

Multivariate statistical approximation of the in situ gamma-ray spectrometry of the State of Zacatecas, México

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Abstract

The environmental radioactivity evaluation is a key point in the assessment of the environmental quality. Through this, it can be found possible radioactive contamination, locate possible Uranium and Thorium deposits and evaluate the primordial isotopes concentration due to human activities. A radioactive map of the State of Zacatecas, México is under construction based on *in situ* gamma-ray spectrometry. The present work reports the results of the multivariate statistical approximation of the measured activity data. Based on Pearson's correlation, the ^{228}Ac and ^{208}Tl activities are statistically significant, while the ^{214}Bi and ^{214}Pb activities are not statistically significant. These can be due to the existence or not of secular equilibrium in the Thorium and Uranium series.

1. INTRODUCTION

An environmental radioactivity map is under construction, in order to evaluate the possibility of a radiological risk, and in case of, its origin and nature. The preliminary radioactivity map results [1] have shown no risk at all. The resulting specific activity values in Bq/kg are 24433.61 ± 4.65 for ^{40}K , 84.85 ± 0.86 for the ^{232}Th series and 60.91 ± 0.33 for the ^{238}U . These values are in the order of the data found in literature, as well the corresponding average dose rate of 180.87 ± 0.58 nGy/hr. On the other hand, the average specific activity of ^{137}Cs of 4.78 ± 0.19 Bq/Kg is between limits.

Whilst *in situ* and mobile gamma spectrometry techniques have been shown to provide rapid and spatially representative estimates of environmental radioactivity across a range of landscapes [2], the technique has been criticized for the occasional lack of correspondence with data derived from conventional analyses of soil samples. Through these effort, *in situ* and laboratory gamma-ray spectrometry have been used with the aid of Neutron Activation Analysis [3] for the assessment and calibration of data.

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In the present work, a multivariate statistical analysis of the in situ gamma-ray activities is presented. The multivariate statistics which imply the Principal Components Analysis (PCA) and the Cluster Analysis (CA), were used to determine the correlations in the measured activities. The analysis were supported by univariate statistics and linear regression relationships. Data were processed through the normalized Varimax method for the PCA evaluation. SPSS.23 software were used and cluster analysis and Pearson correlation were obtained in order to highlight the relationships between the variables and the distribution of the radionuclides under study.

2. EXPERIMENT AND DATA ANALYSIS

The in situ gamma-ray spectrometric system is based on a CanberraGeRe-3522 detector mounted in a BIG MAC 7 portable cryostat. An AccuSpec A computer board is used to the acquisition of the gamma-ray spectra using Genie software and a sequence of analysis customized for the environmental gamma-ray spectra is used to determine the activities of interest.

^{40}K were determined using its characteristic line of 1460.822 keV. For the Thorium series, ^{208}Tl (583.187 and 860.56 keV) and ^{228}Ac (911.196, 968.960 keV) were used. The Uranium series were evaluated trough ^{214}Pb (351.932 keV), ^{214}Bi (295.224 and 609.312 keV) [4]. The acquisition time for every spectrum was set to 7200 s, since it is enough time to achieve a standard error less than 2% under the ^{40}K photopeak. Energy and efficiency calibration of the spectrometer were done using a multinuclide standard with ^{241}Am (59.5 keV); ^{109}Cd (88.3 keV); ^{137}Cs (661.6 keV) and ^{60}Co (1173.23 y 1332.51 keV).



Figure 1. Zacatecas State location

2.1. Location and types of measurement points

The state of Zacatecas is located in the central-north region of the Mexican Republic, neighboring with the Jalisco and Aguascalientes states by the south, San Luís Potosí at east, Coahuila at north and Durango at northwest and west. The state has a surface of 75,040 km², about 3.9% of the whole country, and an average height of 2,100 m above sea level (asl). The extreme coordinates are 25°09' (25.15) N, 21°04' (21.066) S, 100°49' (100.82) E and 104°19' (104.31) W. Zacatecas

has a surface which 39.06 % comes from the Cenozoic age, Quaternary period mainly composed by Sedimentary rocks. A 38.66 % belongs to the Cenozoic age, Tertiary period composed by Extrusive Igneous rocks. The remaining 22.28 % belongs to the Mesozoic age, Tertiary period, composed by metamorphic rocks [5]. Figure 1 shows the Zacatecas location. Fourteen different measurement sites have been included in this work, Table I shows the location, altitude and type of the measured points. ^{137}Cs and other rainfall radionuclides have been used worldwide over the last five decades to evaluate soil redistribution spatial patterns and magnitude since they have been deposited globally. [6-7]

Table I. Measuring points coordinates and type

Site	Longitude	Latitude	Altitude asl (m)	Type
C. de la Virgen	-102.5695	22.7417	2666	Urban
La Condesa	-102.4969	22.7602	2275	
La Zacatecana	-102.4865	22.7224	2236	
Siglo XXI	-102.644	22.7739	2307	
Vetagrande	-102.5523	22.8255	2632	
Altavista	-103.9447	23.4780	2172	Archaeological
La Quemada	-102.8204	22.4553	2121	
Las Lajas	-102.8455	22.6838	2151	Rural
San Benito	-101.7213	23.9030	1957	
San Ramón	-102.525	22.6765	2312	
Sombrerete	-103.6606	23.6570	2440	
Villa de Cos	-102.3592	23.3026	1979	
Ojocaliente	-102.5835	22.5835	2108	
Pánfilo Natera	-102.5571	22.3899	2090	

A descriptive statistical study was carried out on the activities measured in the selected sites. Table II shows the results of the analysis.

Table II. Statistics of the activities

	^{214}Bi	^{214}Pb	^{228}Ac	^{208}Tl	^{137}Cs	^{40}K
Number of data	14	14	14	14	14	14
Mean (Bq)	70.922	68.996	98.096	27.280	4.629	2269.151
Mean standard error	15.373	6.891	16.64	4.439	0.709	279.316
Median	55.077	71.2	95.17	26.89	3.412	2367.41

Mode	11.979 ^a	13.909 ^a	9.320 ^a	2.848 ^a	2.052 ^a	445.461 ^a
Standard deviation	57.52	25.784	62.26	16.608	2.651	1045.105
Variance	3308.517	664.828	3876.241	275.831	7.029	1092243.548
Asymmetry	2.945	-0.032	0.725	0.619	1.236	-0.010
Standard error of asymmetry	0.597	0.597	0.597	0.597	0.597	0.597
Kurtosis	10.022	1.252	0.391	0.167	0.934	-0.778
Standard error of Kurtosis	1.154	1.154	1.154	1.154	1.154	1.154
Range	246.013	103.624	223.389	58.891	8.868	3587.460
Minimum	11.979	13.909	9.320	2.848	2.052	445.461
Maximum	257.992	117.533	232.709	61.739	10.921	4032.921

By applying the PCA, a Pearson's correlation was obtained (Table III)

Table III. Correlation matrix

Correlation	¹³⁷ Cs	⁴⁰ K	²¹⁴ Bi	²¹⁴ Pb	²²⁸ Ac	²⁰⁸ Tl
¹³⁷ Cs	1.000	-0.377	0.603	0.550	-0.056	-0.044
⁴⁰ K	-0.377	1.000	-0.354	0.311	0.407	0.403
²¹⁴ Bi	0.603	-0.354	1.000	0.449	0.540	0.554
²¹⁴ Pb	0.550	0.311	0.449	1.000	0.549	0.549
²²⁸ Ac	-0.056	0.407	0.540	0.549	1.000	0.999
²⁰⁸ Tl	-0.044	0.403	0.554	0.549	0.999	1.000

The ²²⁸Ac and ²⁰⁸Tl relationship is highly significant, which indicate that the secular equilibrium in the Thorium series has a minor perturbation. For the ²³⁸U case, one can observe that its daughters activities, ²¹⁴Bi and ²¹⁴Pb, don't keep a constant relationship, which indicate a disequilibrium condition for this series. On the other hand, ¹³⁷Cs and ⁴⁰K seem to behave randomly, since they did not sympathize with the rest.

The Rotated component matrix shown in Table IV show for the ²¹⁴Bi and ²¹⁴Pb radioisotopes that the first component coincides while the second does not agree, indicating the disequilibrium condition. For the ²²⁸Ac and ²⁰⁸Tl case, there is a clear indication of secular equilibrium for the ²³²Th series. ¹³⁷Cs and ⁴⁰K are far apart, since there is no relationship between them and do not share any similarity with the other components. All these can be seen in Figure 2.

The graph of rotated components nods a more panoramic view of the isotopes according to their rotation where we see that the ²²⁸Ac and ²⁰⁸Tl are placed on one another.

Table IV. Rotated component matrix

	Component		
	1	2	3
²¹⁴ Bi	0.722	-0.643	0.121
²¹⁴ Pb	0.738	0.066	0.057
²²⁸ Ac	0.942	0.187	-0.071
²⁰⁸ Tl	0.946	0.176	-0.046
¹³⁷ Cs	-0.019	0.055	0.994
⁴⁰ K	0.434	0.864	0.134

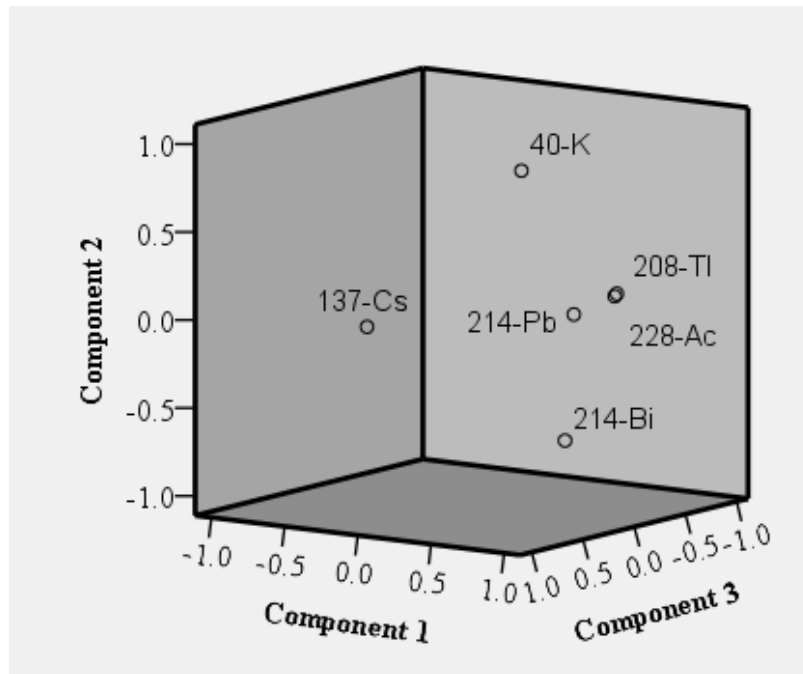


Figure 2. Components graph in rotated space

One of the multivariate techniques used to identify and classify groups with similar characteristics in a new type of observations is the cluster analysis [7]. Applied over the measurement sites results in Figure 3. Zero separation distance indicates that the clusters are 100% similar in their sampling measurement, while for very despair grouping areas the similarity is of 0%. The cluster analysis was performed through axes to identify similar characteristics between the radioisotopes and the radiologic parameters in the different grounds of the state. The dendrogram is made up by three, a first one containing the majority of the sites indicating soil similarities, while Veta Grande, La Quemada and Ojocaliente are the isolated ones.

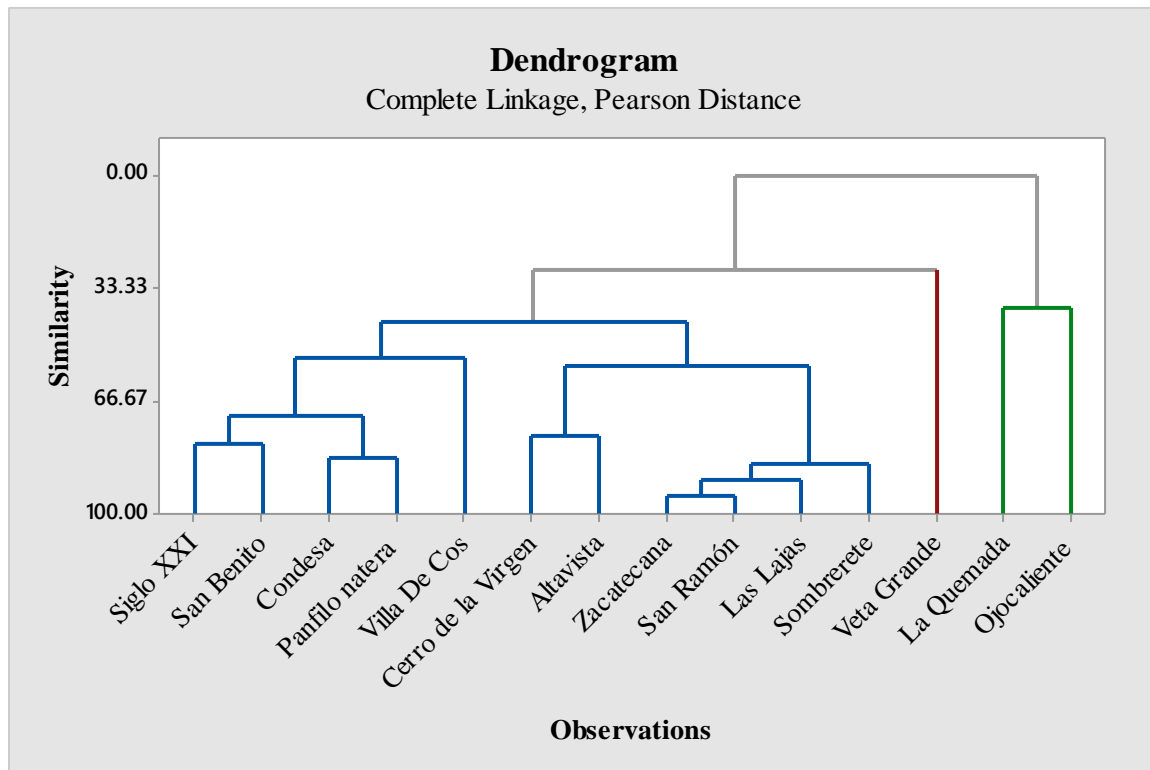


Figure 3. Dendrogram

3. CONCLUSIONS

Measurements of in situ gamma-ray spectrometry in the state of Zacatecas were carried out in sites of the type Archaeologic, Urban, and Rural indicate a perturbation in the secular equilibrium of the Uranium (^{238}U) radioactive series, while the perturbation in the Thorium series is not observable. Statistical results in Table III and IV show the high significant for ^{228}Ac and ^{208}Tl . Disequilibrium in ^{238}U series could be due to the ^{222}Rn long half-life (3.8235 d) compared to the one of ^{220}Rn (18 ms). Rainfall differences together with soil type account for the ^{137}Cs presence

The present work shows how the statistical analysis can be used to disclose the differences in the environmental radioactivity measurements.

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