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The Effects of Low and Very Low Doses of Ionizing Radiation
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Dose estimate and risk assessment due to the ingestion of ^{226}ra, ^{228}ra and ^{222}rn in drinking water supplies of São Paulo State - Brazil

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Abstract : This study aims to determine the activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn in drinking water supplies of São Paulo State. Concentrations up to 235 mBq L^{-1} 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 mSv y^{-1}, 0.6 mSv y^{-1} and 3.2 mSv y^{-1} were estimated for the critical organs, for the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn, respectively. The corresponding committed effective doses reached values of $6x10^{-3}$ mSv y^{-1}, $2x10^{-2}$ mSv y^{-1} and $3x10^{-1}$ mSv y^{-1}, for the same radionuclides. The lifetime risk of radiation-induced cancer due to the ingestion of these radionuclides is also discussed. In the worst case, a total of 1 radium-induced cancer (1 bone sarcoma) was predicted per million exposed persons, for the ingestion of ^{226}Ra and ^{228}Ra. For radon, a higher incidence of fatal stomach cancers found is 373 cases per million exposed persons. These estimates suggest that chronic ingestion of radium and radon at the levels observed in these supply waters would result in an increase in the fatal cancer rate up to 0.2% and 8%, respectively, above the background incidence rate observed in the Southeast region of Brazil.

Keywords : natural radioactivity, drinking water, radium isotopes, radon, dose evaluation, risk assessment.

1. Introduction

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 spring waters and bottled mineral waters (Szikszay and Sampa, 1983; Lauria and Godoy, 1988; Pires do Rio *et al.*, 1988; Oliveira *et al.*, 1994; Mazzilli *et al.*, 1998; Oliveira *et al.*, 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. This paper reports a preliminary study carried out in São Paulo State, which started in 1994 and covered 53.5% of the 574 existing counties (IBGE, 1996). This survey involved the determination of the activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn in surface and groundwater public water supplies.

The Brazilian authority Ministério da Saúde established current drinking water standards for the presence of radioactivity as follows (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 also further analyses in order to determine the radionuclides occasionally present in drinking water and their respective specific activities. The results obtained should be evaluated by comparison with the ALI (annual limit of intake) established for each radionuclide in the Reference Guide CNEN-NE

3.01(CNEN, 1988). Since the levels of natural radioactivity in Brazilian water supplies were never measured before, the interim regulation suggests also carry out a preliminary survey.

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 y generated through decay of ^{238}U, and ^{228}Ra, a shorter-lived beta-emitter (half-life 5.7 y), which is generated directly by the ^{232}Th decay. When humans ingest radium, about 20% is absorbed into the bloodstream. Radium absorbed is initially distributed to soft tissues and bone, but its retention is mainly in growing bone (ICRP, 1993). For ^{226}Ra, the following two types of malignancy can be induced: bone sarcomas and head carcinomas. For ^{228}Ra, which does not produce ^{222}Rn gas, the risk from head carcinomas is regarded as trivial compared to the risk from bone sarcomas (EPA, 1991a).

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 season and 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. Radon-222 is transported by water and 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).

This study aims, therefore, to determine the activity concentrations of ^{226}Ra, ^{228}Ra and ^{222}Rn in drinking water supplies of São Paulo State, since they are considered the most critical natural alpha and beta emitters as far as internal irradiation is considered. In order to evaluate the health hazards, dose estimates and risk assessment due to the ingestion of these waters are also discussed.

2. Material and methods

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. Samples were collected at 452 different locations, geographically distributed over 8 regions, as follows: Vale do Paraiba -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. The sampling program covered 53.5% of the 574 existing counties, 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 the ^{226}Ra and ^{228}Ra determination, 10 L of raw water sample was collected in polyethylene bottles at each site, in which 20 mL of HNO_3 65% was added to get a final pH lower than 1.5.

The radiochemical procedure adopted for ^{226}Ra and ^{228}Ra determination is described with more detail by the same authors in a previous paper (Oliveira *et al.*, 1994). A volume of 3 L of water sample was used for the analysis, which was performed always in duplicate. The water volume was reduced by evaporation to 1L, 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, 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 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. 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 decay of ^{224}Ra and of ^{223}Ra; that is, after 21 days. The ^{228}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 ^{228}Ra beta-particles (40 keV). Typical lower limits of detection 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).

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

Table 1. Natural radioactivity levels in drinking water supplies of São Paulo State during 1994 - 1997

Sampling Location (n)	^{226}Ra range (mBq L^{-1})	Arith. Mean ± stand. dev.	^{228}Ra range (mBq L^{-1})	Arith. Mean ± stand. dev.	^{222}Rn range (Bq L^{-1})	Arith. Mean ± stand. dev.
IV - Vale do Paraíba (56)	< 2.2 - 213	14 ± 32	13 - 78	30 ± 12	0.96 - 274	16 ± 41
IT - Baixo Tietê/Grande (158)	< 2.2 - 42	3.2 ± 4.8	9.1 - 92	24 ± 7.1	0.87 - 23	5.6 ± 3.2
LB - Baixada Santista (13)	< 2.2 - 3.9	1.9 ± 0.9	22 - 38	25 ± 4.1	2.3 - 5.0	3.1 ± 0.6
LR - Vale do Ribeira (39)	< 2.2 - 235	27 ± 53	< 3.7 - 131	34 ± 23	0.40 - 315	43 ± 79
LN - Litoral Norte (20)	< 2.2 - 4.8	1.2 ± 1.0	21 - 27	24 ± 1.9	1.2 - 3.5	2.1 ± 0.5
IM - Médio Tietê (28)	< 2.2 - 66	11 ± 18	17 - 64	30 ± 14	2.6 - 24	8.8 ± 5.7
IB - Baixo Paranapanema (107)	< 2.2 - 30	2.9 ± 4.3	18 - 73	26 ± 7.4	1.3 - 19	5.3 ± 2.7
IA - Alto Paranapanema (31)	< 2.2 - 74	12 ± 18	20 - 61	28 ± 9.4	1.8 - 135	16 ± 27

Legend: n = number of measured samples per region; (LLD): ^{226}Ra = 2.2 mBq L^{-1}; ^{228}Ra = 3.7 mBq L^{-1}; ^{222}Rn = 0.19 Bq L^{-1}

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 concentration 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. The ^{222}Rn activity concentration, on the other hand, ranged from 0.40 Bq L^{-1} to 315 Bq L^{-1}.

Although there are other natural radionuclides, such as ^{232}Th, ^{230}Th and ^{210}Pb, which can be considered more critical than ^{226}Ra and ^{228}Ra intake from drinking water ingestion if their respective dose conversion factors are taken into account, these radionuclides usually represent a small fraction of the gross alpha and beta activities, since their solubility in water is very low. Therefore, in a

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

Sampling Location	^{226}Ra Hb (mSv y^{-1})	^{226}Ra He (mSv y^{-1})	^{228}Ra Hb (mSv y^{-1})	^{228}Ra He (mSv y^{-1})	^{222}Rn Hs (mSv y^{-1})	^{222}Rn He (mSv y^{-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

Legend: Hb = committed dose to the bone
Hs = committed dose to the stomach
He = effective dose

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

Sampling Location [a]	September/ 1997	
	10^3 inhab	(%)
IV	1,048	96.7
IT	480	99.6
LB	1,283	98.1
LR	147	74.9
LN	137	77.7
IM	797	92.2
IB	679	98.8
IA	531	96.4
Total	5,102	

[a] See Table 1

Table 4. Population-weighted averages(Cpwa) of ^{226}Ra, ^{228}Ra and ^{222}Rn in drinking water supplies of São Paulo State

	^{226}Ra(mBq L^{-1})	^{228}Ra(mBq L^{-1})	^{222}Rn(Bq L^{-1})
Cpwa(Sept/97)	7.7	27	9.5

Table 5. Lifetime cancer risk due to ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn from drinking water supplies of São Paulo State

Sampling Location [a]	Bone Sarcomas per person - year	Head Carcinomas per person - year	Fatal Stomach Cancers per person - year
IV	4.2×10^{-7}	4.7×10^{-8}	132×10^{-6}
IT	3.0×10^{-7}	1.1×10^{-8}	27×10^{-6}
LB	3.0×10^{-7}	6.4×10^{-9}	48×10^{-6}
LR	5.4×10^{-7}	9.5×10^{-8}	373×10^{-6}
LN	2.9×10^{-7}	4.2×10^{-9}	18×10^{-6}
IM	4.1×10^{-7}	3.9×10^{-8}	76×10^{-6}
IB	3.2×10^{-7}	1.0×10^{-8}	46×10^{-6}
IA	3.9×10^{-7}	3.9×10^{-8}	137×10^{-6}

[a] See Table 1.

conservative approach, ^{226}Ra and ^{228}Ra can be considered as singly responsible for the 0.1 Bq L^{-1} for gross-alpha activity or 1 Bq L^{-1} for gross-beta activity, from the consumption of drinking water.

In parallel to this work, the gross alpha and beta activities were also determined in the same water samples (Oliveira *et al.*, 1997). These results showed that in only four of the 452 samples analysed, the gross-alpha activity was 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 Paraíba -IV, two in Vale do Ribeira -LR and one in Baixo Tietê/Grande -IT. The corresponding results obtained for ^{226}Ra also presented activities above this limit. These results, however, are below 1/50 of the specific annual limit of intake (ALI) recommended for ^{226}Ra (CNEN, 1988), if a consumption of 2 L per day 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 analysis in these locations will be carried out to check if the results are consistent. Therefore, it can be concluded that the consumption of such waters will not imply in any additional health risk to the population.

Based upon measured concentrations in water supplies of São Paulo State, committed doses to the critical organs and committed effective doses were performed for ^{226}Ra, ^{228}Ra and ^{222}Rn. Doses were estimated by considering a daily consumption of 2 L (WHO, 1993) and the arithmetic mean obtained for each region (see Table 1). The arithmetic mean was chosen instead of the geometric mean, since the activity concentrations obtained correspond to a single measurement at each point and the statistical distribution of the observed data does not correspond, in most cases, to a log-normal distribution. The annual dose 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 to the ingestion of radon in water (Kendall *et al.*, 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 mSv y^{-1}, 0.6 mSv y^{-1} and 3.2 mSv y^{-1} were estimated for the critical organs, considering the ingestion of ^{226}Ra, ^{228}Ra and ^{222}Rn, respectively; while the corresponding committed effective doses reached values of 6×10^{-3} mSv y^{-1}, 2×10^{-2} mSv y^{-1} and 3×10^{-1} mSv y^{-1} for the same radionuclides. The expected doses estimated for the ingestion of these radionuclides by the population of São Paulo are of the same order of magnitude as 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.

According to the data published by IBGE (1996), the population and percentage of people served by SABESP with 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 concentration of ^{226}Ra, ^{228}Ra and ^{222}Rn observed in each region, the population-weighted averages (Cpwa) for these radionuclides in drinking water supplies of São Paulo State were determined. Population-weighted averages are calculated by summing the product of activity or concentration (Ci) and population (Pi) over all sites and dividing by the sum of the population over all sites. According to Aieta *et al.* (1987), Cpwa is one parameter that expresses occurrence information to estimate the health risk to the population from radionuclides. The population-weighted average activities for ^{226}Ra, ^{228}Ra and ^{222}Rn obtained in this study are presented in Table 4. These values are the same order of magnitude of those observed by EPA in some typical United States counties (Cothem and Rebers, 1990).

The lifetime risk of radiation-induced cancer due to the ingestion of radium isotopes and radon were also predicted for each region studied. In order to quantify the carcinogenic effects of ^{226}Ra and ^{228}Ra ingestion from drinking water, the risk assessment in the present study was carried out using the methodology proposed by Mays *et al.* (1985). This model fits the dose-response relationship for the incidence of bone sarcomas and head carcinomas among radium dial painters exposed to ^{226}Ra and/or ^{228}Ra. Mays *et al.* (1985) analysed the predicted incidence of bone sarcomas and head carcinomas among low-dose dial painters following the studies performed by Rowland *et al.* (1983). The analysis of epidemiological data was done considering the total intake of radium to blood, weighting ^{228}Ra 2.5 times more than ^{226}Ra. In both cases, ingestion of radium was converted to intake into the blood using a gastrointestinal absorption factor of 20% (ICRP, 1993).

The incidence of bone sarcomas and head carcinomas due to the ingestion of ^{226}Ra and ^{228}Ra were evaluated taking into account that the life expectancy in Brazil is 65 years. For the incidence of head carcinoma, only the intake of ^{226}Ra during the first 55 years was considered effective, since the minimal latency period for this kind of cancer is 10 years. For the incidence of bone sarcoma, it was assumed that intake of ^{226}Ra and ^{228}Ra occurred during the first 60 years, since in this case the latency period is 5 years. The results obtained for the excess of lifetime risk due to the ingestion of ^{226}Ra and ^{228}Ra are presented in Table 5. In the worse case, a total of 1 radium-induced cancer (1 bone sarcoma) was predicted per 10^6 exposed persons. The background incidence of this type of cancer in the Southeast region of Brazil is 7.0 x 10^{-6} per year (Brumini *et al.*, 1982), which means 455 cases per million, assuming a life expectancy of 65 years. These predictions suggest that chronic ingestion of radium at the levels observed in São Paulo State supply waters would result in an increase of fatal cancer rate up to 0.2% above the background incidence rate in the Southeast region of Brazil.

The stomach cancer was the primary health effect of concern for the ingestion of ^{222}Rn (EPA, 1991b). The lifetime risk from ingestion of radon was estimated assuming an ingestion rate of 2 L per day per person, using the biokinetic model proposed by Crawford-Brown (1991). In these calculations, it was also considered that 20% of the initial radon water content is lost during the process of filling a glass and drinking the water. For ^{222}Rn the higher incidence of fatal cancers found is 373 cases per million of exposed persons. The background incidence rate of stomach cancer in the Southeast region of Brazil is 6.8x10^{-5} year (Brumini *et al.*, 1982), which means 4,437 cases per million, considering a life expectancy of 65 years. Therefore, an excess of 8% in the background incidence rate can be expected due to the ingestion of ^{222}Rn present in the waters.

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