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Radon Concentration Levels Estimation in Some Drinking Water Samples from Communities around Lumwana Mine in North Western Province of Zambia

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Abstract: The difference in the concentration of dissolved radon (^{222}Rn) in drinking water from wells and streams around Lumwana area in Zambia remains of interest because of the radiation-induced public health hazards. A total of nine (09) communities around Lumwana mine were selected for this study because their sources of drinking water are wells and streams. The underlying geology of this area is predominantly high grade, metamorphosed, intensely mylonitised, recrystallised muscovite–phlogopite–quartz–kyanite schists with disseminated sulphides (typically < 5%), dominated by chalcopyrite and bornite with known elevated concentrations of uranium, which ultimately decays to radon gas. The main aim of this paper is to estimate the contribution of (^{222}Rn) to public exposure due to natural radioactivity in drinking water. The concentration of radon levels in the collected water samples was analyzed using an Alpha Spectrometer called RTM (radon thoron monitor 2200) and the average results were found to vary from 4.44 Bq/L to 32.13 Bq/L. The obtained values are lower than the WHO (world health organization) recommended guidelines for drinking-water quality value of 100 Bq/L. The annual effective dose for the adults in these communities were in the range of 16.21 to 117.28 µSv/a slightly above the WHO recommended guideline reference dose level of 100 µSv/a.

Key words: Radon, drinking water sources, radon thoron monitor, alpha spectrometer.

1. Introduction

Radon (^{222}Rn) is a radioactive gas with a half-life of 3.82 days. It is the immediate progeny of radium (^{226}Ra), in the decay series of uranium (^{238}U). Estimation of total radiation dose to the world population has shown that about 96% is from natural sources while 4% is from artificial sources [1]. Globally, not much attention was given to public exposure to radon gas (^{222}Rn) until the 1980s when dangerous radon (^{222}Rn) levels were reported inside homes and schools in the United States of America [2]. Human beings are exposed to radon through inhalation or ingestion. Dissolved radon is released to air during usage of water, which adds to the dose received from inhalation of airborne radon emanating from the ground. In 1991, USEPA (the United States environmental protection agency) proposed a NPDWR (national primary drinking water regulation) for (^{222}Rn) Radon with a MCL (maximum contaminant level) of 11 Bq/L [3] which was revised by the NAS (national academy of sciences) and established an AMCL (alternative maximum contamination level) to 146 Bq/L [4]. In Zambia, according to the Environmental Management Licensing Regulations of 2013, there is no specific limit assigned to radon gas in drinking water. The limit for uranium discharge to the environment is 0.03 mg/L and zero discharge for any other radioactive materials.

The quality of groundwater used as a source of drinking water can vary widely. Groundwater is generally a drinking water source of good quality but the concentrations of radionuclides of natural origin are higher than those in surface water. Water can

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dissolve natural uranium interacting with rocks, soils, or mineralized bodies, and, as a consequence, anomalous uranium concentrations in ground water have been reported and attributed to a particular rock composition, mineralogy, geologic structure and ground water chemistry [5]. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels in the soils of each region in the world [6].

The geology of Lumwana Mine and the surrounding areas as shown in Fig. 1 contains uranium mineralogy hosted within the Mwombezhi Dome, which is a north-east trending basement dome in the western arm of the Neoproterozoic Lufilian Arc thrust

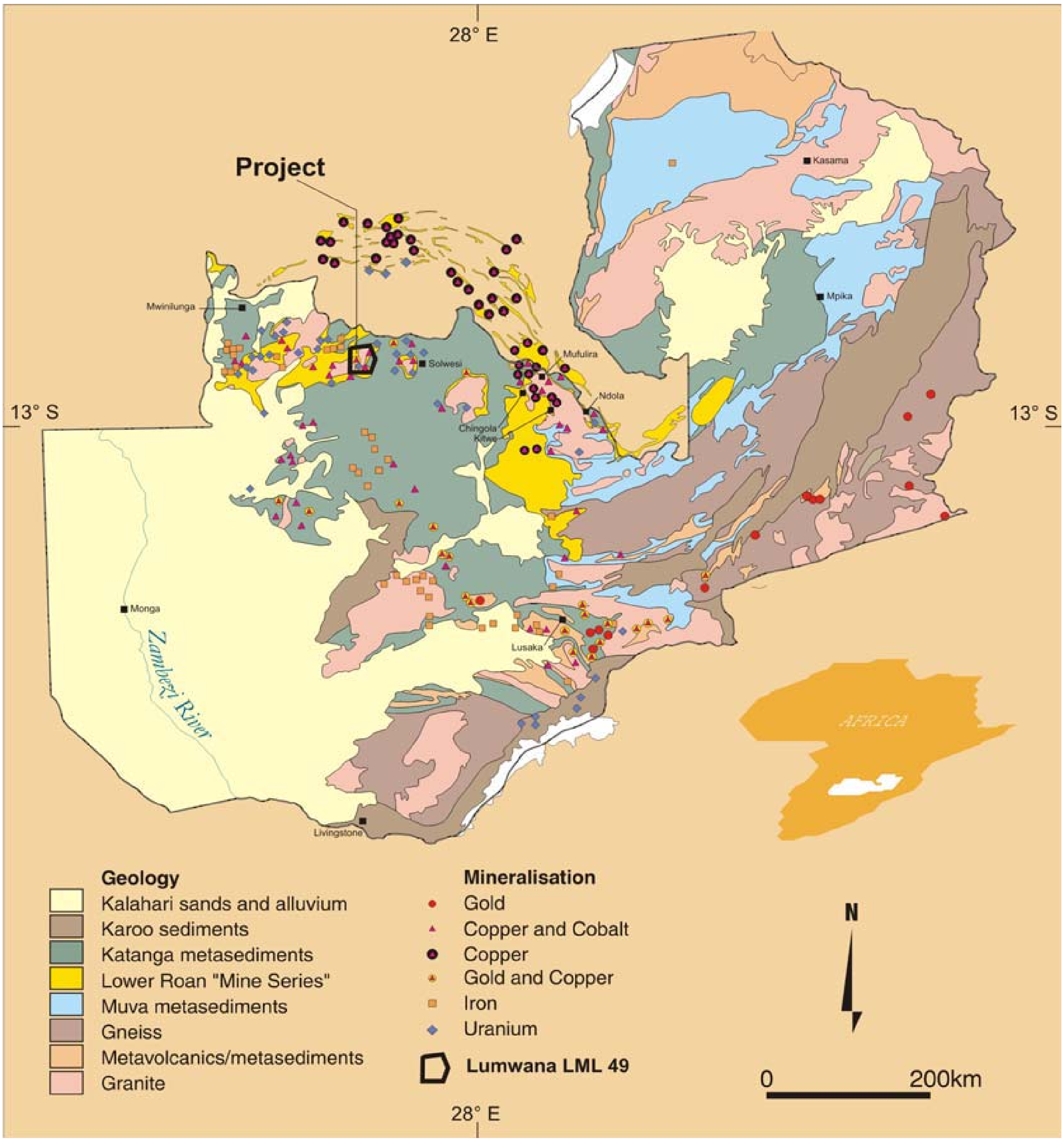

Fig. 1 Geological map of Lumwana area

Source: Extract from EIA-Lumwana mine project, report 5249/30, 2008.

fold belt. The Lufilian Arc is a major tectonic province characterized by broadly north-directed thrust structures and antiformal basement inliers or domes surrounded by Katanga metasediments which host the Central African Copperbelt [7]. In these areas with elevated concentration of uranium ore, the water quality may be compromised due to the presence of radon gas, a radioactive decaying daughter of uranium. Water passing through and over rocks and soil formations dissolves many compounds and minerals, including uranium; therefore, different amounts of it are present in some water sources. The primary health risk from radon is the development of lung cancer from the inhalation of air-born radon [8].

The main source of drinking water for the communities around Lumwana Mine as shown in Fig. 2 is mainly ground water from both shallow and deep wells. Water from Lumwana East River and its tributaries is not usually used for drinking for fear of being contaminated due to mining activities taking place at Lumwana Mine. The swallow wells are about 1 meter deep dug out along seasonal streams in marshlands and deep wells are about 10 meters found on higher grounds.

From the Lumwana Environmental Impact Assessment [9] baseline study conducted as a component of the BSF (bankable feasibility study) in 2003, chemical analyses of groundwater samples from wells at Mafuta village within the study area along T5 main road indicated that the drinking water was generally of good quality, although a pH of 5.8 was recorded in one well. Another well had a relatively high TDS (total dissolved solid) due to the presence of calcium, magnesium, potassium and sodium. The Lumwana East River and its major tributaries, the Malundwe stream and Chimiwungo stream, are the main water courses in the Lumwana mine project and surrounding areas. Surface water quality across the Lumwana mine area is similar and generally of good quality. However, aluminium, selenium, lead, uranium

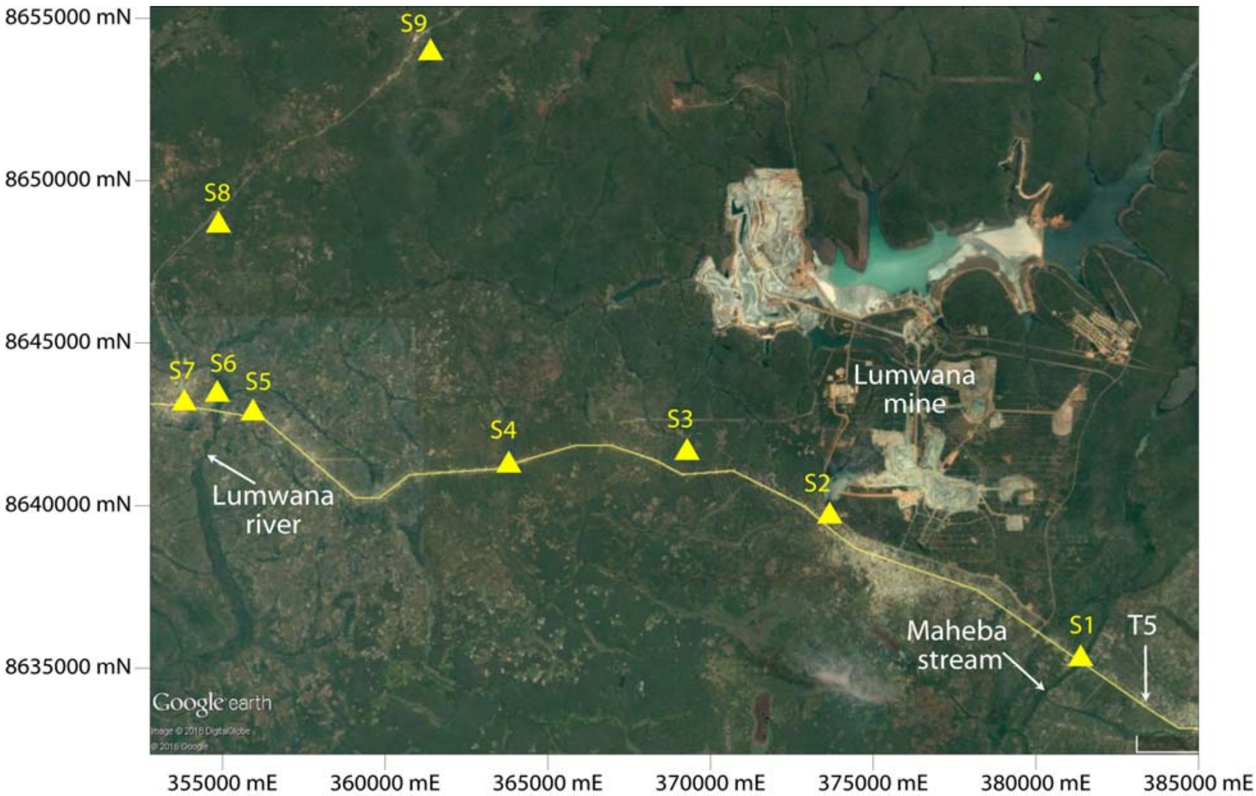

Fig. 2 Map of the sampling sites around Lumwana mine.

and cadmium concentrations occasionally exceed Zambian Drinking Water Standards Quality Standards [9].

In recent months, uranium mining in Zambia has become a matter of intense public debate and scrutiny. Real and perceived environmental pollution risks due to uranium mining and stockpiling at Lumwana Mine in North-Western Zambia have led to intense and sometimes heated public debates. These debates are not based on any sound scientific findings about the environmental pollution risk associated with mining activities at Lumwana mine. In order to better evaluate the radiological risks due to drinking water affecting the inhabitants of this region, a deeper knowledge of the radon concentration in the water is required, aimed at strengthening safety concerning drinking water quality as stipulated by the WHO (world health organization) guidelines [10], which have been adopted by Zambia. The WHO reference dose level for adults is 100 µSv from one year's drinking water consumption (730 L). The recommended screening levels for drinking water below which no further action is required are 0.5 Bq/L for gross alpha activity and 1 Bq/L for gross bêta activity.

In Zambia, while attention has been given to general water quality aspects, little has been done in relation to radiological aspects and a national limit for radon gas in drinking water has not been established. In this study, the dissolved radon levels in a number of potable water sources for communities around Lumwana mine were estimated. As far as we know, this is the first study to estimate the dissolved radon concentrations in drinking water in Zambia.

2. Methodology and Instrumentation

2.1 Sampling and Instrumental Setup

Dissolved radon levels in drinking water largely depend on the extent of aeration. Hence, to measure the actual public exposures levels for the inhabitants of Lumwana communities, water samples from the wells and rivers sources were collected. Radon concentrations in these samples were measured using RTM (radon thoron monitor) 2200, an electronic radon detector. Fig. 3 shows the schematic diagram of the RTM 2200.

In the setup, the RTM 2200 detector was used for measuring radon in water by connecting it with a bubbling kit which enables to degas radon from a water sample into the air in a closed loop. The radon concentration at the beginning of each measurement is zero. To ensure this, the monitor, all connection tubes were flushed with fresh air for at least 15 minutes using the internal pump of the monitor.

The equilibrium state between the air and the water activity concentration was attained after approximately 30 minutes of bubbling. Therefore, the measurements were taken at the earliest after 30 minutes of bubbling.

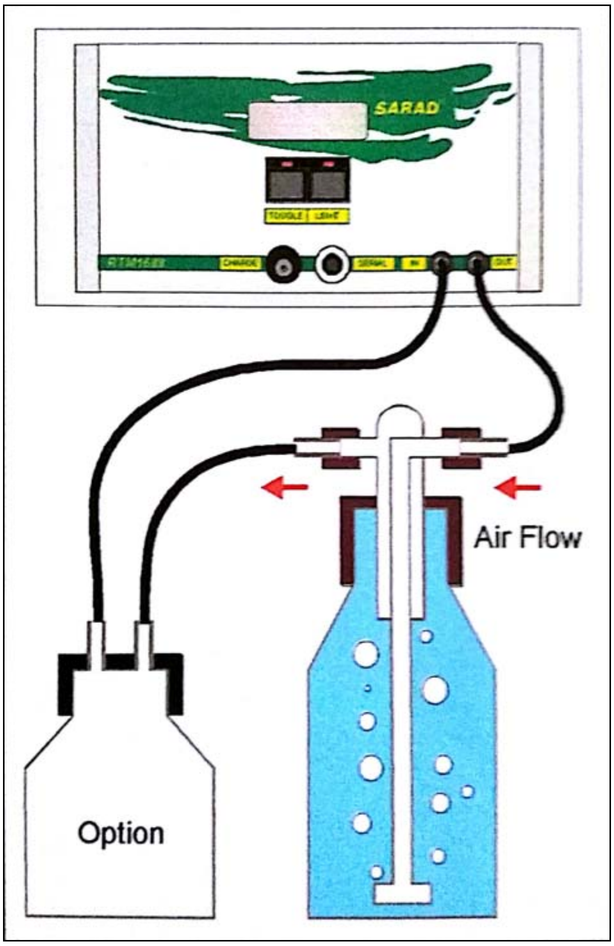

Fig. 3 Schematic diagram of RTM 2200 sampling setup.

A 500 mL sample of drinking water was taken in gas tight polythene sample bottles from each water source between November, 2013 and September, 2015. A total of 12 samples were collected and measured from each water source.

2.2 Measurement Theory of Radon Concentration of Water Samples

The Radon (Rn-222) gas concentration was measured by the short living daughter products, generated by the Radon decay inside the chamber. Directly after the decay, the remaining Po-218 nuclei become charged positively for a short period, because some shell electrons are scattered away by the emitted alpha particle. These ions are collected by electrical field across on the surface of a semiconductor detector. The number of collected Po-218 ions is proportional to the Radon gas concentration inside the chamber. Po-218 itself decays with a half life time of only 3.05 minutes and about 50% (particles emitted towards the detector surface) of all decays are registered by the detector.

The equilibrium between the Radon decay rate and Po-218 detector activity is given after about 5 half life times (15 minutes). This time span defines the minimum achievable response time to a Radon concentration step. The decay is continued by the both beta emitters Pb-214 and Bi-214 followed by another alpha emitter Po-214, which is delayed about 3 hours because of the superposed half life times of these nuclides. That means, each Po-218 decay causes one more detectable decay by Po-214.

The ratio between the activity concentrations depends only on the temperature of the water sample. This dependence can be expressed by the so-called Oswald coefficient which points out that the solubility of the Radon in water decreases if the water temperature increases.

That means, higher water temperatures result in higher Radon concentrations within the air as Eq. (1).

$$K_{Oswald} = C_{Rn}(Water)/C_{Rn}(Air)V \qquad (1)$$

The Oswald-coefficient can be approximated for the temperature range between 0 to 40 °C by the following Eq. (2):

$$K_{Oswald} = 0.425 * EXP\,(0.05 * \text{Temperature in °C}) + 0.1 \qquad (2)$$

The absolute concentrations within the sealed system are dependent on the original concentration of the water sample and on the ratio between the water and air volumes. The higher the water volume is compared to the air volume, the higher the expected activity concentration within the air volume is. Since the total activity within the system can be considered as a constant over the measurement period, the relationship can be stated by Eq. (3):

$$A\,(Water) = A_1\,(Water) + A\,(Air) \qquad (3)$$

with:

A (Water) = Total activity of the water sample before the de-gassing.

A_1 (Water) = Remaining activity of the water sample after the de-gassing.

A (Air) = Activity in the air volume.

Since Eq. (4):

$$C_A = A / V \qquad (4)$$

The following Eq. (5) can be expressed as:

$$C_A\,(Water) * V\,(Water) = C_{A1}\,(Water) * V\,(Water) + C_A\,(Air) * V\,(Air) \qquad (5)$$

with:

C_A (Water) = Activity concentration of the water sample before the de-gassing

V (Water) = Volume of the water probe (constant)

C_{A1} (Water) = Activity concentration of the water sample after the de-gassing

V (Air) = Air volume within the sealed system

C_A (Air) = Activity concentration of the air within the system after the de-gassing

Inserting Eq. (1) in Eq. (5) results in Eq. (6):

$$C_A\,(Water)*V\,(Water) = K_{Osward}*C_A\,(Air)*V\,(Water) + C_A\,(Air)*V\,(Air) \qquad (6)$$

Leading to Eq. (7):

$$C_A\,(Water) = \{C_A(Air) * [K_{Oswald} * V(Water) + V(Air)]\} / V(Water) \qquad (7)$$

The estimated annual effective dose by ingestion was calculated because of habitual consumption of water by the members of the communities around Lumwana mine. It was computed using the formula in Eq. (8):

$$DW = C_W * C_{RW} * Dcw \quad (8)$$

where DW is the annual effective dose (μSvy^{-1}) due to ingestion of radionuclide from the consumption of water; C_w is the concentration of radon in the ingested drinking water ($Bq{\cdot}L^{-1}$); C_{RW} is the annual intake of drinking water ($1\ y^{-1}$), and Dcw is the ingested dose conversion factor for Radon ($Sv{\cdot}Bq^{-1}$).

3. Results

The activity concentrations expressed in Becquerel per litre (Bq/L) of radon in drinking water from swallow wells, deep wells and rivers are shown in Table 1. The detection limit for the sample volume of 500 mLs with air volume of 300 mLs for tubing and internal Radon monitor volume was 0.033 Bq/L at 2-sigma with sensitivity of the radon monitor of 3 kBq/m^3.

The committed effective doses for adults shown in Table 2 via the consumption of drinking water from wells and rivers were calculated from the activity concentration results of Table 1. The effective dose is a product of the dose coefficient, the activity concentration and the amount of water consumed annually [11]. A dose conversion factor of $5*10^{-9}$ $Sv{\cdot}Bq^{-1}$ was used as suggested by the United Nation Scientific Committee on the Effects of Atomic Radiation [6]. An average of 730 L of drinking water was estimated for an adult (age 18 years) [12]. Based on the average activity concentrations for each water source for the period of 12 months (see Table 1), the annual committed effective doses to the adult population for each water source were assessed and shown in Table 2. The average activity concentrations and annual effective doses are also presented graphically in Figs. 4 and 5, respectively.

Table 1 **Radon activity concentrations in drinking water for communitties around lumwana mine (Bq/L).**

Sampling date	Maheba river	Wamukonji village	Kiwala village	R5 dw	Dan shimo village	Shinda school	Samuhanga village	Lumwana east river	Kamwana village	Lumwana east clinic	Mukwemba village	Mukumbi lubinga school
20/11/2013	7.87 ± 0.08	3.73 ± 0.02	15.14 ± 0.03	16.06 ± 0.11	15.63 ± 0.07	6.56 ± 0.09	4.84 ± 0.03	3.52 ± 0.04	3.77 ± 0.04	4.40 ± 0.06	12.92 ± 0.05	30.06 ± 0.05
10/02/2014	6.46 ± 0.07	4.39 ± 0.04	43.81 ± 0.02	31.46 ± 0.11	6.72 ± 0.09	3.82 ± 0.13	8.08 ± 0.10	5.87 ± 0.04	5.91 ± 0.11	2.53 ± 0.05	22.56 ± 0.03	30.38 ± 0.11
24/04/2014	5.61 ± 0.09	3.69 ± 0.06	68.96 ± 0.13	23.52 ± 0.12	19.36 ± 0.11	18.42 ± 0.10	9.28 ± 0.03	6.48 ± 0.11	5.42 ± 0.10	3.16 ± 0.12	15.91 ± 0.08	20.05 ± 0.12
25/05/2014	3.73 ± 0.15	3.52 ± 0.07	56.47 ± 0.04	17.44 ± 0.11	32.15 ± 0.08	7.43 ± 0.05	16.03 ± 0.02	4.97 ± 0.07	6.54 ± 0.01	3.02 ± 0.12	13.36 ± 0.03	22.05 ± 0.02
16/06/2014	3.94 ± 0.04	3.14 ± 0.06	31.16 ± 0.07	15.16 ± 0.07	24.22 ± 0.02	15.22 ± 0.08	3.71 ± 0.07	5.20 ± 0.05	5.54 ± 0.04	5.73 ± 0.09	15.89 ± 0.08	20.18 ± 0.03
28/07/2014	3.19 ± 0.03	4.57 ± 0.03	16.64 ± 0.03	29.42 ± 0.05	6.53 ± 0.03	22.30 ± 0.03	16.30 ± 0.03	11.97 ± 0.04	8.54 ± 0.03	13.34 ± 0.03	11.42 ± 0.03	23.60 ± 0.03
06/10/2014	3.85 ± 0.01	7.68 ± 0.01	16.73 ± 0.02	11.54 ± 0.01	11.53 ± 0.03	17.08 ± 0.02	13.49 ± 0.03	10.09 ± 0.03	6.03 ± 0.02	10.62 ± 0.03	35.04 ± 0.02	24.32 ± 0.17
01/12/2014	6.61 ± 0.01	6.36 ± 0.01	19.41 ± 0.03	18.32 ± 0.02	7.45 ± 0.07	7.93 ± 0.01	6.28 ± 0.01	4.66 ± 0.01	2.78 ± 0.01	4.92 ± 0.01	16.29 ± 0.07	11.24 ± 0.08
09/02/2915	1.61 ± 0.02	3.09 ± 0.05	20.39 ± 0.03	14.79 ± 0.05	20.77 ± 0.02	11.62 ± 0.02	13.05 ± 0.02	17.23 ± 0.04	11.53 ± 0.06	26.95 ± 0.01	34.93 ± 0.03	47.44 ± 0.09
04/05/2015	6.16 ± 0.04	7.79 ± 0.03	26.22 ± 0.02	8.08 ± 0.06	14.96 ± 0.05	13.96 ± 0.07	7.79 ± 0.08	7.55 ± 0.06	16.22 ± 0.05	15.13 ± 0.03	35.86 ± 0.06	31.63 ± 0.04
29/06/2015	2.12 ± 0.01	6.95 ± 0.01	39.04 ± 0.02	18.08 ± 0.01	12.03 ± 0.01	13.96 ± 0.02	3.21 ± 0.01	6.32 ± 0.01	9.65 ± 0.02	12.37 ± 0.02	49.79 ± 0.02	25.80 ± 0.01
29/09/2015	2.15 ± 0.01	3.90 ± 0.01	31.60 ± 0.01	11.22 ± 0.01	14.69 ± 0.02	16.71 ± 0.01	2.08 ± 0.01	3.50 ± 0.01	1.49 ± 0.01	6.41 ± 0.01	19.47 ± 0.01	23.31 ± 0.01
Average	4.44 ± 0.05	4.90 ± 0.04	32.13 ± 0.04	17.92 ± 0.04	15.50 ± 0.03	12.92 ± 0.04	8.68 ± 0.03	7.28 ± 0.03	6.95 ± 0.04	9.05 ± 0.04	23.62 ± 0.03	25.84 ± 0.05

Table 2 **Annual average effective doses (μSv/a).**

NO.	Sampling site	Annual average effective dose (μSv/a)
1	Maheba river	16.21
2	Wamukonji village	17.89
3	Kiwale village	117.28
4	R5 deep well	65.42
5	Dan simo village	56.59
6	Shinda village	47.15
7	Sakuhang’a village	31.67
8	Lumwana east river	26.57
9	Kamwana village	25.37
10	Lumwana east clinic	33.01
11	Mukwemba village	86.21
12	Mukumbi lubinga school	94.33

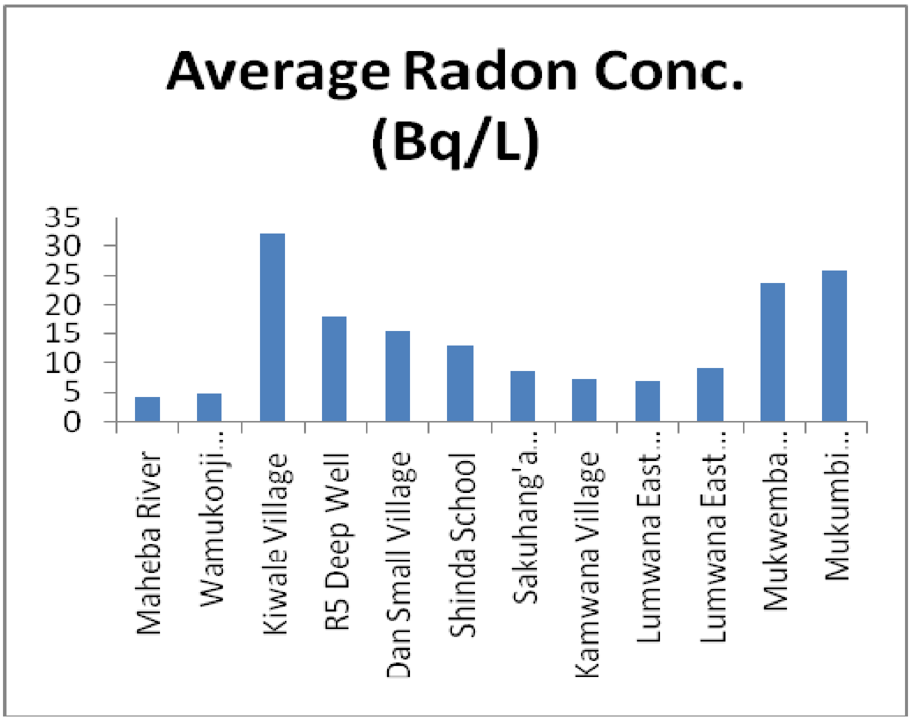

Fig. 4 Average radon concentration (Bq/L).

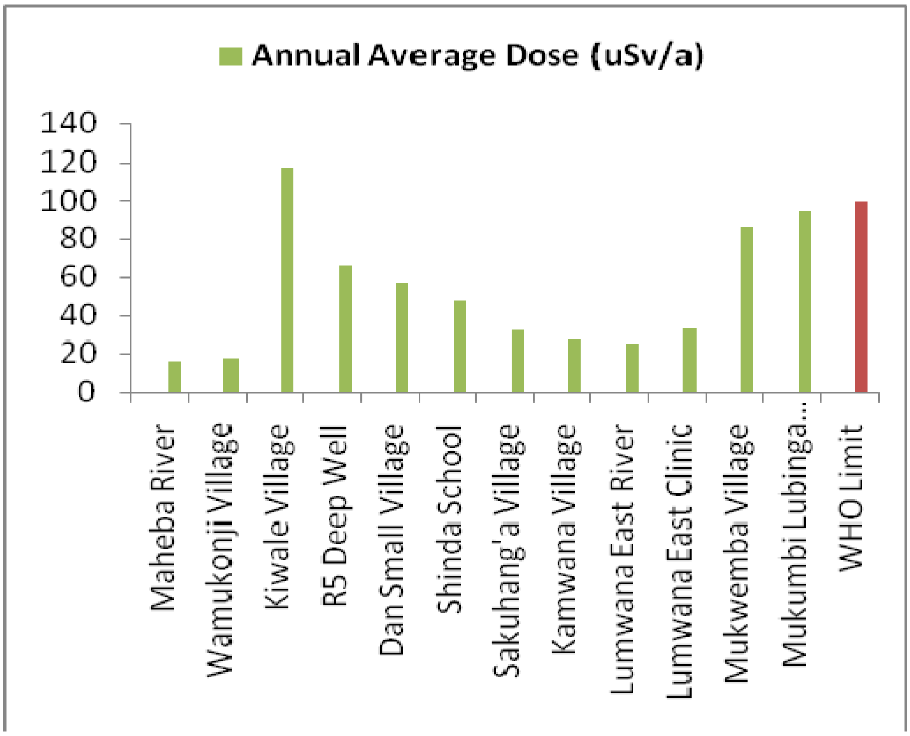

Fig. 5 Annual average effective dose (μSv/a).

4. Discussion

The study results show that the activity concentrations of radon in drinking water from the three sources, swallow wells, deep wells and rivers can vary over a wide range; Maheba River: 1.61-7.87 Bq/L, Wamukonji Village (deep well): 3.14-7.79 Bq/L, Kiwale Village (swallow well): 16.64-68.96 Bq/L, R5 (deep well): 8.08-31.46 Bq/L, Dan Simo (swallow well): 6.53-32.23 Bq/L, Shinda School (deep well): 3.82-22.30 Bq/L, Sakuhang, a Village (deep well): 2.08-16.03 Bq/L, Kamwana Village (deep well): 1.49-16.22 Bq/L, Lumwana East River: 3.52-17.23 Bq/L, Mukwemba Village (swallow well): 11.42-49.79 Bq/L, and Mukumbi Lubinga School (swallow well): 11.24-47.44 Bq/L.

The results indicate that 100% of the sampling sites in this study had radon water concentration below the international standard limits such as the currently debated U.S. AMCL (alternative maximum concentration level) of 146 Bq/L as an upper limit for drinking water in the United States [4] and the European Union Reference level of 100 Bq/L [13]. 50% of the sampling sites had radon concentration levels above the U.S. Environmental Protection Agency limit of 11 Bq/L. The finding is similar to the situation reported on the radon concentration in groundwater [14-16].

5. Conclusions

It can be concluded that generally water from swallow wells had elevated concentrations of radon compared to water from deep wells and rivers. This could be attributed to the geological variance of the sampling sites. Kiwale Village (swallow well) with the highest annual effective dose of 117.28 μSv above WHO guideline [17] value of 100 μSv lies almost in the same geological grid with Chimiwungo open pit at Lumwana mine with uranium mineralogy. This suggests that citing of wells in this area should be based on the good knowledge of the geological map of the area to avoid uranium rocks underground.

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