Brazil’s Participation in the Galaxy Serpent Exercise

Jorge Eduardo Souza Sarkis, Isabella Cristina Antunes Costa Bordon,
Rafael Cardoso Baptistini Pestana, and
Rafael Coelho Marin
Instituto de Pesquisas Energéticas e Nucleares, São Paulo, Brazil

Abstract

The process of nuclear fission is governed by well-understood physical laws that may cause specific and predictable changes in the nuclear fuels resulting from, for instance, the type of nuclear reactor, fuel type and its irradiation history, or aspects associated with energy production through the buildup of fission products and the transmutation of heavy metals.

All such information, compiled in a national nuclear forensic library (NNFL), can be an important tool during the identification of a seized unknown sample, allowing linking of information concerning its irradiation history, the type of reactor or even the origin of the sample.

The usefulness of an NNFL depends on not only the quantity or quality of the available data, but also on the capacity of the investigators to identify, correlate, and interpret the main characteristics identified, or measured, in the seized sample. This paper describes the strategy adopted by the Brazilian team during the virtual, web-based Galaxy Serpent Exercise,[1] coordinated by the Nuclear Forensics International Technical Working Group, where an NNFL was developed and used to identify a hypothetical unknown sample. Our experience demonstrated the importance of knowledge of nuclear reactions in order to identify parameters that are most relevant during the technical evaluation. Then, using these, the importance of simple isotopic correlations can be used to verify the consistency of the available information before invoking more complex multivariate statistical techniques. Based on our investigation the hypothetical seizure, which was determined to come from a boiling water reactor, was conclusively found not to have originated from a reactor in our model NNFL.

Introduction

Since nuclear energy was first utilized, the risk related with criminal or unauthorized acts involving nuclear or other radioactive (RN) materials have been a serious concern for the entire global nuclear community. During the last decades, the possibility that terrorists or other criminals might obtain RN materials for malicious use has become a real threat to global security especially after the collapse of the Soviet Union, at the end of the 1980s, when several tons of these materials were stolen.[2] This concern was enhanced in the early years of the 1990s when several attempts of illicit trafficking of these materials were identified.[3] The inclusion of nuclear materials into classical forensic investigations led the definition of a new science called “nuclear forensic science.”[4] Nuclear forensic science is a branch of criminology that deals with crimes involving nuclear or other radioactive materials. The main objective of this new science is the identification of the nature and origin of the seized material, and of any intent to use it, and requires collaborating efforts of different technical and scientific expertise.

Once seized, the RN material is typically sent to a nuclear research laboratory where material characterization is performed that involves measurements of chemical composition, physical characteristics, and isotopic abundance.

The next step is to identify its possible origin. For this purpose, one method is to compare the obtained results with those existing in a nuclear database, which may be included in a national nuclear forensic library (NNFL). The NNFL gathers all information (measured or modeled) of nuclear and other radioactive material produced, used, stored, or transported within a nation. In this way, in the event of an actual investigation, materials data obtained with evidence can be easily and quickly compared, traced, associated or even identified among materials data already cataloged.[5]

However, the usefulness of an NNFL depends not only on the quantity but also the quality of the catalogued data, as well as the ability of the nuclear forensic examiner to use and adequately interpret the data and relationships arising from the investigation. With this objective, the Nuclear Forensics International Working Group organized the Galaxy Serpent international virtual, web-based, tabletop exercise[1] and, this paper will describe the experience and results obtained by the Brazilian team.

Method

During the exercise, the Brazilian team was designated by the galaxy code name “Draco.” Draco’s team received a database of fuel composition and burnup values for three hypothetical reactors: Hyperion (twelve samples), Daphnis (eighteen samples), and Ijiraq (two samples). The database was used to construct an NNFL. However, the provided data had discrepancies related not only with the number of samples for each reactor, but also the number of parameters provided for each reactor (circa eighty for Daphnis, forty-three for Hyperion, and eighty-two for Ijiraq). The initial enrichment values of the fuel samples were not provided.

The strategy employed for the exercise was divided in three main steps: the first step was to identify, among the available data, classical isotopic correlations found in the literature in order to verify the consistency of data provided in the fuel sample database of the three nuclear reactors. The second step involved the use of multivariate statistical analysis to identify, in the database, the main parameters that could be used to distinguish spent fuel from one reactor from the other reactors. The last step was to add to the model the samples to be investigated and use the canonical discriminant analysis in order to confirm the results.

Isotopic Correlation Technique

The isotopic correlation technique (ICT) was developed during 1970-1980 for safeguards purposes.[6,7] It is based on the fact that nuclear fission is governed by very well-known physical processes. Such processes involve parameters such as type of reactor, fuel, and irradiation history, and more importantly, several of these parameters can be correlated, basically, by first order differential equations. Thus, the ICT can be an important tool used in precursor data analysis during the initial screening stage of a nuclear forensic investigation.

Multivariate Statistical Analysis (MSA)

MSA refers to any statistical technique used to analyze data that arises from more than one variable. These are powerful tools to identify dominant groups of variables in a set of data.[8]

In this work, the data interpretation was performed using two different MSAs approaches: Principal Component Analysis (PCA) and Canonical Correlation.

PCA is one of the most popular MSA methods used for data pre-processing (i.e., exploratory data) and reduction from a larger set of variables. In general, the results are present in two- or three-dimensional plots of the data for visual examination and interpretation. In the context of this work, PCA was used to reduce the numbers of the parameters and select the most representative ones for the establishment of the overall model of data distribution (or grouping).[9]

The remaining parameters were analyzed by Canonical Correlation Analysis (CCA). In the context of this work, CCA is used as an additional procedure for assessing the relationship, or differences, between variables, classes or groups of variables.[10]

Results and Discussion

The consistency, and quality, of the provided data were evaluated using some classical isotopic equations, present in the literature. 2,3,4 It is important to note that our data sets were also not consistent and did not have a complete set of information of each reactor. This is due largely due to the data sets used in the exercise originating from the online, public domain Spent Fuel Compositions (SFCOMPO) database,[11] which was designed to provide post-irradiation data to validate fuel depletion methodologies, and not with nuclear forensics applications in mind. However, it is noted that for actual nuclear forensics data that is repurposed from existing data, coverage, completeness, and consistency will vary. As a result, it was clear that the direct use of the available equations would have to be considered in a conservative way.

The second step was to consider not the previous mathematical equations, but the correlation existing among the parameters resulting from the chemical and isotopic characterization (IC) of the fuels present in the training set. Several first and second order differential equations were tested and correlations were identified with high degree of consistency.

In order to demonstrate the usefulness of this assessment a few examples were selected and are presented below.

The first IC to be evaluated is^{242} PU/ 240 Pu versus^{240} Pu/ 239 Pu as proposed by Christensen and others[12] for all types of PWR reactors (Figure 1).

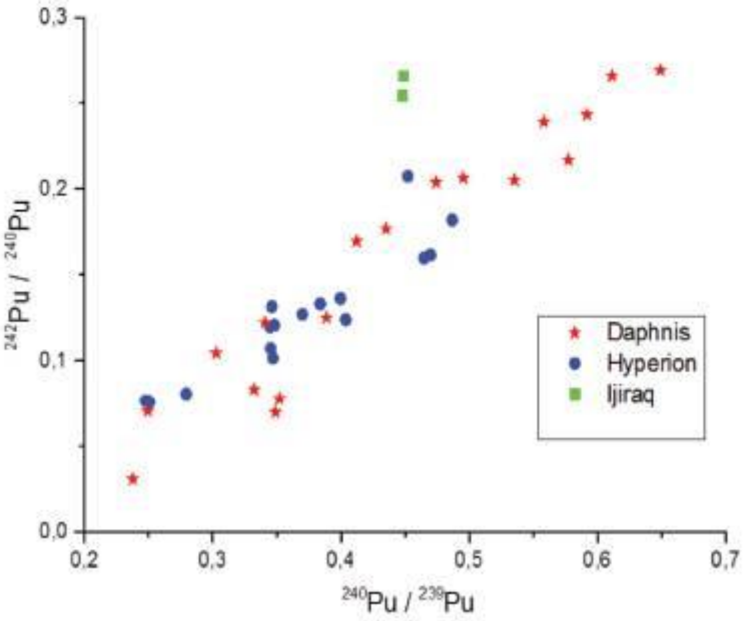

Figure 1. Isotope Correlation:^{242} Pu/ 240 Pu versus^{240} Pu/ 239 Pu

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As shown in Figure 1, the parameters correlated well except for Ijiraq reactor, which is likely due to the low number of samples available. However, after a more careful inspection of Figure 1, for Daphnis and Hyperion there is a slight difference in the alignment amongst the distribution of the data throughout the plot.

This characteristic was confirmed in other IC plots involving plutonium isotopic data, such as one more recently defined by Moody and others,[13] which was applied here and presented in Figure 2 with the confidence band of 95 percent. In order to better understand these graphs, Figure 1 was split in two separate graphs containing the samples of each reactor presented individually, as shown in Figure 3. Figure 3 shows three different groups of samples in Daphnis, and two groups in the Hyperion data set, respectively.

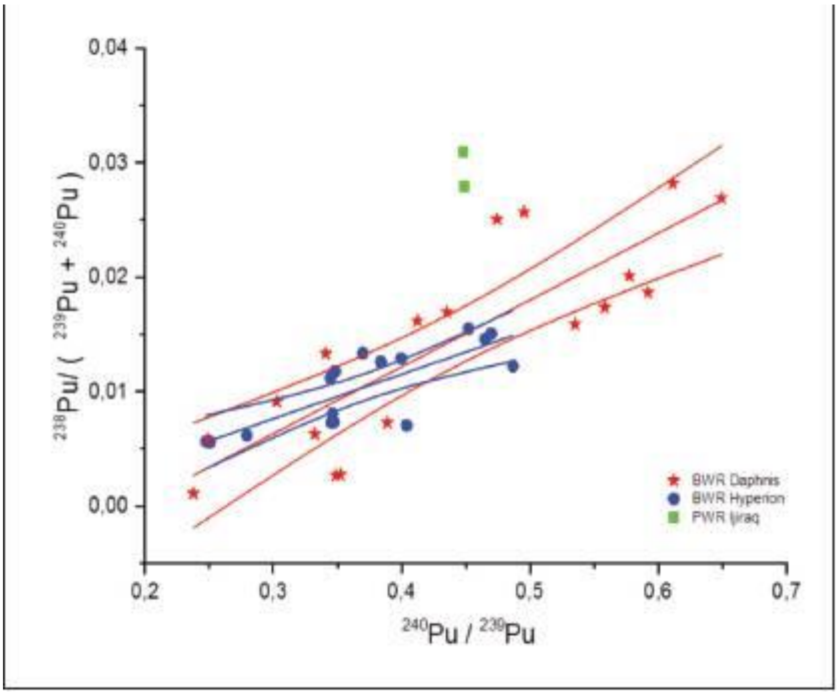

Figure 2. Isotopic Correlation:^{238} Pu/ (^{239} Pu+^{240} Pu) versus^{240} Pu/ 239 Pu

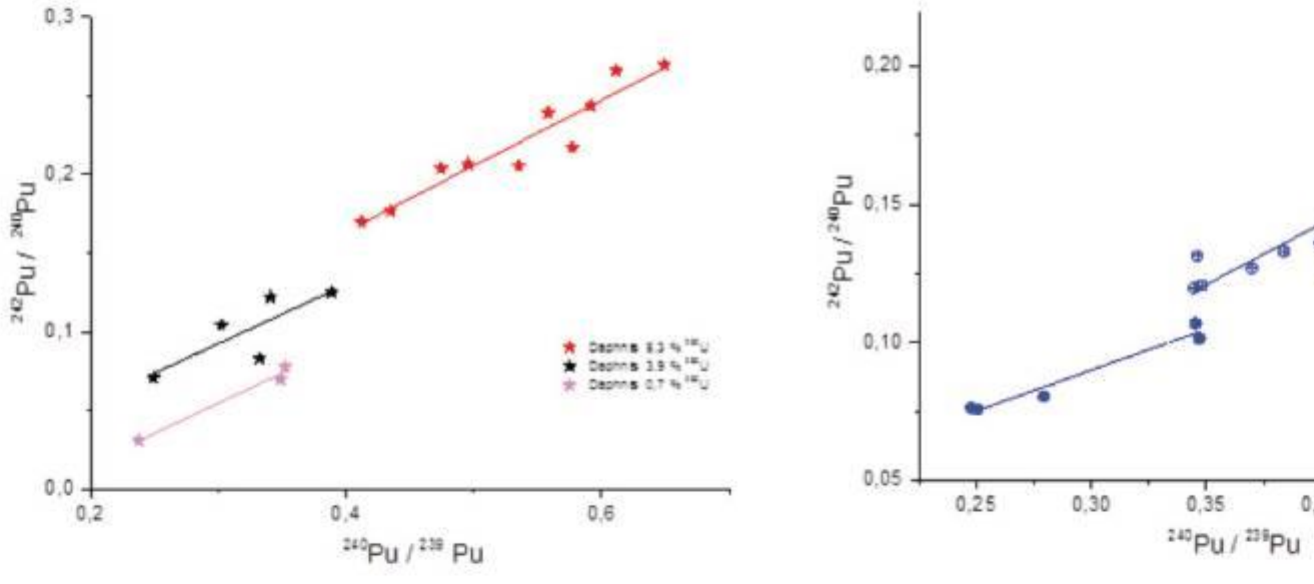

Figure 3. Daphnis and Hyperion:^{242} Pu/ 240 Pu versus^{240} Pu/ 239 Pu

Although the initial enrichment was not provided, these data were, initially, considered important for the samples classification. Thus, the initial enrichment of samples was estimated based on the correlation among isotope ratios 235 U/ 238 U versus^{236} U/ 238 U (Figure 4).

As shown in Figure 4, Daphnis' samples presented three different enrichments and, except for the ones with natural uranium (fuel poison), exhibited burnup levels from 16.7 to 44 GWD/MT. Such a range of burnup values were likely due to exposure in different regions of the nuclear fuel assembly. Hyperion's samples have the same initial enrichment, but were separated in two groups with different burnup levels and plutonium profiles. There is a possibility that the samples may originate from the same fuel rod, but from different positions along the active length of the rod.

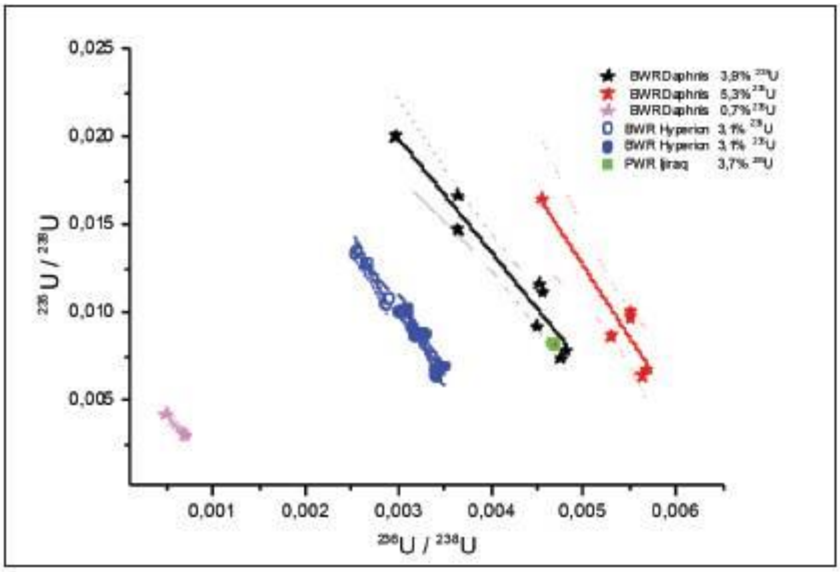

Figure 4. Estimates of initial sample enrichments based on isotope correlation

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Based on our experience, for this portion of the exercise some partial conclusions have been identified. The use of the former IC's mathematical equations has to be considered in a conservative way, because majority of them were developed more than thirty years ago for different purposes than nuclear forensics. Consistency of the data present in the database was demonstrated. Although the mathematical correlations obtained from the data set did not completely agree with those found in the literature, the parameters themselves exhibited the same behavior as described in previous documents. The use of isotopic correlations for a preliminary evaluation of the database was also demonstrated. Several correlations were evaluated and the results were quite consistent. Despite the quantity of the available information, subtle differences in the initial enrichment as well as in the irradiation history can be identified. It is also clear that the quality of these evaluations is directly related to the amount and the quality of the provided data.

Principal Component Analysis

After data reduction, a PCA model was developed with the following parameters:^{236} U/ 235 U,^{235} U/ 238 U,^{236} U,^{238} Pu,^{239} Pu,^{240} Pu, 241 Pu, 242 Pu, Pu-total, 240 Pu/ 239 Pu, 241 Pu/ 239 Pu, 242 Pu/ 239 Pu, 241 Pu/ 240 Pu,^{242}

Pu/ 240 Pu,^{242} Pu/ 241 Pu and is shown in Figure 5. As shown, the PCA model was able to distinguish, with a high degree of confidence, each reactor as well as each class of samples.

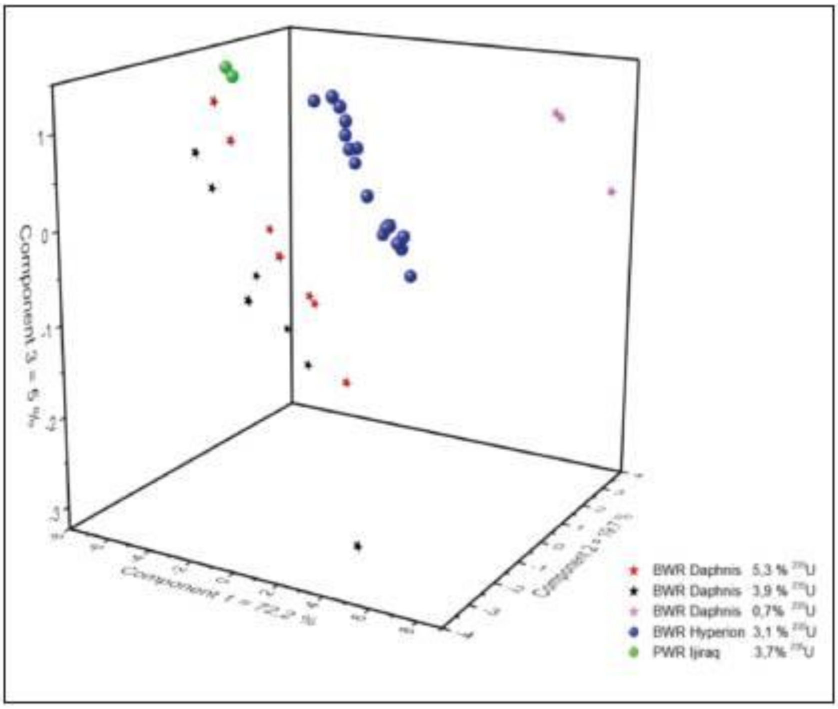

Figure 5. PCA plots for the NNFL reactors

Seized Material: Clio

In Phase 2, the hypothetical seizure, denoted as Clio, was distributed with circa twenty-five parameters. Initial enrichment was calculated based on^{235} U/ 238 U versus^{236} U/ 238 U correlations (circa 3 percent). Isotopic correlations were developed and compared. Results from a few examples are presented in Figures 6 and 7. The correlations shown in Figure 7 indicate that Clio most likely originates from a BWR, as suggested by its grouping with the BWR reactors Hyperion and Daphnis. PCA was performed with the same data as the previous model (Figure 8).

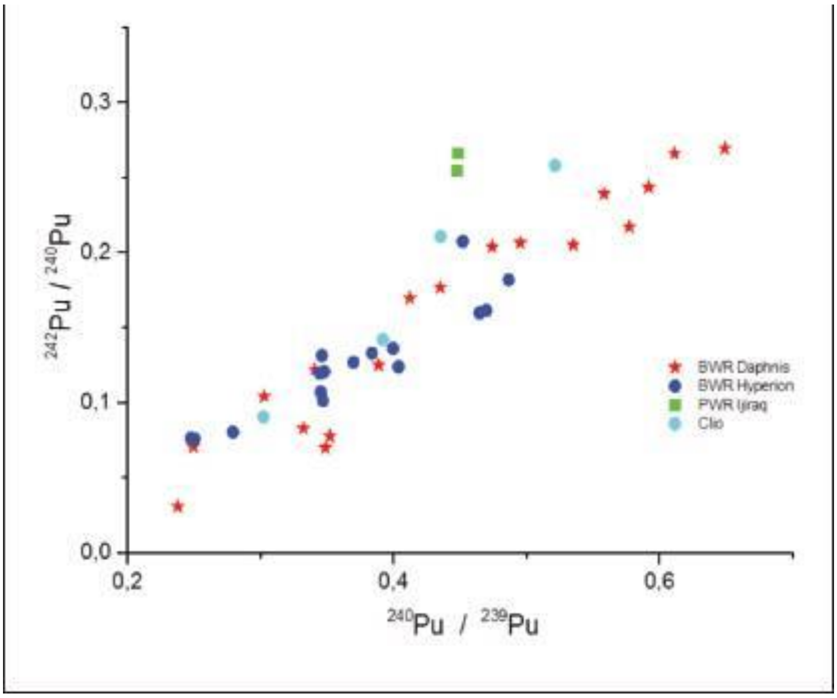

Figure 6. Isotope Correlation:^{242} Pu/ 240 Pu versus^{240} Pu/ 239 Pu for NNFL reactors and Clio seizure

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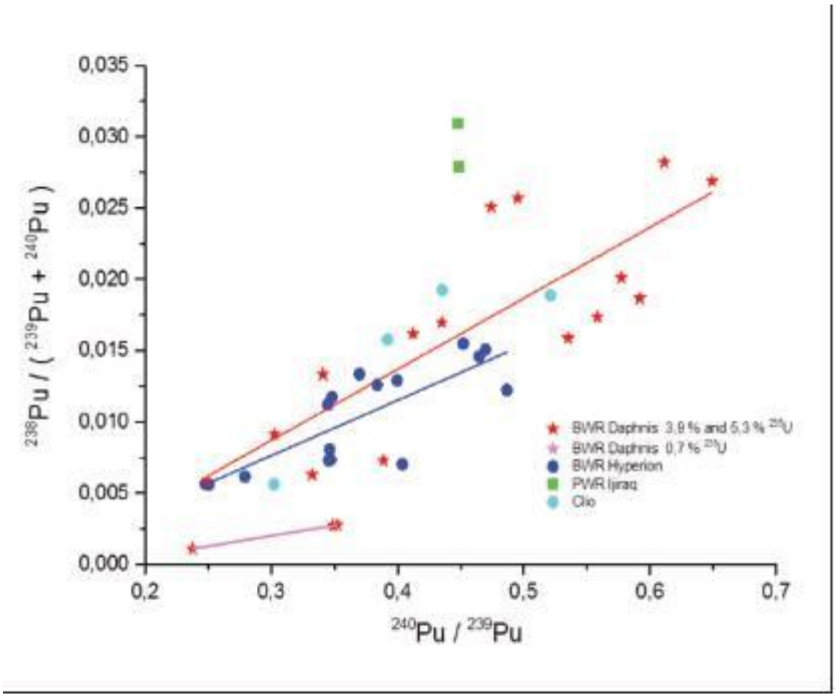

Figure 7. Isotope Correlation:^{238} Pu/(^{239} Pu +^{240} Pu) versus^{240} Pu/ 239 Pu for NNFL reactors and Clio seizure

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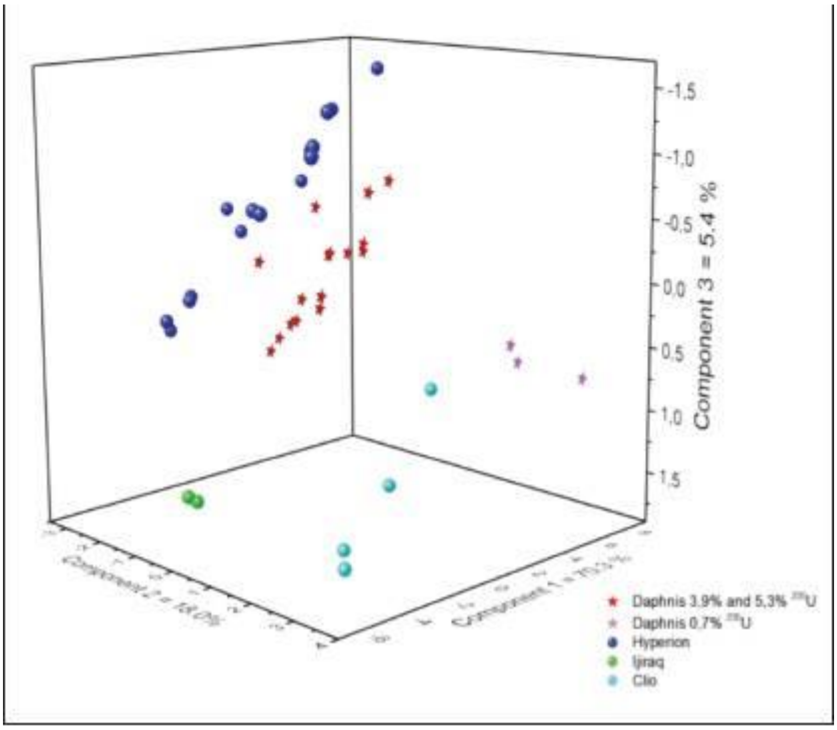


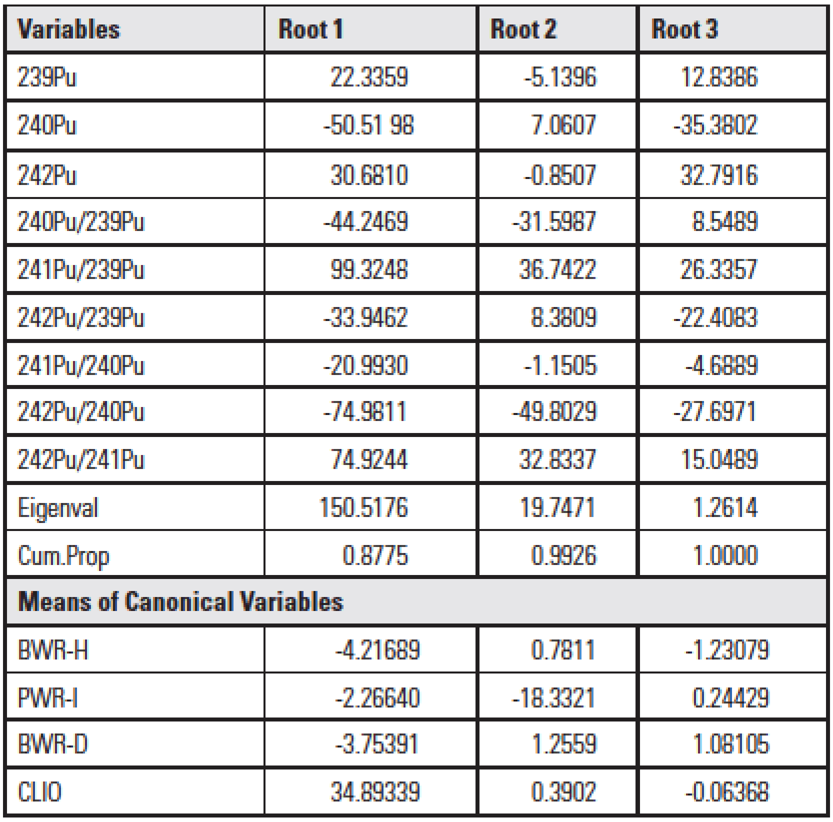
Figure 8. PCA of the NNFL's reactors and Clio

As shown in Figure 9, the variability of the Clio fuel samples tended to group in a different region compared with other fuel compositions from our NNFL, and, based on this analysis, it seems that the Clio fuel composition is not consistent with any reactor fuel in our NNFL.

Canonical Discriminant Analysis

The final evaluation was made using a canonical discriminant analysis using only the main parameters that influenced the previous PCA models. The main results obtained with this analysis are presented in the table below (Table 1). As shown in Table 1, based on the available data set, the main correlated parameters identified with this statistical tool to distinguish the Clio samples from the other fuels were plutonium isotope ratios. These results are in good agreement with the existing literature 15,16 and are shown in Figure 9.

Table 1. Standardized coefficients for canonical variables



Variables	**Root 1**	**Root 2**	**Root 3**
239Pu	22.3359	-5.1396	12.8386
240Pu	-50.51 98	7.0607	-35.3802
242Pu	30.6810	-0.8507	32.7916
240Pu/239Pu	-44.2469	-31.5987	8.5489
241Pu/239Pu	99.3248	36.7422	26.3357
242Pu/239Pu	-33.9462	8.3809	-22.4083
241Pu/240Pu	-20.9930	-1.1505	-4.6889
242Pu/240Pu	-74.9811	-49.8029	-27.6971
242Pu/241Pu	74.9244	32.8337	15.0489
Eigenval	150.5176	19.7471	1.2614
Cum.Prop	0.8775	0.9926	1.0000
Means of Canonical Variables			
BWR-H	-4.21689	0.7811	-1.23079
PWR-I	-2.26640	-18.3321	0.24429
BWR-D	-3.75391	1.2559	1.08105
CLIO	34.89339	0.3902	-0.06368

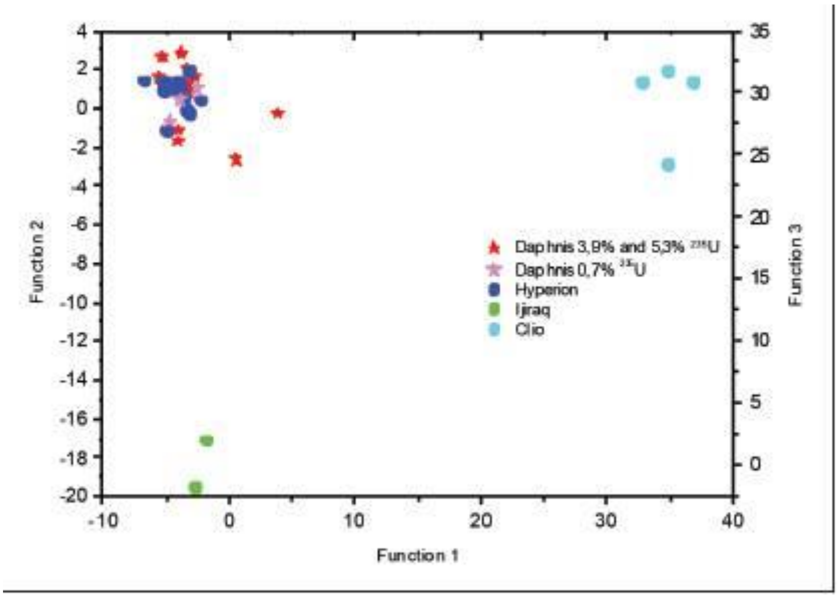


Figure 9. Canonical distribution: NNFL and Clio sample

Conclusions and Lessons Learned

Based on our evaluation it was possible to determine the reactor class (BWR or PWR) pertaining to the hypothetical seizure Clio, and also whether Clio was or was not consistent with a reactor in our developed NNFL. Using ITWG established confidence levels,[13] the Clio sample originated from a BWR reactor with a “suggestive positive” confidence, and the Clio sample was found to not originate from a reactor in the NNFL created by the Brazilian team with a “conclusive negative” confidence level.

Furthermore, the participation of the Brazilian team in this exercise brought more learning and conclusions.

As it was demonstrated, an NNFL can have probative value as part of a nuclear forensic investigation, whether comprised of basic information concerning the isotopic profile of the investigated material, or involving more complex statistical approaches formed from larger data sets. In practical terms, this means that is it possible to advance an actual investigation with even limited information.

However, the most important conclusion is related to the significance of the exercise itself. Namely, considering that global nuclear security is the responsibility of all nations and the transnational nature of many smuggling events, a nuclear security event will likely involve more than one country. It is important to establish a common approach with respect to issues such as how to face a threat, which information can or may need to be exchanged among the involved parties, the quantity and quality of the available information and, most importantly, how to work together. Without a common strategy, or having to begin such collaboration during an actual crisis, our capability to effectively respond will be significantly impacted, so that it may be prejudiced, or even not productive.

In such a scenario, the establishment of a national nuclear forensic library prior to a nuclear security event may be an important tool to start the dialogue among all parties involved to help address key questions pertaining to such an event and also provide a framework which facilitates effective cooperation and procedures.

Jorge Eduardo de Souza Sarkis is senior scientist of the Nuclear and Energy Research Institute (IPEN), Brazil, and associated professor in nuclear science, at São Paulo’s University. He has more than thirty years of experience in nuclear science and analytical chemistry and is author of more than 160 scientific publications, including peer-reviewed articles, book chapters, conference papers, and approximately 100 conferences abstracts. From 1984 to 1986 he was a research fellow at the Institute for Transuranium Elements (ITU) of the European Commission Joint Research Centre, Karlsruhe, German. In 1999, he was a postdoctoral researcher at Los Alamos National Laboratory, New Mexico, USA. Since 2008 he has been the coordinator of the Brazilian Network of Laboratories on Nuclear Forensic Science.

Isabella Cristina Antunes Costa Bordon has an MSc in biology and received her PhD in nuclear technology in 2014, from the Nuclear and Energy Research

Institute (IPEN), São Paulo's University. Her current research interest is biomonitoring.

Rafael Cardoso Baptistini Pestana is a chemist at the State University of Maringá, Brazil, and has a master's degree in nuclear technology from the Nuclear and Energy Research Institute (IPEN), University of São Paulo (USP). He has been working in the identification of nuclear material signatures in environmental samples for safeguards and nuclear forensics purposes.

Rafael Coelho Marin is a physicist at the University of São Paulo and has a master's degree in nuclear technology from the Nuclear and Energy Research Institute (IPEN), University of São Paulo, Brazil. He has been working in identification of nuclear material signatures in environmental samples for safeguards and nuclear forensics purposes.

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