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Radiation dose assessment and radon removal efficiency of water treatment phases in some selected table water production factories in Ado-Ekiti, Southwest, Nigeria

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Abstract

The natural occurrence of radon in drinking water can be hazardous to human air pathways and stomach. Assessment of radon removal efficiencies of drinking water processing phases for public consumption is therefore very important. Measurement of radon concentration has been carried in drilled wells sources for table water production and across the three treatment phases at ten water plants in Ado Ekiti, Nigeria. Average activity concentrations of ^{222}Rn of drilled well source, aeration/dosing, sand-granular activated carbon (GAC) filtration and ultraviolet sterilization phases are 63.6, 27.0, 25.8 and 17.1 Bq L^{-1} respectively. Cumulative average radon removal efficiencies at the three phases are 60.0%, 64.2% and 80.7%, respectively. Radon concentrations in all the processing phases exceed the 0.1 Bq L^{-1} maximum permissible level set by Standard Organization of Nigeria. Estimated radiation dose to the stomach through table water ingestion and occupational dose to water treatment plant workers are 0.0247 mSv y^{-1} and 0.044 mSv y^{-1} respectively.

Keywords: Radon, GAC filtration, UV, radon removal efficiency

1. Introduction

Human exposure to natural radiation sources is an inevitable aspect of life. Man is exposed from cosmic radiation which have its origin from outer space and primordial radionuclides which exist in all environmental media such as groundwater, surface water, soil and air. The primordial radionuclides include those in the uranium (^{238}U), thorium (^{232}Th) and actinium series and non-series potassium (^{40}K). According to the United Nations Scientific Committee on the Effects of Atomic Radiation, the total radiation dose received by man from these natural sources is estimated as 2.4 mSv per year with radon(^{222}Rn) accounting for about half of this radiation dose (UNSCEAR, 2000).

Radon is a radioactive gas which exists naturally in low concentration almost everywhere on earth. It is a colourless and odourless gas which exhibit fair solubility (230 cm^3/L at 20 ℃) in water (Wojcik et al., 2017). Radon is a decay product of ^{226}Ra which is present in uranium bearing rocks. Due to the moderate solubility of ^{226}Ra in water, it enters into groundwater system by desorption from uranium bearing rocks. The ^{226}Ra ($t_{1/2} = 1600\ y$) decays into ^{222}Rn ($t_{1/2} = 3.8\ d$) by alpha particle emission. Radon is therefore present in groundwater with varying concentrations depending on the geology of the environment (Al-Shereideh et al., 2006; Barbosa-Lorenzo et al., 2017; Minda et al., 2009). The radioactive decay products of ^{222}Rn are short-lived radioisotopes which emit alpha and beta particles. Over half of radiation exposure received from natural sources is due to radon (ICRP, 1993). Water-borne radon is a known carcinogen with the potential of causing considerable level of radiation exposure to the stomach (Auvinen et al., 2005) through ingestion and when released into the air is a potential cause of lung cancer (López-Abente et al., 2018; Messier & Serre, 2017). In the last two decades, production factories of table water has sprung up in most places in Nigeria due to the shortage of safe drinking portable water supply in Nigeria (Edema *et al*., 2011; Adekunle *et al*., 2004). As water passes through the different water treatment phases in the production factories, the level of radon concentration in the water reduces through mechanical process, thermally or by natural decay. The present study is therefore posed to assess the radon removal efficiency of the water production phases in the water processing plants, determine the radiation dose from ingestion of water to public and occupational hazard to water production plant workers. Sequence of table water production in the study area is from drilled well to aeration/dosing to sand-GAC filtration to UV sterilization (Fig. 1). The objectives of the aeration/dosing phase is to allow the groundwater to have more contact area with air thereby diffusing unwanted gaseous pollutant present in the water and oxidising the natural organic matter (Ghernaout, 2019). The sand-GAC system acts to filter out sediments from the water with the activated carbon section removing gaseous content of the water. Generally, the sand-GAC system is employed in potable water treatment plants for monitoring of odour and synthetic organic compounds. It has however proved viable for remediation of radon in water. When radon-borne water passes through the GAC system, radon sorbs to the GAC. The GAC, usually designed as a cylindrical column, presents more surface area to the influx of water. As the flow of water advances through the GAC column, the radon is adsorbed to the carbon until all the accessible interfacial area is saturated. The UV treatment phase is a coupled system of UV lamp and thermal pouch sealer which primarily serves to kill microorganisms in the water by scrambling the organisms' DNA. In addition, the thermal energy from the system tends to evaporate some of the dissolved radon in water before bottling.

The maximum contaminant level (MCL) from radon in drinking water for the public is regulated in different parts of the world. In the United States, the US Environmental Protection Agency puts the MCL at 11.1 Bq l^{-1}. In Nigeria, the Standard Organization of Nigeria puts the MCL for radionuclides in drinking water at 0.1 Bq l^{-1} (SON, 2007). The World Health Organisation (WHO) also puts the MCL at 100 Bq L^{-1} for groundwater (WHO, 2008). Most groundwater sources in the area have radon concentrations exceeding the MCL, which implies the water sources would have to be treated in order to drastically reduce or completely remove the radon content.

2. Description of the study area

The study area, Ado-Ekiti is the administrative headquarters of Ekiti State. The area is located in the Southwest region of Nigeria and has a population of about 2,400,000. It lies within the coordinate $7^{o}40^{'}N$ and $5^{o}15^{'}E$ (Fig. 2). The area is underlain by the Precambrian Basement Complex rocks. The basement rocks are characterised with relatively thick overburdens. The major lithology includes granite gneiss, undifferentiated schist, migmatite, porphyritic granite, biotite granite, granodiorite and older granite. The complex formation of the basement complex rocks impacts permeability and porosity which are essential for a robust groundwater occurrence. However, the deformation of the Basement Complex has resulted in the formation of joints and fractures which significantly enhance the groundwater occurrence. The hydrogeology of the area can broadly be classified into surface water and groundwater. The groundwater is widely used in table water production. The occurrence of groundwater is usually within the pore spaces of sediments and also in weathered and fractured basement columns (Bayowa et al., 2014). In the area, water table falls progressively throughout dry season. Most of hand dug wells (within < 20 m depth) dries up during the dry season. Thus many households and water production plants rely on drilled wells (with depths > 40 m) for constant supply of drinking water.

3. Experimental detail

Ten water treatment plants across Ekiti State, Nigeria were sampled to assess the radon removal efficiencies of water processing phases. Four water production phases were considered: drilled well source, dosing/aeration, sand-granulated activated carbon (GAC) filtration and ultraviolet sterilization. Water samples from deep wells were collected directly at the well sites. To obtain representative water samples from the deep wells, water was pumped out of the well for several minutes before the samples were taken. The water samples were obtained at elbow joint of the pipe while avoiding contact with air. Each water sample was fed into a 250 ml Rad-H_2O sample vial. Water samples collected at the dosing/aeration phase were obtained at the base of the dosing tanks in similar way as that of the samples from deep wells. However, the samples were taken at (>30 minutes) after dosing to ensure chemical concentration equilibrium. Water samples which have passed through the GAC system were obtained from the flushing tap of the GAC system. It was ensured that no bubbling took place while obtaining the sample. Water samples of packaged table water which have passed through the UV sterilization machine were also taken. Water samples from the different water production phases were measured for radon concentration using RAD7 radon detector with RADH_2O accessories.

3.1 Measurement technique of radon concentration in water

Fig. 3 shows a schematic diagram describing the set-up for measurement of radon concentration in water. Typically, the set-up comprises of the RAD7 unit, drying agent ($CaSO_4$) and water aerator kits. The RAD7 detector is a solid state detector which has a 0.7 litre hemispherical sample cell. The internal surface of the hemispheric cell is coated with an electrical conductor. At the centre of the hemispheric sampling cell is located a solid state ion-implanted planar silicon alpha detector. A high voltage between 2000 V and 2500 V supplied by the electronic circuitry of the device is applied between the detector and the hemispheric conductor to create electric field throughout the volume of the sampling cell. Before commencement of measurement, the Rad7 unit was connected with the laboratory drying unit and purged with fresh and dry air until the relative humidity in the sample cell dropped below 6% in order to ensure the relative humidity does not rise above reasonable level throughout the measurement period. For the measurement, the RAD7 unit was connected in closed loop with the water aerator kits and was operated in WAT-250 protocol. This protocol is programmed such that the water sample in the sample vial is aerated for 5 minutes thereby degassing the radon content of the water into the sample cell of the RAD7. This protocol has a radon extraction efficiency of about 95%. After the degassing process the RAD7 unit rest for a period of 5 minutes so that radioactive equilibrium would be reached between ^{222}Rn and its daughter ^{218}Po ($t_{1/2} = 3.04$ minutes). In a subsequent four 5-minute cycles, the Rad7 determines the ^{222}Rn concentrations from the ^{218}Po decay events within the sample cell of the detector. It thereafter determines the average concentration of the four radon concentrations.

3.2 Computation of radon removal efficiency

The radon removal efficiency (RRE) is the percentage of radon which has been removed by a phase of water production. It is computed using the following expression:

$$RRE = \frac{Original\ Radon\ Conc. - New\ Radon\ Conc.}{Original\ Radon\ Conc.} \times 100\% \qquad (1)$$

3.3 Estimation of radiation dose

Ingestion of radon-borne water presents radiation risk to the stomach. Determination of radiation dose to stomach is therefore important. The annual effective dose to stomach from ingestion of water, D_{Stom}, was estimated using the Equation (2) as given in (UNSCEAR, 2000).

$$D_{Stom} = C_{Rn} \times V \times F \qquad (2)$$

where C_{Rn} is the experimentally determined radon concentration in water in $Bq\ l^{-1}$, V is water consumption per year ($l\ y^{-1}$), and F is dose conversion factor in $Sv\ Bq^{-1}$. For computation of D_{ing} for infants, children and adult, water consumption rate of 230, 330 and 730 $l\ y^{-1}$ were used respectively. The respective radon F values used are 23, 5.9 and 3.5 nSv Bq^{-1}.

Operators of drinking water treatment plants are also at risk of radon exposure from radon released into air during water treatment activities that cause the water to be agitated such as backwashes, aeration or other agitation. Occupational radiation dose due to inhalation is evaluated using equation (3) as given in (UNSCEAR, 2000):

$$D_{Occ} = C_{Rn} \times AWR \times OF \times Eq \times DCF \qquad (3)$$

where AWR is the air-water concentration ratio of 10^{-4}, OF is the occupancy factor of 1920 $h\, y^{-1}$ (estimated from 40 h workweek), Eq is the equilibrium factor of 0.4, and DCF is the dose conversion factor of 9 $nSv\ (Bq\ h\ m^{-3})^{-1}$.

4. Results and Discussions

The ^{222}Rn concentration in the four phases of water production across the ten water production plants considered for the study is presented in Table 1. The measured radon concentrations for the drilled well source range from 13.4 Bq L^{-1} at Plant 5 to 251.2 Bq L^{-1} at Plant 3 with average value of 63.6 $\pm$ 70.5 Bq L^{-1}. The distribution exhibits high standard deviation due to high spread of the data. The raw groundwater sample obtained at Plant 3 exceeds the European Union's parametric value of 100 Bq L^{-1} (European Commission, 2013). At the aeration/dosing phase, the concentration range is from 6.1 Bq L^{-1} to 139.8 Bq L^{-1} with average value of 27.0 $\pm$ 40.7 Bq L^{-1}. In the Sand-GAC phase, the radon concentration values range from below detectable limit (BDL) to 142.1 Bq L^{-1} with average value of 25.7 $\pm$ 42.3 Bq L^{-1}. At the UV packaging phase, the concentration values range from 1.2 Bq L^{-1} to 116.9 Bq L^{-1} with average value of 17.1 $\pm$ 35.3 Bq L^{-1}. Generally the activity concentration values across the different phases of production exhibit high standard deviation. This is attributable to the high variability of radon concentration values from Plant to Plant. 20% of the assayed packaged water product have concentration above the maximum contamination limits of US Environmental Protection Agency and World Health Organization (WHO, 2008).

The mechanical flow of groundwater through different phases of table water production can reduce the radon concentration in the water. The cumulative radon removal efficiency (RRE) of the different phases of table water production in the study area is presented in Table 2. The cumulative RRE of aeration/dosing phase varies from 40.3% to 88.1% with average value of 60%. RRE for Sand-GAC phase varies from minus 21.6% to 100% with average value of

64.2%. In the UV sterilization/packaging phase, the cumulative RRE ranges from 53.5% to 96.5% with an average value of 80.7%. As presented in Fig. 4, a test of relationship between radon concentration level and RRE revealed a very weak positive relationship of $r = 0.153$. Table 3 presents the percentage radon residue in table water available for public consumption. Across the ten treatment plants, the percentage radon residue varies from 3.5% to 46.5% with an average value of 19.3%. This average value indicates that the water treatment phases possess the capability of removing more than 80% of the dissolved radon content of water. The obtained value in this study is comparable to an average of 74.9% obtained in water treatment plants in South Korea (Cho et al., 2020). Fig. 5 shows a histogram displaying the percentage radon removal for the water processing phases and their corresponding percentage radon residue. As seen from the figure, all the contributions from the water processing phases and the residual radon sums to 100%. Generally, radon concentration reduces across the water processing phases (Fig. 6), although there are relatively few spikes which are attributable to radon released from trapped ^{226}Ra atoms within the sand-GAC profile.

Evaluation of annual effective dose to the stomach D_{Stom} from ingestion of radon-in-water revealed that the D_{Stom} for infants ranges from 0.001 to 0.094 mSv y^{-1} with an average of 0.014 ± 0.027 mSv y^{-1}. In children, the D_{Stom} range from 0.001 to 0.135 mSv y^{-1} with an average value of 0.020 ± 0.039 mSv y^{-1}. For adults, the D_{Stom} values range from 0.003 to 0.295 mSv y^{-1} with an average value of 0.044 ± 0.086 mSv y^{-1}. Although some of the D_{Stom} values obtained for children and adults exceed the recommended reference dose level of 0.1 mSv set by World Health Organization (WHO, 2008), the average D_{Stom} values for infants, children and adults are below the reference level.

Occupational radiation dose due to inhalation of radon, D_{Occ}, assuming a 40-hour workweek range from 0.009 to 0.174 mSv y^{-1} with an average value of 0.044 ± 0.046 mSv y^{-1}. Although, an occupational dose limit has not been set in Nigeria, a comparison with the existing limit of 10 mSv y^{-1} set by the International Commission on Radiological Protection (ICRP, 2014) shows that the estimated occupational dose is much lower.

5. Conclusion

The result from present study revealed the presence of residual radon in table water. The cumulative average radon removal efficiencies for aeration/dosing, GAC filtration and ultraviolet sterilization water processing phases are 60.0%, 64.2% and 80.7% respectively. Table water products exhibit average residual radon of 19.3%. Residual radon concentrations range from very low to levels above recommended limits. The average radiation dose from water samples from the treatment plants is below the recommended limit.

Nigeria water quality regulatory agency is yet to propose permissible limit of radon concentration in drinking water. A nation-wide survey is therefore necessary to enable the regulatory agencies come up with informed guidelines to limit public exposure to deleterious effect of radiation from radon.

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Authors Contributions

Dr. Yinka Ajiboye and Dr. Matthew O. Isinkaye both designed the study and were both involved in the writing of the manuscript.

Table 1: Radon concentration in water processing phases

s/n	**Water Plant Code**	**Drilled well source ($Bq\ l^{-1}$)**	**Aeration/Dosing ($Bq\ l^{-1}$)**	**Sand-GAC coupled filter ($Bq\ l^{-1}$)**	**UV Sterilization ($Bq\ l^{-1}$)**
1	Plant 1	40.3 ± 2.5	13.3 ± 0.8	12.5 ± 0.6	6.1 ± 0.9
2	Plant 2	33.9 ± 3.4	6.1 ± 1.4	3.3 ± 0.2	1.2 ± 0.3
3	Plant 3	251.2 ± 5.8	139.8 ± 4.5	142.1 ± 7.3	116.9 ± 5.3
4	Plant 4	16.2 ± 3.6	8.0 ± 2.6	9.6 ± 2.8	6.5 ± 2.3
5	Plant 5	13.4 ± 3.8	8.0 ± 2.6	10.5 ± 2.9	3.0 ± 1.8
6	Plant 6	85.0 ± 8.0	10.1 ± 2.9	5.7 ± 2.2	4.7 ± 2.0
7	Plant 7	71.4 ± 8.0	29.2 ± 4.9	37.1 ± 5.5	15.2 ± 3.5
8	Plant 8	13.7 ± 3.4	6.3 ± 2.3	BDL	1.2 ± 1.0
9	Plant 9	65.3 ± 8.2	32.9 ± 4.3	25.8 ± 5.4	8.6 ± 2.0
10	Plant 10	45.6 ± 3.6	16.2 ± 1.4	11.2 ± 1.0	7.4 ± 1.5
	Range	**13.4 – 251.2**	**6.1 – 139.8**	**BDL – 142.1**	**1.2 – 116.9**
	Average	**63.6 ± 66.9**	**27.0 ± 38.7**	**25.8 ± 40.1**	**17.1 ± 33.5**

BDL – Below Detectable Limit

Table 2: **Cumulative radon removal efficiency of water production phases**

s/n	Water Plant Code	Aeration/Dosing (%)	Sand-GAC coupled filter (%)	UV Sterilization (%)
1	Plant 1	67.0	69.0	85
2	Plant 2	82.0	90.3	96
3	Plant 3	44.3	43.4	53
4	Plant 4	50.6	40.7	60
5	Plant 5	40.3	21.6	78
6	Plant 6	88.1	93.3	94
7	Plant 7	59.1	48.0	79
8	Plant 8	54.0	100.0	91
9	Plant 9	49.6	60.5	87
10	Plant 10	64.5	75.4	84
	Min	**40.3**	**21.6**	**53.5**
	Max	**88.1**	**100.0**	**96.5**
	Average	**60.0**	**64.2**	**80.7**
	Std Dev	**15.7**	**25.9**	**14.2**

Table 3: Percentage radon removal and radon residue

s/n	Plant	Drilled well (%)	Aeration/Dosing (%)	Sand-GAC (%)	UV/Packaging (%)	Total removed radon (%)	Residual radon (%)
1	Plant 1	100	67.0	2.0	15.9	84.9	15.1
2	Plant 2	100	82.0	8.3	6.2	96.5	3.5
3	Plant 3	100	44.3	-0.9	10.0	53.5	46.5
4	Plant 4	100	50.6	-9.9	19.1	59.9	40.1
5	Plant 5	100	40.3	-18.7	56.0	77.6	22.4
6	Plant 6	100	88.1	5.2	1.2	94.5	5.5
7	Plant 7	100	59.1	-11.1	30.7	78.7	21.3
8	Plant 8	100	54.0	46.0	-8.8	91.2	8.8
9	Plant 9	100	49.6	10.9	26.3	86.8	13.2
10	Plant 10	100	64.5	11.0	8.3	83.8	16.2
	Min	100.0	40.3	-18.7	-8.8	53.5	3.5
	Max	100.0	88.1	46.0	56.0	96.5	46.5
	Average	100.0	60.0	4.3	16.5	80.7	19.3

Table 4: **Annual ingestion and inhalation doses**

s/n	Plant ID	UV/Packaging (Bq L^{-1})	Ingestion Dose mSv/y (Infant)	Ingestion Dose mSv/y (Children)	Ingestion Dose mSv/y (Adult)	Occupational Inhalation Dose (mSv/y)
1	Plant 1	6.1	0.005	0.007	0.016	0.028
2	Plant 2	1.2	0.001	0.001	0.003	0.023
3	Plant 3	116.9	0.094	0.135	0.299	0.174
4	Plant 4	6.5	0.005	0.008	0.017	0.011
5	Plant 5	3	0.002	0.003	0.008	0.009
6	Plant 6	4.7	0.004	0.005	0.012	0.059
7	Plant 7	15.2	0.012	0.018	0.039	0.049
8	Plant 8	1.2	0.001	0.001	0.003	0.009
9	Plant 9	8.6	0.007	0.010	0.022	0.045
10	Plant 10	7.4	0.006	0.009	0.019	0.032
	Average	**17.1**	**0.014**	**0.020**	**0.044**	**0.044**
	Std Dev	**33.5**	**0.027**	**0.039**	**0.086**	**0.046**

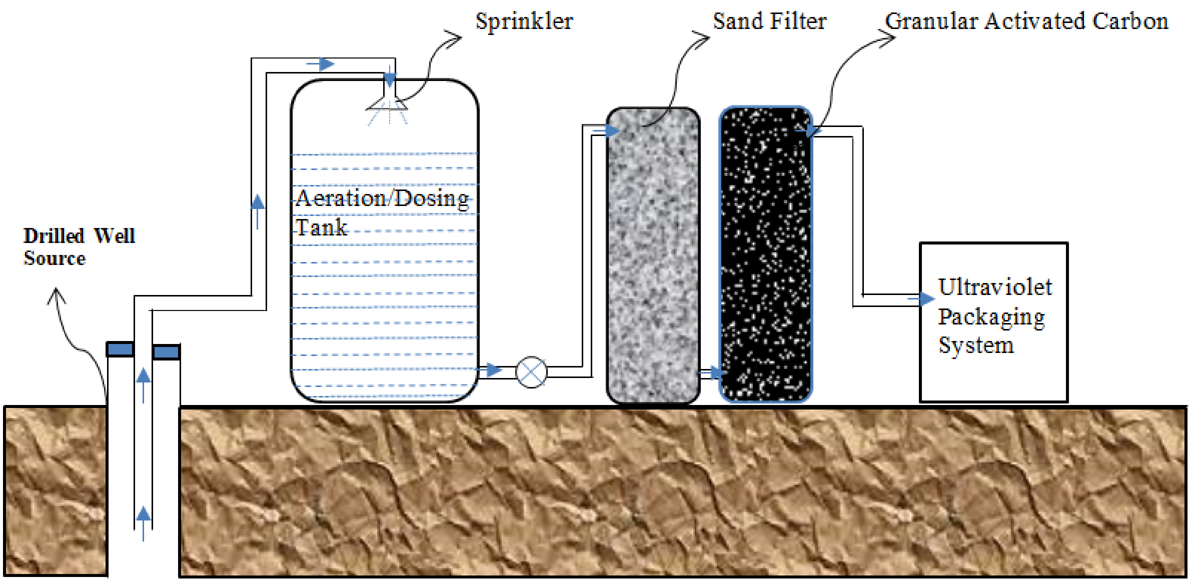

Figure 1. Schematic diagram showing the sequence of table water production in the study area

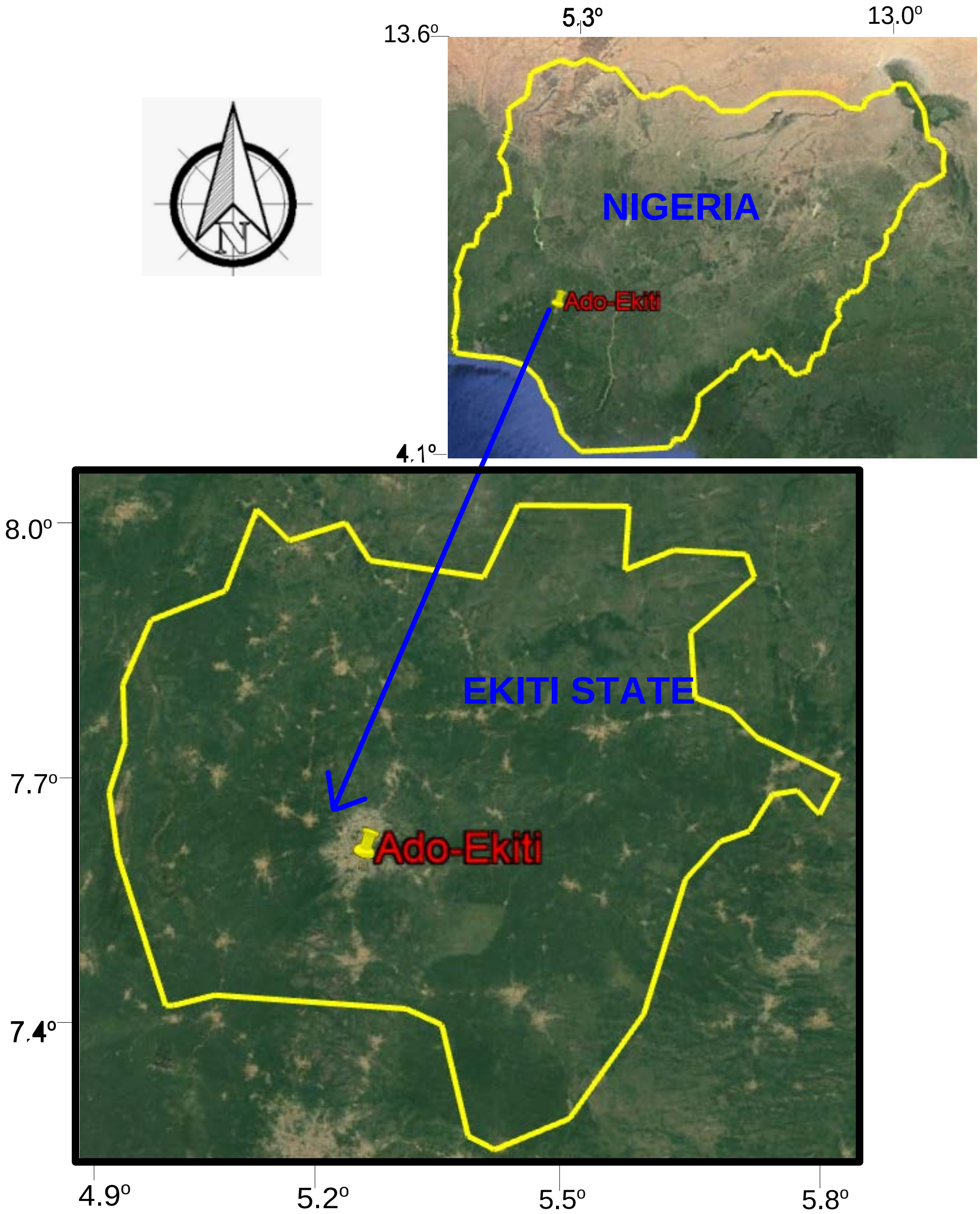

Figure 2. Map of study area (Inset: Map of Nigeria)

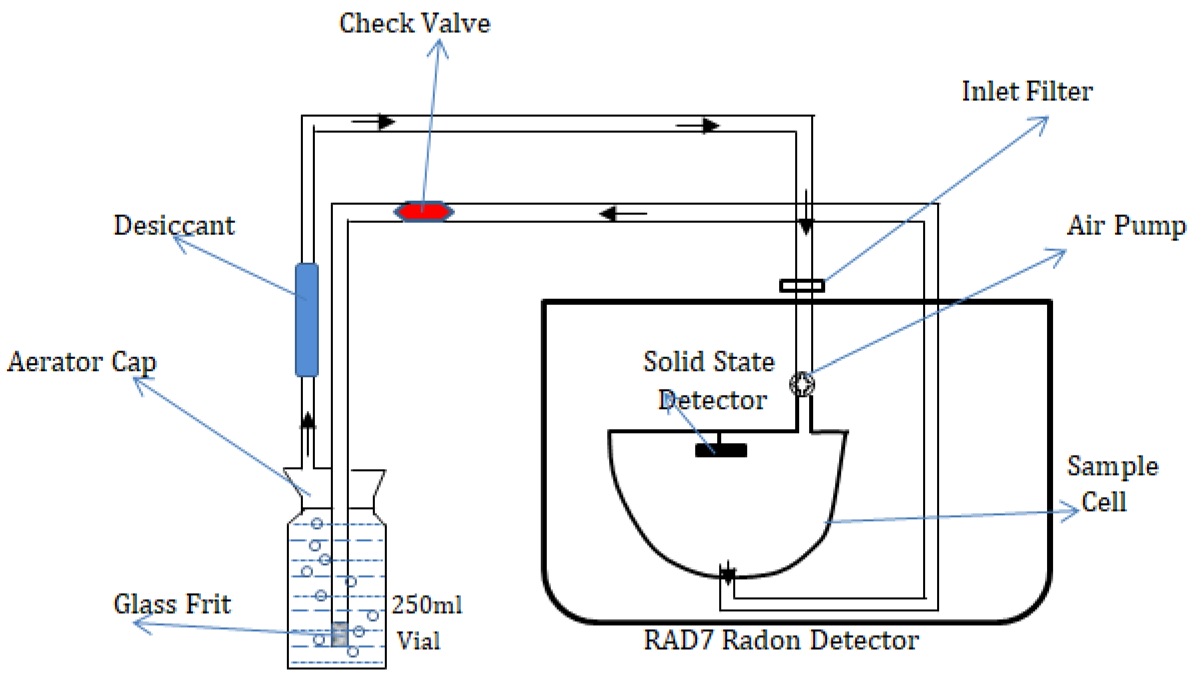

Figure 3. Schematic diagram showing set-up for radon-in-water measurement

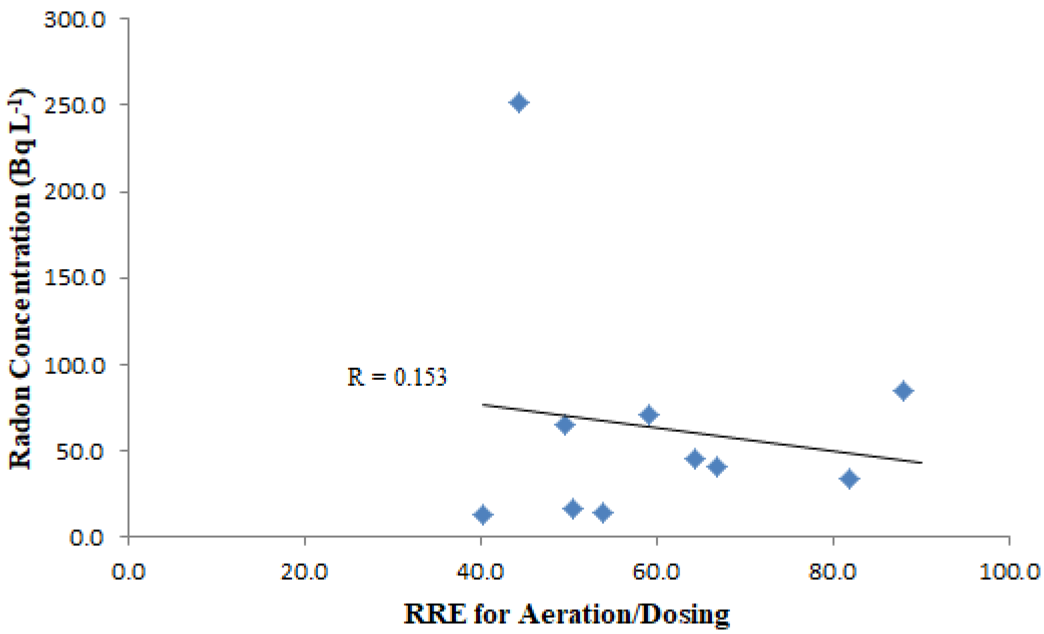

Figure 4. Scatter Plot showing relationship between radon concentration and radon removal efficiency

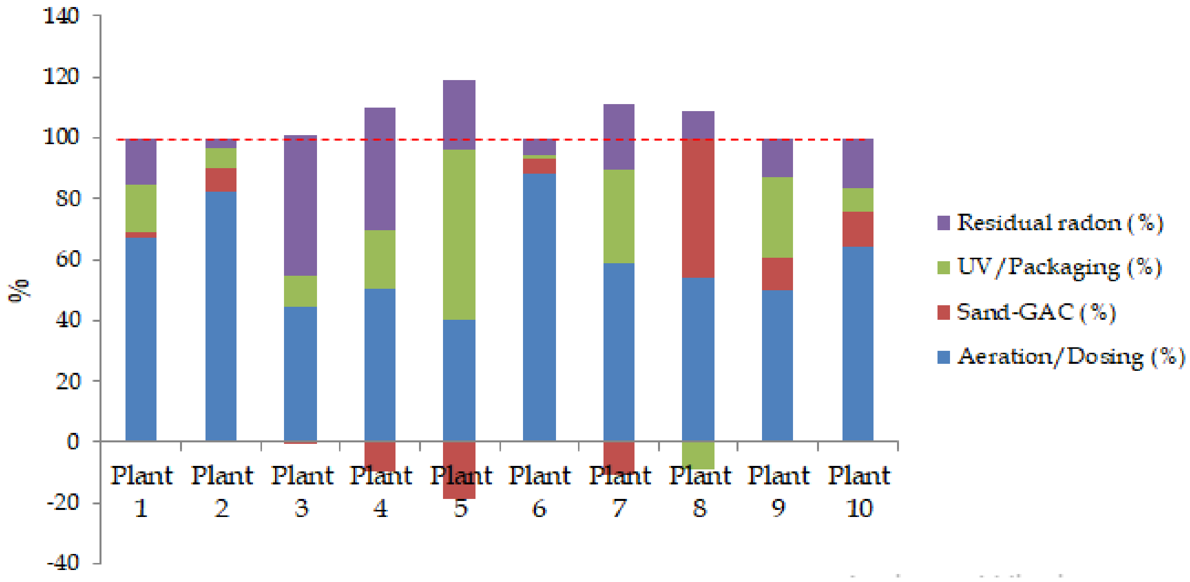

Figure 5. Histogram showing radon removal efficiency of the water processing phases in the ten water plants

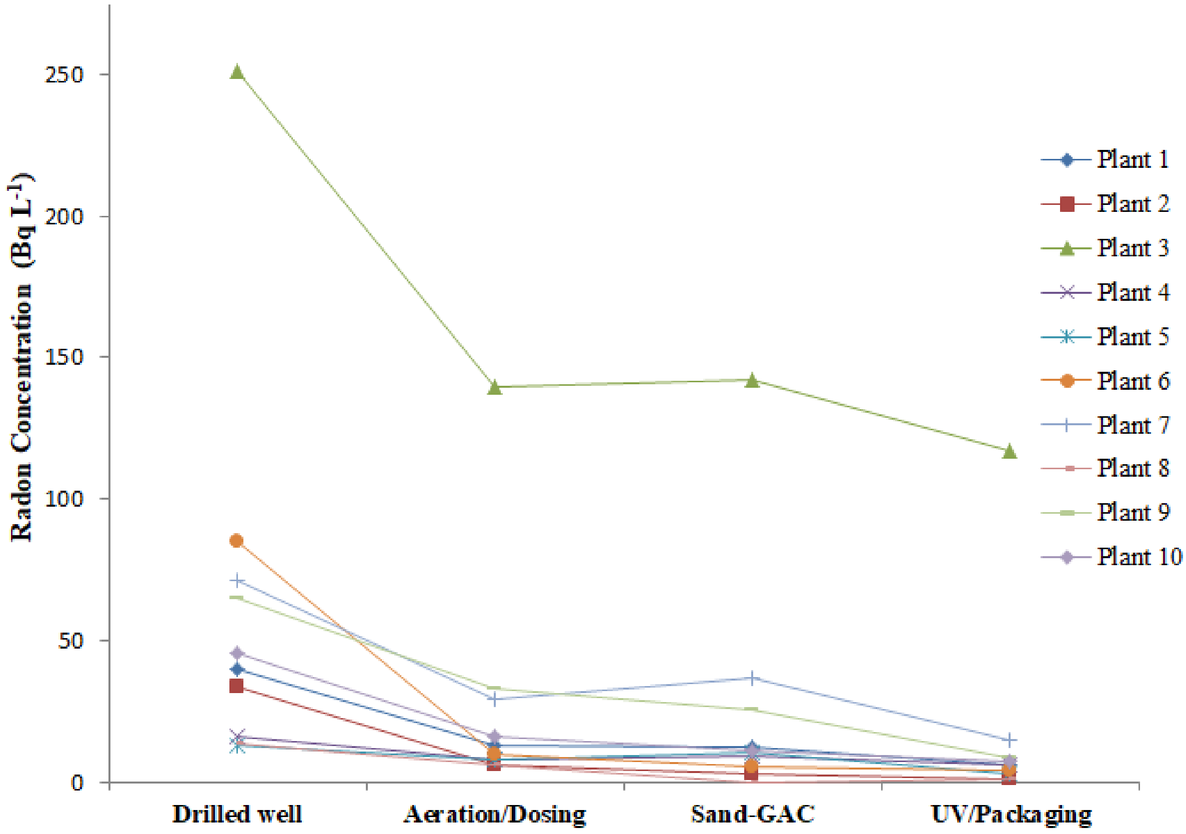

Figure 6. Trend of radon concentration reduction across water treatment phases