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Natural radionuclides in drinking water supplies of São Paulo State, Brazil and consequent population doses

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Abstract

The activity concentrations of ^{228}Ra, ^{226}Ra and ^{222}Rn have been analysed in 452 drinking water supplies of São Paulo State. This study started in 1994 and covered 54% of the 574 existing counties. Concentrations up to 235 and 131 mBq l^{-1} were observed for ^{226}Ra and ^{228}Ra, respectively, whereas ^{222}Rn concentrations reached 315 Bq l^{-1}. Radiation doses up to 0.3, 0.6 and 3.2 mSv yr^{-1} were estimated for the critical organs, for the ingestion of ^{226}Ra, ^{226}Ra and ^{222}Rn, respectively. The corresponding committed effective doses reached values of 6×10^{-3}, 2×10^{-2} and 3×10^{-1} mSv yr^{-1}, for the same radionuclides. These results indicate that ^{222}Rn makes the highest contribution to the total effective dose.

Keywords: Radioactivity; Radium; Radon; Drinking water; Brazil; Radiation dose

1. Introduction

Measurements of natural radioactivity in drinking water have been performed in many parts of the world, mostly for assessment of the doses and risk resulting from consuming water. It was observed that most natural radioactivity present in

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groundwater consists of ^{222}Rn, which varies over a wide range, depending markedly on its origin. The second source of natural radioactivity in drinking water is ^{226}Ra (UNSCEAR, 1993; WHO, 1993).

In Brazil, few data are available concerning the occurrence of natural radionuclides in public water supplies. The only data related to natural radioactivity levels in drinking water already published are concerned with mineral springs and bottled mineral waters (Szikszay & Sampa, 1983; Lauria & Godoy, 1988; Pires do Rio, Godoy and Amaral, 1988; Oliveira, Moreira and Mazzilli, 1994; Mazzilli et al., 1998; Oliveira, Mazzilli, Sampa and Silva, 1998a). The natural radionuclides most studied were ^{226}Ra, ^{228}Ra, ^{222}Rn and ^{210}Pb, because they deliver the highest doses to humans after their intake via ingestion of drinking water.

The recommendations of the Guidelines for Drinking-Water Quality, published by the World Health Organisation (WHO, 1993), can be used to guide the competent authorities in determining whether the water is of an appropriate quality for human consumption. According to this guideline:

- The recommended reference level of committed effective dose is 0.1 mSv from 1 year's consumption of drinking water. This reference level represents less than 5% of the average effective dose attributable annually to natural background radiation.
- Below this reference level of dose, the drinking water is acceptable for human consumption and any action to reduce the radioactivity is not considered necessary.
- For practical purposes, the recommended guideline activity concentrations are 0.1 Bq l^{-1} for gross-alpha and 1 Bq l^{-1} for gross-beta activity.

These recommendations apply to routine operational conditions of water supply systems and do not differentiate between natural and man-made radionuclides.

The recommended radioactivity drinking water standards established by the Brazilian authority Ministério da Saúde are the following (Ministério da Saúde, 1990):

- The gross-alpha activity (including ^{226}Ra) should not exceed 0.1 Bq l^{-1};
- The gross-beta activity should not exceed 1 Bq l^{-1}.

If the measured values are above these limits, the Brazilian standards require further analyses to determine the specific radionuclides present in drinking water and their respective concentrations. The results obtained should be evaluated by comparison with the annual limit on intake (ALI) established for each radionuclide in Reference Guide CNEN-NE 3.01 (CNEN, 1988).

Radium and its daughter products are important contributors to natural environmental radiation exposure. Since ingestion is a major pathway, apart from inhalation of radon, for internal irradiation, the measurement of radioactivity in drinking water is relevant in assessing the contribution of these environmental radiation hazards. Radium has two natural isotopes which are of concern in public

water supplies: ^{226}Ra, an alpha-emitter with a half-life of 1622 yr generated through the decay of ^{238}U, and ^{228}Ra, a shorter-lived beta-emitter (half-life 5.7 yr), which is generated directly by ^{232}Th decay. The distributions of ^{226}Ra and ^{228}Ra in water are a function of the Th and U contents in the aquifer, the geochemical properties of the aquifer solids, and the half-lives of each isotope. Radium enters groundwater by dissolution of aquifer solids, by direct recoil across the liquid–solid boundary during its formation by radioactive decay of its parent in the solid and by desorption. The Ra content of surface water is usually very low and standard water treatment methods are known to remove Ra. When humans ingest radium, about 20% is absorbed into the bloodstream. Absorbed radium is initially distributed to soft tissues and bone, but its retention is mainly in growing bone (EPA, 1991a; ICRP, 1993).

Radon is a water-soluble inert gas. Its occurrence in waters is controlled by physical variables such as pressure, temperature, emanation of radon from rocks, as well as by the geochemistry of its parent, ^{226}Ra. The only radon isotope with a long enough half-life to be considered in drinking water is ^{222}Rn, which is the decay product of ^{226}Ra and has a half-life of 3.84 days. High activity of Rn is associated with granite rocks, U minerals, such as uraninite, with tailings from phosphate fertiliser processing and U mines. Water transport rather than diffusion from the matrix rock in most cases governs the movement of Rn in water. Radon-222 transported by water can lead to public exposure from direct water consumption; the stomach wall is the tissue that receives the greatest radiation dose (EPA, 1991b; ICRP, 1993). In Brazil, there is no federal standard for Rn in water, although Lima (1996) published studies on ingestion doses and inhalation doses.

This study determined the activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn in drinking water supplies of São Paulo State, since these radionuclides are considered the most critical natural alpha and beta emitters as far as internal irradiation is considered. In order to evaluate potential health hazards, doses due to ingestion of these waters were estimated to assess the contribution of these radionuclides to public exposure from natural radioactivity.

2. Materials and methods

A map of Brazil showing the political divisions in States is presented in Fig. 1. The three main geological formations of São Paulo State are the sedimentary region of Paraná Basin (where basaltic and sandstone aquifers can be found), the region of Crystalline Complex (including granitic and metamorphic rock aquifers) and the regions covered by Cenozoic sediments (Fig. 2).

The water samples were collected by SABESP — Companhia de Saneamento Básico do Estado de São Paulo, which is the state agency responsible for the water collection, treatment and supply of São Paulo State. Raw water samples were collected at 452 different locations, geographically distributed over 8 regions (Fig. 3), as follows: Vale do Paraıba — IV, Baixo Tietê/Grande — IT, Baixada Santista — LB, Vale do Ribeira — LR, Litoral Norte — LN, Médio Tietê — IM, Baixo Paranapanema — IB and Alto Paranapanema — IA.

Fig. 1. Map of Brazil with the political division in States.

Among the 452 water samples studied, 73% were groundwater supplies and 27% were surface water (reservoirs, streams, rivers and water treatment stations). Four of the groundwater supplies studied presented depths greater than 1000 m, while the remaining presented depths ranging from 49 to 355 m. The sampling program was started in 1994 and covered 54% of the 574 existing counties (IBGE, 1996), corresponding to the public systems managed routinely by SABESP. The sampling procedure was established according to the methodologies recommended by the "Standard Methods for the Examination of Water and Wastewater" (APHA, 1985). For ^{226}Ra and ^{228}Ra determination, 10 l of raw water sample was collected in polyethylene bottles at each site. About 20 ml of HNO_3 (65%) was added, to prevent losses by sorption of the radionuclides in the bottles.

The radiochemical procedure adopted for ^{226}Ra and ^{228}Ra determination is described in more detail in Oliveira (1993). A 3 l water sample was used for the analysis, which was performed in duplicate. The water volume was reduced by evaporation to 1 l, then 20 mg of Ba^{2+} and Pb^{2+} carriers were added as yield tracers. The radiochemical separation was accomplished by addition of 3 M H_2SO_4,

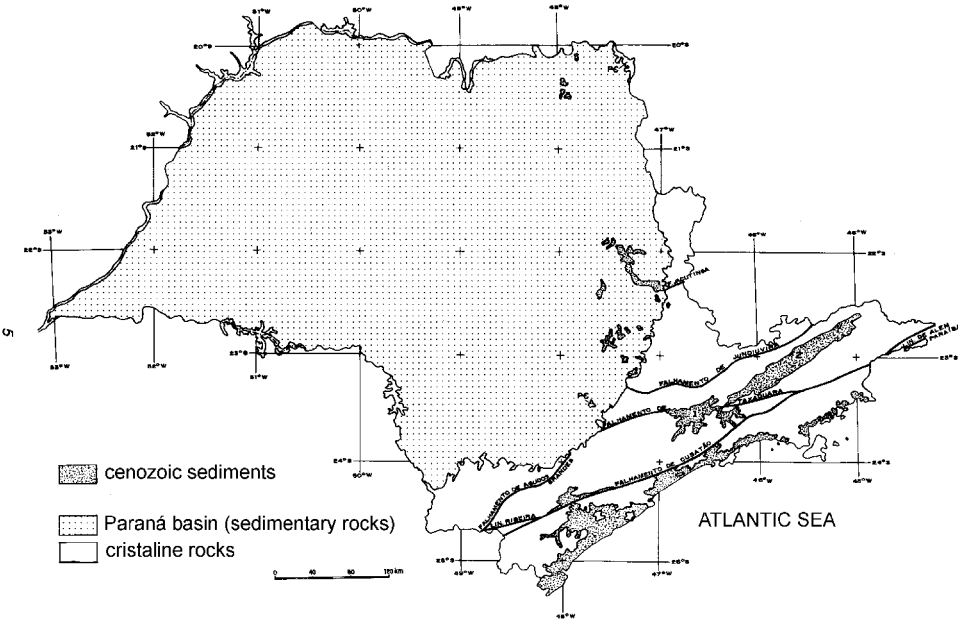

Fig. 2. Main geological formations of São Paulo State (IPT, 1981).

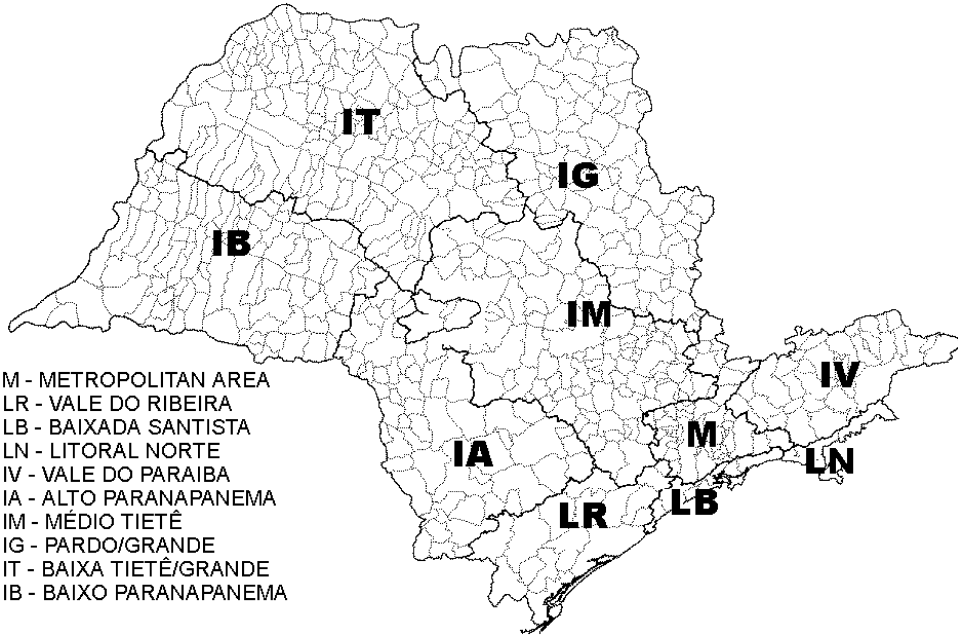

Fig. 3. Political map of São Paulo State with subdivisions in counties, showing the geographic distributions of the regions studied.

under heating. The precipitate of barium, lead and radium sulphate was dissolved with EDTA/NH_4OH and the pH of this solution was adjusted to 4.5–5.0 by the addition of glacial acetic acid. The radium was then co-precipitated as $Ba(Ra)SO_4$, whereas lead remained in solution as a strong complex with EDTA. The $Ba(Ra)SO_4$ precipitate was filtered and the chemical yield determined gravimetrically. Considering that 20 mg of Ba^{2+} carrier was added to the water samples at the beginning of the radiochemical procedure, the total recovery of $Ba(Ra)SO_4$ precipitate should be 34.8 mg. The chemical recovery of $Ba(Ra)SO_4$ precipitate ranged between 85 and 95%. The determination of ^{226}Ra was carried out by measuring the gross-alpha activity of the $Ba(Ra)SO_4$ precipitate using a low-background gas-flow proportional counter, after the decay of ^{224}Ra and of ^{223}Ra; that is, after 21 days. The self-absorption factor for the four alpha particles emitted in secular equilibrium with ^{226}Ra (^{226}Ra — 4.8 MeV; ^{222}Rn — 5.5 MeV; ^{218}Po — 6.0 MeV; ^{214}Po — 7.7 MeV) was determined following the radiochemical procedure described above, by adding to the water samples a ^{228}Ra standard solution, with activity concentrations ranging between 8.7 and 20.3 Bq. The respective value of the self-absorption factor obtained was 0.3001 ± 0.0065. The ^{226}Ra was determined by gross-beta counting of the same precipitate, measuring the activity of its daughter product ^{228}Ac, since it emits beta-rays of higher energy (1.2 and 2.1 MeV) in contrast to the lower energy of ^{226}Ra beta-particles (40 keV). Typical lower limits of detection (LLD) for these methods were 2.2 mBq l^{-1} for ^{226}Ra and 3.7 mBq l^{-1} for ^{228}Ra, at a 95% confidence level (Oliveira, 1998b).

^{222}Rn concentrations were determined by a liquid scintillation method. For this analysis, a volume of 10 ml of raw water sample was carefully transferred directly from the source to the counting vials, in which the same volume of the universal LSC-cocktail Instagel XF had been previously added (Szikszay & Sampa, 1983). The samples were measured in a liquid scintillation analyser, where the spectrum of ^{222}Rn and daughter products were determined. Typical lower limit of detection (LLD) for ^{222}Rn determination was 1.9×10^{-1} Bq l^{-1}, at a 95% confidence level.

3. Results and conclusions

The activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn determined over the 8 regions studied in São Paulo State are presented in Table 1. The concentrations of ^{226}Ra and ^{228}Ra varied from <2.2 to 235 mBq l^{-1} and from <3.7 to 131 mBq l^{-1}, respectively. ^{222}Rn, on the other hand, was found in higher activity concentrations, ranging from 0.40 to 315 Bq l^{-1}. The higher activity concentrations of the radionuclides studied were observed in the Vale do Ribeira — LR region.

The statistical analysis of the results obtained for ^{226}Ra and ^{222}Rn showed a wide dispersion around the mean value (coefficient of variation of 256%), whereas the ^{228}Ra mean activity concentration did not present such a large variation (coefficient of variation of 34%). This high dispersion for ^{226}Ra and ^{222}Rn is probably due to the fact that the regions studied cover a large area with distinct aquifer lithologies and consequent differences in radionuclide solubilities and mobilities.

Table 1
Natural radioactivity levels in drinking water supplies of São Paulo State during 1994–1997[a]

Sampling location (n)		^{226}Ra (mBq l^{-1})	^{228}Ra (mBq l^{-1})	^{222}Rn (Bq l^{-1})
IV — Vale do	Range	<2.2–213	13–78	0.96–274
Paraıba (56)	Arithmetic mean	14	30	16
	Geometric mean	4.6	28	6.3
IT —Baixo	Range	<2.2–42	9.1–92	0.87–23
Tietê/Grande (158)	Arithmetic mean	3.2	24	5.6
	Geometric mean	2.0	24	4.8
LB — Vale do	Range	<2.2–3.9	22–38	2.3–5.0
Ribeira (39)	Arithmetic mean	1.9	25	3.1
	Geometric mean	1.7	254	3.0
LR — Vale do	Range	<2.2–235	<3.7–131	0.40–315
Ribeira (39)	Arithmetic mean	27	34	43
	Gelometric mean	5.7	30	7.0
LN — Litoral	Range	<2.2–4.8	21–27	1.2–3.5
Norte (20)	Arithmetic mean	1.2	24	2.1
	Geometric mean	0.9	24	2.0
IM– Médio	Range	<2.2–66	17–64	2.6–24
Tietê (28)	Arithmetic mean	11	30	8.8
	Geometric mean	3.9	28	7.3
IB — Baixo	Range	<2.2–30	18–73	1.3–19
Paranapanema (107)	Arithmetic mean	2.9	26	5.3
	Geometric mean	1.9	25	4.8
IA — Alto	Range	<2.2–74	20–61	1.8–135
Paranapanema (107)	Arithmetic mean	12	28	16
	Geometric mean	4.3	27	6.4

[a] n = number of measured samples per region. Lower limit of detection (LLD): ^{226}Ra = 2.2 mBq l^{-1}; ^{228}Ra = 3.78 mBq l^{-1}; ^{222}Rn = 0.19 Bq l^{-1}.

An attempt was made to correlate the activity concentrations of ^{226}Ra with ^{228}Ra and of ^{222}Rn with ^{226}Ra, considering the data obtained in each region separately. No good correlation was found between the two radium isotopes. It was observed that in most cases, ^{228}Ra concentrations were higher than ^{226}Ra concentrations in the groundwater samples (the observed ^{226}Ra/^{228}Ra ratio varied from 0.2 to 12). However, in a few cases at Vale do Ribeira — LR and Vale do Paraıba — IV regions, this isotopic ratio was higher than 1, indicating that the area is enriched with ^{238}U relative to ^{232}Th.

The statistical analysis showed that there is a reasonable correlation between ^{222}Rn and ^{226}Ra in Vale do Ribeira — LR (correlation coefficient of 0.7, at the 95% confidence level) and in Vale do Paraıba — IV (correlation coefficient of 0.7). It was

observed that the $^{222}Rn/^{226}Ra$ activity ratio decreases with increasing ^{226}Ra and ^{222}Rn contents. In most cases, the ^{222}Rn activity concentrations were three orders of magnitude higher than the ^{226}Ra activity concentrations.

The gross alpha and beta activities were also determined in the same water samples (Oliveira et al., 1997). These results showed that in only 4 of the 452 samples analysed, was the gross-alpha activity above the limit of 0.1 Bq l^{-1} recommended for public water supplies (Ministério da Saúde, 1990). One of them was located at Vale do Paraıba — IV, two in Vale do Ribeira — LR and one in Baixo Tietê/Grande — IT. The corresponding results obtained for ^{226}Ra also revealed activities above this limit. These results, however, are below 1/50 of the specific annual limit on intake (ALI) recommended for ^{226}Ra (CNEN, 1988), if a consumption of 2 l day^{-1} is considered (WHO, 1993). For the gross-beta activity, only one water sample from Baixo Paranapanema — IB, presented an activity concentration greater than 1 Bq l^{-1}. However, the corresponding ^{228}Ra concentration did not exceed this limit. The main contributor to the beta activity is possibly ^{40}K. Further analyses in these locations will be carried out to determine if the results are consistent. Overall, it can be concluded that the consumption of such waters does not imply any measurable health risk to the population.

Based upon the measured concentrations in the water supplies of São Paulo State, committed doses to the critical organs and committed effective doses were evaluated for ^{226}Ra, ^{228}Ra and ^{222}Rn ingestion. Doses were estimated by considering a daily consumption of 2 l (WHO, 1993) and the arithmetic mean activity concentration obtained for each region (see Table 1). The annual doses per unit of activity ingested for ^{226}Ra and ^{228}Ra used in these calculations (in Sv Bq^{-1}) were taken from ICRP (1993). For the ingestion of radon, there are no internationally accepted dose factors. The application of a modified ICRP model for the ingestion of radon in water (Kendall, Fell and Phipps, 1988) leads to a value of 10^{-8} Sv Bq^{-1} for the committed effective dose per unit intake, with all the dose coming from the gas rather than the decay products (UNSCEAR, 1993). The estimates for the dose to stomach per unit activity of ingested radon vary between 5×10^{-8} and 2×10^{-7} Sv Bq^{-1} (UNSCEAR, 1982, 1984); an intermediate value of 10^{-7} Sv Bq^{-1} was adopted here. The results obtained are presented in Table 2. Doses up to 0.3, 0.6 and 3.2 mSv yr^{-1} were estimated for the critical organs, considering the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn, respectively. The population consuming this water, at a rate of 2 l day^{-1}, are exposed to an annual committed effective dose of 6×10^{-3}, 2×10^{-2} and 3×10^{-1} mSv yr^{-1} due to the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn, respectively. The expected doses estimated for the ingestion of these radionuclides by the population of São Paulo were lower than those obtained, by the same authors, in a previous paper (Mazzilli et al., 1998), where the doses were evaluated for the ingestion of the radionuclides of the uranium series in spring waters from a high background region of Brazil. These doses were also lower than those estimated by Fernandez, Lozano and Gomes (1992) for ingestion of groundwater in Western Spain, and those predicted from the ingestion of drinking water in Finland, Slovenia and Syria (Asikainen & Kahlos, 1980; Kobal, Vaupotic & Mitic, 1990; Othman & Yassine, 1996).

Table 2
Expected committed doses to the critical organs and committed effective doses from the consumption of São Paulo State drinking water[a]

Sampling location	^{226}Ra Hb (mSv yr^{-1})	^{226}Ra He (mSv yr^{-1})	^{228}Ra Hb (mSv yr^{-1})	^{228}Ra He (mSv yr^{-1})	^{222}Rn Hs (mSv yr^{-1})	^{222}Rn He (mSv yr^{-1})
IV	0.1	2.8×10^{-3}	0.6	0.1×10^{-3}	1.1	0.1
IT	0.3×10^{-1}	0.7×10^{-3}	0.4	0.1×10^{-1}	0.4	0.4×10^{-1}
LB	0.2×10^{-1}	0.4×10^{-2}	0.5	0.1×10^{-1}	0.2	0.2×10^{-1}
LR	0.3	0.6×10^{-2}	0.6	0.2×10^{-1}	3.2	0.3
LN	0.1×10^{-1}	0.2×10^{-3}	0.4	0.1×10^{-1}	0.2	0.2×10^{-1}
IM	0.1	0.2×10^{-2}	0.5	0.1×10^{-1}	0.6	0.6×10^{-1}
IB	0.3×10^{-1}	0.6×10^{-3}	0.5	0.1×10^{-1}	0.4	0.4×10^{-1}
IA	0.1	0.2×10^{-2}	0.5	0.1×10^{-1}	1.1	0.1

[a] Hb = committed dose to the bone; Hs = committed dose to the stomach, He = effective dose.

Table 3
Population and percentage of people served with the SABESP water supply in the 8 regions studied in São Paulo State (IBGE, 1996)

Sampling location[a]	September/1997	
	10^3 inhab	(%)
IV	1048	96.7
IT	480	99.6
LB	1283	98.1
LR	147	74.9
LN	137	77.7
IM	797	92.2
IB	679	98.8
IA	531	96.4
Total	5102	>

[a] See Table 1.

According to the data published by IBGE (1996), the population and percentage of people served by the SABESP water supply in the 8 regions studied in São Paulo State are shown in Table 3. Based on statistical data of September 1997 and the corresponding arithmetic mean concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn observed in each region, the population-weighted average concentrations (C_{pwa}) for these radionuclides in the drinking water supplies of São Paulo State were determined. According to Aieta Singhley, Trussel, Thorbjarnarson and McGuire (1987), C_{pwa} is one parameter that expresses occurrence information to estimate the health hazards to the population from radionuclides. The population-weighted average activity concentrations for ^{226}Ra, ^{228}Ra and ^{222}Rn were obtained in this study using the

following equation:

$$\left[C_{\mathrm{pwa}} = \frac{\sum_{i=1}^{N} C_i\, P_i}{\sum_{i=1}^{N} P_i}\right],$$

where: C_{pwa} is the population-weighted average activity concentration in mBq $^{-1}$; C_i is the arithmetic mean activity concentration of ^{226}Ra, ^{228}Ra and ^{222}Rn observed in each region in mBq l^{-1} and P_i is the population of each region.

Based on the data presented in Tables 2 and 3, the São Paulo State population-weighted average concentrations for ^{226}Ra, ^{228}Ra and ^{222}Rn were 7.7, 27 and 9.5 Bq l^{-1}, respectively. These values are similar to those observed by EPA in some typical United States counties (Cothern & Rebers, 1990).

Considering these population-weighted averages, the corresponding committed effective doses reached values of 1.6×10^{-3}, 1.3×10^{-2} and 6.9×10^{-2} mSv yr^{-1} for ^{226}Ra, ^{228}Ra and ^{222}Rn, respectively. This result indicates that ^{222}Rn makes the highest contribution to the total effective dose. The total committed effective dose due to the consumption of São Paulo State drinking water is 8.4×10^{-2} mSv yr^{-1}. If we consider that 5102×10^{3} inhabitants are served by SABESP with water supply in the 8 regions studied, a collective dose of 429 man Sv could be expected for the population of São Paulo State.

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