

Qualitative Assessment of Leaching Possibilities of Heavy metals, Air Quality and Traffic Emissions on Harvested Rainwater Quality and Its Associated Human Health Risk in Akwa Ibom State, South-South, Nigeria

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ABSTRACT

Increasing demand for water usage due to numerous factors affecting the water supply has made rainwater harvesting important for humans in the world at large especially those residing in the developing countries. This led to this study which assesses the physico-chemical and microbiological characteristics, heavy metals, metal composition of the storage vessels/roof type and air quality of the study area. Thirty-six rainwater samples were harvested in the months of April-July, 2017 in Akwa Ibom State, Nigeria in the areas with high and less industrial activities. The results were compared with USEPA, WHO and NSDWQ for water quality standards. The ratings of the air quality and harvested rainwater were evaluated using Air Quality index (AQI) and Water Quality Index (WQI) respectively. Heavy metals were assessed using Flame-AAS and standard methods were used for the analysis of other parameters. The air quality assessment was done using Gasman Air Monitoring Meter and Air Ae Steward Meter. Descriptive and two-way ANOVA statistical analyses were used for the analysis of the results. The obtained values of the rainwater samples gave mean values ranging from 5.75-6.80 for pH, E. coli (not detected), Conductivity (12.50-102.70 $\mu\text{S}/\text{cm}$) and colour (0.43-7.65 TCU). Results (mg/L) for TDS gave mean values of 4.00-27.00, TSS (12.30-38.70), sulphate (0.79-7.88), phosphate (0.31-4.51) and heavy metals (mg/L) gave 0.00-0.40 for Fe, Pb (0.00-0.25), Cr (0.00-0.45), Zn (0.01-0.23), Al (0.00-3.36).

The metal concentrations of the digested roof and storage vessels gave values ranging from 0.06-2.30 for Pb, 0.00-2.12 (Al), 63.40-63.78 (Fe), 5.89-5.94 (Zn) and 1.02-1.23 (Cr). The air quality records ($\mu\text{g}/\text{m}^3$) gave 117.67-139.70 (PM₁₀), 72.67-77.33 (PM_{2.5}). Other results (ppm) were NO₂ (0.30-0.43), SO₂ (0.47-0.57), CO₂ (10,566.67 $\pm$ 1650.50-24,466.70 $\pm$ 2218.86) and CO (22.33-28.33). The values of the analyzed rainwater samples were below, within and above the water quality set standard by USEPA, WHO and NSDWQ and these results were significantly different at ($P < 0.05$). The air quality assessments by AQI ratings gave moderate to hazardous air quality in both areas. The WQI ranged from 11.29-2,276.42, indicating that the harvested rainwater rated from excellent water quality to unfit for drinking. It was concluded that harvested rainwater should be treated before being used for drinking and other domestic/irrigation purposes and some useful recommendations were proposed for positive effect in Akwa Ibom State in the entire South-South region and in Nigeria as a whole.

Keywords: Harvested Rainwater; Air Quality; Traffic Emission, Heavy Metals; Storage Vessels; Air Quality Index; Water Quality Index

Abbreviations: AQI: Air Quality Index; WQI: Water Quality Index; SD: Standard Deviation; SPSS: Software Statistical Package for the Social Sciences

Introduction

Rainwater contamination is a serious global challenge facing the world at large and it is emanating from natural and anthropogenic factors [1,2]. Rainfall deposits most of the fresh water in the world for humans and other living beings for drinking and other purposes [3] as shown in Figure 1 below. Harvested rainwater is usually collected through rooftops or from the sky directly without any shield when rain is falling, and then stored in containers for usage but the quality of the harvested rainwater may be influenced by the materials used

in producing the roofs and industrial/environmental pollution [4-12]. Rainwater can get polluted by sulphur dioxide, nitrogen oxides, particulate matter, and heavy metals [4,13] and these substances come from air quality, industrial and traffic emissions and fuel combustion which can affect the rainwater quality by making it to become toxic [13-19]. The type of storage mediums and roofing material can affect the quality of the rainwater during catchment and storage [1,20-24]. The quality of the rainwater, if it is acidic can dissolve heavy metals and other impurities from the materials of catchment and storage mediums to affect the water quality [25-29].

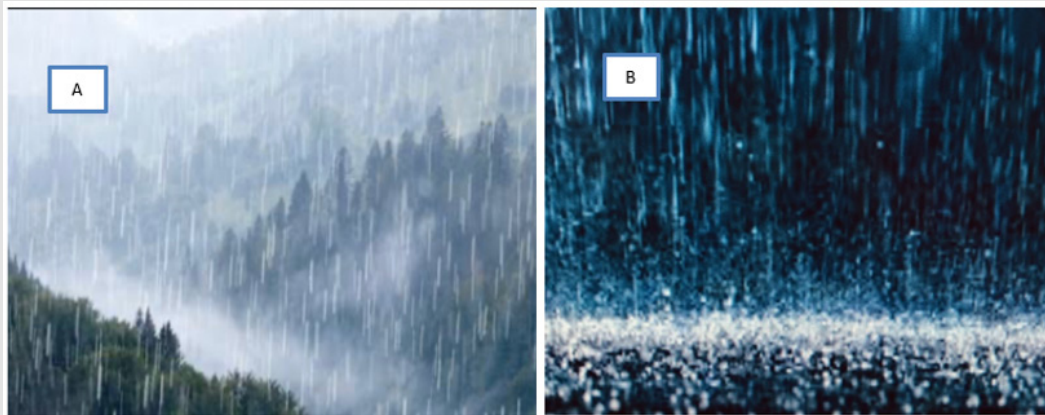


Figure 1: Plates A and B are Pictures of Rainfall.

Water quality can be affected by numerous factors from the environment and the standard ratings for drinking water quality by the water quality index (WQI) values are in five categories which are: class (A) WQI, 0-25 represents excellent water quality and the possible usage is drinking, irrigation and industrial, class (B) WQI 26-50 represents good water quality and is suitable for domestic, irrigation and industrial, class (C) WQI 50-75 represents poor water quality and the possible is irrigation and industrial, class (D) WQI 76-100 denotes very poor water quality and is suitable for irrigation, class (E) WQI > 100 represents unfit for drinking water quality and is restricted for irrigation use and proper treatment is required before usage [30]. Water quality has to do with the general environmental status of any area depending on the activities of such an area having influence on the air quality, as presented and published in numerous academic scientific journals. When some gases exist in the atmosphere above the set limits thereby decreasing the quality of air in the environment, which could be dangerous to the health of humans, it is simply called air pollution [31,32]. Air quality is simply the condition of the air within our surrounding, and the environment can get pollution from these sources which are; nitrogen oxides, sulphur oxides, carbon monoxide, particulate matter, photochemical oxidants (for example; ozone) and lead, along with a variety of airborne heavy metals and volatile organic compounds which are as a result of industrialization and traffic

emissions which pollute the atmosphere [32-36]. Several researchers have reported atmospheric contamination of rainwater by various pollutants that harbour in the air which are from industries, automobiles and other factors [1,2,6,32,34,37-46]. Since, the industrial activity and materials used in producing the roofs and storage containers can have effect on the rainwater quality; therefore, there is need for qualitative evaluation and assessment, to check, if the rainwater quality is in line with the set standards for drinking water [47-49].

Materials and Methods

Proper quality assurance procedures were observed and taken in this work in order to ensure the accuracy of the results according to the standard methods [50,51].

Study Area

This research work was carried out for four months (April -July, 2017) in the wet season in the urban and rural areas of Akwa Ibom State, Nigeria. The study areas with their coordinates were; Ikot Ekpene road, Uyo in Uyo local government area (urban) Latitude 5.03903 oN and Longitude 7.91541oE and in Eneh Awa village, Ibeno local government area (rural) Latitude 4.54446 oN and Longitude 8.00480 oE as shown in Figure 2 below.

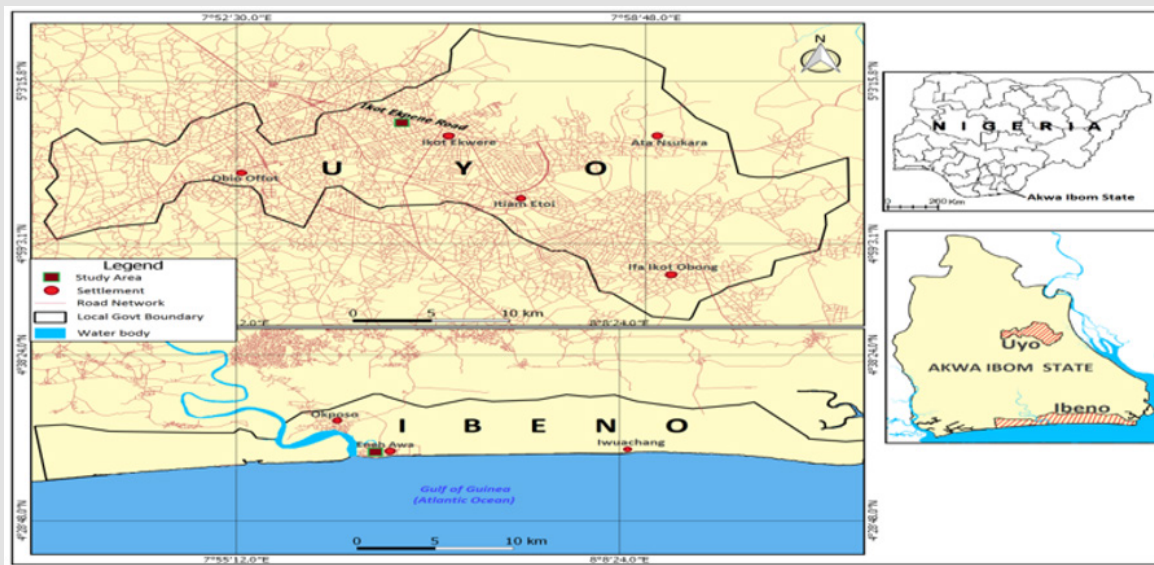


Figure 2: Map of the Study Area with their Coordinates.

Industrial Activity of the Study Area

Ikot Ekpen Road, Uyo in Uyo local government area is populated by government offices. It is where the University of Uyo, Uyo (Uniuyo) is situated. Foot Bridge, taxi loading bays and industrial motor parks are equally located on this road while Eneh Awa village, Ibeno local government area is a highly industrious area because it is where Exxon Mobil Corporation oil firm industry and Ibeno beach resort centres are situated which can produce acidic gases to pollute the environment.

Sample Collection

Thirsty-six harvested rainwater samples were collected from two different sampling locations in the urban and rural areas of Akwa Ibom State, Nigeria between the months of April-July, 2017. Control samples was collected randomly by installing a sterilized rainwater collector 1-2 metres high above the ground to avoid contaminations

and was covered with a sieve of 0.45 micron in diameter in each designated sampling point/locations during rainfall out in the open, weekly without any shield on the sky for three weeks to constitute triplicate. Immediately after the first rain flush out from the roof run-offs, rainwater was collected from Cameroun zinc roof run-off weekly for three weeks for its triplicate. Rainwater samples stored for one month to a year in storage containers were also considered and harvested. Rainwater samples stored in storage containers were collected weekly for triplicate purposes from roofs run-offs using PVC pipes and aluminium funnels connected on the long span aluminium rooftop at the different sampling locations channeling the rainwater into the household PVC and galvanized iron tanks respectively.

Sample Codes and Groupings

The collected samples were grouped and named as HRWAAU/R and HRWAFU/R which were divided into two portions as shown in Table 1 below.

Table 1: Sampling places, sample code, areas and sources of the harvested rainwater.

Sample code	Sampling Place	Areas	Sources of the Harvested Rainwater
HRWAA _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Direct rainwater from the sky (control)
HRWAA _R	Eneh Awa Village, Ibeno local government area	Rural	Direct rainwater from the sky (control)
HRWAB _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Harvested rainwater stored in PVC tank for one month
HRWAB _R	Eneh Awa Village, Ibeno local government area	Rural	Harvested rainwater stored in PVC tank for one month
HRWAC _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Harvested rainwater stored in PVC tank for one year
HRWAC _R	Eneh Awa Village, Ibeno local government area	Rural	Harvested rainwater stored in PVC tank for one year
HRWAD _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Harvested rainwater from Cameroun zinc roof
HRWAD _R	Eneh Awa Village, Ibeno local government area	Rural	Harvested rainwater from Cameroun zinc roof
HRWAE _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Harvested rainwater stored in Galvanized iron tank for one month
HRWAE _R	Eneh Awa Village, Ibeno local government area	Rural	Harvested rainwater stored in Galvanized iron tank for one month
HRWAF _U	Ikot Ekpene Road, Uyo, Uyo local government area	Urban	Harvested rainwater stored in Galvanized iron tank for one year
HRWAF _R	Eneh Awa Village, Ibeno local government area	Rural	Harvested rainwater stored in Galvanized iron tank for one year

Analytical Methodology

All the reagents used in this study were of analytical grade and from Sigma-Aldrich in the USA.

Digestion and Analysis of the Collected Samples

Heavy metals analysed in the collected rainwater samples were done using a Flame-Atomic Absorption Spectrophotometer (Dell, ICE 3000) as described in the standard method [51,52] and in the previous findings by [1]. Physico-chemical properties carried out in this study were evaluated using UV-Visible Spectrophotometer 752P (Model: 752Pro15054) according to standard methods [51,52]. The collected samples were later transferred into properly labelled sample plastic containers which were tightly covered to avoid contamination. Some quantities of the samples were added 3.0 cm³ of Conc. HNO₃ for preservation. These samples were kept in the refrigerator at 4 °C immediately after the determination of temperature, pH, TDS and EC at the collection point. Heavy metals were analysed using Flame-AAS as described in the standard method. All other physico-chemical properties were determined according to standard methods [50-52].

Control and Quality Assurance

Standard solution of 1000 ppm by Sigma-Aldrich, USA, was used for the spiking and calibration standards. The blank samples and standard solutions were analysed to ensure the results accuracy and precision [53].

Determination of Chemical Composition of Roof Type and Storage Vessels

Determination of chemical composition of the various roof types and storage containers was done according to the standard method [51,52].

Determination of *Escherichia Coli*

Determination of *Escherichia Coli* was determined in the samples according to the standard methods [51,52].

Determination of Air Quality Assessment

The air quality assessment in the urban (Ikot Ekpene Road, Uyo in Uyo L.G.A (KeKe loading Bay and Round about Metropolis) and rural (Eneh Awa Village, Ibeno L.G.A (where Exxon Mobil Corporation is situated) areas of Akwa Ibom State, Nigeria was carried out on an hourly basis for 3 h per sampling station; the periods of measuring data from selected sites were as follows for four working days (morning; peak hours (8:00 am), afternoon; off-peak period (2:00 pm) and Evening; peak hours (4:30 pm). This was done in situ by determining the air pollutants using Gasman potable digital Air Quality monitoring pieces of equipment (Models 1200-19831) and Air Ae Steward (Air Quality Monitor; Handheld (Haz-Dust Tm; PM_{2.5} and PM₁₀, Model: HD1000) after calibration according to the manufacturer’s specification. At each designated position, the instrument was held at arm’s length in an open space. The knob was adjusted to TEST position and allowed to stand for 2 min. The instrument was then adjusted to the GAS position and reading was taken in triplicates when the display on LCD was steady as previously done by Ukpung [54]; Nicholas and Ukoha [32,34].

Calculation of Water Quality Index (WQI)

The calculation of WQI ratings was done using weighted arithmetic method. This method classifies the water quality according to the degree of purity using the most commonly measured water quality variables and it is mostly used by several researchers [1,2,30,55-61]. The calculation of WQI was made by using the following Eq. (1) according to Tripaty and Sahu:

$$WQI = \frac{\sum qn \times Wn}{\sum Wn} \quad (1)$$

$$Wn = \frac{1}{(sn)} \quad (2)$$

$$qn = 100 \frac{(Vn - Vid)}{(sn - Vid)} \quad (3)$$

Where,

Wn = the weightage unit of each parameter obtained as shown in Eq.(2) according to the WHO set standard values; Sn = denotes the WHO set standard values for the n th parameter; qn represents the quality ratings obtained using Eq. (3). Vn represents the n th parameter of the given sampling station and Vid is the ideal value of the n th parameter in pure water (Vid for $pH = 7$ and zero (0) for all other parameters).

Air Quality Index (AQI) Ratings

Air quality index (AQI) rating is a number used by government agencies to communicate to the public how the air currently is or how the air is forecasted to become polluted and as the air quality index increases due to an increase of air pollutants (Examples are working or rush hour's traffic or when there is an upwind forest fire), an increasingly large percentage of the population is likely to experience rapidly severe adverse health effects [31]. The United States Environmental Protection Agency (USEPA) [62-64] has classified these air pollutants into six principal category and the classifications of these air pollutants for the air quality index (AQI) ratings as shown in Table 2 below.

Table 2: Classifications of the air pollutants for the air quality index (AQI) value ratings.

S/N	Classifications	Air Quality Index Category	Air Quality Index Ratings
1.	A	0-15	Very Good
2.	B	16-31	Good
3.	C	32-49	Moderate
4.	D	50-99	Poor
5.	E	100	Very Poor
6.	F	>100	Hazardous

Statistical Analysis

Values generated in this study were calculated using Microsoft office excel 2010 for mean and standard deviation (SD). Software statistical package for the social sciences (SPSS), version 20.0 was used for the analysis of obtained results using descriptive and two-way ANOVA statistical analysis.

Results

The results of the physico-chemical parameters namely; pH, temperature, total dissolved solids, total suspended solids, electrical conductivity, colour, turbidity, nitrate, phosphate, sulphate and heavy metals (iron, zinc, lead, chromium, aluminium) and E.coli evaluated in the harvested rainwater from Cameroun zinc roof, stored harvested rainwater in galvanized iron and PVC tanks and direct rainwater from the sky (control) are shown in Tables 3-5 & Figures 3-11, air quality assessment records and Air Quality Index (AQI) ratings in Table 6 and the Water Quality Index (WQI) ratings in Table 7 & Figure 12 below.

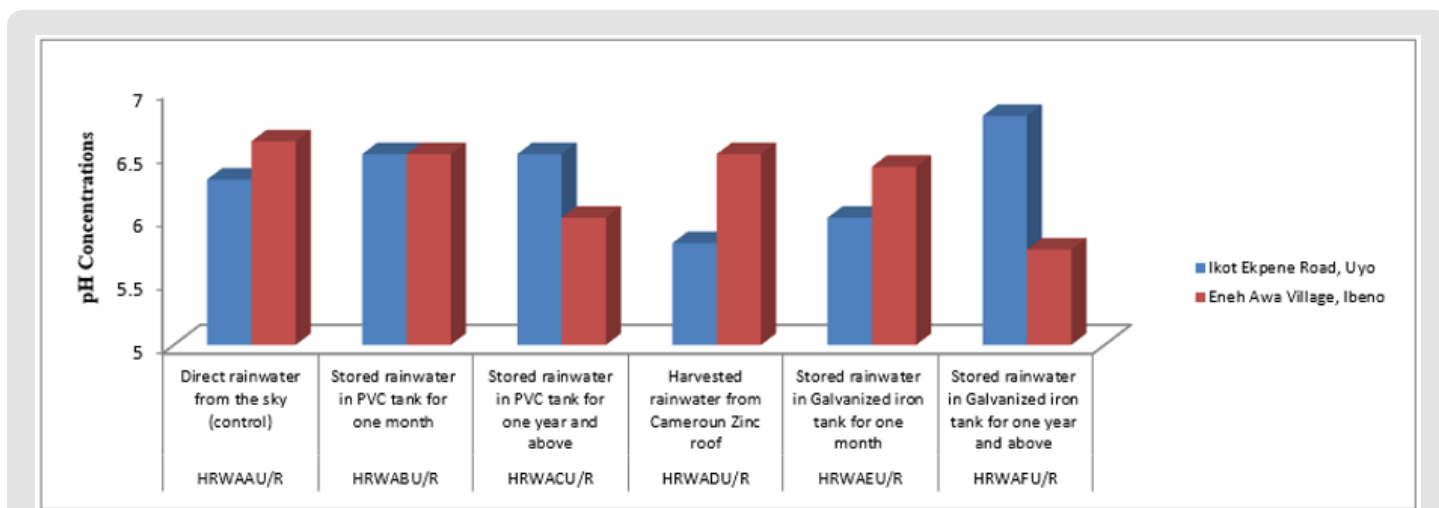


Figure 3: pH values of analyzed samples in Akwa Ibom State, Nigeria.

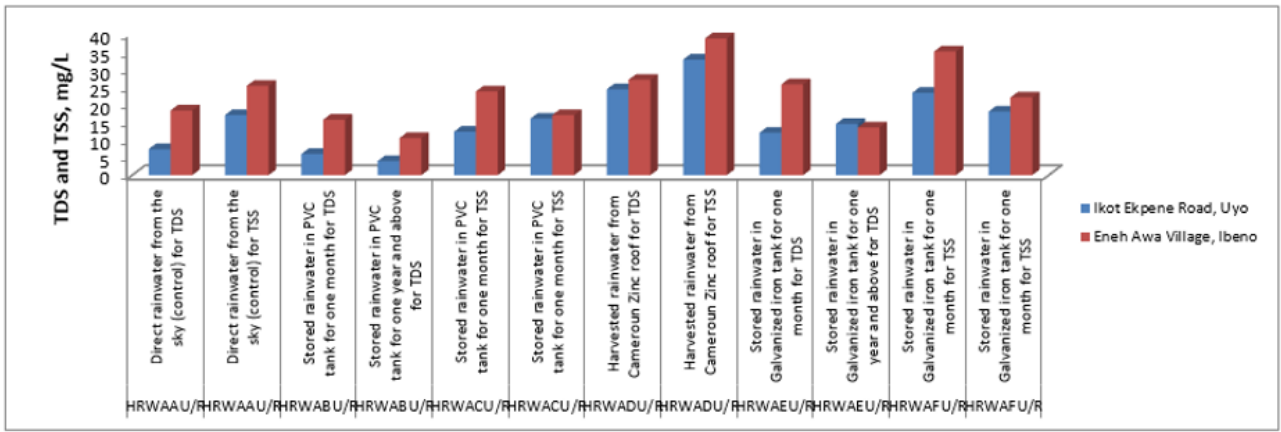


Figure 4: TDS and TSS, mg/L values of analyzed samples in Akwa Ibom State, Nigeria.

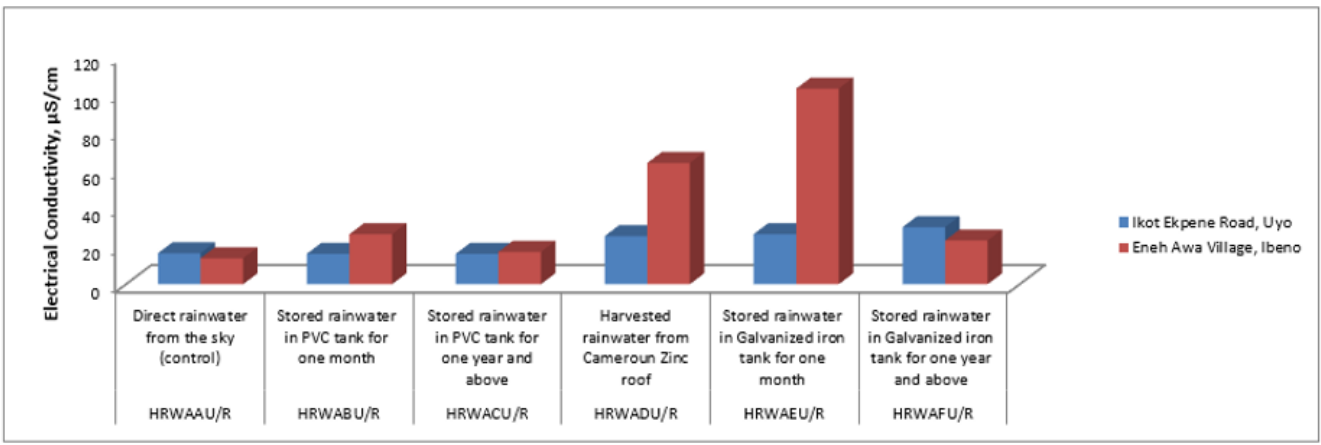


Figure 5: Electrical Conductivity (µS/cm) values of analyzed samples in Akwa Ibom State, Nigeria.

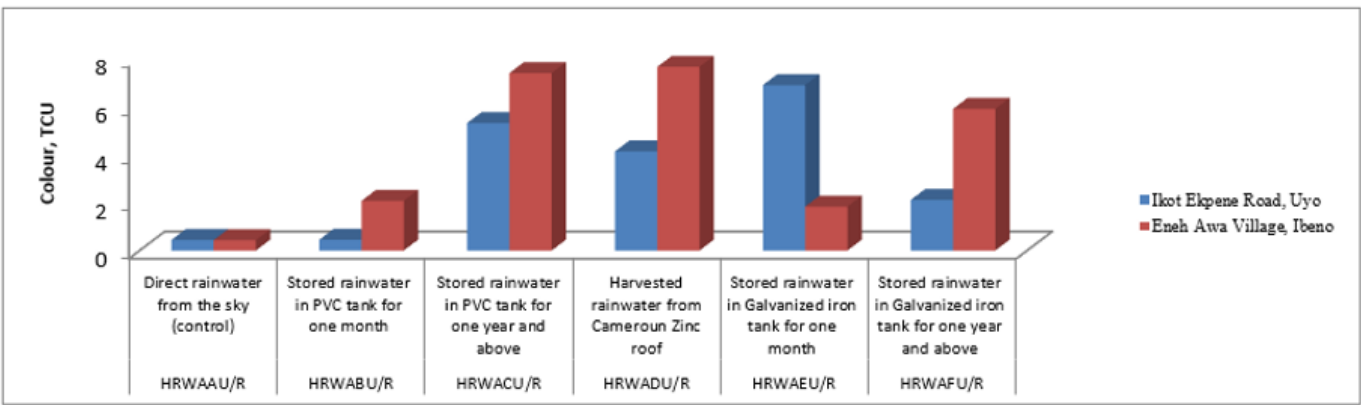


Figure 6: Colour (TCU) values of analyzed samples in Akwa Ibom State, Nigeria.

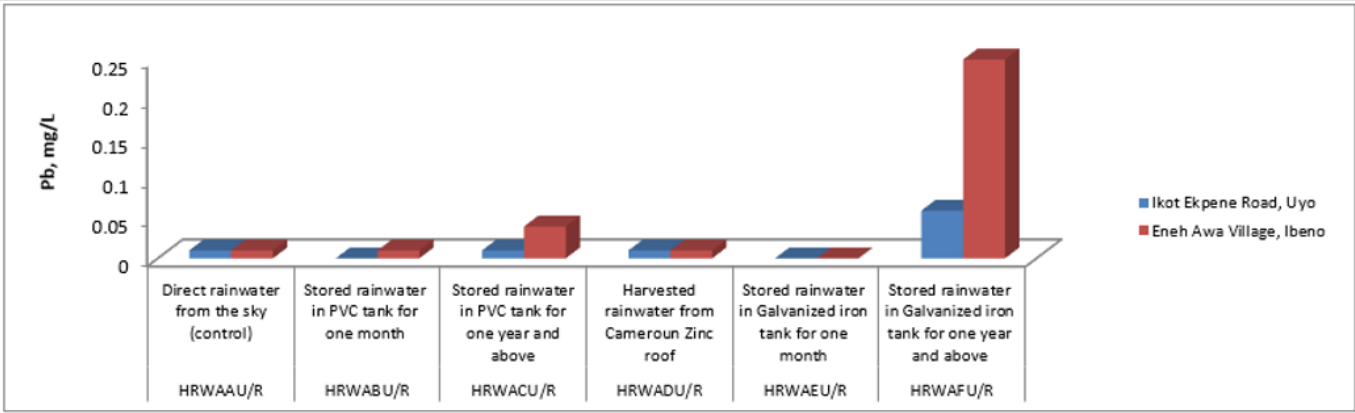


Figure 7: Pb (mg/L) values of analyzed samples in Akwa Ibom State, Nigeria.

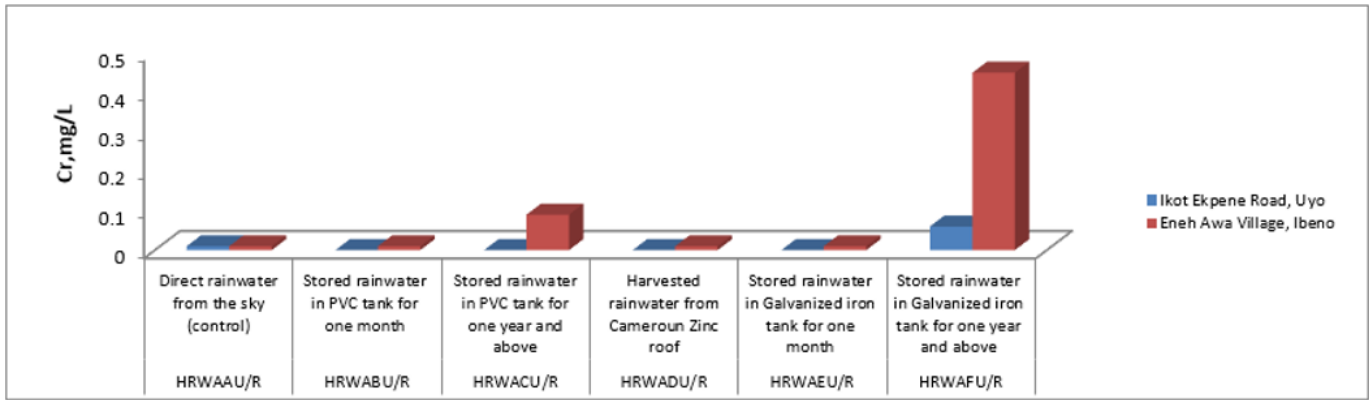


Figure 8: Cr (mg/L) values of analyzed samples in Akwa Ibom State, Nigeria.

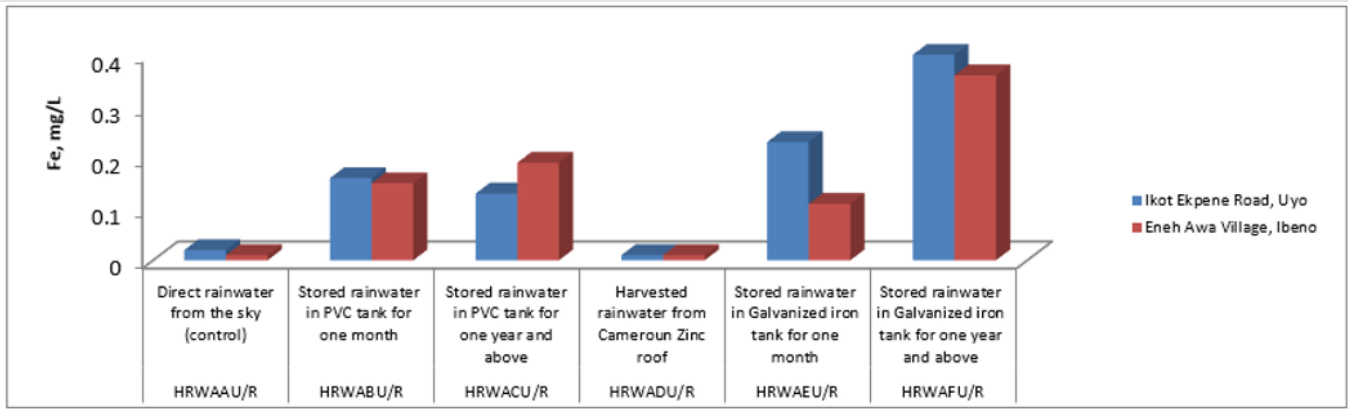


Figure 9: Fe (mg/L) values of analyzed samples in Akwa Ibom State, Nigeria.

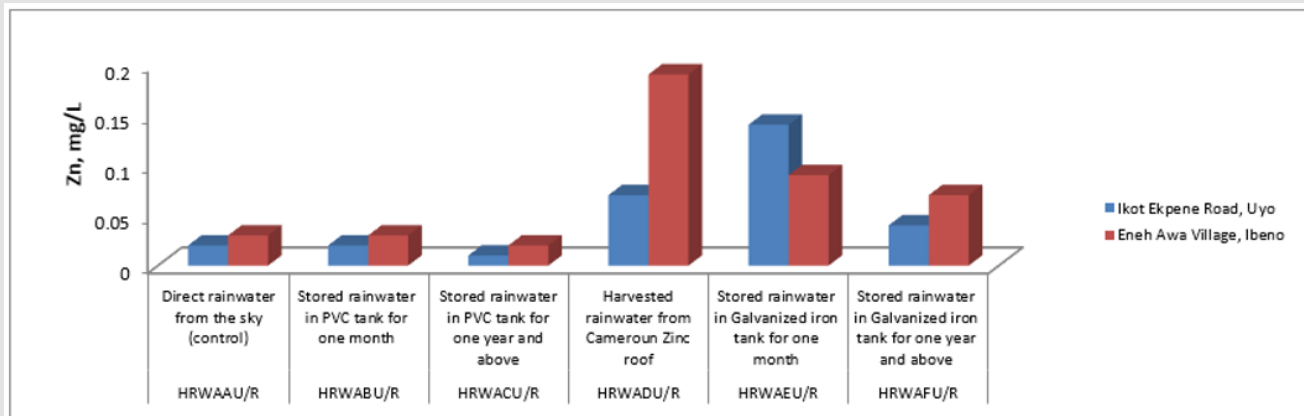


Figure 10: Zn (mg/L) values of analyzed samples in Akwa Ibom State, Nigeria.

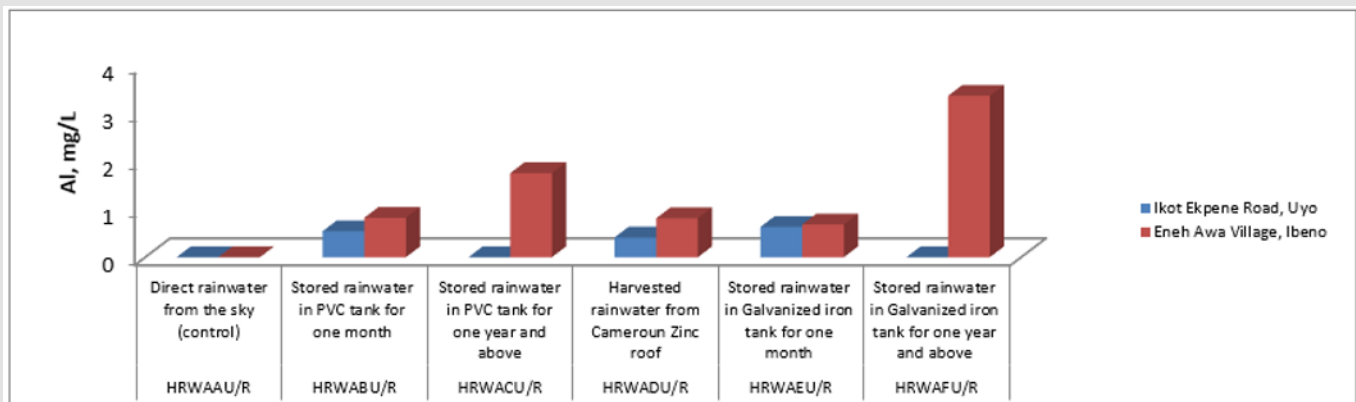


Figure 11: Al (mg/L) values of analyzed samples in Akwa Ibom State, Nigeria.

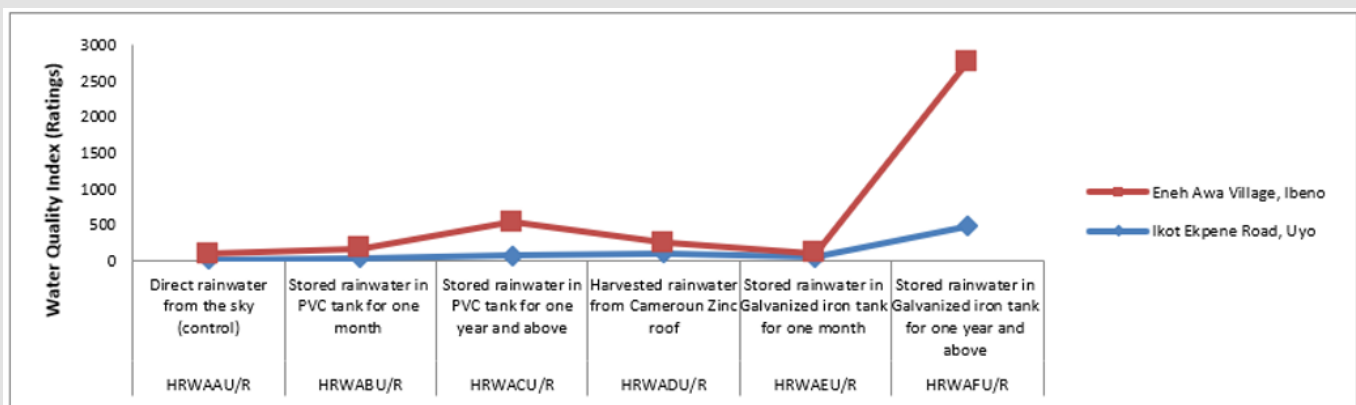


Figure 12: Water Quality Index (WQI) ratings of analyzed samples in Akwa Ibom State, Nigeria.

Table 3: Physico-chemical characteristics, heavy metals and *E. coli* of harvested rainwater directly from the sky and stored rainwater in PVC tank for one month and a year and above in Akwa Ibom State, Nigeria.

S/N	Parameters	Direct rainwater from the Sky (control)		Harvested rainwater stored in PVC tank for one month		Harvested rainwater stored in PVC tank for one year		WHO, 2011	USEPA, 2012	NSDWQ, 2015
		HRWAA _U	HRWAA _R	HRWAB _U	HRWAB _R	HRWAC _U	HRWAC _R			
1.	pH	6.30±0.20	6.60±0.30	6.50±0.20	6.50±0.30	6.50±0.14	6.00±0.35	6.5-8.5	6.5-8.5	6.5-8.5
2.	Temp., °C	21.60±1.15	22.30±0.57	21.30±1.53	20.60±1.15	23.50±0.71	25.00±1.41	30	Ambi-ent	Ambient
3.	TDS, mg/L	7.36±3.05	18.30±2.52	6.00±2.00	15.70±5.86	4.00±1.41	10.50±6.36	250	500	500
4.	TSS, mg/L	17.00±5.57	25.30±7.58	12.30±3.51	23.70±8.02	16.00±2.12	17.00±4.24	250	500	500
5.	Electrical Conductivity, $\mu\text{S}/\text{cm}$	16.30±2.52	13.67±2.52	16.00±2.65	26.30±3.05	12.50±3.54	13.50±4.95	1200	1000	1000
6.	Colour, TCU	0.44±0.01	0.43±0.01	0.45±0.04	2.07±1.84	5.31±0.73	7.39±1.30	5	15	15
7.	Turbidity, NTU	1.80±1.06	4.16±0.29	4.16±1.19	6.73±2.31	6.84±0.57	8.40±1.53	5	5	5
8.	Phosphate, mg/L	1.35±0.69	0.31±0.08	1.73±1.16	4.51±1.45	1.60±1.56	3.19±0.76	5	5	5
9.	Sulphate, mg/L	0.79±0.03	1.64±0.98	1.61±1.23	2.12±1.36	0.98±0.46	1.59±1.38	250	200	100
10.	Nitrate, mg/L	0.43±0.01	0.42±0.01	0.45±0.04	2.07±1.84	4.22±0.81	1.46±1.29	50	50	50
11.	<i>E. Coli</i> Count, cfu/mL	ND	ND	ND	ND	ND	ND	0	0	0
12.	Lead, mg/L	0.01±0.00	0.01±0.00	ND	0.01±0.00	0.01±0.01	0.04±0.02	0.01	0.01	0.01
13.	Chromium, mg/L	0.01±0.00	0.01±0.00	ND	0.01±0.00	ND	0.09±0.01	0.05	0.05	0.05
14.	Iron, mg/L	0.02±0.01	0.01±0.00	0.16±0.05	0.15±0.04	0.13±0.06	0.19±0.12	0.3	0.3	0.3
15.	Zinc, mg/L	0.02±0.00	0.03±0.00	0.02±0.00	0.03±0.00	0.01±0.00	0.02±0.01	3.0-5.0	3.0	3.0
16.	Aluminium, mg/L	ND	ND	0.54±0.04	0.82±0.06	ND	1.75±0.94	0.1-0.2	0.2	0.2

Table 4: Physico-chemical characteristics, heavy metals and *E. coli* of harvested rainwater from Cameroun Zinc roof, stored rainwater in Galvanized iron tank for one month and a year and above in Akwa Ibom State, Nigeria.

S/N	Parameters	Harvested rainwater from Cameroun Zinc Roof		Harvested rainwater stored in Galvanized iron tank for one month		Harvested rainwater stored in Galvanized iron tank for one year and above		WHO, 2011	USEPA, 2012	NSDWQ, 2015
		HRWAD _U	HRWAD _R	HRWAE _U	HRWAE _R	HRWAF _U	HRWAF _R			
1.	pH	5.80±0.10	6.50±0.20	6.00±0.20	6.40±0.30	6.80±0.21	5.75±0.21	6.5-8.5	6.5-8.5	6.5-8.5
2.	Temp., °C	27.30±0.57	21.60±2.08	24.30±2.52	26.60±1.53	26.00±2.83	24.50±0.71	30	Ambi-ent	Ambient
3.	TDS, mg/L	24.30±10.40	27.00±3.00	12.00±3.00	25.70±12.50	14.50±2.12	13.50±4.95	250	500	500
4.	TSS, mg/L	32.70±7.57	38.70±7.09	23.30±5.03	35.00±8.19	18.00±2.83	22.00±2.82	250	500	500
5.	Electrical Conductivity, $\mu\text{S}/\text{cm}$	25.30±4.51	63.70±6.51	26.30±4.04	102.70±24.60	30.50±4.95	23.00±4.24	1200	1000	1000
6.	Colour, TCU	4.13±2.50	7.65±2.38	6.89±2.78	1.83±1.51	2.11±0.04	5.90±0.17	5	15	15
7.	Turbidity, NTU	8.36±2.57	5.34±0.68	7.19±2.84	4.24±1.24	2.00±0.19	6.26±0.11	5	5	5
8.	Phosphate, mg/L	3.85±0.18	4.04±0.36	2.72±0.73	1.96±1.26	2.12±0.09	3.23±0.49	5	5	5
9.	Sulphate, mg/L	3.81±1.32	5.55±2.15	2.63±1.57	7.88±0.63	2.87±0.86	5.03±0.91	250	200	100
10.	Nitrate, mg/L	4.12±2.49	7.65±2.37	6.88±2.77	1.83±1.51	6.70±1.20	9.40±1.69	50	50	50

11.	<i>E. Coli</i> Count cfu/ mL	ND	ND	ND	ND	ND	ND	0	0	0
12.	Lead, mg/L	0.01±0.00	0.01±0.00	ND	ND	0.06±0.03	0.25±0.09	0.01	0.01	0.01
13.	Chromium, mg/L	ND	0.01±0.00	ND	0.01±0.00	0.06±0.002	0.45±0.08	0.05	0.05	0.05
14.	Iron, mg/L	0.01±0.01	0.01±0.00	0.23±0.07	0.11±0.03	0.40±0.11	0.36±0.09	0.3	0.3	0.3
15.	Zinc, mg/L	0.07±0.00	0.19±0.00	0.14± 0.00	0.09± 0.00	0.04±0.02	0.07±0.02	3.0-5.0	3.0	3.0
16.	Alumini- um, mg/L	0.41±0.04	0.81±0.06	0.63±0.06	0.68±0.07	ND	3.36±1.87	0.1-0.2	0.2	0.2

Table 5: Metals Concentrations of the Roof Type and Storage Vessels.

Concentration (mg/kg)							
S/N	Sample	Pb	Al	Fe	Zn	Cr	
1.	Cameroon zinc sheet	2.30	ND	63.78	5.94	1.02	
2.	Galvanized iron pan sheet	0.06	2.12	63.40	5.89	1.23	
3.	PVC Tank	ND	ND	ND	ND	ND	

Table 6: Air quality assessment records in Akwa Ibom State, Nigeria.

Parameters							
Areas	PM _{2.5} (µg/m³)	PM ₁₀ (µg/m³)	SO ₂ (ppm)	NO ₂ (ppm)	CO (ppm)	CO ₂ (ppm)	Temperature, °C
Urban	77.33±23.02	139.70±28.02	0.47±0.15	0.30±0.10	22.33±1.52	10,566.67±1650.35	29.50±0.10
Rural	72.67±8.08	117.67±10.97	0.57±0.21	0.43±0.15	28.33±1.52	24,466.70±2218.86	27.63±0.55
AQI Ratings							
Urban	Poor (D)	Very Poor (E)	Very Poor (E)	Very Poor (E)	Moderate (C)	Hazardous (F)	
Rural	Poor (D)	Poor (D)	Very Poor (E)	Very Poor (E)	Moderate (C)	Hazardous (F)	

Table 7: Water quality indices (WQI) of rainwater in storage vessels, Cameroun Zinc roof and directly from the sky (control) in Akwa Ibom State.

Sample code	Areas	Sample Source of Collection	Water Quality Index (WQI)	Quality Rating
HRWAA _U	Urban	Direct rainwater from the sky (control)	11.79	Excellent
HRWAA _R	Rural	Direct rainwater from the sky (control)	79.54	Very Poor
HRWAB _U	Urban	Harvested rainwater stored in PVC tank for one month	40.72	Good
HRWAB _R	Rural	Harvested rainwater stored in PVC tank for one month	137.03	Unfit for drinking
HRWAC _U	Urban	Harvested rainwater stored in PVC tank for one year and above	76.28	Very Poor
HRWAC _R	Rural	Harvested rainwater stored in PVC tank for one year and above	461.89	Unfit for drinking
HRWAD _U	Urban	Cameroun Zinc roof	106.26	Unfit for drinking
HRWAD _R	Rural	Cameroun Zinc roof	139.15	Unfit for drinking
HRWAE _U	Urban	Harvested rainwater stored in Galvanized iron tank for one month	48.93	Good
HRWAE _R	Rural	Harvested rainwater stored in Galvanized iron tank for one month	55.01	Poor
HRWAF _U	Urban	Harvested rainwater stored in Galvanized iron tank for one year and above	476.85	Unfit for drinking
HRWAF _R	Rural	Harvested rainwater stored in Galvanized iron tank for one year and above	2276.42	Unfit for drinking

Discussion

Physiochemical Parameters

pH levels of Harvested Rainwater: The level of pH recorded in control samples in the urban and rural areas of Akwa Ibom State were 6.30 ± 0.20 and 6.60 ± 0.30 respectively (Table 3 & Figure 3) showed slight acidity. All other analysed samples indicated acidity in the results which were in support with the air quality assessment record values ranging from 117.67 - 139.70 $\mu\text{g}/\text{m}^3$ for PM₁₀, 72.67 - 77.33 $\mu\text{g}/\text{m}^3$ for PM_{2.5} and other results (ppm) were: NO₂ (0.30-0.43), SO₂ (0.47-0.57) and CO (22.33-28.33) as shown in (Table 6) above and due to high rate of vehicular emissions and atmospheric pollutions and the dissolution of the particulate matter in the rainwater samples promote acidity. The obtained results in this study were also in agreement with the earlier findings of slight acidity (6.2-7.6) of harvested rainwater as reported by Emerole et al. [21] for harvested rainwater samples from Owerri, Imo State and Moses et al. [22] who also reported harvested rainwater to be slightly acidic in Uyo, Akwa Ibom State and that of Ojo [19] in Akure, Ondo State. pH values in this study were below and within the permissible limits of 6.5-8.5 for drinking water [47-49] as shown in Tables 4 & 5 above.

TDS and TSS levels of Harvested Rainwater

TDS and TSS levels gave mean values (mg/L) for control samples ranging from 7.36 ± 3.05 - 18.30 ± 2.52 and 17.00 ± 5.57 - 25.30 ± 7.58 respectively in Ikot Ekpene Road, Uyo L.G.A and in Eneh Awa Village, Ibeno L.G.A of Akwa Ibom State (Table 3 & Figure 4) above. Cameroun zinc roof rainwater gave mean values ranging from 24.30 ± 10.40 - 27.00 ± 3.00 mg/L for TDS and 32.70 ± 7.57 - 38.70 ± 7.09 mg/L (TSS) in both areas. Stored harvested rainwater in PVC tank for one month to one year and above gave mean values ranging from 4.00 ± 1.41 - 15.70 ± 5.86 mg/L for TDS in both areas. TSS showed mean values ranging from 12.30 ± 3.51 - 16.00 ± 2.12 mg/L in the urban area and 17.00 ± 4.24 - 23.70 ± 8.02 mg/L in the rural area as shown in (Tables 3 & 4). Findings by some researchers reported TDS and TSS for harvested rainwater from Oko in Anambra State and Maiduguri in Borno State having a range of 35.3-43.6 and 95.32-150.32 mg/L respectively by Dinrifo et al. [28] and Chukwuma et al. [37] which were higher than the values of the present discoveries and were within the permissible limits by WHO. Obtained values for TDS and TSS were all within the drinking water set standard of 250-500 mg/L [47-49]. The potability of water with a total dissolved solids (TDS) level of less than about 500 mg/L is generally acceptable to be excellent and suitable for drinking purposes [48].

Electrical Conductivity levels of Harvested Rainwater

The level of electrical conductivity recorded in control samples in Ikot Ekpene Road, Uyo L.G.A (urban) and in Eneh Awa village, Ibeno L.G.A (rural) of Akwa Ibom State were 16.30 ± 2.52 and 13.67 ± 2.52 $\mu\text{S}/\text{cm}$ respectively (Table 3 & Figure 5). Rainwater from Cameroun

zinc roof gave mean values ranging from 25.30 ± 4.51 - 63.70 ± 6.51 $\mu\text{S}/\text{cm}$ in both areas. Stored harvested rainwater in galvanized iron tanks for one month to one year and above had mean values of conductivity ranging from 26.30 - 30.50 $\mu\text{S}/\text{cm}$ (urban) and 23.00 - 102.70 $\mu\text{S}/\text{cm}$ (rural) whereas that of stored rainwater in PVC tank ranged from 12.50 - 16.00 $\mu\text{S}/\text{cm}$ (urban) and 13.50 - 26.30 $\mu\text{S}/\text{cm}$ (rural) which were of lower concentrations as seen in Tables 3-4 & Figure 5 above. The lower levels of conductivity found in harvested rainwater as stated by earlier researchers were in supports with the values obtained in this study. These results were in support of the findings by Nicholas et al. [1]; Ojo, [20]; Emerole et al. [21]; Waziri et al. [44]; Moses et al. [22] who reported lower concentrations of electrical conductivity in harvested rainwater samples from Delta and Enugu States; Ondo State; Imo State; Maiduguri and in Akwa Ibom State respectively. The values obtained were all below the set standards of 1000-1200 $\mu\text{S}/\text{cm}$ for drinking water [47-49].

Colour Levels of Harvested Rainwater

The level of colour recorded in control samples in both areas of Akwa Ibom State ranged from 0.43 ± 0.01 - 0.44 ± 0.01 TCU (Table 3 & Figure 6). Rainwater in PVC tank had values ranging from 0.45 - 5.31 TCU for colour in the urban area and 2.07 - 7.49 TCU in the rural area while that of galvanized iron tank ranged from 2.11 - 6.89 TCU for colour in the urban and 1.83 - 5.90 TCU in the rural area as shown in Tables 3 & 4. The high levels of colour seen in the stored harvested rainwater are caused by micro-pollutants, dust particles and vehicular emissions from these areas. The colour values of control samples, stored rainwater in PVC tank for one month were within the set standard of 5 TCU (colour) for drinking water [48] while the values for stored rainwater in PVC tank for one year and above in both areas and that of stored rainwater in galvanized iron tanks for a month giving mean values of 6.89 TCU (urban) and 5.90 TCU (rural) were above the standard of 5-15 TCU for colour in drinking water [47-49].

Nitrate levels of Harvested Rainwater

The level of nitrate recorded in control samples in both areas of Akwa Ibom State were 0.43 ± 0.01 and 0.42 ± 0.01 mg/L respectively (Table 3). Cameroun zinc roof rainwater gave mean values ranging from 4.12 ± 2.49 - 7.65 ± 2.37 mg/L in both areas. However, stored harvested rainwater in galvanized iron tank collected from Cameroon zinc roof had mean nitrate values ranging from 6.70 - 6.88 mg/L in Ikot Ekpene Road, Uyo (urban) and 1.83 - 9.40 mg/L in Eneh Awa village, Ibeno (rural) while PVC tank had 0.45 - 4.22 mg/L in both areas as seen in Tables 3 & 4. The air quality assessment record of the urban and rural areas had values ranging from 0.30 - 0.43 ppm for NO₂ as shown in Table 6 above. Comparing the values of the air quality assessment record with the results of the rainwater samples, significant difference ($p < 0.05$) was seen and it was at where Exxon Mobil Corporation oil producing industry and Ibeno beach resort centre in situated, which as a result of atmospheric/anthropogenic pollution, flared

gases and vehicular emissions in the environments that settled at the rooftop and later transferred in the harvested rainwater on storage duration. The values obtained were within the set standard of 15-50 mg/L for drinking water [47-49].

Sulphate Levels of Harvested Rainwater

The mean sulphate concentrations recorded in control samples in both areas of Akwa Ibom State were 0.79 ± 0.03 and 1.64 ± 0.98 mg/L respectively (Table 3). Cameroon zinc roof rainwater gave mean values ranging from 3.81 ± 1.32 - 5.55 ± 2.15 mg/L in both areas. Stored rainwater in PVC tanks had mean sulphate values ranging from 0.98 ± 0.46 - 1.61 ± 1.23 mg/L (urban) and 1.59 ± 1.38 - 2.12 ± 0.96 mg/L (rural) while galvanized iron tanks had values ranging from 2.63 ± 1.57 - 2.87 ± 0.86 mg/L (urban) and 5.03 ± 0.91 - 7.88 ± 0.63 mg/L (rural) as seen in Tables 2 & 3. The air quality assessment of the urban and rural areas had values ranging from 0.47-0.57 ppm for SO₂ as shown in Table 5 above. These results were in support with the findings of Nicholas and Ukoha, [2,5] who reported lower values of sulphate in Rivers State and in Imo State, Nigeria but not in agreement with the values of 484.81 ± 115.02 mg/L for sulphate in rainwater as reported by Emerole et al. [21] in Owerri, Imo State, Nigeria. Comparing the air quality assessment values with that of the rainwater samples, it was seen that, there was influence on the rainwater quality and this is likely due to less organic micro-pollutants present in water which originated mainly from atmospheric pollution and exhaust gas from vehicles which had minimal effect on the samples. Sulphate mean values obtained were all within the set standard of 100-250 mg/L for drinking water [48-49].

Microbiological Parameters

E. Coli levels of the Analyzed Harvested Rainwater

Mean values of *E. coli* ranged from 0.00-0.00 cfu/mL as shown in Table 3 above, indicating that *E. coli* was not present in the analysed samples. Result obtained was not in line with the earlier findings by Nicholas et al. [1] who gave the values of 1.00 ± 0.00 cfu/mL for *E. coli* in rainwater samples in Delta State, Nigeria. The values obtained for coliform level in this study were within the set standard of 0 cfu/mL for drinking water [47-49].

Air Quality Assessment Record

The air quality assessment in Ikot Ekpene road, Uyo L.G.A and in Eneh Awa village, Ibeno L.G.A had mean values ranging from 117.67-139.70 µg/m³ for PM₁₀, 72.67-77.33 µg/m³ for PM_{2.5}, Temperature (27.63-29.50 °C) and other results (ppm) were: NO₂ (0.30-0.43), SO₂ (0.47-0.57), CO₂ (10,566.67±1650.50-24,466.70±2218.86) and CO (22.33-28.33) as shown in Table 6 above. These results were in agreement with the findings of Nicholas and Ukoha, [32,34] on air quality assessments in the urban and rural areas of Delta and Rivers States in Nigeria. The values obtained for air quality assessment were within and above the set standard by USEPA [62-64].

Heavy Metals Concentrations of the Harvested Rainwater

Heavy metals levels of control samples gave no detection of Al and very insignificant quantities (mg/L) of Pb and Cr. Fe had mean values between 0.16 and 0.15 mg/L in both areas of Akwa Ibom State and Zn had mean values between 0.02 and 0.03 mg/L respectively (Tables 3 & 4 & Figures 7-11). Roof run-offs of Cameroon zinc showed very low quantities of 0.01 mg/L for Fe, Pb in both areas and Cr (rural) whereas Cr was not found in Ikot Ekpene Road samples. The air quality assessment records of the urban and rural areas had values ranging from 117.67-139.70 µg/m³ for PM₁₀, 72.67-77.33 µg/m³ (PM_{2.5}) and CO (22.33-28.33 ppm) as shown in Table 7 above. Nicholas and Ukoha, [2] reported in their study that, there was no detection of Pb, Cr and Al in the analyzed direct harvested rainwater from the sky (control) while Fe gave mean values ranging from 0.01 ± 0.00 - 0.02 ± 0.01 mg/L and 0.01 ± 0.00 - 0.02 ± 0.00 mg/L for Zn in the urban and rural areas of Rivers State and these results were not in supports with the values in this study. The results of heavy metals in galvanized iron tank for a year and above showed higher concentrations (mg/L) of 0.40 ± 0.11 (urban) and 0.36 ± 0.09 (rural) for Fe, 0.06 ± 0.03 (urban) and 0.25 ± 0.09 (rural) for Pb, 0.06 ± 0.02 (urban) and 0.45 ± 0.08 (rural) for Cr and 3.36 ± 1.87 for Al (rural).

However, Cameroon zinc roof harvested rainwater gave higher values of 0.07 ± 0.00 - 0.19 ± 0.00 (Zn) and 0.41 ± 0.04 - 0.81 ± 0.06 (Al) and stored rainwater in PVC tank for a year and above also gave higher values (mg/L) of 0.04 ± 0.02 (rural) for Pb, 1.75 ± 0.94 (rural) for Al and 0.09 ± 0.01 (rural) for Cr. The metal concentrations of the digested roof and storage vessels gave heavy metals values ranging from 0.06-2.30 for Pb, 0-00-2.12 (Al), 63.40-63.78 (Fe), 5.89-5.94 (Zn) and 1.02-1.23 (Cr) as shown in Table 4 above. Findings by Nicholas and Ukoha, [2] also reported mean values (mg/L) of heavy metals ranging from 0.18 ± 0.11 - 0.22 ± 0.17 for Fe, Cr (0.04 ± 0.02 - 0.12 ± 0.04), Zn (0.01 ± 0.01 - 0.03 ± 0.01) and Pb (0.09 ± 0.01 - 0.23 ± 0.06) and Al (0.10 ± 0.06 - 1.61 ± 0.68) for stored rainwater in galvanized iron tank for one year and above in the urban (Nwaja, Trans Amadi Ind. Layout, P/H L.G.A) and Umuazu village, Igbo-Etche, Etche L.G.A (rural) areas of Rivers State respectively and these results were in agreement with the values obtained in this study which are of higher concentrations. Earlier study by Okudo et al. [7] also reported high values of Pb (Emene: 0.58 ± 0.11 and Iva Valley: 0.48 ± 0.04) and Cr (Emene: 0.10 ± 0.02) in rainwater samples from Enugu State and that of Nanji et al. (2023) who also reported higher concentrations of Cr in the road run-offs harvested rainwater in Nsukka, Enugu State, Nigeria. Emerole et al. [21] who reported higher values of heavy metals in direct rainwater and roofs run-offs harvested rainwater (mg/L) of Fe 2.12 ± 1.17 , Al 1.70 ± 1.83 and Pb 0.44 ± 0.36 from Owerri, Imo State were above the permissible limits [47-49] and were not in agreement with the control samples values obtained in this study.

Comparing the rainwater values with that of the digested roof / storage vessels (Table 5) and the air quality assessment record (Table 6) above, there was significant difference ($p < 0.05$) in the values of the control samples for (Fe) and in stored rainwater in galvanized iron tank for one year and above (Fe, Zn and Al) which was as a result of atmospheric pollution and heavy metals fragments deposition in the form of particulate matter. The rust on roof tops is likely the reason for increase of metal burden, but the effect will be minimal, if the roof tops are properly washed by heavy rain before collecting of harvested rainwater for domestic use and it also shows that, the roof components and storage vessels (Tables 3-5) may leach into the rainwater in the presence of acid rain. This shows that the accumulative effect of the metals on standing as dust particles, flared gases, atmospheric pollution, vehicular emissions, silt, debris and leaching of the storage vessels will impact on the harvested rainwater quality. Though, PVC tank has no metal component as shown in the results of the digested analysed sample (Table 5) above, there was an indication of heavy metals present in the stored rainwater in PVC tank which was acquired from the rooftop run-off on collected and accumulation of particles on storage and also from associated contaminants. This can be eliminated by allowing heavy rain to wash the roof at least five times before harvesting the rainwater for domestic usages. The PVC tank should be properly covered to keep dust particles from settlements in the tank as they could lead to various levels of metals [1,2,22]. The obtained values in this study were within and above the permissible limits of 0.30 mg/L (Fe), 0.01 mg/L (Pb), 0.05 mg/L (Cr), 3.0-5.0 mg/L (Zn) and 0.1-0.2 mg/L (Al) for drinking water [47-49].

Water Quality Index (WQI) Ratings

Comparing the analysed rainwater samples with the NSDWQ, WHO and ICMR [30,47,48] set standards, the control samples (urban) gave values of 11.79 (Excellent water quality) and 79.54 (Very Poor water quality) in the rural area. Stored rainwater samples in galvanized iron tank for one month gave the values of 48.93 (Good water quality) and 55.01 (Poor water quality) in both areas respectively while stored rainwater for one year and above in galvanized iron tank gave values of 476.85 and 2,276.42 (Unfit for drinking quality) in both areas. Cameroun zinc roof rainwater gave values ranging from 106.26-139.15 indicating unfit for drinking water quality in both areas. PVC tank stored rainwater for one month gave values of 40.72 (Good water quality) and 137.03 (unsuitable for drinking water quality) in Ikot Ekpene Road, Uyo and in Eneh Awa village, Ibeno respectively whereas stored rainwater in PVC tank for one year and above gave 76.28 (Very Poor water quality) and 461.89 (Unfit for drinking water quality) in both areas (Table 6 & Figure 12).

Conclusion

Based on the results obtained, this present study revealed the leaching tendencies of heavy metals (Fe, Pb, Cr, Zn and Al) and effect of air quality and traffic emissions on harvested rainwater quality in Akwa Ibom State, South-South, Nigeria. This research work showed

the extent of which the metal components of the roof type/storage tanks and the dissolved metals could corrode or leach into the harvested rainwater and are as follows; Fe, Al, Zn, Cr and Pb (metal tank > Cameroun Zinc roof > PVC tank). The values obtained in Ikot Ekpene Road, Uyo and in Eneh Awa village, Ibeno for PM_{2.5}, PM₁₀, NO₂, SO₂, CO and CO₂ gave moderate to hazardous air quality by AQI ratings. Rainwater from Cameroun zinc roof, direct samples (control) and stored rainwater in PVC and Galvanized iron tanks showed excellent water quality in location with less industrial activities and very poor to unfit for drinking water quality in the location with heavy industrial activities by the WQI ratings. It was concluded that, rainwater should be treated before being served for domestic and potable usages because when the obtained results of the rainwater were compared with the values of the different digested roof types, galvanized iron and PVC tanks and also with the result of the air quality assessment records, it was clearly shown that the industrial activities, air quality, traffic emissions and environmental pollution can affect the quality of the rainwater and thereby having a bad effects on the health of humans.

Recommendations

The following suggestions are recommended;

- i. There should be an adoption of proper environmental monitoring workforce in these locations in order to control the atmospheric pollution and anthropogenic emissions.
- ii. Rainwater collected for possible domestic usage should be stored in plastic PVC tanks to avoid negative influence on the rainwater quality.
- iii. Rainwater collected from metal roofs run-off should be done with caution since the metal components of the roof type could corrode and leach into the harvested rainwater thereby altering the rainwater quality resulting from acidic rainfall due to traffic emissions and industrial activities of the location, most especially at where oil and gas companies are situated.

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Competing Interests

The authors declare that they have no known competing personal relationships or financial interests that could affect the study reported or presented in this paper.

Availability of Data and Materials

Anywhere it is applicable, data will be made available on request and the reference section contains all the irrelevant citations.

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