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TRITIUM AS AN INITIAL TOOL FOR AQUIFER VULNERABILITY ASSESSMENT IN A KARST REGION (MINAS GERAIS, BRAZIL)

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Abstract: This study conducts an initial investigation of tritium concentrations at the *Centro Nacional de Pesquisa de Milho e Sorgo* (CNPMS) belonging to EMBRAPA, in Sete Lagoas, Minas Gerais (Brazil), to highlight tritium as a tool for contributing to the assessment of local aquifer vulnerability. Located in a karst region, the area is susceptible to contamination from anthropogenic activities, including agriculture. Water samples were collected in August 2024 from three wells and two lakes and analyzed by liquid scintillation spectrometry at CDTN. Minimum groundwater residence times were determined using the tritium radioactive decay equation, considering the regional background. Results indicate groundwater residence times ranging from 34 to 50 years, suggesting interaction with recent recharge. In contrast, one surface water sample showed a younger age (20 years). Notably, the other surface water sample exhibited a residence time of 33 years, indicating possible hydraulic connectivity with shallow aquifer zones and increased vulnerability to contamination. These preliminary findings offer initial insights fundamental for aquifer protection zoning and land-use management, while underscoring the potential of tritium as a key tracer in such evaluations. Future work will incorporate a broader isotopic dataset for a more comprehensive vulnerability assessment.

Resumo: O presente estudo realiza uma investigação inicial das concentrações de trítio no Centro Nacional de Pesquisa de Milho e Sorgo (CNPMS) da EMBRAPA, em Sete Lagoas, Minas Gerais (Brasil), visando destacar o trítio como uma ferramenta para contribuir na avaliação da vulnerabilidade do aquífero local. A região é cárstica, tornando-a suscetível à contaminação de atividades antrópicas, incluindo as práticas agrícolas. Amostras de água foram coletadas em agosto de 2024 de três poços e dois lagos, e analisadas por espectrometria de cintilação líquida no CDTN. O tempo de residência mínimo da água foi determinada pela equação do decaimento radioativo para o trítio, levando em consideração o background do isótopo para a região. Os resultados revelaram tempos de residência de 34 e 50 anos para as águas subterrâneas, indicando interação com a recarga recente. Em contraste, uma das amostras de água superficial mostrou idade mais jovem (20 anos). Notavelmente, a outra amostra de água superficial apresentou tempo de residência de 33 anos, indicando possível conectividade hidráulica com zonas aquíferas rasas e maior vulnerabilidade à contaminação. Tais descobertas preliminares oferecem percepções iniciais importantes para o zoneamento de proteção do aquífero e o gerenciamento do uso da terra, enfatizando o potencial do trítio como um traçador chave nessas avaliações. Trabalhos futuros incorporarão um conjunto de dados isotópicos mais amplo para uma avaliação de vulnerabilidade mais abrangente.

Keywords – tritium, groundwater vulnerability, karst aquifers.

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INTRODUCTION

Water is a fundamental prerequisite for human and economic development and for the maintenance of ecosystems. However, inadequate governance, investment, and mismanagement have left many without essential water services, intensifying pressure on resources, economic competition, and regional conflicts (OECD, 2009). The vital yet scarce freshwater comprises only 2.5% of global water resources; the rest is saline and essentially unusable or uneconomical for most purposes. Groundwater constitutes about 30% of this freshwater, nearly 100 times greater than that found in rivers and lakes, underscoring its critical role as a water supply source (Madramootoo, 2012).

Groundwater is integral to the hydrological cycle, with atmospheric and surface waters infiltrating to become groundwater, then reemerging in surface waters, discharging to oceans, or returning to the atmosphere through evaporation. Globally, groundwater provides half of domestic water, drinking water for most rural populations without centralized systems, and about 25% of irrigation water (UN, 2022). Nevertheless, groundwater is often undervalued, misunderstood, and mismanaged; the UN (2022) highlights overexploitation and the need for integrated management. Given its role in human health, livelihoods, economic development, and ecosystem stability, the sustainable use of groundwater is imperative in the context of increasing global water scarcity.

Karst aquifers are a critical global groundwater resource, extensively used for water supply and potable water (Stevanović, 2019; Vías et al., 2006; Andreo *et al*., 2008; Goldscheider & Drew, 2014; Goldscheider, 2003; Ford & Williams, 2008). Underlying 10–15% of Earth's land, karst aquifers supply water to an estimated 9.2% to 25% of the global population, though figures vary due to data uncertainties (Goldscheider *et al*., 2020; Ford & Williams, 2008; Stevanović, 2019). In many regions, karst aquifers constitute the sole freshwater source (Vías *et al*., 2006) and represent significant economic resources (Bakalowicz, 2005). Climate change and the degradation of surface water quality are expected to increase the reliance on karst aquifers further. However, the unique hydrogeological characteristics of karst systems make them highly susceptible to contamination from anthropogenic activities, necessitating effective protection strategies (Iván & Mádlszőnyi, 2017).

Karst is defined by distinctive hydrology and geomorphology, resulting from highly soluble rocks and well-developed secondary porosity (fractures). Sinking streams, caves, enclosed depressions, fluted rock surfaces, and large springs characterize these landscapes. High rock solubility alone does not guarantee karst formation; factors such as rock structure, lithology, and fracture density are equally critical (Ford & Williams, 2008). Karst aquifer characterization is complex due to high heterogeneity, with voids and conduits enabling rapid flow and long-distance connectivity (Bakalowicz, 2005; Bocanegra *et al*., 2005; Andreo *et al*., 2008; Foster *et al*., 2013).

According to Foster *et al*. (2013), the concept of groundwater vulnerability was first introduced by Margat (1968) and has since evolved. Albinet & Margat (1970) developed early maps of pollutant infiltration and propagation in shallow aquifers. Zwahlen (2004) refined the definitions of groundwater vulnerability by distinguishing between intrinsic vulnerability and specific vulnerability. Intrinsic vulnerability is based on hydrogeological characteristics, without considering the type of pollutant or contamination scenario. On the other hand, specific vulnerability incorporates both the intrinsic vulnerability and the properties of contaminants, providing a more comprehensive assessment. “Groundwater vulnerability” is an aquifer's susceptibility to adverse effects caused by surface-introduced contaminants. It is not universally quantifiable, as all aquifers are susceptible to contaminants (e.g., brine, nitrates), and it depends on intrinsic aquifer properties and specific contaminant nature (Foster *et al*., 2013; Zwahlen, 2004). In this context, environmental isotopes have become essential tools in hydrogeological studies, offering valuable insights into aquifer dynamics, vulnerability assessments, and water resource management.

Isotopic techniques are particularly effective in tracing the hydrological cycle and identifying contamination sources, providing independent and complementary information beyond the scope of conventional hydrogeological methods (Tazioli, 2022). Tritium (^3H), a hydrogen isotope (half-life 12.43 years), is a widely used radioactive tracer (Clark & Fritz, 1997; Canada, 2009; Calmon *et al*., 2001; Silva *et al*., 2021). It is naturally produced by nuclear reactions triggered by high-energy cosmic rays interacting with nitrogen and oxygen in the upper atmosphere (Calmon *et al*., 2001) and from anthropogenic sources, such as nuclear weapons testing. Although anthropogenic tritium represents a smaller fraction of overall radioactivity than natural background levels, it tends to be more concentrated (Silva *et al*., 2021). Significant atmospheric tritium releases from nuclear testing in the 1950s-1960s increased environmental levels, enabling its use for dating young groundwater (up to ~100 years) (Clark & Fritz, 1997; Mook, 2000; Silva *et al*., 2021). Tritium decays via beta (β^-) emission and antineutrino (ν) release, producing a stable isotope of helium (^3He) and liberating 18.6 keV of energy (Mook, 2000):

$$ {}^3_1H \rightarrow {}^3_2He + \beta^- + \overline{\nu} \quad (1)$$

Tritium behaves conservatively in hydrological systems, following water movement without undergoing chemical reactions, adsorption processes, or dissolution/precipitation dynamics. As a result, it retains the geochemical signature of its source (Telloni, 2022). Due to these distinctive properties, tritium is one of environmental studies' most widely used radioactive tracers. Its incorporation into water molecules makes it a quasi-ideal tracer for tracking water movement and estimating residence times in hydrological systems (Silva, Moreira & Cota, 2021). When tritium levels in groundwater are comparable to those in contemporary atmospheric precipitation, it indicates recent recharge. Conversely, lower tritium concentrations suggest limited or delayed recharge due to the absence of meteoric infiltration or extended groundwater residence times (Telloni, 2022).

Recent studies have emphasized the role of tritium in assessing aquifer vulnerability. Podgorski (2024) demonstrated the effectiveness of combining tritium measurements with machine learning techniques to map aquifer vulnerability in the western Sahel region. The study revealed that arid areas with pronounced precipitation seasonality, higher permeability, and deeper groundwater tables generally contain older water and exhibit lower vulnerability to contamination. Conversely, regions with higher tritium concentrations indicated recent recharge and greater susceptibility to surface-derived pollutants, including organic compounds, industrial chemicals, untreated sewage, fertilizers (nutrients), and pesticides and herbicides used in agricultural activities. Although the infiltration rates of various contaminants depend on their specific physical and chemical properties, recently recharged water generally signifies increased vulnerability.

Similarly, Hagedorn (2018) reported that the overuse of nitrogen fertilizers in California has exacerbated groundwater vulnerability, particularly in aquifers recharged within the past ~60 years, where recent infiltration increases contamination risks. Tritium-based methods have proven effective in identifying these vulnerable zones. Various groundwater vulnerability assessment methods have been developed, evaluated, and applied at test sites worldwide (Iván & Mádlszőnyi, 2017). These include the DRASTIC method (Aller et al., 1987) and the GOD method (Foster & Hirata, 1988; Foster *et al*., 2013), both commonly used for porous media and karst terrains, as well as the EPIK method (Doerfliger *et al*., 1999), which was explicitly designed for karst environments.

Hilal *et al*. (2024), for instance, further validated the use of tritium in vulnerability assessments by applying the Susceptibility Index (SI) method, derived from the DRASTIC model, to a region in Morocco. Their study highlighted the spatial heterogeneity of tritium concentrations, identifying distinct recharge zones and confirming that areas with younger groundwater correlate with higher vulnerability indices. Combining tritium analyses with models like DRASTIC enhances aquifer vulnerability assessments' spatial and temporal resolution, supporting more effective groundwater protection strategies (Mountassir *et al*., 2021).

Understanding the relationship between land use and water resource impacts is fundamental for developing preventive and corrective measures. Agricultural activities have been associated with negative impacts linked to the application of agrochemical inputs. Nonetheless, these compounds' environmental behavior and fate under tropical conditions have not been sufficiently scrutinized (EMBRAPA, 2016).

This knowledge gap hinders the development of region-specific mitigation strategies, particularly in regions where rapid infiltration pathways, such as karst systems, exacerbate groundwater contamination risks. Therefore, this exploratory study aims to investigate tritium concentrations at the *Centro Nacional de Pesquisa de Milho e Sorgo* (CNPMS) with the primary objective of showcasing tritium as a valuable initial tool for contributing insights towards evaluating the groundwater vulnerability of the local aquifer. While a comprehensive vulnerability assessment will require additional data, including other isotopic tracers and detailed hydrochemical analyses, this work specifically calls attention to the utility of tritium and the insights derivable even from simplified interpretative models, setting a baseline for future, more detailed investigations.

METHODOLOGY

Study area

The study was conducted at EMBRAPA's CNPMS (2,000 hectares) in Sete Lagoas, Minas Gerais (MG-424, km 65), ~70 km from Belo Horizonte. In addition to being situated within a karst region potentially vulnerable, the CNPMS hosts experimental agricultural activities, making it suitable for research on environmental impacts. The CNPMS is entirely within the Jequitibá River sub-basin, which is part of the middle section of the Velhas River basin, itself a sub-basin of the São Francisco River watershed. The area is traversed by two perennial streams belonging to the Jequitibá River sub-basin: the Matadouro Stream, which crosses the northwestern portion of the area, and the Marinheiro Stream, which crosses its southwestern portion.

The CNPMS is situated within the São Francisco Craton, where clay-carbonate sediments from the Sete Lagoas and Serra de Santa Helena formations were deposited, forming the Bambuí Group (Ribeiro *et al*., 2003). These sedimentary units overlie the basement rocks of the Belo Horizonte Complex, which is composed primarily of gneissic rocks associated with granitic and migmatitic zones. The Sete Lagoas Formation includes the Pedro Leopoldo Member (fine-grained limestones, dolomites, marls, pelites with dissolution macrostructures) and the upper Lagoa Santa Member (medium-grained dark limestones with dissolution features in caves and caverns, indicating the presence of paleo-conduits (Galvão *et al*., 2015). The Serra de Santa Helena Formation comprises slates, marbles, siltstones, claystones, and quartz veins. These sedimentary units are overlain by unconsolidated Cenozoic deposits consisting of sandy-clayey sediments with gravel layers, semi-consolidated clayey-sandy materials, and fine to coarse sands (Tuller *et al*., 2010; Galvão *et al*., 2016).

The geomorphology reflects karst geology, characterized by sinkholes, caves, lakes, ponds, and poorly developed surface drainage systems. The occurrence of sinkholes is considered indirect evidence of subsurface karstified limestone. The landscape is marked by undulating and hilly relief, with drainage lines often confined between cliffs and valleys, particularly in areas underlain by rocks of the Serra de Santa Helena Formation (Pessoa, 1996).

Karst, fractured, and granular are the three main types of aquifers in the study area (Pessoa, 1996). Karst aquifers are the most significant and are developed within the Sete Lagoas Formation, which comprises fine-grained limestones from the Pedro Leopoldo Member and medium-grained limestones from the Lagoa Santa Member. Recharge is autogenic (sinkholes, caves) and allogenic (limestones that are overlain by unconsolidated Cenozoic sediments), primarily from precipitation concentrated between October and December (Pessoa, 1996; Galvão *et al*., 2017).

Sampling points

Two samples were obtained from surface water bodies (lakes), and three were collected from groundwater sources (wells) (Figure 1). The physical-chemical parameters of all samples were measured in the field using a portable multiparameter probe. The water samples collected in August 2024 (dry season) were analyzed at the Laboratory of Environmental Tritium (LTA) at CDTN. Tritium content can be expressed either in conventional radioactivity units (Bq/L, Ci/L) or in Tritium Units (TU), where 1 TU corresponds to a $\frac{^3H}{^1H}$ ratio of 10^{-18} (Clark & Fritz, 1997; Canada, 2009).

Laboratory analysis

Due to the low tritium concentrations in water and the low energy of its beta emissions, the isotope must be enriched prior to analysis via electrolysis. Before electrolysis, samples undergo preliminary distillation to reduce electrical conductivity to 3.5 $mS \cdot cm^{-1}$. The mass difference among tritium (^3H), deuterium (^2H), and protium (^1H) leads to different hydrogen evolution reaction rates during the electrolysis-induced decomposition of water molecules. This results in the enrichment of heavier isotopes in the remaining liquid phase. After electrolysis, the sample goes through a second distillation step before being incorporated into the measurement system. The tritium concentrations obtained correspond to the levels of this isotope present in the water at the time of sampling.

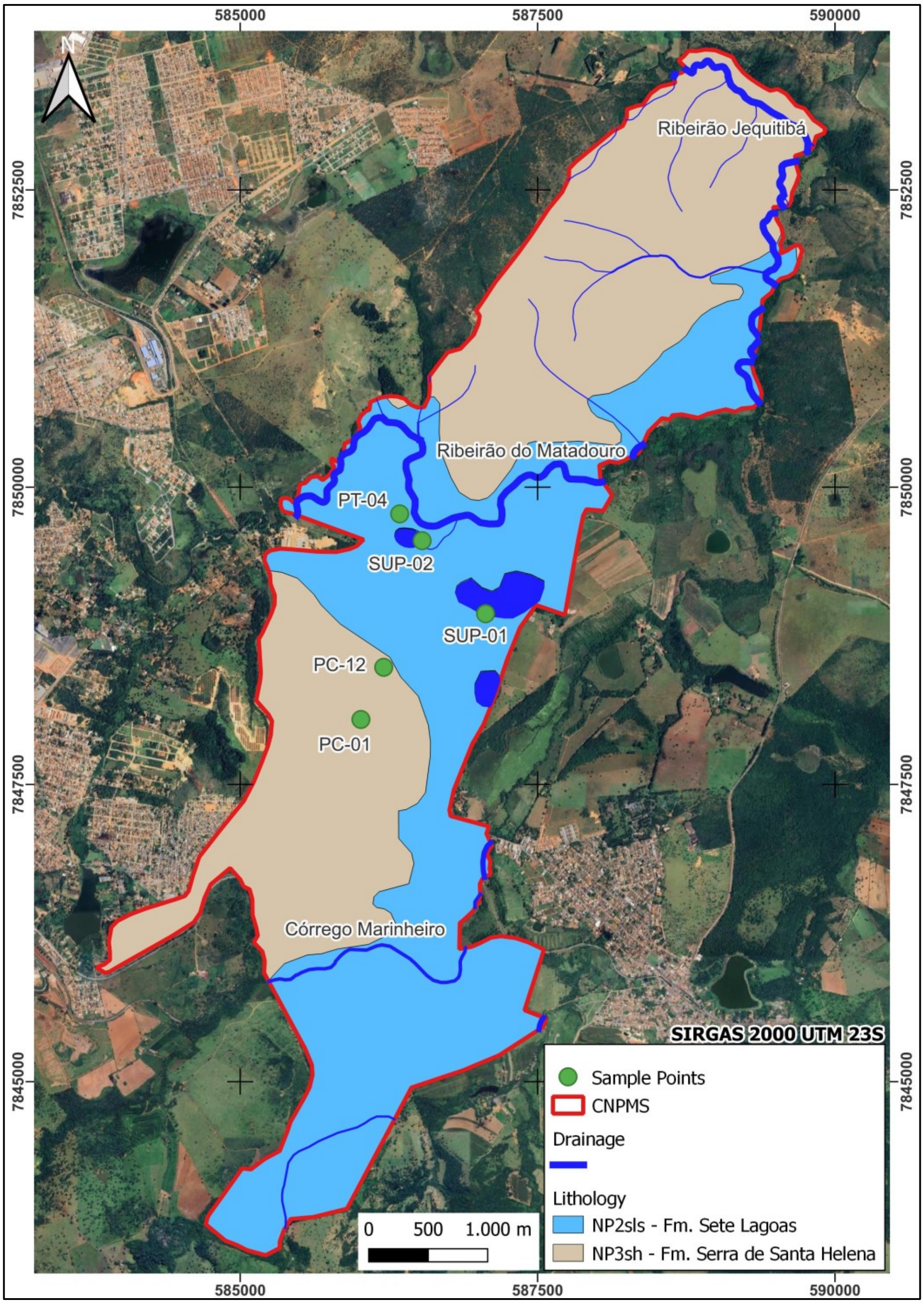
The analytical technique applied was liquid scintillation spectrometry using the Quantulus™ 1220. This instrument is recognized for its high efficiency in detecting low-energy, low-penetration beta radiation emitted by tritium (detection limit of 0.3 TU). During liquid scintillation counting, the sample is mixed with a cocktail that emits photons upon interaction with the β-radiation emitted by tritium. These photons are detected by photomultiplier tubes and converted into electrical pulses. The methodology employed to determine the minimum water residence time for the collected samples was based on the radioactive decay equation for tritium.

The approach involved using the measured tritium concentration in the samples ($CT_{^3_1H}$) in combination with a background tritium concentration ($BG_{^3_1H}$) value, derived from Silva & Cota (2021), as a reference for the initial isotope concentration. These authors reconstructed the tritium concentration curve for the CDTN station, which is relevant to the study area, based on data provided by the International Atomic Energy Agency (IAEA) through the Global Network of Isotopes in Precipitation (GNIP) monitoring program:

$$Minimum_{Residence\ Time} = -\frac{\ln\left(\left[{CT_{^3_1H}}/{BG_{^3_1H}}\right]\right)}{\lambda_{^3_1H}} \qquad (2)$$

It is important to note that this method, based on the simple radioactive decay equation, estimates the *minimum* residence time. This approach offers a valuable first approximation, particularly for highlighting recent recharge. However, it does not account for complexities such as groundwater mixing or variations in initial tritium concentrations beyond the assumed background. Therefore, while these results are instrumental in this exploratory phase to demonstrate tritium's applicability, they represent a simplification. Future analyses will aim to incorporate Lumped Parameter Models (LPMs) for a more refined interpretation of residence times, considering such hydrological complexities.

Figure 1 – Sampling points



RESULTS AND DISCUSSION

The sample description and results are summarized in Table 1. The PC-01 well sample (<0.30 TU), with a tritium concentration below the detection limit, indicates the oldest minimum groundwater residence time, approximately 50 years. This provides strong evidence that the water has remained isolated from recent recharge for an extended period. The PC-12 (0.74 TU) and PT-04 (0.64 TU) well samples yield estimated minimum residence times of approximately 34 and 36.5 years, respectively. These results are consistent with groundwater systems characterized by relatively slow renewal rates. For PC-01, Linhares (2017), using LPM, reported longer residence times of 135 years (0.32 TU) and 120 years (0.35 TU).

The SUP-01 lake sample (1.63 TU) has an estimated minimum residence time of approximately 20 years. This outcome reflects the dynamic nature of surface water bodies, as lakes typically undergo constant and rapid exchange with the atmosphere and precipitation. Consequently, these systems generally exhibit short water residence times and younger water ages. For the same point, Linhares (2017) reported much shorter residence times—2 years (1.88 TU) and 5 years (1.56 TU).

Conversely, the SUP-02 lake sample (0.77 TU) presents an estimated residence time of approximately 33 years, the highest calculated among the lake samples, with a tritium concentration comparable to that observed in PC-12. This result may suggest a significant contribution of older groundwater to the lake or, alternatively, a slightly longer residence time than lakes with faster hydrological turnover. Supporting this, Linhares (2017) found a residence time of 46 years (0.85 TU) for the same lake area.

Linhares (2017) also analyzed groundwater samples from wells not included in the present study, reporting the following residence times: PC-06 — 9 years (1.60 TU); PC-07 — 19 years (1.33 TU) and 33 years (1.13 TU); PC-13 — 23 years (1.28 TU); PC-17 — 28 years (1.19 TU and 1.20 TU); and PC-22 — 16 years (1.42 TU). Differences between the results may be attributed to the methodological approaches employed in each study, particularly the use of LPM versus simple decay equations. It underscores the sensitivity of age estimations to the chosen model, emphasizing that the minimum ages reported here are indicative and primarily demonstrate tritium's utility as a tracer. The discrepancies further justify the planned future application of LPMs to this study's expanded dataset for a more robust comparison and refined age interpretations.

Table 1 – Water minimum residence time

Code	Source type	E.C. (μS/cm^2)	T (°C)	pH	Tritium (TU ± 2σ)	Minimum Residence Time (years)
PC-01	Well	325	25.62	4.43	<0.30 ± —	≈50
PC-12	Well	8	25.70	4.44	0.74 ± 0.21	≈34
PT-04	Well	346	27.9	7.42	0.64 ± 0.15	≈36.5
SUP-01	Lake	106	30.76	7.48	1.63 ± 0.20	≈20
SUP-02	Lake	325	29.07	7.3	0.77 ± 0.16	≈33

The minimum groundwater ages calculated for the wells, ranging from approximately 34 to 50 years, indicate that groundwater has relatively short residence times. This suggests that water requires a few decades to infiltrate from the surface and reach the sampled portions of the aquifer. The predominance of modern groundwater, as suggested by these initial estimations using a simple decay model, may be viewed as a negative indicator of water quality concerning current surface contamination. Especially in the karst context, it implies that contaminants could reach these groundwater reserves fast, and remediation would be technically challenging due to the aquifer's complexity. Furthermore, the minimum age of 33 years observed for one of the lake samples, comparable to one groundwater sample, suggests a possible hydraulic connection between the lake and the underlying aquifer. This interaction could increase the vulnerability of a shallow aquifer influenced by the lake, which is more directly exposed to surface contamination risks.

CONCLUSIONS

This exploratory study successfully demonstrated the utility of tritium as an initial tool for assessing groundwater dynamics and potential vulnerability indicators at the CNPMS. Tritium analyses revealed minimum groundwater ages, calculated using a simplified radioactive decay model, ranging from 34 to 50 years, indicating interaction with recent recharge. While these ages are considered preliminary and represent minimums due to the model's inherent limitations (e.g., not accounting for mixing), they effectively flag areas with relatively younger groundwater. Detecting a lake sample (SUP-02) with residence time comparable to groundwater (PC-12) indicates possible hydraulic connectivity between surface water and shallow aquifer zones, increasing their susceptibility to contamination. These initial findings are critical for informing future, more detailed groundwater protection zoning and land-use management. Special attention should be given to recharge areas supplying older groundwater, as contamination in these zones may have long-lasting impacts. To build upon this foundational work, future investigations will involve expanding the tritium dataset, incorporating analyses of other environmental isotopes ($\delta^{18}O$, $\delta^{2}H$, and noble gases), and integrating hydrochemical data. The application of LPM is planned to refine residence time estimates, which will be compared against existing studies to enhance hydrogeological assessments and provide a more robust basis for a detailed vulnerability evaluation. This study, therefore, serves as an important first step, highlighting the importance of isotopic monitoring, particularly tritium, for aquifer dynamics elucidation and sustainable groundwater management, calling attention to its value even in preliminary assessments and setting the stage for more comprehensive investigations.

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