

Characterization of radon in drinking water of Villy and the associated radiological hazards

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ABSTRACT

In the present study, the activity concentrations of radon were measured in twenty-seven (27) water samples collected from Villy, a village of west central region which presents uranium anomalies. The associated radiological hazards were then assessed. The radon activity concentrations were in the range of 5.77 ± 0.74 to 441.76 ± 4.45 Bq.L⁻¹ with an average of 157.31 ± 2.30 Bq.L⁻¹. The lower detection limit of the instrument SARAD EQF 3200 is 0.106 Bq.L⁻¹. The average total annual effective dose due to radon was 1.06 mSv.y⁻¹ with the range of 39 μ Sv.y⁻¹ to 2.99 mSv.y⁻¹. 70.37 % of the measured radon concentrations are found to be higher than 100 Bq.L⁻¹, the reference levels of World Health Organization and European Union but were lower than the European Union action level of 1000 Bq.L⁻¹. Accordingly, it does not pose any significant health hazards to the populace of Villy.

RESUME

Dans la présente étude, les concentrations d'activité du radon ont été mesurées dans vingt-sept (27) échantillons d'eau prélevés à Villy, un village de la région du centre-ouest qui présente des anomalies d'uranium. Les dangers radiologiques associés ont ensuite été évalués. Les concentrations d'activité du radon se situaient entre $5,77 \pm 0,74$ et $441,76 \pm 4,45$ Bq.L⁻¹, avec une moyenne de $157,31 \pm 2,30$ Bq.L⁻¹. La limite inférieure de détection de l'instrument SARAD EQF 3200 est de 0,106 Bq.L⁻¹. La dose efficace annuelle totale moyenne due au radon était de 1,06 mSv.an⁻¹, avec une plage de 39 μ Sv.an⁻¹ à 2,99 mSv.an⁻¹. 70,37 % des concentrations de radon mesurées sont supérieures à 100 Bq.L⁻¹, niveaux de référence de l'Organisation Mondiale de la Santé et de l'Union Européenne, mais inférieures au seuil d'intervention de l'Union Européenne de 1000 Bq.L⁻¹. En conséquence, il ne pose pas de risques significatifs pour la santé de la population de Villy.

I. INTRODUCTION

The radionuclides in the environment can be divided into three groups: the primordial radionuclides, the cosmogenic radionuclides and the anthropogenic radionuclides. Among the primordial radionuclides, there are four long series of genetically related radioactive nuclides beginning with thorium (Th), uranium (U) or Neptunium (Np) and ending with lead

(Pb) and bismuth (Bi). In the uranium decay series (starting with U-238), the thorium decay series (starting with Th-232) and the actinium decay series (starting with U-235), there is a gaseous element called radon with the respective isotopes Rn-222 (radon), Rn-220 (thoron) and Rn-219 (actinon). Radon is a naturally occurring radioactive gas which may be found in high concentrations in water. Thanks to its fairly solubility in water it comes to the water

from the decay of radium in soil or rocks adjacent to these reservoirs (Bem et al., 2014). The largest proportion of human exposure to radiation comes from natural sources and the largest fraction of natural radiation exposure comes from radon (Kumar et al., 2016).

Radon is one of the leading causes of lung cancer (3 to 14%) (WHO, 2023). It is odorless, tasteless, and colorless, and therefore cannot be detected by the human senses (Kumar et al., 2016). Actinon has a half-life of 3.4 s, thoron a half-life of 54 s and radon a half-life of 3.825 days. Due to the longer half-life of radon (Rn-222), it is detected everywhere in the environment (in indoor air, outdoor air, soil gas, and water samples). Radon in water presents two pathways of exposure for individuals: ingestion of drinking water and inhalation of air containing radon released from underground water through some pipes. To protect the health of the public from radon in drinking water, different reference levels have been established. For waters intended for human consumption the Euratom Drinking Water Directive established parametric values in 2013, World Health Organization (WHO) (WHO, 2008) used guidance level and the United States introduced maximum contaminant levels (Jobbágy et al., 2017). The country concerned establishes guidance levels and parametric values on the basis whether that value poses a risk to human health from a radiation protection point of view or not (i.e. if further remediation action is needed or not) (Jobbágy et al., 2017).

Some studies on radon (Rn-222) in drinking water have been performed in many parts of the world for the radiological impact assessment of radon on the populace (Bem et al., 2014; Kumar et al., 2016). Indeed, In Iran, Meghdad Pirsaeheb et al, Yadolah Fakhri et al (Fakhri et al., 2015; Pirsaeheb et al., 2015), in Nigeria Yinka Ajiboye et al and Janet A. Ademola and Oluwaferanmi R. Ojeniran (Ademola & Ojeniran, 2017; Ajiboye et al., 2022), in China, Yunyun Wu et al (Wu et al., 2018), in Turkey, Halime Kayakökü (Kayakökü, 2021), in India Suresh S et al and Sudhir Mittal et al (Mittal et al., 2016; Suresh et al., 2020), published papers on radon in drinking water. None of these studies was performed in Burkina Faso. The choice of Villy as study area was because it is an area of uranium anomalies (Beogo et al., 2019) and then the subsoil could be rich in uranium. Knowing that radon (Rn-222) resulted from the decay of uranium therefore the underground water could be rich in radon thanks to the dissolution property. The objective of this work was to assess the level of radon in underground water of Villy and find out if it does not pose any significant radiological hazards to the population.

II. MATERIAL AND METHODS

2.1. Study area

The present study was carried out at Villy, in the Boulkiemde province, west central region, Burkina Faso. It lies on the latitude 12°16'46" N and the longitude 2°09'53" W.

The Boulkiemde province belongs mainly to the Precambrian D formations and to a lesser extent the Precambrian C (BANGOU et al., 2021; SAWES, 2006). Magmatites, granites and a few greenstone intrusions make up the bulk of the geological formations of the area (BANGOU et al., 2021; SAWES, 2006).

Underground water constitutes the main source of drinking water resources in the village of Villy. Indeed, according to the local development plan 2018-2022 of Koudougou, there is about fifty-five (55) boreholes in Villy. There is a dam in the village which is used for irrigation and livestock watering. People only drink water from boreholes and wells. A total of twenty-seven water samples were collected and analyzed with the monitor SARAD EQF 3200 GmbH.

The locations of the sampling sites were identified using the GPS Garmin 64 s. Thanks to the software ArcGIS 10.3, a map of the sampling sites in Villy was done and presented in Figure 1.

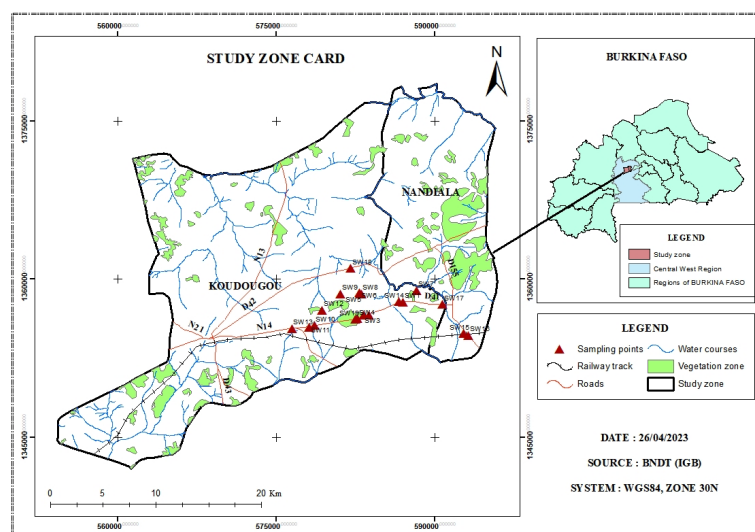


Figure 1: Location of the water sampling sites.

2.2. Sample collection

Water samples were collected from boreholes and wells. No water was collected from the dam because it is not consumed by the population. 0.5 L and 1.5 L of clean polyethylene bottles were used for that purpose. Before sampling, they were rinsed three times with the same water prior to filling. The water was filled up to the brim in a manner that no bubble remains in the bottles. It was then tightly closed with the cap and some cellotape. The collected water samples were then brought to the laboratory within 24 h and explored for dissolved Rn-222 using the Emanometry method (Mayya et al., 1998).

At the laboratory, the samples were put in the bubbling flask one after another for the analysis using the radon monitor SARAD EQF 3200.

2.3. Methodology of radon measurement in water

The bubbling flask containing the water to measure is connected to the radon monitor SARAD EQF 3200 through air connection tubes as shown in the figure 2 below.



Figure 2: Experimental set-up for the measurement of radon in water.

The radon monitor SARAD EQF 3200 contains an internal pump which, when activated, engender a certain pressure that leads to the circulation of air in the closed loop. This enables the radon gas dissolved in the water to be degassed. The radon gas is then transferred in the sealed radon chamber for the measurement process. The concentration equilibrium between water and air is reached after about 30 Minutes (SARAD GmbH, 2007a). After this equilibrium, the measurement is stopped and restarted for the determination of radon concentration in air. The radon activity concentration in water $C_{Rn}(Water)$, within the system after the de-gassing, is related to the radon activity concentration in air $C_{Rn}(Air)$ by the following equation:

$$C_{Rn}(Water) = \frac{C_{Rn}(Air) * [K_{Oswald} * V(Water) + V(Air)]}{V(Air)} \quad (1)$$

Where $V(Water)$ is the Volume of the water, $V(Air)$ is the volume of air within the sealed system, K_{Oswald} is the Oswald-coefficient.

The Oswald coefficient is the ratio of activity concentration of radon in water by the activity concentration of radon in air. It is given by the equation 2 :

$$K_{Oswald} = 0.425 * e^{(-0.5T)} + 0.1 \quad (2)$$

The radon activity concentration in water can easily be determined using the software « Radon In Water Calculator ».

The radon monitor has a protection flask, inserted between the air inlet of the instrument and the outlet of the bubbling flask (SARAD GmbH, 2007a), which prevent water from entering in it.

The measurement of radon activity concentration in underground water samples at a later time in the laboratory does not truly represent the original concentration. It is therefore important to correct measured concentration to

concentration at the time of sampling (Ajiboye et al., 2022) using the decay law.

2.4. Measurement process

The schematic diagram of the set-up realized for the measurement of radon using SARAD EQF 3200 is shown in Figure 3. As previously mentioned, the air pump enables the air flow in the closed loop resulting in the creation of bubbles. The bubbles contain the radon gas.

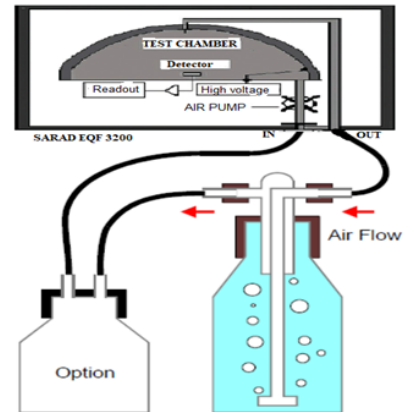


Figure 3 : Schematic diagram of the experimental set-up for radon measurement in water.

The Radon gas ($Rn-222$) will then enter the measurement chamber called test chamber where it will decay into its short living daughter products. The radon activity concentration is measured thanks to these daughter products, generated inside the test chamber (SARAD GmbH, 2007a). The decay of $Rn-222$ gives $Po-218$ which interacts with the emitted alpha particles in the medium. The interaction causes the ionization of $Po-218$. After the ionization, the positively charged $Po-218$ are collected by electrical field forces on the surface of a semi-conductor detector presented in Figure 3 (SARAD GmbH, 2007b). The number of collected $Po-218$ ions is proportional to the radon gas concentration inside the chamber (SARAD GmbH, 2007b).

$Po-218$ itself decays with a half-life time of only 3.05 minutes and about 50 % of the emitted alpha particles move towards the detector surface and interact with the detector. The interaction produces an electrical signal proportional in strength to the energy of the alpha particle. The signal is amplified to the level at which it can be counted. The number of decays registered by the detector is proportionnal to the amount of $Po-218$ collected.

The radon activity concentration in the sealed radon chamber of the SARAD EQF 3200 is given by the following equation:

$$\frac{dC_{Rn}(t)}{dt} = -\lambda_{Rn} C_{Rn}(t) \quad (3)$$

The polonium activity concentration in the same chamber is as follows :

$$\frac{dC_{Po}(t)}{dt} = \lambda_{Po} C_{Rn}(t) - \lambda_{Po} C_{Po}(t) \quad (4)$$

Considering the secular equilibrium between radon (Rn-222) and polonium (Po-218), ($T_{Rn} \gg T_{Po}$ and $\lambda_{Rn} \ll \lambda_{Po}$), which is reached approximately at 5 half-lives time (SARAD GmbH, 2007b), about 15 minutes, the radon activity concentration is then estimated from the number of polonium decays registered during this time. The time of 15 minutes defines the minimum achievable response time to a radon activity concentration measurement step (SARAD GmbH, 2007b) and is observed in Figure 4.

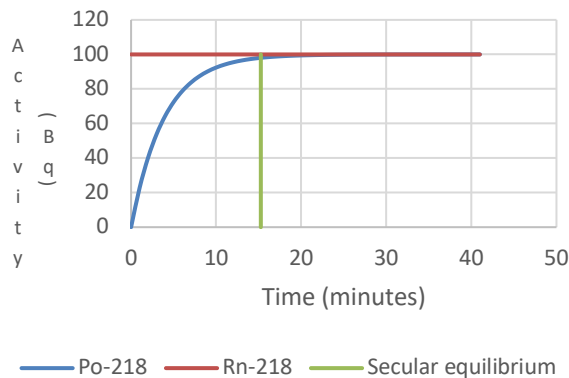


Figure 4: Secular equilibrium between radon (Rn-222) and Polonium (Po-218).

The graph of Figure 4 represent the resolution of the differential equations (3) and (4) using the Euler-Cauchy method with the initial conditions : $C_{Rn}(0) = 100$, $C_{Po}(0) = 0$ for the range $t = 0$ (1) 41.

2.5. Detection limits

The lower detection limit depends on many parameters: the volume of the sample, the sensitivity of the monitor, the duration of the sampling.

For the determination of the lower detection limit, some distilled water is assessed for radon by following the same measurement process as usual. The calculated lower detection limit is 0.106 Bq.L^{-1} .

2.6. Uncertainty calculation

The relative statistical error E for a chosen confidence interval of k -Sigma can be predicted from the number of detected counts N by the equation (SARAD GmbH, 2007b) :

$$E[\%] = 100 \times k \times \frac{\sqrt{N}}{N} \quad (5)$$

The number of counts N within an integration interval T is determined using the following equation:

$$N = C_{Rn} \times T \times S \quad (6)$$

Where C_{Rn} is the measured Radon concentration and S is the sensitivity of the SARAD equipment.

2.7. Effective dose calculation

The total annual effective dose E_{Rn} for general population caused by occurrence of radon in drinking water and its domestic use is the sum of the effective doses due to radon

ingestion with water E_{ing} and inhalation from waterborne radon E_{inh} (Bem et al., 2014).

The effective dose due to radon ingestion with water E_{ing} is obtained using the equation (7) (Ajiboye et al., 2022) :

$$E_{ing} = DCF \times A \times V \quad (7)$$

Where DCF (dose conversion factor) is the dose coefficient in Sv.Bq^{-1} , A is the average activity concentration of radon in drinking water in Bq.L^{-1} and V is the weighted estimate of consumption in L .

Since radon is readily lost from water by heating or boiling, the total annual water intake for the so called ICRP Standard Man equals to 2 L per day or 730 L per year is not used for the dose calculation. A proposed realistic value of 60 L for the weighted direct annual consumption of tap water has been given by UNSCEAR 2000 Report and this value of volume has been used in this work (Bem et al., 2014; United Nations, 2000).

The dose from inhalation of water-borne radon is calculated using the following equation (Ajiboye et al., 2022) :

$$E_{inh} = DCF \times A \times T \times F \times t \quad (8)$$

Where DCF is the dose conversion factor or dose coefficient in Sv.Bq^{-1} , A is the average activity concentration of radon in drinking water in Bq.L^{-1} , T is the radon transfer from water to air coefficient in $\text{dm}^3.\text{m}^{-3}$, F is the indoor radon-daughters equilibrium factor and t is the average annual indoor occupancy in h (Bem et al., 2014).

2.8. Simulation of the contribution of underground water in indoor radon of a three-bedroom house constructed in Villy.

The contribution of household's water in indoor radon is sometimes not negligible. Indeed, water can be a source of elevated radon levels and even more, a source of very high concentrations of indoor radon (IAEA, 2021). This may happen when the water is supplied from a private borehole in an area enriched with radium in the bedrock (IAEA, 2021). The degassing routes of radon from water include splashing of water, heating of water, water splitting, the use of pressurized pipes to conduct the water.

In this section, the prediction of the contribution of each borehole to the increase of radon levels in a three-bedroom house (Figure 5) dependent solely on the borehole is given.

For the estimation of the contribution of household water supplied by each borehole or well of the study area to indoor air of a three-bedroom house, the assumptions made are as follow :

- the daily water consumption of one personne is 50 L to live decently (UN, OHCHR, UN-Habitat, WHO, 2010) ;
- ten persons are living in the three-bedroom house ;

- the height of the house is 2.6 m ;
- the parameters e_i and W_i proposed for this study are given in the table 1 hereafter.

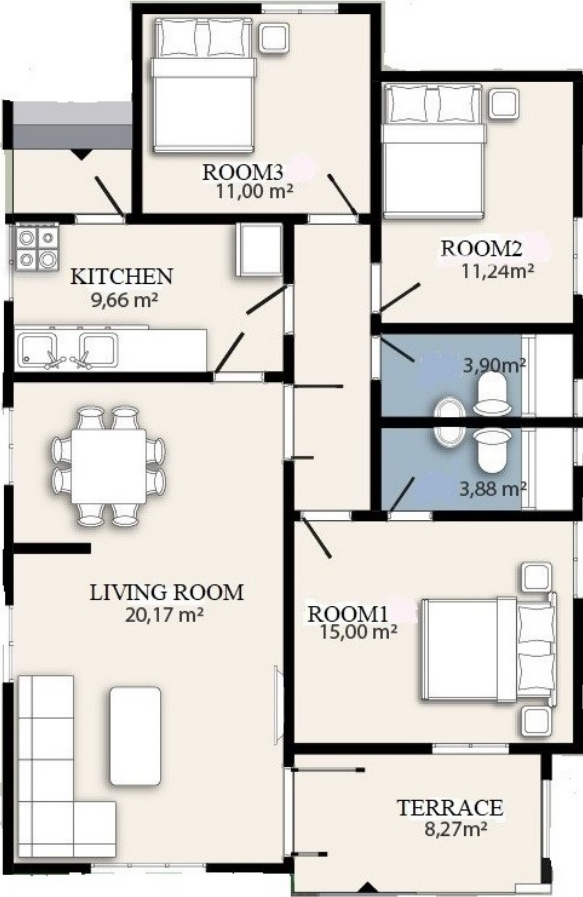


Figure 5: Three-bedroom house.

Table 1: Radon emanation to indoor air and corresponding volume for each purpose

Use	e_i (%)	W_i (m³)
Drinking water	45	0.03
Shower	65	0.25
Dishwashing	95	0.07
Bath	40	0.03
WC	30	0.05

III. RESULTS AND DISCUSSION

The radon activity concentrations in water samples collected from Villy and the corresponding annual ingestion (E_{ing}) and inhalation doses (E_{inh}) are given in Table 2. The total annual effective dose (E_{Total}) is also given in the same Table 2.

Laundry	92.5	0.07
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The values of e_i were chosen among the range of values of e_i given by IAEA in the IAEA-TECDOC-1951 (IAEA, 2021).

The radon exhalation from the water into a building is estimated using the following equation (IAEA, 2021) :

$$C_v = \frac{C_w}{24.(\lambda+n).V} \sum_i e_i \cdot W_i \quad (9)$$

Where

C_v is the radon contribution from household water usage to indoor air in Bq.m⁻³;

C_w is the radon concentration in water in Bq.m⁻³ ;

λ is the radon decay constant in h⁻¹ ;

n is the air exchange in the building in h⁻¹ ;

V is the building volume in m³ ;

W_i is the volume of water used daily for purposes in m³.day⁻¹ ;

e_i is the share of radon that merges into the indoor air in percentage.

Table 2: Radon activity concentrations in water and associated annual ingestion doses and annual inhalation doses.

Sample Code	Radon Concentration in Water (Bq.L ⁻¹)	Average annual ingestion dose E _{ing} (Sv)	Average annual inhalation dose E _{inh} (Sv)	Total annual effective dose E _{Total} (Sv)
WS1	112.19±2.28	6.73E-05	6.91E-04	7.58E-04
WS2	376.64±4.17	2.26E-04	2.32E-03	2.55E-03
WS3	220.02±4.18	1.32E-04	1.36E-03	1.49E-03
WS4	76.16±2.70	4.57E-05	4.69E-04	5.15E-04
WS5	197.06±4.34	1.18E-04	1.21E-03	1.33E-03
WS6	7.56±0.71	4.54E-06	4.66E-05	5.11E-05
WS7	30.51±1.56	1.83E-05	1.88E-04	2.06E-04
WS8	17.26±1.28	1.04E-05	1.06E-04	1.17E-04
WS9	441.76±5.42	2.65E-04	2.72E-03	2.99E-03
WS10	312.42±4.98	1.87E-04	1.92E-03	2.11E-03
WS11	107.41±2.92	6.44E-05	6.62E-04	7.26E-04
WS12	15.69±1.07	9.41E-06	9.66E-05	1.06E-04
WS13	280.35±5.17	1.68E-04	1.73E-03	1.90E-03
WS14	5.77±0.74	3.46E-06	3.55E-05	3.90E-05
WS15	14.21±1.01	8.53E-06	8.75E-05	9.61E-05
WS16	277.28±2.05	1.66E-04	1.71E-03	1.87E-03
WS17	219.02±1.84	1.31E-04	1.35E-03	1.48E-03
WS18	108.51±1.29	6.51E-05	6.68E-04	7.34E-04
WS19	223.67±1.86	1.34E-04	1.38E-03	1.51E-03
WS20	126.78±1.39	7.61E-05	7.81E-04	8.57E-04
WS21	107.65±1.29	6.46E-05	6.63E-04	7.28E-04
WS22	211.78±1.97	1.27E-04	1.30E-03	1.43E-03
WS23	217.93±2.00	1.31E-04	1.34E-03	1.47E-03
WS24	151.47±1.68	9.09E-05	9.33E-04	1.02E-03
WS25	188.98±1.87	1.13E-04	1.16E-03	1.28E-03
WS26	169.71±1.76	1.02E-04	1.05E-03	1.15E-03
WS27	29.68±0.67	1.78E-05	1.83E-04	2.01E-04
Average	157.31±2.30	9.44E-05	9.69E-04	1.06 E-03
Range	5.77-441.76	3.46E-06-9.44E-05	3.55E-05-2.72E-03	3.90E-05-2.99E-03

The average radon activity concentration was 157.31±2.30 Bq.L⁻¹ with the range of 5.77±0.74 to 441.76±4.45 Bq.L⁻¹.

All the calculated annual ingestion doses were found to be lower than the annual inhalation doses. In fact, this confirm the statement of Henryk Bem et al. that generally, it is not the ingestion of natural radionuclides with water but

inhalation of the radon escaping from water which is a substantial part of the radiological hazard due to the presence of the natural radionuclides from the uranium and thorium series in the drinking water (Bem et al., 2014). The calculated average annual ingestion (E_{ing}) dose due to absorption of radon in drinking water was 94.4 μSv.y⁻¹ with the range of 3.46 μSv.y⁻¹ to 265 μSv.y⁻¹. The

calculated average annual inhalation dose (E_{inh}) of water-borne radon was $969 \mu\text{Sv.y}^{-1}$ with the range of $35.5 \mu\text{Sv.y}^{-1}$ to 2.72 mSv.y^{-1} . So, the average total annual effective dose due to radon was 1.06 mSv.y^{-1} with the range of $39 \mu\text{Sv.y}^{-1}$ to 2.99 mSv.y^{-1} . The average radon activity concentration in underground water in the present study was compared to those of similar studies performed in different parts of the world. These values are given in Table 2. It is observed that the average

Table 3: comparison of average radon activity concentrations in water from this study with similar works.

Country	Locality	Number of samples N	Radon average activity concentration (Bq.L ⁻¹)	Reference in Bibliography
Burkina Faso	Villy	27	157.31±2.30	This work
Iran	Aliabad Katoul	16	2.90±0.57	(Adinehvand et al., 2016)
China	---	73	11.8	(Wu et al., 2018)
Nigeria	Southwest region	145	35.9	(Ajiboye et al., 2022)
India	Rajasthan	60	113	(Duggal et al., 2020)
Portugal	Covilha	33	352.8	(Inácio et al., 2017)
UNSCEAR	-	-	40	(Ajiboye et al., 2022)
WHO	-	-	100	(WHO, 2008)
European Union (EU)	-	-	100-1000	(Euratom, 2013)

And approximatively seventy percent of the measured radon activity concentrations in underground water, in this study, were above the reference levels of WHO and EU. However, these values were lower than the EU action level

radon activity concentration in this study is higher than those from Iran, China, Nigeria and is slightly higher than the average value of radon activity concentration in India. However, the average radon activity concentration in this study is lower than that obtained in Portugal. About eighty-one percent of the measured radon activity concentrations in underground water in the present study were higher than the limit set by UNSCEAR as presented in Table 3.

of 1000 Bq.L⁻¹. Accordingly, it does not pose any significant health hazards to the populace of Villy. The predicted radon contributions of each of the household waters, supplied by the different boreholes or wells studied at Villy, to the indoor air of a three-bedroom house were assessed and presented in figure 6.

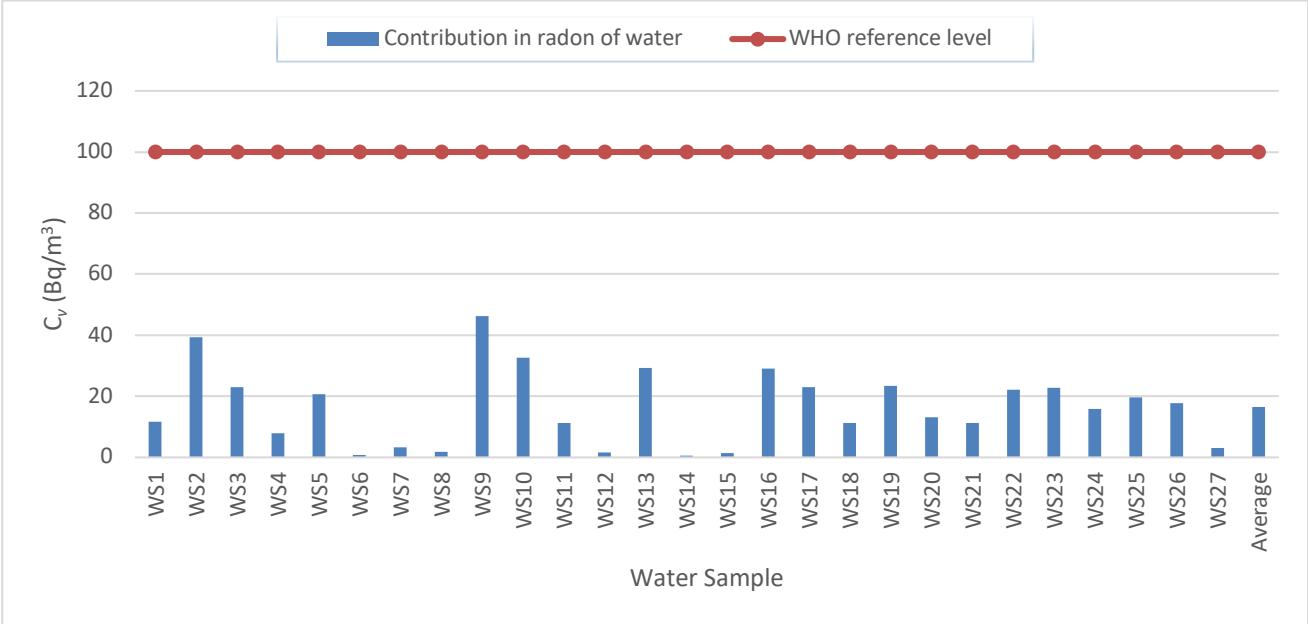


Figure 6: Contribution to indoor radon of each studied watering hole.

The predicted radon contributions of WS6, WS14 and WS15 were very low compared to those of WS2 and WS9. So, the more radon-rich the borehole, the more radon it

generates inside homes. Almost all the values were not negligible. Forty percent of the predicted radon contributions of the household waters to indoor air were above 20 Bq.m⁻³. The maximum value obtained is 46.2

Bq.m⁻³ which is almost the half of WHO reference level of radon in indoor air. Taking into consideration the emanation of radon from soil and building materials which are relatively important, the indoor radon could easily exceed the reference level of WHO at Villy. So, there may have the need to establish a country reference level of indoor radon.

IV. CONCLUSION

The average value of radon activity concentration in drinking water was 157.31±2.30 Bq.L⁻¹ with the range of 5.77±0.74 to 441.76±4.45 Bq.L⁻¹. The variation of radon concentration in underground water may be due to the depth of the water source and the geological structure of the studied area.

About seventy percent of the measured radon activity concentrations in underground water of Villy were above the reference levels of WHO and EU but were lower than the EU action level of 1000 Bq.L⁻¹. Accordingly, it does not pose any significant health hazards to the populace of Villy.

The average annual effective dose due ingestion of radon in drinking water and the average annual effective dose due inhalation of water-borne radon were respectively 94.4 µSv.y⁻¹ and 1.06 mSv.y⁻¹ with the respective ranges of 3.46 µSv.y⁻¹ to 265 µSv.y⁻¹ and 39 µSv.y⁻¹ to 2.99 mSv.y⁻¹.

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