Application of Multivariate Statistic of U, Th and Pb Concentrations and Pb Isotopic Signatures in the Assessment of Geogenic and Anthropogenic Sources in a U-Mineralized Area

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Abstract

This work presents a Principal Component Analysis (PCA) study using Pb-isotope signatures and U, Th and Pb concentrations from groundwater, sediments and rocks (granites and orthogneisses) of the Complex of Lagoa Real (Bahia, Brazil). This area is naturally enriched in U and Th, with the occurrence of Pb derived from the radioactive decay of the elements $^{238}\text{U}$, $^{235}\text{U}$ and $^{232}\text{Th}$ in the form of their stable isotopes $^{206}\text{Pb}$, $^{207}\text{Pb}$ and $^{208}\text{Pb}$ in addition to the natural isotope $^{204}\text{Pb}$. Sampling was carried out in the rainy season (December to January) and the points were selected according to regional hydrology and geology. Thirty samples were analyzed: 12 of groundwater (AP) and 18 of sediments (S). The results show that the use of isotopic ratios allows discrimination between geogenic and anthropogenic samples. This information is not obtained using only the analysis of concentration data. Statistically, the isotopic data of Pb stand out as an efficient tool in the characterization of sources in the scenario investigated, allowing an effective environmental monitoring and a better management of the mining activities.

Keywords

Environmental Monitoring, Statistical Analysis, Lead Stable Isotopes, Anthropogenic Sources, Geogenic Sources

1. Introduction

The Uraniferous Province of Lagoa Real (Bahia, Brazil) is an area naturally

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enriched in U and Th, with the occurrence of Pb derived from the radioactive decay of U ($^{238}\text{U}$ e $^{235}\text{U}$) and Th ($^{232}\text{Th}$), in the form of their stable isotopes $^{206}\text{Pb}$, $^{207}\text{Pb}$ and $^{208}\text{Pb}$ respectively, in addition to the presence of the natural isotope $^{204}\text{Pb}$. These mineralizations in uranium are the basis of the mining activity of this element, largely responsible for the economic development of the region. In these areas, the groundwater represents the main source of water for the population, since water scarcity is significant.

According to Licht (2001), most of the natural or anthropic processes occurring on the Earth’s surface are characterized by associations of chemical elements (characteristic of the generating process) and geographically associated to their area of occurrence. However, these geochemical signatures may be masked by the superposition of other ones from different sources. Thus, the identifiable geochemical signal, e.g. in waters and sediments, would represent a sum of contributions originated from the natural environmental processes and human action. According to this author, when a complex geochemical signal is interpreted using a multidisciplinary approach, it is possible to decompose it, and, hence, natural or anthropogenic processes can be identified and isolated.

In order to investigate the correlation between a given element and the source that originated it (natural or anthropogenic), works from the 1960s onwards showed the applicability of the methodology of Pb isotope ratios in environmental studies (Nriagu, 1989). Considering that physical or chemical processes do not affect the isotopic composition of Pb in the environment (Doe, 1970; Bollhöfer et al., 1999), the Pb signature tends to be identical to the source that originated it. Therefore, the isotopic signatures of Pb of anthropogenic provenance are distinct from the isotopic signature of natural origin, and can be used as a tracer of the possible sources of Pb in the environment (Bollhöfer et al., 1999; Bollhöfer and Rosman, 2000; Bollhöfer and Rosman, 2001; Chen et al., 2005; Gioia et al., 2006; Gulson et al., 2007; Komárek et al., 2008; Gioia et al., 2010; Vecchia, 2015; Vecchia et al., 2015; Vecchia et al., 2017a and 2017b).

In the area of the Gneissic-Granitic Complex, Cordani et al. (1992) obtained data on isotopic composition of Pb in granite rocks of the São Timóteo type (non-deformed granites) and in orthogneisses (deformed granites and associated with uraniferous mineralization). Subsequently, Iyer et al. (1999) demonstrated the applicability of Pb isotopes in the quantitative determination of U and Th mobility in different geological formations present in the São Francisco Craton. These authors studied several geological environments belonging to the São Francisco Craton, among which is the Lagoa Real Gneissic-Granitic Complex, using the isotopic data of Pb published by Cordani et al. (1992).

According to Vecchia (2015), Vecchia et al. (2015) and Vecchia et al. (2017a and 2017b), the use of the Pb isotopic tool to study environmentally impacted areas requires knowledge of the Pb isotopic signatures of geographic samples as a determinant factor in the investigative process of environmental matrices. Therefore, this study uses the Pb isotopic data of geographic samples of the study area obtained by Cordani et al. (1992) along with the U, Th and Pb concentra-
tion data from Iyer et al., 1999. The isotopic data of Pb produced by Cordani et al. (1992), containing information on the preserved geological environment, were compared to the isotopic data of Pb from Vecchia et al. (2017b), in order to diagnose influences of geogenic and/or anthropogenic sources in this area.

Statistical analyses allow the specification of the variables that best characterize the geogenic influences coming from the geological context and/or the anthropic activities in the environment. The multivariate statistical methods allow the extraction of useful information capable of reducing the dimensional representation of the data, organizing them in a structure that facilitates the visualization of the whole data set. Principal Component Analysis (PCA) is among the most widely used with this purpose. It compacts the data into a smaller number of variables (principal components) whose variance (and importance) is given by their eigenvalues. These components are mathematical functions that generate scores that have embedded the contributions (and importance) of each variable with its weight (loadings) (Manly, 2005; Mingoti, 2007; Zhao et al., 2011; Cavalcante et al., 2013; Sun et al., 2013; Siepak and Sojka, 2017). It is important to emphasize that the results obtained with these techniques should, whenever possible, be confronted with information related to the research context, seeking an association on the possible processes in the study region (Vecchia, 2015; Vecchia et al., 2015; Vecchia et al., 2017a and 2017b).

This work presents a statistical study based on the PCA, using Pb isotopic ratios, and U, Th and Pb concentrations from groundwater and sediments in the Lagoa Real Uranium Complex (Brazil). The main objective is to refine the information from the investigations carried out by Vecchia et al. (2017b) by analyzing the contributions of each variable in the diagnosis of possibly impacted areas in the Uraniferous Province area. This work aims to diagnose not only the water quality and sediments in the study area, but also to identify the areas possibly affected by the mining activity.

2. Study Area

The study area, Southeastern Brazil, is located in the semi-arid region characterized by a marked water deficit. It is considered the main active site of uranium in Brazil and South America. Pb isotopic signature of samples was determined in an area of 1200 km² at Lagoa Real Uranium Complex, Bahia State, Brazil (Figure 1). This complex is geologically identified as a Gneissic-Granitic Complex with ages between of 1.72 and 1.77 Ga (Cordani et al. 1992). Granite type São Timóteo (undeformed granites), orthogneiss (deformed granites and associated U-mineralization) and U-albitites (main hosts of the uranium mineralization) are the principal lithologies of the region and represents the geogenic sources (Vecchia et al., 2017b).

The average annual rainfall is around 700 mm, with high variability in the spatial and temporal distribution (annual seasonality) (Vecchia, 2015). According to Lamego Simões et al. (2003), the climatological and hydrological characteristics,
Figure 1. Simplified geologic map with the main lithologies representative of the geogenic sources in the Lagoa Real Uranium Complex: Albitites, Orthogneiss and São Timóteo Granites and the sample locations of sediments and groundwater. Modified by Vecchia et al. (2017b).

associated to the conformation of the regional relief with flows to Atlantic slope, give rise to a hydrographic network in which intermittent drainage is common (Figure 1). Complementary hydrological data and geology of the study area can be found in Vecchia (2015).

3. Experimental Methods

3.1. Sampling

Sampling was carried out according to Vecchia et al. (2017b). Groundwater and sediment sampling points were selected according to regional hydrology and geology, including points at the mining area and points located upstream and downstream of the U anomalies (contents higher than 1500 ppm). The deficit of surface water led to the choice of groundwater and sediment as study matrices.
Sampling was carried out in the rainy season (December to January) because of the discontinuous flow of the rivers and the low precipitation level in the region (around 700 mm). The location of sampling was devised aiming to achieve a reliable geographical representation and to ensure a good spatial distribution of the data. Thirty samples were analyzed; 12 of groundwater (AP) and 18 of sediments (S) as shown in Figure 1. The sampling points were georeferenced, using GPS (Global Positioning System).

The sampling and preservation of water samples were carried out according to the Environmental Agency of the State of São Paulo directions—CETESB (2011) and Veridiana et al. (2008). More details in Vecchia et al. (2017b).

Approximately 1 kg of sediments samples, representative of the regional lithologies (Figure 1), were collected in depth from 50 cm to 80 cm, using a cylindrical collector. This material was transferred to low-density polyethylene (LDPE; Nalgon®) containers. Subsequently, these samples were dried, disaggregated, homogenized and sieved. Fractions < 250 Mesh Tyler were sent to isotopic Pb and chemical analysis.

### 3.2. Pb Isotopic Geochemical Analytical Procedure

Pb isotopic ratios of groundwater and sediments were determined through Thermal Ionization Mass Spectrometry (TIMS), using a multi-collector Finnigan MAT 262 at the Geochronological Research Center of USP (University of São Paulo) according to the Vecchia et al. (2017b).

### 3.3. Pb, U and Th Geochemistry Analytical Procedure

U, Th and Pb were analyzed in water and sediments samples by Inductively Coupled Plasma Mass Spectrometry, ICP-MS (PerkinElmer - ELAN DRC-e) at the Center for Development of Nuclear Technology (CDTN), according to the Vecchia et al. (2017b) and procedures recommended by the U.S. EPA 2008.

### 3.4. Multivariate Analysis

Data were submitted to a Principal Component Analysis aiming to establish relations between U, Th and Pb contents and Pb isotopic ratios (206Pb/204Pb, 207Pb/204Pb, 208Pb/204Pb, 206Pb/207Pb, 208Pb/207Pb) to the environment and their origin (geogenic or anthropogenic). The whole dataset (Tables 1-3 in Annex) consists of 12 sediment samples, 12 water samples (11 groundwater samples and 1 surface water sample) and 15 samples of Cordani et al. (1992) (07 São Timóteo Granite samples and 08 orthogneiss samples). Minitab® software, version 16.1.1.0, was used for statistical analysis.

### 4. Results and Discussion

**Multivariate Analysis**

Data (Tables 1-3 in Annex) were submitted to a Principal Component Analysis. This study was conducted in three steps. In the first one, U, Th and Pb
concentrations and Pb isotopic ratios of orthogneisses samples were analyzed together with data of groundwater and sediment samples. In the second step, only data of isotopic ratios from the same samples were used. In the third step, data of Pb isotopic ratios of São Timóteo granite were used together with data of groundwater and sediment samples. It is important to point out that these two types of rocks are characteristic of the geological formation of the study area and involve the genesis of uranium mineralization. Thus, they are important references of the geographic sources of Pb in the region. The use of these data aims to determine the type of rock that most influences the Pb isotopic signatures of groundwater and sediments and, therefore, which one better discriminates the environmental matrices investigated.

1) PCA using Pb isotopic ratios and U, Th and Pb concentrations in sediments, groundwater and orthogneiss samples

Table 1 presents the loading values of each variable in each of the components as well as their respective eigenvalues. These data can be interpreted in terms of two main components, PC1 and PC2, which account for 81.2% of the data variability. The first component (PC1) with an eigenvalue of 4.6145 accounts for 51.3% of the data variance. It is mainly dominated by isotopic ratios $^{206}\text{Pb}/^{204}\text{Pb}$, $^{207}\text{Pb}/^{204}\text{Pb}$, $^{206}\text{Pb}/^{207}\text{Pb}$, $^{206}\text{Pb}/^{208}\text{Pb}$ and by U. This component can be interpreted as representing the contribution of this element and its decay to the geochemistry of sediments and water. The second component (PC2) with an eigenvalue of 2.6974 accounting for 30.0% of the data variance is dominated by the isotopic ratios $^{208}\text{Pb}/^{204}\text{Pb}$, $^{208}\text{Pb}/^{207}\text{Pb}$ and by Th and Pb. This component can be understood as representing the contribution of Th and its decay in the general characterization of sediments and waters.

Table 1. Loading values of each variable in each of the principal components for samples of water, sediment and orthogneisses of the uraniferous province of Lagoa Real BA, for PCA carried out with all variables.

| Variable          | PC1  | PC2  |
|-------------------|------|------|
| $^{206}\text{Pb}/^{204}\text{Pb}$ | 0.440 | −0.152 |
| $^{207}\text{Pb}/^{204}\text{Pb}$ | 0.445 | −0.104 |
| $^{208}\text{Pb}/^{204}\text{Pb}$ | 0.212 | 0.480 |
| $^{206}\text{Pb}/^{207}\text{Pb}$ | 0.441 | −0.141 |
| $^{208}\text{Pb}/^{206}\text{Pb}$ | −0.416 | 0.230 |
| $^{208}\text{Pb}/^{207}\text{Pb}$ | −0.002 | 0.567 |
| U                 | 0.274 | −0.104 |
| Th                | 0.243 | 0.413 |
| Pb                | 0.247 | 0.401 |
| Eigenvalue        | 4.6145 | 2.6974 |
| Proportion        | 0.513 | 0.300 |
| Cumulative        | 0.513 | 0.812 |
Figure 2 shows the score plot of the first two components (PC1 and PC2) for water, sediment and orthogneiss samples. The score of a sample represents the contribution of each variable measured and its respective weights to the characteristics of the sample. Sediment and rock samples are grouped around the origin of the diagram, showing little difference from each other, which shows the geogenic character of the sediments, since in the process of weathering they did not undergo enrichment, i.e., anthropic influence. Sample P19S stands out from this group. Its high score values of PC1 and PC2 suggests enrichment of U and Th whose increase in concentration leads to an increase in the score value for the score of the samples in each component. Also noteworthy is the sample P35S with positive score value of PC1 and negative score value of PC2. In this case, the sample is enriched in U rather than Th, being the U enrichment due to anthropic influence. The water samples stand out from the group with negative score values for both PC1 and PC2. This means that, in general, the values of the variables for this matrix are low, which is a reflex of the low solubility of U, Th and Pb species (and their isotopes) in water. Samples P22AP is enriched in these species while P18A is enriched in Th. Samples P22AP and P18AP stand out from the rest of the groundwater samples, resembling the sediment and orthogneiss group of samples, probably because their isotopic signatures are under the geogenic influence of the orthogneiss.

2) PCA using Pb isotopic ratios in sediments, groundwater and orthogneiss samples

The same procedure was applied to the same samples, removing U, Th and Pb concentrations, i.e., working only with the Pb isotopic ratios as variables. Table 2 shows the loading values for both components. Concerning the isotopic ratios there is no difference between the results obtained with the two approaches, i.e., calculations with and without U, Th and Pb concentrations. Figure 3 and Figure 4 reflect this result. The distributions of samples in both diagrams are the same.
Table 2. Loading values of each variable in each of the principal components for samples of water, sediment and orthogneisses of the uraniferous province of Lagoa Real (BA), for PCA carried without U, Th and Pb concentrations.

| Variable          | PC1    | PC2    |
|-------------------|--------|--------|
| $^{206}$Pb/$^{204}$Pb | 0.499  | 0.003  |
| $^{207}$Pb/$^{204}$Pb | 0.685  | 0.000  |
| $^{208}$Pb/$^{204}$Pb | 0.150  | 0.693  |
| $^{206}$Pb/$^{207}$Pb | 0.499  | 0.019  |
| $^{208}$Pb/$^{206}$Pb | -0.485 | 0.099  |
| $^{208}$Pb/$^{207}$Pb | -0.094 | 0.714  |
| Eigenvalue        | 3.9889 | 1.8926 |
| Proportion        | 0.665  | 0.315  |
| Cumulative        | 0.665  | 0.980  |

Figure 3. Score plot of PC1 and PC2 for samples of water, sediment and orthogneisses of the uraniferous province of Lagoa Real BA, without U, Th and Pb concentrations, using Pb isotopic ratios.

Figure 4. Score plot of PC1 and PC2 for samples of water, sediment and granite of the uraniferous province of Lagoa Real BA, using Pb isotopic ratios.
Exception is sample P19S for which the concentrations of these three elements are especially higher. This suggests that the isotopic ratios are sufficient to assess the characteristics of samples. An additional observation due to the absence of the concentration variable in this step is the increase in the percentage of explanation of the variability of the data of the principal components PC1 and PC2, evidencing the importance and efficacy of the isotopic ratios in the study of environmental samples.

3) **PCA using Pb isotopic ratios data for sediments, groundwater and São Timóteo Granite samples.**

For this matrix, only data of Pb isotopic ratios were used. They refer to the samples of São Timóteo type granite, groundwater and sediments. **Table 3** shows the loading values of each variable in each of the main components.

These data can be interpreted in terms of two principal components, PC1 and PC2, which account for 97.8% of the data variability. The first component (PC1) with an eigenvalue of 3.9180 accounts for 65.3% of the data variance. It is mainly dominated by isotopic ratios $^{206}\text{Pb} / ^{204}\text{Pb}$, $^{207}\text{Pb} / ^{204}\text{Pb}$, $^{206}\text{Pb} / ^{207}\text{Pb}$, $^{208}\text{Pb} / ^{206}\text{Pb}$ the latter with negative loading value. This component can be interpreted as representing the contribution of U represented by its decay isotopes to the geochemistry of sediments. The second component (PC2) with an eigenvalue of 1.9517 accounting for 32.5% of the data variance is dominated by the isotopic ratios $^{208}\text{Pb} / ^{204}\text{Pb}$, $^{208}\text{Pb} / ^{207}\text{Pb}$, which are the decay products of Th. It represents the contribution of Th in the general characterization of sediments.

**Figure 4** shows the score plot of the first two components (PC1 and PC2) for water, sediment and granite samples. In general, the same observations made with the previous PCA study (**Figure 3**) can be summarized here: no separation of sediment and rock samples leading to their geogenic character, separation of water samples, separation of samples P22AP, and P18AP, P19S and one sample (P35S) due to anthropogenic enrichment.

According to Vecchia et al. (2017b) there is a great variation of Pb isotopic

**Table 3.** Loading values of each variable in each of the principal components for samples of water, sediment and granite of the uraniferous province of Lagoa Real BA.

| Variable       | PC1   | PC2   |
|----------------|-------|-------|
| $^{206}\text{Pb} / ^{204}\text{Pb}$ | 0.503 | −0.016 |
| $^{207}\text{Pb} / ^{204}\text{Pb}$ | 0.488 | 0.026 |
| $^{208}\text{Pb} / ^{204}\text{Pb}$ | 0.161 | 0.678 |
| $^{206}\text{Pb} / ^{207}\text{Pb}$ | 0.504 | −0.003 |
| $^{208}\text{Pb} / ^{207}\text{Pb}$ | −0.477 | 0.176 |
| $^{208}\text{Pb} / ^{206}\text{Pb}$ | −0.040 | 0.713 |
| Eigenvalue     | 3.9180| 1.9517|
| Proportion     | 0.653 | 0.325 |
| Cumulative     | 0.653 | 0.978 |
signatures for both sediment and groundwater samples related to the geogenic sources (representative of the regional lithologies). However, the variation for the sediments is significantly higher. Moreover, among the geogenic sources it was observed that the Pb isotopic signatures for sediments and groundwater, from the same geographical coordinate, indicated a natural enrichment in U and Th. The values of the majority of the Pb isotopic signatures for groundwater are considered low, in spite of being in accordance with the geogenic sources of the region, which are under the influence of granites. One sediment sample (P35S), collected in the U mining area, presented isotopic Pb signature typical of U anthropogenic sources (high $^{206}$Pb/$^{204}$Pb ratio compared to $^{208}$Pb/$^{204}$Pb ratio), which means U contamination. However, the PCA analysis proved that this was not the behavior evidenced by the P19S sample, which presented isotopic Pb signature in accordance with the geogenic sources of the region.

5. Conclusion

One of the major vulnerabilities of the nuclear area and/or mining activities to public opinion refers to frequent suspicions that nuclear and/or mining industry always generate(s) large environmental impacts, especially contamination of groundwater. These kinds of worries can find legal resonance even without the necessary pre-operational background data, and most of the decisions consider the analytical results of radionuclide concentrations from the operational phase as the main incriminating technical argument. The present study showed that the determination of the concentration of the contaminants is not enough to characterize environmental contamination. It also shows that the most effective analytical method to elucidate the role of anthropogenic interferences in the environment is the determination of isotopic ratios. It can even be used in environments with high concentrations of radionuclides.

This tool becomes even more relevant in regions where pre-operational data are scarce and incomplete, if not inexistent, or even in regions, such as Caetité (Bahia), where water sampling is very difficult due to low water availability. In addition, it has been proved that the Pb isotopic tool, besides being used in any type of environmental matrices, is still promising in the investigation of industrial environmental impacts from mining as well as from physical, chemical and thermal processes where the generation of NORM (Natural Occurring Radioactive Material) wastes is related.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.
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