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RADON SYSTEMATICS IN THE HIGH BACKGROUND RADIATION AREA (HBRA) GEOTHERMAL WATERS OF WINNAM GULF (KENYA)

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Abstract: The activity concentration of radon in ground and surface waters associated with a geothermallyactive high background radiation area (HBRA) of Winnam Gulf (Kenya) was measured using liquid scintillation counting (LSC) spectrometry. The results were analyzed using chemometric techniques viz principal components analysis (PCA), hierarchical cluster analysis (HCA) and soft independent modeling of class analogy (SIMCA). The goal of the study was to estimate the levels as well as explore the multivariate alterations of radon in relation to geothermal activity in typical HBRA. PCA was used for dimensionality reduction of the analyzed data and exploration of alterations of radon concentration in the waters. HCA was used to partition the analyzed data into non overlapping clusters and display them inform of a dendrogram. SIMCA was used and was successful in identifying local models for possible groups of geothermal waters which may be used to predict a probable class membership for new observations from similar geological areas based on the activity concentration of radon. PCA and HCA successfully delineated the radon levels with respect to whether they belonged to HBRA or non-HBRA. SIMCA modeling was 64%, and the modeling power of the activity concentration of radon was closer to 100%.

Keyword: HCA, LSC, PCA,Radon and SIMCA

1. INTRODUCTION

Radon is a non reactive naturally occurringradioactive gas which is colorless, odorless, tasteless and eight times heavier than air. This noble gas is a decay product of naturally occurring uranium and radium in soils and rocks. It has half-life of 3.825 days [1], [2]. Radon may be generated in geothermal areas due to the presence of small quantities of radioactive rocks containing uranium that lie in the path of the passage of geothermal waters and it may be transported a long with an influx of magnetic gases such as CO_2, He, H_2, and $CH_4$from a deeply burned magma chamber [3], [4]. Geothermal activities in the environment normally alter the permeability of rocks and raise the temperature of the waters, which in turn affect the level of radon in waters. The Winnam Gulf of South Western Kenya is a unique setting comprising of both geothermally active HBRA and non-geothermal HBRA regions. The region has a series of cone sheets of carbonatite alkaline rocks and ijolite (alkaline igneous rocks) which are known to contain above background levels of radioactivity[5], [6].Geothermal activity in the region occurs at Kanam, Kanjera and Bala which lie on the southern shores of the Winnam Gulf through a few widespread hot/warm springs, gas release in the springs and the *Fimbristylis exilis* 'geothermal grass' which is widespread in the area and is also common in other geothermal areas within the main Kenya Rift Valley. The soils of these areas are mainly clay and in some parts clay mixed with sand.

In this study, LSC technique was used to quantify the concentration of radon in water samples from both geothermally active and non-geothermally active high background radiation area (HBRA) of Winnam Gulf (Kenya). LSC was used because it has high sensitivity, requires small sample volumes of 10 ml and takes small counting time [7]. Principal components analysis (PCA) was used for data dimensionality reduction and deciphering clusters of radon concentration within the set of analyzed data. Hierarchical cluster analysis (HCA) was used to partition the activity concentration of radon into non overlapping clusters in order to assess whether they are consistent with the clusters displayed by PCA. Soft independent modeling of class analogy (SIMCA) was used to identify local models for possible groups of water samples and to predict a probable class membership for new observations from the same geological area. These chemometrics techniques were aimed to apportion the analyzed data according to the source.

2. METHODOLOGY

2.1. Sample collection and handling

Ground and surface waters were sampled in the Winnam Gulf region of Kenya indicated in Fig. 1. The sampling points were systematically selected, but their locations were determined using the global positioning system (GPS).
Water samples from each sampling point were collected using 100 ml standard EPA type water collection bottles. Samples taken at each location were given identification codes while noting time and date of sampling. The samples were transported to the Kenya Radiation Protection Board laboratory while keeping track of the transportation time.

Figure 1: Geology of Winnam Gulf of Kenya.

2.2. Liquid scintillation counting

10 ml of Ultima Gold™ cocktail was measured and placed in 24 ml plastic vials. A needle of a 20 ml hypodermic syringe was inserted below the water surface and several milliliters of water withdrawn and discarded. This rinse was repeated several times. 12 ml of water was withdrawn slowly to minimize air bubbles. The syringe was inverted to eject any air bubbles and retain 10 ml of water. The syringe needle was placed under the surface of 10 ml of Ultima Gold™ cocktail contained in a plastic scintillation vial and water ejected slowly from the syringe into the cocktail [8]. The vial was tightly capped and vigorously shaken. During shaking almost all ^{222}Rn is extracted from water to the cocktail, while other radionuclides, elements or minerals remained in the water [9]. The above procedure was repeated three times for each sample. The radon partition coefficient for water:Ultima Gold™ cocktail:air is 1:48:4. The prepared samples were left for at least four hours for radon to equilibrate with its daughter nuclide before they were quantitated by LSC [10].
The activity concentration of radon was calculated as follows:

$$RnC = \frac{100(N)\exp^{(\lambda t)}}{60\times5\times0.949} \quad (1)$$

where RnC is ^{222}Rn concentration at the time of sample collection (Bq L^{-1}); N is the sample total count rate (count min.$^{-1}$); t is the elapsed time between sample collection and counting (min.); λ is ^{222}Rn decay constant (1.26 x 10^{-4} min.$^{-1}$); 100 is a conversion factor of 10 ml to per liter (L^{-1}); 60 is conversion factor from min. to sec. (s. min.$^{-1}$); 5 (500 %) is the number of emissions per disintegration of ^{222}Rn (3α and 2 β, assuming 100 % detection efficiency for each); and 0.949 is the fraction of ^{222}Rn in 10ml of Ultima Gold™ cocktail in a vial of 24 ml total capacity.

2.3. Principal components analysis (PCA) of activity concentration of radon in relation to pH and temperature

PCA was performed on the activity concentration of radon in ground and surface waters from both geothermally active and non-geothermally active HBRA using Unscrambler Camo 9.7 software. A plot of principal components displayed patterns which enabled us to view interrelationships between the different variables, and detect and interpret sample, groupings, similarities or differences [11], [12]. Before the PCA was performed, the data was pre-processed through taking the logarithms of the variables so that the distribution of skewed variables is improved, smoothing the variables using median filter in order to remove noise from the variables, and mean centering of the variables so that the result is interpretable in terms of variation around the mean. The pre-processed data was transformed into a product of two matrices according to equation 1, one of which contains information about the samples and the other about the variables.

$$\mathbf{X = T \cdot P^t + E} \quad (2)$$

where X is the data matrix, T is the factor scores, P is the factor loadings, P^t is the transposed matrix of P, E is residuals as a result of reduction of dimensionality [13 — 15].

The PCA biplot showed the variance, the scores and the loadings in the same plot. The variance revealed how much of the information in the data table was described by the model whereas the loadings described the data structure in terms of variable correlation. The scores described data structure in terms of sample patterns, and they showed sample differences and similarities. Samples with close scores along the same PC were similar (they had close values for the corresponding variables). Conversely, samples with score differences much higher were quite different from each other with respect to those variables [16].

2.4. Hierarchical cluster analysis (HCA) of activity concentration of radon

HCA plot was performed on the activity concentration of radon in waters from both geothermally active and non-geothermally active HBRA using Matlab software. The HCA technique examined interpoint distances between samples and radon levels and presented the information in the form of a dendrogram [17]. The goal of HCA was to present the data in the form that facilitates the use of human pattern-recognition abilities by forming similarity groups which have a physical interpretation such as for example source apportionment of the waters. An advantage of HCA method over the PCA lies in its use of all the variation in the data in determining similarity; in PCA, the dimension of the data is first reduced before approximating similarity.

2.5. Soft independent modeling of class analogy (SIMCA)of activity concentration of radon

SIMCA was performed on the activity concentration of radon data using Unscrambler Camo 9.7 software. Validated PCA models were built for each class (waters: ground, surface; regions: Bala, Kanam, Kanjera) which describe the structure of those classes as accuratelyas possible [18], [19]. The data set in each class was systematically divided into two parts, calibration samples and test samples. The test samples were used to perform the role of unknown samples.

The modeling power of the activity concentration of radon in geothermal waters was determined. This was aimed at determining the influence of radon levels on Bala, Kanam and Kanjera classification models. The modeling power was calculated using equation 2.

$$mp = 1 - \sqrt{\frac{\sigma_r^2}{\sigma_t^2}} \quad (3)$$

where σ_r^2 and σ_t^2 are residual and total variances respectively.

Avariable with modeling power of more than 0.3 is considered to be more relevant for that model; a variable with modeling power less than 0.3 is considered to be less relevant for the model and it should be left out when constructing that model [20].

3. RESULTS AND DISCUSSION

3.1Activity concentration of radon

Results of the activity concentrations of radon in groundwaters and surface watersfrom geothermally active HBRA and non-geothermally active HBRA of Winnam gulf are summarized in Table 1 and Table 2. The minimum activity concentrations of radon observed in groundwaters, and surface waters were 19.8 ± 1.9 Bq/L and 8.7± 0.6 Bq/L respectively, and the maximum values were 46.3± 1.4 Bq/L and 24.1± 0.3 Bq/L respectively. The mean concentrations of radon in groundwaters from geothermally active HBRA and non-geothermally active HBRA were 23.2 Bq/L and 43.8 Bq/L respectively, and the mean concentrations in surface waters from geothermally active HBRA and non-geothermally active HBRA were 9.2 Bq/L and 21.9 Bq/L respectively.

Table 1: The activity concentration of radon in ground waters

GE0THERMALLY ACTIVE HBRA		NON-GEOTHERMALLY ACTIVE HBRA	
SAMPLE	^{222}Rn (Bq/L)	SAMPLE	^{222}Rn (Bq/L)
H3	26.7 ± 1.9	H2	43.0 ± 3.5
H8	24.5 ± 1.1	H5	43.9 ± 0.8
H10	25.3 ± 0.7	H7	45.2 ± 3.4
H14	24.6 ± 2.7	H9	44.9 ± 1.9
H17	22.2 ± 0.7	H12	45.3 ± 0.9
H22	21.4 ± 3.8	H20	42.4 ± 1.3
H23	21.3 ± 2.3	H21	41.3 ± 2.3
H31	23.3 ± 2.7	H27	46.3 ± 1.5
H34	19.8 ± 2.0	H32	42.7 ± 3.4

Table 2: The activity concentration of radon in surface waters

GEOTHERMALLY ACTIVE HBRA		NON-GEOTHERMALLY ACTIVE HBRA	
SAMPLE	^{222}Rn (Bq/L)	SAMPLE	^{222}Rn (Bq/L)
H4	9.5 ± 0.8	H1	21.3 ± 0.8
H11	9.5 ± 0.7	H6	21.0 ± 0.9
H13	9.1 ± 0.8	H15	20.7 ± 1.3
H16	9.0 ± 1.8	H18	24.2 ± 0.4
H24	9.4 ± 1.4	H19	21.5 ± 1.6
H29	9.3 ± 0.8	H25	22.9 ± 1.0
H30	8.7 ± 0.7	H26	23.4 ± 1.9
H33	9.2 ± 0.8	H28	20.5 ± 0.9

It was observed that the activity concentrations of radon in water samples from geothermally active HBRA were higher in ground water samples than in surface water samples. This is because the sources of these ground water samples are in contact with rock matrix containing radium which is the source of radon; hence more radon atoms will dissolve into the ground water samples. The lower concentrations of radon in surface water samples is as a result of the partitioning of radon into any separate volatile phase making it to degas from waters exposed to

the atmosphere. Radon is four times soluble in air than in water [9], [10].

Figure 2 indicates that PC1 in the score plots capture 99% of the variation contained in the original data and PC2 explains only 1% of the variation which may be regarded as noise. It is observed that the groundwater samples are classified into two distinct clusters: geothermally active HBRA and non-geothermally active HBRA. From the loadings plot, it is observed that radon concentration loads heavily to PC1. This shows that the distinction of the two clusters is based on the levels of radon concentration inground water samples.

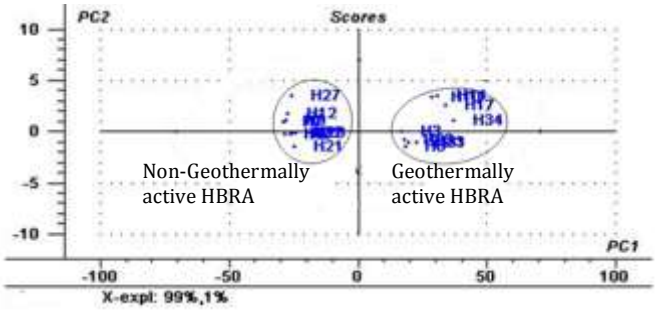

Figure 2: PCA score plots of the activity concentration of radon in ground water samples from Winnam Gulf (Kenya).

Figure 3 indicates that PC1 in the score plots explain 99% of the variation contained in the original data and PC2 explains only 1% of the variations which is regarded as noise. There are also two distinct clusters; geothermally active HBRA and non-geothermally active HBRA.These clusters are based on the activity concentrations of radon in surface water samples. The clusters are similar to those in Figure 2. This means that PCA can apportion the analyzed water samples according to the source on the basis of radon concentration.

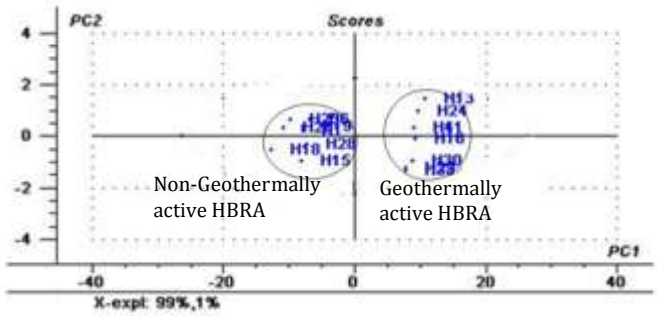

Figure 3: PCA score plots of the activity concentration of radon insurface water samples from Winnam Gulf (Kenya).

It is observed that PCA is a powerful tool in revealing patterns and alterations in radon which are distinct with respect to geological sites. In this study, the clusters which are a characteristic of geothermal activity, are clearly distinguished using only PC1. In the two cases (both ground and surface waters) PC1 explains more than 95% of the variation in the original data.

Hierarchical cluster analysis was performed on the activity concentration of radon in ground water and surface water samples in order to assess whether the samples will cluster in relation to the geological sites (source apportionment of the waters). The HCA results obtained are presented in Figures (4-5).
Two major clusters are observed in Figure 4 which are formed on the basis of the activity concentration of radon. These clusters are like in the case of PCAgeothermally active HBRA and non-geothermally active HBRA clusters. The geothermally active HBRA cluster has three sub clusters (H22, H23, H17, H31; H8, H14, H10; and H3, H34). The non-geothermally active cluster has also three sub clusters (H2, H20, H32, H5; H7, H12, H9; and H21, H27). This sub-clustering did not follow a specific trend indicating that other factors could be influencing the clustering. Thus, supporting the fact that the variability in the activity concentration of radon in ground waters is a function of the geology of the area, depth of the sampled waters and season [21].

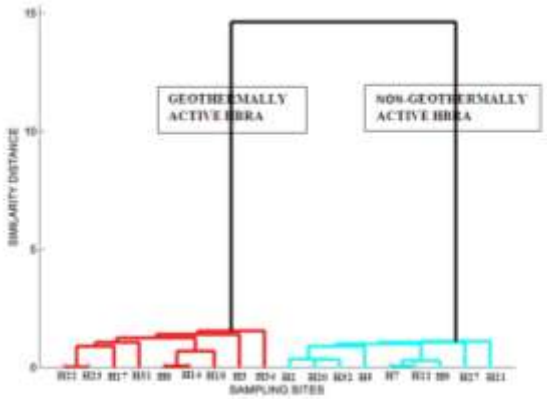

Figure 4: Dendrogram of the activity concentration of radon in ground water samples from Winnam Gulf (Kenya).

Figure 5 also reveals two major clusters for surface water samples. These clusters are, similarly, geothermally active HBRA cluster and non-geothermally active HBRA cluster. The non-geothermally active HBRA cluster has two sub clusters (H1, H19, H6, H15, H28; and H18, H25, H26). These clusters also indicate that the level of radon in water among other factors such as temperature and salinity [22].

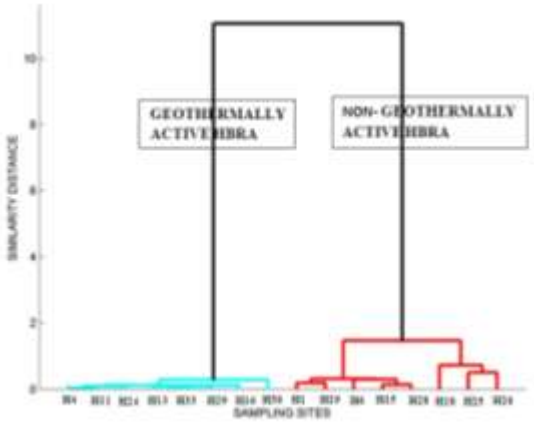

Figure 5: Dendrogram of the activity concentration of radon in surface water samples from Winnam Gulf (Kenya).

3.2.Soft independent modeling of class analogy (SIMCA)for Winnam Gulf geothermal waters

From the PCA and HCA results, it was observed that geothermal water samples exhibit distinct clusters based on the activity concentrations of radon. These clusters are a characteristic of either geothermally active HBRA or non-geothermally active HBRA. Therefore, there was the need to make SIMCA classification models based on the activity concentrations of radon in water samples, which can be used to predict and classify samples into probable class membership. This was aimed at to identify the type of water (ground, surface) in a typical HBRA field based on the activity concentrations of radon.

In order to construct SIMCA, Unscrambler Camo 9.7 software was used to construct PCA models for ground and surface. The PCA models for water samples from Bala, Kanam and Kanjera were also constructed. Before these models were constructed, the water samples were divided systematically into training set and test set. The models were validated using cross validation approach. Test samples were projected onto each model. The samples were classified as members if they were similar enough in terms of the activity concentration of radon to the corresponding model. The results obtained are presented in Table 3.

Table 3: SIMCA classification of Winnam Gulf geothermal watersbased on radon concentration (* indicates where the sample is classified)

Sample	GROUND	SURFACE	BALA	KANAM	KANJERA
H7	*		*		
H10	*		*		
H22	*				*
H31	*		*	*	
H9		*	*	*	
H11		*	*	*	*
H25		*	*	*	*
H32		*	*	*	*
H1					
H16					*
H18					*

Samples H1, H7, H10 and H22 were classified as true positive while samples H9, H11, H16, H18, H25, H31 and H32 were classified as false positive. In order to determine the model in which the false positive samples belong to, the Cooman's plot was performed on Bala, Kanam and Kanjera models [20], [23]. Table 4 shows the result obtained after the Cooman's plot.

Table 4: SIMCA classification of Winnam Gulf geothermal watersbased on radon concentration after the Cooman's plot.

Sample	GROUND	SURFACE	BALA	KANAM	KANJERA
H7	*		*		
H10	*		*		
H22	*				*
H31	*		*		
H9		*	*	*	
H11		*	*	*	*
H25		*	*	*	*
H32		*	*	*	*
H1					
H16					
H18					

Table 4 shows that only samples H16, H18 and H31 are now additionally assigned to correct models after the Cooman's plot. Samples H9, H11, H25 and H32 were incorrectly assigned to one or two wrong classes that were not always neighbors. This misidentification of the water samples could indicate that other variables also influence the levels of radon in geothermal waters. The SIMCA classification was only 64% efficient based on the test samples which were assigned to the correct models. This indicates that the SIMCA classification is not sufficient to classify samples using the PCA models constructed in this

work. Therefore, there was the need to diagnose the models of the geothermally active HBRA using the modeling power plot of the activity concentrations of radon. This was aimed at assessing how the radon levels influence the classification of Bala, Kanam and Kanjera models.

After the diagnosis it was observed that the activity concentrations of radon has a modeling power of more than 0.95. This suggests that, the only chance to improve on the classification of samples in order to diagnose geothermal potential in a typical HBRA based on the activity concentrations of radon is to measure some additional variables such as SiO_2, B, Na, K, Ca, Mg, Fe, Al, CO_2, SO_2, H_2S, Cl, F, Li and NH_3 which determine the chemical composition of thermal waters and include them in the construction of classification models [24], [25].

4. CONCLUSION

Liquid scintillation counting method was used to measure the activity concentration of radon in ground and surface waters from geothermally active and non-geothermally active HBRA of the Winnam Gulf, Kenya. The mean and maximum values of the activity concentration of radon in water samples from geothermally active and non geothermally active HBRA are 16.5± 2.0Bq/L, 18.5± 3.5 Bq/L and 39.7± 1.3 Bq/L, 43.9± 0.8 Bq/L respectively. The activity concentration of radon in non geothermally active HBRA was higher than the activity concentration of radon in geothermally active HBRA. This was attributed to the variation temperature of the geological sites as a result of geothermal activities.

Principal components analysis (PCA) of groundwaters and surface waters showed distinct clusters based on the activity concentration of radon which are a characteristic of the geological sites. Waters from geothermally active HBRA and non-geothermally active HBRA was segregated into distinct clusters. Hierarchical cluster analysis (HCA) results were similar to those of PCA even though these results were based on the activity concentration of radon before pre-processing. The additional value of the HCA result is that it re-affirmed group consistency based on the activity concentration of radon;therefore, the activity concentration of radon could be used to classify geological sites by successfully segregating dissimilar data.

Soft independent modeling of class analogy (SIMCA) was used to construct classification models aimed at classifying samples according to geological sites based on the activity concentration of radon. The prediction and classification efficiency of SIMCA was 64% indicating that the models are unreliable; hence more variables are required.Future measurements of radon should include gas and steam condensates from fumaroles. There is also need to measure some additional variables such as chemicals and isotopic components in geothermal waters and use them together with the radonactivity concentrations to construct multivariate chemometrics models which could diagnose geothermal potential and reservoir characteristics based on radon levels.

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