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Pollution assessment of light rare earth metals in sediments of the Ogun River

Diayi M. A.[1], Gbadebo A. M.[1*], Arowolo T. A.[1] and Awomeso J. A.[2]

[1]Department of Environmental Management and Toxicology, Federal University of Agriculture, Abeokuta, P.M.B. 2240, Abeokuta 110001, Nigeria.
[2]Department of Water Resources Management and Agrometeorology, Federal University of Agriculture, Abeokuta 110001, Nigeria.

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This research was aimed at determining the concentrations of light earth metals in sediments of the Ogun River as well as their pollution and contamination status. Sediment samples collected using a graduated hand held sediment grab were stored in polythene bags and transported to the laboratory in an ice chested cooler. At the laboratory, the sediment samples were air dried, pulverized and sieved. Induced Couple Plasma Mass Spectrometry (ICP/MS) was used to determine the concentrations of light rare earth metals in the samples. Results obtained showed that the Cs values obtained which ranged from 21.10 to 121.40 mg kg^{-1} were higher than the Average Shale Value (ASV) of 5.00 mg kg^{-1}. Similarly, La ranged from 47.60 to 56.20 mg kg^{-1}; a value which exceeded the ASV value of 43.00 mg kg^{-1}. Values obtained were subjected to Geochemical Pollution Intensity (Igeo) and Contamination Factor (CF) analyses where all Cs values were seen to be very strongly contaminated while Igeo values for Cs which ranged from 3.20 to 4.00 showed highly polluted status. Cs is of great concern to sediment quality in areas surveyed. Likewise, light rare earth metals from Arakanga New Scheme (dam area) and Lafenwa (residential area) should be closely monitored to avoid adverse effects on aquatic or human lives due to high values observed.

Key words: Light rare earth metals, sediment, river, concentration, contamination.

INTRODUCTION

Sediments can be defined as loose sand, clay, silt and other soil particles that are deposited at the bottom of water bodies or accumulated at depositional sites. Their emergence can likely be from bedrocks, erosion, deposition of plants and animals or even soil (Andem et al., 2015). Through these processes as well as anthropogenic factors like industrial emissions and indiscriminate dumping of refuse, metals can be released into water bodies. These metals when in aquatic systems settle as bottom sediments. Metals can be considered as serious pollutants of the aquatic ecosystem due to their environmental persistence, toxicity and ability to be incorporated into food chains (Mashiatullah et al., 2011). Metals have toxic properties which results in deleterious

*Corresponding author. E-mail: dmanslem06@yahoo.com. Tel: +234-8063517259.

health effects on humans and ecosystem (Mahfuza et al., 2012).

Rare earth metals have been utilized in a number of industrial, medical and agricultural or zootechnical applications due to their specific properties such as high electrical conductivity and high luster capabilities. Fertilizers which are enriched with rare earth metals are released into cultivated lands from where they can find their way into water bodies and eventually, sediments due to run-off (Zhuang et al., 2017). Additionally, rare earth metals in water bodies like rivers may be due to mining activities.

Rare earth elements in water and sediments enter into humans through multiple exposure pathways majorly via food ingestion. The environmental impact of rare earth elements mining has been associated with bioaccumulation (Thomsen, 2016). Continuous exposure to low levels of rare earth elements on human health have been raising concerns because they tend to accumulate in the blood, brain and bone after entering the body (Zaichick et al., 2011). Long term exposure to rare earth elements could lead to health issues like changes in the brain and bone. It could also result in decreased Intelligence Quotient (IQ) and memory loss when children are exposed to them (Zhuang et al., 2017).

Additional implications of rare earth elements extraction and refining activities which make the activity a major environmental concern is the fact that strong acids are used at various stages of ore processing and refining. This results in the release of acidic effluents which can affect downstream water bodies (USEPA, 2012).

The Ogun River basically serves as means of water supply (for drinking and domestic purposes) to the people of Ogun State, South West Nigeria, with an estimated population of 5,217,716 (Wikipedia, 2021). Fishes from this river are also consumed by the residents and others from neighboring states. Since rare earth metals settle in bottom river sediments and can be released into water bodies which have the potential to increase concentrations of the metals in the environment, it is important to assess their levels in the Ogun River sediment so that information generated can serve as a basis for monitoring. Additionally, information on this area of study/research is limited therefore the study was embarked on to provide significant contribution to knowledge on rare earth metal levels (in the Ogun River sediments) which would be useful in policy/decision making on areas identified as culpable sources of release of these metals into the water body.

MATERIALS AND METHODS

Sampling area

The sampling locations include: Ikere-gorge, Iseyin, Olokemeji, Ofiki, Opeki, Iganagan, Oyan, Lafenwa, Arakanga Old Scheme, Arakanga New Scheme, Mokoloki, Oke-Oko and Kara (Figure 1). Using a calibrated sediment grab, sediment samples were collected from 13 locations along the Ogun River course namely: Ikere-gorge, Iseyin, Olokemeji, Ofiki, Opeki, Iganagan, Oyan, Lafenwa, Arakanga Old Scheme, Arakanga New Scheme, Mokoloki, Oke-Oko and Kara between January and March, 2013. Samples were placed in polythene bags after which they were transported to the laboratory via ice chested coolers. The sediment samples were then sun dried to remove moisture, sieved using 2 mm sieve and pulverized in order to obtain finer particles. Three sediments samples were collected per location for analysis. Sediment metals were determined using the ICP/MS technique.

Determination of metal concentration

A 0.5 g sample (sediment) was digested in aqua regia at 90°C in a controlled microprocessor digestion block for 2 h. Digested samples were diluted and analyzed by Perkin Elmer Sciex Elan 9000 ICP/MS. Based on the ICP/MS technique, sediment samples were introduced into argon plasma as aerosol droplets. The plasma dried the aerosol, dissociated the molecules, and then removed an electron from the components, thereby forming singly-charged ions, which were directed into a mass filtering device known as the mass spectrometer. A quadrupole mass spectrometer which rapidly scans the mass range was then used. At any given time, only one mass-to-charge ratio was allowed to pass through the mass spectrometer from the entrance to the exit. Upon exiting the mass spectrometer, ions stroke the first dynode of an electron multiplier, which served as a detector. The impact of the ions released a cascade of electrons, which were amplified until they became a measureable pulse. The software compared the intensities of the measured pulses to those from standards, which made up the calibration curve, to determine the concentration of the element. The ICP/MS analysis was carried out at Actlab, Canada.

Data generated were subjected to Analysis of Variance (ANOVA) while various means obtained were separated by Duncan Multiple Range Test using Statistical Package for the Social Sciences version 20. Other analysis carried out using generated data includes: Geochemical Pollution Index analysis was determined using Muller (1979) and mathematically expressed as:

$$\mathrm{Igeo} = \log_2\left(\frac{\mathrm{Cn}}{1.5\mathrm{Bn}}\right) \tag{1}$$

where Cn represents heavy metal concentration in sediment of the study area, Bn is the geochemical background value of element of interest in average shale (background value) and 1.5 is the background matrix correction factor due to lithogenic effects. This index is classified into various classes for easy identification of sediment pollution status. If the value obtained falls within 0-1, the pollution intensity is termed unpolluted while 1-2 is classified as moderately polluted to unpolluted, 2-3 is moderately unpolluted, 3-4 is moderately to highly polluted, 4-5 is highly polluted while 5 and above is very highly polluted.

Contamination factor analysis was determined using the formula:

$$\mathrm{CF} = \frac{(\mathrm{Cheavy\ metal})}{(\mathrm{Cbackground})} \tag{2}$$

CF stands for the Contamination Factor while $C_{heavy\ metal}$ stands for the concentration of the heavy metal in a location and $C_{background}$ stands for the background concentration (in this case, the world shale value of the element). Based on this, contamination levels were classified on their intensities on a scale ranging from 1-6, where 0= none, 1= none to medium, 2 = moderate, 3 = moderate to strong, 4 = strongly polluted, 5 = strong to very strong, 6 = very strong.

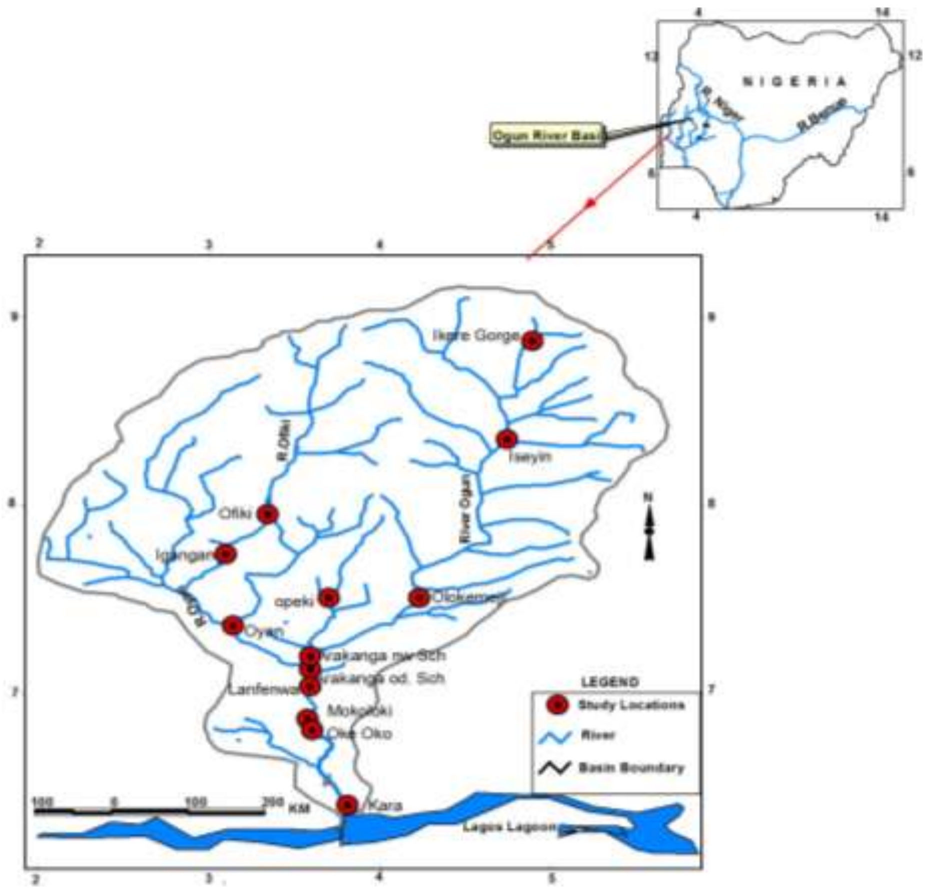

Figure 1. Map of the Ogun River.
Source: Author

Quality assurance

Three samples were taken per location for analysis while glasswares used were soaked in a 1% HCl solution after which they were washed using Alconnox. The wares were then rinsed 5-6 times with distilled water and oven dried at 160°C for proper sterilization. A series of standard reference materials used at the Activation Laboratory for sediment metal concentration analysis include: GXR-1 Meas, GXR-1 Cert, GXR-2 Meas, DH-1a Meas, DH-1a Cert, GXR- 4 Meas, GXR-4 Cert, GXR-6 Meas, GXR-6 Cert, SAR-M (U.S.G.S) Meas and SAR-M (U.S.G.S) Cert, while TILL-1 Meas, TILL-1 Cert, TILL-2 Meas and TILL-2.

RESULTS

Values obtained for Lanthanum (La) analysis showed that the highest concentration of the metal was obtained at Arakanga New Scheme (56.20 mg kg^{-1}), while the least value of 9.00 mg kg^{-1} was obtained at Igangan (Table 1). When compared with the average shale value standard of 43.00 mg kg^{-1}, La values from Lafenwa (47.60 mg kg^{-1}), Ofiki (48.60 mg kg^{-1}), and Arakanga New Scheme (56.20 mg kg^{-1}) exceeded the Average Shale Value (ASV). When subjected to the geochemical pollution intensity (Igeo) analysis, all values obtained for La ranged from -2.80 to -0.20 (Table 2). Contamination Factor (CF) values obtained from Ikere, Iseyin, Olokemeji, Opeki, Igangan, Arakanga Old Scheme, Mokoloki, Oke-Oko and Kara were less than 1 while Lafenwa, Ofiki and Arakanga New Scheme had values that fell between 1 and 2 (Table 3).

The Caesium (Cs) analysis carried out showed a range of 21.10 mg kg^{-1} (Iganagan) to 121.40 mg kg^{-1} (Arakanga New Scheme) (Table 2) which exceeded the ASV standard of 5.00 mg kg^{-1} (Table 1). Igeo values ranged from 1.50 (Igangan) to 4.0 (Arakanga New Scheme) while CF values ranged from 4.20 to 24.30 (Table 3). All Praseodynium (Pr) values obtained were lower than the ASV standard of 9.80 mg kg^{-1} except for those from Ofiki (11.90 mg kg^{-1}), Oyan (10.10 mg kg^{-1}), Lafenwa (12.00 mg kg^{-1}) and Arakanga New Scheme (15.30 mg kg^{-1}) (Table 1).

Neodymuim (Nd) values obtained ranged from 7.10 mg kg^{-1} (Iganagan) to 54.20 mg kg^{-1} Arakanga New Scheme (Table 1). Values which exceeded the ASV standard value of 33.00 mg kg^{-1} were obtained from Ofiki (41.40 mg kg^{-1}), Oyan (35.20 mg kg^{-1}), Lafenwa (42.70 mg kg^{-1}), Arakanga Old Scheme (33.80 mg kg^{-1}), Mokoloki (33.70 mg kg^{-}1) and Arakanga New Scheme (54.20 mg kg^{-1}) (Table 1).

Table 1. Mean and standard deviation of light rare earth element from the Ogun River .

Location	La	Cs	Pr	Nd	Sm	Eu	Gd
				mg kg^{-1}			
Ikere	20.40 ± 12.00	52.30 ± 14.40	4.80 ± 2.95	17.40 ± 10.85	3.20± 2.10	0.40 ± 0.30	2.80 ± 1.80
Iseyin	15.00 ± 12.80	55.10 ± 11.30	3.50 ± 3.10	12.50 ± 11.20	2.20 ± 2.20	0.30 ± 0.20	1.90 ± 1.60
Olokemeji	14.20 ± 2.90	32.70 ± 7.20	3.20 ± 0.60	11.60 ± 2.30	2.00 ± 0.40	0.30 ± 0.10	1.60 ± 0.30
Opeki	36.00 ± 1.40	99.70 ± 1.10	8.70 ± 0.30	30.20 ± 1.20	4.00 ± 0.20	0.50 ± 0.10	2.80 ± 0.15
Ofiki	48.60 ± 7.50	100.00 ± 12.50	11.90 ± 1.80	41.40 ± 6.00	6.20 ± 0.85	0.50 ± 0.10	4.30 ± 0.60
Igangan	9.00 ± 1.80	21.10 ± 3.80	2.00 ± 0.40	7.10 ± 1.69	1.00 ± 0.20	0.20 ± 0.00	0.80 ± 0.15
Oyan	42.50 ± 11.30	100.00 ± 25.40	10.10 ± 3.00	35.20± 10.5	5.50 ± 1.60	0.40 ± 0.10	3.60 ± 0.90
Lafenwa	47.60 ± 8.10	105.10 ± 17.85	12.00 ± 2.10	42.70 ± 7.60	6.70 ± 1.05	0.60 ± 0.10	4.70 ± 0.75
Arakanga Old Scheme	40.50 ± 9.30	83.10 ± 15.20	9.60 ± 2.30	33.80 ± 7.90	5.30 ± 1.00	0.60 ± 0.10	3.80 ± 0.55
Arakanga New Scheme	56.20 ± 27.60	121.40 ± 60.95	15.30 ± 8.15	54.20 ± 29.60	8.10 ± 4.30	0.40 ± 0.20	5.10 ± 2.70
Mokoloki	38.40 ± 8.80	79.30 ± 9.80	9.40 ± 2.30	33.70 ± 8.60	5.60 ±1.50	1.00 ± 0.35	4.4 ± 1.25
Oke Oko	32.70 ± 1.90	69.90 ± 4.30	7.90 ± 0.50	27.8 ± 1.95	4.60 ± 0.25	0.60 ± 0.10	3.40 ± 0.25
Kara	19.90 ± 2.30	27.40 ± 20.00	4.80 ± 0.60	16.7 ± 2.20	2.70 ±0.30	0.20 ± 0.10	2.00 ± 0.25
ASV	43.00	5.00	9.80	33.00	6.20	1.20	5.10
ECSQG	NS	NS	NS	NS	NS	NS	NS

ASV- Average Shale Value, ECSQGS – Environment Canada Sediment Quality Guideline Standard, NS – Not Detected.
Source: Author

Table 2. Geochemical pollution index values (Igeo) of light rare earth element in sediments samples from the Ogun River.

Location	La	Cs	Pr	Nd	Sm	Eu	Gd
Ikere	-1.70	2.80	-1.60	-1.50	-1.50	-2.20	-1.70
Iseyin	-2.12	2.90	-2.11	-2.00	-2.10	-2.60	-2.30
Olokemeji	-2.20	2.10	-2.30	-2.10	-2.20	-2.60	-2.50
Opeki	-0.80	3.70	-0.70	-0.70	-1.20	-1.83	-1.70
Ofiki	-0.42	3.70	-0.30	-0.25	-0.60	-1.83	-1.10
Igangan	-2.80	1.50	-2.80	-2.80	-3.20	-3.20	-3.50
Oyan	-0.60	3.70	-0.50	-0.49	-0.80	-2.20	-1.10
Lafenwa	-0.40	3.80	-0.30	-0.10	-0.50	-1.60	-1.00
Arakanga Old Scheme	-0.70	3.47	-0.60	-0.60	-8.80	-1.60	-1.30
Arakanga New Scheme	-0.20	4.00	0.50	0.10	-0.20	-2.20	-0.80
Mokoloki	-0.70	3.40	-0.60	-0.60	-0.70	-0.80	-1.10
Oke Oko	-1.00	3.20	-1.60	-0.25	-1.00	-1.60	-1.40
Kara	-1.70	1.90	-0.90	-1.60	-1.80	-3.20	-2.20
Average Shale Value	43.00	5.00	9.80	33.00	6.20	1.20	5.1

Source: Author

All Europium (Eu) and Gadolinium (Gd) values were below the ASV standards of 1.20 and 5.1, respectively except for Gd value of 5.1 from Arakanga New Scheme which was of same value with the standard (Table 1).

Pr values obtained from the research ranged from 2.00 to 15.30 mg kg^{-1} (Table 1), while Igeo values ranged from -2.80 (Igangan) to 0.50 (Arakanga New Scheme) (Table 2) with CF values ranging from 0.20 (Igangan) to 1.60 (Arakanga New Scheme) (Table 3). Similarly, Nd values ranged from 7.10 to 54.20 mg kg^{-1} with the least value from Igangan and highest from Arakanga New Scheme (Table 1). Igeo and CF values for Nd ranged from -2.80 to 0.10 (Table 2) and 0.20 to 1.60 (Table 3), respectively. Igangan and Arakanga New Scheme had the least and highest values, respectively for Igeo and CF, respectively. All Eu and Gd values obtained were not higher than the ASV standards of 1.20 and 5.10, respectively.

DISCUSSION

With La values from Lafenwa (47.60 mg kg^{-1}), Ofiki (48.60 mg kg^{-1}), and Arakanga New Scheme (56.20 mg

Table 3. Contamination factor values of light rare earth element in sediments samples from the Ogun River.

Location	La	Cs	Pr	Nd	Sm	Eu	Gd
Ikere	0.50	10.50	0.50	0.50	0.50	0.30	0.55
Iseyin	0.35	11.00	0.40	0.40	0.35	0.25	0.40
Olokemeji	0.30	6.50	0.30	0.35	0.30	0.25	0.30
Opeki	0.80	19.90	0.90	0.90	0.65	0.40	0.55
Ofiki	1.10	20.00	1.20	1.30	1.00	0.40	0.84
Igangan	0.20	4.20	0.20	0.20	0.10	0.20	0.20
Oyan	1.00	20.00	1.00	1.10	0.90	0.30	0.70
Lafenwa	1.10	21.00	1.20	1.30	1.10	0.50	0.90
Arakanga Old Scheme	0.90	16.60	1.00	1.00	0.90	0.50	0.75
Arakanga New Scheme	1.30	24.30	1.60	1.60	1.30	0.30	1.00
Mokoloki	0.90	15.90	1.00	1.00	0.90	0.80	0.90
Oke Oko	0.80	14.00	0.80	0.80	0.70	0.50	0.70
Kara	0.50	5.50	0.50	0.50	0.40	0.20	0.40
Average Shale Value	43.00	5.00	9.80	33.00	6.20	1.20	5.1

Source: Author

kg^{-1}) exceeding the 43.00 mg kg^{-1} average shale value standard the metal is seen to be a threat to sediment quality in those locations. The Igeo results obtained (-2.80 to -0.20) from all locations were classified as unpolluted since values obtained were below 0. Similarly, Ikere, Iseyin, Olokemeji, Opeki, Igangan, Arakanga Old Scheme, Mokoloki, Oke-Oko and Kara were seen not to be contaminated based on the CF values obtained which were less than 1 while Lafenwa, Ofiki and Arakanga New Scheme were seen to have medium contamination since their values fell between 1 and 2 when subjected to Contamination Factor analysis. Lanthanum accumulates in different organisms and thus may pose a hazard to species belonging to higher levels of the food chain (Henning et al., 2016). La values obtained from Gora River which ranged from 5.00 to 21.00 mg kg^{-1} were lower than the ASV standard of 43.00 mg kg^{-1}. Similarly, research by Dilioha and Onwualu-John (2016) showed that all La values ranging from 8.30 to 38.00 mg kg^{-1} were observed to be lower than the ASV standard.

According to Das et al. (1988), Lanthanum (La), is bioavailable in its trivalent form (La^{3+}) and has a high risk of biological effects while Dave et al. (1991) asserted that La^{3+} could compete for binding sites in biological systems with Ca^{2+} thereby inhibiting channels for calcium within cell membranes thus affecting the work of cells and tissues. Hua et al. (2017) also reported that La^{3+} has toxic effects on gills and livers of rare minnows. Additionally, high affinity of La to phosphate was observed by Herrmann et al. (2016) where insoluble lanthanum phosphate complexes were formed. This could have adverse effect on algal growth as a result of phosphate limitation. Similarly, high La concentrations inhibited growth of *Tetrahymena shanghaiensis,* a ciliate which grazes on planktonic bacteria while low La concentrations stimulated growth (Herrmann et al., 2016).

Caesium values obtained were seen to be at least 4 to 20 times higher than the ASV standard of 5.00 mg kg^{-1}. This presents a very high risk to biota in the aquatic environment. Igangan and Kara Igeo values (1.50 and 1.90) were observed to be unpolluted to moderately polluted, while values from Olokemeji (2.10), Ikere (2.80) and Iseyin (2.90) showed moderate pollution. Oke-Oko (3.20), Mokoloki (3.40), Opeki (3.70), Ofiki (3.70), Oyan (3.70), Lafenwa (3.80) and Arakanga New Scheme (4.00) were observed to be highly polluted. Similarly, all Cs values obtained showed very strong contamination when subjected to Contamination Factor analysis. Ren et al. (2007) reported that increased CE concentrations resulted in increased bioaccumulation intracellular and extracellular processes, which in turn lead to a decrease in lichen viability. Animals which are exposed to extremely high doses of cesium showed changes in behavior which are manifested in form of increased or decreased activity (www.lenntech.com).

Praseodymium (Pr) values from Oyan (10.10 mg kg^{-1}), Ofiki (11.90 mg kg^{-1}), Lafenwa (12.00 mg kg^{-1}), and Arakanga New Scheme (15.30 mg kg^{-1}) are a threat to sediment quality since they exceeded the ASV standard of 9.80 mg kg^{-1}. Igeo values obtained which ranged from -2.80 (Igangan) to 0.5 (Arakanga New Scheme) showed unpolluted status for all locations while CF analysis showed medium contamination for Ofiki (1.2), Lafenwa (1.2) and Arakanga New Scheme (1.6). Values obtained at the Southern Benue Trough ranging from 2.18 to 8.54 were observed to be lower than the ASV standard (Dilioha and Onwualu-John, 2016).

Effects of Pr on *Spirodela polyrhiza* at a concentration range of 0 to 60 µM for 20 days, in which it was observed that significant increase in cell death occurred with a reduction in photosystem II activity (Xu et al., 2016). This is indicative of the presence of Pr-inducing oxidative stress. Similarly, significant increase in cell death was observed in Pr-treated plants. Praseodymium can be a threat to the liver when it accumulates in the human body (www.lenntech.com).

Mokoloki (33.80 mg kg^{-1}), Ofiki (41.40 mg kg^{-1}), Lafenwa (42.70 mg kg^{-1}) and Arakanga New Scheme (54.20 mg kg^{-1}) Nd values which exceeded the 33.0 mg kg^{-1} ASV standard were seen as threats to sediment quality. Igeo values obtained ranged from -2.80 (Igangan) to 0.10 (Arakanga New Scheme) which indicates unpolluted status while CF values which ranged from 1.00 (Arakanga Old Scheme and Mokoloki) to 1.60 (Arakanga New Scheme) were observed to be moderately contaminated. According to Lenntech (2018), Nd causes damage to cell membranes of aquatic organisms which could have several negative influences on reproduction and the functions of the nervous system. Nd values from the Southern Benue Trough which ranged from 8.00-31.50 were found to be lower than the ASV standard of 33.00 mg kg^{-1} (Dilioha and Onwualu-John, 2016).

All Igeo values for Eu and Gd were observed to have shown unpolluted status. Similarly, all CF values for Eu and Gd were observed to be uncontaminated except for that of Arakanga Ne Scheme (1.00) which showed moderate pollution. Eu values obtained from the Southern Benue Trough which ranged from 1.29 to 1.46 were found to be higher than the ASV standard of 1.20 mg kg^{-1} (Dilioha and Onwualu-John, 2016). This showed that Eu values are of great concern to sediment quality in that area. Similarly, Gd values obtained from River Gora, Minna, Niger State, which ranged from 8 to 26 mg kg^{-1} were observed to be higher than the ASV standard of 5.1 mg kg^{-1} (Obaje et al., 2015). This poses a threat to sediment quality as well as aquatic organisms. Wilde et al. (2002) stated that Gd can be toxic to selected microorganisms such as algae and bacteria at very high concentrations (average 80,000 mg/L, maximum 259,000 mg/L).

Conclusion

Eu and Gd sediment metals were of no threat to sediment quality due to the unpolluted status observed while Cs should also be monitored closely bearing in mind that values obtained from all locations were 4 to 20 times higher than the ASV standard. This showed high Cs contamination status in all locations. Arakanga New Scheme and Lafenwa sediments are a concern to sediment quality in view of metals values obtained which exceeded ASV standards in these locations except for Eu and Gd values. Metals assessments in these locations are of high importance in order to avoid adverse effect occurrences on human and aquatic lives since dam for public water supply is situated at Arakanga while Lafenwa has high human residential areas that depend on the river water for various purposes.

CONFLICT OF INTERESTS

The authors have not declared any conflict of interests.

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