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Chemical and Radiological Risk Assessment of Uranium in Surface and Groundwater in Ogoni Land, Rivers State, Nigeria

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Abstract: The chemical and radiological risks associated with uranium in the well, tap/borehole and river/creek waters within three communities that host Ogoniland oilfields was here assessed and measured with gamma ray spectroscopy. The results obtained were used to evaluate chemical and radiological risk over lifetime ingestion by the inhabitants in the area. The activity concentrations of uranium in the water supply sources were found to range from 2.42±0.45 to 12.77±1.12Bq/l. The uranium mass concentration was found to range from 97.52±18.13 to 514.71±45.14µg/l. These uranium mass concentration values of the three water supply sources was found to be over 5 times higher than the recommended international permissible limits. The radiological risks for cancer mortality and morbidity risks were found to be lower than permissible standard which respectively ranged from $9.08x10^{-5}$ to $4.79x10^{-4}$ and $1.39x10^{-4}$ to $7.34x10^{-4}$. However, the chemical toxicity which was estimated using lifetime average daily dose (LADD) and hazard quotient (HQ) was found to vary respectively from 2.67 to 14.09µg/kg/day and 4.45 to 23.48. The LADD values exceeded the acceptable reference dose level of 0.6µg/kg/day and the international threshold daily intake value of 1.0µg/kg/day. The HQ was greater than unity implying significant potential risk of uranium in water due to chemical toxicity. Human risk arising from ingestion of uranium in water was therefore attributed to the chemical toxicity of uranium as heavy metal rather than radiological risks. Ion-exchange pre-treatment and reverse osmosis treatment technique should therefore be adopted to the three water supply sources in order to reduce and remove uranium as heavy metal before consumption.

Keywords: Cancer Morbidity, Cancer Mortality, Chemical Toxicity Risk, Hazard Quotient (HQ), Lifetime Average Daily Dose (LADD), Radiological Risk

Introduction:

Radionuclides are unstable forms of chemical elements that radioactively decay spontaneously from radioactive materials, causing emission of nuclear radiation. Crude oil and gas contains naturally occurring radioactive materials (NORM) or radionuclides. The release of ionizing radiation such as ^{238}U, ^{232}Th and ^{40}K during the decay of radionuclides may pose risks to the environment and human health, in particular increasing cancer risk. Besides its radiological health risk, uranium-238 is also chemically poisonous. Natural uranium is therefore classified as both a radiological and chemo-toxicological agent and it is the only radioactive substance for which chemical toxicity is the limiting factor in risk assessment (Hakonson-Hayes *et. al*., 2002).

Contamination of water by uranium from NORM generating industries is a major environmental problem in terrestrial and water ecosystems (Kumar *et. al*, 2012). Amakom and Jibiril (2010) opined that the presence of uranium in water is of serious environmental concern due to its ability to bond with oxygen to form the uranyl ion, or uranium oxide, which is soluble in groundwater under aerobic conditions. WHO (2011) also established that high concentration of uranium greater than 30µg/l in drinking water may present harmful biological effects in humans. The fact that uranium is predominantly an alpha-emitting radionuclides, utilization of water from these sources of water supplies has raised concerns of potential radiological and chemo-toxicological risks to human consumers as there have been claims and counter claims of cancer, leukemia, eye cataracts, kidney dysfunction, potential DNA damage and other health related radiation induced sicknesses attributed to oil and gas exploration/exploitation activities, gas flaring and oil spillages caused by ruptured pipelines or pipeline vandalism, by the host communities of the study area. For over four decades, Ogoniland has been a sink of pollution associated with oil exploitation and exploration. Oil spillage and gas flaring from oil production have continuously increased the pollution matrix of the area, leaving a severely degraded environment. It is true that the United Nations Environment Programme (UNEP) in 2010 undertook an assessment of oil pollution in Ogoniland, yet oil spillage, gas flaring, oil bunkering and artisanal refining of crude oil, and disposal of radioactive wastes by oil and non-oil companies are recorded in the area in recent time thereby adding to the pollution in the area. There was however, no detailed report on the uranium content of the polluted waters in the UNEP findings. Apart from the direct impacts of oil spills on water bodies, emissions from gas flaring are dispersed into communities around the oil facilities

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where they settle on communities' sources of water (hand-dug wells, creeks, rivers). Jabbar *et. al.* (2010) asserts that such water could be radioactive and chemically toxic to the extent that could be harmful when ingested by people. Studies on the radiological and chemical toxicity of uranium in water have been carried out by many authors. Amakom and Jibiri (2010) showed that the radiological risks of cancer mortality and morbidity for uranium in borehole and well water for the inhabitants of Odeda community in Ogun State, Nigeria were low while the chemical toxicity risks due to uranium consumption in the water sources were higher than the recommended safe level by international organizations. Agbalagba and Osakwe (2013) assessed the radiological and chemical risk of Uranium-238 in well, borehole and river waters in rural communities in OML 30 and 26 oilfields in Delta State. The study showed that human risk due to uranium content in water supplies that will result from ingestion may be attributed to chemical toxicity of uranium as heavy metals rather than radiological risk. The rural communities of the study area depend on waters from hand-dug wells, tap/borehole, and river/creek as sources of drinking water. With the limited literature on the effect of uranium consumption in water supply sources of crude oil polluted water in Ogoniland, this study therefore aim at assessing the chemical and radiological health risks of uranium consumption from surface and groundwater by the inhabitants of the area. This is important in view of the water pollution by oil spills from oil pipelines at Bodo, Bunu-Tai, Gio (Giokoo) and Eleme communities as reported by Scoop Newspaper (2013), Daily Post (2013), Daily Times (2016), Vanguard (2016) and Nigerian Nation (2016) as well as the on-going illegal refining of crude oil and bunkering, gas flaring and effluents discharge into water bodies by companies.

The Study Area:

Ogoniland is in the south-eastern part of Nigeria in the Niger Delta basin. It is situated approximately within Latitudes 04°45´44.1´´N and 04°45´48.7´´N of the equator and Longitudes 007°06´26.1´´E and 007°06´33.0´´E of the Greenwich Meridian (Figure 1). The general topography is relatively flat lying and consists of terrestrial and marine environment. Due to the crude oil spills that polluted the land, the terrestrial environment has patchy regenerating vegetation which consisted mostly of scanty and secondary type residual grasses.

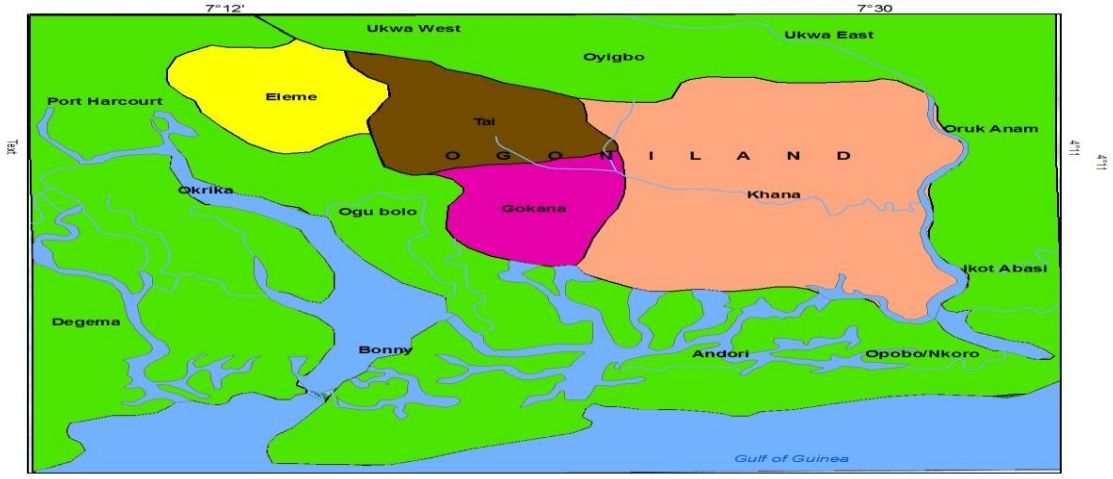

Figure 1: Map of Niger Delta Showing Ogoniland, the Study Area
Source: UNEP (2011)

Regional Geology:

Ogoniland falls within the Niger Delta region which is made up of thick clastic sedimentary sequence with age ranging from Eocene to Recent (Tattam, 1943). It sits astride the Niger flood plains, which overlies the Benin formation that is often called the coastal plain sand (Tattam, 1943). This formation consists predominantly of coarse grained sandy soils with few shale intercalations. The unconsolidated, highly porous sands of the Benin formation is a fresh water bearing sands zone (Amajor, 1991), and all aquifers in this region are located within this lithiostratigraphic unit. The ground characteristics of Ogoniland are consistent with deltaic environments where erosion and deposition of sediments constantly shift the course of channels, tributaries and creeks (Amadi *et. al.*, 1989). Two groundwater regimes exist in the area viz the salt water and fresh water. Probably due to salt water intrusion, a depth interval of 26.7 to 32 metres underground contains salt water while the portion below this level contains fresh water (Offordile, 1971 and 2002).

Materials and Methods

Sample Collection and Preparation:

Eight water samples were collected from 3 communities that host oilfields with history of crude oil spill, gas flaring and oil bunkering activities (table

1 and figure 2). The simple random sampling method was adopted. Water samples were collected from rivers, taps/borehole and hand-dug wells that serve as community water supply sources. Selection of sample locations was based on proximity of water source to oilfield, spill site and industrial area. Three water samples used as control were collected from river, tap/borehole and hand-dug well from non-oil bearing communities (with no history of oil production and oil spillages) located at 55km away from Ogoniland. Well water samples were collected manually in the early hours of the day while river/creek water samples were collected near the middle of the water body in the early hours using wading gear and a 2-metre extendable metal grab. Samples were collected against the flow of water by submerging a water container to a depth of 10-20 cm under the surface. Tap/borehole water samples were collected with treated containers at laminar flow rate after first turn on at full capacity for 5 minutes to purge the plumbing system of any water that might contaminate the sample to reduce radon loss.

To minimize contamination, the collection of the water samples from the 3 water supply sources was done each with 2-litre container. The 2-litres containers were first rinsed three times with water and acid washed with 20ml±1ml Conc. HNO_3 before using it to collect the samples so as to avoid adsorption of the radionuclides on the walls of the containers. For each sample, about 1% airspace was left for thermal expansion before it was tightly sealed and taken to the laboratory to store for 30 days to ensure that no radon loss occurs, and to reach a state of circular equilibrium between radium isotopes and their daughters. Samples` were then transferred into a 1-litre Marinelli beaker after filtration to remove all solid particles in the water. It was further processed through evaporation until 0.5 litres remained in the beaker and was then stored in a desiccator to allow it cool and to prevent it from absorbing moisture
.

Table 1: Locations where water samples were collected

	Oilfield	Community	River Water	Tap/borehole	Hand-dug well
1	Korokoro	Bunu-Tai	Oo-a-naana River	Bunu-Tai Tapwater	Bunu-Tai Well
2	Bodo West	Bodo	Sugi-Bodo River	Bodo Tapwater	Bodo Well
3	Onne	Eleme	Ochanai creek	Alode Tapwater	-

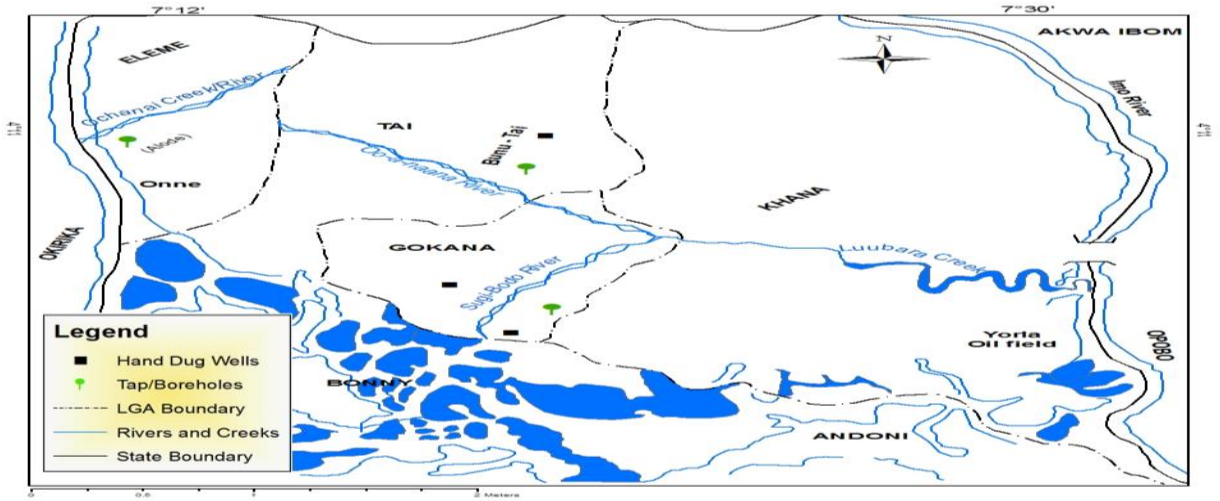

Figure 2: Map Showing Water Sampling Points in the Study Area
Source: NGSA (2010).

Sample Analysis:
The activity of the natural radiouclide of the prepared water samples were counted at the Centre for Energy Research and Training, Zaria with gamma ray spectrometer detector for 29000 seconds at 900V to produce strong peaks at gamma emmiting energies. The detector is a Thallium activated Canberra 7.6cm x 7.6cm sodium iodide [NaI(TI)] detector (model 803 series) coupled to a Canberra series 10 plus Multichannel-Analyzer through an ORTEC 456 amplifier base. The detector, enclosed in a 10 cm thick lead shielding lined with 1.5mm thick cadmium and 0.8mm thick copper, was connected to a computer program Maestro window that matched gamma energies to a library of possible isotopes. The lead shield was to reduce environmental background radiation. Using disc-type reference standard source, the gamma ray spectrometer was calibrated up to 3MeV. Background measurement and efficiency calibration of the system was made using ^{137}Cs and ^{60}Co standard sources from IAEA. The natural radionuclide tested was ^{238}U. The activities of ^{238}U

were determined from the average activities of ^{214}Pb at 352kev and ^{214}Bi at 609Kev. The background spectra measured under the same conditions for both the standard and sample measurements were used to correct the calculated sample activity concentrations. The activity concentration (Cs) of ^{238}U in the samples was calculated after subtracting decay correction by applying the following equation (Arogunjo et. al., 2005):

$$C_s = \frac{C_a}{P_\gamma (M_s/V_s)\varepsilon_\gamma t_c} (Bq/l) \quad \ldots\ldots 1$$

Where C_s is the sample concentration, C_a is the net peak area of a peak at energy, ε_γ is the efficiency of the detector for a γ-energy of interest, M_s/V_s is the sample mass/ volume of water, t_c is the total counting time, and P_γ is the abundance of the γ-line in a radionuclide.

Risk Assessment of Uranium

The activity concentrations of uranium (Bq/l) in all the water sources were converted to uranium mass concentration (µg/l) using the conversion factors (Amakom and Jibiri, 2010):

$$1Bq/l = 27.0pCi/l; 1\mu g/l = \frac{1pCi/l}{0.67} \quad \ldots. 2$$

Equation 2 can be re-written as:

1Bq/l = 27.0pCi/l = 40.2985µg/l; 1µg/l = 0.02481Bq/l …….....3

And 1pCi/l =1.4925µg/l ……..4

Using equation 3, risk assessment was made for (i) the radiological risk due to intake of uranium from the 3 water sources, and (ii) the chemical toxicity risk due to intake of uranium from the 3 sources of water in the study area.

(i) Radiological Risk Assessment

The lifetime cancer mortality and morbidity risks $R_{(mt,mb)}$ associated with ingestion of uranium in water were evaluated using the relation (Amakom and Jibiri, 2010):

$R_{(mt,mb)} = r \times A \times C \times T$ ……5

Where A is the activity concentration of uranium in water in Bq/l; C is the consumption rate of water in Lyr^{-1} which is 730 litres/yr (WHO, 2011); T is the average life expectancy which is 45.5 years in Nigeria (WHO, 2011) and r is the applicable risk coefficient which was taken from USEPA (1989), EPA (1999) and UNSCEAR (2008) as $1.13x10^{-9}$Bq for cancer mortality and $1.73x10^{-9}$Bq for cancer morbidity.

(ii) Chemical Toxicity Risk Assessment

The lifetime average daily dose (LADD) of uranium intake in water and the hazard quotient (HQ) were evaluated using the relations (Amakom and Jibiri, 2010):

$$LADD = \frac{EPC \times IR \times EF \times ED}{AT \times BW} \quad \ldots\ldots 6$$

Where IR is the water ingestion rate taken as 2 litres per day, EPC is the exposure point concentration (µg/l), EF is the exposure frequency taken as 365 days per year, ED is the total exposure duration which is taken as 45.5 years in Nigeria (WHO, 2011), AT is the average time (i.e 45.5 years x 365 days) = 16,607.50 days, BW is the weight for a standard man taken as 70kg (WHO, 2012; ICRP, 2012; UNSCEAR, 2008).

HQ is defined as the ratio of the chronic daily uranium intake to its reference dose (RfD), that is:

$$HQ = \frac{LADD}{RfD} \quad \ldots\ldots 7$$

Where RfD is defined as the daily ingestion of uranium to which the population is exposed without any appreciable risk during lifetime, and it is the reference dose or an acceptable level of chemical toxicity risk due to uranium in water whose value is 0.6µg/kg/day (Ye-shin *et. al.*, 2004; Amakom and Jibiri, 2010). This standard for uranium is used in several organizations and thereby produces a hazard quotient (Amakom and Jibiri, 2010) such that:

HQ < 1 implies that adverse effects are very unlikely to occur with the ingestion of uranium.

HQ > 1 implies that there are significant potential risks associated with the metal (uranium) due to chemical toxicity. That is the LADD of the metal (uranium) exceeded the reference dose level (RfD) (USEPA, 1993).

Discussion:

Determination of Activity and Mass Concentration of Uranium

The results of the radiological and chemical toxicity doses associated with ingestion of uranium from the communities' water supply sources in the study area are shown in table 2. The cancer mortality is the term used for the number of people who died within a population due to cancer and it is usually expressed as the number of deaths due to cancer per 100,000 populations (Balvinder *et. al.*, 2014). The cancer morbidity refers to the state of being unhealthy due to cancer within a population (Balvinder *et. al.*, 2014). From the table, the lifetime cancer mortality risk due to intake of uranium from all the 3 water sources ranged from $4.48x10^{-4}$ to $4.79x10^{-4}$ for the hand-dug well water, $1.43x10^{-4}$ to $9.08x10^{-5}$ for the tap/borehole water and $2.56x10^{-4}$ to $3.31x10^{-4}$ for the river/creek waters. These values of lifetime cancer mortality risks although higher than the control values, are lower than the international acceptable limit of $1.0x10^{-3}$ for radiological risk according to Ye-shin *et. al.* (2004). Therefore, there was no radiological health risk that would lead to having lifetime cancer mortality by consuming uranium from the 3 sources of water in the study area. The lifetime cancer morbidity risk from the 3 sources of water

supply varied from $6.86x10^{-4}$ to $7.34x10^{-4}$ for hand-dug well water, $1.39x10^{-4}$ to $3.05x10^{-4}$ for tap/borehole water and $3.91x10^{-4}$ to $5.07x10^{-4}$ for the river/creek water. Again these values of the lifetime cancer morbidity risks are higher than the control, but lower compared to the maximum permissible limit of $1.0x10^{-3}$ for radiological risk (Ye-shin *et. al.*, 2004). This confirms that the water supply sources are radiologically safe for uranium consumption. However, these values are slightly above the reported values for well water and boreholes in Ogun State, Southwestern Nigeria by Amakom and Jibiri (2010) and the reported values by Agbalagba and Osakwe (2013) for well, borehole and river waters in Delta State, Nigeria.

The chemical toxicity health risks associated with the consumption of uranium in drinking water from the 3 sources of water supplies in the study area was done by assessing uranium as a heavy metal, and then compares the uranium mass concentration (µg/l), LADD and HQ with the international acceptable limit of uranium in drinking water. HQ is defined as the ratio of the chronic daily uranium intake (ie LADD) to its reference dose (RfD) while RfD is defined as the daily ingestion of uranium to which the population is exposed without any appreciable risk during lifetime, and it is the reference dose or an acceptable level of chemical toxicity risk due to uranium in water (Ye-shin *et. al.*, 2004; Amakom and Jibiri, 2010). The study showed that the values of the mass concentration of uranium (µg/l) for the hand-dug well water, tap/borehole water and river/creek water are over 5 times higher than the control and the 20µg/l and 30µg/l permissible limit of uranium in drinking water (WHO, 2011; USEPA, 2003; Health Canada, 1999; Australia/New Zealand, 1998) for chemical toxicity risk as shown in table 2 and figure 3. This implies that the waters from the 3 sources are chemically toxic due to high uranium content. The values of LADD for hand-dug well water, tap/borehole water and river/creek water respectively exceeded the control values and it is over 2.67µg/kg/day higher than the accepted international threshold daily intake value of 1.0µg/kg/day by WHO (2011) (See figure 4). The LADD values obtained in this study are higher than those obtained by Amakom and Jibiri (2010) in borehole and well waters in Ogun State; Agbalagba and Osakwe (2013) for well, borehole and river waters in oilfields rural communities in Delta State; and Balvinder et. al. (2014) for uranium in groundwater from Western Haryana, India; Omeje *et. al.* (2017) for groundwater based drinking at Immigration Headquarters, Gosa and Federal Housing Lugbe area of Abuja. North Central Nigeria; Alausa *et. al.* (2017) for pipe borne water from some waterworks in Lagos metropolis, Nigeria; and the reported values by Abojassin and Neama (2020) for groundwater samples collected from Al-Kufa area, Iraq. The LADD values were also observed higher in the hand-dug well water samples than in the river/creek and in the tap/borehole water samples. This high value obtained for hand-dug well waters in relation to the tap/borehole and river/creek waters can be attributed to the recent oil spill at the Bunu-Tai and Gio (Giokoo) communities. It could also be attributed to the frequent deposition of crude oil wastes/particulate matter emission deposits that contain radioactive uranium on the water bodies at Bodo community. The emission of particulate matter and crude oil wastes in Bodo and Giokoo communities are due to illegal artisanal oil

Table 2: Radiological and Chemical Toxicity Risk of Uranium in the Water Samples

S/No	Sample Name		Chemical Toxicity Risk			Radiological Risk	
		^{238}U (Bq/l) (Uranium Activity Concentration)	^{238}U (µg/l) (Uranium mass Concentration)	LADD (µg/kg/day)	Hazard Quotient	Lifetime cancer Mortality Risk	Lifetime cancer Morbidity Risk
1	Eleme Well Water	-	-	-	-	-	-
2	Bunu-Tai Well Water	12.77±1.12	514.71±45.14	14.09	23.48	$4.79x10^{-4}$	$7.34x10^{-4}$
3	Bodo Well	11.94±1.12	481.16±45.13	13.18	21.96	$4.48x10^{-4}$	$6.86x10^{-4}$
4	Alode (Eleme) Tap/borehole	2.42±0.45	97.52±18.13	2.67	4.45	$9.08x10^{-5}$	$1.39x10^{-4}$
5	Bunu-Tai Tap/borehole	3.82±2.20	153.94±88.65	4.22	7.03	$1.43x10^{-4}$	$2.19x10^{-4}$
6	Bodo Tap/borehole	5.31±0.80	213.98±32.24	5.86	9.76	$1.99x10^{-4}$	$3.05x10^{-4}$
7	Ochanai Creek (Eleme)	6.81±0.80	274.43±32.24	7.52	12.53	$2.56x10^{-4}$	$3.91x10^{-4}$
8	Oo-a-naana River (Bunu-Tai)	8.48±1.84	341.72±74.15	9.36	15.60	$3.18x10^{-4}$	$4.87x10^{-4}$
9	Sugi-Bodo River (Bodo)	8.82±1.20	355.43±48.36	9.74	16.23	$3.31x10^{-4}$	$5.07x10^{-4}$
	CONTROL WATER SAMPLES						
1	Hand-dug Well	1.32±0.14	53.20±5.64	1.52	2.53	$4.95x10^{-5}$	$7.58x10^{-5}$
2	Tap/borehole	2.26±0.08	91.09±3.22	2.60	4.33	$8.48x10^{-5}$	$1.29x10^{-4}$
3	River/Creek	2.94±0.10	118.50±4.03	3.38	5.63	$1.10x10^{-4}$	$1.68x10^{-4}$
Ye-shin et. al.(2004); Amakom and Jibiril (2010)				**0.6**	**< 1.0**	**1.0x10-3**	**1.0x10-3**
WHO (2011); USEPA (2003)			**30**				
ICRP (1993)			**1.9**				
Health Canada (1999); Australia and New Zealand (1998)			**20**				
Threshold Daily Intake TDI (WHO, 2011)				**1.0**			

refining activity in the area. Thus, the deposited uranium on the topsoil finds its way into underground hand-dug well water through infiltration and leaching resulting to enhance uranium concentration and the chemical toxicity of the water samples in the hand-dug wells whose depths are less than 10metres because the geology of the area is characterized by high water level. A comparison of the lifetime average daily dose (LADD) obtained in this study with the reference dose (RfD) of 0.6µg/kg/day as the acceptable level, shows that the chemical toxicity risks due to uranium in water samples were all above the reference dose. This therefore confirms that the health risks associated with intake of uranium by the inhabitants in the study area through drinking water from the 3 water supply sources are mainly due to the chemical toxicity risk. The calculated hazard quotient (HQ) using reference dose (RfD) value of 0.6µg/kg/day showed that HQ values are greater than unity (i.e HQ>1.0) as in figure 5, indicating significant potential risk due to chemical toxicity of uranium in water (USEPA, 1993). The study therefore confirm that human risk due to uranium content in water supplies that will result from ingestion may be attributed to chemical toxicity of uranium as heavy metal rather than radiological risk.

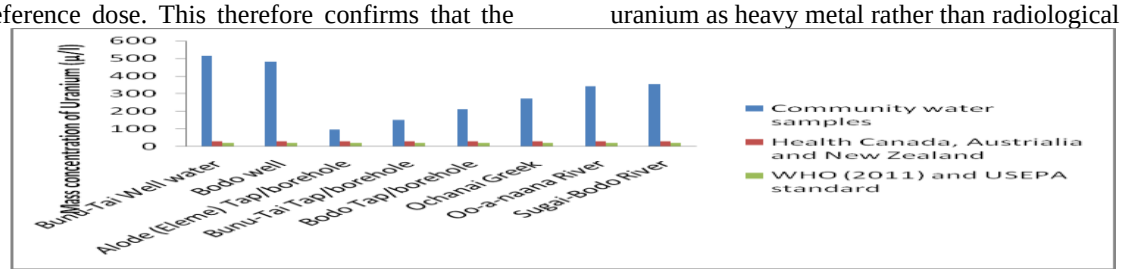

Figure 3: Comparison of Uranium mass concentration in the 3 water supply sources with International Standards

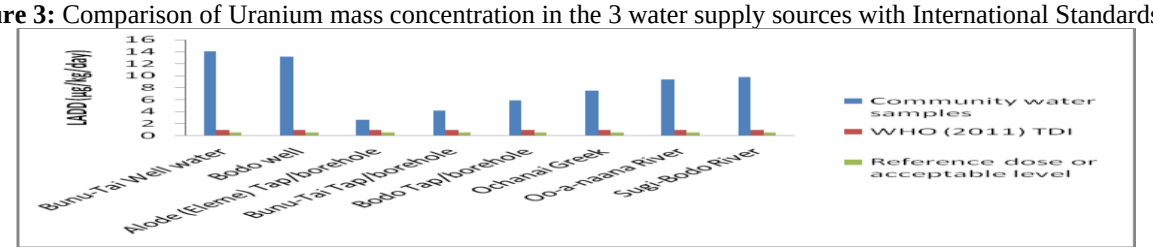

Figure 4: Comparison of Lifetime average daily dose due to intake of uranium from the 3 water supply sources with WHO Permissible Standards

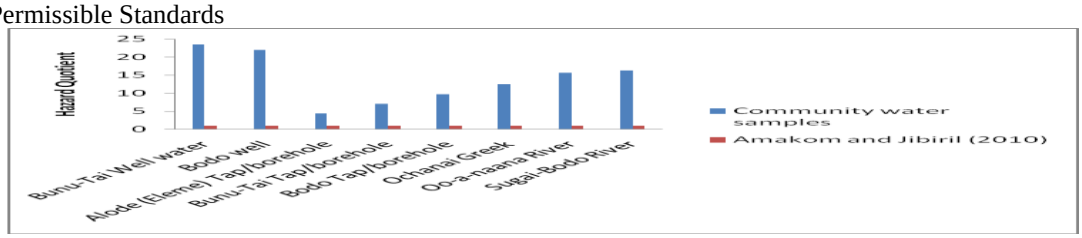

Figure 5: Comparison of Hazard Quotient due to intake of uranium from the 3 water supply sources with Standards

Conclusion:

The radiological and chemical toxicity risks associated with the consumption of uranium from surface and groundwater in oilfield communities in Ogoniland have been established. The value of uranium mass concentration (µg/l) in the 3 water supply sources sampled were found to be relatively higher compared with the recommended safe limits by various international organizations. The radiological dose of ^{238}U in the hand-dug well, tap/boreholes and rivers/creek was estimated on the basis of the cancer mortality and morbidity risks due to intake of uranium in drinking water. It was found that the chances of the inhabitants of the community and its environs having a lifetime cancer mortality and morbidity risks due to intake of uranium from the 3 drinking water sources are low as the calculated risks are much lower than the permissible risk limits of $1.0x10^{-3}$ respectively. The cancer mortality and morbidity risk values are in agreement with other reported values within and outside the country and are well below accepted standard. But the chemical toxicity risk of uranium in the water samples from the 3 water supply sources as estimated by LADD and HQ showed that their values were above the threshold daily index (TDI) of 1.0 (WHO, 2011), the reference dose level (RfD) of 0.6µg/kg/day (i.e the accepted level) and the HQ > 1. According to USEPA (1993) this indicated a significant health risk due to chemical toxicity in uranium. It could therefore be concluded that human risk due to uranium content in water supplies that will result from ingestion in the study area may be attributed to chemical toxicity of uranium as heavy metals rather than radiological risks. It is therefore recommended that an ion exchange pre-treatment and reverse osmosis treatment technique be adopted to the water supply sources as this will help to reduce and remove heavy metals and radionuclide such as uranium from the water before consumption.

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