification of β⁻-emitters radionuclides. Then, the remaining sample volume of 12 mL was dried in a 1" diameter aluminum can that was inserted in a vacuum chamber for the alpha readings. The alpha spectrometry was based on the

Table 2
Statistical evaluation of the data obtained for gross alpha and gross beta measurements in spas groundwaters from southeastern Brazil.

Range of measured gross alpha values (mBq/L)	Average value (mBq/L)	Frequency	Frequency percentage (%)	Cumulative percentage (%)
0.68–1.87	1.0	34	45.3	45.3
1.87–5.13	2.74	5	6.7	52.0
5.13–14.09	7.52	11	14.7	66.7
14.09–38.69	20.65	14	18.7	85.3
38.69–106.20	56.68	7	9.3	94.7
106.20–291.55	155.61	3	4.0	98.7
291.55–800.36	427.19	1	1.3	100
Range of measured gross beta values (Bq/L)	**Average value (Bq/L)**	**Frequency**	**Frequency percentage (%)**	**Cumulative percentage (%)**
0.06–0.12	0.08	1	1.3	1.3
0.12–0.24	0.16	8	10.7	12.0
0.24–0.48	0.32	29	38.7	50.7
0.48–0.96	0.64	24	32.0	82.7
0.96–1.92	1.28	8	10.7	93.3
1.92–3.84	2.56	1	1.3	94.7
3.84–7.68	5.12	4	5.3	100

direct measurement of the α-particles generated in the ^{238}U decay series. The α-counting was realized with four 0.1 mm depletion depth, 450 mm^2 area, Passivated Implanted Planar Silicon (PIPS) detectors. The counting time was approximately 1 day for each distinct reading. The critical level of detection (L_c) of both techniques has been estimated according to the procedure described by Currie (1968) that is widely used in nuclear spectroscopy. It corresponded to a number of counts of 834, a count rate of 0.008 cps and an activity of 30 mBq for the gross beta measurements, whereas the activity ranged from 0.5 up to 3 mBq (average = 1 mBq) for the gross alpha measurements (Bonotto et al., 2009).

Different methods were used for characterizing the dissolved ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{238}U, ^{234}U, ^{210}Pb and ^{210}Po in the same water sources as reported by Bonotto (2014, 2015, 2016, 2017) and Bonotto and Oliveira (2017). The radon dissolved in water was analyzed on site using RAD7 alpha particles detector coupled to accessory $RADH_2O$ from Durridge Co. The RAD7 utilizes a solid state alpha detector, comprising a Si semiconductor material that converts the energy of the alpha particles into an electrical signal (Durridge, 2009). The ^{226}Ra activity concentration was evaluated from ^{222}Rn readings after waiting at least 25 days for ^{222}Rn to reach radioactive equilibrium with ^{226}Ra (Zereshki, 1983). The ^{222}Rn activity concentration was measured using the device Alpha Guard PQ2000PRO (Genitron GmbH) equipped with an appropriate drive (Aquakit), following the protocol suggested by the manufacturer (Genitron, 2000; Schubert et al., 2006). Two aliquots were used for ^{228}Ra analysis, one chemically processed with addition of the ^{133}Ba radioactive tracer. After following the procedure described in Bonotto (2015), they were submitted to gamma rays spectrometry through a 3"×3" NaI(Tl) well-type scintillation detector that provided readings of the 338 keV and 911 keV ^{228}Ac photopeaks for yielding the ^{228}Ra activity concentration data. Different aliquots of the same groundwater samples were subjected to several radiochemical steps for analysis of ^{238}U, ^{234}U, ^{210}Pb and ^{210}Po, which involved co-precipitation with $Fe(OH)_3$, Fe^{3+} removal, ion exchange in a strong chloride anion exchanger (Dowex 1-X8 resin), ^{210}Po deposition onto copper discs suspended in hydroxylamine hydrochloride + sodium citrate solution placed on hot plate magnetic stirrer, or U-isotopes electrodeposition on stainless steel planchets after 3 hours in a Teflon cell at a current density of 1 Acm^{-2}. Alpha spectrometry with four EG&G ORTEC Model BU-020-450-AS ULTRA-AS Ion-Implanted Detectors with B-Mount allowed quantifying the activity concentration of ^{238}U, ^{234}U, ^{210}Pb and ^{210}Po (Bonotto, 2017; Bonotto and Oliveira, 2017).

3. Results and discussion

Table 1 reports the acquired database for the spas groundwaters focused in this study. The gross alpha activity range was <1–428 mBq/L, with 30 groundwater samples exhibiting an activity concentration value lower than the detection limit of 1 mBq/L. However, none sample exceeded the maximum WHO (2011) guideline reference value of 500 mBq/L (Fig. 1). Table 1 shows that the gross beta activity range was 0.08–5.22 Bq/L, with 13 groundwater samples exhibiting activity concentration that exceeds the maximum WHO (2011) guideline reference value of 1 Bq/L (Fig. 1).

The whole data set for the gross alpha and beta activities was submitted to a statistical treatment, considering class intervals arranged in geometric progression, due to the great variability of the values obtained (Table 2). Lognormal distribution was found for the gross beta database (Fig. 1) as also reported by Bonotto (2014, 2015, 2017) and Bonotto and Oliveira (2017) for other radionuclides (^{222}Rn, ^{226}Ra, ^{238}U, ^{234}U, ^{210}Po and ^{210}Pb) in the same spas groundwaters. The median, mode, and mean values were, 0.33, 0.47, and 0.40 Bq/L, respectively. The gross alpha database did not adjust to the lognormal distribution because of the high frequency of values < 1.0 mBq/L.

3.1. *Major relationships of the gross radioactivity database*

Potassium is a major alkaline element widely distributed in crustal rocks like Ca-enriched granites that may contain up to 2.5% K (Cox, 1991), occurring in various minerals (such as the feldspars orthoclase and microcline) and clays. Weathering processes may cause its dissolution and transfer into the liquid phase where it reacts rapidly and intensely with water, forming a colourless basic potassium hydroxide solution and hydrogen gas (Greenwood and Earnshaw, 2002).

^{40}K is the unique radioactive isotope of natural potassium, occurring in an abundance of 0.0119% from total K (Davis, 1963). ^{40}K decays directly to ^{40}Ca in the ground state through β^- emission (89%) and also to ^{40}Ar in a 1.46 MeV excited state followed by a prompt 1.46 MeV gamma emission through electron capture (11%) (Adams and Gasparini, 1970). As a consequence of water/rock-soil interactions, ^{40}K is released

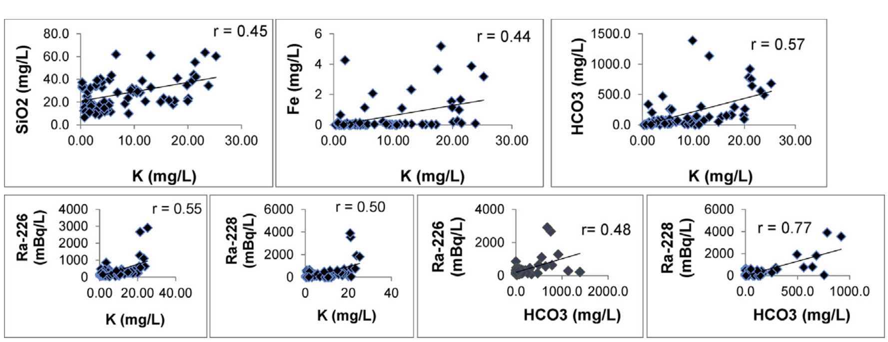

Fig. 2. The dissolved potassium concentration in the spas groundwaters from southeastern Brazil plotted against the dissolved silica, iron, bicarbonate, ^{226}Ra and ^{228}Ra. The relationship of dissolved bicarbonate with the ^{226}Ra and ^{228}Ra activity concentration is also plotted.

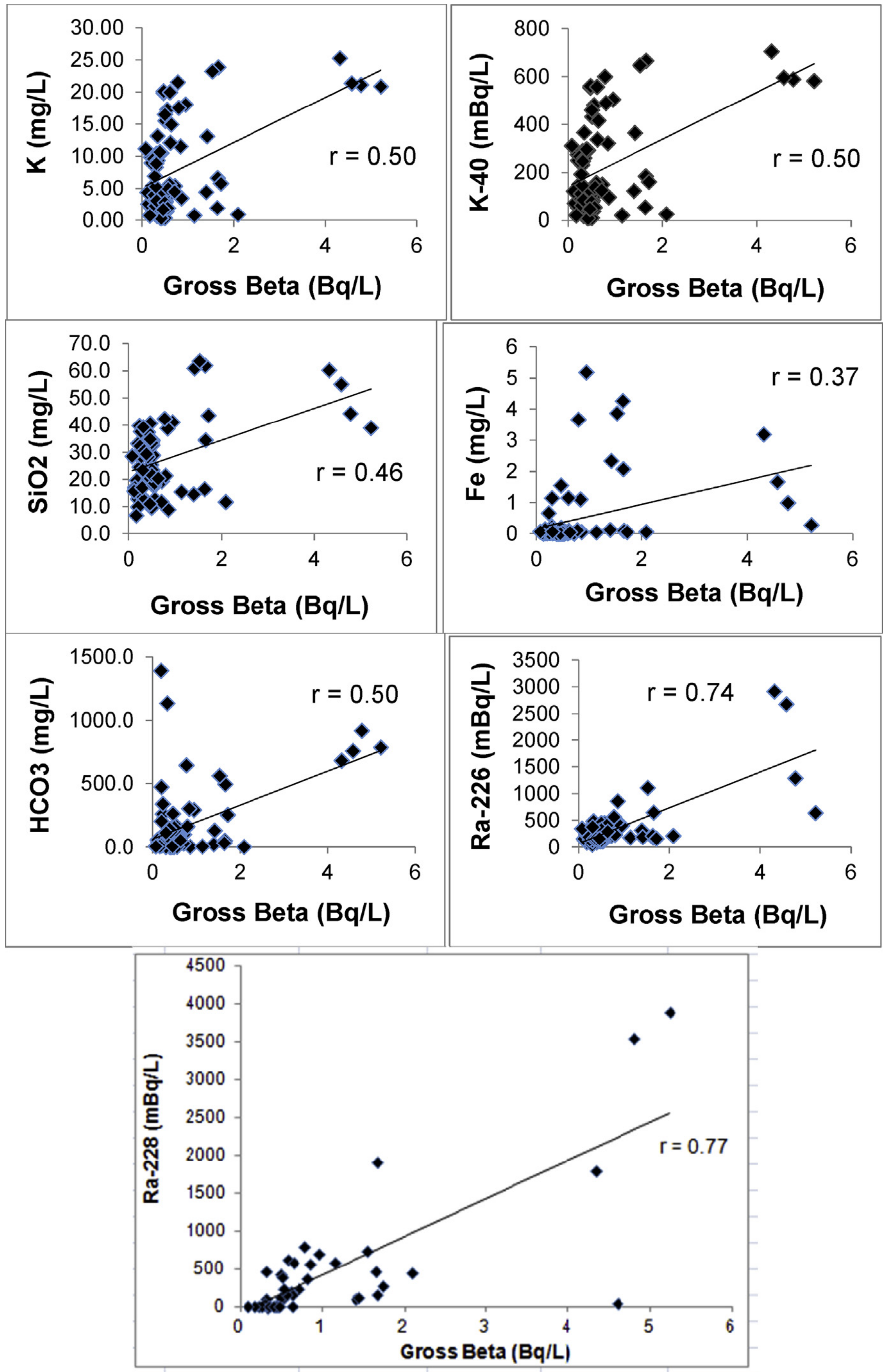

Fig. 3. Plots of the gross beta activity in the spas groundwaters from southeastern Brazil against the dissolved potassium (and ^{40}K activity concentration), silica, iron, bicarbonate, ^{226}Ra and ^{228}Ra.

to water bodies, contributing to the presence of radioactive constituents in drinking water. The chemical isolation of ^{40}K from solution is difficult and the gamma-rays analysis is of low sensitivity to determine the ^{40}K activity concentration in a water sample. Thus, WHO (2011) recommended the chemical analysis of potassium by traditional methods and the use of an appropriate factor to convert the total K to ^{40}K activity concentration, i.e. 27.9 Bq of beta activity per gram of total potassium.

The following parameters in the water sources of this study have been reported and interpreted by Bonotto (2016): temperature, pH, electrical conductivity, redox potential Eh, dissolved gases (O_2, CO_2, H_2S), dry residue (~TDS, total dissolved solids), alkalinity (bicarbonate, carbonate, hydroxide), major cations (Na, K, Ca, Mg), major anions (sulfate, chloride, nitrate, fluoride, phosphate), silica, and iron (total Fe, Fe^{2+}). Statistical tests of correlation between these parameters and the dissolved potassium content in the spas groundwaters indicated significant values for silica (r = 0.45), iron (r = 0.44), bicarbonate (r = 0.57), ^{226}Ra (r = 0.55), and ^{228}Ra (r = 0.50), as shown in Fig. 2. Water-rock/soil interactions affecting micas, feldspars, clays, iron oxides and/or other primary/secondary minerals occurring in the aquifers strata and unsaturated zone in the spas studied would cause the congruent dissolution of K, silica and Fe that are introduced into the liquid phase as very fine colloidal particles and Fe oxyhydroxides, thus, yielding the significant correlation of K with SiO_2 and Fe (Fig. 2).

Bonotto (2016) reported the bicarbonate ions dominance in these

Table 3
Radiation dose due to ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{210}Po, ^{210}Pb, ^{238}U, ^{234}U and respective doses CED and rCED in spas groundwaters from southeastern Brazil. Assuming a drinking water consumption rate = 2 L/day (WHO , 2011). The adopted Dose Conversion Factor (DCF) is reported in footnote.

Sample Code	^{222}Rn [1] (μSv/yr)	^{226}Ra [2] (μSv/yr)	^{228}Ra [3] (μSv/yr)	^{210}Po [4] (μSv/yr)	^{210}Pb [5] (μSv/yr)	^{238}U [6] (μSv/yr)	^{234}U [7] (μSv/yr)	CED (mSv/yr)	rCED (mSv/yr)
ALS	9.8	32.0	Nc	8.5	0.2	0.008	0.03	0.05	0.04
GIO	12.2	66.1	21.4	40.6	0.1	0.14	0.72	0.14	0.13
JUV	0.1	19.2	62.0	4.6	0.5	0.06	0.15	0.09	0.09
PLA	178.3	29.9	Nc	4.8	0.5	0.10	0.88	0.21	0.04
POL	35.1	44.8	43.2	1.1	0.5	0.24	3.11	0.13	0.09
VIT	388.9	57.6	42.4	10.6	0.2	0.42	1.86	0.50	0.11
BOI	545.6	12.8	Nc	0.08	15.3	0.03	0.14	0.57	0.03
PTA	169.6	17.1	Nc	1.4	0.1	0.01	0.08	0.19	0.02
VIL	761.7	175.0	978.4	14.6	2.5	0.22	0.86	1.93	1.17
PDE	41.0	42.7	Nc	0.7	3.7	0.004	0.008	0.09	0.05
SIL	161.2	25.6	5.4	0.2	3.3	0.03	0.06	0.20	0.03
FIL	57.0	23.5	Nc	3.5	0.4	0.05	0.12	0.08	0.03
BEL	60.5	21.3	Nc	2.1	3.4	0.08	0.15	0.09	0.03
SRE	36.2	32.0	Nc	0.3	2.8	0.05	0.18	0.07	0.03
COM	43.5	98.2	3.1	0.2	1.4	0.06	0.16	0.15	0.10
LIN	53.7	66.1	Nc	38.4	1.8	0.02	0.10	0.16	0.11
CUR	67.2	23.5	64.6	4.0	0.5	0.008	0.01	0.16	0.09
SJO	72.6	38.4	22.5	1.4	1.7	0.002	0.003	0.14	0.06
SCA	78.6	36.3	91.2	3.4	2.8	0.004	0.009	0.21	0.13
ITA	42.4	32.0	22.1	0.8	17.4	0.008	0.02	0.11	0.07
SLU	182.6	23.5	138.4	53.8	0.2	0.002	0.004	0.40	0.22
SAT	229.1	21.3	Nc	47.3	10.9	0.002	0.003	0.31	0.08
BRU	220.2	23.5	Nc	26.7	0.04	0.004	0.02	0.27	0.05
LAN	45.1	40.6	Nc	12.1	0.4	0.0008	0.002	0.10	0.05
SAA	174.8	21.3	2.6	0.1	16.1	0.004	0.01	0.21	0.04
SBE	75.1	38.4	Nc	8.1	6.1	0.04	0.05	0.13	0.05
BIO	17.7	29.9	Nc	0.8	1.5	0.002	0.005	0.05	0.03
JOR	3.6	21.3	93.2	21.5	0.5	0.008	0.01	0.14	0.14
ADB	19.8	17.1	Nc	24.7	0.09	0.14	0.21	0.06	0.04
CGO	24.5	42.6	Nc	9.3	0.07	0.38	0.42	0.08	0.05
SRC	26.1	25.6	57.2	4.2	1.8	0.05	0.06	0.11	0.09
SEI	17.4	34.1	Nc	4.3	1.6	0.008	0.01	0.06	0.04
BMU	9.8	16.4	1.7	4.7	0.1	1.96	4.68	0.04	0.03
LA1	59.7	68.3	107.7	30.9	0.3	0.008	0.03	0.27	0.21
LA2	75.3	91.8	96.2	3.5	2.5	0.04	0.05	0.27	0.19
LA3	98.6	81.1	119.7	19.9	10.0	0.008	0.01	0.33	0.23
LA4	69.9	91.8	56.6	16.8	1.8	0.003	0.009	0.24	0.17
LA5	57.5	8.5	53.7	0.2	4.7	0.008	0.03	0.12	0.07
LA6	61.8	64.0	59.8	1.4	3.9	0.06	0.07	0.19	0.13
SL7	12.0	64.0	70.1	3.4	0.8	0.02	0.08	0.15	0.14
SL5	8.0	49.1	225.7	0.6	1.7	0.02	0.02	0.28	0.28
SL6	30.2	81.1	356.9	0.5	2.9	0.04	0.08	0.47	0.44
SL3	32.4	93.9	289.9	2.0	4.2	0.03	0.05	0.42	0.39
SL4	29.4	51.2	88.6	6.2	0.2	0.02	0.05	0.18	0.15
SL1	6.6	55.5	81.5	3.9	1.0	0.01	0.02	0.15	0.14
SL10	35.1	132.3	965.0	1.2	4.4	0.008	0.02	1.14	1.10
SL9	26.0	47.0	191.0	5.9	0.3	0.06	0.06	0.27	0.24
ROR	11.1	66.1	319.5	23.6	0.3	0.008	0.01	0.42	0.41
REW	33.2	36.3	298.5	1.9	2.0	0.003	0.006	0.37	0.34
CAF	21.7	42.7	235.0	1.7	2.1	0.002	0.002	0.30	0.28
FEP	97.5	36.3	83.1	0.2	15.4	0.004	0.01	0.23	0.13
MAR	11.9	38.4	61.2	0.2	10.0	0.003	0.006	0.12	0.11
SLI	5.8	42.7	241.2	3.3	1.6	0.004	0.004	0.29	0.29
GFL	21.8	262.5	1785.3	8.1	7.7	0.020	0.04	2.08	2.06
VEN	34.8	130.2	1964.0	27.7	0.2	0.002	0.01	2.16	2.12
MAY	257.9	83.2	Nc	81.7	7.0	0.003	0.01	0.43	0.17
EGU	73.4	546.3	23.6	3.0	27.6	0.02	0.05	0.67	0.60
VIO	334.9	57.6	128.9	104.7	4.8	0.008	0.009	0.63	0.30
DPE	297.1	66.1	202.7	24.5	26.6	0.02	0.03	0.62	0.32
BZA	50.2	595.4	912.6	351.7	1.0	0.02	0.12	1.91	1.86
DXE	58.1	115.2	401.2	4.0	10.1	0.02	0.03	0.59	0.53
LEO	134.5	32.0	139.5	23.6	2.3	0.24	0.35	0.33	0.20
ISA	38.6	226.2	379.7	18.9	8.4	0.008	0.05	0.67	0.63
QUI	61.3	64.0	Nc	6.0	1.7	0.07	0.10	0.13	0.07
NOV	118.3	44.8	24.7	11.7	5.1	0.01	0.05	0.20	0.09
MAC	14.9	21.3	20.3	17.3	0.3	0.008	0.02	0.07	0.06
SIN	259.0	27.7	4.9	89.2	7.6	0.02	0.05	0.39	0.13
FRA	169.1	49.1	Nc	10.5	0.08	0.08	0.14	0.23	0.06
PEB	128.6	70.1	10.7	4.1	0.8	0.008	0.02	0.21	0.08
RIV	265.2	32.0	Nc	24.2	9.1	0.004	0.008	0.33	0.06
SMA	75.6	27.7	Nc	21.2	2.7	0.002	0.008	0.13	0.05
SJO	299.8	32.0	Nc	5.2	3.4	0.02	0.05	0.34	0.04

(*continued on next page*)

Table 3 (*continued*)

Sample Code	^{222}Rn [1] (μSv/yr)	^{226}Ra [2] (μSv/yr)	^{228}Ra [3] (μSv/yr)	^{210}Po [4] (μSv/yr)	^{210}Pb [5] (μSv/yr)	^{238}U [6] (μSv/yr)	^{234}U [7] (μSv/yr)	CED (mSv/yr)	rCED (mSv/yr)
AMO	229.1	32.0	Nc	153.4	3.5	0.0004	0.0006	0.42	0.19
DBJ	821.1	76.8	239.4	6.8	13.0	0.06	0.12	1.16	0.34
AJU	50.8	59.6	296.3	6.8	6.1	0.02	0.08	0.42	0.37

Nc = not calculated.
[1] DCF = 10^{-8} Sv/Bq (Kendall et al., 1988).
[2] DCF = 2.8×10^{-7} Sv/Bq (WHO, 2011).
[3] DCF = 6.9×10^{-7} Sv/Bq (WHO, 2011).
[4] DCF = 1.2×10^{-6} Sv/Bq (WHO, 2011).
[5] DCF = 6.9×10^{-7} Sv/Bq (WHO, 2011).
[6] DCF = 4.5×10^{-8} Sv/Bq (WHO, 2011).
[7] DCF = 4.9×10^{-8} Sv/Bq (WHO, 2011).

water sources from the use of the Aquachem 4.0 software (Waterloo Hydrogeologic, 2003). Consequently, this hydrochemical facies justifies the significant relationship of HCO_3^- with K, ^{226}Ra and ^{228}Ra (Fig. 2). The importance of (bi)carbonate ions has been already recognized for the formation of aqueous Ra complexes (e.g. Langmuir and Reise, 1985; Encian, 2014; Porcelli et al., 2014; Matyskin, 2016).

^{40}K is a well-known beta-particles emitter, whose activity concentration has been estimated in this paper from the dissolved K concentration in the spas groundwaters (Table 1). In this study, both parameters (K concentration and ^{40}K activity concentration) correlated significantly with the gross beta activity as shown in Fig. 3. Therefore, the significant relationships of K with SiO_2, Fe, HCO_3^-, ^{226}Ra and ^{228}Ra also implied on significant correlations of the gross beta activity with SiO_2, Fe, HCO_3^-, ^{226}Ra and ^{228}Ra (Fig. 3) .

3.2. *Gross radioactivity and radiation dose*

Radiation dose calculations are helpful to integrate the activity concentration data obtained for all natural radionuclides analyzed in the spas groundwaters (Table 1). WHO (2011) proposed a guidance level of 0.1 mSv for the Committed Effective Dose (CED) from 1 year's consumption of drinking water at an ingestion rate of 2 L/day. The adoption of some dose conversion factor (DCF) (IAEA , 1996; WHO, 2011) is required to estimate the CED due to the dissolved radionuclides in waters. WHO (2011) reported the following DCF values: ^{226}Ra = 2.8×10^{-7} Sv/Bq; ^{228}Ra = 6.9×10^{-7} Sv/Bq; ^{210}Po = 1.2×10^{-6} Sv/Bq; ^{210}Pb = 6.9×10^{-7} Sv/Bq; ^{238}U = 4.5×10^{-8} Sv/Bq; ^{234}U = 4.9×10^{-8} Sv/Bq. Despite there is no consensus on the DCF value for ^{222}Rn, the application of a modified ICRP model for the ingestion of ^{222}Rn in water allowed Kendall et al. (1988) suggest DCF = 10^{-8} Sv/Bq that is the value adopted in this paper.

The spas groundwaters in this study have been utilized or directly consumed in springs discharging in the touristic cities of their occurrence. It is a traditional practice of people visiting them to ingest large amounts of waters as they have been considered "good for health" in the common sense. Additionally, by the same reason, the local population prefers utilizing those waters for drinking purposes rather than that provided by the public water-supply systems. Thus, it is reasonable to perform the CED calculation taking into account an annual consumption of 2 litres of water per day.

Thus, considering the activity concentration of each radionuclide dissolved in the water sources as reported in Table 1 and its corresponding DCF value, it is possible estimate a total CED range of 0.04–2.16 mSv/yr (Table 3). The distribution of annual average radiation exposure for the world population indicates that radon is a naturally occurring source whose mean dose is the highest (1.26 mSv) compared to other natural and artificial sources (medical and other) (WHO, 2011). The acquired radionuclides database for the spas groundwaters also indicates that radon takes a major role on the dose estimate as its contribution to the total CED was the highest for 31 water sources (41.3%). It is followed by ^{228}Ra (28 water sources – 37.3%) and ^{226}Ra (15 water sources – 20%), whilst the dissolved ^{210}Po level of ADB spring (Termas de Ibirá – SP) showed the highest contribution to the total CED in this water source.

Fig. 4 shows that there is significant relationship of the CED with the gross alpha and beta activities, highlighting the experimental database and related calculation. Additionally, Fig. 4(c) indicates that the dose guideline reference level of 0.1 mSv/yr established by WHO (2011) was surpassed in 62 water sources (83%) rather than the 13 groundwater samples exhibiting gross beta activity concentration above the maximum of 1 Bq/L (WHO, 2011) as shown in Fig. 1. Such number is very high, denoting failure of the screening tests based on the gross alpha and gross beta readings for indicating some level of risk drinking water due to the presence of dissolved radionuclides. On the other hand, these findings show the relevance of the radiological surveys, detailing as much as possible the dissolved radionuclides present in the potable waters, despite the analytical difficulties and costs involved.

The inclusion of radon on the total CED calculation is a subject that deserves some caution. In the case of bottled waters consumed days or even weeks after the collection of the waters, the dose estimate may not be representative because most of the radon dissolved determined in the *in situ* measurement has decayed. However, the spas groundwaters in this study are not bottled as they have been utilized in homes or directly consumed after their discharge in taps installed in the sites of their occurrence. In such situations, the waters consumption is performed in a short time interval after collection, but, on the other hand, some of this Rn escapes from the waters in the process of handling, aeration and even in the process of the flasks filling. Thus, it is convenient disregard the radon contribution into the doses estimation and the parameter represented by rCED in Table 3 expresses this new calculation. Under this scenario, Fig. 4(d) indicates that the dose guideline reference level of 0.1 mSv/yr established by WHO (2011) was surpassed now in 41 water sources (55%) rather than 62 water sources (83%). Despite the decrease, this number is still high, indicating that even without radon, the other analyzed radionuclides dissolved in the natural spas groundwaters yielded dose values above 0.1 mSv/yr in a significant number of water sources.

4. Conclusion

Groundwater samples (75) of spas groundwaters from São Paulo (SP) and Minas Gerais (MG) states, Brazil, have been analyzed in terms of the gross alpha/beta activity and natural radioactivity due to ^{40}K and radionuclides belonging to the ^{238}U and ^{232}Th decay series. The gross alpha activity range was <1–428 mBq/L, with 30 groundwater samples exhibiting an activity concentration value lower than the detection limit of 1 mBq/L. None water source exceeded the maximum guideline reference value of 500 mBq/L established by the WHO in 2011. Lognormal distribution was found for the gross beta database, yielding median, mode, and mean values of 0.33, 0.47, and 0.40 Bq/L, respectively. The gross beta activity range was 0.08–5.22 Bq/L, with 13 groundwater samples exhibiting activity concentration that exceeds the maximum WHO guideline reference value of 1 Bq/L. In such cases, it is highly recommended not to drink the waters. The dissolved K

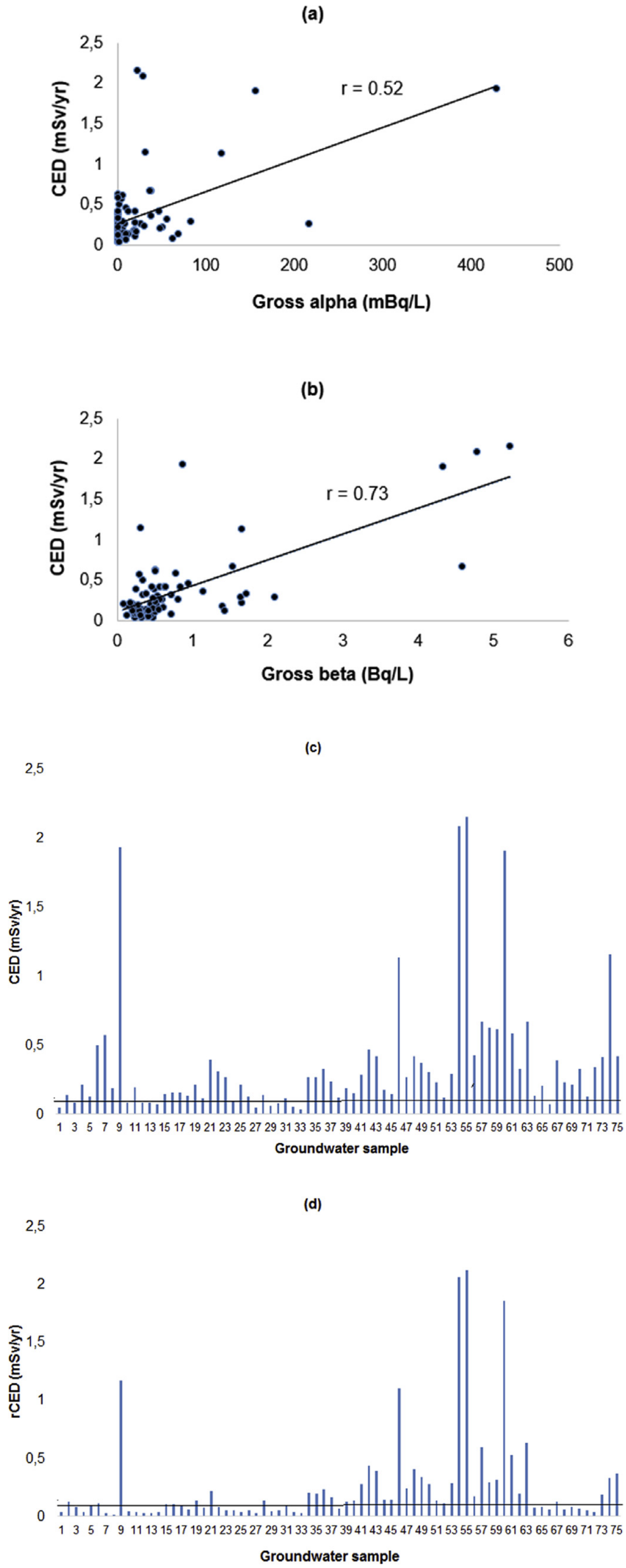

Fig. 4. Plots of the total Committed Effective Dose (CED) against (a) the gross alpha activity, and (b) the gross beta activity in the spas groundwaters from southeastern Brazil. The CED and rCED values are compared in (c) and (d), respectively, with the WHO (2011) guideline reference limit of 0.1 mSv/yr.

concentration (and ^{40}K activity concentration) in the spas groundwaters correlated significantly with the gross beta activity, which also exhibited significant relationships with SiO_2, Fe, HCO_3^-, ^{226}Ra and ^{228}Ra due to congruent dissolution of K, silica and Fe from the aquifers strata that are introduced into the liquid phase as very fine colloidal particles and Fe oxyhydroxides. Radiation dose calculations allowed integrate the activity concentration data obtained for all natural radionuclides analyzed in the spas groundwaters, permitting determine a total Committed Effective Dose (CED) range of 0.04–2.16 mSv/yr. Radon took a major role on the dose estimate as its contribution to the total CED was the highest (41.3%), followed by ^{228}Ra (37.3%), ^{226}Ra (20%), and ^{210}Po (1.3%). Despite CED correlated significantly with the gross alpha and beta activities, the dose guideline reference level of 0.1 mSv/yr established by WHO was surpassed in 62 water sources (83%) that is a number much higher than that above the screening gross beta value of 1 Bq/L. Such number decreased to 41 water sources (55%) when the radon contribution was disregarded in the doses calculation. Therefore, detailed radiochemical analysis revealed more level of risk in drinking water rather than traditional gross alpha and beta screening tests.

Declarations

Author contribution statement

Daniel Marcos Bonotto: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Competing interest statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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