URANIUM, RADIUM, AND RADON IN GROUNDWATERS OF THE PHOSPHATIC REGION OF NORTHEASTERN BRAZIL

Lima, R. A.; Hazin, C. A.; Khoury, H. J. (*)
Pecequilo, B. S. (**)

(*) IPEN/CNEN
(**) DEN/UFPE
Brazil

Abstract.

This study aimed to determine the concentration of ^{226}Ra, natural uranium and ^{222}Rn in groundwaters of the phosphatic region of Pernambuco, a State in the Northeast of Brazil. In this region there are two aquifers that provide water for consumption and irrigation. Water from 43 dug wells tapping the superficial aquifer and from 10 drilled wells tapping the lower aquifer was analyzed. The results showed concentration of natural uranium ranging from 2.25±0.14 mBq/L to 130.8±4.4mBq/L. For ^{226}Ra, concentrations ranged from 6.4±3.4 mBq/L to 2029±19 mBq/L. Radon-222 concentrations, on the other hand, ranged from 0.3 Bq/L to 40.3 Bq/L. The highest concentrations, for each of the radionuclides investigated, were found in water from wells tapping the uppermost aquifer.

1. Introducción.

Uranium and other members of the uranium series are commonly associated with phosphorite sediments which are found in several parts of the world. Phosphate deposits normally occur at shallow depths, and are often underlain by water bearing strata. Water movement within the aquifer region contributes to extract radionuclides from the sediments increasing, therefore, their concentration in groundwater. Current interest in radioactivity in groundwater arises from the potential hazards of the presence of radioisotopes such as uranium, radium, and radon in drinking water. The concern is justified by the fact that natural radioactivity accounts for almost 70% of the total dose received by the population.

The zone selected for this study is a 4-km wide land strip located in the costal region of the Northeast of Brazil (Fig.1). The area is known for its phosphate deposits and contains uranium in concentrations ranging from 150 to 300 ppm of uranium oxide. The region, which has been partially mined, is underlain by two aquifers: the free uppermost aquifer which is accessed through dug wells; the lower aquifer, on the other hand, is confined and supplies water through drilled wells. Water from these aquifers is used for consumption, as well as, for irrigation, resulting in the incorporation of radionuclides in the human body. Wells selected for this study are known to open to discrete water-bearing zones, so that the radioactivity content of each of the aquifers can be characterized.

This study aims to determine the concentrations of natural uranium (U_{nat}), ^{226}Ra, and ^{222}Rn in waters from dug and drilled wells in a selected area, in order to provide data to be used to calculate radiation doses received by people living in the region under investigation.

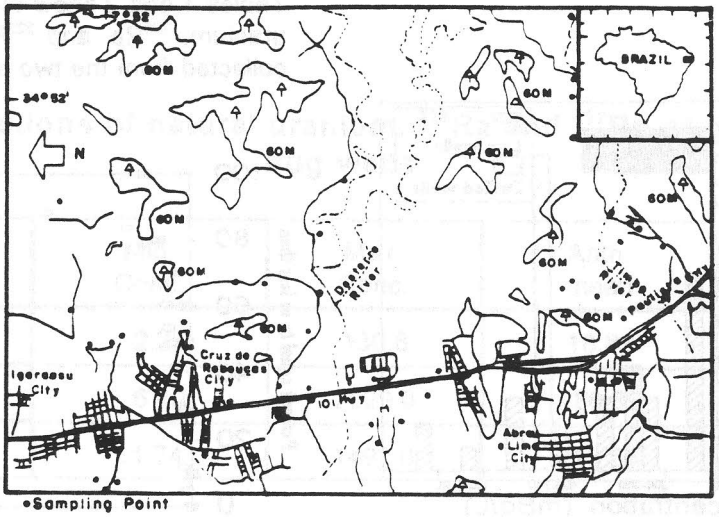

Fig. 1. Location of the investigated area with the position of the wells analyzed.

2. Methodology.

Water samples were collected from 43 dug wells and 10 drilled wells of the phosphate district. Its uranium content was determined through the fluorophotometry technique, which consists in the extraction of uranium with an organic extractor (TOPO), followed by deposition on NaF/LiF disks previously prepared in platinum crucibles. The uranium spiked disks were then fused at 9000 °C, and cooled according to the procedure given by Price et alli. [1]. The fluorescence of each disk was measured in a fluorometer (Jarrel-Ash Mod. 27-000) and related to the uranium content of the water sample, through a previously prepared calibration curve.

Radium-226 concentrations were determined through the radon emanation technique. Radium was concentrated and scavenged by co-precipitation with a barium carrier. The precipitate was dissolved with EDTA reagent to end up with a volume of approximately 16 mL. The solution was transferred to a radon bubbler, deemanated by passing compressed air through the bubbler, and stored to allow for ingrowth of ^{222}Rn. At the end of the storage period each sample was de-emanated into an evacuated scintillation cell attached to the bubbler. After waiting for at least three hours to ensure equilibrium between radon and radon daughters, each cell was counted for 10 minutes in a radon counter.

Radon-222, on the other hand, was determined through the liquid scintillation technique, in a procedure similar to the one described by Prichard and Gesell [2]. The cocktail used was prepared by adding 7g of PPO and 0.75g of POPOP to 1L of scintillation grade p-xylene and stirring until complete dissolution was achieved. Each sample was prepared by pipetting 12mL of scintillator into a glass vial, injecting 12mL of well 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 an indication of inappropriate preparation. After sample collection, the vials were set aside for a minimum of three hours, to allow the ingrowth of radon daughters, placed in a liquid scintillation counter (Beckman Mod. LS1701), and counted for 50 minutes. The counting efficiency was determined through the use of a ^{226}Ra standard. At least two vials for background and two vials with the ^{226}Ra standard were counted with each batch of samples.

3. Results and Discussion.

Figure 2 shows the ^{226}Ra concentration distribution, as a bar chart, in the dug and drilled wells. It was found that 97.7% of the wells investigated have a radium-226 concentration equal or exceeded 6.7 mBq/L (0.18 pCi/L), the minimum reporting level (MRL) [3]. About 65% of the wells showed ^{226}Ra concentration below 5 pCi/L (185 mBq/L), the interim maximum concentration level for this radionuclide.

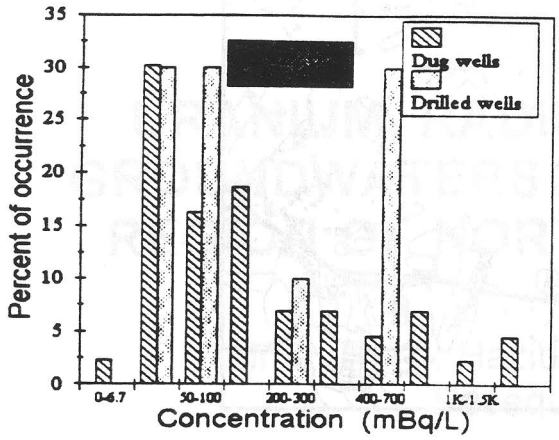

Fig. 2. Distribution of ^{226}Ra concentrations in dug and drilled wells.

Figures 3 and 4 show the probability of occurrence versus the uranium concentration obtained for the waters from dug and drilled wells, respectively. All of the samples present natural uranium concentrations higher than the MRL (0.08 mg/L). For ^{222}Rn all the dug wells investigated present concentrations higher than the MRL (100 pCi/L), while 10% of the drilled wells have a concentration less than the-MRL.

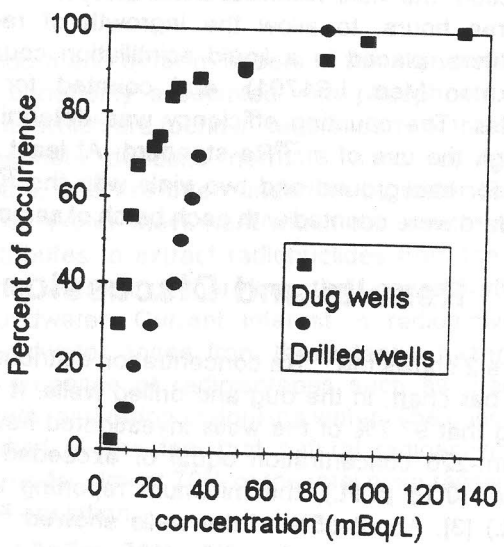

Fig. 3. Probability acumulated of Unat concentrations in dug and drilled wells.

Tables 1 and 2 show the concentrations of natural uranium, ^{226}Ra, and ^{222}Rn in 53 samples of water collected from the two aquifers

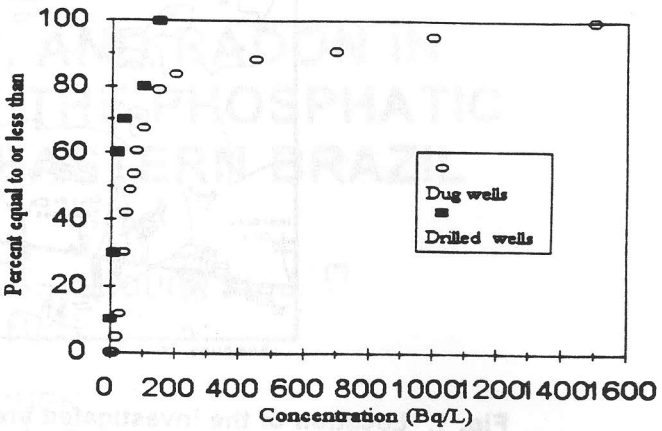

Fig. 4. Probability acumulated of ^{222}Rn concentrations in dug and drilled wells.

Analyses of the data (tables 1 and 2) showed that the highest concentrations, for each of the radionuclides investigated, were found in water from wells tapping the uppermost aquifer. This was expected since the first aquifer penetrates, in some locations, the phosphate bearing strata.

4. References.

1. Price, G. R.; Ferreti, R. J., and Schwartz, S.: "Fluorophotometric determination of uranium". Analytical Chem., 25(2):323–31, 1953.

2. Prichard, H.M., and Gesell, T. M.: " Rapid measurements of ^{222}Rn concentrations in water with a commercial liquid scintillation counter", Health Phys. 33:577-81, 1977.

3. Cothern,C.R and Rebers,P.A, "Radon, Radium and Uranium in drinking water", Lewis Publishers, 1990.

Table 1. Concentrations of natural uranium, ^{226}Ra and ^{222}Rn of ground waters in dug wells.

Radionuclide	Min. Conc.	Max. Conc.	Arith. mean	Geom. mean
U_{nat} (mBq/L)	2.25	130.8	18.81	10.68
^{226}Ra (mBq/L)	6.40	2029.0	286.85	103.54
^{222}Rn (mBq/L)	11.74	1491.0	193.56	86.18

Table 2. Concentrations of natural uranium, ^{226}Ra and ^{222}Rn of ground waters in drilled wells.

Radionuclide	Min. Conc.	Max. Conc.	Arith. mean	Geom. mean
U_{nat} (mBq/L)	7.58	58.70	202.97	22.02
^{226}Ra (mBq/L)	20.60	521	202.97	112.16
^{222}Rn (mBq/L)	2.40	148.70	49.19	22.02