Radon in Groundwater Public Supplies in the Metropolitan Area of Recife, Brazil

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INTRODUCTION

Man's exposure to natural radiation accounts for around 70% of the total dose received by the population. Most of this exposure is due to ²²²Rn, a member of the ²³⁸U decay series, which is produced by the direct decay of ²²⁶Rn. Being a noble gas, radon produced in the rock or soil matrix can easily diffuse to the water trapped in the pore spaces and migrate to ground water, where it may be present in concentrations several orders of magnitude higher than those of its predecessor.

Hydrological investigations concerning the presence of radionuclides in public and private drinking water supplies have been mainly related to groundwater sources, because the radon content of surface waters is typically low due to its desorption by aeration.

In Brazil there are only a few studies concerning the presence of radionuclides in water (1,3). Recently, the Brazilian Ministry of Health issued new interim regulations concerning the quality of water for human consumption (4). As far as the levels of radioactive contaminants are concerned, it was stated that the gross alpha and beta activities should not exceed 0.1 Bq/L and 1 Bq/L, respectively. The regulation states that further analyses shall be carried out to identify the specific radionuclides present in the water, as well as their individual concentration, whenever the measured values are above limits those limits.

In order to verify if the levels of radioactive contaminants in the water from public supplies are in compliance with the limits set by the Ministry of Health, a survey has been carried out in the metropolitan area of Recife, Brazil. This paper reports the results of this survey, as far as ²²²Rn is concerned.

MATERIALS AND METHODS

The region chosen for this study encompasses 13 counties that form the metropolitan region of Recife, one of the most populated cities in Northeastern Brazil. Water from all groundwater sources that supply water for public consumption (97 drilled wells) was collected and analyzed for their radon content. These wells are under the jurisdiction of COMPESA - Companhia Pernambucana de Saneamento, a state owned water company.

Radon concentration was determined by the liquid scintillation counting technique. This detection method relies on the very high solubility of radon in certain organic solvents when compared to water (the ratio of the Ostwald coefficient for radon in toluene and radon in water is roughly 50 at 20°C). In contrast to the standard procedure in liquid scintillation counting, there is no requirement for having sample and scintillation cocktail forming a well-mixed single phase. Rather, the fact that the organic scintillator and water form a twophase mixture is advantageous because, in this case, the relative solubilities will determine which radionuclides will be present in the scintillator phase. The method used in this research is similar to the one developed by Prichard and Gesell (5). The cocktail used was prepared by adding 7 g of PPO (2,5 diphenyloxazole) to 1 L of scintillation grade p-xylene, stirring to dissolve, adding 1.5 g of bis-MSB (p-bis-[o-Methylstyryl]-benzene) and stirring until complete dissolution was achieved. Each sample was prepared by pipetting 11 mL of scintillator into a glass vial, injecting 11 mL of water under the scintillator, closing the vial, and agitating vigorously to speed up the transfer of radon from the water to the organic phase. Each sample was prepared in duplicate, so that large discrepancies observed between paired samples could be used as indication of inappropriate sample preparation. After sample collection, the vials were set aside for a minimum of three hours to allow the ingrowth of radon progeny, placed in the liquid scintillation counter (Beckman, mod.) and counted for 50 minutes. The counting efficiency was determined through the use of a ²²⁶Ra standard (a known activity of ²²⁶Ra added to a vial containing 11 mL of scintillator and 11 mL of water). At least two vials for background (11 mL of scintillator and 11 mL of distilled water) and two vials with the ²²⁶Ra standard were counted in each batch of samples.

The assessment of the dose derived from the ingestion of radon bearing water was carried out according to the biokinetical model developed by Crawford-Brown (6). In this model, the human body is divided in several compartments, the dose rate to each compartment being a function of the time elapsed since water ingestion. The model assumes that the ingested radon enters the bloodstream via the gastrointestinal tract (GI), reaching the liver and the lungs. Radon in the alveolar blood emanates to the airspace in the alveolar region until a condition of equilibrium is reached. This "equilibrium condition" persists as long as a flow of radon from the GI exists. Radon that accumulates in the alveolar region leaves the body mainly through exhalation. The air exchange rate in the alveolar region for an adult is taken as 7.5 L.min⁻¹.

A detailed model for the movement of radon that enters the human body through ingestion is shown in Figure 1. The time-dependent radon concentrations for each compartment were determined from the experimental data obtained by Correia et al. (7).



Figure 1. Biokinetic model for radon ingestion developed by Crawford-Brown (6).

The time-dependent concentration functions for specific organs, obtained from the biokinetic model are: Stomach:

$$C(t) = 2.5 \times 10^{-3} \times \left(0.998 \, e^{-0.29t} + 0.002 \, e^{-0.0026t}\right) \tag{1}$$

Small intestine:

$$C(t) = 2.4 \times 10^{-4} \times \left(0.9 \ e^{-0.02t} + 0.012 \ e^{-0.0045t}\right)$$
(2)

Upper large intestine:

$$C(t) = 1.4 \times 10^{-4} \times \left(0.85 e^{-0.015t} + 0.15 e^{-0.006t}\right) + \left(1 - e^{0.099t}\right)$$
(3)

Lower large intestine:

$$C(t) = 3.3 \times 10^{-5} \times e^{-0.0048t} \times (1 - e^{-0.099t})$$
(4)

Liver:

$$C(t) = 3.3 \times 10^{-5} \times \left(0.87 e^{-0.022t} + 0.13 e^{-0.0039t}\right)$$
(5)

Muscle:

$$C(t) = 1.1 x 10^{-5} x e^{-0.0029t} x \left(1 - e^{-0.023t}\right)$$
(6)

Fat:

$$C(t) = 1.1 \times 10^{-5} \times e^{-0.005t} \times (1 - e^{-0.099t})$$
(7)

General body tissue:

$$C(t) = 1.1 \times 10^{-4} \times \left(0.92 \, e^{-0.022 t} + 0.08 \, e^{-0.0035 t}\right) \tag{8}$$

The above concentration functions correspond to the ingestion of 1 Bq at t = 0. The concentrations C(t) are expressed in Bq/cm³ and t is expressed in minutes. The model assumes that the biological half-life is not age-

$$H_i = 3.821 x 10^{-6} x A \int_0^t C(t) dt$$
(9)

dependent and that the density of each organ or tissue is 1 g/cm^3 . The organ volumes used in the calculations are those for the reference man, as given in ICRP-23 (8).

The equivalent dose for a specified organ or tissue was calculated by the equation:

where A is the ingested radon activity (Bq). The effective dose was then calculated by summing the weighted equivalent doses for all tissues and organs included in the model.

The risk was estimated by assuming an average life expectation of 65 years for the inhabitants of the region and by using the risk coefficient of 0.05 Sv^{-1} given in ICRP-60 (9) for somatic effects (averaged for sex and age). The latency period for the manifestation of cancer was assumed to be equal to ten years. Therefore, only the dose received during the first 55 years of life will be considered for calculating the risk. The cumulative excess risk $R_{(Rn)}$ during the lifetime will be given by:

$$\mathbf{R}_{(\mathrm{Rn})} = 0.05 \text{ x } \mathbf{E}_{55} \tag{10}$$

where E_{55} is the cumulative effective dose received in 55 years of life.

RESULTS

The radon content of water samples ranged from 5.3 to 83.7 Bq/L, with an average concentration of 28.0 Bq/L. For calculating the rate of ingestion of radon, some specific considerations are necessary, due to the fact that radon can easily emanate from water, depending on the way water is used. It is well known that that the rate of radon emanation is a function of water temperature, surface area of water exposed to the air and to the degree of agitation. The rate of radon release from water increases dramatically as the water temperature is increased, so that by boiling the water all its content of radon will be released. Therefore, water used for making coffee, tea, soup and cooked food will have no radon. For this reason, the consumption of one liter of water per day was adopted for calculating the rate of radon ingestion.

Based on these considerations, the annual rate of radon ingestion by the population living in the metropolitan area of Recife was found to range from 1.48×10^3 to 4.73×10^4 Bq. The corresponding effective doses ranged from 0.36 to 2.25 mSv/yr. The average concentration of radon found in this study is four times higher than the one found in a similar study carried out in North Carolina.

The estimated probability of cancer incidence varied from 1.0 x 10⁻⁴ to 6.2 x 10⁻³, corresponding to the

occurrence of 100 to 6,200 additional cases for a population of one million inhabitants. This range corresponds to an estimated increase between 0.17% and 3.25% of the normally expected cases of death from cancer (10).

CONCLUSION

The relatively high concentrations of radon in water found in this study emphasize the necessity of further studies aiming to assess the contribution of other radionuclides to the dose received by the population.

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