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Gamma-Ray Spectrometric Determination And Multivariate Analysis Of Radionuclide Fluxes In Shore Sediment At Port Victoria, Kenya

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***Abstract*—Increased human and industrial activities discharge effluents containing TENORM while geological processes results to NORM directly onto land and in water bodies in L. Victoria basin. River Nzoia is the largest river draining the basin and forms an integral part of the lake shoreline at Port Victoria. In this study, bottom sediments from the shoreline at Port Victoria were sampled and analyzed for radionuclide activity using NaI(Tl) and chemometric techniques; principal component analysis (PCA) and hierarchical cluster analysis (HCA). The mean concentration levels of the radionuclides K-40, U-232 and Th-232 were 523.21±26.53, 66.23±8.55 and 76.23±7.32 Bq/kg which were found to be above the world wide accepted average values of 420, 33 and 45Bq/kg respectively. PCA performed on the measured data indicates that U-238 and Pb-214 are mostly TENORM while Th-232, K-40 and Ac-228 as mostly NORM. HCA apportioned applied fertilizers and oil spills in the Lake basin is the main source of U-238 and Pb-214 while Th-232, Ac-228 and K-40 to be mostly from the geological weathering of the igneous and sedimentary rocks that characterize the Victoria catchment. Through biomagnifications and associated food chains in the aquatic system, radionuclides directly or indirectly ionize the living cells of humans causing somatic effects hence an urgent need to control pollution by radionuclides in the Lake Victoria basin is called for in this study.**

Keywords—TENORM, NORM, NaI(Tl), PCA, HCA

1. INTRODUCTION

Lake Victoria is the second largest fresh water Lake in the world. It stretches 412 km from North to South between latitudes 0° 30' N and 3°, 12' S and 355 km from West to East between longitudes 31°, 37'and 34°,53 'E (COWI, 2002). The shoreline of Port Victoria is directly linked to Lake Victoria basin by river Nzoia which is the largest river in the basin. Several processing industries such as pulp in Webuye, sugar industries (Mumias, Nzoia, West Kenya, Butali) are located along this river. The high population density in the Lake basin (World Bank, 1996) engage in small and large scale farming of both food and cash crops for subsistence and commercial use. Due to the reduction in soil fertility, farmers opt for inorganic fertilizers to boost crop production in the region. Some phosphatic fertilizers contain an activity of 3700 – 5550 Bq/kg of U-238 (Bliss, 1978; Bolivar *et al*., 2002) hence continuous application of such fertilizers in the farms without repetitive ploughing to disperse the radionuclides leads to the accumulation of technologically enhanced natural occurring radioactive material (TENORM) in the plough layer (Meissel *et al.*, 1991). Petroleum spills in the Lake basin contain technologically enhanced natural radioactive material (TENORM) (Salman and Amany, 2008) while the presence of igneous and sedimentary rocks in some areas of the basin is a source of NORM (Achola *et al*., 2009).

Through washouts, NORM and TENORM from the lake basin are deposited at the shoreline of Port Victoria via river Nzoia. Sediment deposition results from reduced velocity of the discharging river relative to Lake Victoria water (Hakason and Jason, 1983). The coarse particles of sediments are usually deposited near discharge points while the finer sediments are usually deposited far in deep regions of the water body. The bottom sediments acts as the main sink for radionuclide and their enrichment is usually more pronounced than in water (Windom *et al*., 1989; Irion, 1991; Bubb and Lester, 1996). Through biomagnifications and food chains, radionuclides ionize human cells causing somatic effects such as cancer (ICRP, 1991; Leo, 1948). There is inadequate knowledge about environmental impact due to pollution by immobilized NORM and TENORM in the Lake Victoria basin (Bliss, 1978; Mwamburi, 2009; Salman and Amany, 2008; Foster and Charlesworth, 1996). This is due to the scanty research studies as well as the commonly used univariate techniques which often fail to provide information about complex environmental data. In this study, levels of K-40, Th-232, U-238, Ac-228 and Pb-214 in bottom sediment samples were determined using NaI(Tl) spectrometry technique in combination with multivariate chemometric techniques; principal component analysis (PCA) and the hierarchical cluster analysis (HCA).

2.MATERIALS AND METHODS

2.1 Study area and sampling

Using the global positioning system (GPS), locations of the selected sampling points were determined at intervals of 1 km along a shoreline of 10 km as shown in Fig 2.1. Each sampling point was reached by boat and two bottom sediment samples of at least mass 500 g were collected at each site at a uniform depth of (0-10 cm) within a sampling area of 1 m^2. The samples were then put in well labeled polythene bags and then transported to the laboratory for preparation and analysis.

2.2 Sample preparation and measurements

The sediment samples from each site were oven-dried for 48 hrs to a constant weight before being crushed in the mortar using a pestle into fine particles. Sieving was done using a 60 µm sieve to ensure sample homogeneity and then sample weights of mass 500 g were portioned into well labeled polythene bags which were hermetically sealed to allow formation of radon gas. The samples were then stored for a minimum period of one month to allow secular equilibrium between (^{238}U and ^{232}Th) and their decay products in their respective series to be attained (Maina, 2007). The radioactive measurements were made using lead shielded 76 mm x 76 mm NaI(TI) detector with an Oxford PCA-P card operating on Windows system to acquire and analyze data using counting period of 10 hours.

2.3 Energy calibration in NaI(Tl) spectrometry

Using IAEA procedures (IAEA, 1987), the output of the detector was adjusted to agree with the known values of the IAEA standard radionuclides. This was done at every start of the measurements to cater for changes in weather, vibrations and heating up of the detector. Sample photon energies were measured simultaneously with some well known gamma lines for instance, RGU-1 spectrum was determined by means of its progeny photo peaks; ^{214}Bi (609 keV), ^{214}Bi (1765 keV) and ^{214}Pb (352 keV). RGTh -1 spectrum was determined by its progeny photo peaks of ^{208}Tl (2615 keV) and ^{228}Ac (911.2 keV) while RGK -1 was determined directly through its 1461 keV photo peak as shown in Fig 2.3

2.4 Gamma ray spectral data analysis

To identify and quantify the radionuclides in the collected samples, the spectrum of IAEA-RGMIX-2 standard reference material was used (IAEA, 1987). The spectral data acquisition time was ≈ 10 hrs. The typical gamma ray spectrum of a sediment sample collected from Port Victoria was then plotted (Figure 2.4).

2.5 Activity Concentrations.

The specific activity for each detected radionuclide was determined using method of comparison (Mustapha, 2002) in equation 2.5.

$$\frac{A_s.M_s}{I_s} = \frac{A_R.M_R}{I_R} \qquad (2.5)$$

where, A_S is the activity of the radionuclide in the sample, M_S is the mass of the sample to be analyzed, I_S is the intensity of the radionuclide in the sample to be analyzed, A_R is the activity of the radionuclide in the reference sample, M_R is the mass of the reference sample, I_R is the intensity of the radionuclide in the reference sample.

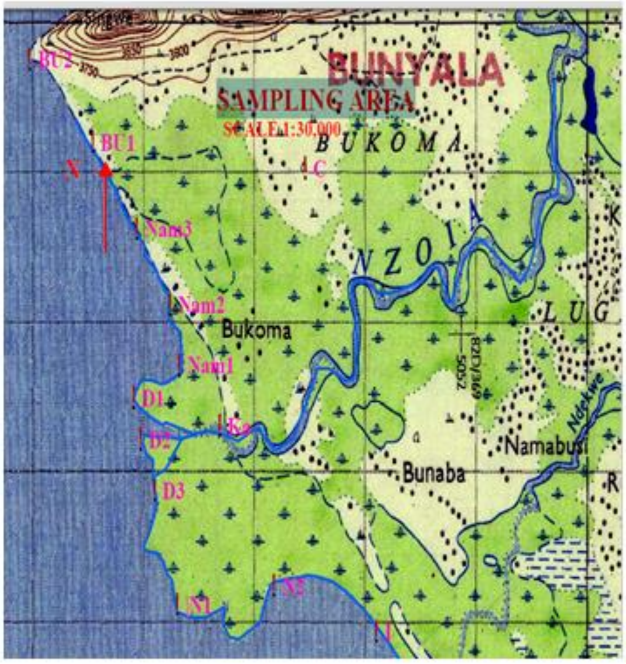

Figure 2.1: Map of Port Victoria shoreline. (Map drawn to scale by the Institute of Survey Nairobi, Kenya). Note the sampling sites along the shoreline: Ka, D1, D2, D3, N1, N2, I, Nam1, Nam2, Nam3, Bu1 and Bu2. The labeling of the sites is as follows: Ka= Kabras, D1, D2 and D3= Active deltas of River Nzoia, N1and N2= Nanganda, I= Indufu, Nam1, 2, 3= Namugerwa (blocked deltas of River Nzoia), Bu1, 2= Bukoma beaches. Also note River Ndekwe discharging at site I.

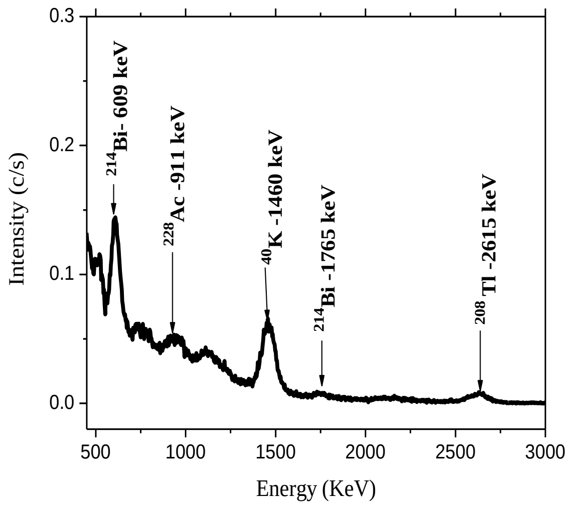

Figure 2.4: A typical gamma ray spectrum of a sediment sample collected from Port Victoria. The live time used was 36,000 s.

2.6 Multivariate Chemometric Analysis

Multivariate techniques; Principal component analysis and Hierarchical cluster analysis were used. This was because the highly dynamic environment presents complex data which can only be analyzed effectively using multivariate techniques; PCA and HCA respectively (Einax *et al.*, 1997). MATLAB software version7.1 was used.

2.6.1 Principal Component Analysis (PCA)

Principal component analysis presented information present in many variables in the raw data matrix using a few principal components (Loska and Wiechula, 2003). The extracted principal components were ordered in the sequence of decreasing variance (Einax *et al.*, 1997) and therefore only the first few principal components (PCs) were used for interpreting relationships among the observed variables (Spanos *et al*., 2008, Kaiser, 1960) while the rest were considered as noise (Lu *et al*., 2009). The original data matrix was transformed into a product of two matrices (Praveena *et al*., 2008), one of which contained the information about the samples (eg sites) and the other about the features (eg concentration) as shown in equation 2.6.1

$$X = T.P^t + E \qquad (2.6.1)$$

where, X is the original data matrix (m x n) with m in this work representing sampling sites and n represents measured activity concentrations in bottom sediment samples. T is the score matrix (m x a) with a representing the necessary number of principal components (Pcs) containing maximum information. This matrix characterized the sampling sites by the new transformed coordinates of the data in space and it had the same number of rows as the original data matrix. The score matrix described the data structure in terms of patterns (similarities/ differences) as well as the identification of the outliers

2.6.2 Hierarchical Cluster Analysis (HCA)

This method helped to examine the inter point distances between all sampling sites at Port Victoria shoreline and measured activity concentrations of bottom sediment samples. Dendrograms presented the data from a high dimensional row space in a form that facilitated the use of human pattern recognition abilities. Each radionuclide was regarded as a cluster before a similarity distance (Euclidean) was determined between all the clusters (Simeonova *et al.,* 2010). For example, if two clusters (radionuclides) are taken to be i and i′, then the similarity distance $d_{ii'}$ exists only if $d_{ii'}=d_{i'i}$ <0 where $d_{i'i}=1$ when $X_i=X_{i'}$. The row vectors in the data matrix X are given as X_i and $X_{i'}$ as in equation 2.6.2

$$d_{ii'} = \sum_{j=1}^{j}\left(x_{ij} - x_{i'j}\right)^2 \qquad (2.6.2)$$

Where, j is the number of repetitive measurements between the clusters.

3. Results and Discussions

3.1 Radioactivity Concentration in the Shore Sediment

The activity concentrations of K-40, U-238, Th-232, Pb-214 and Ac-228 were calculated in Bq/kg by method of comparison and recorded in Table 3.1. The below detection limits were used where activity values were below the detection limits for the purpose of multivariate analysis.

3.1: Activity concentration of (K-40, U-238, Th-232, Pb-214 and Ac-228) in Bq/kg

Site	K-40	U-238	Th -232	Pb-214	Ac-228
Ka	659.59 ±17.69	47.26± 0.77	125.74± 6.83	32.9±8 .53	16.17±4 .19
D1	568.23 ±16.45	77.52± 7.97	81.53±4 .43	BDL	17.84±2 .56
D2	918.64 ±26.53	BDL	70.06±2 .72	13.92± 3.22	27.63±3 .48
D3	209.36 ±6.05	BDL	BDL	21.23± 6.32	BDL
N1	218.97 ±6.33	BDL	BDL	36.52± 8.03	BDL
N2	605.18 ±17.49	79.15± 8.14	112.18± 6.06	20.26± 8.56	5.72±1. 95
I	627.33 ±18.13	178.46 ±18.29	60.59±3 .27	BDL	BDL
Nam1	500.98 ±14.48	83.13± 8.55	80.91±4 .37	27.16± 9.57	BDL
Nam2	465.96 ±13.47	57.36± 5.97	74.67±4 .63	BDL	4.12±0. 32
Nam3	230.22 ±6.68	74.84± 7.69	BDL	20.66± 6.56	BDL
Bu1	733.79 ±21.21	53.91± 5.51	135.56± 7.32	26.88± 5.49	BDL
Bu2	540.23 ±15.61	BDL	96.99±3 .08	35.66± 7.71	BDL

The activities in Bq/kg ranged widely from 209.36±6.05-918.64±4.56 for K-40, 15.55±0.032 -178.24±1.98 for U-238 and 36.68±0.29-135.56±0.06 for Th-232. The mean values of K-40, U-238 and Th-232 were 523.21±26.53, 66.23±8.55 and 76.23±7.32 Bq/kg which were found to be above the global accepted average values of 420, 33 and 45Bq/kg respectively (UNSCEAR, 2000). This indicates that TENORM from Lake Victoria basin have contributed to the elevated natural levels at Port Victoria shoreline (Bliss, 1978).

Large variations were observed in the activity concentrations at different sampling points. This is due to different discharge rates of the three deltas of River Nzoia (D1, D2 and D3) in Fig 2.1 during both wet and dry seasons as well as the influence of water tides which results in fresh deposition and sweeping away of light materials that are sinks for the radionuclides (Ramasamy *et al.*, 2008). The hydraulic processes during transportation of the radionuclides and variation in grain size distribution were also attributed to contribute to variations in concentration levels at different sampling points (Axtman and Luoma, 1991).

3.2 Principal Component Analysis (PCA) of the measured radionuclides

PCA was performed on (K-40, Th-232, U-238, Pb-214 and Ac-228) to establish similarities and differences among them (Tahir *et al.*, 2005). Varimax rotation method and normalization by Kaiser Method were carried out to enhance data interpretation (Drystad, 1998) and to reduce the influence of some measured radionuclides over others. The principal components and the percent of variance explained by each PC were then calculated as in Table 3.2.

Table 3.2: Factor loadings on principal components

Element	Loadings on principal components		
	PC1	PC2	PC3
K-40	0.10	0.67	0.51
U-238	0.90	0.01	0.11
Th-232	-0.03	0.22	0.94
Pb-214	0.89	0.09	-0.12
Ac-228	-0.04	-0.94	-0.12
Eigen Value	2.00	1.56	0.61
% explained variance	39.96	31.27	12.24
% cumulative	39.96	71.23	83.47

Extraction method: Principal component analysis. Rotation converged in five iterations.

Two principal components with eigen values greater than one (accounting for 71.23 % of total variation) were used for data analysis about the measured radionuclides in this work (Kaiser, 1960; Zhu and Ghodsi, 2006). The first factor explained 39.96 % of total variance. U-238 and Pb-214 were found to have strong loadings of 0.90 and 0.89 respectively on this first component and were regarded to be TENORM. The second factor explaining 31.27 % of total variation moderately and strongly loaded on by K-40 (0.67) and Ac-228 (-0.94). These radionuclides were found to be NORM with different sources in the Lake Basin (Tahir, 2005). The eigen value for the third factor was less than one and it was discarded during analysis (Kaiser, 1960). These results are consistent with those presented by correlation coefficients. In Fig 3.2, the cluster mostly TENORM comprises of U-238 and Pb-214 with significant correlation of 0.61. These radionuclides were found to have positive loadings (0.90 and 0.89) respectively on PC1. These radionuclides were found to characterize site I in Fig 2.1. The concentration levels of these radionuclides (particularly U-238) were found to be elevated at this site. This was attributed to the discharge by the nearby R. Ndekwe that traverses Bunyala rice fields where phosphatic fertilizers are used on large scale. K-40 and Th-232 formed a mostly NORM cluster while Ac-228 with a negative loading on PC2 is uncorrelated with all radionuclides analyzed in this work. Its source may be different from those of Th-232 and K-40.Its source may be different from those of Th-232 and K-40.

Though Th-232 could not be accounted for by PC2 (with eigen value less than one), the 3D plot clustered it with K-40. This indicated that K-40 and Th-232 are correlated (i.e mostly NORM) in the estuarine of Lake Victoria.

3.3 Hierarchical Cluster Analysis (HCA) of radionuclides

In this work, the concentration data matrix (5 x 12) ie five radionuclides and twelve sampling sites of the measured radionuclides was standardized by means of Z scores. Euclidean distances were calculated before applying the Ward's method on the standardized data. The results in Fig 3.3 present two main clusters of the measured radionuclides. The two main clusters were found based on the source of origin in the lake basin. The cluster Mostly TENORM may be attributed to the use of phosphatic fertilizers for farming and spilled oil that is transported from the deports in the catchment areas of Lake Victoria (Bolivar *et al*., 2002; Meissel *et al*., 1991; Bliss, 1978; Salman and Amany, 2008).

Th-232 and K-40 (with mean levels which were found to be above the natural background levels) were due to artisanal mining activities and natural weathering of the rocks in the lake basin. Ac-228 which was uncorrelated to all radionuclides (with negative loading on PC2) was attributed to emanate from the decay of its parent Th-232 in Lake Victoria basin. This HCA interpretations are consistent with those of correlation coefficient and the PCA done in this work.

4.0 Conclusion

The principal component analysis and the hierarchical analysis have been successfully utilized to reveal the relationships among the radionuclides in this work. U-238 and Pb-214 were found out to be mostly TENORM while Th-232, K-40 and Ac-228 were found to be mostly NORM in the Lake Victoria basin.

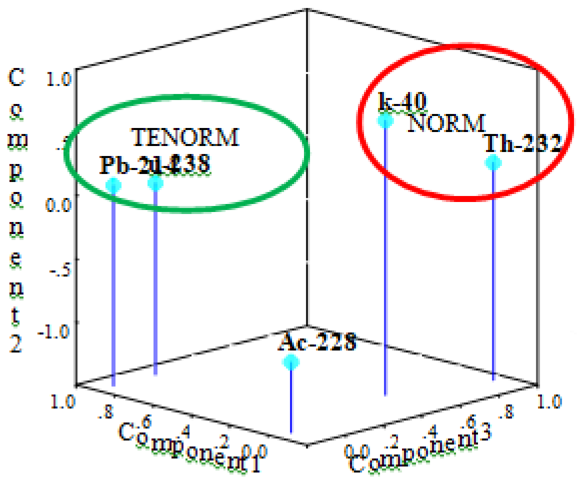

Figure 3.2: PCA of radionuclides in three dimensional plot

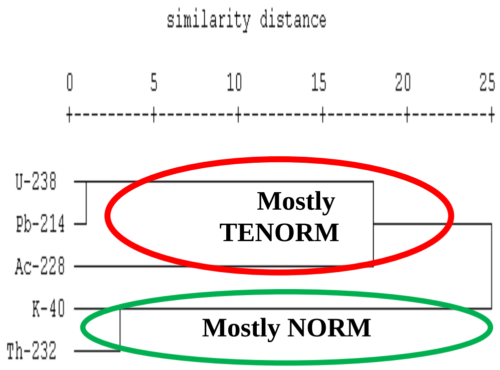

Figure 3.3: Hierarchical cluster analysis of (Pb-214, Ac-228, Th-232, U-238 and K-40)

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