



## Research article

## Risk assessment of drinkable water sources using gross alpha and beta radioactivity levels and heavy metals

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## ARTICLE INFO

## Keywords:

Gross alpha  
Gross beta  
Heavy metals  
Risk  
Environmental analysis  
Environmental assessment  
Environmental chemistry  
Environmental hazard  
Environmental risk assessment  
Environmental science  
Chemistry

## ABSTRACT

The increase in economic activities as a result of population surge in Dutsin-Ma has culminated into increase in waste and environmental quality degradation. Hence, this study examined the risk associated with different drinking water sources (dam water (DM), borehole water (BW), Hand dug well water (WW) and tap water (TW)) in Dutsin-Ma, Katsina using concentrations of gross alpha ( $\alpha$ ) and gross beta ( $\beta$ ) activity, and heavy metals cadmium (Cd) and lead (Pb) in the water. A total of Thirty six (36) samples were collected from the study area. Standard methods were deployed for the determination of pH, concentration of gross alpha and gross beta activity and concentrations of Cd and Pb in the water samples were done using chemical fraction method. The results of the pH showed values not within the recommended limits for drinking water quality with exception of 8.30 (WW). Also, the measured activity concentrations of gross alpha in all water samples are below 0.1 Bq/L with few exceptions, while the measured activity concentrations of gross beta in 50% of the total water samples collected are higher than 1.0 Bq/L permissible limit. The mean annual dose equivalent all the water sources is higher than 0.1 mSv recommended dose for radionuclides in water, hence the life cancer risk assessment showed higher values, indicating the water are unsafe for drinking. The results in this study showed that Cd and Pb in all the water fractions (mobile, dissolved, total and particulate) are above the WHO recommended limit of 0.003 and 0.01 mg/L in drinking water. Similarly, the hazard index (HI) for the water samples are greater than one (1), hence, a possible concern for potential carcinogenic effect for consumer of the water sources.

## 1. Introduction

The importance of water to human is numerous such that its availability in quantity and quality becomes crucial to human survival. Water is one of the essentials that support all forms of plant and animal life (Vanloon and Duffy, 2005) and it is obtained from two principal natural sources; surface water such as fresh water lakes, rivers, streams and ground water such as borehole water and well water (Mendie, 2005).

However, the availability of quality water is of interest to the scientist, government and relevant stakeholders. Some of contaminant that affects the quality of water includes heavy metals, radionuclides and gaseous emission etc. The natural radionuclides and their decay products are usually present in all types drinking water sources (Rajamannan et al., 2013).

Availability of Alpha and beta radiations known as high Linear Energy Transfer (LET) radiations in drinking water is considered to be an

important factor in increasing the natural radiation exposure in humans (UNSCEAR, 2000). These dependent upon the amount of radionuclides present in source rock, soils and other natural and artificial radionuclide materials that the water comes in contact (Darko et al., 2014). Therefore, there is the need to determine the concentration of alpha and beta radiations in water. The gross alpha and beta counting is the preliminary test, as stipulated in the World Health Organization guideline for water quality determination (WHO, 2004). Also, due to its relatively stability, low costs and simplicity, it has become a veritable tool for determination of relative radioactivity levels in water (Jobbágy et al., 2011; Sahin et al., 2017).

In addition, heavy metals become toxic when their concentrations are above the threshold recommended thereby damaging the life functions of an organism (Albergoni and Piccinni, 1983). Metals in natural waters can exist in truly dissolved colloidal and suspended forms. The proportion of these forms varies with metals and for different water bodies.

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Received 3 June 2020; Received in revised form 23 June 2020; Accepted 5 August 2020

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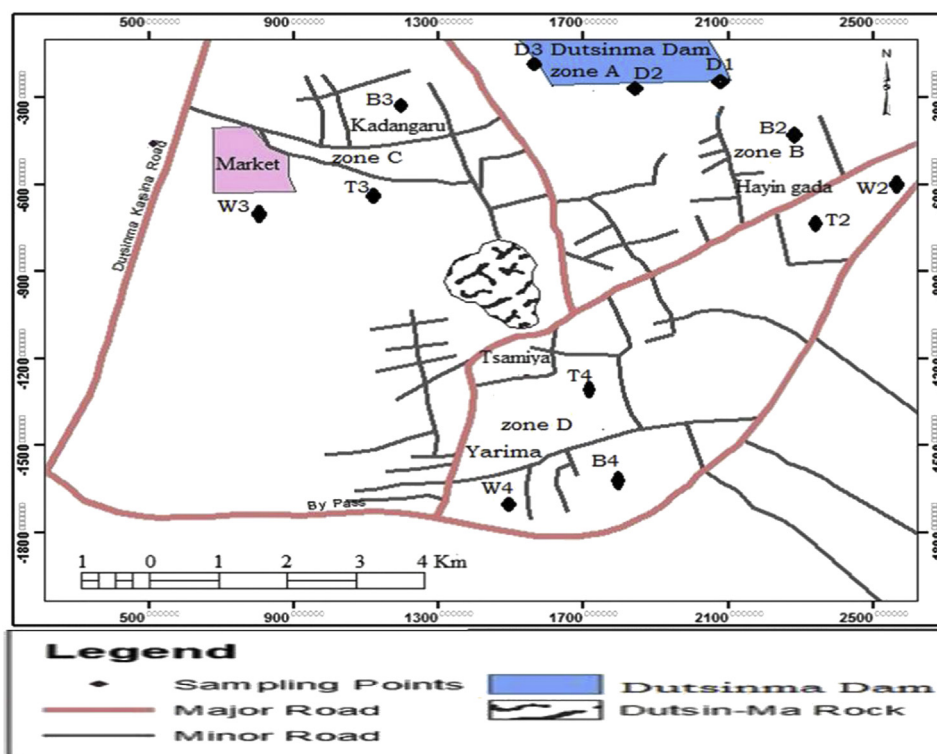


Figure 1. Map of the study area showing the sampling points.

Consequently, the toxicity and sedimentation potential of metals change depending on their forms (Marcovachia et al., 2007). Non-essential metals often exert their action through their chemical similarity to essential elements for example, cadmium with copper or zinc (Marco-vachia et al., 2007).

Dustin-Ma water resources of recent have been threatened by the population growth due to the location of the Federal University coupled with other economic activities that follows. This has culminated into generation of large waste worsen by poor disposal system which has increased the level of heavy metals in water source to the water works (Okunola and Mainasara, 2016).

Survey of literature showed that there is no published information on data related to radioactivity and heavy metal forms in different drinking water sources in Dutsin-Ma especially since the coming o Federal University Dutsin-Ma. Hence, the present study was conceived to give a baseline data information on these. Therefore, the present study aimed to determine natural radioactive using activities of gross alpha and gross beta and heavy metals (Cd and Pb) concentrations in available drinking water sources in Dutsin-Ma. The data were used to calculate the carcinogenic health risks that could emerge from the natural radioactivity and heavy metals accumulation in Dutsin-Ma drinking water.

## 2. Materials and methods

### 2.1. Study area and sampling location

Dutsin-Ma town is the headquarters of Dutsinma Local Government Area in Katsina State, Nigeria. The Local Government has an area of about 527 Km<sup>2</sup>. The coordinates of Dutsin-Ma town lie between 12° 27' 18" N and 7° 29' 29" E. Dutsinma LGA share its borders with Kurfi to the north, Charanchi to the northeast, Kankia to the east, Matazu to the southeast, Danmusa to southwest and Safana to the west.

Water samples were collected from four different sources of water in Dutsin-ma, this includes; raw water from the dam (DW), Pipe-borne (TW), borehole (BW) and hand dug well water (WW). The samples were collected in the month of June, 2016 during the early hour of the

day between 6:30 and 11:57 am. For the samples collection; Dutsin-ma town was stratified into four zones. Zone A is the Dutsinma Dam, Zone B is the Hayin Gada, Zone C is the Dutsin-Ma Market/Kandangaru; and Zone D is the Tsamiya/Yarima as shown in Figure 1. Four (4) grab samples were collected from each zone per source of water, then pooled together to give composite sample for a source. Samples were analyzed in duplicates.

Random sampling technique was employed for selecting hand-dug-wells, bore holes and tap water, except for Zone A, where three water samples of the dam water only were collected at different points. On the whole, a total of Thirty six (36) samples were collected. Three (3) samples were collected from each sample location as follows; three sampling

Table 1. pH of different water sources. Gross alpha and beta concentrations in water samples.

Sample	pH
DW1	4.66
DW2	4.87
DW3	4.70
<b>Mean ± SD</b>	<b>4.74 ± 0.11</b>
WW1	8.30
WW2	7.58
WW3	7.30
<b>Mean ± SD</b>	<b>7.73 ± 0.52</b>
BW1	5.57
BW2	5.80
BW3	5.80
<b>Mean ± SD</b>	<b>5.72 ± 0.13</b>
TW1	6.51
TW2	7.02
TW3	6.70
<b>Mean ± SD</b>	<b>6.74 ± 0.26</b>
<b>WHO (2011)</b>	<b>8.2–8.5</b>

**Table 2.** Gross Alpha and Beta Activity in different water samples.

Sample	Mean activity of gross alpha (Bq/L)	Mean activity of gross beta (Bq/L)
DW1	0.001 ± 0.003	0.185 ± 0.142
DW2	0.016 ± 0.007	0.991 ± 0.323
DW3	0.038 ± 0.027	2.685 ± 1.07
<b>Mean</b>	<b>0.018 ± 0.012</b>	<b>1.287 ± 0.512</b>
WW1	0.012 ± 0.031	1.511 ± 1.47
WW2	0.010 ± 0.007	0.036 ± 0.269
WW3	0.005 ± 0.003	0.035 ± 0.001
<b>Mean</b>	<b>0.009 ± 0.014</b>	<b>0.527 ± 0.580</b>
BW1	0.034 ± 0.000	3.126 ± 1.170
BW2	0.146 ± 0.034	4.917 ± 0.900
BW3	0.043 ± 0.030	4.157 ± 1.230
<b>Mean</b>	<b>0.074 ± 0.021</b>	<b>4.067 ± 1.100</b>
TW1	0.003 ± 0.006	0.112 ± 0.293
TW2	0.002 ± 0.008	0.008 ± 0.003
TW3	0.037 ± 0.03	2.014 ± 1.27
<b>Mean</b>	<b>0.014 ± 0.015</b>	<b>0.711 ± 0.522</b>
<b>*Standard Reference</b>	<b>0.1</b>	<b>1</b>

\* WHO (2011).

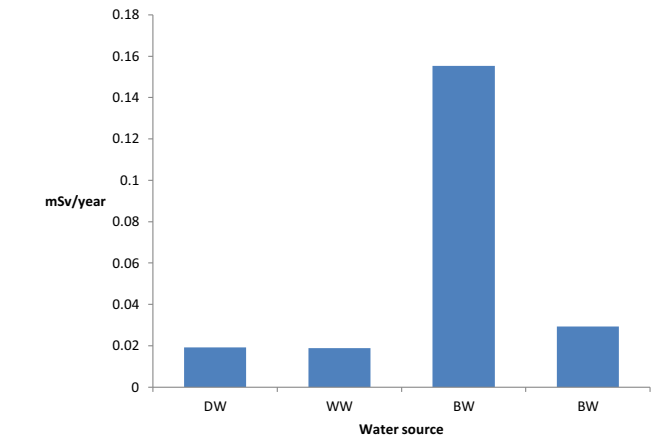
location each for the different sources of water (hand-dug-wells, bore holes, tap water and dam water). Each of the grab samples at each location for a type of water source was pooled together to give a composite sample. Hence a total of twelve (12) composite samples were obtained Two (2) liters of High Density Polyethylene (HDPE) bottles were used in the sample collection and in each, 1% (v/v) of 68.5–69% purity of concentrated nitric acid AnalaR was measured into the container and made to volume with the sampled water and covered tightly using clean cellophane and the bottle cover as soon as possible to avoid absorption of radionuclides on the walls of the containers. The water samples were kept in the laboratory until analysis was carried out.

**2.2. Determination of pH**

The pH of samples was taken *in-situ* using the HANNA instrument HI 8014. The sensor was held in the sample until the pH value was stabilized within a one decimal range. Between the readings, the sensor was cleaned or rinsed in distilled water. The pH meter was calibrated before use using buffer solution of pH 4 and 9.

**2.3. Sample preparation for gross alpha and gross beta determination**

Sample preparation and analysis was done at the Center for Energy Research and Training (CERT) Ahmadu Bello University, Zaria, Kaduna



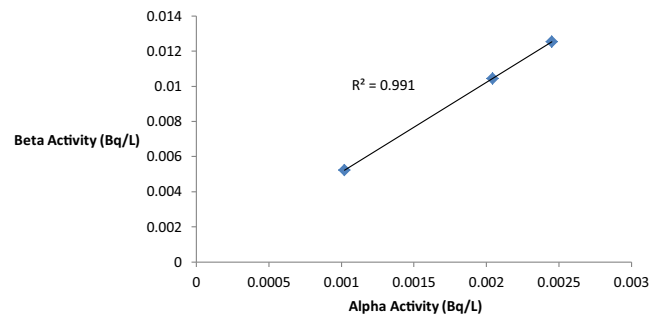
**Figure 3.** Mean effective dose equivalent for gross beta in different water sources. Relationship between beta and alpha activity concentrations.

State. Each of the 2 L water sample was evaporated using a hot plate. When the sample in question were almost dried, it was transferred to the crucible which was placed under an infrared radiator until constant weight was achieved. All the sample preparation and analysis procedures, and equipment calibration were done according to Nwoke (2006) and Onoja et al. (2007).

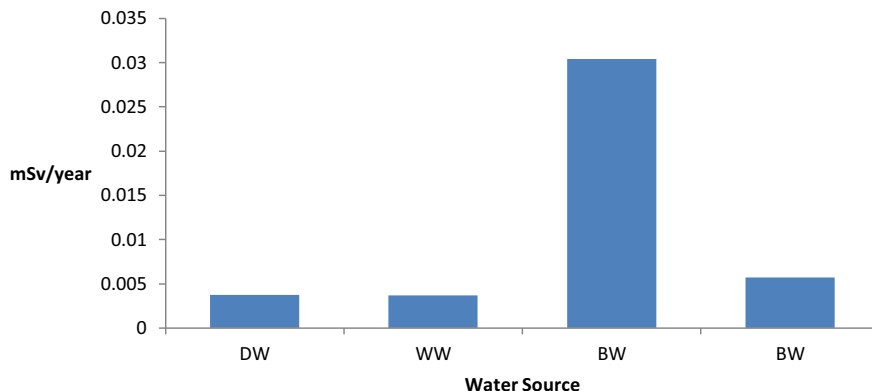
The gross  $\alpha$  and  $\beta$  activities of the water samples were estimated according to Saleh and Shayeb (2014) as shown in Eq. (1)

$$A_{\alpha,\beta} = \frac{N}{60 \times \frac{Eff_{\alpha,\beta}}{100} \times V_s} \tag{1}$$

N is the separately net gross alpha or beta count rate (cpm),  $Eff_{\alpha,\beta}$  is separately gross alpha or beta counting efficiency (in percent)  $V_s$  is the volume of sample aliquot (in L) and 60 is the conversion factor.



**Figure 4.** Correlation between gross beta and gross alpha in dam water.



**Figure 2.** Mean effective dose equivalent for gross alpha for different water sources.

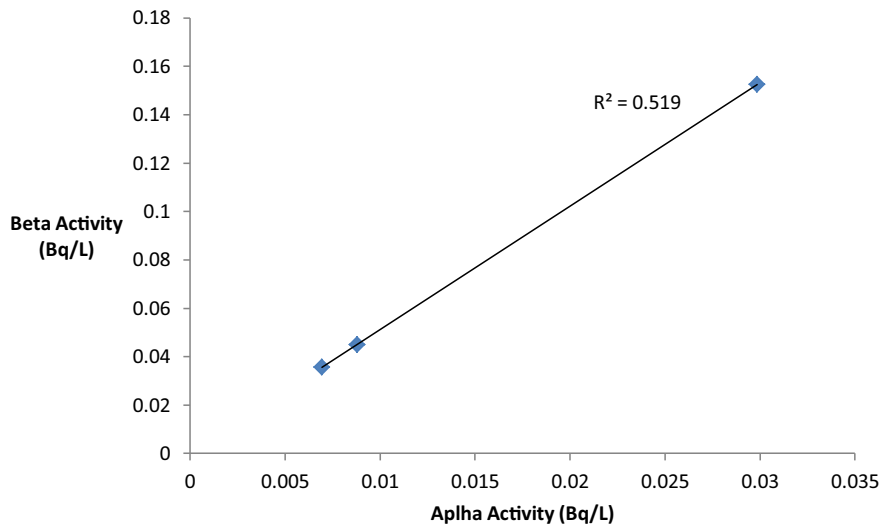


Figure 5. Correlation between gross beta and gross alpha in hand dug well water.

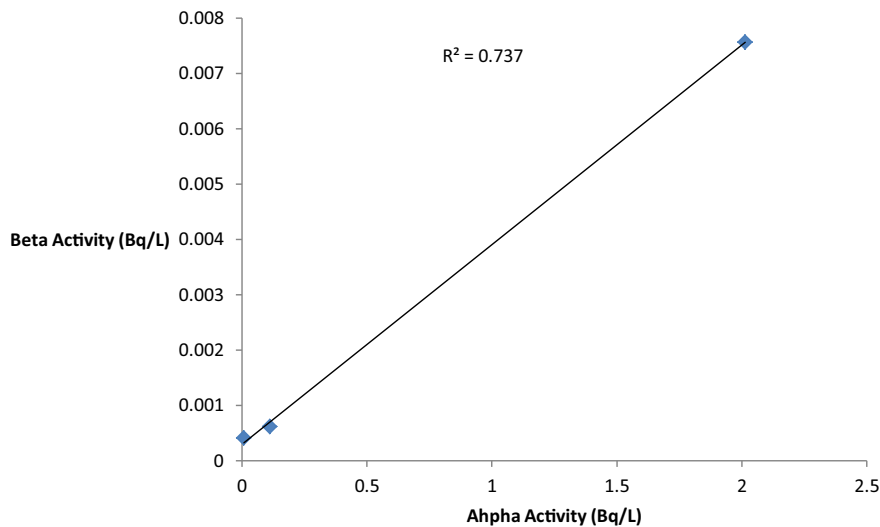


Figure 6. Correlation between gross beta and gross alpha in bore-hole water.

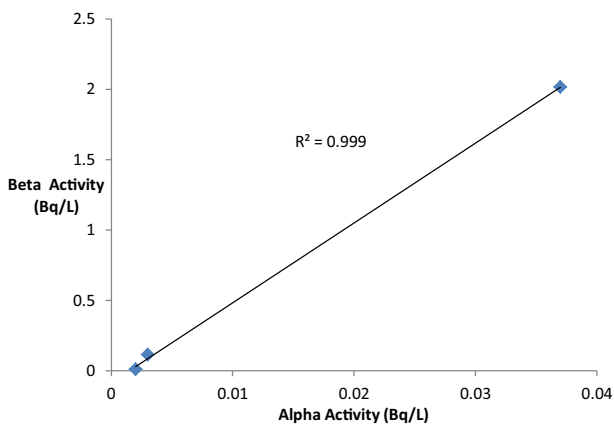


Figure 7. Correlation between gross beta and gross alpha in tap water.

#### 2.4. Chemical fractionation of samples

Chemical fractionation of water samples was carried out on the principle proposed by Bäckström et al. (2003). Samples were subjected to extraction separately using 3.2 ml of 65 %<sup>v/v</sup> concentrated HNO<sub>3</sub> to give 2% HNO<sub>3</sub> acidification in 100 ml of sample. The extraction was aimed at differentiating fractions in three stages as follows: **Dissolved fraction**, **Mobile fraction** and **Total Fraction**. The particulate concentration was calculated as the difference between **Fraction III** and **Fraction I**.

#### 2.5. Effective dose equivalent and lifetime risk assessment

The effective dose equivalent (DR<sub>w</sub>), total effective equivalent dose (TEED) and lifetime risk index using concentrations of gross alpha and beta activity were calculated using ICRP (1991), UNSCEAR (2000) and Karahan et al. (2018).

$$DR_{w(\alpha/\beta)} = A_{w(\alpha/\beta)} \times DCF_{(\alpha/\beta)} \times 730 \tag{2}$$

where DR<sub>w(α/β)</sub> is the dose equivalent effective (Sv/year), A<sub>w(α/β)</sub> is activity (Bq/L), A daily water intake of 2 L/Day according to WHO (2011) results in annual consumption rate of 730 L/year. DCF<sub>(α/β)</sub> is the

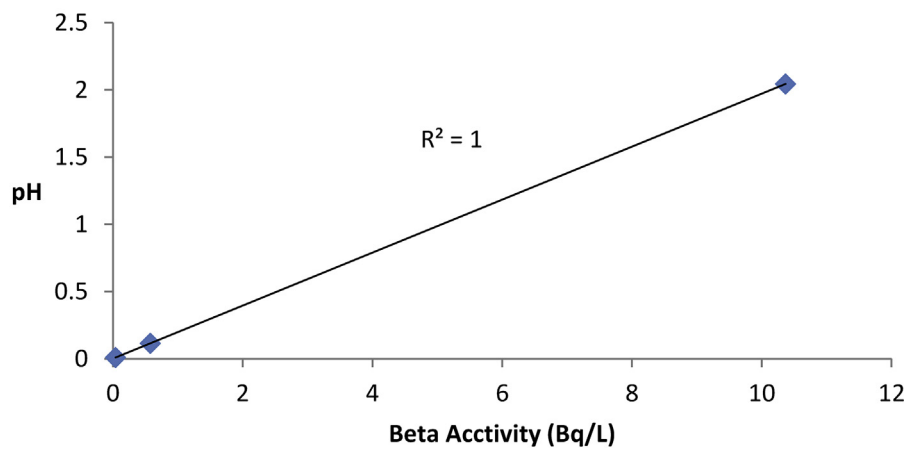
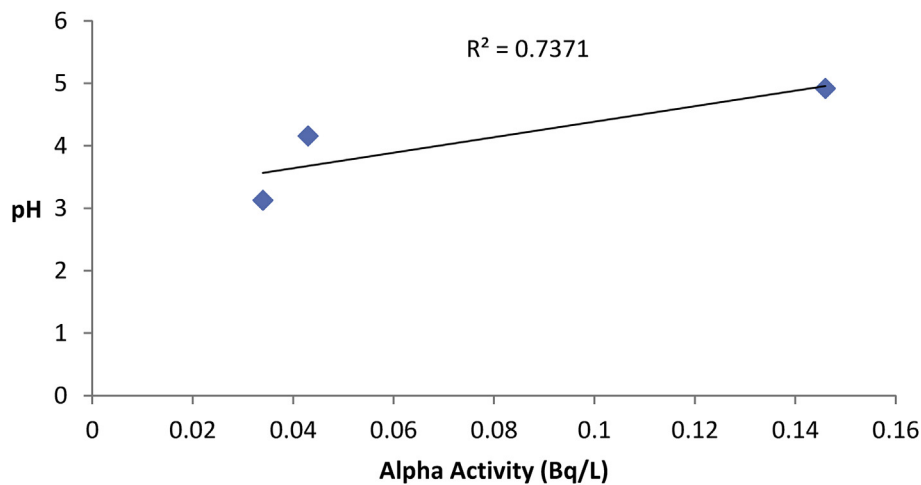


Figure 8. Relationship between gross alpha and gross beta and pH in Dam water.

dose conversion factor for ingestion of the individual natural radionuclides for adult,  $\alpha = 2.80 \times 10^{-4} \text{ mSvBq}^{-1}$  and  $\beta = 6.90 \times 10^{-4} \text{ mSvBq}^{-1}$  (WHO, 2004).

$$\text{Total Equivalent Effective Dose (mSv) (TEED}_{(\alpha+\beta)}) = DR_{w\alpha} + DR_{w\beta} \quad (3)$$

$$\text{Lifetime Risk}(LR_{\alpha}) = DR_{w(\alpha/\beta)} \times DL \times RF \quad (4)$$

where DL is duration of life (70 years) and RF is risk factor recommended as  $7.3 \times 10^{-2} \text{ Sv}^{-1}$  (ICRP, 1991).

### 2.6. Risk assessment study for heavy metals

Average daily intake of metals (ADI), Hazard quotients (HQ), excess lifetime cancer risk (ELCR) and Hazard Index were calculated accordingly using Karahan et al. (2018).

$$ADI = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (5)$$

where C is the concentration of heavy metal in water (), IR is the daily intake of water, 2 L/day (WHO, 2011). ED is the exposure duration, 70 years. EF is the exposure frequency 365 days/year. BW is the body weight of the exposed individual (adult), 70 kg. AT is the time period over which the dose is averaged,  $365 \times 70$  days for carcinogens and non-carcinogens.

$$HQ = \frac{ADI}{RfD} \quad (6)$$

where RfD is the chronic reference dose values of heavy metals, Cd =  $5.0 \times 10^{-4} \text{ mg/kg per day}$ ; Pb =  $3.5 \times 10^{-3} \text{ mg/kg per day}$

$$ELCR = \sum_{k=1}^n ADIK \times SFk \quad (7)$$

where K is the heavy metals (Cd and Pb), SFk are the average daily intake and the cancer factor, respectively for the k, n is the number of heavy metals. The SF for Cd = 15 and Pb =  $8.5 \times 10^{-1} \text{ mg/kg per day}$ .

## 3. Results and discussion

### 3.1. pH of water samples

The pH of water sample from different sources of water in Dutsin-Ma is presented in Table 1. The pH ranged from 4.66 to 4.87 with a mean value of  $4.74 \pm 0.11$  for raw water from the dam (DW), and from 7.30 to 8.30 with an mean value of  $7.73 \pm 0.52$  for hand-dug well water (WW), and from 5.57 to 5.80 with a mean value of  $5.72 \pm 0.13$  for borehole water (BW), and from 6.51 to 7.02 with an average value of  $6.74 \pm 0.26$  for tap water (TW). The lowest pH value 4.66 was obtained in water from the dam and maximum pH value 8.30 was obtained in well water.

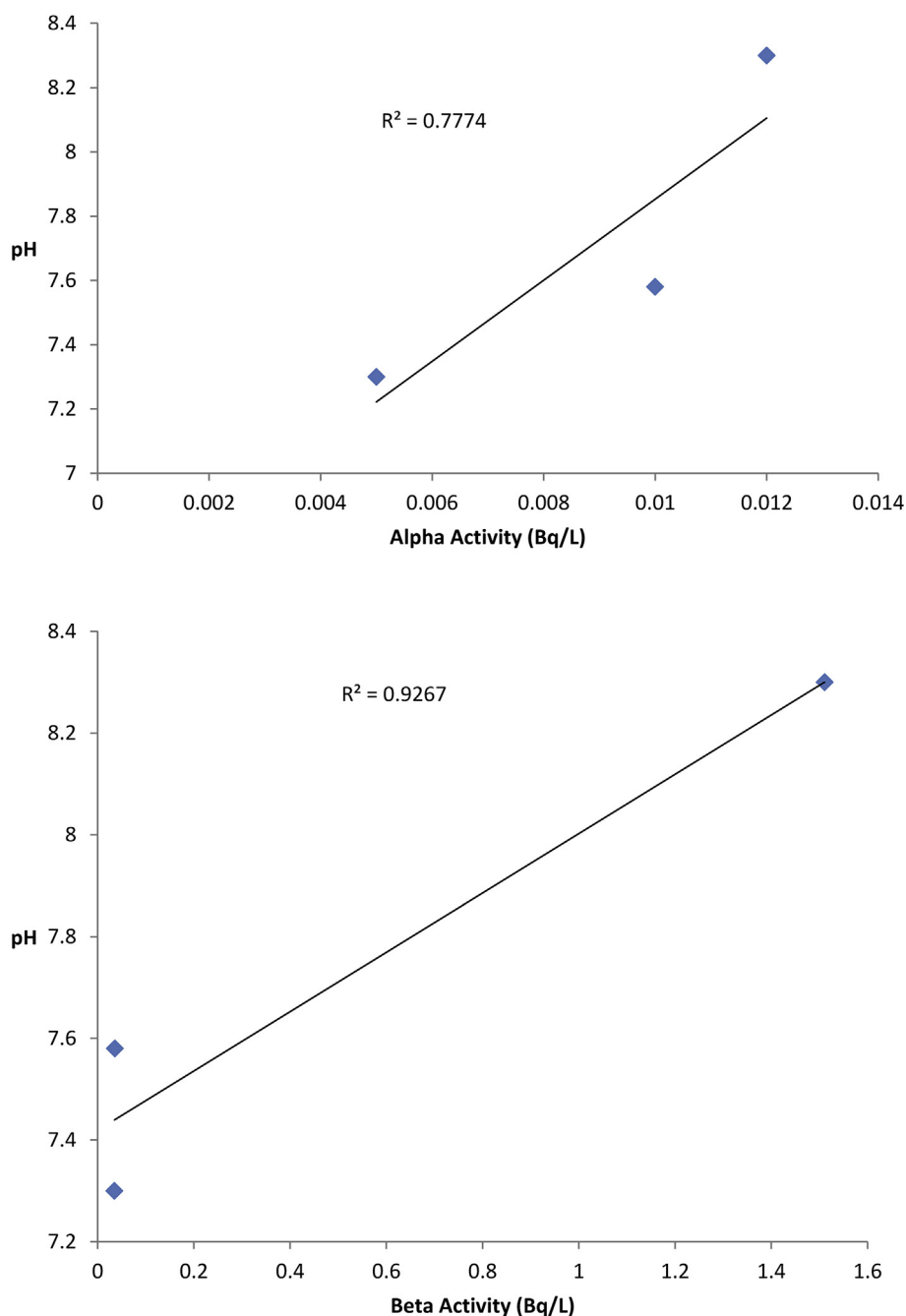


Figure 9. Relationship between gross alpha and gross beta and pH in hand-dug well water.

The WHO guideline for drinking water quality prescribed pH range of 8.2–8.8 (WHO, 2011), however, the values obtained in this study with exception of 8.30 (WW1) are all below the recommended limits for drinking water quality. pH can be influenced by various chemicals, and biological processes, geographical and environmental processes. It may be concluded that drinking water from dam, boreholes, and tap-water in Dutsin-Ma metropolis are contaminated and not suitable for drinking and other domestic uses. The analysis of variation (ANOVA) test shows significant different in pH values at various point. The mean pH values along the point are similar to typical values as (4.5–8.5) reported by (Mccutchen et al., 1992), but otherwise lower than 7.3–8.1 recorded for tap by Korkmaz et al. (2016).

### 3.2. Gross alpha and beta concentrations in water samples

The activity concentrations of gross alpha and gross beta in water samples collected from different sources in Dutsin-Ma town are given in Table 2. As shown in Table 2 the activity concentration of gross alpha in dam water (DM) ranged from  $0.001 \pm 0.003$  Bq/L to  $0.038 \pm 0.027$  Bq/L with an average value of  $0.018 \pm 0.012$  Bq/L, hand-dug well water (WW) varied from  $0.005 \pm 0.003$  to  $0.012 \pm 0.031$  Bq/L with an average value of  $0.009 \pm 0.014$  Bq/L. The minimum and maximum gross alpha activity concentration for borehole water (BW) was  $0.034 \pm 0.000$  Bq/L and  $0.146 \pm 0.034$  Bq/L, respectively, and the mean value was  $0.074 \pm 0.021$  Bq/L. Also, the range of  $0.002 \pm 0.008$  Bq/L, to  $0.037 \pm 0.03$  Bq/L, was

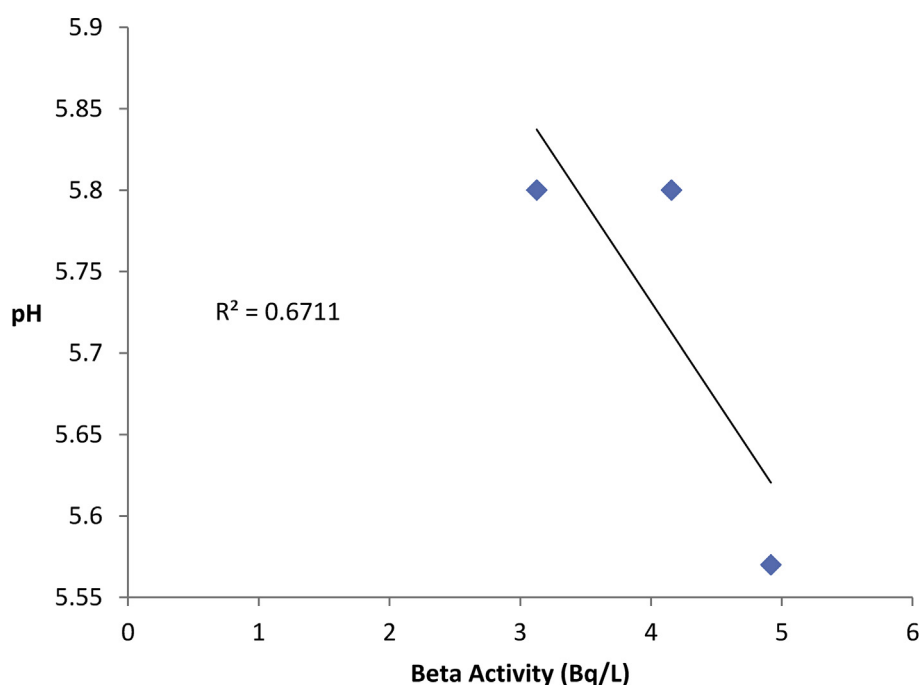
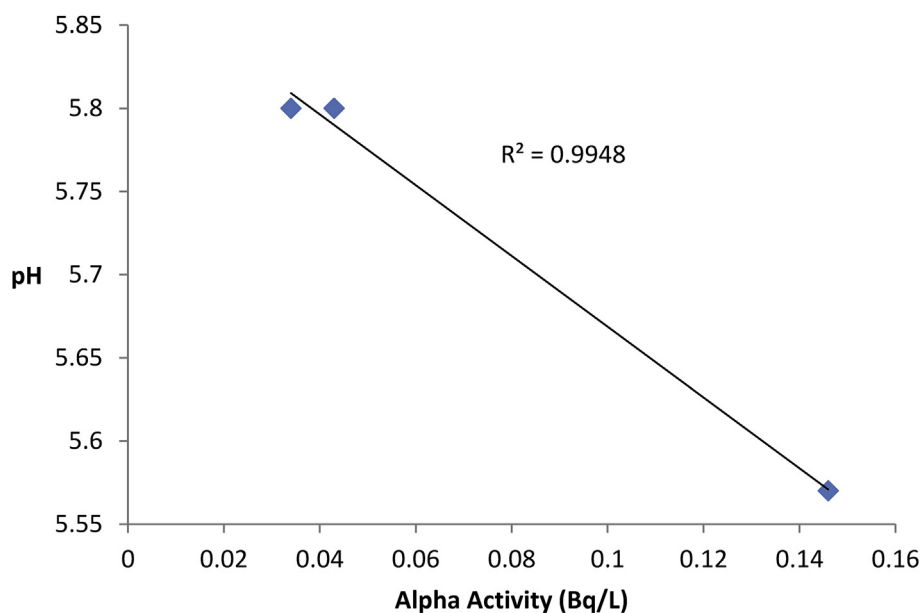


Figure 10. Relationship between gross alpha and gross beta and pH in borehole water.

recorded for tap water (TW) with an average value of  $0.014 \pm 0.015$  Bq/L.

The highest gross alpha activity in the water was measured in BW2 as shown in Figure 2. This could be due to the geological setting of the underground water source. The lowest gross alpha activity in water samples was found in DW1, a surface water supply. The gross alpha activity in water samples is primarily comprised uranium decay products such as  $^{226}\text{Ra}$  (Malanca et al., 1998), and national and world recommends the gross alpha activity concentration to be 0.1 Bq/L (SON, 2007; WHO, 2011). Hence, the results obtained in this study showed that the measured activity concentrations of gross alpha in all water samples are

lower than 0.1 Bq/L, except for BW2 which is slightly above the permissible limit.

The gross beta activity concentration ranged from  $0.185 \pm 0.142$  Bq/L to  $2.685 \pm 1.07$  Bq/L with an average value of  $1.287 \pm 0.512$  Bq/L for samples DW. For the activity concentration of sample WW, lowest and highest values of  $0.035 \pm 0.001$  Bq/L and  $1.511 \pm 1.47$  Bq/L was recorded respectively with an average value of  $0.527 \pm 0.580$  Bq/L. The gross beta activity concentrations of BH and TP water samples ranged:  $3.126 \pm 1.170$  Bq/L -  $4.917 \pm 0.900$  Bq/L and  $0.008 \pm 0.003$ – $2.014 \pm 1.27$  Bq/L with average values of  $4.067 \pm 1.100$  Bq/L and  $0.711 \pm 0.522$  Bq/L, respectively. The highest gross beta activity in the water samples

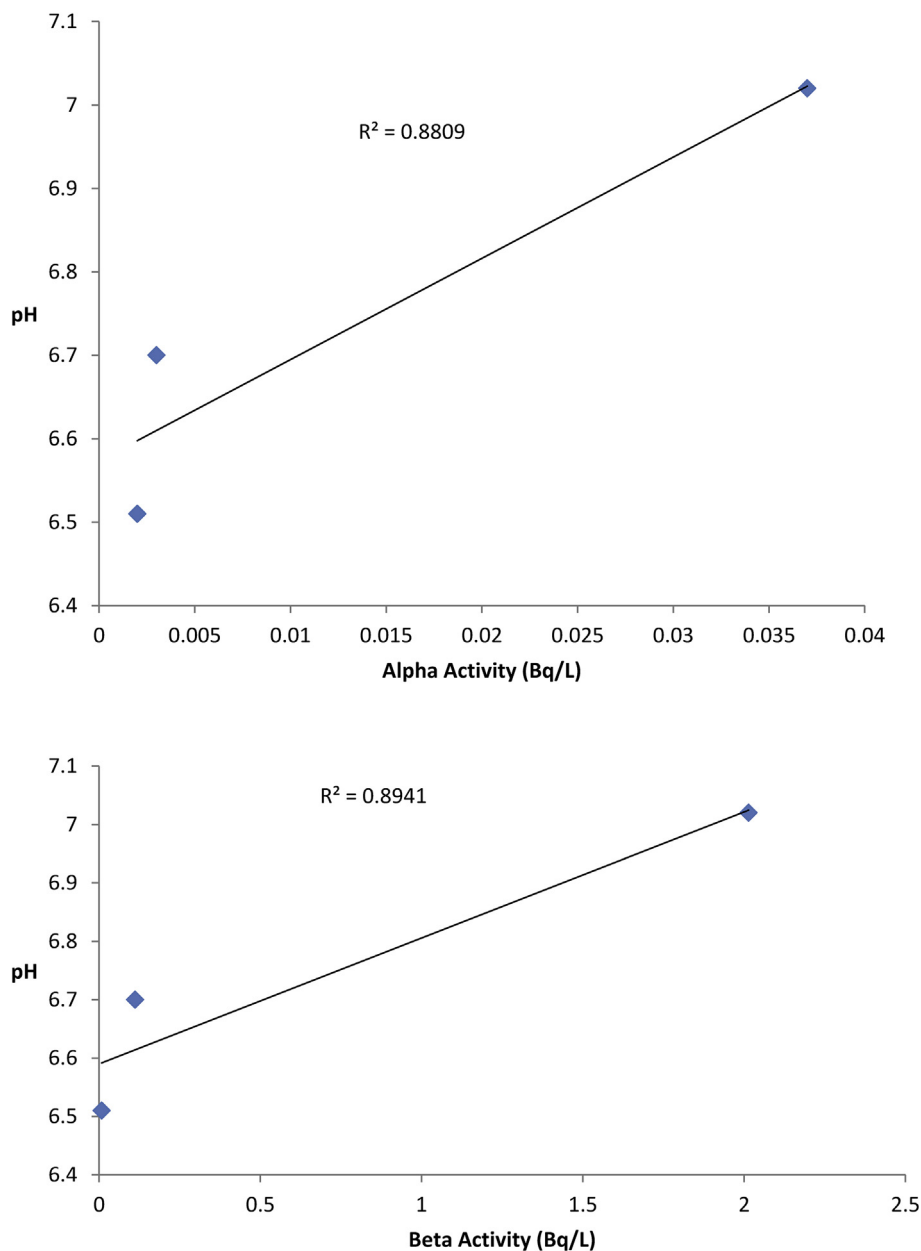


Figure 11. Relationship between gross alpha and gross beta and pH in tap water.

was found in sample BW2 as shown in Figure 3. This could be due to high level of agricultural activities around the area releasing phosphate into water. While the lowest gross beta activity in water samples was found in TW2. The variation in water sample between water sources or among the same water source could be origin of water resources and environmental factor.

The gross beta activity in water sample is primarily contributed from  $^{228}\text{Ra}$  and  $^{210}\text{Pb}$  (WHO, 2004). and national and world recommends the gross alpha activity concentration to be 1.0 Bq/L (SON, 2007; WHO, 2011). Hence, the results obtained in this study showed that The results obtained show that the measured activity concentrations of gross beta in six water samples; DW3, WW1, BW1, BW2, BW3, and TW3 representing 50% of the total water samples collected are higher than 1.0 Bq/L permissible limit.

### 3.3. Relationship between beta and alpha activity concentrations

The correlation between activity concentration of gross beta and alpha in dam water, hand-dug well water, borehole water and tap water are presented in Figures 4, 5, 6, and 7, respectively. The analysis was done to ascertain availability of the same radionuclides for the beta and alpha activities of i,n the water of the same source (Agbalagba et al., 2013). The figures showed the linear correlation of alpha and beta concentration in the water sources. The results showed significant  $R^2$  values: 0.991, 0.519, 0.737 and 0.999 for dam water, hand-dug well water, borehole water and tap water, respectively. This indicates that same radionuclides are responsible for the activities of beta and alpha particles in the water samples. Also, this validates the prediction of gross alpha activity concentration in the samples. Similar reports are made by Agbalagba et al. (2013) and Alomari et al. (2019).

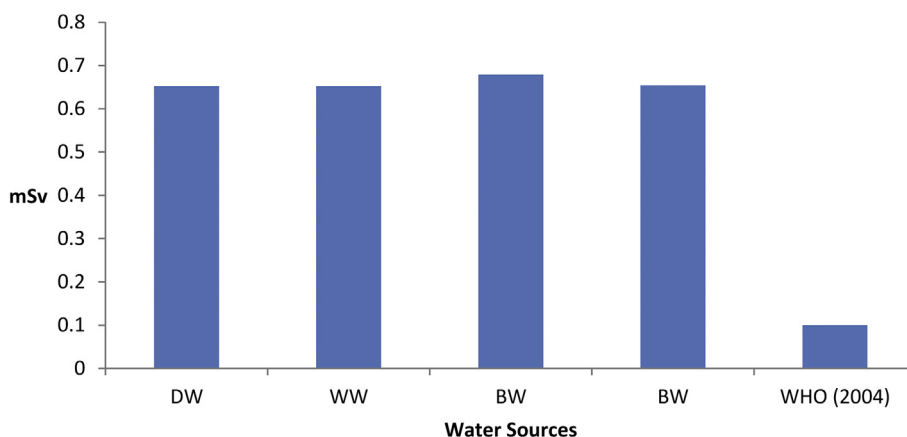


**Table 3.** Comparison of the gross alpha and beta activity concentration in this study and other studies.

Concentration of gross alpha (Bq/L)	Concentration of gross beta (Bq/L)	Type of water	References
0.001–0.038	0.991–2.685	Dam water	This study
0.005–0.012	0.035–1.511	Well water	This study
0.034–0.146	3.126–4.917	Borehole water	This study
0.002–0.037	0.008–2.014	Tap water	This study
31.46	50.14	bottled drinking water	Ismail et al. (2009)
1299	582	Groundwater	Fasae (2013)
174	222.5	portable drinking water	Fasae (2015)
45.9	91.2	Natural spring	Kobyia et al. (2015)
1.57	1.62	Ground water	Alomari et al. (2019)
0.18	0.21	Drinking water	Forte et al. (2007)
0.0002–0.015	0.0252–0.2644	Tap water	Damla et al. (2006)
0.006–0.125	0.001–0.667	Tap ans river water	Korkmaz et al. (2016)
0.012	0.234	Tap water	Saleh et al. (2015)
0.11–16	0.10–16.90	Thermal spring water	Sahin et al. (2017)
0.0157–0.1427	0.0893–0.400	Borehole and well water	Darko et al. (2014)

**Table 4.** The Annual effective dose equivalent lifetime cancer risk and total equivalent effective dose for different water sources.

Sample	Annual Effective Dose Equivalent (mSv)		Lifetime Cancer Risk (mSv/year)	Total Equivalent Effective Dose (mSv)
	DR $\alpha$	DR $\beta$	LR $\alpha$	TEED $\alpha+\beta$
DW1	0.0002	0.0932	0.0010	0.0934
DW2	0.0033	0.4992	0.0167	0.5024
DW3	0.0078	1.3524	0.0397	1.3602
<b>Average</b>	<b>0.0037</b>	<b>0.6483</b>	<b>0.0191</b>	<b>0.6520</b>
WW1	0.0025	0.7611	0.0125	0.7635
WW2	0.0020	0.0181	0.0104	0.0202
WW3	0.0010	0.0176	0.0052	0.0187
<b>Average</b>	<b>0.0018</b>	<b>0.2656</b>	<b>0.0094</b>	<b>0.2675</b>
BW1	0.0069	1.5746	0.0355	1.5815
BW2	0.0298	2.4767	0.1525	2.5065
BW3	0.0088	2.0939	0.0449	2.1027
<b>Average</b>	<b>0.0152</b>	<b>2.0484</b>	<b>0.0776</b>	<b>2.0636</b>
TW1	0.0006	0.0564	0.0031	0.0570
TW2	0.0004	0.0040	0.0021	0.0044
TW3	0.0076	1.0145	0.0386	1.0220
<b>Average</b>	<b>0.0029</b>	<b>0.3583</b>	<b>0.0146</b>	<b>0.3612</b>



**Figure 12.** Comparison of average total equivalent effective dose for difference water sources.

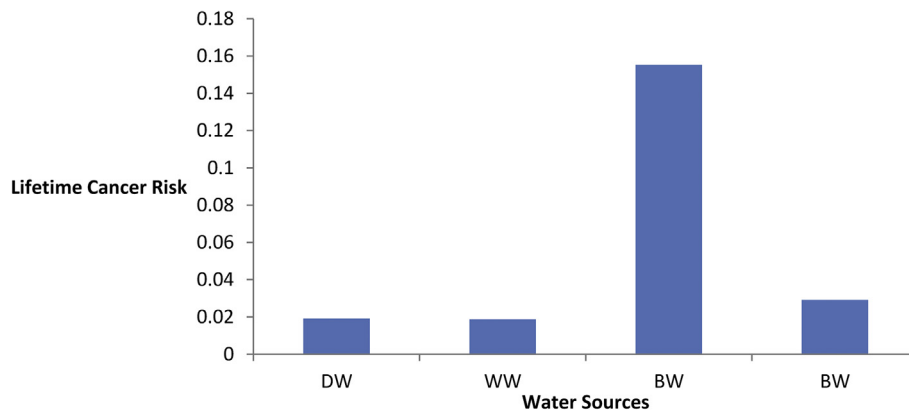


Figure 13. Comparison of average lifetime cancer risk in difference water sources r.

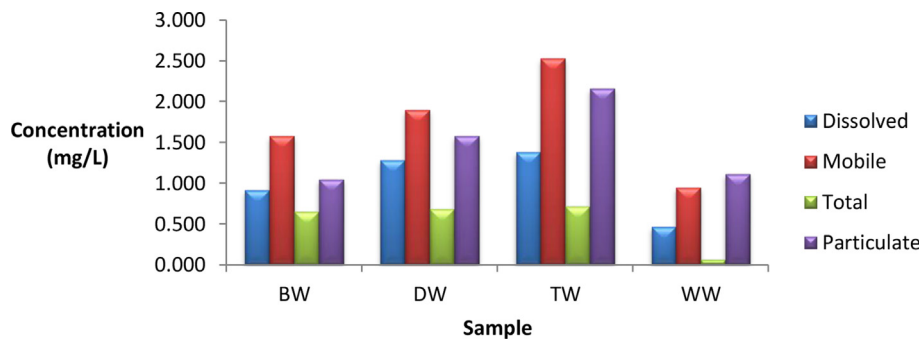


Figure 14. Concentration of Cd in different fraction of water sources.

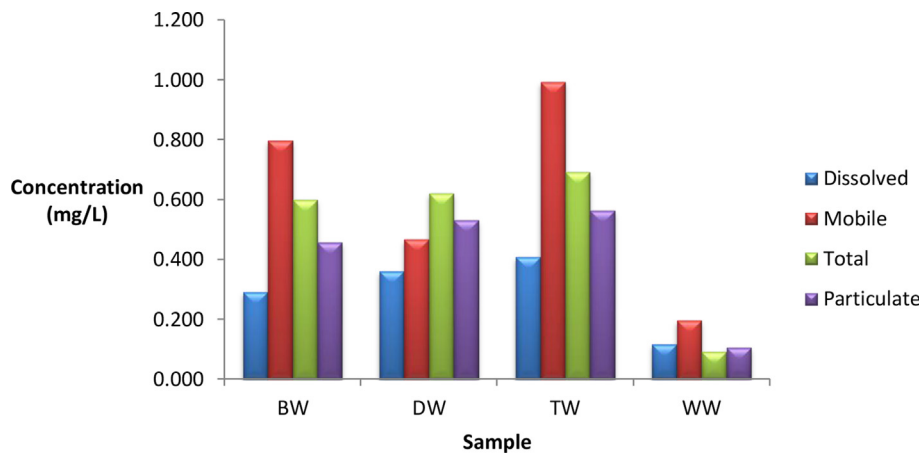


Figure 15. Concentration of Pb in different fraction of water sources.

Table 5. Correlation of Cd among fractions in water.

Parameter	Cd <sub>Dissolved</sub>	Cd <sub>Mobile</sub>	Cd <sub>Total</sub>	Cd <sub>Particulate</sub>
Cd <sub>Dissolved</sub>	1.000			
Cd <sub>Mobile</sub>	0.857**	1.000		
Cd <sub>Total</sub>	0.822*	0.958**	1.000	
Cd <sub>Particulate</sub>	0.456	0.773*	0.880**	1.000

\*\* Correlation is significant at the 0.01 level (2-tailed).

\* Correlation is significant at the 0.05 level (2-tailed).

### 3.4. Relationship between gross alpha and beta activity concentrations and pH

The correlation between pH and activity concentrations of gross beta and alpha in dam water, hand-dug well water, borehole water and tap water are presented in Figures 8, 9, 10, and 11, respectively. From the figures, R<sup>2</sup> values of 0.737 and 1.000 (DW), 0.777 and 0.926 (WW), 0.994 and 0.671 (BW) and 0.880 and 0.894 (TW) for pH vs activity concentrations of gross alpha and pH vs activity concentrations of gross beta in the samples. High positive R<sup>2</sup> between the parameters indicates that pH is responsible for the activity concentrations of gross alpha and beta in the samples. This report showed similar trends of significant relationship between the pH and gross alpha and gross beta activity at pH

**Table 6.** Correlation among Pb fractions in water.

Parameter	Pb <sub>Dissolved</sub>	Pb <sub>Mobile</sub>	Pb <sub>Total</sub>	Pb <sub>Particulate</sub>
Pb <sub>Dissolved</sub>	1.000			
Pb <sub>Mobile</sub>	0.400	1.000		
Pb <sub>Total</sub>	0.937**	0.574	1.000	
Pb <sub>Particulate</sub>	0.022	0.579	0.369	1.000

\*\* Correlation is significant at the 0.01 level (2-tailed).

< 8.3 (Alomari et al., 2019). This shows that pH of the samples are responsible for the activity concentration of the gross alpha and beta.

Comparison of the results obtained in this study with literatures is presented in Table 3. The table below shows that the activity concentration of gross alpha and beta are lower than reported studies in Nigeria (Fasae, 2013, 2015). The tap water results for concentration of gross alpha and beta in this study is higher than similar studies reported in literature (Damla et al., 2006; Darko et al., 2014; Saleh et al., 2015 Korkmaz et al., 2016). For the ground water: borehole and well water, the activity concentration of gross alpha and beta are higher and lower than reports of Kobya et al. (2015) and Alomari et al. (2019), respectively as shown in Table 3. The higher or lower activity concentrations of gross alpha and beta in this study compare to other studies is due to the geological properties of the underground source of water and the activities carried out around the water source.

The radiological risk indexes of different drinking water sources are presented in Table 4. The indexes Includes annual effective dose equivalent lifetime cancer risk and total equivalent effective dose. The annual effective equivalent dose for alpha (DR $\alpha$ ) ranged from 0.0002 to 0.0298 mSv with the highest and lowest values obtained for borehole water and dam water, respectively. Also, the annual effective equivalent dose due to beta activity (DR $\beta$ ) ranged from 0.0040 to 2.4767 mSv with the lowest recorded in tap water and the highest in borehole water. The total equivalent effective dose in the water sources showed mean value of 0.6520, 0.2675, 2.0636 and 0.3612 mSv for dam water, well water, borehole water and tap water, respectively with the lowest mean value recorded in the well water and highest mean value recorded in broehole water as shown in Figure 12. The value obtained in this study is lower than values recorded by Agbalagba et al. (2013) and higher than 0.0678

**Table 7.** Health Risk due to concentration of Cd in different water sources.

Sample	Fractions	ADI	HQ	ELCR
BW	Dissolved	0.026	5.237	0.393
	Mobile	0.037	7.331	0.550
	Total	0.040	7.903	0.593
	Particulate	0.013	2.666	0.200
<b>Mean</b>		<b>0.029</b>	<b>5.784</b>	<b>0.434</b>
DW	Dissolved	0.045	9.046	0.678
	Mobile	0.054	10.854	0.814
	Total	0.072	14.474	1.086
	Particulate	0.027	5.429	0.407
<b>Mean</b>		<b>0.050</b>	<b>9.951</b>	<b>0.746</b>
TW	Dissolved	0.019	3.714	0.279
	Mobile	0.020	3.903	0.293
	Total	0.020	4.094	0.307
	Particulate	0.002	0.380	0.029
<b>Mean</b>		<b>0.015</b>	<b>3.023</b>	<b>0.227</b>
WW	Dissolved	0.030	6.000	0.450
	Mobile	0.045	9.046	0.678
	Total	0.062	12.380	0.929
	Particulate	0.032	6.380	0.479
<b>Mean</b>		<b>0.042</b>	<b>8.451</b>	<b>0.634</b>

reported for drinkable water in Nigeria (Ogundare and Adekoya, 2015). The mean value obtained for all the water sources is higher than 0.1 mSv recommended dose for radionuclides in water. The implication of this is possible health risks to consumers of this water sources in Dutsin-Ma.

The lifetime cancer risk assessment in the water samples as shown in Table 4 revealed the range of 0.0010–0.0397 with a mean value of 0.0191, 0.0052 to 0.0125 with a mean value of 0.0094, 0.0355 to 0.1525 with a mean value of 0.0736 and 0.0021 to 0.0386 with a mean value of 0.0146 for dam water, well water, borehole water and tap water, respectively. The highest average value was obtained in borehole water and the least in well water as shown in Figure 13. Comparison of the results with other study shows that values obtained in this study is higher than mean of  $10.12 \times 10^{-5}$  reported for drinking water in Turkey (Kobya et al., 2015). Generally, the results show that there is risk for consumption of the waters, hence, the waters are unsafe for drinking.

### 3.5. Concentration of Cd in the water fractions

The results of chemical fractionation of different water samples for Cd is presented in Figure 14. The concentrations of Cd ranged from 0.650 – 1.53 mg/L, 0.683–1.900 mg/L, 0.717–2.533 mg/L and 0.066–1.117 mg/L for dissolved, mobile, total and particulate fractions. The highest concentration of the fractions for all samples was obtained in dam water (DW) sample with exception of particulate fraction which presented highest concentration for well water. Analysis of variation (ANOVA) among the samples fractions showed the statistical differences of Dissolved = 0.079, Mobile = 0.025, Total = 0.005, and Particulate = 0.074 of different fractions. The results in this study showed that Cd is above the recommended limit of 0.003 mg/L (WHO, 2011). Analyses of relationship among the fractions for Cd are shown in Table 5. Significant correlations ( $p < 0.05$ ) observed among the fractions indicates that the sources Cd in these fractions are same.

### 3.6. Concentration of Pb in the water fractions

The mean concentration of Pb in different fractions of the water using chemical fractionation methods are presented in Figure 15. Analysis of variation (ANOVA) among the fractions showed statistical differences; Dissolved is 0.582, Mobile is 0.912, Total is 0.504, and Particulate is 0.904. The concentrations of Pb varied from 0.292 mg/L (BW) – 0.798

**Table 8.** Health Risk due to concentration of Pb in different water sources.

Sample	Fractions	ADI	HQ	ELCR
BW	Dissolved	0.008	2.384	0.007
	Mobile	0.010	2.947	0.009
	Total	0.012	3.339	0.010
	Particulate	0.003	0.955	0.003
<b>Mean</b>		<b>0.008</b>	<b>2.406</b>	<b>0.007</b>
DW	Dissolved	0.023	6.510	0.019
	Mobile	0.013	3.816	0.011
	Total	0.028	8.114	0.024
	Particulate	0.006	1.604	0.005
<b>Mean</b>		<b>0.018</b>	<b>5.011</b>	<b>0.015</b>
TW	Dissolved	0.017	4.902	0.015
	Mobile	0.018	5.073	0.015
	Total	0.020	5.657	0.017
	Particulate	0.003	0.755	0.002
<b>Mean</b>		<b>0.014</b>	<b>4.097</b>	<b>0.012</b>
WW	Dissolved	0.013	3.731	0.011
	Mobile	0.015	4.339	0.013
	Total	0.016	4.600	0.014
	Particulate	0.003	0.869	0.003
<b>Mean</b>		<b>0.012</b>	<b>3.385</b>	<b>0.010</b>

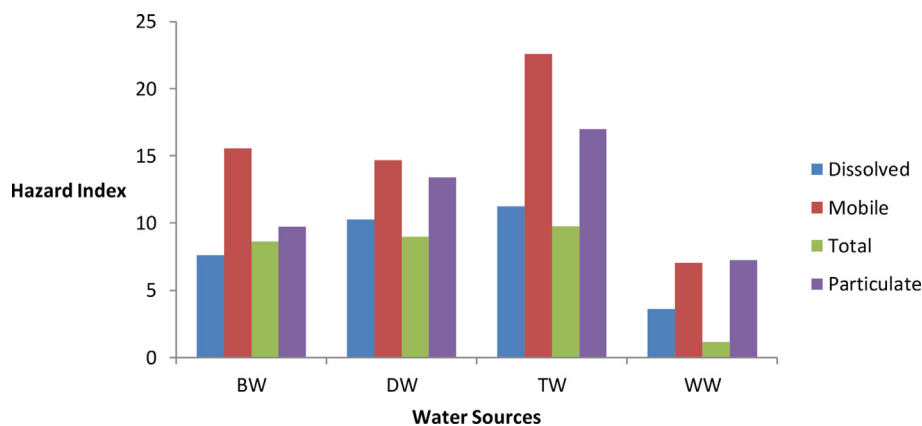


Figure 16. Hazard Index of fractions of different water sources in Dutsin-Ma.

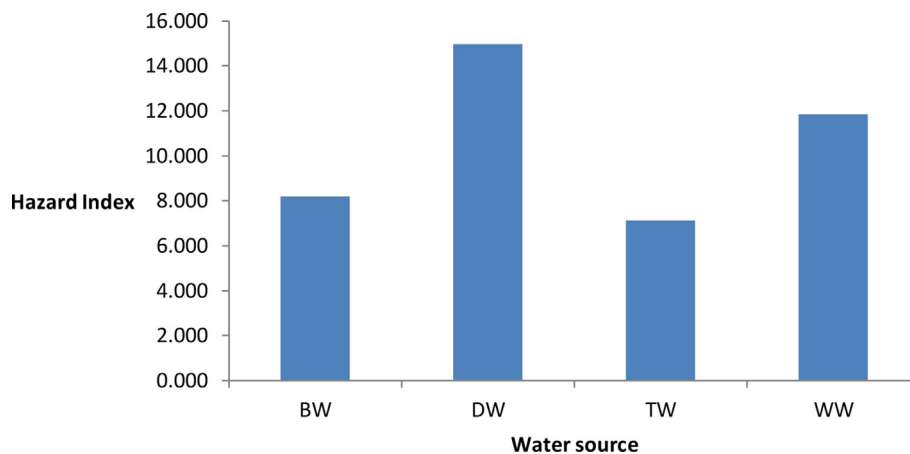


Figure 17. Average hazard Index of different water sources in Dutsin-Ma.

mg/L (DW) for dissolved fraction, 0.361 mg/L (BW) – 0.622 mg/L (TW) for mobile fraction, 0.409 mg/L (BW) – 0.994 mg/L (BW) for total fraction and 0.093 mg/L (TW) – 0.192 mg/L (DW) for particulate fraction. The highest concentration of 0.994 mg/L was recorded for sample DW of total fraction. Similarly, the values obtained in different water fractions for Pb is above recommended limit of 0.01 mg/L for drinking water (WHO, 2011). According to Watershed Protection Plan Development Guidebook, (2005) Lead sources are batteries, gasoline, paints, caulking, rubber, and plastics. Lead can cause a variety of neurological disorders. In children, it inhibits brain cell development. The analyses of relationship among the fractions for Pb are shown in Table 6. Significant correlations ( $p < 0.05$ ) observed among the fractions indicates that the sources of these fractions are same.

### 3.7. Health risk assessment of Cd and Pb in the fraction of the water samples

The calculated ADI, HQ and ELCR of Cd and Pb in different water fraction are shown in Tables 7 and 8. The results of ADI showed that particulate fraction and total fractions has the lowest and highest values of Cd, respectively in the drinking water sources except for well water (WW) that showed the lest concentration in the dissolved fraction. The highest value of 0.072 was recorded for dam water (DM) with mean value of 0.050 for the fractions. The HQ decreases in the following order DW > WW > BW > TW. The highest and lowest values of 14.474 and 0.380 were recorded for total and particulate fractions, respectively. The excess lifetime cancer due to Cd calculated showed that sample DW has the highest risk index compare to other water samples. The values

obtained in this study for ADI, HQ and ECLR is higher than values reported for the same indexes by Karahan et al. (2018) for drinking water.

Also, the results of ADI of Pb showed that particulate fraction and total fractions has the lowest and highest values, respectively in the drinking water sources. The highest value of 0.028 was recorded for dam water (DM) with mean value of 0.018 for the fractions. The mean HQ decreases in the following order DW > TW > WW > BW. The highest and lowest values of 8.114 and 0.755 were recorded for total and particulate fractions, respectively. The excess lifetime cancer due to Pb calculated showed that sample DW has the highest risk index compare to other water samples. Similarly, the values obtained in this study for ADI, HQ and ECLR is higher than values reported for the same indexes by Karahan et al. (2018) for drinking water.

### 3.8. Comparison of hazard index (HI) among the drinking water sources

The hazard index due to the Cd and Pb loads in fractions of the drinking water sources are presented in Figures 16 and 17. The results showed value higher than one for all the fractions in the water sources. Hence, since the HI value exceeds one, there may be concern for potential carcinogenic effects (Karahan et al., 2018) on consumers of this water in Dutsin-Ma.

## 4. Conclusion

This study is the first of its kind in the area of study to determine the radioactivity levels in various sources of drinking water using concentrations of gross alpha and gross beta activity and determine the

concentrations of Cd and Pb in different fractions of water using chemical fractionation. From the results obtained in the study, it clearly showed that drinking water in Dutsin-Ma is contaminated and pose great health risk to the rising population of students and workers in the town. The calculated risk indices showed the profile of DW > WW > BW > TW with respect to the potential hazard exposure due to drinking of the water. Given the significant level of Pb and Cd in the water consumed by members of the community studied, there is no doubt that appreciable amounts would have accumulated in the bodies of these individuals. Hence, a public health challenge because of the association of elevated systemic Pb and Cd with significant morbidities. The mean annual effective dose value obtained for all the water sources is higher than 0.1 mSv recommended dose for radionuclides in water. The implication of this is possible radiological health risk to consumers of this water sources in Dutsin-Ma. Finally, data obtained in this study is a baseline information that can be used for future evaluation of drinking water in the area.

## Declarations

### Author contribution statement

Oluwole J. Okunola, Olayinka B. Popoola: Conceived and designed the experiments; Analyzed and interpreted the data; Wrote the paper.

Mark O. A. Oladipo: Conceived and designed the experiments; Performed the experiments; Wrote the paper.

Theophilus Aker: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

### Funding statement

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

### Competing interest statement

The authors declare no conflict of interest.

### Additional information

No additional information is available for this paper.

## Acknowledgements

The authors wish to acknowledge the assistance and support provided by Professor Adeyemo and Mr. I. A. Bappah of the Center for Energy Research and Training (CERT), Zaria-Nigeria in sample analysis and data interpretation.

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