



Radiological Hazards Assessment of Produced Water from Some Oil and Gas Flow Stations in Delta State, Nigeria

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Authors' contributions

This work was carried out in collaboration between authors EEO and OAG. Author EEO designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors EEO and OAG managed the analyses and the literature searches of the study. Both authors read and approved the final manuscript.

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ABSTRACT

The radiological hazards of produced water collected from seven oil and gas flow stations (fields onshore) of Delta State, Nigeria were estimated using thallium activated 3"x3" Sodium iodide [NaI(Tl)] detector. Twenty- one samples of produced water from seven flow stations waste pit were collected within the oil fields using standard methods. From the radionuclide results, the radiological hazards were computed. Comparing the computed average values for representative gamma index, annual effective dose equivalent (Outdoor) annual effective dose equivalent (indoor) which are 0.104 mSv/y, 0.0229 mSv/y and 0.03276 mSv/y respectively with the standard of 1.0 mSv/y, it was observed that the results are all below the recommended limit. The computed absorbed dose rate ranged from 4.570 nGyh⁻¹ to 10.088 nGyh⁻¹ with an average value of 6.68 nGyh⁻¹ was found to be higher than the acceptable standard of 1.5 mSv⁻¹. The results obtained in this study provides a baseline map of radiological hazard levels of produced water that may likely

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be discharge in to the environment and may be used as reference information to assess any changes in this studied area. This subject is important in environmental radiological protection since produced water are widely been discharged in the environment during oil and gas productions.

Keywords: Radionuclides; concentration; produced water; radioactivity; health hazard.

1. INTRODUCTION

The knowledge of radionuclides distribution and radiation levels in the environment is important for assessing the effect of radiation exposure due to both terrestrial and extraterrestrial sources. Terrestrial radiation is due to radioactive nuclides present in varying amounts in rock, building materials, water, soils and atmosphere. Some of these radionuclides from these sources are transferred to man through food chain or inhalations [1,2]. Naturally occurring rocks are generally permeated with fluids such as water, oil, or gas. Thus, reservoir rocks normally contain both petroleum hydrocarbons (liquid and gas) and water. The less dense hydrocarbons migrated to trap locations, displacing some of the water from the formation in becoming hydrocarbon reservoirs. This water may be formation water trapped within the hydrocarbon reservoir for millions of years, or a mixture of seawater and formation water when seawater has been injected in order to maintain pressure in the reservoir during oil and gas production [3]. Produced water usually is the wastewater generated in the largest volume during the production of oil and gas wells that are brought to the surface of the earth. Produced water contains a wide variety of chemicals and naturally occurring radioactive elements such as uranium, thorium and potassium (^{238}U , ^{232}Th and ^{40}K) that have been dissolved or dispersed from the hydrocarbon and the geologic reservoir containing the formation water [4]. Produced water generated may be treated and reinjected, disposed or discharged into the environment depending on local environmental regulations and available technology.

Discharge of produced water into the environment during exploration and production of oil and gas activities have been a subject of major concern with respect to environmental pollution including possible radiation exposure to the Niger Delta region of Nigeria. This produced water contains some level of naturally occurring radionuclides [5], and the water disposed of by the oil and gas operating

companies into nearby rivers water, lakes and through underground injection where the TENORMs find their way into underground aquifers thus contaminates water bodies, soil, vegetations and related organisms.

Despite treatment before discharge to satisfy regulatory limitations on oil content, produced water contains a certain amount of Naturally Occurring Radioactive Materials (NORM) such as ^{226}Ra and ^{228}Ra . NORM are difficult to remove from produced water, which make the assessment of their effects on human health important to the oil and gas producing industries [6]. The long term exposure to radionuclide such as radium and thorium through inhalation and ingestion has severe health effects such as chronic lung diseases, acute leucopenia, anemia and necrosis of the mouth [7]. Radium causes bone weakening, cranial and nasal tumors. Other diseases caused by radioactivity exposure include lung cancer, pancreas, hepatic, bone, skin, kidney cancers, cataracts, sterility, atrophy of the kidney and leukemia [8].

Knowledge of natural radioactivity present in water, produced water, soil and river sediments enables one to assess any possible radiological hazard to mankind by the uses of such materials. Hence, the objective of this study is to evaluate the radiological hazards from produced water that is being discharged in to the Niger Delta environment by the oil and gas companies operating in the study area.

2. MATERIALS AND METHODS

2.1 Description of Study Area

This study was carried out within Delta state onshore Niger delta of Nigeria. The study area lies within latitude $5^{\circ}18'\text{N}$ and $5^{\circ}68'\text{N}$ and longitude $5^{\circ}33'\text{E}$ and $6^{\circ}40'\text{E}$ South-West of Niger delta region of Nigeria. The geology of the study area has been reported earlier [9]. The lithological log correlation showed that the topsoil layer, which is



Fig. 1. Map of the study area

composed of plastic clay, has a thickness ranging from 30 ft-35 ft, which is capable of protecting the underlain aquifer unit from being contaminated by surface toxic discharge. A silt sand/sandy layer directly underlies this, which form the aquifer unit of the study area. The groundwater table ranges between 8 ft-10 ft. The natural water system of the area depending on the location and depth and belong to two extremes of fresh and salt water with an intermediary represented by the blackish water [10]. Thus there is the likelihood of overflowing of this produced water from their waste pits, leaching and seepage into surface and underground waters outside the illegal disposal of these untreated wastes into the environment.

2.2 Sample Collection and Preparation

Twenty one produced water samples were collected from the seven selected flow stations which includes: Oghini (PWO), Oweh (PWO), Ewreni (PWE), Afiesere (PWA), Eriemu (PWE), Otorogun (PWO) and Olomoro- Oleh (PWO)

At the point of sampling, 2-liter plastic container was used for the collection of the samples with about 1% air space of the container left for thermal expansion. Sample containers were raised three times with sampled produced water to minimize contamination from the original

content of sample container. Produced water samples collected from the flow stations were immediately acidified with 10ml±1ml of 11 M HCl per liter of samples collected to avoid absorption of radioactivity on the walls of the containers [11,12]. The samples containers were then tightly covered with container cover and kept in the laboratory until analysis. Thereafter, samples collected were sent to the Centre for Energy Research and Development (CERD), Obafemi Awolowo University Ile-Ife, Nigeria for analysis.

At the radioactivity measurement laboratory centre for energy research and development (CERD), Obafemi Amolowo University Ile-ife Nigeria beaker of one (1) liter volume capacity washed, rinsed with a dilute sulfuric acid and dried to avoid contamination with the sample containers were filled with known volume of the produced water samples were firmly sealed for four weeks to ensure that loss of radon does not occur and ensuring secular equilibrium to be established before the gamma-ray analysis.

2.3 Sample Analysis

The samples were analyzed at CERD Gamma Ray Spectrometry Laboratory, Obafemi Amolowo University Ile-ife, using a thallium activated 3"x3" Sodium iodide [NaI(Tl)] detector connected to ORTEC 456 amplifier. The

detector, enclosed in a 100mm thick lead shield, was connected to a computer program SAMPO 90 window that matched gamma energies to a library of possible isotopes. Since the accuracy of the quantitative measurements is depended on the calibration of the spectrometry system and adequate energy. Background measurement and efficiency calibration of the system was made possible using Cs-137 and Co-60 standard sources from IAEA, Vienna. Spectrum were accumulated for background for 29000s at 900volts to produce strong peaks at gamma emitting energies of 1460keV for ^{40}K ; 609keV of ^{214}Bi and 911keV of ^{228}Ac , which were used to estimate the concentration of ^{238}U and ^{232}Th , respectively. The energy resolution of the detector using Cs-137 and Co-60 standards is 39.5% and 22.2% respectively while the activity of the standards at the time of calibration is 25.37KBq for Cs-137 and 4.84KBq for Co- 60. The background spectra, measured under the same conditions for both the standard and sample measurements, were used to correct the calculated sample activities concentration in accordance with [13]. The activity concentration (C) in Bq l^{-1} of the radionuclides in the samples was calculated after subtracting decay correction using the expression:

$$C_s (\text{Bq/kg}) = \frac{C_a}{\sum \epsilon_{E\gamma} \times M_s \times t_c \times P_\gamma} \quad (1)$$

Where C_s = Sample concentration, $N_{E\gamma}$ = net peak area of a peak at energy, $\epsilon_{E\gamma}$ = Efficiency of the detector for a γ -energy of interest, M_s =Sample volume, t_c = total counting time, P_γ =Emission probability of radionuclide of interest.

2.4 Radiation Hazard Indices Calculation

2.4.1 Estimation of gamma radiation absorbed doss rate (D)

UNSCEAR, [14] has given the dose conversion factors for converting the activity concentrations of ^{238}U , ^{232}Th and ^{40}K into doses (nGy.h^{-1} per Bq l^{-1}) as 0.462, 0.621 and 0.0417, respectively.

The gamma radiation population doses of those living in the area are given as:

$$D=0.462A_u + 0.621 A_{Th} + 0.0417A_k \quad (2)$$

where D is the dose rate in nGy h^{-1} and A_u , A_{Th} and A_k are the concentrations of uranium, thorium and potassium, respectively.

2.4.2 Representative level index (I_{yr})

In order to examine whether the samples meets these limits of dose criteria, Another radiation hazard index, the representative level index, I_{yr} was used to estimate the level of γ - radiation hazard associated with the natural radionuclide's in specific investigated samples. This is defined as [15].

$$I_{yr} = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \quad (3)$$

where A_{Ra} , A_{Th} and A_k are the concentrations of ^{226}Ra , ^{232}Th and ^{40}K , respectively in Bq l^{-1}

2.4.3 Annual effective dose rate (outdoor)

The annual effective dose (mSv yr^{-1}) was calculated using the formula [14].

$$\text{Annual effective dose rate (mSv yr}^{-1}) = D(\eta \text{Gyrh}^{-1}) \times 8760 \text{hr}^{-1} \times 0.7 \times (10^3 \text{mSv}/10^9) \eta \text{Gy} \times 0.2$$

$$E_{ff} \text{ Dose} = D1.2264 \times 10^{-3} \quad (4)$$

where D is effective dose rate, [14] has recommended 0.7 Sv/Gy as the conversion coefficient from the absorbed dose in air to effective dose and 0.2 as the value for the outdoor occupancy factor.

2.4.4 The Annual effective dose rate (indoor)

The Annual effective dose rate (indoor) was calculated using the expression in the equation 5.

$$\text{Effective dose(mSv yr}^{-1})=D(\eta \text{Gyrh}^{-1}) \times 8760 \text{hr}^{-1} \times 0.7 \times (10^3 \text{mSv}/10^9) \eta \text{Gy} \times 0.8 \times 10^{-6} \quad (5)$$

The United Nation Scientific Committee on the effect of Atomic Radiation [14] has recommended 0.7 Sv/Gy as the conversion coefficient from absorbed dose in air to effective dose and 0.8 (19/24) as the value for the indoor occupancy factor.

2.4.5 The indoor Gamma dose rate

The indoor gamma rate (D_{in}) due to the emissions of gamma – ray from the

radionuclides (^{226}Ra , ^{232}Th and ^{40}K) in the produced water samples was calculated using equation 6 for a standard room dimension [14].

$$D_{in} = 0.92A_{Ra} + 1.1A_{Th} + 0.080A_K \quad (6)$$

where A_{Ra} , A_{Th} and A_K are the activity concentration of ^{226}Ra , ^{232}Th and ^{40}K respectively.

2.4.6 Excess Lifetime cancer risk (ELCR)

Excess lifetime cancer risk (ELCR) can be defined as the excess probability of developing cancer at a lifetime due to exposure level of human to radiation. The significance of exposure from natural radioactivity in produced water and the potential risk for causing health problem, especially cancer, have not received the desired attention in Niger Delta. Therefore there is need to determine the excess cancer risk over a lifetime (ELCR). Excess lifetime cancer risk is calculated using the equation below.

$$\text{ELCR} = \text{AEDR} \times \text{DL} \times \text{RF} \quad (7)$$

where AEDR, DL and RF are respectively the annual effective dose equivalent, duration of life (70 years) and risk factor (Sv-1), fatal cancer risk per sievert. For stochastic effects, ICRP 60 uses values of 0.05 for the public [8,16].

3. RESULTS AND DISCUSSION

Table 1 present the mean values of the three (^{40}K , ^{226}Ra and ^{232}Th) natural radionuclide isotopes present, excess lifetime cancer risk and hazard indices in the produced water analyzed. The average activity concentration of ^{40}K , ^{226}Ra and ^{228}Ra in produced water from the studied area are 48.78 ± 13.67 Bq/l, 6.04 ± 2.48 Bq/l and 5.18 ± 2.14 Bq/l respectively.

The representative gamma index mean values range from 0.070 mSvy $^{-1}$ at Otorogun gas plant to 0.156 mSvy $^{-1}$ at Olomoro flow station with average value of 0.104 mSvy $^{-1}$. The obtained results when compared with standard of 1.0 mSvy $^{-1}$ [14] as shown in Fig. 2 revealed that the values are less than the world permissible value of unity [17]. This indicates that the values will not lead to respiratory diseases such as asthma and

cancer and external diseases such as erythema, skin cancer and cataracts.

The absorbed dose rate in Olomoro flow station has the highest value 10.088 mSvy $^{-1}$ while Otorogun gas plant has the lowest value 4.570 mSvy $^{-1}$ with an average value of 6.679 mSvy $^{-1}$. The obtained results when compared with standard of 1.5 mSvy $^{-1}$ [14] as shown in Fig. 3 revealed that the average values of absorbed dose rate in all the produced water samples are higher than world standard limit. This could again be attributed to geology formation of the study area and the various radionuclide induce activities of the exploration companies.

The calculated values of excess lifetime cancer risk have its highest at Olomoro flow station as 0.0434 mSvy $^{-1}$ and lowest at Otorogun gas plant as 0.0196 mSvy $^{-1}$ with average value of 0.0288 mSvy $^{-1}$. Comparing obtained values of produced water for all the flow stations with the world average standard (0.29×10^{-3}) [8,16] as shown in Fig. 4, it was observed that the obtained values are higher than the world allowable average. This could be attributed to the high activity concentrations of ^{40}K , ^{226}Ra and ^{228}Ra that are present in the measured produced water and also could be attributed to the activities of oil and gas companies operating in the area. This implies that the chances of having cancer by staff of oil and gas companies working in flow station and the public are significant. These high concentrations pose significant health threat to both human system and the environment.

The Annual effective dose rate AEDR (outdoor) and Annual effective dose rate AEDR (indoor) are calculated using equation 4 and 5 respectively. The obtained average results of AEDR (outdoor) and AEDR (indoor) are 0.0229 mSvy $^{-1}$ and 0.3276 mSvy $^{-1}$ respectively. When compared with [18,19] standard of allowable limit of 1.0 mSvy $^{-1}$ as shown in Fig. 5 and 6 respectively, it was observed that the obtained results are below the internationally accepted limit. The obtained values are also less than unity. Hence, from a radiological health standpoint, the obtained values of effective doses do not pose a significant threat to both human system and the environment. Hence, the produced water will do more harm than good to both man and the environment. Therefore, it must be properly managed to avoid pollution.

Table 1. Mean Specific activity concentration of ²²⁶Ra, ²²⁸Ra and ⁴⁰K and the excess lifetime cancer risk and health hazard indices of the sampled produced water

S/N	CODE	K-40	U-238 (Ra-226)	Th-232 (Ra-228)	(I _γ) Bq.Kg-1	ADR (D) nGyh ⁻¹	(ELCR)	AEDR (Outdoor)	AEDR (Indoor)
1	PWOG	14.23±6.53	3.87±1.63	3.99±1.64	0.079	4.858	0.0209	0.0060	0.2383
2	PWOW	34.14±9.40	5.58±2.26	9.31±3.70	0.122	7.854	0.0337	0.0100	0.3853
3	PWEV	84.84±21.34	3.50±1.18	5.14±2.24	0.084	5.080	0.0220	0.0062	0.2492
4	PWAF	42.58±11.58	8.20±3.68	5.22±2.11	0.134	8.809	0.0377	0.0108	0.4321
5	PWOL	82.14±21.35	10.81±4.24	5.37±2.33	0.156	10.088	0.0434	0.0124	0.4949
6	PWER	52.94±15.51	5.29±2.14	3.85±1.25	0.086	5.494	0.0240	0.0068	0.2695
7	PWOT	30.57±10.01	5.01±2.20	3.40±1.68	0.070	4.570	0.0196	0.0056	0.2242
Average		48.78±13.67	6.04±2.48	5.18±2.14	0.104	6.679	0.0288	0.0229	0.3276

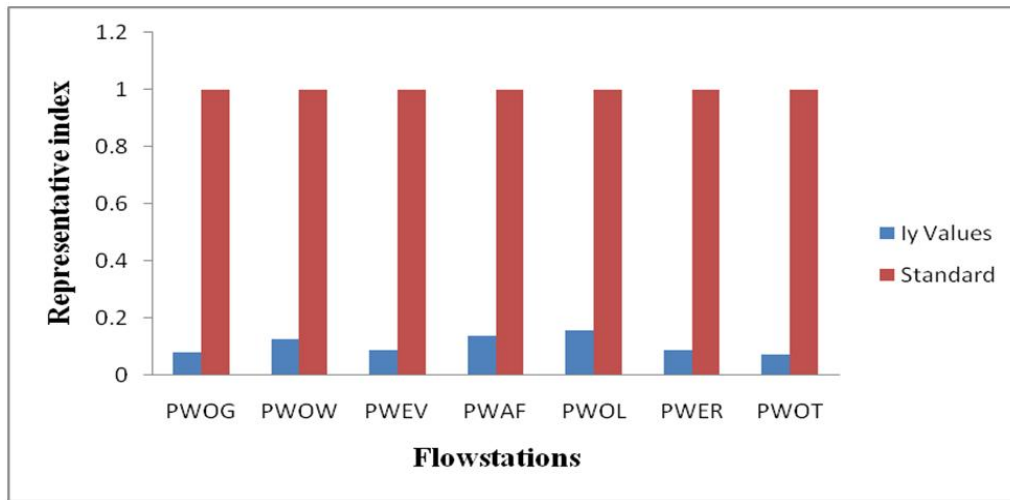


Fig. 2. Comparison of gamma representative index of the studied samples (Bq.Kg-1) with standard in studied flowstations

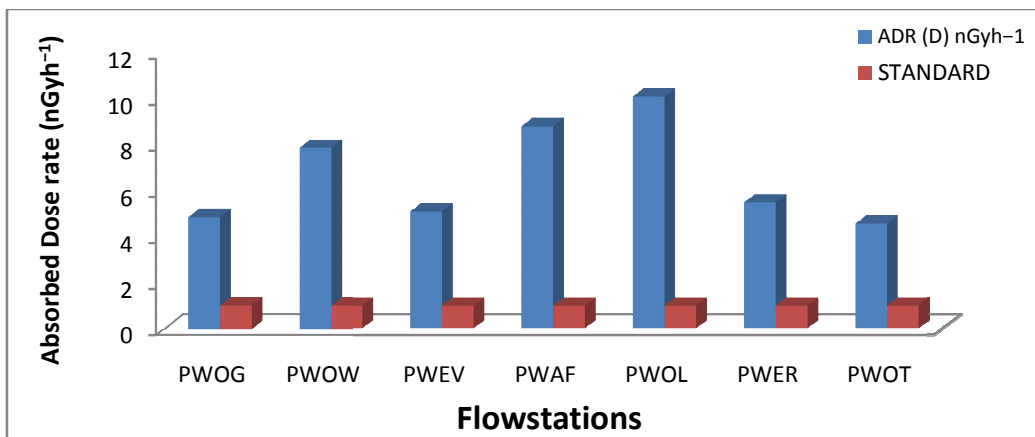


Fig. 3. Comparison of absorbed dose rate (nGyh⁻¹) with standard in studied flowstations

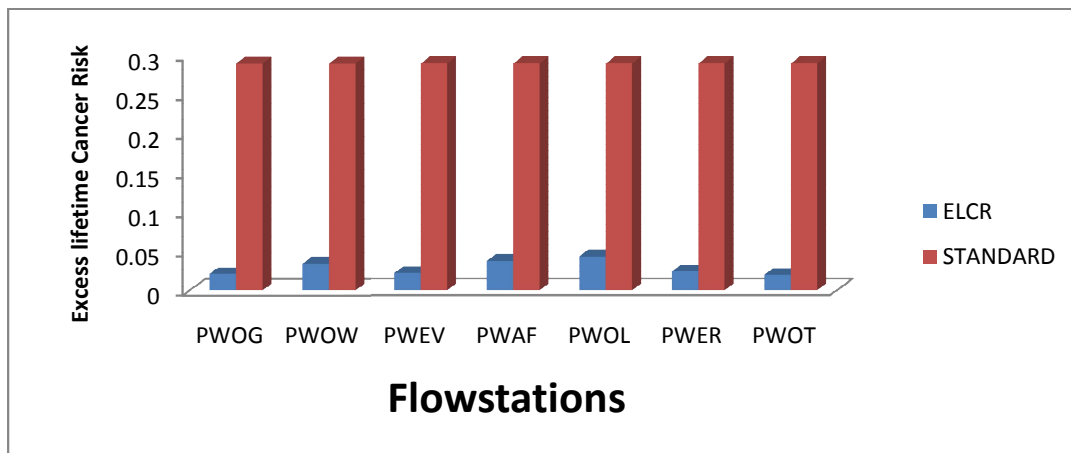


Fig. 4. Comparison of excess lifetime cancer risk with standard in studied flowstations

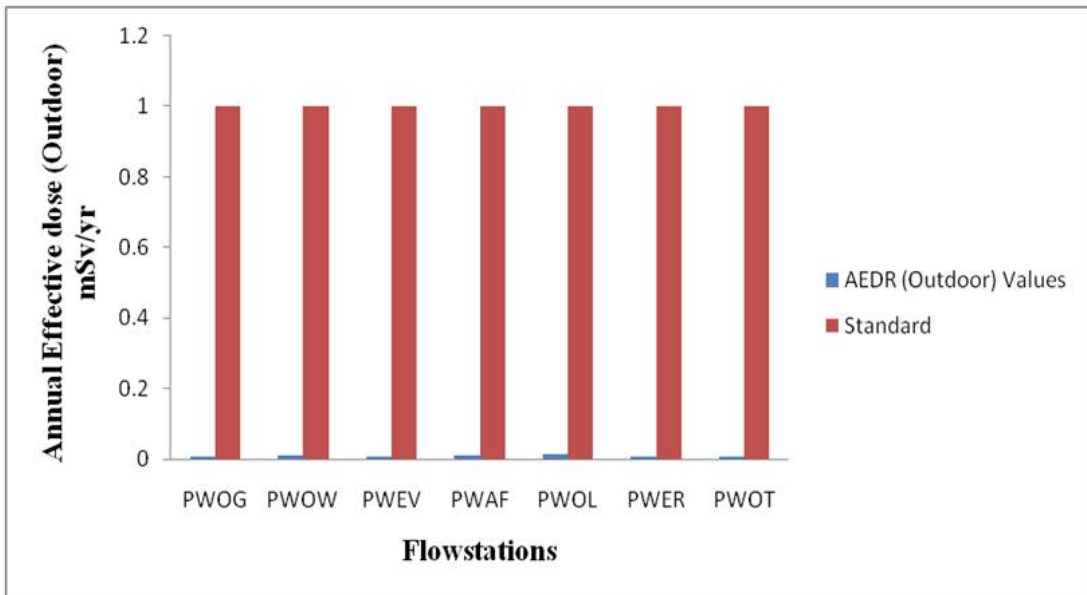


Fig. 5. Comparison of annual effective dose (Outdoor) (mSv^{-1}) with standard in studied flowstations

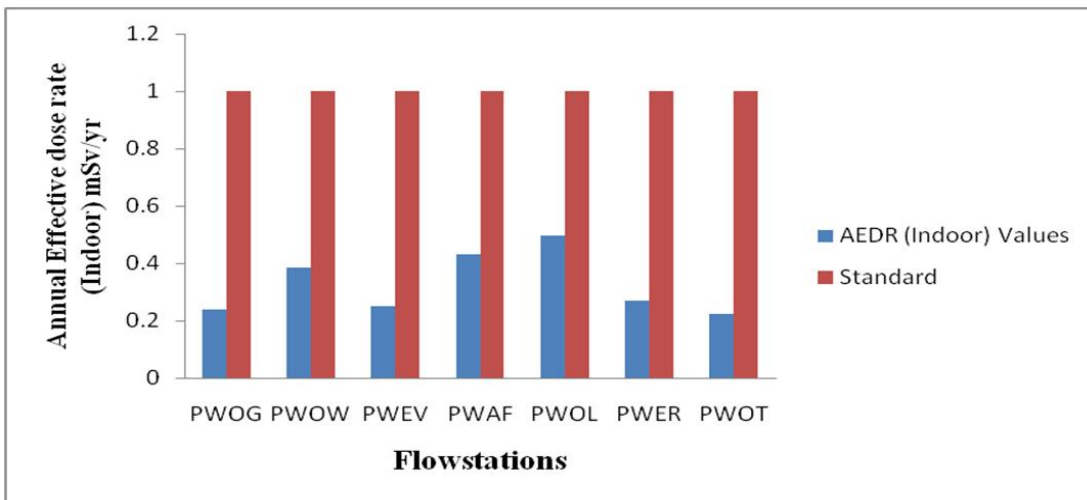


Fig. 6. Comparison of annual effective dose (Indoor) (mSv^{-1}) with standard in studied flowstations

4. CONCLUSION

This work is aimed at computing radiological hazard and excess lifetime cancer risk produced water in oil and gas flow stations in Delta State, Nigeria. The results of the evaluation indicated that the average values of excess lifetime cancer risk and absorbed dose rate are higher than standard values and this can be attributed to the impact of oil and gas exploration and exploitation activities on the

environment. This could result from age-long contamination of aquifers with formation water and effluent discharge at various magnitudes in the oil fields. Representative gamma index, Annual effective dose rate AEDR (outdoor) and Annual effective dose rate AEDR (indoor) are below the acceptable limit (Safety Limit). Though immediate health implication for the oil and gas workers and public users may not be observed at the present level, but long-term health effects are probably

in the nearest future. We, therefore, recommended as follows:

- The oil and gas operating companies in these areas should put in place means of reducing their radionuclide input during oil and gas production.
- Health insurance policies should be acquired for employees and contract staff working within the flow stations, to take care of their long-term health problems.
- All oil and gas installations should meet all known local and international standard.
- There should be a regular monitoring of radiation/radiological levels in these environments.
- Government agencies should enforce all the existing radiation/radiological legislation on the oil and gas companies.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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