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Article

Source Characterization of Metals in Rainwater: Case Study of Akure, Ondo State, Nigeria

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Abstract: One source of water is rainwater. It is a pure solvent if not polluted. It has diverse functions in humans, animals and other materials. For rainwater not to be polluted, it means the values of cations, anions and particulate matter should be below water permissible limits. In this paper, we have characterized metals in rainwater harvested in Akure, Ondo State, Nigeria using standard methods of analyses. The physico-chemical parameters and metals were below WHO water guidelines. The variation of the metals was as follows: Ca>K>Na>Mg>Zn>Fe>Cu>Pb>Cr. Cd was absent and the Pb content was low. Principal Component Analysis showed that factors 1, 2 and 3 showed high loadings for Cr and Zn; Pb and Ca; and Cu, Mn and Mg respectively. Sources of these metals were due to anthropogenic activities.

Keywords: Rainwater, harvest, trace metals, permissible limits, PCA, anthropogenic sources.

1. Introduction

Water has diverse functions in humans, animals, plants and other substances. Without water it is impossible for life to thrive. One of the sources of water is rainwater. It is highly pure if not contaminated by pollutants (Mehta, 2011). According to Cerqueira *et al.*, (2014) the chemical component of water is depicted by the environment where it falls. This statement was confirmed by Akoto *et al.*, (2011).

Trace metals are abound in the environment so also rainwater, According to WHO report of 2002, almost 1.1 billion consumers drink unsafe water and this has resulted into different ailments (WHO, 2002). When water (rainwater) has values below permissible limit of trace metals the water is considered safe. However, it is always advisable to constantly monitor. Thus, it calls for appropriate interventions by stakeholders to constantly put up awareness programmes and improve all existing infrastructures. If all these are put in place, there is the possibility of reducing the potential health risk of the consumers (Yasin *et al*., 2015).

Akure is the capital of Ondo State, Nigeria it is a fast growing city. The population, housing, transportation and industries are on the increase. No doubt the possibility of environmental pollutants would be on the increase too. Researches on water samples have been on going in Akure (Akinnusotu *et al*., 2015) most of the results dealt with the physico-chemical parameters and trace metals, but none dealt with finding the sources of the pollutants in the area (source apportionment).

In this present work, we studied the physico-chemical properties, metal concentrations, identified and apportioned the emission sources.

2. Materials and Methods

2.1. The Study Area

2.1.1. Ondo State

Ondo State, the sunshine state is one of the states in Nigeria created in February 1976. According to 2006 census a total of 1, 745, 057 and 1,715,820 were recorded for men and women respectively. The state has a land size of 15,195.177 km^2 and 18 Local Government Areas (NPC, 2010). The state is Located in the South West Geo-Political Zone of the country. The mineral resources the state is endowed with are large deposit of bitumen and other mineral resources such as marble gold, gemstone, clay, diorite and lignite. Some of the tourist attractions in the state include the Idanre Hills, Owo Museum, Olumirin Water Falls, Ebomi Lake and Oke Marie Hills. The people of Ondo State are lovers of traditional arts and crafts especially ivory carvings and bronze works (NPC, 2010).

2.1.2. Akure

Akure is regarded as the largest city in Ondo State and it is the capital of the state. It is situated in south-western part of Nigeria. The city has a population of 421,100. The people are of the Yoruba ethnic group. The geographical coordinates of Akure, Ondo, Nigeria are 7° 15' 0" North, 5° 12' 0" East at an elevation/altitude of meters. The average elevation of Akure, Nigeria is 353 meters. The time zone for Akure is Africa/Lagos (GeoNames Geographical Database, 2012). Tradition has it that Akure was founded by Omoremi Omoluabi, a grandson of the Emperor Oduduwa. The title of Akure king is known as the Deji of Akure and is supported by six (6) high chiefs (Iwarefa) in his or her domain. The city of Akure is fast growing. According to the National population census, it has a total of 421,000 people (NPC, 2010). This town is the trade center of cash crops, has radio and television stations, federal and state primary, secondary and tertiary institutions, shopping activities, two stadia and other recreational and tourist attractions. The Christianity, Islam and Traditional or Totemistic worshipers live in peace with one accord.

2.2. Research Design

A number of different analytical approaches have been applied in air quality management in order to establish the overall levels of pollution in the air. To achieve the research objectives, these methodologies were used.

2.3. Sampling Area

The area is Akure, Ondo State, Nigeria (Fig 1). The sites were divided into eleven (11) locations comprising of forty (40) sites (Table 1)**.**These sites represented different industrial, urban and rural settings. Samples were collected for a period of one month (August, 2015). At the end of the sampling period 40 samples were collected. The location (Longitude and Latitude) of the sampling sites were determined using GPS Map 76CSX (Garmin Ltd, Taiwan).

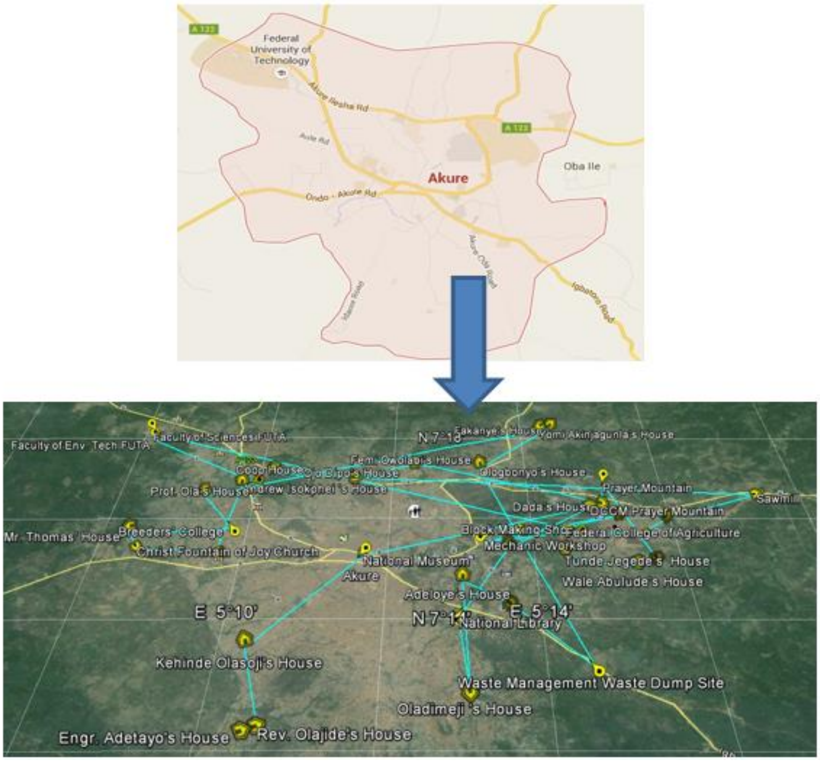

Fig. 1: Sampling locations

Table 1: Sample locations

Sites No	Location	No of Samples in each location	Description of Sites
1 - 3	Oba Ile Estate	3	Residential
4 - 9	Expressway Area	7	Mountain Top, Residential, Traffic, Farm Settlement, Mechanic Workshop, Abattoir, Industry, Quarry.
11 – 12	Owode Area	2	Sawmill, Traffic, Residential.
13 – 18, 40	Oba Ile Area	6	Residential, Market, Traffic, Welding, Mechanic Workshop, Construction of Road, Petrol Station, Block Industry.
19	Federal College of Agriculture	1	Residential, Traffic, Road Construction, Farming Activities
20 – 23, 10	Federal University of Technology Area	4	Residential, Industry, MTN Mast, Traffic, Petrol Station, School of Science (Laboratories), School of Environmental Technology.
24 – 26	Idanre Road Area	3	Residential, Traffic
27 - 31	Ondo Road Area	5	Sawmill, Traffic, Incinerator, Residential, College Compound, Mechanic Workshop
32 – 34	Igoba Road Area	3	Residential, Traffic
35 - 37	Igbatoro Road Area	3	Incinerator, Waste Dump, Traffic, Residential
38 - 39	Oda Road Area	2	Residential, Traffic

2.4. Sample Collection

The rain water samples were collected once a month, using appropriate sampler (Fig 2). A simple system made with a high-density polyethylene (HDPE) bottle (5L) connected to a HDPE funnel. The container was placed on sampling stand at a height above 1.5 m ground in order to prevent lichen forming during the sampling period. After rainwater samples were collected, they were filtered using Whatman ashless filter paper (11.0cm, Cat. No. 14442 110).

The prepared samples were subjected to appropriate determinations using standard methods of analyses.

Fig. 2: Sampler for the rainwater samples

2.5. Analyses

2.5.1. Physico-chemical properties

Total Dissolved Solids (TDS), temperature and Electrical Conductivity (EC) were determined in the samples using a 3 in 1 tester (EZ – 1 TDS & EC, China). pH was measured with a pen type pH meter (pH – 009 (1) CE, ROHS, China). All these parameters were taken at the sampling sites using manufacturers' specifications and methods. The free CO_2 was determined using the methods of Limgis (2001). The determination was done immediately on arrival to the laboratory. All chemical evaluations were performed within 12 hours after sampling.

2.5.2. Metals

The samples were subjected to wet ashing using concentrated acid (APHA, 1998). Standard methods of analyses were employed. The instrumentation were done with AAS Buck Scientific 210 VGP (Cd, Pb, Cr, Ni, Cu, Co, Fe, Mn, Zn, Mg) and Flame Photometer FP902 (Na, Ca, K).

2.6. Statistical Analyses

Basic descriptive statistics (mean, std error, error of mean (SE), std deviation (SD), variance, coefficient of variation, minimum, maximum and skewness), Factor Analysis (FA) and Principal Component Analysis (PCA) were done with Minitab version 16 software.

3. Results and Discussion

3.1. TDS

The range in mg/L was between 2.0 and 24.0 with a mean of 7 and coefficient of variation in percent 71.65(Table 2). This showed that the TDS in the samples were highly varied. The results obtained in this report were in agreement with those reported for rainwater in Warri, Nigeria (19.37 – 33.38mg/L, Olowoyo, 2011). Also these results followed the same trend with the results (6.8 – 24.1mg/L) recorded for some rainwater harvested in Ghana (Akoto *et al*., 2011). According to Chughtai *et al*., (2014), the higher the value of TDS in a water sample, the more the suspended and dust particles in it. The WHO (2006) guideline for water standard is 500mg/L, when we compared our results with WHO standard it showed that the rainwater obtained for this work were of high quality.

Table 2: Physico-chemical Properties of rainwater samples

	TDS (mg/L)	Temp (^oC)	pH	EC (μS/cm)	Free CO_2 (mg/L)
Mean	7.00	27.67	6.26	13.79	35.46
SE Mean	0.80	0.10	0.12	1.57	7.77
Std Deviation	5.02	0.62	0.72	9.83	48.51
Variance	25.16	0.39	0.52	96.64	52.99
Coeff. Variation	71.65	2.25	11.46	71.26	136.79
Minimum	2.00	27.00	4.30	4.00	12.00
Maximum	24.00	29.00	7.10	48.00	56.00

3.2. Temperature

Low variability (0.3.386 variance; 2.25% coefficient of variation) were recorded. For the results. The minimum value was 27 and maximum 29^oC. High values were recorded in the dry periods. This was expected because of the high relative humidity. The high temperature in this work was due to high solar radiation. As usual, the temperature here was in consonant with values from India (27^oC, Umerfaruq and Solanki, 2015), Ethiopia (22.79 – 24.53^oC, Yasin *et al*., 2015) and Nigeria (25.00 – 28.22^oC, Waziri *et al*., 2012). The values made available here were in conformity with WHO standard (55 – 50^oC). The temperature of water including rainwater depends on season, geographical location and sampling periods

(Venkatesharaju *et al*., 2010). It affects rates of chemical reactions in the water body, increases taste and colour and reduces solubility of gases in water.

3.3. pH

The pH of samples ranged between 4.3 and 7.1 with a variance of 0.515. This showed that the variation is minimal. About 25% of the values were slightly acidic, while about 75% were acidic in nature. The sites showed no significant variation in the pH values. Greater percent of the pH values fell short of the WHO standards of 6.5 – 9.5. The reason might be due to the washing of atmospheric pollutants into the containers for sampling.

3.4. Electrical Conductivity

Table 2 depicted the mean value of 13.79μS/cm. The values were highly varied (96.64). The values ranged between 4.0 and 48.0 μS/cm. These results were far less than 1131 - 1278 μS/cm recorded for waste waters in Pakistan. The differences may have been caused by the use of pesticides, insecticides and fertilizer in the agricultural purposes in the surrounding areas (Mastoi *et al*., 2014). Conductivity is expressed as the ability of water to conduct an electric current. Electrolytes in a solution dissociate into positive (cations) and negative (anions) and impact conductivity (Limgis, 2001).

3.5. Free Carbon-Dioxide (CO_2)

The mean value for free $CO_2$was highest at sample no 34 with 56mg/L while the lowest was 12mg/L. All these were residential areas. The free CO_2 when dissolved in rainwater contributes to the harness of water or rainwater. Free CO_2reacts with water partly to form calcium bicarbonate and in the absence of bicarbonate gets converted to carbonate releasing carbon-dioxide. There were not much differences in the values obtained in all the sites.

3.6. Metals

Figs 2 and 3 depicted the pie and histogram charts of the metal variables. The charts showed the variations in the following order: Ca>K>Mg>Zn>Mn>Fe>Cu>Pb>Cr. In all expect Mg there were high variations in the results obtained for the metals. The values of Na, Ca, K were higher than those of the trace metals. This is a normal phenomenon. All the macro metals in this work were within WHO permissible limits for drinking water. It is gratifying to note the content of Pb was low. This is mainly due to the removal of lead-gasoline vehicles from the roads due to improved technology in manufacturing of engines and treatment of petroleum products. The overall result is evident that the rainwater and environment were not polluted. In general, our results were in good agreement with results of Akoto *et al*., (2011) and Al-Momani (2003) in analyzed rainwater samples.

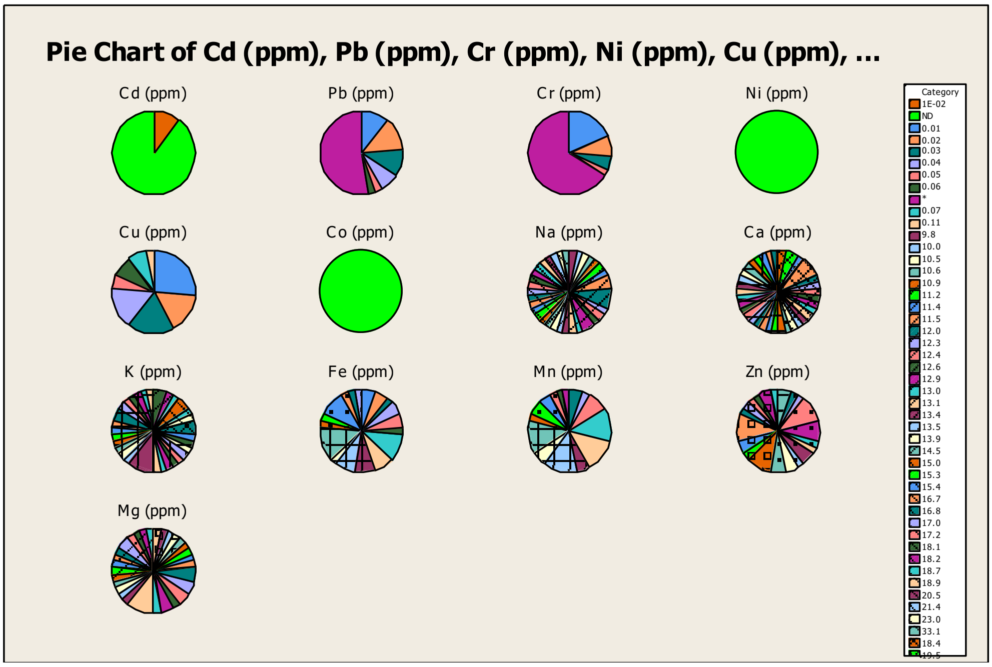

Fig. 2: Pie Chart of the metal variables

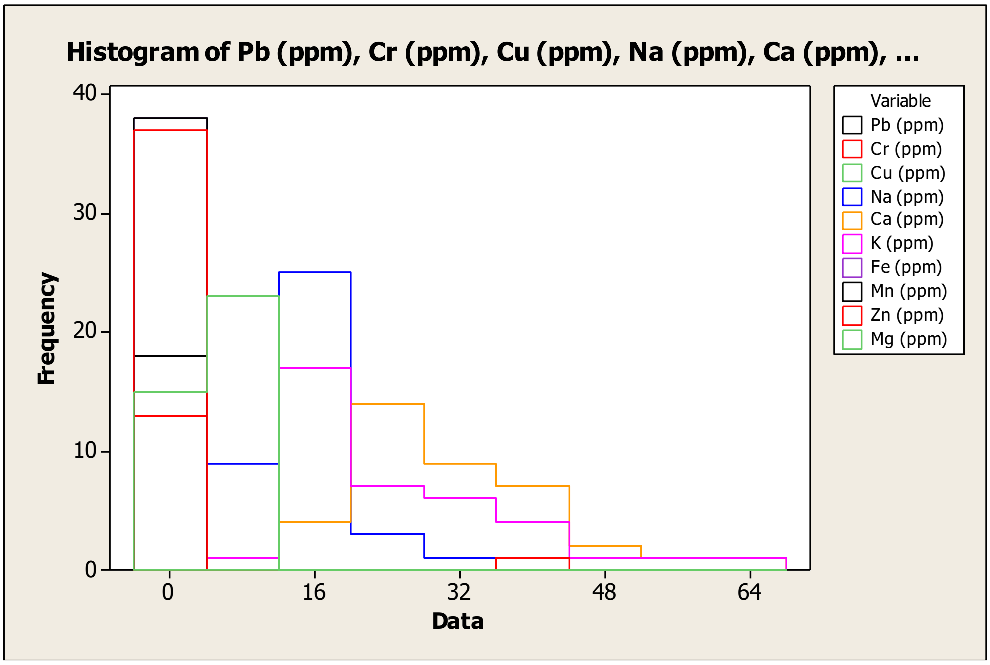

Fig. 3: Histogram of the metal variables

According to Cerqueira *et al*., (2014), when metals are present in soluble form, trace metals are made available. Rates of hydrolysis of trace metals in rainwater depend on the balance of anion and cation, pH of water, size distribution, the redox potential and chemical nature of the depositing particles (Morselli *et al*., 2004).

The high toxicity of some trace heavy metals have been used as index of pollution (Omoigberale and Ogbeibu, 2005). Table 3 depicted the correlation matrix of the metals. Although positive correlations were recorded by all metals, but Ni and Co were not detectable (ND), 10% of the samples had Cd (0.01mg/L) values. The results showed strong correlations between Cu:Pb, Cu:Cr, Ca:Cr, Zn:Cu, Zn:Mn, Mg:Mn and Mg:Zn others showed low and weak correlations.

Table 3: Correlation matrix of the metals

	Pb	Cr	Cu	Na	Ca	K	Fe	Mn	Zn
Cr	0.180								
Cu	**0.863**	**0.804**							
Na	0.239	0.631	0.310						
Ca	**0.793**	**0.845**	**0.794**	0.000					
K	0.068	0.053	0.372	0.010	0.019				
Fe	0.105	0.265	0.118	0.281	**0.731**	**0.770**			
Mn	0.492	0.060	0.056	0.430	0.505	**0.790**	0.111		
Zn	0.386	0.500	**0.869**	0.530	**0.769**	0.635	0.084	**0.817**	
Mg	0.620	0.672	0.574	0.005	0.0.009	0.060	0.417	**0.965**	**0.953**

3.7. Principal Component Analysis (PCA)

PCA with varimax rotation was utilized and presented in Table 4. The factor loading for PCA analysis of the metals were shown in this table. Factor loading above 0.1 have been shown in the table, while values above 0.5 were bold. Eigenvalues greater than 0.9 was employed to identify major metals associated with the different sources.

Table 4: PCA Varimax Rotation

Variable	**Factor1**	**Factor2**	**Factor3**
Pb	0.455	**0.833**	0.020
Cr	**0.904**	0.351	0.131
Cu	0.401	-0.011	**0.838**
Na	-0.856	-0.473	-0.069
Ca	-0.504	**0.737**	-0.417
K	0.464	0.010	0.370
Fe	-0.219	-0.971	0.011
Mn	0.260	0.025	**0.951**
Zn	**0.892**	-0.087	0.345
Mg	-0.311	-0.582	**0.746**
Eigenvalue	5.157	3.463	0.988
Proportion	0.516	0.346	0.099
Cumulative	0.516	0.862	0.961
Variance	5.1573	3.4636	0.9881
% Var	0.516	0.346	0.099

Factor 1 component contained high loadings for Cr and Zn amounting to 5.2 of variance. These metals were mostly provided by soil and road dust.

Factor 2 showed a strong loading for Pb and Ca, these are mainly provided by anthropogenic sources (Table 5) like automobile exhaust, incinerators, waste burning and maybe industrial processes.

Factor 3 had a strong loading for Cu, Mn and Mg this could be attributed to emissions from industry and vehicles. According to Moreno *et al*., (2006), industrial activities have been related to pollution from chemicals.

Table 5: Anthropogenic sources of heavy metals in atmospheric particulate matter

Heavy Metals	Sources	References
As	Coal Combustion	Tian *et al*., 2010
	Smelting furnace	Yang et al., 2003
Cd	Steel, plastic and pigments production	Tian *et al*. 2010
	Tyre wearing	Hjortenkrans *et al*., 2007
Cr	Coal and oil combustion	Tian *et al*., 2010
	Rubber type wear and vehicle (as an active agent in catalytic Converters	Galvgno *et al*., 2002 Pastuszka *et al*., 2010
Cu	Smelting furnace burning	Yang *et al*., 2003
	Vehicle emission (diesel combustion and brake lining wear)	Weckwerth, 2001 Manoli *et al*., 2002, Xia and Gao, 2011
	Coal combustion	Thurston and Spengler, 1985
Mn	Steel smelting	Querol *et al*., 2006
	Coal combustion	Deng *et al*., 2014
Ni	Petroleum and coal combustion	Cercasov *et al*., 1998 Tian *et al*., 2012
	Production and recycling of nickel-cadmium batteries	Morselli *et al*., 2003
Pb	Steel, plastic and pigments production	Li *et al*., 2012
	Coal combustion	Zhang *et al*., 2009
	Lead gasoline	Yang *et al*., 2003
	Waste incinerator	Zhang *et al*., 2002
V	Mining & Smelting of vanadium	Hope, 1997
	Oil combustion	Cercasov *et al*., 1998
Zn	Steel smelting	Querol *et al*., 2006
	Burning of incinerators coal-fired boiler	Thurston & Spengler, 1985 Yang *et al*., 2003
	Waste incinerator	Deng *et al*., 2006
	Vehicle emission (gasoline engine emissions and tire wearing	Salvador *et al*., 2006 Fang *et al*., 2006

Source: Dai*et al*., 2015

4. Conclusion

As part of environmental campaign in Nigeria, contamination assessment of rainwater samples were performed in Akure, Ondo State. Physico-chemical properties of the samples were analysed and also the metals were determined. All the metals values were within permissible limits of WHO. It is gratifying to note the low level of Pb, Cd was not detectable, but it is necessary to constantly monitor these elements because of the effect on human and animal health. PCA was used to determine the sources of the metals, from the results, three factors were highlighted namely anthropogenic, vehicular emission, corrosion and wear of vehicle parts. Part of the anthropogenic source (industrial activities) confirmed the increase in industrial development in Akure, the state capital of Ondo State, Nigeria.

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