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# Radon and heavy metal risk assessments of drinking water sources

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**ABSTRACT**

In this study, the concentrations of radon and heavy metals were measured in drinking water samples from the Pusat-Özen Dam (Hafik Dam), Sivas, Turkey. The measurements were conducted using an active radon gas analyser and energy dispersive X-ray fluorescence. The annual mean radon concentration and the annual effective dose equivalent were found to be 0.103 Bq $L^{-1}$ and 0.266 μSv $year^{-1}$, respectively. The results were compared with the international recommended values. All measured radon concentrations were below the safe limit as recommended by the World Health Organization and the United States Environmental Protection Agency. Via an elemental analysis studies, 20 different elements in the drinking water were evaluated for health risks. The annual mean values of Al, Ar, Cr, Fe, Ni, Mn and Se are higher than the recommended permissible limits. The results of this study provide a data baseline for future studies and subsequent evaluations of possible environmental contamination in Sivas.

*Keywords:* Drinking water; Heavy metals; Radon; Risk assessment; Water quality

## 1. Introduction

Radon is a naturally occurring radioactive gas resulting from the radioactive decay of $^{226}Ra$. High levels of $^{222}Rn$ play a vital role in delivering doses of radon to the public via terrestrial exposure, primarily via inhalation of its short-lived decay products and secondarily via ingestion. Radon is a soluble gas in water; therefore, the radon content in water sources depends on the radium concentration in the rocks and soils underground, where uranium-rich rocks are situated. According to the United States Environmental Protection Agency (US EPA) [1], the primary health risks from radon in drinking water are lung cancer from inhaling radon discharged from water used in the home (because radon and its progenies are $\alpha$ and $\beta$ emitters, exposure of the population to its progenies for relatively longer periods can lead to functional respiratory changes) and stomach cancer from ingesting radon in drinking water [1]. Accordingly, evaluation studies of radiation doses from radon ingestion and inhalation from drinking water are routinely conducted. Multiple studies have been conducted worldwide to determine the concentration of radon in water to minimise its adverse effects on humans [2]. These studies help improve our understanding of the environmental processes that effect radon exposure.

In addition to radon, increased levels of micropollutants in the environment are of great concern in freshwater systems. Chemical contamination in public water reservoirs has become increasingly prevalent and may have long-term effects on the ecosystem and human health [3].

Heavy metals also are introduced into the water via rock erosion, dust carried by wind, volcanic activity and the burning of forests and vegetation. Significant amounts of chemical pollutants are introduced into aquatic environments via the atmosphere because elements that are in the atmosphere are transferred into the water over time via wind and rain; these pollutants then have an impact on the aquatic systems [4–6]. Metals are present in water in free ions, organic and inorganic compounds and adsorbed onto particulate matter [7].

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It is said that the collapse of adsorbed heavy metals has toxic effects via various physical and chemical events. Most heavy metals are lethal at the 1-ppm limit [8]. Bruzzoniti et al. [9] assessed the problems with water analyses and reviewed all the pollution problems that may be present in water. Their study indicated that the sources of the factors that cause water pollution, their effects and their chemical structures are widely variable. The main pollutants that cause water pollution are organic and inorganic substances, salts, microorganisms, detergents, pesticides, heavy metals, suspended solids, radioactivity, fats, petroleum products and waste heat [9–11]. Surface water used for different purposes, especially drinking and irrigation, suffer according to the quality and quantity of heavy metals introduced from the atmosphere and industrial and urban waste [12]. Water pollution occurs due to domestic, industrial and agricultural activities and mines. Numerous different pollutants reach aquatic ecosystems from these sources. These contaminants include heavy metals and pesticides.

Heavy metals are defined as elements with densities greater than 5 g $cm^{-3}$ that are not vitiated biologically [13]. In addition, heavy metals accumulate in tissues and can cause immediate and long-term damage to living organisms [14].

The major heavy metals that cause serious health problems and environmental pollution are arsenic (As), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni) and zinc (Zn). Health problems caused by water contamination due to these elements have been reported by many studies worldwide [2,6,15,16].

The aim of this study is to describe the findings of the $^{222}$Rn activity concentrations and heavy metal contents of water samples from the Pusat-Özen Dam for drinking and irrigation purposes and to determine the annual effective dose due to waterborne radon in Hafik/Sivas, Turkey. The dam region is also part of a second-degree earthquake zone [17]. The results obtained from this study will contribute to a database of environmental radioactivity measurements and heavy metal pollution. In addition, these results will be useful for assessing the safety of drinking water.

## 2. Materials and methods

### 2.1. Study area

Sivas is a city on the Central Anatolia Region of Turkey and is in an earthquake zone (Fig. 1). Sivas has a total area of 28,619 $km^2$ and is bordered by the provinces of Giresun, Ordu, Tokat, Erzincan, Malatya, Kahramanmaraş, Kayseri and Yozgat. The population of the city is approximately 624,000, and the latitude and longitude of Sivas are 39°45′N and 37°1′E at an altitude of 1,285 m above sea level. The province is established on a high plateau with mountains and is the coldest province in the Central Anatolia Region [18]. In Sivas, the Pusat-Özen Dam is a major supplier of household water and is filled by water from surrounding streams and rainwater.

### 2.2. Radon activity in water

In this study, over a 1-year period in 2015, samples were collected monthly at five different points on the Pusat-Özen Dam. A total of 60 water measurements were made to determine the radon concentrations of the water samples in the research area. Water samples were collected from the dam in 0.5-L linear polypropylene bottles, which were slowly and completely filled and then immediately closed tightly underwater to avoid air bubbles. All the water sample measurements were made in the Nuclear Physics Laboratory at Kastamonu University. Laboratory measurements were performed within

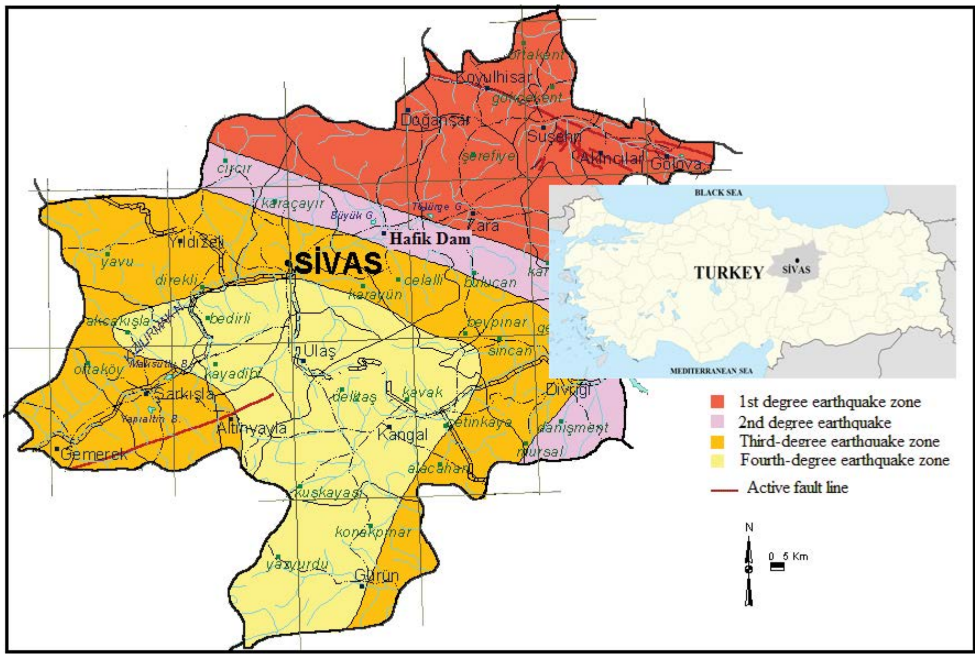


Fig. 1. Locations of the sampling sites and an earthquake map of Sivas [17].

3 d after drawing the samples. The results were recorded in units of Bq m$^{-3}$ and converted to units of Bq L$^{-1}$.

The radon concentrations of the water samples were measured using a professional portable radon monitor AlphaGUARD PQ2000 PRO, which is an ionisation chamber (0.62 L) designed to measure radon in the air, soil and water.

AlphaGUARD has an ionisation chamber that is also part of the gas cycle. Radon was expelled from the water samples (placed in an emanation vessel) using a pump in a closed gas cycle. Then, the water was injected into the emanation vessel, and the AlphaGUARD and AlphaPUMP were switched on. The flow rate of the pump was 0.3 L min$^{-1}$. After 10 min, the pump was switched off and the AlphaGUARD remained switched on for another 20 min, continuing the radon measurement. The AlphaGUARD monitor worked in 'flow' mode for 1 min. Prior to each water sample measurement, the background of the empty set-up was measured for a few minutes. Calibration of the measuring system was carried out by Saphymo (Genitron Instruments, Germany), with a guaranteed stability and accuracy for 5 years [19].

The following equation was used to determine the radon concentrations in the water samples [20]:

$$A_{\text{water}} = \frac{A_{\text{air}}\left(\frac{\left(V_{\text{system}} - V_{\text{sample}}\right)}{V_{\text{sample}}} + k\right) - A_0}{1,000} \tag{1}$$

where $A_{\text{water}}$ is the radon concentration in the water sample (Bq m$^{-3}$), $A_{\text{air}}$ is the radon concentration in the set-up after expelling the radon from the water (Bq m$^{-3}$), $A_0$ is the background radon level (Bq m$^{-3}$), $V_{\text{system}}$ is the interior volume of the measurement set-up (mL), $V_{\text{sample}}$ is the volume of the water sample (mL) and $k$ is the radon distribution coefficient:

$$k = 1.105 + 0.405e^{-0.502\times T(^\circ C)} \tag{2}$$

### *2.3. Heavy metal concentrations in water*

The collection period for the water samples was 1 year (2015), and the measurements were taken in four 3-month surveys. A total of 20 water samples were collected to determine the heavy metal content in the Pusat-Özen Dam. The collected water samples were stored in presterilised bottles for the heavy metal analysis. The seasonal variations in the heavy metal contents of the water samples were ascertained.

The elemental analysis survey was conducted using energy dispersive X-ray fluorescence [21] via a very versatile EDXRF spectrometer, which optimises the excitation using polarisation and secondary targets. The spectrometer has an auto sampler for up to 12 items and software modules and uses a 50-W end-window X-ray tube to excite the samples. The target changer, with up to eight polarisation and secondary targets, offers many different excitation conditions ensuring the optimum determination of all elements from $^{11}$Na to $^{92}$U. The measurements are conducted in a helium gas atmosphere. A spectral resolution of less than 155 eV at Mn $K_\gamma$ was achieved. The sample chamber was equipped with a sample spinner for 40-mm sample cups [21].

## 3. Results and discussion

### *3.1. Radon measurement values and evaluations in terms of health*

The mean radon activity concentration values varied from 0.075 ± 0.002 to 0.133 ± 0.001 Bq L$^{-1}$ during the 1-year period, and the annual mean value was found to be 0.103 ± 0.001 Bq L$^{-1}$. The monthly variations in the radon concentration in the water samples are given in Table 1. Fig. 2 shows a graphic representation of the monthly variation in the mean radon concentrations in the study area during the 1-year period.

The US EPA [22] has recommended a maximum contamination level of radon in drinking water of 11 Bq L$^{-1}$ (300 pCi L$^{-1}$) [22], while the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) [23] has given a range of 4–40 Bq L$^{-1}$. All the recorded values from the Pusat-Özen Dam water samples were lower than the permissible limits suggested by both the US EPA [22] and UNSCEAR [23]; therefore, the water collected from the study area is safe for drinking purposes in terms of the concentration of radon activity.

Radon in water may cause exposure via the ingestion of drinking water and the inhalation of radon released into the air when the water is used. The annual mean effective doses

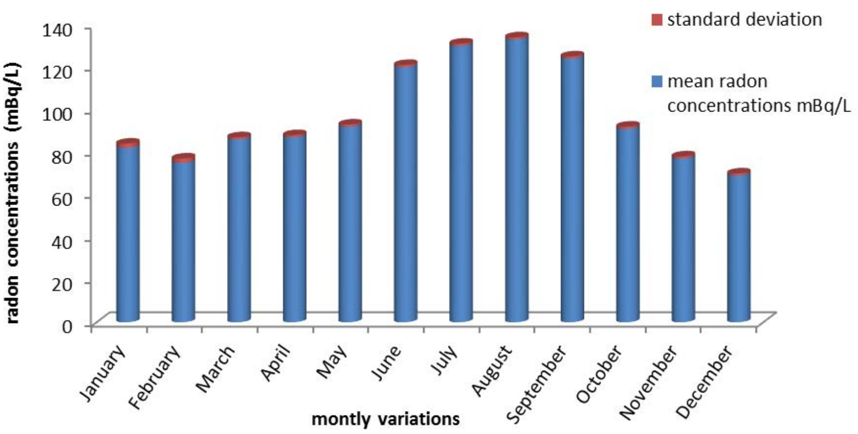


Fig. 2. The monthly variations in the mean radon concentrations.

Table 1
The monthly variations in the radon concentrations and the annual effective doses

| Months | Sample number | Mean $^{222}$Rn activity concentrations ± SD (Bq $L^{-1}$) | Annual effective dose equivalent (μSv $year^{-1}$) | | | Annual effective dose equivalent (μSv $year^{-1}$) (ingestion) | | |
|---|---|---|---|---|---|---|---|---|
| | | | Inhalation | Ingestion | Total | Infants | Children | Adults |
| January | 5 | 0.082 ± 0.002 | 0.207 | 0.017 | 0.224 | 0.029 | 0.022 | 0.014 |
| February | 5 | 0.075 ± 0.002 | 0.189 | 0.016 | 0.205 | 0.026 | 0.020 | 0.013 |
| March | 5 | 0.086 ± 0.001 | 0.218 | 0.018 | 0.236 | 0.030 | 0.023 | 0.015 |
| April | 5 | 0.087 ± 0.001 | 0.220 | 0.018 | 0.238 | 0.031 | 0.023 | 0.015 |
| May | 5 | 0.092 ± 0.001 | 0.232 | 0.019 | 0.252 | 0.032 | 0.024 | 0.016 |
| June | 5 | 0.120 ± 0.001 | 0.303 | 0.025 | 0.329 | 0.042 | 0.032 | 0.021 |
| July | 5 | 0.130 ± 0.001 | 0.328 | 0.027 | 0.355 | 0.046 | 0.034 | 0.023 |
| August | 5 | 0.133 ± 0.001 | 0.334 | 0.028 | 0.362 | 0.046 | 0.035 | 0.023 |
| September | 5 | 0.124 ± 0.001 | 0.312 | 0.026 | 0.338 | 0.043 | 0.032 | 0.022 |
| October | 5 | 0.091 ± 0.001 | 0.229 | 0.019 | 0.248 | 0.032 | 0.024 | 0.016 |
| November | 5 | 0.077 ± 0.001 | 0.195 | 0.016 | 0.211 | 0.027 | 0.020 | 0.014 |
| December | 5 | 0.069 ± 0.001 | 0.174 | 0.015 | 0.189 | 0.024 | 0.018 | 0.012 |
| The annual mean | | 0.103 ± 0.001 | 0.245 | 0.020 | 0.266 | 0.034 | 0.026 | 0.017 |

for ingestion and inhalation were calculated according to the parameters introduced by the UNSCEAR [23] report and are calculated as follows.

For inhalation,

$$\text{AEDE } (\mu\text{Sv year}^{-1}) = A\ (\text{Bq L}^{-1}) \times 10^{-4} \times 7{,}000\ \text{h year}^{-1} \times 0.4 \times 9\ \text{nSv}\ (\text{Bq m}^{-3}\ \text{h})^{-1} \times 1{,}000\ \text{L m}^{-3} \quad (3)$$

where $A$ is the $^{222}$Rn activity concentration in the water (Bq $L^{-1}$), $10^{-4}$ is the air–water concentration ratio, the indoor occupancy is 7,000 h $year^{-1}$, 0.4 is the equilibrium factor between radon and its decay products and 9 nSv (Bq $m^{-3}$ h)$^{-1}$ is the dose conversion factor for radon exposure [22].

For ingestion,

$$\text{AEDE } (\mu\text{Sv year}^{-1}) = A\ (\text{Bq L}^{-1}) \times 60\ \text{L year}^{-1} \times 3.5\ \text{nSv Bq}^{-1} \quad (4)$$

where 60 L $year^{-1}$ is the weighted estimate of the water consumption and 3.5 nSv $Bq^{-1}$ is the effective dose coefficient for ingestion [23].

According to the UNSCEAR [23] report, the ingestion of water estimated in the US EPA [22] report was 100, 75 and 50 L $year^{-1}$ for infants, children and adults, respectively. Assuming the proportion of these groups in the population to be 0.05, 0.3 and 0.65, respectively, the weighted estimate of consumption was determined to be 60 L $year^{-1}$ [23].

The mean annual effective dose equivalent AEDE (μSv $year^{-1}$) for inhalation, ingestion and the total of the two were calculated monthly using the information given earlier. The annual effective dose equivalent values for inhalation, ingestion and the total varied from 0.174 to 0.334 μSv $year^{-1}$, from 0.015 to 0.028 μSv $year^{-1}$ and from 0.189 to 0.362 μSv $year^{-1}$, respectively, and the mean values were found to be 0.245, 0.020 and 0.266 μSv $year^{-1}$, respectively (Table 1).

As can be seen from Table 1, the maximum estimated total annual effective dose was 0.362 μSv $year^{-1}$ in August and the minimum was 0.189 μSv $year^{-1}$ in December. For water samples, the safe limit for the annual effective dose per person as recommended by the World Health Organization (WHO) is 100 μSv $year^{-1}$ [24]. The results of this study indicate that all the measured values were well below this limit for radon in drinking water. Therefore, the water can be used as drinking water. For the radon exposure from ingestion, the annual effective dose equivalents were ascertained separately for adults, children and infants (Table 1).

The drinking water samples analysed for heavy metal concentrations (Table 2) showed that the heavy metals are well within the safe limits as given by the WHO [25] and the European Commission (EC) [26]. The recommended levels according to these agencies are given in Table 3.

### *3.2. Heavy metal risk assessments*

The mean value of Al was measured at 7.52 mg $L^{-1}$ for the 1-year period. The lowest mean value was 4.81 mg $L^{-1}$ in the autumn, while the highest mean value was 12.6 mg $L^{-1}$ in the summer. The annual mean value of Al is higher than the permissible limits recommended by the WHO [25] and EC [26]. The As concentration varies from 0.01 to 0.02 mg $L^{-1}$ in the study area. The mean values of As in the summer and autumn seasons are the same as the permissible limits; however, the mean values are slightly higher in the spring and summer seasons. The mean values of Cd, boron (B) and Pb are within the permissible limits recommended by the WHO [25] and EC [26], and the mean values of U and Zn are lower than the permissible limits in all seasons. The Cd concentration (0.002 mg $L^{-1}$) in the lake can be attributed to the artificial phosphate fertilisers used for agricultural activities around the dam. The annual variation in the Cu values was found to be 0.5–2.3 mg $L^{-1}$. The reason for this increase is thought to be the drainage into the dam water via rain of Cu, which accumulates in the soil due to the dense usage of copper sulphate during maintenance and pruning processes in fruit gardens [27]. The mean values of Cu are lower than the permissible limits except during the winter season.

Table 2
Heavy metal concentrations (mg L$^{-1}$) in the water samples

| Elements | Spring | Summer | Autumn | Winter |
|---|---|---|---|---|
| Aluminium (Al) | 7.13 | 12.6 | 4.81 | 5.52 |
| Arsenic (As) | 0.02 | 0.01 | 0.01 | 0.02 |
| Boron (B) | 0.4 | 0.4 | 0.4 | 0.4 |
| Cadmium (Cd) | 0.002 | 0.002 | 0.002 | 0.002 |
| Calcium (Ca) | 89.1 | 92.9 | 85.8 | 46.8 |
| Chloride (Cl) | 2.0 | 2.0 | 2.0 | 2.0 |
| Chromium (Cr) | 0.05 | 0.08 | 0.07 | 0.06 |
| Copper (Cu) | 0.5 | 0.7 | 1.1 | 2.3 |
| Iron (Fe) | 1.21 | 1.29 | 1.25 | 1.24 |
| Lead (Pb) | 0.01 | 0.01 | 0.01 | 0.01 |
| Magnesium (Mg) | 264 | 425 | 601 | 854 |
| Manganese (Mn) | 0.9 | 0.7 | 1.1 | 1.0 |
| Nickel (Ni) | 0.04 | 0.04 | 0.04 | 0.04 |
| Phosphorus (P) | 6.4 | 9.7 | 5.5 | 3.9 |
| Potassium (K) | 10.1 | 8.4 | 6.3 | 7.5 |
| Selenium (Se) | 0.5 | 0.5 | 0.5 | 0.5 |
| Sodium (Na) | 162 | 159 | 139 | 916 |
| Sulphur (S) | 32.1 | 42.7 | 20.1 | 18.6 |
| Uranium (U) | 1.0 | 1.0 | 1.0 | 1.0 |
| Zinc (Zn) | 0.4 | 1.3 | 2.3 | 0.5 |

Table 3
The recommended levels according to various agencies and the results for the study area

| Elements | Standards (mg L$^{-1}$) | | |
|---|---|---|---|
| | WHO (2003) | EC (2005) | [a]Pusat-Özen Dam |
| Aluminium (Al) | 0.2 | 0.2 | 7.52 |
| Arsenic (As) | 0.01 | 0.01 | 0.02 |
| Boron (B) | 0.3 | 1.0 | 0.4 |
| Cadmium (Cd) | 0.003 | 0.005 | 0.002 |
| Calcium (Ca) | – | – | 78.7 |
| Chromium (Cr) | 0.05 | 0.05 | 0.07 |
| Copper (Cu) | 2.0 | 2.0 | 1.2 |
| Iron (Fe) | – | 0.2 | 1.25 |
| Lead (Pb) | 0.01 | 0.01 | 0.01 |
| Manganese (Mn) | 0.5 | 0.05 | 0.9 |
| Nickel (Ni) | 0.02 | 0.02 | 0.04 |
| Selenium (Se) | 0.01 | 0.01 | 0.5 |
| Sodium (Na) | 200 | 200 | 344 |
| Uranium (U) | 1.4 | – | 1.0 |
| Zinc (Zn) | 3.0 | – | 1.1 |

[a]Study area.

The annual mean values of Cr, Fe, Mn, Ni and Se are higher than the permissible limits recommended by the WHO [25] and EC [26].

The concentrations of Fe increased during the summer season due to widespread wheat planting around the lake. Due to the use of ferrous-containing agricultural pesticides used to increase the grain productivity of wheat plants, ferrous-containing water and particles may enter the dam water via rain [27].

It is known that heavy metals constitute an important pollutant group and that they tend to accumulate within the bodies of living organisms and have significant toxic and carcinogenic effects. Heavy metals having strong poisonous effects even at very low concentrations may inhibit the self-cleaning processes of natural water and may negatively affect the usability of water sources for human consumption and domestic use by urban and rural populations [27].

The radon concentrations in the drinking water samples from the Pusat-Özen Dam (Sivas, Turkey) are below the safe limits recommended by Engin et al. [5] and UNSCEAR [23]. The mean annual effective dose estimated from 1 year of measurements was also found to be well below the safe limit of 100 μSv year$^{-1}$ provided by the EC [26]. In general, the concentrations of the heavy metals were well within the recommended limits. The outcome of this study will provide a good source for environmental management and further monitoring.

## 4. Conclusions

This study focused on the effects of the concentrations of radon and heavy metals measured in drinking water samples taken from the Pusat-Özen Dam in Sivas, Turkey. The following conclusions can be drawn.

- The radon concentrations of the water samples were measured using a professional portable radon monitor. All measured radon concentrations were lower than the permissible limits in drinking water. Therefore, the water can be used as drinking water.
- An elemental analysis was conducted using an EDXRF spectrometer. The heavy metal contamination in the drinking water was evaluated for health risks. The annual mean values of Cr, Fe, Mn, Ni, Se and Na are higher than the permissible limits. The concentrations of the other heavy metals are well within the recommended limits.
- The results of this study provide a data baseline for future environmental radioactivity measurements and monitoring of heavy metal contamination. In addition, these results will be useful for assessing of safety of the drinking water.

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