2017 International Nuclear Atlantic Conference - INAC 2017
Belo Horizonte, MG, Brazil, October 22-27, 2017
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR – ABEN

Use of Environmental Tritium in Groundwater Dating in the Upper Jequitibá River Basin, Municipality of Sete Lagoas, Minas Gerais, Brazil

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ABSTRACT

Tritium is a natural radioactive isotope that can be used in dating modern groundwater. Due to the increase of this radionuclide content in the atmosphere during the nuclear tests in the 1960s, it became possible to determine the age of recent groundwater. Such a measurement is important inasmuch as it sheds light upon groundwater circulation and the renewability of aquifers. The area where this research was carried out is located at the upper section of the Jequitibá river basin, geologically dominated by limestone rocks of the Bambui Group. At his region the karstic aquifers are responsible for the water supply of the cities of Sete Lagoas and Prudente de Moraes. The tritium activity was determined in samples from wells and the analytic results allowed the calculation of the ages of the water using the Exponential Flow Model, which considers that there was a mixture of more recent waters along the travelled path in the subsoil. The obtained results showed that the water of the deep aquifer is older, between 200 and 60 years, while waters of the free shallow aquifer are less than 37 years old. These results indicate the renewal time in the aquifers and can contribute to the better management of the water resources in regions with water availability problems.

1. INTRODUCTION

Tritium is the radioactive isotope of hydrogen that has three atomic masses (^3H), whose half-life is 12.43 years (CLARK & FRITZ, 1997; CANADA, 2009; CALMON & GARNIER-LAPLACE, 2010). This isotope may be part of the water molecule, and it is considered as a conservative tracer, except for its decay in the scale of tens of years. As an environmental tracer, it is important for the dating of modern groundwater, that is, the groundwater that has infiltrated the aquifer during the last decades.

Tritium is produced naturally by nuclear reactions due to the incidence of high energy cosmic rays on nitrogen and oxygen atoms in the upper atmosphere, where 99% of this formed tritium is oxidized to tritiated water (HTO) and dispersed to the hydrosphere (GARNIER LAPLACE, 2010). However, during the nuclear tests mainly between the years 1945 and 1963, many radionuclides, including tritium, were released into the atmosphere, generating a signal that marks the waters that were recharged during this time and thereafter.

According to Clark & Fritz (1997) and also the report organized by the Government of Canada (CANADA, 2009), the standard unit of measurement for concentration of tritium in waters is the Tritium Unit (TU) which equals 1 atom of tritium for each 1018 hydrogen atoms, which corresponds to an activity of 0.0118 Bq or 0.319 pCi.

The dating of groundwater is based on the premise that the activity of tritium at the time of recharge is known and that the value obtained by analysis of the water of the aquifer corresponds only to the radioactive decay.

According to Mourão (2007) and Jurgens *et al.* (2012), the residence time is commonly calculated by two methods: piston flow (Equation 1) and exponential flow (Equation 2).

The piston flow model considers that the recharge water does not mix, preserving its isotopic identity. This model assumes that a tracer is transported from the recharge area to the exit area (well or spring, for example) without hydrodynamic dispersion. The piston flow model may be applicable to hydrogeological configurations where dispersion is low, linear velocity is high, or the path of the refueling flow to discharge is short (JURGENS et al., 2012). It is represented mathematically by Equation 1.

$$\mathrm{C_s}\,(t)= C_0\, e^{-\lambda t} \qquad (1)$$

Where:

C_s (t) - concentration of tritium at time t at which analysis was performed [UT];
C_0 - initial concentration [UT];
λ - tritium decay constant [t^{-1}];
t - period of time between recharge and analysis [t].

The exponential flow model takes into account water mixtures during the residence in the aquifer. It is applicable to homogeneous and unconfined aquifers of constant thickness and receiving a uniform recharge. This situation is represented by a vertical stratification of the groundwater age, with ages starting from zero in the water table and tending to infinity at the

base of the aquifer (Appelo and Postma, 2006). This model may be suitable for fully penetrating wells or aquifers discharging into springs. The exponential model is applied when the transverse dispersion does not occur along the flow lines and the mixing occurs inside the well or spring rather than the aquifer (Jurgens et al, 2012).

The equation of this model is represented in Equation 2.

$$C_s(t) = \sum_{t'=-\infty}^{t-\Delta t} C_0(t')\frac{1}{\tau}\frac{1}{\frac{1}{\tau}+\lambda}\left\{Exp\left[-\left(\frac{1}{\tau}+\lambda\right)(t-t'-\Delta t)\right] - Exp\left[-\left(\frac{1}{\tau}+\lambda\right)(t-t')\right]\right\} \quad (2)$$

Where:
Cs (t) - Concentration at time t at which the sample [TU] was analyzed;
C_0 - initial concentration [TU];
λ - tritium decay constant [t^{-1}];
τ - residence time of water in the aquifer [t];
(t-t ') - time between the year in which C_0 is considered until the time of analysis [t].

2. STUDY AREA

This research was carried out around the Experimental Station of the National Center for Research on Corn and Sorghum (CNPMS) of the Brazilian Agricultural Research Corporation (EMBRAPA). The Upper Jequitibá river basin, where the CNPMS area is located, is in the municipalities of Sete Lagoas and Prudente de Moraes, 70 km northwest of the State capital, Belo Horizonte. The figure 1 show the study area.

The area of interest is located in the karstic region of the Hydrogeological Province of the São Francisco River, and according to Galvão et al. (2015), is constituted by neoproterozoic limestones of the Sete Lagoas Formation, where karst conduits were developed, giving rise to the homonymous aquifer, which is covered by inconsolidated Cenozoic sediments and neoproterozoic metasedimentary rocks of the Serra de Santa Helena Formation.

Peculiarities involving karstic aquifers have justified numerous studies in their areas of occurrence, since these aquifers are of great importance as a source of water, but are also vulnerable to contamination (MACHADO, 2011).

3. HYDROGEOLOGICAL ASPECTS

There are four types of aquifers in the area (Machado, 2011):
- Granular aquifer: formed by the cover soils and saprolites.
- Karstic Aquifer: formed in the rocks of the Sete Lagoas Formation., which is quite productive and confined, showing flowing artesian wells in the area.
- Fractured-Karstic Aquifer: Associated with the presence of intercalations of pelitic rocks and calcareous lenses occurring in the Serra de Santa Helena Formation. Deeper portions behave like aquitards.
- Fractured Aquifer: formed in the gneisses of the Belo Horizonte Complex.

4. MATERIALS AND METHODS

4.1. Sampling Points

The groundwater samples were collected in seven shallow monitoring wells on Serra de Santa Helena Formation, six deep pumping wells, 4 of which capture water from the karstic aquifer of the Sete Lagoas Formation, and two capture water from the fractured aquifer of the Belo Horizonte Complex. Some water samples were also collected from a manual excavated well, called cistern. The sampled points is presented in the Figure 1.

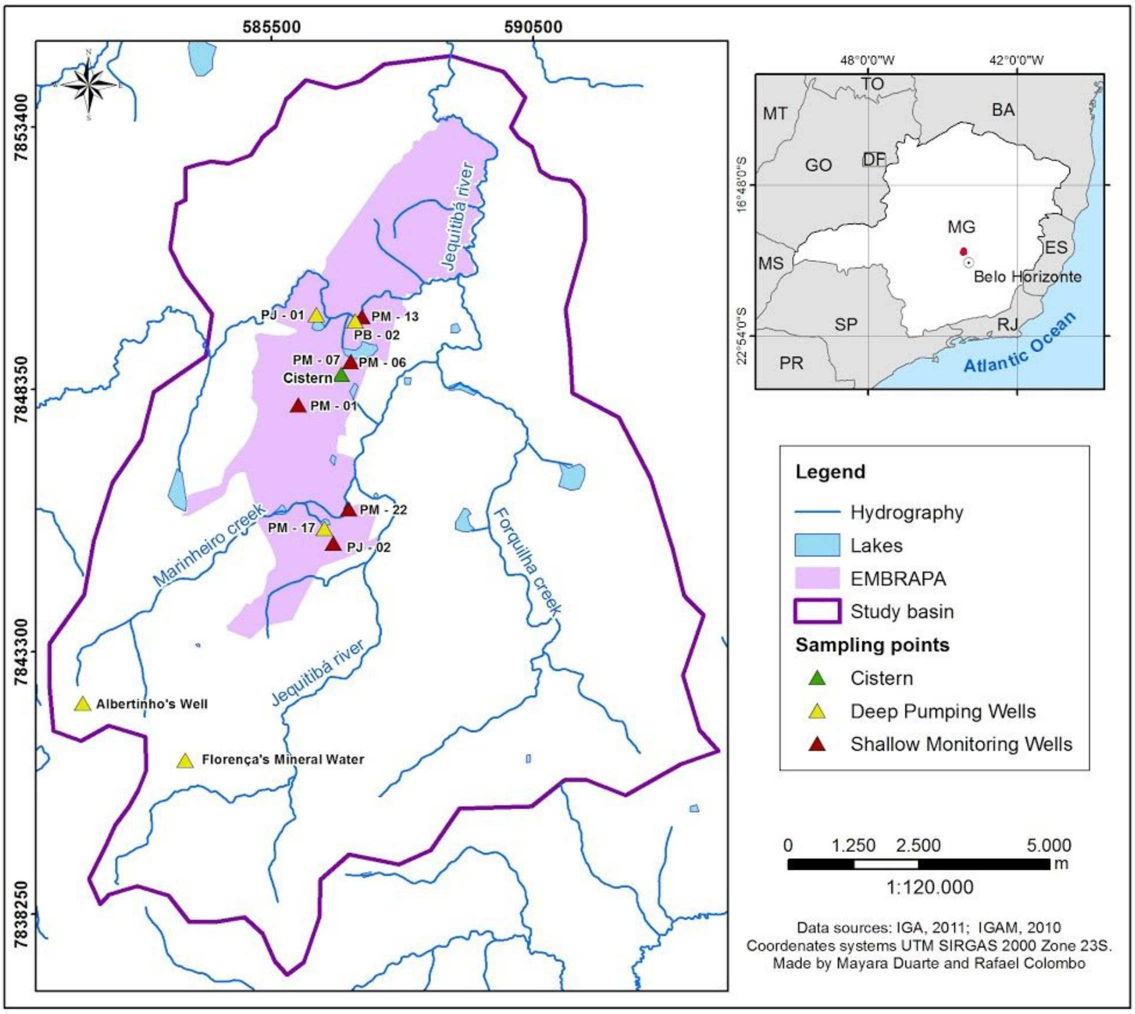

Figure 1 - Map showing the study basin and the sampled points.

4.2. Laboratory Isotopic Analysis of Tritium

The isotope analyzes were performed in the CDTN laboratories. The tritium samples were analyzed at the CDTN Environmental Tritium Laboratory, concentrated by the electrolytic enrichment process and counting by liquid scintillation spectrometry in the Perkin-Elmer Quantulus 1220 spectrometer (Davis, 2006). They were analyzed in the period between May and June of 2015 year.

4.3. Water Age Determination

For the determination of the water age in the aquifers of the study area, the exponential model was used, and the estimates calculated by Mourão (2007) and reviewed by Carvalho (2012), as input values for tritium were used. Values measured in the precipitations of several stations in Brazil and abroad, and were obtained the mean weighted average concentrations for the area of the Belo Horizonte region between the years of 1953 and 2013.

The figure 2 shows the values of tritium activity used as input values for the aquifers of the study area. The data presented by Mourão (2007) and Carvalho (2012) were used, which were recalculated and complemented until the year 2015. One can observe the peak of tritium activity for the years 1964 and the decay to the present day, where tritium values are close to pre-nuclear test values.

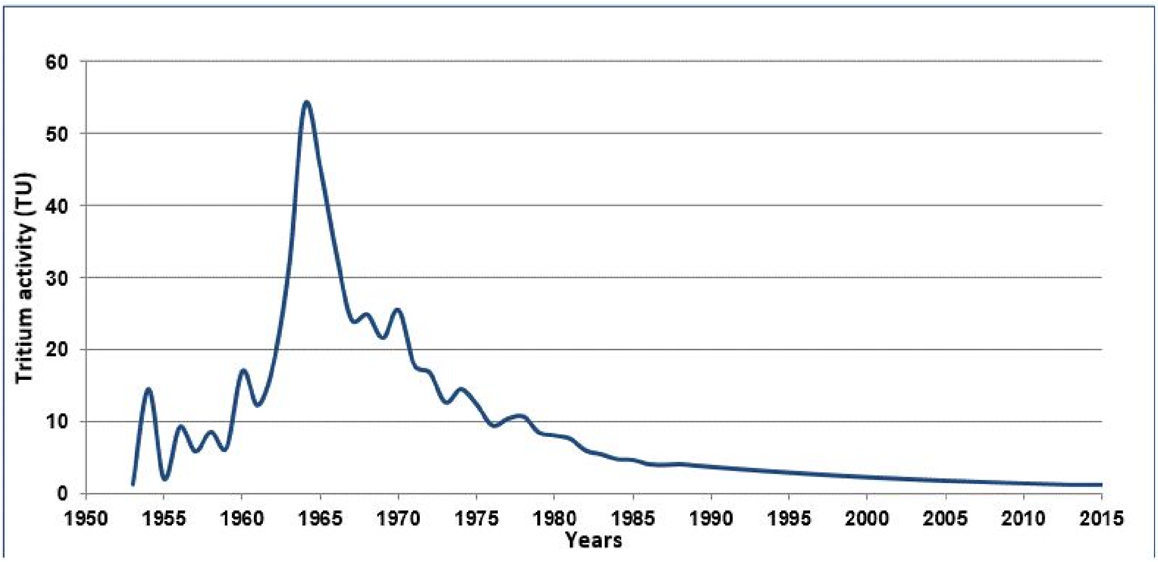

Figure 2 - Input tritium values used for the calculation.

From the exponential model of Equation 2, the renewal curve for the tritium activities was calculated as a function of the water renewal time in the aquifer, using the estimated values of input for the interval between the years 1953 and 2015. This curve is shown in Figure 3.

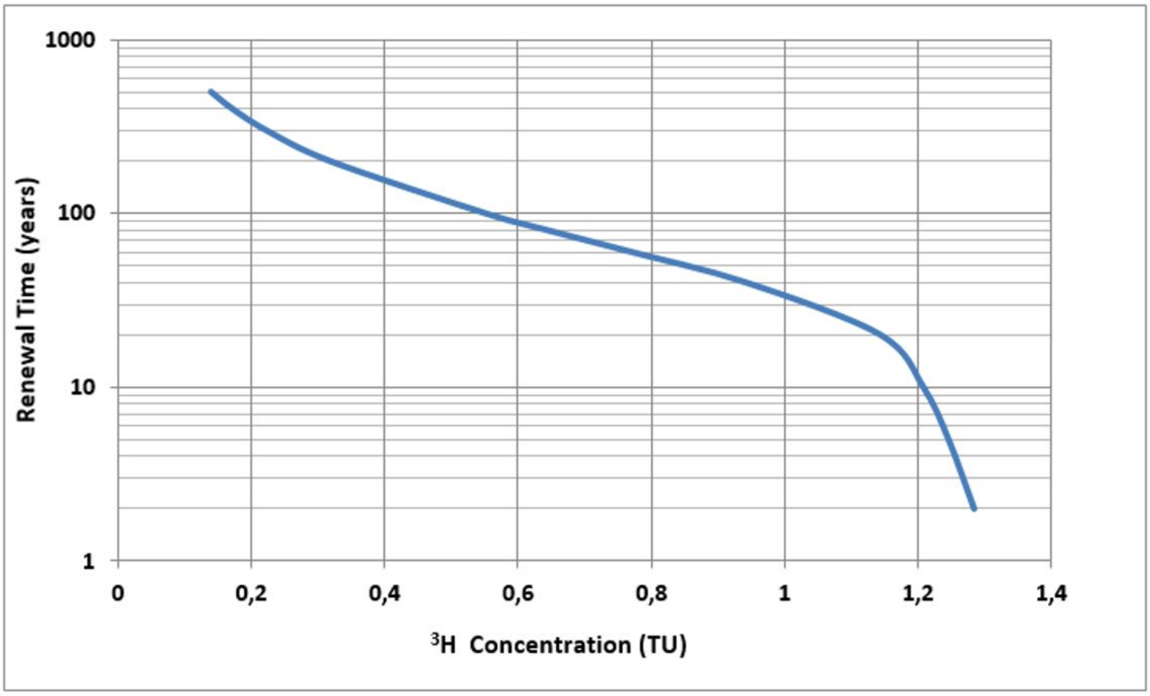

Figure 3 – Renewal time curve as a function of the ^{3}H concentration.

5. RESULTS

For the determination of ages, the tritium values measured at the groundwater sampling points were plotted on the renewal time *versus* tritium concentration curve. These results are presented in the Figure 3. It shows that the waters presenting the smallest tritium activities and, therefore, a longer time of renovation, were waters from the tubular well of the Florença Mineral Water and the Monitoring Well PM-01, with renewal times of 220 and 200 years, respectively.

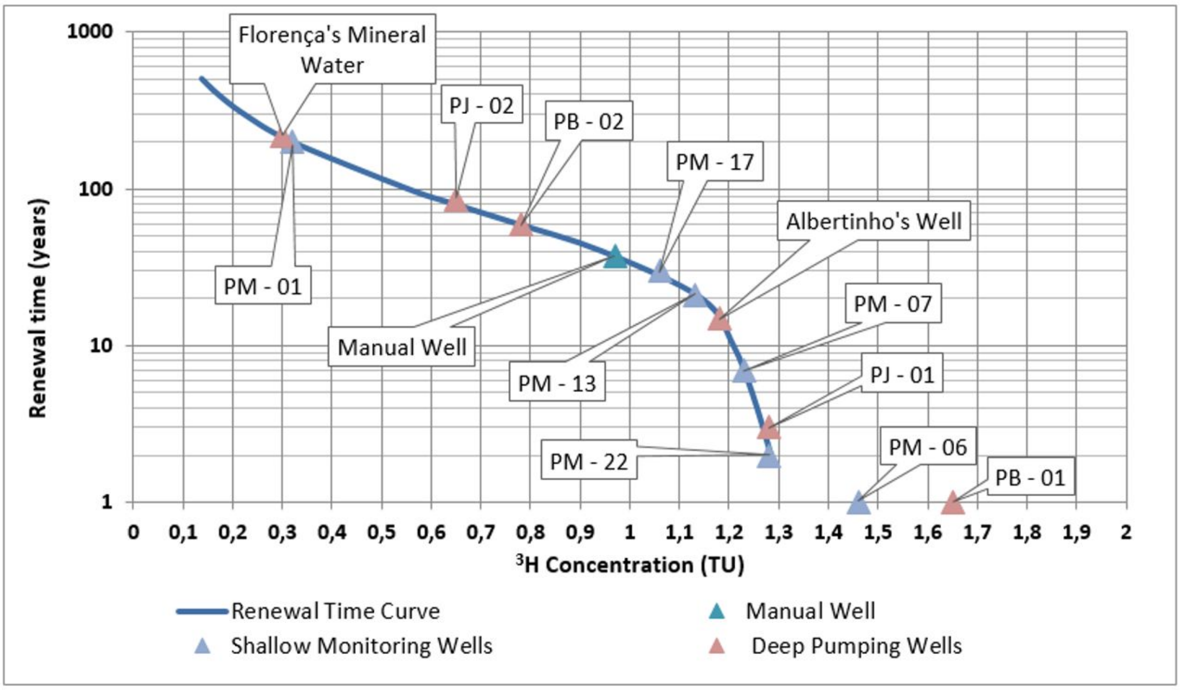

Figura 3 – Tritium results in the renewal curve.

According to Figure 3, the deep tubular wells PJ-02 and PB-02, which extract water from the confined aquifer of the Sete Lagoas Formation, presented renewal times of 85 and 60 years, respectively. Although the deep well PJ-01 also draws water from the Sete Lagoas Formation, presenting a renewal time of three years. It is assumed that part of the recharge water of this well had a value of concentrated tritium, probably coming from the lagoon Olhos d'Água (1.41 T.U.), adjacent to this well.

The deep pumping well of of Albertinho's farm, which extracts water from the fractured aquifer of Belo Horizonte Complex, presented a renewal time of 15 years, contrasting with the 220 year time of the Florença Mineral Water, which captures water from the same aquifer. This difference may be related to the differences of flows pumped by these wells. It indicates that the most productive and commercial Florença Mineral Water well has already captured the most recent waters and currently captures deeper water and with greater renewal times. The Albertinho's farm uses the well's water only for domestic use, capturing smaller volumes and, therefore, more recent waters, from the free granular aquifer superimposed to the fractured gneisses.

The waters of the shallow monitoring wells in the Serra de Santa Helena Formation, PM-17, PM-13, and manual well (cistern), placed on the free aquifer, presented renewal times in the decade's scale, with ages of 30, 21 and 37 years, respectively. The shallow monitoring wells PM-07 and PM-22 had waters with the renewal time of seven and two years.

The monitoring well PM-06 and the deep well PB-01 showed high tritium activities relative to the input value for the year 2015 (1.3 T.U.), and it was not possible to determine the time of renewal of these waters. It is possible that part of the waters captured by these wells evaporated and, therefore, the concentration of tritium in these waters has been increased in

relation to the other groundwaters.

6. CONCLUSIONS

The model results showed that the waters of PM-01 well and Florença Mineral Water are of the order of 200 years. The PJ-02 and PB-02 wells, which capture water from the deep aquifer of the Sete Lagoas Formation, are 85 and 60 years old, respectively. The monitoring wells show smaller ages, between 37 and 21 years, for wells PM-17, PM-13 and cistern. The wells PM-07 and PM-22 have the smallest ages, calculated to be seven and two years, after the recharge. The technique of water dating using radioactive isotopes is very useful in determining the renewability of water in the aquifer. This work may be useful for the better management of the water resources by the governmental agencies.

ACKNOWLEDGMENTS

Thanks to the National Nuclear Energy Commission (CNEN) for providing the scholarship for this study, to the Center for the Development of Nuclear Technology (CDTN) for its support and infrastructure, to the Brazilian Agricultural Research Corporation (EMBRAPA) and to the Florença Mineral Water, that allowed the sampling of its well.

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