# 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 Varimx 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 las 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 | |
| PánfiloNatera | -102.5571 | 22.3899 | 2090 | |

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

The Rotated component matrix shown in Table IV show for the $^{214}$Bi and $^{214}$Pb radioisotopes that the first component coincides while the second does not agree, indicating the disequilibrium condition. For the $^{228}$Ac and $^{208}$Tl case, there is a clear indication of secular equilibrium for the $^{232}$Th series. $^{137}$Cs and $^{40}$K 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 $^{228}$Ac and $^{208}$Tl 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 |

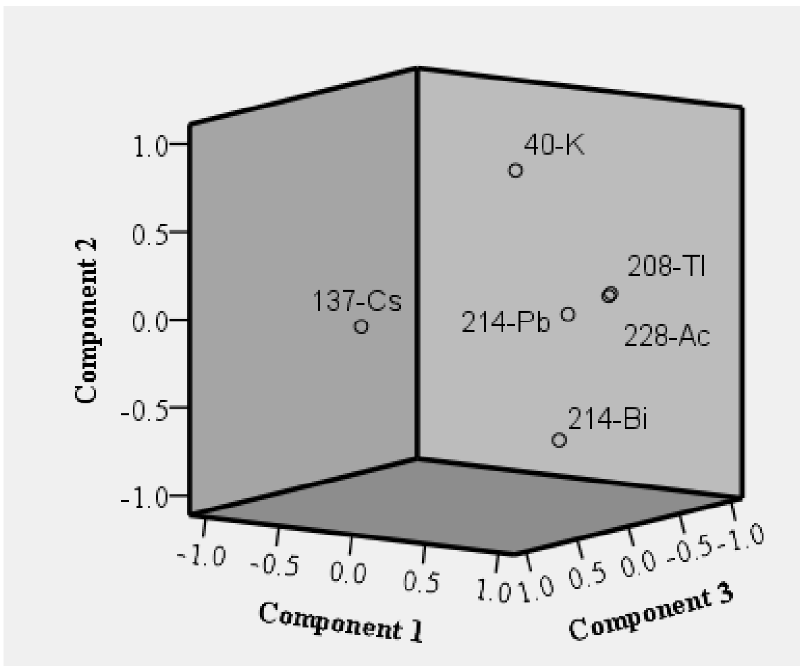


**Figure 2. Components graph in rotated space**

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. Cero 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.

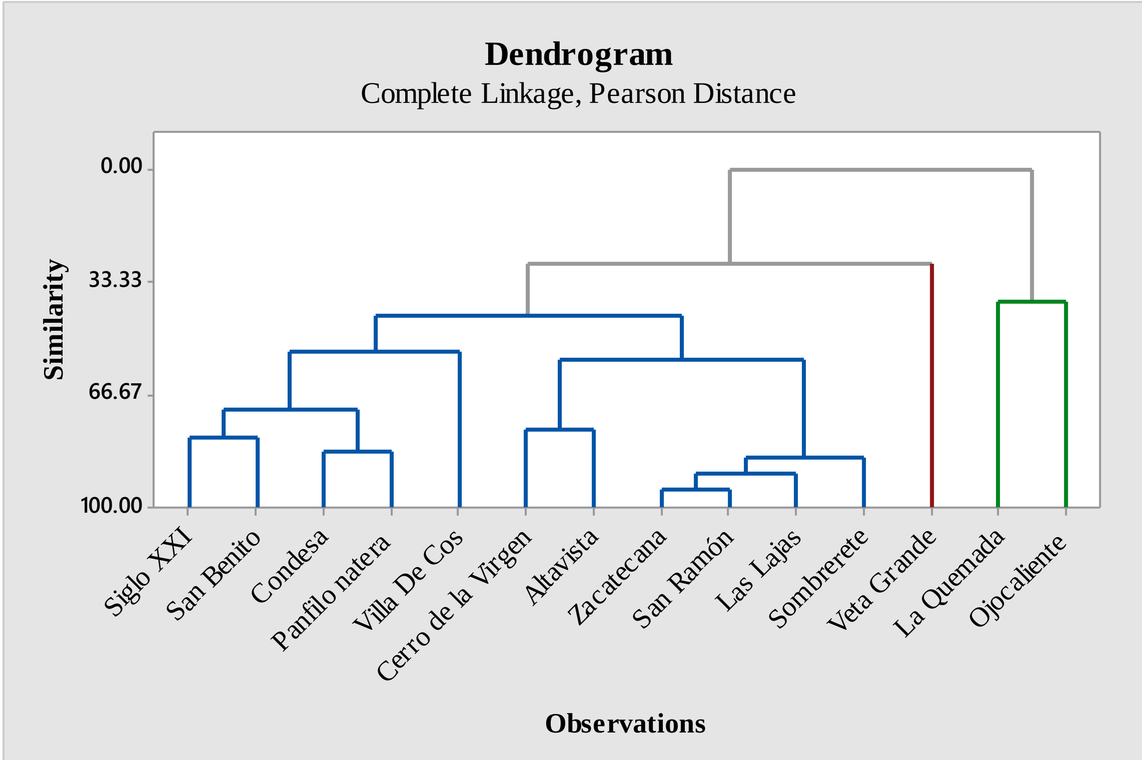


**Figure 3. Dendogram**

## 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. Statiscal 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 220Rn (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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