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Potential applications of Water Stable Isotopes in Nuclear Facility Environmental Monitoring Programs

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1. Introduction

Nuclear facilities follow very strict operation procedures, rules, and safety guidelines[1], [2]. However, these facilities could impact the environment in unforeseen events. In 2014, an IAEA/ UNESCO meeting addressed the problem of contaminated groundwaters near Fukushima-Daiichi Nuclear Power Plant after the earthquake in March 2011[3]. This meeting highlighted the need of a detailed hydrogeological settings of nuclear facilities worldwide. In 2016, IBAMA defined specific environmental licensing procedures for nuclear and radioactive. facilities (NRF) [4] In 2018, it was defined that a Simplified Environmental Report (RAS) or a more detailed EIA - Environmental Impact Study (RIMA) should be presented [5]. In both cases, the NRF influence area must be determined and delimited. The influence area is affected in a large extent by groundwater mixing and recharging zones [6]. Since 2006, Ipen publishes the results of Environmental Monitoring Plan of non-radioactive compounds (PMA-Q). This activity consists of liquid effluent and groundwater systematic monitoring. In 2022, in a collaboration among Ipen, CDTN and UNA-Costa Rica, with the support of IAEA [7], the water stable isotopes were systematically taken at Ipen facility. The present paper identifies opportunities where water stable isotopes are applicable to assist Environmental Monitoring Plans (EMP's) and increase safety and security in nuclear facilities. The present paper includes basic concepts and principles of water stable isotope techniques application to groundwater conceptual models.

1.1 Principle and concepts

The stable isotopes are atoms of an element with larger number of neutrons than the most abundant isotope, that are stable presenting no radioactive decay. The hydrogen isotopes ^{1}H (protium) and ^{2}H (deuterium or D) are the stable isotopes, while tritium is unstable with beta decay [8]. There is no nomenclature for oxygen isotopes, the symbols are ^{16}O, ^{17}O and ^{18}O. Water stable isotopes are suitable for hydrological studies as tracers because they are not affected by rock-water interactions, like the major ions. Stable isotope values are affected by processes like evaporation or mixing, which make them a suitable tool for identifying sources of water, interactions between surface and groundwater, precipitation, etc. No concentration unity is used for stable isotopes. Instead, values of relative isotope ratios in units of per mil (‰) are used as delta (δ) notation expressed in Eq.1:

$$\delta\ {}^{i}E = \left[\frac{R_{Sample}}{R_{Reference}} - 1\right] \times 1000\ ‰ \qquad (1)$$

$$\delta D = 8 \times \delta^{18}O + 10 \quad (2)$$

where: ^{i} is the heavier isotope of the element E, R is the ratio of the abundance of the heavier to the lighter isotope of the element E, in a sample and in a reference material. For δ^{18}O and δ D, the usual reference material is the Vienna Standard Mean Ocean Water (VSMOW).

Precipitation exhibits a relationship between of δD and δ^{18}O values that is called Global Meteoric Waterline (GMWL)[8], [9], [10], expressed at Eq.2. Deviations from GMWL due to local effects derived from spatial variations are named local meteoric water line (LMWL).

2. Methodology

2.1 Stable Isotopes Data in precipitation

A database of stable isotope composition in precipitation samples from Brazil was obtained at the GNIP website [6], a data repository managed by IAEA with freely available curated data of worldwide isotopic composition. The retrieval isotopic data for Brazil precipitation samples was straightforward and easily applied for the present study. The database has 1,097 entries of D and ^{18}O from 1961 up to 2022. The isotopic composition is expressed in delta (δ) values which are used to express the relative difference of a ratio of two isotopes in a specimen compared with that of a reference, commonly an international measurement standard, as presented in eq. 1.

Data from groundwater collected at Ipen facility was used to illustrate the application of water stable isotopes in groundwater vulnerability studies. A total of 6 monitoring wells were sampled on two periods: at the middle (February) and at the end (May) of the wet season. Samples were analyzed by IRMS at CDTN.

3. Results and Discussion

3.1 Local meteoric water line

Figure 1 presents LMWL from several Brazilian cities, with data from 1961 up to 2022 [13]. This is the first step in hydrological studies [12], [14], as LMWL is the input function to groundwaters and surface waters. Deviation from the LMWL presents the signal by evaporation or specific recharge timing and mechanisms. The data from Sao Paulo was very limited (n= 44, from 1989- 1990) as in Table I. In the tropics, δ^{18}O in rainfall showed a weak dependence on air temperature (T)[8]. Precipitation event-based δ^{18}O also reflected weak relationship with (P) amount, but monthly weighted average showed a much stronger correlation, than an integrated event. From the δ^{18}O and δ^{2}H values, many studies observe that rainfall (>10 mm) is the main source of groundwater recharge, [15], [16], [17].

Table I: δD versus δ^{18}O in precipitation in Sao Paulo state and groundwater I (February) and II (May).

Line	δD vs. δ^{18}O	R^2	Period	n	Reference
LMWL-Sao Paulo	$7.59 * \delta^{18}O + 9.91$	0.918	1989-1990	44	[13]
LMWL- Rio Claro Monthly	$8.33 \pm 0.10 * \delta^{18}O + 16.43 \pm 0.80$	N/A	2013-2018	65	[14]
LMWL- Rio Claro Daily	$7.96 \pm 0.12 * \delta^{18}O + 12.93 \pm 0.80$	N/A	2016-2017	69	[14]
Groundwater I	$5.68 * \delta^{18}O - 2.27$	0.887	February, 2023	6	Present study
Groundwater II	$8.18 * \delta^{18}O + 12.05$	0.989	May, 2023	6	Present study

N/A- Not available.

3.2 Groundwater isotopic composition

The groundwater isotopic composition at the middle and at the end of the wet season (see Table I) identify seasonal variations in the groundwater recharge mechanism of the study area. The direct δ^{18}O comparison of local rainfall to groundwater reveals that (Figure 2) that groundwater recharge is strongly influenced by the more intense monthly rainfall during the wet season with a slope ~8 (very close to the GMWL slope). Moreover, despite the evaporation effects observed at the middle of wet season in some samples (plotting below the GMWL), the isotopic composition of groundwater indicates minimum influence of secondary evaporation enrichment, because of rapid infiltration processes via preferential pathways such as soil macropores and probably high atmospheric humidity in the study area [18]. As these are the preliminary assessments of the groundwater samples at Ipen, this work will continue under the scope of IAEA CRPF31007 for at least two years more. By that way we will be able to assess the full isotopic seasonality.

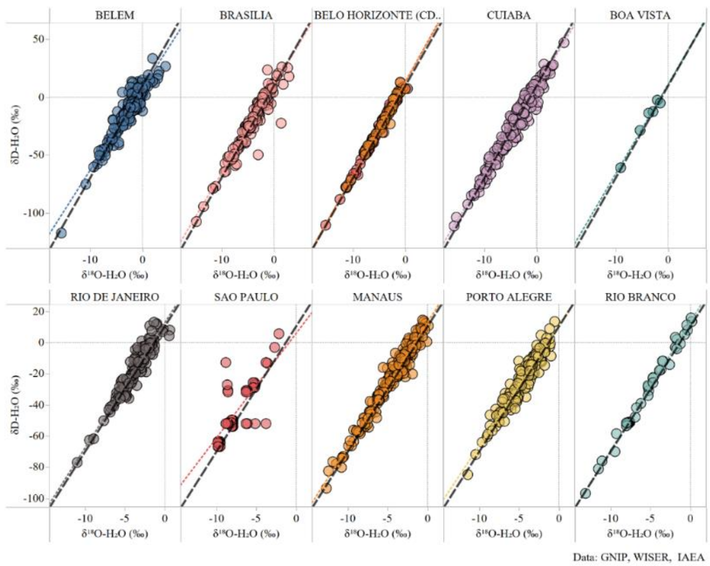

Figure 1: Local meteoric water lines from different cities in Brazil [13].

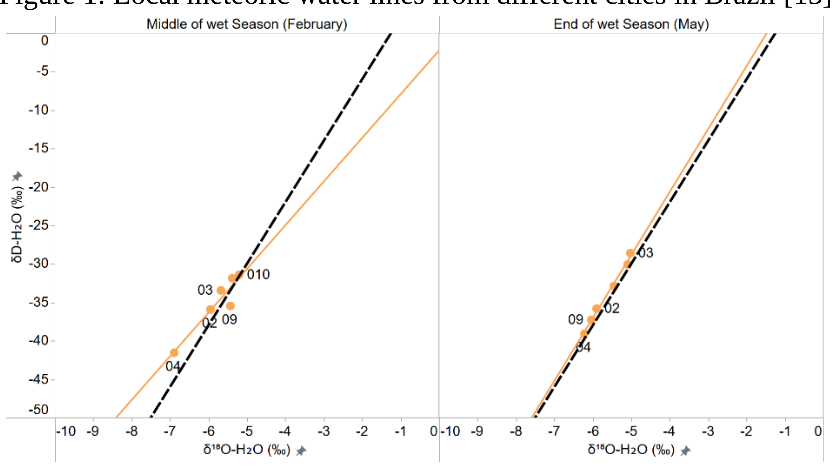

Figure 2: Groundwater isotopic signal at the middle and at the end of the wet season

(Dashed line is LMWL).

4. Conclusions

This paper presents the use of water stable isotopes ($\delta^{18}O$ and δD), in EMP supporting hydrological studies. The historical data of Sao Paulo LMWLs was compared with 6 monitoring wells isotopic composition in two seasons. $\delta^{18}O$ and δD values in groundwater samples compared to precipitation could indicate the amount rainfall contribution to the recharge. In the middle of wet season, the precipitation isotopic composition was promptly altered by high rate of evaporation, while in the wet season the intense precipitation and rapid infiltration led to similar isotopic signals among precipitation and groundwater. As a first assessment, this could indicate fast recharge, and hence, groundwater vulnerability to pollutants infiltration.

Future isotopic studies should be coupled single rain events signal to the ground water recharge and with point-source investigations such as water level monitoring, pan evaporation, infiltration, rain gauge measurements, and tracer tests to understand local-scale groundwater movement. Such studies are important for decision makers to present sustainable measures to avoid further deterioration of the groundwater.

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