Seasonal Fluctuations in the Concentration of Natural Radionuclides in Groundwater of Al Dhahirah Governorate, Oman

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This study investigated the influence of seasonal variations on radionuclide concentrations in groundwater from eight wells in Al Dhahirah Governorate, Oman. To evaluate the long-term radiological impact on the region's drinking water quality, samples were taken monthly from December 2021 to November 2022. A gamma spectrometry system with a hyper-pure germanium detector (HPGe), with a 60% efficiency, measured the activity concentrations of U-238, Th-232, and K-40. The results showed a clear trend: monsoon seasons (September–November) led to a significant rise in radionuclide concentrations due to increased interaction between water and rock formations. Conversely, dry periods (March–May) displayed the lowest concentrations because of the limited water available for such interactions. Importantly, the annual average concentrations of U-238 (0.042 Bq/L), Th-232 (0.042 Bq/L), and K-40 (0.038 Bq/L) were below the safety limits set by the World Health Organization's (WHO). This indicates that the groundwater in these wells is currently safe for drinking. However, the study emphasizes the importance of considering seasonal variations when monitoring groundwater quality. Additionally, it highlights the need for further research into potential long-term health effects from exposure to these radionuclides and the development of effective water management strategies, particularly during monsoon seasons.

Keywords: Groundwater, Leaching, Monsoon rains, Radionuclide concentration, Seasonal variations.

1. Introduction

Groundwater, as a sustainable source of fresh water, strategically fulfills the growing needs of an expanding population. Its pivotal role is especially notable in arid and semiarid areas where surface water is scarce. However, climate change, with its anticipated changes in precipitation patterns and increased frequency of droughts, is imposing significant pressure on groundwater reserves [1, 2]. Consequently, evaluating and continuously monitoring the quality status of these resources is critical to ensuring a clean and sustainable water supply for human consumption. Human actions and certain natural occurrences both contribute to the pollution and alteration of water quality. Water pollution extends beyond the discharge of substances like chemicals or microorganisms and can also involve the emission of energy, such as radioactivity or heat, into water sources [3].

Radioactive substances are naturally present in the environment, contributing to environmental radiation from a diverse range of both natural and anthropogenic sources. Naturally occurring radioactive materials (NORMs), mostly made up of U-238, Th-232 series, and K-40, as well as their decay products [4], are the main sources of gamma radiation in natural water sources. The radionuclides that occur naturally in groundwater come from the minerals in the soil and bedrock. The hydrogeological and physicochemical characteristics of the aquifer primarily affect the levels of these radionuclides in the water [1, 5]. Furthermore, precipitation can carry radioactive particles from the atmosphere to the soil and into the water supply. Precipitation can become radioactive if the atmosphere is polluted with radioactive particles. This leads to pollution of the soil and water supplies [6]. The occurrence of radionuclides in groundwater can lead to health issues if they accumulate in the human body through the consumption of contaminated water. Dissolved radionuclides can emit various alpha and beta particles as well as photons in water, potentially causing substantial effects on the tissues and cells within the human body [7]. When exceeding particular concentrations, radioactive materials in groundwater can cause serious carcinogenic health problems, with bone cancer being the most frequent consequence [8]. The World Health Organization (WHO) suggests a maximum permissible concentration of 30 μg/L for U-238 and 250 μg/L for Th-232 in drinking water [9].

Numerous studies from various countries show that seasonal fluctuations in both rainfall and aquifer capacity can cause significant variations in groundwater chemistry [2, 4, 10–12]. Consequently, fluctuations in seasons can directly impact the quality of water, and seasonal changes can also have a considerable impact on the concentration of radionuclides in aquatic systems [4, 8, 12–14].

The study area is defined by its geological features, primarily consisting of rocky desert plains and mountains. Additionally, the prevailing weather conditions are characterized by high temperatures and relatively low levels of precipitation. Therefore, this present study focused on the fluctuation of radionuclide concentrations in drinking water derived from underground water resources in the Al Dhahirah Governorate concerning seasonal variations, marking an initial effort of its kind within this geographic area.

2. Methodology

2.1 Study area

The Al Dhahirah Governorate located in the north-central region of Oman, at 23° 12′ 58.2

North and 56° 29′ 26.5" East [15], as illustrated in Figure 1. This region's diverse geology, encompassing Precambrian rocks to sedimentary formations, makes it a unique location to study seasonal variations in groundwater radionuclide concentrations. This region relies heavily on groundwater, making it crucial to understand the potential impact of seasonal monsoons on the mobilization of naturally occurring radioactive materials within the aquifers. The underlying geological formations themselves may harbor these radioactive elements, highlighting the importance of investigating how seasonal variations in rainfall and water flow might influence groundwater quality and the safety of this vital freshwater resource for local communities. The depths of the selected wells range from 92.5 m to 150 m. In Oman, rainfall is scant and sporadic across most of the country. On average, rainfall in the coastal plains and deserts is less than 50 mm per year, while in the mountains it can exceed 350 mm [16]. Al Dhahirah experiences a hot desert climate with distinct seasonal variations. Summer months are scorching, with high temperatures and limited rainfall. In contrast, winter brings milder temperatures and occasional precipitation, with the monsoon season typically occurring between July and September [17]. This seasonal variation in rainfall is a significant factor influencing groundwater recharge and potentially impacting radionuclide mobilization.

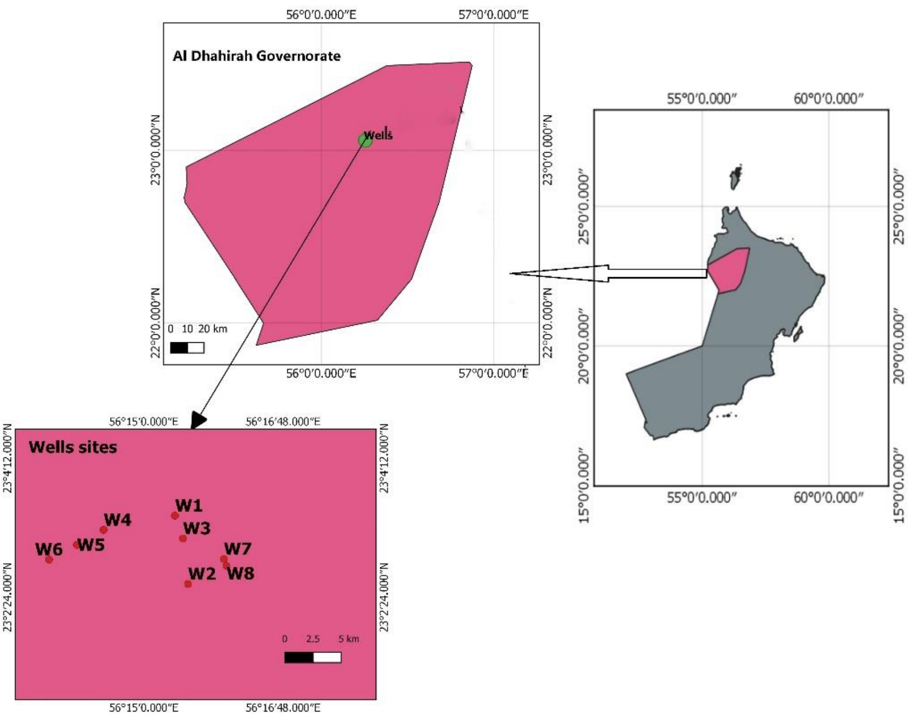

Figure 1: Map created with Quantum Geographic Information System (QGIS) software showing a map of Oman and the specific locations of the samples

2.2 Sample collection and preparation

To ensure consistent and reliable water quality data, sample collection procedures in this study followed the established guidelines set forth by the American Public Health Association (APHA) [18]. In this investigation, a total of eight wells (W1 to W8) situated within the Al

Ain Valley (Al Masaraat Basin) were selected for groundwater analysis. The study utilized data from 96 samples collected monthly between December 2021 and November 2022. A systematic random sample method was used for groundwater sampling. The groundwater samples were collected in sealed polyethylene bottles (labeled with location, date, and time) from each well after purging for 10 minutes and transported in a chilled polystyrene box to the laboratory for analysis. Upon arrival at the Sultan Qaboos University Gamma Spectroscopy Laboratory (College of Science), the samples underwent preparation for analysis. This involved transferring each sample to a pre-cleaned 1-liter Marinelli beaker. These beakers were meticulously decontaminated with a 10% hydrochloric acid solution, rinsed thoroughly with de-ionized water, and then air-dried. To minimize CO_2 interference and radionuclide loss, bottles were filled to capacity before secure sealing within the beakers using masking tape [19]. A 30-day storage period allowed for radioactive decay and establishment of secular equilibrium between parent and daughter radionuclides [20].

2.3 Radioactivity Measurements

Analysis of the water samples was performed utilizing a gamma spectrometry system with a hyper-pure germanium coaxial detector (Baltic Scientific Instruments [BSI]). This detector's high efficiency (60%) and energy resolution (1.8 keV at 1332 keV, with a peak of Co-60) enabled accurate detection and quantification of gamma-emitting radionuclides. Cooled with liquid nitrogen, the detector operated at +3700 V and offered a wide detection range for gamma rays up to several MeV. Regular energy and efficiency calibrations were ensured using certified mixed gamma standards (Eckert & Ziegler Analytics). Background radiation was routinely assessed through blank measurements under identical sample settings. Subsequently, each sample's gamma spectrum was corrected for background interference. Genie 2000 (version 3.0) software (Canberra) facilitated data acquisition, display, and analysis of the acquired gamma-ray spectra. The samples were counted for a prolonged duration (72,000 seconds) to enhance sensitivity. Following data acquisition, spectra were meticulously examined for the presence of radionuclides.

Direct detection of U-238 and Th-232 activity proved challenging due to their low-energy photon emissions and slow decay rates. Consequently, an indirect approach was employed. The photopeak gamma energies of their progeny were analyzed, assuming they were in secular equilibrium. For U-238, the average relative intensities of gamma-rays from Pb-214 (295.21 keV and 358.91 keV) and Bi-214 (609.3 keV) were used to estimate its activity. Similarly, Th-232 activity was determined using the weighted mean of gamma-ray intensities from Ac-228 (911.1 keV), Pb-212 (238.6 keV), and Tl-208 (583 keV). Conversely, K-40 activity was directly measured through its characteristic gamma-ray line at 1460.8 keV [21].

3. Results and Discussion

The most reliable method for detecting and examining the gamma radiation that radioisotopes emit is gamma-ray spectroscopy. To explore how varying seasons affect the concentrations of radionuclides, a series of four 3-month sampling periods were carried out between December 2021 and November 2022 (detailed in Table 1). Monthly samples were collected from each location to ensure adequate data for subsequent statistical analysis. These samples were then used to calculate the mean concentrations of radionuclides in each period. Table 1 provides an overview of the mean specific activity concentrations of U-238, Th-232, and K-40 in all locations for each specific period during the year-long study (12 months). Additionally, it presents the mean, minimum, maximum, and standard deviation values associated with these measurements.

The data showed notable fluctuations in the average radionuclide activity levels in the water samples examined during this study. Notably, there were increases observed in the fourth period, aligning with the monsoon season from September 2022 to November 2022. During this particular period, there was a substantial increase in the concentrations of U-238, Th-232, and K-40; the activity concentrations ranged from 0.049 to 0.092 Bq/L for U-238, 0.053 to 0.099 Bq/L for Th-232, and from 0.020 to 0.088 Bq/L for K-40. The monsoon season typically begins in mid-August, and the initial rainfall during this time has the potential to cause significant variations in the observed concentrations compared to the pre-rain readings. However, it's in September that monsoon activity reaches its peak, with precipitation amounts ranging from 25 to 61 mm [22]. Various research studies [6, 12, 23, 24] show good agreement with the current findings, indicating that during periods of precipitation, there is a notable increase in radionuclide concentrations. Monsoonal rains significantly influence the concentration of naturally occurring radionuclides in groundwater due to enhanced leaching and potential geological interactions [12, 25]. Monsoonal rains can be likened to a geochemical process that enhances the dissolution of radioactive materials within the geological matrix. This mobilized radioactive material is then transported by the increased water flow, leading to a rise in radionuclide concentrations in groundwater sources. Additionally, the monsoon's interaction with geological formations might further promote radionuclide release. For instance, increased water flow could expose fresh rock surfaces containing higher radioactive material or facilitate chemical reactions that release these elements [12].

In contrast, the second period (March-May 2022), coinciding with the dry meteorological period, witnessed the lowest activity concentrations. Concentrations of U-238, Th-232, and K-40 ranged from 0.010 to 0.033 Bq/L, 0.010 to 0.016 Bq/L, and from 0.009 to 0.017 Bq/L, respectively. Drier periods experience less leaching as there's limited water to dissolve the radioactive materials. Furthermore, existing groundwater might become diluted by minimal rainfall, lowering the overall concentration of dissolved radionuclides [12, 25].

In the case of rainfall, peaks in radionuclide levels are often noticeable as sharp, temporary surges, quickly reverting to normal background levels or even dipping momentarily below them, as indicated in Figure 2. These increases can be attributed to the leaching of radionuclides during the infiltration of rainwater [25]. As per the literature [4, 6, 12, 13, 23, 25, 26], radiation levels are affected by numerous factors beyond just precipitation. One significant factor is the presence of radon (originating from the U and Th series) and its decay products, naturally occurring substances that can be captured by precipitation and subsequently deposited on the ground, resulting in a temporary increase in radiation levels.

Table 2: Average specific activity concentrations (Bq/L) of U-238, Th-232, and K-40 in each period over the entire study year in all studied groundwater samples

Sample ID	Concentrations of radionuclides (Bq/L)											
	P1 (Dec 2021- Feb 2022)			P2 (March 2022- May 2022)			P3 (Jun 2022- Aug 2022)			P4 (Sept 2022- Nov 2022)		
	U-238	Th-232	K-40	U-238	Th-232	K-40	U-238	Th-232	K-40	U-238	Th-232	K-40
W1	0.057	0.049	0.079	0.026	0.015	0.013	0.036	0.026	0.049	0.064	0.097	0.088
W2	0.053	0.053	0.067	0.033	0.013	0.016	0.054	0.018	0.051	0.062	0.066	0.073
W3	0.037	0.074	0.046	0.024	0.014	0.017	0.037	0.055	0.058	0.087	0.077	0.079
W4	0.062	0.036	0.067	0.016	0.015	0.01	0.041	0.021	0.017	0.092	0.074	0.070
W5	0.035	0.037	0.019	0.01	0.016	0.009	0.016	0.039	0.012	0.049	0.054	0.038
W6	0.056	0.045	0.032	0.012	0.01	0.014	0.018	0.020	0.045	0.061	0.087	0.061
W7	0.032	0.063	0.027	0.025	0.012	0.015	0.031	0.017	0.019	0.073	0.099	0.046
W8	0.037	0.052	0.018	0.011	0.012	0.010	0.031	0.013	0.015	0.073	0.053	0.020
mean	0.046	0.051	0.044	0.020	0.013	0.013	0.033	0.026	0.033	0.070	0.076	0.059
minimum	0.032	0.036	0.018	0.010	0.010	0.009	0.016	0.013	0.012	0.049	0.053	0.020
maximum	0.062	0.074	0.079	0.033	0.016	0.017	0.054	0.055	0.058	0.092	0.099	0.088
Standard deviation	0.012	0.013	0.024	0.009	0.002	0.003	0.012	0.014	0.019	0.014	0.018	0.023

*P: Period, *W: Well

In summary, it was observed that weather conditions, particularly seasonal variations, influence the radionuclide contents. Conducting research during or immediately after heavy rainfall yields artificially elevated radionuclide levels. Conversely, conducting research during the summer season, when there is no precipitation, results in decreased radionuclide measurements. The monsoon period intensifies various hydrological and geological processes, such as leaching, erosion, and groundwater recharge, which significantly influences the concentrations of U-238, Th-232, and K-40 in water sources. These processes collectively result in increased levels of radionuclides during the monsoon season. However, in dry weather, geological stability increases, leading to reduced radionuclide release from geological formations into the water. Moreover, dry weather typically means decreased levels of aerial deposition, which could introduce radioactive particles and ions to the ground and possibly elevate radionuclide levels in water. These factors collectively contribute to the observed minimum levels of radionuclide activity concentrations during dry meteorological periods.

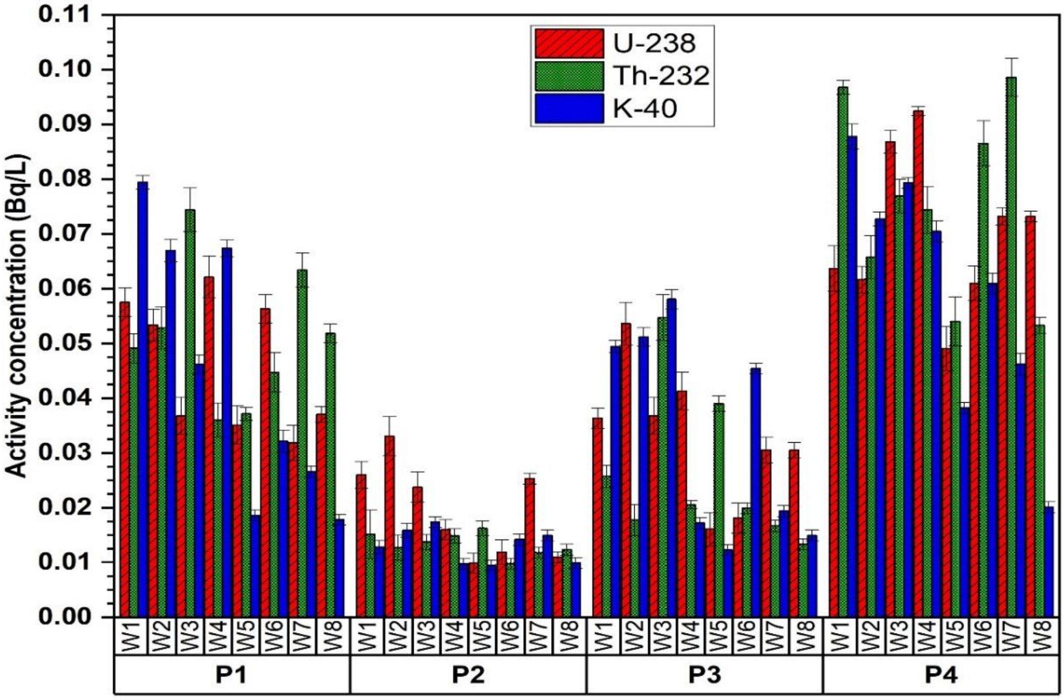

Figure 2: Comparison of average activity concentrations of U-238, Th-232, and K-40 in all locations during different periods

Furthermore, an annual assessment of radionuclide concentrations was conducted for all months across various sites, with detailed results presented in Table 2 and Figure 3. The analysis revealed that the annual average concentrations of the three radionuclides (U-238, Th-232, and K-40) were measured at 0.042, 0.042, and 0.038 Bq/L, respectively. Importantly, these measurements were found to be below the WHO's recommended limit for drinking water [27]. In this study, U-238 concentrations ranged from 0.028 to 0.053 Bq/L, with the lowest concentration observed in water samples from Well-5 and the highest in samples from Well-4. However, the calculated annual average activity concentrations of Th-232 ranged from a maximum of 0.055 Bq/L in Well-3 to a minimum of 0.033 Bq/L in Well-8. Notably, there were no significant differences in the annual average activity concentrations of U-238 and Th-232. This consistency can be attributed to the similar geological characteristics of the sampled locations, as geological composition stands as the primary factor influencing the presence of radionuclides in water [28]. However, the annual average activity concentrations of K-40 in the groundwater samples showed a wider range, varying from 0.016 to 0.057 Bq/L. Among the samples, Well-1 exhibited the highest activity concentration, while Well-8 recorded the lowest. The presence of K-40 in drinking water can be traced back to the release of K-40 into water bodies due to interactions between water and geological materials, including rocks and soil [29]. Overall, the levels of U-238, Th-232, and K-40 found in all the examined samples were far below the WHO's maximum allowable limits for drinking water [27].

Table 3: Annual average activity concentrations (Bq/L) of U-238, Th-232, and K-40 from all months of the study in all studied groundwater samples

Sample ID	Annual concentration (Bq/L)		
	U-238	Th-232	K-40
W1	0.046	0.047	0.057
W2	0.051	0.038	0.052
W3	0.046	0.055	0.050
W4	0.053	0.037	0.041
W5	0.028	0.037	0.020
W6	0.037	0.041	0.038
W7	0.040	0.048	0.027
W8	0.038	0.033	0.016
mean	0.042	0.042	0.038
minimum	0.028	0.033	0.016
maximum	0.053	0.055	0.057
standard deviation	0.008	0.008	0.015

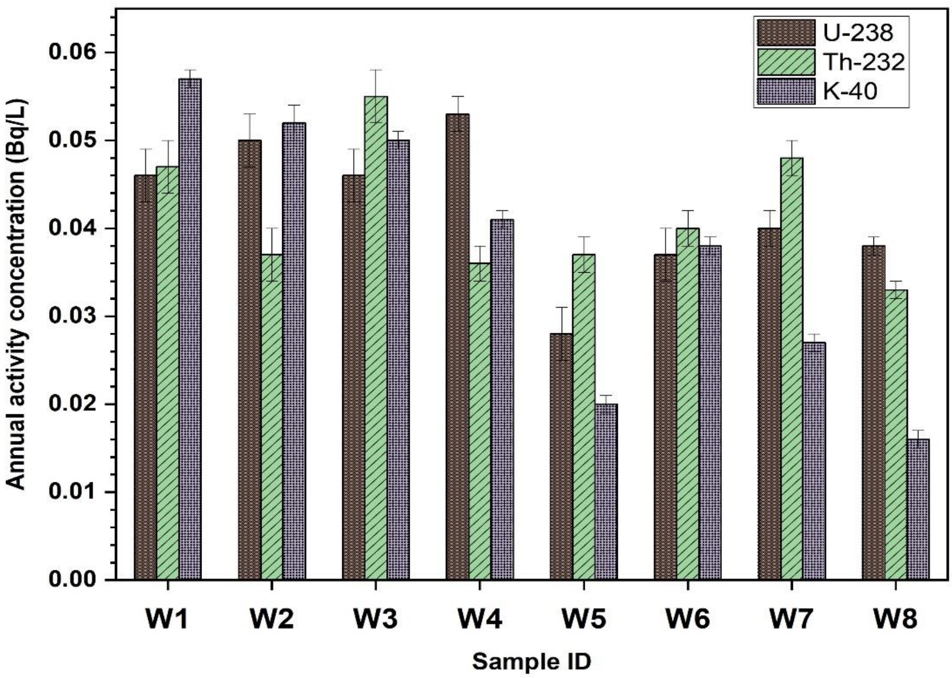

Figure 3: Average activity levels of U-238, Th-232, and K-40 throughout the year, derived from groundwater samples analyzed across all months

The study revealed fluctuations in radionuclide levels in water samples, with significant increases during the monsoon season due to leaching and interactions with geological formations, while drier periods exhibited lower radionuclide concentrations. These results show that weather conditions, especially seasonal changes, affect the radionuclide content. Additionally, annual measurements showed that the levels of U-238, Th-232, and K-40 in the examined samples were below the WHO's recommended limits, which means that the underground water sources are suitable for both household and agricultural purposes.

4. Conclusion:

This study examined the seasonal fluctuations of radionuclide levels (U-238, Th-232, and K-40) in groundwater samples from eight wells in Al Dhahirah Governorate, Oman, over one year (December 2021-November 2022). The findings highlight the significant influence of seasonal changes, particularly during the monsoon season, on the dynamic movement of radionuclides within the studied aquifers. Seasonal variations affected radionuclide levels, with the monsoon season (September-November 2022) showing higher concentrations of U-238, Th-232, and K-40 due to increased leaching and potential geological interactions. In contrast, the dry period (March-May 2022) exhibited lower levels of radionuclides due to the limited water availability for leaching and potential dilution effects. Importantly, the annual average concentrations of U-238, Th-232, and K-40 remained below the safety limits set by WHO for drinking water, indicating the safety of these groundwater sources for consumption. However, the observed seasonal increases in radionuclide levels during the monsoon season emphasize the need for further investigation into potential long-term health impacts and the formulation of effective water management strategies, especially during periods of increased rainfall. This study contributes valuable insights into the influence of seasonal variations on radionuclide levels in Oman's groundwater, contributing to a wider understanding of factors affecting groundwater quality and guiding sustainable water management practices in the region.

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CONFLICTS OF INTEREST

The authors have no conflicts of interest to declare.

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